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THE FERNALD DOSIMETRY RECONSTRUCTION PROJECT

Tasks 2 and 3

Radionuclide Source Terms and Uncertainties

Submitted to the Centers for Disease Control and Prevention
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by

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June 1995

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GLOSSARY OF TERMS AND ACRONYMS

Each term or acronym is in italics the first time it appears in the text.

Activation products are radionuclides that result from the absorption of neutrons by uranium and other materials present in a nuclear reactor. An example is plutonium-239 produced following neutron absorption by uranium-238.

AMAD – Activity median aerodynamic diameter, a measure of particle size.

AMS – Air monitoring stations

Anisokinetic sampling –refers to a mismatch between the air or fluid velocity in the sampling probe and that in the stack releasing airborne effluents. It is a source of bias in effluent sampling. In contrast, *isokinetic sampling* results in an unbiased sample of the stack effluent.

Assessment Domain is the region surrounding a facility for which radiation doses to people are calculated; for this project, a circular region with a radius of 10 kilometers (km) (6.25 mi.) with its center in the FMPC production area.

Background Radioactivity – refers to radioactive elements in the natural environment including those in the crust of the earth (like radioactive potassium, uranium and thorium isotopes) and those produced by cosmic rays.

Bias is a systematic distortion of measurements that makes the results inaccurate.

CDC – Centers for Disease Control and Prevention, who funded the Fernald study.

Chemical Symbols are abbreviations for different elements and compounds. Examples of elements include U for uranium, O for oxygen, N for nitrogen and F for fluorine. Examples of compounds include UF_4 for uranium tetrafluoride (green salt) and UO_3, or uranium trioxide (orange oxide).

Contamination refers to unwanted radioactive material, or to the deposition of radioactive material in the environment or in any place where it may make surfaces or equipment unsuitable for some specific use.

Decay (daughter) products refer to the isotopes or radionuclides that result from radioactive decay of isotopes, such as the uranium and thorium isotopes. In most of the feeds received by the FMPC, the uranium had previously been separated chemically from the other decay products. As a result, the facility's effluents consisted primarily of uranium, and decay product radionuclides were generally present in small quantities. In naturally-occurring uranium ores, the decay products include isotopes of uranium, protactinium, thorium, radium, radon and radon daughter products. *Radon daughter products* that are derived from uranium are the short-lived decay products from radon-222, and include polonium-218, lead-214, bismuth-214 and polonium-214.

Denitration – a process in Plant 2/3 in which nitrates were driven off by heating uranyl nitrate hexahydrate (UNH) to produce uranium trioxide (UO_3, or orange oxide).

Derbies are masses of uranium metal fabricated in Plant 5. The derbies were then remelted and cast into ingots of metallic uranium.

Direct exposure – refers to one pathway of exposure of people to radiation from the FMPC. In this exposure pathway, penetrating radiation emitted from radioactive material is partially absorbed by individuals exposed to it. The amount of exposure decreases with distance from the source. An example is gamma radiation from the K-65 silos that resulted in low-level exposure of nearby residents.

DOE – U.S. Department of Energy

Dose is a general term denoting the quantity of radiation or energy that is absorbed by the body. There are technical terms with specific definitions, such as absorbed dose, dose equivalent, effective dose, etc.

Dust Collector is one type of filtration system for airborne effluents used at the FMPC to remove airborne particulate material before it was discharged through the stack to the outside. The filtering medium is similar to that used for large fiber vacuum cleaner bags.

Effluent is a gas or liquid containing contaminants that flows from a process, building or the site into the surrounding environment.

Empirical values are values which are measured (as opposed to theoretically determined or calculated values).

Enrichment of uranium – a process by which the relative abundances of the isotopes of uranium are altered, thereby producing a form of the element that has been enriched in one particular isotope and depleted in its other isotope. For example, natural or "normal" uranium contains 0.72% ^{235}U. Enriched uranium contains more than the natural concentration of ^{235}U, while depleted uranium contains significantly less than 0.72% ^{235}U.

Entrainment is a process in which the uranium-containing liquid droplets in a scrubber are carried by the exhaust air stream and are vented to the atmosphere with the exhaust gases.

Environmental exposure – exposure to radiation through environmental pathways.

Epidemiology – the study of diseases in human populations.

Fission products are radionuclides that result from the splitting of heavy elements like uranium in a nuclear reactor. Examples are strontium-90 (^{90}Sr), technetium-99 (^{99}Tc), ruthenium-106 (^{106}Ru) and cesium-137 (^{137}Cs).

FDRP – Fernald Dosimetry Reconstruction Project

FEMP – Fernald Environmental Management Project, the new name of the FMPC beginning in 1991.

FMPC – Feed Materials Production Center

GM – Geometric Mean, or median, the central point of a distribution. Half of the values are larger than the median value and half are smaller.

GSD – Geometric Standard Deviation, a measure of the spread of a distribution. A large GSD indicates a wide range of measured or calculated values.

Grab samples – samples, usually of relatively small volume, taken at random or at preselected frequencies. These samples define the concentration of a contaminant at the specific time when they are collected and differ from continuous or proportional samples which are intended to reflect the time averaged value.

Great Miami River is the major water flow near the Feed Materials Production Center (FMPC) that receives most of the liquid effluents from the FMPC. The river, located about a mile east and south of the FMPC, runs in a southerly direction and enters the Ohio River approximately 18 miles (29 km) downstream of Cincinnati. Upstream of the FMPC on the Great Miami River lie the communities of Fairfield, Hamilton, Middletown, and Dayton. The flow of the river at the Hamilton gauge averages 3300 cubic feet per second (cfs) (93.4 $m^3\ s^{-1}$) with a maximum of 352,000 cfs (9970 $m^3\ s^{-1}$) measured in March 1913 and a minimum of 100 cfs (2.8 $m^3\ s^{-1}$) measured in September 1941.

Green salt is the common name for uranium tetrafluoride (UF_4), the product from the Plant 4 operations that was sent to Plant 5 for conversion to derbies.

Gulping operations refers to a process in Plant 2/3 in which orange oxide (uranium trioxide, or UO_3) from the denitration pots was transferred by a vacuum hose to a storage hopper. It appeared that the hose was "gulping" the orange oxide.

IH&R – Industrial Hygiene and Radiation Department at the FMPC

ICRP – International Commission on Radiological Protection

IT – International Technology Corporation

K-65 Silos – The *K-65 Storage Silos* are large concrete tank-like structures that store residues from the extraction of uranium from ores that were processed during the early years of FMPC operations.

kilo – a prefix that multiplies a basic unit by 1000. For example, 1 kilogram = 1000 grams.

Lognormal distribution – If the logarithms of a set of values are distributed according to a normal ("bell-shaped") distribution the values are said to have a lognormal distribution, or be distributed "lognormally".

MTU – abbreviation for metric ton of uranium; one MTU equals 1,000 kg or 2,200 pounds

NCRP – National Council on Radiation Protection and Measurements

NKES – Northern Kentucky Environmental Services

NLO – National Lead Company of Ohio, the contractor for the FMPC through the end of 1985.

NO_x – nitrogen oxides, such as NO_2 and NO_3.

ODH – Ohio Department of Health

Orange oxide – abbreviation for uranium trioxide (UO_3), the product from the Plant 2/3 refinery that was sent to Plant 4 for further processing.

OSTI – the Office of Scientific and Technical Information, located in Oak Ridge, Tennessee, is the national center for worldwide literature on scientific and technical energy-related matters. It was one of the sources of information that *RAC* used for completion of the project.

Paddy's Run – a small intermittent stream lying along the west boundary of the site that joins the Great Miami River approximately 3 kilometers south of the FMPC. The flow in Paddy's Run, which generally exists only during January to May, averaged 2 to 4 cfs (0.065 to 0.1 $m^3\ s^{-1}$). Since flow in Paddy's Run is dependent upon rainfall, discharges from the site to Paddy's Run generally occurred during periods of heavy rain and runoff when the storm sewer outfall overflowed, or when runoff from the west side the of site flowed into the Paddy's Run.

pico – a prefix that multiplies a basic unit by 1/1,000,000,000,000 or 1×10^{-12}. For example, one picocurie (pCi) equals 1×10^{-12} curie (Ci).

RAC – *Radiological Assessments Corporation* was the group chosen by CDC to do the Fernald Dosimetry Reconstruction Project.

Recycled uranium is uranium that had been irradiated in nuclear reactors, where finished uranium products were used. As a result, when the uranium was recovered and returned to the FMPC, small amounts of fission and activation products were introduced into the process stream.

Reentrainment is a process whereby the exhaust airflow creates new droplets from liquid that had been previously collected by a screen type filter.

Scrubber – a type of treatment system for airborne effluents that uses liquid droplets to remove particulate matter and reactive gases from airborne waste streams before they were discharged through the stack to the outside. At the FMPC, scrubbers were used in Plant 2/3 (refinery) and in Plant 8 (scrap recovery).

Scrub Liquor – the *scrub liquor* is the liquid in a scrubber that cleans or scrubs the exhaust air from certain plant operations. The liquid removes reactive gases and particles in the airstream before the airstream is discharged to the atmosphere.

South Plume –refers to the groundwater that has been contaminated by uranium from the FMPC. It extends southward from a point south of the waste pits and reflects the movement of contaminated groundwater.

Source Term – refers to the quantity, and chemical and physical form of radioactive materials released to the environment from various locations onsite.

SSOD – The Storm Sewer Outfall Ditch is a drainage ditch that runs south from the FMPC production area near the storm sewer lift station to Paddy's Run.

TLD – A thermoluminescent dosimeter is a device used at the FMPC to measure the amount of external radiation in the environment. These devices measure both radiation from naturally-occurring radioactivity in the soil and from the K-65 silos.

TRU – transuranic nuclides refer to isotopes heavier than uranium that are created by neutron capture by heavy elements.

Uncertainty –term used to describe probable bounds on, or how much evidence we have to support, our key findings. Uncertainty can result from two process: the first is due to random variations in sampling, measurement, and operational procedures. The second type of uncertainty occurs because of a lack of information about particular processes. This may occur because the right measurements were not done during part or most of the period of facility operation.

UF_4 – uranium tetrafluoride, or green salt was the product from Plant 4 that was sent on to Plant 5 for conversion to derbies.

UNH – uranyl nitrate hexahydrate was an intermediate step in the denitration process in Plant 2/3; nitrates were removed from UNH to produce uranium trioxide (UO_3, or orange oxide).

UO_3 – uranium trioxide, often called orange oxide, was produced in the Plant 2/3 refinery and was sent to Plant 4 for further processing.

$UO_2(NO_3)_2$ – uranyl nitrate was a product of the digestion phase in the Plant 2/3 refinery.

USGS – United States Geological Survey

Validation is the comparison of available measurements of the radionuclides in the local environment during the period of study with corresponding predictions from mathematical models.

WMCO – Westinghouse Materials Company of Ohio, the FMPC site contractor from 1986 through 1992.

EXECUTIVE SUMMARY

The purpose of the Fernald Dose Reconstruction Project (*FDRP*) is to estimate radiation doses to people who lived near the Fernald (Ohio) Feed Materials Production Center (*FMPC*) during its years of operation from 1951 to 1988. Exposures resulted from both planned and unplanned releases of radionuclides to the environment. The study was conducted for the Centers for Disease Control and Prevention.

The project was divided into seven tasks. The goal of Task 2 was to determine the radionuclide source term for the facility; that is, to determine both the amounts of radioactive material released to the environment and the variability of release rates. The Task 3 objective was to determine the uncertainties associated with those past releases.

This final report describes our estimates for source terms for the period 1951–1988. In finalizing this report, *RAC* has considered comments and suggestions received from a number of sources on our draft report (Voillequé et al. 1993). Initially we examined a three-year period in the early sixties to develop the methods that would be applicable to all years (Voillequé et al. 1991).

Our calculations are based on a thorough search of records documenting operations and effluent and environmental monitoring at the FMPC. In some cases, effluent measurement data from which estimates could be derived directly were not available. These situations were handled using statistical methods that simulate a possible range of values that could have existed. Source terms were divided into three categories of release: emissions to air, emissions to surface water, and *contamination* of groundwater.

The principal activity at the FMPC was processing uranium (U), with some thorium processing occurring at various times. In the early years, uranium ore was processed, and the waste materials were stored in drums and silos onsite. These waste materials are a source of radon and its decay products. Consequently, this report focuses primarily on emissions of uranium, and radon and its decay products. Some uranium was recycled, which is uranium that had been returned to the FMPC from other weapons material processing facilities. As a result, other radionuclides were also released at the site. Thus, release estimates are given for thorium, and selected activation products (plutonium-238, plutonium-239,240, neptunium-237), fission products (strontium-90, technetium-99, ruthenium-106, cesium-137), and decay products of uranium (radium-226) and thorium (radium-228). Table ES–1 summarizes the most important (uranium and radon) source term estimates and their uncertainties for 1951–1988.

Airborne waste streams were typically treated prior to release to the environment using either dust collectors (filters) or scrubbers (treatment systems employing liquids to remove particulate matter from gaseous waste streams). The efficiency of both of these methods varied greatly with the state of the technology at the time, maintenance of the system, and plant throughput. For dust collectors, our estimates accounted for anisokinetic sampling and sample line losses. Anisokinetic sampling occurs when the sampling probe in the dust collector stack does not record the stack exhaust gas velocity accurately. Losses of particles in the sampling line before they are detected at the sampler can significantly affect estimates of releases from stacks at the plant. These factors were not considered in previous studies.

Table ES–1. Summary of Median Uranium and Radon Releases Estimates From the FMPC for 1951–1988 With Uncertainty Bounds[a]

Release Point	Median Release Estimate	5th–95th Percentile Range
Uranium to atmosphere		
Dust collectors	140,000	120,000–170,000
Plant 2/3 scrubbers	66,000	56,000–78,000
Plant 8 scrubbers	81,000	56,000–130,000
Miscellaneous Sources[b]	16,000	9300–28,000
Total: airborne sources	310,000	270,000–360,000
Uranium to surface water		
Manhole 175	82,000	71,000–94,000
Paddy's Run	17,000	14,000–20,000
Total: surface water	99,000	85,000–120,000
Radon to Atmosphere		
K-65 Silos		
Radon	170,000 Ci	110,000–230,000 Ci
Radon decay products[c]	130,000 Ci	87,000–190,000 Ci

[a] Values are in kg of uranium, except for releases from the K-65 Silos which are reported in units of activity, called curies, Ci.
[b] Unmonitored and accidental releases.
[c] The release quantities for radon and its decay products are given in units of activity, curies (Ci); quantities of each of the short-lived decay products, polonium-218, lead-218, bismuth 214, and polonium-214.

Estimates of releases from the denitration processes scrubbers in Plant 2/3 (refinery) and from the scrubbers in Plant 8 (scrap recovery) were made considering uncertainty and variability in parameters that affect scrubber performance. Relevant site-specific data were used as much as possible. Monte Carlo techniques allowed us to sample the parameter uncertainty distributions to make the release estimates. The distributions represent uncertainties associated with these individual parameters and can be combined to form a distribution that characterizes the overall range of potential scrubber releases, in contrast to the point estimates of previous studies. Our estimates of releases from Plant 8 scrubbers relied heavily on data reporting monthly amounts of uranium found in the scrubber liquid residue (called scrub liquor) and measurements of scrubber penetration of uranium. The Plant 8 scrubbers dominated the uranium releases in the 1960s, with approximately 47,000 kg U released in that decade, compared to 21,000 and 19,000 kg U for the dust collectors and Plant 2/3 scrubbers, respectively. In the 1970s, the Plant 2/3 scrubbers were relatively more important. In the 1950s and 1980s, the dust collectors contributed most to the total uranium releases, although the magnitude of all releases in the 1980s was significantly less than in the 1950s.

A thorough evaluation of atmospheric releases of uranium from unmonitored sources (incinerators, building ventilation, lab hoods, unmonitored process emissions and waste pits) and accidental releases (fires, spills and episodic releases) indicates that these were relatively minor compared to the three primary sources of atmospheric emissions (dust collectors, Plant 8 and Plant 2/3 scrubbers). However, the detailed assessments of these sources provide thorough documentation of their magnitude with uncertainties. These release estimates are included as part of the total atmospheric source term.

Radon releases were calculated for the K-65 silos, located near the west side of the site, and for drummed K-65 material temporarily stored on the Plant 1 Pad in the early 1950s. The silos contained K-65 material, a waste from the extraction processing of uranium ore. This material contains high concentrations of radium-226, and thus, acts as a continuous source of radon-222, a highly mobile radioactive inert gas. Release estimates were complicated by a lack of data describing characteristics of the material in the silos, and by structural changes that occurred over the years. Our estimates of radon and radon decay product releases were derived from measurements found in the historical records and from previous studies. The rate of radon release from the K-65 Silos for 1959–1979 is greater than for other periods, and significantly greater than for later periods. Radon releases from the Plant 1 Pad drums were insignificant contributors to the total radon releases for the period 1951–1988, but were important contributors for 1951 and 1952.

Radioactive material left the site in liquid effluents at two key points: through Manhole 175 (MH 175), a final junction point for major effluent streams onsite to the Great Miami River, and, periodically, through the storm sewer outfall to Paddy's Run. Effluent concentrations and volumes were measured regularly at both locations, and records were used to reconstruct these source terms. More uncertainty is encountered with the release estimates to Paddy's Run because the frequency of sampling was less than at MH 175, and there were discharges to the stream that were not monitored. Nevertheless, estimates of releases of uranium in liquid discharges are relatively well known, and uncertainties are generally smaller than with releases to air.

An evaluation of the groundwater plumes underlying the FMPC indicated that, at the present time, three offsite wells are contaminated, and only a small number of people would have potentially received radiation doses from contaminated groundwater. Consequently, a simple model is used to estimate concentrations of uranium in the contaminated plume, based on recent measurements in the three offsite wells and on quantities of uranium released to the storm sewer outfall ditch and to Paddy's Run since the 1950s. Based on this simple model, it is likely that uranium contamination in the groundwater would not have reached the offsite wells prior to 1968.

There have been several previous assessments of uranium releases from the FMPC. Previous estimates of uranium discharged in liquid effluent fall within the uncertainty range of our estimates. Source terms from previous studies of airborne uranium releases have all fallen outside our uncertainty range except for one study. Exhaustive comparisons have not been made; however, reasons for our higher estimates include:

- the time to conduct a comprehensive review of historical documents, in particular original records, related to the FMPC operations;
- the use of a distribution of scrubber efficiencies for Plant 8 scrubbers;

- accounting for uranium losses from miscellaneous unmonitored sources and accidents;
- accounting for biases from sample line losses and o her sampling deviations in the calculation of dust collector losses.

Our results report not only best estimates of releases (as a median value) but also associated uncertainties that were calculated as an integral part of the estimates. This approach represents a significant improvement in the state-of-the-art of source term analysis. This depth of analysis was not undertaken in earlier estimates of releases. These source term estimates will be used in Task 6 to calculate radiation doses to people who live near the FMPC.

TASKS 2 AND 3

RADIONUCLIDE SOURCE TERMS AND ASSOCIATED UNCERTAINTIES FOR 1951–1988

INTRODUCTION AND OVERVIEW

The purpose of the Fernald Dose Reconstruction Project *(FDRP)* is to estimate radiation doses to people who lived near the Fernald (Ohio) Feed Materials Production Center *(FMPC)* during its years of operation from 1951 to 1988 (Figure 1). Exposures resulted from both planned and unplanned releases of radionuclides to the environment. The study was conducted for the Centers for Disease Control and Prevention.

The project was divided into seven tasks. The goal of Task 2 is to determine the radionuclide source term for the facility; that is, to determine both the amounts of radioactive material released to the environment and the variability of release rates. This information is fundamental to the assessment of radiation doses to persons in the vicinity of the site. The Task 3 objective was to determine the uncertainties associated with those past releases.

An interim Task 2/3 report (Voillequé et al. 1991) initially determined the source term for the years 1960, 1961 and 1962. This shorter time period was selected because environmental samples and records were available and there was a relatively consistent level of emissions. The pilot study tested and presented our methods for estimating the amounts of radioactive materials released and for assessing the uncertainties associated with those estimates. Based on the methods described in the interim Task 2/3 report, we estimated the amounts of radioactive materials released to air, surface water and in groundwater throughout the history of the Fernald plant's operation. Those results were presented in a draft report (Voillequé 1993). The draft report was reviewed, and comments were received from a number of people and organizations, including the CDC, members of the public, current employees at the FEMP, and former employees of NLO. All comments were considered in finalizing this current report, which reflects those changes and represents the final Task 2/3 report for this project. In addition to minor editorial changes, the main revisions to this report from the draft version include:

- Annexes listing the types of documents found in Central Files at the FMPC and of the boxes of contaminated documents that were examined in the Plant 4 storage area (Appendix A)
- Revised screening calculations using updated NCRP screening factors (Appendix D)
- Re-evaluation of the attachment fraction of particles in the calculation of sampling line losses for dust collector releases (Appendix G and E).
- Recalculation of discharges from the Plant 2/3 denitration operations using additional scrub liquor concentration data; determination of effect of alternative calculation of the outage fraction on Plant 2/3 scrubber releases (Appendix H)
- Two alternative calculations of releases from the Plant 8 scrubbers to test the effect of different modeling choices on the results. (Appendix I, page I–37)

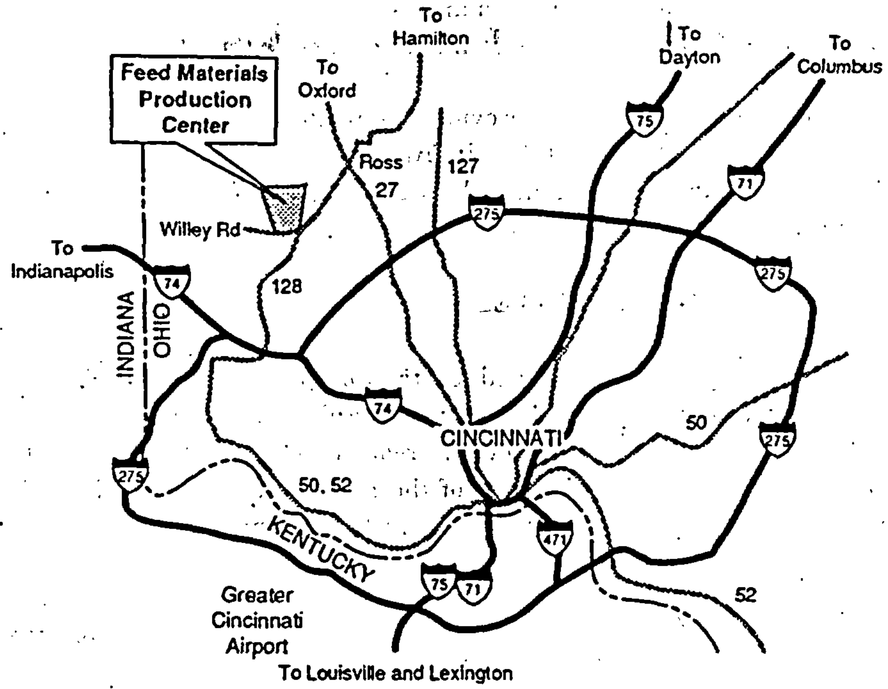

Figure 1. Location of the Fernald Feed Materials Production Center.

- An alternative calculation of radon and radon daughter product releases from the K-65 silos using a conventional methodology of radon releases from bulk quantities of ^{226}Ra-bearing materials (Appendix J, page J–73).
- Revision of fugitive emissions calculations for the waste pits using an improved model (Appendix K).
- Use of an empirical model to estimate uranium concentrations in offsite contaminated wells for years when no measurements were made; the model uses available uranium measurements in well water and considers the uranium released to Paddy's Run and the storm sewer outfall ditch (Appendix M).

This report is divided into this summary and 13 appendices. Each appendix is in bold type when it first appears in the discussion of that appendix. The appendices are:

Appendix A	Sources of Information
Appendix B	Plant Processes and Wastes
Appendix C	FMPC Production Information
Appendix D	Other Radionuclide Releases
Appendix E	Effluents from Dust Collector Exhausts
Appendix F	Fitting Particle Size Distributions for FMPC Dust Collectors
Appendix G	Estimates of Bias in Effluent Sampling for Particles
Appendix H	Discharges from Plant 2/3 Denitration Operations
Appendix I	Releases from Plant 8 Scrubber Systems
Appendix J	Releases of Radon, Radon Decay Products and Gamma Radiation from the K-65 Silos
Appendix K	Other Sources and Episodic Releases to the Atmosphere

Appendix L Surface Water Discharges
Appendix M Groundwater Contamination Outside the FMPC

The goal of this report is to provide the reader with a clear picture of the FMPC operations from 1951 through 1988. It explains the generation of effluents from those operations, and estimates effluent releases using relevant measurements and related information.

PLANT PROCESSES AND WASTES

The FMPC is a government-owned, contractor-operated facility whose primary purpose was to convert uranium (U) feed stocks to uranium metal ingots for machining or for extrusion into tubular form. Production reactor fuel cores and target elements were fabricated. Figure 2 gives an overview of the main features of the FMPC area. An aerial photograph shows the environs of the FMPC in 1965 (Figure 3).

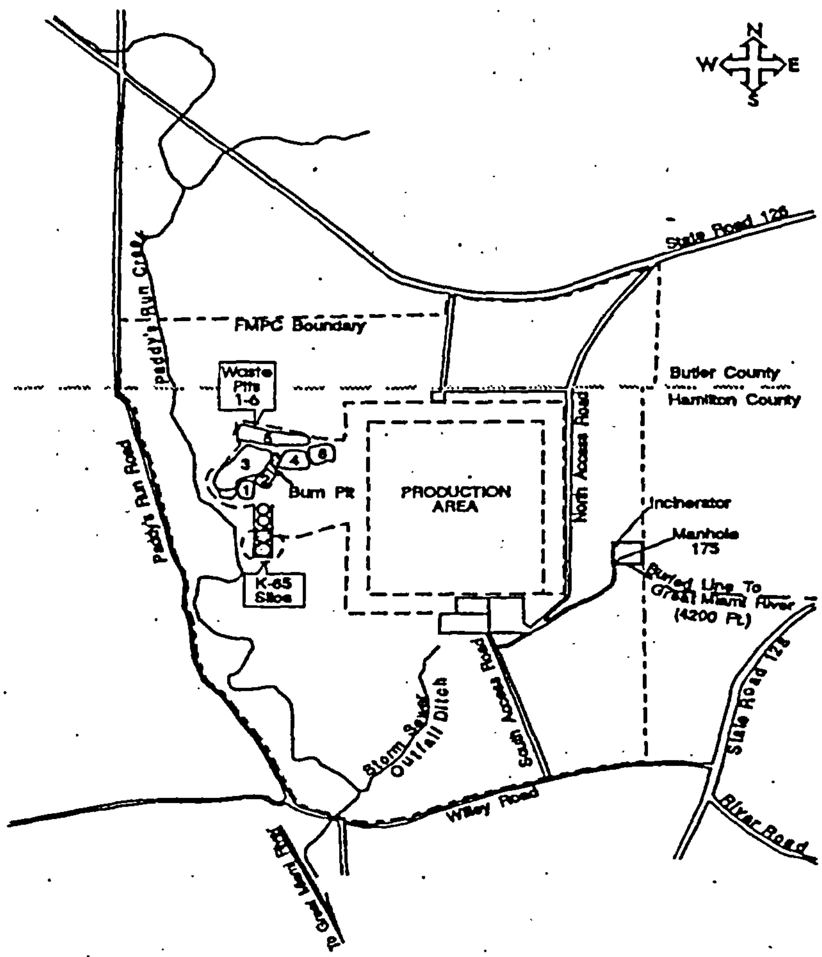

Figure 2. Overall view of the FMPC facility. The width of the production area is about 700 meters from east to west (inner fence).

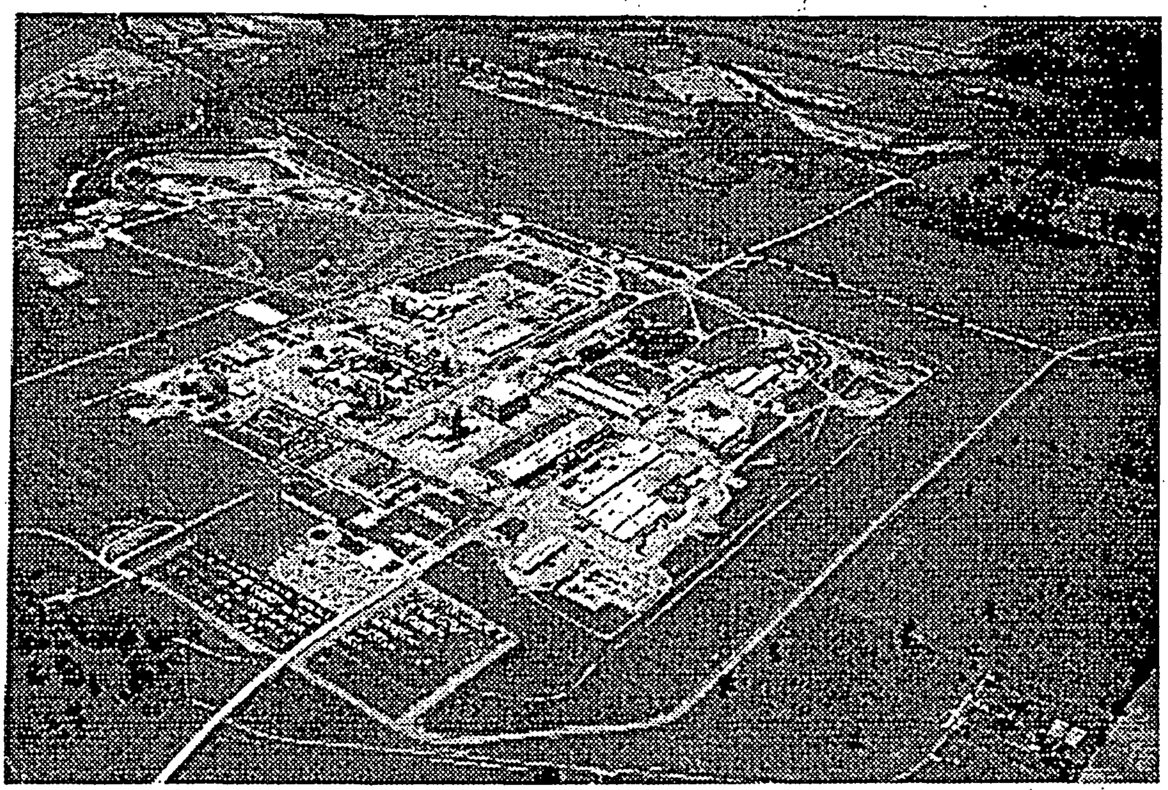

Figure 3. Aerial photograph taken from the southeast of the Feed Materials Production Center in 1965, showing the production area and general land features (digitized from DOE 1965). The area within 5 miles (8 km) from the center of the FMPC is populated with farm houses, small communities, and the small town of Ross, Ohio, with land use being primarily grazing and farming.

Appendix B describes the plant functions in some detail by following the flow of uranium through the various facilities as it was converted from raw material to finished products. Although uranium was the primary product at the FMPC, lesser amounts of thorium were produced intermittently during the mid-1950s, and from 1964 through 1980. In addition, the FMPC began processing materials recycled from other stages of the nuclear fuel cycle in 1962.

Figure 4 is a material flow diagram which shows the movement of incoming raw and recycled material (called feed materials) into the FMPC at Plant 1, the Sampling Plant, and their passage through various chemical and physical processes before leaving the site as finished products. Historic records and discussions with plant staff revealed that the same basic processing scheme was employed throughout all years of operation.

From Plant 1, the materials passed to Plant 2/3, the Refinery, where the uranium in the various feed materials was converted to uranium trioxide (UO_3, called orange oxide because of its color). The UO_3 was converted to uranium tetrafluoride (UF_4, called green salt) in Plant 4, and then sent to Plant 5, Metals Production. There the UF_4 was converted to uranium metal derbies or ingots. From Plant 5 the ingots were shipped offsite, or were sent to either Plant 6 (where the metal was fabricated into finished products) or to Plant 9 (where special products were machined).

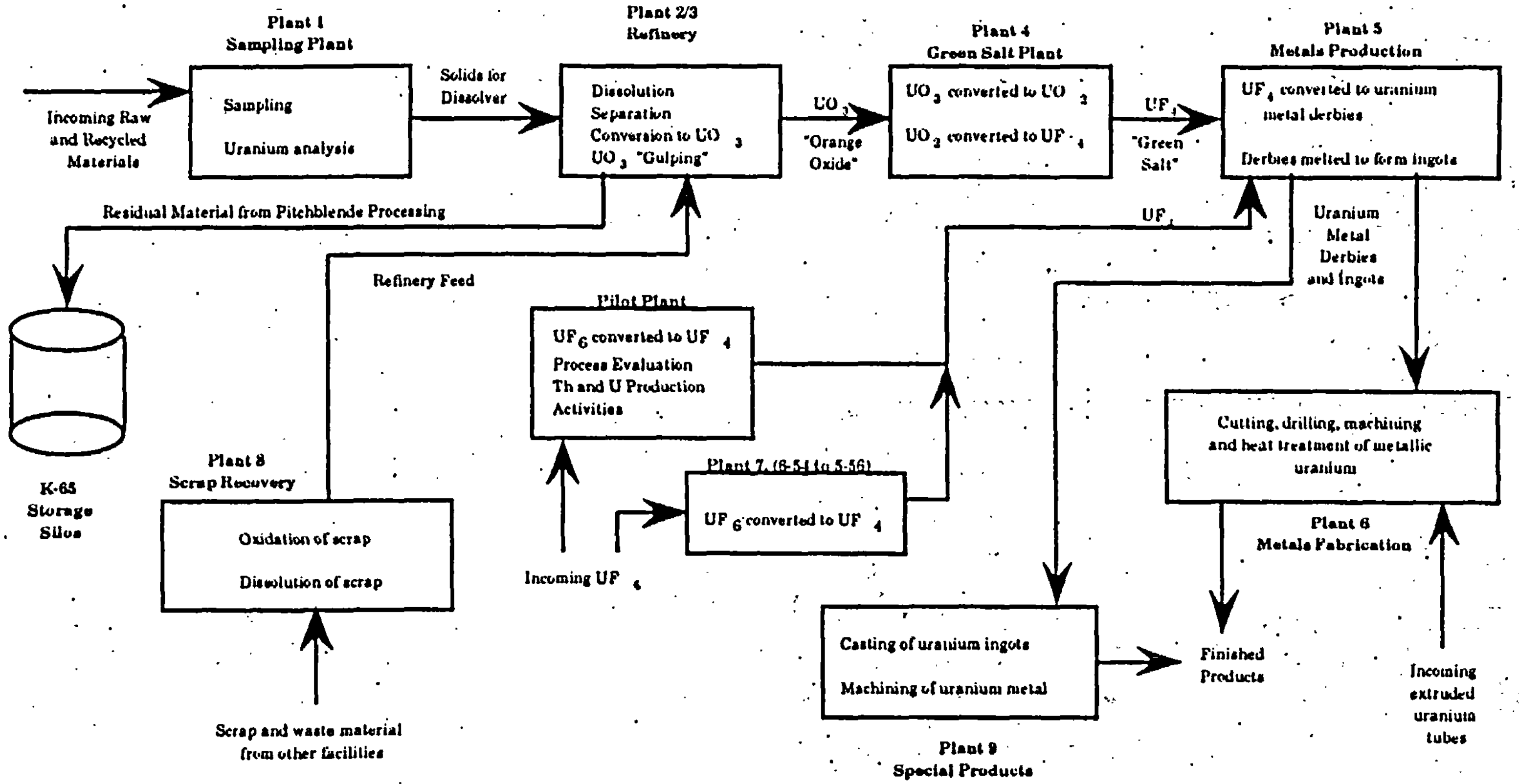

Figure 4. Schematic Diagram of Flows of Uranium Between Facilities at FMPC. Flows of recycled material were more complex than can be illustrated simply. For example, recycled uranium received as uranium trioxide would not be processed in Plant 2/3, but would be feed for Plant 4.

In Plant 8, the Scrap Recovery Plant, waste materials and metal scraps from the production processes were heated to remove impurities before being sent back through the Refinery (Plant 2/3) and the production process. The Pilot Plant was used for the direct conversion of incoming enriched UF_6 (uranium hexafluoride) to UF_4 (green salt).

Much of the thorium production activity at the FMPC took place in the Pilot Plant, beginning in 1954. Plant 7 operated only from 1954–1956 in converting UF_6 to UF_4. Waste materials from these processes were treated in various ways at the FMPC depending upon their physical form. The K-65 Storage Silos, large concrete tank-like structures, store residues from the extraction of uranium from ores that were processed during the early years of FMPC operations. Liquid effluents were collected and treated at the general sump before being discarded to the waste disposal pits. Liquids from the clearwell portion of the waste pit, along with the storm sewer runoff and sewage treatment plant effluent were piped to the Great Miami River from Manhole 175 on the eastern boundary of the site. Solid waste materials were sent directly to the waste pits, or they were burned in the incinerator located near the eastern edge of the facility or in the burn pit near the waste pits. The FMPC also operated a graphite burner from 1965 to 1984, an oil burner from 1962 until 1979, and an incinerator for liquid organic wastes that was installed in 1983. Releases from these latter facilities are described in Appendix K.

FMPC PRODUCTION INFORMATION

Production information provides a guide to the magnitude of FMPC activities over the years. In the absence of other data, it can be used to help estimate releases from the facility to the environment. **Appendix C** contains details of the receipts and shipments of uranium at FMPC along with specific production data for each plant for the time period 1951–1988.

These records of shipment and receipts, and plant production provide several key pieces of information. First, they specify the level of *"enrichment"* of processed uranium, which relates to the concentration of uranium-235 (^{235}U) relative to uranium-238 (^{238}U).

- "Natural" uranium contains 0.72% ^{235}U.
- "Depleted" uranium contains less ^{235}U; typically 0.14–0.20% at FMPC.
- "Enriched" uranium contains more ^{235}U; typically, 0.95–1.25% at FMPC.

While most of the enriched uranium was in the above range, some processing of 2% enriched uranium occurred in the 1960s. The capability to digest 5% enriched uranium was added to Plant 1 in 1970.

Second, records of receipts of material by FMPC and shipments from FMPC provide a rough indication of production rates. Comparisons of the data on receipts and shipments indicate that material was received, processing occurred, and products were shipped on a fairly regular schedule during much of the time. During fiscal year (FY) 1952 through 1980, the FMPC received about 362,000 metric tons (MT; 1 MT = 1,000 kg = 2,200 pounds) of uranium and shipped about 358,000 MT to offsite locations (Audia 1977; FMPC 1988). Approximately 54% of the receipts and shipments were natural uranium, about 20% were enriched uranium, and some 26% were depleted uranium. Uranium shipments tended to follow the pattern of receipts during most of the years of operation.

Third, plant-specific production rates are useful for estimating releases of radioactive materials from specific facilities. Processing rates in each plant were increased or reduced because of changes in the demand for intermediate materials and finished metal products. Figure 5 summarizes the total production quantities in metric tons of uranium (MTU) for 1951 to 1988. In some plants, there was no production during certain years. For example, there was no production of UF_4 from UF_6 in the Pilot Plant from 1968 to 1984. Data on the enrichment categories of products are presented in Appendix C.

Thorium production at the FMPC was estimated to have been only about 0.4% of the uranium production. Processing was limited to a few facilities and to specific time periods. Some of the uranium received at the FMPC was recycled, that is, it had other radionuclides as contaminants in the uranium.

OTHER RADIONUCLIDE RELEASES

Radioactive decay of uranium and thorium isotopes produces series of other radionuclides that are collectively referred to as *decay or daughter products*. In most of the feeds received by the FMPC, the uranium had previously been separated chemically from the other decay products. As a result, the facility's effluents consisted primarily of uranium. Other radionuclides were generally present in small quantities. Early processing campaigns treated ores that contained nearly equilibrium amounts of the decay products. The wastes from that early processing were placed in the K-65 Storage Silos. Releases from the silos are discussed in Appendix J.

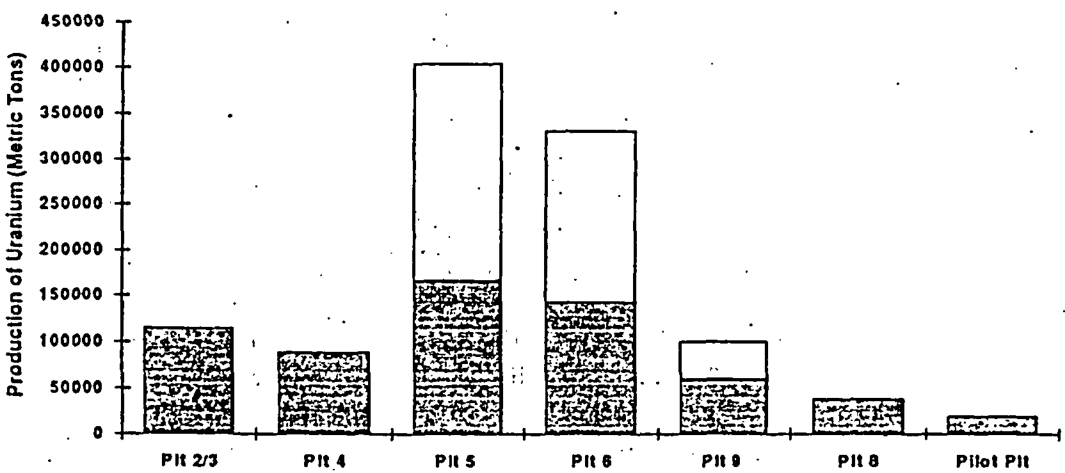

Figure 5. FMPC plant production for 1952 through 1988. Each plant produced a different product: uranium trioxide in Plant 2/3, uranium tetrafluoride in Plant 4, metal derbies (dark bar) and ingots (light bar) in Plant 5, machined (dark) or rolled products (light) in Plant 6, uranium ingots (light) and machined products(dark) in Plant 9, uranium recovered from scrap materials in Plant 8, and uranium tetrafluoride in the Pilot Plant.

Because *recycled feed materials* were sometimes used, small amounts of other radionuclides called *fission and activation products* were also introduced into the process stream and later released. Recycled uranium was not processed at the FMPC prior to October 1962, so releases of fission and activation products did not occur prior to that time. Measurements of the amounts of these radionuclides, relative to uranium, were not performed until years later. These products were measured from airborne effluents (in scrub liquor or dust collectors) at only one time in 1985.

Appendix D provides the measurement data of fission and activation products in particulate materials done at that time. The concentration of fission products— strontium-90, technetium-99 and cesium-137—were highly variable. The transuranic nuclides - neptunium-237, plutonium-238, and plutonium-239,240 were detected in all of the samples analyzed, but the observed concentrations varied over a wide range within the plants and from plant to plant. Only the short-lived decay products of uranium-238 were found in consistent concentrations. The concentrations of thorium isotopes and their radium decay products were found to be consistent in samples from some plants but not from others. In some plants, the concentrations of transuranic nuclides (*TRU*) were clearly affected by the processing of material containing unusually high concentrations of TRU between 1980 and 1985.

Measurements of radionuclides other than uranium in liquid effluents are available for a longer time period than for airborne effluents. There was no processing of thorium during the time periods 1952–1953, 1958–1963, or since 1980. Relative concentrations of thorium with respect to uranium were measured in the mid-1950s, and again beginning in 1967. Beginning in 1976, the concentrations of plutonium, neptunium, radium and the fission products, cesium-137, ruthenium-rhodium-106, technetium-99, and strontium-90, were measured relative to uranium. The concentrations of these other radionuclides in liquid effluents are shown in Appendix D. Estimates of the amounts discharged in liquid effluents are presented in **Appendix L.**

The relative importance of various radionuclides as potential contributors to offsite radiation doses was assessed using a methodology developed by the *NCRP* (National Council on Radiation Protection and Measurements) (NCRP 1989). These calculations show that releases of uranium are by far the most important contributors to the potential doses from releases to the atmosphere at the FMPC. For liquid releases, the radium isotopes were found to be of primary importance, depending upon the pathway considered.

FMPC RELEASES TO THE ENVIRONMENT: FACTORS TO CONSIDER

Several factors regarding source term estimates were considered at the outset of the project. These included the initial period of study, characteristics of radionuclide releases, the uncertainties involved in making source term estimates, and the sources of information that would be used for this process. To apply resources most efficiently, it was necessary to assign priorities to the three source terms— airborne effluents, liquid waste discharges, and inputs to the groundwater — according to their importance. The greatest emphasis was given to those releases that had the largest potential impact on the population residing in the vicinity of the FMPC. All the evidence, which will be documented throughout the report, indicates that airborne releases deserve the greatest attention. That conclusion influenced the level of detail of the investigations and the corresponding reports in this series.

Period of Time Studied

Although radionuclide source terms are reported here for the entire operating history of the FMPC (1951–1988), our initial effort focused on a shorter time period (Voillequé et al. 1991). Originally, we considered examining 1955, the year of the highest reported releases to the atmosphere (Boback et al. 1987). During a September 1990 site visit, it was confirmed that the installation of effluent sampling equipment was not complete during 1955. The quality and variability of results from an operational effluent sampling system are needed to estimate source term uncertainty, needed for Task 3.

Other factors indicated that a time period in the early 1960s was the best focal point for the initial work on source terms and their uncertainties. These included the availability of environmental samples and records along with a level of emissions which make uncertainty analysis workable. We were also able to locate other documentation that was needed to derive source term estimates.

Analysis of data from a period of relatively consistent operation (1960, 1961 and 1962) has provided a basis for estimating source terms for other periods when fewer measurements were made and when there were more unmonitored release points. The interim draft Task 2/3 report addressed releases to the atmosphere, to surface water and to groundwater by the FMPC for the period 1960–1962. In the current report, we use the same methods of investigation to derive annual source term estimates for uranium and other radionuclides released in air, surface water and ground water from the FMPC for the entire period 1951–1988.

Characteristics of Radionuclide Releases

Initially, it is important to identify specific attributes of the radionuclide release, or source term, to be documented. The most important parameters that are common to all releases include:

- nature of release: Was it routine or episodic?
- magnitude or size of the release
- radionuclides released

For the surface water source term, the discharged radionuclides in waste water were either in solution or in suspension as finely divided particles. In either case, the radioactivity was carried from the FMPC site via a pipeline to the Great Miami River or in the storm sewer overflow via Paddy's Run, a small stream at the west boundary of the site. Paddy's Run joins the Great Miami River approximately 3 kilometers south of the FMPC (Figure 2).

Radioactivity reached the groundwater by infiltration in a form similar to that in liquid discharges. The radiation doses from consumption of water from either source depend on the amounts released and upon the dilution in the river or the aquifer before withdrawal for human use.

For radionuclide releases to the atmosphere, there are two other factors, besides release rate and dispersion, that are important determinants of the radiation doses to members of the public. These are:

- the chemical form of the discharge
- its physical characteristics, primarily the size distribution of the released particles

Human metabolism of radionuclides that have been inhaled is dependent upon the chemical form of the radionuclides. Soluble compounds are readily taken up into the blood stream and are rapidly distributed throughout the body. Chemical forms that are insoluble in body fluids tend to be retained in the lung for a longer time and are only gradually transported to other tissues. The chemical form of the discharges are presented in the appendices describing atmospheric releases.

The particle-size distribution is important for calculating the amounts of radioactive material that were deposited on the ground following release. Particle size is also important for estimating the radiation dose from inhalation of the particles.

Uncertainties in Estimating Releases

Results of scientific investigations are, by their nature, uncertain, and it is a common practice for investigators to provide some estimate of uncertainties that affect their estimates. Estimating the uncertainties associated with the source term estimates (Task 3) is, therefore, an important part of this work. The absence of uncertainty estimates is a weakness in the previous source term information.

Knowledge of several parameters, or numbers, is required to define a radionuclide release. None of them is known exactly, and most are contributors to the overall uncertainty associated with the release estimate. Two types of parameter uncertainty affect the overall source term uncertainty (Hofer and Hoffman 1987). The first is due to random variations in sampling, measurement, and operational procedures. For example, estimates of uranium releases to the atmosphere are based upon analytical measurements of the sample mass, the percent of the collected mass that is uranium, the flow rate through the sampler, the flow rate through the stack, etc. The physical dimensions of the sampling probe and the exhaust duct are also factors. Although the latter two quantities are fixed and relatively well known, each of the other measurements is rather more uncertain, for various reasons. This uncertainty contributes to the overall uncertainty of a particular release estimate.

A second type of uncertainty occurs because of a lack of knowledge about particular parameters. This may occur because the parameters were not measured during part or, in some cases, most of the period of facility operation. Examples of this type are periods when the stack sampler flow rate was not measured, and periods when the stack flow rate was not measured. In these cases, estimates of the values of those parameters during the periods between measurements will be necessary. In the absence of definitive information, subjective judgment of experts can be used to estimate the range and distribution of values for the unknown parameters during such periods.

The technique of using a computer to draw many random samples from the parameter distributions and combining these sample releases to obtain information about the distribution of the releases is an example of what is called a Monte Carlo procedure. Figure 6 illustrates this process.

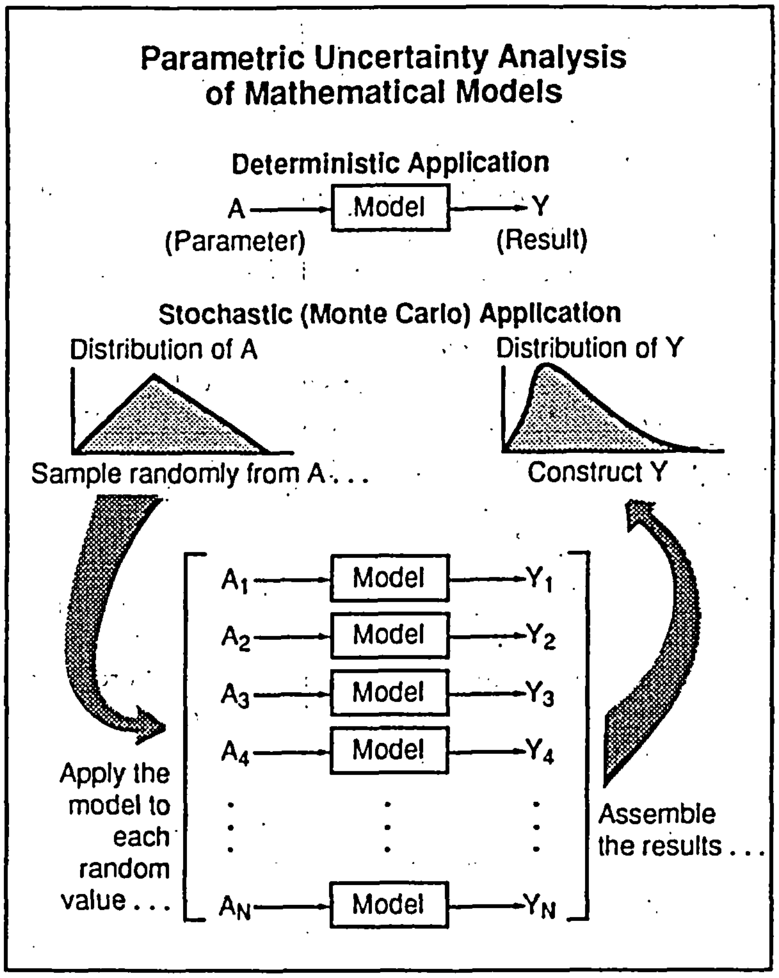

Figure 6. Schematic presentation of Monte Carlo methods for propagating a parametric uncertainty distribution through a model to its results. In this simplified illustration, A is an input parameter to the model, and Y is the result, or output, corresponding to A. For each specific value of A, the model produces a unique output Y. Such an application of the model is deterministic, because A determines Y. But A may not be known with certainty. If uncertainty about A is represented by a distribution, such as the triangular one in the figure, repeatedly sampling the distribution at random ad applying the model to each of the sample input values A_1, A_2... gives a set of outputs Y_1, Y_2,..., which can be arranged into a distribution for Y. The distribution of Y is then our estimate of the uncertainty in Y that is attributable to uncertainty in A. This is a stochastic, or Monte Carlo application of the model.

Our use of a Monte Carlo procedure to estimate releases explicitly recognizes that those estimates are uncertain because of variability or lack of knowledge of the parameters upon which the estimates depend. This procedure applies our best estimates of the distributions of parameter values to produce a distribution of results. Our approach contrasts with one in which a calculation is based upon point estimates of the various parameters and yields a single result. The Monte Carlo calculation carries the underlying uncertainty in the parameters forward and displays it in the breadth of the distribution of results.

This process was illustrated in the interim Task 2 and 3 report (Voillequé et al. 1991) by examining the distributions involved in the calculation of releases from the Plant 8 scrubbers for May 1961, and for that entire year. The estimated release from those scrubbers depends upon two parameters: the amount of uranium collected in the scrub liquor and the penetration of uranium through the scrubbers. The Monte Carlo procedure for estimating the Plant 8 scrubber releases involves independent selection of values of the two parameters and the use of the selected values to compute an estimate of the release. This procedure was performed repetitively (5000 times in the current example) and yielded a distribution of results.

Just as these source term estimates reflect the underlying variability and lack of knowledge about individual parameters, the radiation dose calculations, performed in a subsequent task (Task 6), will consider the range of source term values for a given year. They will also incorporate uncertainties about meteorological dispersion, particle deposition, and other parameters to produce distributions of estimated doses to people residing near the FMPC.

Sources of Information

A major effort in the Fernald Dosimetry Reconstruction Project has been searching for, and reviewing, hundreds of documents related to the operation of the Feed Materials Production Center since operations began in 1951. It has been our practice to trace the information back to original sources whenever possible. In the Task 1 report, issued in January 1991 (RAC 1991), we outlined the general approaches that we have taken to obtain this information. These five methods, which have formed the foundation for the project in providing the technical data for this study, are:

- site visits to the FMPC facility;
- investigation of records and scientific literature pertaining to the FMPC;
- retrieval and review of documents from NLO, Inc. using their computer database of document titles;
- examination of engineering diagrams, site blueprints, historic photographs and maps; and
- discussions with current and former longtime employees.

Because we realized the importance of retrieving documents from a wide range of sources, considerable time has been spent identifying types and locations of reports and records pertinent to the completion of this project. We visited a number of locations around the country to review documents that might provide background information on FMPC operations (Figure 7). Generally, this documentation of FMPC operations and releases comes from two broad areas: (a) from National Lead Company of Ohio, Inc. (NLO), the former operator of the site, the Westinghouse Materials Company of Ohio (WMCO), the site operator from January 1, 1986 through 1992, and the Department of Energy (DOE); and (b) from FMPC-independent sources. **Appendix A** provides a detailed look at the sources and locations of documents used for the project.

While not all the original records are still available, many original documents remain in the files at the FMPC facility, in the library of the NLO offices, and in storage facilities utilized by WMCO. Many hours have been spent examining original plant documents, particularly those related to effluent discharge measurements and procedures. The information sources can be categorized as follows:

- processes descriptions for the various facilities
- plant operating procedures
- effluent sampling procedures
- daily and monthly reports of liquid effluent discharges
- monthly reports of airborne effluent discharges
- original analytical data sheets recording sample concentrations
- plant operating process logbooks
- nuclear materials control reports
- daily sump discharge logbooks
- topical reports related to effluent characteristics
- reports of ventilation system tests and evaluations
- incident reports
- investigation reports
- letter reports of operational problems
- production records for specific processes

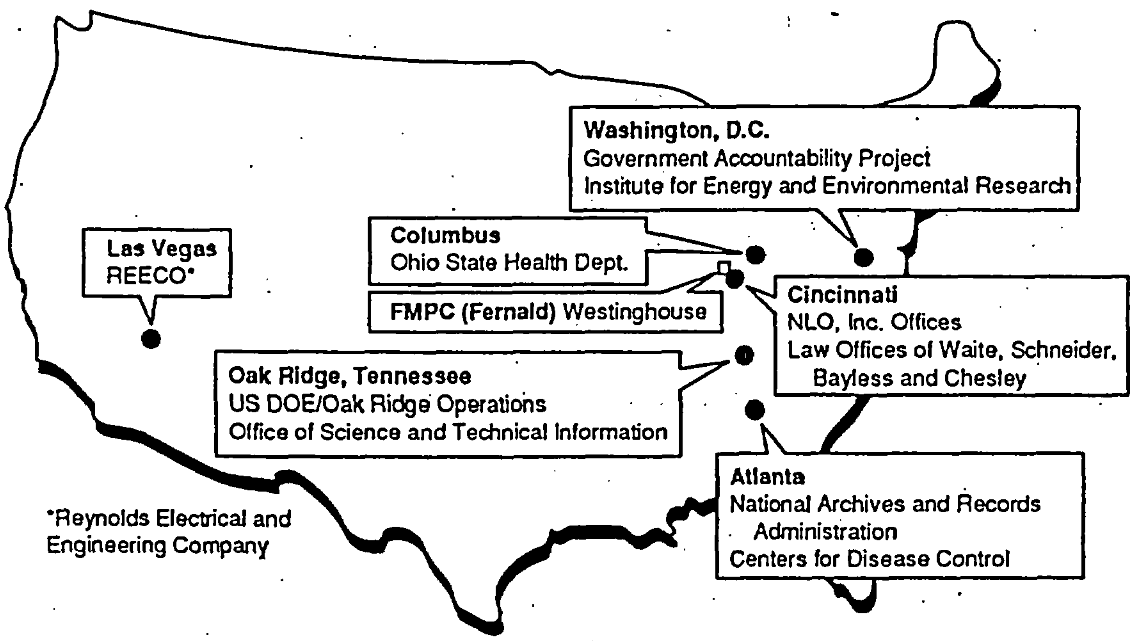

Figure 7. Locations visited in obtaining FMPC-related documentation and information.

Discussions with long-time employees and retirees from the FMPC provided another source of information for the project (RAC 1991). Their recollections on processes and procedures that routinely occurred since facility start-up served to identify sources and locations of documentation. Documents used in the construction of the source terms are referenced in the appropriate section of the text, with the references listed at the end of the appendix or section. In addition, we have maintained a collection of all documentation that we have reviewed since the Fernald Dosimetry Reconstruction project began in 1990. Appendix A lists all documents that have been added to the RAC Document Repository up to this time.

In general, data from original records used in this study are reported in the same units that appeared in the source documents. For example, the uranium concentrations in liquid effluents and volume measurements, compiled in Appendix L, are reported in mg L^{-1} and gallons, respectively. In contrast to some of the original sources of information, our final release estimates and results of other calculations are reported to only two significant figures.

ATMOSPHERIC RELEASES FROM DUST COLLECTORS

Atmospheric releases from FMPC operations came from buildings where uranium processing took place and from outside areas such as the waste pits and incinerators. Appendix K reviews estimates of emissions of uranium from miscellaneous unmonitored processes, non-routine events, and episodic releases. Some airborne effluents were treated with one of the two treatment systems used at the FMPC: scrubbers or dust collectors. These treatment systems are discussed in **Appendix B**. Dust collectors employed bag filters to remove airborne particulates from an exhaust stream. Information on effluents from dust collectors is presented in **Appendix E**. The key points of the dust collector operation and our estimates are presented here.

Dust Collector Operation

Process area ventilation air was ducted to dust collectors where airborne particulate material was removed before discharge through the stack to the outside. The dust collectors recovered valuable uranium that would otherwise be lost and reduced worker exposure in the process area. When operating as designed, the dust collector systems could be quite efficient (Drinker and Hatch 1956, Ross and Boback 1971).

The sampling systems installed in the dust collector stacks were simple in concept. A schematic diagram of the sampling system is drawn in Figure 8. Air was drawn from the exhaust stack through a sampling line to a pleated cellulose filter for collection of particulate material in the sample of discharged air. The filters were periodically changed and submitted for analysis. Details of the design and operation of these systems and of the sample analysis and data reporting are given in Appendix E.

Distribution to all the plants of an initial stack sampling procedure seems to have occurred in February 1956 (Starkey 1956). Later that year a formalized procedure was developed (Boone 1956). Initial sampling frequencies were weekly, biweekly, or monthly depending on the magnitude of the previous effluent measurements. Monthly reports of releases were made to plant management by the Industrial Hygiene and Radiation (*IH&R*) group.

The sequence of reports itself documents the onset and growth of the dust collector effluent sampling program. Periodic sampling of some stacks was performed as early as

1953; however, the continuous sampling program did not begin until April 1955. Initiated in seven stacks in Plant 4 and 5, the sampling program grew fairly rapidly to encompass thirty stacks six months later. There were increases in the 1950s to a maximum of 50 sampling systems in May 1958.

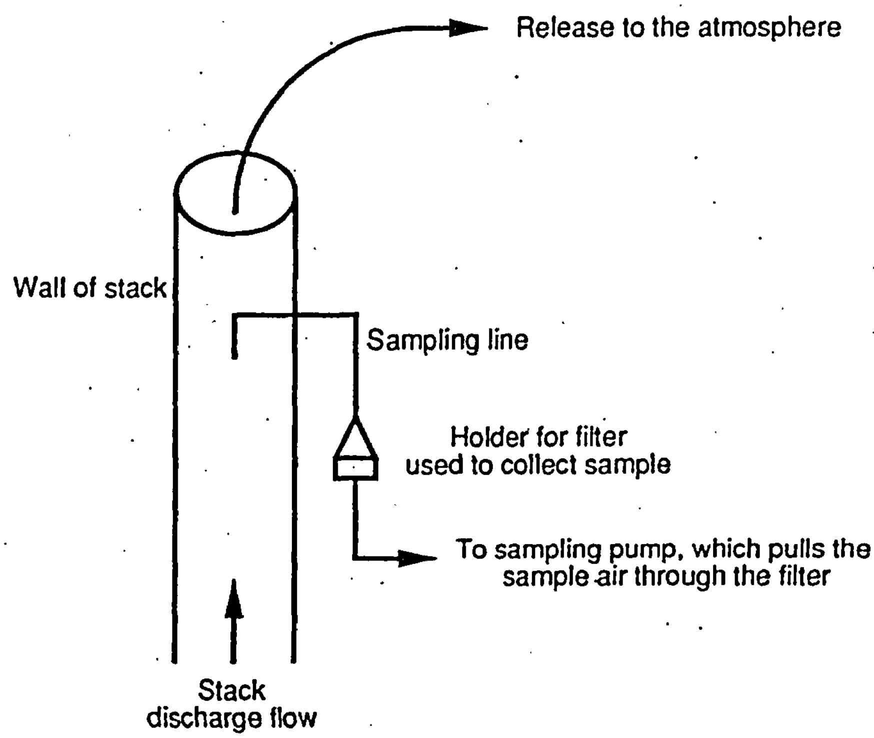

Figure 8. A schematic diagram of the dust collector stack sampling system.

At the start of 1960, there was a decline to 44 samplers for dust collector exhaust due to shutdown of systems in Plant 1 and in the Pilot Plant. At that time, the most common sampling interval was one month, although a few stacks were sampled more frequently. In the 1960s, sampling intervals were occasionally as long as six weeks for discharge points that were minor contributors to plant uranium releases. In later years, both plant production and staff were reduced. Intervals between sample analyses were greater and routine reports contained less detail. Filters were no longer changed and analyzed regularly. Filter changes and analysis occurred primarily when the filter had collected a visually detectable amount of particulate material.

Current Estimates of Release From FMPC Dust Collectors

Estimates of releases from individual dust collectors at the FMPC were tabulated from original records, which were usually monthly reports of the measurements. Review of the reported results revealed periods when samplers were not in operation and other times when the releases were too low to be detected. Estimates were made for these periods based on other sampling results and information about the sampling and analysis procedures.

Estimates were also made for years before monitoring was established as a routine procedure. These estimates were based either upon normalized release rates soon after routine monitoring was established or representative measurements during the mid- to late-1950s. In some cases, evaluations of unmonitored effluents led to significant increases over previous release estimates.

There are two major deficiencies in the tabulations of reported releases in the monthly reports. The first is that the release estimates were incomplete. Release estimates were not provided for sampling periods when samplers were not installed or were not operational for the entire period. The second deficiency in the tabulations is the failure to properly account for undetected releases. If no material was detected on the filter from a dust collector exhaust sample, the reported release was shown as zero. There were entire months when either no samples were collected or no releases were detected in the dust collector exhausts because the total reported releases from some of the plants were zero. To develop a better estimate of the releases for this report, it was necessary to estimate the unmonitored and undetected releases by determining the maximum release that could have occurred when none was detected. The details of this method are given in **Appendix E.**

In addition to correcting for unmonitored and undetected releases, the initial releases estimates are subject to further revision to account for biases in the effluent measurements themselves. While the design of the sampling systems was generally well conceived, three types of deviations from ideal sampling conditions may have biased the dust collector discharge estimates.

- Nonrepresentative sampling may have occurred when particles were not uniformly mixed in the exhaust at the location of the sampler. This is more likely to happen when the exhaust ducts are greater than 15 cm in diameter. The ANSI (1969) guide recommends multiple sample withdrawal points for ducts greater than 15 cm in diameter. The reason for multiple probes is to provide assurance that the samples will not be biased because of a nonuniform distribution of the contaminant in the stack. The sample extracted from the center of a dust collector exhaust stack would be representative if the particles were uniformly mixed in the exhaust or if the concentration on the centerline happened to be equal to the average concentration in the stack. When this is not the case, the sample is not representative of the material being discharged. The bias introduced may be positive or negative. A qualitative assessment of nonrepresentative sampling in presented in **Appendix G.**

- Anisokinetic sampling may have occurred. This occurs when there is a mismatch between the fluid velocity in the probe and that in the stack. If the velocities are not the same, over- or under-sampling of particles of various sizes could occur. The possible effects of anisokinetic sampling conditions were calculated using the methods described in Appendix G. That appendix contains example calculations and the basis for parameters used in Monte Carlo calculations of bias due to anisokinetic sampling.

- Losses of particles in the sampling line can occur when particles are deposited on the walls of the line, or when they are impacted due to the presence of bends in the lines between the probe and the collection filter. Neither topic has been addressed in previous analyses of the uranium release data. It should be emphasized that sample line losses lead only to underestimates of the effluent releases. The magnitudes of such losses depend upon particle size and density (**Appendix F**), the configuration of the sampling line, and the operating conditions for the line. These relationships are described in Appendix G.

A Monte Carlo procedure was used to estimate the sampling biases and their uncertainties. The calculations considered the three sources of bias identified above to obtain a measure of overall sampling bias. Major contributors to the uncertainty were the velocity of air in the sampling probe and in the duct, the bias due to nonrepresentative sampling, and a parameter used in computation of the attachment fractions. There is no simple way to reduce the largest uncertainties, which principally reflect the absence of information about conditions of past operations and sampling. Corrections for these biases are applied in estimating the dust collector uranium losses in **Appendix E**.

Once released from the stack, the physical and chemical characteristics of the uranium are important in the transport and deposition of released uranium and in the estimation of the radiation dose due to uranium inhalation.

- Particle size distributions were measured for some of the effluent streams in 1985. Those data and information about other uranium processing facilities have been used to estimate particle size distributions for the dust collector exhausts in this report (See Appendix F and Appendix E). Particle-size distributions for the stack emissions measured in 1985 are included as a part of the source-term characterization for stacks for all years because the plant processes served by the stacks have not changed significantly since the start of FMPC operations. Appendix F contains information on the reported measurements done in 1985. The distributions cover wide ranges of particle sizes and are not truly lognormal. The ranges of particle sizes have been subdivided into intervals and representative sizes are used in the calculations. Average particle-size distributions for both the inlet and the outlet ducts for stacks emitting UF_4 and U_3O_8 were derived from the data in Appendix F. The average distributions and distributions obtained from similar facilities are used for FMPC exhausts for which particle size measurements were not made. In spite of some substantial variations from stack to stack, the particles were relatively large.

- The chemical form of the materials discharged from the dust collectors affects the particle density, the transport and deposition of released uranium, and the estimation of the radiation dose due to uranium inhalation. The predominant uranium species emitted from each stack was identified from FMPC reports and engineering drawings of process equipment. About three-fourths of the releases from the dust collectors were in the form of uranium oxides.

The process of developing revised estimates of releases from the FMPC dust collectors is complex. Reported releases were incomplete because sampling was not initiated when production began. The reported releases do not include estimates of releases that were undetected by the analytical procedure or because a sampling system was temporarily out of service. The three sources of possible bias in the reported results, discussed above, have been estimated as part of this effort.

The first step in the approach adopted was to return whenever possible to the original release reports that were prepared routinely by the IH&R department. In the early years of full operation of the effluent sampling program, these reports contained a great deal of information about sample collection and about operational problems in all the plants. These detailed reports made it possible to estimate the magnitudes of undetected releases. Later reports of results, when production rates and releases were lower, were not as detailed and were much less helpful in this regard. In general, inclusion of undetected releases does not have a large effect on the estimates for early years when releases were large. In plants whose releases were relatively small (tens of kilograms of uranium per year) the relative contribution of estimates of releases that had gone undetected was greater.

Overall, corrections for unmeasured releases and for sampling bias led to revised release estimates that were about 50% higher than previous estimates of dust collector releases. Table 1 shows that the median estimate of total releases from the FMPC dust collectors from 1951 to 1988 was about 140,000 kg uranium. Most releases occurred during the 1950s. Principal contributors to the releases during that decade were Plants 4, 7, and 5. Plant 8 also contributed significantly to the total, but most of those releases occurred over a longer period of time. Although releases from the other facilities were not small, those releases were not major fractions of the total release. However, some of the releases from plants that were lesser contributors to the total were important in individual years.

Table 1. Summary Release Estimates for FMPC Dust Collectors

Period	Best estimate of release (kg U)	Other percentiles in distribution of release estimates (kg U)			
		5th percentile	25th percentile	75th percentile	95th percentile
1950s	120,000	96,000	110,000	130,000	150,000
1960s	21,000	18,000	19,000	22,000	24,000
1970s	3,100	2,500	2,800	3,400	3,800
1980s	2,100	1,700	1,900	2,400	2,700
1951–1988	140,000	120,000	130,000	160,000	170,000

DISCHARGES FROM PLANT 2/3 DENITRATION OPERATIONS

The air emitted from release points not equipped with dust collectors was cleaned through scrubbers. Scrubbers used either acid or caustic solutions to scavenge particles from the air stream being discharged to the atmosphere. Most of the particles are scavenged by mist droplets, which, for the most part, are collected by mist-eliminating devices and recycled to the liquid reservoir. This liquid (*scrub liquor*) is changed periodically. The uranium-containing droplets accumulate on the mist-eliminators, and some of the liquid is agglomerated into larger droplets and escapes back into the exhaust gas stream in a process called *reentrainment*. Figure 9 illustrates these processes. In this manner, the scrubbers of Plant 2/3 and Plant 8 emitted liquid droplets of reentrained scrub liquor of varying uranium concentration.

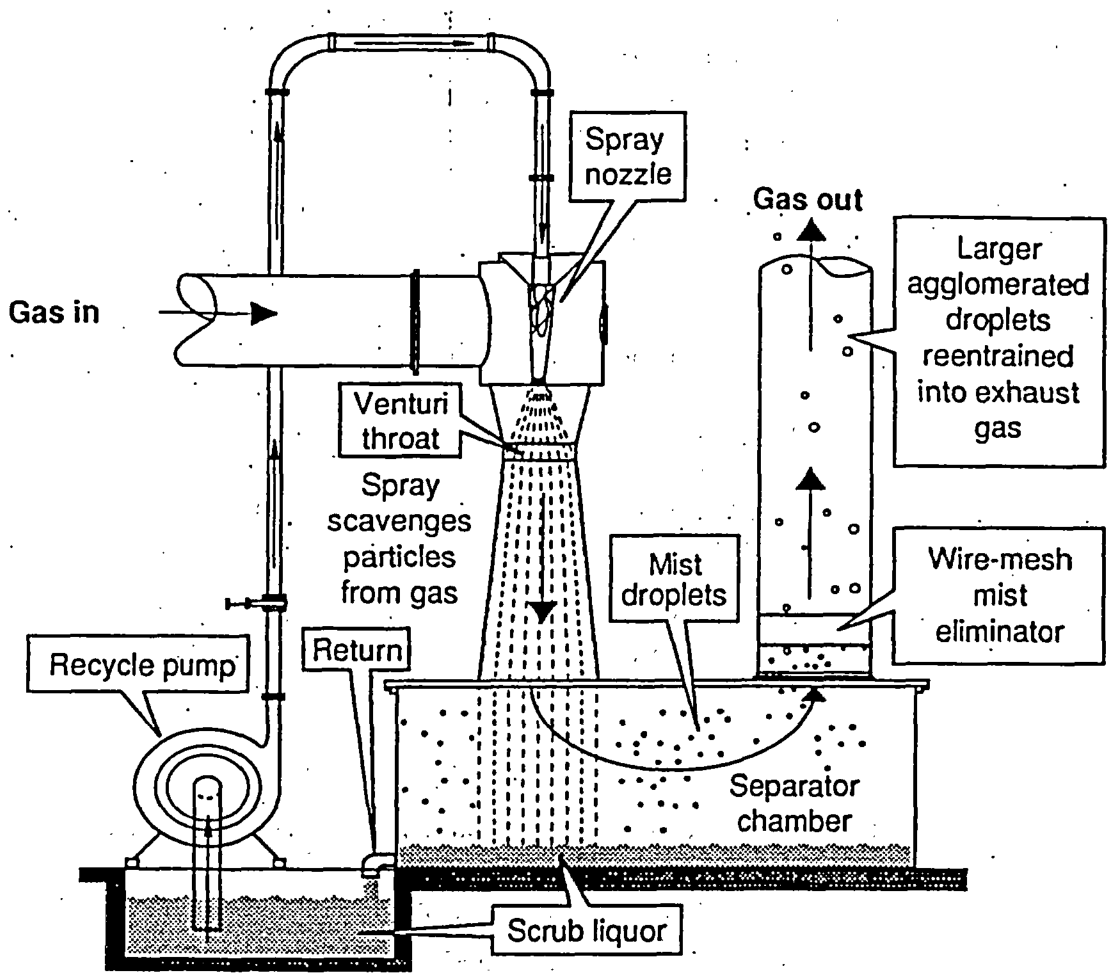

Figure 9. Scrubber schematic. Exhaust gas entering the scrubber is forced through a liquid spray into a Venturi tube. The gas then passes through a separator chamber and into the outlet duct. The spray entrains most particles into liquid droplets. Most of the liquid (or scrub liquor) is collected in the separator chamber and returns to a reservoir from which it is recycled. The scrub liquor of the Plant 2/3 and Plant 8 scrubbers was changed periodically and uranium was recovered from it. To inhibit the escape of the uranium-containing droplets various mist-eliminating systems were used. The figure indicates a wire mesh mist eliminator in the outlet duct (as in Plant 2/3), which would trap most droplets. But some of the trapped liquid was reentrained into the gas stream as large agglomerates and escaped to the atmosphere. Evaporation of the liquid produced relatively large solid particles.

After 1956, exhausts from the *denitration* process in Plant 2/3 were treated by a wet scrubber prior to discharge to the atmosphere. In the denitration process, nitrates were removed from uranyl nitrate hexahydrate (UNH) to produce uranium trioxide (UO_3, or orange oxide). Fumes of oxides of nitrogen that were produced during denitration were routed to the scrubber system. In a second process, orange oxide from the denitration pots was transferred by vacuum or *"gulping"* to a storage hopper. The releases of uranium from the scrubber exhausts were not sampled, even periodically, until recently. In June 1988, an investigation of higher than expected environmental radioactivity measurements led to the conclusion that releases from Plant 2/3 processing activities were the source of the observed higher offsite air concentrations (Investigation Board 1988). **Appendix H** provides details of the scrubber exhaust system, our current approach to estimating releases from the Plant 2/3 scrubbers, and previous release estimates. Because information is lacking on early operations with dust collectors, releases for those years are estimated using the same model used for years when the scrubbers were in operation.

Current release estimates are based upon a review of the following:

- previous release estimates (Semones and Sverdrup 1988);
- plant operating data from 1969, 1970 and 1973;
- the Shift Foremen's Logs for 1956–1962 and 1967; and
- uranium trioxide production data.

The log sheets and logbooks contained information on parameters important for the calculation of releases due to gulping operations. Uranium released from the Plant 2/3 scrubbers is composed of releases due to scrub liquor *entrainment* and to particles of UO_3 in the air stream that pass through the scrubber. Independent estimates of releases from the Plant 2/3 scrubber system were performed using models of scrubber penetration by particles and mist reentrainment that were based upon the recent effluent measurements. Monte Carlo techniques were then used to sample the parameter distributions and the randomly selected parameter values were used to make the release estimates. The parameters considered in calculating the releases estimates are:

- scrubber outage fraction
- scrub liquor concentration
- entrainment release factor
- amount of UO_3 in a pot
- gulping time
- gulping release factor.

Estimates of Plant 2/3 scrubber releases obtained from the Monte Carlo calculations are shown in Table 2 by decade. Median estimates of releases during three of the four decades of operation are comparable, about 20,000 kg, while the value for the 1980s was much lower. The median release estimate for the entire period of operation was 66,000 kg uranium. This estimate was bounded by 5th and 95th percentile values of 56,000 and 78,000 kg uranium, respectively. The highest annual releases were estimated for the period 1957–1961.

Table 2. Summary Release Estimates for Plant 2/3 Scrubbers

Period	Best Estimate of Release (kg U)	Other percentiles in distribution of release estimate (kg U)			
		5th	25th	75th	95th
1950s	24,000	18,000	21,000	26,000	32,000
1960s	19,000	14,000	17,000	21,000	25,000
1970s	22,000	17,000	20,000	25,000	29,000
1980s	980	730	850	1,100	1,600
1953–1988	66,000	56,000	62,000	71,000	78,000

About 25% of the release is estimated to have been small particles of UO_3 that penetrated through the scrubber. The larger fraction (~75%) would have been uranyl nitrate hexahydrate (UNH). The estimated size range for these particles is 19–100 µm. An alternative calculation of releases from the Plant 2/3 denitration operations, based on a change in the outage fraction, is described in **Appendix H.**

RELEASES FROM PLANT 8 SCRUBBERS

Descriptions of Plant 8 operations, scrubber efficiency measurements, and the basis for both previous and current release estimates are given in **Appendix I** of this report and in the Task 4 report (Killough et al. 1993). Ten air scrubbing systems in Plant 8 cleansed, or scrubbed, the exhaust air by contact with droplets of caustic liquid. Six of the scrubbers—the rotary kiln, oxidation #1, the caustic or primary calciner, uranium ammonium phosphate (UAP) furnace, the oxidation #2 or NPR, and the green salt reverter—handled hot exhaust gases from the kiln and furnaces. The other four scrubbers—old digester, new digester, the ammonium diuranate (ADU), and the leach tank—treated ventilation air collected above the digestion and other process tanks. Some of the key findings that affect the current release estimates are:

- The exhausts from these systems were not sampled on a regular basis. Periodic measurements of discharge concentrations and of scrubber efficiencies were performed by the Industrial Hygiene and Radiation Department. A number of their measurements for the caustic, kiln, UAP, and NPR scrubbers were made during the early 1960s, a period of substantial concern about releases of uranium from these systems. In the early 1980s, when Plant 8 production was lower, measurements were made to determine emission factors for the Plant 8 scrubber discharges.

- There were no reported measurements of the sizes of the particles or liquid droplets released to the atmosphere from the Plant 8 scrubbers. A theoretical analysis of Plant 8 scrubber operations was conducted to estimate these particle size distributions [see Appendix D of the Task 4 report (Killough et al. 1993)]. About 30% of the total uranium emitted from the Plant 8 scrubbers included solid particles of U_3O_8 of less than 10 micrometers in diameter. The remainder of the released uranium from the scrubbers escaped as large droplets (80 to 180 µm in diameter) of reentrained scrub liquor. Evaporation of the liquid produced relatively large solid particles.

Previous estimates of releases from the Plant 8 scrubber systems were reviewed. An important difficulty with previous estimates of the Plant 8 scrubber releases was the assumption of a constant scrubber efficiency. Just as with these previous estimates, current estimates require knowledge of scrubber efficiencies and uranium concentrations in the scrubber liquor. Plant records were found in storage that provided data on the amounts of uranium scrubbed from the airborne effluents during periods ranging from one month to one year. Plant 8 production (uranium recovery) data were compiled to indicate the changing scale of plant operations. Memoranda and analytical data sheets were located that described measurements of scrubber efficiencies performed in Plant 8, primarily during 1961–1965. These data were compiled for each scrubber for use in calculations of releases from 1953 through 1981. Data collected in the 1980s on short-term measurements of release rates from the various stacks were also compiled and used for calculations for this later period.

For the years 1953–1981, annual uranium releases from the Plant 8 scrubbers and the uncertainties associated with them were estimated by applying a simple model to each scrubber. The calculations used the following plant-specific data:

- Plant 8 production (uranium recovery) data;
- amounts of uranium found in scrub liquor;
- the amount of uranium in scrub liquor per unit production;
- the use and performance of the scrubbers serving the calciner, rotary kiln, UAP furnace and the two oxidation furnaces.

For the latter years of FMPC operation (1982–1988), release estimates were based upon the operating times for the various scrubbers and release rate measurements that had been made during scrubber operation. For both time periods, simple models of releases were applied to individual scrubbers. When information on scrub liquor collections was not available, the 6- to 12-month average ratio of plant production to the amount of uranium collected in scrub liquor was found to be a reasonable link between production data and scrubber operations.

Monte Carlo calculations were performed to estimate uranium releases from the Plant 8 scrubbers. The ranges of all of the parameters used in calculations were relatively broad, owing both to variability and to limited historic data. Table 3 contains summary release estimates by decade and for the entire period from 1953 through 1988. The table illustrates the importance of the releases during the 1960s when plant production was highest. The median estimate for the 1950s was second highest, about 60% of that for the following full decade of operation. Alternative calculations of releases from the Plant 8 scrubbers, performed to test the effect of different modeling choices on the results, are described in Appendix I. The first alternative used correlations between scrubber penetration and the accumulation of uranium in the scrub liquor for the calculation. The second alternative approach was based on ratios of release to production for the early 1960s, when the scrubbers were studied most intensively. These ratios were applied to the entire period of operation.

The release estimates for the Plant 8 scrubbers that are summarized in the table are higher than previous FMPC estimates. The fundamental reason for the difference is that the present calculations consider ranges of individual scrubber performance that are broader than the single collection efficiency of 83 percent that had been assumed for all of the scrubbers.

Analysis of the Plant 8 scrubber releases suggests that two distinct types of particles were present in the emissions. The first type consisted of solid particles of U_3O_8 of less than 10 micrometers in diameter which penetrated the scrubber systems. The second type was droplets of entrained scrub liquor that contained suspended uranium particles. During the first two decades, when releases were highest, it is estimated that about 25% of the releases were of small particles of U_3O_8 and that the remainder were the result of entrainment of contaminated scrub liquor containing suspensions of uranium compounds.

Table 3. Summary Release Estimates for Plant 8 Scrubbers

Period	Best Estimate of Release (kg U)	Other percentiles in distribution of release estimate (kg U)			
		5th	25th	75th	95th
1950s	29,000	17,000	23,000	37,000	53,000
1960s[a]	47,000	30,000	39,000	57,000	78,000
1970s	1,700	1,000	1,400	2,100	2,700
1980s	1,400	980	1,200	1,600	2,000
1953–1988	81,000	56,000	69,000	95,000	130,000

[a] In making these estimates it was assumed that the bypass for the UAP scrubber operated 10 per cent of the time between September 1963 and April 1966.

To put these atmospheric releases into perspective, Figure 10 compares the uranium released annually from the dust collectors, the Plant 8 scrubbers, and the Plant 2/3 denitration processes. The dust collectors dominated the releases in the 1950s with 120,000 kg of uranium released, with a maximum of 54,000 kg of uranium released from them in 1955 alone. In the 1960s, the Plant 8 scrubbers dominated the releases, with approximately 47,000 kg uranium released during that decade, compared to 21,000 and 19,000 kg U for the dust collectors and Plant 2/3 scrubbers, respectively. In the 1970s, the Plant 2/3 scrubbers were relatively more important, discharging 22,000 kg U, compared to 3,100 and 1,700 kg U, respectively for the dust collectors and Plant 8 scrubbers. Again in the 1980s, the dust collectors contributed most to the total uranium releases, although the magnitude of all releases in the 1980s was significantly less than at any other time.

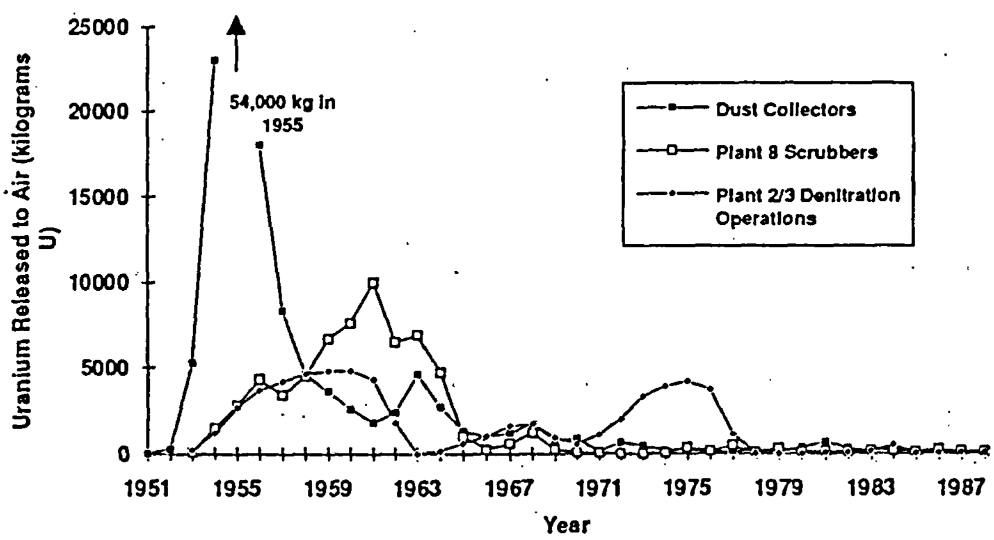

Figure 10. The best estimates of annual releases of uranium to the atmosphere from the Plant 8 scrubbers (square), the dust collectors (triangle) and the Plant 2/3 scrubbers (circle). The relative importance of each of these sources to the total atmospheric uranium release changes with each decade.

OTHER SOURCES AND EPISODIC RELEASES TO THE ATMOSPHERE

Appendix K addresses other miscellaneous unmonitored sources and accidental releases to the atmosphere. The unmonitored sources include emissions from:

- five waste incinerators,
- building exhaust and lab hood ventilation,
- miscellaneous unmonitored process emissions, and
- the waste pits.

Accidental releases include:

- non-routine events, and
- episodic releases.

Episodic releases are actual accidental releases which occurred in the past, and which were large enough to be given special treatment in terms of environmental transport and dose assessment. In addition to actual episodic releases, non-routine releases from other events, such as spills, fires and leaks of gaseous uranium hexafluoride and uranyl nitrate, were estimated in a generic way based on the frequency of occurrence of such events. Table 4 presents the total release estimates from the miscellaneous unmonitored sources. In addition, the table illustrates the difference between our reconstructed source terms and those previously developed by the FMPC contractor. In contrast to previous estimates, the reconstructed source terms all carry some estimate of uncertainty and are well documented.

Releases from these sources were more thoroughly examined than they had been in the interim source term report (Voillequé et al. 1991). There, only a few revised source term estimates were developed. Although releases from these sources were believed to be relatively minor compared with the dust collectors and scrubber emissions, the documentation to support that conclusion was lacking in most cases, and some of the previous methods used to estimate releases needed improvement. The detailed assessments in Appendix K provide thorough documentation of the magnitude of these sources, with uncertainties.

Miscellaneous Unmonitored Emissions

The agreement between past and revised release estimates is good for the incinerators. Of all incinerators at the FMPC, the old solid waste incinerator had the highest total release of uranium, with a median estimate of 2200 kg. The reconstructed median release estimate from building ventilation or exhausts (4100 kg U) is over ten times higher than the previous estimate, due to two main reasons:

(1) the use of lower dilution factor for building make-up air, and
(2) the use of higher in-plant airborne contamination levels, measured in the 1950s, to make a forward projection through 1970.

The median release estimate for non-routine releases (1300 kg U) is less than that previously calculated by Vaaler and Nuhfer (1988), although the 5th and 95th percentile range encompasses the previous estimate. The median estimate of releases from the waste pits (3000 kg U) was about twice as high as previous results, because we used a model (i.e., the resuspension algorithms found in MILDOS) that was highly sensitive to soil particle size which varied greatly among the pits.

Table 4. Summary of Total Estimated Releases of Uranium from Miscellaneous Unmonitored and Accidental Sources at the FMPC

		Total Release Estimate (kg U)		
Source	Inclusive Dates	Median	5th–95th Percentile Range	Previous Estimate[a]
Miscellaneous Unmonitored Releases				
Old Solid Waste Incinerator	1954–1979	2200	1600–2900	2471
Oil Burner	1962–1979	370	270–470	467
Graphite Burner	1965–1984	230	61–730	129
New Solid Waste Incinerator	1979–1986	8	0.6–90	14
Liquid Waste Incinerator	1983–1986	4	0.9–9	12[d]
Building Ventilation	1954–1987	4100	970–15,000	390
Unmonitored Process Emissions	1953–1988	[b]	110–970[c]	324
Lab Hoods	1953–1987	[b]	20–200[c]	66.5
Waste Pits	1953–1988	3000	900–12,000	1560
Accidental Releases				
Non-routine Releases[e]	1952–1988	1300	780–2900	2784
Episodic Releases[f]	1953, 1960, 1966, 1978, 1979, 1983	1700[f]	1300–2100[f]	Not defined previously

[a] From FMPC operating contractor. See individual sections of Appendix K for sources of information.
[b] Not reconstructed; estimate developed previously by the FMPC contractor.
[c] Subjective uncertainty of a factor of 3 applied to previous estimate.
[d] Based on maximum processing rate.
[e] Includes fires, spills, and leaks of uranium hexafluoride and uranyl nitrate.
[f] Does not include the November 1960 episodic release from the Pilot Plant dust collectors, which is included in the total dust collector source term. Does include two accidental releases of uranium hexafluoride and three releases (unknown sources) identified from ambient air monitoring.

Accidental Releases

Accidental releases are frequently characterized as increases in the effluent discharge rates due to unplanned and non-routine events. In previous historic reports, typical events included spills, fires, and cleanup system failures. However, when the frequency of the unusual events is high, one questions whether the adjective "accidental" is correct. Similarly, when a large release is the result of a conscious operational decision, it hardly qualifies as unplanned. Such situations complicate the definition of the term accidental releases; so the term "episodic releases" has been defined and used in the Fernald Dose Reconstruction Project. Criteria for an episodic release, discussed fully in **Appendix K**, that were used to determine whether special evaluation of a release from a particular event is warranted include:

- the event under consideration caused the composite release rate of the FMPC to increase by a factor of ten or more above the value that would otherwise have been observed, and
- the duration of the high release rate caused by the particular event was less than 10 days.

Six incidents involving releases of uranium were identified which met our criteria for special treatment as episodic releases. It should be emphasized that all known releases are included in the total source term estimates, but only a small number are truly episodic releases, by our definition. Three episodes, documented in incident reports, occurred on November 7, 1953, in November 1960, and on February 14, 1966. The remaining three episodes were identified by air monitoring data, although documentation could not be found to identify the sources. These events occurred sometime during the weeks ending on September 28, 1978, February 8, 1979, and September 20, 1983. In terms of total quantity of uranium released, the dust loss episode in November 1960 had the most impact. However, the episode on February 14, 1966 had the largest release rate, releasing 750 kg U in one hour. A release of about 30 Ci of radon occurred on April 25, 1986, from unauthorized venting of the K-65 silos. This source term may also be treated separately as an episodic release.

Figure 11 compares the relative importance of the various unmonitored sources with releases from the dust collectors, the Plant 2/3 denitration operations and the Plant 8 scrubbers. It is clear that the magnitude of uranium releases from the miscellaneous unmonitored sources is minor relative to the three major sources of atmospheric emissions from the FMPC (Figure 10). When all of the miscellaneous sources investigated in Appendix K are combined, using appropriate statistical measures, the grand total of the releases is 16,000 kg (median estimate), with a 5th–95th percentile range of 9,300 to 28,000 kg. This total does not include the November 1960 dust loss from the Pilot Plant, which is included with the total dust collector source term.

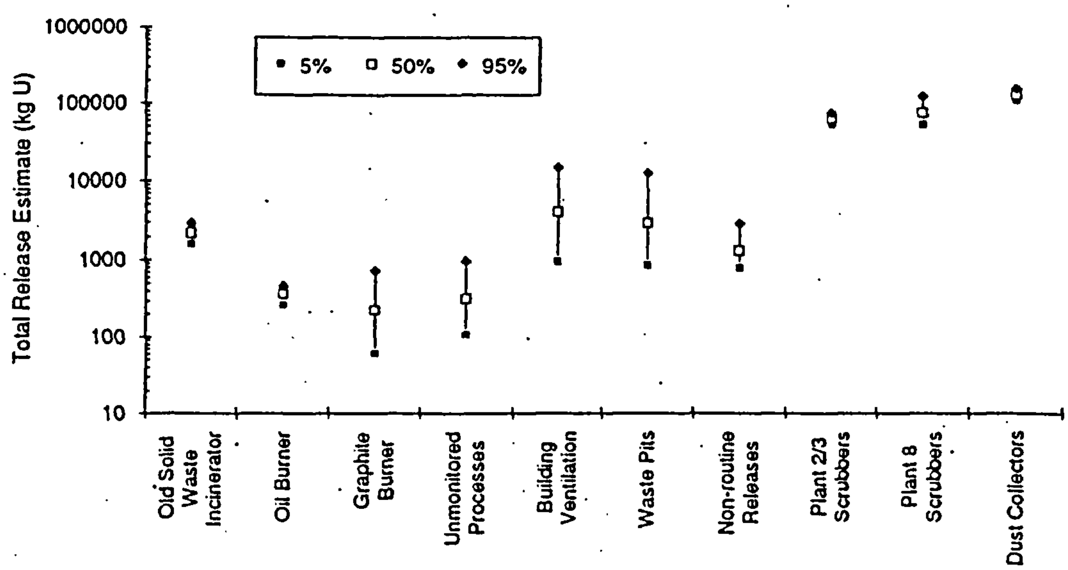

Figure 11a. *Note logarithmic scale.*

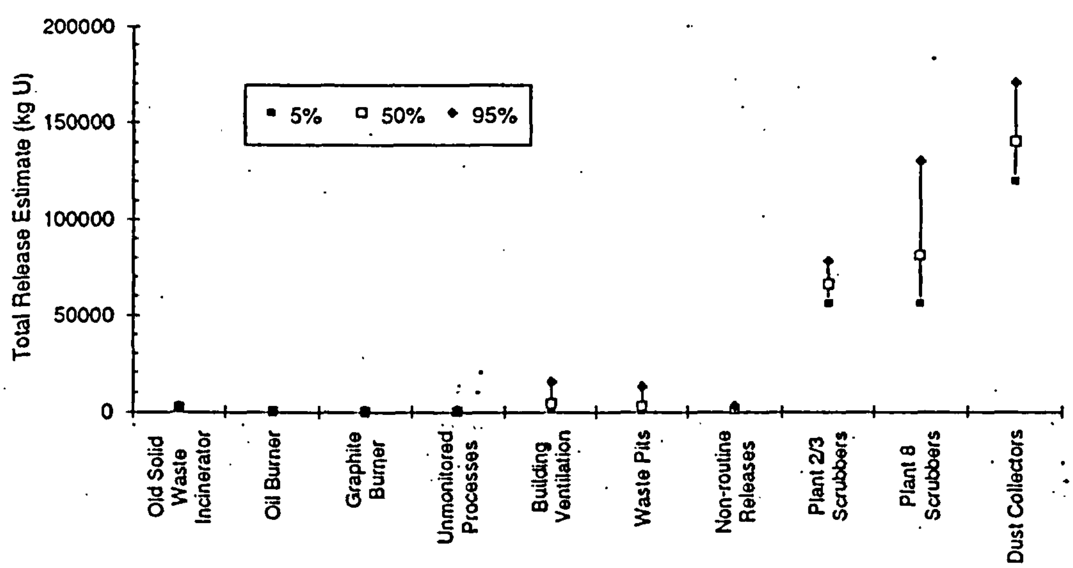

Figure 11b. *Note linear scale.*

Figure 11. Relative importance of miscellaneous unmonitored sources of atmospheric releases of uranium compared with releases through scrubbers and dust collectors. The 50% point represents the median (best estimate). The 5% and 95% points encompass a 90% probability range on the total estimates. Figure 11a is plotted on a logarithmic scale, so that the uncertainty distributions can be seen more clearly, while Figure 11b is plotted using a linear scale, which more accurately illustrates the true relative magnitude of these sources.

RADON AND DECAY PRODUCT RELEASES FROM K-65 SILOS AND MATERIALS

The main source of radon-222 release from the FMPC is material stored in the K-65 silos, which contain residue, called K-65 material, from the extraction of uranium from pitchblende or other uranium ores. Originally, the waste residues from the processing, including the K-65 material, were to be returned to the supplier, the African Metals Corporation. On an "interim" basis, the wastes were stored at processing facilities, where they remain. The K-65 material contains very high concentrations of radium-226, and consequently, is a significant source of radon-222 emissions.

The K-65 material at the FMPC has primarily been stored in large concrete storage tanks, called the K-65 Silos, located in the waste storage area of the site. Figure 2 shows the location of the K-65 Silos, as well as two other waste storage silos. Silo 3, the Metal Oxide Silo, contains the metal oxide waste material, another waste residue from the extraction processing of uranium ores. The metal oxide material is also contaminated with radioactivity, but the concentration of radium-226 is much lower than in the K-65 material. Silo 4 has never been used, and contains only a small quantity of water with very low levels of radioactive and chemical contaminants. The Metal Oxide Silo and Silo 4 are not considered significant sources of radon-222 releases. Belgian Congo uranium ores were also processed at the Mallinckrodt Chemical Works (MCW) facility in St. Louis. Due to insufficient storage capacity at MCW, K-65 material from MCW was shipped to the FMPC, beginning in 1951, before construction of the K-65 Silos was complete. That K-65 material was stored in 55-gallon drums on the storage pad around Plant 1.

Appendix J contains the detailed descriptions of the radon-222 and radon daughter release estimates, including more information about the K-65 and metal oxide materials and storage silos; a summary of previous estimates of radon releases, by others; a discussion of potential radon sources at the FMPC; descriptions of our calculational strategies for current estimates of releases; models and calculated releases for the different time periods assessed; and a discussion of an alternative calculation, for comparison with current estimates. The following sections provide some information about the history of K-65 materials at the FMPC, and our estimates of radon-222 and radon decay releases from the site.

History of K-65 Silos and K-65 Material at the FMPC

The K-65 Silos were constructed in August 1951 through July 1952 for storage of K-65 materials. However, MCW began shipping K-65 material to the FMPC before construction of the FMPC silos was complete. By the end of July 1952, about 13,000 55-gallon drums of K-65 material (equal to about half the capacity of one Silo) had been received at the FMPC. Before disposal in the Silos began, the drummed K-65 material was stored on the concrete ore storage pad around Plant 1, the Sampling Plant, for the period September 1951–mid-June 1953. The K-65 material was added to the Silos from July 1952 through September 1958. We thus calculate radon-222 and radon decay product releases from:

- the K-65 Silos, and
- stored drums of K-65 material on the storage pad near Plant 1 for 1951–1953.

The K-65 Silos have had problems of deterioration, almost since the time of construction. Significant cracking in the walls and seepage of the contents was noted from the 1950s (Wunder 1954; Martin 1957). Because of these problems, repairs and improvements to the

Silos occurred from the 1960s through the 1980s. Not all of the changes to the Silos would have had a significant effect on the releases of radon. The most important change, for radon emissions, was the sealing of penetrations of the Silo domes in 1979. This action would have significantly reduced the ventilation of the silo air spaces, and thus also reduced the radon releases from the Silos. The addition of an exterior foam layer on the silo domes in 1987 may have further reduced the emissions of radon. Earthen berms were built around the Silos in 1964. However, at that time the radon releases occurred primarily through openings in the silo domes, so the addition of the berms would not have altered the releases.

Based on these changes to the K-65 Silos and on the operational periods of them, we estimate radon and radon daughter releases from the silos separately for each of the following periods:

- mid-July 1952 to mid-June 1953 (operational period for Silo 1)
- mid-June 1953 to mid-September 1958 (operational period of Silo 2)
- mid-September 1958 to June 1979 (both silos inactive; prior to sealing penetrations),
- July 1979-to December 1987 (both silos inactive; after sealing penetrations), and
- 1988 (1988 is the last year of concern for this project).

Current Estimates of Radon Releases

For some other releases at the FMPC, extensive data sets of direct measurements of release quantities are available. However, for radon and radon decay product releases there are no direct measurements of release quantities. In addition, until the 1980s there were very few measurements of parameters that can be used indirectly to calculate radon releases. Because of this limited availability of data, we use models to estimate radon release quantities.

The traditional model used to estimate radon releases from radium-226-bearing material, such as uranium mill tailings, involves calculations of the quantity of radon formed in the material, and the subsequent diffusion of the radon through the material to the outside air. For the K-65 materials, measurements have not been made of the radon diffusion coefficient and radon emanation fraction, which are two key parameters in this traditional calculation. Literature values can be obtained for these parameters, but without site-specific values, the uncertainty ranges are extremely large. To reduce the uncertainties in our results, we have used different models, which we believe make the best use of the limited data that are available. Appendix J describes the available, useful information; the information lacking, that would be useful to improve estimates; and the general approach to estimating radon releases. The methods used for 1980–1987 are generally similar to those used in previous release estimates (Borak 1985; IT 1989; Grumski 1987; Boback et al. 1987), though additional data have been obtained and used.

There are no direct data available for estimating releases of radon decay products. Thus, radon decay product releases are calculated to be equal to radon releases multiplied by two correction factors. The first correction factor accounts for the expected ratio of radon decay product concentrations in the silo air to the radon concentration (equilibrium fraction). The second is a fractional release factor, that accounts for deposition of radon decay products along the release path (such as cracks in the silo domes, or penetrations in the domes), which reduces the quantities of decay products released.

As for other releases, we use Monte Carlo methods to perform the calculations of radon and radon decay product releases, so that uncertainties are calculated along with best estimates. The estimated release rates from the K-65 Silos are plotted versus time in Figure 12. The cumulative quantity of radon released from the K-65 Silos for 1959–1979 is larger than for other periods, due to the length of this period and the higher release rate for the period. Releases for this period may also be important in terms of potential doses to offsite people. The predicted radon release rate from the K-65 Silos remained elevated through most of the 1970s, while uranium releases to air generally decreased through the 1970s compared to the 1960s (see Figure 10 and Table 11).

The predicted total quantities of radon released from the FMPC for 1951–1988, are summarized in Table 5. From this summary, it can be seen that radon releases from the drummed K-65 material stored on the Plant 1 pad are relatively insignificant contributors to the total radon releases for the period 1951–1988. However, the radon releases from the drummed K-65 material occurred when operations at the FMPC were just beginning and releases of uranium were relatively small. Consequently, radon releases from the drummed K-65 material may be significant contributors to site-wide releases of all radionuclides from 1951–1953.

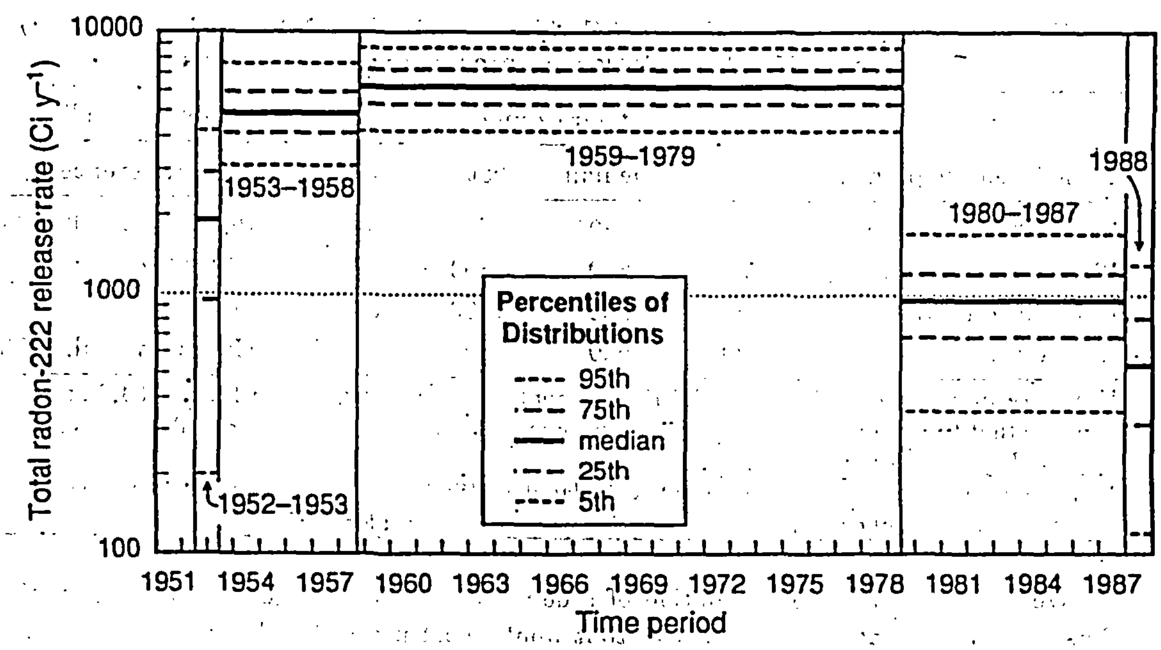

Figure 12. Estimated radon-222 release rates from the K-65 Silos as a function of time. The periods indicated are only the nominal periods; the more precise dates are given in Appendix J.

Table 5. Summary of Predicted Total Radon and Radon Decay Product Release Quantities (Ci) from the FMPC for the Period 1951–1988

	Radon released			Decay products released[a]		
Source of releases	5th	median	95th	5th	median	95th
K-65 Silos	110,000	170,000	230,000	87,000	130,000	190,000
Drummed K-65 material stored on Plant 1 pad	54	720	3,400	4.5	130	880
Both sources	110,000	170,000	230,000	87,000	130,000	190,000

[a] The release quantities for radon-222 decay products are release quantities of each of the short-lived decay products, polonium-218, lead-214, bismuth-214, and polonium-214.

Table 6 presents a comparison of our results with previous estimates of the emissions of radon from the K-65 Silos. The other studies did not report uncertainties associated with the release rate estimates. However, results of the other studies generally fall within, or close to, our 90% probability interval (5th to 95th percentile) of release rates.

Table 6. Comparisons of Current Estimates of Radon Release Rates (Ci y^{-1}) from K-65 Silos to Release Rates from Other Studies

	Percentiles of our estimates			Results of other studies	
Period, release pathway	5th	median	95th	Value	Reference
1980–1987, diffusion	72	130	240	60 [a]	Borak 1985; IT 1989[b]
1980–1987, air exchange	230	810	1600	1023 [a]	IT 1989 [b]
1980–1987, total	360	950	1700	1083 [a]	IT 1989 [b]
1988, total	120	540	1300	1150 [b]	Hamilton et al. 1993

[a] These results were considered by IT (1989b) to apply to the complete period 1953–1984, but we believe that the conditions and parameters used to develop the estimates were only valid for the period July 1979–1987.

[b] This result was the average release rate calculated for 1989–1990. We compare it to our results for 1988 because we believe conditions of the Silos were unchanged for 1988–1991.

We did an alternative calculation of radon releases using more conventional methods. This method estimates radon releases that would exist if the Silo domes did not cover the K-65 material. The results of the alternative method are generally consistent with, but not as satisfactory as the current methodology because of very large uncertainties and the apparent underprediction of the radon releases.

DIRECT EXPOSURES FROM GAMMA RADIATION FROM THE SILOS

Radium-226 and other radionuclides in the materials stored in the K-65 and Metal Oxide Silos produce emissions of gamma radiation, which may have exposed people outside the FMPC. In our Task 4 Report (Killough et al. 1993), we described the methodology to be used to calculate exposures and doses due to this direct radiation. Exposure rates will be calculated using the MicroShield 4 computer software (Negin and Worku 1992). In Appendix J, we provide additional information, necessary to complete the exposure calculations that will be reported in the Task 6 report.

The two K-65 (Silos 1 and 2) and the Metal Oxide (Silo 3) Silos are the only significant sources of direct radiation exposures to people outside the FMPC boundary. This conclusion is based on the results of aerial radiation surveys of the FMPC site and surrounding area, and results of penetrating radiation monitoring performed by the FMPC along the site boundary. Additional information is used for direct exposure calculations, including:

- concentrations of radionuclides in the Silos 1,2 and 3,
- concentrations of radionuclides in the air space of the K-65 Silos,
- densities and moisture content of the materials stored in the Silos, and
- information about the time-history of filling of the K-65 Silos.

LIQUID WASTE DISCHARGES FROM FMPC

Liquid wastes that are generated at the FMPC come from three main sources: process water via the clearwell portion of the waste pit, sanitary sewage, and storm water. Figure 2 shows that liquid effluent streams from FMPC are released to the offsite environment at two locations. These are (1) the combined sewer outfall which discharges through Manhole 175 into the Great Miami River at a point almost directly east of the plant site, about three miles upstream from New Baltimore and (2) the storm sewer outfall which discharges into a branch of Paddy's Run onsite. **Appendix L** provides more detailed descriptions of the principal contributors to liquid discharges from the FMPC and the types of documentation used to tabulate the discharges.

Releases of Uranium in Liquid Effluents from the FMPC

To the Great Miami River. Manhole 175, located on the eastern side of the facility, is the discharge point for waste water leaving the site through the main effluent line to the Great Miami River. It is the final junction point of the major waste effluent streams from the facility. The discharge flow to the Miami River was continuously measured. A composite sample was collected and analyzed for uranium on a daily basis. These daily uranium measurements were found for most years in the 1950s and 1960s. Daily flow rate measurements were located for 1958–1964, and monthly totals were available for later years. When specific information was not located for a particular month, an average value, based on the other months in the same year, was used.

The quantity of uranium released to the river is the product of the uranium concentration multiplied by the flow volume. Sources of uncertainty for these estimates of uranium losses through Manhole 175 to the Great Miami River come primarily from the analytical errors in measuring effluent flow, and in sampling and measuring uranium concentrations in the water.

To Paddy's Run. Runoff water collected in the storm sewer system passed through the storm sewer lift station before release through Manhole 175 to the river. Since the storm sewer lift station was not connected to any process, all the uranium lost through it was assumed to be from leaks and spills (Ross, 1972). When the capacity of the storm sewer lift station was reached, water overflowed through the storm sewer outfall to Paddy's Run. The volume of storm water that overflowed the storm sewer lift station was related to rainfall amounts and patterns.

Estimates of uranium losses from the storm sewer outfall to Paddy's Run were based upon analytical data sheets and monthly reports which listed the individual outfall events occurring during that month. There are three major components of uncertainty associated with estimation of uranium losses to Paddy's Run:

- the analytical errors associated with determining uranium concentration and water flow before discharge to Paddy's Run.
- time periods when rainfall, and consequently runoff, were quite high and the capacity of the storm sewer lift station flow meter and v-notch weir at Paddy's Run was exceeded.
- unmeasured losses from the site above the point where the storm sewer outfall enters Paddy's Run (where the measured losses were recorded).

Figure 13 shows the annual uranium release estimates to the Great Miami River and to Paddy's Run for all years. The magnitude of the uranium releases to the river peaked in 1961 with 7300 ± 140 kg uranium. From 1974 onward, the annual releases were below 1000 kg. The uranium losses to Paddy's Run show much more month-to-month variation than do the uranium losses to Manhole 175 (MH 175). However, the average quantity of 500 kg uranium discharged through Manhole 175 to the Great Miami River each month during the early 1960s was roughly five times greater than the average quantity of 100 kg of uranium lost to Paddy's Run during that same time.

Other Radionuclides Released in Liquid Effluents

Release estimates for thorium, radium-226, radium-228, and fission and activation products are based on correlations between the total annual releases of uranium and those of the other radionuclides. These ratios of releases, computed for years when measurements were made, provide a basis for estimating the release of the other radionuclides for years when they were not measured. This methodology is described in **Appendix D** in the present report, and in Appendix C of Task 4 (Killough et al. 1993). Ratios of the annual average activity of a radionuclide (or quantity of thorium) to the annual uranium quantity were calculated for years when data were available. The measured concentrations at MH 175 reported in analytical data sheets were used to calculate the ratio for some years. Annual average concentrations of radium, thorium and the fission and activation products in liquid effluents were reported by the FMPC in historic release reports (Boback et al. 1987), and in annual environmental monitoring reports beginning in 1976. The variability of the release ratio from year to year was considered in deriving the uncertainty associated with the estimated releases of these other radionuclides. The release estimates and uncertainty analysis were computed using Monte Carlo techniques in the Crystal Ball® program (Decisioneering 1993).

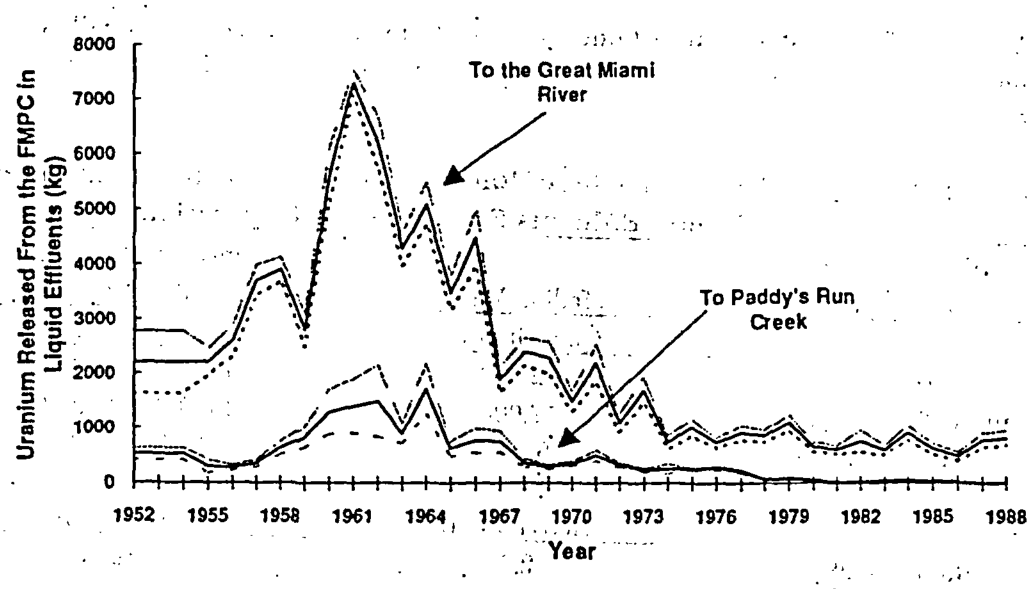

Figure 13. Uranium losses to the Great Miami River via Manhole 175 and to Paddy's Run from the FMPC from 1952–1988. The uncertainty of each estimate is described by the 95th percentile (top, broken line), and the 5th percentile (lower, dotted line).

Table 7 summarizes our estimates for releases of materials in liquid effluents from the FMPC for all years of operation. Our best estimate of uranium released to the Great Miami River for all years is 82,000 kg. The 5th to 95th percentile uncertainty range is 71,000 to 94,000 kg of uranium. Some estimates of uranium in liquid wastes have been made by others on an annual basis (Boback 1971), or, in summary reports evaluating the past discharge history of the facility (Rathgens 1974, Boback et al., 1985). These estimates of uranium to surface water from 1951 through 1984 range from 74,000 to 77,000 kg (Boback et al. 1987, Galper 1988) and fall within the uncertainty range of our estimates. Revisions to historic discharge reports generally focused on amending estimates of uranium loss to airborne effluents, and did not include updated figures for liquid effluents (Boback et al. 1985, Boback et al. 1987).

The total release estimate for uranium to Paddy's Run via the storm sewer outfall ditch and runoff is 17,000 kg of uranium. The 5th to 95th percentile uncertainty range is 14,000 to 20,000 kg of uranium. Losses to Paddy's Run show much more month to month variation than do the uranium loss estimates to the Great Miami River. The highest annual releases of uranium occurred from 1960 to 1964, when the average quantity of uranium discharged through MH 175 to the river was approximately 500 kg each month, about 3 to 4 times greater than the average quantity of uranium lost to Paddy's Run each month.

The other materials released at various times over the years include decay, fission and activation products of uranium, thorium and recycled uranium. Recycled uranium was not processed at the site until late 1962, so releases of fission and activation products would not have begun until that time. Releases of thorium, and one of its decay products, radium-228, occurred when thorium was processed at the site in 1954–1957, and 1964–1988. Releases of radium-226 occurred throughout the history of the site; and the total release is estimated at

18,000 mCi or 18 Ci, with an uncertainty range of 15 to 22 Ci. These values will be used to calculate radiation doses to the population in the vicinity of the FMPC in our final task report.

Table 7. Summary of Total Estimates of Radioactive Materials Released From the FMPC in Liquid Effluents For All Years of Operation

Material Released to Great Miami River	Median Value	Uncertainty Range (5th %ile to 95th %ile)
	Quantity (kg)	Quantity (kg)
Uranium	82,000	71,000 to 94,000
Uranium (To Paddy's Run)	17,000	14,000 to 20,000
Thorium	5,800	3800 to 9400
	Activity (Ci)	Activity (Ci)
Radium-228	2.7	0.33 to 20
Radium-226	18	15 to 22
Plutonium-239,240	0.0088	0.0019 to 0.033
Plutonium-238	0.00028	0.00016 to 0.0034
Neptunium-237	0.0044	0.0011 to 0.018
Cesium-137	0.54	0.14 to 1.9
Ruthenium-106	0.056	0.014 to 0.22
Technetium-99	300	110 to 800
Strontium-90	6.0	1.5 to 24

The chemical form of uranium in liquid effluents is not known with certainty, but several uranium species of both the +4 and +6 oxidation states may have been present in solution in liquid waste streams during this period. The ratios of these various ionic species in the process waste streams, in Paddy's Run, or in the main effluent pipeline to the river, would be a function of the pH of the water. The presence of suspended solids in the liquid wastes is considered in assessing the relative solubility of uranium in liquid releases. Daily measurements of total suspended solids (TSS) were made on 24-hour composite effluent samples at MH 175 beginning in 1956 (NLCO 1956). Among the suspended solids may have been very small particulates of the insoluble U_3O_8 and UO_2. Not all the suspended solids measured on a daily basis were uranium, but the average monthly values may provide an upper bound, or conservative estimate, for the amount of insoluble uranium that was released in liquid effluent. Furthermore, some uranium-containing suspended solids that were released into the waste streams might have dissolved during dilution downstream from the FMPC.

URANIUM CONTAMINATION IN GROUNDWATER OUTSIDE THE FMPC

Contamination of the groundwater could occur either by direct discharge of waste waters to it or by infiltration of contaminated water through the soil. No evidence of direct discharges to the groundwater from the facility has been found in review of historic documents. Concern about the infiltration pathway has been evident in FMPC documents since the late 1950s, and a variety of studies and analyses have been conducted from that time to the present day (Eye 1961, Dove and Norris 1951, Hartsock 1960, Spieker and Norris 1962). Recent reports describe the measured contamination levels in groundwater, primarily to the south and southwest of the FMPC that have resulted from infiltration of water bearing uranium to the aquifer (GeoTrans 1985, ASI-IT 1990). Uranium contamination of groundwater outside the FMPC has been known since late 1981, when the first samples of water from private wells were analyzed. The significant offsite uranium contamination in groundwater is south of the site, and is now called the "South Plume." There are additional known areas of groundwater contamination on the FMPC site, but only the South Plume area extends outside the site boundary. Since this dose reconstruction project is concerned with past doses to people around the site, the groundwater contamination under consideration here is limited to the South Plume. Figure 14 shows the estimated area of the South Plume contamination, as of 1991. Also shown are the locations of the private wells sampled by the FMPC monitoring program.

In our Task 4 report (Killough et al. 1993), we examined the potential importance of the groundwater contamination for doses to people around the FMPC. It was shown that only three of the private wells monitored, numbers 12, 15, and 17, have had measured uranium concentrations above the range of background. Although well 26 is within the area of groundwater contamination, it is installed deeper in the aquifer, and the uranium concentrations are at background levels. We concluded that because of the limited area of the South Plume, only a small number of people would have potentially received radiation doses from contaminated groundwater. Toward the main objective of this project, the determination of the feasibility of an epidemiological study, doses to these people would be less significant to the collective population dose than doses through other pathways. For this reason, we further concluded that a detailed assessment of the groundwater transport of radionuclides, and detailed assessments of doses to individuals potentially exposed through groundwater pathways, are not warranted. For other project objectives, it is still important to estimate potential doses through the groundwater pathway, so instead we use simple methods to estimate concentrations of uranium in the three contaminated wells. **Appendix** M contains details of our groundwater assessments.

Potential Sources of Groundwater Contamination

The status of groundwater contamination in the vicinity of the FMPC has been investigated. Appendix M describes a special study that was conducted to determine the primary transport pathway for uranium deposited on soil around the FMPC. The study compared uranium migration due to infiltration, surface soil erosion, and surface water runoff. Results of the study show that uranium deposited on soils is primarily transported by infiltration and that soil erosion transports the least amount of uranium. There are two potential sources of groundwater contamination originating on the FMPC site (see Figure 14): (1) historical releases of uranium-contaminated water to Paddy's Run and to the Storm Sewer Outfall Ditch (SSOD), and (2) possible releases from the solid and liquid waste pits in the waste storage area.

Of these two potential sources, the principal source of uranium contamination in the South Plume has been determined to be the historical releases to Paddy's Run and the SSOD (DOE 1990). The bottom sediments of Paddy's Run and the SSOD are very permeable in the area north and west of the South Plume, so these areas are recharge areas for the regional aquifer. Thus, uranium contamination in Paddy's Run and the SSOD percolates downward through the permeable sediments to ultimately reach the groundwater.

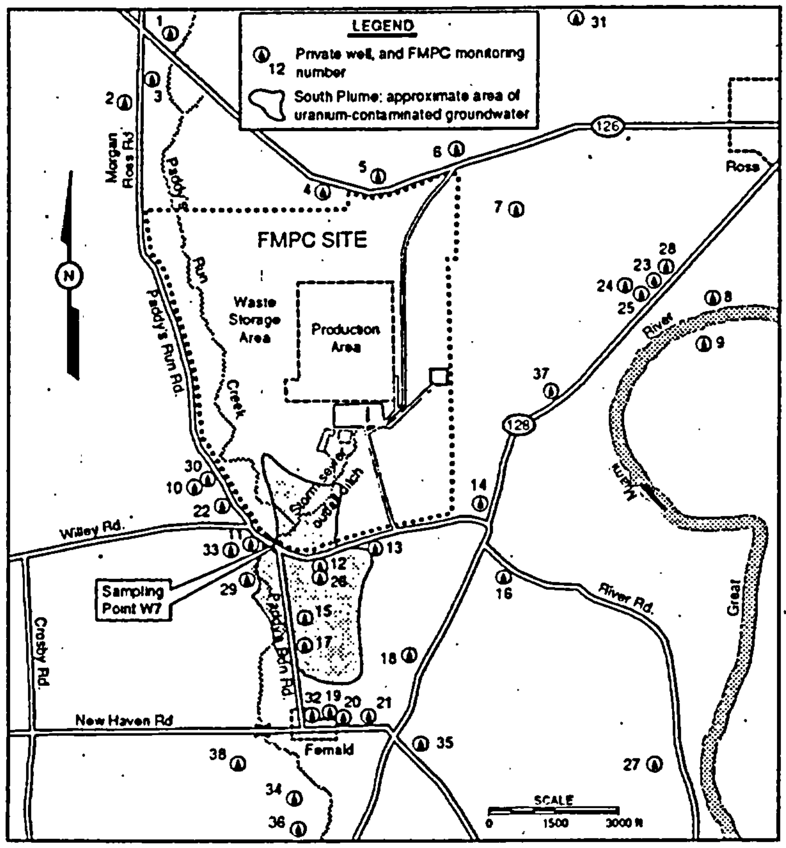

Figure 14. Approximate area of uranium contamination in the South Plume at the end of 1991, and locations of the private wells around the FMPC sampled in the FMPC routine monitoring program. Sampling point W7 is a location for sampling the surface water in Paddy's Run, at the Willey Road bridge.

Estimated Uranium Concentrations in Private Wells

A preliminary investigation of the movement of contaminated groundwater was performed, to determine the transport times required for uranium contamination to move from the source (waters in Paddy's Run and the SSOD) to offsite locations. The study is described more fully in Appendix M. Based on results of this preliminary assessment, we concluded that the South Plume would not have reached the offsite private wells in the South Plume area until after 1962. Thus, exposures of people using wells in the South Plume might have occurred from 1963 onward.

Monitoring of the three contaminated wells (wells 12, 15 and 17) was initiated in late 1981. Routine monitoring of these wells, as well as other private wells, has been performed by the FMPC since 1982. We obtained results of monthly measurements of uranium concentration in well water for the three contaminated wells for late 1981 through 1992. Annual average uranium concentrations are shown in Table 8. The annual average concentrations for 1982–1988 will be used as the basis of dosimetry calculations for these years.

For the period 1963–1981, for which well monitoring was not performed, we used models to estimate concentrations of uranium that might have existed in well water of the South Plume. We first developed an estimated upper bound on the annual average uranium concentration that could have existed in wells 12, 15, and 17. As mentioned above, the primary source of uranium contamination of the South Plume has been determined to be uranium-bearing waters released into Paddy's Run and the SSOD. Thus, uranium concentrations in the groundwater are expected to be at the most, equal to concentrations in Paddy's Run and the SSOD. Uranium concentration data for Paddy's Run and the SSOD were obtained and compiled in Appendices L and M. Uranium concentrations were higher in the SSOD than in Paddy's Run. In the SSOD, the maximum concentration of uranium was 8,300 pCi L^{-1}, for the year 1960. Thus, this value is used as the upper bound of the annual average uranium concentration that might have existed in the contaminated wells during 1963–1981.

We recognize that this upper bound is an extremely conservative estimate (that is, the estimated value is too high) of the uranium concentrations in the three contaminated wells for 1963–1981. The conservatism results because: (1) the maximum annual average concentration was used to represent the concentrations for the complete period, (2) dilution of the uranium with water from Paddy's Run (with lower concentrations than that of the SSOD) was ignored, and (3) dilution in the groundwater (from other groundwater sources) was also ignored. For the dosimetry calculations, we believe the use of the upper bound uranium concentration of 8,300 pCi L^{-1}, to represent concentrations in private wells of the South Plume area for 1963–1981, is unrealistically conservative.

Table 8. Annual Average Concentrations of Uranium (pCi L^{-1}) in the Three Contaminated Wells[a]

Year	Well 12	Well 15	Well 17
1982	170	320	45
1983	180	290	39
1984	170	220	36
1985	140	200	31
1986	150	190	31
1987	200	200	40
1988	170	190	38
1989	170	190	27
1990	130	180	30
1991	100	170	27
1992	100	150	25

[a] The range of long-term average, background concentrations of total uranium in private well water around the FMPC is 0.09 to 1.3 pCi L^{-1} (Shleien et al. 1993).

Thus, we developed an empirical model to estimate uranium concentrations in the contaminated wells. An empirical model is one based primarily on measurement data, rather than on theory, to explain the particular conditions. In this case, the data we used are the annual average measured uranium concentrations in the contaminated wells for 1982–1992, and the calculated quantities of uranium released to Paddy's Run and the SSOD for 1952–1988 (these releases are discussed in Appendix L). Details of the model are described in Appendix M. We think that the use of this model provides more realistic, though still somewhat conservative, estimates of uranium concentrations that might have existed in the contaminated wells for 1963–1981.

Table 9 summarizes the uranium concentrations in well water from the South Plume, that will be used for the dosimetry calculations (Task 6). The values for 1963–1981 are based on the empirical model. Based on the empirical model calculations, it is likely that uranium contamination in the groundwater would not have reached the offsite wells prior to 1968 (estimated concentrations are zero prior to 1968). The values for 1982–1988 are the annual averages based on measurements for well 15. Concentrations from well 15 are used in this assessment because they are the highest concentrations of the three contaminated wells.

Table 9. Values of Uranium Concentration (pCi L^{-1}) Used to Represent Annual Average Concentrations in Contaminated Wells of the South Plume Area

Year	Concentration	Year	Concentration	Year	Concentration
1951–1967[a]	0	1975	490	1983	290
1968	180	1976	580	1984	220
1969	230	1977	620	1985	200
1970	230	1978	620	1986	190
1971	230	1979	570	1987	200
1972	240	1980	510	1988	190
1973	290	1981	460		
1974	370	1982	320		

[a] The concentration listed is applied to each year in this range.

TASK 2 AND 3 SUMMARY AND CONCLUSIONS

The purpose of the Fernald Dosimetry Reconstruction Project is to estimate doses to the public who lived near the Feed Materials Production Center near Fernald, Ohio from the radionuclides released to the environment during operation of the facility. This report describes our best estimates of releases to the atmosphere and to surface water from FMPC operations, and from the K-65 Silos, during the period 1951–1988. Table 10 provides a summary of our best estimates these results.

Figure 15 shows the relative contributions of uranium released from the major sources at the FMPC facilities during the period. These major sources are uranium released to the atmosphere, uranium released in liquid effluents, and releases of radon gas and its decay products. They are shown in three main sections separated by vertical lines. Numerical values of the best estimate of release are shown next to the heavy bars that represent them.

The methods used to determine these release estimates are described carefully and fully in the accompanying appendices.

Table 10. Summary of Median Uranium and Radon Release Estimates From the FMPC for 1951–1988 With Uncertainty Bounds[a]

Source	Median release estimate	5th percentile	95th percentile
U to Atmosphere			
Dust Collectors	140,000	120,000	170,000
Plant 2/3 Scrubbers	66,000	56,000	78,000
Plant 8 Scrubbers	81,000	56,000	130,000
Miscellaneous Sources[b]	16,000	9,300	28,000
Total: airborne sources	310,000	270,000	360,000
U to Surface Water			
To the Great Miami River	82,000	71,000	94,000
To Paddy's Run	17,000	14,000	20,000
Total: surface water	99,000	85,000	120,000
Radon to Atmosphere			
K-65 Silos			
Radon-222	170,000 Ci	110,000 Ci	230,000 Ci
Radon-222 decay products[c]	130,000 Ci	87,000 Ci	190,000 Ci

[a] Values are in kg of uranium, except for releases from the K-65 silos which are reported in units of activity, called curie, Ci. Median estimates of releases from the various sources cannot be directly added to obtain a corresponding total median release estimate for all sources because medians do not have the additive properties that are associated with arithmetic means. See discussion on uncertainty in release estimates on page 10.

[b] These estimates do not include the November 1960 release from the Pilot Plant which is included in the dust collector releases.

[c] The release quantities for decay products are quantities of each of the short-lived decay products, polonium-218, lead-214, bismuth-214, and polonium-214.

It should be noted that uncertainties associated with the parameters used to determine these values vary considerably. In some cases, detailed measurements had been made and were located. An example is the uranium discharged in liquid effluent to the Great Miami River. In other cases, however, measurements of uranium losses were not made, and current release estimates are based on other information (for example, the Plant 8 scrubber releases). The median release estimates do not stand alone. The statistical parameters reported with these values in the appendices are an integral part of the release estimates; they should always be reported with them. The table and figure include ranges of estimates as well as the *best* estimates to provide a general comparative overview of annual release estimates for these years.

For the operational period of the FMPC, the total releases from atmospheric sources (dust collectors, Plant 2/3 scrubbers, Plant 8 scrubbers and miscellaneous sources) are 310,000 kg uranium, with the 5th to 95th percentile range of 270,000 to 360,000 kg. The predicted total quantities of radon and radon decay products released from the FMPC through 1988 are 170,000 Ci (5th to 95th percentile range of 110,000 to 230,000 Ci), and 130,000 Ci (5th to 95th percentile range of 87,000 to 190,000 Ci). For releases of uranium in liquid effluents, the median release estimate to the Great Miami River during this time period, is 82,000 kg (5th to 95th percentile range of 71,000 to 94,000 kg), while that to Paddy's Run is 17,000 kg, with the 5th to 95th percentile range of 14,000 to 20,000 kg.

It is important to realize that median estimates of releases from various sources may *not* be directly added to obtain a corresponding median estimate of the annual total release for all sources. The reason is that the medians do not have the additive properties that are familiar to most people from dealing with (arithmetic) means. We have chosen to use median estimates because they represent the 50th percentile of their distributions. For nonsymmetric distributions such as those encountered in this work (principally lognormal or approximately so), the mean is larger than the median by an amount that increases with the weight of extremely large values. For this reason, the median is considered a more stable measure of the central tendency of the distribution, and it is generally used in this study to represent best estimates of uncertain quantities.

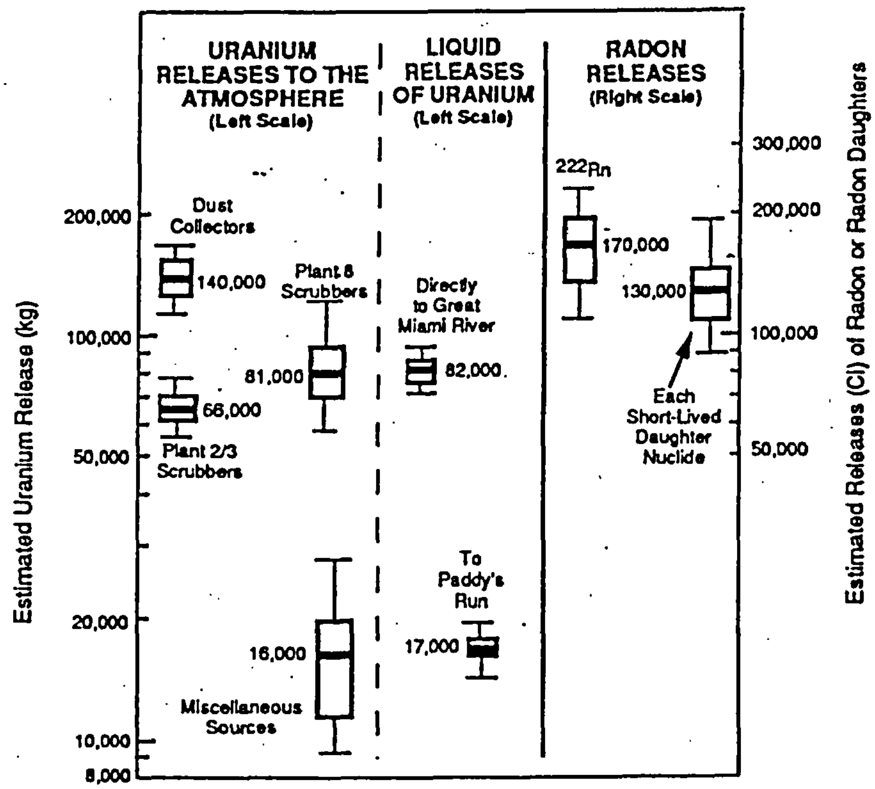

Figure 15. Summary of release estimates from the FMPC for the years 1951–1988. Releases are divided into three main sections which are separated by vertical lines. The center square represents the median or *best* estimate. The dark square on top represents the 95th percentile value, while the lower diamond represents the 5th percentile value. Ninety percent of the estimates lie within the range defined by top and bottom values that surround the best estimate.

Our work strongly supports the conclusion that atmospheric releases account for the greatest fraction of uranium released from the FMPC facility. Table 11 summarizes the grand medians and percentile values for the releases by decade for the three primary sources—the dust collectors, the Plant 8 scrubbers, and the Plant 2/3 scrubbers. The total releases estimate for 1951–1084 is a summary for all release points, including the unmonitored and accidental releases. The unmonitored releases are relatively minor compared to the three major sources, contributing only 16,000 kg uranium over the 47-year time span (Figure 15). Uranium releases to the atmosphere were highest in the 1950s with 175,000 kg uranium released from the three primary sources, and declined to almost half that in the 1960s. Total release estimates for the 1970s and 1980s are significantly less at 30,000 and 4,400 kg, respectively.

Table 11. Summary of Uranium Release Estimates for the Airborne Sources

Period	Best Estimate (kg U)[a]
1950s	175,000
1960s	90,000
1970s	30,000
1980s	4,400

[a] Releases by decade are releases from the dust collectors, the Plant 8 scrubbers and the Plant 2/3 denitration processes.

There have been several previous attempts at determination of uranium releases from the FMPC. Estimates of uranium discharged in liquid effluent were have been made by others on an annual basis (Boback 1971), or in summary reports evaluating the past discharge history of the facility (Rathgens 1974, Boback et al., 1985). These estimates of uranium to surface water from 1951 through 1984 range from 74,000 to 77,000 kg (Boback et al. 1987, Galper 1988) and fall within the uncertainty range of our estimates. Revisions to historic discharge reports generally focused on amending estimates of uranium loss to airborne effluents, and did not include updated figures for liquid effluents (Boback et al. 1985, Boback et al. 1987).

Previous reports of airborne uranium releases which have been used to estimate radiation doses in the offsite population around the FMPC have been reviewed for this project (Shleien 1991). Table 12 summarizes estimates of atmospheric releases of uranium which have been presented by others previously. These previous studies to determine the releases of radionuclides from the FMPC have yielded source terms which are less than our median or best estimates described in the present report. Our uncertainty ranges do not encompass these estimates except for that of the IEER. Exhaustive comparisons have not been made; however, reasons for our higher estimates include:

- the time to examine numerous documents, in particular original records, related to the FMPC operations;
- the use of a distribution of scrubber efficiencies for Plant 8 scrubbers;

- accounting for uranium losses from miscellaneous unmonitored sources and accidents;
- accounting for biases from sample line losses and other sampling deviations in the calculation of dust collector losses.

Table 12. Summary of Previous Atmospheric Uranium Release Estimates

Years (inclusive)	Uranium (kg)	Reference
1953–1984	96,000	Data for EPA estimate[a]
1951–1985	135,000	FMPC-2082 report[b]
1951–1987	179,000	Addendum to FMPC-2082 Report; IT report[c]
1951–1985	390,000	Reports prepared by Institute for Energy and Environmental Research for litigation involving the US DOE[d]

[a] From Kennedy 1985 and Meyers, no date; no specific documentation for estimate is provided.
[b] From Boback et al. 1985; report estimated airborne uranium releases from plant operations only.
[c] From Clark et al. 1989 and IT 1989; addendum also included uranium releases from Plant 2/3 scrubber operations, unmonitored releases and accidental releases. The IT report used the source term from the Addendum to the 2082 report.
[d] From Makhijani and Franke 1989; this estimate from their "alternative #2" calculations included additional scrubber losses from Plant 8 based on 70% efficiency for scrubbers instead of 85%.

Our methodology represents a significant improvement in the state-of-the-art of source terms analysis over previously reported data. It involves estimating a median, or best estimate of the releases in addition to a formal uncertainty analysis of parameters associated with these estimates. The Monte Carlo procedure uses our best estimates of the distributions of parameter values to produce a distribution of results. This process has resulted in obtaining a distribution of release estimates, instead of determining a single point estimate of the various parameters, with a single result. As a result, the source term has been characterized by a distribution of uncertainty for each year's releases.

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APPENDIX A

SOURCES OF INFORMATION FOR THE FERNALD DOSIMETRY RECONSTRUCTION PROJECT

A major effort in the Fernald Dosimetry Reconstruction Project has been searching for, and reviewing thousands of documents related to the operation of the Feed Materials Production Center (FMPC) since the facility opened in 1951. It has been our practice to trace the information back to original sources whenever possible. In the Task 1 report, issued in January 1991 (RAC 1991), we outlined the general approaches that we have taken to obtain this information. These five methods, which have formed the foundation for the project in providing the technical data for this study, include site visits to the FMPC facility; investigation of records and scientific literature pertaining to the FMPC; the retrieval and review of documents from NLO, Inc. using their computer database of document titles; examination of engineering diagrams, site blueprints, historic photographs and maps; and discussions with current and former longtime employees. The employees' recollections on plant processes, and procedures that routinely occurred since facility start-up served to identify sources and locations of documentation. Many of these individuals had been at the facility since the early fifties and sixties, and had served in various capacities, including maintenance, engineering, production, and plant management.

Because we realized the importance of retrieving documents from a wide range of sources, considerable time has been spent identifying types and locations of reports and records pertinent to the completion of this project. Generally, this documentation of FMPC operations and releases comes from two broad areas: (a) those produced by National Lead Company of Ohio, Inc. (NLCO), the former operator of the site, the Westinghouse Materials Company of Ohio (WMCO), the site operator from January 1, 1986 through 1992, and the Department of Energy (DOE); and (b) those issued by FMPC-independent sources.

The purpose of this appendix is to outline these sources of information and the types of documents that were found. For each source or location of documents, we have described the broad types and dates of documents, and have maintained detailed records of the reports and records that we have obtained at each location. In addition, *RAC* has maintained a detailed bibliography of all documents that we have gathered for the project. The documents in the *RAC* Document Repository are organized by topic and listed in Annex C of this appendix. All documents in the *RAC* Document Repository have been kept at a single location throughout the active phase of this project, but will be transferred to CDC at the conclusion of the project. Table A-1 lists the general sources of documents, dates, and comments. Each category or location is described briefly.

FMPC SITE

Many official monthly and annual FMPC reports, analytical data sheets, records, logbooks, and personal notes and diaries for most years of operation still exist. These various types of records at the FMPC site are found in the main records storage area

(Central Files), the Library, and individual plants or buildings. The great majority of documents in the Central Files area have been tabulated in the FMPC Records Storage Inventory list of documents. Although individual documents are not listed, most records are grouped together by department, individual's personal records, plant processes or building location. The documents on the list are distributed among four locations: in the Central Files vault at the FMPC site, at the Federal Storage Center in Dayton, in local storage in Cincinnati, or in the process area (Plant 9 or Plant 4) where contaminated records are stored. There is an index card for each file folder of records which indicates its location, date, and box number holding the documents.

Records and documents in the Central Files and Vault at FMPC are listed in the WMCO FMPC Records Storage Inventory list. The list is organized by topic or document type under a particular department. The actual record related to each topic may be located in a single folder in a box (about 13" x 16"), or may require many boxes, each with records related to the main topic.

We used the WMCO FMPC Records Storage Inventory list to select documents of interest. In the following table, document topics or types are listed in the first column with the box number following. The topics are arranged by department similar to the FMPC Records Storage Inventory list from which we were working. Each topic or document type is listed by box number, if known, and outcome or status. The "not useful" comment indicates that the information was not helpful to us at the time. Other comments were added when available. All copied documents are part of the *RAC* document repository. Annex A of this appendix lists the types and status of documents that we reviewed from the Central Files storage area.

The Library is a source of logbooks kept by individuals or as a record for various processes or departments, and some classified documents. All documents are listed in a card catalogue, and stored in a secure vault there. The library has an index, and copies of all FMPC Quarterly and Topical Reports published since operations began. Although many logbooks and diaries are descriptive in nature, a few also provide quantitative data on operation times and duration, production amounts, or concentrations and volumes of materials released. Many of these have been reviewed and copied.

The card file of classified documents was examined. Many of the classified documents were compilations of abstracts of classified research that had been performed over the years by the AEC. During construction of a new building, the Library vault was closed temporarily. During that time, the plant logbooks were sent to Central Files, to storage offsite, or to 4 onsite if the logbooks were contaminated with low-level radioactivity. RAC has compiled a list of all FMPC logbooks that have been examined up to this time.

The repository for contaminated documents and logbooks is located on the third floor of Plant 4. Over the course of the project we spent ample time examining the documents that were sent there from other areas onsite, and noted records of interest. Annex B to this appendix lists the box numbers, dates and the types and status of the documents in the contaminated box repository at the FMPC.

Table A–1. Sources and Locations of Documentation for the Fernald Dosimetry Reconstruction Project

Source and Location of Documents	Dates of Documents	Comments
FMPC Site		
Central Files	All	WMCO provided list of all document categories. RAC checked hundreds of documents in dozens of boxes; a listing of all files/boxes examined has been kept.
Library	All	FMPC quarterly and topical reports, logbooks and diaries. Classified document card file examined; no significant content. Listing of logbooks examined here and in Plant 9.
Plant 4	All	Contaminated logbooks and documents in boxes from Central Files and Library.
Plant 6	All	Seven 4-drawer file cabinets with files from "metal" and "chemical" plants. Most files related to process testing and Test Authorizations since FMPC startup, their status, and final report, if done. List is available.
FMPC Public Affairs Reading Room	1980s and 1990s	Hundreds of documents on environmental procedures, investigation reports, safety, hazardous materials and waste, and general information about FMPC.
National Lead Company of Ohio, Inc., Cincinnati.	All	Over 200,000 documents gathered by NLO, Inc. for litigation purposes. Using a computer database file of document titles, RAC has requested and received hundreds of documents from this source.
DOE Oak Ridge Operations (ORO) and Office of Scientific and Technical Information (OSTI), Oak Ridge, TN.	1970s and 1980s	DOE Oak Ridge Records Retention Center has documents grouped by shipment number. Environmental Division records checked. Some classified documents reviewed; requested declassification. OSTI has a computer listing of all FMPC-related documents.
National Archives and Record Center, Atlanta, GA.	1940s and 1950s	Listing of documents in two shipments from DOE/ORO reviewed; one shipment of 28 boxes from 1947 to 1954, the other shipment of 84 boxes from 1943 to 1964.
Ohio State Health Department, Columbus, OH.	Various	Found a few boxes of reports related to FMPC discharges; follow-up visit showed FMPC information is not easy to locate.

(Continued on next page)

Table A–1. Sources and Locations of Documentation for the Fernald Dosimetry Reconstruction Project (cont'd)

Source and Location of Documents	Dates of Documents	Comments
U.S. Geological Survey (USGS)	1951–1985	Reports in early 1950s, 1962, 1968, and 1985 on groundwater movement and stormwater collection onsite.
Open scientific literature	1958–present	Database systems used to search for FMPC-related reports include HP QUEST, GRATEFUL MED, and Toxline.
FMPC Area Residents; FRESH	Various	RAC and CDC have asked for leads in finding sources of documents.
Offices of Waite, Schneider, Bayless & Chesley, Cincinnati, OH	All	Documents related to FMPC discharge history; generally the same as those at the NLO, Inc. offices.
GAP (Government Accountability Project), Washington, D.C.	1954–1986	Non-profit organization representing FMPC workers' interests. Have 5 boxes of documents related to FMPC.
IEER (Institute for Energy and Environmental Research, Tacoma Park, MD	All	Performed release and dose estimates for Waite, Schneider, Bayless & Chesley; have documents related to FMPC operations.
Universities	1960–1990	Reports by professors from University of Cincinnati, Miami University, Colorado State University, and University of Rochester.
Private Companies	1970–1990	PEDCo Environmental, Cincinnati; EG&G; L. Lehman & Associates, Inc.; Roy F. Weston, Inc., West Chester, PA and others have studied and reported on the FMPC site and vicinity.

NLO, INC.

Over 200,000 documents pertaining to the FMPC operations were gathered by NLO, Inc. for litigation purposes. These documents are stored at the NLO office in Cincinnati, and each has been assigned a unique identifying inventory control number (ICN). For each of the 200,000 documents, NLO has listed the ICN, the title, the author(s), and the date in a computer database file. RAC received a computer database file of these documents through the Centers for Disease Control and Prevention (CDC) at the beginning of the project in

1990, and an update to the database in January 1993. We have used the database in several ways to identify and sort documents that pertain to the dose reconstruction project. Several thousand documents have been retrieved and reviewed for their relevance to the project. We have obtained copies of many of these documents for the RAC Document Repository.

OAK RIDGE: DOE ORO and OSTI

The Department of Energy Oak Ridge Operations Office (DOE/ORO) in Oak Ridge, Tennessee oversees the operations of the Fernald facility, and documents have been sent from FMPC to DOE/ORO over the years. The DOE/ORO Records Retention Center (RRC) lists boxes of documents by shipment number only so there is currently no timely or logical method for searching for FMPC-related documents. Nevertheless, inventory files in the RRC were reviewed by year and division in an attempt to locate Fernald related documents. Environmental Division records were studied more closely than others. Ultimately, documents from the DOE Records Retention Center in Oak Ridge are sent to the Federal Archives in Atlanta for permanent storage.

The Office of Scientific and Technical Information (OSTI) is the national center for scientific and technical information for DOE. OSTI encompasses not only DOE-originated information but also worldwide literature on scientific and technical energy-related matters, and maintains computerized energy-information databases that can be accessed through computer retrieval systems. At OSTI, a computer listing was available for all documents related to Fernald. These documents were reviewed relative to their usefulness to the project and important documents were copied.

FMPC-INDEPENDENT SOURCES

Locating independent sources of documents has been particularly important in verifying the data and records from the FMPC site. All avenues were explored to find pertinent monitoring data on environmental releases that may have been gathered by individuals or organizations not directly involved with FMPC operations. In the following discussion, examples of this work are cited and referenced. The listing is not comprehensive.

The Ohio State Health Department had some historical records and environmental monitoring data to substantiate information we had gathered previously. In addition, CDC has kept the Ohio State Health Department informed of our activities at FMPC for the Fernald Dosimetry Reconstruction Project.

Among the earliest independent studies were those conducted by the U.S. Geological Survey (USGS). Reports on ground water conditions in the Fernald area were prepared in the early fifties (Dove & Norris 1951) and sixties (Spieker & Norris 1962, Spieker 1968). The Ohio Division of Water has also performed hydrologic studies (Dove 1961).

Searches for publications related to the FMPC in the open scientific literature were performed using the bibliographic computer database systems, HP QUEST, GRATEFUL MED, and Toxline. The database HP QUEST includes publications devoted to radiation protection, while GRATEFUL MED is the National Medical Library search system. Various search criteria and keywords were employed to locate FMPC-related documents, specifically those from independent sources. A number of useful documents were found in this way.

Over the years of FMPC operation, professors at several universities around the country have completed a diversity of projects at FMPC. For example, in the early 1960s, Professor J. D. Eye, in the Department of Civil Engineering at the University of Cincinnati reviewed the potential of groundwater pollution at FMPC in several research reports (for example: Eye 1961a, 1961b). In 1985, T. B. Borak from Colorado State University studied the emission of radon from the K-65 silos (Borak 1985). Several private companies have also prepared various reports on the status of FMPC from the 1970s to the present. In 1976, 1977, and 1985, EG&G completed aerial radiological surveys of FMPC and surrounding areas (Feimster 1979; Shipman 1985). In 1988, L. Lehman & Associates, Inc. of Minneapolis reviewed literature pertaining to FMPC, and proposed a mechanism for groundwater contamination near FMPC (Lehman and Hansen 1988).

Efforts to find FMPC-related documentation have led to numerous meetings and phone conversations with knowledgeable individuals, such as Mr. Van Clay, the Assistant Attorney General for the State of Ohio, and Professor Roy Eckert of the University of Cincinnati. In addition, we talked with former employees and retirees from the FMPC. RAC has visited the attorneys in the office of Waite, Schneider, Bayless & Chesley in Cincinnati, who have assembled hundreds of documents for litigation related to the discharge history and past practices of FMPC. Although much of the documentation is similar to that retrieved from NLCO offices, some documents relevant to the dose reconstruction project were identified and copied.

We also visited the Government Accountability Project (GAP) office in Washington, DC on two occasions to review documents which they had obtained from FMPC personnel, and from DOE and WMCO through Freedom of Information Act requests. The GAP is a non-profit organization with the stated purpose of representing workers' interests. The five boxes of documents related to FMPC were checked for their application to this study.

Finally, we met with Arjun Makhijani of the Institute for Energy and Environmental Research, in Takoma Park, Maryland regarding studies on release estimates and radiation doses that they had completed in 1988 and 1989 (Franke 1988, Makhijani 1988, Makhijani and Franke 1989). They have a large number of documents similar to those found in offices of Waite, Schneider, Bayless & Chesley.

In summary, RAC has determined that there still exist a large number of reports, production records, and monitoring data related to FMPC operations. Although the record of operations is more complete for the seventies and eighties, a large number of analytical data sheets, monthly reports, letters, memoranda, photographs and drawings have been located for the fifties and sixties. Original logbooks have been useful; however, not all plant processes were documented in detail in logbooks.

RAC has gathered thousands of these documents for careful review in the preparation of the all project-related task reports. We are maintaining an ongoing list of the documents we have gathered in the *RAC* Document Repository. This documentation process will continue throughout the entire dose reconstruction project, with the final collection of *RAC* documents stored at CDC.

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ANNEX A TO APPENDIX A

TYPES OF DOCUMENTS IN THE CENTRAL FILE STORAGE AREA AT THE FEED MATERIAL PRODUCTION CENTER

TOPIC	BOX NO.	STATUS
ANALYTICAL		
Report of chem. analysis-Plant 1 (1/81-12/87)	40618	Not useful; lab data sheets with no report or sample key
Report of Isotopic analysis - all areas (1983)	41029	Not useful; raw data without report or sample key
EMERGENCY PREPAREDNESS		
Emergency. prepare. records and correspondence	45212	Not useful
	45103	Not useful
	46096	Not useful
ENGINEERING SERVICES		
Documents from REECO database; air, water emissions inf.; all have ICN #-can request through NLO; total of 12 boxes-looked all	46521 46523 46524	Some Copied at Central Files; some copied at NLO;
INDUSTRIAL HYGIENE		
K. Ross - Fallout, River, Grass/Soil, Air Boundary Sampling	44584	Useful, copy later
K. Ross - Stack Sampling Newsletter	44584	Not useful
Miami Valley Water Quality Committee Correspondence	A43540	Not useful
Incident Observation Reports	A23814	Not useful; daily log of technician.
Report of Fume Release (50-65) amounts given; Health/Safety narrative of accidents	A17936	Not useful; small accident reports - no
Plant Reports (1961)- occupational. exp. studies Plants 1-8	A17936	Not useful
Plant reports/ Radiation and effluent - Jan-Jun monthly reports; MH 175 & storm sewer	A17936	Copied
Major Incident Investigations (75-85)	34736	Copied

MAINTENANCE

Job Orders - 1985 ->	A45214	Copied
- 1988-89	A46758	Not useful;
PMS - Work Orders.	A46752	Not useful; canceled jobs.

MATERIALS CONTROL & ACCOUNTABILITY

Nuclear Materials Mngmnt Reports		FY62-FY87: very good mtrl rec'd, beg. & ending inven. by month & yr. 1961 available.
Physical Inventories/WIP		Inventories & mtrl. balance dif. (9/77-FY88) with details on losses & inventory; nuclear mtrl. production reports for 9/77-FY88.
SS Receiving Log A-Z (1965-75)		Examined
NMC Files - Bernie Gessiness	38577	Has file on Plt 8 loss back to 60's to 80s; efforts to control losses; maps of all manholes & connections.
Routine Operating Losses - By type discard (52-72)	A41492	Copied
Plant 2 refinery discard (53-77)	A41492	Copied; process information from 1964 to 74; invent./ship. inf. some early 60's & 70's, much 1977 information.
Routine Operating Losses (81-82)	A41492	Copied; records prior to 1965 destroyed but 1960-63 at NLO all copied
ROL's VVB (wet & dry pits)	A41492	Copied (52-72)
Discards by plants (64-77)	A41492	Copied
B-PIDS (Book-Physical Inventory differences - 7/61-7/62)	A41492	Tables I-V in front Copied; others copied
Write-off Correspondence	A41492	Copied Notes on Plt. 8 Trailer Cake
	A41492	Copied; discharge & losses from Plant 8 in 70's.

PLANT 1

SS Material Receiving Report		Destroyed prior to 65; (1965-75) Checked- type of mtrls, date rec'd, shipped, mailed, account #; Many handwritten.

NUCLEAR & SYSTEM SAFETY		
Enriched Material Incident		Checked; (4/61 - 8/64) enr. mtrl. incident reports beginning Apr 61
PRESIDENT'S OFFICE		
Historical Reports (1964-1985)		Checked list of docs-not useful
Production Supplies (1962 -84)		Offsite at BIS
PROPERTY ACCOUNTING		
409A-Special Reactor Materials		Checked: Inventory (12/59-6/62) cost of inven./no wt., no U costs; has Be, Y, some Ra.
PERFORMANCE ASSESSMENT		
Mike Boback's Misc. Files		Copied (1951-1985)
Historical Discharge report -notes and data re: solubility -Tom Borak reply to comments -Letter re: raincaps -PO for analyses of dust coll. -Inf. re: Rn meas. near K-65	47225	Copied
Historical discharge Report(F2) -Misc. re: 2082 report -UO_3 gulping operations	47225	Copied
Historical discharge Report (F-3) -Boback review of NLO dbase doc. -MC&A comments on 2082	47225	Copied
FMPC-2082 - Misc. material -Data on Th & Ra in feed mtrl -Particle size of U compounds -Memo Koch to Herman, 17 Apr 1985 particle size distribution - dust collector material	47225	Copied
FMPC-2082 Tables 13, 14, 88	47225	Copied
Northern Ky - dust collector efficiency	47225	Copied
Major Emission Stacks -Letter to Reafsnyder, 20 Sep 85 data compilation for historical dose estimates -Letter to Spenceley, 16 Jul 85, data for 15 major emission stacks -Letter to Reafsnyder, 16 Jul 85, partial data for major emission. stacks. -Data sheets of annual composites of boundary air dust samples 1982-84.	47225	Copied

Epidemiology Study -Self-absorp. fac./air fltr	47225	Copied
NLCO-1093-Re: graphite and oil incinerator	47225	Copied
DOE-ORO Sites Discharge Reports -Report on historic U releases from current DOE ORO facility., 24 Jun 85	47225	Copied

RADIOLOGICAL SAFETY

DOE Misc. Correspondence (6/63-7/79)	A43540	Copied
DOE Annual Reports 2/5/71 ->	A43540	Copied
Accounting: Loss of Material 2/8/71 -> 12/80	A43540	Copied
DOE Misc. - Soils 1970-71	A43540	Copied
Equipment & Mtrl. Pass July 81->	A43540	Not useful
Daily Monitoring Records (Vehicle) 8/2/82->	A43540	Not useful
Stack sampler flow rate and stack loss conversion	45539	Copied

RADIOLOGICAL SAFETY (DOSIMETRY AND INSTRUMENTATION)

Incident Investigation Reports	37188	Copied most

Analytical Data Sheets
(1954-68, 1970-86)
[There is a box for each year with air and water data: estimated stack losses for each plant, offsite air dust, misc. file, daily or weekly water samples for sewage plant, storm sewers, Manhole 175, Miami River at Venice & New Baltimore Bridges, fluoride pit, Paddy's Run water treatment. Data sheets grouped Jan-Jun, and Jul-Dec in separate folders for each location.]

Data sheets for 1961	0-000-535-291	Copied Paddy's Run, MH175, water treat; Fluoride pit, sewage plant; storm sewer, river copied at NLO
	0-000535-292	Carbon copies of Box 0-000-535-291
Data Sheets for 1960	0-000-535-290	MH 175, Paddy's Run Copied
	.0-000-535-290	Offsite air dust copied
	0-000-535-289	Carbon copies of Box 0-000-535-290

Box No. 17936
Industrial Hygiene Chronological File - Radiation and Effluent Control (1/58-6/58 only) Contains miscellaneous memos, letters, reports and monthly reports from the Radiation and Effluent Control section. Mostly not useful, but all sorts of things are in these files, occasionally something useful. Only had 1958 in this location.

Box No. 17936
Chronological File - Engineering and Special Problems Section (1/58-6/58 and 7/58-12/58)
See above comment.

Box No. 17936
Chronological File - Survey Section (1/58-6/58 and 7/58-12/58)
See above comment.

Box No. 17936
Report of Fume Release
Previously reviewed by others, nothing useful found.

Box No. 17936
Investigation of Injury
Nothing useful.

Box No. 44584
EPA Method 5 Stack Sampling - Power Plant and Kelley Waste Incinerator - K. Ross
Nothing useful on the incinerator.

Box No. 44584
Files containing rainfall sampling data - K. Ross
Rainfall measurements and analysis results for radioactivity in the rainfall, for mid-1960s? Copied some typical documents for Kathleen Meyer.

Box No. 37188
Drawings of PERMs
Not useful.

Box No. 37188
River sample summaries
Not useful.

Box No. 37188
Technical Lab and Lab Machine Shop 1954-1969
Surveys, miscellaneous. Not useful.

Box No. 37188
Miscellaneous Surveys 1964-1968
Not useful.

Box No. 37188
Plant 2/3
Surveys, miscellaneous. Not useful.

Box No. 37188
Pilot Plant
Surveys, miscellaneous. Copied 2.

Box No. 37188

Radiation Exposure Investigations 1964-1965
Not useful.
Plant 9

Box No. 37188
Surveys, miscellaneous. Not useful.

Box No. 37188
Plant 6
Survey results and miscellaneous. Copied one document regarding samples taken during a chip fire.

Box No. 37188
Incident Investigations 1959-1969

Box No. 37188
Services for Offsite Work to 1961
Survey results and other reports. Not useful.

Box No. 37188
Knoxville Iron Company
Reports about work and surveys for Knoxville Iron. Not useful.

Box No. 37188
Plant 8
Contains some documents about the Plant 8 scrubbers. None were copied; believe we already have copies of same documents.

Box No. 37188
AEC Audits
Not useful.

Box No. 37188
Plant 4 1962-1969
Various survey activities for Plant 4, copied one document.

Box No. 37188
Plant 5 Air Dust Surveys

Box No. 37188
Disposals to Waste Pit 1964-1968
Contains information about material sent to the burning pit. None copied at this time.

Box No. 37188
Three reports of exposure studies in Plants 5 and 8 for 1967 and 1968
Not useful.

Box No. 21936
IH & R Monthly Reports 1963

These are potentially useful. Monthly progress reports for Survey section, Engineering and Special Problems section, Radiation and Effluent Control section, and for IH & R department. All available copied.

Box No. 21936
Stack Loss Reports 1963
Set of stack loss reports for 1963, except does not include December. All copied.

Box No. 21936
Daily Monitoring Records
Completed forms for radiation surveys of shipments.

Box No. 21936
Inspection and Service Reports 1963
Records of inspections of radiation detection alarms (RDAs). Not useful.

Box No. 21936
Radiation Monitoring Records 1963
Records of surveys of equipment, presumably before release. Not useful.

Box No. 21936
Equipment and Material Passes
Tags to apply to equipment to show survey results and recommendations. Not useful.

Box No. 21936
Miscellaneous Correspondence 1957, 1958, 1959, 1961, 1962, and 1963
Various memos, letters, and reports. Found useful K-65 silos survey with Rn concentration measurements.

Box No. 23814
Trip Reports for 1964 and 1965
Not useful.

Box No. 23814
Reports of Fume Releases - NLO-H&S-1538 - 1964 and 1965
Records of investigations of fume releases relative to worker protection - mostly chemical, some radioactivity releases. Not useful.

Box No. 23814
Reports of Injury and Ambulance Service 1964 and 1965
Not useful.

Box No. 23814
Report of Plant Fires

Box No. 23814
Equipment and Material Passes
Not useful.

Box No. 23814
Inspection and Service Reports 1964 and 1965

Records of inspections of RDAs and nuclear accident dosimeters. Not useful.

Box No. 23814
Correspondence on Fume and Dust Control Committee 1962 through 1964.
Not useful.

Box No. 23814
Monthly Reports for 1964
Reports for the three sections in IH & R. Copied

Box No. 23814
Daily Monitoring Records 1964
See earlier comment. Not useful.

Box No. 23814
Estimated Stack Loss Reports 1964

Box No. 23814
Notice of Contamination Source 1959 through 1963
Not useful.

Box No. 23814
Radiation Monitoring Records 1964 and 1965
See earlier comment.

Box No. 23814
Miscellaneous Correspondence 1962, 1963, 1964, 1965
Could be useful. Memos, letters, reports from IH & R department. Various pieces were copied.

Box No. 44583
RDA Test Evaluation
This file relates to test evacuations. Not useful.

Box No. 43207
Radiological Safety (Dosimetry and Instrumentation)
Historical Radiation Reports 1953-1983
Compilation of radiation exposures to personnel. Not useful.

Box No. 46404
Beta and Gamma Exposure Readings 1959
Film badge records for 1960 and part of 1959. Not useful.

Box No. 37192,... etc.
Radiological Safety
Radiation Monitoring Record 1961-1962. Contains routine survey records: Daily Monitoring Record, Radiation Monitoring Records (equipment after decon), Reports of Fume Release, Inspection and Service Reports (RDAs), and IH & R form 492 (equipment for disposal). There are other boxes (not reviewed) with similar files, for various years in the 1960s, 1970s, and 1980s. Not useful.

Box No. 45539
Miscellaneous monitoring files for: ERMT Class, Contamination Surveys, Environmental Rad. Man. Qual. Check, Plant 5, K-65, Tank Farm, D & D Facility, Stack Sampler Flow Rates & Stack Loss, Stack - Jan. and March, Stack - April and June, Report of Chemical Analysis, SRPD Logs, Stack Results 1986, Smears, K-65 Paddy's Run.
Mostly not useful. The K-65 Paddy's Run files contain records of external gamma radiation surveys performed along Paddy's Run Road, at points closest to the K-65 silos. These may be useful for calculating gamma doses due to the silos. Copied a representative sample of these files.

Box No. 45539
Sample Result Correspondence 1986
This file not useful. Similar files exist for earlier years - from 1956, which could have useful information.

Box No. 46573
Miscellaneous routine survey and other routine records for: K-65 Area, Lab HFM Survey, Laundry, Locker Room, Maint #107, Medical Emergency, Men's Locker Room, N.A.D. Inspection, Outgoing Vehicles, Paddy's Run Road, P.P. Office, Radon/Thoron Samples, R.S. Trailer, Respirator Trailer, RIMIA, Rust Building 3045, Radiation Work Permit, RDA, Personnel Contamination, RGM #2 Operational Checks, and Radiation Monitoring Record.
Mostly not useful. There was additional data from the Paddy's Run Road gamma survey program, which could be useful. None copied.

ANNEX B TO APPENDIX A

BOXES OF CONTAMINATED DOCUMENTS FROM CENTRAL FILES AND LIBRARY AT THE FMPC EXAMINED BY *RAC*

Boxes of Contaminated Documents from Central Files and the Library at the FMPC

Box No.	Plant	File	Date	Status
A 36715		Fire and safety work permits	no date	Rev. by F. Rogers, CDC, 6/2/94; no interest
A44099		Chemical analysis reports	1984-85	Reviewed by F.Rogers, CDC, 6/2/94
B 39374	Plant 8	Work Records-Plt. 8	Jul 56 - Sep 57	No- list of personnel and job assignments
		Foreman's Log-Chip Furnace-Plt. 8	Feb - Oct 1953	
		Area Foreman's Notes to Foremen-Plt. 8	Aug 53 - May 56	Copied Feb, Mar,Jun, Jul, Aug 53 (oper. summary); 30 steno notebooks plus folders with notes; some reference to scrubbers
39408	Prod\Plt 8	Work Record-Plt. 8 NLO-PRO-1868		No copies- personnel and job assignments
		Daily dust col. check.-Plt. 8 NLO-PRO-1868	May 68-Sep 68	Copied few; dc bag configuration for ea plant; notes on operation or bag failure
			Feb 67- Jul 67	
		Work Record - Project labor Pool-H&S-1015	Feb 62 - Dec 62	No- list of personnel and job assignments
			Dec 67- May 68	
		Work Record NLO-PER 1567-Laundry	1963	No- list of personnel job assignments
39375	Plant 8	Foreman's handwritten notes-Wet Area-Plt. 8	Jan 57-Nov 57	No copies; 1953 logs; requires time and effort to read all notes.
	3	Foreman's handwritten notes-Dry Area-Plt. 8	May 56 - Oct 57	
		Area Foreman's notes to shift foremen -Plt. 8	May 56- Sep 57	
39379	Plant 8	Work Record-H&S 1015-Plt. 8	Oct 57 - Jun 59	No copies- list of personnel and job assignments
		Handwritten shift foreman's log/notes-Plt. 8	Oct 57-Dec 58	
39403	Plt. 5 ,6, 8	Work Record-Plt.8-H&S-1015	Sep - Dec 60	No
		Work Record-Plt 9-H&S	Jan - Dec 60	No
		Foreman's Log Sheet-Plt. 9	May 59 - Jan 60	
		Daily dust collector check sheets-Plt.9	Oct-Dec 59	No; separate form with dc and bag configuration for each plant; notes on operation or bag failure
		Daily dust collector check sheets-Plt.8	Nov 57-Dec 58	" "
		Daily dust collector check sheets-Plt.8	Oct 55 - May 56	" "

Box No.	Plant	File	Date	Status
39403	Plt. 5 ,6, 8	Daily dust collector check sheets-Plt.5	Jan - Dec 62	" "
	Plt. 7	Plant 7 notebooks: Vaporizer; Refrigeration;	Dec 55- May 56	No
		Plant 7 notebooks: Leaderman's Log	Mar - May 56	Copied; small binder with norm & depl quantities on shift by shift basis; may be useful for Plt 7 production.
A 45875	Pilot /Plt. 7	Misc. records & correspondence	1951 - 1968	Copied: loss of materials; dc bags & filter types; enr. UF4 prod. Aug56-Apr 57; dc loading & efficiency tests; fume release in 1955.
39342	Plant 2&3	Leaderman's Log-Extraction Area-Ore Ref.	Jan-Jun57	No
		Shift Foreman's Log -Plt 3-Recovery	Jan-Jun 57	No
		Shift Foreman's Log-Ore Ref.- Denitration	Jan- Jun 57	Copied-"gulping" operations-3 shifts/day
		Operator's Shift Log-Ore Ref.-Denitration	Jul-Dec 57	Copied-"gulping" operations-3 shifts/day
		Work Record & Shift Foreman's Log-PRO-664	Jul-Dec 57	No
		Shift Log-Denitration Area	Aug-Oct 57	Copied-"gulping" operations log -3 shifts/day
		Shift Foreman's Log Recovery Area-Ore Ref.	Nov, Dec 57	No
		Denitration Operators Shift Log	Jan-Dec 57	Copied Apr to Sep -"gulping" operations
39346	Plant 2&3	Shift Foreman's Log-Denit. Operators Log	Jun 58-Mar 59	Copied-"gulping" operations-3 shifts/day
		"On-stream Factor Report"	Jun 58-Mar 59	Copied-summary of all operational logs: shift foreman's extraction, dinitration, sump technician's log, nitric acid recovery
39345	Plant 2&3	Sump Technician's Log-PRO-1039-Ore Ref.	Jan-Dec 58	Copied-Daily discharges to general sump &MH 175
		Shift Foreman's Log-Denit. & Recovery Area	Jul-Nov 58	Copied-"gulping" operations- 3 shifts per day
		Operators Shift Log-Denit. Area-PRO-1175	Jul-Dec 58	Copied-"gulping" operations log-3 shifts/day
A48689		Hydrolysis UNO3 test log 903 1955-56; U techniques	1955-61	No; reviewed by F. Rogers, CDC, 6/2/94
A 47394		Chemical analysis cards, water	1989	No; reviewed by F. Rogers, CDC, 6/2/94
A48700	Notebooks	Log book 2853, gulping amounts 7 day/shift 1975-76	1975-76	Reviewed by F. Rogers, CDC, 6/2/94
		Log book 2855; gulping oper. 1975-76	1975-76	Reviewed by Felix Rogers, 6/2/94

Box No.	Plant	File	Date	Status
A48696	Notebooks: handwritten from library	625: Metallurgy Dept.; cost comparison PP & Plt. 4 products; 30x and 500 x photos.	1954 - 55	
		627: Solvent extrac U nitrate;purification of thorium in cellulose column.	1953 - 55	
		692: Development of moving bed reac UF4	Oct-Nov 54	
		699: U metal quality; photos of slugs 100x	1954 - 59	
		700: Pilot Plant - Furnace operation	1954	No; nothing on releases
		705: Plant 4, UO3 processing	1954 - 55	
		708: Salt heat treating of U; diff. therm. anal	1954 - 57	
		3087: Pilot Plt operations & procedures	1978	No; nothing on releases
		3088: Pilot Plt operations & procedures	1978 - 79	No; nothing on releases
A48696	Notebooks: handwritten from library	3100: Refinery; boil down operations	1981 - 82	No copies
		3102: Supervisor's instructions, refinery	1981 - 85	No copies
		3099: Ray Bauer's notebook; mold specs.	1978 - 84	No copies
		3105: Pilot Plt-procedures and operations	1979	No; nothing on releases
		3108: Pilot Plt-procedures and operations	1979	No; nothing on releases
		3109: Pilot Plt-procedures and operations	1979	No; nothing on releases
		3113: Pilot Plt-procedures and operations	1979	No; nothing on releases
		3118: Pilot Plt-procedures and operations	1979	No; nothing on releases
		3117: Plant 1-operations & daily log	1979 - 80	No copies
		3119: Plant 4	1979	No; nothing on releases
		3120: Plant 4-operations & procedures	1979-80	No; nothing on releases
		3105: Pilot Plt-operations	Jan-Apr 79	No; nothing on releases
		3108: Pilot Plt-shift & activities log	Apr-79	No; nothing on releases
		3109: Pilot Plt-shift & activities log	May-79	No; nothing on releases
		3118: Plant 4-shift log	Nov-79	No; nothing on releases
A42673	Plant 6	Environmental Safety & Health-MC&A daily production reports-Plant 6	1974 - 84	No copies
B39213		Daily time sheets	19598	No; reviewed by F. Rogers, CDC, 6/2/94
54200		Purchase Orders	1985	No; reviewed by F. Rogers, CDC, 6/2/94
39219	Metals area	Shift Foreman's Daily Log Sheets-Plt. 9- 2nd and 3rd shifts	Oct - Dec 58 Apr 59-Aug 60	No copies
		Foreman's Daily Log-PRO-909-Plt. 6	Jun 59-May 60	

Box No.	Plant	File	Date	Status
39219	Metals area	Daily time sheets-PRO 909-Plt. 9	Apr 59-Jun 60	
B 39341	Plant 2/3	Foreman's Log-Ore Refinery		
		Foreman's Log-Denit /Acid Recovery	Jan-Dec 56	Detailed shift logs for "gulping" operations;
		Digestion/Extraction Logs	1956	No
A48688	2543	Metallurgical Dept. handwritten lab books	1968-70	
	2464	"	1968-69	
	2525	"	1968-69	
	433	Maurice Atwell	1953-1957	
	2427	Metallurgical Dept. handwritten lab books	1967-68	
	2455	"	1967-70	
	2485	"	1967-69	
	2364	"	1966-68	
	2322	"	1965	
	2249	"	1965-66	
	2246	"	1965-66	
	2250	"	1965-67	
	2254	"	1965-69	
	2253	"	1966-68	
	2266	Handwritten carbide tool development	1966-67	
	2333	Metallurgical Dept. handwritten lab books	1965-78	
	2271	Metallurgical Dept. handwritten lab books	1964-77	
	2284	Project log- extraction	1964-66	
	2499	R.C. Kispert	1967	
	2626		1968-69	
	2344	Plant 2/3 denitration log (# pots gulped)	May-Sep 1973	copied
	2279	C.W. Huntington	1964-66	
	2284	Thorium extraction test log-Kispert	1964-66	
	2333	SS Material weight log	1965-78	
	2271	SF materials shipping log	1964-77	
B39240	Plant 2/3	Foreman's daily log sheets	Jan 59-Sep 60	copied
A48691	1936	General sump log for 1963	1963	copied
	3335	Operators' shift log for Plt 8; feed/prod. wt.	3/83 to 9/83	Furnace oper.; drums to furnace; clean out

Box No.	Plant	File	Date	Status
A48691	3345	Plant 8 shift log; furnace operation	6/83 to 8/83	Operator's log kiln; R.L. Gardner
	3337	R.L. Gardner; Plt 8; operators log	4/83 to 6/83	"
	3350	Opeators' shift log; Plant 8	4/83-6/83	"
	3351	P.A. Shanks-Chemical production tech.		"
A44040	Plant 8	Misc. chemical analysis;notebooks		
		UO3 production for Paducah	Mar73-May77	
		Lot marking system	1968-70	
		Sample log summary	1975-77	
		Refinery and Plt 8 analysis	May-Nov'75	Box furnace product-Plant 8
		Plant 8 analysis; trailer cake	1971	
		UO3 analysis	1975	
		Plant 8 oxidation furnace product	May-Nov'75	
		Plant 8 rotary kiln product	"	
		Analysis of refinery feeds	"	
A45875		Misc. receipts, correspondance	1951-1968	Reviewed by F. Rogers, CDC, 6/2/94
A39343		Foreman's logs	1958	Reviewed by F. Rogers, CDC, 6/2/94
A427574		Disouted weight cards	no date	Reviewed by F. Rogers, CDC, 6/2/94
A48697	918	Lab notebook; W.E. Palmer	1955-57	
	928	Logbook	1955	
	934	Logbook	1955-56	
	942	Logbook	1955-56	
	944	Logbook	1955-56	
	956	Logbook	1955-57	
	970	Logbook	1955-57	
	973	Ray Bauer's chemical lab notebook	1955-57	
	974	Logbook	1955-57	
	979	Logbook	1956	
	2087	Logbook	1963	
	2901	Logbook		
	2903	Logbook	1973-74	
	2904	Logbook	1973-74	
	2921	Logbook	1973-74	
	2922	Calciner shift log	1974	

Box No.	Plant	File	Date	Status
	2927	Calciner shift log	1975	
	2928	Plant 9 test logbook	1974-75	
	2939		1974-78	
	2979		1975	
	2978		1975-78	
B39226	Plts 5,6,9	Foreman's log sheets	Jan-Jun 1962	
		Daily time sheets	Jan 63-Jun83	
B39376	Plant 8	Shift foreman's log notes and log	Oct 57-Dec 58	
		Work record; H&S;	Oct 57-Jun 59	
B39351	Plt 2/3	Work record	Jan 57-Dec 68	
B39351		Work record; shift foreman's log-digestion & extraction	1967	Some copied
		Operators' shift log all areas-ore refinery	1967	Copied
		Foreman's instructions log-ore refinery	May 67-May68	
B39344	Plant 2/3	Work record; shift foreman's log-digestion & extraction	Jan 58-Dec 59	
		Operators' shift log all areas-ore refinery	Jan 57-Mar 60	Copied
		Area foremens' notes; cold weather shutdown	Nov 60-Dec 60	
B39340	Plant 2/3	Foreman's log-ore refinery, digestion, extraction areas	Dec 53-Dec 57	Some copied
B39351				
A44584	ES&H	Stack sampling newsletter	1974-78	
		Exhaust ventilation surveys	1965-67	
		Fallout, river, grass/soil, air sampling	1960-1969	Keith Ross' box; all copied
		Correspondance of Keith Ross files	1980-85	Copied
		River water survey at 15 locations	1960s	copied
B39217	Plt. 6	Foremans' daily log sheets; mechanical	Jun 58-Feb 59	
		dept. Daily time sheets	Jan - Dec 1959	
B39343	Plt. 2/3	Work record; shift foremans' log for refinery, extraction	Jan-Dec 1957	Copied
		Work record; shift foremans' log for refinery, extraction	Dec 56-May 57	Copied
		Work record; shift foremans' log	Jan-Jun 1958	Copied

Box No.	Plant	File	Date	Status
		for refinery, denitration		
68000		Purchase orders	1985	No; reviewed by F.Rogers, CDC, 6/2/94
48693		Misc. lab notebooks-research	1984	Reviewed by F. Rogers, CDC, 6/2/94
84400		Purchase orders	1987	Reviewed by F. Rogers, CDC, 6/2/94
39378		Time cards, assignment sheets	1955-60	Reviewed by F. Rogers, CDC, 6/2/94
Boxes outside of Storage room on 3rd floor of Bldg. 4				
40 boxes from Pilot Plant; routine operations				
1	Pilot	Misc. records; 1987 investigation rpt; of HF release on Sept 29, 1987; study environs of P P July 3, 1987	1987	
2		Foremans' office; minor events; vessel inspections	1986-87	
3		Production files		
32 boxes from Plant 2/3; unnumbered;some indications of content written on top				
	Plt 2/3	Area clock cards	1987	
		Pot control records	1987	
		Job orders	1987	
		Vacatin records		
		Daily records	1984-87	
		Refinery/extraction records; slag leach data sheets; production	1984-87	
		consumption worksheets. Safety meeting	1981-1983	
		minutes. plant test authorizations, nucelar material custodian activities	1981-1983	

ANNEX C TO APPENDIX A

RAC DOCUMENT COLLECTION FOR THE FERNALD DOSIMETRY RECONSTRUCTION PROJECT

The following list identifies the documents that we have located and reviewed for the Fernald Dosimetry Reconstruction Project. All documents in the *RAC* Repository are grouped by topic and kept at a single location. Additional materials are added regularly to the collection as the project proceeds.

TABLE OF CONTENTS

AERIAL SURVEYS / PHOTOGRAPHS / MAPS

Feimster, E. L., June 1979. An Aerial Radiological Survey of the Area Surrounding The Feed Materials Production Center, Fernald, Ohio, EGG-1183-1680. EG&G Energy Measurements Group. Date of survey: August 1976/May-June 1977.

FMPC Series of Aerial Photographs of FMPC Site and Process Buildings in 1954, 1960, 1965, 1987 - 1990.

Ross, K. Late 1950. Plan of General Area Property, Incinerator. Drawing No. 8-4001. (24 x 36 inch map that PGV copied in sections from K. Ross files in Central files. Includes sampling locations for gumpaper, fallout, soil and grass in late 50s and early 1960s). National Lead Company of Ohio.

Shipman, G. R., October 1985. An Aerial Radiological Survey of the Feed Materials Production Center and Surrounding Area, Fernald, Ohio, EGG-10282-1084. EG&G Energy Measurements. Date of Survey, April 1985.

Stern, R. J. Comments on Aerial Radiation Survey of the Feed Materials Production Center, Fernald, Ohio.. Report to W.A. Vaughn regarding the EG&G aerial surv ey of the FMPC. Department of Energy, Environment, Safety and Health. PE-222. ICN 2144793.

AIR SAMPLING/BIOASSAY/GUMFILM

Bipes, R.L. Ventilation Survey of the Paint Spray Booth–Mechanical Shop. Memorandum to D.E. Diehl. Cincinnati, OH: National Lead Company of Ohio. 19 October 1962.

Blase, E.F. Air dust samples in wet side of pilot plant., 3013. Memorandum to R.C. Heatherton. Cincinnati, OH: National Lead Company of Ohio. 22 April 1952.

Blase, E.F. Air dust survey - reversing mill. Memorandum to R.C. Heatherton. Cincinnati, OH: National Lead Company of Ohio. 26 November 1952.

Boback, M.W. and R.C. Heatherton. 28 September 1964. Recent Bioassay Activities at National Lead Company of Ohio. NLCO-933. (Summarizes recent nonroutine problems for bioassay department including use of infrared heating, U in lung and lymph node tissue, U slivers in hand, radium isotopes from remelt operation, U in fecal samples). Prepared for presentation at the Tenth Annual Bio-assay and Analytical Chemistry Conference, Cincinnati, Ohio. October 8-9, 1964. National Lead Company of Ohio. Health & Safety Routine Reports

Boback, M.W. Procedure for Treatment and Analysis of Gumpapers. Letter to Felix Rogers. Memo No: 94-026. Cincinnati, OH: Fernald Environmental Managment Project. 18 May 1994.

Dodd, A.O. Monthly Progress Reports. Memorandum to R.H. Starkey. Have 1958: March, April, May, June, July, August, September, Oct. (Report film badge exposure data, special external radiation investigations, ground contamination surveys, plant liquid effluent, outplant air dust and fallout).

Dodd A.O. Annual Report - Radiation and Effluent Control Section. Memorandum to R.H. Starkey. (2 pages, personnel monitoring data, effluent control, air dust and fallout studies, enriched materials movement coordination, residues surveillance; nothing quantitative). Cincinnati, OH: National Lead Company of Ohio. 12 February 1958.

Heatherton, R.C., M.W. Boback. and J.A. Quigley. A Continued Program of Analysis for Uranium in Human and Animal tissues. NLCO-895. (Comparisons are made of uranium concentrations in tissues of 16 exposed and unexposed persons). Prepared for presentation at the Ninth Annual Bioassay and Analytical Chemistry Conference, San Diego, California October 10-11, 1963. National Lead Company of Ohio. 20 September 1963.

Held, B.J. and E. Chenault. Perimeter Air dust survey. Memorandum to R.C. Heatherton. (Results of survey on October 30 and 31, 1956 and December 27, 1956 around perimeter. Cincinnati, OH: National Lead Company of Ohio. 8 November 1956.

Kessler, L. W. Air Contamination In Plant 8, Project No. P-23000-15, Short Order Completion Report for Production Engineering Department, NLO/ICN 2225348. (Equipment contributing to high airborne contamination in Plant 8 identified and action taken outlined; future activity reported under Project P-20000-22). 17 February 1959.

Klein F.J. Memorandum to R.L. Fischoff. Uranium Fallout Study in Adjacent Vicinity of the Oil Burner and The Incinerator. (Lists concentration range and averages at five locations). NLO/ICN 2118894. Cincinnati, OH: National Lead Company of Ohio. 1 May 1964.

Ross, K.N. and F.J. Klein. Monthly FMPC fallout data using variety of techniques and Abbe Observatory rainwater data for 1965. Letter to C.E. Schumann (City of Cincinnati). Cincinnati, OH: National Lead Company of Ohio. 8 February 1966.

Starkey, R. H. Compilation of High Air Dust Exposure Operations. Report to J. A. Quigley, P. G. DeFazio & C. R. Chapman. Cincinnati, OH: National Lead Company of Ohio. (7 pages, compilation of air dust exposure operations for all plants in excess of 3 x MAC, and status of corrective action). 20 October 1960.

Starkey, R. H. Compilation of High Air Dust Exposure Operations. Report to J. A. Quigley, P. G. DeFazio, C. R. Chapman and S. Marshall. (9 pages, compilation of air dust exposure operations for all plants in excess of 3 x MAC, and status of corrective action as of 12/13/62). Cincinnati, OH: National Lead Company of Ohio. 2 January 1963.

Starkey, R. H. Compilation of High Air Dust Exposure Operations. Report to J. A. Quigley, P. G. DeFazio, C. R. Chapman and S. Marshall. (8 pages, compilation of air dust exposure operations for all plants in excess of 3 x MAC, and status of corrective action as of 4/18/63). Cincinnati, OH: National Lead Company of Ohio. 24 May 1963.

Starkey, R.L. Correlation between two-stage air sampling data and the excretion of uranium in urine. For presentation at the American Industrial Hygiene Conference, 9 May 1963, and the health Physics Society meeting 11 June 1963. Cincinnati, OH: National Lead Company of Ohio. NLCO-869. 1963.

Twitty, B.L. and M.W. Boback. 1970. Rapid Determination of Thorium in Urine by Thermal Neutron Activation Analysis. Analytical Chimica Act20, 49: 19-24.

Wing, J. F., K. N. Ross and R. G. Wissman, Exposure Study of Technical Laboratory personnel to Radioactive Airborne Dust. (Brief summary and discussion of existing air dust levels at Technical lab. Bldg, 10 pages). 30 November 1961.

Wing, J. F. to H. M. Beers. Air Dust Samples From Outside Mill, Plant 8, NLO/ICN 2225349. (Results grossly in excess of MAC), 1 August 1958.

Wing, J.F. Simultaneous Air Samples and Face Velocity Measurements. Memorandum to E.D. Leininger, R.G. Wissman and R.L. Ruhe. Cincinnati, OH: National Lead Company of Ohio. 20 January 1965.

ANALYTICAL DATA SHEETS / CHEMICAL ANALYSIS

Boback, M.W. 22 March 1967. Neptunium in NFS Uranium. Memorandum to J.A. Quigley. (Analyses for Np in NFS Dresden material, shipment 24 and 30). National Lead Company of Ohio.

Boback, M.W. 17 May 1967. Neptunium in Plant Materials. Memorandum to J.A. Quigley. (4 tables contain results of Np in NFS Dresden uranyl nitrate; attempt to follow particular batch of Np-containing uranium through plant processes). National Lead Company of Ohio.

Gustavson, S.R. NLO-NBL Assay Comparisons on NLO Inventory Samples. Letter to C.H. Weldon. (Tabulation of U assay results on various process samples incl. scrubber acid, digestor liquor, Q-11). US Atomic Energy Commission, New York Operations Office. 8 February 1954.

Air-Inplant Sampling

NLO (National Lead Company of Ohio). Analytical data sheet of alpha in air dust and smear samples taken in Laboratory Building collected 1/9, 1/12, 1/13, 1/14, 1/16, 1/19, 2/5, 3/4, 4/5, 3/9, 3/10, 3/11, 3/18, 3/24, 3/26, 3/31, 4/2, 4/3, 4/6, 4/14, 4/17, 4/20, 4/27, 4/28, 4/29, 5/1, 5/4, 5/6, 5/7, 5/22, 5/26, 5/27, 6/9, 6/10, 6/15, 6/17, 6/24, 7/1, 7/13, 7/14, 7/16, 7/17, 7/21, 8/2, 8/7, 8/15, 8/17, 8/20, 9/2, 9/11, 10/12, 10/19, 10/26, 11/7, 11/9, 12/15, 12/16/53. From FERMCO Box 535-279. Cincinnati, OH: National Lead Company of Ohio. 1953.

NLO (National Lead Company of Ohio). Contamination survey sheet of various rooms in Laboratory Building collected 8/2, 8/7, 8/10, 8/19, 8/27, 9/4, 9/14, 10/19, 10/26/53. From FERMCO Box 535-279. Cincinnati, OH: National Lead Company of Ohio. 1953.

NLO (National Lead Company of Ohio). Analytical data sheet of alpha in air dust and smear samples taken in Laboratory Building collected 1/11, 2/11, 2/15, 2/16, 3/9, 3/11, 3/15, 4/7, 4/16, 4/26, 4/28, 4/29, 4/30, 5/7, 12/10/54. From FERMCO Box 535-280. Cincinnati, OH: National Lead Company of Ohio. 1954.

NLO (National Lead Company of Ohio). Analytical data sheet of alpha in air dust samples taken in Health and Safety Service Building collected 2/22, 2/24, 5/12, 5/13, 6/24/55. From FERMCO Box 535-283. Cincinnati, OH: National Lead Company of Ohio. 1955.

NLO (National Lead Company of Ohio). Analytical data sheet of alpha in air dust samples taken in Health and Safety Service Building collected 2/2/56. From FERMCO Box 535-283. Cincinnati, OH: National Lead Company of Ohio. 1956.

NLO (National Lead Company of Ohio). Analytical data sheets of alpha in wipe samples in Laboratory Building collected 1/2, 1/21, 2/28, 3/4, 3/6, 6/17, 6/18, 6/19, 6/21, 6/23, 6/25, 6/26, 6/27, 7/1, 7/2, 7/3, 7/5, 7/8, 7/11, 7/12, 7/15, 7/16, 7/18, 7/30, 8/15, 8/19, 9/3, 10/23, 10/24, 10/29, 11/11, 11/13, 12/16/57. From FERMCO Box 535-286. Cincinnati, OH: National Lead Company of Ohio. 1957.

NLO (National Lead Company of Ohio). Analytical data sheets of alpha in air dust samples from Laboratory Building collected 2/6, 2/11,2/15, 3/3, 3/5, 3/15, 12/18, 12/19, 12/23, 12/24, 12/29/58. From FERMCO Box 535-287. Cincinnati, OH: National Lead Company of Ohio. 1958.

NLO (National Lead Company of Ohio). Analytical data sheets of alpha in air dust samples from Health and Safety Building collected 1/24, 8/16, 8/17, 9/30, 10/1/58. From FERMCO Box 535-288. Cincinnati, OH: National Lead Company of Ohio. 1958.

NLO (National Lead Company of Ohio). Analytical data sheets of alpha in air dust samples from Laboratory Building collected 1/13, 2/9, 3/10, 3/11, 6/19, 10/1, 10/9, 10/12, 10/13, 10/14, 10/29, 11/6,11/21/59. From FERMCO Box 535-288. Cincinnati, OH: National Lead Company of Ohio. 1959.

NLO (National Lead Company of Ohio). Analytical data sheets of alpha in air dust samples from Health and Safety Building collected 1/19, 1/23, 3/31/59. From FERMCO Box 535-288. Cincinnati, OH: National Lead Company of Ohio. 1959.

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ENGINEERING DRAWINGS

(Note: Many of these engineering drawings were produced for construction projects at the FMPC site. Because of this, a project number, such as a CP number, is often noted on the drawing. The project numbers have not been listed here, but can be easily located. The dates listed in the formal reference are the dates of the latest revision of the drawing. If the date of the latest revision differs significantly from the date of the original drawing, the "Revision 0" date is noted in the parenthetical remarks. Some of the titles may look odd, but they are given as recorded on the drawing, to the extent feasible.)

Plant 1 Drawings

WMCO (Westinghouse Materials Company of Ohio). Plant 1. Preliminary. Four sheet set of drawings showing the layouts of the first, second, third, and fourth floors. Cincinnati, Ohio: WMCO. circa 1990. (Drawings for recent planned construction activities? Useful for locating some interior equipment, and for locations of numbered and lettered support columns.)

Plant 2/3 Drawings

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NLCO (National Lead Company of Ohio). Plant 4, south end, dust collection alterations, flow diagram. Revision 1. Fernald, Ohio: NLCO. drawing number 4-4353; FMPC drawing index codes 04A-5500-F-01496 and 04A-5500-H-01497. circa 24 July 1959.

NLCO (National Lead Company of Ohio). Exhaust stack, dust collectors G4-3 & G4-7. Fernald, Ohio: NLCO. sk. number M4-244; FMPC drawing index code 04A-5502-H-01072. 21 April 1961. (Shows dimensions of stacks, including original-style rain cap, for dust collectors G4-3 and G4-7.)

Roberts, B. N. Feed Materials Production Center, Fernald area, process flow diagram, Green Salt Plant, sump recovery system. Philadelphia, Pennsylvania: Catalytic Construction Company. drawing number P-3004-86-F; FMPC drawing index code 04X-1450-F-00434. 24 July 1952.

Smith, R. W. Feed Materials Production Center, Fernald area, engineering flow diagram, heating & ventilation, general vacuum cleaning system, Green Salt Plant. Revision 1. Philadelphia, Pennsylvania: Catalytic Construction Company. drawing number 3004-P-03-A; FMPC drawing index codes 04A-1450-H-00416 and 04A-1450-F-00417. 16 April 1952.

Smith, R. Feed Materials Production Center, Fernald area, engineering flow diagram, heating & ventilating, vacuum conveying system, Green Salt Plant. Revision 2. Philadelphia, Pennsylvania: Catalytic Construction Company. drawing number 3004-P-05-A; FMPC drawing index codes 04A-1450-H-00419 and 04A-1450-F-00420. 16 June 1952.

Smith, R. W. Feed Materials Production Center, Fernald area, engineering flow diagram, heating & ventilating, dust control, process heating & ventilating, Green Salt Plant. Revision 1. Three sheets. Philadelphia, Pennsylvania: Catalytic Construction Company. drawing number 3004-P-04-R; FMPC drawing index code 04X-1450-H-00418. 6 August 1952.

Stull, H. Plant 4, south end, dust collection alterations, plans & elevation. Fernald, Ohio: National Lead Company of Ohio. drawing number 4-4354; FMPC drawing index code 04A-5500-H-01498. 30 October 1958.

Stull, H. Plant 4, dust collection alterations, east side duct work, elevations. Fernald, Ohio: National Lead Company of Ohio. drawing number 4-4393; FMPC drawing index code 04A-5500-H-01540. 13 April 1959.

Turkowitz, L. Plant 4, reactor area, ventilation of blender & packaging station, ductwork for dust collector G4-12, elevations A–A & B–B. Revision 1. Fernald, Ohio: National Lead Company of Ohio. drawing number 4-4496; FMPC drawing index code 04A-5500-H-01632. 14 November 1985. (Revision 0 dated 23 April 1963. Revision 1 indicates replacement of rain cap, with new style, on stack for dust collector G4-12.)

WMCO (Westinghouse Materials Company of Ohio). Plant 4, composite 1988 thru 1992, project construction periods interior. Preliminary. Seven sheet set of drawings showing the layouts of the various elevations (floors) in the plant. Cincinnati, Ohio: WMCO. circa 1990. (Drawings for recent planned construction activities? Useful for locating some interior equipment, and for locations of numbered and lettered support columns.)

Plant 5 Drawings

Catalytic Construction Company. Feed Materials Production Center, Fernald area, architectural, elevations, Metals Plant. Revision 3. Philadelphia, Pennsylvania: Catalytic Construction Company; drawing number 3005-C-1002-A; FMPC drawing index code 05X-1450-A-00009. 13 June 1988. (Revision 0 dated 13 December 1951. Shows planned replacements of weather caps (rain caps) for stacks for dust collectors G5-249, G5-250, G5-251, G5-253, G5-260, and G5-261.)

Dickson. Feed Materials Production Center, Fernald area, sections "A–A" & "B–B," heating & ventilating, Metals Plant. Revision 2. Philadelphia, Pennsylvania: Catalytic Construction Company; drawing number 3005-P-56-A; FMPC drawing index code 05X-1450-H-01087; 14 November 1985. (Revision 0 dated 31 March 1952. Shows planned replacements of weather caps (rain caps) for stacks for dust collectors G5-249 and G5-251. Also shows locations of stack sampling connections into these stacks.)

Dickson. Feed Materials Production Center, Fernald area, sections "E–E" & "F–F," heating & ventilating, Metals Plant. Revision 2. Philadelphia, Pennsylvania: Catalytic Construction Company; drawing number 3005-P-58-A; FMPC drawing index code 05X-1450-H-01089; 14 November 1985. (Revision 0 dated 30 April 1952. Shows planned replacements of weather

caps (rain caps) for stacks for dust collectors G5-254 (?) and G5-261. Also shows locations of stack sampling connections into these stacks.)

Dickson. Feed Materials Production Center, Fernald area, sections "G–G" & "S–S," heating & ventilating, Metals Plant. Revision 2. Philadelphia, Pennsylvania: Catalytic Construction Company; drawing number 3005-P-60-A; FMPC drawing index code 05X-1450-H-01091; 14 November 1985. (Revision 0 dated 1952. Shows planned replacements of weather caps (rain caps) for stacks for dust collectors G5-258 and G5-260. Also shows locations of stack sampling connections into these stacks.)

Singmaster & Breyer. Metals Plant no. 5 – new wings, elevations, architectural drawing. Revision 3. New York: Singmaster & Breyer; drawing number 5250-5A-5036; FMPC drawing index code 05X-7000-A-00075; 16 May 1986. (Revision 0 dated 22 March 1955.)

WMCO (Westinghouse Materials Company of Ohio). Plant 5, October 1989 thru 1990. Preliminary. Two sheet set of drawings showing the layouts of the first and second floors in the plant. Cincinnati, Ohio: WMCO; 18 December 1989. (Drawings for recent planned construction activities? Useful for locating some interior equipment, and for locations of numbered and lettered support columns.)

WMCO (Westinghouse Materials Company of Ohio). Plant 5. Preliminary. Two sheet set of drawings showing the layouts of the first, second, and third floors. Cincinnati, Ohio: WMCO; circa 1990. (Useful for locating some interior equipment, and for locations of numbered and lettered support columns.)

Plant 6 Drawings

NLCO, Boies, R.B., Engineering Drawing Plant 6; NLCO Plant Layout; 27 May 1965.

WMCO (Westinghouse Materials Company of Ohio). Plant 6. Preliminary. One sheet drawing showing the interior layout. Cincinnati, Ohio: WMCO; circa 1990. (Useful for locating some interior equipment, and for locations of numbered and lettered support columns.)

Plant 7 Drawings

Catalytic Construction Company. Production plant, Fernald area, equipment layout–process bldg. Set of seven drawings; five of plans at various interior elevations and two of cross sections. Philadelphia, Pennsylvania: Catalytic Construction Company; drawing numbers 3241-A103 through 3241-A109; FMPC drawing index codes 07X-1450-M-00004 through 07X-1450-M-00010. (Presumed to be original construction drawings. Originally drawn 15 June 1953, most with revisions through late 1953 or early 1954, and two with revisions in 1987. Show equipment layout.)

Plant 8 Drawings

Bunk, L. W. Plant 8, area "C," replacement of box furnace dust collector. Fernald, Ohio: National Lead Company of Ohio; FMPC drawing index code 08X-5500-H-02307; 22 November 1968. (Shows plans for replacing the Sly 6A Dynaclone dust collector with a Day 44AC10 dust collector, to serve the box furnace. Shows location of exhaust stack for this dust collector and locations of two nearby scrubbers.)

Jurkonitz, L. Plant 8 Calciner Bldg.,. first floor, drumming station for Hoffman unit, location and duct work connections, drawing number 8-4990. FMPC drawing index code 39A-5500-

H-0059 and 08D-5500-H--1889. Cincinnati, OH: National Lead Company of Ohio. 5 June 1963.

NLCO (National Lead Company of Ohio). Plant 8, area C, scrubber for box furnace, plan. Fernald, Ohio: NLCO; FMPC drawing index code 08X-5500-H-02312; circa 1968. (Ductwork and piping.)

NLCO (National Lead Company of Ohio). Plant 8, area C, scrubber for box furnace, sections "A–A" &"B–B." Fernald, Ohio: NLCO; FMPC drawing index code 08X-5500-H-02313; circa 1968. (Shows locations of stacks for scrubber (without rain cap) and dust collector.)

NLCO (National Lead Company of Ohio). Plant 8, crusher area, outside crusher dust collector replacement, demolition, plans & section. Revision 3. Fernald, Ohio: NLCO; drawing number 8-4213; FMPC drawing index code 08X-5500-H-02315; 16 January 1969. (Revision 0 dated circa 1958. Shows the stack for dust collector G43-44C without a rain cap. Indicates the dust collector was to be removed circa 1969.)

WMCO (Westinghouse Materials Company of Ohio). Plant 8, January 1990 to January 1991. Preliminary. Two sheet set of drawings showing the layouts of the first and second floors in the plant. Cincinnati, Ohio: WMCO; 12 February 1990 and 24 January 1990. (Drawings for recent planned construction activities? Useful for locating some interior equipment, and for locations of numbered and lettered support columns.)

Plant 9 Drawings

George A. Fuller Company. Location and detail of exhaust stacks thru roof – new Material Feed Production building #3542. Cincinnati, Ohio: George A. Fuller Company; drawing number 3542-H-GAFCo-7-sA; FMPC drawing index code 09X-3595-H-00275; 4 November 1952. (Shows locations of stacks on roof and heights. But, stacks appear to be numbered by stack number (?), or some other number not directly related to dust collector number.)

WMCO (Westinghouse Materials Company of Ohio). Plant 9. Preliminary. One sheet drawing showing the interior layout. Cincinnati, Ohio: WMCO; circa 1990. (Useful for locating some interior equipment, and for locations of numbered and lettered support columns.)]

Pilot Plant Drawings

WMCO (Westinghouse Materials Company of Ohio). Pilot Plant, interior. Preliminary. One sheet drawing showing the interior layout. Cincinnati, Ohio: WMCO; circa 1990. (Useful for locating some interior equipment, and for locations of numbered and lettered support columns.)

K-65 and Metal Oxide Silos Drawings

Anonymous. FMPC drawing index code 34X-1450-F-00020. (Drawing showing water piping for K-65 drum dumping and washing and slurry to silos. Sheet 1 of 2.)

Catalytic Construction Company. Process flow diagram, hot raffinate storage system. Philadelphia, Pennsylvania: Catalytic Construction Company; drawing number 3034-H-01-F; FMPC drawing index code 34X-1450-F-00037; 14 June 1951.

Clift, W. K-65 storage farm, K-65 handling & storage, enlarged K-65 storage farm flow diagram. Fernald, Ohio: National Lead Company of Ohio; FMPC drawing index code 34X-5500-F-00061. 4 December 1956.

Creter, L. W. Process flow diagram K-65 handling & storage. Two sheets. Philadelphia, Pennsylvania: Catalytic Construction Company; drawing number P-3034-37-F; FMPC drawing index code 34X-1450-F-00038. 5 May 1952.

Geesner, T. J. K-65 storage, K-65 tank embankment stabilization, general layout. Revision 1. Fernald, Ohio: NLO, Inc.; FMPC drawing index code 34X-5500-G-00084; 14 July 1983. (Shows expansion of the silo berms.)

Glenn, F. J. Engineering flow diagram metals oxide. Philadelphia, Pennsylvania: Catalytic Construction Company; drawing number 3035-H-02-A; FMPC drawing index code 35X-1450-F-00017; 20 June 1952.

Locke, M. K-65 silos, K-65 radon treatment system, 'as built,' plan, details and sections. Fernald, Ohio: Westinghouse Materials Company of Ohio; FMPC drawing index code 34X-5500-M-00116; 5 October 1990.

NLCO (National Lead Company of Ohio). K-65 slurrying system. Fernald, Ohio: NLCO; sk. number P-34-6; FMPC drawing index code 34X-5506-F-00043; 8 May 1952.

NLCO (National Lead Company of Ohio). Tank unloading & drumming system for 'K-65' material. Fernald, Ohio: NLCO; sk. number E-34-3; FMPC drawing index code 34X-5500-F-00051; 23 April 1958. (for proposed removal of K-65 material from silos.)

NLCO (National Lead Company of Ohio). K-65 storage, earth embankment at K-65 tanks, plan. Fernald, Ohio: NLCO; drawing number 34-4013; FMPC drawing index code 34X-5500-M-00066; 2 December 1963.

Preload (Preload Enterprises, Inc.). Two 125,000 c.f. slurry storage tanks - type K65, Atomic Energy Commission, Fernald, Ohio, Catalytic Construction Co. Revision 4. New York: Preload; drawing number 51T20-3; FMPC drawing index code 34X-1450-A-00086; 15 September 1951. (Drawing from original constructor of K-65 silos. Useful for construction details, characteristics.)

Preload (Preload Enterprises, Inc.). Two 125,000 c.f. slurry storage tanks - type K65, Atomic Energy Commission, Fernald, Ohio, details. Revision 3. New York: Preload; drawing number 51T20-7; FMPC drawing index code 34X-1450-P-00090; 25 September 1951. (Drawing from original constructor of K-65 silos. Useful for locations and details of hatches and other penetrations.)

Preload (Preload Enterprises, Inc.). Two 125,000 c.f. tanks, Atomic Energy Commission, Fernald, Ohio, engrs: Catalytic Const. Co. Revision 6. New York: Preload; drawing number 51T29-2; FMPC drawing index code 34X-1450-A-00092; Drawing from original constructor of K-65 silos. Cincinnati, OH: National Lead Company of Ohio. 20 November 1951.

Seiwert, J. F.M.P.C. Fernald, Ohio, protective cover system for K-65 tanks, dome plans & details. Cincinnati, Ohio: Camargo Associates, Limited; FMPC drawing index code 34X-5500-S-00109; 16 December 1985.

Smith, E. Process flow diagram K-65 (handling & storage. Preliminary. Two sheets. Philadelphia, Pennsylvania: Catalytic Construction Company; drawing number 3034-H-02-F; FMPC drawing index code 34X-1450-F-00019; 12 July 1951.

Stull, H. P. K-65 storage farm, K-65 handling & storage flow diagram. Fernald, Ohio: National Lead Company of Ohio; drawing number 34-4005; FMPC drawing index code 34X-5500-F-00060; 7 February 1956.

"Old" Solid Waste Incinerator and Sewage Treatment Area Drawings

Bosum, D. Sewage Treatment Area Classified Paper Destroyer Installation, drawing No. 39-4002. Cincinnati, OH: National Lead Company of Ohio. 27 August 1957.

Briscoe, D. Incinerator Building, drawing number MX-44. FMPC drawing index code 39X-5502-S-00011 and 00A-5502-A-00126. Cincinnati, OH: National Lead Company of Ohio. 28 November 1961.

Bunk, L. W. Sewage treatment area, incinerator, modifications to plant incinerator. Fernald, Ohio: National Lead Company of Ohio; FMPC drawing index code 39X-5500-G-00009; 24 April 1969. (Shows size of existing stack.)

NLCO (National Lead Company of Ohio). Waste paper and chemical carton incinerator, drawing number 39-5000; FMPC drawing index code 39X-5500-X-00007. Cincinnati, OH, 31 March 1952

NLCO (National Lead Company of Ohio). Incinerator. Preliminary. Fernald, Ohio: NLCO; drawing number 39-4000; FMPC drawing index code 39X-5500-X-00002; 2 October 1953. (Appears to be original plans for installing the incinerator. However, the as-built location of the incinerator differs from that shown on this drawing.)

NLCO (National Lead Company of Ohio). Foundation plan, details, incinerator. Revision 2. Fernald, Ohio: NLCO; drawing number 39-4001; FMPC drawing index codes 39X-5500-A-00003 and 39X-5500-S-00004; circa 1954. (Revision 1 dated 3 August 1954. Appears to be original plans for installing the incinerator. The as-built location of the incinerator shown on this drawing appears to match the actual location.)

NLCO (National Lead Company of Ohio). Incinerator details. Revision 1. Fernald, Ohio: NLCO; drawing number 39-4002; FMPC drawing index codes 39X-5500-A-00005 and 39X-5500-S-00006; 1 April 1954.

Richards, A. Sewage treatment plant expansion, plot plan. Revision 2. New York: Singmaster & Breyer; drawing number 5250-25M-4019; FMPC drawing index code 25A-7000-F-00033; 14 July 1960. (Revision 0 dated 26 May 1955. Useful for showing location of the incinerator, Manhole 175, and the sewer sampler station on the effluent line to the Great Miami River.)

Schultheis, R. Incinerator, plans & sections. Revision 1. Fernald, Ohio: National Lead Company of Ohio; FMPC drawing index code 39X-5500-M-00012; 28 September 1976. (Revision 0 dated 21 September 1972. Shows size of stack (new).)

Schultheis, R. Incinerator building, Solid Waste Incinerator Piping and electrical plan and details, FMPC drawing code 02F-5500-P-02808 and 02F-5500-E-02809. National Lead Company of Ohio. 3 March 1979.

Schultheis, R. Incinerator building, Solid Waste Incinerator general arrangement plan and elevation, FMPC drawing code 02F-5500-M-02805. National Lead Company of Ohio. 26 January 1979.

Other Buildings Drawings

Geesner, T. J. Bldg 39, liquid waste incinerator, conduit & cable layout. Fernald, Ohio: National Lead Company of Ohio; FMPC drawing index code 39X-5500-E-00041; 10 June 1982.

Singmaster & Breyer. (Raffinate calciner bldg.) incinerator bldg., plans – elevations. Revision 5. New York: Singmaster & Breyer; drawing number 5250-3A-5318; FMPC drawing index codes 02F-7000-A-00957 and 39A-5500-A-00054; 4 August 1988. (Revision 0 dated 11 April 1956. Shows locations of exhaust stacks.)

WMCO (Westinghouse Materials Company of Ohio). Incinerator Building, Floor Plan, drawing number 39A-5500-A-00063. Approved by G. Paul. Cincinnati, OH, 10 August 1990 (?).

FMPC General Area Drawings

Locke, M. Production area site plan. Fernald, Ohio: Westinghouse Materials Company of Ohio; 26 March 1990. (Filename ZFAI:[100,4]PRODMAP.DGN. General map of production area, with buildings and some features labeled.)

WMCO (Westinghouse Materials Company of Ohio). Feed Materials Production Center. Fernald, Ohio: WMCO; 27 June 1989. (R.E.S.-1238. Filename ZFAI:[100,4]RCRA.DGN. General map showing complete FMPC site and surrounding roads and houses. Shows coordinates of the FMPC Site Origin, in USGS, OSP, and site coordinates. Also shows direction of north for each of these coordinate systems.)

Woolpert Consultants. Fernald facility, Department of Energy, Fernald, Ohio. Seventeen sheets. Dayton, Ohio: Woolpert Consultants; FMPC drawing index codes 75X-5500-G-00112 through 75X-5500-G-00128; circa 1988. (Topographic maps, prepared for Weston, of much of the FMPC facility, at scale of 1 in = 50 ft. Very useful for locating points on the site. Uses Ohio State Plane coordinate system.)

Miscellaneous Drawings

Cooper, D. P. 1985 site runoff characterization survey. Fernald, Ohio: NLO, Inc.; FMPC drawing index code 18X-5500-X-00658; 16 September 1985. (This "drawing" consists of copies of Attachments 1, 2, and 3 to an unidentified report, presumably regarding site water runoff and calculations of required capacity of storm catch basin. Includes a reference to availability of backup data on site characterization.)

Ellis, J. J. General stack sampler assembly & detail. Revision 4. Fernald, Ohio: NLO, Inc.; FMPC drawing index code 00X-5500-N-01368; 18 January 1989. (Revision 0 dated 11 January 1983. Shows filter holder, tubing and connections, and sampling probe designs.)

NLCO (National Lead Company of Ohio). Sketch and text describing bag filter specifications, for purchasing department. Revision 1. Fernald, Ohio: NLCO; sk. number GS-4-5982, B-4-1; FMPC drawing index code 04X-5505-M-01778; 26 July 1957. (Revision 0 undated. For Ensinger Filter, G4-39.)

NLCO (National Lead Company of Ohio). Sketch and text describing bag filter specifications, for purchasing department. Fernald, Ohio: NLCO; sk. number GS-4-2611, B-4-2; FMPC drawing index code 04X-5505-M-01779; undated. (For dust collectors G4-2 through G4-7 and (unknown) item G1-208.)

NLCO (National Lead Company of Ohio). Sketch and text describing bag filter specifications, for purchasing department. Revision 1. Fernald, Ohio: NLCO; sk. number GS-X-450, B-4-3; FMPC drawing index code 04X-5505-M-01780; 22 July 1957. (Revision 0 undated. For G4-11. Was superseded 23 October 1958.)

NLCO (National Lead Company of Ohio). Sketch and text describing bag filter specifications, for purchasing department. Fernald, Ohio: NLCO; sk. number GS-X-450; FMPC drawing index code 04X-5505-M-01781; undated. (For GS-5-247 Hoffman unit. Was superseded 23 October 1958.)

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Berger, J.D., G.S. Gist, C.M. Morrow, D.J. Niederkorn, D.T. Robinson (ORAU). Environmental Program Review of the Feed Materials Production Center Fernald, Ohio. (100 pg review of air monitoring, water treatment and monitoring, groundwater, soil, sediment, vegetation, monitoring, waste management, analytical procedures, QA, dose assessment, emergency preparedness). Prepared by Radiological Site Assessment Program. Oak Ridge Associated Universities, Oak Ridge, TN. October 1985.

Dolan, L.C. Action Plan in Response to ORAU Environmental Program Review. Letter to J.A. Reafsnyder. (Lists of short-term, 67 action items, and long-term, 10 action items, priority items). Westinghouse Materials Company of Ohio. 15 January 1986.

Dugan, T. A., G. L. Gels, J. S. Oberjohn, and L. K. Rogers, FMPC Annual Environmental Report for Calendar Year 1989. FMPC-2200, Special. Environmental Management Department, Westinghouse Materials Company of Ohio. October 1990.

Facemire, C. F., D. L. Jones and R. W. Keys, FMPC Environmental Monitoring Annual Report for 1984, NLCO-2028, Special. National Lead of Ohio, 1985. 15 July 1985.

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APPENDIX B

PLANT PROCESSES AND WASTES

In this appendix the functions of each of the major processing facilities at the Feed Materials Production Center (FMPC) are discussed briefly. The major activities in each facility are highlighted and the chemical conversions that occurred are presented (WMCO 1988). Discussions with plant staff have indicated that the basic processing scheme was employed throughout all years of operation. The chemical forms of the radioactive materials are particularly important to the estimation of doses from material released to the atmosphere. More detailed information about uranium processing is available in Harrington and Ruehle (1959).

The FMPC was primarily concerned with processing uranium. Most of the uranium received at the FMPC had been separated from the naturally occurring daughter radionuclides, including ^{226}Ra. Appendix J describes the disposition of wastes from processing of raw uranium ores early in the history of the FMPC. Relatively small amounts of thorium were also processed at various times. General descriptions of the thorium processes are given in a later section. Cuthbert (1958) provides more detailed information on thorium processing.

To reflect the emphasis on uranium processing at the FMPC, presentation of plant functions in this appendix follows the flow of uranium through the various facilities as it was changed from the incoming material to finished products. The primary processing sequence involved Plant 1, Plant 2/3, Plant 4, Plant 5, Plant 9, and Plant 6. These facilities are discussed in that order. Figure B–1 illustrates flows of materials between facilities. Figure B–2 shows the layout of the plants within the Production Area at the FMPC.

Uranium that was recycled from other facilities entered the FMPC production scheme at a location consistent with its chemical form. For example, uranium delivered to the FMPC as UO_3 could be fed directly to Plant 4 without processing in Plant 2/3. Recycled uranium was separated from fission and activation products prior to shipment to the FMPC; however, some of those radionuclides were present as contaminants. Appendix D contains the results of recent measurements performed to identify levels of fission and activation products in materials at the FMPC. Near the end of this appendix, the functions of other facilities and waste management activities are described.

PLANT 1 — THE SAMPLING PLANT

Incoming materials for the FMPC were weighed, sampled, and analyzed for uranium (U) content in Plant 1. Initially, the plant handled large quantities of uranium ore and concentrates and had a crushing, grinding, and blending capacity of more than 9 metric tons per hour (about 20,000 pounds per hour).

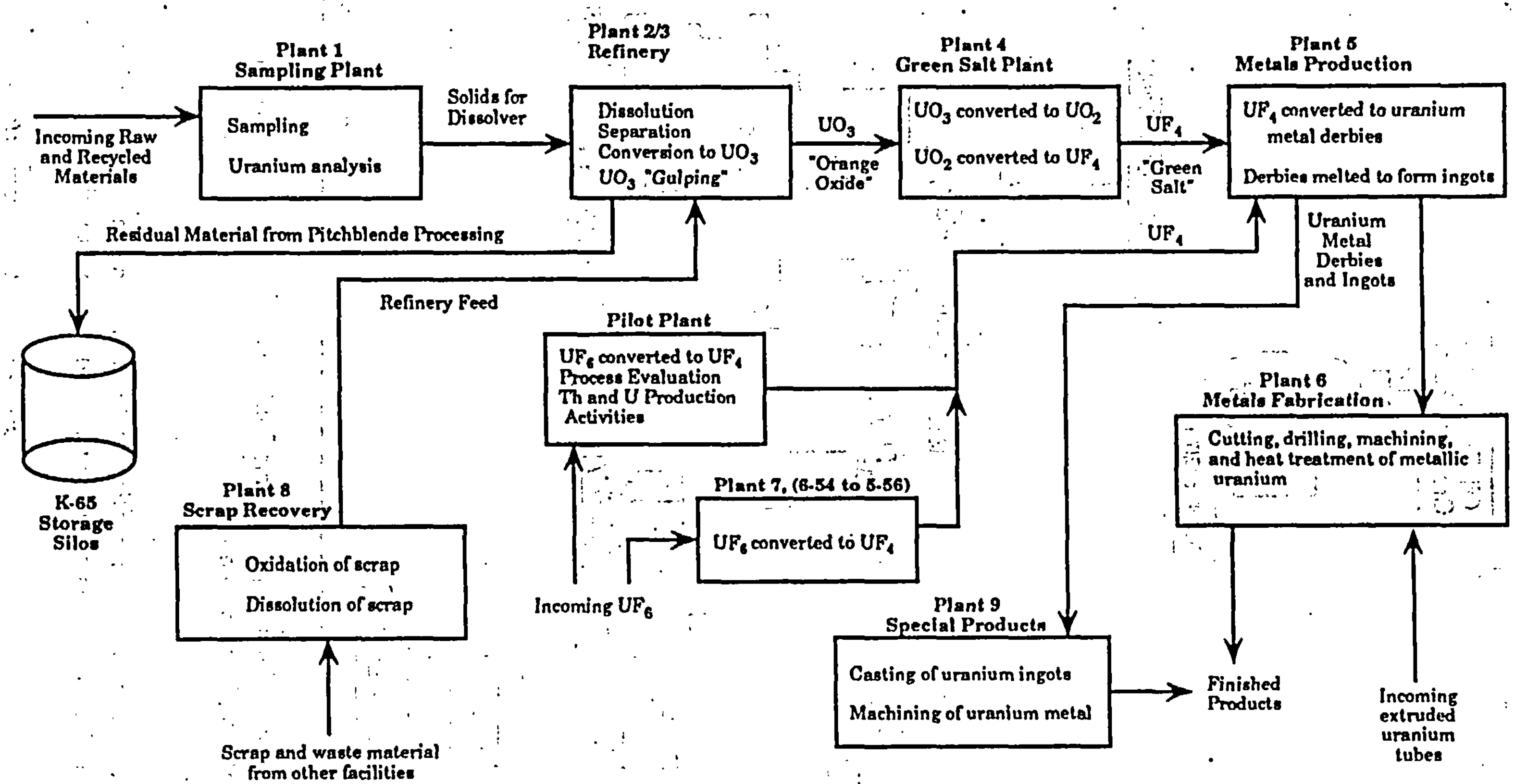

Figure B–1. Schematic diagram of flows of uranium Between Facilities at FMPC. Flows of recycled material were more complex than can be illustrated simply. For example, recycled uranium received as uranium trioxide would not be processed in Plant 2/3, but would be feed for Plant 4.

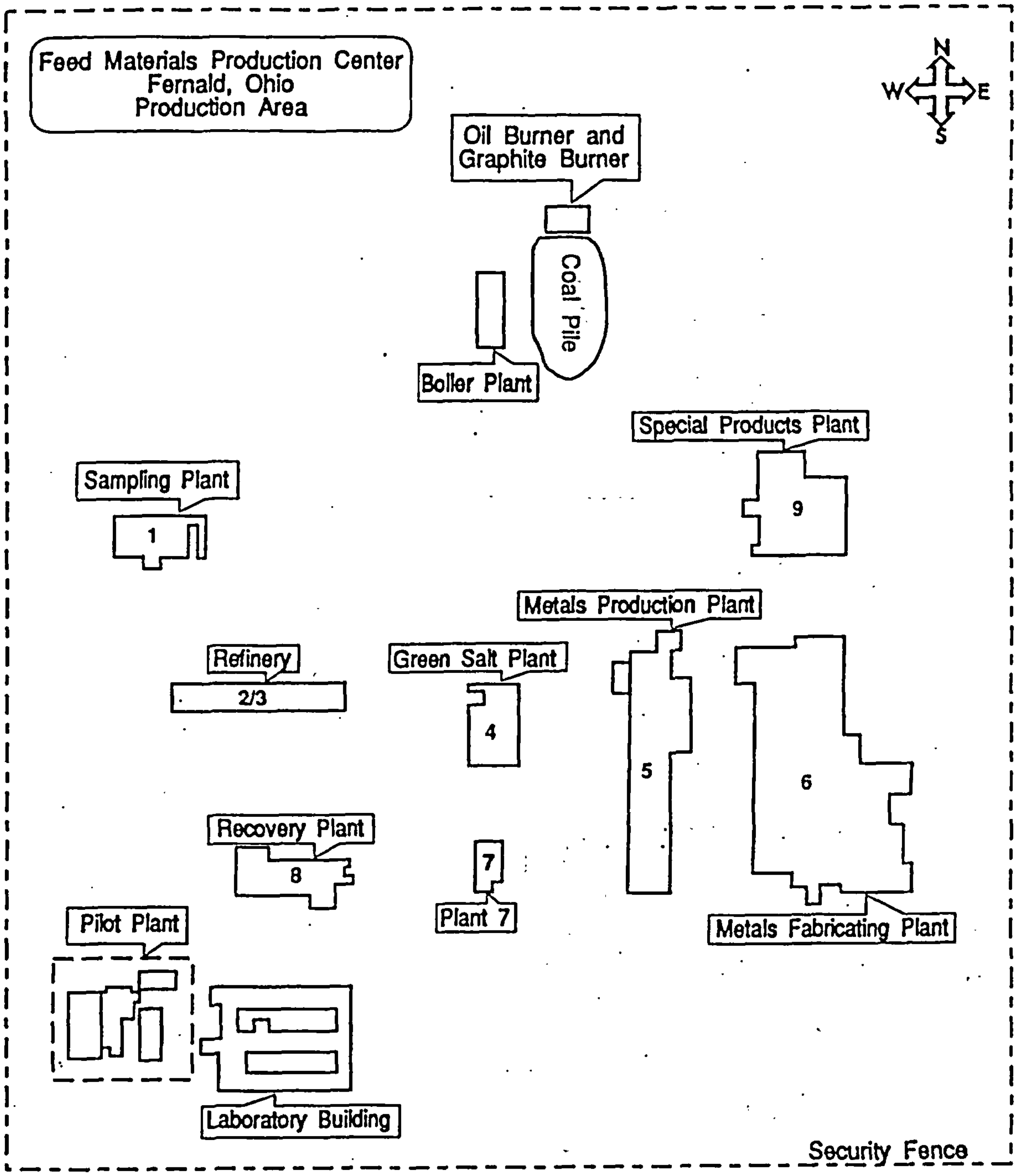

Figure B–2. Locations of major processing facilities within the production area at the Feed Materials Production Center.

In 1970, a digestion system was installed in Plant 1 to permit processing of uranium enriched to as much as 5% ^{235}U. This system was used intermittently in the later years of plant operation.

Thus two types of processes were used in Plant 1:

- mechanical treatment: feed materials were dried, crushed, milled, ground, classified, and blended if necessary
- digestion: feed materials were dissolved in acid solutions

Material prepared in Plant 1 was transferred to Plant 2/3 for processing as described in the following section.

PLANT 2/3 — THE REFINERY

The function of the refinery was to separate the uranium contained in various feed materials and convert it to a high-purity product, uranium trioxide (UO_3, called orange oxide because of its color). This was accomplished using three chemical processes: digestion, extraction, and calcination (or denitration).

- digestion: uranium bearing materials were digested with nitric acid in an agitating tank to produce a slurry containing uranyl nitrate, $UO_2(NO_3)_2$, and nitric acid in solution and insoluble impurities.
- extraction: the aqueous slurry was mixed with an organic solvent consisting of tributyl phosphate dissolved in kerosene. The uranyl nitrate was preferentially extracted out of the aqueous phase into the organic solvent while the impurities and most of the nitric acid remained in the aqueous slurry. The purified uranyl nitrate was then preferentially extracted from the organic solvent into deionized water in the absence of nitric acid.
- calcination or denitrification: the uranyl nitrate solution was concentrated in a closed evaporator system and by further heating in large tanks. The concentrated material was then transferred to denitration pots. The pots were fired for several hours to convert the uranyl nitrate to uranium trioxide and drive off the volatile nitrogen oxides (NO_x). The heat induced decomposition process is: $UO_2(NO_3)_2 \rightarrow UO_3 + NO_x$. The denitration pots were vented through a scrubber system so the nitrogen oxides could be recovered, as nitric acid, which was recycled to the digestion area.

The UO_3 product was transferred from the pots using a vacuum line. This transfer process, called "gulping" the pot of UO_3, carried the product through cyclone separators to storage hoppers. The UO_3 was then ground and packaged for shipment to Plant 4 or offsite.

Some experimental work with thorium was conducted in the Refinery during the late 1960s. No production scale thorium operations were undertaken in the facility.

PLANT 4 — THE GREEN SALT PLANT

Plant 4 was named for its product, uranium tetrafluoride (UF_4, called green salt because of its color). The conversion of UO_3 to UF_4 was a two-stage process.

- reduction: powdered UO_3 was heated in stainless steel fluid bed reactors with dissociated ammonia (H_2 and N_2) at temperatures ranging from about 530 to 590°C. The reduction of uranium trioxide to uranium dioxide (UO_3 to UO_2) is: $UO_3 + H_2 \rightarrow UO_2 + H_2O$. The uranium product of this reaction was called brown oxide.

- hydrofluorination: the UO_2 produced by the reduction process was then reacted with anhydrous hydrogen fluoride in a series of three counter current flow screw reactors. The temperature in the metal reactors increases from about 150°C for the first to about 650°C for the last reactor. The hydrofluorination reaction is: $UO_2 + 4\,HF \rightarrow UF_4 + 2\,H_2O$. The UF_4 product was weighed, blended if necessary, and packaged for shipment to Plant 5.

Production of thorium tetrafluoride in Plant 4 occurred soon after startup of the facility in 1954. The reaction of thorium dioxide with HF is similar to the uranium reaction shown above.

PLANT 5 — METALS PRODUCTION

Conversion of UF_4 to masses of uranium metal, called derbies, was accomplished in Plant 5. The derbies were then remelted and cast into ingots of metallic uranium. The reduction process in Plant 5 is described below:

- reduction: green salt was reacted with magnesium metal (Mg) in a steel pot lined with magnesium fluoride (MgF_2) slag. The steel pot was heated in a furnace to a temperature between 650°C and 820°C for 3-4 hours before the reaction occurred. The metal product was a mass of uranium, called a derby, that weighed about 150 kg (~330 lb.). The reaction, which is exothermic, is: $UF_4 + 2\,Mg \rightarrow U + 2\,MgF_2$. The internal temperature of the pot could reach 1650°C, well above the melting point of uranium metal (~1130°C).

After cooling, the derbies were transferred to casting area in Plant 5, to Plant 9, or shipped offsite. Activities in the casting area are described next. The processes in Plant 9 are described in the next section. The other principal activites in Plant 5 were:

- remelting and casting: uranium metal derbies and scrap uranium metal were vacuum melted in graphite crucibles, and the molten uranium metal (~1480°C) was flowed into heated graphite molds to produce ingots weighing up to 650 kg (1440 lb.).

The top 5 cm of each ingot was sawed off to remove cavities and impurities before the ingots were transferred to Plant 9 for drilling and machining. These croppings were subsequently remelted with derby metal.

PLANT 9 — SPECIAL PRODUCTS

Independent processes carried out in the plant included casting of large-diameter ingots from derbies and high-grade recycled metals, drilling and machining of uranium metal ingots for extrusion, and chemical decladding of rejected unirradiated fuel elements from Hanford. These processes are described below:

- casting: ingots that measured up to 33 cm (13 inches) in diameter and weighed up to 900 kg (~1980 lb.) were cast.
- drilling and machining: cropped billets were center drilled and machined, prior to shipment offsite for extrusion.

- chemical decladding: rejected unirradiated fuel elements from Hanford were immersed in dilute nitric acid to remove the outer layer of copper. They were then treated with dilute hydrofluoric acid to remove the Zircaloy-2 cladding from the uranium metal core of the element. The purified uranium metal was recycled to the ingot manufacturing process.

Machined billets were sent to Plant 6 for treatment and inspection before shipment offsite.

Thorium metal was also processed in Plant 9 and thorium scrap metals were formed into briquettes for recycle. These operations occurred during the period 1954–1955.

PLANT 6 — METALS FABRICATION

Uranium metal billets and extruded tubes were heat treated, cut, and machined in this facility. Flat billets were also produced in a rolling mill. The feed stock of round billets was received from Plant 9; extruded tubes were received from offsite locations.

- heat-treatment: uranium metal billets from Plant 9 were heat treated in a neutral salt bath before shipment offsite for extrusion.
- cutting and machining: blanks were produced by lathe cutting extruded tubes to appropriate lengths. These were treated in a hot salt bath and quenched in oil before being automatically machined to specific tolerances. Machined fuel sections were degreased, pickled, rinsed, and dried before final inspection and shipment.
- rolling: a rolling mill was used to produce flat billets that were inspected and shipped offsite.
- recycling: chips and turnings were crushed, pickled, rinsed, dried, and formed into briquettes for use in the Plant 5 casting operation.

Scrap metal produced in Plant 6 was prepared for recycling through Plant 5.

PLANT 8 — SCRAP RECOVERY

Recycling of residues and metal scraps from production processes, at the FMPC and other sites, was designed to return a suitable material to the uranium production stream. Refinery preparation for high grade scraps employed various furnaces to oxidize the material. Low grade residues were processed to yield an ammonium diuranate cake. Initially, conversion of the cake to feedstocks that were acceptable for the refinery was accomplished by offsite contractors. After a plant expansion in 1955, the need for such offsite processing was eliminated. A rotary kiln and vertical hearth furnaces of various sizes were used in Plant 8. The enrichment of materials processed was limited to a maximum of 1.25% ^{235}U.

- refinery preparation: uranium metals, uranium octoxide (U_3O_8, called black oxide), furnace salts, dust collector product, and floor sweepings were roasted in one of three furnace to dry them and to oxidize impurities such as metals, oil, and graphite.
- hydrometallurgical processing: conversion of various low grade residue forms to ammonium diuranate (ADU) cake or uranium ammonium phosphate (UAP) was performed in accordance with feed available and refinery requirements.

Processing steps varied over the years of operation of Plant 8. The history of the recovery operations (Mead 1972) provides additional details on processing and the range of recovery operations undertaken at the FMPC.

Plant 8 was also involved in the production of thorium hydroxide and thorium oxalate and the calcination of sump cakes that contained thorium. These operations occurred between 1966 and 1971.

PILOT PLANT

The Pilot Plant was used for numerous process testing and experimental operations. It was also employed as a production facility for various processes. In the early years, derbies were produced there, in the manner described above for Plant 5. Another process operated on a production scale was the direct conversion of uranium hexafluoride (UF_6) to green salt (UF_4). This production process was operated with UF_6 that contained as much as 2.5% ^{235}U. A two-step procedure was used:

- vaporization of UF_6: solid UF_6 in large cylinders was heated in autoclaves at approximately 110°C to produce gaseous UF_6.
- reduction of UF_6: gaseous UF_6 was mixed with hydrogen gas at 480-650°C in metal reactors to produce UF_4 powder. Hydrogen fluoride (HF) was a valuable byproduct of the reaction, which was: $UF_6 + H_2 \rightarrow UF_4 + 2\,HF$.

Much of the thorium production activity at the FMPC took place in the Pilot Plant. Several processes were operated there, beginning in 1964. Thorium production activities continued until 1980.

PLANT 7 — URANIUM HEXAFLUORIDE CONVERSION TO GREEN SALT

Plant 7 utilized the process for conversion of UF_6 to UF_4 that had bee successfully employed and improved in the Pilot Plant. There were two sets of four reactors in the facility. One set was used for natural uranium; the other was used for depleted uranium. Each reactor was designed for greater capacity than had been available in the Pilot Plant. Normally, three reactors were in operation and one was held as a spare. Under these conditions, production capacity was six tons per day for natural and depleted uranium tetrafluoride production. Plant 7 produced green salt for only two years, from June 1954 through May 1956, before it was closed and subsequently dismantled.

THE K-65 STORAGE SILOS

The K-65 Storage Silos were constructed of concrete in the western portion of the FMPC site in 1951–1952. Their purpose was to store residues from the extraction of uranium from ore concentrates that were processed during the early years of FMPC operation. The residues, containing the daughter products of the uranium decay chains, were transferred to the silos as slurries, primarily between 1953 and 1955. Other waste materials were also slurried to the silos in later years.

The slurried waste solids contain particulate radionuclides including radium (Ra). The nuclide ^{226}Ra decays to ^{222}Rn (radon gas), which can diffuse into the airspace of the silo and be released to the atmosphere. A detailed discussion of this process and of the changes to the silos that affect it is presented in Appendix J of this report.

WASTE INCINERATORS AND BURNERS

An incinerator for solid waste materials was installed near the eastern facility boundary, outside the production area, in 1954. This incinerator was operated intermittently until 1979, when it was found to be out of compliance with applicable codes. In 1980, a new solid waste incinerator was installed in the same area. A detailed chronology is provided in Appendix K.

A graphite burner, an oil burner, and an incinerator for organic liquids were also operated for varying periods. Both the graphite burner and the oil burner were simple arrangements that operated for about twenty years. The graphite burner was operated from November 1965 to September 1984. The oil burner began operation at the end of March 1962 and operated until June 1979. The liquid organic waste incinerator was installed in April 1983 and has operated since that time. Appendix K contains more information about these facilities. Estimated releases from these sources are presented in that appendix.

THE GENERAL SUMP FOR LIQUID WASTES

Physically, the General Sump is a collection of tanks of various sizes, pumps, piping and valves where process wastes from the various plants were received and analyzed. Some liquid wastes were generated in almost every operation at FMPC. The major process areas had individual treatment facilities capable of pretreating the liquid wastes that were peculiar to that particular process step. These plant treatment facilities were simple installations which provided equipment and tanks to collect waste liquors, to adjust pH for precipitation of uranium, and to filter the resultant slurry. Filter cake that resulted from precipitation, was recycled as a process residue, while the filtrate was pumped to the General Sump System. Effluent slurries from Plant 8 were discharged directly to the pits if uranium concentrations were below the discard limit. If the concentration was higher, the slurry was recycled (Johnson et al. 1958, Calhane 1961).

In addition, the Plant 2/3 Refinery had a sump system in place for neutralizing and recovering process materials and effluents. Some wastes received at the General Sump required only settlement and movement through the various tanks prior to discharge of the supernatant liquid and sludges to the wet chemical waste pit. If certain wastes exceeded the discard specifications for the General Sump, however, it was recycled through the Refinery sump in Plant 2/3 for further treatment. Standard Operating Procedures (NLCO 1961) directed that all acidic uranium-bearing wastes be adjusted for pH to obtain maximum precipitation of the radioactive material before being pumped to the wet chemical waste pit.

After settling had occurred in the tanks of the General Sump, the effluent was pumped to the Clearwell. located near the waste pits (Figure B–3). From the clearwell, the effluent was combined with effluents from the storm sewer system, treated sanitary sewer system and water treatment plant effluent before being discharged through Manhole 175 and the main effluent line to the Great Miami River. See Appendix L for more information about liquid waste disposal.

In October 1986, the Storm Water Retention Basin (SWRB) went on-line to help lower the number of storm sewer overflow violations. Runoff from the waste pit 4 was collected in the SWRB and then discharge into the plant effluent via waste pits 5 and 6, and the clearwell. This system helps control the flow of runoff from the waste storage area into Paddy's Run Creek. The SWRB also resulted in a decrease in total uranium discharged to the Storm Sewer Outfall Ditch (SSOD) (WMCO 1988). In 1986, prior to the SWRB operation, there were 3 hexavalent chromium violations at the combined General Sump and clearwell; and 3 total suspended solids (TSS) violations at Manhole (MH)-175; in 1987 there were no violations (Reafsnyder 1987).

Several major changes in treatment of FMPC process wastes occurred in 1987. First, on February 23, 1987, Waste Pit 5 and the clearwell were taken off-line to protect the groundwater as mandated by the Ohio EPA Director's Findings and Orders (DFO). At that time, wastewater was routed from the General Sump to the Biosurge Lagoon (BSL), instead of to Pit 5. The DFO also required that an additional liner be placed in the BSL by September 1988. Consequently, wastewater was emptied from the lagoon, and a larger percentage of the process wastewater was discharged from the General Sump directly to MH-175 during the summer of 1987.

Since the Biodenitrification facility was on-line, the FMPC could meet the National Pollutant Discharge Elimination System (NPDES) discharge limits for nitrates 90% of the time. The biosurge lagoon was taken off-line in October 1987 to prepare for the upgrade of the liner. While the biosurge lagoon was out of service, flows were routed to two nearby temporary tanks designed to hold process waste water flows.

LIQUID AND SOLID WASTE DISPOSAL PITS

Several waste disposal pits have been utilized during the course of the operations at the FMPC. These pits were all located near the western boundary of the site, close to Paddy's Run Creek (see Figure B–3). There were six pits in all; three were used for disposal of dry solid wastes only and three were used to dispose of liquid wastes. The largest amount of waste was disposed in Pit 3, a liquid waste disposal pit. The periods of operation of each of the six pits are shown in Figure B–4. More detailed information about the sizes of the pits and their construction is given in Appendix K.

Wastes in the first four of the pits may have contributed to groundwater contamination. Even though three of these were used for disposal of solid waste, the presence of rainwater and collection of snow melt presented opportunities for downward migration of waste materials. The bottoms of Pits 5 and 6 were both lined with rubber and were therefore less likely to contribute to subsurface soil and groundwater contamination.

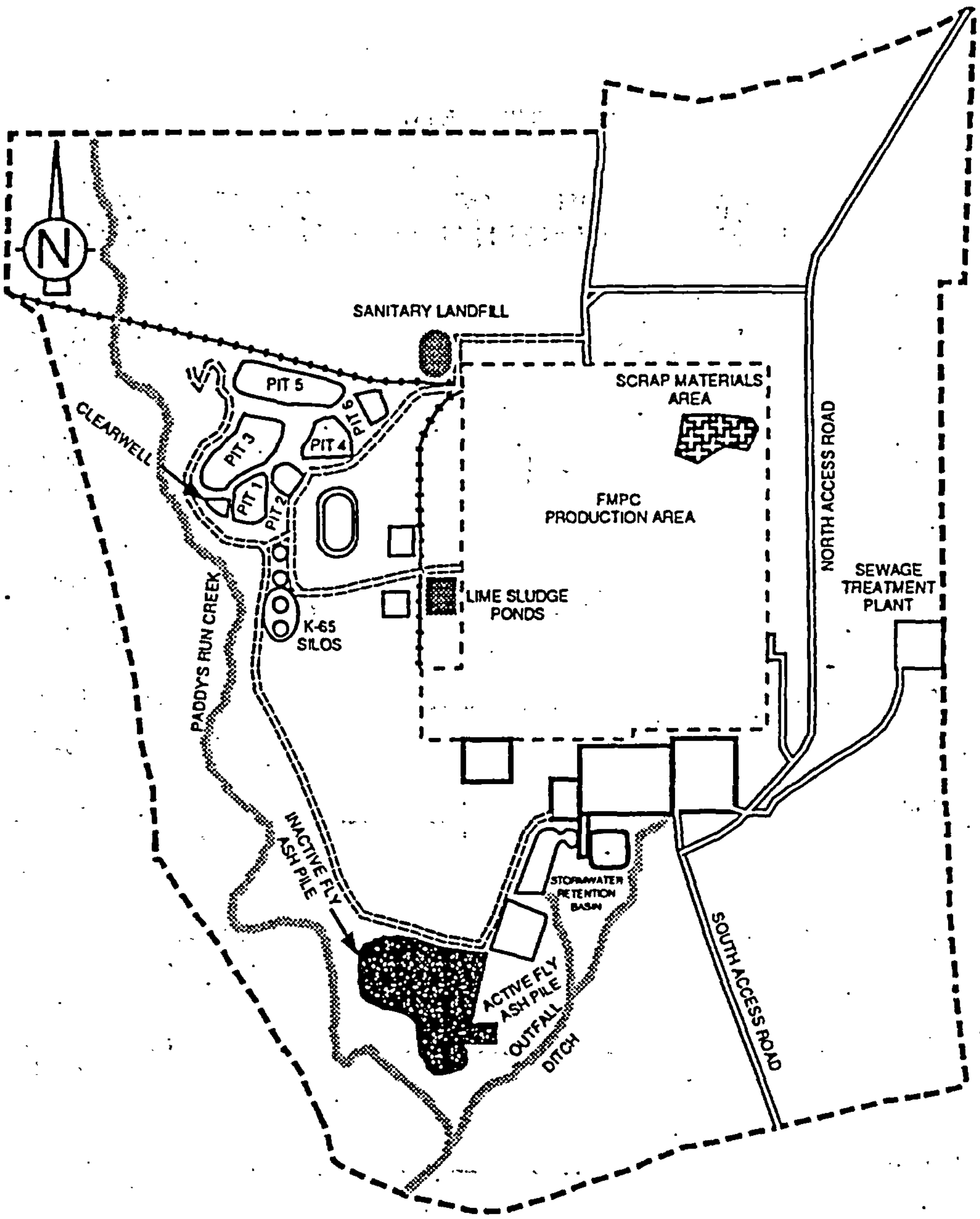

Figure B-3. Diagram of the FMPC showing the waste pits, the active and inactive fly ash pits and scrap materials area.

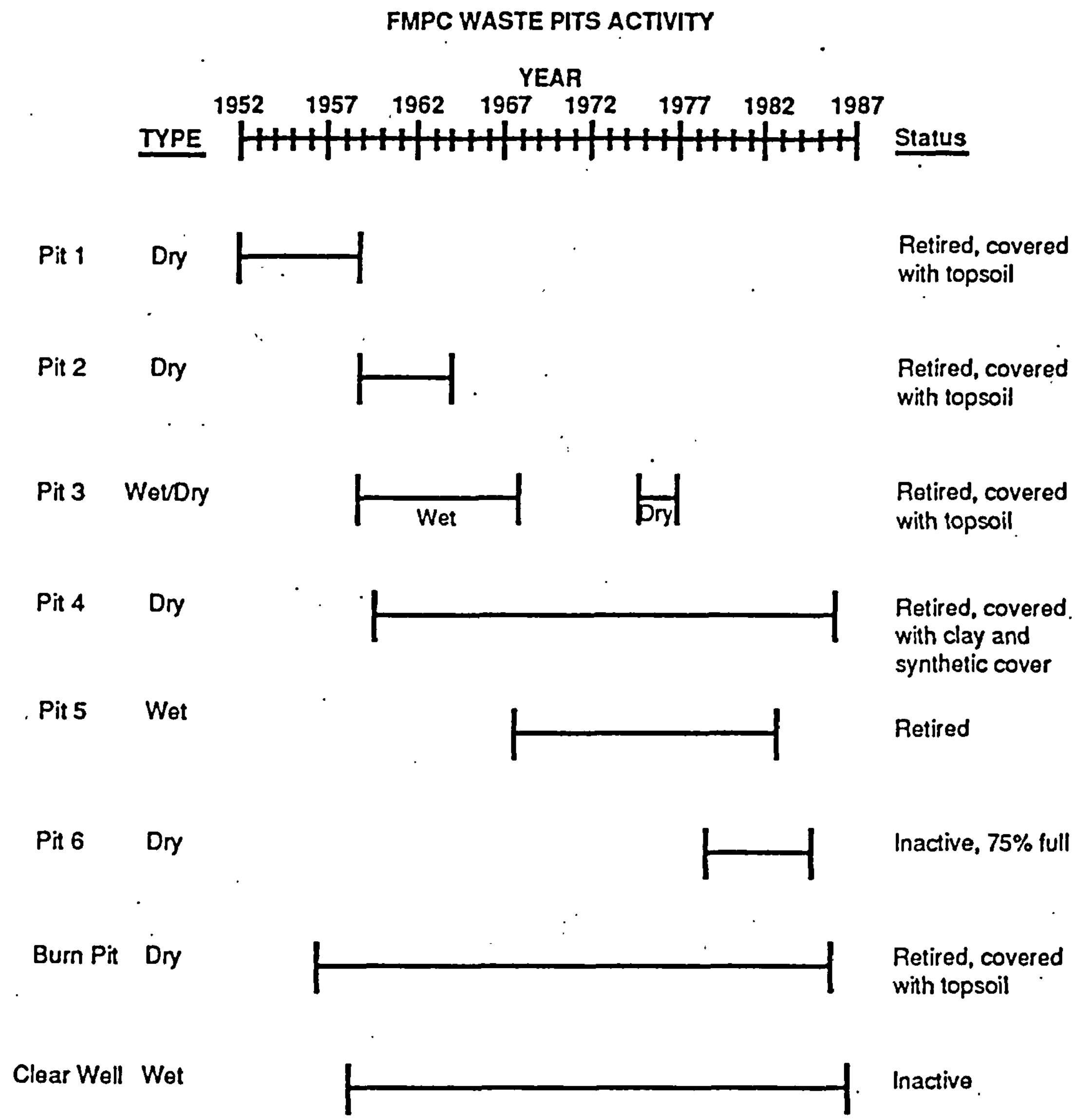

Figure B–4. Historic use and current status of the waste pits, burn pit and clearwell at the FMPC.

GASEOUS WASTE TREATMENT SYSTEMS

The two types of gaseous effluent treatment systems employed at the FMPC were dust collectors and scrubbers. Not all gaseous waste streams were treated by such systems. The incinerator and simple waste burners, building roof exhaust vents, and laboratory hood exhausts are examples of untreated gaseous discharges. Many important process exhausts discharged through dust collectors. Dust collectors employed bag filters to remove airborne particulates from an exhaust stream. A few exhaust streams passed through liquid scrubber systems. Scrubbers employed either acid or caustic solutions to scavenge particles from the air stream being discharged to the atmosphere. Each type of system is discussed below. More detailed information about performance of these systems is given in Appendices E, H, and I.

Dust Collectors

Many of the processes that operated in the FMPC facilities were served by dust collectors. In some cases a single operation, such as an area used for packaging of a solid product, would be ventilated through a collector to remove the airborne dust generated by the packaging process and recover the product material. In other situations, exhaust air from several operations was carried by ventilation ductwork to the dust collector. In some installations, the airborne dust passed through a cyclone separator prior to reaching the dust collector. In the cyclone, particles are removed from the air stream by impaction on the walls as the air flows along a path of circular cross section with constantly decreasing radius.

Although a variety of dust collection equipment was used at the FMPC, the designs shared many common features. Dust collectors contained numerous bag filters through which the air passed before discharge to the environment. The bag filters resembled an array of hollow vertical columns. The filter bags were clamped to supports at both the top and bottom. The collector intake routed the contaminated air into the inside of the columns formed by the filter bags. The air was drawn through the bag material and discharged to the atmosphere by the system's blower. The collector designs included a bag cleaning mechanism that dislodged dust deposited on the inside of the filter bags. The dust fell by gravity into a hopper which was periodically emptied into a drum. The recovered material was then recycled.

The filtration medium used most frequently at the FMPC was virgin wool felt; bags composed of this fabric were manufactured to FMPC specifications. Several different materials were studied and used at various times during the history of particular plants. These materials included other forms of wool felt, polyesters, and most recently Gore-Tex™. Much of the testing of alternative filtration materials was undertaken to try to reduce failures of dust collector bags. Some dust collectors handled exhaust air which was at elevated temperatures and/or contained corrosive compounds, such as HF. Bags deteriorated under such conditions and failures of filter bags were often the reasons for elevated releases (see Appendix E). Such failures could be detected in several ways: by periodic inspection, by the elevated release rate determined by analyzing the effluent sample filter, or by measurements of the pressure drop across the filtration system. In later years, radiation detectors were installed to monitor the filter that collected the effluent sample; the system would alarm when sufficient radioactive material collected on the filter to indicate that an abnormally large release had occurred. When such releases were detected, the exhaust system was shut down and the failed bags were replaced.

Most dust collector exhaust streams were sampled on a continuous basis. Measurements of the amounts of dust collected by the systems were also made when the material was drummed for recycling. The results of some of these operational measurements were reported by Ross and Boback (1971). The data showed that dust collection systems could be highly efficient. Measurements made on four collection systems in Plant 5 between May 1968 and September 1971 yielded estimated efficiencies of greater than 99.9%. However, available data on effluents from dust collectors (Appendix E) show that these systems were not consistently as efficient as they were during the period studied by Ross and Boback (1971).

Scrubber Systems

While all of the FMPC facilities employed dust collectors, the use of scrubber systems for radioactive effluent control was predominantly at two facilities, Plant 2/3 and Plant 8. These systems were used for effluent streams that were corrosive and/or at high temperature. In Plant 2/3, the NO_x fumes driven off during denitration and the airstream from the UO_3 gulping operation were passed through scrubbers that employed a nitric acid solution. The solution became more acidic as the NO_x fumes were collected and was periodically diluted. The nitric acid produced was recycled for use in the Refinery. The uranium collected by the system was also returned to the digestion area. Before entering the scrubber, the particle-laden air stream from the UO_3 transfer operation was passed through a primary cyclone and a secondary cyclone to collect the UO_3 product. The material was stored in a surge hopper prior to grinding and packaging. Appendix H contains more information about the Plant 2/3 scrubbers.

In Plant 8, scrubbers were used to cleanse the exhausts from the rotary kiln, the primary calciner, and various other furnaces. The hot exhaust gases were forced to follow an extended path through the scrub liquor to maximize the contact between the gases and the solution. This arrangement is designed to cool the discharge and to increase the removal of particulates. The scrub liquor for these systems was sodium hydroxide solution. The Plant 8 scrubbers are described in more detail in Appendix I.

Because routine sampling of scrubber exhausts was not performed, periods of elevated releases cannot be identified directly. Measurements of the efficiency of the Plant 8 scrubber systems were performed periodically over the years, beginning in the late 1950s. Those measurements provide some historical evidence of system performance. Measurements of releases from the Plant 2/3 scrubbers were performed only recently. Because the system has changed little over the years, the recent data can be used to estimate releases from that source. The results of the scrubber efficiency evaluations and effluent release measurements are discussed in Appendices H and I.

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APPENDIX C

FMPC PRODUCTION INFORMATION

INTRODUCTION

The FMPC primarily processed uranium and its compounds, but thorium metal and compounds were also produced in relatively small quantities on several occasions. Production information for uranium and thorium provides a guide to the magnitude of FMPC activities over the years and, in the absence of other data, can be used as an aid in estimation of facility releases to the environment. Data on uranium processing by fiscal year are presented in the Annex at the end of this appendix.

Several types of data are presented in this appendix. Information on receipts of material at the FMPC and shipments from it provides a rough indication of production. When available, plant-specific production rates are also presented because they are more useful for estimating releases from specific facilities.

A variable factor during the course of uranium production was the ^{235}U content of the uranium being processed. The concentration of ^{235}U in a sample or batch of uranium is generically referred to as the "enrichment" of the material. Three general categories of uranium enrichment based upon the concentrations of ^{235}U present are defined as follows:

- natural uranium — contains 0.72% ^{235}U; also called "normal" uranium
- depleted uranium — contains significantly less than 0.72% ^{235}U, typically 0.14–0.20% at the FMPC
- enriched uranium — contains more than the natural concentration of ^{235}U, typically 0.95–1.25% at the FMPC

Data on the enrichment of processed uranium are presented using these categories in the figures and tables of this appendix.

While most of the enriched uranium was in the range shown above, some processing of 2% enriched uranium occurred in the 1960s. The capability to digest 5% enriched uranium was added to Plant 1 in 1970.

Some of the uranium received at the FMPC was recycled. That is, it had been recovered from reactor fuel prior to shipment to the FMPC. The enrichment of the recycled material was variable. Processing of the fuel separated the uranium from the bulk of the fission and activation products in the irradiated fuel. However, some of those radionuclides were detectable as contaminants in the uranium. Appendix D contains the results of recent measurements of fission and activation products in materials at the FMPC.

Thorium production at the FMPC was estimated to have been only about 0.4% of the uranium production. Processing was limited to a few facilities and to specific time periods.

URANIUM PRODUCTION

The following assessment of historic uranium production at the FMPC is divided into: (1) gross receipts and shipments of uranium, (2) plant-specific production data, and (3) shipments of specific uranium products.

Gross Receipts and Shipments of Uranium

A general indication of overall plant activity from fiscal year 1952 through fiscal year 1980 is provided by data on the receipt of uranium at the FMPC and the subsequent shipment of uranium products to other locations (FMPC 1988). The information available is generally tabulated on a fiscal year (FY) basis because FMPC budgets followed that schedule. The government's fiscal year changed in 1976 from a July–June to an October–September calendar. In the following data summaries, shipment and production activities during July–September 1976 are included in FY 1976. When making comparisons, it is necessary to remember that fiscal "year" 1976 contains 15 calendar months.

After FY 1980, the accountability system was changed to include onsite transfers between FMPC plants in the total quantities received and shipped. As a consequence, receipts and shipments listed in accountability reports after FY 1980 do not reflect overall FMPC activity and are not included here.

During the fiscal years 1952 through 1980, the FMPC received about 362 thousand metric tons (MT) of uranium and shipped about 358 thousand MT to offsite locations (Audia 1977; FMPC 1988). Approximately 54% of the receipts and shipments were natural uranium, about 20% were enriched uranium, and some 26% were depleted uranium. Table C–1 shows the total receipts and shipments for the three categories of material. Data for individual fiscal years are given in Table C1–1 in the Annex. (It should be noted that some material may have been counted twice, even in these tabulations. For example, billets manufactured at the FMPC were shipped offsite for extrusion, then shipped back for final processing before the finished reactor fuel was sent to the customer.) No detailed time history of the amount of uranium stored onsite at the FMPC has been found.

Table C–1. Quantities (MT) of Uranium Received by and Shipped from the FMPC Between July 1951 and September 1980

Activity	Natural	Enriched	Depleted
Receipts	1.94×10^5	7.14×10^4	9.65×10^4
Shipments	1.95×10^5	6.95×10^4	9.35×10^4

The distributions of uranium receipts and shipments among the three uranium enrichment categories were not constant with time. Figure C–1 shows the fractions of the uranium receipts that were natural, enriched, and depleted uranium during each of the fiscal years. Most of the uranium received at the FMPC during the first decade of operation was natural uranium. Some significant quantities of depleted uranium were received during FY 1955 and FY 1956. Enriched uranium receipts did not exceed 10% of the total until 1961, but then rose steadily until 1966. Following that time, the material received was a highly variable mixture of the three uranium enrichment categories.

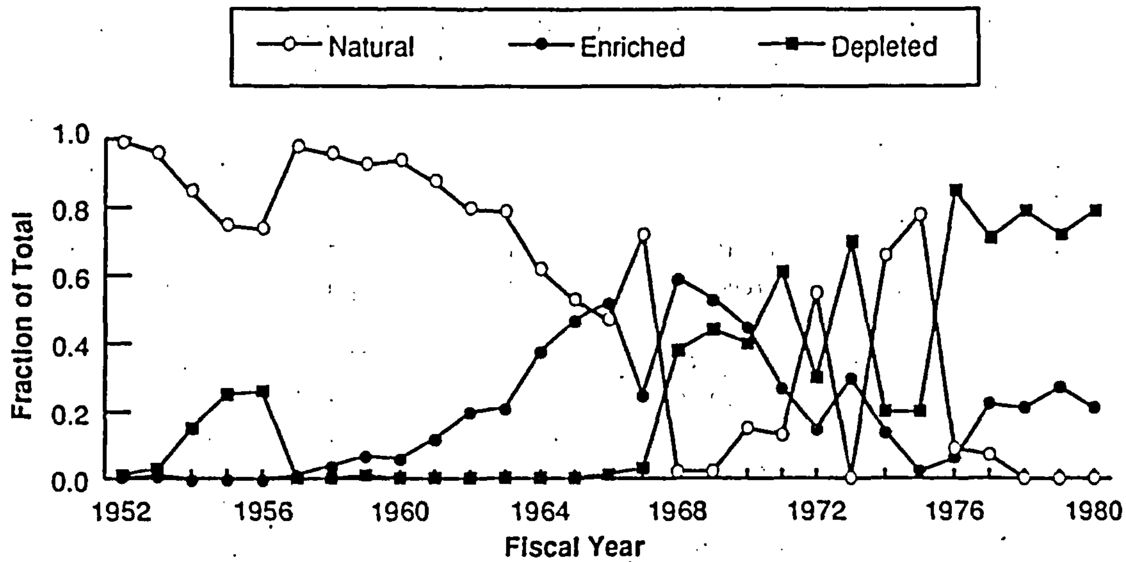

Figure C-1. FMPC uranium receipts during FY 1952–1980 segregated according to ^{235}U content.

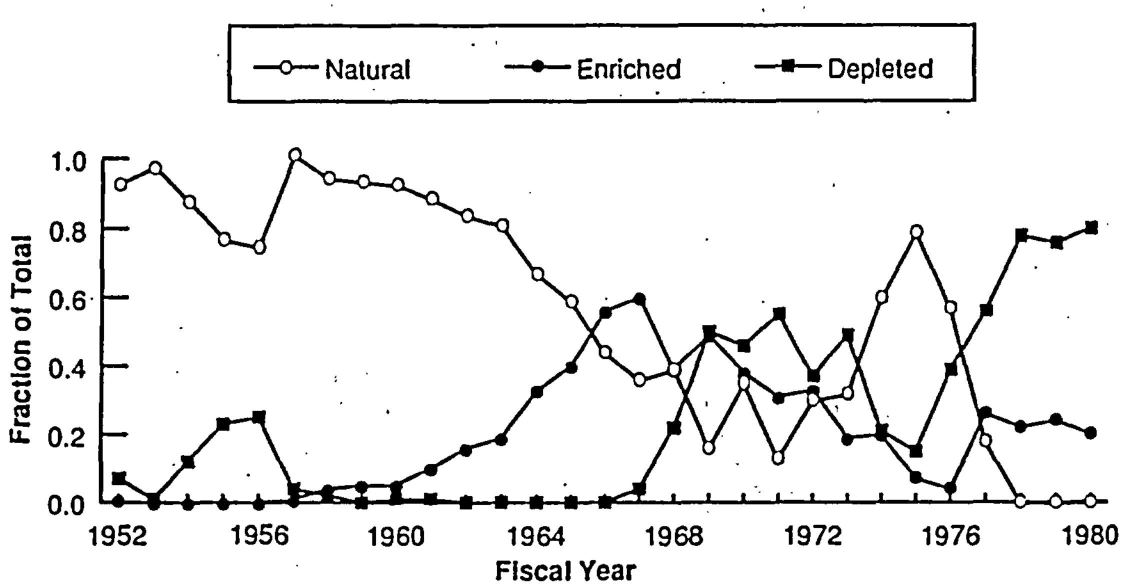

Figure C-2. FMPC uranium shipments during FY 1952–1980 segregated according to ^{235}U content.

Uranium shipments from the FMPC (Figure C-2) tended to follow the pattern of receipts during most of the first 29 years of operation. This was particularly true for enriched uranium for the twenty years between 1955 and 1974; in only three of those years was the difference between the two quantities more than 30%. The ratios of shipments to receipts for depleted uranium were highly variable between 1957 and 1967, but were more consistent before and after that time. For natural uranium, the quantity shipped was always within 30%, and usually within 20%, of the amount received between 1953 and 1966. After that time, there was much greater variability as the inventory that had been built up was converted to finished products and shipped. Comparisons of the data on receipts and shipments indicate that material was received, processing occurred, and products were shipped on a fairly regular schedule.

Figure C–3 is a plot of the total annual shipments, in metric tons of uranium (MTU), of all three categories of uranium. It is an indication of the magnitude of plant operations during the first 29 years of operation. The amount shipped during 1952, about 160 MTU, is not shown clearly in the figure. While the plot gives an idea of the overall site activity for the entire FMPC, it does not address the operation of specific processes. Data for individual plant operations are given in the next section.

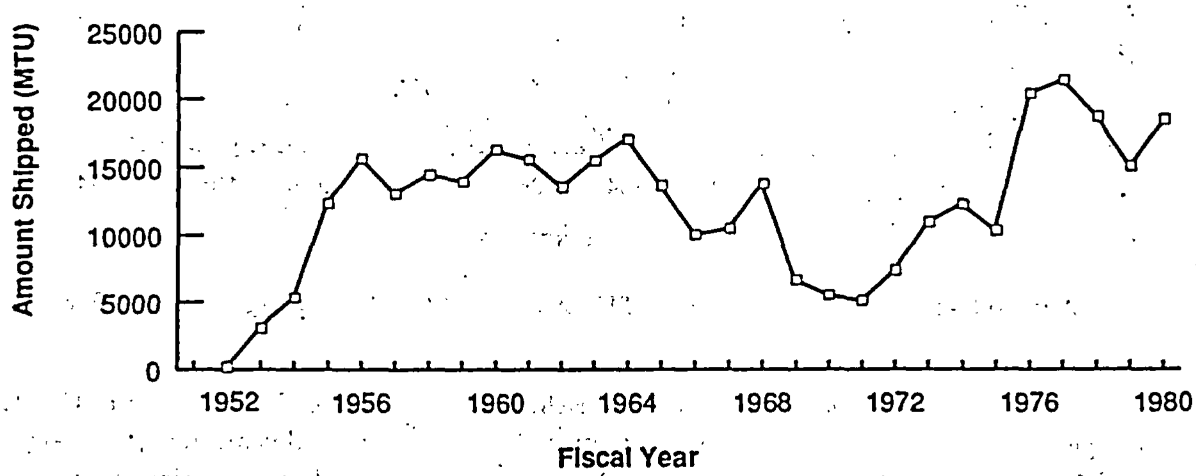

Figure C–3. Total amounts (MTU) of uranium shipped from the FMPC during FY 1952–1980.

Plant-Specific Uranium Production Data

Operation of the various facilities at the FMPC varied with time. Processing rates were increased or reduced because of changes in the demand for intermediate materials and finished metal products. Data on specific material production rates are more directly related to radionuclide releases from individual facilities. The data on uranium processing listed in the Annex and summarized in figures in this section come from several different sources at the FMPC (Audia 1977; Dunaway 1993; FMPC 1988; Rathgens 1985). The plots shown below illustrate the variations in operational histories of specific FMPC facilities.

Plant 2/3. Figure C-4 contains the production rates for uranium trioxide (UO_3) during the fiscal years 1952 through 1988. The plot shows two periods when the annual production of UO_3 exceeded 4000 MTU as well as extended periods of lower production. Data for individual fiscal years are given in Table C1-2 in the Annex. These production rates are important for estimation of releases from the UO_3 vacuum transfer and packaging activities.

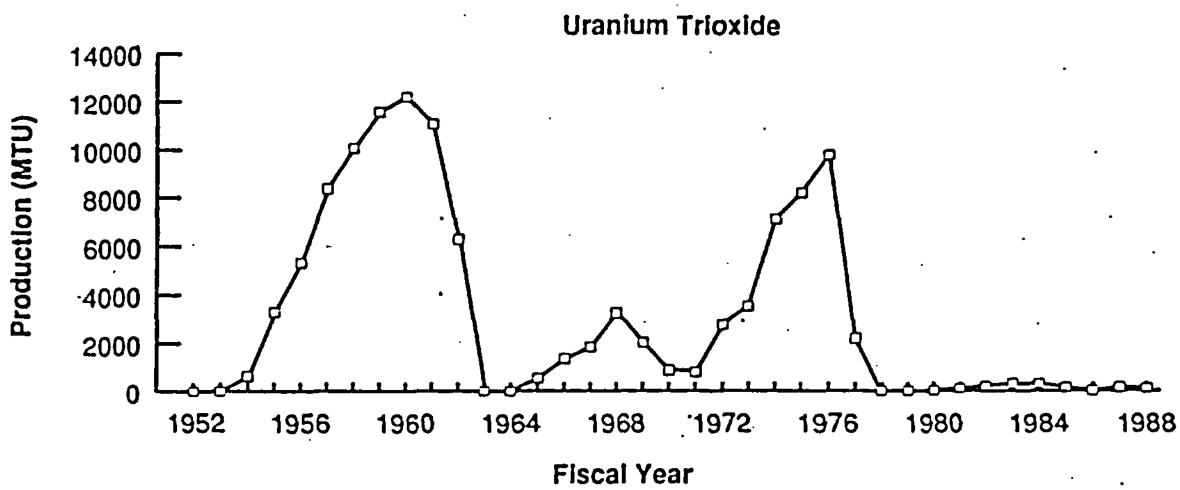

Figure C-4. Annual production (MTU) of UO_3 in Plant 2/3 during FY 1953-1988.

Data on the distribution of the UO_3 by enrichment category are shown in Figure C-5 for the period FY 1953-88. There was no production of UO_3 during fiscal years 1953, 1963-1964, and 1978-1980. During FY 1965, when UO_3 production resumed, only enriched uranium was processed. The fraction that was enriched exceeded 0.5 between FY 1965 and FY 1970, but then declined rapidly. The only identified UO_3 production from depleted uranium was 41 MTU in FY 1970, 4.7% of the total production for that year. This fraction is not shown in Figure C-5, but is reflected in the data shown for that year. Of the approximately 111,000 MTU converted to UO_3 during FY 1952-1976, nearly 94% was natural uranium.

Plant 4. Figure C-6 contains data on production of green salt (UF_4) in Plant 4. The highest annual production, greater than 12000 MTU, occurred in FY 1958. In each of the fiscal years from 1957 to 1963, UF_4 production exceeded 9000 MTU. Production declined steadily during subsequent years. During the period FY 1971-1988, the production of UF_4 was less than 1250 MTU. Data for individual fiscal years are given in Table C1-2 in the Annex. Comparing the UF_4 and UO_3 production curves (Figures C-6 and C-4, respectively) suggests that an offsite source provided some UO_3 feed for Plant 4 prior to FY 1959 and a substantial amount of the feedstock between FY 1963 and FY 1968.

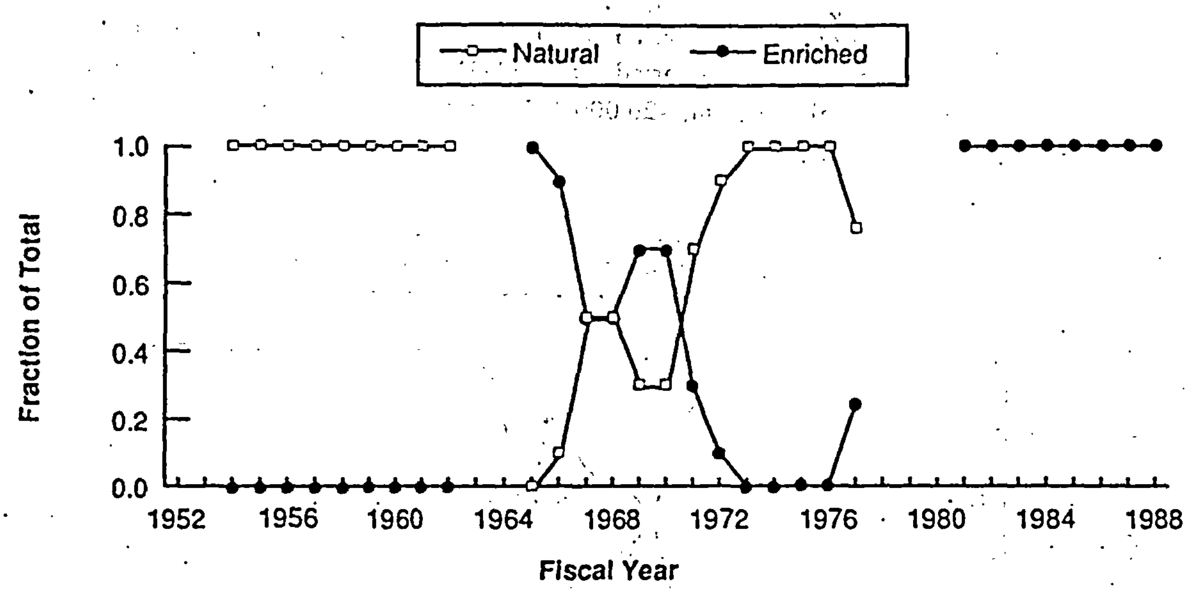

Figure C–5. Fractions of the UO_3 production from natural and enriched uranium during FY 1953–1988.

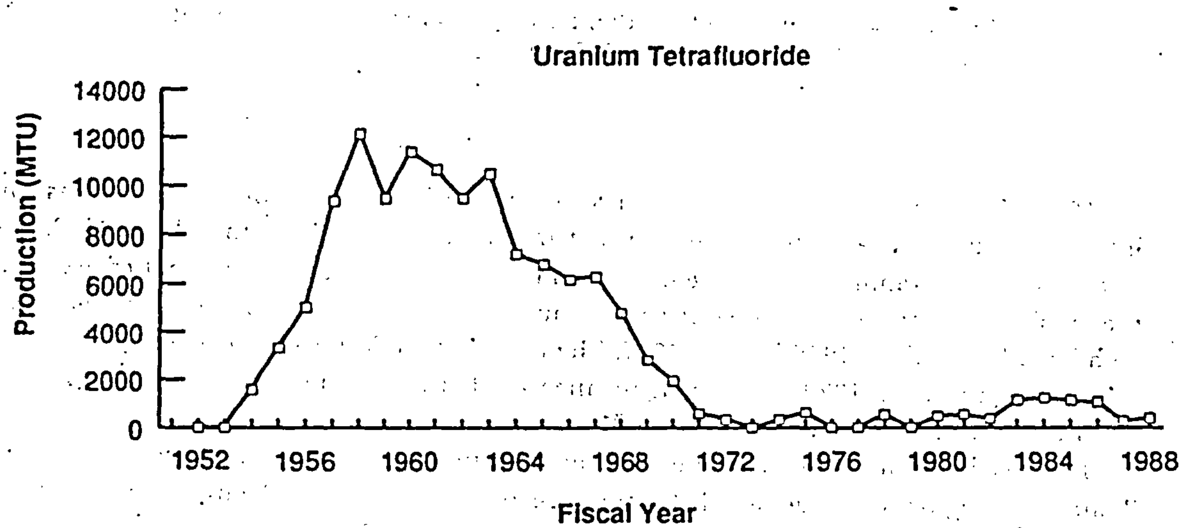

Figure C–6. Annual production (MTU) of UF_4 in Plant 4 during FY 1953–1988.

The fractions of the UF_4 production from natural and enriched uranium are shown in Figure C–7. Before FY 1963, all the UF_4 was produced from natural uranium. After FY 1962, enriched UF_4 production began to increase. Enriched uranium was the dominant material for UF_4 production in the years following FY 1966. Depleted UF_4 (not shown in the figure) was produced in only one year, FY 1970. It accounted for almost 20% of the total production in that year. During the period FY 1952–1976, UF_4 production in Plant 4 was ~89,000 MTU from natural uranium, ~20,000 MTU from enriched uranium, and only ~340 MTU from depleted uranium.

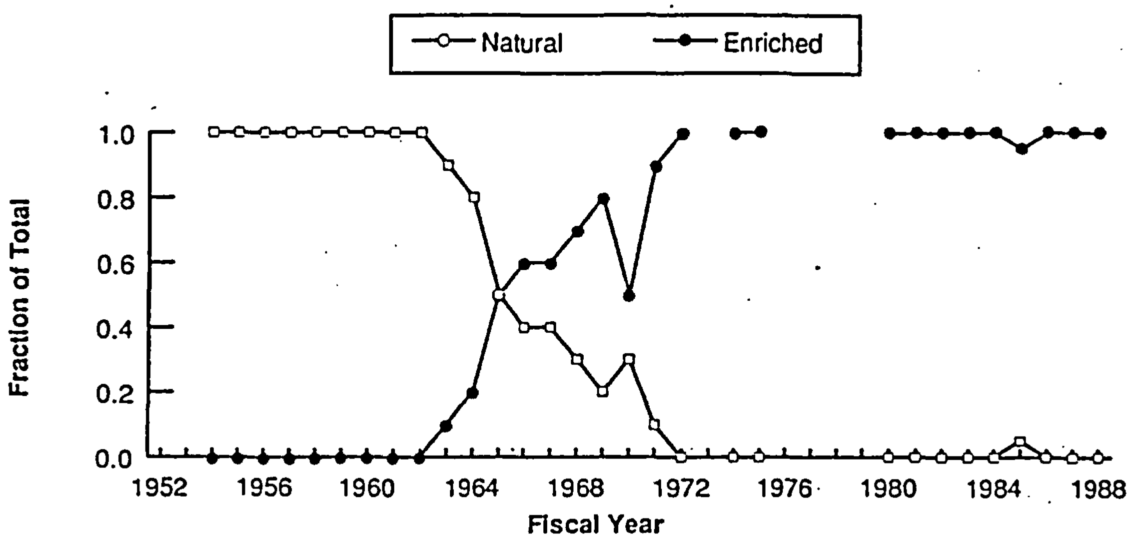

Figure C–7. Fractions of the Plant 4 UF_4 production from natural and enriched uranium during FY 1953–1988.

Plant 5. Figure C–8 contains the data on production of uranium metal derbies and ingots in Plant 5. The derby production plot indicates the amounts, in MTU, of UF_4 that were reduced to uranium metal. The ingot production data show the throughput of the metal remelt furnaces and casting operations. Annual production for both processes was at a high level between FY 1956 and FY 1967. Then production declined to a relatively constant lower level until FY 1979 when it again increased. Data for individual fiscal years are given in the Annex in Tables C1–3 and C1–4.

The distribution of derby production by uranium enrichment category is shown for Plant 5 in Figure C–9. Depleted uranium derbies were produced only after FY 1964. Production of derbies from enriched uranium began in FY 1958.

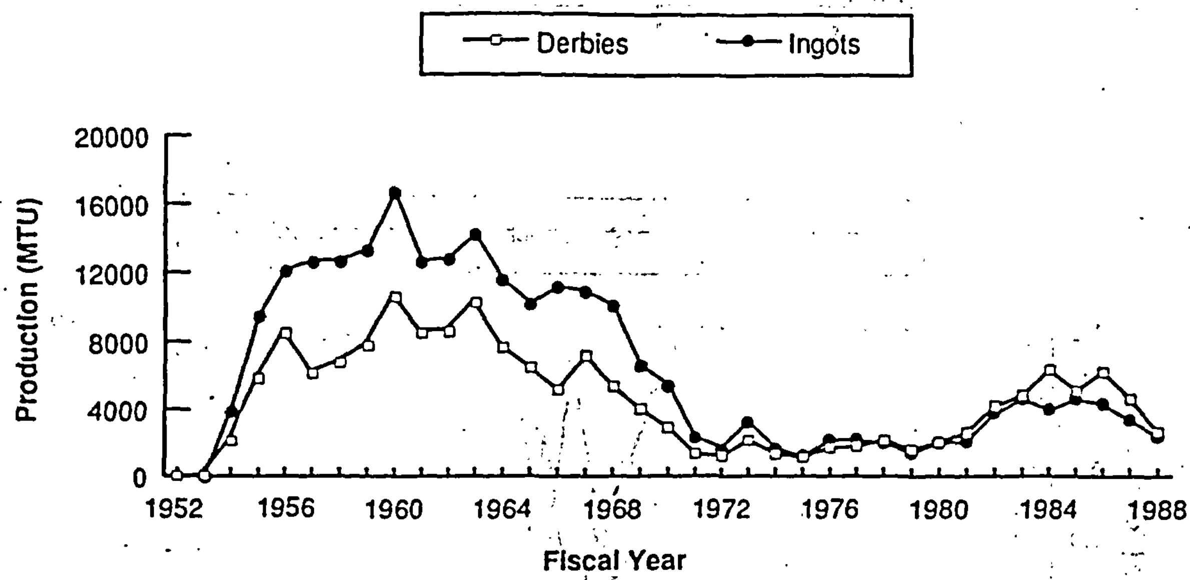

Figure C–8. Annual production (MTU) of uranium metal derbies and ingots in Plant 5 during FY 1953–1988.

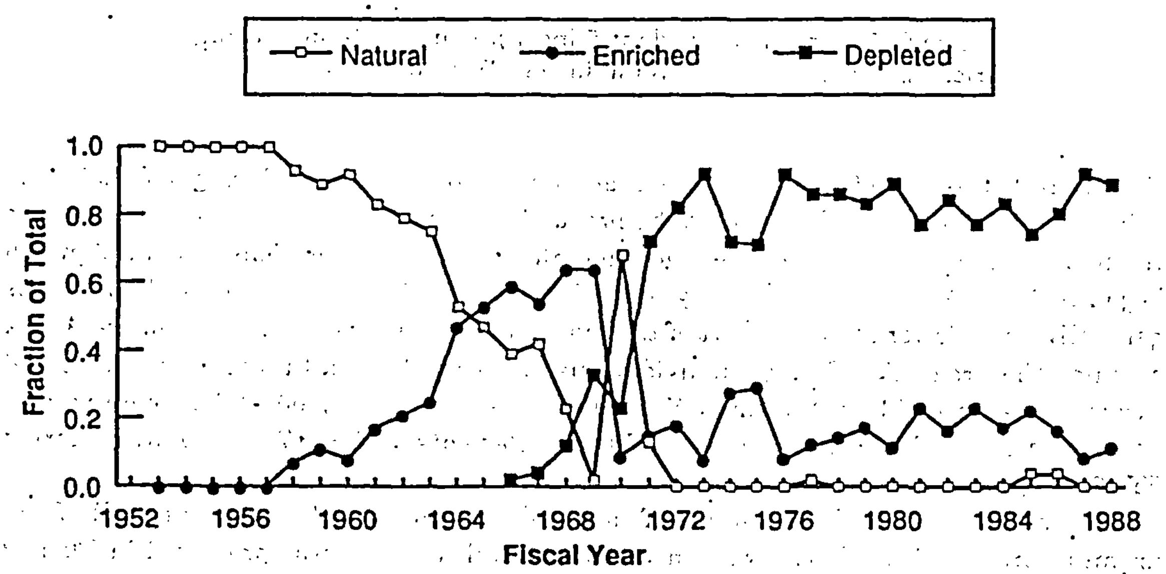

Figure C–9. Fractions of Plant 5 metal derby production from natural, enriched, and depleted uranium during FY 1953–1988.

The prevalence of natural uranium use in the early years of ingot production in Plant 5 is shown in Figure C–10. It was used exclusively until FY 1966. Casting of enriched uranium ingots in Plant 5 occurred between FY 1965 and FY 1969. In FY 1967, enriched uranium ingots accounted for about 30% of the total production. Depleted uranium was the principal form in FY 1969 and FY 1971–1988.

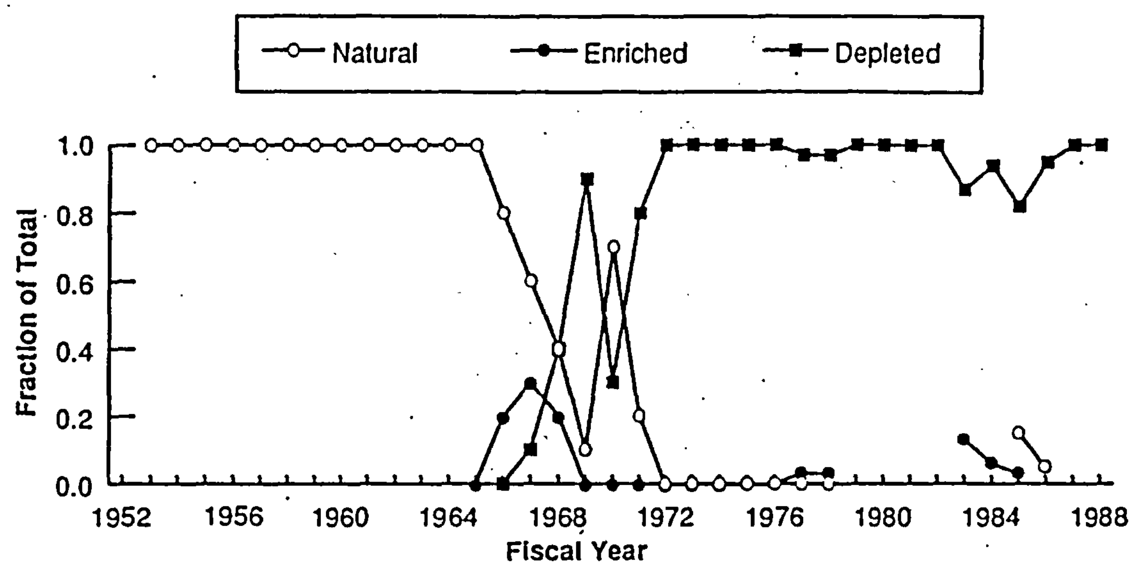

Figure C-10. Fractions of Plant 5 ingot production from natural, enriched, and depleted uranium during FY 1953–1988.

Plant 9. Casting of ingots was also accomplished in Plant 9 during FY 1958 and subsequent years. Figure C–11 shows the production data for that operation. Also shown in the figure is the throughput for the ingot and billet machining operations in Plant 9, which began in 1966. After peaking in the years FY 1964–1965, ingot production in Plant 9 was generally much lower after the start of the machining operations. Table C1–5 in the Annex contains the production data for individual fiscal years. The total production of core and target elements in Plant 9 during FY 1958–1971 was about 22,000 MTU. There was no production of enriched uranium cores and target elements during FY 1953–1957 or during FY 1972–1976.

In contrast with the ^{235}U content of materials handled in Plant 5, nearly all of the uranium processed in Plant 9 was enriched. Natural uranium was processed in Plant 9 only in FY 1973 and accounted for just 3 percent of the throughput during that year. Reactor cores and target elements produced in Plant 9 were also composed of enriched uranium.

Plant 6. Production data for Plant 6 are shown in two curves in Figure C–12. The first of these is the production of rods by the rolling mill. That operation began in FY 1953 and continued until FY 1971. The second curve shows the annual production of machined fuel elements. This activity also declined after FY 1964, but continued at a greatly reduced rate between FY 1971 and FY 1988. Data on production during individual fiscal years are shown in Table C1–5 in the Annex.

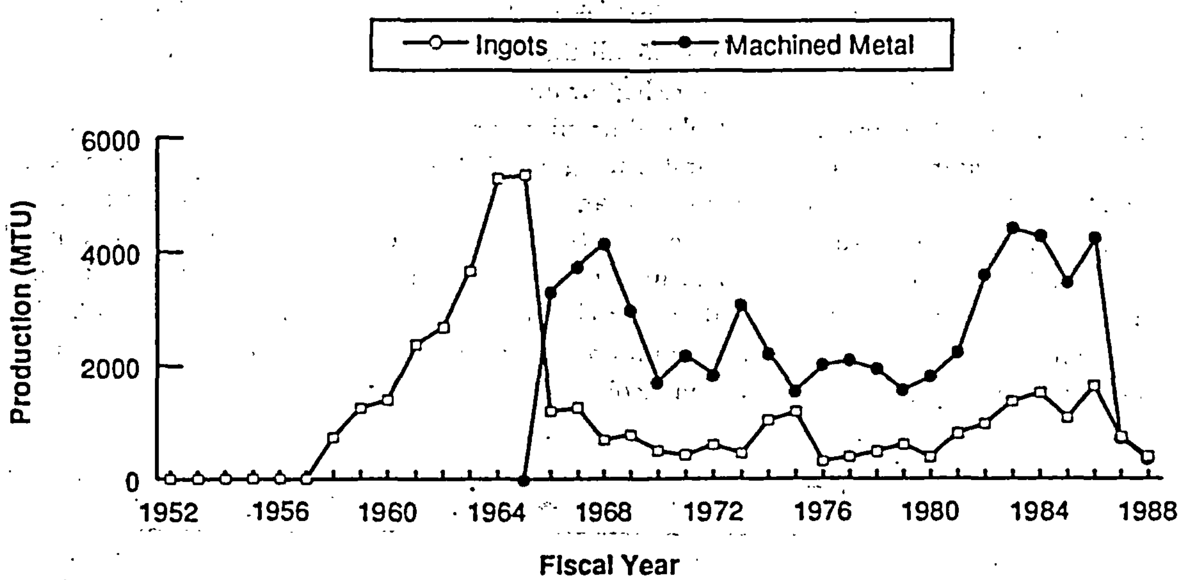

Figure C-11. Annual production (MTU) of uranium ingots and machined metal products in Plant 9 during FY 1952–1988.

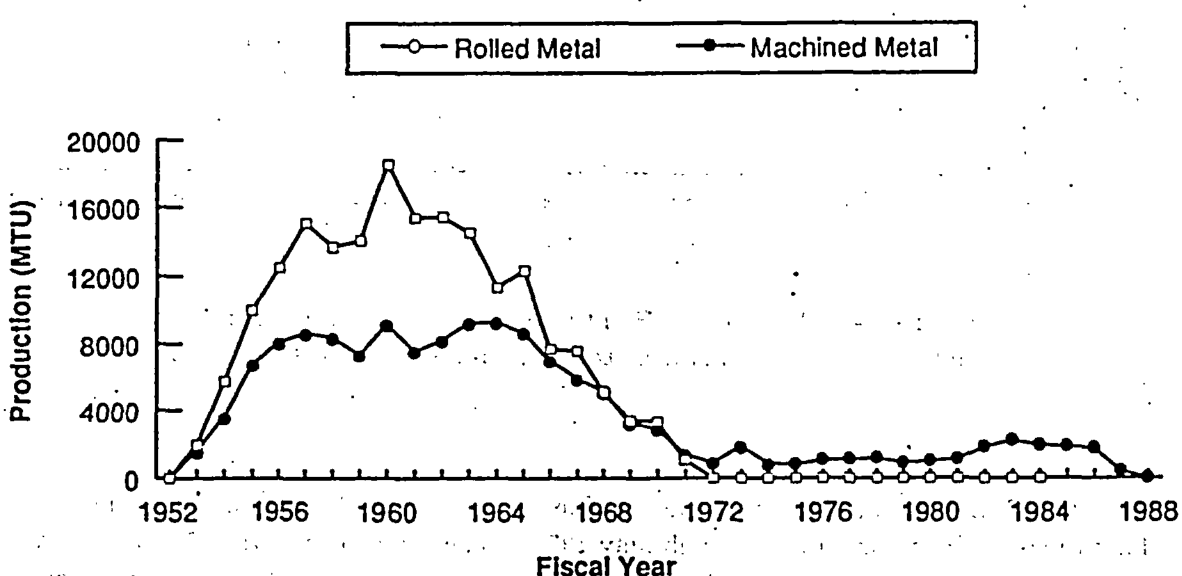

Figure C–12. Annual production (MTU) of rolled and machined metal products in Plant 6 during FY 1953–1988.

Fractional distributions of the products from Plant 6 according to their ^{235}U content are shown in Figure C-13. Prior to FY 1965, all the products were manufactured using natural uranium. During FY 1966–1968, enriched uranium was used for 15–31% of the cores and targets produced. Production of depleted uranium products began in FY 1967 and was the dominant material in FY 1969 and during FY 1971–1976. Table C1–6 in the Annex contains the data for individual fiscal years. This distribution of materials is similar to that for ingots produced in Plant 5 during the same period (Figure C–10). Approximately 107,000 MTU of core and target elements were produced in Plant 6 between FY 1952 and FY 1976. More than 88% of the total was produced from natural uranium; about 9% of the elements were made of depleted uranium. Distribution by enrichment category is not presently available for years beyond 1976.

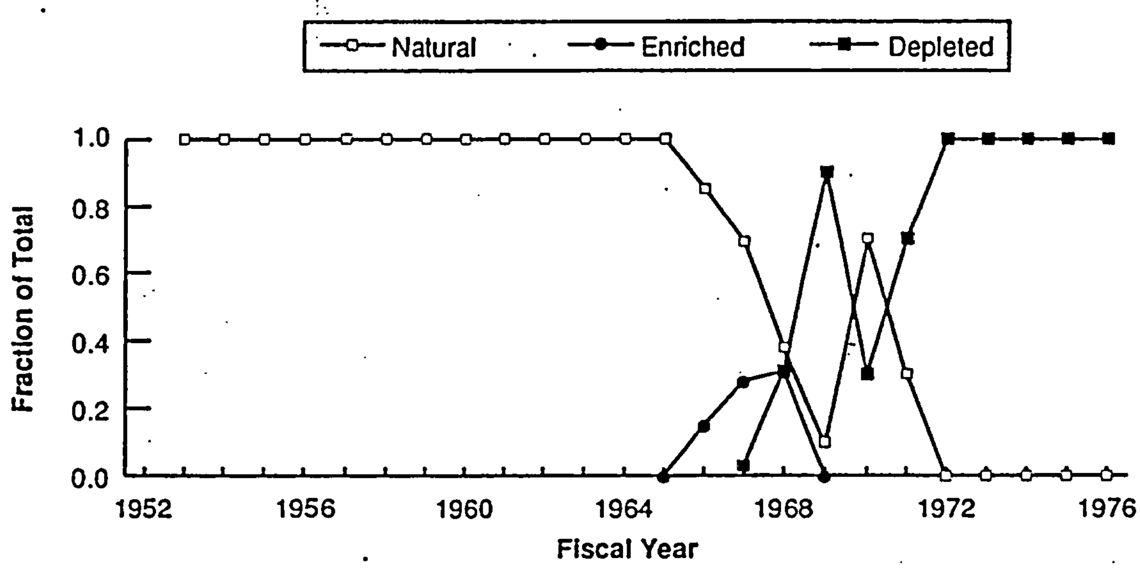

Figure C–13. Fractions of Plant 6 core and target production from natural, enriched, and depleted uranium during FY 1953–1976.

Plant 7. Uranium tetrafluoride was produced by reacting UF_6 with hydrogen in Plant 7 for two years, from June 1954 through May 1956. Its design capacity was 12 tons per day of uranium as UF_4, when six of eight reactors were operating. Depleted and natural uranium products were produced; each bank of four reactors included a spare. Little information on actual production has been found. During January–May 1956 the normal uranium bank of reactors produced 656 MTU of UF_4. During January–April of the same year, the bank handling depleted uranium produced 1114 MTU of UF_4. It may be presumed that all four reactors may have been operating or that three reactors operated well above the initial design capacity during this four-month period. The limited monthly data are given in the Annex.

Plant 8. The recovery of uranium from scrap material in various forms was accomplished in Plant 8. The recovered uranium then became feed for the Plant 2/3 Refinery. Figure C–14 shows the amounts (MTU) of uranium recovery by Plant 8 by fiscal year. During FY 1955–1969, uranium recovery exceeded 1000 MTU per year. During eight years of operation, uranium recovery exceeded 2000 MTU per year. Uranium recovery data for individual fiscal years are given in Table C1–7 in the Annex.

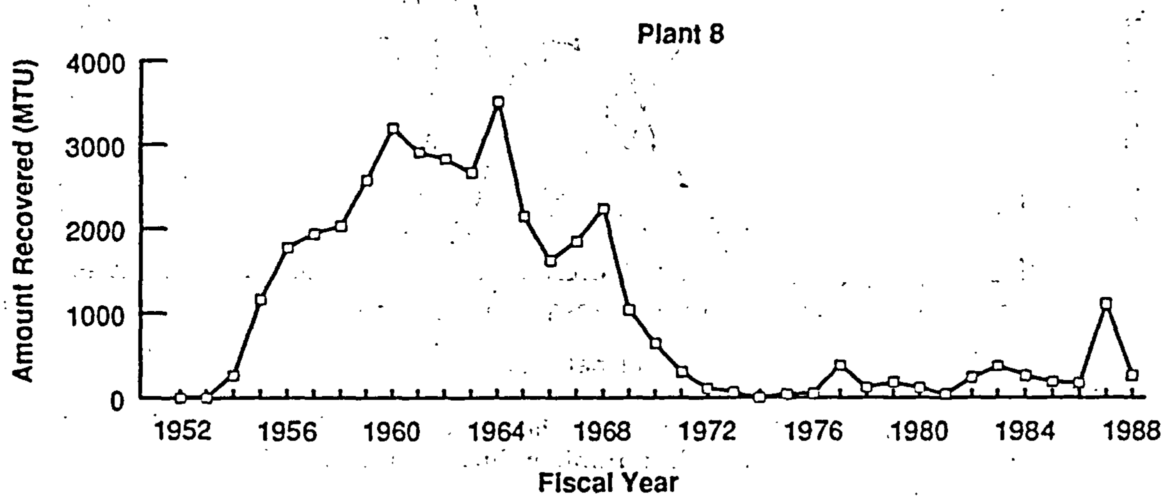

Figure C–14. Annual uranium recovery (MTU) from scrap during FY 1953–1988

Figure C–15 contains the fractions of the total uranium values recovered that were natural and enriched uranium through FY 1988. During the first nine years of operation, natural uranium was the only material recovered. Processing of scrap containing enriched uranium began in 1963. The fraction of the uranium recovered that was enriched was greater than 0.5 during nine of the eleven years during the period FY 1966–1976. A total of ~38,000 MTU was recovered through FY 1988. Approximately 78% of the uranium recovered through FY 1976 was natural uranium. There is currently no information concerning distribution by enrichment category for years from 1977 through 1984.

Depleted uranium comprised less than 0.2% of the total amount of uranium recovered by Plant 8 during the period FY 1953 through FY 1976. For the four years, 1985 through 1988, however, the percentage of depleted uranium was 21, 19, 80, and 69%, respectively (Table C1–7).

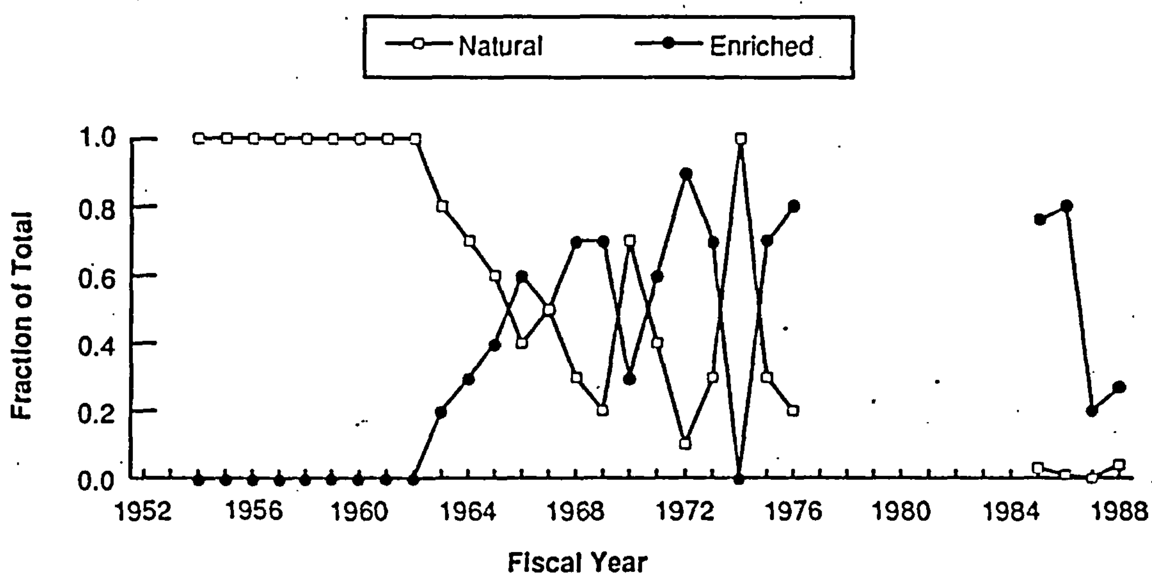

Figure C–15. Fractions of material recovered by Plant 8 that were natural or enriched uranium during FY 1952–1988.

Pilot Plant. The FMPC Pilot Plant was used for production activities as well as process development and testing. Detailed data for the complete range of production campaigns have not yet been uncovered. However, data are available on the production of UF_4 from UF_6 in the Pilot Plant. Annual production figures are plotted in Figure C–16. The figure shows an increase to the peak production of about 3500 MTU in 1964 and the subsequent equally rapid decline in production in later years. Data for UF_4 production during individual fiscal years are shown in Table C1–8 in the Annex. Information about the small quantities produced during the early years (1953-1956) came from Davis (1956). There was no UF_4 production during the years 1968 through 1984, but production was restarted in 1985.

Much of the UF_6 to UF_4 conversion was performed using enriched uranium. The distribution of natural and enriched uranium employed for UF_4 production in the PilotPlant is shown in Figure C–17. Initial production was primarily from enriched UF_6, but production was about evenly divided between the two forms between FY 1962 to FY 1965. Enriched uranium was then used almost exclusively as the feedstock during 1967 UF_4 production. Depleted uranium was used for UF_4 produced during 1986, 1987, and 1988. It accounted for 53, 70, and 100% of total production in those years.

Detailed data on the enrichment of uranium employed in other work at the Pilot Plant has not been uncovered. The record of shipments of uranium to the FMPC indicates clearly that the earliest operations would have employed natural uranium. However, the material usage in later years may have been quite variable.

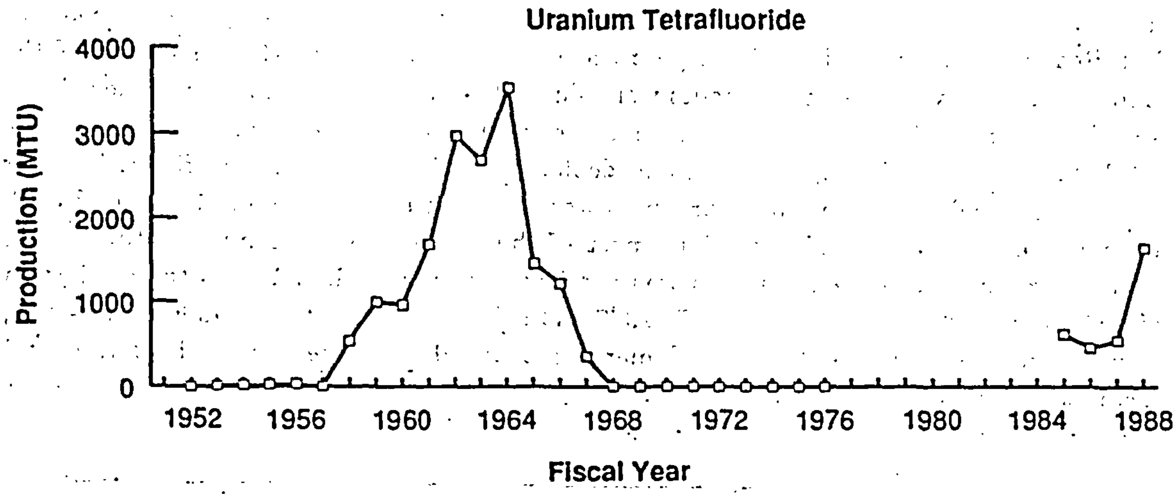

Figure C–16. Annual UF_4 production (MTU) in the Pilot Plant during FY 1952–1988.

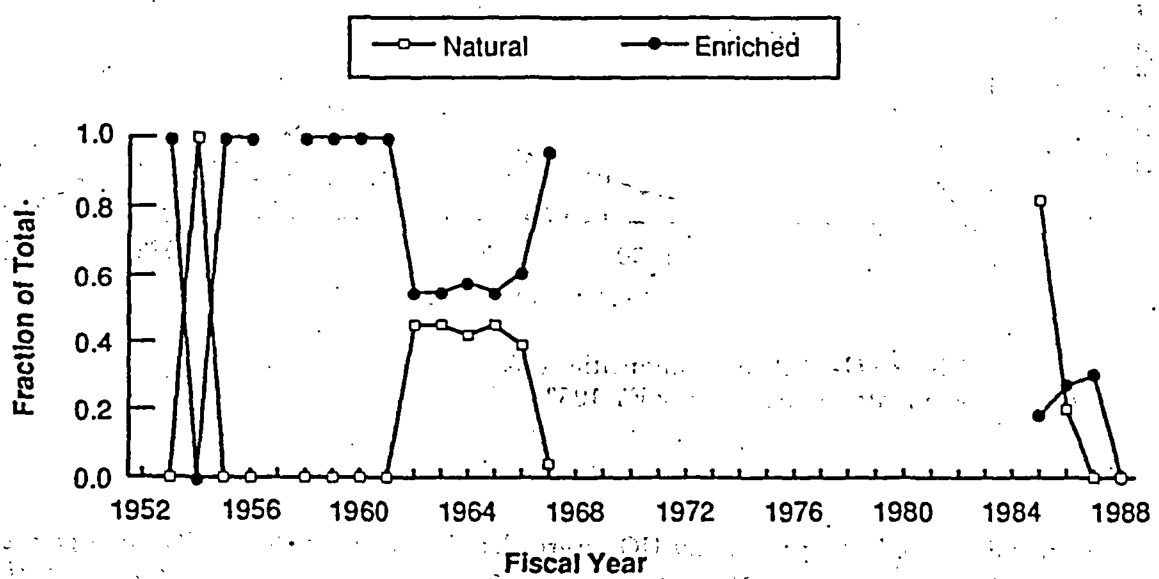

Figure C–17. Fractions of Pilot Plant UF_4 production using natural or enriched uranium during FY 1953–1988.

Shipments of Specific Uranium Products

A compilation of historic data on specific products (Rathgens 1985) provides additional information on the manufacture of products composed of uranium of varying ^{235}U content. Detailed data for individual fiscal years are given in Tables C1–9 and C1–10 of the Annex. Fuel cores and target elements were shipped to both the Richland Operation (RLO) and the Savannah River Plant (SRP) for reactors in operation there. Figure C–18 shows the total shipments of natural, enriched, and depleted uranium, in several configurations, for FY 1952 through FY 1976. These products were finished in Plants 6 and 9 at the FMPC. During the first decade of operation, most of these products were manufactured from natural uranium. Enriched uranium was employed in greater than 10% of the production of fuel and target elements between FY 1967 and FY 1971. During the years FY 1966–1969, enriched uranium fuel and target elements accounted for more than half of the total quantities shipped.

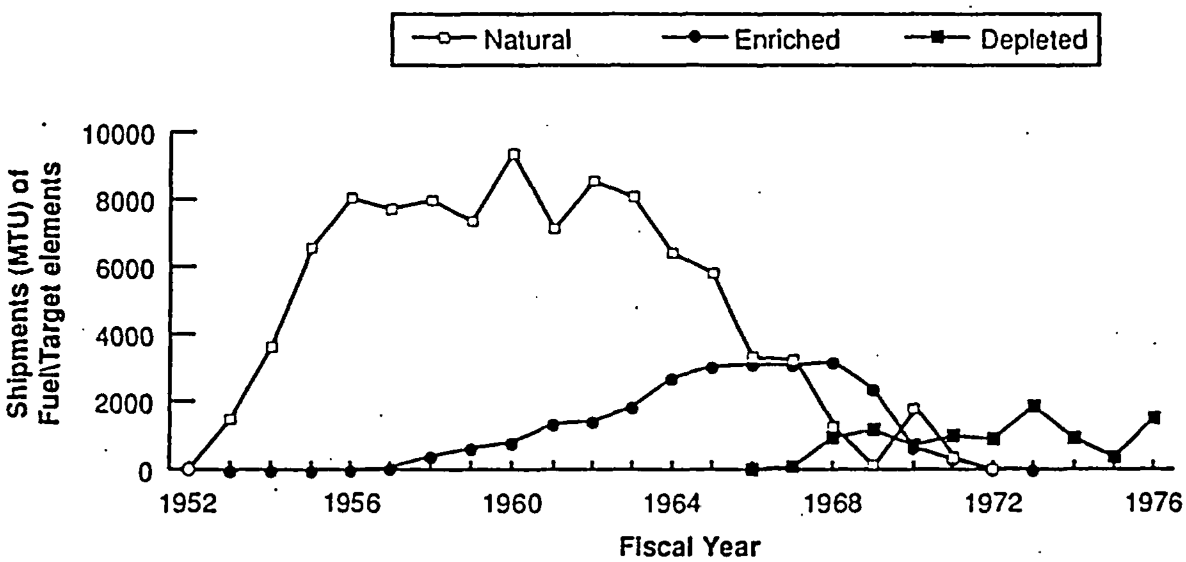

Figure C–18. Annual quantities (MTU) of fuel and target elements shipped during FY 1953–1976.

Intermediate products, principally UO_3, were also shipped from the FMPC. Data on the quantities shipped between FY 1971 and FY 1976 have been located. Figure C–19 summarizes the amounts and timing of the shipments of the two most important intermediate products from FMPC. Shipments of uranium trioxide were by far the largest of any intermediate products. Most of the UO_3 was sent to the Paducah, KY plant but a small amount was sent to Allied Gulf Nuclear Services in South Carolina. About 97% of the UO_3 shipped was natural uranium. The next largest category of intermediate product shipments was ingots of 0.95% enriched uranium. These ingots were sent to RMI

Company, Inc. in Ashtabula, Ohio for preparation of fuel elements for the RLO. The amounts shipped were in the range of 300–1000 MTU during the 6-year period. Depleted derbies and metal pieces were shipped to the Y-12 Plant in Tennessee, the Rocky Flats Plant in Colorado, and the Idaho National Engineering Laboratory. Enriched U_3O_8 and UF_4 were shipped to Goodyear Atomic during the period, but the amounts were small, less than 30 MTU.

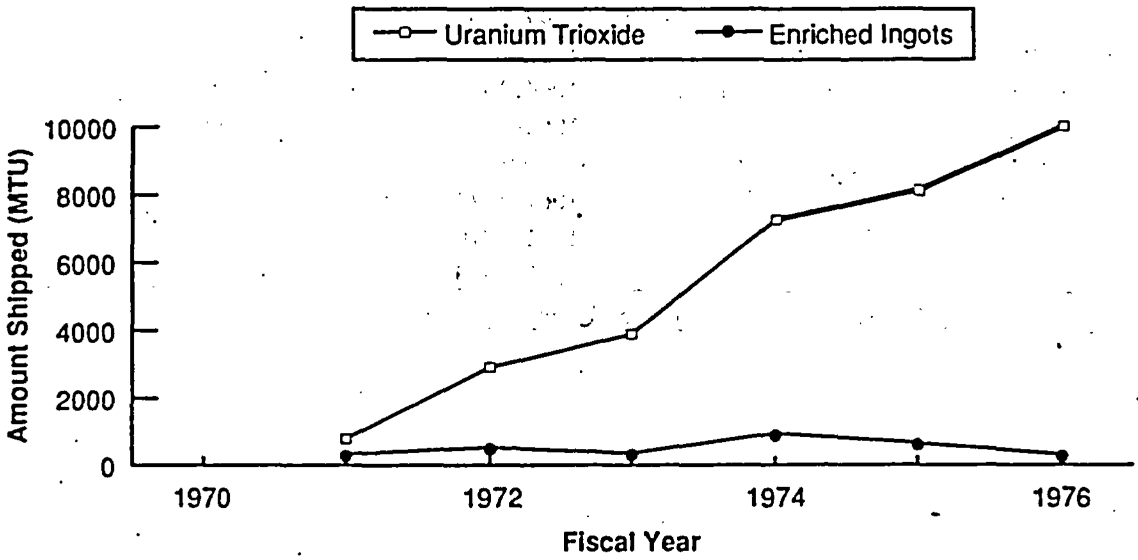

Figure C–19. Annual quantities (MTU) of intermediate products shipped during FY 1971–1976.

THORIUM PRODUCTION

Table C–2 lists the FMPC plants which processed significant quantities of thorium, the chemical form produced, the time period during which the processing was carried out, and the total quantity produced expressed as metric tons (MT) of thorium metal. There has been no thorium processing at the FMPC in more than 10 years, and most of the thorium processing equipment has been removed. Very little information is now available about the equipment which had been used, possibly because many of the records concerning thorium processing were destroyed in the early 1970s. Most of the information listed in Table C–2 came from an addendum (Hill and Dolan 1988; Clark et. al. 1989) to the FMPC radionuclide discharge report (Boback et al. 1987). Some of the information in the table came from production records located by Hill and Dolan which had not been destroyed in the early 1970s. These authors estimated other production information from interviews with past and present FMPC employees, product volume, and yield information.

Table C–2. Thorium Processing in FMPC Plants

Plant	Product	Period	Quantity (MT Th)
2/3	Testing	1968	None
4	Fluoride	1954	460[a]
8	Hydroxide	1966	59
8	Oxalate	1969–1971	310
9	Metal	1954–1955	380
9	Briquetting	1954–1955	76
Pilot	Extraction	1964–1980	790
Pilot	Gel	1964–1970	689
Pilot	TNT Crystals	1966	0.4
Pilot	Metal	1967–1971	51
Pilot	Oxalate	1971–1976	153
Pilot	Gel	1977–1979	350

[a] Assuming this production supported the metal production in Plant 9.

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ANNEX TO APPENDIX C— DATA TABULATIONS

RECEIPTS AND SHIPMENTS

Table C1–1 contains the data on receipts and shipments of natural, enriched, and depleted uranium for the fiscal years from 1952 through 1980. These data form the basis for Figures C–1, C–2, and C–3.

Table C1–1. Receipts and Shipments of Uranium at the FMPC by Fiscal Year

Fiscal	Amounts received (MTU)			Amounts shipped (MTU)		
Year	Natural	Enriched	Depleted	Natural	Enriched	Depleted
1952	861	0	11	148	0	11
1953	3,221	19	106	2,993	12	43
1954	6,015	2	1,034	4,700	4	620
1955	9,124	40	2,996	9,458	30	2,837
1956	12,141	33	4,254	11,681	34	3,896
1957	13,699	179	40	13,246	91	526
1958	13,837	579	41	13,625	514	221
1959	14,740	1,086	97	13,040	760	64
1960	16,125	995	41	15,136	876	217
1961	13,744	1,825	11	13,802	1,501	172
1962	10,715	2,670	2	11,327	2,124	2
1963	14,201	3,727	1	12,399	3,003	2
1964	10,286	6,234	66	11,317	5,653	36
1965	6,325	5,698	0	8,060	5,486	7
1966	5,263	5,917	124	4,388	5,578	25
1967	17,538	6,036	839	3,784	6,236	379
1968	213	5,237	3,367	5,394	5,294	3,038
1969	204	4,381	3,613	1,055	3,257	3,333
1970	1,050	3,104	2,803	1,934	2,118	2,564
1971	518	1,099	2,522	683	1,597	2,827
1972	4,897	1,301	2,689	2,183	2,447	2,686
1973	35	2,157	4,998	3,536	2,111	5,290
1974	8,511	1,816	2,538	7,353	2,380	2,505
1975	8,148	172	2,097	8,119	678	1,521
1976	711	486	7,026	11,725	751	7,908
1977	1,785	5,442	17,366	3,859	5,604	11,938
1978	11	3,811	14,768	4	4,077	14,608
1979	69	3,726	9,835	0	3,589	11,414
1980	8	3,546	13,229	0	3,659	14,785

PRODUCTION OF UO_3 AND UF_4

Data on the production of uranium trioxide in Plant 2/3 are presented in Table C1–2. The use of depleted uranium in production of UO_3 occurred only during FY 1970. In that year, 41 MTU of depleted UO_3 was produced. These data are the basis for Figures C–4 and C–5. Also shown in Table C1–2 is the production data for uranium tetrafluoride in Plant 4. The most recent compilations are given for FY 1952–88 in the column labeled "Total." It is the basis for Figure C–6. The data for production of natural and enriched UF_4 through FY 1976 were from an earlier tabulation. These earlier data in some cases disagree with those in the later tabulation. It is not presently known whether the difference is due to production of depleted UF_4 or to a revision of the production data for some years. The only year for which depleted UF_4 production has been specifically identified is FY 1970 when 343 MTU of that material was produced. Depleted UF_4 from Paducah was repackaged in Plant 4, but that activity is not reflected in the table.

PRODUCTION OF DERBIES AND INGOTS IN PLANT 5

Data on the production of uranium metal derbies and ingots in Plant 5 are presented in Tables C1–3 and C1–4. The total production amounts cover the period from FY 1952 through FY 1988. These data form the basis for Figures C–8, C–9, and C–10.

PRODUCTION IN PLANTS 6 AND 9

Table C1–5 contains information about the production of enriched uranium metal ingots and enriched uranium metal products that were machined in Plant 9. Enriched uranium was used almost exclusively in Plant 9. The only exception was the processing of 37 MTU of natural uranium in Plant 9 during FY 1974. Also contained in Table C1–5 are data on manufacture of rolled and machined uranium products in Plant 6. The distribution of uranium enrichments for Plant 6 products is shown in Table C1–6. These two tables form the basis for Figures C–11, C–12, and C–13.

Table C1–2. Production of Uranium Trioxide and Uranium Tetrafluoride

Fiscal	Production (MTU) of Uranium trioxide in Plant 2/3			Production (MTU) of Uranium tetrafluoride in Plant 4		
Year	Total	Natural	Enriched	Total	Natural	Enriched
1952	0	0	0	0	0	0
1953	0	0	0	0	0	0
1954	642	642	0	1,568	1,568	0
1955	3,288	3,288	0	3,314	3,314	0
1956	5,329	5,329	0	5,029	5,029	0
1957	8,370	8,370	0	9,358	9,358	0
1958	10,039	10,039	0	12,117	11,577	0
1959	11,540	11,540	0	9,454	8,459	0
1960	12,187	12,187	0	11,388	10,426	0
1961	11,039	11,039	0	10,642	8,966	0
1962	6,288	6,288	0	9,468	7,849	0
1963	0	0	0	10,482	7,928	1,075
1964	0	0	0	7,203	4,145	997
1965	543	0	543	6,797	3,117	2,888
1966	1,347	196	1,151	6,174	2,052	3,381
1967	1,835	832	1,003	6,263	2,632	3,283
1968	3,251	1,555	1,696	4,809	1,219	3,588
1969	2,028	665	1,363	2,821	494	2,326
1970	880	259	621	1,923[a]	666	914
1971	809	574	235	580	55	525
1972	2761	2,365	396	347	0	347
1973	3,534	3,533	1	0	0	0
1974	7,114	7,114	0	342	0	342
1975	8,189	8,189	0	634	0	633
1976	9,752	9,752	0	0	0	0
1977	2,191	1673	518	0	0	0
1978	0	0	0	0	0	0
1979	0	0	0	0	0	0
1980	0	0	0	479	0	479
1981	103	0	103	562	0	562
1982	203	0	203	366	0	366
1983	319	0	319	1,145	0	1,145
1984	306	0	306	1,240	0	1,240
1985	145	0	145	1,146	60	1,086
1986	2	0	2	1,068	0	1,068
1987	170	0	170	280	0	280
1988	93	0	93	388	0	388

[a] Includes production of 343 MTU of UF_4 using depleted uranium.

Table C1-3. Production of Uranium Metal Derbies in Plant 5

Fiscal Year	Production (MTU) of uranium metal derbies			
	Total	Natural	Enriched	Depleted
1952	0	0	0	0
1953	45	45	0	0
1954	2,099	2,099	0	0
1955	5,824	5,824	0	0
1956	8,459	8,459	0	0
1957	6,113	6,113	0	0
1958	6,749	6,260	489	0
1959	7,759	6,881	878	0
1960	10,586	9,704	882	0
1961	8,470	7,052	1,418	0
1962	8,563	6,782	1,781	0
1963	10,243	7,655	2,588	0
1964	7,648	4,080	3,568	0
1965	6,432	2,991	3,441	0
1966	5,166	2,018	3,054	94
1967	7,172	2,756	3,547	236
1968	5,339	1,255	3,435	660
1969	4,017	95	2,578	1,344
1970	2,885	1,974	261	650
1971	1,344	172	205	967
1972	1,217	0	225	992
1973	2,139	0	170	1,969
1974	1,317	0	362	954
1975	1,121	0	325	797
1976	1,703	0	140	1,564
1977	1,780	35	219	1,525
1978	2,139	0	291	1,848
1979	1,618	0	272	1,346
1980	2,019	0	213	1,806
1981	2,608	0	588	2,020
1982	4,159	0	682	3,477
1983	4,802	0	1,085	3,717
1984	6,290	0	1,054	5,237
1985	5,075	218	1,111	3,746
1986	6,205	215	1,010	4981
1987	4,606	0	346	4,260
1988	2,667	0	305	2,362

Table C1-4. Production of Uranium Metal Ingots in Plant 5

Fiscal Year	Production (MTU) of uranium metal ingots			
	Total	Natural	Enriched	Depleted
1952	0	0	0	0
1953	90	90	0	0
1954	3,976	3,976	0	0
1955	9,528	9,528	0	0
1956	12,137	12,037	0	0
1957	12,680	12,680	0	0
1958	12,727	12,727	0	0
1959	13,365	13,365	0	0
1960	16,708	16,708	0	0
1961	12,691	12,691	0	0
1962	12,865	12,865	0	0
1963	14,285	14,285	0	0
1964	11,655	11,655	0	0
1965	10,234	10,234	0	0
1966	11,239	6,498	1,376	67
1967	10,969	5,266	2,451	432
1968	10,144	2,503	1,506	2,248
1969	6,638	191	0	2,540
1970	5,425	3,762	0	1,269
1971	2,375	435	0	1,838
1972	1,683	0	0	1,633
1973	3,292	0	0	3,260
1974	1,711	5	0	1,525
1975	1,167	0	0	1,041
1976	2,142	0	0	2,080
1977	2,175	0	61	2,114
1978	1,963	0	53	1,910
1979	1,386	0	0	1,386
1980	1,989	0	0	1,989
1981	2,047	0	0	2,047
1982	3,732	0	0	3,732
1983	4,569	0	610	3,959
1984	3,933	0	239	3,694
1985	4,558	691	125	3,742
1986	4,310	206	0	4,104
1987	4,501	0	0	4,501
1988	3,109	0	0	3,109

Table C1-5. Production of Uranium Ingots and Machined Metal Products in Plants 9 and 6

Fiscal Year	Production (MTU) of uranium metal products			
	Plant 9		Plant 6	
	Ingots	Machined	Rolled	Machined
1952	0	0	0	0
1953	0	0	1,966	1,608
1954	0	0	5,679	3,581
1955	0	0	9,973	6,752
1956	0	0	12,470	8,086
1957	0	0	15,074	8,629
1958	732	0	13,665	8,378
1959	1,251	0	14,033	7,320
1960	1,388	0	18,532	9,131
1961	2,364	0	15,370	7,552
1962	2,663	0	15,430	8,211
1963	3,660	0	14,507	9,232
1964	5,297	0	11,313	9,279
1965	5,361	0	12,310	8,674
1966	1,197	3,296	7,683	6,987
1967	1,258	3,753	7,576	5,837
1968	691	4,165	5,029	5,105
1969	778	2,980	3,380	3,227
1970	499	1,720	3,309	2,882
1971	422	2,182	1,068	1,413
1972	599	1,839	0	922
1973	452	3,067	0	1,881
1974	1,031	2,221	0	870
1975	1,189	1,532	0	797
1976	304	1,996	0	1,065
1977	381	2,074	0	1,110
1978	480	1,932	0	1,172
1979	604	1,558	0	900
1980	380	1,788	0	999
1981	796	2,214	0	1,127
1982	974	3,566	0	1,821
1983	1,366	4,391	0	2,191
1984	1,516	4,254	0	1,924
1985	1,074	3,428	0	1,860
1986	1,640	4,222	0	1,743
1987	745	709	0	426
1988	394	338	0	8

Table C1–6. Production of Uranium Fuel and Target Elements in Plant 6

Fiscal Year	Uranium core and target production (MTU)		
	Natural	Enriched	Depleted
1952	0	0	0
1953	1,608	0	0
1954	3,581	0	0
1955	6,752	0	0
1956	8,086	0	0
1957	8,629	0	0
1958	7,961	0	0
1959	6,660	0	0
1960	8,330	0	0
1961	6,306	0	0
1962	6,906	0	0
1963	7,396	0	0
1964	6,428	0	0
1965	5,665	0	0
1966	3,312	582	0
1967	2,983	1,218	103
1968	1,246	1,024	1,012
1969	131	0	1,154
1970	1,779	0	777
1971	410	0	941
1972	0	0	922
1973	0	0	1,881
1974	0	0	870
1975	0	0	797
1976	0	0	1,065

RECOVERY OF SCRAP URANIUM

Table C1–7 contains data on the recovery of uranium from scrap material in Plant 8 at the FMPC. The recovered uranium was then fed to the Refinery. Most of the material recovered was natural uranium. Recovery of depleted uranium was minimal. Data in the table are plotted in Figures C–14 and C–15.

Table C1-7. Uranium Recovered from Scrap Material in Plant 8

Fiscal Year	Recovery of feed material(MTU) from scrap			
	Total	Natural	Enriched	Depleted
1952	0	0	0	0
1953	0	0	0	0
1954	266	266	0	0
1955	1,160	1,160	0	0
1956	1,764	1,764	0	0
1957	1,927	1,927	0	0
1958	2,018	2,018	0	0
1959	2,568	2,568	0	0
1960	3,188	3,188	0	0
1961	2,902	2,902	0	0
1962	2,820	2,820	0	0
1963	2,657	2,115	542	0
1964	3,505	2,380	1,125	0
1965	2,134	1,182	952	0
1966	1,617	650	967	0
1967	1,837	855	982	0
1968	2,222	687	1,530	5
1969	1,036	256	759	21
1970	649	423	204	22
1971	307	128	172	7
1972	111	7	103	1
1973	66	21	45	0
1974	3	3	0	0
1975	43	11	32	0
1976	51	12	39	0
1977	386[a]			
1978	122[a]			
1979	184[a]			
1980	118[a]			
1981	41[a]			
1982	237[a]			
1983	376[a]			
1984	261[a]			
1985	188	5	143	40
1986	176	2	141	33
1987	1,106	0	223	883
1988	261	10	69	181

[a] No data on enrichment categories are available.

PRODUCTION OF UF_4 FROM UF_6

Table C1–8 contains information about the production of uranium tetrafluoride from uranium hexafluoride in the FMPC Pilot Plant. There was no production during the years 1968 through 1984. Enriched UF_6 was the primary feed for the process. No UF_4 was produced using depleted uranium in the Pilot Plant until 1986.

Table C1–8. Production of Uranium Tetrafluoride in the Pilot Plant

Fiscal Year	Production of UF_4 from UF_6 in the Pilot Plant		
	Total	Natural	Enriched
1953	15		15
1954	~20[a]	~20[a]	
1955	26		26
1956	33		33
1957	0	0	0
1958	540	0	540
1959	995	0	995
1960	962	0	962
1961	1,676	0	1,676
1962	2,961	1,342	1,619
1963	2,676	1,197	1,479
1964	3,529	1,468	2,061
1965	1,450	658	792
1966	1,219	478	741
1967	361	13	348
1968-1984	0	0	0
1985	622	511	111
1986	462[b]	92	125
1987	542[c]	0	160
1988	1,642[d]	0	0

[a] Estimate based on apparent capacity (Davis 1956).
[b] Includes 245 MTU of depleted uranium.
[c] Includes 382 MTU of depleted uranium.
[d] Production was entirely of depleted uranium.

Conversion of UF_6 to UF_4 was also the purpose of Plant 7, which operated for only two years (from June 1954 through May 1956). Natural and depleted uranium were employed in separate sets of reactor banks in the facility. Production of UF_4 from natural uranium feed was 71.3, 138, 104, 227, and 110 MTU during the first five months of 1956. Conversion of depleted UF_6 during the first four months of that year was accomplished at a rate that exceeded the original design capacity of the plant. Monthly production amounts were 311, 230, 265, and 308 MTU, all well above an expected 180 MTU for that half of the plant. Higher than design feed rates, improvements in availability, or use of all four reactors are possible explanations of this high level of production. It is known that a number of improvements had been made to the plant to overcome initial operational difficulties.

SHIPMENTS OF PRODUCTS FROM THE FMPC

Data on shipments of intermediate products from the FMPC have been located for the fiscal years 1971 through 1976. These shipment data are summarized in Table C1–9. The quantities of UO_3 shipped offsite were larger than shipments of any other intermediate product during this period.

Table C1-9. Shipments of Intermediate Products from the FMPC, FY 1971–1976

Fiscal Year	Shipments (MTU) of intermediate products			
	UO_3 (natural)	Ingots (enriched)	Metal (depleted)	UF_4 and U_3O_8 (enriched)
1971	804	342	0	5
1972	2,908	538	0	25
1973	3,885	372	0	2
1974	7,238	941	0	10
1975	8,100	672	193	4
1976	9,998	321	158	14

Table C1–10 contains data on the shipments of fuel and target elements of varying ^{235}U content from the FMPC from FY 1952 through FY 1976. Natural uranium was the most important component of the total until the early 1960s, when enriched uranium began to comprise a significant fraction of the total amounts shipped.

Table C1-10. Quantities of Fuel and Target Elements Shipped from the FMPC

Fiscal Year	Shipments (MTU) of fuel and target elements		
	Natural	Enriched	Depleted
1952	0	0	0
1953	1,476	1	0
1954	3,612	0	0
1955	6,544	0	0
1956	8,033	13	0
1957	7,705	65	0
1958	7,954	409	0
1959	7,332	626	0
1960	9,325	791	0
1961	7,116	1,344	0
1962	8,530	1,414	0
1963	8,062	1,837	0
1964	6,395	2,693	0
1965	5,791	3,033	0
1966	3,312	3,102	0
1967	3,229	3,081	86
1968	1,246	3,170	934
1969	139	2,357	1,161
1970	1,774	655	709
1971	357	318	982
1972	0	28	893
1973	0	0	1,846
1974	0	0	912
1975	0	0	365
1976	0	0	1,496

APPENDIX D

OTHER RADIONUCLIDE RELEASES

Processing of uranium was the principal function of the FMPC. Thorium processing was a secondary activity. Radioactive decay of uranium and thorium isotopes produces series of other radionuclides that are collectively referred to as decay products. The initial decay products for the three decay series of greatest interest are shown in Figures D–1, D–2, and D–3. The first of these illustrates the decay products of uranium-238 (^{238}U), including another important uranium isotope, ^{234}U.

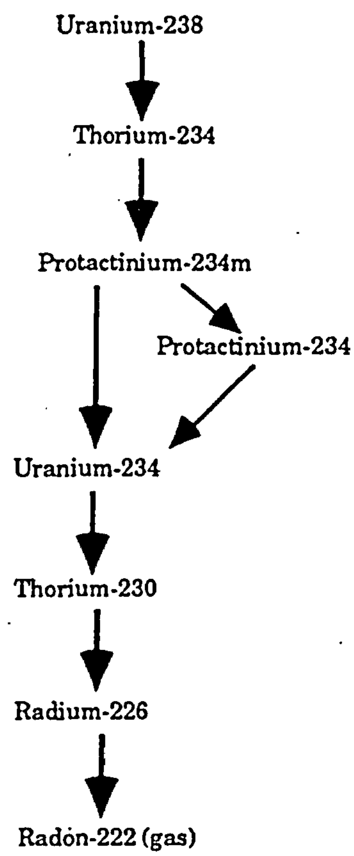

Figure D–1. Decay products of uranium-238, from thorium-234 to radon-222.

In most of the feeds received by the FMPC, the uranium had previously been separated chemically from the other decay products. As a result, the facility's effluents consisted primarily of uranium and other radionuclides were generally present in small quantities. Radioactive decay of uranium after the initial chemical separation from the daughter radionuclides also produced those same nuclides as trace contaminants.

However, early processing campaigns treated ores that contained near equilibrium amounts of the daughter radionuclides through radium. As shown in Fig. D–1, the decay product that follows radium is radon, a gas. The wastes from that early processing were placed in the K-65 Storage Silos (see Appendix J). Releases of radon and other nuclides from the silos are a special case that is treated in Appendix J.

Some thorium was processed at the FMPC. Fig. D–2 shows a comparable sequence of the decay products of thorium-232 (^{232}Th), which includes thorium-228 and two radium isotopes. This sequence also leads to a gaseous radon isotope. Processed thorium would include both thorium isotopes and small residuals of the other solid elements. Radioactive decay after processing would also produce trace contaminants in the thorium.

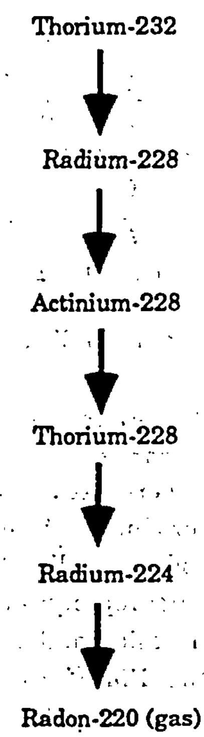

Figure D–2. Decay products of thorium-232, from radium-228 to radon-220.

The third decay chain of interest is that of uranium-235, which is present (0.72%) in natural uranium and in increased amounts (generally less than 1.5%) in enriched uranium processed at the FMPC. This decay sequence (Fig. D–3) also includes an isotope of radon.

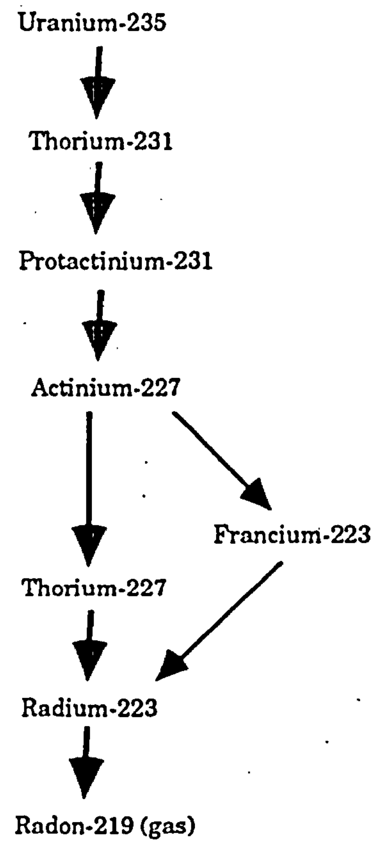

Figure D-3. Decay products of uranium-235, from thorium-231 to radon-219.

In addition to decay products, other radionuclides were released during FMPC operations. These originated in nuclear reactors, where finished uranium fuel and target elements, produced at the FMPC, were used. Fissioning of the uranium atom produces other radionuclides, called fission products. Absorption of neutrons by uranium and other materials present in the reactor produces radioactive activation products. When spent fuel from the reactors was processed at fuel reprocessing plants (not at the FMPC), the uranium was not completely separated from fission and activation products. As a result, recovered uranium that was recycled to the FMPC introduced small amounts of fission and activation products into the process streams at the FMPC.

Receipts of recycled uranium began at the FMPC in fiscal year (FY) 1961. All of the recycled uranium that was received during FY 1961–1963 was in the form of enriched uranium trioxide (UO_3) from Hanford (Spenceley 1985). Production of enriched uranium tetrafluoride (UF_4) from UO_3 in Plant 4 did not begin until FY 1963 (Rathgens et al. 1985), so releases of fission and activation products would not have occurred prior to July 1962. Plant staff involved in the processing of the recycled UO_3 have identified October 1962 as the time that processing of that material began at the FMPC (Bonfer 1991). Measurements of the amounts of these radionuclides, relative to uranium, were not performed until years later. At that time, concentrations of fission and activation products were probably higher than those present in the early years of processing of recycled uranium at the FMPC. However, the later measurements are presently the only guide to the concentrations of those contaminants in effluents. Results of the measurements are discussed below.

AIRBORNE RADIONUCLIDES

In 1985, measurements of fission and activation products in particulate material trapped in scrub liquor and by dust collectors were performed (Boback et al. 1987). Single grab samples were taken from a wide variety of locations and analyzed for fission and activation products. The results of these measurements are presented as radionuclide concentrations per kilogram of uranium in Table D–1. Absence of an entry in the table, as in the column for ruthenium-106 (^{106}Ru, shown in this table as Ru-106), for example, indicates that no result was reported for that radionuclide.

For most radionuclides, the variability, both from one dust collector to another in any particular plant and among plants, was substantial. This can be seen from the arithmetic means and standard deviations that have been computed using the results for each plant. Sampling and analytical uncertainties for these results were not reported in Boback et al. (1987). Only the short-lived daughters of ^{238}U were found in consistent concentrations. The concentrations of thorium isotopes and their radium (Ra) daughter products were found to be consistent in samples from some plants but not from others. The concentrations of fission products — cesium (Cs), strontium (Sr), and technetium (Tc) — were highly variable. For some analyses, ^{90}Sr was not detected; upper bound concentrations are included in Table D–1, but were not considered in the statistical analysis of the ^{90}Sr concentrations. The fission product ^{106}Ru was reported for only one of the samples: 0.084 µCi (kg U)$^{-1}$ in dust from collector G4-2.

Transuranic elements neptunium (Np) and plutonium (Pu) were also measured. The nuclides ^{237}Np, ^{238}Pu, and $^{239/240}$Pu were detected in all of the samples analyzed. The relative amounts of ^{239}Pu and ^{240}Pu cannot be determined by alpha spectrometry (the common analytical technique) because the alpha particles emitted by the two nuclides have very similar energies. The observed concentrations varied over a wide range within individual plants and from plant to plant. An important radionuclide that has not been identified, but would be expected to be present, is ^{241}Am. The energy of the alpha particle emitted by ^{241}Am is virtually the same as that emitted by ^{238}Pu. Because the special chemical separation needed to isolate ^{241}Am was apparently not performed, the results reported for ^{238}Pu no doubt include a contribution from ^{241}Am. Because concentrations of individual transuranic nuclides were not determined, these nuclides have been grouped and referred to as TRU, short for transuranic.

Concentrations of fission and activation products observed in 1985 could have been among the highest ever present because recycled uranium had been processed over a long time period. However, plant operations just prior to the measurements could also have had a substantial effect on the measurements. The annex contains a tabulation of data on the quantities of recycled uranium and the associated plutonium that were shipped to the FMPC for processing. Average plutonium concentrations in the various forms of recycled uranium compounds differed substantially, with plutonium/uranium (Pu/U) ratios that

Table D-1. Results of Analyses for Specific Radionuclides and Uranium in Dusts and Scrubber Liquids at the Feed Materials Production Center in 1985

Source of Sample	Radionuclide Concentration (μCi/kg of uranium)														Uranium	Chemical	U-235	U-236
	Pu-239,240	Pu-238	Np-237	Th-234	Pa-234m*	Th-232	Th-230	Th-228	Ra-228	Ra-226	Cs-137	Ru-106	Tc-99	Sr-90	(gU per g)	Form**	Percent	Percent
Plant 1 Dust, G2-1	0.10	0.011	0.051	438	438	0.12	0.23	0.079	0.012	0.009	0.030		1.1		0.708	UTf, UOct	0.95	0.01
Plant 1 Dust, G2-64	219	8.7	12	381	381	19	3685	8.7	11	39	12		56	2.3	0.160	UOct	0.71	0.01
Plant 1 Dust, G2-76	6.1	0.68	0.79	975	975	1.4	30	1.8	0.79	2.0	30		7.9	24	0.028	UOct	0.84	0.02
Plant 1 Dust, G2-172	1.6	0.29	0.33	629	629	8.8	26	5.5	5.5	3.5	0.031		503		0.238	UOct	0.67	0.02
Plant 1 Dust, G2-235	0.18	0.11	0.077	496	496	0.32	9.3	1.3	0.12	0.50	0.22		5.4	0.30	0.626	UDi, UOct	3.43	0.04
Plant 1 Mean	45	1.9	2.6	584	584	6.0	750	3.5	3.4	9.1	8.5		115	13				
Std. Dev.	97	3.8	5.2	237	237	8.3	1641	3.6	4.6	17	13		218	15				
Plant 4 Dust, G4-1	0.30	0.029	0.22	722	722	0.098	0.30	0.27	0.018	0.16	1.6		69	1.2	0.73	UTri	0.83	0.06
Plant 4 Dust, G4-2	0.054	0.0063	0.10	277	277	0.061	0.090	0.053	0.0026	0.0037	0.026	0.084	82	0.070	0.76	UTf	0.84	0.07
Plant 4 Dust, G4-4	0.19	0.017	0.064	536	536	237	0.42	0.25	0.0072	0.0051	0.032		52	0.044	0.75	UTf	0.51	0.02
Plant 4 Dust, G4-5	0.11	0.018	0.14	605	606	0.077	0.32	0.30	0.015	0.0083	0.28		97	0.20	0.71	UTf	0.80	0.05
Plant 4 Dust, G4-7	0.63	0.047	0.15	610	610	0.17	0.28	0.31	0.0041	0.0047	0.067		106	0.028	0.51	UDi	1.11	0.05
Plant 4 Dust, G4-12	0.013	0.0023	0.0035	293	293	0.035	0.052	0.049	0.0027	0.0012	0.097		2.1		0.75	UTf	1.78	< 0.01
Plant 4 Dust, G4-13	0.048	0.0086	0.17	783	783	0.22	0.31	0.33	0.0053	0.013	0.062		93	0.032	0.82	UOct	0.90	0.02
Plant 4 Dust, G4-14	0.0010	0.0036	0.0070	726	726	0.077	0.13	0.033	0.0044	0.0028	0.038		0.046		0.76	UTf	0.20	< 0.01
Plant 4 Dust, G4-15	0.46	0.039	0.088	721	721	0.16	0.54	0.22	0.0050	0.015	0.020		5.3		0.74	UTf	0.84	0.07
Plant 4 Mean	0.20	0.019	0.11	586	586	26	0.27	0.20	0.0072	0.024	0.25		55	0.27				
Std. Dev.	0.22	0.016	0.073	187	187	79	0.16	0.12	0.0055	0.052	0.52		43	0.47				
Plant 5 Dust, G5-247	3.1	0.52	1.4	1223	1223	1.6	6.3	15	1.9	0.55	11		18	94	0.014	UOct	0.78	0.05
Plant 5 Dust, G2-67	5.2	0.47	1.5	1983	1983	3.6	11	9.5	1.8	0.21	13		34	78	0.023	UOct	0.81	0.05
Plant 5 Dust, G5-248	0.40	0.10	0.25	32653	32653	1.2	1.2	6.3	0.15	0.14	5.6		2.3	1.1	0.015	UOct	0.21	< 0.01
Plant 5 Dust, G5-249	0.015	0.0030	0.015	880	880	0.073	0.17	0.071	0.0071	0.0052	0.054		0.48	1.2	0.47	UTf	0.22	< 0.01
Plant 5 Dust, G5-250	0.0093	0.0028	0.018	1162	1162	0.10	0.35	0.31	0.0058	0.014	0.041		0.17	0.38	0.34	UTf	0.20	0.01
Plant 5 Dust, G5-251	0.90	0.084	0.60	23913	23913	0.30	6.0	1.4	0.13	0.13	4.9		8.7	4.1	0.037	UOct	0.43	0.01
Plant 5 Dust, G5-253	0.37	0.062	0.22	25478	25478	0.61	2.3	0.27	0.18	0.11	10		1.3	76	0.016	UOct	0.39	0.01
Plant 5 Dust, G5-254	0.65	0.093	0.33	18981	18981	0.79	1.1	0.93	0.19	0.069	18		3.0	79	0.022	UOct	0.28	0.01
Plant 5 Dust, G5-256	1.3	0.13	0.53	17004	17004	0.97	1.9	1.1	0.21	0.13	21		4.9	4.5	0.025	UOct	0.27	0.01
Plant 5 Dust, G5-260	0.030	0.011	0.022	34364	34364	0.020	0.26	0.12	0.0055	0.0034	0.024		0.28		0.49	UOct	0.20	< 0.01
Plant 5 Dust, G5-261	0.033	0.0037	0.015	9185	9185	0.0047	0.069	0.060	0.0041	0.0041	0.017		0.28	0.013	0.75	UOct	0.21	< 0.01
Plant 5 Dust, G5-262	1.0	1.0	3.9	4727	4727	68	45	138	1.6	0.90	66		34		0.0011	UOct	0.25	< 0.01
Plant 5 Dust, G5A-100	0.17	0.062	0.33	2913	2913	2.3	2.7	9.3	0.12	0.058	3.1		17		0.052	UOct	0.31	0.01
Plant 5 Dust, G5A-101	0.32	0.074	0.24	24233	24233	1.9	1.8	6.5	0.11	0.053	4.2		1.3	3.9	0.062	UOct	0.22	< 0.01
Plant 5 Dust, Bldg. 55	0.18	0.029	0.25	23558	23558	2.7	4.0	5.3	0.077	0.15	3.3		4.8	1.2	0.021	UOct	0.21	< 0.01
Plant 5 Mean	0.92	0.18	0.64	14817	14817	5.6	5.6	13	0.44	0.17	11		8.7	29				
Std. Dev.	1.4	0.28	1.0	12216	12216	17	11	34	0.71	0.24	17		12	39				

Table D-1 (Continued). Results of Analyses for Specific Radionuclides and Uranium in Dusts and Scrubber Liquids at the Feed Materials Production Center in 1985

	Radionuclide Concentration (μCi/kg of uranium)														Uranium	Chemical	U-235	U-236
Source of Sample	Pu-239,240	Pu-238	Np-237	Th-234	Pa-234m*	Th-232	Th-230	Th-228	Ra-228	Ra-226	Cs-137	Ru-106	Tc-99	Sr-90	(gU per g)	Form**	Percent	Percent
Plant 8 Dust, G43-27	4.7	0.24	0.64	257	257	1.1	101	1.7	0.25	0.18	0.14		36	0.26	0.11	UOct	0.93	0.05
Plant 8 Dust, G43-29	0.64	0.083	0.36	38	38	0.23	1.9	0.81	0.035	0.036	0.022		32	0.041	0.69	UOct	0.91	0.05
Plant 8 Dust, 8035	0.16	0.037	0.067	409	409	0.28	0.59	0.30	0.058	0.0059	0.26		13	0.028	0.54	UOct	0.42	0.02
Plant 8 Dusts Mean	1.8	0.12	0.36	234	234	0.54	34	0.92	0.11	0.075	0.14		27	0.11				
Std. Dev.	2.6	0.11	0.29	186	186	0.49	58	0.68	0.12	0.095	0.12		12	0.13				
Scrub Liquor Samples																		
Plant 8, Box Furnace	7.1	0.95	1.5	320	320	23	30	23	0.53	1.1	13		71		0.0017	UOct	0.63	0.03
Plant 8, Rotary Kiln	3.0	0.33	0.70	607	607	7.7	77	6.2	0.22	0.12	0.65		39	3.4	0.0067	UOct	0.92	0.05
Plant 8, Oxidation Furnace #1	0.036	0.012	0.036	94	94	1.0	1.2	3.6	0.15	0.016	1.3		5.8	0.27	0.036	UOct	0.22	< 0.01
Plant 8 Liquids Mean	3.4	0.43	0.74	341	341	11	36	11	0.30	0.40	5.0		39	1.8				
Std. Dev.	3.6	0.48	0.72	257	257	11	38	10	0.20	0.58	7.0		33	2.2				
Plant 9 Dust, C9N1-1039	1.7	0.13	0.46	13858	13858	0.060	0.27	1.8	0.013	0.44	0.73		69.3	0.22	0.55	UOct	0.93	0.05
Pilot Plant Dust, G-1	0.0023	0.0023	0.0048	81	81	0.073	0.35	0.13	0.0023	0.0036	0.20		0.096		0.75	UTf	0.78	< 0.01
Pilot Plant Dust, G-2	0.0017	0.0035	0.0099	123	123	0.11	0.19	0.13	0.0028	0.0019	0.21		0.17		0.75	UTf	0.74	< 0.01
Pilot Plant Dust, 735-13-7050	0.62	4.9	0.18	782	782	11	9.1	8.6	6.6	0.45	6.2		2.1		0.024	UDi, UOct	0.62	< 0.01
Pilot Plant Mean	0.21	1.6	0.064	329	329	3.8	3.2	3.0	2.2	0.15	2.2		0.78					
Std. Dev.	0.36	2.8	0.098	393	393	6.4	5.1	4.9	3.8	0.26	3.4		1.1					

*Pa-234m taken to be in equilibrium with Th-234; original values had not been corrected for decay since time of sampling.

**Chemical form codes: UTf = uranium tetrafluoride; UDi = uranium dioxide, UTri = uranium trioxide; UOct = uranium octoxide.

ranged from about 0.3 parts per billion (ppb) for receipts of offsite UO_2 to more than 1100 ppb for UO_3 received from Paducah in 1980. Except for the 1980 shipment, the Pu/U ratios of incoming materials, while variable, were less than 10 ppb.

Part of the material from Paducah was repackaged, from hoppers to drums, in Plant 4. To reduce the Pu concentration, it was blended with sump cake in the rotary kiln in Plant 8 and converted to calcium uranate, which was subsequently used as feed for the refinery. Production of UO_3 from this feed stock appears to have begun in May 1982 and 110 lots had been produced by May 1985. The Pu content of each lot was measured and Pu/U ratios ranging from 4 to 46 ppb were found (Spenceley 1985). The ratio generally increased with time, but not monotonically, as the feed with higher Pu content became incorporated into the refinery inventory. Samples of uranyl nitrate hexahydrate (UNH) from 14 tanks in the refinery were analyzed for Pu in April 1985. The measured Pu/U ratios in samples of UNH ranged from 6.5 to 81 ppb.

Processing of the Paducah material was performed in the years just prior to the time concentrations of plutonium and other transuranic nuclides were measured in various samples of dusts and scrub liquors. In Plant 8, where the Paducah material was blended, ratios of Pu/U in samples of scrub liquor averaged about 60 ppb. Dust from primary dust collector for that facility was found to have a Pu/U ratio of about 80 ppb. Samples of dusts collected in Plant 4, which presumably represent historically more typical Pu/U ratios, averaged about 5 ppb. A similar low concentration ratios were also found in the dusts collected from the Pilot Plant. A somewhat higher average Pu/U ratio was found in dusts from Plant 5, but the results appear to be highly dependent upon the specific process exhaust treated. The highest Pu/U ratio was found in a sample of dust from Plant 1; it was about 3600 ppb in dust from collector G2–64. This finding apparently reflects dust from grinding and homogenization of samples of the original Paducah UO_3.

OTHER RADIONUCLIDES IN LIQUID WASTES

Various FMPC monthly reports, environmental monitoring reports, and analytical data sheets have been found to contain data on the presence of radionuclides other than uranium in liquid waste discharges. These data are tabulated in Appendix L to which the reader is referred for a more detailed discussion. Measurements of releases of thorium and ^{226}Ra were made in the mid-1950s. However, monitoring of the two radium isotopes (^{226}Ra and ^{228}Ra) does not appear to have been performed consistently until 1968. Data from measurements of activation and fission products beginning in 1976 have been identified. Concentrations of activation products (^{237}Np, ^{238}Pu, and $^{239/240}$Pu) and of fission products (^{137}Cs, ^{106}Ru, ^{99}Tc, and ^{90}Sr) have been documented in liquid wastes. Other decay products of ^{238}U and ^{232}Th were also present as shown by the data in Table D–1. The releases of other radionuclides in liquid wastes have generally not been related to specific facilities at the FMPC or to particular operations within the plants.

Because the measurements of other radionuclides were not made in every year, it was necessary to develop correlations between the releases of uranium and those of the other radionuclides. Ratios of releases, expressed for example as μCi ^{226}Ra per kg U, were computed for years when measurements were made. These ratios, compiled in Tables L–12 and L–13, provide a basis for estimating releases of the other radionuclides for years when they were not measured. Substantial year to year variability is common for these ratios; the standard deviations are typically larger than the mean values. This variability was considered when deriving the uncertainties associated with the estimated releases of other radionuclides.

RELATIVE IMPORTANCE OF RELEASES

The relative importance of releases of radionuclides to the environment depends upon comparison of three factors. These are the quantities released, the potential for concentration in the environment, and the relative toxicities of the radionuclides, as measured by their dose conversion factors for the several possible modes of exposure. Differences in dispersion and dilution of uranium and the other radionuclides in the atmosphere and in the river are not expected to be significant.

A methodology developed by the National Council on Radiation Protection and Measurements (NCRP) (NCRP 1989) was used to assess the relative importance of the identified radionuclides as potential contributors to offsite radiation dose. The NCRP screening methodology was primarily intended to evaluate compliance with environmental standards. However, the screening factors that were developed for many radionuclides and a variety of exposure pathways can also be used to assess the relative importance of radionuclide releases to the environment. The referenced methodology has been expanded to include liquid pathways; formal publication of that work is expected in 1995.

The screening factors for radionuclides released to the atmosphere or to fresh water address two of the three factors listed above. The potential for concentration in the environment is evaluated by considering environmental pathways that reflect important transport mechanisms. These are buildup of radionuclides in soils and sediments and uptake into the terrestrial and aquatic food chains. The relative toxicity of each radionuclide (and any other radionuclides that may be produced by its radioactive decay) is also reflected in the NCRP screening factors. Data for the third comparison—differences in the quantities released—were available from direct measurements of releases of uranium and other radionuclides and from the measurements of the relative concentrations of other radionuclides in collected dust and scrub liquors presented above.

The relative importance of a particular radionuclide is defined as the fraction that it contributes to the total potential radiation dose from all radionuclides. This parameter was evaluated for releases to the atmosphere and for releases to water. Both surface water and groundwater were considered in the latter category. Mathematically, the relative importance of a particular nuclide (RI_j) is

$$RI_j = \frac{Q_j \; SF_j}{\sum_{j=1}^{n} Q_j \; SF_j}$$

where Q_j and SF_j are the quantity discharged and the screening factor, respectively, for releases to the atmosphere or to water. The summation in the denominator of the equation extends over all (n) of the radionuclides released to the medium of interest.

The relative importance of the radionuclides in FMPC discharges to air and water were evaluated using Monte Carlo techniques. Measured and estimated uranium releases were used, together with data on correlations of releases of other radionuclides to releases of uranium, to develop release estimates for the other radionuclides. The transuranic nuclides were treated as a group, rather than individually, because all individual contributions were not defined. The environmental behavior and toxicity for the TRU group were approximated by parameters applicable to the plutonium isotopes.

Uncertainties in the uranium releases were derived as part of the source term estimates and those uncertainty estimates were used in the present calculations. The concentration ratios obtained for airborne release locations were assumed to be medians of lognormal distributions whose geometric standard deviations were estimated to be 1.5. For the liquid releases, the observed means and ranges of release ratios were used to define triangular distributions for the calculations.

Because the NCRP screening factors were developed to assess compliance with standards, their cautious approach tends to overestimate potential exposures. For the present calculations of *RI*, it was assumed that a triangular distribution with the most probable value equal to the *SF* could be used to define a range of possible estimates. In most cases, the upper bound of the distribution was taken to be 2 times *SF* and the lower bound was 0.1 times *SF*.

The Monte Carlo calculations made to assess the relative importance of radionuclides released to the atmosphere considered inhalation, direct radiation, and ingestion pathways. The results of these calculations are shown in Figure D–4. The most important releases are clearly those of uranium, with an estimated median *RI* of 0.85. The thorium isotopes ^{232}Th and ^{230}Th had median values of *RI* of 0.051 and 0.039, respectively. Median values of *RI* for other nuclides were < 0.02.

The figure clearly illustrates the relative unimportance of other radionuclides compared to uranium. The other nuclides deserve correspondingly less attention in the dose assessment process. Inhalation was the most important exposure pathway, accounting for 91% of the potential uranium dose and about 70% of the doses from thorium isotopes even assuming, as the calculations do, that persons consumed only foods produced near the plant. The contribution of inhalation to the total would have been even greater for persons with typical food supplies.

Three different exposure scenarios were evaluated to ascertain the relative importance of various radionuclide releases to water. The first of these considered all potential exposure pathways. Although rainfall in the Cincinnati area is frequent, some irrigation was considered possible for a demanding crop, such as corn, during crucial growth periods. Based upon a review of precipitation records in the 1960s, it was assumed that supplemental irrigation would be provided for three weeks during July and August to assure adequate moisture for the crop. Some use of river water for irrigation has been reported by nearby residents who were interviewed.

Because exposure from all pathways would be limited to at most a few individuals, alternative calculations were performed for two other exposure scenarios. Table D–2 shows the pathways considered in each case. Scenario 2 considers all pathways but drinking water, a situation that may have been realized along the river near the FMPC. The third scenario considered only drinking water and is relevant for individuals who ingested contaminated groundwater or river water downstream of the plant.

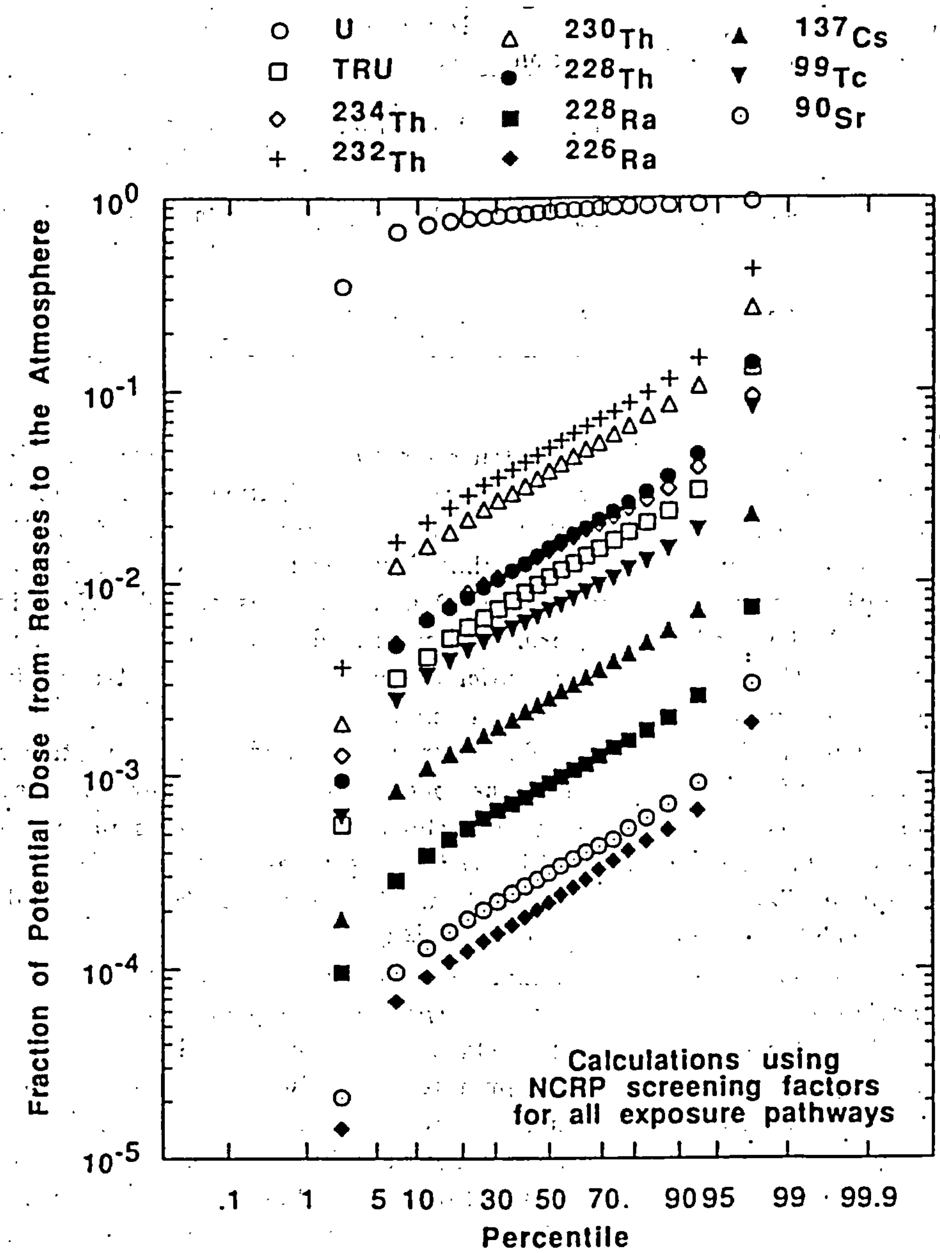

Figure D–4. Contributions of radionuclides released to the atmosphere to the potential dose.

Table D-2. Screening Calculations Performed for Liquid Effluents

	Pathways considered in calculations			Most	
Exposure scenario	Drinking water	Fish consumption	Irrigation water use	important nuclides	Results presented in
1	yes	yes	July, Aug.	^{226}Ra, ^{228}Ra, U, ^{232}Th, ^{234}Th	Fig. D-5
2	no	yes	July, Aug.	^{226}Ra, ^{228}Ra, ^{232}Th, U	Fig. D-6
3	yes	no	no	^{226}Ra, ^{234}Th, ^{228}Ra, U	Fig. D-7

In the first scenario it is assumed that river water is used for drinking, fish from the river are used for food, and river water is used for irrigation of human food crops and plants used for feed for animals that are used in turn for human food. Under these assumptions, the calculations indicate that ^{226}Ra and ^{228}Ra are the most important nuclides with median values of *RI* of 0.60 and 0.19, respectively. Next in order of importance is U, followed by two thorium isotopes; median values of *RI* for these nuclides were between 0.042 and 0.050. Contributions of other nuclides to potential dose for this scenario can be seen in Fig. D-5 to be even smaller.

Figure D-6 contains the results of calculations that address the situation when river water is not used for drinking but the other pathways identified above are assumed to be operative. For this scenario, the same nuclides are identified as important but the rankings are changed somewhat. The radium isotopes ^{226}Ra and ^{228}Ra are again predominant (median values of *RI* were 0.62 and 0.19, respectively); median values for the other three nuclides were in the range 0.025–0.048.

Figure D-7 shows the results for exposure scenario 3 when drinking water is the only complete pathway. Because the groundwater was contaminated by the liquid effluents from the plant, this calculation indicates the relative importance of radionuclides that could be consumed as a result of drinking contaminated groundwater. In these calculations, the lower bound for the triangular screening factor distribution was taken to be 0.5 times *SF* and the upper bound was taken to be 1.1 times *SF*. The most likely value was assumed equal to *SF*. These choices reflect the fact that the average tap water intake is about 1.1 L d^{-1}, compared with the 2.2 L d^{-1} assumed in derivation of the screening factors. Ninety-five percent of a representative population would be expected to consume tap water at a rate < 2.4 L d^{-1} (Roseberry and Burmaster 1992). For the drinking water pathway, ^{226}Ra is again the primary contributor to the dose (median $RI = 0.34$), followed by ^{234}Th (median $RI = 0.23$), ^{228}Ra (median $RI = 0.16$), and uranium (median $RI = 0.15$).

These calculations show that releases of the radium isotopes are quite important for all three scenarios, accounting for roughly 50–80% of the potential dose. The contributions of the uranium and thorium isotopes vary for the three scenarios but are consistently important contributors.

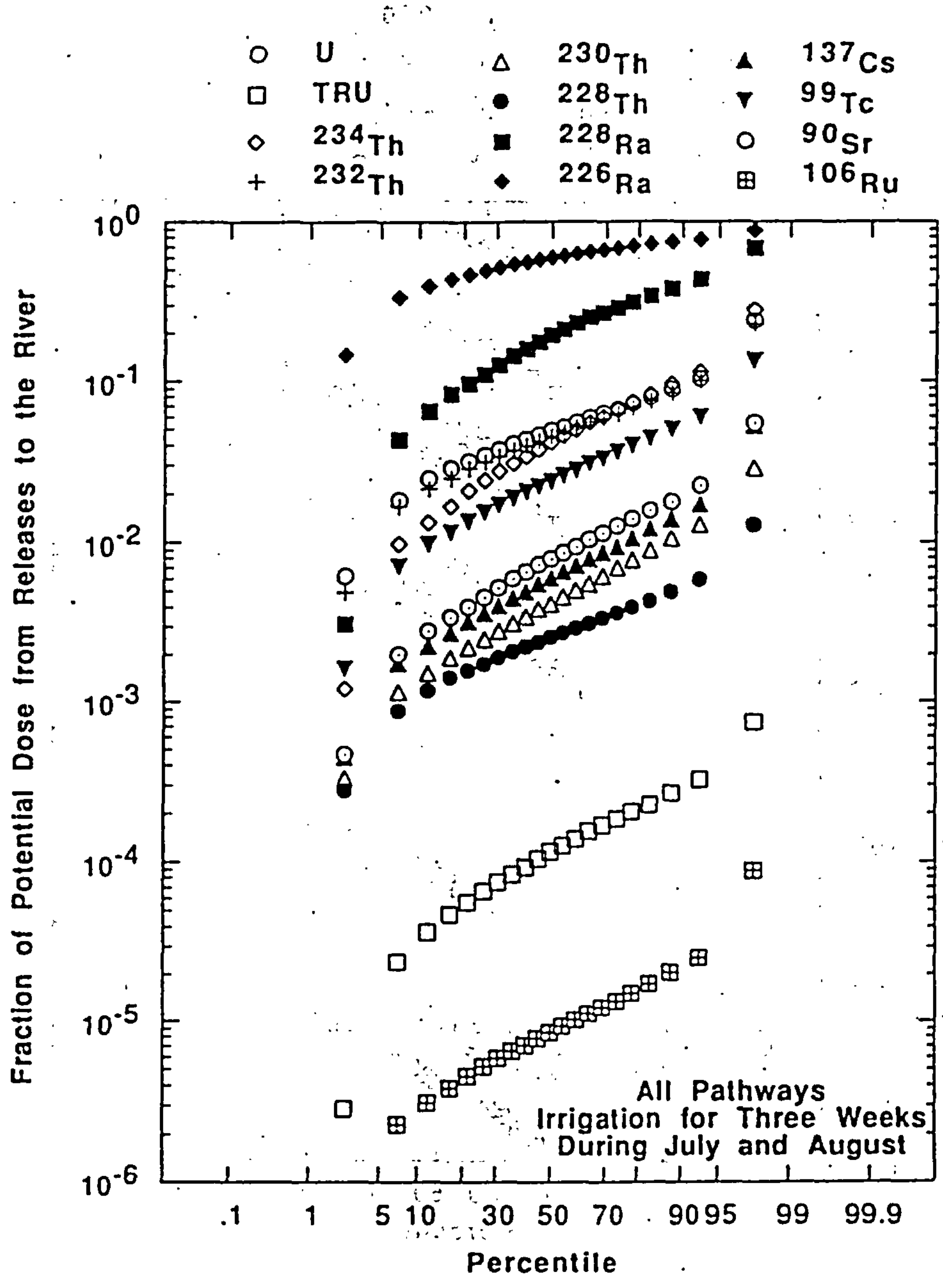

Figure D–5. Contributions of radionuclides released to water to the potential dose from all pathways.

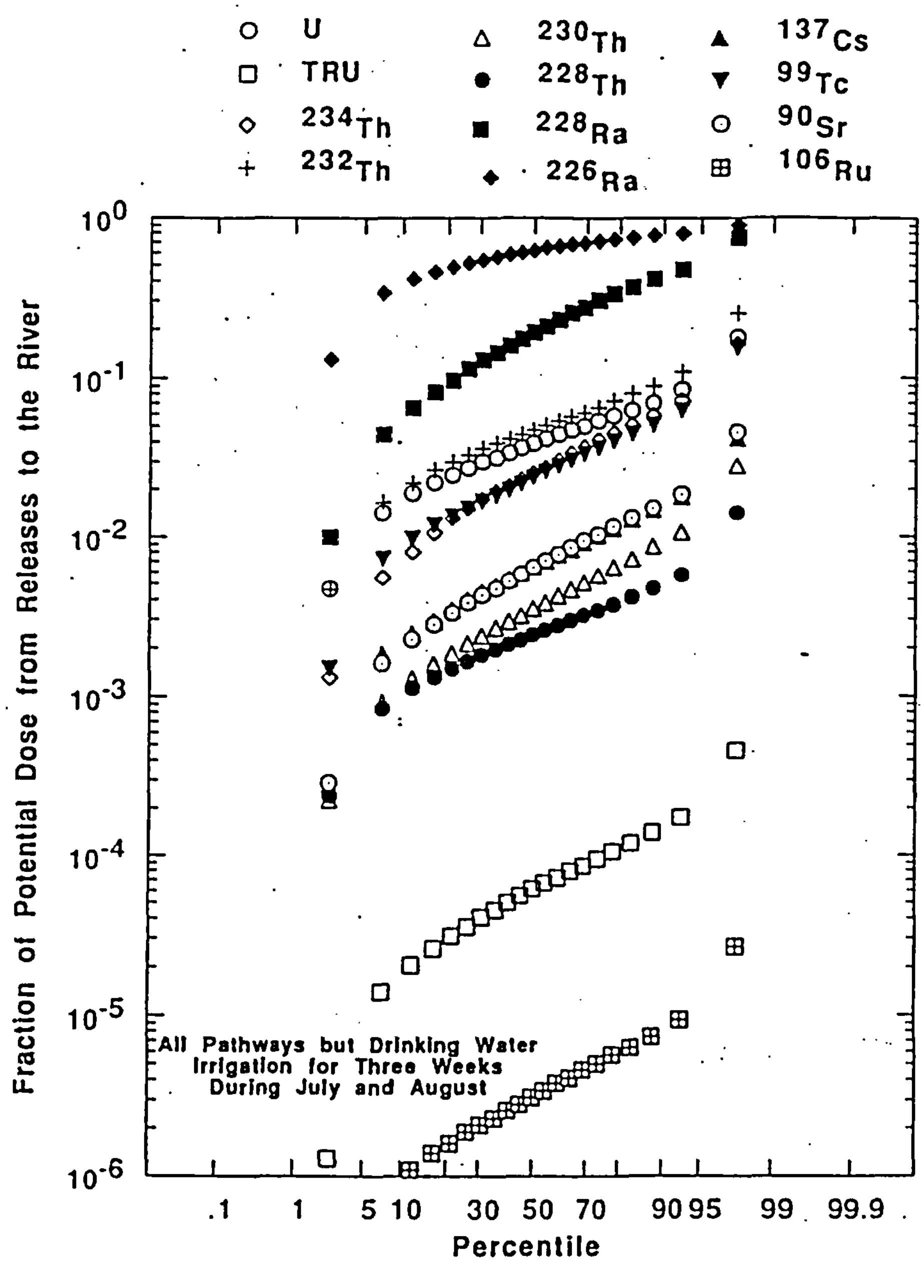

Figure D-6. Contributions of radionuclides released to water to the potential dose from all pathways except drinking water.

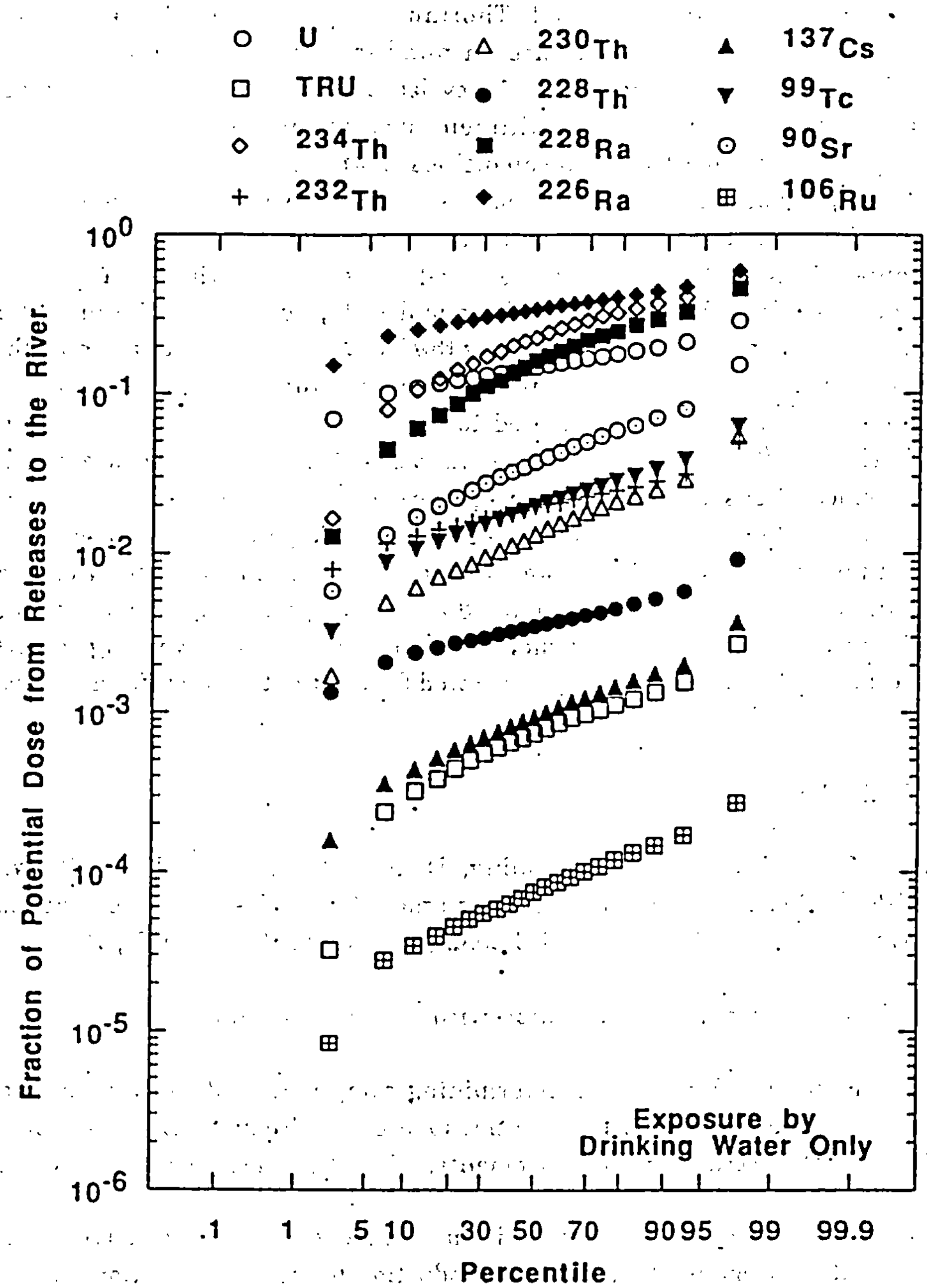

Figure D–7. Contributions of radionuclides released to water to the potential dose from drinking contaminated groundwater or water from the river.

SUMMARY

Uranium was the principal material processed at the FMPC and, on a mass basis, was the primary contaminant released. Thorium was the second largest contaminant on a mass basis. The facility also released a number of activation and fission products that reached the plant as contaminants of recycled uranium. The annex contains a tabulation of receipts of recycled uranium. Effluent monitoring data and the results of special sampling were used to estimate the quantities of other nuclides that were released and their relative importance for dosimetric purposes. The special case of releases of radon and other nuclides from the K-65 silos is treated in Appendix J.

Monte Carlo calculations were used to estimate the relative importance of radionuclides released to the atmosphere and in liquid wastes. The procedure was based upon the screening approach developed by the NCRP. The calculations show that the release of uranium was by far the most important contributor (~85%) to the potential dose from releases to the atmosphere. Estimated to be next in importance for atmospheric releases were ^{232}Th and ^{230}Th. Inhalation was found to be the dominant exposure pathway.

The calculations for liquid releases were more complex. Three exposure scenarios were addressed to reflect various possible water usage patterns. In all scenarios, ^{226}Ra was the most important nuclide; ^{228}Ra was second or third in importance in each case. Overall, the radium isotopes accounted for 50–80% of the potential dose from liquid releases. Specific isotopes of uranium and thorium were found to be of varying importance for the three scenarios, but as a group they accounted for most of the potential dose not attributed to radium.

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ANNEX TO APPENDIX D

The analysis of the relative importance of other radionuclides to radiation doses from releases to the atmosphere at the FMPC showed that uranium was by far the dominant source of dose and that only thorium and transuranic nuclides could make potentially significant contributions to the total dose. In this annex, the results of further investigations into releases of these radionuclides are presented.

RELEASES OF TRANSURANIC NUCLIDES

The FMPC began to receive recycled uranium as enriched UO_3 during the last half of 1961 (Gessiness 1985). Processing of this material in Plant 4 could have occurred during the last half of 1962, but may not have started until FY 1964 or 1965.

Table D1–1 shows the quantities of recycled uranium and plutonium (Pu) received by the FMPC. The quantities of plutonium received in 1961–1964 were estimated using the mean concentrations measured during 1965–1970. The recycled uranium was shipped to the FMPC from several sources and in a variety of forms (Gessiness 1985). The plutonium concentrations in the table are relative to the amount of uranium.

The data show that about half of the plutonium was received in 1980. This material was UO_3 received from Paducah that had originated at the Hanford reservation. The high concentrations of Pu measured in samples from Plants 1, 2/3, and 8 in 1985 (Table D–1) were affected by the processing of the Paducah shipment and are not reflective of processing of the recycled uranium in earlier years. However, concentrations in other facilities appear more representative of historic operations and perhaps a gradual buildup of Pu concentrations over time.

Releases of transuranic nuclides to the atmosphere were estimated using the data in Tables D–1 and D1–1 together with estimates of the uranium releases from particular effluent paths. Except as noted above, the concentration ratios measured in 1985 were taken to be representative of earlier years of operation. The relative amounts of recycled uranium and uranium that had not been previously irradiated were considered, but not on a year by year basis because it was not possible to track when particular batches of material were actually processed.

RELEASES OF THORIUM

Releases of thorium as a contaminant of uranium were based upon the 1985 concentration ratios that were given in Table D–1. Some of these ratios may have been influenced by thorium processing campaigns, but it was not possible to isolate events as specific as the Paducah shipment of high plutonium content that was discussed above and in the main text of the appendix.

Table D1-1. Plutonium Received by FMPC in Recycled Uranium from Various Sources (Data from Gessiness 1985)

Fiscal Year	Recycled Uranium Received (MTU)	Plutonium Received (g)	Plutonium Concentration (parts per billion)
1961	40	0.21[a]	
1962	453	2.4[a]	
1963	367	1.9[a]	
1964	780	4.1[a]	
1965	8.2	0.019	2.318
1966	103	0.698	6.746
1967	413	1.938	4.693
1968	150	0.994	6.624
1969	120	0.805	6.710
1970	1,302	5.305	4.075
1971	68	0.448	6.631
1972	5.8	0.008	1.377
1973	15	0.011	0.737
1974	49	0.123	2.528
1975	37	0.099	2.678
1976	10	0.047	4.526
1977	23	0.007	0.290
1978	15	0.084	5.533
1979	397	2.161	5.439
1980	124	25.512	205.183
1981	423	2.197	5.197
1982	639	3.631	5.680
1983	479	2.207	4.604
1984	838	1.025	1.222
1985	321	0.322	1.002
Total	7,184[b]	56.2[a]	

[a] Estimated using average plutonium concentration between 1965 and 1970.
[b] This total may be compared with 403,000 MTU received during the same period that did not contain plutonium.

Releases of thorium from the processing campaigns are difficult to estimate because of the general lack of information about those activities. The information and production data assembled by Hill and Dolan (1988) was used in making estimates of thorium releases. Normalized release rates for uranium from similar activities were used in calculations of thorium releases. Estimates for the most important activities are shown in Table D1-2. The broad ranges of estimates indicates substantial uncertainty.

Particle sizes for the thorium releases are believed to be comparable to those observed for uranium. A median diameter of about 7 μm with a GSD of about 3 is considered reasonable.

Table D1-2. Estimated Parameters for the Significant Thorium Releases

FMPC Facility	Period of Operation	Chemical Form	Estimated Release Rate (kg Th y^{-1})	Range of Release Rates (kg Th y^{-1})
Plant 9	1954–1956	ThF_4, ThO_2	100	50–200
Plant 4	1954	ThO_2, ThF_4	5	0–10
Pilot Plant	1964–1980	$Th(NO_3)_2$	15	7–30
Pilot Plant	1964–1970	$Th(OH)_4$	50	20–100
Pilot Plant	1969–1971	ThO_2	15	10–30
Pilot Plant	1971–1976	$Th(C_2O_4)_2$	10	5–25
Pilot Plant	1977–1979	$Th(OH)_4$	60	20–100
Plant 8	1966	$Th(OH)_4$	150	50–250
Plant 8	1969–1971	$Th(OH)_4$	400	200–800

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APPENDIX E

EFFLUENTS FROM DUST COLLECTOR EXHAUSTS

INTRODUCTION

Many of the plant processes that were expected to generate airborne particles were serviced by dust collectors. Process area ventilation air was ducted to the collectors where airborne particulate material was removed before discharge. The dust collectors recovered valuable uranium that would otherwise be lost and worker exposure in the process areas was reduced.

A general description of dust collector operation is given in Appendix B. More information is available in pollution control and ventilation handbooks such as Danielson (1973) and CIV (1980). Detailed descriptions of some of the specific systems that were in use at the FMPC are available in ventilation system evaluation reports for the various plants; for examples, see Boies (1965).

When operating as designed, the systems could be quite efficient (Drinker and Hatch 1956, Ross and Boback 1971). However, the effluent sampling program identified many occasions when dust collector performance at the FMPC was not optimal. These cases were documented using sampling systems installed to estimate losses of uranium to the environment. The set of effluent sampling systems and the data they produced are of primary importance in any estimation of effluent releases from the dust collector exhausts. Accordingly, the first sections of this appendix are devoted to a description and analysis of those systems.

The following aspects of the effluent sampling systems that were utilized for FMPC dust collector exhausts are discussed below:

- description of the sampling systems
- operating procedure
- sample analysis
- reports of results

After this introductory information, the historic dust collector effluent measurement results, taken from monthly reports, are presented. The reported releases based on those sampling systems were sometimes incomplete. The reasons for those deficiencies and remedies to them are discussed. Simple interpolation was sufficient to estimate releases when results were unavailable for short periods. Normalized release rates were employed for periods prior to implementation of a routine program of effluent monitoring.

While the design of the sampling systems was generally well conceived, there are possible biases in the results that were given in routine reports. The potential biases are described and methods for estimating the magnitudes of the biases are discussed.

The sizes of particles in the effluents being sampled have an important bearing on the degree of bias in the reported results. Particle size distributions for some of the effluent streams were measured in 1985. Those data and information about other uranium processing facilities have been used to estimate particle size distributions for the dust collector exhausts; the results of that effort are presented. In addition, the chemical forms of materials discharged from the dust collectors are summarized. The chemical form is a determinant of particle density and affects the sampling bias. The transport and deposition of released uranium and the estimation of the radiation dose due to uranium inhalation are also dependent upon particle size and density.

Even with estimates of the particle size distribution and chemical form of the effluent, there is currently insufficient information to make definitive adjustments for sampling bias. The overall sampling bias was estimated using Monte Carlo techniques. These were used to make release estimates for this study. Those estimates, together with the associated uncertainties, are presented in the last section of this appendix.

DUST COLLECTOR EFFLUENT SAMPLING SYSTEMS

The sampling systems installed in the dust collector stacks were simple in concept. Air was drawn from the exhaust duct to a pleated filter for collection of particulate material in the sample of discharged air. The filters were periodically changed and submitted for analysis. Details of the design and operation of these systems and of the sample analysis and data reporting are given below.

Sampler Design and Installation

Design of the sampling systems was generally well conceived and consistent with guidance for good sampling practices. Important features of sampler design and installation (Starkey 1956, Boone 1956b, Bipes 1961) were:

- isokinetic sampling — the air velocity through the sampling probe was designed to be the same as that in the exhaust duct at the sampling location to avoid over- or under-sampling particles of various sizes

- proper location — sampling probes were to be installed 7 to 10 stack diameters downstream of the exhaust fan or major bend with 2 to 3 stack diameters of straight ductwork beyond the sampling point

- short sample lines — the filter holder was located outside the stack at an elevation near that of the sampling probe, so total line lengths were generally less than one meter.

A simple schematic diagram of the sampling system is shown in Figure E–1. It is only intended to illustrate the basic components of the system.

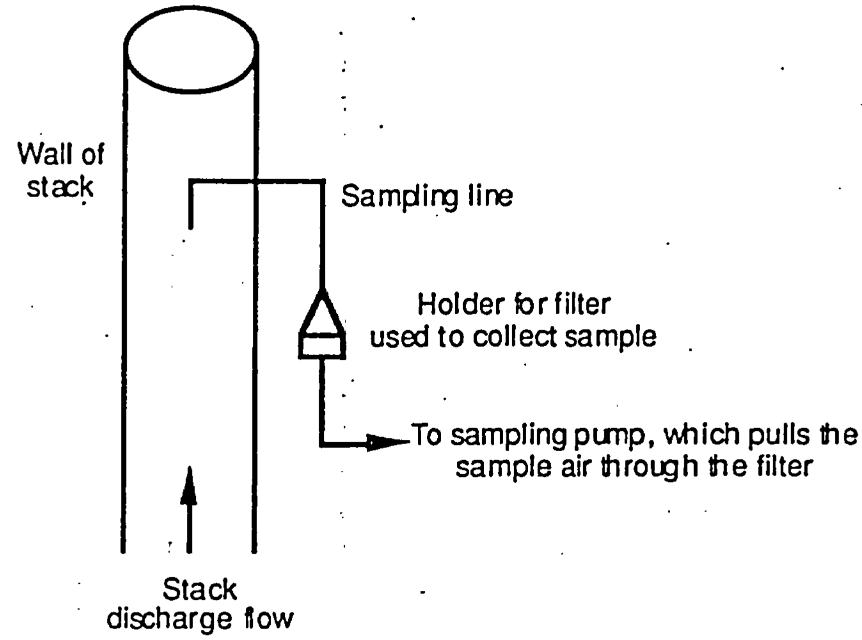

Figure E-1. Schematic diagram of dust collector stack sampling system. Not shown are the support piping outside the stack or the rain caps that were atop stacks.

The design features identified above were all consistent with consensus guidance for stack sampling installations (ANSI 1969). Initially, a preference for sampling locations in laminar flow was indicated (Boone 1956b); however, the feasibility of satisfying that criterion was limited and it was not present in the later installation procedure (Bipes 1961). Other probe location goals may not have been achieved in practice. The September 1956 procedure recognized that compromises may be necessary with regard to sampling location. It guided the installer to approach the optimal location "as nearly as possible" (Boone 1956b). Earlier guidance circulated by Starkey (1956) indicated that it would rarely be possible to satisfy the probe location guidance without locating the sampling probes on the roof. Some samplers were located outside, but the majority were not. The implied limitation on probe placement would likely mean that the probe was closer to the air mover or to a bend in the stack than was recommended.

The initial sampling probe design included a tapered inlet nozzle that had an internal diameter of 0.95 cm (Starkey 1956). Recent investigations (ORAU 1985) revealed that not all of the inlets were tapered, which may have added to impaction loses in some sampling lines. The initially suggested sampling line diameter appears to have been modified to 0.62 cm at an early stage in the development (Boone 1956a, Boone 1956b).

Prior to installation of the probe in the duct, a pitot tube traverse of the stack was performed to determine the air velocity at several points in the stack. Traverse data were obtained periodically after installation as well. This information was necessary to

Prior to installation of the probe in the duct, a pitot tube traverse of the stack was performed to determine the air velocity at several points in the stack. Traverse data were obtained periodically after installation as well. This information was necessary to determine the proper flow rate for isokinetic sampling and for the calculations of releases from the sampling data (Starkey 1956, Boone 1956b, Bipes 1961).

A valve in the line to the vacuum source was provided to make adjustments to the sampling flow rate. Such adjustments were normally made when samples were changed but there was no mechanism for assuring a constant flow rate during the sampling period.

Operating Procedures

Some effluent sampling was performed in the Pilot Plant in 1953, but a routine program for measuring discharges from the facilities was not begun until 1955. Distribution of the initial stack sampling procedure to all the plants occurred in February 1956 (Starkey 1956). Later that year a formalized procedure was promulgated (Boone 1956b). Initial sampling frequencies were weekly, biweekly, or monthly depending on the magnitude of the previous effluent measurements. Pleated "Type S" cellulose filters were used to collect the particulate material. The procedure called for measurement and documentation of the flow rate at the end of the sampling period prior to removal of the filter. The exposed filter was taken from the filter holder, placed in a marked bag, and sealed for delivery to the laboratory. A new pre-weighed filter was placed in the holder. The flow rate was set to the value required for isokinetic sampling and the system was reassembled.

Sample Analysis

Tare weights of filters were determined before they were placed at sampling locations. Exposed filters were weighed to determine the total mass of material collected. If sufficient mass was present, the sample was analyzed for uranium content. Otherwise, the uranium fraction of the total mass was estimated using previous measured uranium fractions for the same exhaust. Laboratory results were provided to the Industrial Hygiene and Radiation (IH&R) group on standard analytical data sheets. If an exposed filter was wet, the normal procedure appears to have been to analyze it for total uranium content. The total amount of material releases and the uranium fraction were not reported in such cases.

Reports of Results

Monthly reports of releases were made to plant management by the IH&R group. These reports usually included results for all the measurement periods during the month. Estimates of releases from the dust collector exhausts at the FMPC relied on isokinetic sampling of the stacks. When the probe velocity (u, cm s^{-1}) and the stack fluid velocity (v, cm s^{-1}) are equal, there is a very simple relationship between the mass of material released from the stack and the mass of material collected on the filter. That relationship, which involves the flows through the sampling probe and the stack, was used to estimate releases from the dust collector exhausts. Because the flow through a tube is the product of the fluid velocity and the cross-sectional area, one way to express the relationship is as follows:

$$Q_r = [v_a / v][A_s / A_p]M_f \qquad \text{(E-1)}$$

where Q_r is the amount of material released (g), v_a is the average fluid velocity (cm s^{-1}) in the stack, v is the fluid velocity at the stack centerline (the point of sampling), A_s and A_p are the areas (cm^2) of the stack and the probe, respectively, and M_f is the amount of material (g) found on the filter. Implicit in Equation (E-1) is the assumption that $u = v$, which reflects the fact that the effluent sampling systems were designed to operate isokinetically. An equivalent expression for Q_r is

$$Q_r = [F_s^* / F_p^*]M_f \qquad \text{(E-2)}$$

where F_s^* and F_p^* are the current "standard" flow rates in the stack and sampling probe, respectively. That is, F_p^* is the computed sampling flow rate that would provide isokinetic sampling for a stack whose measured flow rate was F_s^*. The value of F_p^* for a given stack was adjusted when a new value for F_s^* was obtained from pitot tube measurements in the stack.

When a sample weight was determined but the uranium content was not, it was common practice to assume the last measured value of the fraction of the dust that was uranium to convert the mass released to the amount of uranium released. Sometimes grab samples of the collected dust were analyzed to determine the uranium fraction of the dust.

The monthly reports, which are still available, also contained comments regarding the operation of the facilities, the dust collectors, and the sampling systems. These notes indicate the difficulties that were encountered by the IH&R staff in implementing the sampling program. The discussions related to plant operating conditions are very useful for reconstructing the history of a particular release point.

Early History of the Sampling Program

The sequence of monthly reports documents the onset and growth of the dust collector effluent sampling program. Periodic sampling of some stacks was performed as early as 1953; however, the continuous sampling program did not begin until April 1955. Initiated in seven stacks in Plants 4 and 5, the sampling program grew fairly rapidly to encompass thirty stacks six months later. Subsequent growth was more gradual, as is shown in Figure E-2. Small changes from month to month may reflect either sampling problems or changes in plant operations. The sharp increase early in the second year was due to the installation of more sampling systems in Plant 8. The following month, operation of Plant 7 was terminated and those sampling systems were taken out of service. During the next two years the sampling program grew gradually to a maximum of 50 sampling systems in May 1958. The decline in number of systems after that time was due to the shutdown of systems in Plant 1 and in the Pilot Plant.

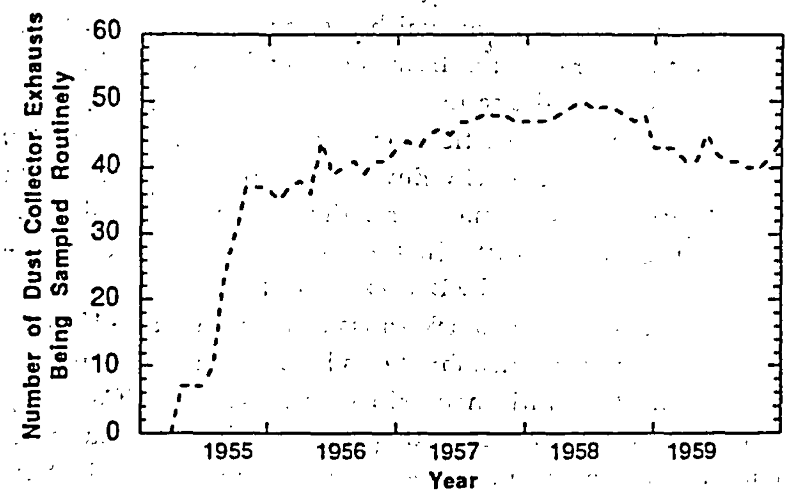

Figure E-2. Growth of the dust collector exhaust stack sampling program during the first years of operation.

At the start of 1960, there were 44 dust collector exhaust sampling systems in operation at the FMPC. At that time, the most common sampling interval was one month, although a few stacks were sampled more frequently. Typically, the time resolution of the data varied from 3–7 days for exhausts with the highest release rates to approximately four weeks for systems with the lowest release rates. When the staff were trying to determine whether a malfunctioning dust collector had been repaired, sampling periods as short as a few hours were used. In the 1960s, sampling intervals were occasionally as long as six weeks for discharge points that were minor contributors to plant uranium releases.

Both plant production and staff were reduced in later years. Intervals between sample analyses were greater and routine reports contained less detail. Filters were no longer changed and analyzed regularly. Instead, sampling systems and filters were inspected routinely, but filter changes and analysis occurred primarily when the filter had collected a visually detectable amount of particulate material.

PARTICLE SIZE DISTRIBUTIONS AND CHEMICAL FORMS OF RELEASES

The chemical and physical characteristics of the uranium that was released to the atmosphere are important for four reasons. In the present context, particle size and density, which is related to chemical form, are important determinants of the transmission factors and of the magnitude of anisokinetic sampling bias. In addition, the chemical form determines the mobility of uranium inhaled by humans and affects its distribution in and clearance from the body. Physical characteristics, primarily particle size, affect two important processes. The size and shape of the particles are both parameters that affect the deposition of discharged radionuclides. In addition, aerodynamic particle size is an important determinant of the fate of an inhaled aerosol in the human respiratory tract.

Larger particles are collected in the upper regions while very small particles penetrate further along the bronchial airways. Many of the larger particles will be swallowed and enter the gastrointestinal (GI) tract from which the radionuclides may be absorbed into the blood. The smaller particles are cleared from the lung directly to the blood and to the GI tract where uptake to blood may also occur.

The only measurements of the particle sizes of stack emissions from the FMPC were conducted in 1985 by Northern Kentucky Environmental Services (NKES) (Reed 1985). In the NKES study, measurements were made for both the inlet ducts and the outlet ducts of 15 major uranium-emitting stacks with dust collectors. The particle-size distributions determined in the study were reported by Boback et al. (1987).

Particle-size distributions for the stack emissions measured in 1985 are included as a part of the source-term characterization for stacks because the plant processes served by the stacks have not changed significantly since the start of FMPC operations. The hydrofluorination process for producing UF_4 (green salt), for example, has remained basically the same over the years with respect to conditions which might affect the particle size distribution of the product. Similarly, the various plant operations which produce U_3O_8 particles also have not changed in a manner which would significantly alter particle size distributions.

The particle size data given in Boback et al. (1987) for inlets to and outlets from the dust collectors have been consolidated in Appendix F, which contains plots of the reported measurement results. Some of the distributions deviate substantially from the expected lognormal shape. For convenience in calculations, polynomial functions have been fit, using least squares techniques, to the reported distributions. These functions, also given in Appendix F, permit computer calculation of the portion of the aerosol in a particular size interval. Particle size distributions for the outlet ducts (or emission stacks) are representative of emissions from dust collectors with intact bag filters. However, when bag failures permit unfiltered air to escape to the atmosphere, the distributions of particles in the inlet ducts would be more representative of the releases.

The predominant uranium species released from each stack was identified from FMPC reports and engineering drawings of process equipment. In some cases more than one uranium species was determined to be emitted from a stack. Nearly all of the dust collector exhaust stacks evaluated by NKES emitted either UF_4 or U_3O_8. One of the stacks studied discharged a mixture of UO_2 and UO_3.

To verify them, results presented by Boback et al. (1987) were compared against original NKES data. That process and resolution of the questions that arose from it are discussed below.

Verification of Particle-Size Measurements

Verified particle size measurements are those for which the reported results are consistent with the original data and which meet the test of physical reality. The latter test is simply the question of whether, as expected, the particle size for the outlet duct of a specific dust collector is less than that for the inlet duct over the entire range of measurements.

Most of the particle sizes listed in Boback et al. (1987) were verified in accordance with these criteria, but discrepancies and omissions were found in some cases. Unverified values were not considered representative of specific stack emissions.

Discrepancies were found for the outlet ducts of G5–251, G5–253, and G5–260. The particle size distributions as reported by Boback et al. (1987) for these cases are not consistent with the original NKES data sheets. The reported distributions were derived from modified data sheets of uncertain origin.

Measured particle sizes for the outlet ducts of G4–5 and G43–27 were greater than those for the corresponding inlet ducts, which is physically unrealistic. The particle size data for the inlet duct of G5–251 were also difficult to accept because they indicated smaller particles than those for the outlet duct for the same collector. However, the latter values were not verified (see above). It was also found in the verification process that reported values (Boback et al. 1987) for the larger particle sizes in the distributions in inlet ducts of G5–254 and G5–256 seem to contain relatively small systematic errors (5–10%). These errors have been corrected and the revised values are included in the verified results.

Table E–1 contains the results of particle size measurements that were verified as part of this study. The calculations of sampling bias employ ten distinct particle sizes — the 5th, 15th, 25th, . . ., and 95th percentile values — to represent the distribution for the dust collector exhaust of interest. These are given in the table. The same calculations also require information on particle density, so the chemical form of the discharged uranium is also of interest. This information has been included in Table E–1 for each duct. The same chemical form assignment applies to both the inlet and outlet ducts.

It should be noted that the bag filters of the dust collectors for FMPC stacks were not all made from the same material during the 1985 NKES study. Some of the dust collectors had wool felt bags, but a change to Gore-Tex bags was in progress over the period of years which included 1985. There were too few stacks with the same uranium species and different bag types to draw definite conclusions about differences in bag collection efficiencies for specific particle sizes ranges.

Inferred Particle Sizes for Other Stacks

The particle size distributions for emissions from some stacks for which no measurements had been made were inferred from the results obtained by NKES (Reed 1985). This was accomplished by relating the uranium chemical species and plant operation(s) serviced by an unstudied exhaust to those of exhausts for which measurements had been made.

The particle size distributions of the stacks which emitted UF_4 produced by the hydrofluorination process were averaged, and this average distribution was assumed to apply to all stacks emitting UF_4 also produced by hydrofluorination but for which reliable measured values are not available. Estimation of the average distributions for UF_4 and a similar average for U_3O_8 are described in a subsection below.

Table E-1. Summary of Verified Information on Particle Size for Dust Collectors

			Equivalent diameter (µm) at specified percentile									
Stack	Form	[a]	5	15	25	35	45	55	65	75	85	95
G4–2	UF_4	O	1.5	4.3	6.1	7.6	9.0	10	12	14	16	20
		I	2.6	4.2	5.3	6.3	7.3	8.3	9.5	11	13	17
G4–5	UF_4	O	0.56	1.4	2.5	3.8	5.4	7.4	10	14	21	36
		I	1.0	1.9	2.7	3.6	4.6	5.9	7.7	10	15	27
G4–7	UO_2	O	0.80	1.6	2.8	4.7	7.3	11	15	20	27	41
G4–12	UF_4	O	2.5	4.5	5.8	6.9	8.0	9.0	10	12	13	17
		I	3.4	5.5	6.9	8.1	9.2	10	11	13	15	18
G4–14	UF_4	O	0.92	3.1	5.0	6.7	8.4	10	12	15	18	24
		I	5.4	8.1	9.9	12	13	14	16	18	20	25
G5–249	UF_4	O	0.13	0.29	2.4	4.6	6.4	8.0	10	12	15	19
		I	2.7	5.2	6.9	8.3	9.7	11	13	14	17	21
G5–250	UF_4	O	0.66	2.4	4.1	5.8	7.5	9.3	11	14	18	25
		I	5.9	8.8	11	12	14	15	17	19	22	27
G5–251	UF_4	I	0.28	0.35	0.40	0.45	0.51	0.58	0.69	0.85	1.2	6.0
G5–253	UF_4	I	0.89	3.4	5.4	6.9	8.4	9.7	11	13	15	19
G5–254	U_3O_8	O	0.63	1.4	2.5	3.7	4.8	5.9	7.2	8.7	11	14
		I	1.7	3.0	4.2	5.5	7.0	8.8	11	15	21	37
G5–256	U_3O_8	O	0.48	0.84	1.6	3.0	4.6	6.1	7.7	9.5	12	16
		I	0.75	3.2	4.8	6.2	7.4	8.6	9.8	11	13	17
G5–260	U_3O_8	I	1.5	3.1	4.2	5.3	6.4	7.6	9.0	11	13	18
G5–261	U_3O_8	O	1.1	2.9	4.2	5.4	6.5	7.7	8.9	11	13	16
		I	2.9	5.3	7.0	8.6	10	12	14	16	19	26
G43–27	U_3O_8	I	1.1	2.6	4.0	5.3	6.7	8.3	10	13	16	23
G9N1–1039	U_3O_8	O	0.34	0.47	0.61	0.79	1.1	1.9	3.8	6.0	8.7	13
		I	1.0	2.3	3.4	4.2	5.0	5.8	6.6	7.6	8.8	11

[a] Distributions are given for the outlet (O) and inlet (I) of the dust collector.

Airborne U_3O_8 is produced in the FMPC as a result of the oxidation of uranium metal surfaces by air. There are two general types of plant operations which can produce airborne U_3O_8 particles:

- foundry operations such as melting and casting of uranium metal, breakout of the uranium derbies and ingots from crucibles, and cleaning of metal surfaces
- the machining of uranium derbies and ingots.

The NKES study included only stacks which served foundry operations in Plant 5. The average particle size distribution based upon the U_3O_8 emission points that were evaluated was assumed to apply to all stacks exclusively serving foundry operations for which no measurements had been made. Surface oxidation of uranium scrap in high-temperature furnaces such as took place in Plant 8 was assumed to be in the same category as foundry operations.

Distributions of particle size for machining operations were inferred from other sources of particle size data. These are presented in a the second subsection. Particle sizes for emissions from dust collectors in Plants 1 and Plant 2/3 were also inferred from other sources and are discussed separately below. In the last subsection, the issue of particle sizes for UF_4 produced by reduction of UF_6 is addressed.

Calculation of Average Distributions for UF_4 and U_3O_8. The average particle-size distributions for both the inlet ducts and the outlet ducts for stacks emitting UF_4 and U_3O_8 were derived from the data in Appendix F. Table E–2 gives the verified particle size distributions for UF_4 in six outlet ducts. The average distribution derived from the six sets of measurements of this type is also shown. Table E–3 contains the verified distributions of UF_4 measured in the inlets to seven dust collectors and the average distribution derived from those measurements. Tables E–4 and E–5 contain the verified and derived average distributions for U_3O_8 in three outlet and four inlet ducts, respectively. In all four tables, results are given in terms of the equivalent aerodynamic diameter, defined as the diameter of a sphere of unit density (1 g cm^{-3}) that has the same gravitational settling velocity as the particle (also assumed to be spherical).

Table E–6 contains the median particle sizes for the inlets and outlets of dust collectors handling UF_4 from hydrofluorination in Plant 4 and U_3O_8 from foundry operations in Plant 5. Also shown in the table are particle sizes of U_3O_8 in air measured during foundry operations at Los Alamos (Hyatt et al. 1959) and at two facilities in the United Kingdom (Vallis 1991; Fishwick 1991). These results agree reasonably well with dust collector inlet values from Plant 5 at the FMPC.

As noted previously, the measured distributions deviate from lognormality and the composite distributions are also not truly lognormal. However, if the central portion of the distribution is used to make an estimate, geometric standard deviations (GSDs) of the composite distributions for the FMPC are about two. A GSD of two was quoted by Fishwick (1991) as typical of the measurements at Springfields.

Table E-2. Size Distributions for UF_4 in Dust Collector Outlet Ducts

Plant	Stack	Percentage of particles in specified size range (µm)[a]						
		0–2.5	2.5–5.0	5.0–7.5	7.5–10	10–15	15–20	20–40
4	G4–2	8.0	12	15	16	29	15	5.0
	G4–5	25	17	14	8.0	13	7.0	16
	G4–12	5.0	15	22	23	25	8.5	1.5
	G4–14	12	13	15	15	21	14	10
5	G5–249	25	13	15	13	19	10	4.5
	G5–250	16	14	15	14	16	14	10
Average		15	14	16	15	20	12	7.8

[a] Ranges are given for equivalent aerodynamic diameters of particles.

Table E-3. Size Distributions for UF_4 in Dust Collector Inlet Ducts

Plant	Stack	Percentage of particles in specified size range (µm)[a]						
		0–2.5	2.5–5.0	5.0–7.5	7.5–10	10–15	15–20	20–40
4	G4–2	5.0	17	26	22	22	5.5	2.5
	G4–5	23	27	14	10	11	5.0	10
	G4–12	3.5	8.5	18	24	29	14	3.0
	G4–14	0.8	3.2	8.0	14	34	22	18
5	G5–249	4.5	9.5	15	20	29	15	7.0
	G5–250	0.7	2.8	6.5	12	28	30	20
	G5–253	12	10	17	18	27	12	4.0
Average		7.1	11	15	17	26	15	9.2

[a] Ranges are given for equivalent aerodynamic diameters of particles.

Table E-4. Size Distributions for U_3O_8 in Outlet Ducts of Dust Collectors Serving Foundry Operations

Plant	Stack	Percentage of particles in specified size range (µm)[a]						
		0–2.5	2.5–5.0	5.0–7.5	7.5–10	10–15	15–20	20–30
5	G5-254	24	22	21	15	10	7.2	0.8
	G5-256	32	16	16	13	17	5	1
	G5-261	13	18	23	19	19	6	2
Average		23	19	20	16	15	6.1	1.3

[a] Ranges are given for equivalent aerodynamic diameters of particles.

Table E-5. Size Distributions for U_3O_8 in Inlet Ducts of Dust Collectors Serving Foundry Operations

		Percentage of particles in specified size range (µm)[a]						
Plant	Stack	0–2.5	2.5–5.0	5.0–7.5	7.5–10	10–15	15–20	20–35
5	G5-254	16	22	14	14	14	8.0	12
	G5-256	5.0	16	24	17	26	10	2.0
	G5-260	11	20	22	18	18	6.5	4.5
	G5-261	4.0	10	13	16	27	16	14
Average		9.0	17	18	16	21	10	8.1

[a] Ranges are given for equivalent aerodynamic diameters of particles.

Table E-6. Composite Median Particle Sizes in Plants 4 and 5 and Some Data From Other Facilities

Species	Source	Location	Median size (µm)
UF_4	Hydrofluorination in Plant 4	Inlet to dust collector	9.5
		Outlet from dust collector	8.1
U_3O_8	Foundry operations in Plant 5	Inlet to dust collector	8.3
		Outlet from dust collector	6.0
	Los Alamos foundry	Airborne particles	7.3
	Aldermaston, UK foundry	Airborne dust in workshop	9.7
	Springfields, UK foundry	Workplace air	5–11

The particle size distributions for dust collector stacks for which no measurements are available were inferred from the available data. Dust collectors handling UF_4 produced by hydrofluorination were assigned the composite distribution for that species. Stacks serving foundry operations were similarly assigned the distribution for U_3O_8 from Table E-6. Estimates for uranium machining operations are discussed below.

Inferred Particle Sizes for U_3O_8 Produced During Machining. Machining operations such as cutting and milling of uranium metal ingots and derbies were conducted in Plant 6 and Plant 9. Studies in other facilities have estimated particle size distributions for releases from machining operations. Hyatt et al. (1959) reported an AMAD of 6.7 µm with a geometric standard deviation (GSD) of approximately 2.7 for uranium machining operations at Los Alamos. A median particle size of 6.9 µm was reported for similar operations at Aldermaston in the United Kingdom (Vallis 1991). The GSD for the Aldermaston distribution was stated to be approximately 3. The distributions are quite consistent considering the great differences in time and location.

Particle size measurements were made for one stack in Plant 9 in 1985. The inlet median diameter was 5.4 µm for dust collector G9N1-1039. The reported median particle size for the outlet was about 1.5 µm, which was atypical of the FMPC results. The reason for the large reduction in size between inlet and outlet of this dust collector is not known.

For other discharges from dust collectors in Plants 6 and 9, a median diameter of 6.8 µm was assumed to apply to inlet ducts for dust collectors serving machining operations in Plant 6 and Plant 9 at the FMPC. A median diameter of 5.1 µm is estimated to apply to the outlet ducts for those operations. This reflects the nominal 25% reduction in median particle size seen in most of the FMPC measurements.

Inferred Particle Sizes for Emissions from Plant 1 and Plant 2/3. A mixture of particles of U_3O_8, UO_3, and UO_2 is assumed to be present in the discharges from dust collector stacks in Plant 1 and Plant 2/3. Those collectors serve areas handling ores and various other feed stocks for the digestors. Because the 1985 NKES study did not include any stacks for these plants, particle-sizes for these emissions must be inferred from measurements made for similar operations elsewhere.

A study of particle sizes of uranium-containing dust from mining and milling operations was performed in the Elliot Lake Area of Canada (Duport and Edwardson, 1985; Duport and Horvath, 1989). Those authors reported AMADs of mill atmosphere aerosols for several processes as shown in Table E–7.

Table E–7. Results of Particle Size Measurements for Uranium Milling Processes

Process	Median size (µm)
Jaw crushing	9.5
Cone crushing	9
Screening	7.5
Grinding	8
Acid precipitation	6
Filtering	10
Concentrate drying	8
Concentrate packing	7.5

The average AMAD for mills (possibly a weighted average) was reported to be about 7 µm. The GSDs for the particle size distributions given in Table E–7 ranged between 3 and 5 (Duport and Hovarth 1989).

On the basis of the data cited above, a median particle diameter of 7 µm with a GSD of 4 is assumed for the U_3O_8 dust emitted from Plant 1 and Plant 2/3 as a result of ore handling. This inferred value applies to inlet ducts of the dust collectors. For the exhaust stacks, a median value of 5.3 µm is assumed. As before, this reflects a nominal 25% reduction in the median particle size due to filtration in the collectors.

Particle Sizes for UF_4 Produced by Reduction of UF_6. There were two sources of releases of UF_4 produced by reduction of UF_6. This process was developed and modified in the Pilot Plant (Davis et al. 1956). During a 2-year period of operation, Plant 7 produced UF_4 in larger reactor vessels that were modeled after the one in the Pilot Plant. Dust collectors in both facilities released product material during operations.

Process particle size measurements were made during the development process (Davis et al. 1956). The system to remove HF from the offgas employed two cyclone collectors, two filters, and a KOH scrubber. Particle size data from the two cyclones indicate median particle sizes between 9 and 11 µm. These measurements are consistent with the median of the composite UF_4 size distribution for dust collector inlets (Table E–7), indicating that the particle size of airborne UF_4 is not strongly dependent upon the production process. The composite particle size distributions for UF_4 were used for the Pilot Plant and Plant 7 dust collector systems that handled UF_4.

Summary of Inferred Particle Size Distributions. As noted previously, it is convenient to summarize information on both particle size and chemical form together. Table E–8 contains the information for the composite and inferred particle size distributions just discussed. Two particle size distributions are given, one for the inlet (I) to the dust collector and one for the outlet (O). The chemical form is the same for both the inlet and the outlet. The dust collectors to which the composite distributions were applied are listed in the footnotes to the table. Particle size distributions that were derived from other sources are given for the machining operations in Plants 6 and 9 and for operations with uranium feed stocks that generated airborne dusts in Plant 1 and Plant 2/3.

Table E–8. Composite and Inferred Particle Size Distributions

			Equivalent diameter (µm) at specified percentile									
Stack	Form	[a]	5	15	25	35	45	55	65	75	85	95
Composite[b]	UF_4	O	1.0	2.6	4.2	5.8	7.5	9.4	11	14	17	22
		I	2.0	4.2	6.0	7.8	9.4	11	12	15	17	23
Composite[c]	U_3O_8	O	0.6	1.2	2.8	3.9	5.1	6.5	7.9	9.6	12	16
		I	1.8	3.4	4.9	6.3	7.8	9.2	11	14	17	24
Plant 6 Plant 9	U_3O_8	O	0.82	1.6	2.4	3.3	4.4	5.8	7.8	11	16	32
		I	1.1	2.2	3.3	4.4	6.0	7.8	10	15	22	43
Plant 1 Pl. 2/3	U_3O_8	O	0.63	1.3	2.1	3.1	4.5	6.4	9.1	14	21	52
		I	0.7	1.7	2.8	4.2	6.0	8.5	12	18	30	72

[a] Distributions are given for the inlet (I) and outlet (O) of the dust collector.
[b] Composite UF_4 distribution applied to dust collectors G4–1, G4–4, G4–5(I), G4–8, G4–13, G4–15, G4–7001, G5–251(I), G5–252, G5–253(O), G20–20, G4–2507, G4–2508, G4–2509, G4–2510.
[c] Composite U_3O_8 distribution applied to dust collectors G5–247, G5–248, G5–258, G5–259, G5–260(O), G55–E100, G5A–100, G5A–101, G43–27 (O) and other U_3O_8 discharges from Plant 8 and the Pilot Plant.

PREVIOUS RELEASE ESTIMATES

In this section, previous release estimates, based primarily on routine operational measurements, are presented. The deficiencies in these estimates and possible biases in the reported values are discussed.

Routine Measurements

Results of the measurements described above were reported routinely by the IH&R staff at the FMPC. Although monthly reports were prepared, the period for which data were presented did not correspond to the beginning and end of the calendar month. Typical reporting periods began and ended between the 20th and 25th day of the month. The results presented in a particular report could cover sampling periods with greater variation in start and stop times, depending upon which analyses were completed by the date of report preparation. Copies of many of those reports have been retained to the present day.

Previous FMPC release estimates for the dust collection systems gave annual totals that were largely based upon those reports. Table E–9 contains the annual releases from dust collector exhaust presented by Boback et al. (1987). The reported releases for each plant reflect samples collected from as many as eighteen different dust collector exhausts.

About 35% of the uranium discharges from dust collectors reported by Boback et al. (1987) came from Plant 4. Plant 5 was estimated to contribute about 28% of the total. Although it operated for only two years, Plant 7 was estimated to have released about 14% of all the uranium discharged by dust collectors. Plant 8 (with 11%) was the only other facility estimated to contribute more than ten percent of the total. The other five facilities were estimated to have made minor contributions. None accounted for as much as 4% of the total and the group was estimated to contribute about 12%.

Deficiencies in Reported Release Estimates

There are two major deficiencies in the tabulations of reported releases in the monthly reports that form the basis for Table E–9. The first and most important is that the data are incomplete. In the early years of operation, no release estimate for a particular duct was made until a sampler was installed. Annual totals for those years must therefore be viewed with caution. There were also no estimates for times when the sampler malfunctioned during the sampling period. In later years, when production declined from the peak years in the early 1960s, the level of detail in the monthly reports was greatly decreased. Sample filters were changed much less frequently and detailed information about the sampling program was no longer included.

The second deficiency in the tabulations was the failure to account properly for undetected releases. If no material was detected on the filter from a dust collector exhaust sample, the reported release was shown as zero. Actually, the release was between zero and an upper bound computed using a variant of Equation (E–1):

$$Q_{rm} \leq [v_a / v][A_s / A_p] MDA_f \qquad \text{(E–3)}$$

Table E–9. Releases of Uranium from Dust Collectors Reported in FMPC-2082 (Boback et al. 1987)

	Reported releases (kg U) from FMPC facilities									
Year	1	2/3	4	5	6	7	8	9	Pilot	All
1951	0	0	0	0	0		0		123	123
1952	0	0	0	0	6		0		493	499
1953	4	6	1473	90	12		0		493	2,078
1954	46	71	5,890	4,119	28	4,261	201		271	14,887
1955	46	363	12,450	10,410	53	7,268	877		443	31,910
1956	43	228	5,145	3,501	27	1,743	1,316		32	12,035
1957	49	980	814	3,664	35		791	0.4	18	6,352
1958	407	220	661	715	161		875	679	27	3,745
1959	46	119	1,428	478	127		260	417	36	2,911
1960	20	213	212	203	269		298	219	718	2,152
1961	53	67	262	76	118		209	67	174	1,026
1962	14	67	703	356	77		618	135	174	2,144
1963	83	0	1,469	783	163		994	159	52	3,702
1964	24	0	545	330	34		1,051	252	13	2,249
1965	4	13	335	226	43		390	68	10	1,089
1966	16	54	228	77	11		328	49	18	781
1967	26	27	280	148	3		417	76	12	989
1968	1	10	267	88	30		900	121	4	1,421
1969	35	8	49	119	3		424	13	4	655
1970	6	47	30	53	0		569	14	0	718
1971	11	26	0	0	0		91	0	0	128
1972	56	410	9	33	0		5	24	0	537
1973	2	186	57	79	0		14	15	0	353
1974	1	15	24	40	0		11	38	0	130
1975	6	65	120	19	0		2	0	0.4	212
1976	3	9	26	14	2		8	3	0	65
1977	1	6	12	53	0		5	0	10	87
1978	2	0	12	29	0		0	72	2	117
1979	1	0	46	12	0		0	2	0	62
1980	13	3	134	90	0		5	0	3	247
1981	1	0	432	135	0		0	0	0	568
1982	2	2	21	122	0.5		81	5	0	234
1983	6	0	43	41	0		25	0	0	115
1984	12	4	40	84	1		8	171	3	323
Total	1,042	3,218	33,217	26,189	1,204	13,272	10,774	2,599	3,133	94,646

in which Q_{rm} is the maximum release for the sampling period (g), MDA_f is the minimum detectable amount of material on the filter (g), and the other terms are as defined previously.

A review of the analytical sheets for dust collector effluent sampling has indicated that the smallest reported amount of material on a filter was 0.1 g. No indication of the MDA_f has been found on those forms reviewed. In the absence of other information, a value of 0.05 g has been used for M_f in Eq. (E–1) to estimate of the release during a sampling period when no release was detected. This is equivalent to assuming that the filter could have contained any amount of uranium between zero and MDA_f and, in the long-term, the sum of such release estimates will be an unbiased approximation of the true release.

An example in which undetected releases were important for some dust collectors is shown in Table E-10. The measured releases for Plant 6 during 1960 were primarily from the South Precipitron. (Plant 6 employed some electrostatic precipitators; releases from them are included in the dust collector releases). That stack was sampled more frequently

Table E-10. Measured and Estimated Releases from Plant 6 in 1960

Sampling Period		Estimated release from South Precipitron	Sampling Period		Estimated releases from G6-6057	Estimated releases from G6-86	Estimated releases from G6-88
Start	Stop	(kg U)	Start	Stop	(kg U)	(kg U)	(kg U)
12-15-59	1-21-60	11[a]	12-15-59	1-21-60	1.0[b]	0.17[b]	0.19[b]
1-21-60	2-16-60	11[a]	1-21-60	2-16-60	1.0[b]	0.17[b]	0.19[b]
2-16-60	3-6-60	12	2-16-60	3-22-60	1.0[b]	0.17[b]	0.19[b]
3-6-60	3-15-60	11[a]	3-22-60	4-20-60	1.0[b]	0.17[b]	0.19[b]
3-15-60	3-22-60	11[a]	4-20-60	5-14-60	33.5	0.17[b]	0.19[b]
3-22-60	4-4-60	11	5-14-60	6-24-60	1.0[b]	0.17[b]	0.19[b]
4-4-60	4-12-60	10	6-24-60	7-22-60	1.0[b]	0.17[b]	0.19[b]
4-12-60	4-20-60	25	7-22-60	8-23-60	1.0[b]	0.17[b]	0.19[b]
4-20-60	4-29-60	20	8-23-60	9-23-60	1.0[b]	0.17[b]	0.19[b]
4-29-60	5-14-60	39	9-23-60	10-24-60	1.0[b]	0.17[b]	0.19[b]
5-14-60	5-17-60	1.0[b]	10-24-60	11-30-60	1.0[b]	0.17[b]	0.19[b]
5-17-60	6-2-60	6.5	11-22-60	12-5-60	1.0[b]	0.17[b]	0.19[b]
6-2-60	6-9-60	1.0[b]	12-5-60	12-30-60	1.0[b]	0.17[b]	0.19[b]
6-9-60	6-20-60	9.1		Total	46	2.2	2.5
6-20-60	7-5-60	1.1[b]					
7-5-60	7-18-60	16					
7-18-60	7-26-60	7.1					
7-26-60	8-3-60	1.1[b]					
8-3-60	8-8-60	4.1					
8-8-60	8-19-60	9.1					
8-19-60	9-6-60	9.1					
9-6-60	9-22-60	6.1					
9-22-60	10-13-60	2.0					
10-13-60	10-24-60	1.1[b]					
10-24-60	11-4-60	1.1[b]					
11-4-60	11-14-60	13					
11-14-60	11-22-60	1.1[b]					
11-22-60	11-30-60	1.1[b]					
11-30-60	12-5-60	2.0					
12-5-60	12-30-60	1.1[b]					
	Total	250					

[a] Estimated release based on operational data.
[b] Estimated release for period when no release was detected.

than the other three release points, whose discharges were generally not detected by the samplers. Estimates for the undetected releases vary because the exhaust flows from the four ducts are different.

When sampling equipment was not installed or failed to operate or if no analytical result was available, interpolation, using releases measured during previous and subsequent sampling periods, was used to estimate the release. This was necessary for four of the sampling periods for the South Precipitron.

For the period shown, inclusion of estimates for periods of unmeasured and undetectable releases led to a total of 305 kg. This may be compared with the reported total of 269 kg in Table E–9; the difference between the two estimates is about 13%. The relative importance of undetected and unmonitored releases depends upon the magnitude of facility releases. Releases from Plant 1, which were relatively small, were underestimated by about 30% because of unmonitored and undetected releases during 1960–1962. However, underestimations for most facilities were smaller, comparable to those for Plant 6 during that period.

Table E–9 shows several years when the stated releases for various facilities were zero. In some cases, this occurs because processes were not operating. Plant 2/3 was closed throughout 1963 and, as noted, Plant 7 operated for only a brief period. On the other hand, Plant 6 is reported to have produced no less than 800 MTU of rolled or machined uranium during each of the years between 1970 and 1986, but annual releases were reported to be about 2 kg U or less. For times when sampling of dust collector exhausts was less complete, or less frequent and poorly described in the routine records, plant releases were estimated using release rates normalized to production rates, described in the next major section of this appendix.

Initial estimates of releases, including those corrected for unmonitored and undetected releases and those based upon normalized release rates and production rates, are subject to further revision to account for biases in the effluent measurements themselves. The potential biases that have been identified and quantified are discussed next.

Possible Biases in Release Estimates

Assessing the magnitude of biases in the sampling results is a difficult and important problem. Although much information was recorded about dust collector operation and the associated sampling, detailed records of stack and sampler flow rates are not available. Assessments of sampling losses have not been found in plant archives, although anecdotal information about problems with plugged sampling lines was recorded. However, revision of the previous release estimates requires quantification of the biases in sampling the dust collector exhausts and of the uncertainties associated with the revised estimates.

Three types of deviations from ideal sampling conditions may have biased the dust collector discharge estimates. These are inhomogeneous distribution of effluent particles in the exhaust duct, mismatch of sampling flow and duct flow, and losses of material in the sampling line. Each is discussed briefly in a subsection below.

Nonrepresentative Sampling. One design feature that was not consistent with standard guidance for sample collection from exhaust ducts was the use of a single sampling probe in larger ducts. The ANSI (1969) guide recommends multiple sample withdrawal points for ducts greater than 15 cm in diameter. The reason for multiple probes is to provide assurance that the samples will not be biased because of a nonuniform distribution of the contaminant in the stack. The sample extracted from the center of a dust collector exhaust stack would be representative if the particles were uniformly mixed in the exhaust or if the concentration on the centerline happened to be equal to the average concentration in the stack. When this is not the case, the sample is not representative of the material being discharged. The bias introduced may be positive or negative, depending upon the actual relationship between the centerline and average concentrations. Quantitative assessment of this question requires tracer measurements in the exhaust stacks. Such an effort is well beyond the scope of this work and is not feasible for many of the exhausts. A qualitative assessment was made and is presented in Appendix G.

Anisokinetic Sampling. The second type of sampling bias that may have occurred is that due to anisokinetic sampling; that is, when the fluid velocity in the sample probe (u, cm s^{-1}) differs from the fluid velocity in the exhaust stack (v, cm s^{-1}). The samplers were set up to obtain isokinetic samples of the stack exhausts, by adjusting the sampling flow to the rate that would make $u = v$. However, the samplers were not equipped with constant flow rate control mechanisms and, as a result, sampler flow rate could vary during the sampling period. The stack flow rate may also vary from the most recently measured value, which was used to determine the isokinetic sampling rate for the stack.

The effect of deviations from isokinetic conditions depends not only upon the ratio of the fluid velocities (u / v), but also on the size and density of the particles (see above and Appendix F), the sampling probe diameter, and, to a lesser degree, on the air temperature and pressure. Anisokinetic conditions can also be produced by misalignment of the sampling probe.

The possible effects of anisokinetic sampling conditions were calculated using the methods described in Appendix G. That appendix contains example calculations and the basis for parameters used in Monte Carlo calculations of bias due to anisokinetic sampling. Anisokinetic sampling can produce either a positive or negative bias in sampling results depending upon whether $u < v$ or $u > v$. The upper bound value for the bias depends upon the aerodynamic diameter of the particle, but can be as great as (v / u) for large particles.

Losses of Particles in the Sampling Lines. Two processes lead to losses of particles in the sampling lines. These are deposition of particles on the walls of the line and impaction of particles due to the presence of bends in the lines. The transmission factor for an aerosol through the sampling line is the ratio of the concentration at the outlet of the line, the sample collection point, to that at the inlet in the stack. A low transmission factor indicates large losses due to deposition and impaction. Unlike the biases due to nonrepresentative and anisokinetic sampling, losses due to deposition and impaction of particles in the sampling line lead only to underestimates of the effluent release. The magnitudes of such losses depend upon particle size and density (see above and Appendix F), the configuration of the sampling line, and the operating conditions for the line. These relationships are described in Appendix G, which contains example calculations and the basis for parameter values used in Monte Carlo calculations of sampling bias due to these processes.

Application of Monte Carlo Techniques to Assess Sampling Bias

The Monte Carlo calculational procedure that was used to estimate sampling biases and their uncertainties is summarized briefly in this subsection. The basis for the calculations is given in Appendix G to which the reader is referred. The calculations employed the measured and inferred particle size distributions described in a previous section of this appendix.

There are several parameters relevant to the estimation of sampling bias for a particular exhaust duct. None of these parameters is known with certainty. The Monte Carlo procedure utilizes information about the expected values and distributions of possible parameter values to make a series of estimates of quantities that depend upon the parameters. The calculations considered the three sources of bias identified above to obtain a measure of overall sampling bias.

The following steps were performed to apply this calculational technique. Distributions of the relevant parameters were developed that reflect the uncertainties associated with the parameters. The distributions were then sampled and the selected parameter values were used to estimate the overall sampling bias for a particular dust collector exhaust. This process was performed repeatedly to obtain a distribution of estimates of the overall sampling bias; the distributions obtained were approximately lognormal. The central values, medians or geometric means (GMs), of the distributions and the associated geometric standard deviations (GSDs) were used to generate revised release estimates and their uncertainties.

Median estimates of overall bias for individual stacks ranged from 0.82 to 0.98, with GSDs ranging from 1.4 to 1.6. The 90% confidence intervals for the estimates of overall bias indicate that, in general, releases may have been underestimated by as much as a factor of two or overestimated by as much as a factor of 1.6. The bounds of possible over- or underestimation are somewhat greater for the very large diameter ducts, notably in Plants 6 and 9. For those exhausts, underestimation by a factor of about 2.5 was possible as was overestimation by a factor of about 1.8. These estimates of overall bias differ from those made in the draft report, primarily because a better treatment of the attachment fraction was developed (see Appendix G).

Major contributors to the uncertainty were the velocity of air in the sampling probe and in the duct, the bias due to nonrepresentative sampling, and a parameter used in computation of the attachment fractions. There is no simple way to reduce the largest uncertainties, which principally reflect the absence of information about conditions of past operations and sampling.

RELATIONSHIPS BETWEEN REPORTED RELEASES AND PRODUCTION

Because there are periods when routine sampling data are not available, it is useful to determine whether there are relationships between reported releases and plant production that could be used to make preliminary discharge estimates for those times. Dust collector releases reported by Boback et al. (1987) (Table E–9) and the plant production data, in metric tons of uranium (MTU), given in Appendix C were used. The period examined was 1956–1984. As shown in Figure E–1 installation of effluent samplers was incomplete before 1956. The plant total release estimates were used because release rates for individual exhausts were interpolated for some years by Boback et al. (1987) and because the utilization of specific dust collectors varied over the years of operation.

Figures E-3 and E-4 show annual production (P, MTU) and reported dust collector releases (Q, kg U) for Plants 4 and 8, respectively. These figures show that, while there is substantial variability in the reported releases for a given level of production, the values were lower during periods of reduced production.

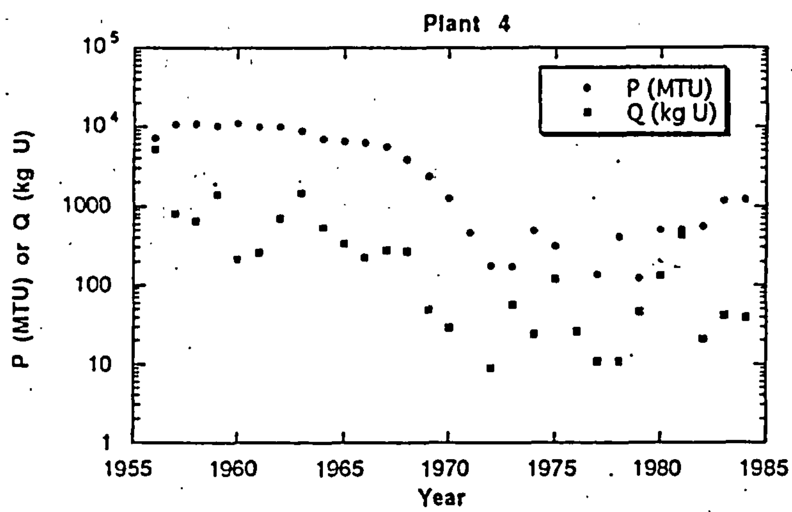

Figure E-3. Plant 4 production data (P, MTU) and reported releases from dust collectors (Q, kg U).

The ratio of the reported release for the year to the plant production during the same period is termed the normalized release rate. This ratio, (Q/P), has units kg U MTU^{-1} and is useful for estimating releases for periods when data are incomplete or unavailable. In Figure E-5 the normalized release for Plants 4, 5, and 6 are plotted as functions of time. Lines connecting the points are provided only to aid the eye of the reader, not as interpolations for years when the reported releases were zero and no points are plotted.

Figure E-6 contains normalized release estimates for Plants 8 and 9. The normalized release estimates for Plants 8 drop rather sharply during the first few years, suggesting that efforts to reduce the dust collector releases were succeeding. However, the normalized releases returned to higher levels in the 1960s. After an initially low value for the first year of uranium production, a general downward trend is shown for Plant 9. Similar patterns are shown for the early years of operation of Plants 4 (Figure E-5).

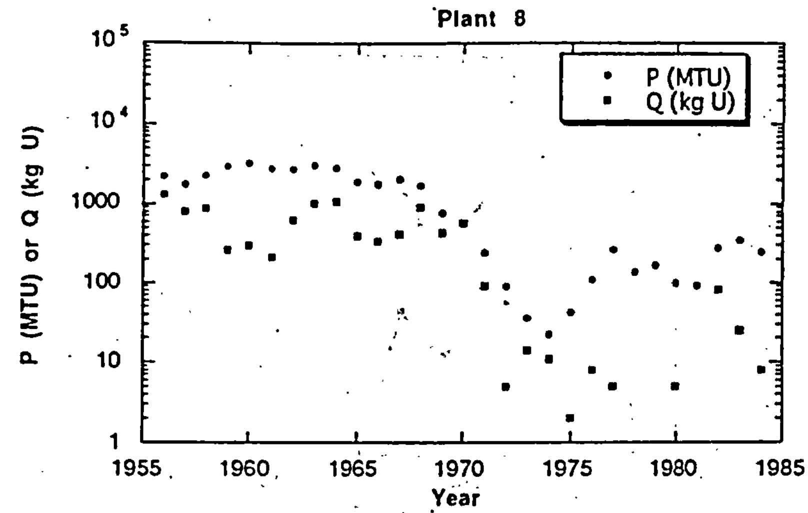

Figure E–4. Plant 8 production data (P, MTU) and reported releases from dust collectors (Q, kg U).

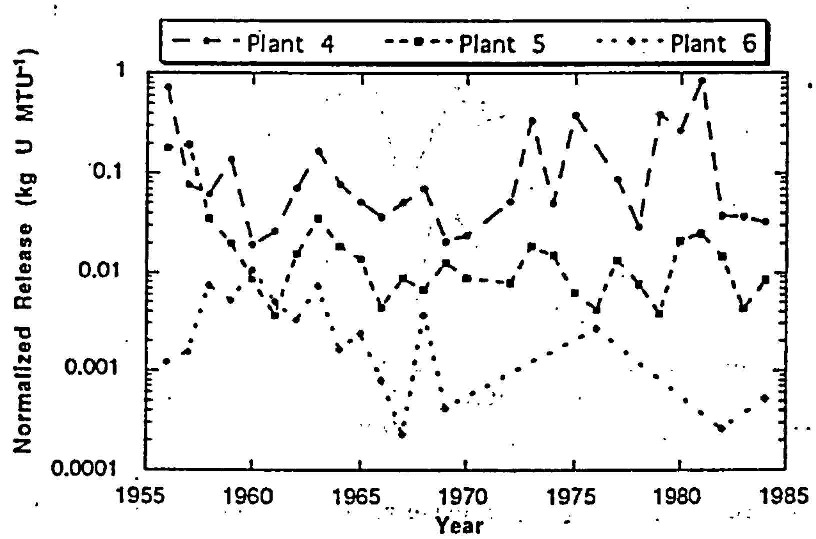

Figure E–5. Normalized release rates (Q/P) for Plants 4, 5, and 6.

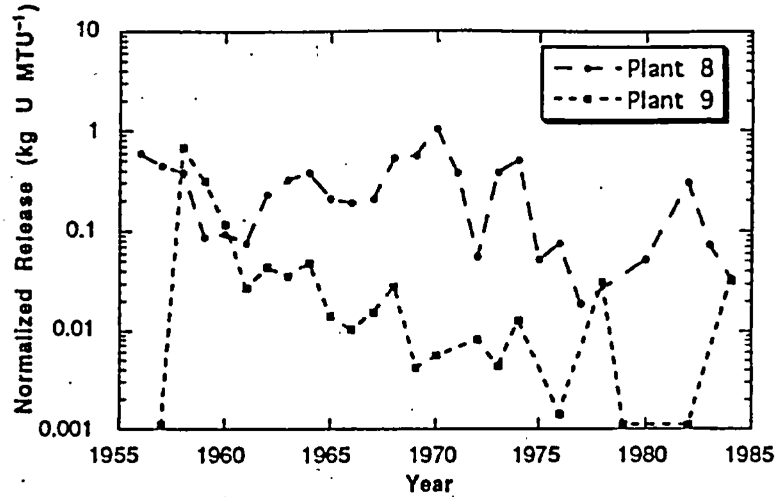

Figure E-6. Normalized release rates (*Q/P*) for Plants 8 and 9.

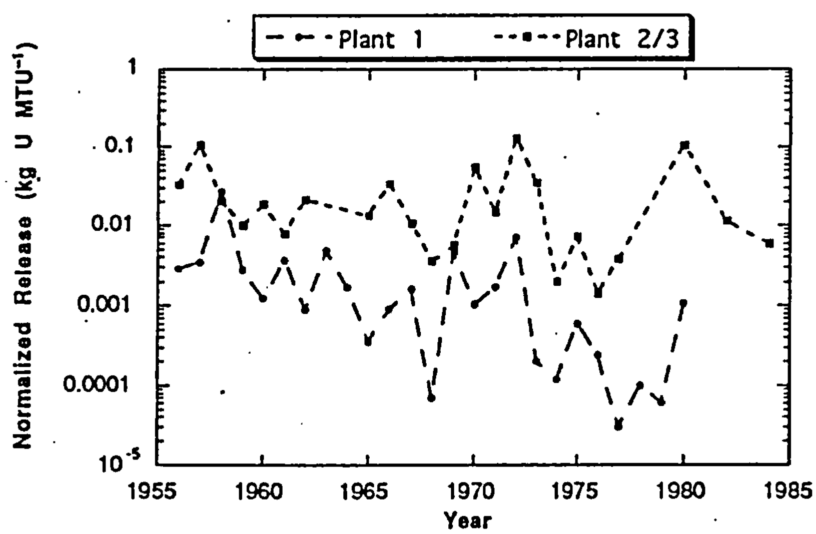

Figure E-7. Normalized release rates (*Q/P*) for Plants 1 and 2/3.

Figure E-7 contains normalized release estimates for Plants 1 and 2/3. The amount of uranium received by Plant 1 during a year was used as a surrogate for production in calculations of (*Q/P*). The normalized releases from Plant 1 show a generally declining trend but with some oscillations. Normalized releases for Plant 2/3 periodically returned to values near those of 1956–1959; normalized releases from Plants 4 and 8 show similar patterns. In contrast, normalized releases from Plants 5, 6, and 9 were generally lower in the later years. Because there is evidence that normalized releases often decreased with time, the entire distribution of estimates cannot be used to make estimates of releases prior to 1956.

Normalized release rates for Plant 7 and the Pilot Plant have not been estimated because production data are not well defined, missing, or incomplete. It would be expected that normalized release rates for Plant 7 and the Pilot Plant would be relatively high. In both those facilities, as in Plants 4 and 8, hydrofluoric acid fumes and high temperature exhausts were constant threats to the integrity of some of the dust collectors.

REVISED RELEASE ESTIMATES

Introduction

The process of developing revised estimates of releases from the FMPC dust collectors is complex. Reported releases were incomplete because sampling was not initiated when production began. The reported releases do not include estimates of releases that were undetected by the analytical procedure or because a sampling system was temporarily out of service. Three sources of possible bias in the reported results, discussed above, have been estimated as part of this effort. Details are provided in Appendix G.

The first step in the approach adopted was to return whenever possible to the original release reports that were prepared routinely by the IH&R department. In the early years of full operation of the effluent sampling program, these reports contained a great deal of information about sample collection and about operational problems in all the plants. These detailed reports made it possible to estimate the magnitudes of undetected releases. Later reports of results, when production rates and releases were lower, were not as detailed and were much less helpful in this regard. In general, inclusion of undetected releases does not have a large effect on the estimates for early years when releases were large. In plants whose releases were relatively small (tens of kilograms of uranium per year) the relative contribution of estimates of releases that had gone undetected was greater.

The reported releases, together with production data, were used to compute normalized releases that offered some guidance for making initial estimates of releases when no effluent sampling was performed. However, such estimates necessarily reflect the biases in reported releases that were identified above and also require correction for them.

Calculational Procedure

Summation of measured or estimated releases, such as those in Table E–10, can be performed using ordinary arithmetic. However the estimates of overall bias in the sampling are approximately lognormally distributed, which introduces complexities into the calculations. The special procedures required are described below.

A particular release measurement (E_1) was assumed to be upon a sample from a normal distribution. The one-sigma uncertainty for that measurement is designated S_1. Parameters of the equivalent lognormal distribution are the median or geometric mean (GM_1) and the geometric standard deviation (GSD_1). These were computed using the following equations:

$$GM_1 = \exp[\mu_1] \text{ in which } \mu_1 = \ln[E_1/\sqrt{(1+(S_1/E_1)^2)}] \quad \text{(E–4)}$$

$$GSD_1 = \exp[\sigma_1] \text{ in which } \sigma_1 = \sqrt{[\ln(1+(S_1/E_1)^2)]} \quad \text{(E–5)}$$

The central estimate of the lognormal distribution of computed values of overall sampling bias characteristic of the exhaust being sampled is designated GM_{ob}. The distribution of such estimates is characterized by the value of the GSD_{ob}. To make the correction for biases, we define $\mu_{ob} = \ln GM_{ob}$ and $\sigma_{ob} = \ln GSD_{ob}$. The geometric mean revised release estimate is: $\exp(\mu) = \exp(\mu_1 - \mu_{ob})$ and the corresponding geometric standard deviation is: $\exp(\sigma) = \exp(\sqrt{(\sigma_1^2 + \sigma_{ob}^2)})$.

Composites of these revised release estimates cannot be obtained by simple addition because the central estimates are medians of lognormal distributions. The correct procedure for determining the median of a sum of such distributions is described below (Dunning and Schwarz 1981, Hoffman and Gardner 1983).

First, each geometric mean revised estimate was used to compute the corresponding arithmetic mean value (m) using the following equation:

$$m = \exp(\mu + 0.5\,\sigma^2) \quad \text{(E–6)}$$

where $\mu = \ln GM$ and $\sigma = \ln GSD$ for the distribution of revised estimates. The variance (s^2) associated with a particular value of m was computed using

$$s^2 = [\exp(2\mu + \sigma^2)][\exp(\sigma^2) - 1] \quad \text{(E–7)}$$

The means and variances of the revised releases to be summed are denoted by m_i and s_i^2, respectively. The arithmetic sums $M = \Sigma\, m_i$ and $S^2 = \Sigma\, s_i^2$ were computed. These were then used to calculate the geometric mean composite release (GM_a) and its geometric standard deviation (GSD_a) using Eqs. E–8 and E–9.

$$GM_a = \exp[\mu_a] \text{ in which } \mu_a = \ln[M/\sqrt{(1+(S/M)^2)}] \quad \text{(E–8)}$$

$$GSD_a = \exp[\sigma_a] \text{ in which } \sigma_a = \sqrt{[\ln(1+(S/M)^2)]} \quad \text{(E–9)}$$

The same basic procedure was used to composite results from multiple time periods for an exhaust point, to obtain a median estimate of the releases from many ducts in a single plant, and to obtain median release estimates for the collection of individual plants. Uncertainties associated with the geometric mean total releases are generally smaller than those associated with the individual contributions to the total.

Estimates for Plant 1

Plant 1 was not a production facility in the classical sense. As an alternative, receipts of uranium have been used to indicate the level of activity. Figure E–8 shows the increase in uranium receipts in the early years. Uranium was received prior to startup of Plant 1 operations, which did not begin until December 1953.

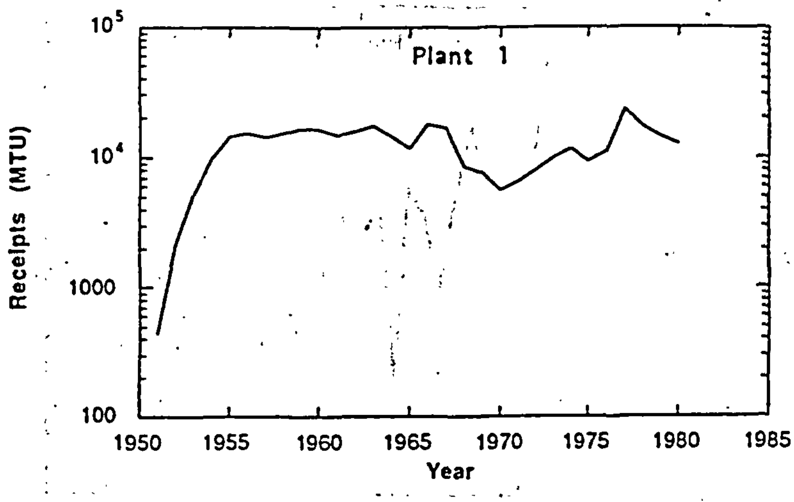

Figure E–8. Receipts of uranium in Plant 1.

Figure E–9 shows the median annual release estimates developed in this study. In the earliest years of operation, releases were not monitored. The average monthly releases measured during the period September–December 1955 were used to make initial estimates of releases during the years 1953–1955. Releases were variable from year to year, being highest in 1958. Releases from Plant 1 were not a large fraction of the total FMPC releases during that year. The median release from Plant 1 for the entire period was estimated to be 1,300 kg U oxides (GSD = 1.2).

Estimates for Plant 2/3

Plant 2/3 also began production late in 1953. Figure E–10 shows the substantial variability in UO_3 production over time in that facility. Effluent sampling systems were not in place until August of 1955. Initial estimates of releases for that year were obtained by scaling the results for the 5-month period for which data were available. The average normalized release rates for 1955–1957 were used to make initial release estimates for 1953 and 1954 when no effluent measurements were made.

The estimated median annual releases, shown in Figure E–11, generally follow the pattern defined by the changes in production, but there is additional variability. The highest releases from the Plant 2/3 dust collectors were in 1958. Over the entire period of operation, the median estimate of releases from Plant 2/3 dust collectors was about 4,000 kg U oxides (GSD = 1.2). This contribution to overall dust collector releases is about three times larger than that from Plant 1, but nonetheless is not a major fraction of the total.

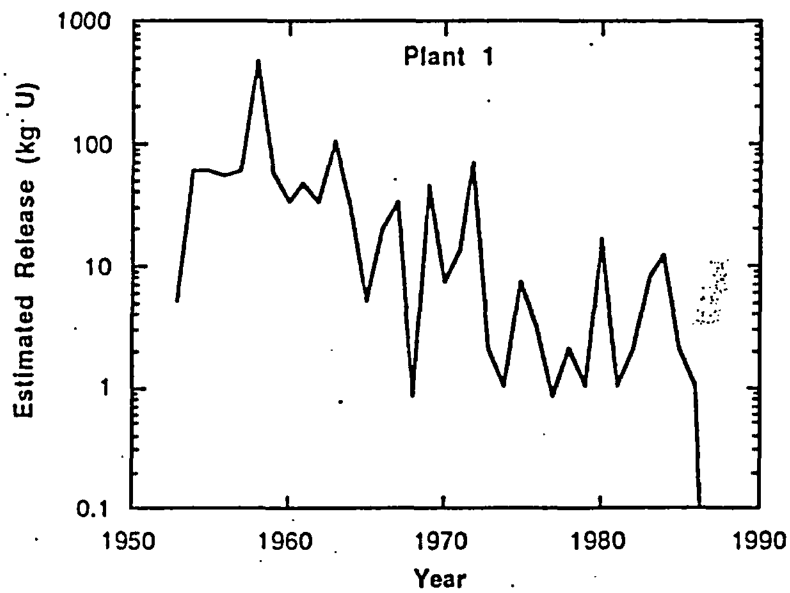

Figure E-9. Estimated median annual releases from Plant 1 dust collectors.

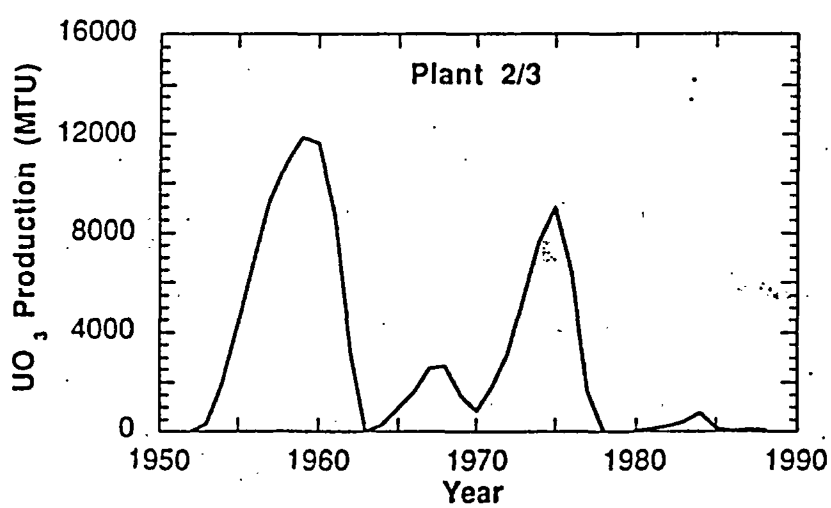

Figure E-10. Production of uranium trioxide in Plant 2/3.

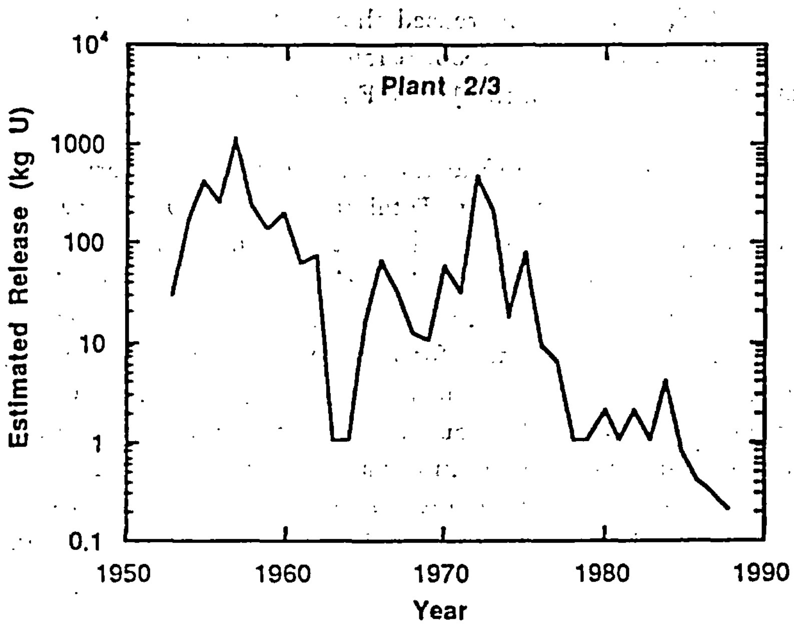

Figure E–11. Estimated median annual releases from Plant 2/3 dust collectors.

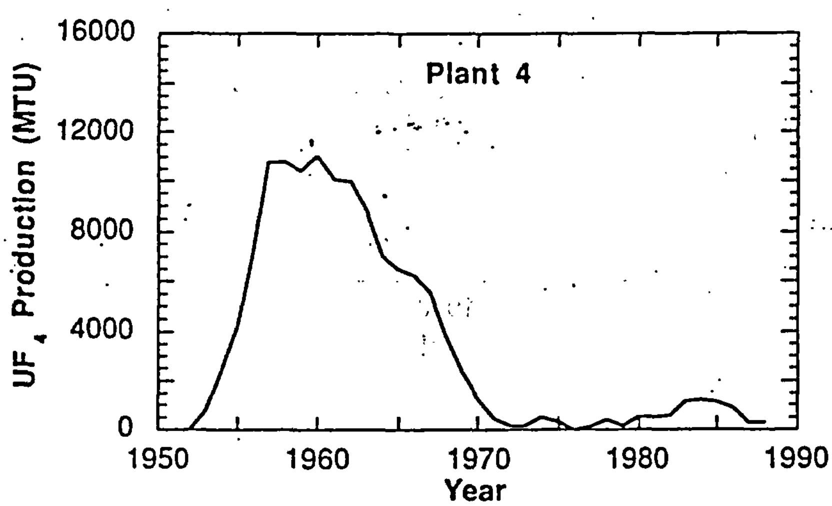

Figure E–12. Production of uranium tetrafluoride in Plant 4.

Estimates for Plant 4

Production of UF_4 in Plant 4 increased sharply to more than 10,000 MTU in the first four years after startup in October 1953. It fell below 1000 MTU in 1971 and was slightly above that level for three years in the 1980s. Figure E–12 shows the time history of Plant 4 production.

Partial monitoring of Plant 4 effluents was established in April 1955; more samplers were installed in August of that year. Total releases for 1955 were estimated from the measurements and the normalized release rate for that year was used, together with production data, to estimate releases during 1953 and 1954. The estimate for 1953 was in good agreement with measurements made in November and December of that year.

Estimated median annual releases for Plant 4 dust collectors are shown in Figure E–13. Estimated releases were very high during the 1950s with a peak in 1955. Relatively high releases occurred early in the 1960s, but declined sharply toward the end of that decade. Releases in more recent years were quite variable, with a peak in 1981. For the entire period, the median release from the Plant 4 dust collectors was about 41,000 kg U (GSD = 1.2), about 80% of which was uranium oxides. Plant 4 was one of three primary contributors to total dust collector releases.

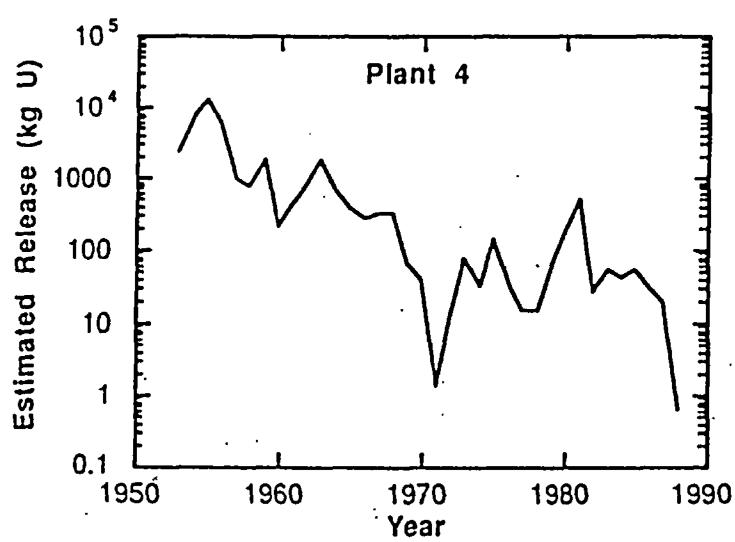

Figure E–13. Estimated median annual releases from Plant 4 dust collectors.

Estimates for Plant 5

Uranium metal and ingot production in Plant 5 is shown in Figure E–14. After startup in May 1953, production rose sharply initially and remained at a high level until the late 1960s. A secondary peak in production occurred in the 1980s.

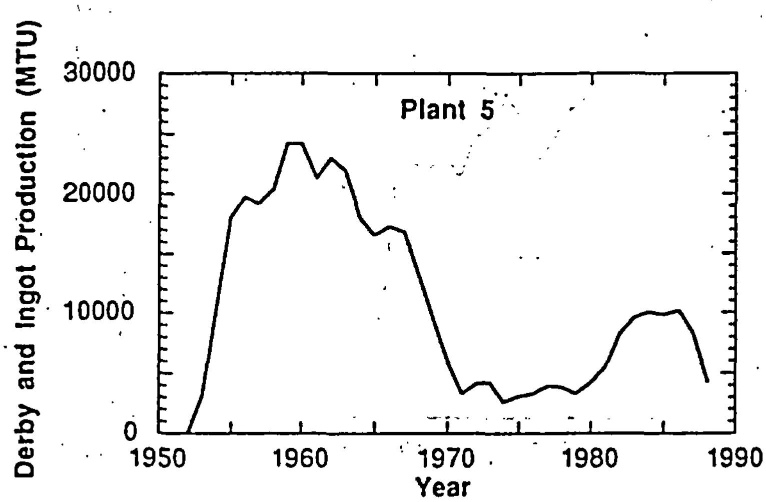

Figure E–14. Production of uranium metal derbies and ingots in Plant 5.

Partial effluent monitoring coverage was initiated in April of 1955 and a more complete program was in place by October of that year. Initial estimates of releases for 1955 were based upon extrapolations of the partial data for that year. The estimate of the normalized release for 1955 and production data were used to project releases for 1953 and 1954.

The estimated median annual release for all years are shown in Figure E–15. As was the case with Plant 4, the highest releases from Plant 5 occurred during the 1950s and were relatively high during the 1960s before declining to the much lower levels of the 1970s and 1980s. Plant 5 was another of the principal contributors to total releases from the FMPC dust collectors. The median estimate of release from Plant 5 for the entire period was about 38,000 kg U oxides (GSD = 1.2). This release is somewhat lower than but comparable to the amount estimated for Plant 4.

Estimates for Plant 6

Machining operations in Plant 6 were initiated in July of 1952 and the rolling mill began production the following month. Combined production figures from the two operations are plotted in Figure E–16. Production rose rapidly after startup and remained high for many years, exceeding 20,000 MTU for all years between 1956 and 1964. Production levels were much lower after 1970, with maximum levels of about 2,000 MTU for three years in the 1980s.

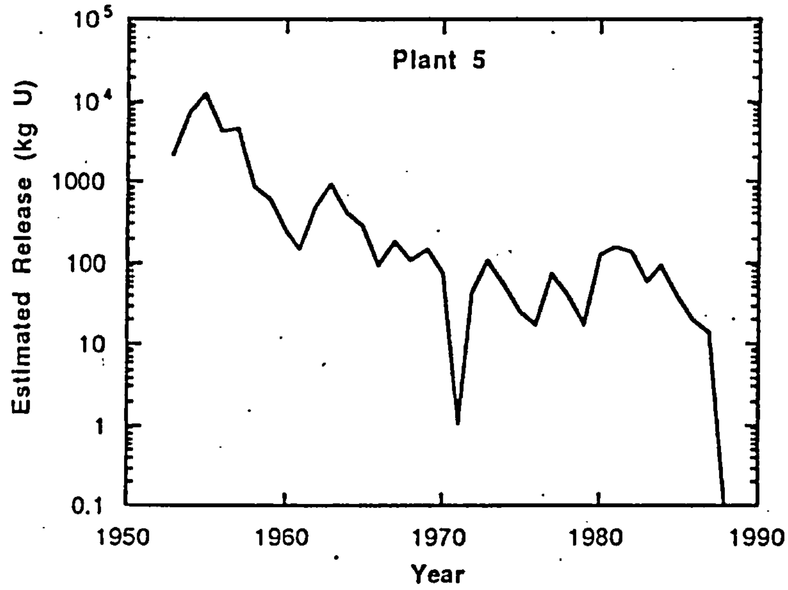

Figure E-15. Estimated median annual releases from Plant 5 dust collectors.

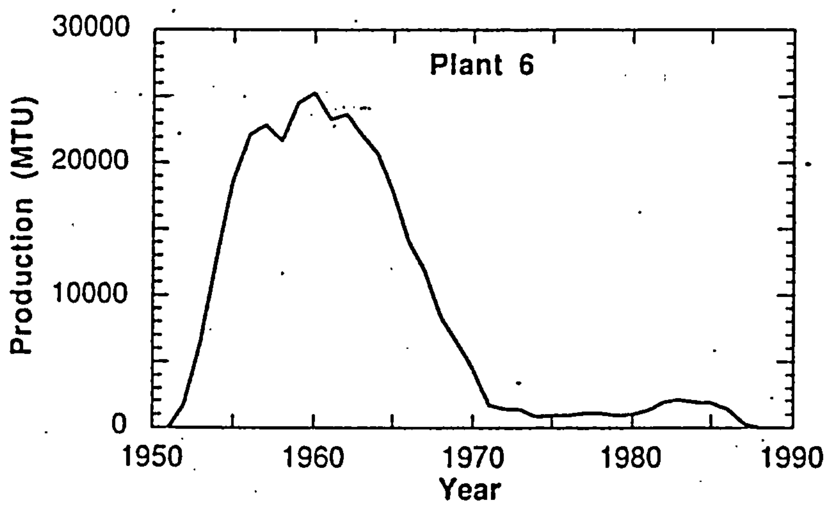

Figure E-16. Production of machined and rolled uranium in Plant 6.

Although effluent sampling was initiated in Plant 6 in August 1955, a full complement of sampling systems was not in place until mid-1957. The average normalized release rate in 1958 and 1959 was used, together with production data, to estimate uranium releases from Plant 6 for the years 1952 through 1957. Figure E–17 shows median annual release estimates for all years. The releases follow the general pattern of the production levels in Plant 6 with peak values during the years of highest production and low values after 1970.

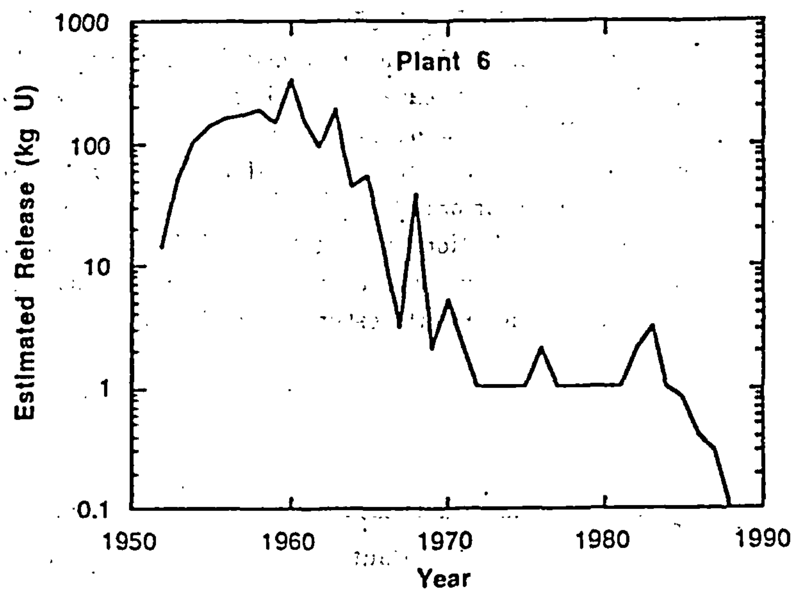

Figure E–17. Estimated median annual releases from Plant 6 dust collectors.

The figure shows that Plant 6 was not a major contributor to total FMPC dust collector releases. After 1970, estimated releases were all less than 10 kg U and some were less than 1 kg U. (Some exhausts from Plant 6 were treated by electrostatic precipitators. Releases from those stacks are included in these totals in the "dust collector" category). The median estimate of the total Plant 6 release was about 2,100 kg U oxides (GSD = 1.1), about 1.5 times the release from Plant 1.

Estimates for Plant 7

Plant 7 was turned over to the Production Division in late June of 1954. Initial operations were completed in the third quarter of that year. The plant ceased operation in May 1956. Although the capacity of Plant 7 is known, operational data for the facility appear to have been lost or destroyed.

Effluent monitoring began in Plant 7 in September of 1955. All four dust collector exhausts were sampled during the next eight months (until shutdown). Results for about one-third of the samples collected in 1955 were considered unreliable because of trouble with the vacuum system which pulled air through the samplers and because some sampling lines were plugged with UF_4. Sampler operation in 1956 was satisfactory and the improvement was noted in the sampling data record.

Whenever possible, measurement data were used as the initial estimates of releases from Plant 7. For other periods, two operating scenarios and corresponding release rates were developed for each of the dust collectors. One scenario reflected relatively good performance of the collectors while the other was used for periods when degraded performance was identified or suspected. During initial startup operations, when the systems were being tested and checked, and for the remainder of 1954 it was assumed that the first scenario applied. However, in July 1955 there were identified problems with loss of materials from Plant 7 (Cuthbert 1955). It was presumed that these difficulties began at the start of the 1955 and that the higher release rate estimates applied from January to late September (when actual sampling data were available).

Figure E–18 shows the estimated median annual releases from the Plant 7 dust collectors. Even though it operated for only two years, the observed and projected releases from the plant made a major contribution to the total FMPC dust collector releases. The median estimate of Plant 7 releases was about 35,000 kg U, primarily UF_4, (GSD = 1.4), which is lower than but still comparable to the estimate for Plant 5.

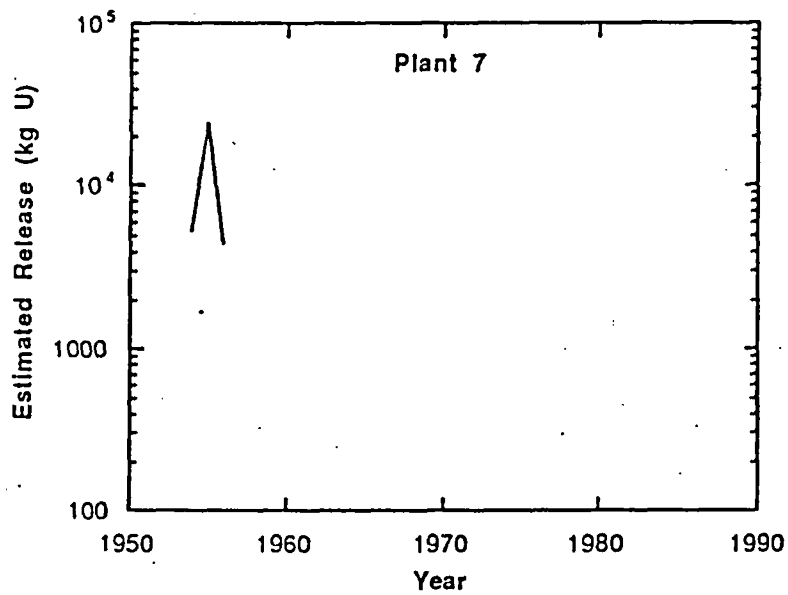

Figure E–18. Estimated median annual releases from Plant 7 dust collectors.

Estimates for Plant 8

Uranium recovery operations in Plant 8 began in November 1953. Figure E–19 shows the history of that work over time. As was the case for other facilities at the FMPC, the highest activity occurred during the late 1950s and the 1960s. Annual uranium recovery increased rapidly during the first three years of operation and exceeded 1500 MTU for all years during the period 1957–1968. Uranium recovery during later years was substantially lower, but did rise to nearly 900 MTU in 1987.

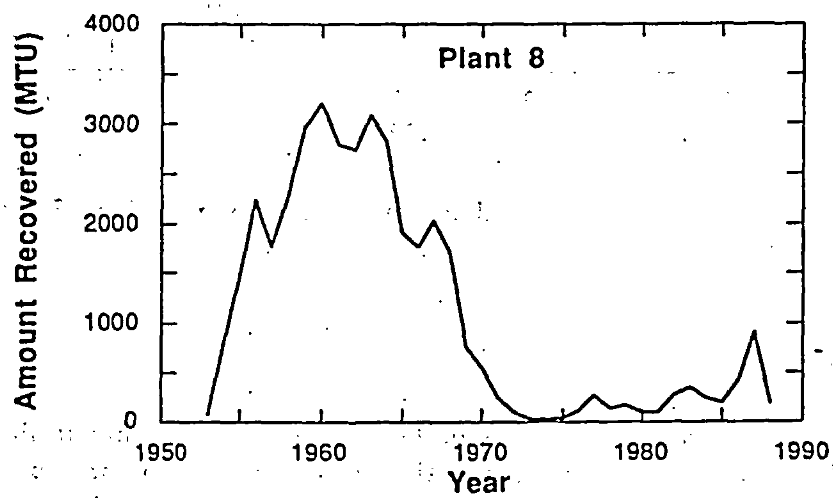

Figure E–19. Recovery of uranium from scrap in Plant 8.

Effluent monitoring was initiated in Plant 8 in July of 1955, but comprehensive monitoring was not established until the following year. Results from 1955 and 1956 were used to establish a normalized release rate that was used, together with production data, to make initial estimates of releases in late 1953 and during 1954 and 1955.

Estimated median annual releases from the dust collectors in Plant 8 are shown in Figure E–20. The highest estimated releases occurred between 1955 and 1970. Releases in later years were generally below 50 kg U with the exception of 1982. Because of the early releases — estimated to exceed 1000 kg U in each of ten years before 1971 — Plant 8 is one of the major contributors to total dust collector releases. Overall, the median Plant 8 dust collector release was about 14,000 kg U oxides (GSD = 1.1), lowest of the four major sources of releases.

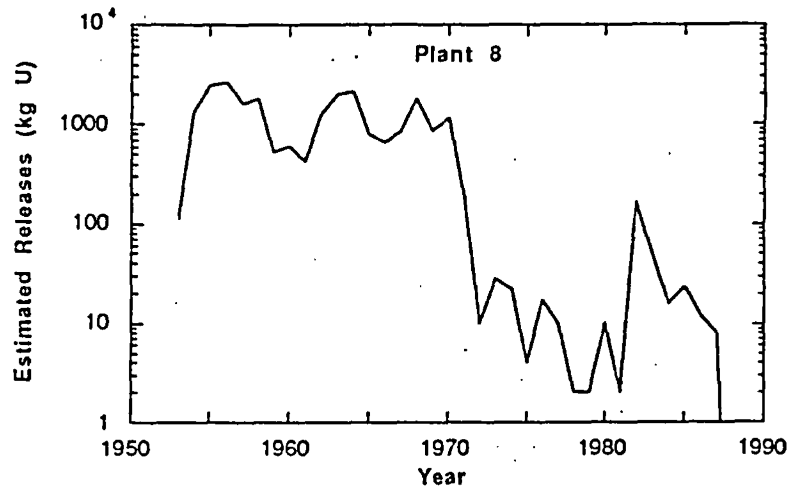

Figure E-20. Estimated median annual releases from Plant 8 dust collectors.

Estimates for Plant 9

Plant 9 began uranium operations in 1957. Production for that facility, shown in Figure E-21, includes both ingot production and machining of uranium metal. The production rate plot for Plant 9 is somewhat unique among the FMPC facilities because the production in later years was comparable to that in the mid-1960s.

Effluent monitoring had already been started in 1957 but was more complete in 1958. The normalized release rate for 1958 was used with production information to estimate the releases in 1957. Median annual release rate estimates for all years are shown in Figure E-22. Releases were highest during the early years but were estimated to exceed 100 kg U twice after 1975. Overall, the median release estimate for Plant 9 dust collectors was about 3,300 kg U oxides (GSD = 1.1), a contribution to the total roughly comparable to that from Plant 2/3.

Estimates for the Pilot Plant

Work was started in the Pilot Plant in October 1951. Many different operations were performed there, not all of which were documented in terms of "production." Conversion of UF_6 to UF_4 was an initial production activity as was the reduction of UF4 to metal. Data on the first process were available but the metal production information was not as well preserved. Tests run to evaluate operational problems and recommend changes also had some attributes of production runs, but no accessible chronicle of these efforts was found.

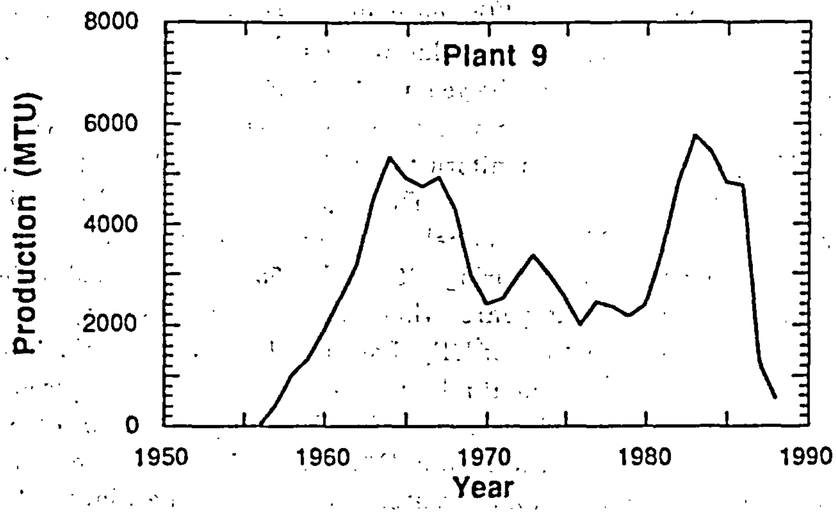

Figure E–21. Production of uranium ingots and machined products in Plant 9.

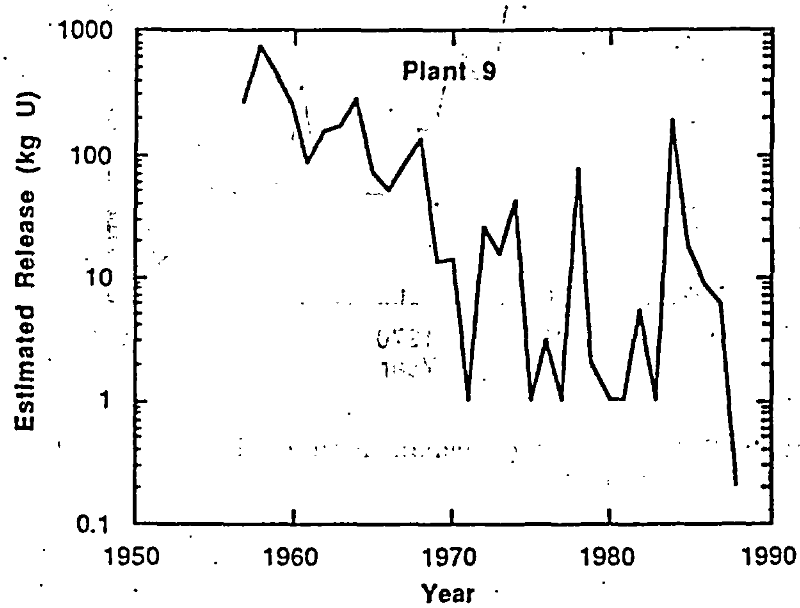

Figure E–22. Estimated median annual releases from Plant 9 dust collectors.

There was early monitoring of effluents from the Pilot Plant. Releases from the UF_6 to UF_4 production area were monitored during June–August 1953. Routine monitoring was started in the last quarter of 1955 and the normalized release rates estimated from the two sets of data were consistent. The mean value was used with production data to estimate releases from this part of the plant for the years 1952–1955.

Releases from the metal production area were also measured in 1953 and were found to be about 80 kg U per year. Routine monitoring in 1955 and 1956 showed that these releases had been reduced to about 3 kg U per year. The higher rate was used to estimate releases from October 1951 through 1952. Lower releases were estimated for 1953, when Plant 5 production was started, and releases during 1954 and 1955 were assumed to decline further.

Most of the releases during years when monitoring was only occasional were estimated to be due to the production of UF_4. Estimated median annual releases for all years are shown in Figure E-23. Annual releases are estimated to have exceeded 1000 kg U during four early years, but were less than 50 kg U in all years after 1963. Overall, the median Pilot Plant contribution to the total FMPC dust collector releases was estimated to be about 3,900 kg U (GSD = 1.2), approximately the same as the Plant 2/3 dust collector releases.

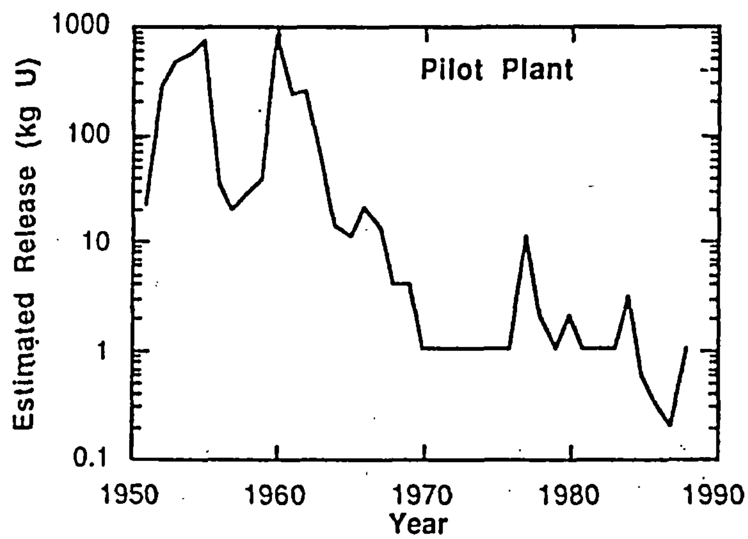

Figure E-23. Estimated median annual releases from the Pilot Plant dust collectors.

Summary of Dust Collector Releases

Estimated median annual releases from all FMPC dust collectors (including, as noted previously, the releases from stacks treated by electrostatic precipitator in Plant 6) are shown in Figure E–24. The plot shows that the highest releases were in 1955, with dominant contributions from Plants 7, 4, and 5. The figure illustrates the relative importance of releases during the early years of FMPC operations. Median annual releases exceeded 1000 kg U in only three years of the decades of the 1970s and 1980s, whereas they were consistently above that level during all but one of the years from 1954 to 1968.

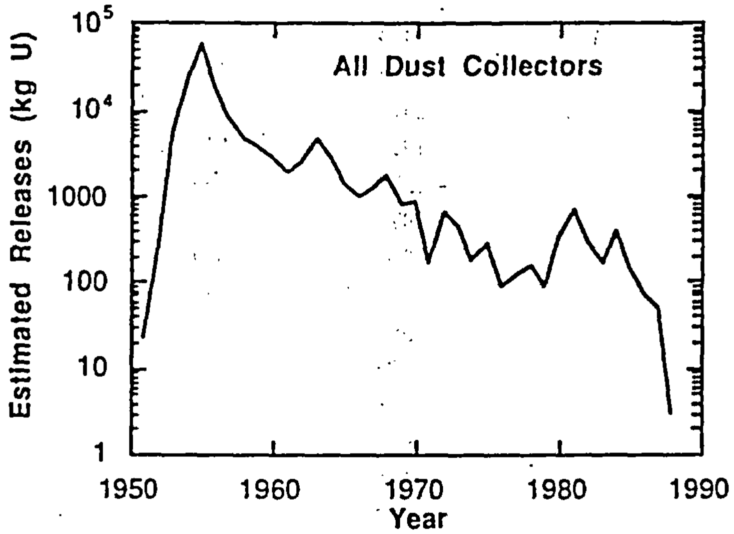

Figure E–24. Estimated median annual releases from all dust collectors.

Table E–11 contains the best (median) estimates of the releases from FMPC dust collectors for each year of operation. These values, which have been rounded to two significant figures, were computed (as were the sums for individual plants) using the special procedure described earlier in this appendix. Also shown in the table are the geometric standard deviations for the lognormal distributions of release estimates.

Table E–12 contains the estimates of releases for each decade during which operations occurred. In addition to the median estimates, values for various percentiles of the distributions are shown. The table shows clearly the predominance of releases during the early years of operation of the facility.

CONCLUSIONS

Many exhaust stacks at the FMPC were served by dust collectors that recovered uranium that would otherwise be lost. Routine sampling of the dust collectors was begun in April 1955 and expanded rapidly during the next two years. The results of these measurements form the basis for initial estimates of releases from the dust collectors.

Table E-11. Median Annual Release Estimates for FMPC Dust Collectors

Year	Median estimate of release (kg U)	Geometric Standard Deviation	Year	Median estimate of release (kg U)	Geometric Standard Deviation
1951	22	1.5	1970	850	1.4
1952	273	1.5	1971	160	1.4
1953	5,300	1.3	1972	640	1.4
1954	23,000	1.3	1973	440	1.3
1955	54,000	1.3	1974	170	1.2
1956	18,000	1.3	1975	270	1.3
1957	8,300	1.3	1976	85	1.2
1958	4,600	1.2	1977	120	1.3
1959	3,600	1.3	1978	150	1.3
1960	2,600	1.2	1979	90	1.4
1961	1,800	1.2	1980	320	1.3
1962	2,400	1.2	1981	680	1.4
1963	4,600	1.3	1982	280	1.3
1964	2,700	1.3	1983	160	1.3
1965	1,300	1.3	1984	360	1.3
1966	950	1.3	1985	140	1.3
1967	1,200	1.3	1986	71	1.3
1968	1,700	1.3	1987	48	1.3
1969	780	1.3	1988	3	1.2

Table E-12. Summary Release Estimates for FMPC Dust Collectors

Period	Best estimate of release (kg U)	Other percentiles in distribution of release estimates (kg U)			
		5th percentile	25th percentile	75th percentile	95th percentile
1950s	120,000	96,000	110,000	130,000	150,000
1960s	21,000	18,000	19,000	22,000	24,000
1970s	3,100	2,500	2,800	3,400	3,800
1980s	2,100	1,700	1,900	2,400	2,700
1951–1988	140,000	120,000	130,000	160,000	170,000

Physical and chemical parameters of the releases are important to the dispersion and dose calculations. Extensive review of measurements of particle size made in 1985 was performed and information was obtained from other facilities. Particle size distributions have been assigned to the various stacks based on direct measurement results or similarity of function to other stacks or areas in which measurements were performed. In spite of some substantial variations from stack to stack, it can be stated that the particles were relatively large. Plant processes have been reviewed to evaluate the chemical forms that would be released from the various stacks. About three-fourths of the releases from the dust collectors were in the form of uranium oxides.

Previous estimates of releases from individual dust collectors at the FMPC were tabulated from original records. In the early years, there were monthly reports of the measurements. Review of the reported results revealed periods when samplers were not in operation and other times when the releases were too low to be detected. Estimates were made for these periods based on other sampling results and information about the sampling and analysis procedures. Estimates were also made for years before monitoring was established as a routine procedure. These estimates were based either upon normalized release rates soon after routine monitoring was established or representative measurements during the mid- to late-1950s. In some cases, evaluations of unmonitored effluents led to significant increases in release estimates.

Possible biases in the sampling results were investigated and a Monte Carlo procedure was developed to estimate their magnitudes (see Appendix G). The procedure was applied to each of the dust collector exhaust sampling systems. The estimates of bias were used to compute revised release estimates for the dust collector exhausts. The magnitudes of the estimates of overall bias varied among the stacks from 0.82 to 0.98. Corrections for unmeasured releases and for sampling bias led to revised release estimates that were about 50% higher than previous estimates of dust collector releases.

The median estimate of total releases from the FMPC dust collectors was about 140,000 kg U. The preponderance of the releases occurred during the 1950s. Principal contributors to the releases during that decade were Plants 4, 7, and 5. Plant 8 also contributed significantly to the total, but most of those releases occurred over a longer period of time. Although releases from the other facilities were not small, those releases were not major fractions of the total release. However, some of the releases from plants that were lesser contributors to the total were important in individual years.

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APPENDIX F

FITTING PARTICLE-SIZE DISTRIBUTIONS FOR FMPC DUST COLLECTORS

INTRODUCTION

The distributions of particle size in releases of particulate radioactive material from a nuclear facility are important in two particular respects for estimating transport and radiation dose. First, atmospheric deposition processes are sensitive to particle aerodynamic properties determined by size, shape, and density, and thus successful estimation of the rate of depletion of the plume depends on making reasonable assumptions about these distributions. Second, the use of the respiratory model of the International Commission on Radiological Protection (ICRP) requires the assumption of activity median aerodynamic diameters (AMAD) appropriate to the material.

In 1985, the Feed Materials Production Center (FMPC) and subcontractor Northern Kentucky Environmental Services (NKES) performed sampling operations on the inlet and outlet ducts of 15 dust collectors. Uranium particle-size distributions and isotopic fractions for these samples are presented by Boback et al. (1987), by NKES in an unpublished report (Reed 1985), and in the original data sheets. These data provide essentially all of the usable information on distributions of particle size in FMPC stack emissions that has come to light at the date of this writing (September 11, 1991), and this information will have to be taken as the basis of generic representations of particle size in the source term for purposes of transport simulation and dose reconstruction.

Each measured diameter is reported as an *equivalent aerodynamic diameter*, which is the diameter of a sphere of unit density ($\rho = 1$ g cm^{-3}) that has the same gravitational settling velocity as the particle. Physical diameter is equal to equivalent aerodynamic diameter divided by the square root of the density (expressed in units of g cm^{-3}) of the particular compound of uranium for diameters in the ranges encountered in these tables. We confine this presentation to equivalent aerodynamic diameters.

In order to make use of the distributions, some extrapolation is necessary, because some of the distributions leave 30% or more of the probability unaccounted for in the region of the largest particles. Some degree of smoothing will be required for some of the distributions. And in all cases it will be necessary to interpolate between the tabular values. The large number of release points at the FMPC may also make it desirable to consolidate the particle-size distributions into a smaller number of "generic" distributions that can be applied to the simulations of release, transport, and uptake by inhalation. These requirements point to the need for a method of fitting a cumulative distribution function (CDF) to the empirical distributions. Such needs clearly are not new, and methods have been discussed in the literature (e.g., DallaValle et al. 1951).

In only a few of the cases are the data adequately represented by a lognormal distribution. Of the remainder, some but not all are of bimodal form (i.e., the density or frequency function has two maximum points).

The purpose of this appendix is twofold:

- to explain a method that we have found satisfactory for fitting cumulative distribution function (CDF) curves to empirical particle size distributions with small numbers of observed cumulative mass fractions
- to present the results of applying the method to those empirical particle-size distributions for inlet ducts and emission stacks given by Boback et al. (1987) as corrected, in two instances, with data from the original worksheets.

Figures F–1 through F–15 show plots of the distributions. For each dust collector, plots of the distributions for the inlet duct and the emission stack are shown on the same chart relative to log-probability axes; the observed cumulative mass fractions are plotted as discrete points, and the fitted CDF functions are shown as curves. Accompanying each plot is output from the plotting program that shows the input distribution, the fitting parameters determined by the method, the observed and predicted cumulative probabilities, summary statistics of the fitted distribution, and the diameter corresponding to each extreme value of the density function. These data are shown side-by-side for the inlet duct and the emission stack.

> PLEASE NOTE: The empirical distributions discussed here are mass distributions. We frequently substitute the word "probability" because it is the usual term in mathematical discussions. Also, the density (or frequency) function (PDF) is the derivative of the cumulative distribution function.

REGRESSION FUNCTION AND DETERMINATION OF PARAMETERS

The method of regression is based on the application of a one-to-one transformation $z = T(\zeta)$ to the domain of the standard normal cumulative distribution function

$$P(z) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{z} e^{-t^2/2}\, dt \qquad \text{(F–1)}$$

If the transformation T is increasing for all ζ, the composite function $P(T(\zeta))$ is also a cumulative distribution. For distributions that may be considered distortions of the lognormal, it is reasonable to try the further substitution

$$\zeta = \ln(x)$$

and seek to determine the transformation $T(\zeta)$ so that the composite function

$$\Phi(x) = P(T(\ln(x))) \qquad \text{(F–2)}$$

represents the data as closely as possible by some chosen criterion of fit.

By choosing $T(\zeta)$ to be a cubic polynomial and determining the coefficients by nonlinear least-squares regression, we have found that the resulting regression function (Eq. F–2) gives qualitatively appropriate representations of the empirical distributions. Algorithmically, the procedure is as follows. We are given diameters x_i and cumulative probabilities p_i, $i = 1, \ldots, N$. The regression is

$$t_i \approx T(\zeta_i) \qquad \text{(F–3a)}$$

where the transformed data points (ζ_i, t_i) are

$$t_i = P^{-1}(p_i), \quad \zeta_i = \ln x_i, \quad i = 1, \ldots, N \tag{F–3b}$$

We note that Eqs. F–2 and F–3 depend on numeric evaluation of the functions $P(z)$ and $P^{-1}(p)$, where P is the standard normal CDF and P^{-1} is its inverse. Rational approximations to these functions are provided by Abramowitz and Stegun (1968; Eqs. 26.2.18 and 26.2.23, respectively).

One might begin by requiring that $T(\zeta)$ have the general cubic polynomial form

$$T(\zeta) = c_0 + c_1\zeta + c_2\zeta^2 + c_3\zeta^3 \tag{F–4}$$

so that the regression problem is to determine the coefficients $c_0, \ldots, c_3$:

$$\sum_{i=1}^{N} [t_i - T(\zeta_i; c_0, \ldots, c_3)]^2 = \text{minimum}. \tag{F–5}$$

Because the coefficients enter the expression for $T(\zeta)$ linearly, the polynomial least squares regression problem posed by Eq. F–5 is linear. Once the coefficients c_i are determined, Eq. F–2 gives the distribution function $\Phi(x)$.

But this procedure is invalid if the polynomial $T(\zeta)$ fails to represent a one-to-one increasing transformation. For the cubic, a simple test may be derived and applied after the coefficients have been determined. The necessary criteria are met if and only if

$$c_2^2 - 3c_1c_3 \le 0 \quad \text{and} \quad c_1 > 0. \tag{F–6}$$

Unfortunately, the data from several of the inlet ducts produce linear regressions that fail to satisfy the criterion of Eq. F–6; the polynomials produced in these cases fail to be everywhere increasing. An alternative procedure is necessary to avoid this difficulty.

By rewriting Eq. F–4 as

$$T(\zeta; \alpha, \beta, \gamma, \delta) = \alpha^2(\zeta - \beta)^3 + \gamma^2\zeta + \delta \tag{F–7}$$

we retain the functional form of a cubic polynomial, but with coefficients that depend on the parameters α, β, γ, and δ. The derivative

$$T'(\zeta) = 3\alpha^2(\zeta - \beta)^2 + \gamma^2$$

is clearly everywhere nonnegative, enforcing the condition that the polynomial $T(\zeta)$ be everywhere increasing (except that it will have zero slope at $\zeta = \beta$ if $\gamma = 0$). But the parameters α, β, and γ enter the expression for $T(\zeta)$ in a nonlinear manner, complicating the regression procedure. We express the nonlinear regression as

$$\sum_{i=1}^{N} [t_i - T(\zeta_i; \alpha, \beta, \gamma, \delta)]^2 = \text{minimum}. \tag{F–8}$$

We have solved this nonlinear least-squares regression problem for α, β, γ, and δ with the FORTRAN subroutine LMDER1, which implements the Levenberg-Marquardt algorithm (Moré et al. 1980). This procedure has been successful with all of the FMPC particle-size data.

Once α, β, γ, and δ are known for a given distribution, we may relate the coefficients $c_0, \ldots, c_3$ of Eq. F–4 to these parameters as follows:

$$c_0 = \delta - \alpha^2\beta^3 \tag{F–9}$$

$$c_1 = 3\alpha^2\beta^2 + \gamma^2 \tag{F–10}$$

$$c_2 = -3\alpha^2\beta \tag{F–11}$$

$$c_3 = \alpha^2. \tag{F–12}$$

It is $c_0, \ldots, c_3$ as given in Eqs. F–9 through F–12 that we report in the tables accompanying Figs. F–1 through F–15.

Summary statistics on each fitted distribution are obtained by numerical methods. The median (which may be read from the 50th percentile of the graph) is calculated from iterated interval bisections, and the mean and standard deviation are calculated from numerical algorithms for the integrals

$$\mu = \int_{x_{min}}^{x_{max}} x \, d\Phi(x) \tag{F–13}$$

$$\sigma = \left[\int_{x_{min}}^{x_{max}} (x-\mu)^2 \, d\Phi(x)\right]^{\frac{1}{2}} \tag{F–14}$$

where the numbers $x_{\min}$ and $x_{\max}$ replace the infinite limits of integration for practical computation (the values 0 and 100 were appropriate for the data treated here).

Interest in whether a distribution has multiple modes may exist, and it is not difficult to answer the question numerically for the distribution $\Phi(x)$. We used a partition of the interval $[x_{\min}, x_{\max}]$ and checked the derivative $\phi(x) = \Phi'(x)$ for changes from increasing to decreasing or from decreasing to increasing from one subinterval to the next. Using subinterval length 0.01, all distributions were analyzed and the diameter corresponding to each extremum was recorded. The existence of three extrema (maximum, minimum, maximum) implies a bimodal distribution.

It is important to realize that parameter estimates (median, mean, standard deviation) based on distributions fitted by this regression technique may be expected to differ from their counterparts presented in the NKES report (Reed 1985). The methodology of the NKES report is based on the assumption of lognormality, which our results indicate is seldom justified in the case of these data. In the case of the median of a distribution, our estimate is based on the diameter at which the fitted curve, plotted on log-probability paper, crosses the 50th percentile. The linear fit (lognormal assumption) in general does not intersect the 50th percentile line at the same diameter as does the cubic fit. The geometric standard deviation (GSD) has meaning only in the context of the lognormal distribution and thus is generally not applicable to the distributions based on the cubic regression (but this formulation does include the lognormal distribution as a special case, namely for $\alpha = 0$). Dispersion information such as the first and third quartiles can be read directly from the plots of these fitted distributions. In using a single number, such as a standard deviation or an interquartile range ($Q_3 - Q_1$), as a measure of dispersion for these distributions, one must keep in mind the lack of symmetry of the distributions when one tries to interpret such a parameter.

CAVEATS: Some cautions need to be observed in dealing with the particle-size data from the NKES 1985 study and the fitted distributions described in the present report. The first is that each fitted distribution is based on just eight points of an observed cumulative distribution function (i.e., cumulative mass fraction). Second, we observe that extrapolation is at best a questionable exercise, and the use of the fitted distributions outside the diameter regions spanned by the data must be undertaken only with this realization in mind. The polynomial transformation introduced into the regression is, of course, arbitrary, but no more so than other functional forms that are commonly applied to empirical data analysis. We consider that such extrapolations will be necessary in the course of the dose reconstructions, and it is our judgment that the errors introduced will be less serious than those which would result from the use of artificially truncated distributions. The fitted distributions must be regarded in the same way that any model which makes predictions beyond available data is regarded. At a more fundamental level, the question of the extent to which one may generalize the observed and fitted distributions — based only on processes sampled in 1985 — to the longer history of the FMPC must be considered. If satisfactory answers to this question cannot be found, relevant components of uncertainty in the predicted doses must be estimated from the 1985 data.

RESULTS

This appendix shows input and output information for the particle-size distribution at the inlet duct and the emission stack for each of the fifteen dust collectors as presented by Boback et al. (1987) but corrected, in two cases (the inlet ducts for G5-254 and G5-256), from the original data sheets. For each distribution in Figs. F–1 through F–15, the accompanying data table gives the identification of the plant and dust collector, the calculated coefficients (i.e., the linear representation, corresponding to Eq. F–4) of the polynomial $T(\zeta)$, the predicted and observed cumulative probabilities (cumulative mass fractions) corresponding to the diameters of the input distribution, the summary statistics of the fitted distribution, and the number and locations (i.e., diameters) of extrema of the density function (i.e., the derivative of $\Phi(x)$). Each plot shows the fitted distribution in relation to the points of the empirical distribution, plotted with reference to log-probability axes. The extent of departure of the plot from linearity is indicative of the extent of departure of the distribution from lognormality. The number of modes is not obvious from these plots; in particular, these examples demonstrate that a curvilinear distribution on log-probability paper does not necessarily correspond to a bimodal distribution. All but perhaps two of the distributions considered here show some curvilinear trend (usually sigmoid). For these two inlet ducts (G4-5 and G4-7), the coefficients of the ζ^2 and ζ^3 terms of the cubic are small (decisively so in the former case), so that $\Phi(x)$ approximates a lognormal distribution.

CONCLUSIONS

In this appendix, we have demonstrated a cubic transformation method, based on nonlinear least-squares polynomial regression, for fitting a CDF to the particle-size distributions measured at the FMPC in 1985. The method is useful as an instrument for interpolation, extrapolation, and smoothing of the distributions. The resulting numeric data and plots are shown in Figs. F–1 through F–15.

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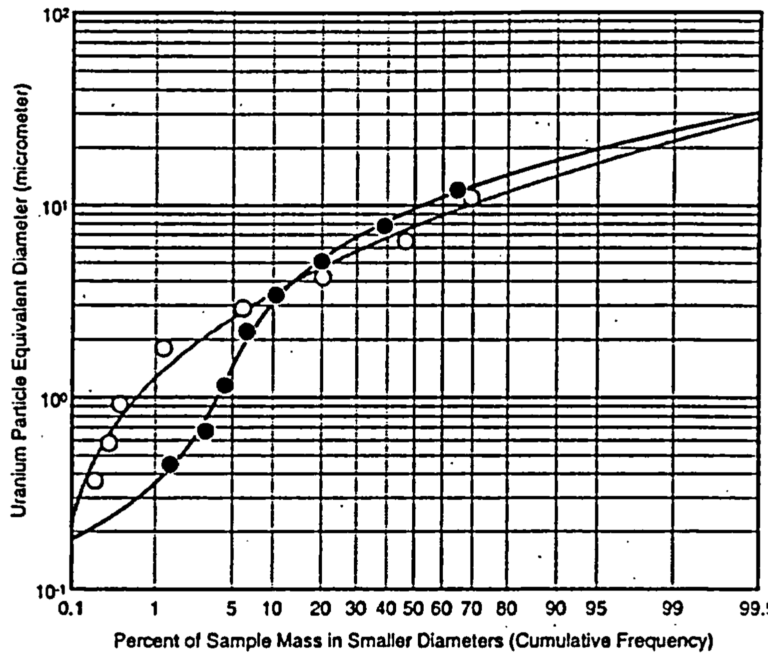

Figure F–1. Particle equivalent diameter distributions for inlet duct and emission stack G4-2.

PLANT 4 DUST COLLECTOR G4-2: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	-2.49664
1	0.667467
2	0.21843
3	0.0238272

Equivalent Diameter	Predicted Probability	Observed Probability
11	0.754384	0.693
6.5	0.372323	0.466
4.2	0.154222	0.202
2.9	0.0655723	0.06
1.8	0.0214851	0.012
0.92	0.00537394	0.004
0.58	0.00256086	0.003
0.37	0.0014998	0.002

Median diameter based on regression: 7.79

Mean of fitted distribution: 8.47
Standard deviation of fitted distribution: 4.41

Number of extrema detected = 3

Diameter	Type of extreme
0.01	max
0.05	min
6.43	max

PLANT 4 DUST COLLECTOR G4-2: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	-1.77972
1	0.364822
2	-0.0553174
3	0.106503

Equivalent Diameter	Predicted Probability	Observed Probability
12	0.662541	0.649
7.8	0.366683	0.39
5.1	0.191719	0.199
3.4	0.11106	0.104
2.2	0.0702061	0.064
1.15	0.0418582	0.043
0.67	0.026097	0.03
0.45	0.0153661	0.014

Median diameter based on regression: 9.63

Mean of fitted distribution: 10
Standard deviation of fitted distribution: 5.49

Number of extrema detected = 3

Diameter	Type of extreme
0.35	max
1.49	min
8.9	max

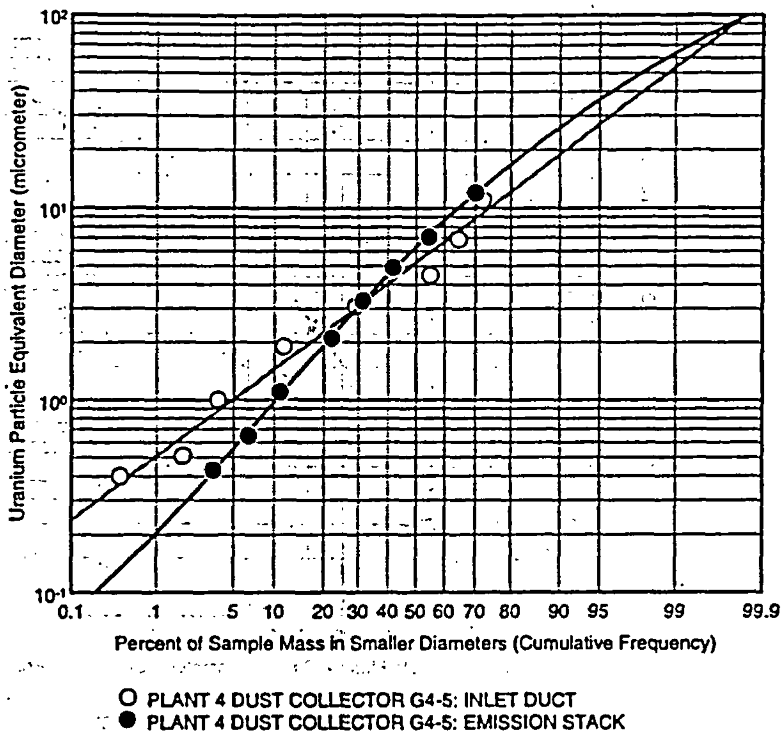

Figure F–2. Particle equivalent diameter distributions for inlet duct and emission stack G4-5.

PLANT 4 DUST COLLECTOR G4-5: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	-1.65107
1	0.99859
2	-1.96857E-07
3	5.28563E-08

Equivalent Diameter	Predicted Probability	Observed. Probability
11	0.771393	0.722
6.9	0.609388	0.646
4.5	0.440731	0.551
3.1	0.301091	0.29
1.9	0.156218	0.115
1	0.049362	0.037
0.51	0.0100771	0.018
0.4	0.0051428	0.004

Median diameter based on regression: 5.22

Mean of fitted distribution: 8.4
Standard deviation of fitted distribution: 9.86

Number of extrema detected = 1

Diameter	Type of extreme
1.92	max

PLANT 4 DUST COLLECTOR G4-5: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	-1.26806
1	0.640205
2	0.00340749
3	0.0122341

Equivalent Diameter	Predicted Probability	Observed Probability
12	0.702479	0.7
7.1	0.536659	0.546
4.9	0.423513	0.418
3.3	0.316314	0.313
2.1	0.215875	0.22
1.1	0.113715	0.109
0.65	0.0612699	0.065
0.43	0.0348924	0.034

Median diameter based on regression: 6.32

Mean of fitted distribution: 10.6
Standard deviation of fitted distribution: 12.5

Number of extrema detected = 1

Diameter	Type of extreme
0.57	max

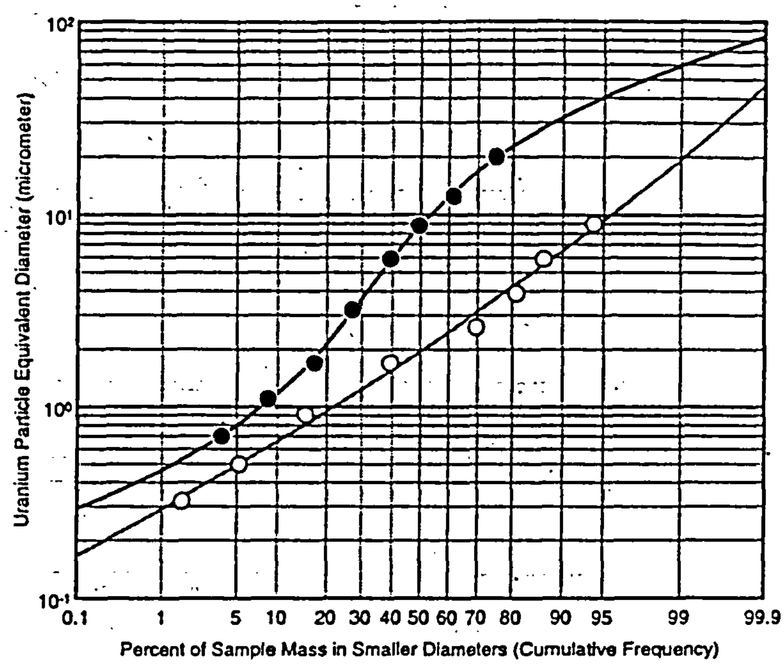

Figure F-3. Particle equivalent diameter distributions for inlet duct and emission stack G4-7.

PLANT 4 DUST COLLECTOR G4-7: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	-0.775328
1	1.19451
2	-0.0523289
3	0.00076522

Equivalent Diameter	Predicted Probability	Observed Probability
9	0.945727	0.94
5.9	0.88185	0.868
3.9	0.774987	0.809
2.6	0.62511	0.696
1.7	0.437974	0.393
0.91	0.18715	0.151
0.5	0.0516888	0.052
0.32	0.0137109	0.016

Median diameter based on regression: 1.95

Mean of fitted distribution: 3.12
Standard deviation of fitted distribution: 4.05

Number of extrema detected = 1

Diameter	Type of extreme
0.9	max

PLANT 4 DUST COLLECTOR G4-7: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	-1.42257
1	0.933117
2	-0.272263
3	0.0658473

Equivalent Diameter	Predicted Probability	Observed Probability
20	0.757941	0.754
12.5	0.601927	0.616
8.8	0.498533	0.493
5.9	0.399024	0.393
3.2	0.273605	0.27
1.7	0.16005	0.17
1.1	0.0907661	0.086
0.7	0.0364854	0.037

Median diameter based on regression: 8.85

Mean of fitted distribution: 13.3
Standard deviation of fitted distribution: 13.5

Number of extrema detected = 1

Diameter	Type of extreme
0.86	max

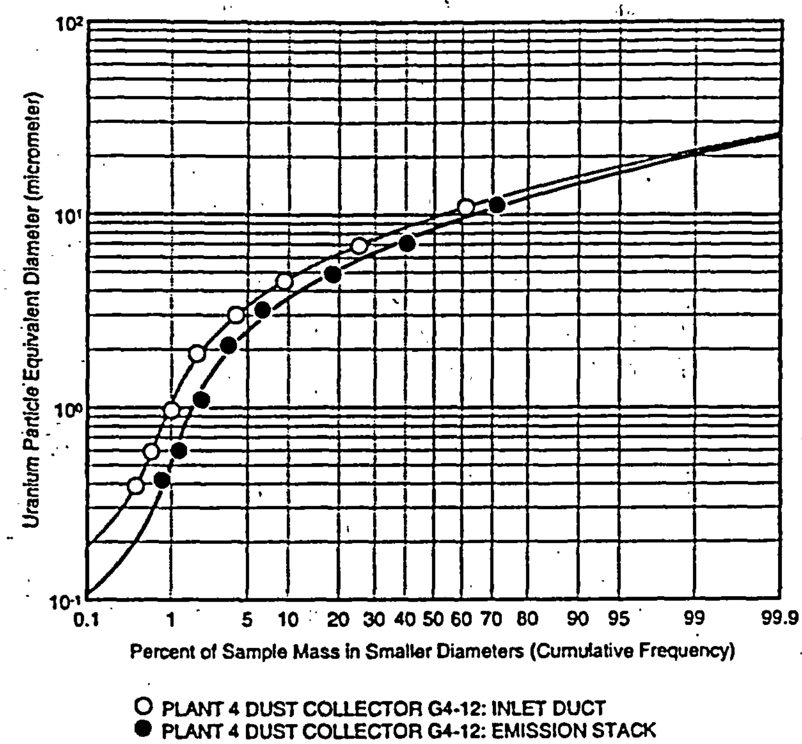

Figure F–4. Particle equivalent diameter distributions for inlet duct and emission stack G4-12.

PLANT 4 DUST COLLECTOR G4-12: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	-2.34342
1	0.324196
2	0.088823
3	0.097744

Equivalent Diameter	Predicted Probability	Observed Probability
11	0.614991	0.612
6.9	0.247777	0.253
4.5	0.0930367	0.093
3	0.0400207	0.04
1.9	0.0190911	0.018
0.97	0.00930586	0.01
0.59	0.00613794	0.006
0.39	0.00400619	0.004

Median diameter based on regression: 9.71

Mean of fitted distribution: 10
Standard deviation of fitted distribution: 4.38

Number of extrema detected = 3

Diameter	Type of extreme
0.24	max
0.85	min
9.08	max

PLANT 4 DUST COLLECTOR G4-12: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	-2.13727
1	0.342225
2	0.139168
3	0.078725

Equivalent Diameter	Predicted Probability	Observed Probability
11.2	0.729691	0.713
7.1	0.367326	0.404
4.9	0.177248	0.185
3.2	0.0767844	0.065
2.1	0.0379824	0.035
1.1	0.0177192	0.02
0.6	0.0111195	0.012
0.42	0.00863733	0.008

Median diameter based on regression: 8.48

Mean of fitted distribution: 8.86
Standard deviation of fitted distribution: 4.27

Number of extrema detected = 3

Diameter	Type of extreme
0.15	max
0.69	min
7.73	max

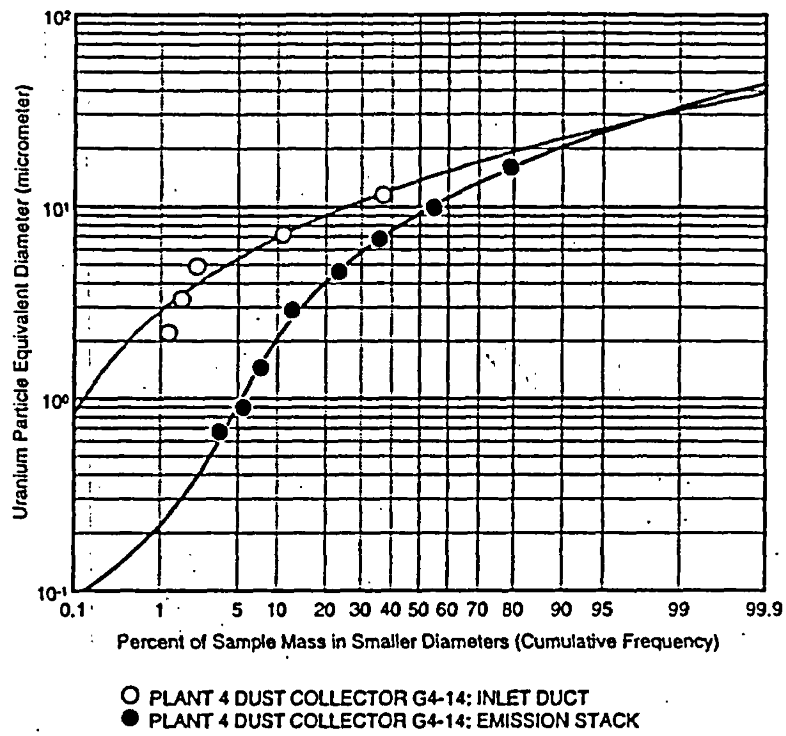

Figure F–5. Particle equivalent diameter distributions for inlet duct and emission stack G4-14.

PLANT 4 DUST COLLECTOR G4-14: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	-3.00214
1	0.475575
2	0.0837776
3	0.0656336

Equivalent Diameter	Predicted Probability	Observed Probability
11.5	0.350237	0.37
7.2	0.108991	0.109
4.9	0.0382558	0.022
3.3	0.0137899	0.016
2.2	0.00549654	0.012
1.1	0.00155823	0.001
0.66	0.000711381	0.0005
0.44	0.000372606	0.0005

Median diameter based on regression: 13.7

Mean of fitted distribution: 14.4
Standard deviation of fitted distribution: 6.17

Number of extrema detected = 1

Diameter	Type of extreme
12.47	max

PLANT 4 DUST COLLECTOR G4-14: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	-1.61264
1	0.403673
2	0.0318019
3	0.0504584

Equivalent Diameter	Predicted Probability	Observed Probability
16	0.795739	0.79
9.9	0.535023	0.55
6.8	0.356978	0.357
4.6	0.228673	0.233
2.9	0.138763	0.125
1.45	0.0727413	0.075
0.9	0.0489744	0.055
0.67	0.0381604	0.035

Median diameter based on regression: 9.26

Mean of fitted distribution: 10.5
Standard deviation of fitted distribution: 7.35

Number of extrema detected = 3

Diameter	Type of extreme
0.21	max
1.34	min
6.74	max

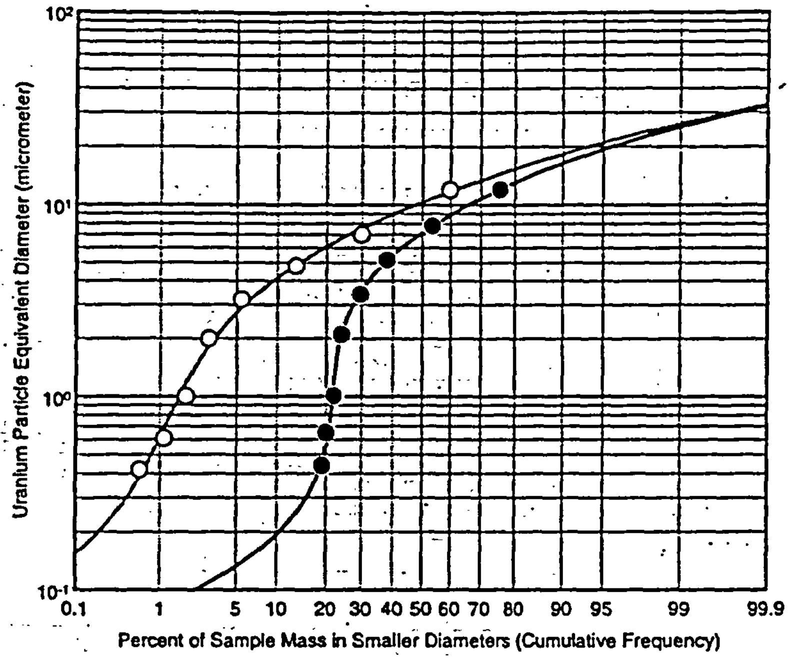

Figure F–6. Particle equivalent diameter distributions for inlet duct and emission stack G5-249.

PLANT 5 DUST COLLECTOR G5-249: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	-2.15589
1	0.369058
2	0.0669841
3	0.0729165

Equivalent Diameter	Predicted Probability	Observed Probability
12	0.615471	0.596
7	0.258874	0.298
4.8	0.129085	0.132
3.2	0.0640987	0.054
2	0.0326201	0.029
1	0.0155463	0.018
0.61	0.00988324	0.011
0.42	0.00669466	0.006

Median diameter based on regression: 10.3

Mean of fitted distribution: 10.9
Standard deviation of fitted distribution: 5.59

Number of extrema detected = 3

Diameter	Type of extreme
0.23	max
0.88	min
9.2	max

PLANT 5 DUST COLLECTOR G5-249: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	-0.784878
1	0.0638725
2	-0.00260951
3	0.0871555

Equivalent Diameter	Predicted Probability	Observed Probability
12	0.756477	0.758
7.8	0.536138	0.534
5.2	0.383574	0.38
3.4	0.290856	0.295
2.1	0.240925	0.24
1.01	0.216449	0.22
0.65	0.206152	0.2
0.44	0.187458	0.19

Median diameter based on regression: 7.19

Mean of fitted distribution: 7.83
Standard deviation of fitted distribution: 6.32

Number of extrema detected = 3

Diameter	Type of extreme
0.13	max
1.14	min
7	max

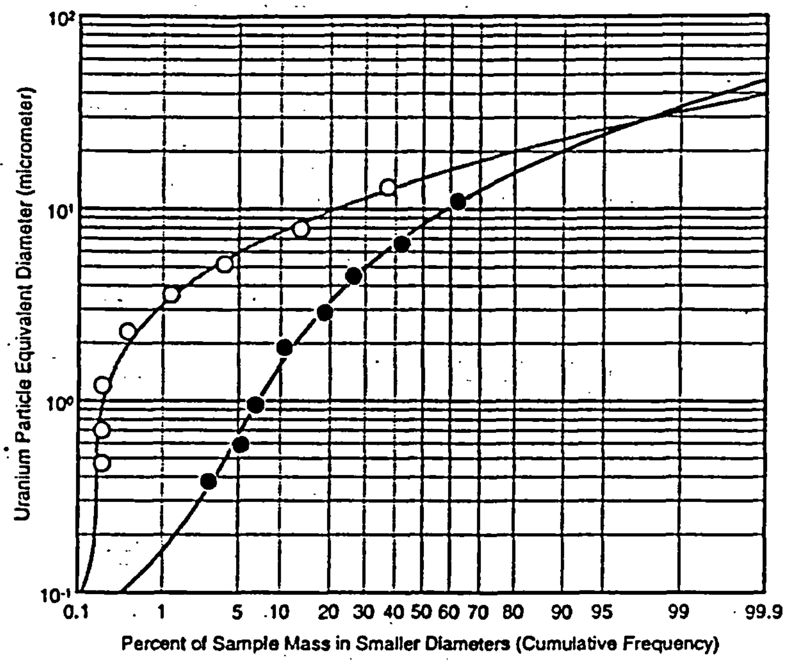

Figure F-7. Particle equivalent diameter distributions for inlet duct and emission stack G5-250.

PLANT 5 DUST COLLECTOR G5-250: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	-2.86696
1	0.178617
2	0.17733
3	0.0586841

Equivalent Diameter	Predicted Probability	Observed Probability
13	0.400562	0.373
7.9	0.110836	0.134
5.2	0.0338114	0.038
3.6	0.0130788	0.012
2.3	0.00521454	0.004
1.2	0.00234079	0.002
0.7	0.00180256	0.002
0.47	0.00171669	0.002

Median diameter based on regression: 14.5

Mean of fitted distribution: 15.1
Standard deviation of fitted distribution: 6.19

Number of extrema detected = 3

Diameter	Type of extreme
0.06	max
0.37	min
13.33	max

PLANT 5 DUST COLLECTOR G5-250: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	-1.47564
1	0.419789
2	0.0436889
3	0.0406511

Equivalent Diameter	Predicted Probability	Observed Probability
11	0.634072	0.625
6.6	0.399468	0.421
4.5	0.271894	0.264
2.9	0.17616	0.185
1.9	0.119507	0.106
0.95	0.0671878	0.067
0.59	0.0454234	0.052
0.38	0.0302079	0.028

Median diameter based on regression: 8.35

Mean of fitted distribution: 9.9
Standard deviation of fitted distribution: 7.61

Number of extrema detected = 3

Diameter	Type of extreme
0.16	max
1.47	min
5.01	max

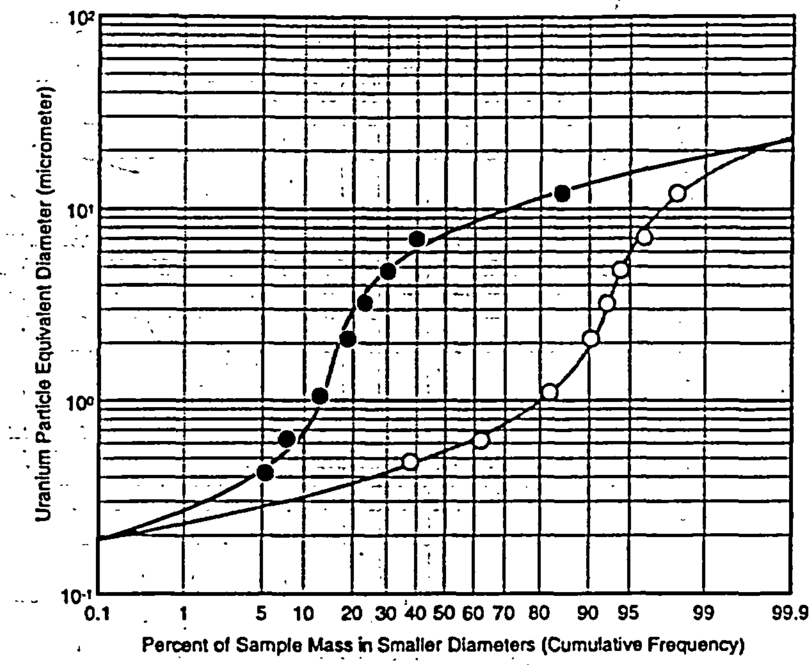

Figure F–8. Particle equivalent diameter distributions for inlet duct and emission stack G5-251.

PLANT 5 DUST COLLECTOR G5-251: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	0.846798
1	0.98466
2	-0.576711
3	0.154204

Equivalent Diameter	Predicted Probability	Observed Probability
12	0.982074	0.981
7.1	0.957499	0.962
4.8	0.941501	0.941
3.2	0.9271	0.925
2.1	0.907061	0.902
1.1	0.825245	0.823
0.62	0.589967	0.623
0.48	0.402236	0.38

Median diameter based on regression: 0.544

Mean of fitted distribution: 1.28
Standard deviation of fitted distribution: 2.65

Number of extrema detected = 1

Diameter	Type of extreme
0.39	max

PLANT 5 DUST COLLECTOR G5-251: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	-1.12925
1	0.297224
2	-0.231645
3	0.181877

Equivalent Diameter	Predicted Probability	Observed Probability
12	0.833889	0.85
7	0.464985	0.4
4.75	0.294427	0.302
3.25	0.211011	0.231
2.1	0.168036	0.186
1.05	0.132366	0.127
0.63	0.0911078	0.076
0.42	0.046464	0.053

Median diameter based on regression: 7.42

Mean of fitted distribution: 7.45
Standard deviation of fitted distribution: 4.62

Number of extrema detected = 3

Diameter	Type of extreme
0.38	max
1.78	min
7.68	max

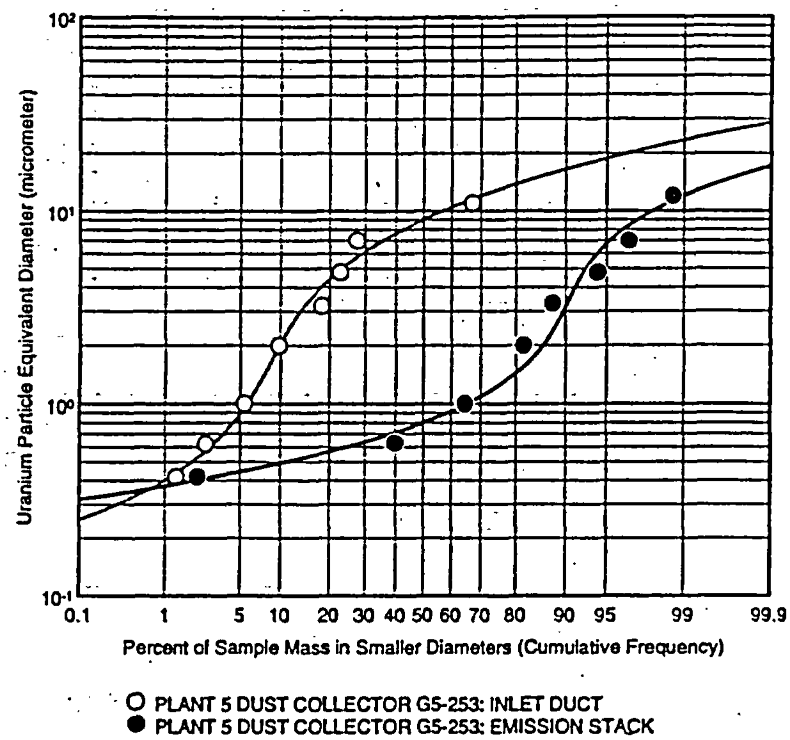

Figure F-9. Particle equivalent diameter distributions for inlet duct and emission stack G5-253.

PLANT 5 DUST COLLECTOR G5-253: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	-1.58257
1	0.504374
2	-0.218532
3	0.143409

Equivalent Diameter	Predicted Probability	Observed Probability
11	0.635932	0.672
7	0.354982	0.275
4.8	0.218993	0.227
3.2	0.143237	0.181
2	0.0984905	0.097
1	0.0567598	0.054
0.62	0.0294271	0.025
0.42	0.0113575	0.013

Median diameter based on regression: 9.05

Mean of fitted distribution: 9.35
Standard deviation of fitted distribution: 5.42

Number of extrema detected = 3

Diameter	Type of extreme
0.51	max
2.09	min
8.71	max

PLANT 5 DUST COLLECTOR G5-253: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	0.395111
1	1.5468
2	-0.991084
3	0.27408

Equivalent Diameter	Predicted Probability	Observed Probability
12	0.98995	0.987
7	0.952713	0.967
4.8	0.925164	0.941
3.3	0.90244	0.878
2	0.860458	0.817
1	0.65362	0.648
0.63	0.288363	0.396
0.42	0.0306367	0.021

Median diameter based on regression: 0.801

Mean of fitted distribution: 1.55
Standard deviation of fitted distribution: 2.27

Number of extrema detected = 1

Diameter	Type of extreme
0.59	max

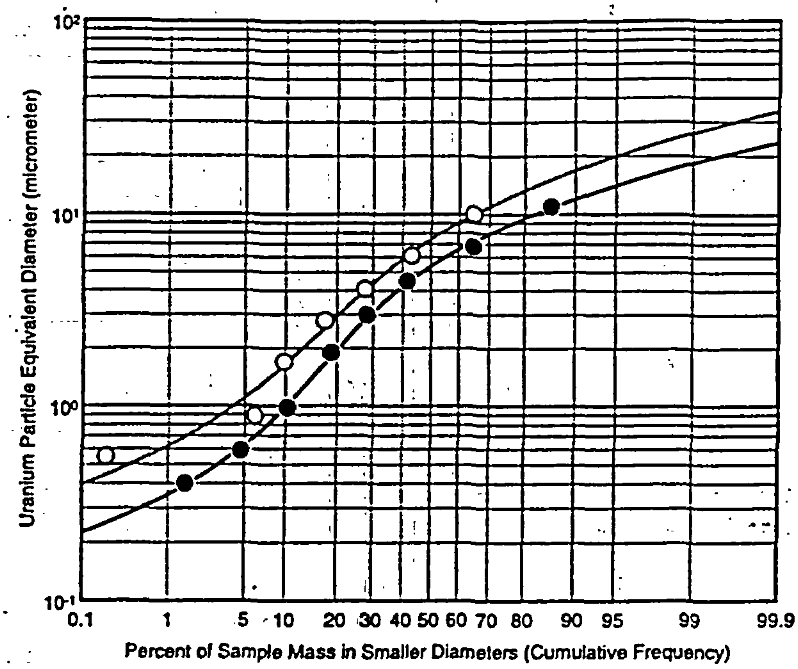

Figure F–10. Particle equivalent diameter distributions for inlet duct and emission stack G5-254.

PLANT 5 DUST COLLECTOR G5-254: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	-1.74205
1	1.051
2	-0.326252
3	0.117889

Equivalent Diameter	Predicted Probability	Observed Probability
10	0.650774	0.649
6.1	0.416333	0.432
4.1	0.281811	0.277
2.8	0.190214	0.172
1.7	0.104086	0.098
0.89	0.0308016	0.06
0.55	0.00599949	0.002
0.37	0.000629015	0.001

Median diameter based on regression: 7.4

Mean of fitted distribution: 8.51
Standard deviation of fitted distribution: 6.04

Number of extrema detected = 1

Diameter	Type of extreme
1.13	max

PLANT 5 DUST COLLECTOR G5-254: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	-1.27232
1	0.699124
2	-0.167917
3	0.120992

Equivalent Diameter	Predicted Probability	Observed Probability
11	0.865807	0.858
6.8	0.619082	0.651
4.5	0.425057	0.416
3	0.292363	0.283
1.9	0.194682	0.187
0.98	0.099131	0.103
0.59	0.0440296	0.047
0.4	0.0158973	0.015

Median diameter based on regression: 5.36

Mean of fitted distribution: 6.04
Standard deviation of fitted distribution: 4.31

Number of extrema detected = 3

Diameter	Type of extreme
0.57	max
2.8	min
4.03	max

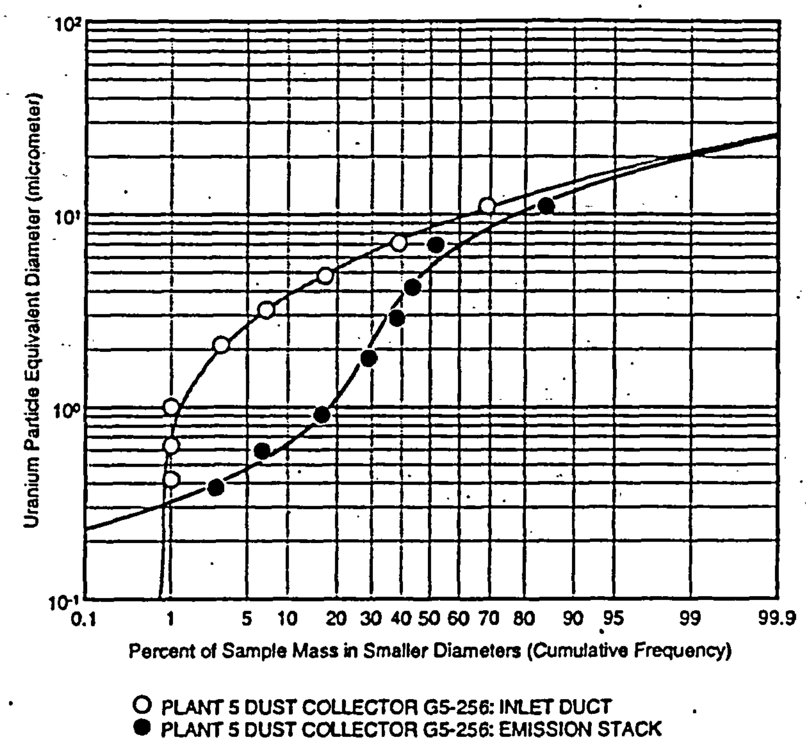

Figure F–11. Particle equivalent diameter distributions for inlet duct and emission stack G5-256.

PLANT 5 DUST COLLECTOR G5-256: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	-2.23783
1	0.321723
2	0.224742
3	0.0523317

Equivalent Diameter	Predicted Probability	Observed Probability
11	0.707948	0.69
7.1	0.363288	0.39
4.8	0.163988	0.17
3.2	0.0698094	0.07
2.1	0.0318662	0.03
1	0.012616	0.01
0.63	0.00954764	0.01
0.42	0.00861032	0.01

Median diameter based on regression: 8.53

Mean of fitted distribution: 8.99
Standard deviation of fitted distribution: 4.36

Number of extrema detected = 3

Diameter	Type of extreme
0.03	max
0.24	min
7.6	max

PLANT 5 DUST COLLECTOR G5-256: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	-0.913069
1	0.660854
2	-0.331578
3	0.156728

Equivalent Diameter	Predicted Probability	Observed Probability
11	0.822766	0.847
6.9	0.600922	0.519
4.2	0.426869	0.437
2.9	0.345991	0.382
1.8	0.271807	0.289
0.91	0.16392	0.163
0.59	0.0842422	0.065
0.38	0.0224867	0.027

Median diameter based on regression: 5.34

Mean of fitted distribution: 6.18
Standard deviation of fitted distribution: 5.02

Number of extrema detected = 3

Diameter	Type of extreme
0.52	max
3.2	min
5.65	max

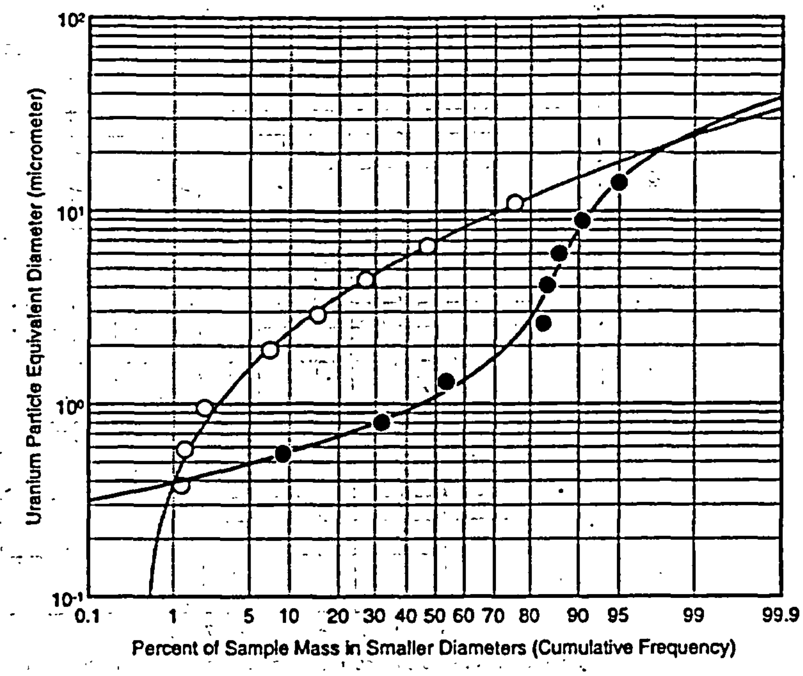

Figure F–12. Particle equivalent diameter distributions for inlet duct and emission stack G5-260.

PLANT 5 DUST COLLECTOR G5-260: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	-1.92417
1	0.577013
2	0.176517
3	0.0179997

Equivalent Diameter	Predicted Probability	Observed Probability
11	0.76503	0.758
6.6	0.465827	0.469
4.4	0.266564	0.27
2.9	0.1383	0.15
1.9	0.0699279	0.072
0.95	0.0253921	0.02
0.58	0.0142981	0.013
0.38	0.00981042	0.012

Median diameter based on regression: 7

Mean of fitted distribution: 8.01
Standard deviation of fitted distribution: 5.19

Number of extrema detected = 3

Diameter	Type of extreme
0.01	max
0.04	min
5.02	max

PLANT 5 DUST COLLECTOR G5-260: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	-0.132047
1	1.54057
2	-0.72514
3	0.149104

Equivalent Diameter	Predicted Probability	Observed Probability
14	0.947791	0.95
8.9	0.907918	0.905
6	0.876562	0.866
4.1	0.845388	0.841
2.6	0.790455	0.831
1.3	0.588979	0.537
0.8	0.303773	0.319
0.55	0.0894604	0.09

Median diameter based on regression: 1.09

Mean of fitted distribution: 2.94
Standard deviation of fitted distribution: 5.08

Number of extrema detected = 1

Diameter	Type of extreme
0.68	max

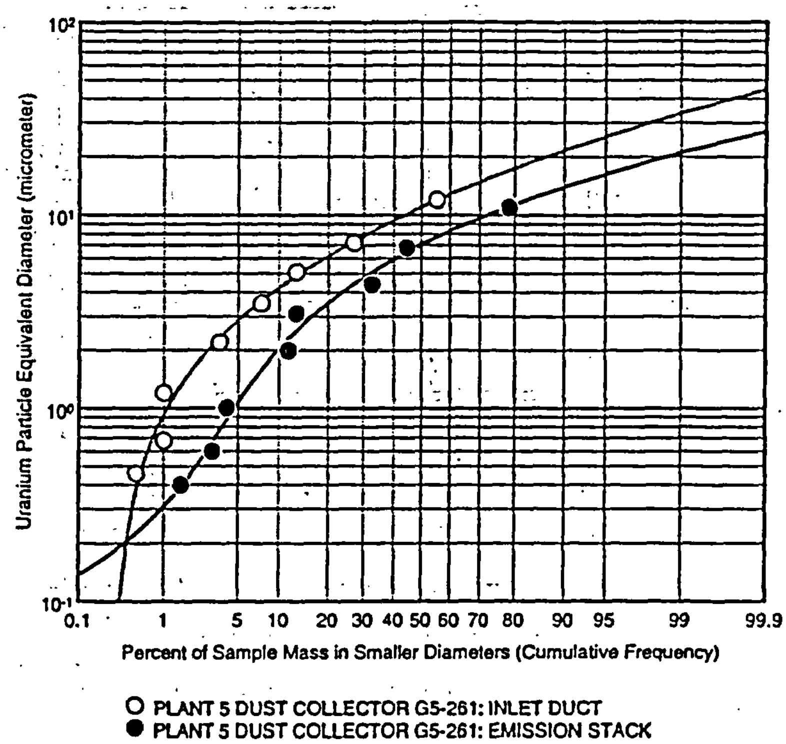

Figure F–13. Particle equivalent diameter distributions for inlet duct and emission stack G5-261.

PLANT 5 DUST COLLECTOR G5-261: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	-2.29694
1	0.415779
2	0.161617
3	0.0266991

Equivalent Diameter	Predicted Probability	Observed Probability
12	0.557189	0.555
7.2	0.260782	0.27
5.1	0.141172	0.13
3.5	0.0707907	0.075
2.2	0.0317587	0.035
1.2	0.0133596	0.01
0.68	0.00745039	0.01
0.46	0.00562473	0.005

Median diameter based on regression: 11

Mean of fitted distribution: 12.2
Standard deviation of fitted distribution: 7.06

Number of extrema detected = 3

Diameter	Type of extreme
0.01	max
0.22	min
8.71	max

PLANT 5 DUST COLLECTOR G5-261: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	-1.68163
1	0.491472
2	0.0411743
3	0.0753348

Equivalent Diameter	Predicted Probability	Observed Probability
11	0.780035	0.785
6.8	0.477049	0.445
4.4	0.268268	0.325
3.1	0.167583	0.13
2	0.0974717	0.115
1	0.0463207	0.04
0.6	0.0266811	0.03
0.4	0.0155676	0.015

Median diameter based on regression: 7.07

Mean of fitted distribution: 7.67
Standard deviation of fitted distribution: 4.68

Number of extrema detected = 3

Diameter	Type of extreme
0.31	max
1.01	min
5.91	max

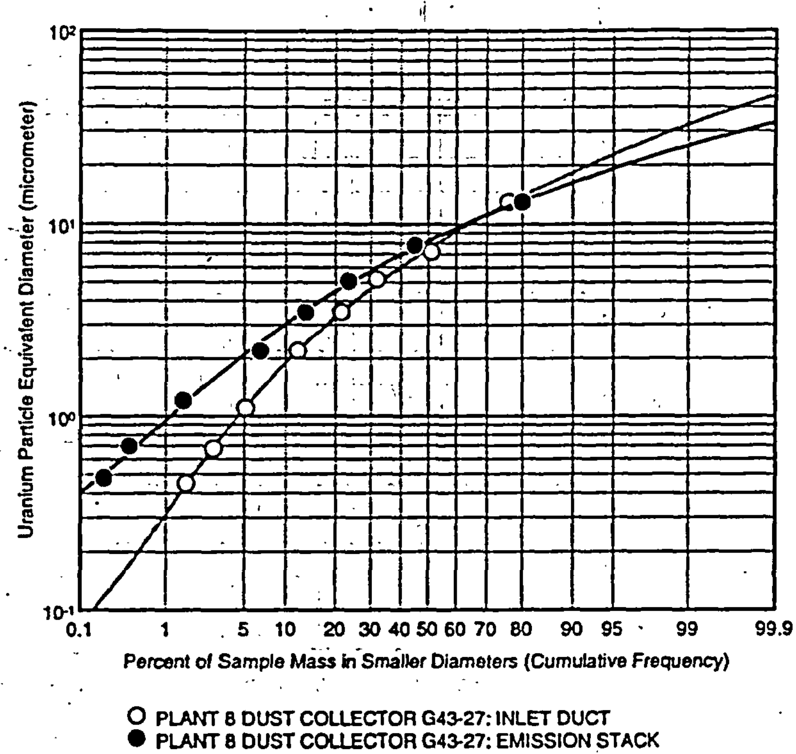

Figure F–14. Particle equivalent diameter distributions for inlet duct and emission stack G43-27.

PLANT 8 DUST COLLECTOR G43-27: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	-1.69976
1	0.592724
2	0.0746656
3	0.0251448

Equivalent Diameter	Predicted Probability	Observed Probability
13	0.76916	0.764
7.2	0.481949	0.512
5.2	0.342029	0.318
3.5	0.21459	0.215
2.2	0.120262	0.12
1.1	0.0502364	0.051
0.68	0.0275121	0.028
0.45	0.0162484	0.016

Median diameter based on regression: 7.48

Mean of fitted distribution: 9.11
Standard deviation of fitted distribution: 7.02

Number of extrema detected = 1

Diameter	Type of extreme
4.06	max

PLANT 8 DUST COLLECTOR G43-27: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	-2.28575
1	0.83555
2	-0.00280343
3	0.0576776

Equivalent Diameter	Predicted Probability	Observed Probability
13	0.791675	0.8
7.8	0.467583	0.45
5.1	0.247481	0.234
3.5	0.129238	0.134
2.2	0.0547524	0.066
1.2	0.0164563	0.015
0.7	0.00484433	0.004
0.48	0.00173144	0.002

Median diameter based on regression: 8.21

Mean of fitted distribution: 9.11
Standard deviation of fitted distribution: 5.36

Number of extrema detected = 1

Diameter	Type of extreme
6.33	max

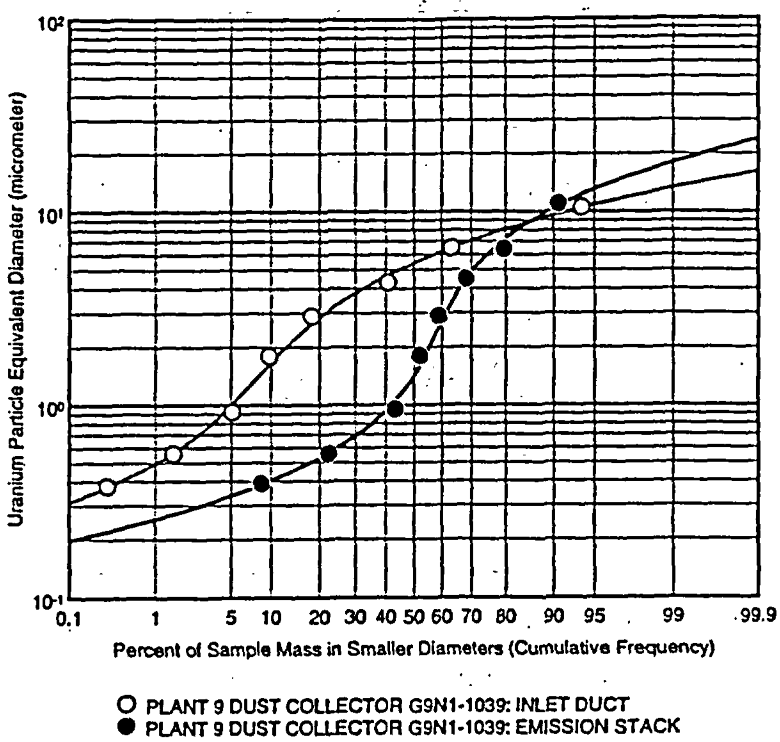

Figure F-15. Particle equivalent diameter distributions for inlet duct and emission stack G9N1-1039.

PLANT 9 DUST COLLECTOR G9N1-1039: INLET DUCT

Coefficients of polynomial

i	coeff[i]
0	-1.64175
1	0.752471
2	-0.195005
3	0.192257

Equivalent Diameter	Predicted Probability	Observed Probability
10.5	0.939296	0.937
6.5	0.634708	0.632
4.3	0.358512	0.407
2.9	0.203384	0.179
1.8	0.109764	0.097
0.92	0.044008	0.051
0.56	0.0145888	0.015
0.38	0.00319984	0.003

Median diameter based on regression: 5.42

Mean of fitted distribution: 5.6
Standard deviation of fitted distribution: 2.99

Number of extrema detected = 3

Diameter	Type of extreme
0.72	max
1.45	min
5.23	max

PLANT 9 DUST COLLECTOR G9N1-1039: EMISSION STACK

Coefficients of polynomial

i	coeff[i]
0	-0.200583
1	0.696287
2	-0.406941
3	0.161866

Equivalent Diameter	Predicted Probability	Observed Probability
11	0.913231	0.907
6.4	0.76579	0.794
4.5	0.683265	0.681
2.9	0.608271	0.588
1.8	0.540209	0.52
0.95	0.406177	0.432
0.56	0.219861	0.222
0.39	0.0881633	0.085

Median diameter based on regression: 1.42

Mean of fitted distribution: 3.77
Standard deviation of fitted distribution: 4.41

Number of extrema detected = 1

Diameter	Type of extreme
0.44	max

APPENDIX G

ESTIMATES OF BIAS IN EFFLUENT SAMPLING FOR PARTICLES

INTRODUCTION

This appendix treats three possible sources of bias in particle sampling results for dust collector exhaust stacks at the FMPC. These are (a) nonrepresentative sampling, due to use of a single sampling probe; (b) anisokinetic sampling, due to a mismatch between the fluid velocity in the probe and that in the stack; and (c) losses of particles, due to deposition or impaction on the wall of the sampling line between the probe inlet and the collection filter. These issues have not been addressed in previous analyses of the uranium release data.

The effect of using a single sampling probe in large ducts can only be estimated quantitatively. Calculation methods for estimating anisokinetic sampling bias and line losses are presented together with example calculations for uranium aerosols. The distributions used to characterize specific variables for Monte Carlo calculations of corrections for previous sampling conditions (Appendix E) are described. Results of some of the calculations and of a sensitivity analysis are presented.

NONREPRESENTATIVE SAMPLING

One design feature that was not consistent with standard guidance for sample collection from exhaust ducts was the use of a single sampling probe in larger ducts. The guide developed by the American National Standards Institute (ANSI, 1969) guide recommends multiple sample withdrawal points for ducts greater than 15 cm in diameter. The reason for multiple probes is to provide assurance that the samples will not be biased because of a nonuniform distribution of the contaminant in the stack. The sample extracted from the center of a dust collector exhaust stack would be representative if the particles were uniformly mixed in the exhaust or if the concentration on the centerline happened to be equal to the average concentration in the stack. When this is not the case, the sample is not representative of the material being discharged. The bias introduced may be positive or negative, depending upon the actual relationship between the centerline and average concentrations. Quantitative assessment of this question requires tracer measurements in the exhaust stacks. Such an effort is well beyond the scope of this work and is not feasible for many of the exhausts, so a qualitative assessment must be made.

Two features favor a well mixed exhaust stream at the point of sample extraction: the exhaust systems were generally simple and the sampling point was downstream of the discharge fan. Operation of the fan tends to mix exhausts that may be inhomogeneous at the inlet to the fan. Combinations of exhausts from several individual discharge fans are frequently not well mixed, so a complex exhaust duct arrangement would indicate greater potential for inhomogeneity. On the other hand, the exhaust stack diameters, some greater than 1 m, increase concern about possible nonrepresentative sampling. Only one of the ducts sampled was less than 15 cm in diameter (*RAC* 1991).

It is assumed that the variation of the concentration distribution is greater for stacks with larger diameters. Thus, the chance that the concentration determined from a single sample of the exhaust stack will differ from the average concentration in the stack increases with stack size. Estimated ranges of possible bias (B) for six stack diameter categories are shown in Table G–1. These estimates are based on practical experience in measuring tracer concentration profiles in stacks and ducts. As noted above, the bias may have been in either direction. In the lower portion of the table are the stacks whose diameters (or effective diameters for the few rectangular stacks) lie within the ranges shown. The range of uncertainty at the top of the column is applied to all stacks listed in that column. Not shown in the table is stack G2–235, whose diameter was less than 15 cm. For that stack the fractional uncertainty for concentration inhomogeneity is estimated to be 8%.

ANISOKINETIC SAMPLING

Accurate sampling of the particulates in gaseous effluents often requires that the fluid velocity in the sampling probe (u, cm s^{-1}) be the same as the velocity of the stack gas at the point of sampling (v, cm s^{-1}). When this condition is achieved, the sampling is termed "isokinetic." Deviations from this condition, called "anisokinetic sampling," can lead to bias in the sample. The bias may be high or low, depending upon whether the sampling flow rate yields a probe fluid velocity $u < v$ or $u > v$. The method for estimating the bias is discussed below.

General Approach

The approach developed by Durham and Lundgren (1980) was used to assess the potential effects of deviations from isokinetic sampling conditions. The consequences of such deviations depend upon the ratio of the fluid velocities (u/v), the size and densities of the particles to be sampled, the diameter of the sampling probe, and, to a much lesser extent, the air temperature. Improper alignment of the sampling probe along the streamlines of flow in the stack can also lead to sampling biases. However, if the probe axis is within 15° of the proper position, the effects of misalignment are small, ~5% (Durham and Lundgren 1980). In this assessment, it was assumed that alignment of the sampling probe was sufficiently accurate to make any bias from that source small compared with measurement uncertainties.

For a properly aligned sampling probe, the ratio (R) of the sampled concentration of particulates to the concentration actually present in the stack can be computed using the following equations.

$$R = 1 + [(v/u) - 1][1 - (1/k)] \qquad \text{(G–1)}$$

with

$$k = 1 + [2 + 0.62\,u/v]\,\mathrm{Stk} \qquad \text{(G–2)}$$

Table G-1. Estimated Bias (B) Due to Nonrepresentative Sampling of Uranium Concentration in Dust Collector Exhaust Stacks

Categories of stack diameters (D_s, cm)					
$15 < D_s \leq 30$	$30 < D_s \leq 46$	$46 < D_s \leq 61$	$61 < D_s \leq 76$	$76 < D_s \leq 91$	$91 < D_s \leq 122$
Estimated bias (B) due to nonrepresentative sampling in stack of diameter D_s					
±10%	±15%	±20%	±30%	±35%	±50%
Dust collector exhaust stacks grouped by size category					
G2–1	G2–68	G2–63	G4–3	Mid ESP	G6–6057
G2–171	G2–172	G2–64	G4–7	G9N1–1039	North ESP
G2–174	G1–94	G2–67	G4–14	735–13–7050	South ESP
G2–6042	G1–754	G2–76	G5–A100	G5–259	G9E2–400
G4–1	G1–856	G2–77	G43–27	G5–261	
G4–4	G5–247	G1–252	G42–615		
G4–5	G5–248	3–N	G42A–100		
G4–12	G6–86	3–S	108843		
G4–13	G6–88	G4–2			
G4–15	G4–2509	G4–8			
G4–7001	G4–2510	G5–249			
G55–E–100	G3A–2	G5–250			
G8–7	G8–1	G5–251			
6018	G8N1–1000	G5–252			
6019	G43–29	G5–253			
8002	G43–44C	G5–254			
8021	8035	G5–256			
8024	8057	G5–258			
8083	G37–5011	G5–260			
2102		G5–A101			
G–1		G4–2507			
G–2		G4–2508			
		G8–2			
		G8–3			
		G8–4			
		G20–20			
		G6–93A			
		735–13–7041			

where u and v are the velocities defined above and Stk is the Stokes number for the particles. It is given by

$$\mathrm{Stk} = [\rho d^2 C_c v]/[18 \eta \delta] \tag{G-3}$$

where ρ and d are the density (g cm^{-3}) and physical diameter (cm) of the particle, respectively, C_c is the dimensionless Cunningham slip correction factor for the particle, η is the viscosity of the exhaust air (dyne s cm^{-2}) and δ is the diameter (cm) of the probe opening. The factor C_c can be calculated using the following empirical equation (Hinds 1982):

$$C_c = 1 + 2\,[\,6.32 + 2.01\,\exp\,(-0.1095\,P\,d\,10^4\,)]\,/\,(P\,d\,10^4) \qquad \text{(G–4)}$$

where d is the particle diameter, as defined above, P is the absolute pressure (cm Hg), and the factor of 10^4 is just the conversion from cm to µm. Calculations show that C_c is quite close to unity for particles with diameters greater than 1 µm.

Results for Typical Conditions

The sequence of equations given above were used to compute the effects of deviations from isokinetic sampling conditions. The calculations did not address an exhaustive list of sampling conditions that were present in the many exhaust stacks. The actual conditions prevailing when deviations from isokinetic sampling occurred are not known. Based upon a review of effluent discharge points at the FMPC (RAC 1991), a representative set of conditions (listed below) was chosen for the calculations.

Air temperature: 20°C
Air pressure (P): 76 cm Hg
Air viscosity (η): 1.81×10^{-4} dyne s cm^{-2}
Air density (ρ_a): 1.2×10^{-2} g cm^{-2}
Stack gas velocity (v): 1500 cm s^{-1}
Sampling probe diameter (δ): 0.62 cm

Calculations were performed for four chemical forms of uranium that were present in FMPC effluents: U_3O_8, UO_3, UF_4, and UO_2. Results are presented for spherical particles of UF_4 and UO_2, whose densities (4.7 and 10.9 g cm^{-3}, respectively) bound the range for the chemical forms considered (Hodgman et al. 1959).

The results in Table G–2 show that deviations from isokinetic conditions have little effect upon sampling for small particles. For aerodynamic diameters that are less than 2 µm, the bias introduced is 25% or less. As the particle diameter increases, the Stokes number increases, k becomes smaller, and the concentration ratio R approaches the limiting value of (v/u).

Increasing the density of the particles increases the aerodynamic diameter, $[\,d\,\sqrt{\rho}\,]$, and Stokes number. Table G–3, which contains results for UO_2, shows the more rapid approach to the limiting value of R as the physical diameter of the particles increases. Results for particles of U_3O_8 and UO_2 are qualitatively similar to those shown in Tables G–2 and G–3 but reflect the different densities of those materials. The concentration ratio for a specific aerodynamic diameter is the same, regardless of the chemical form of the particle.

Method Used to Estimate Corrections for Previous Sampling

As the tabulations show, anisokinetic sampling may lead to either an overestimate or an underestimate of the amount of material in the air being discharged. The discharge would be underestimated if the sampling flow rate exceeded the appropriate value for a par-

Table G-2. Calculated Ratios of Probe Concentration to Stack Concentration for Anisokinetic Sampling of UF_4 Particles

Diameter (µm)		Concentration ratio (R) for specific (u/v)					
Physical	Aerodynamic	0.25	0.50	0.75	1.25	1.5	2.0
0.1	0.22	1.01	1.00	1.00	1.00	1.00	1.00
0.2	0.43	1.02	1.01	1.00	1.00	1.00	1.00
0.3	0.65	1.03	1.01	1.00	1.00	1.00	0.99
0.5	1.1	1.07	1.03	1.01	0.99	0.99	0.98
0.8	1.7	1.16	1.06	1.02	0.99	0.98	0.96
1	2.2	1.24	1.09	1.03	0.98	0.96	0.94
2	4.3	1.74	1.26	1.09	0.94	0.90	0.84
3	6.5	2.25	1.43	1.15	0.90	0.84	0.74
5	11	2.98	1.67	1.23	0.86	0.76	0.63
8	17	3.49	1.84	1.28	0.83	0.71	0.56
10	22	3.65	1.89	1.30	0.82	0.70	0.54
20	43	3.90	1.97	1.32	0.81	0.67	0.51

Table G-3. Calculated Ratios of Probe Concentration to Stack Concentration for Anisokinetic Sampling of UO_2 Particles

Diameter (µm)		Concentration ratio (R) for specific (u/v)					
Physical	Aerodynamic	0.25	0.50	0.75	1.25	1.5	2.0
0.1	0.33	1.02	1.01	1.00	1.00	1.00	1.00
0.2	0.66	1.04	1.01	1.00	1.00	0.99	0.99
0.3	0.99	1.07	1.03	1.01	0.99	0.99	0.98
0.5	1.7	1.16	1.06	1.02	0.99	0.98	0.96
0.8	2.6	1.36	1.13	1.04	0.97	0.95	0.92
1	3.3	1.51	1.18	1.06	0.96	0.93	0.88
2	6.6	2.29	1.45	1.15	0.90	0.83	0.73
3	9.9	2.87	1.64	1.22	0.86	0.77	0.64
5	16	3.45	1.83	1.28	0.83	0.71	0.56
8	26	3.76	1.92	1.31	0.81	0.69	0.53
10	33	3.84	1.95	1.32	0.81	0.68	0.52
20	66	3.96	1.97	1.33	0.80	0.67	0.50

ticular duct or if the flow rate in that duct was actually lower than that upon which the standard sampling rate was based. Overestimates may be due to a reduced sampling flow rate or an increase in the duct discharge rate (due, for example, to repair of the exhaust fan).

Estimates of releases from the dust collector exhausts at the FMPC relied on isokinetic sampling of the stacks. When the probe velocity and the stack fluid velocity are equal, there is a very simple relationship between the mass of material released from the stack and the mass of material collected on the filter. That relationship, which involves the flows through the sampling probe and the stack, was used to estimate releases from the dust collector exhausts. Because the flow through a tube is the product of the fluid velocity and the cross-sectional area, one way to express the relationship is as follows:

$$Q_r = [v_a / v][A_s / A_p]M_f \tag{G–5}$$

where Q_r is the amount of material released (g), v_a is the average fluid velocity (cm s^{-1}) in the stack, v is the fluid velocity (cm s^{-1}) at the stack centerline (the point of sampling), A_s and A_p are the areas (cm^2) of the stack and the probe, respectively, and M_f is the amount of material (g) found on the filter. Implicit in Equation (G–5) is the assumption that $u = v$, which reflects the fact that the effluent sampling systems were designed to operate isokinetically. An equivalent expression for Q_r is

$$Q_r = [F_s^* / F_p^*]M_f \tag{G–6}$$

where F_s^* and F_p^* are the current "standard" flow rates in the stack and sampling probe, respectively. That is, F_p^* is the computed sampling flow rate that would provide isokinetic sampling for the stack whose measured flow rate was F_s^*. The value of F_p^* for a given stack was adjusted when a new value for F_s^* was obtained from pitot tube measurements in the stack.

At this time, previous deviations from isokinetic conditions can only be estimated based upon knowledge of the systems and measured variability of operating flow rates. Flow rates were recorded at the beginning and ending of a sampling period (Boone 1956b), but those data are unavailable. The sampling flow rate was established by setting a flow control valve to achieve a desired rotameter setting at the start of sampling period. Vibration could cause the valve opening to increase or decrease during the period. The flow was checked again at the end of the period; however the procedure does not specify use of the flow rate data to make corrections in the standard release estimate. The pleated filters used for sample collection had a large surface area. This would reduce the chance of sampling flow reduction due to loading of the filter; however, there were times when filter loadings were substantial and flow reductions may have resulted.

To account for possible variations in parameters that would produce anisokinetic sampling conditions, distributions were developed to characterize each of the parameters. Given the chemical form and particle size distribution for the uranium effluent from a particular stack (Appendix E), an estimate of the density and a set of independently selected, representative particle diameters were obtained. This information, together with randomly selected, independent estimates of F_p^* and F_s,* was used to estimate the effect of anisokinetic sampling by computing R, using Equation (G–1), for each representative particle size in the distribution. The distributions chosen for particular parameters are described in the subsections that follow. The bounds and shapes of the distributions are best estimates given the present knowledge.

Chemical Form. The chemical form(s) of uranium released from particular stacks in the various plants have been discussed in Appendix E. The particle density is dependent on the chemical form of the material being released. As noted above, the range of densities for the uranium compounds of greatest interest range from 4.7 to 10.9 g cm^{-3}. The purity of the material will also affect its density; mixtures of MgF_2 and UF_4 will have densities lower than the value of 4.7 g cm^{-3} for pure UF_4. In the absence of definitive data on the chemical purities of the exhaust streams, a right triangular distribution with lower limit of 0.9 ρ^*

and upper limit and mode of ρ^* was selected. The quantity ρ^* is the density (g cm^{-3}) of the pure form of the expected uranium compound.

Particle Size Distribution. Most of the particle size data obtained in 1985 indicate median aerodynamic diameters in the range of 2 to 10 µm, although the distributions are not necessarily lognormal (see Appendix F). It has been suggested, because there were no substantial changes in plant processes, that those data are representative of the historic size distributions of aerosols released from the stacks studied. Composites of distributions have been produced for UF_4 and U_3O_8 and other available data have been utilized for some processes for which no measurements at the FMPC are available (Appendix E). Ten particular particle sizes are used to represent an effluent particle size distribution in calculations. Each of these is chosen to represent 10% of the particles in the distribution. The variability in measured size distributions of FMPC produced UF_4 and U_3O_8 (Appendix E) suggest that it is appropriate to consider a range of values from 0.8 d_{ri} to 1.2 d_{ri}, where d_{ri} is one of the representative particle diameters used in calculations. A uniform distribution of diameters of that range was assumed.

Stack Flow Rate. During the time between evaluations of the stack flow rate, the actual value (F_s) might be higher or lower than the current standard (F_s^*). Data from recent stack traverse data sheets were evaluated to determine the likely magnitude of such variability. There were many stacks whose normal flow rate had been measured two or more times between 1984 and 1989 (RAC 1991). Examination of these data led to the choice of a triangular distribution for F_s with bounds of 0.5 F_s^* and 1.5 F_s^* and a mode of F_s^*.

Sampler Flow Rate. Similarly, the actual flow through the sampling probe (F_p) could vary from the standard value (F_p^*) and could, at any time, be higher or lower than the standard value of F_p^*. However, the effect of filter loading would increase the probability that the sampling flow would be less than the desired value. In the absence of any definitive information to describe this variable, a triangular distribution with bounds of 0.5 F_p^* and 1.25 F_p^* and a mode of F_p^* was chosen.

LOSSES OF PARTICLES IN THE SAMPLING LINE

Introduction

There are indications that collection of particles in sampling systems was a problem at the FMPC. In the monthly reports of effluent releases, there are references to plugged sampling lines. These comments do not define the reason(s) for the plugging; some may have been due to unusual operating conditions. For example, a high moisture content in the sampled air could lead to condensation in cold weather or to particle agglomeration and greatly increase the rate of accumulation of material in the sampling line. There may have been other factors, such as high concentrations of reactive gases, that could create similar effects.

In the evaluation of the FMPC monitoring program by Oak Ridge Associated Universities (ORAU 1985), it was noted that "All probes inspected were partially clogged with material . . . ." While the observed buildup may be due in part to impaction on the relatively blunt ends of the probes (the report noted that the probes were not knife-edged), some of it was surely due to deposition. In the fall of 1985, a complete inspection of all sampling systems was undertaken. The exhaust stack flow rates were measured and the sampling probes were removed for cleaning. Some of the probes were replaced (NLCO 1985–1986).

Another complete refurbishment of the sampling probes appears to have been completed in 1988-1989. Probes were disassembled, checked, repaired, and cleared of deposited material. There were numerous indications of internal contamination of the sampling probes, lines, and the cones above the filter holders. In some cases the probe decontamination task required hot soapy steam or hydrochloric acid and was accomplished in the decontamination facility (WMCO 1988–1989).

There are two primary mechanisms that can lead to losses of particles in the sampling line itself. These are deposition on the walls of the vertical and horizontal sections of the line and impaction of particles on the walls due to the presence of bends in the line. A competing process which may mitigate these effects is resuspension of material deposited on the walls of the sampling line. Treatments of the problem of sampling line losses generally do not account for resuspension. Fluid velocities in sampling lines are often below those at which significant reentrainment of deposited material has been shown to occur (Corn 1965; Sehmel 1970).

In the next section, the sampling system is described. In sections that follow, the methodology that was developed to analyze deposition and impaction losses of particles from the sampled air stream between the stack and the filter is presented. Each of those sections contains results of some generic calculations and discussion of them. Distributions that reflect the variability of the important parameters are presented.

FMPC Particle Sampling System

Detailed descriptions of individual dust collector exhaust sampling systems are not available. Many of the process exhaust systems have been modified or removed and the sampling systems have been removed as well. Other systems are presently inaccessible. The sampling lines employed for exhaust stacks were relatively simple (Ross and Boback 1971). Figure G–1 is illustrative of a typical sampling arrangement for a vertical stack. The probe, constructed of copper tubing, faced into the exhaust air stream. Outside the stack, the small diameter copper sampling line was enclosed in a protective and supportive pipe. There were two rounded 90° bends in the sampling line between the probe and the collection filter.

Measurements of the lengths of sampling lines remaining in Plants 4 and 5 in June 1991 showed that for most systems the horizontal section was 10–12 cm long. However, three systems had horizontal sections with lengths of about 30 cm because there were obstacles near the exhaust duct and a shorter line was not feasible. Outside the duct, the vertical line sections were about 14 cm long at which point the line enters an 8-cm tall conic expansion leading to the filter holder. The base of the conic section is about 8 cm in diameter and attaches to the filter holder. The pleated collection filters were about 10 cm in diameter.

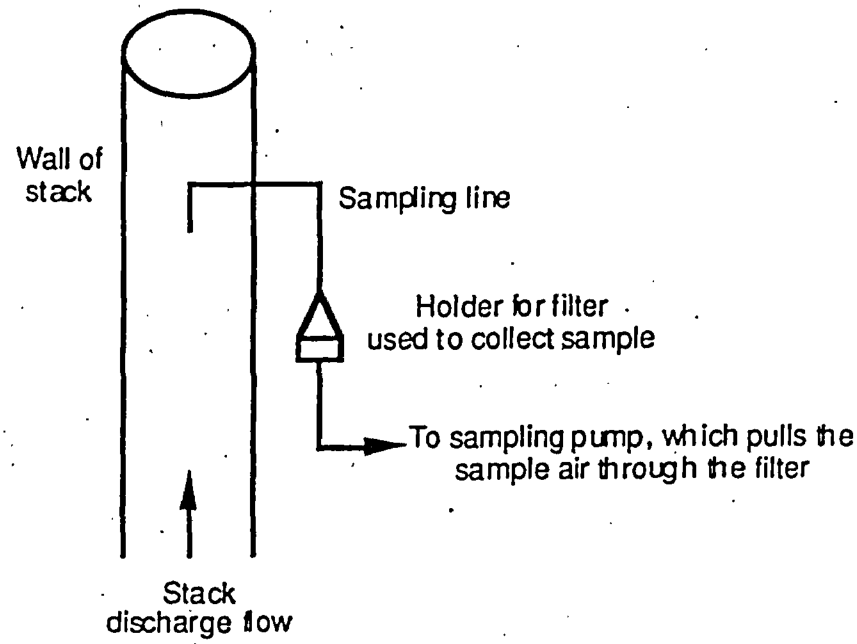

Figure G-1. Schematic diagram of dust collector stack sampling system. The diagram does not show the support piping that was located outside the stack or the rain caps that were atop dust collector stacks.

These dimensions are in general agreement with a drawing prepared at the time the first samplers were being installed (Boone 1956a). That drawing shows that the vertical portion of the line inside the duct was 10 cm in length and that the sampling line internal diameter was small, 0.62 cm. The sample extraction point was the center of the exhaust duct, so the horizontal length of tubing inside the duct varied as a function of the size of the duct being sampled.

Estimates of Particle Deposition

The topic addressed first in this section is the nature of the flow in the sampling system. The typical linear velocity ($\upsilon = 1500\ \text{cm s}^{-1}$), sampling probe opening ($\delta = 0.62$ cm), air density ($\rho_a = 1.2 \times 10^{-3}\ \text{g cm}^{-3}$), and air viscosity ($\eta = 1.81 \times 10^{-4}$ dyne s cm^{-2}) used in the previous section were also employed here. For these parameter values, the Reynolds number (Re) for the sampling line would be:

$$\mathrm{Re} = (\upsilon\ \delta \rho_a) / \eta = \sim 6200 \qquad \text{(G-7)}$$

Flow in such a line would be turbulent. Transport of particles to the wall by turbulent diffusion would be much more important than transport by Brownian diffusion.

Gravitational settling would also be of limited importance because of the brief transport time through the horizontal section of the line.

Vincent (1989) has summarized results from a number of studies of deposition in lines under turbulent conditions. He defines the "penetration" to be the ratio of the particle concentration at the outlet of the line to that present at the line inlet. This ratio has also been called the sampling line "transmission factor" for the particles and that terminology will be employed here. This presentation follows the treatment given by Vincent; however, there are other expositions of some of the same ideas, including Schwendiman and Postma (1961), the ANSI guide (ANSI 1969), and other references cited below.

The relevant equation for the transmission factor that reflects losses due to deposition (TF_D) is:

$$TF_D = \exp[-4\,(w / v)\,(L / \delta)] \tag{G–8}$$

where w is the deposition velocity (cm s^{-1}) appropriate for the specific particles in the sampling line, L is the length (cm) of the sampling line, and v and δ have been previously defined.

Using a figure from Liu and Agarwal (1974), Vincent illustrates that satisfactory agreement among three theoretical approaches and experimental data can be shown by plotting the normalized deposition velocity (w^*) against a normalized relaxation time (τ^*) that reflects particle size. The dimensionless normalized parameters are:

$$w^* = w / [(f / 2)^{0.5}\, v] \tag{G–9}$$

and

$$\tau^* = [\,\tau \rho_a\,(f / 2)\, v^2\,] / \eta \tag{G–10}$$

where τ is the relaxation time and f is the Fanning friction factor. The relaxation time is defined by

$$\tau = [\,\rho d^2 C_c\,] / [\,18\,\eta\,] \tag{G–11}$$

For Reynolds numbers less than 10^5, Perry et al. (1984) give the following expression for the friction factor:

$$f = 0.0791\,\mathrm{Re}^{-0.25} \tag{G–12}$$

A good approximation to the best theoretical relationships between w^* and τ^* can be obtained using power functions. For these calculations, two power functions were used. For values of τ^* between 0.1 and 10:

$$w^* = (5.40 \times 10^{-4})\, \tau^{*(1.974)} \tag{G–13}$$

For values of τ^* between 10 and 300:

$$w^* = (2.45 \times 10^{-2})\ \tau^{*(0.3178)} \tag{G–14}$$

The normalized deposition velocity is approximately constant (0.15) for values of τ^* that are greater than 300. Use of these functional relationships allowed a closer correspondence to the experimental data when estimating w^* and then w for particles of various uranium compounds.

The deposition velocities derived from these relationships were used to compute transmission factors for a 1-m sampling line having an inside diameter of 0.62 cm, with a fluid velocity of 1500 cm s^{-1}. Results of the calculations are shown in Table G–4 as a function of the physical diameter of the particles. The calculated losses due to deposition of particles begin to be substantial for physical diameters of 2 µm or greater.

The estimated deposition losses calculated as described above are higher than those given in the ANSI Guide (ANSI 1969). Back calculation of deposition velocities from the results presented in Table B3 of (ANSI 1969) indicates that substantially lower values of w were used in those calculations. The deposition velocities used in the ANSI guide appear to be based on a correlation developed and later published by Sehmel (1970). The correlation relates the normalized deposition velocity to particle density, the ratio of the particle diameter to the tube diameter, and the Reynolds number. The dependence on Re is much stronger than in the correlation described above.

Table G–4. Estimated Transmission Factors for Particles of Pure Uranium Compounds, 1-meter Sampling Line

Physical diameter (µm)	Calculated transmission factors for a 1-m line			
	UF_4 particles	UO_3 particles	U_3O_8 particles	UO_2 particles
0.1	1.0	1.0	1.0	1.0
0.2	1.0	1.0	1.0	1.0
0.3	1.0	1.0	1.0	1.0
0.5	1.0	0.99	0.99	0.99
0.8	0.99	0.97	0.96	0.94
1	0.97	0.93	0.92	0.86
2	0.68	0.40	0.31	0.13
3	0.16	0.087	0.079	0.063
5	0.054	0.023	0.020	0.014
8	0.020	0.011	0.009	0.006
10	0.011	0.006	0.005	0.003
20	0.002	0.002	0.002	0.002

Sehmel's experimental results for deposition velocity are among the smaller values that have been measured (Gieseke et al. 1980) and are uniformly lower than predicted values from Friedlander and Johnstone (1957). The Friedlander and Johnstone theoretical values and the predictions of Beal (1970) and Liu and Ilori (1974) are reasonably representative of the set of rather variable data (Gieseke et al. 1980, Vincent 1989). Theoretical estimates developed by Davies (1965) follow the same general trend of increasing w^* with τ^*, but the specific values of w^* are much lower than experimental results and the predictions of the three other theoretical analyses.

At least some of the variability in the data is due to differences in measurement goals. Some measurements were made to determine the flux of particles to the surface and efforts were made to assure that particles transported to the wall of the line did not reenter the air stream. Other measurements made with liquid drop aerosols would similarly be free of the effects of particle bounce at impact or subsequent resuspension. The former appears to be the most important mechanism for flows with Reynolds numbers considered in this context. Resuspension of previous deposits apparently requires Re ~ 5 x 10^4 or greater (Corn 1965; Sehmel 1970).

To address the fact that not all particles striking the wall of the sampling line will stick to it, an attachment fraction for deposition (a_d) is introduced. For the purpose of estimating line losses, an effective deposition velocity is defined as the product [$a_d\ w$] and is used in Eq. (G–8). Marshall et al. (1982) compared experimental measurements against predicted sampling line losses for particle diameters of 5–20 µm and found that the latter values consistently exceeded those that were measured. Beal (1978) also compared computed deposition velocities with measured ones to assess the probability that a particle would stay on the wall. However, these comparisons may only illustrate bias in the calculational models.

Direct evidence for incomplete attachment of particles on tubing walls is provided by a figure presented by Sehmel (1967). It shows mean deposition velocities to tacky surfaces for particles with aerodynamic diameters between 2 and 30 µm to be factors of 5 to 20 higher than those to untreated surfaces. The data were for Re = 36,000 in a large diameter line, which initially clouded their utility for the small diameter and lower Reynolds numbers characteristic of sampling lines at the FMPC. However, the analysis by Beal (1978) of data generated by Sehmel (1966, 1968) suggests a method that can be used. He found the dimensionless stopping distance (S^+) to be a useful parameter for developing correlations.

For particles in the probe and sampling line, the stopping distance (S, cm s^{-1}) is defined as

$$S = 0.05\, u\, d\, \rho\, (f/2)^{0.5} / \eta + 0.5\, d \qquad \text{(G–15)}$$

in which the parameters all have the meanings that were defined above. The dimensionless stopping distance (S^+)

$$S^+ = S\, u\, (f/2)^{0.5}\, \rho_a / \eta \qquad \text{(G–16)}$$

is used to relate the attachment fraction to the particle size and flow parameters.

Direct comparisons between measurements of deposition onto tacky tubing walls and deposition onto untreated walls form the basis for the estimation of the attachment fraction. In this analysis the tacky surface is regarded as a perfect sink; that is, it is assumed that the applied film holds all the particles that reach the wall of the line. Estimates of the attachment fraction and dimensionless stopping distance for the measurement conditions were computed from Sehmel's data. There is substantial variability in these estimates, which are plotted in Figure G–2. Also shown in the figure are the central estimates of the attachment fraction, described by the following functions

$$a_d = c \quad \text{for } S^+ \leq 1.5$$
$$a_d = (0.1/S^+)^e f \text{ or } S^+ > 1.5 \qquad \text{(G–17)}$$

Central estimates of c and e are 0.20 and 0.59, respectively. The experimental deposition measurements upon which these estimates are based were made using vertical tubes. In our analysis of sampling line losses, Eq. (G–17) was used for both horizontal and vertical tube sections. In the Monte Carlo calculations (discussed below), estimates of a_d are made by sampling distributions of these two parameters. Figure G–2 shows the ranges of values of a_d computed for three values of S^+.

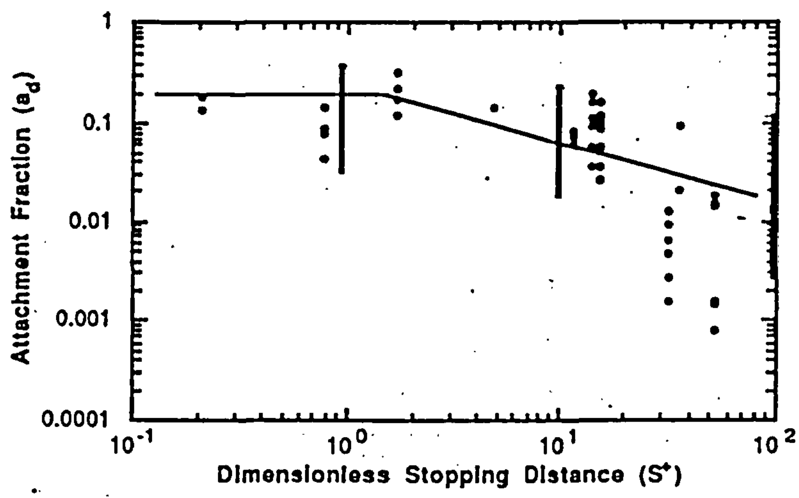

Figure G-2. Dependence of the attachment fraction upon the dimensionless stopping distance. Points are observed values from experimental comparisons of tacky and untreated surfaces. The thin lines show the functional form of the relationship given by Eq. (G–17). The vertical lines show the ranges of computed attachment fractions considering the uncertainties in c and e.

Estimates of Particle Impaction in Bends

Also contributing to the loss of material between the point of sample extraction and the collection filter would have been impaction of large particles in the two 90° bends in the sampling line. Vincent (1989) presents an analysis that indicates that the probability of particle impaction on the wall of the line (ε_i) is a complicated function of the Stokes number, but independent of the radius of curvature of the bend in the line. Other investigators have developed relationships in which the impaction probability is a linear or slightly curvilinear function of the Stokes number (Yeh 1976, Crane and Evans 1977). Cheng and Wang (1981) consider the radius of curvature of the bend (R_b) using Dean's number (De), which is defined by

$$De = Re / (2 R_b / \delta) \qquad \text{(G–18)}$$

where, as before, Re is the Reynolds number for the flow in the sampling line and δ is the internal diameter of the line. In this formulation, higher flow rates and tighter bends lead to greater impaction probabilities. For a particular value of De, the impaction probability is a function of the Stokes number and thus reflects both the physical diameter and the density of the particles. For Reynolds numbers in the range 100–1000, the shape of the impaction probability curve is sigmoid. For higher Reynolds numbers it approaches the curvilinear form for idealized flow (Cheng and Wang 1981). The curve for idealized flow was defined for 90° bends of differing radii of curvature by Cheng and Wang (1975).

Experimental confirmation of their approach has been reported for the conditions De = 400 and Re = 1000 (Pui et al. 1987). However, the experimental data for De = 35 and Re = 100 do not agree with the theoretical values of Cheng and Wang. For fully turbulent flow, the theory also failed to match the measurement results (Pui et al. 1987).

The latter case is of great interest for the current problem. Measurements were made with lines having internal diameters of 0.50 and 0.85 cm at Re of 6,000 and 10,000. No dependence on Re or d can be seen in the results. The data fit the function

$$\varepsilon_i = 1 - 10^{-0.963\,\mathrm{Stk}} \qquad \text{(G–19)}$$

very closely at nearly all values of Stk studied, from 0.03 to 1.35.

A set of estimates of the impaction probabilities is presented in Table G–5 for selected Stokes numbers. The functional relationship given above can be used to obtain impaction probabilities for other values of Stk in the experimental range. The transmission factor that reflects losses due to impaction of particles is designated TF_I. For a sampling line having n 90° bends

$$TF_I = [\,1 - \varepsilon_i\,]^n \qquad \text{(G–20)}$$

Table G–5. Estimated Impaction Probability for a Single 90° Bend as a Function of Stokes Number

Stokes number (Stk)	Impaction probability (ε_i)	Stokes number (Stk)	Impaction probability (ε_i)
0.03	0.064	0.7	0.79
0.05	0.10	0.8	0.83
0.1	0.20	0.9	0.86
0.2	0.36	1.0	0.89
0.3	0.49	1.1	0.91
0.4	0.59	1.2	0.93
0.5	0.67	1.3	0.94
0.6	0.74	1.35	0.95

The results in Table G–5 are for $n = 1$. Table G–6 contains estimated transmission factors for particles of various uranium compounds for passage through two 90° bends. These estimates are based on the data of Pui et al. (1987). The calculations indicate that losses due to impaction begin to be substantial for particle diameters of 2 μm and increase sharply with increasing particle size. The effect of particle density can be seen by reading across a row for particle diameters greater than 0.3 μm.

Table G–6. Estimated Transmission of Particles of Pure Uranium Compounds Through Two 90° Bends

Physical diameter (μm)	Estimated Transmission Through Two 90° Bends			
	UF_4 particles	UO_3 particles	U_3O_8 particles	UO_2 particles
0.1	1.0	0.99	0.99	0.99
0.2	0.99	0.98	0.98	0.97
0.3	0.98	0.97	0.96	0.95
0.5	0.95	0.92	0.91	0.89
0.8	0.89	0.83	0.81	0.76
1	0.84	0.76	0.73	0.66
2	0.51	0.36	0.31	0.21
3	0.23	0.10	0.075	0.033
5	0.019	0.002	0.001	< 0.001
8	< 0.001	< 0.001	< 0.001	< 0.001
10	< 0.001	< 0.001	< 0.001	< 0.001

The impaction probabilities and transmission estimates in Tables G–5 and G–6 are based upon measurements of uranine traced liquid aerosols. The deposition in the bend was physically removed and analyzed. Solid particles of uranium compounds will not attach to the tubing walls as effectively as the oleic acid aerosol used in the experiments. An effective impaction parameter was defined as the product $[a_i \varepsilon_i]$ and used in Eq. (G–20) to estimate impaction losses. Because measurements that could be used to derive attachment fractions specific for impaction are unavailable, relationships comparable to those in Eq. (G–17) were used to define a_i

$$a_i = c \quad \text{for } S^+ \leq 1.5$$
$$a_i = (0.1/S^+)^e \quad \text{for } S^+ > 1.5 \qquad \text{(G–21)}$$

The nominal values of c and e are 0.20 and 0.59. Parameter distributions that were sampled as part of the calculation of a_i are discussed below.

Estimates of Overall Transmission Factors

Overall particle transmission will reflect losses due to both deposition and impaction. For a given size and density, the overall transmission factor for a particle (TF_P) is the product

$$TF_P = TF_D \times TF_I \qquad \text{(G–22)}$$

where TF_D and TF_I are the transmission factors for deposition and impaction, respectively.

It was noted earlier that the measured deposition velocities for particles in sampling lines exhibit variability, both within and among sets of experimental results. There is a smaller uncertainty associated with the measured impaction probabilities. In both cases, the possibility that retention on the wall of particles that are transported to it may be different for uranium aerosols produced by plant processes than for those generated for laboratory experiments is a concern.

To address the uncertainties associated with deposition and impaction of the uranium aerosols in FMPC sampling lines, distributions of important parameters were developed for use in the Monte Carlo calculation procedure for the particle transmission factors. The important parameters and the chosen distributions are discussed below.

Particle Density. The density of the released particles is primarily determined by the chemical form of the uranium and the degree of purity of the discharged material. The same right triangular distribution with bounds of 0.9 ρ^* and ρ^* defined previously was used for the particle transmission calculations.

Particle Size Distribution. As noted earlier, the particle size distributions are treated in calculations using a representative particle diameter (d_{ri}) for each tenth of the particles in the distribution. As for the evaluation of previous deviations from isokinetic sampling conditions, a uniform distribution between 0.8 d_{ri} and 1.2 d_{ri} was used for each of the representative diameters to reflect the uncertainty in the particle size distribution.

Fluid Velocity in Sampling Line. Once the diameter of the sampling line is fixed (and no evidence has been found that the early sampling lines were constructed of anything but the small diameter copper tubing previously described) the fluid velocity depends only on the sampler flow rate. The distribution given previously, namely triangular with a mode of F_p^* and bounds of 0.5 F_p^* and 1.25 F_p^*, was also used to compute u for the particle transmission calculations.

Sampling Line Configuration. The basic physical layout of the sampling lines appears to have been highly uniform. As noted the line diameter and material do not appear to have changed and all lines that have been inspected or seen in photographs had two 90° bends. For the particle impaction calculations, n was always equal to two.

The lengths of the sampling lines varied. However, for a particular exhaust duct the only uncertainty was associated with the length of the horizontal section outside the stack. The inner and outer vertical segments were taken to be 10 and 22 cm in length, respectively, and the length of the horizontal section inside the duct was half the duct diameter. These dimensions are based on physical observation of the sampling systems and the drawing (Boone 1956a) that describes them. The length of the exterior horizontal section was represented by a right triangular distribution with a minimum value and mode of 10 cm and a maximum value of 30 cm. The total length of each stack sampling line was computed using the known dimensions and the randomly selected sample of the one variable segment length.

Effective Deposition Velocity. The aforementioned variations in particle density, diameter, and velocity were used in the calculation of the normalized relaxation time τ^* and the dimensionless stopping distance S^+. The relationships between τ^* and w^* and between w and w^* are given in Eqs. (G–13, –14) and in Eq. (G–9), respectively. The attachment fraction was estimated using Eq. (G–20). For $S^+ \leq 1.5$, the value of $a_d = c$ was randomly selected from a triangular distribution with bounds of 0.03 and 0.38 and a mode of 0.20. For larger values of S^+, the distribution of the exponent (e), whose nominal value is 0.59, was taken to be uniform with bounds of 0.32 and 0.86. This procedure yields a distribution of values of a_d that is comparable to estimates derived from experimental measurements. Then the effective deposition velocity, $[a_d\ w]$, was used in place of w in Eq. (G–8).

Effective Impaction Efficiency. The particle density, diameter, and velocity derived from the distributions described above were used in the calculation of the Stokes number. The value of Stk is the fundamental determinant of the impaction efficiency ε_i. The attachment parameter for impaction (a_i) was computed and the quantity $[a_i\ \varepsilon_i]$ was used in place of ε_i in Eq. (G–17) to compute TF_I. Estimates of a_i were made using Eq. (G–21). The distribution of c was assumed to be triangular with a mode of 0.20 and bounds of 0.03 and 0.38. A uniform distribution was sampled for values of e; its bounds were 0.32 and 0.86.

OVERALL SAMPLING BIAS

For a particular sampling system, the estimate of overall sampling bias was the product of three factors: B, R, and TF_P. The appropriate operating conditions and inlet particle size distribution, either measured or inferred, for that line were used in a single Monte Carlo calculation to make estimates of the overall sampling bias and its uncertainty. Estimates of the bias (B) due to nonrepresentative sampling were taken from Table G–1. It was assumed that a symmetric uniform distribution applied and that B was equally likely to overestimate or underestimate the true concentration. The bias due to anisokinetic sampling (R), computed using Eq. (G–1), could also be either positive or negative depending upon the randomly selected values of duct and probe air velocities. The calculations just described led ultimately to a value of TF_P computed using Eq. (G–22). In the Monte Carlo procedure, the overall bias, $B\ R\ TF_P$, was calculated repeatedly to obtain a distribution of estimates.

Examples of the results of calculations of overall bias are shown below for selected exhaust ducts. Figure G–3 shows the distribution of estimates for the G4–3 exhaust in Plant 4. The distribution is approximately lognormal with a median value of 0.92 and a geometric standard deviation (GSD) of about 1.5. The 5th and 95th percentile values are 0.49 and 1.6, respectively. These results indicate that there is less than a 5% chance that releases from this stack were underestimated by more than a factor of two and an equal probability that they were overestimated by more than a factor of 1.6.

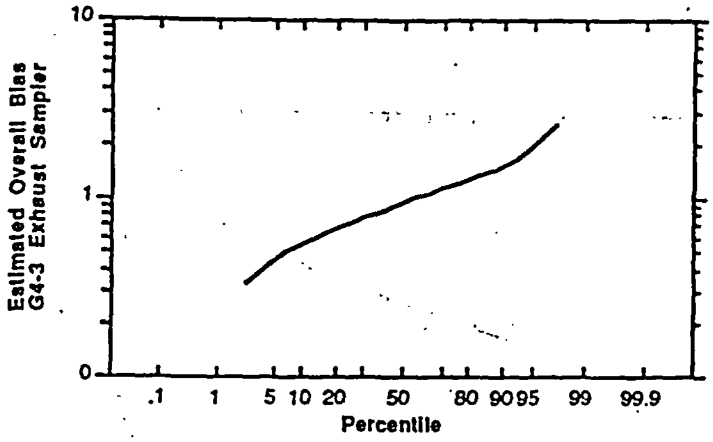

Figure G–3. Distribution of estimates of overall bias for the sampler serving the G4–3 exhaust stack in Plant 4.

An analysis was performed to determine the sensitivity of the estimates of bias to the parameters used in the calculation. Four parameters ranked much higher than any of the others: v, B, u, and e. The relatively large uncertainties in these variables were discussed above. For G4–3, the estimated range for B is ±30%. There is, unfortunately, no simple way to reduce these uncertainties, which primarily relate to unknown conditions of past operations.

Figure G–4 shows the distribution of estimates of overall bias for the G4–2509 exhaust in Plant 7. The distribution is approximately lognormal with a median of 0.88 and a GSD of about 1.4. The 90% confidence interval ranges from 0.52 to 1.4. The sensitivity analysis revealed the importance of the same four parameters, but in different order: u, v, e, and B. For G4–2509, the estimated range for B is smaller, ±15%.

Figure G–5 shows the distribution of estimates of overall bias for the G5–260 exhaust in Plant 5. This distribution is also approximately lognormal with a median of 0.85 and a GSD of about 1.5. The 90% confidence interval lies between 0.52 and 1.4. The sensitivity analysis revealed the importance of the same four parameters, but in a third sequence: e, u, v, and B. For G5–260, the estimated range for B is ±20%.

CONCLUSIONS

Three possible sources of bias in dust collector effluent sampling have been investigated. They are nonrepresentative sampling, anisokinetic sampling conditions, and losses of particles from the sampled air stream due to deposition and impaction. All could affect the reported releases from the dust collector exhaust stacks at the FMPC. Nonrepresentative sampling and anisokinetic sampling could produce either a high or low bias in the reported results, depending upon inhomogeneities in the concentration and

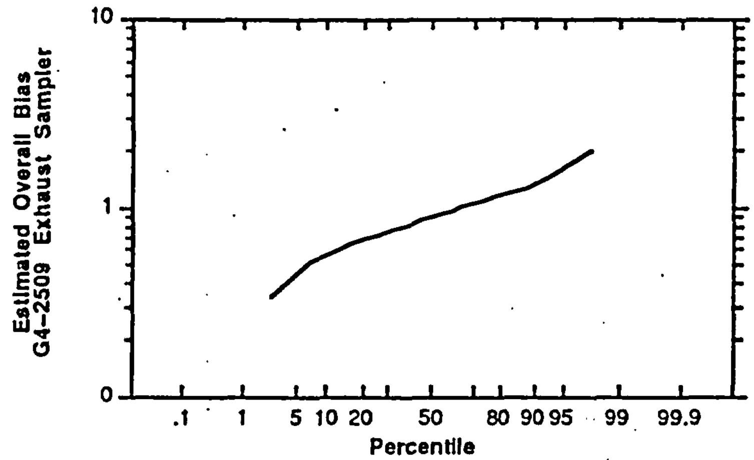

Figure G-4. Distribution of estimates of overall bias for the sampler serving the G4-2509 exhaust stack in Plant 7.

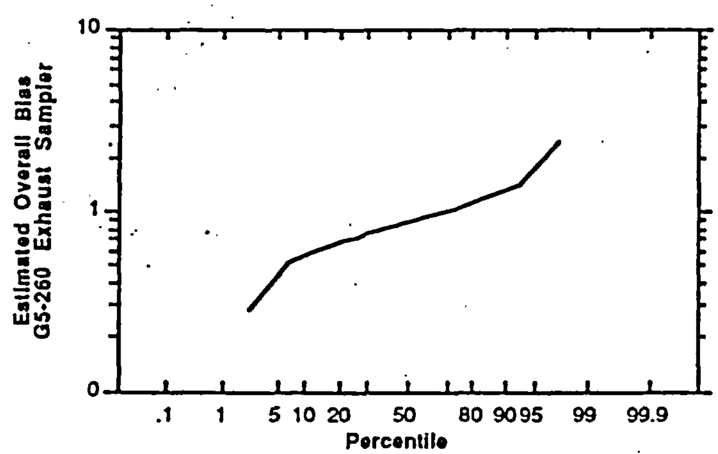

Figure G-5. Distribution of estimates of overall bias for the sampler serving the G5-260 exhaust stack in Plant 5.

possible changes in flow rates in the duct and sampling line. The effect of losses due to deposition and impaction is one-sided, always leading to an underestimation of the amounts released.

The overall bias reflects all three factors and is dependent upon a number of variables. Distributions of the parameters required for bias estimation were developed. These distributions are used in Monte Carlo calculations of the sampling biases for FMPC sampling systems. The confidence bounds for these estimates of overall bias indicate that releases may have been underestimated by as much as a factor of two or overestimated by as much as a factor of 1.6. The results of these calculations are employed in Appendix E.

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APPENDIX H

DISCHARGES FROM PLANT 2/3 DENITRATION OPERATIONS

INTRODUCTION

After 1956, exhausts from the denitration operations in Plant 2/3 were treated by a wet scrubber prior to discharge. The releases of uranium from the scrubber exhausts were not sampled, even periodically, until recently. In June 1988, an investigation of environmental radioactivity measurements led to the conclusion that releases from Plant 2/3 processing activities were the source of the observed higher offsite air concentrations (Investigation Board 1988). In sections that follow the scrubber exhaust system is described and the previous estimates of releases are discussed. Results of a review of the denitration operations are then presented. The approach to estimation of releases from the denitration operations is described and the results of its implementation are presented. Because information is lacking on early operations with dust collectors, releases from those years are estimated using the same model used for years when the scrubbers were in operation.

SYSTEM DESCRIPTION

Two processes are of primary interest in describing the Plant 2/3 denitration scrubber system. The first of these is the denitration process itself. As the name implies nitrates were removed from the uranyl nitrate hexahydrate (UNH) to produce uranium trioxide (UO_3, also called orange oxide). Fumes of oxides of nitrogen that were produced during denitration were routed to the scrubber system. Absorption of these gases in aqueous solution produced nitric acid that was recycled to the digestion area of the plant. The second process was the vacuum transfer of the orange oxide from the denitration pots to a storage hopper. This process was called "gulping" the orange oxide. That term, derived from the fact that the snakelike tool appeared to swallow the UO_3, is employed in subsequent discussion.

There were two parallel lines of denitration pots, located along the north and south sides of the building. Each of the pot lines had a storage hopper, a product mill, and a product packaging station. The suction of the vacuum system pulled the product out of the pot and carried it through two sequential cyclone separators that removed most of the product from the stream. The exhaust of the second cyclone, which contained the UO_3 that had not been removed by the separators, was routed to the scrubber. Uranium captured by the scrubber was routed to the digestion area when the scrub liquor collection tank was pumped to the digestion area (Cahalane 1957, Hicks 1957, Semones and Sverdrup 1988).

Wet scrubbers are a class of devices that have long been used to remove particles or mists from air streams. Detailed technical descriptions of scrubbers can be found in the literature (Perry, Green and Maloney 1984; ECT 1978). The scrubbers for the UO_3 vacuum transfer or gulping process employed a nitric acid solution. First contact of the scrub solution with the airstream occurs in the throat of the venturi scrubber under highly turbulent conditions. The liquid droplets remove particles from the airstream by impingement. After the impingement section, the air stream containing liquid droplets and the remaining free particles enters the separator section. This is typically a chamber containing baffles or other surfaces upon which the large droplets are deposited by impaction. The air leaving the separator contains small droplets of scrub liquor and the free particles. In the Plant 2/3 scrubber system, there was a wire mesh demister in the stack that was used to remove more droplets and particles from the exhaust air. Limited data on both the particle fraction and the entrained liquor fraction of the release are available. The nature of the releases from the Plant 2/3 scrubber system is discussed in more detail in a later section.

PREVIOUS RELEASE ESTIMATES

Estimates of historic releases from the Plant 2/3 denitration system scrubbers were added to the list of FMPC source terms in 1989 (Clark et al. 1989). Those estimates rely on the analysis of Semones and Sverdrup (1988) who investigated the scrubber emission source. Their report includes results of the only known effluent sampling results for the Plant 2/3 scrubber system exhausts. The experimental measurements of releases of uranium during scrubber operation were related to the scrub liquor uranium concentration during the test operations. Release estimates were obtained for mist entrainment when UO_3 was not being transferred and for particulate losses during gulping. The estimated release factor was used to make estimates of the releases from the scrubber systems over the years of operation of the system.

In addition to the release factor developed from the effluent testing effort, other parameters were required to estimate releases. A nominal processing cycle was developed; the time required for pot gulping was taken to be one hour. The number of pots gulped in a year was estimated using the annual production of UO_3 and a pot capacity of 843 kg of uranium. The frequency of pumping of the scrub liquor to the digestion area affects the average concentration of uranium in the scrub liquor and the discharge of uranium entrained in mist during scrubber operations. A mean concentration of 41 g U L^{-1} of scrub liquor, partly based on measurements between 1985 and 1988, was used in calculations of releases for earlier years.

An operational history for the denitration process was estimated. For the years 1953–1964, it was assumed that the process operated during 3 shifts per day, 50 weeks per year. The scrubber was assumed to operate during 90% of the available hours during 1960, the year of maximum UO_3 production. Scrubber operation was prorated according to the total annual UO_3 production for other years.

These assumptions led to estimates of Plant 2/3 scrubber releases that were presented by Clark et al. (1989). Over the entire period of operation, inclusion of the Plant 2/3 scrubber source term estimates raised the earlier total uranium release estimate by about 28% (Clark et al. 1989).

URANIUM TRIOXIDE PRODUCTION

Uranium trioxide production data have been summarized on a fiscal year basis in Appendix C. Short term production data were generally not available, except for the very detailed data in Shift Foreman's logs that were found for some years (see below). Average values for calendar years were estimated approximately using the tabulation in Appendix C. Figure H-1 shows the average annual uranium trioxide production amounts for each year of operation. The plot shows that production in metric tons of uranium (MTU) varied markedly from year to year. The years of maximum production were 1959–1961, when about 12,000 MTU of UO_3 was produced. The period of greatest activity was soon followed by a shutdown, which began in July 1962 and continued for two years. Two other secondary production peaks occurred in later years.

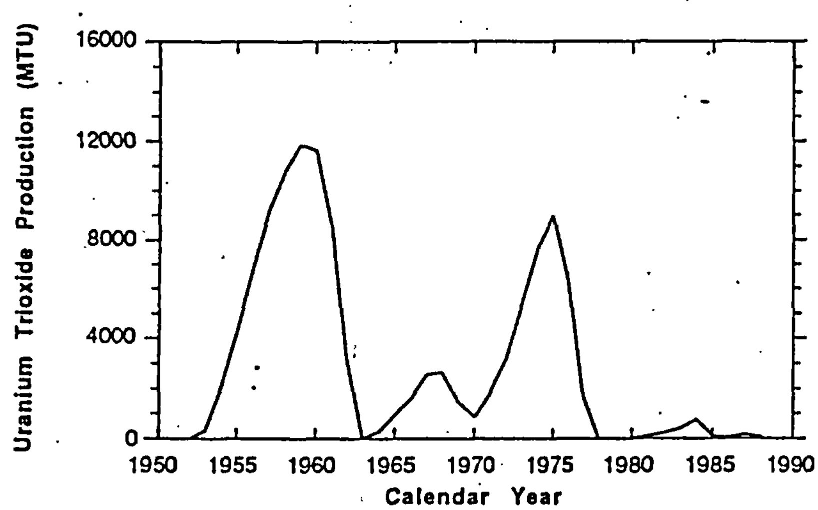

Figure H-1. Production of uranium trioxide (UO_3) in Plant 2/3.

REVIEW OF DENITRATION OPERATIONS

Because the analysis of Semones and Sverdrup (1988) indicated that the Plant 2/3 scrubber releases accounted for about 30 percent of the total releases to the atmosphere from the FMPC, a review of the basis for those estimates was undertaken. The review of denitration operations and the release estimates was conducted in several steps. Each part of the review focused upon a particular set of the relevant technical reports or historic records that had been found. The following subsections treat particular aspects of the denitration process and the releases that resulted from its operation. Some large collections of original records were found that greatly assisted the investigation.

Review of Logbooks from 1969, 1970, and 1973

The initial review of records focused on logbooks from the denitration area of Plant 2/3 that covered operations during the years 1969, 1970, and 1973. This review indicated that actual plant operating data differed from parameter values that had been assumed in the analysis of Semones and Sverdrup (1988).

Plant logbook data for 5 May through 26 September 1973 showed that the denitration process operated nearly continuously (NLCO 1973). This contrasted with the operating schedule of five days per week that had been assumed by Semones and Sverdrup (1988) for the years from FY 1965 through FY 1982.

Production data were available on a shift by shift basis in the logbook, which permitted evaluation of a question about the relative releases at different times of the day. During the night (12–8), day (8–4), and afternoon (4–12) shifts, the plant denitrated 898, 816, and 799 pots of UNH, respectively. The amounts of UO_3 packaged were 989, 917, and 996 MTU for the three shifts, respectively. The processing and packaging rates did not vary greatly during this time period. The production data indicate that nocturnal releases from the Plant 2/3 scrubbers would not have exceeded those during the day shift by more than about 10%.

During May–September 1973, 2513 pots of UNH were denitrated and 2902 MTU were packaged as UO_3 (NLCO 1973). The gross average amount of product per pot was 1.15 MTU, as opposed to the assumed quantity of 843 kg U. Calculation of the amount of product per pot on a daily basis and averaging of the daily values yielded an estimate (± sample standard deviation) of 1.17 ± 0.15 MTU packaged per pot denitrated.

The time required to transfer the UO_3 in a pot using the gulping system is another variable employed in the analysis of releases. Two sets of plant logbook entries yielded information about this parameter. During May–August 1969, 2% enriched UNH was being denitrated to UO_3 and packaged in Plant 2/3. From data for 27 production batches, the mean time (± standard deviation) to transfer a pot of UO_3 was 81 ± 22 minutes (NLCO 1969). In 1970, 34 batches of enriched UNH were denitrated during a 2-month period. Transfer times were recorded for transfers of 28 pots of UO_3 (NLCO 1970). The mean UO_3 transfer time for a pot of product was 93 ± 18 minutes. Both sets of observations led to transfer times longer than the 60-minute duration assumed in Semones and Sverdrup (1988). It is possible that the transfer times were longer because of extra care taken with the more valuable enriched uranium. Overall production was low during the periods cited and that may have been an influence as well. The recorded heating times for 19 batches of UNH lead to an average of 9.3 ± 0.4 hours, compared to the "standard" estimate of 6.0 hours given by Semones and Sverdrup (1988). This suggests a more leisurely pace of operations during the enriched uranium operations. Transfer times were not recorded during the period of higher production in 1973 cited above.

Limited data on scrub liquor uranium content and acidity were also found during the review of plant logbooks. When scrub liquor was pumped to the digestion area, the uranium concentration and acidity were reduced by an average of 29% (NLCO 1970). This is less than the 50% reduction assumed by Semones and Sverdrup (1988). This observation suggests that there may have been a consistent low bias (of 15–20%) in the estimation of the component of releases that is due to carryover of scrub liquor during operation.

Review of logbook data showed that production of uranium trioxide was not constant throughout the year. For example, between 9 September 1970 and 5 January 1971, there was no denitration of UNH (NLCO 1970). In FY 1973, 25% of the total production occurred in two months (NLCO 1973). These data indicated that if detailed time resolution is needed for dose estimates the generally available annual production rates could be misleading. Annual production data do not indicate periods of variable production or of plant outages.

As a result of the initial review, it was determined that further investigation was needed to determine appropriate parameters for the calculations of release rates from the Plant 2/3 scrubbers. This entailed review of additional detailed data on denitration operations in Plant 2/3.

Review of Operational Data for 1960–1962

Historic records on the denitration of UNH in Plant 2/3 during 1960–1962 were located in storage. The Shift Foremen's Logs that were kept during those years (NLCO 1960–1962) contained information on the number of pots charged with UNH, the number of processed pots of UO_3 that were gulped, and the number of drums or hoppers of milled product that were packaged during each shift. The log sheets also contained some information on the amount of scrub liquor pumped to the digestion area and on the normality of the liquid transferred. Although data on the uranium concentration in the scrub liquor at the time of transfer had been found in the logbooks for later years, little information on that parameter was found on the log sheets for 1960–1962. Although sample log sheets were used to record information on uranium concentrations in scrub liquor, no substantial collection of these forms has been located. It is known that many original analytical laboratory data sheets, another possible source of such data, have been incinerated.

Data from the Shift Foremen's Logs were compiled and analyzed. Daily production and packaging rates have been computed from the values recorded for each shift. The number of pots gulped and the reported production of orange oxide were used to estimate the quantity packaged per pot gulped. For the three years 1960, 1961, and 1962, the average production amounts per pot were 1.11, 1.06, and 1.14 MTU. These values are somewhat lower than the value (1.17 MTU) found for 1973.

The day to day variability in the amount of material transported by the gulping system was also examined. Plant 2/3 UO_3 production was full time five days a week but much lower on the weekends. There was generally more production on Saturday than on Sunday and there were outages for holidays. These data differ from those found for later years (see above).

The daily shift log sheets (NLCO 1960–1962) also provided some information on the amount of time the scrubber systems operated. In some cases there were notations in the log that the scrubbers were shut down or started up during a particular shift. During shifts when there were no UO_3 gulping activities recorded in the log, the scrubbers may have been off; however, without a notation on the log sheet it is uncertain. The fraction of the time that the other scrubbers may have been shutdown, termed the maximum scrubber outage fraction, was computed on a monthly basis using the denitration log sheet data. This fraction generally increased with time while the production rate declined. It was estimated that the actual outage time was about 75% of the maximum value derived from the logsheets. Using this assumption, the average scrubber outage fraction for the 3-year period was 0.10, which is consistent with operator estimates for periods of high production (Semones and Sverdrup 1988).

The relationship between the production rate and the scrubber outage fraction was evaluated for production rates between 13 and 37 MTU d^{-1} during the 30 months of operation in 1960–1962. In Figure H–2, the estimated scrubber outage fraction (f_o) is plotted as a function of the average daily production. The best fit line through the data points (lower line in figure) has an intercept of 0.287 ± 0.035 and a slope of -0.00586 ± 0.00123 (d MTU^{-1}). The correlation is not extremely strong ($r^2 = 0.45$).

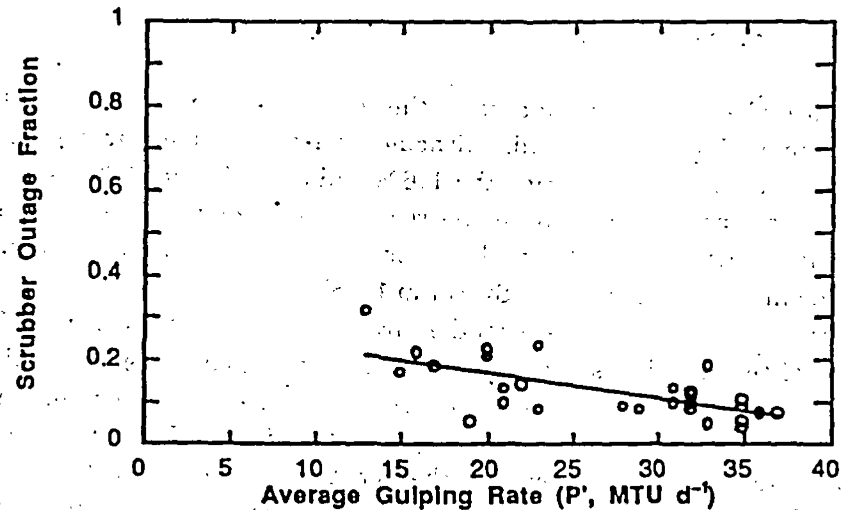

Figure H-2. Observed dependence of the scrubber outage fraction (f_o) on the average amount of UO_3 gulped (P', MTU d^{-1}). The points are monthly average values from 1960–1962, with the best-fit line shown. The equation of the line is $f_o = 0.29 - 0.0059\,P'$.

The scrubbers did not operate during periods when the plant was shut down ($f_o = 1$). Estimation of the outage fraction for production rates between zero and those shown in Figure H–2 is discussed in a later section.

Review of Operational Data for 1956–1959 and 1967

Nearly complete sets of Operator's Shift Logs for the years 1959 (NLCO 1959) and 1967 (NLCO 1967) were found in the archives. These were reviewed to ascertain if additional information on the duration of the gulping process for a pot of orange oxide. Data for 1959 were of particular interest because only natural uranium was processed during that year. Two pot lines and the associated scrubbers were operating in both years. The times at which gulping and charging were completed were recorded, but, unfortunately, times for the start of gulping were not. The logsheets do support heating times that are longer than the value of 6 hours that had been previously assumed by Semones and Sverdrup (1988).

Partial records from the denitration area of Plant 2/3 were found for the years 1956, 1957, and 1958. These logsheets contained information on the numbers of pots gulped and hoppers of product filled, but no information on the times required to perform operations. Notations on the logsheets indicated that denitration times were sometimes lengthened by lowering the heat input to the pots.

The logs indicated the collection of samples, but no analytical results were included. Some data on the concentrations of uranium in the UNH charge to the pots. However, like the heating time, these data are not critical to the calculations of releases from the Plant 2/3 scrubbers.

Review of Effluent Measurements

No evidence of early measurements of the discharge of uranium from the Plant 2/3 scrubber exhausts has been found. Although numerous measurements of scrubber efficiency were made in Plant 8 during the 1960s and again in the 1980s (see Appendix I) it appears that no comparable measurements were made in Plant 2/3. At this time, the only known effluent monitoring results available for the Plant 2/3 scrubber exhausts are those that were made in 1988 (Semones and Sverdrup 1988).

Two operational conditions were studied by Semones and Sverdrup (1988). The first was operation of the scrubber system alone without operation of the UO_3 gulping system. Because the scrubber system was also used to remove oxides of nitrogen and convert them to nitric acid that was recycled, the system operated while the UNH was being heated to produce UO_3. Uranium was present in the scrub liquor at these times and could be discharged to the environment as the result of entrainment of mist from the scrubber with the exhaust airstream. The second condition studied was operation of the UO_3 vacuum transfer system in concert with the scrubber system. During gulping operations, penetration of UO_3 particles through the scrubber would also contribute to the quantity released. The total release would then be the sum of the two components. Only a limited number of tests were conducted to measure the reentrainment and particle penetration releases in Plant 2/3. Results of these few measurements are discussed below.

The results of two measurements during scrubber operations alone are shown in the upper part of Table H–1. Three measurements were made of the discharge rate when the scrubbers were operating alone, but isokinetic sampling conditions were not achieved during the first run. The estimated release rate for that test, which was anomalously low, was not included in average release rate used by Semones and Sverdrup (1988) and is not considered here either. The normalized entrainment release rate [Q_e, (kg U h^{-1}) per (kg U L^{-1})] was obtained by dividing the uranium release rate by the uranium concentration in the scrub liquor. The average of the two estimates of Q_e (± the standard deviation) of the two estimates is 3.40 ± 0.38 (kg U h^{-1}) per (kg U L^{-1}).

Also shown in Table H–1 are the results of measurements of the uranium release rate when both the gulping system and the scrubber system were operating. Again, one of the three measurements was deemed invalid by Semones and Sverdrup (1988) and two results are available. The net normalized releases from UO_3 gulping (last column) are the differences between the gross values and the average normalized release rate for operation of the scrubber alone. The products of these normalized release rates and the corresponding scrub liquor concentrations yield two estimates of the net particle release due to operation of the UO_3 gulping system (Q_g, kg U h^{-1}). These estimates are shown in the lower portion of the first column. The mean (± standard deviation) of the two estimates of Q_g is 0.130 ± 0.026 kg U h^{-1}.

Table H–1. Results of Monitoring Uranium Discharges from the Plant 2/3 Scrubber System

Operating mode	Release rate (kg U h^{-1})	Scrub liquor concentration (g U L^{-1})	Normalized release rate (kg U h^{-1}) per (kg U L^{-1})	
Scrubber only	0.150	48.0	3.13	
	0.176	48.0	3.67	
		Average	3.40	
			Gross	Net due to gulping
UO_3 gulping with scrubber	0.245	38.8	6.31	2.91
	0.209	18.1	11.55	8.15
UO_3 Gulping Net Values	0.113			
	0.148			
Average	0.130			

Measurement uncertainties were not presented in the original report (Semones and Sverdrup 1988). If the 1-σ uncertainties associated with the release rate and scrub liquor concentration measurements were about 10% of the values, then the propagated uncertainties in the normalized release from entrainment of scrub liquor and the release rate from gulping alone would be comparable to the variability of the two estimates of each of those quantities.

Review of Scrub Liquor Concentration Data

Although the concentration of uranium in scrub liquor was measured routinely, most of the records of this information appear to have been lost. Records of scrub liquor concentration measurements were found for parts of five years of operation: 1970, 1982, 1983, 1985, and 1987. Data from the periods of highest production would obviously be preferred, but no records for those periods have been located. Table H–2 summarizes the scrub liquor concentration data, which are included in the annex to this appendix.

Table H-2. Summary of Available Scrub Liquor Concentration Measurements

	Individual Measurements of Scrub Liquor Concentration (g U L^{-1})					
	1970	1982	1983	1985	1987	Composite
Mean	34	52	134	47	63	68
Median	35	43	139	45	59	57
Range	15–47	<1–134	34–242	28–72	4–124	<1–242
	(12)[a]	(66)[a]	(34)[a]	(32)[a]	(89)[a]	(221)[a]
	Set of Averages of Scrub Liquor Concentration (g U L^{-1})					
		37 (33)[a]	134 (34)[a]	47 (32)[a]	53 (30)[a]	
		66 (33)[a]			75 (30)[a]	
					59 (29)[a]	

[a] The number of measurements of scrub liquor concentrations for each year or subset of data is shown in parentheses.

Based upon the measurements that were found, the average concentration of uranium in scrub liquor was estimated to be 0.068 kg U L^{-1}. The median of the 221 values was 0.057 kg U L^{-1}. As the table shows, there was substantial variation of values within each of the years. This is to be expected because of the nature of the process. Some of the concentrations were very low, <5 kg U L^{-1}, indicating that the tank was completely drained at some times.

The desired distribution of long term average concentrations is not available. To approximate that distribution, sets of concentration measurements, shown in the lower part of Table H–2, were assembled. There were 34 and 32 measurements in 1983 and 1985, respectively. The sequential measurements in 1982 and 1987 were divided into groups of comparable size (29-33 measurements) and averaged. The seven average values were used to estimate the distribution of the average scrub liquor concentration, shown in Figure H–3. Because there were only 12 measurements in 1970, that average was not used in constructing this distribution. We believe that the breadth of the distribution is sufficient to reflect the uncertainty in our knowledge of the average scrub liquor concentration and is satisfactory for estimating annual releases from the Plant 2/3 scrubbers. Sampling from this distribution yields a mean concentration of 0.067 kg U L^{-1}, comparable to the true mean, and a median value of 0.057 kg U L^{-1}, the same as the median measured concentration.

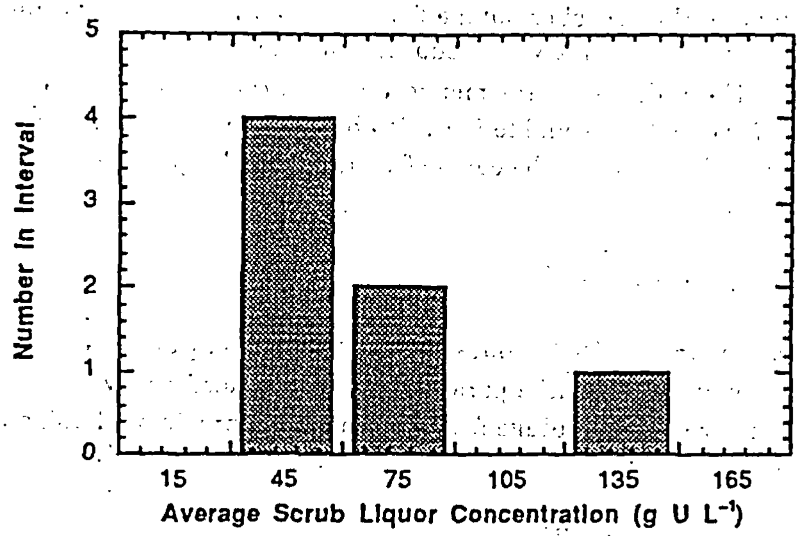

Figure H–3. Approximated distribution of average scrub liquor concentrations (g U L^{-1}). The distribution reflects the set of average concentrations given in the lower part of Table H–2.

ESTIMATES OF RELEASES

The methods used to estimate releases from the scrubbers are described below. The distributions used to characterize individual parameters are presented and discussed in the second subsection. The last subsection contains the release estimates for the Plant 2/3 scrubbers.

Calculational Methods

The release of uranium from the Plant 2/3 scrubbers is composed of releases due to scrub liquor entrainment and those due to particles of UO_3 in the airstream that pass through the scrubber. Releases due to entrainment of scrub liquor (Q_s, kg U) were computed using Eq. (H–1).

$$Q_s = n\,(1 - f_o)\,N_h\,C_s\,Q_e \qquad \text{(H–1)}$$

in which:

- n is the number of scrubbers operating
- f_o is the scrubber outage fraction, which depends upon the production rate
- N_h is the number of hours in the period
- C_s is the average concentration of uranium in the scrub liquor
- Q_e is the entrainment release per unit scrub liquor concentration (kg U h^{-1}) per kg U L^{-1}).

For the years after 1977 when the plant was operating, there was normally one scrubber running; thus, $n = 1$ for those years. For other years of operation, the calculations use $n = 2$.

Before calculating the scrubber outage fraction, the average gulping rate is computed. Let P be the amount of UO_3 that was produced and transferred using the gulping system during the year. The units of P are metric tons of uranium (MTU). Then the average gulping rate, P' (MTU d^{-1}), is equal to P divided by 365, the number of days in the year.

The equation used to compute the outage fraction depends upon the magnitude of P'. For values of $P' \geq 15$ MTU d^{-1}:

$$f_o = a_1 - m_1 P' \tag{H–2}$$

From the best fit line in Fig. H–2, values of the parameters a_1 and m_1 are 0.29 and 0.0059, respectively. Distributions for these parameters are described below. It was assumed that for lower production rates, the outage fraction could be approximated using a straight line. Thus, for values of $P' \leq 15$ MTU d^{-1}:

$$f_o = a_2 - m_2 P' \tag{H–3}$$

was used with $a_2 = 1$ (no scrubber operation when the plant was shut down) and $m_2 = 0.053$. Distributions for these parameters are described below. For $P' = 15$ MTU d^{-1}, the two equations yield the same value, 0.20, within the uncertainty caused by rounding.

Figure H–4 illustrates the range of application of these two equations. Eq. (H–2), which is based upon the data plotted in Figure H–2, is used to estimate f_o for the higher average production rates. The assumed linear decline in f_o as P' increases from zero to 15 MTU d^{-1} is shown by the dashed line.

Because the calculations are performed on an annual basis, the number of hours in the period (N_h) is 8760. The average scrub liquor concentration (C_s) and the entrainment release factor (Q_e) used in Eq. (H–1) are obtained by sampling from their distributions, which are described below.

The release due to particle escape during gulping operations (Q_g, kg U) depended upon the duration of those operations and on the release factor that reflects particle penetration only (Q_g, kg U h^{-1}). That release was estimated using Eq. (H–4).

$$Q_\gamma = N_p \, T_g \, Q_g \tag{H–4}$$

in which N_p is the number of pots of UO_3 that were gulped during the period and T_g is the time (h) required to transfer all the UO_3 from a pot to the storage hopper.

The number of pots gulped is computed from the production, P (MTU) and the parameter k, which is the amount of uranium trioxide per pot. The relationship is $N_p = P/k$. The gulping time and the release fraction during gulping were obtained from their distributions, discussed below.

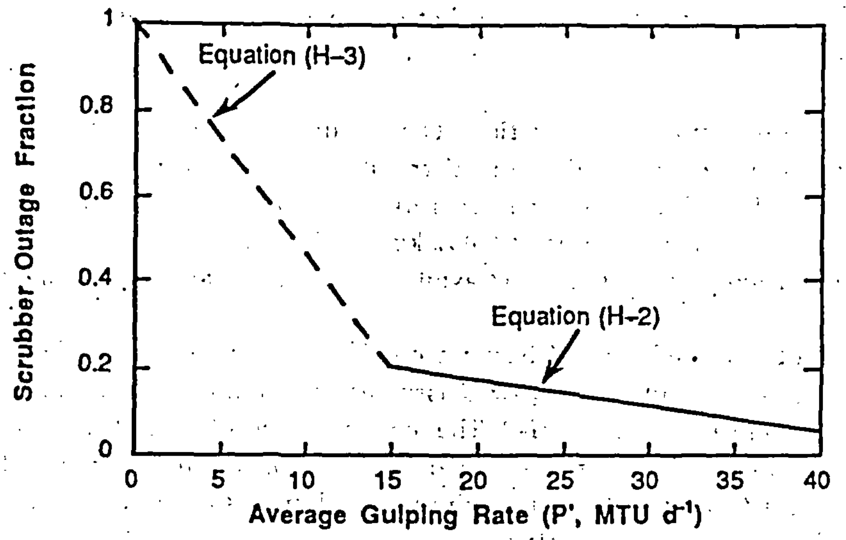

Figure H–4. Approximated distribution of average scrub liquor concentrations (g U L^{-1}). The distribution reflects the set of average concentrations given in the lower part of Table H–2.

The total uranium release ($Q_e + Q_\gamma$) for each year of denitration operations was estimated by performing Monte Carlo calculations using Crystal Ball® (DI 1991). The program sampled each of the parameter distributions and repeatedly performed the calculations just described to produce a set of estimates of the release during each year. Totals for each decade were computed as part of the same set of calculations. The distributions of parameters used in the calculations are described below.

Parameter Distributions

The distributions that were used for parameters in the calculations described above are presented below in the order of appearance of the parameters in the discussion. As noted, specific integer values are used for the number of scrubbers in operation, the number of hours in the year, and the outage fraction when there was no production.

Scrubber Outage Fraction Calculations. For the relatively high production and gulping rates, $P' \geq 15$ MTU d^{-1}, the calculation of f_o using Eq. (H–2) requires two parameters, a_1 and m_1. The intercept for the best fit line (a_1) was represented by a triangular distribution with a mode of 0.29 and bounds of 0.22 and 0.36. The slope of the best-fit line (m_1) was also represented by a triangular distribution; the mode was 0.0059 (d MTU^{-1}) and the bounds were 0.0084 and 0.0033 (d MTU^{-1}).

For gulping rates lower than 15 MTU d^{-1}, Eq. (H–3) requires a different intercept and slope, a_2 and m_2, respectively. The value of a_2 was taken to be one; that is, when there was no production the scrubbers did not operate. The slope of the estimation line (m_2) was represented by a triangular distribution with a mode of 0.053 (d MTU^{-1}), between bounds of 0.045 and 0.061 (d MTU^{-1}). This distribution produces an uncertainty range for a gulping rate of 15 MTU d^{-1}, that is very close to that obtained using the best fit line for the same gulping rate.

Average Scrub Liquor Concentration. The annual average concentration of uranium in scrub liquor was approximated by the distribution of short term average measurements shown in Figure H–4. Sampling from this distribution yields a mean concentration of 0.067 kg U L^{-1} and a median value of 0.057 kg U L^{-1}, both of which are comparable to the corresponding parameters for the set of available concentration measurements.

Entrainment Release Factor. There were only two measurements of the entrainment release factor. The distribution for Q_e was assumed to be uniform with a mean equal to the average of the two results in Table H–1. The mean value was 3.4 (kg U h^{-1}) per (kg U L^{-1}). A standard deviation of 0.6 (kg U h^{-1}) per (kg U L^{-1}), which is 50% greater than that computed from the two results, was used in the calculations to reflect the limited amount of information about this release fraction.

Amount of UO_3 in a Pot. The distribution of k, the amount of UO_3 per pot, was taken to be uniform in shape with boundaries of 1.06 and 1.17 MTU per pot. The bounds of the distribution were determined by the maximum and minimum of the four estimates of the ratio derived from Plant 2/3 logsheets and logbooks. The mean of the four values was 1.12 MTU per pot, which is consistent with the distribution selected.

Gulping Time. In the review of denitration production data, about fifty recorded values of the gulping time per pot were recorded. The mode was in the interval 70–80 minutes. The minimum time was 50 minutes. The maximum time was 130 minutes; however times in excess of 100 minutes are inconsistent with most UO_3 production rates. For these calculations T_g was described by a triangular distribution with a mode of 75 minutes and a range of 50 to 100 minutes. Because the data upon which the distribution is based were recorded during processing of enriched uranium, it is possible that they may overestimate T_g appropriate to other periods. However, the distribution is based upon the best available information.

Gulping Release Factor. Only two measurements of the release rate during UO_3 gulping have been made (Table H–1). The resulting estimates of the mean and standard deviation of the underlying distribution of release rates are uncertain. A uniform distribution was chosen to represent the release factor. The mean of the distribution was taken to be the mean of the two measured values, 0.13 kg U h^{-1} (Table H–1). The standard deviation was taken to be 0.04 kg U h^{-1} . which is 50% greater than the value computed from the two estimates, to reflect our lack of knowledge of the actual distribution.

Results of Plant 2/3 Scrubber Release Calculations

Estimates of Plant 2/3 scrubber releases obtained from the Monte Carlo calculations described above are presented in Table H–3. These estimates have been rounded to two significant figures. The best estimate for each year is given in column two of the table and is the median of the distribution of estimates for that year. Calculated releases for the first years of operation with dust collectors, prior to scrubber installation, have the added uncertainty that the present calculations are only surrogates. The highest releases are estimated to have occurred during the 5-year period between 1957 and 1961. Annual releases were nearly as high again in 1974–1976.

The median lies in the center of the distribution; half of the estimates were higher than the median and half were lower. The 5th, 25th, 75th, and 95th percentile values for each distribution are also given in Table H–3. These values, which have also been rounded to two significant figures, show the spread of the distributions and the range of release estimates for each year. There is a 50 percent chance that the release lies between the 25th and 75th percentile values and only a 10 percent chance that it lies outside the range defined by the 5th and 95th percentile values.

Cumulative probability distributions of release estimates for three years are presented as examples in Figure H–5. The vertical dotted line marks the 50th percentile or median values that are the best estimated given in Table H–3. The central parts of the three distributions are approximately straight lines. If the distributions of releases were lognormal, the plots of cumulative probability would be true straight lines. The tails of the distributions deviate more from the central slope of the lines, indicating deviations of the distributions from lognormality. The slopes of all three lines are comparable, which implies that the uncertainties are about the same for these years, and this is true for other years as well.

The median release estimates for each year between 1952 and 1988 are plotted in Figure H–6. As noted, the largest releases are estimated to have occurred during the late 1950s and early 1960s, with another period of substantial releases in the mid-1970s. The plot of annual release estimates is similar to that for annual production amounts (Fig. H–1).

Table H–4 contains summary release estimates by decade and for the entire period from 1952 to 1988. The estimates for each decade, also rounded to two significant figures, were obtained by summation as part of the Monte Carlo calculations of the annual releases. The distribution of estimates for the period 1952–1988 is also the result of Monte Carlo calculations using the distributions of releases for each of the four decades. These estimates do not correspond to simple arithmetic sums of medians or particular percentile values. The shapes of the distributions of the sums for each decade and (especially) for the whole period approach the normal distribution. The table shows that releases during the first three decades were comparable. Releases for 1978 and later years were relatively small and the total for the 1980s is much smaller than for the other decades.

Table H-3. Annual Release Estimates for Plant 2/3 Scrubbers

Year	Best estimate (kg U)	Other percentiles in distribution of release estimates (kg U)			
		5th percentile	25th percentile	75th percentile	95th percentile
1953	200	120	160	270	460
1954	1,200	750	990	1,600	2,800
1955	2,700	1,700	2,200	3,700	6,000
1956	3,700	2,300	3,000	5,000	8,100
1957	4,200	2,600	3,500	5,500	8,800
1958	4,600	2,800	3,800	5,800	9,400
1959	4,800	3,000	3,900	6,100	9,700
1960	4,800	3,000	3,900	6,100	9,600
1961	4,300	2,600	3,500	5,600	9,100
1962	1,800	1,100	1,500	2,300	3,900
1963	0[a]	a	a	a	a
1964	170	100	140	230	390
1965	590	370	480	800	1,300
1966	1,000	610	800	1,300	2,300
1967	1,600	960	1,300	2,100	3,600
1968	1,700	1000	1,300	2,200	3,800
1969	930	550	730	1,200	2,000
1970	540	320	420	710	1,200
1971	1,100	670	890	1,500	2,600
1972	2,000	1,200	1,600	2,600	4,400
1973	3,300	2,000	2,700	4,400	7,400
1974	3,900	2,400	3,200	5,100	8,300
1975	4,200	2,600	3,400	5,500	8,700
1976	3,700	2,300	2,900	4,900	8,000
1977	1100	620	830	1,400	2,300
1978–9	0[a]	a	a	a	a
1980	10	6	8	13	20
1981	51	30	41	65	100
1982	81	51	66	100	160
1983	160	98	130	200	310
1984	470	280	380	620	1,100
1985	43	26	35	55	85
1986	17	10	14	22	33
1987	60	38	49	75	120
1988	29	18	24	37	58

[a] There was no production of uranium trioxide during these years.

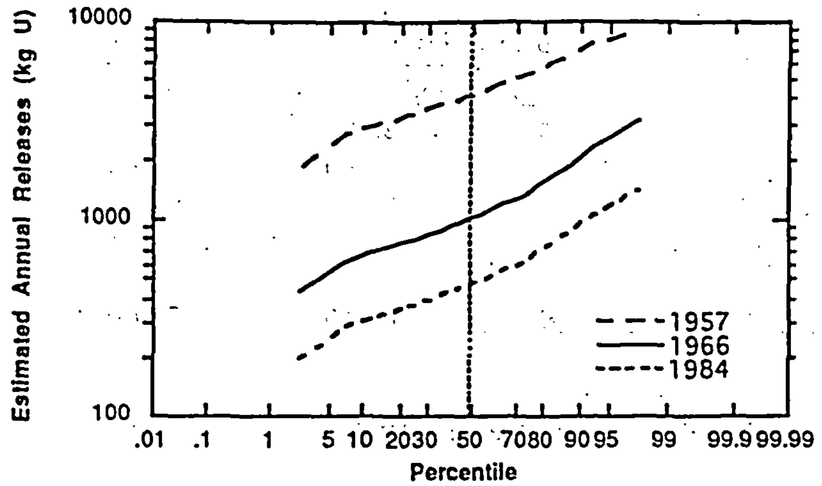

Figure H–5. Cumulative probability distributions for estimated releases from Plant 2/3 scrubbers during the years 1957, 1966, and 1984.

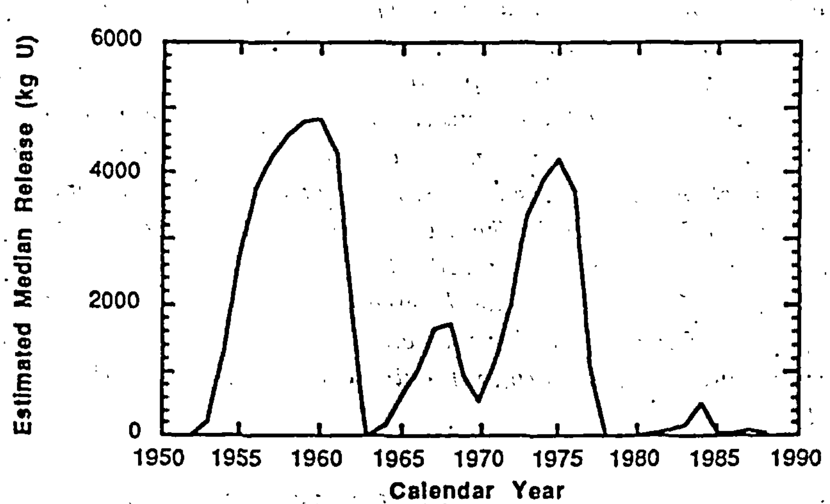

Figure H–6. Median estimates of annual releases from the Plant 2/3 uranium trioxide gulping operations.

Table H-4. Summary Release Estimates for Plant 2/3 Scrubbers

Period	Best estimate of release (kg U)	Other percentiles in distributions of release estimates (kg U)			
		5th percentile	25th percentile	75th percentile	95th percentile
1950s	24,000	18,000	21,000	26,000	32,000
1960s	19,000	14,000	17,000	21,000	25,000
1970s	22,000	17,000	20,000	25,000	29,000
1980s	980	730	850	1,100	1,600
1952–1988	66,000	56,000	62,000	71,000	78,000

ALTERNATIVE CALCULATION OF THE OUTAGE FRACTION

Alternative outage fraction calculations were performed to evaluate the effect of changing the range of applicability of Eq. (H–2). As shown in Figure H–4, for the results reported above, it was applied for $P' > 15$ MTU d^{-1}.

- *To test the effect of extrapolating the best-fit line to lower values of P', alternative calculations were performed. For these calculations, it was assumed that Eq. (H–2) applied for $P' > 8$ MTU d^{-1}. The alternative calculations did not require changes in the distributions of parameters a_1, m_1, or a_2; however, it was necessary to revise the slope (m_2) in Eq. (H–3) to fit its new domain of $0 < P' < 8$ MTU d^{-1}. A triangular distribution, with a mode of 0.095 d MTU^{-1} and bounds of 0.083 and 0.11 d MTU^{-1}, was used for the parameter m_2 in the alternative calculations.*

  In general, the alternative approach produced somewhat higher release rates because some of the estimated outage fractions were lower and the corresponding entrainment release estimates (Eq. (H–1)) were higher. The greatest change of a median estimate for a decade was from 19,000 to 24,000 kg U for the 1960s. This was due primarily to differences in the estimates for 1962, 1967, and 1968. For those years, the production rates were near 8 MTU d^{-1} and the differences in computed values of f_o were largest. All of the alternative median estimates lie within the bounds of the distributions of the original estimates presented in Tables H–3 and H–4.

PHYSICAL CHARACTERISTICS OF THE RELEASED URANIUM

There are two principal components to the release of material from the Plant 2/3 scrubbers to the atmosphere. The first is the particles of UO_3 that penetrate the system. The calculations of scrubber releases described above indicate that this component accounts for about one-fourth of the total U release from the scrubbers. In years of low production, it is estimated that the fraction was as low as 0.2. For years of high production, the corresponding fraction of the U release is estimated to be 0.3.

There are no reported measurements of the particle size distribution of the effluents from the Plant 2/3 scrubbers. The geometric mean physical diameter of the UO_3 aggregates transferred in the Plant 2/3 gulping operation was reported to be 22 µm (Semones and Sverdrup 1989). The particle size of material entering the scrubber would be small relative to the UO_3 product because of passage through the two cyclone separators upstream of the scrubber system. To estimate penetration of particles through the two cyclones that were upstream of the scrubber, it was assumed that the geometric standard deviation of the product particle size distribution was 3. Using data on cyclone performance from Lund (1971), it was estimated that the two cyclones should have removed about 90% of particles having a diameter of 5 µm and greater percentages of larger particle size fractions. Based upon the efficiencies reported for particle removal by venturi scrubbers (Lund 1971), about 90% of the UO_3 particles that passed through the scrubbers would have physical diameters less than 2 µm. The diameter of more than 99% of the particles is estimated to be less than 5 µm. The median diameter was estimated, by extrapolating the slope of the censored distribution, to be about 0.5 µm.

The wire screen mist eliminator used in the Plant 2/3 scrubber stacks was estimated to have a peak efficiency of 99% at an exhaust velocity of 3–4 m s^{-1}. Thus nearly all of the small droplets of uranyl nitrate exiting the scrubber would be expected to impact the screen in the mist eliminator. However, the operating velocities were 6–12 m s^{-1}, which would have increased reentrainment of liquid from the mist eliminator and reduced the overall efficiency. It was estimated that an overall efficiency of about 60% was appropriate for the operating conditions that prevailed (Semones and Sverdrup 1988). Nearly all of the release observed when only the scrubber was operating (see Table H–1) would have been due to reentrainment of liquid that had been trapped in the mist eliminator. The release calculations indicate that about three-fourths of the total U release was by this mechanism. The range of that fraction is estimated to be from 0.7 to 0.8 for years of high and low production, respectively.

Reentrained mist droplets are reported to be generally greater than 100 µm in diameter (Black and Strauss 1981). Droplets from the Plant 2/3 scrubber system exhaust contained uranyl nitrate in nitric acid solution. They would have shrunk during downwind plume travel as a result of evaporation from their surfaces. Complete loss of liquid would leave solid uranyl nitrate hexahydrate (UNH) crystals in the plume. Calculations indicate that even the larger reentrained droplets would have dried by the time the plume had traveled a few hundred meters. The rapid crystallization of UNH from the liquid would result in a polycrystalline mass that could break apart during plume travel.

Calculations were made to estimate the size of solid particles that could be produced from the reentrained mist. It was assumed that the reentrained droplets had diameters that ranged from 80 to 180 µm. Table H–2 shows that uranium concentration in the scrub liquor that are known have a median value of 57 g L^{-1}. The geometric standard deviation of the distribution of those measurements is about 1.8. The uranium concentration range within two GSDs of the median is 17–180 g L^{-1}. Using a density of 2.8 g cm^{-3} for UNH, that range of concentrations, and the stated range of droplet diameters, the diameters of solid UNH spherical particles that could be produced was estimated to vary between 29 and 53 μm, with a central value of 41 μm.

CONCLUSIONS

Releases from the scrubbers serving the Plant 2/3 UO_3 gulping system were recognized only recently and were not sampled prior to 1988. Limited measurement data from that time formed the basis of models of effluent release processes. Plant 2/3 Foreman's log sheets and logbooks were found that contained information on parameters important for the calculation of releases due to the gulping operation. Data on scrub liquor uranium concentrations, required to estimate part of the releases, were also recovered for portions of five years of operation.

Independent estimates of releases from the Plant 2/3 scrubber system were performed using models of scrubber penetration by particles and mist reentrainment. Monte Carlo calculations produced distributions of release estimates for each year, each decade, and for the entire period, 1952–1988. Median estimates of releases during three of the four decades of operation were comparable, about 20,000 kg U, while the value for the 1980s was much lower. The median release estimate for the entire period of operation was 66,000 kg U. This estimate was bounded by 5th and 95th percentile values of 56,000 and 78,000 kg U, respectively.

About 25% of the release is estimated to have been small (≤ 5-μm) particles of UO_3 that penetrated through the scrubber. The larger fraction (~ 75%) would have been UNH particles produced by evaporation of entrained droplets of scrub liquor. The approximate size range for these particles is estimated to be between 29 and 53 μm. The physical stability of these large particles during transport in the atmosphere is not known.

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ANNEX TO APPENDIX H — SCRUB LIQUOR DATA TABULATION

Table H1–1 contains the data on uranium concentrations in Plant 2/3 scrub liquor that were found in laboratory data records for 1982. Sampling times were not generally given so the sequence of multiple samples on the same day is not known and should not be presumed from the ordering of the data in the table.

Table H1–2 contains the data on uranium concentrations in Plant 2/3 scrub liquor that were found in laboratory data records for 1983 and 1985. Sampling times were generally not given so the sequence of multiple samples on the same day is not known and should not be presumed from the ordering of the data in the table.

Table H1–3 contains most of the data on uranium concentrations in Plant 2/3 scrub liquor that were found in laboratory data records for 1987. Sampling times were generally not given so the sequence of multiple samples on the same day is not known and should not be presumed from the ordering of the data in the table. The remainder of the data from 1987 and a smaller number of values form 1970 are contained in Table H1–4.

Table H1–1. Concentrations of Uranium in Plant 2/3 Scrub Liquor in 1982

1982 Date	Concentration ($g\ U\ L^{-1}$)	1982 Date	Concentration ($g\ U\ L^{-1}$)
19 June	64.9	22 September	76.8
19 June	50.3	23 September	<1
21 June	46.1	23 September	87.7
22 June	31.9	23 September	99.3
22 June	26.6	24 September	<1
23 June	29.2	24 September	106
23 June	26.6	24 September	111
23 June	24.1	24 September	<1
24 June	31.3	25 September	107
24 June	36.8	25 September	82.1
25 June	34.7	25 September	<1
25 June	39.3	18 October	8
25 June	30	19 October	26.3
26 June	32.1	20 October	58.3
26 June	35	21 October	93.3
26 June	35.1	22 October	125
28 June	23.7	26 October	105
28 June	29.2	27 October	134
29 June	21.6	28 October	46.8
29 June	35.4	29 October	78.9
29 June	37.1	1 November	34.1
30 June	24.3	2 November	66.2
30 June	23.5	4 November	71.2
2 July	21.8	5 November	78.1
2 July	16.9	8 November	79.3
6 July	25.4	9 November	71
6 July	26.2	10 November	106
7 July	28.7	10 November	49.1
15 September	59.7	11 November	46.5
17 September	99.2	12 November	54.6
19 September	104	15 November	67.4
21 September	73.4	16 November	77.7
21 September	<1		
21 September	51.1		

Table H1-2. Concentrations of Uranium in Plant 2/3 Scrub Liquor in 1982 and 1985

1983 Date	Concentration ($g\ U\ L^{-1}$)	1985 Date	Concentration ($g\ U\ L^{-1}$)
1 March	33.8	27 February	31.3
15 March	58.4	27 February	33.3
16 March	67.1	27 February	53.2
17 March	78.4	27 February	41.6
18 March	58.4	28 February	32.6
18 March	81.5	28 February	28.1
21 March	85	28 February	34.4
22 March	90.8	1 March	30.5
23 March	130	1 March	40.4
24 March	157	4 March	44.9
25 March	178	4 March	59.5
28 March	242	4 March	46.8
28 March	178	5 March	63.1
29 March	194	5 March	72.4
29 March	204	6 March	44.9
31 March	129	6 March	45.3
11 April	158	7 March	50.4
14 April	184	7 March	60.3
15 April	177	7 March	44.9
4 May	110	8 March	53.2
4 May	114	11 March	65.2
5 May	150	11 March	36.5
6 May	188	12 March	45.3
9 May	188	12 March	50.5
10 May	197	13 March	43.5
11 May	132	14 March	40.3
12 May	149	15 March	37.3
13 May	192	18 March	49.8
1 June	203	19 March	59.9
3 June	145	20 March	55.1
3 June	133	21 March	52.5
3 June	86.7	22 March	47
6 June	47.8		
7 June	52.2		

Table H1-3. Concentrations of Uranium in Plant 2/3 Scrub Liquor in 1987

1987 Date	Concentration (g U L^{-1})	1987 Date	Concentration (g U L^{-1})
5 May	39	22 May	91.8
7 May	29.3	28 May	90
7 May	22.1	28 May	93.9
11 May	76.3	29 May	88.5
11 May	74.1	29 May	89.8
12 May	124	29 May	96.9
13 May	33.9	1 June	89
13 May	36.1	1 June	98.1
13 May	37.8	1 June	88.1
13 May	41	2 June	114
13 May	40	2 June	115
13 May	14.9	2 June	114
14 May	25.4	2 June	114
14 May	27.9	2 June	117
15 May	40.3	3 June	117
15 May	39.1	6 July	81.2
15 May	38.4	6 July	25
15-May	114	6 July	25.6
15 May	44.4	7 July	38.8
18 May	43.1	7 July	30.5
18 May	42.3	7 July	38.4
19 May	50.1	8 July	56.8
19 May	58.7	8 July	41
19 May	42	8 July	43.3
20 May	56.4	9 July	60.8
20 May	59.2	9 July	51.2
20 May	49.5	9 July	55.3
21 May	81.3	10 July	71.4
22 May	102	10 July	68
22 May	109	10 July	81.2

Table H1–4. Concentrations of Uranium in Plant 2/3 Scrub Liquor in 1987 and 1970

1987 Date	Concentration (g U L^{-1})	1970 Date	Concentration (g U L^{-1})
11 July	67.6	11 August	15
11 July	63.8	13 August	18
11 July	74.8	14 August	24
11 July	3.8	16 August	29
12 July	60.6	17 August	35
12 July	60.5	30 August	34
13 July	67.6	31 August	42
13 July	65.2	2 September	45
13 July	57.6	3 September	43
14 July	71	5 September	47
14 July	66.6	6 September	35
15 July	72.8	9 September	37
15 July	62.4		
15 July	59.2		
16 July	51.4		
16 July	46		
16 July	54.4		
20 July	74		
7 August	29.3		
8 August	31		
8 August	48		
9 August	32		
9 August	52		
9 August	74.4		
10 August	68.6		
13 August	98.4		
13 August	39		
13 August	97		
15 August	62.4		

APPENDIX I

RELEASES FROM PLANT 8 SCRUBBER SYSTEMS

INTRODUCTION

Several of the high temperature and other exhausts from Plant 8 were discharged through scrubber systems. In these systems the exhaust air is cleansed, or scrubbed, by contact with droplets of liquid. This liquid, called the scrub liquor, scavenges reactive gases and particles that are in the airstream. Table I-1 contains a listing of the Plant 8 scrubbers and the process equipment serviced by each of them. The first six scrubbers listed handled hot exhaust gases from the kiln and furnaces. Scrubbers from this group were among the most important sources of uranium releases to the atmosphere from Plant 8 and are the subject of this appendix.

Table I-1. Exhaust Air Scrubbing Systems for Plant 8

Scrubber designation	Equipment number	Scrubber type	Equipment served
Rotary kiln	735-43-9F	Ejector-venturi	Rotary kiln
Oxidation #1	D43-205	Ejector-venturi	Oxidation furnace #1
Caustic (primary calciner)	F43-6	Ejector-venturi altered by NLO	Primary calciner; box, muffle, and Graphite furnaces
UAP furnace	735-43-1F	NLO special design ejector-venturi	UAP furnace
Oxidation #2 (NPR)	D-8N1-1000 or 735-43-8031	Turbulaire-Doyle	Oxidation furnace #2
Green salt reverter		Ejector-venturi	Green salt reverter furnace
Old digester	735-43-16B	Ejector-venturi	Leach tank
New digester	FG-101	Ejector-venturi	Digester
ADU	W8-42	Packed tower	ADU system digester
Leach tank	W8-36	Packed tower	Leach tank

The last four scrubbers in the table treated ventilation air collected above digestion and other process tanks. Packed towers, with counter-current flow of exhaust gases and scrub liquids, are particularly useful for removal of gases from the exhaust stream. Fumes collected above the digesters and leach tanks could be effectively removed by such systems. Packed tower scrubbers can be plugged by dust and are not suitable for exhausts containing high concentrations of particulate material (Danielson 1973, CIV 1980). The much smaller releases from these scrubbers are considered in Appendix K.

DESCRIPTIONS OF PLANT 8 SCRUBBER SYSTEMS

Early FMPC Manufacturing Standards, which contained process and equipment descriptions and drawings, show that furnace discharges were generally routed through a cyclone or knockout drum to remove large particles from the exhaust gas before it entered the scrubber. One system not having this design feature was the Box Furnace, which was serviced by the large caustic scrubber. The scrubbers were installed for two purposes: to neutralize acidic off-gases from the furnaces and to scrub out any entrained solids for recovery of uranium. However, the first purpose, fume scrubbing, was mentioned most prominently in the early descriptions of system operation. Caustic soda solution, with an initial concentration of 10%, was used as the scrub liquor (Calhane 1958a, 1958b, 1962; Harvey, Heareth and Hicks 1958; Hicks 1958a, 1958b, 1958c, 1958d, 1958e, 1958f).

The operation of ejector-venturi scrubbers is described by Perry, Green, and Maloney (1984), who note that these devices are widely used as gas adsorbers. (In some earlier editions of Perry's Handbook, the device is referred to as a water-jet scrubber). The jet of scrub liquor from the spray nozzle provides a draft that draws the air to be cleaned into the scrubber. At the FMPC, the downward facing nozzle and exhaust gas-droplet contact section was described as the obnoxious vapor condenser (OVC). Impaction of droplets and pollutants in the exhaust air results in pollutant removal and collection in the scrub liquor, which entered at one end of the scrubber hotwell. Exhaust fans withdrew the scrubbed air at the opposite end of the hotwell. Although not shown in the generic drawing in Perry, Green, and Maloney (1984), barriers to carryover of scrub liquor droplets were components of most of the scrubber systems of interest.

Table I–2 contains reported feed rates for the furnaces in Plant 8. Three sources of information on furnace capacities have been found and are tabulated. The first is the set of FMPC Manufacturing Standards prepared in 1959 by the Quality Control Group (1959a, 1959b, 1959c, 1959d, 1959e, 1959f, 1959g). These contain rated uranium feed rates for seven pieces of equipment. The only capacity stated for the calciner was for total material, namely 21,600 kg d^{-1}.

The second and third sets of capacity data are taken from the history of residue recovery operations in Plant 8 (Mead 1972) and from a report by Savage (1975) that specifies FMPC equipment capacities. These sets of estimates show variations in capacity depending upon feed type. While of historical interest and included in Mead (1972), no estimates for the graphite furnace or green salt reverter are given in the 1975 data set. The graphite furnace was shut down in 1960 and the reverter only operated between 1956 and 1958 (Mead 1972).

The largest difference among the estimates is the very large rated feed rate given for the UAP Furnace in the 1959 specification, which is about 2.5 times larger than the values given later. Levy's transmittal memo (1975) for the Savage report states that the capacities were based on "actual previous experience" and may better reflect the true capacity of the equipment. Problems with operation of the UAP furnace were attributed in part to overloading it (Noyes 1962). Noyes states that the original capacity of that furnace for ammonium diuranate (ADU) cake was 108 lb U h^{-1} (1170 kg U d^{-1}), but that the throughput in June 1962 was 380 lb U h^{-1} (4100 kg U d^{-1}).

Table I-2. Estimated Production Capacities for Equipment in Plant 8

Equipment description	Rated feed rate (kg U d^{-1})[a]	Feed type[b]	Capacity (kg U d^{-1})[c]	Capacity (kg U d^{-1})[d]
Box furnace	380	B	270	270
Muffle furnace	330	A, B, C	320	320
Graphite furnace	370	A, B	360	e
Calciner (for ADU)	e	C	1620	1620
Rotex screen output	e	B	7660	7660
Hydromet. prep.	e	C	e	5400
UAP furnace	3600–4500	C	1620	1620
Rotary kiln	600	A	1690	1690
	e	B	3380	3380
	e	C	1180	1180
Oxidation furnaces	1150	A	900	900
	e	B	1170	1180
Green salt reverter	630	D	720	e

[a] Rounded estimate from rated capacities (lb U d^{-1}) in FMPC Manufacturing Standards (Quality Control Group 1959a-g); variation with feed type not specified.
[b] A: high grade metallic sludges; B: high grade residues and compounds; C: low grade residues and compounds and "unlimited;" D: scrap UF_4.
[c] Rounded estimate based on equipment capacities (U tons d^{-1}) in Mead (1972).
[d] Rounded estimate based on stated capacities (U tons d^{-1}) in Savage (1975).
[e] No estimate provided.

Processing rates are included in reports of recent stack exhaust measurements (Rakiewicz, Jackson, and Phoenix 1988a–e). Extrapolated to a 24-hour schedule, these correspond to about 270 kg U d^{-1} for the oxidation furnace, 650 kg U d^{-1} for the box furnace, 9800 kg U d^{-1} for the rotary kiln, and a maximum of 8900 kg U d^{-1} for the calciner. The estimated throughput for the oxidation furnace is lower than the values in Table I–2, while the values for the box furnace and rotary kiln are two to three times the tabled values. The calciner throughput was increased consistently as the tests progressed. A report by Adams (1975) indicates an ore concentrate calcining rate of about 12,000 kg d^{-1} for the rotary kiln.

The discharges from the scrubber system exhaust blowers were not sampled on a regular basis. Periodic measurements of discharge concentrations and of scrubber efficiencies were performed by the Industrial Hygiene and Radiation Department. A number of their measurements for the caustic, kiln, UAP, and NPR scrubbers were made during the early 1960s, a period of substantial concern about releases of uranium from these systems. These are discussed later in this appendix.

In the early 1980s, when Plant 8 production was lower, measurements were made to determine emission factors for the Plant 8 scrubber discharges (Ross 1982, 1983). Application of the factors was based on the duration of furnace charging operations (Bardo 1985, 1986). It was assumed that most of the releases occurred during charging. Several source emission tests using EPA Method 5 were conducted for scrubber exhausts just prior to shutdown of the FMPC operations (Rakiewicz, Jackson, and Phoenix 1988a–e). These recent measurements led to the development of new emission factors (Beirne 1988, Bonfer 1988, Hill 1989). Emission factor estimates are also discussed in a later section.

Various scrubber efficiency estimates have been used to estimate releases to the atmosphere from the Plant 8 scrubbers during the early years. The following sections discuss those estimates, plant operational information related to release estimates for these systems, the efficiency measurements, and calculation of release estimates for this report.

PREVIOUS RELEASE ESTIMATES

The calculation of most of the releases from the Plant 8 scrubbers has employed estimates of scrubber efficiency together with measurements of the amounts of uranium that were collected in the scrub liquor. Let M_s be the amount of uranium (kg) found in the scrub liquor at the end of a specified period and let ε be the efficiency of the scrubber during the period. If these two quantities are known, then the release from the scrubber to the atmosphere during the period (Q, kg) can be calculated from mass balance considerations. If I is the amount of airborne uranium (kg) that entered the scrubber, then

$$Q = I - M_s \tag{I–1}$$

For operation at a constant efficiency, $M_s = \varepsilon I$; therefore,

$$Q = M_s \frac{(1 - \varepsilon)}{\varepsilon} \tag{I–2}$$

A similar expression can be derived in terms of the penetration, p, the fraction of the material entering the scrubber that passes through the system and is discharged in the exhaust ($p = 1 - \varepsilon$). It has been pointed out (Randle 1971, Anonymous 1989) that this calculation, made with no knowledge of either I or Q, is susceptible to substantial undetected error if ε is much less than the presumed value. For small values of ε, the equation is also unreliable because $(1 - \varepsilon)/\varepsilon$ increases without limit as ε approaches zero. This leads to large overestimates of Q; in reality, Q can not exceed I.

Many estimates of uranium releases from the Plant 8 scrubbers were based on an estimated scrubber efficiency of 83%. This value, which is the midpoint of the range of 70 to 95 percent efficiency specified by one of the manufacturers, was used by the FMPC for an extended period of time for all of the scrubbers (Vath 1964c, Randle 1971, Diehl 1980). Although it was recognized that the feed material and operational conditions had an effect on scrubber efficiency, those effects were not quantified and considered on a routine basis. In an initial investigation, Vath (1964a) considered the range of quoted efficiencies (70 to 95%) as well as the average value. His estimate for the average value was a monthly release of about 610 kg (1340 lb); his estimated range of monthly releases was about 160 to 1270 kg (about 340 to 2800 lb).

The results of special measurements were used in some contemporary estimates of releases. Measured efficiencies of 95, 69, and 79 percent for the rotary kiln, UAP, and caustic scrubbers, respectively, were used by Starkey (1961) to estimate a monthly release of about 460 kg (1005 lb) from the scrubbers to the atmosphere. Starkey (1964) estimated a total monthly loss from Plant 8 of about 660 kg (1460 lb). A comparable monthly release estimate of 1500 lb (~ 680 kg) is quoted in a contemporary letter to the Atomic Energy Commission by Noyes (1964) related to material accountability difficulties in Plant 8. Vath (1964c) also developed a revised estimate of 680 kg per month in a subsequent memorandum related to uranium accountability. Hill (1989) reported that at the end of fiscal year (FY) 1964, an inventory difference of about 58,000 kg of uranium was attributed to scrubber losses during previous years of operation.

In the compendium of FMPC releases, document FMPC-2082 (Boback et al. 1987), the total Plant 8 scrubber releases in early periods of operation are given by fiscal year. During a subsequent review, the calculations for FMPC-2082 could not be located, but it was reported to the reviewer that the amount written off at the end of FY 1964 had been distributed over the fiscal years 1954-1964 (Hill 1989). The sum of the releases reported in Boback et al. (1987) for those fiscal years is about 20,000 kg.

The rounded values from a handwritten tabulation of inventory adjustments (Courtney 1964) are shown in Table I-3. The inventory adjustments were assigned to FY 1964, but it was recognized that the losses had occurred over a number of years of previous operations. The amount attributed to unmeasured scrubber and vent losses was about 37,000 kg, roughly 64% of the total write-off in that category reported by Hill (1989) and 181% of the total scrubber emissions presented in Boback et al. (1987). The totals given agree approximately with amounts determined from the formal inventory withdrawal records (Gessiness 1964, Noyes 1964). For natural uranium, the FY 1964 write-off was about 130,000 kg U; for enriched uranium about 32,000 kg were removed from the inventory. The enriched uranium losses were later distributed by Vath (1966) to the fiscal years 1961-1965. The fractions of the loss attributed to the four fiscal years were 0.142, 0.157, 0.190, and 0.511, respectively.

Table I-3. Compilation[a] of Inventory Adjustments to Account for Unmeasured Losses During Previous Years Prior to FY 1964

	Plant 8 inventory adjustment (kg)[b]	
Adjustment category	Natural U	Enriched U
Liquid effluent	33,000	17,000
Barren filter cake	23,000	4,000
Burn pit losses	44,000	3,000
Scrubber and vent losses	29,000	8,000
All categories	129,000	32,000

[a] Courtney (1964).

[b] Values have been converted from original tabulations in pounds of uranium and rounded to the nearest thousand kilograms. Although original values were given to four or five significant figures, it is clear that the specific values and the distributions by category are not known with great precision. An evaluation during May 1964 showed the Plant 8 volume and uranium concentration measurements to be substantial underestimates, by factors of 4 and 11, respectively (Vath 1964a). Comparison sampling of dry discards showed better results; Plant 8 values were low by about 33% (Vath 1964b).

Hill (1989) reported an upper limit estimate of scrubber losses during the FY 1954–FY 1964 period. In this calculation it was assumed that the monthly release rate of 680 kg (1500 lb) occurred in FY 1964, the year of highest production. Hill references Vath (1964c) who suggested that release rate in 1964, based upon an average scrubber efficiency of 83%. The releases for other fiscal years were then computed using a scaling factor that was the ratio of the production in the particular year to the production in FY 1964.

The results of Hill's (1989) calculations and the estimates from FMPC-2082 (Boback et al. 1987) both track the Plant 8 production data (see Appendix C). The total scrubber release for FY 1954–1964 estimated by Hill (1989) is 57,712 kg, the exact amount reported to have been written off at the end of FY 1964 (Hill 1989). The estimates from FMPC-2082 are consistently a factor of 2.85 lower than the estimates of Hill (1989). That difference could correspond to selection of scrubber penetration 2.85 times smaller than the 17% assumed by Hill, or ~ 6%. As discussed later, some measurements of penetration of uranium through the scrubbers have been that low. However, the data show that consistent performance at that level was not realized.

DATA DESCRIBING PLANT 8 OPERATIONS

Production Data

Plant 8 production data have been summarized on a fiscal year basis in Appendix C. Monthly production data were not generally available although they were found for most of the period 1956–1960 in plots in a history of FMPC uranium inventories (Anonymous, circa 1973–1976). When monthly data were available, calendar year average recovery rates could be computed exactly. For most years, average values for calendar years were estimated approximately using the tabulation in Appendix C. Figure I-1 shows the average monthly uranium recovery amounts estimated for each calendar year of operation. During 1953 operations were conducted only in November and December; average recovery for those months was 37 metric tons of uranium (MTU).

Uranium Collected in Scrub Liquor

The Plant 8 scrubbers were charged with a 10% sodium hydroxide (NaOH) solution. The NaOH concentration was gradually reduced during scrubber operation. When it reached 1%, or after 1–2 weeks of operation, the scrubber solution was changed. The reaction of the NaOH solution with the U_3O_8 particles captured in the scrubbers is very complicated chemically. The reaction produces a complex mixture of sodium uranates with low solubility. An excellent study of phase relations in the sodium oxide–uranium trioxide–water system (Ricci and Loprest 1955) showed that sodium uranates exhibit wide ranges of solid solution and that the solubility of uranium trioxide in the ternary solution is only about 10 mg L^{-1}.

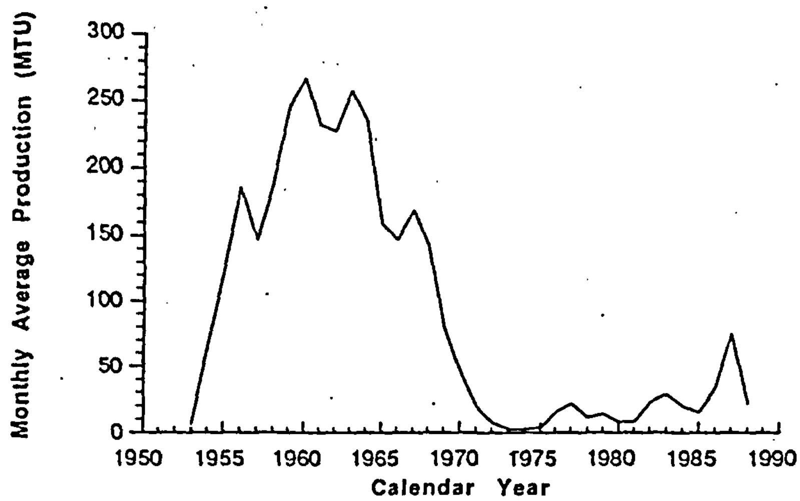

Figure I–1. Estimated average monthly production (uranium recovery) for operations in Plant 8.

The recirculating scrub liquor carried the sodium uranate and uranium oxide solids in suspension. The solids gradually settled out in the main scrub liquor storage tank and were periodically removed. Reprocessing of the insoluble solids or "scrubber cake" was performed at the FMPC to recover the uranium.

After it was recognized that significant amounts of uranium were being discharged to the environment from the Plant 8 scrubbers, the method described above was used to estimate uranium releases. That method depended on knowledge of the amounts of uranium that were collected in scrub liquor. Detailed data on scrub liquor content were compiled and used to estimating losses of uranium from scrubber discharges. Unfortunately, nearly all of the detailed data has been lost. Monthly data on the amounts of uranium collected in scrub liquor have only been located for twenty months between 1959 and 1964 (Beers 1961, Vath 1964c). Table I–4 contains most of the data on uranium in scrub liquor as well as individual monthly or average data on production in Plant 8 (Rathgens 1970, Rathgens et al. 1985). One additional isolated value for scrub liquor content is 2964 kg in February 1964.

The contributions of individual scrubbers to the total amounts of uranium in scrub liquor were recorded in detail at the time. The breakdowns for October 1959 (Marshall 1959) and February 1964 (Vath 1964c) are given in Table I–5. These data indicate that the scrubbers serving the calciner, rotary kiln, and UAP furnace were the most important for estimation of releases to the atmosphere at that time. These data are generally consistent with the equipment capacity data given in Table I–2, but it would have been desirable to have additional empirical breakdowns of the contributions to the total scrub liquor.

Table I-4. Average Monthly Production and Scrub Liquor Uranium Content, October 1959–June 1962

Year	Month	Monthly production (MTU)	Scrub liquor content (kg U)
1959	October	249	2139
	November	236	4345
	December	227	1588
1960	January	204	562
	February	249	4804
	March	295	5466
	April	299	
	May	299	
	June	318	
	July	259	
	August	304	
	September	240	
	October	268	
	November	227	
	December	240	
1961	January	a	
	February	a	
	March	a	
	April	a	
	May	a	
	June	a	2849
	July	b	1624
	August	b	485
	September	b	3075
	October	b	1066
	November	b	12615
	December	b	6731
1962	January	b	2159
	February	b	3561
	March	b	889
	April	b	3443
	May	b	2617
	June	b	

[a] Average monthly production during the last half of FY 1961 was 275 metric tons of uranium (MTU).
[b] Average monthly production during FY 1962 was 242 MTU.

Table I-5. Contributions from Individual Scrubbers to Total Recovery of Uranium in Scrub Liquor

Scrubber for	Uranium (kg) collected in scrub liquor	
	October 1959	February 1964
Calciner, muffle and box furnaces	1370	522
Rotary kiln	569	1310
UAP furnace	184	1124
Digesters	1	9
Oxidation furnaces	15	
All equipment	2139	2965

Additional information on collection of uranium in scrub liquor was obtained from the records of withdrawal of uranium from the FMPC inventory (McCreery and Gessiness 1959–1978). The monthly data collected on uranium retention in the scrubbers were summed to make an estimate of the losses via scrubber discharges during each fiscal year. Typically, estimates of anticipated losses were made at the beginning of a 6- or 12-month period and the projections were revised once or more as better information became available or changes in processing schedule were determined. Table I–6 contains information on withdrawals from inventory for enriched uranium between July 1964 and June 1973. Average production figures are available for these periods.

Table I-6. Withdrawals of Uranium from Inventory to Account for Scrubber Losses From FY 1965 Through FY 1973

Period	Withdrawals (kg U) for scrubber losses		
	Enriched U	Natural U	Total
July 1964–June 1965	2735	3075	5810
July–December 1965	454	225	679
January–June 1966	21	225	246
July–December 1966	182	484	666
January–June 1967	641	483	1124
July–December 1967	714	362	1076
January–June 1968	1644	363	2007
July–December 1968	2717	[a]	2717
January–June 1969	406	[a]	406
July–December 1969	592	37	629
January–June 1970	[a]	37	37
July 1970–June 1971	203	338	541
July 1971–June 1972	[a]	[a]	[a]
July 1972–June 1973	34	5	39

[a] No withdrawal for period.

The total amounts withdrawn from inventory had been estimated using measured amounts of uranium collected in scrub liquor and the assumed efficiency of 83% in Eq. (I–2). This procedure was used at the FMPC until 1982 (Diehl 1980, Bardo 1985). The reverse procedure was used to compute the amounts that were measured in scrub liquor using the withdrawal data from Table I–6. Those values were then used together with average uranium recovery tonnages for those periods to estimate the ratio of the quantities in scrub liquor to the uranium recovery.

The inventory withdrawal records reviewed (McCreery and Gessiness 1959–1978) did not explicitly identify any scrubber losses after June 1973. Uranium recovery activity in Plant 8 was lowest during the period 1973–1974 (see Figure I–1), which may account for decreased attention to that facility. Some other sources of information were located for the years 1975–1981. These are discussed below.

Although not reflected in uranium recovery data for the plant, approximately 92,000 kg of ore concentrates were calcined in the rotary kiln between 30 June and 11 July 1975. At the end of this processing campaign approximately 2300 kg U were unaccounted for (Adams 1975). About 1850 kg U were removed during a thorough cleaning of the scrubber as part of the search for missing uranium. Although the possibility of loss to the atmosphere was not addressed in the report, that release path was probably important.

The importance of the rotary kiln to Plant 8 processing was continued during the next several years. Handwritten data on collections of uranium in scrub liquor (Anonymous 1980) show it to be the principal source of that material, accounting for 80–100 percent of the total. The box furnace, with its own scrubber, and an oxidation furnace continued to operate throughout the 1970s. The muffle furnace operated periodically until early 1977 and does not appear to have been used regularly again until 1982. The kiln, box furnace, and oxidation furnace were operating when Ross (1979, 1980, 1982, 1983) began measurements of releases from the scrubber exhausts that formed the basis for the emission factors used in the 1980s.

Table I–7 summarizes information on the collection of uranium in scrub liquor and the corresponding estimates of the ratio of scrub liquor content to production. Because only average uranium recovery rates are known for many periods, most of the estimates of the ratio (R, kg U MTU^{-1}) of scrub liquor collection to plant production were made for periods of six months or more. The exception is the value for the short period when ore concentrates were calcined in the kiln in 1975. The four estimates for periods between October 1959 and June 1965 lie near the center of the range of values from subsequent periods. The data in the table provide a basis for estimating quantities of uranium in scrub liquor for periods when such information is unavailable. Their application is discussed in the section describing calculations of releases.

Quantities of uranium in scrub liquor and data on scrubber performance can be used together to make estimates of releases from the scrubbers. Scrubber performance can be characterized by a collection efficiency or its complement, the amount of penetration through the scrubber. In the following section data on scrubber performance from two sources are discussed.

Table I-7. Estimates of the Ratio of Uranium Collection in Scrub Liquor to Production (Uranium Recovery) in Plant 8

Period	Quantity (kg U) collected in scrub liquor	Plant 8 production (MTU)	Ratio (R) (kg U MTU^{-1})
Oct 1959–Mar 1960	18904	1460	12.9
June–Nov 1961	21714	1485	14.6
Dec 1961–May 1962	19400	1452	13.4
July 1964–June 1965	28336	2134	13.3
July–Dec 1965	3315	838	3.95
Jan–June 1966	1201	838	1.43
July–Dec 1966	3252	920	3.54
Jan–June 1967	5488	920	5.97
July–Dec 1967	5253	1111	4.73
Jan–June 1968	9799	1111	8.82
July–Dec 1968	13265	593	22.4
Jan–June 1969	1982	593	3.34
July–Dec 1969	3071	365	8.42
Jan–June 1970	181	365	0.50
July 1970–June 1971	2641	370	7.15
July 1972–June 1973	190	66	2.89
June–July 1975	1850	92	20.2
July 1975–Jan 1977	727	154	4.73
Feb 1977–Jan 1978	2112	246	8.57
Feb–Nov 1978	356	112	3.18

MEASUREMENTS OF SCRUBBER PERFORMANCE

During the years 1958–1965 simultaneous measurements of concentrations of U in the exhaust and intake air were obtained by members of the Industrial Hygiene and Radiation staff at the FMPC. Two sources of information about these measurements have been located. The first source of data is a set of analytical data sheets from the Health and Safety Division's radiochemical laboratory (NLCO 1958–1965). Laboratory data sheets, which were used to record information about the samples and the analytical results, were found for fifty measurements of scrubber performance during the period. Records that contained enough information to estimate scrubber efficiencies were primarily for measurements made during 1961–1965. The second source of information was FMPC internal memoranda summarizing measurements of scrubber efficiency. Plant memoranda contained information about nineteen sets of scrubber performance measurements.

Performance Estimates Based on Data from Laboratory Analytical Sheets

The laboratory analytical sheets typically included descriptive information about the scrubber being measured and the intake and exhaust samples taken, and results of the uranium analyses, in units of total uranium measured and concentration of uranium in the air (NLCO 1958–1965). Estimates of scrubber efficiency or penetration were not recorded on the data sheets, but these quantities can be computed using the recorded information.

Three categories of estimates of scrubber performance were established. The first category consists of estimates based upon measurements of concentrations of uranium in the inlet and outlet streams and of the corresponding flow rates. Then the inlet and outlet mass loadings can be computed. If F_i and F_o are the inlet and outlet flow rates ($m^3\ s^{-1}$), carrying uranium concentrations ($g\ m^{-3}$) of C_i and C_o, respectively, then the corresponding mass loadings (M_i and M_o, $g\ s^{-1}$) are

$$M_i = F_i\, C_i \text{ and } M_o = F_o\, C_o \tag{I–3}$$

and the penetration is computed using

$$p = M_o / M_i \tag{I–4}$$

and the efficiency is $\varepsilon = 1 - p$. Performance estimates of this type, based on the inlet and outlet mass loadings, are preferred and were used whenever possible.

If mass loadings could not be determined, estimates based upon uranium concentrations alone were used. If C_i ($g\ m^{-3}$) is the concentration measured at the inlet and C_o ($g\ m^{-3}$) is the outlet concentration, then the efficiency can be estimated using

$$p = C_o / C_i \tag{I–5}$$

This calculation assumes that the flow rate of air into the scrubber equals the flow rate exhausting the scrubber. There were two alternative concentration-based estimates. The best estimates of this type are those in which the concentrations were corrected to standard conditions of temperature and pressure (295 K and 1 atmosphere). Unfortunately, the data on temperatures and pressures that were available at the time were frequently not recorded on the analytical sheet. This fact required the definition of the third, and least preferred, type of estimate, one based on uncorrected concentration data.

Table I–8 contains estimates of apparent uranium penetration through the scrubbers based on results recorded on the analytical data sheets (Schmidt 1992). When the available data permitted more than one type of estimate, all of the estimates are shown. A blank in either of the last two columns indicates that it was not possible to make the corresponding type of estimate because of lack of information.

The term *apparent penetration* is used because carryover of uranium in drops of scrub liquor seems to have been an important process. When the inlet air concentration is relatively low but the scrub liquor is carrying high uranium concentrations, just prior to regeneration for example, carryover of droplets of the scrub liquor can lead to high outlet uranium concentrations and apparent penetrations that exceed 100 percent. The importance of carryover seems also to be supported by the finding, presented later, that the apparent penetrations were generally lower when the uranium concentrations in inlet air stream were high.

Table I-8. Apparent Uranium Penetration Through Plant 8 Scrubbers Based on Measurement Results Recorded on Analytical Data Sheets, 1961–1965

Date sampled	Apparent penetration (%) by estimation method: Uncorrected concentration	Corrected concentration	Mass loading
	Scrubber for the primary calciner		
3-15-61	0.26	0.20	0.19
3-17-61	0.18		
3-29-61	13		
3-30-61	20		
3-19-62	2.0	2.0	
3-20-62	1.5		1.5
3-22-62	5.7	4.0	4.2
3-22-62	6.0	5.8	5.3
6-3-65	76		100
6-4-65	9.9		14
	Scrubber for rotary kiln		
3-20-61	8.0		
3-21-61	100		
3-27-61	5.3		
3-28-61	1.1		
5-23-61	2.2		1.9
5-25-61	6.9	5.4	
5-26-61	16		
7-6-61	11		
7-6-61	10		
11-3-61	1.2	0.74	0.83
12-5-61	2.2	1.8	2.7
7-30-62	22		
8-1-62	2.1		
8-2-62	7.6		
8-3-62	11		
	Scrubber for NPR furnace		
3-21-63	86	59	82
3-27-63	100	70	110
3-28-63	260	190	240
11-7-63	11	7.2	13
11-8-63	32	20	38
11-13-63	28	19	34

Table I-8 (Continued). Apparent Uranium Penetration Through Plant 8 Scrubbers Based on Measurement Results Recorded on Analytical Data Sheets, 1961–1965

Date sampled	Apparent penetration (%) by estimation method: Uncorrected concentration	Corrected concentration	Mass loading
	Scrubber for UAP furnace		
3-24-61	160		
3-24-61	33		
9-18-62	33	21	29
9-20-62	54		
7-9-63	11	6.9	
7-10-63	29	19	
7-11-63	26	26	
8-29-63	13	8.7	15
8-29-63	7.8	5.2	8.8
11-15-63	1.1		
11-19-63	0.38	0.23	
12-18-63	28	18	32
12-19-63	7.9	5.0	9.2
12-20-63	70	45	88
8-10-65	60	45	
8-11-65	3.7	2.5	
8-12-65	2.9	2.0	
9-8-65	0.97	0.52	
	Scrubber for oxidation furnace #1		
8-11-61	190		

Droplet carryover is of course not the only reason for poor collection efficiencies by the scrubbers. It was noted previously that overloading of the UAP furnace was considered an important reason for the difficulties with its operation (Noyes 1962). Variability in scrubber performance was also a factor. Although penetration of uranium through the scrubbers for the primary calciner and for the rotary kiln was usually found to be relatively low, some very high apparent penetrations were also observed. Performance of the other scrubbers was even more variable.

Performance Estimates Reported in Plant Memoranda

There were several internal plant memoranda that contained information on the performance of the Plant 8 scrubbers. These were prepared to document measurements, support estimates of uranium losses, and to support proposals to upgrade the scrubber discharge treatment equipment. Table I–9, which includes the calculation method and the reference for each estimate, summarizes these performance estimates. Most results were reported as scrubber efficiencies that were computed using the mass loading method. In one memorandum the method used was not stated. Four of the results had been based on concentrations measurements alone, but better estimates have been derived from the data (Schmidt 1992). Those and other estimates in Table I–9 have been presented in terms of uranium penetration through the scrubbers.

Table I-9. Estimates of Uranium Penetration Through Plant 8 Scrubbers Based on FMPC Memoranda

Scrubber for	Date sampled	Calculation basis	Penetration[a] (percent)	Reference
Rotary kiln	[b]	Mass loading[b]	5[b]	Starkey (1961)
UAP furnace	[b]	Mass loading[b]	31[b]	Starkey (1961)
Primary calciner	[b]	Mass loading[b]	21[b]	Starkey (1961)
UAP furnace	3-24-61	Mass loading	33	Bipes (1963b)
UAP furnace	3-24-61	Mass loading	55	Bipes (1963b)
Rotary kiln	7-30-62	Mass loading[c]	20[c]	Bipes (1962)
Rotary kiln	8-1-62	Mass loading[c]	2.2[c]	Bipes (1962)
Rotary kiln	8-2-62	Mass loading[c]	6.7[c]	Bipes (1962)
Rotary kiln	8-3-62	Mass loading[c]	8.1[c]	Bipes (1962)
UAP furnace	9-18-62	Mass loading	47	Bipes (1963b)
UAP furnace	9-20-62	Mass loading	79	Bipes (1963b)
UAP furnace	7-9-63	Mass loading	12	Bipes (1963b)
UAP furnace	7-10-63	Mass loading	29	Bipes (1963a,b)
UAP furnace	7-11-63	Mass loading	23[d]	Bipes (1963a,b)
UAP furnace	8-29-63	Mass loading	8	Bipes (1963a,b)
UAP furnace	8-29-63	Mass loading	11[d]	Bipes (1963a,b)
NPR furnace	11-7-63	Mass loading	12	Bipes (1963c)
NPR furnace	11-8-63	Mass loading	21	Bipes (1963c)
NPR furnace	11-13-63	Mass loading	30	Bipes (1963c)
UAP furnace	12-18-63	Mass loading	27	Bipes (1964)
UAP furnace	12-19-63	Mass loading	72	Bipes (1964)
UAP furnace	12-20-63	Mass loading	7	Bipes (1964)
UAP furnace	Spring 1964[e]	Not indicated	26–29[e]	Vath (1964b)

[a] The memos reported scrubber efficiencies in percent. The penetrations shown here were computed using $p\,(\%) = 100\% - \varepsilon\,(\%)$.

[b] The memo, dated 11 April 1961, states that the measurements had "recently been completed." This would indicate that they were performed in the previous month or two. Scrubber inlet loading data are given and it is presumed that they were used to estimate the efficiency. The memo also indicates that the values were averages; the number of samples was not given. The result for the calciner is also in a memo by Chenault (1961).

[c] The memo reported efficiencies based on concentrations. However, it also contained enough data to calculate penetration on the basis of inlet and outlet mass loadings (Schmidt 1992). The latter results are presented here.

[d] In Bipes (1963a), the efficiencies measured on 11 July and 29 August are stated to be 74% and 87%, respectively; no data are given. Examination of the data included in Bipes (1963b) indicates that the correct values for efficiency were 77% and 89%, respectively.

[e] The memo does not provide dates or the number of measurements.

The measurements for the UAP furnace scrubber that were performed in the summer of 1963 followed refurbishment of that scrubber. The feed to the furnace during the July measurements was at the normal rate of five drums per hour; however, at the end of August it was reduced to three drums per hour to test the effect of that action. There was a noticeable improvement in the scrubber's performance during operation at the reduced feed rate (Bipes 1963a). That reference does not include the result for 9 July, the first after the modifications were completed; the feed rate on that day is not known.

The measurements of scrubber efficiency for the rotary kiln scrubber were performed after the scrubber had been refurbished (June 1962) to reduce losses. Uranium fines were being processed during the measurements. The report of results (Bipes 1962) is detailed and contains information on inlet and exhaust flow and loading rates.

The reductions in efficiency observed during sequential measurements for the NPR Furnace scrubber in November 1963 were attributed to the buildup of solids in the scrubber. The first measurement was made after cleaning the scrubber cone; continued operation prior to the second and third measurements had resulted in an increasing buildup of material (Bipes 1963c).

Summary

Both data sources were used to characterize uranium penetration through the Plant 8 scrubbers. Estimates based on mass loading were preferred over those based upon concentration data alone. When two estimates based on mass loading differed, the mean of the two values was used. Estimates based on corrected air concentration data were used whenever possible instead of those based upon concentrations that had not been converted to standard conditions. The following sections summarize the preferred estimate for each of the scrubber performance tests. These short-term test results are the only basis for estimating long-term average values needed for calculations of scrubber releases. In the summaries, mean values of apparent penetration (p) are given with (±) the standard deviation of the mean.

Uranium penetration through the primary calciner scrubber. The estimates for uranium penetration through the scrubber that served the primary calciner and other small furnaces were derived from information recorded on analytical data sheets. There were ten estimates in all; preferred values and the method used to derive them, from Schmidt (1992), are shown in Table I–10. Six of the ten estimates were based on mass loading calculations. One estimate was based on concentrations corrected to standard conditions. Typical scrubber inlet mass loadings were in the range of 30 to 110 kg d^{-1}. One estimate of apparent penetration through the calciner scrubber was greater than 20%. It was obtained during testing of a new nozzle under conditions when the mass loading was only 0.2 kg d^{-1}, far below the normal operating condition. For that reason it was not included in the calculation of the mean penetration of 6.7 percent or in the distribution of individual penetration estimates shown in Figure I–2.

Uranium penetration through the rotary kiln scrubber. Table I–11 contains the preferred values and the basis used to estimate uranium penetration through the scrubber serving the rotary kiln. Seven of the fifteen estimates were based on mass loading calculations and one was based on corrected concentration data. Nearly all of the estimates of penetration were less than 20%, but one indicated complete penetration. The mass loading during the measurement that yielded an apparent penetration of 100% is not known, although other measurements near the same time were performed at a loading of more than 20 kg d^{-1}. The mean of all the measurements of penetration through the rotary kiln scrubber was 13 percent. The distribution of these short-term measurements is shown in Figure I–3.

Table I-10. Summary of Short-Term Measurements of Uranium Penetration Through the Primary Calciner Scrubber

Date	Method[a]	p (%)	Date	Method[a]	p (%)
3-15-61	ML	0.19	3-22-62	ML	4.2
3-17-61	U	0.18	3-22-62	ML	5.3
3-29-61	U	13	6-3-65	ML	100[b]
3-30-61	U	20	6-4-65	ML	14
3-19-62	C	2.0			
3-20-62	ML	1.5	Mean[c]	(n = 9)	6.7 ± 2.5

[a] ML: mass loading; C: corrected concentration, U: uncorrected concentration.
[b] Measured at very low loading; not included in average.
[c] Mean value ± the standard deviation of the mean; n: number of measurements.

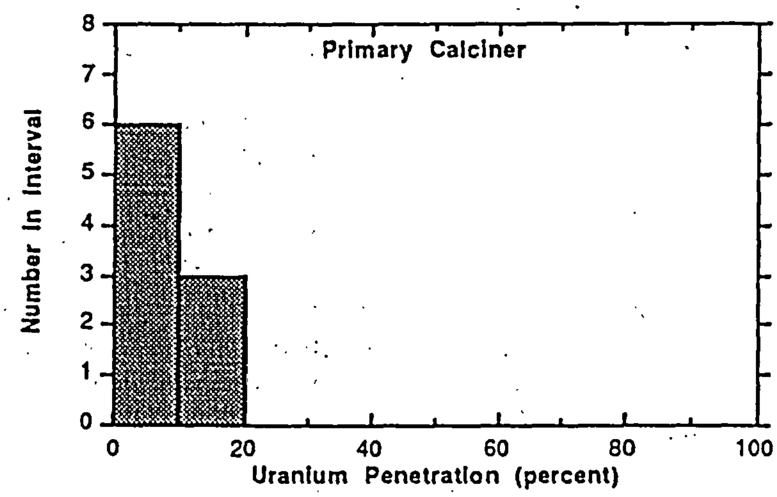

Figure I–2. Distribution of results of short-term uranium penetration measurements for the calciner scrubber made during 1961–1965.

Table I–11. Summary of Short-Term Measurements of Uranium Penetration Through the Rotary Kiln Scrubber

Date	Method[a]	p (%)	Date	Method[a]	p (%)
3-20-61	U	8.0	7-6-61	U	10
3-21-61	U	100	11-3-61	ML	0.83
3-27-61	U	5.3	12-5-61	ML	2.7
3-28-61	U	1.1	7-30-62	ML	20
5-23-61	ML	1.9	8-1-62	ML	2.2
5-25-61	C	5.4	8-2-62	ML	6.7
5-26-61	U	16	8-3-62	ML	8.1
7-5-61	U	11	Mean[b]	(n = 15)	13.3 ± 6.6

[a] ML: mass loading; C: corrected concentration, U: uncorrected concentration.
[b] Mean value ± the standard deviation of the mean; n: number of measurements.

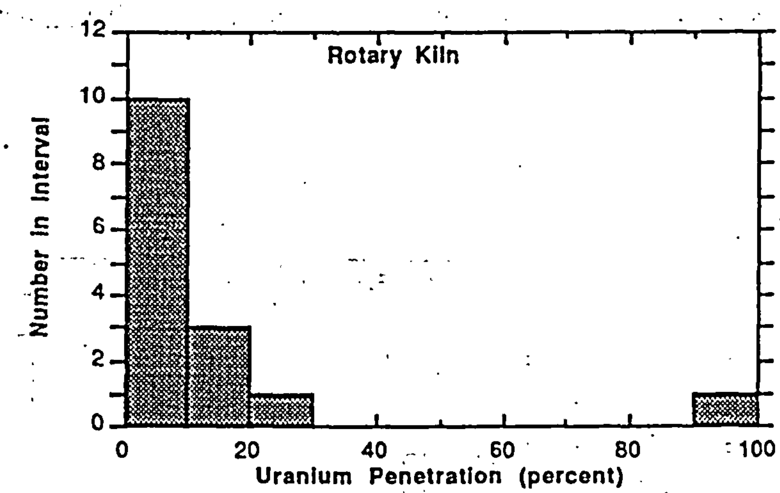

Figure I–3. Distribution of results of short-term uranium penetration measurements for the rotary kiln scrubber made during 1961–1962.

Uranium penetration through the UAP furnace scrubber. There were eighteen short-term measurements yielding estimates of uranium penetration through the scrubber that served the UAP furnace. Information about these measurements is given in Table I–12. Twelve of the estimates were based upon mass loading calculations and five were based on corrected concentration data. Mass loadings were typically less than 8 kg d^{-1} although two measurements were made at 12 kg d^{-1} and one was made at 20 kg d^{-1}. The range of apparent penetrations was wide, ranging from 0.2 to 80 percent. Half of the estimates were between 20 and 80 percent. The mean value was 25 percent. The data indicate that the performance for the UAP furnace scrubber was generally poorer than that of the scrubbers serving the primary calciner and rotary kiln.

Table I-12. Summary of Short-Term Measurements of Uranium Penetration Through the UAP Furnace Scrubber

Date	Method[a]	p (%)	Date	Method[a]	p (%)
3-24-61	ML	55	11-19-63	C	0.23
3-24-61	ML	33	12-18-63	ML	29
9-18-62	ML	38	12-1-63	ML	8.1
9-18-62	ML	79	12-20-63	ML	80
7-9-63	ML	12	8-10-65	C	45
7-10-63	ML	29	8-11-65	C	2.5
7-11-63	ML	23	8-12-65	C	2.0
8-29-63	ML	13	9-8-65	C	0.52
8-29-63	ML	8.4			
11-15-63	U	1.1	Mean[b]	(n = 18)	25.5 ± 6.2

[a] ML: mass loading; C: corrected concentration, U: uncorrected concentration.
[b] Mean value ± the standard deviation of the mean; n: number of measurements.

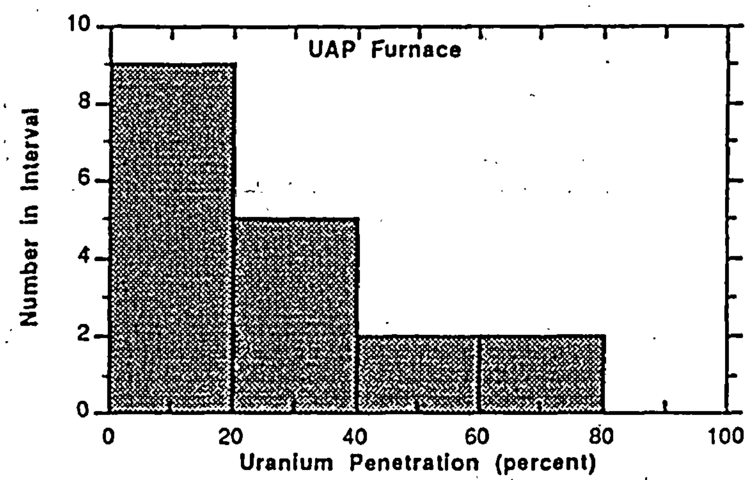

Figure I–4. Distribution of results of short-term uranium penetration measurements for the UAP furnace scrubber made during 1961–1965.

Uranium penetration through the oxidation furnace scrubbers. Most of the oxidation furnace scrubber measurements were made in the discharge from furnace #2, called the NPR furnace. Table I–13 contains information on uranium penetration through that scrubber. All of the estimates of apparent penetration were based on mass loading computations. Mass loadings for the NPR scrubber were generally low, averaging about 3 kg d^{-1}. The smallest apparent penetration measured was 12%. Two of the six values exceeded 100%, indicating substantial carryover of uranium in scrub liquor droplets during the measurement. The single measurement for the other oxidation furnace, based upon uncorrected concentration data, indicated a penetration greater than 100%, again indicating substantial carryover of droplets during the measurement. Although the true penetration of uranium through these scrubbers can not be reliably determined from the data, it was clearly substantial, with an average estimated to lie between 50 and 60 percent.

Table I-13. Summary of Short-Term Measurements of Uranium Penetration Through Scrubbers for the NPR Furnace and Oxidation Furnace #1

Date	Method[a]	p (%)	Date	Method[a]	p (%)
3-21-63	ML	82	11-8-63	ML	30
3-27-63	ML	> 100	11-13-63	ML	32
3-28-63	ML	> 100	8-11-61	U	> 100
11-7-63	ML	12			

[a] ML: mass loading, NPR Furnace; U: uncorrected concentration, Oxidation Furnace #1.

Entrainment of scrub liquor. As noted above and elsewhere in this section, entrainment or carryover of scrub liquor appears to have been an important factor that influenced overall performance of the scrubbers. This conclusion is based in part upon the observations of very high apparent penetrations, as in Table I–13, and upon comparison of measured performance with expected performance.

Removal efficiencies of venturi scrubbers for various inlet particle sizes given in Lund (1971) exceed 99% for particles with diameters greater than 5 µm. Using a distribution of furnace off-gas particle sizes based upon plant documents, it was estimated that scrubber efficiencies should have been about 97% (Killough et al. 1993, App. D). The preceding tables show that most measured apparent penetrations were above 3%. This comparison also suggests that entrainment of droplets containing suspended uranium particles contributed substantially to the releases. Entrainment appears to be a more likely explanation than the alternative that the diameters of particles entering the scrubbers were very much smaller than is indicated in the limited available documentation.

DEPENDENCE OF PERFORMANCE ON SCRUBBER LOADING

Examination of the scrubber performance data suggested that uranium penetration through the scrubbers was dependent upon the concentration of uranium in the scrubber inlet duct (Schmidt 1992). Table I–14 contains all the penetration estimates obtained from mass loading calculations. For these tests the amounts of uranium entering the scrubber, referred to as the scrubber inlet loading, have been computed and are shown in the third column. The apparent penetrations are the best estimates from the data sheets and plant memos, evaluated as described previously.

For the calciner, inlet loadings ($\acute{I}$, kg d^{-1}) generally varied between about 30 and 110 kg d^{-1}. The unusually low loadings in June 1965 were during a test of a new scrubber spray nozzle. Inlet loadings for the scrubber serving the rotary kiln were higher in 1961 than in tests conducted after scrubber refurbishment in 1962. The UAP furnace scrubber generally handled less than 10 kg d^{-1}, although in three instances the loadings were higher than that. Loadings for the oxidation furnace scrubbers were even lower, about 3 kg d^{-1}.

The relationship between ln p and ln C_i (the inlet concentration) seen by Schmidt (1992) suggested that there would be a similar relationship between ln p and ln $\acute{I}$ and that was observed: This implied that a potentially useful relationship between penetration and the rate of buildup of uranium in scrub liquor was also present. Values of this variable (M_s', kg d^{-1}) were computed from the data in Table I-14 and are the basis for Figure I-5.

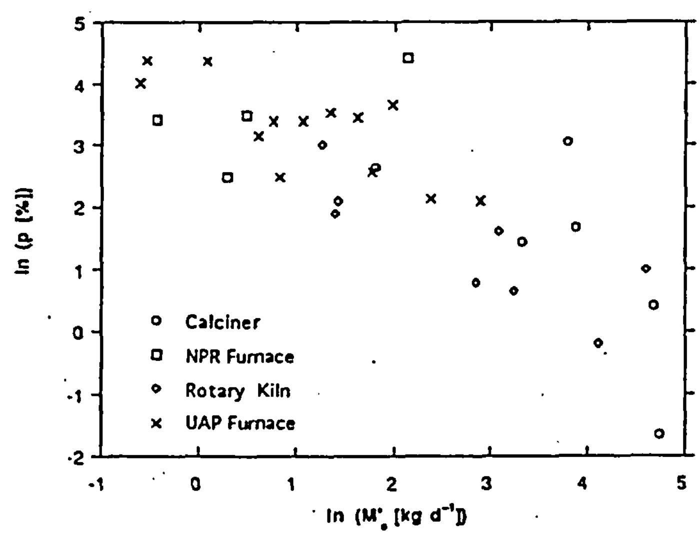

Figure I-5. Plot of the logarithm of the apparent penetration through scrubbers and the logarithm of the computed rate of uranium accumulation in scrub liquor.

These observations are consistent with the earlier observation that carryover of droplets of scrub liquor is an important factor. When the challenge aerosol concentrations are relatively low, the uranium carried in the droplets can be the controlling factor in determining the apparent penetration. This is perhaps best illustrated by the measurement made for the calciner when the inlet loading was only 0.2 kg d^{-1}.

In Figure I-5 the logarithm of the apparent penetration of uranium through the scrubbers is plotted against the logarithm of the rate of accumulation of uranium in scrub liquor. This plot shows that ln p generally decreases as ln M_s' increases; however, there is substantial variability for all of the scrubbers. The limited data for the NPR furnace scrubber run counter to the general trend.

Table I-14. Estimates of Apparent Uranium Penetration Through Plant 8 Scrubbers Based on Mass Loading Data

Date sampled	Equipment [illegible]d [illegible]ber	Scrubber inlet loading (kg d^{-1})	Apparent uranium penetration (%)
Feb–Mar 1961	[illegible]er	56	21
3-15-61	[illegible]ner	110	0.19
3-20-62	[illegible]ciner	110	1.5
3-22-62	Calciner	29	4.2
3-22-62	Calciner	51	5.3
6-3-65	Calciner	0.2	100
[illegible]-65	Calciner	7.0	14
Feb–Mar [illegible]	Rotary Kiln	23	5
5-[illegible]	Rotary Kiln	26	1.9
[illegible]	Rotary Kiln	62	0.83
[illegible]-61	Rotary Kiln	102	2.7
[illegible]-62	Rotary Kiln	4.4	20
[illegible]-62	Rotary Kiln	18	2.2
[illegible]-2-62	Rotary Kiln	4.3	6.7
[illegible]-3-62	Rotary Kiln	4.5	8.1
Feb–Mar 1961	UAP Furnace	7.3	31
3-24-61	UAP Furnace	5.7	33
3-24-61	UAP Furnace	1.2	55
9-18-62	UAP Furnace	12	38
9-20-62	UAP Furnace	5.1	79
7-9-63	UAP Furnace	2.6	12
7-10-63	UAP Furnace	3.0	29
7-11-63	UAP Furnace	2.4	23
8-29-63	UAP Furnace	6.7	13
8-29-63	UAP Furnace	12	8.4
12-18-63	UAP Furnace	4.1	29
12-19-63	UAP Furnace	20	8.1
12-20-63	UAP Furnace	2.9	80
3-21-63	NPR Furnace	46	82
3-27-63	NPR Furnace	6.1	110
3-28-63	NPR Furnace	3.5	240
11-7-63	NPR Furnace	1.5	12
11-8-63	NPR Furnace	0.9	30
11-13-63	NPR Furnace	2.4	32

When the penetration was apparently 100% or greater the rate of accumulation of uranium in scrub liquor is zero or negative; thus, those points cannot be shown on the figure. However, the correlation discussed below does include cases of complete penetration at low inlet mass loadings.

The equation of the line describing the correlation between the variables is:

$$\ln (p\,[\%]) = A - B \ln (M_s'\,[\mathrm{kg\,d^{-1}}]) \qquad \text{(I–6)}$$

The two best-fit coefficients with their standard errors are A = 3.81 ± 0.26 and B = 0.71 ± 0.10. The overall correlation coefficient (r) was 0.79. The relationship is applicable for scrubber uranium accumulation rates ranging from about 0.33 kg d^{-1} (predicted value of p = 100%) to about 110 kg d^{-1}. Evaluation of a data set that does not include the four results for the scrubber serving the NPR furnace improves the correlation slightly (r = 0.83) and changes the parameters of the line by about 10%. Correlations for individual scrubbers were also derived for possible use in making estimates of scrubber releases. The results of application of these correlations are discussed in a later section.

OTHER FACTORS AFFECTING RELEASES FROM PLANT 8 SCRUBBERS

The ranges estimates from short-term measurements of uranium penetration, discussed above, illustrate the variability in performance. Examination of the apparent penetration of uranium through scrubbers as a function of time of measurement did not reveal any notable trends. There were numerous repairs of scrubber system components that were exposed to the corrosive alkaline scrub liquor, but the short-term performance data give no indication of significant improvement or degradation of performance of any of the scrubbers with time during the period 1961–1965.

The most important factor, besides performance variability, that affected scrubber releases seems to have been due to a change in the Plant 8 process. The "UAP furnace" was originally installed in mid-1955 to roast ammonium diuranate (ADU), not uranium ammonium phosphate (UAP). After the process was changed to produce UAP in 1958, problems with the off-gas system occurred. These included blow-back of furnace gases into the work area because of plugging of the off-gas system. Reducing the feed rate to the furnace reduced the problem but did not eliminate it (DeFazio 1966; Mead 1972).

At some point a scrubber bypass line was installed in the ventilation system. This would avoid the backflow of furnace exhaust while the blockage was being cleared and permit workers to continue to operate the furnace. The earliest documents found in FMPC records that cite release estimates from this source were from 1964. Release estimates of 25 lb U per 24 hours of furnace operation (DeFazio 1964) and 750 lb U per month (Starkey 1964) were given. The estimates are consistent for continuous operation of the UAP Furnace. Documentation of the basis for these release estimates has not been found in analytical data sheets. It appears that the emergency system could have been used fairly frequently, perhaps as much as 10% of the time. Such action would increase the average penetration from 25% to 33% and lead to correspondingly greater releases to the environment.

An early schematic drawing of the UAP furnace off-gas system shows no bypass and comparable diagrams for other scrubbers also show no direct pathways to the atmosphere (Calhane 1958b). A later schematic drawing (GFA 1966) contained in a Plant 8 ventilation survey also shows no bypass of the scrubber serving the UAP furnace. However, an undated drawing of the NPR furnace and associated systems in a procedure for calcining enriched uranium (Egart 1962–1963) shows emergency dampers that could release furnace

For comparison, the data in Table I–14 were used to develop a set of release estimates for the earlier period when production was quite high. The results of those calculations are shown in Table I–16. The release rates measured during the 1961–1965 period were uniformly higher than those measured during years of low production. The mean release rate for the rotary kiln was about three times greater than that found during low production. The result for the NPR furnace was about eight times greater than a later value for another oxidation furnace. The mean calciner release rate was more than 100 times higher during the earlier, high production years.

Table I–16. Results of Scrubber Discharge Measurements During Years of High Production in Plant 8

	Measured uranium release (kg U d^{-1})			
Year	Rotary kiln	UAP furnace	NPR furnace	Primary calciner
1961	1.2	2.3	[a]	12
	0.49	1.9		0.21
	0.51	0.66		
	2.8			
1962	0.88	4.6		1.6
	0.40	4.0		1.2
	0.29			2.7
	0.36			
1963		0.31	3.8	
		0.87	6.7	
		0.55	8.4	
		0.87	0.18	
		1.0	0.27	
		1.2	0.77	
		1.6		
		2.3		
1965				0.98

[a] Blanks in the table mean that no measurements were made.

CALCULATIONS OF RELEASES FROM THE PLANT 8 SCRUBBERS

Introduction

The period of operation of the Plant 8 scrubbers was divided into two time periods and release estimates were computed differently for each period. The first period extended from plant startup through 1981. The second began in 1982 and ended in 1988. During the first period, release estimates were based upon estimated long-term average scrubber efficiencies; in the later years, the estimates depend upon release factors derived from measurements shown in Table I–15. The release factor approach is considered more reliable; however, sufficient data on plant operations are not available to apply that approach in earlier years.

The general approach employed is described briefly in the next section. Details of the calculations are discussed subsequently for each period.

General Calculational Technique

The calculations of releases from the Plant 8 scrubbers were performed using Crystal Ball®, a Monte Carlo spreadsheet program (DI 1991). The use of a Monte Carlo procedure explicitly recognizes that there is uncertainty due to variability and lack of knowledge of the individual parameters upon which the calculational result depends. This approach is greatly preferred over a calculation that is based upon point estimates of the various parameters and yields a single result that does not reflect the underlying uncertainties. The Monte Carlo calculation carries the underlying uncertainties in the parameters forward and displays its magnitude in the breadth of the distribution of results. The procedure employs distributions of model parameters and produces a distribution of results.

Calculations of Releases for 1953—1981

For the years 1953–1981, annual uranium releases from the Plant 8 scrubbers and the uncertainties associated with them were estimated by applying a simple model to each scrubber. The calculations employ plant-specific data that were presented above. The types of data used were:

- Plant 8 production (uranium recovery) data
- amounts of uranium found in scrub liquor
- the distribution of the amount of uranium in scrub liquor per unit production
- estimates of utilization and performance of the scrubbers serving the calciner, rotary kiln, UAP furnace, and the two oxidation furnaces

Releases from each scrubber were estimated using a variant of Eq. (I–2) that employs penetration rather than efficiency and rates rather than total amounts.

$$Q' = M_s' \frac{p}{(1-p)} \qquad \text{(I–7)}$$

In this equation, Q' is the release rate and M_s' is the rate of accumulation of uranium in scrub liquor. Because average production information is being converted from fiscal to calendar years and because most information on accumulation of uranium in scrub liquor and its relationship to production is best defined for 6- to 12-month periods, six months was chosen as the basic time interval for these calculations. Some deviations from this schedule were necessary to accommodate calculations that considered the bypass of the

UAP scrubber and the availability of data on M_s'. The sequence of calculations for each 6-month period is described below.

- The first step was to determine the rate of accumulation of uranium in scrub liquor during the period (M_s'). It could be known (Table I–7) or computed from the average production using the ratio R (kg U MTU^{-1}). The value of R was selected from the distribution of values computed from plant data (shown below).
- The next step was to apportion the scrub liquor uranium content among four scrubbers: primary calciner, the rotary kiln, the UAP furnace, and the oxidation furnaces (grouped together for the calculations). Values of two of the apportionment fractions were determined by random selection from distributions described below. Two other fractions were obtained by calculation to assure that the sum of the four fractions chosen was one.
- The third step was to select an estimate of the long-term average uranium penetration through each of the four scrubbers. The distributions used to make these estimates, based upon measurements described previously, are discussed below.
- The fourth step was to compute the release estimate for each scrubber using Eq. (I–7) and to total those for the period. Estimates for calculational periods within a calendar year were then summed to obtain the total release for that year.
- In the last step, estimates of releases for each decade and, subsequently, for the entire period of operation were calculated using Monte Carlo procedures.

It was previously noted that Eq. (I–2) is unreliable for low scrubber efficiencies. The same applies to the analogous equation (I–7) for high scrubber penetrations. As p approaches one, the predicted release increases without limit. To avoid this difficulty, the maximum values of the long-term average scrubber penetration distributions were limited to values of 95% Even though short-term measurements indicated penetrations higher than 95%, such high values are highly unlikely to have persisted for a 6-month period. Scrubber release estimates were checked to assure that the highest predicted annual releases were credible; that is, that they did not exceed the quantity of material that could have entered the scrubber during the year.

In each of the steps requiring selection of a parameter value, that was accomplished using the Monte Carlo calculational procedure. Distributions of each of the parameters required for the calculations are presented next.

Scrub Liquor Accumulation Rate. For some periods, the scrub liquor accumulation rate was known (Table I–7). For those periods, values employed in the calculations were obtained by sampling a uniform distribution with bounds of 0.9 M_s' and 1.1 M_s'. If M_s' was not known, it was computed using the ratio R.

The Ratio R (kg U MTU^{-1}). The ratio of the total amount of uranium collected in scrub liquor (kg U) per unit production (MTU) provides a link between furnace operations and plant production. Figure I–6 shows the composite distribution of the twenty estimates of the ratio R (kg U MTU^{-1}) in Table I–7. The overall distribution covers a fairly broad range of values; the mean value was 8.2 kg U MTU^{-1}. The distribution in Figure I–6 was sampled to obtain estimates of R used, with production data, to compute an estimate of M_s' for the period of interest.

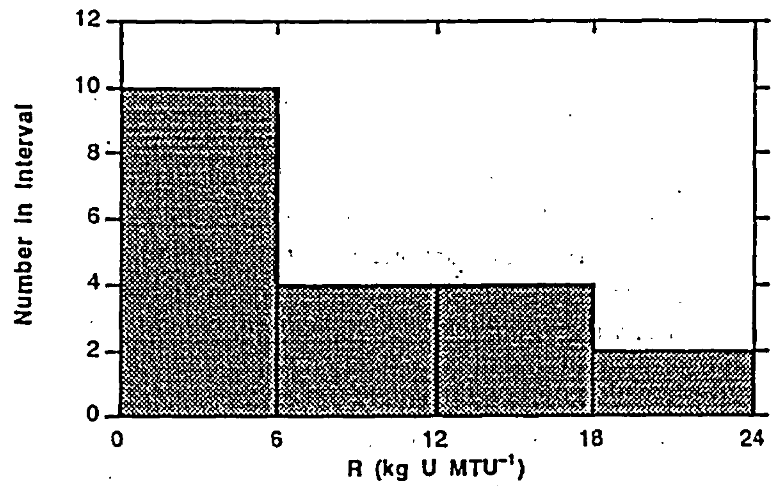

Figure I-6. Distribution of ratios of scrub liquor accumulation to production for Plant 8.

Production Rates. Average production rates were derived from information previously tabulated by the FMPC on a fiscal year basis. Conversion from fiscal years to calendar years was performed in 6-month blocks until 1976. Thereafter, nine months of a fiscal year were in the following calendar year. The 6-month average production estimate was described by a uniform distribution that ranged from 0.9 to 1.1 times the tabulated average for the period. This spread corresponds approximately to the standard deviation of such mean values.

Scrub Liquor Apportionment. Apportionment of the uranium in scrub liquor to the different scrubbers was based on very limited data on individual scrub liquor collections and upon equipment capacities. Data on actual collection of uranium in scrub liquor (Table I-5) provides information on utilization of the furnaces at two times during the early years of operation. Capacity data were listed in Table I-2. The primary calciner and rotary kiln were physically larger and had greater capacities than the other furnaces employed in Plant 8. Stated furnace capacities greatly exceeded actual plant production, so they are only indicative. Average utilization fractions for the furnaces were estimated from information in Tables I-2 and I-5 and discussions with plant personnel.

For years prior to 1975, it was assumed that half of the scrub liquor uranium came from the primary calciner and that about 28% came from operation of the rotary kiln. Uniform distributions were assumed for both fractions, with ranges of 0.4–0.6 and 0.2–0.35, respectively. These fractions were selected independently. The sum of the four utilization fractions was constrained to be unity; thus, between 5 and 40 percent of the uranium remained to be assigned to the other furnaces. Fractions for the UAP furnace and oxidation furnaces were computed. The UAP furnace fraction was determined by multiplying the remainder by a parameter selected from a symmetrical triangular distribution with bounds of 0.55 and 0.85. The fraction for the oxidation furnaces was then calculated by difference. The resulting distributions for these two utilization fractions were symmetrical and triangular with ranges of 0.035–0.28 and 0.015–0.012, respectively.

Furnace utilization changed in the 1970s. In the last half of 1975, the special ore concentrate calcining in the rotary kiln was by far the largest amount of processing that was accomplished. In the first half of 1975, and in subsequent years through 1979, the rotary kiln fraction was taken to be an average of 0.9. This fraction was represented by a uniform distribution with values between 0.8 and 1.0. During this period, utilization fractions for the UAP furnace and oxidation furnaces were computed as described above. The resulting distributions of utilization factors were both symmetrical and triangular with ranges of 0–0.14 and 0–0.06 for the UAP furnace and oxidation furnaces, respectively.

For 1980 and 1981, data indicated that the box furnace, which used the same scrubber as the calciner, would account for 22% of the loading. For 1980, the fraction for the oxidation furnaces was determined by difference. All the scrub liquor uranium was distributed between these two scrubber categories. The distributions were both taken to be uniform, with ranges of 0.20–0.24 and 0.76–0.80, respectively. For 1981, the rotary kiln was estimated to account for 29% of the loading. Distributions of fractions for the box furnace and rotary kiln were sampled independently. Uniform distributions with ranges of 0.20–0.24 and 0.26–0.29, respectively, were used. The fraction for the oxidation furnaces was computed, with a resulting distribution that was uniform between 0.44 and 0.54.

• **Average Penetration Through the Scrubbers.** Table I–17 contains information about the distributions used to estimate values of the long-term average scrubber penetrations used in the Monte Carlo calculations. The basis for these distributions was the short-term measurements summarized previously (Tables I–10 through I–13). For each distribution the lower bound and upper bound are listed; for the triangular distributions the mode is also given.

In the last two columns of the table are the computed mean values and standard deviations of the penetrations derived from these distributions. These resultant values are those that one obtains by sampling the specified distributions of penetration. The mean resultant penetrations correspond to the average penetrations measured during 1961–1965. The resultant standard deviations are all larger that the estimates from the measurements. This is considered appropriate when contemplating the uncertainties involved in estimating penetrations in other years and for a range of operating conditions that occurred before and after the period 1961–1965.

In the lower part of the table are descriptions of the distributions used to estimate average penetration through the UAP furnace scrubber during the period when the bypass line was installed. A bypass fraction (BF) of 0.10 was taken to be the base case for the period September 1963–March 1966. For BF = 0.10, the average penetration is estimated to be 33%. Distributions used to reflect alternative values of the BF are also shown in the lower portion of the table; these were used in calculations performed to show the effect of varying the bypass fraction. For bypass fractions of 0.05 and 0.15, the long-term average penetrations of uranium through the UAP scrubber would be 29% and 37%, respectively.

Table I-17. Distributions of Long-Term Average Penetrations (%) Used to Estimate Uranium Releases from Plant 8 Scrubbers, 1953–1981

	Distribution Characteristics[a]				Resultant Values[b]	
Scrubber	Shape	LB	Mode	UB	Mean	Std. Dev.
Calciner	Triangular	0.2	2.5	17.5	6.7	4.0
Rotary kiln	Triangular	0.8	5.0	34	13.3	7.6
UAP furnace	Triangular	0.2	10	65	25	14
Oxidation furnaces	Uniform	15		95	55	23
UAP Furnace Scrubber Calculations, September 1963–March 1966[c]						
BF = 0.05	Triangular	0.2	12	75	29	16
BF = 0.10	Triangular	0.2	14	85	33	19
BF = 0.15	Triangular	0.2	16	95	37	21

[a] For most time periods; LB: lower bound; UB: upper bound.
[b] Mean values and standard deviations of scrubber penetrations derived from sampling the distribution.
[c] Three different values of the bypass fraction (BF) were considered; the central estimate was computed using the distribution for BF = 0.10.

Estimates of Plant 8 Scrubber Releases During 1982–1988

For the latter years of FMPC operation, release estimates were based upon the operating times for the various scrubbers and measurements that had been made of release rates during scrubber operation. Only the set of relatively recent release rate measurements (Table I–15) was used in the calculations of releases for this time period. The values in Table I–16, from the earlier high production period, are distinctly different from those obtained in years of low throughput.

Figures I–7 through I–9 contain the distributions of measured release rates for the rotary kiln scrubber, the box furnace scrubber, and the oxidation furnace scrubbers, respectively. There is no record of measurements of releases from the muffle furnace. In estimating releases, plant personnel assigned a release rate equal to the largest average value that was used by them for other Plant 8 furnaces. In recent years, the muffle furnace release rate was set equal to that for the rotary kiln. In calculations for this time period, the distribution of release rates for the rotary kiln were also applied to the muffle furnace.

The variations in the release rates are a primary source of uncertainty in release estimates for the latter years of plant operation. Figures I–7 through I–9 illustrate the relatively broad range of release rates measured in recent years. The operating times for the furnace operations were recorded by plant staff for the purpose of making release estimates. There is much less uncertainty associated with those data than with the release rate measurements.

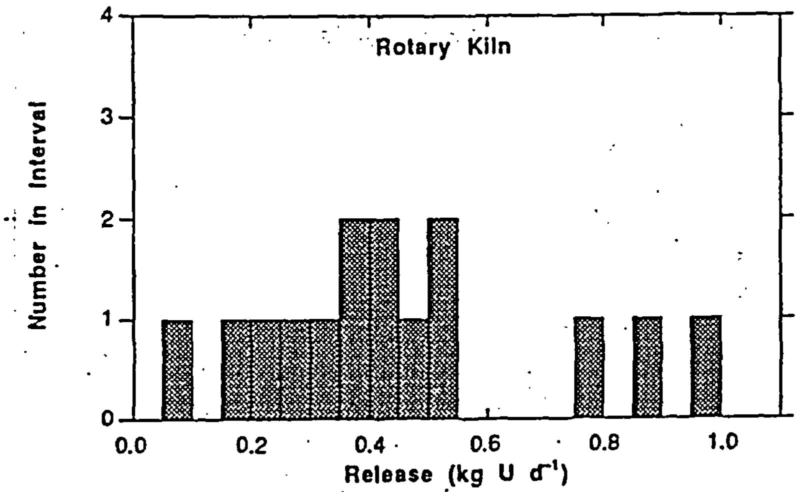

Figure I–7. Distribution of release rates from the scrubber serving the rotary kiln measured between 1970 and 1988. The distribution was used in calculations of releases during 1982–1988.

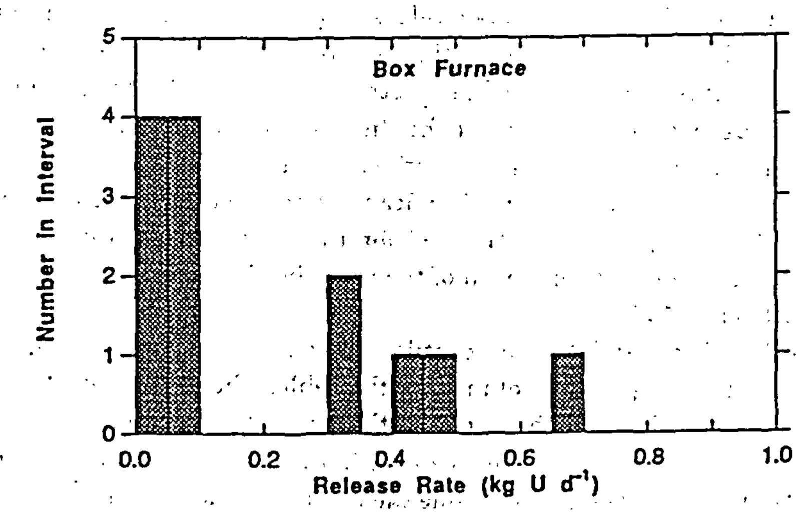

Figure I–8. Distribution of measured release rates from the scrubber serving the box furnace measured between 1979 and 1988. The distribution was used in calculations of releases during 1982–1988.

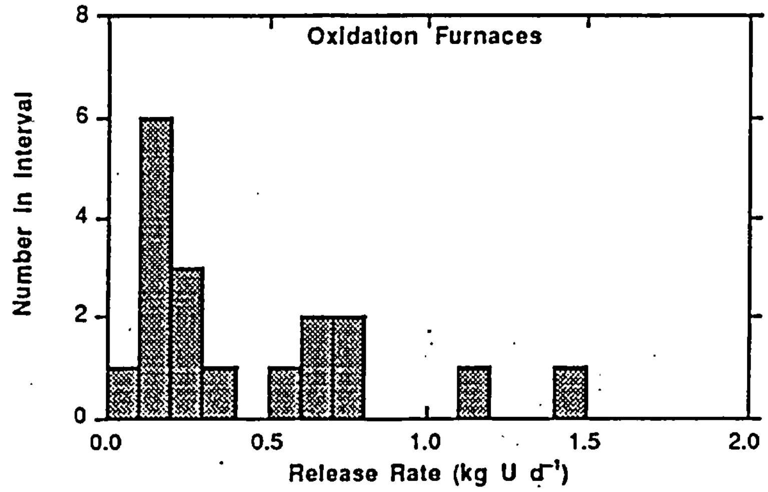

Figure I-9. Distribution of release rates from the scrubber serving the oxidation furnaces measured between 1981 and 1988. The distribution was used in calculations of releases during 1982–1988.

Results of Plant 8 Scrubber Release Calculations

Best estimates of Plant 8 scrubber releases are presented for each year in the period 1953–1988 in Table I-18. The best estimate value for a particular year is given in the second column and is the median of the distribution of estimates computed for that year. The median value lies in the center of the distribution of estimates for the year; half the estimates were higher than the median value and half were lower. These median estimates have been rounded to two significant figures. The largest release from the Plant 8 scrubbers, 10,000 kg U, was estimated for the year 1961. Estimated median annual releases exceeded 5,000 kg during each of the years between 1959 and 1963 and were nearly that large in 1958 and 1964.

The 5th, 25th, 75th, and 95th percentile values for each distribution, also rounded, are given in the third through sixth columns of the table. These percentile values indicate the spread in the distribution and the range of the estimates produced. There is a 50 percent chance that the yearly release lies between the 25th and 75th percentiles. There is only a 10 percent chance that the release lies outside the bounds defined by the 5th and 95th percentile results. Cumulative probability distributions for four years are presented as examples in Figure I-10. The vertical line marks the 50th percentile or median values reported as the best estimates. The central portions of these distributions are approximately linear, an indication that the distributions are approximately lognormal.

Table I-18. Annual Release Estimates for Plant 8 Scrubbers

Year	Best estimate of release (kg U)	Other percentiles in distribution of release estimates (kg U)			
		5th percentile	25th percentile	75th percentile	95th percentile
1953	100	13	43	210	570
1954	1,500	340	810	2,600	5,800
1955	2,800	660	1,600	4,900	11,000
1956	4,300	1,100	2,500	7,400	16,000
1957	3,400	820	1,900	5,900	13,000
1958	4,500	1,100	2,500	7,400	16,000
1959	6,700	2,400	4,400	10,000	21,000
1960	7,600	2,900	5,000	12,000	24,000
1961	10,000	4,500	7,100	15,000	29,000
1962	6,500	2,400	4,200	9,500	20,000
1963[a]	6,900	2,200	4,200	11,000	21,000
1964[a]	4,700	1,800	3,100	7,800	17,000
1965[a]	1,000	470	700	1,600	3,100
1966[a]	240	120	180	340	640
1967	570	290	430	830	1,500
1968	1,200	640	910	1,700	3,300
1969	270	140	200	380	700
1970	73	36	53	110	240
1971	62	27	43	96	230
1972	5	2	3	7	16
1973	5	2	4	8	17
1974	34	5	16	72	180
1975	330	100	200	510	800
1976	150	65	110	200	330
1977	460	130	280	700	1,200
1978	76	28	51	110	180
1979	280	63	150	460	900
1980	90	26	50	180	400
1981	120	35	68	250	530
1982	77	36	558	110	160
1983	180	79	130	260	430
1984	150	59	100	210	310
1985	110	44	68	170	290
1986	210	93	150	310	510
1987	120	51	84	180	310
1988	110	44	67	170	290

[a] In making these estimates it was assumed that the bypass for the UAP scrubber operated 10 percent of the time between September 1963 and March 1966.

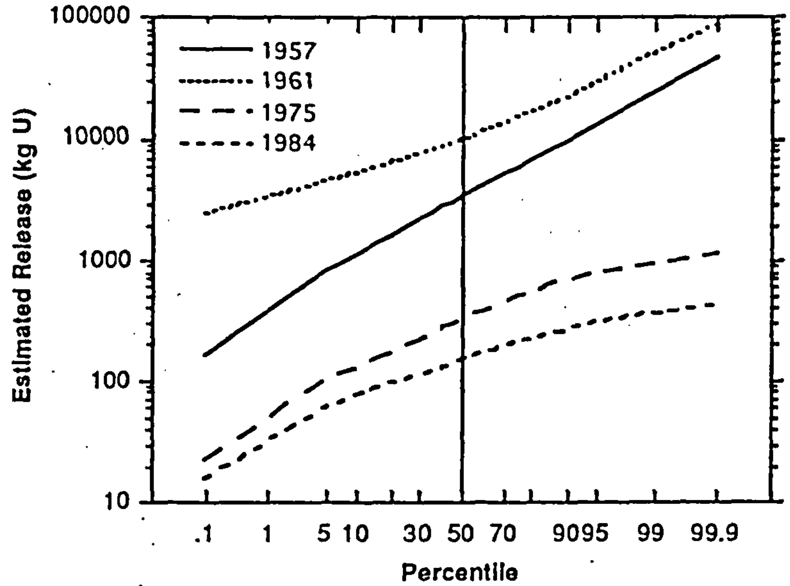

Figure I- 10. Probability distributions of estimated releases from Plant 8 scrubbers for 1957, 1961, 1975, and 1984.

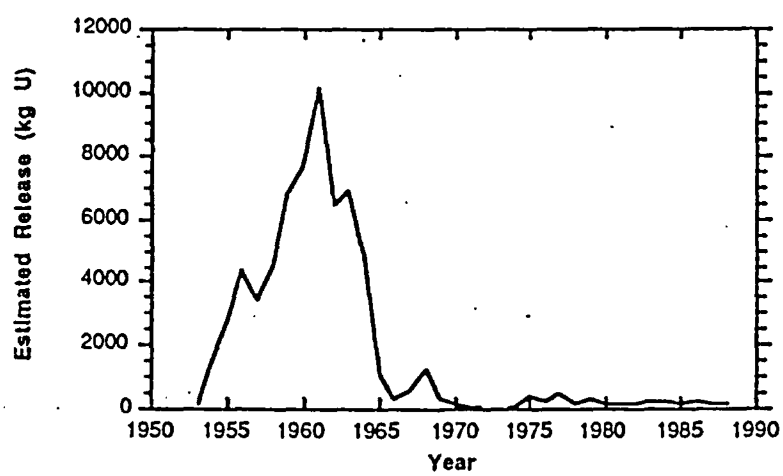

Figure I-11. Median estimates of release from Plant 8 scrubbers.

The tails of the distributions in Figure I-10 deviate from the slope of the line defined by the center of the distribution. As noted previously, the upper ends of the distributions are subject to distortion by the instability (for high values of penetration) of Eq. (I-7), which was used for most of the computations. The distribution of estimates for 1957 is broader and the slope of the line is greater than for the other distributions shown. Both facts indicate greater uncertainty in the estimate for that year. In contrast, the distribution for 1984 is relatively flat. Lower uncertainties in estimates for years in the 1980s is due to the fact that operating data were well known for those years.

The median estimates of the yearly releases from the Plant 8 scrubbers are shown in Figure I-11. The central estimates for the years 1963-1966 correspond to results in Table I-18 and reflect a bypass fraction (BF) of 0.1 for the UAP scrubber. Table I-19 compares the results for alternative values of BF equal to 0.05 and 0.15. The value of BF affects the average penetration for that scrubber directly, but has little effect on the median release estimates for all scrubbers. The effects on total releases were greatest in 1964 and 1965 when the bypass operated throughout the year.

Table I-19. Effect of Bypass of UAP Scrubber on Penetration and on Total Releases from Plant 8 Scrubbers

BF[a]	Mean UAP scrubber penetration	Median release (kg U) from all Plant 8 scrubbers			
		1963	1964	1965	1966
0.05	29	6500	4500	940	230
0.10	33	6900	4700	1000	240
0.15	37	7000	5200	1100	250

[a] BF: bypass fraction for UAP scrubber during September 1963-March 1966; increasing values raise the long-term average penetration through that scrubber (Col. 2).

Table I-20 contains summary release estimates by decade and for the entire period from 1953 through 1988. These estimates are also rounded to two significant figures. The releases for each decade were computed as part of the Monte Carlo procedure used for the annual release estimates. The distribution for the total for 1953-1988 was obtained by Monte Carlo calculations using the distributions of releases during each of the four decades. The shapes of the distributions of the sums, particularly the one for all years, approach that of a normal distribution.

The table illustrates the importance of the releases during the 1960s when plant production was highest (see Figure I-1). The median estimate for the 1950s was second highest, about 60% of that for the following full decade of operation. Since the early 1970s, when the plant was placed on a reduced operating schedule and subsequently shut down, its utilization was only periodic and releases have been much lower than they were during the early years of operation.

Table I-20. Summary Release Estimates for Plant 8 Scrubbers

Period	Best estimate of release (kg U)	Other percentiles in distribution of release estimates (kg U)			
		5th percentile	25th percentile	75th percentile	95th percentile
1950s	29,000	17,000	23,000	37,000	53,000
1960s[a]	47,000	30,000	39,000	57,000	78,000
1970s	1,700	1,000	1,400	2,100	2,700
1980s	1,400	980	1,200	1,600	2,000
1953–1988[a]	81,000	56,000	69,000	95,000	130,000

[a] In making these estimates it was assumed that the bypass for the UAP scrubber operated 10 percent of the time between September 1963 and April 1966.

The release estimates for the Plant 8 scrubbers that are summarized in the tables and graphs above are higher than previous FMPC estimates. The fundamental reason for the difference is that the present calculations consider ranges of individual scrubber performance that are broader than the single collection efficiency of 83 percent that had been assumed for all of the scrubbers. Sometimes the performance was better; at other times it was much worse than had previously been assumed. Inclusion of the variations in performance and of uncertainties in other parameters of the model has led to a relatively wide range of estimates for any particular year. Uncertainties for estimates of releases in the most recent years of operation are primarily associated with variations in the release rate factors for the individual scrubbers.

ALTERNATIVE CALCULATIONS OF RELEASES

Alternative calculations of releases from the Plant 8 scrubbers were performed to test the effect of different modeling choices on the results. The first of these involved the use of correlations between scrubber penetration and the accumulation of uranium in the scrub liquor. The second alternative approach was to derive release to production ratios for the early 1960s, when the scrubbers were studied most intensively, and to apply them to the entire period of operation. These two modeling approaches are described below and the results of the calculations are summarized.

- *Alternative 1. Correlations of the type shown in Eq. (I-5) were used as part of the procedure to estimate releases for the years 1953–1981. Alternatives that employed a single correlation for all scrubbers or separate correlations for each individual scrubber were considered. The results of calculations that employed such correlations were counter-intuitive. While estimated releases during years of high uranium recovery were both lower and higher than those presented above, releases during years when Plant 8 was virtually shut down were predicted to be substantial. This is shown in Figure I-12. The correlation cannot be applied to the many later years when uranium recovery operations had* *declined substantially. A second factor is that the correlation is logarithmic; at low rates of uranium accumulation in scrub liquor, there can be large variations in penetration estimates obtained from the correlation.*

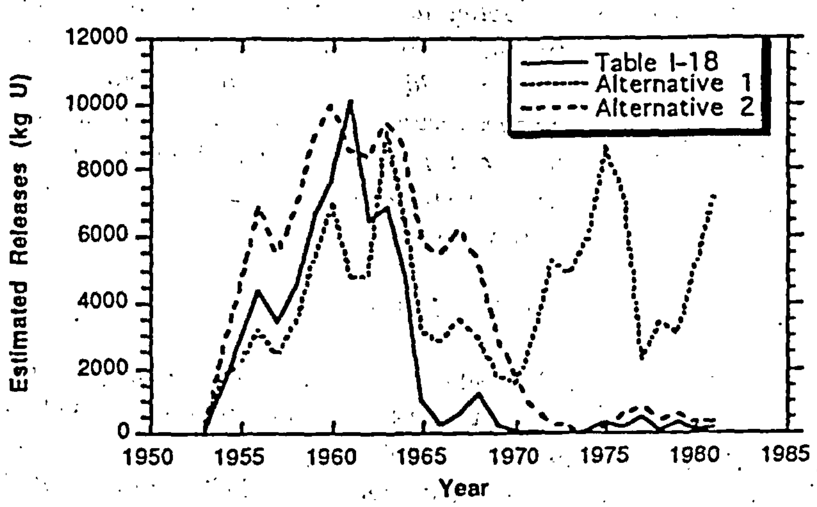

Figure I-12. Comparison of alternative median estimates of annual releases with those given in Table I–18. Penetration estimates for Alternative 1 employed a correlation between scrubber penetration and scrub liquor accumulation rate (page I–22). With this approach, predicted releases estimated for years when Plant 8 was virtually shut down were very high and lack credibility. Alternative 2 employed a normalized release rate derived for 1960–1963. The normalized release rate for a period of high production and many operational problems is not considered a good predictor during years of lower production.

- *Alternative 2. A normalized release rate based upon results for the early 1960s, when most of the scrubber efficiency measurements were made, was also used to estimate releases in other years. The normalized release rate was defined as the total scrubber release rate (kg U y^{-1}) divided by the Plant 8 production rate (MTU y^{-1}). Calculations of releases for the period 1961–1963, prior to installation of the bypass line for the UAP scrubber, were used to develop the normalized release rate. Calculations of the normalized release rates were performed as part of the main simulation for the 1960s and thus reflect the 3- and 6-month time intervals that were used during those three years to fit available scrub liquor data and to isolate the period of bypass installation. The median normalized release rate was 3.1 kg U MTU^{-1}. The geometric standard deviation of this parameter was about 1.6; extreme values were 0.71 and 16 kg U MTU^{-1}. Results of release calculations using this approach for the period 1953–1981 are also shown in Figure I–12.*

Predicted releases using this approach are generally higher than the values in Table I-18. However, for years before 1964, the median estimates obtained for Alternative 2 lie within the central half (25th–75th percentiles) of the base case estimates in Table I-18. The differences between the two approaches during 1961–1963 reflect the different bases for the calculations and the application of a normalized release rate derived over the period to individual years within it. During later years, the normalized release approach yields overpredictions because it does not take advantage of the known scrub liquor uranium collection data. The normalized release rate for a period of high throughput and many operational problems is not considered as good a predictor of releases during periods of lower production.

PHYSICAL CHARACTERISTICS OF THE DISCHARGES

Analysis of the Plant 8 scrubber releases suggests that two distinct types of particles were present in the emissions. The first type consisted of solid particles of U_3O_8 of less than 10 micrometers in diameter which penetrated the scrubber systems. The second type was droplets of entrained scrub liquor that contained suspended uranium particles.

No reports have been found of measurements of the sizes of the particles or liquid droplets released to the atmosphere from the Plant 8 scrubbers. Limited information on the distributions of particle sizes in furnace exhausts was used, together with scrubber performance data (Lund 1971) to estimate the distribution of particle sizes that would not be captured by the scrubber. The analysis is described in Appendix D of the Task 4 report (Killough et al. 1993). Table I-21 contains the calculated size distribution for these particles.

Table I-21. Calculated Size Distribution of U_3O_8 Particles Expected to Penetrate the Scrubbers

Diameter Range (μm)	Frequency (percent)
0–1	71.4
1–2	11.8
2–5	14.6
5–10	2.1

Using information on the expected collection efficiencies for the scrubbers, it was estimated that about 30% of the total uranium emitted from the Plant 8 scrubbers would have been small particles of U_3O_8 (Killough et al. 1993). A second analysis was performed, using the same approach with more complete compilations of scrubber inlet loading data (Table I-14) and scrubber penetration estimates (summarized in Table I-17). The results of the second stochastic calculations indicated a median penetrating particle fraction of 0.25. The distribution had a GSD of 1.5. The mean value would be about 0.27, not greatly different from the initial estimate.

The median fraction due to entrainment of scrub liquor was estimated in the second analysis was 75%. Based upon information in the literature, the scrub liquor droplets produced by entrainment were assumed to follow a lognormal distribution with a median diameter of 140 µm and a GSD of 1.4. Evaporation during downwind plume travel in the atmosphere would have reduced the original entrained droplets to hypothetical solid polycrystalline spheres with diameters about 46% of the diameters of the original spherical liquid droplets (Killough et al. 1993).

The solid polycrystalline spheres would consist of a mixture of various salts such as sodium chloride, sodium phosphate, sodium carbonate, and sodium uranates as well as sodium hydroxide. The densities of a typical liquid droplet and a typical polycrystalline solid particulate resulting from evaporation of the droplet were calculated to be approximately 1.1 and 2.0 g cm^{-3}, respectively (Killough et al. 1993).

CONCLUSIONS

Several high temperature and fume bearing exhausts in Plant 8 were treated using scrubbers that employed caustic solutions. Furnace exhausts likely to contain substantial concentrations of airborne particles were passed through ejector-venturi scrubbers. The scrubber releases were not measured routinely. However, special measurement efforts undertaken during the early 1960s and unexplained losses of uranium from the facility led to the conclusion that scrubber exhausts were an important source of routine operating losses.

Previous estimates of releases from these scrubber systems were reviewed. Plant records were found in storage that provided data on the amounts of uranium scrubbed from the airborne effluents during periods ranging from one month to one year. Plant 8 production (uranium recovery) data were compiled to indicate the changing scale of plant operations. Memos and analytical data sheets were located that described measurements of scrubber efficiencies performed in Plant 8, primarily during 1961–1965. These data were compiled for each scrubber for use in calculations of releases from 1953 through 1981. Data from measurements of release rates from the various stacks collected in later years were also compiled and used for calculations for the period 1982–1988. Measured releases during these years were much lower than those observed during years of high production.

For both time periods, simple models of releases were applied to individual scrubbers. The 6- to 12-month average ratio of the amount of uranium collected in scrub liquor to plant production was found to be a reasonable link between production data and scrubber operations when information on scrub liquor collections was not available.

Monte Carlo calculations were performed to estimate uranium releases from the Plant 8 scrubbers. The ranges of all of the parameters used in calculations were relatively broad, owing both to variability and to limited historic data. The results of the calculations reflect these uncertainties. The 90-percent confidence interval for the release during a particular year is relatively wide, typically from a factor of about 2–3 lower to a factor of about 2–3 higher than the median.

Overall, it is estimated that the Plant 8 scrubbers released about 81,000 kg of uranium; the 90-percent confidence interval for this estimate is 56,000 to 130,000 kg of uranium. The decade of highest releases was the 1960s when production peaked. High releases were also estimated for the 1950s. Estimated releases during the other two decades were very much lower.

During the first two decades, when releases were highest, it is estimated that about 25% of the releases were of small particles of U_3O_8 and that the remainder were the result of entrainment of contaminated scrub liquor containing suspensions of uranium compounds. The importance of the latter process during this period is supported by the actual data on scrubber penetration and comparison of the measured performance with that expected for the scrubbers.

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APPENDIX J

EMISSIONS OF RADON, RADON DAUGHTERS, AND GAMMA RADIATION FROM THE K-65 SILOS

INTRODUCTION

This appendix provides detailed descriptions of the current estimates of emissions of ^{222}Rn and Rn daughters from the FMPC, and detailed information related to emissions of gamma radiation from the FMPC. These releases were primarily from the K-65 Silos and from other stored K-65 material. First, however, we present characteristics of and general information about the K-65 Silos and material (page J-2). Since the current estimates of releases utilize the same general methods for air exchange and diffusion releases as the previous estimates, we next describe the previous estimates (page J-12).

Then we discuss the current estimates of releases of ^{222}Rn and daughters that have been developed in the Fernald Dosimetry Reconstruction Project, including the models used, the distributions chosen to represent parameter uncertainties, the mechanics of the calculations, and the results (page J-16). Most of the effort on Rn releases has been toward estimates of releases from the K-65 Silos for the periods 1959–1979 and 1980–1987. These two periods encompass most of the time under consideration in this Project. And, it appears that the relative impacts of Rn releases, compared to releases of uranium to air, will be highest in the 1970s and 1980s. Releases of Rn in the early 1950s from drummed K-65 material, stored at the FMPC, may also be relatively significant, compared to uranium releases, because operations at the site were just beginning in this early period. The discussion about our current estimates of ^{222}Rn and Rn daughter releases, in this Appendix, includes the following subsections:

We also discuss an alternative calculation, performed to corroborate current estimates of releases (page J–73). The last sections of the Appendix describe data that will be used later in the Project, for transport and exposure calculations: (1) parameters to be used for application of the building wake effects model to the K-65 Silos and associated berms (page J–85), and (2) parameters to be used for the assessment of direct exposures to gamma radiation emitted from the Silos (page J–86). The section on parameters for direct exposures to gamma radiation includes detailed information about the radiation sources. Calculations of exposure rates at various distances from the Silos will be discussed in the report of Task 6 of this Project. Following the reference list (page J–97) are two annexes. The first presents a detailed data table, used in our analyses (page J–104). The second provides a summary of the general approaches to the calculations, equations used, and parameters used for the current estimates of ^{222}Rn and Rn daughter releases (page J–105).

In the period of consideration, 1951–1988, we have identified one potential episodic Rn release from the K-65 Silos. This release, which occurred April 25, 1986, is discussed in Appendix K.

Note that in this Appendix, when "Rn" or "radon" is used, we refer to ^{222}Rn.

CHARACTERISTICS OF K-65 AND METAL OXIDE SILOS AND MATERIALS

For the current estimates of ^{222}Rn releases from the FMPC, the K-65 Silos (Silos 1 and 2) and drummed K-65 material stored on the site are considered, for reasons discussed later in this Appendix (see page J–17), to be the only significant sources of Rn releases. The previous estimates of ^{222}Rn source terms from the FMPC considered the K-65 Silos to be the only sources. The current estimates of direct exposures to people outside the FMPC boundary from radioactive materials on the site consider the K-65 Silos and the Metal Oxide Silo, Silo 3, to be the only significant sources of gamma radiation (see page J–87). Thus, the characterization information in this section focuses primarily on the K-65 and Metal Oxide Silos.

Facility Description

Four large concrete storage tanks, called silos, are located in the waste disposal area of the FMPC, as shown in Figure J–1 below. These silos are in a north-south line, and are about 1000 ft west of the production area. The silos are numbered one to four, with the southernmost silo being Silo 1. Silos 1 and 2 contain K-65 waste raffinate material from the extraction processing of uranium ore, and are thus referred to as the K-65 Silos (DOE 1990). The K-65 material was slurried from the refinery (Plant 2/3) through pipes into the Silos, and decanted with the use of baffles and weirs located along the height of the Silo walls. The K-65 material originated both from onsite ore processing and from processing at the Mallinckrodt Chemical Works in St. Louis. The ore processed was pitchblende from the Belgian Congo, having very high uranium (and thus uranium-chain radionuclides) concentrations. The K-65 material contains very high concentrations of ^{226}Ra (DOE 1990), which decays to form ^{222}Rn. The K-65 material has long been known as a significant source of ^{222}Rn (Strattman 1955; Boback 1979; and others).

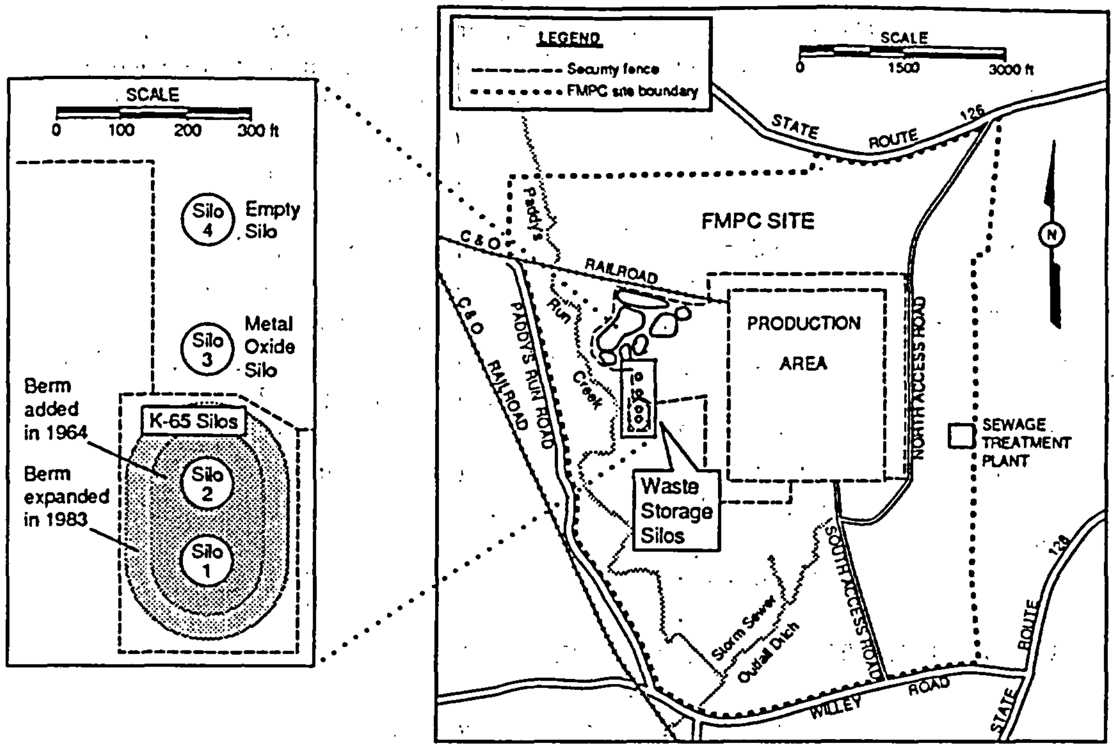

Figure J–1. Location of the waste storage silos on the west side of the FMPC site.

Silo 3, the Metal Oxide Silo, contains the metal oxide waste raffinate material from the extraction processing of uranium ores and concentrates. Unlike the K-65 material, the metal oxide material was dewatered and spray calcined to produce a dry, powder-like material (DOE 1990). The metal oxide material was conveyed from Plant 2/3 by high-pressure air, through pipes, to the Silo. All of the metal oxide material originated from onsite processing. The metal oxide material is also contaminated with radioactivity, but the concentration of ^{226}Ra is much lower than in the K-65 material. Silo 4 has never been used, and contains only a small amount of water with very low levels of radioactive and chemical contaminants (DOE 1990).

A set of large scale topographic drawings of the FMPC site (Woolpert circa 1988) shows the locations of the silos and also includes the Ohio State Plane (OSP) coordinate system. We scaled approximate coordinates for the centers of the silos from these drawings. These coordinates are shown in Table J–1.

As shown in Figure J–2, each silo is 80 ft in diameter with an overall height of 36 ft, of which about 26 ft 8 in is the tank wall and about 9 ft 4 in is the domed silo roof (Preload 1951a; Shanks and Vogel 1988). The walls are 8 inch thick concrete and the domes are nominally 4 inch thick concrete (Preload 1951a; Shanks and Vogel 1988). The total volume of each silo is about 160,000 ft^3 (4500 m^3). The K-65 Silos are roughly two thirds full (see later discussion, page J–29). In 1972 the Metal Oxide Silo was estimated to contain 150,000 ft^3 of material (Nelson 1972b). This is almost equal to the total volume, so the Metal Oxide Silo is considered full. Figure J–3 shows the Silos as they appeared in 1965 (DOE 1965a).

Table J–1. Approximate Coordinate Locations of the Waste Storage Silos

	OSP Coordinates (ft)	
Silo	East	North
Silo 1 (K-65)	1,378,484	480,400
Silo 2 (K-65)	1,378,486	480,522
Center between Silos 1 and 2	1,378,485	480,461
Silo 3 (Metal Oxide)	1,378,492	480,730
Silo 4	1,378,499	480,941

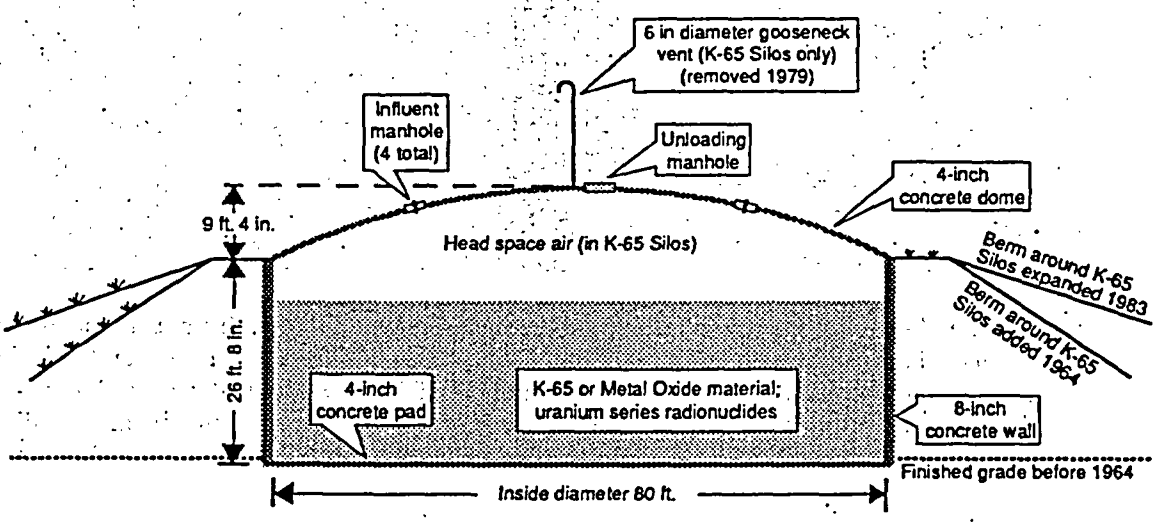

Figure J–2. General cross section of the K-65 and Metal Oxide Silos.

History of K-65 Disposal

The K-65 Silos were constructed in 1951 to 1952 for the temporary storage of K-65 materials (Catalytic circa 1950s(a); NLCO 1962; Grumski 1987a; Shanks and Vogel 1988). Originally the K-65 residues were to be returned to the African Metals Corporation, which provided the pitchblende ore, called Q-11, processed for its high uranium content. The K-65 residue is one particular waste product from the extraction of uranium from pitchblende ores, and contains high concentrations of ^{226}Ra, gold, and other metals.

The FMPC-2082 report indicated that K-65 materials were added to the silos from 1953 until 1955 (Boback et al. 1987). Additional documentation about the history of disposal of K-65 at the FMPC has been obtained. This information is summarized in Table J–2 and indicates K-65 materials were added to the silos from July 1952 through September 1958.

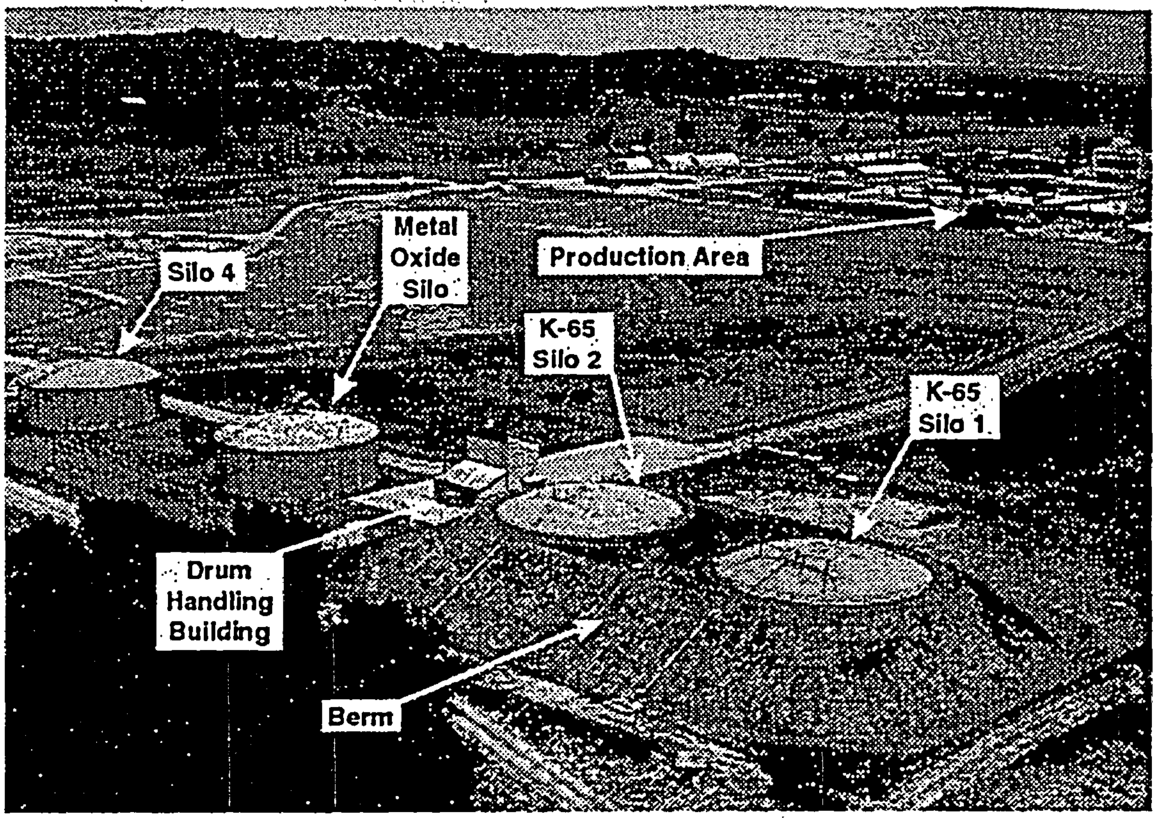

Figure J-3. Photograph of the waste storage silos in 1965, from the southwest (labels added to image digitized from DOE 1965a). The appearance of the Silos would have been similar to this for the period 1964 (after installation of berms) to 1979 (before removal of piping and sealing of penetrations). The drum handling building was removed in 1983 when the berms were enlarged (Geesner 1983).

History of Silo Structural Characteristics Affecting Rn Releases

The K-65 Silos have had problems of deterioration, almost since the time of construction. Significant cracking in the walls and seepage of the contents was noted from the 1950s (Wunder 1954; Martin 1957). Because of these problems, repairs and improvements to the Silos have occurred from the 1960s through the 1980s.

The project records for FMPC project 34-9 indicate that initial construction of the berms (at slope of 1½:1, horizontal to vertical) was complete by June 1, 1964 (NLCO 1984). In 1979, planning was in progress to close and seal all penetrations in the Silo domes (Heatherton 1979). An FMPC memorandum indicates that, on the Silo domes, the gooseneck vent pipe was removed, all openings were sealed, and metal covers were sealed with gaskets and bolted shut, in June 1979 (Boback 1980b). In the report of Task 5 of this Project, we analyzed Rn measurements taken at the boundary air monitoring station BS-6, west of the Silos (Shleien et al. 1993). A fairly abrupt decrease in Rn concentrations indicated that the sealing of these Silo dome penetrations probably occurred at the end of June, 1979. The

Table J–2. Disposal History of the K-65 Silos

Dates	Activities (reference)
August 1951–July 1952	Construction of the silos (Catalytic circa 1950s(a); NLCO 1962; Grumski 1987a; Shanks and Vogel 1988).
September 1951	Mallinckrodt Chemical Works (MCW), in St. Louis, began shipping drummed K-65 material to the FMPC (Blythe 1951; Walden 1952).
July 1952	Disposal of MCW-generated, drummed K-65 material, by slurrying to Silos, began (Davis 1952).
About June 1953	The south silo, Silo 1, was full, and storage of material in Silo 2 had begun (derived from Strattman 1953).
October 1955–January 1956	Campaign 1 of FMPC Q-11 processing and K-65 production and disposal (Lynch circa 1958).
November 1955	Drummed K-65 continued to arrive from MCW, and was still being added to silo (Madoffori 1955a; Madoffori 1955b; Madoffori 1955c).
August–October 1956	Campaign 2 of FMPC Q-11 processing and K-65 production and disposal (Lynch circa 1958).
March–April 1957	Campaign 3 of FMPC Q-11 processing and K-65 production and disposal (Lynch circa 1958).
May 1957	Campaign 4 of FMPC Q-11 processing and K-65 production and disposal (Lynch circa 1958).
September 1957	Campaign 5 of FMPC Q-11 processing and K-65 production and disposal (Lynch circa 1958).
December 1957	Campaign 6 of FMPC Q-11 processing and K-65 production and disposal (Lynch circa 1958).
March 1958	Campaign 7 of FMPC Q-11 processing and K-65 production and disposal (Lynch circa 1958).
June–September 1958	Campaign 8 of FMPC Q-11 processing and K-65 production and disposal (Lynch circa 1958).
September 1958	The north silo, Silo 2, was removed from service and decanted (Noyes 1958; NLCO 1962).

records for project 34-9 also show that the earthen berms were expanded to a slope of 3:1 (horizontal to vertical) in June 1983 (NLCO 1984). The significant changes to the Silos are summarized in Table J–3.

Characteristics of K-65 Material

Various characterization studies have been undertaken on the K-65 materials in the past. Currently, the K-65 Silos are included as Operable Unit 4 in the FMPC Remedial Investigation and Feasibility Study (RI/FS) of the Department of Energy. Table J–4 summarizes the results of these studies for some parameters pertinent to estimates of ^{222}Rn generation rates.

The draft Remedial Investigation Report (DOE 1990) indicates that the 1989 sampling was considered inadequate for characterization purposes, because sample cores had very low

Table J–3. Summary of Historical Changes to the K-65 Storage Silos

Date	Repairs or Improvements (reference)
May 1964	Cracks in silo walls were patched, waterproofing sealant applied, and earthen berm constructed to counterbalance material inside silos (NLCO 1984; Shanks and Vogel 1988; Noyes 1964).
end of June 1979	Openings in silo domes, including the gooseneck pipe and other penetrations, were sealed, with gaskets installed, to prevent Rn emissions (Boback 1980b; Shleien et al. 1993; Boback 1980a; Grumski 1987a).
June 1983	The earthen berms were enlarged to correct erosion problems (NLCO 1984; Grumski 1987a; Shanks and Vogel 1988).
Early 1986	Dome covers added to protect the center sections of the silo domes; neoprene membrane layer applied to part of Silo 2 (Grumski 1987a; Shanks and Vogel 1988).
November 1987	Radon Treatment System installed to treat displaced Rn during work on Silos (not continuously operated) (Grumski and Shanks 1988; Shanks and Vogel 1988).
December 1987	Rigid, polyurethane foam layer and urethane coating applied to exterior of silo dome surfaces to weatherproof the Silos (Grumski and Shanks 1988; Shanks and Vogel 1988).
November 1991	Addition of layer of bentonite on top of K-65 material in Silos (WEMCO 1992).

recoveries (fraction of the intended sample retained in the sampling device), mostly from 4 to 30%. Because of this, additional sampling was performed in 1991 by the ASI and IT team using a different sampling device. According to the field geologist for the 1991 sampling program, the 1989 sampling used a sampling device that vibrated in the vertical direction only, which allowed material to easily plug the sampler, resulting in the low recoveries (Jarvis 1992).

If the sampling device for the 1989 program was easily plugged, the material recovered may have been primarily from the upper layers of the silo contents. It is known that the material in the Silos is not homogeneous, as material was deposited in layers in the Silos, and the ^{226}Ra content of the K-65 material varied with time (and thus should vary with depth in the Silos). In describing the 1989 sampling episode, the draft Remedial Investigation Report (DOE 1990) stated: "Previous attempts to sample the silo contents were unsuccessful because a continuous, representative sample core could not be recovered for inspection and analysis. The variability and inconsistency of results from previous sampling efforts precludes [*sic*] the use of the data for fully characterizing the silo contents." We thus think that the samples from the 1989 sampling episode were not representative of random samples from the Silos (i.e. the locations may be biased toward the uppermost material in the Silos), and thus should not be used to estimate the average ^{226}Ra concentrations in the Silos (the averages are what we desire).

The 1991 program used an improved sampling device that vibrated in all directions, allowing better penetration of the soft K-65 material, without plugging (Jarvis 1992). The average recovery for the 1991 sampling was roughly 64% (Jarvis 1992). We conclude that

Table J–4. Summary of Some Characteristics of K-65 Material

Time of study	Concentration ^{226}Ra [a]	Moisture content (dry weight %)	Density (g cm^{-3})	Reference
1952	0.3 ppm	30[b]	1.179[c]	Earlier study reported by DOE (1990)
1972	0.28 and 0.36 ppm	65 and 65[d]	0.53–0.72[e]	^{226}Ra from Litz (1974) treatability study
1980	0.2 ppm	—	—	Earlier study reported by DOE (1990)
1988	0.13–0.21 ppm	—	—	Earlier work reported by DOE (1990)
1988	—	43 and 90[f]	—	Gill (1988)
1989	657–192,600 pCi g^{-1}; mean 110,000 pCi g^{-1}; GM 76,000 pCi g^{-1}; GSD 4.4.[g]	21.8–73.5[b]	—	RI/FS sampling effort (DOE 1990)
1991	134,900–890,700 pCi g^{-1}; mean 420,000 pCi g^{-1}; GM 380,000 pCi g^{-1}; GSD 1.6.[g]	—	—	RI/FS sampling effort (ASI/IT 1992)

[a] Concentrations are presented in the units given in the reference. For conversion between units for ^{226}Ra concentrations, 1 ppm = 0.989 µCi g^{-1}.

[b] The basis of the unit (%) was not given in the reference; we assume it to be dry weight.

[c] The reference did not state whether the value is dry or wet density. We assume dry density.

[d] Moisture contents in percent dry weight for these samples were calculated, in this present work, from values of moisture loss on drying (fraction of wet weight), obtained from a laboratory analytical data sheet (NLCO 1972). The calculated moisture contents agree with those calculated (here) from "as received" [wet] and dry U concentrations reported by Nelson (1972b).

[e] Dry bulk densities were calculated, in this present work, from values reported by Nelson (1972b), which we determined to be wet densities. Three values were reported by Nelson; a value of 60 lb ft^{-3} for both silos was attributed to Cotter Corporation, and values of 54.3 and 73.7 lb ft^{-3} for Silo 1 and Silo 2, respectively, were attributed to NLO.

[f] Moisture content in percent dry weight were calculated, in this present work, from values of moisture loss on drying (fraction of wet weight), from the reference.

[g] GM and GSD are the geometric mean and geometric standard deviation, calculated in this present work.

the results from the 1991 sampling more closely approximate random samples from the entire contents of the Silos, and are thus preferred for estimating the average ^{226}Ra concentrations in the Silos. (We note that in the final Remedial Investigation Report for the Silos (DOE 1993), ^{226}Ra concentrations from the two sampling programs are averaged together. Based on the above information, we do not agree with this approach.)

Table J–5 tabulates the radionuclide concentrations from this 1991 sampling. For the calculated means and standard deviations we ignored samples with "not detected" results. This could result in slight positive biases to the means, but should not be significant for our uses of the data. These results were obtained from an ASI/IT database (ASI/IT 1992), and are also in the final Remedial Investigation Report for the Silos (DOE 1993). Sample locations from this program are described by the zone and manhole from which the sample was obtained. Each zone refers to roughly one-third of the K-65 material in the Silo, with zone A the top third, zone B the middle third, and zone C the bottom third (Jarvis 1992). Sampling was performed through the four former influent manholes on top of each Silo, which are described by their direction from the center of the Silo (NE, SE, SW, and NW). In some cases more than one sample was obtained from a given zone of a given manhole, although in these cases information about the sample location within the zone is not available. From the information we have obtained, the 1991 sampling program did not include analyses of additional, pertinent parameters such as density, moisture content, Rn emanation fraction, and Rn diffusion coefficient. As seen in the range of ^{226}Ra concentrations found in the recent sampling of the silos (DOE 1990 and ASI/IT 1992), the K-65 material is not homogeneous. The color of the material also varied greatly (DOE 1990).

The range of moisture contents reported for the 1989 sampling is a summary of eight measurements (DOE 1990). Of the eight, five were 20–35%, one was roughly 50%, and two were between 70% and 75%. It seems likely that the two highest values were for saturated material. We note that the DOE report did not indicate the vertical location in the silos for the samples. In fact, because of the very low sample recoveries, the vertical locations can probably not be determined. Thus, no information about the vertical profile of moisture content in the K-65 material can be discerned from these recent data.

We note that the densities measured in the early studies seem anomalously low, when compared to typical values for uranium mill tailings or soils. The basis (wet versus dry bulk density) of the results for the 1952 study is not given by DOE (1990). The letter report by Nelson (1972b) does not indicate the method of determining the densities. Usually one would assume the values to be dry bulk densities. However, in the report (Nelson 1972b) the densities were used in a calculation of the total weight of U in the silos, as if they were "as received," or wet, densities. We have assumed they were wet densities, and the values presented in Table J–4 have been converted to dry densities, using the moisture content (65% dry weight) calculated from information on the related laboratory analytical data sheet (NLCO 1972). The calculated dry bulk densities of about 0.53 to 0.72 g cm^{-3} and the value of 1.179 g cm^{-3} (basis unknown) seem quite low, relative to a more typical value of 1.5 g cm^{-3} for uranium mill tailings or soils. However, no results of bulk density measurements were reported in the recent RI/FS sampling (DOE 1990; ASI/IT 1992).

No specific values were reported for the porosity of the K-65 material. However, the report of the 1989 sampling (DOE 1990) reports specific gravity for eight samples (of which two are composites) to be between 2.58 and 3.37, with mean 2.98 and standard deviation 0.29 (about 10%). Porosity can be calculated from bulk density and specific gravity. Using the range of densities reported in the early studies (assumed to be dry bulk densities) and the mean specific gravity, results in relatively high (compared to typical uranium mill tailings) nominal estimates of porosity from about 0.6 to 0.8.

Table J–5. Results of Radionuclide Analyses on K-65 Material Obtained by ASI/IT from 1991 Sampling[a]

Sample identification[b]			Radionuclide concentrations in K-65 material (pCi g^{-1})									
number	zone	location	^{227}Ac	^{210}Pb	^{210}Po	^{226}Ra	^{228}Th	^{230}Th	^{232}Th	^{234}U	$^{235,6}U$	^{238}U
						Silo 1						
099728	C	SE	6870	235,200	267,000	601,600	nd[c]	105,372	981	1548	57.4	861
099743	A	NE	5623	117,700	144,000	394,900	nd	59,274	1106	750	105	677
099870	A	SE	8486	126,800	296,000	367,600	nd	54,050	nd	1466	43.7	650
099885	A	NW	4320	77,860	154,000	306,800	nd	33,100	735	489	19.1	387
099909	B	NE	17,390	144,300	269,000	397,900	nd	64,400	661	875	39.4	719
099930	B	NW	10,700	191,300	237,000	680,900	2280	52,300	nd	1089	42.1	673
099939	C	NW	8118	235,900	273,000	510,400	nd	83,627	nd	936	31.5	564
099948	C	NW	6054	r[d]	232,000	r	835	r	835	508	nd	486
099966	B	SE	11,130	381,400	434,000	890,700	nd	75,370	982	721	29.2	680
099975	B	SE	7016	248,100	276,000	503,300	nd	50,917	702	608	22.0	649
100004	B	NE	9931	200,900	174,000	571,700	nd	54,521	nd	758	29.2	631
100025	C	NE	9012	183,600	230,000	520,600	nd	97,353	nd	696	90.0	717
100039	C	NE	5194	182,300	166,000	550,600	nd	99,494	nd	746	39.9	687
Mean for Silo 1[e]			8450	194,000	242,000	525,000	1560	69,100	857	861	45.7	645
Standard deviation[e]			3420	78,700	77,100	158,000	1020	22,900	169	330	26.5	116
						Silo 2						
099355	B	SE	5448	125,000	168,000	404,800	nd	93,399	nd	1945	48	943
099356	B	SE	3407	161,000	164,000	414,000	nd	95,892	1785	na[f]	na	na
099359	B	SE	7517	194,700	188,000	481,000	nd	90,495	nd	na	na	1925
099710	C	SE	8258	129,700	104,000	285,400	nd	43,600	nd	841	93.3	810
099721	C	SE	6722	76,210	692,000	219,700	nd,r	37,300	nd,r	1792	74.8	2299
099774[g]	C	NW	7357	179,500	93,400	252,100	nd	25,200	nd	783	35.6	608
099788[g]	C	NW	6210	121,700	57,900	191,600	nd	160,000	2140	852	98.5	857
099802	C	NW	5641	125,900	90,600	176,900	622	37,000	985	586	92.0	595
099811	A	NE	4474	58,160	55,300	134,900	798	20,500	nd	671	73.8	688
099831	B	NE	5649	74,650	132,000	179,500	nd	35,500	nd	1408	80.9	818
099846	C	NE	10,450	127,800	209,000	368,200	nd	74,200	983	1429	130	1265
099861	C	NE	9668	133,000	241,000	405,500	7360	99100	nd	1465	172	1356
Mean for Silo 2[e]			6730	123,000	193,000	299,000	2930	65,400	1470	1220	92.4	1140
Standard deviation[e]			2130	40,300	175,000	119,000	3840	30,500	583	500	37.3	572
						Silos 1 and 2 considered together						
Mean for Silos 1 and 2[e]			7660	160,000	220,000	417,000	2380	67,400	1080	1010	65.7	861
Standard deviation[e]			2980	71,600	131,000	179,000	2860	26,200	464	435	38.8	452

[a] Ref: ASI/IT 1992. Analyses were also performed for ^{231}Pa, ^{224}Ra, and ^{228}Ra. Since these radionuclides were not detected in any samples, we do not include them in this table.

[b] Zone A refers to the top one-third of a complete core (thus in the top one-third of the K-65 material), zone B to the middle one-third, and zone C to the bottom one-third. The locations are the manholes, by direction, through which the sample was obtained.

[c] "nd" means not detected. The ASI/IT table reported a less-than value, which we do not give here.

[d] "r" means the data validation code, in the ASI/IT data table, indicates the analysis result was rejected (though a value was given by ASI/IT, we do not present it or use it here).

[e] For our calculations of the mean and standard deviation, we ignored samples with "not detected" results.

[f] "na" means no analysis result was reported by ASI/IT.

[g] Samples 099774 and 099788 were field duplicates. We averaged the results before calculating means and standard deviations.

Information searches and discussions with the RI/FS Operable Unit 4 staff at the FMPC have indicated that measurements of ^{222}Rn emanation fraction from and diffusion coefficient in the K-65 material have not been performed (as of September 1992).

Characteristics Of Metal Oxide Material

The metal oxide material of Silo 3 has been characterized by a few studies in the past. Currently, the Metal Oxide Silo is included as part of Operable Unit 4 in the FMPC RI/FS of the Department of Energy. The 1989 RI/FS sampling included extensive sampling of the metal oxide material (DOE 1990). The sample core recoveries for this program ranged from 28% to 35%. Because all of the material in Silo 3 was produced at the FMPC in the same waste stream, and was dried and conveyed by air into the Silo, the material was expected to be relatively homogeneous. The Remedial Investigation Report (DOE 1990) concludes that the samples obtained should be adequate to characterize the material. The 1989 sampling also recovered many more samples than previous efforts.

The Remedial Investigation Report (DOE 1990) reports the radionuclide concentrations in samples from Silo 3, from the 1989 sampling effort, and those concentrations are tabulated below in Table J–6. For the calculated means and standard deviations of concentrations we ignored samples with "not detected" results. This could result in slight positive biases to the means, but should not be significant relative to our uses of the data. Sampling was performed through three of the four former influent manholes (on top of the Silo), located to the northeast, southeast, and northwest of the Silo center. The Remedial Investigation Report does not identify the location for the individual samples.

Table J–6. Radionuclide Analyses on Metal Oxide Material from 1989 Sampling of Silo 3[a]

	Radionuclide concentrations in metal oxide material (pCi g-1)											
number	^{227}Ac	^{231}Pa	^{228}Th	^{230}Th	^{232}Th	^{224}Ra	^{226}Ra	^{228}Ra	^{210}Pb	^{234}U	$^{235,6}U$	^{238}U
21	523	521	907	41,911	1451	453	2589	525	2437	1934	152	2043
22	416	401	nd[b]	33,881	nd	451	2192	559	2221	1618	117	1649
23	234	266	554	21,010	815	64	467	82	454	348	nd	320
24	1363	na[b]	nd	71,650	911	213	6435	nd	6427	1524	127	1600
25	534	556	459	40,968	411	295	3073	392	2493	1467	54	1392
26	706	889	859	41,555	nd	335	1862	441	1910	1910	76	1860
27	421	458	nd	53,227	nd	370	1518	325	1084	1317	80	1243
28	412	na	996	63,649	755	106	3702	nd	2589	1052	42	994
29	443	564	537	61,190	672	137	4169	117	3553	1843	158	1951
30	773	931	nd	68,759	581	449	2240	360	1942	1643	75	1574
33	566	431	949	65,488	672	313	4451	415	3674	1600	118	1878
mean[c]	581	557	752	51,200	784	290	2970	357	2620	1480	99.9	1500
stdev[c]	298	220	226	16,400	309	142	1650	164	1570	456	40.1	503

[a] Ref: DOE 1990.

[b] "nd" means not detected. "na" means not analyzed for this radionuclide.

[c] For our calculations of the mean and standard deviation (stdev), we ignored samples with "not detected" results.

Information about moisture content in the metal oxide materials was obtained in the 1989 sampling program (DOE 1990) and in a study conducted in 1972 (Nelson 1972b). In the

1989 program, five samples, including one composite sample, were analyzed for moisture content. Results ranged from 3.7% to 10.2%, with a mean for the four individual samples of 6.9%. Though the basis of the units is not stated by DOE (1990), we assume the results are in dry weight percent. The samples were obtained from the top one-third and bottom one-third of the material in the Silo, so they are probably fairly representative of all of the material in the Silo. For the 1972 study, Nelson (1972b) reports that for the sample that was analyzed for U concentration "Material was free flowing — drying was not needed." We interpret this statement to indicate that the moisture content was very low, which corroborates the low values seen in the 1989 program.

The only source of bulk density information we have located is the letter report by Nelson (1972b) of the 1972 study. Nelson reports two densities, a "free flowing" density and a density for material "tapped to maximum density." As the metal oxide was carried into the Silo by air, we think the free flowing density is likely to be more representative of the *in situ* bulk density. The measured densities were 40.02 lb ft^{-3} (or 0.64 g m^{-3}) for the free flowing density and 63.68 lb ft^{-3} (or 1.02 g m^{-3}) for the maximum density.

The report of the 1989 sampling (DOE 1990) reports specific gravity for five samples, including one composite sample, to be from 2.08 to 2.75. For the four individual samples, the mean was 2.32 and the standard deviation was 0.21. Porosity can be calculated from specific gravity and bulk density.

EVALUATION OF PREVIOUS ESTIMATES OF RADON RELEASES FROM K-65 SILOS

The source term for ^{222}Rn emissions from the K-65 storage silos was previously evaluated by IT Corporation, in their assessment of doses from historical releases from the FMPC (IT 1989). This assessment by IT Corporation did not include original calculations; rather it summarized and revised calculations from two other sources. Two pathways for emissions of Rn from the silos were considered: (1) diffusion of Rn from the K-65 residue into the silo air space and subsequent diffusion through the concrete domes into the surrounding air, and (2) free air exchange between the silo air and surrounding air, through cracks in the domes. The estimate of diffusion emissions was taken directly from the calculations of Borak (1985). It is noted that the FMPC-2082 report estimated the Rn emissions from only the diffusion pathway, and incorporated the Borak report as its Appendix A (Boback et al. 1987). In the IT report, the estimate of air exchange emissions was taken from a WMCO feasibility investigation report, with minor modification (Grumski 1987a; IT 1989). Detailed descriptions of these previous assessments of Rn releases from the K-65 Silos follow.

Diffusion Releases

The calculations by Borak (1985) of diffusion releases of ^{222}Rn were based on one-dimensional steady-state diffusion equations obtained from a National Bureau of Standards (NBS) summary technical report (Collé et al. 1981). The concentration of ^{222}Rn in the silo air space was first calculated from characteristics of the K-65 waste material and dimensions of the silos:

$$C_a = \frac{\phi}{\lambda_{Rn}}\left(\frac{\varepsilon_w l_w}{\varepsilon_w l_w + h}\right) \quad \text{(J–1)}$$

where:

C_a = concentration of ^{222}Rn in the silo air,

ϕ = production source term of ^{222}Rn in pores of K-65 material,

λ_{Rn} = decay constant of ^{222}Rn,

ε_w = total porosity of the K-65 waste material,

l_w = diffusion length of ^{222}Rn in the K-65 waste material, and

h = height of the air space in the silos above the waste material.

This equation for the Rn concentration applies to a closed container over the Rn source, with no losses other than radioactive decay (Collé et al. 1981). Since there are releases from the silo air space, these conditions are not met for the K-65 Silos.

The production source term of ^{222}Rn in pore spaces was determined by:

$$\phi = \frac{[Ra] EF \rho_w \lambda_{Rn}}{\varepsilon_w} \quad \text{(J–2)}$$

where:

[Ra] = concentration of ^{226}Ra in K-65 waste material (activity per mass),

EF = emanation fraction of ^{222}Rn production in K-65 material, and

ρ_w = bulk density of K-65 waste material.

From the concentration of ^{222}Rn in the silo air, the flux of ^{222}Rn diffusion through the concrete dome was calculated by:

$$J = \frac{\varepsilon_c \lambda_{Rn} l_c C_a}{\sinh\left(\frac{L}{l_c}\right)} \quad \text{(J–3)}$$

where

J = ^{222}Rn flux from the dome surfaces to the surrounding air (pCi m^{-2} s^{-1}, or similar units),

ε_c = total porosity of the dome concrete,

l_c = diffusion length of ^{222}Rn in the dome concrete, and

L = thickness of the dome concrete.

The total release rate is then the product of the ^{222}Rn flux and the surface area of the domes. It was assumed that the domes approximate circles of 40 ft radii, and thus the surface area of each dome is about 5030 ft^2, or 467 m^2.

The parameter values used by Borak are given in Table J–7. However, we note that the sources of these values were not documented (Borak 1985). The results of these diffusion release calculations were a ^{222}Rn concentration in the silo air of 3×10^7 pCi L^{-1}, and a total release rate to the atmosphere of 60 Ci y^{-1} (Borak 1985).

Free air exchange

As part of the investigations of the K-65 silos for controlling ^{222}Rn emissions, the FMPC performed temperature and pressure monitoring of the silos. Measurements of temperature at two depths into the silo air space, on the surface of the concrete domes, and in ambient air near the silos, and measurements of differential pressure between the silo air space and the atmosphere were obtained from March 13 to May 15, 1987 (Grumski 1987a). Due to instrument problems, much of the data was not usable. However, usable data were obtained for 11 complete days, including a three-day period, May 8 to 11, during which the daily increases in temperature were large (Grumski 1987a; Shanks 1991).

Table J-7. Parameter Values Used in Previous Assessment of Diffusion Releases[a]

Parameter	Units	Value used
λ_{Rn}	s^{-1}	2.1×10^{-6}
ε_w		0.3
l_w	cm	150
h	cm	300
[Ra]	pCi g^{-1}	2×10^5
EF		0.2
ρ_w	g cm^{-3}	1.6
ε_c		0.3
l_c	cm	12
L	cm	10

[a] Ref: Borak 1985.

For this three-day period, the internal gas temperatures for both silos showed a maximum daily increase of about 35 °F. Using the Ideal Gas Law, it was estimated that a closed tank of air initially at a pressure of 2117 PSF (pounds ft^{-2}) (or 14.7 psi) and temperature of 63 °F would undergo an internal pressure increase of about 142 PSF if the internal temperature was increased 35 °F (Grumski 1987a). For this monitoring period, the pressure monitoring indicated that Silo 2 held a maximum positive differential pressure of 7.6 PSF and a maximum negative pressure of 4.9 PSF. The maximum differential pressure was about 5% of what would be expected for a sealed system. Silo 1 showed negligible differential pressure with these temperature variations. It was concluded that the silos can not hold any significant pressure and thus that increases in the temperature of the internal silo air resulted in the volumetric expansion of the air and the release of "excess" volume to the atmosphere (Grumski 1987a).

The calculation of free air exchange emissions of ^{222}Rn from the silos was based on the expansion of the silo gases with warming of the gases due to warming of exterior air. The Ideal Gas Law was used to calculate the volume of air that would be emitted from the silos (Grumski 1987a):

$$PV = nRT \tag{J–4}$$

where

P = pressure of the gases within the silo,

V = volume of the silo air space (not including pore spaces of the K-65 material),

n = number of atoms of the gases,

R = ideal gas constant, with appropriate units, and

T = temperature, in units of an absolute scale (K or °R).

The calculations (Grumski 1987a) assumed that the internal gas pressure does not change. If this is the case, the volume of a given quantity of gas will be directly proportional to the temperature of the gas. Thus, the change in volume for a temperature change was calculated as:

$$\Delta V = (\Delta T / T_0) V_0 \tag{J–5}$$

where

ΔV = change in air space volume per day,

ΔT = change in temperature per day,

T_0 = the initial temperature, and

V_0 = the initial air space volume.

It was then assumed that, in the case of rising temperatures during a typical day, the complete increase in volume, ΔV, is released from the silo to the surrounding air. In this case the ^{222}Rn released, Q, is simply calculated as:

$$Q = C_a \Delta V \tag{J–6}$$

For this calculation, the ^{222}Rn concentration used was the value of 3×10^7 pCi L^{-1} calculated earlier by Borak (Grumski 1987a; IT 1989). The value used for ΔV was 1000 ft^3 d^{-1}, based on a value of ΔT of 20°F d^{-1}. The first calculations in the feasibility study used a silo air volume of 25,000 ft^3 (Grumski 1987a). This resulted in a calculated release rate of 600 Ci y^{-1}, for the two silos combined. The sources of the parameters used were not documented (Grumski 1987a).

However, the silo volume was revised to 43,758 ft^3 in the IT dose assessment (IT 1989). This increased the estimated release rate to 512 Ci y^{-1} for each silo, or a total of 1023 Ci y^{-1} (IT 1989).

CURRENT ESTIMATES OF RADON AND RADON DAUGHTER RELEASES FROM K-65 SILOS AND DRUMMED K-65 MATERIAL

In this section we first describe the sources of Rn releases at the FMPC for which we calculate releases, and the less important sources for which releases are not calculated. We discuss the general methodology used in current estimates of ^{222}Rn releases from the K-65 Silos, followed by a justification of the specific approaches to calculating releases from the Silos for different periods. The methods used to implement the calculations are reviewed. Then, in separate subsections for the different types of releases, we thoroughly discuss the models (equations) used for the calculations, the distributions chosen to represent the uncertainty of the parameters, and the calculation results. A summary of the predicted releases concludes this part of the Appendix.

Sources of ^{222}Rn Releases at the FMPC

The apparent source of the majority of the ^{222}Rn and Rn daughter releases from the FMPC is the K-65 Silos, in the waste storage area of the site. However, there are other potential sources that must at least be considered. In this section we briefly discuss the reasons for including some sources in our calculations, and for considering other sources to be negligible.

As discussed in Appendix B, most of the uranium received at the FMPC had been separated from its naturally occurring daughter radionuclides, including ^{226}Ra. The primary source of ^{226}Ra, and thus ^{222}Rn emissions, is the uranium ore received and processed in the early years of operation. The majority of this ore was from the African Metals Corporation (Afrimet), and the agreement with Afrimet stipulated that ^{226}Ra from the ores was to be retained for eventual return to Afrimet (Consiglio 1952; DOE 1990). Thus, the waste material from the uranium extraction processing of these ores was retained. The wastes were of two separate forms, the K-65 material and the metal oxides, and were stored in the two K-65 Silos and in the Metal Oxide Silo. Because of this storage, large quantities of ^{226}Ra are not expected to exist in other areas of the FMPC site, such as the waste pits. Other areas may have received small quantities of ^{226}Ra, both unrecovered radium from the ore processing and radium as a contaminant in other feed materials.

The two K-65 Silos and the Metal Oxide Silo have all received wastes from the processing of uranium ores, and thus contain significant quantities of ^{226}Ra. Thus, all three Silos are considered potential sources of ^{222}Rn releases. Recent sampling of the K-65 Silos and the Metal Oxide Silo has measured the concentrations of ^{226}Ra in the K-65 and Metal Oxide materials (see page J–6). From the 1989 sampling, the average concentrations of ^{226}Ra were determined to be about 110,000 pCi g^{-1} in the K-65 Silos, and about 2900 pCi g^{-1} in the Metal Oxide Silo (DOE 1990). Earlier sampling of the K-65 Silos had indicated concentrations of ^{226}Ra averaging about 350,000 pCi g^{-1} (Litz 1974). From the 1991 sampling, the average ^{226}Ra concentration was about 420,000 pCi g^{-1}. Additional sampling was not performed for the Metal Oxide Silo. From these measurements, the concentration of ^{226}Ra appears to be at least 40 times higher in the K-65 Silos than in the Metal Oxide Silo. Thus, a rough estimate is that the K-65 Silos have the potential for generating about 40 times more ^{222}Rn (per silo) than the Metal Oxide Silo. Thus, for this assessment, the Metal Oxide Silo, Silo 3, is considered an insignificant contributor to the ^{222}Rn releases. Silo 4 has never been used, and contains only a small amount of water with very low levels of radioactive contaminants (DOE 1990). Since Silo 4 is essentially empty, it is not considered a source of Rn releases. Thus, for this assessment, of the four waste storage silos, the K-65 Silos, Silos 1 and 2, will be considered the only significant sources of Rn releases.

As discussed earlier (see Table J–2), the K-65 Silos were completed in July 1952. However, K-65 material had been shipped to the FMPC from the Mallinckrodt Chemical Works (MCW) in St. Louis, starting in September 1951, and quite a large inventory of drummed K-65 material, about equal to half the capacity of one Silo, had been accumulated at the FMPC prior to operation of the Silos (Walden 1952). This drummed K-65 material was stored on the Plant 1 storage pad (Belmore 1951). Prior to operation of the K-65 Silos, the drummed K-65 material stored on the Plant 1 pad was apparently the only potential source of Rn releases from the FMPC. In this time period we expect that uranium releases from the site would have been quite low, since much of the site was still under construction. Thus, for this time period, Rn releases from the drummed K-65 material are likely to have been relatively significant, compared to other releases. We thus calculate releases for this stored, drummed K-65 material (see page J–55).

The majority of the K-65 material placed into the K-65 Silos was the material shipped to the FMPC from MCW (Lynch circa 1958). An operating manual for the K-65 area (Dougherty and Jennings circa 1951) indicates that the drums of K-65 material were opened and dumped, by inverting the opened drum, into a slurry tank, for makeup of a slurry for slurrying into the Silos. The dumping of the drums occurred in the drum handling building, which was located in the waste storage area generally between the Metal Oxide Silo and Silo 2. The dumping process is a source of Rn releases. A simple calculation can be performed to estimate an upper bound on the quantity of Rn that might have been released during these dumping operations.

An upper bound on the Rn released during dumping of the drums would be the quantity of Rn present in the pore spaces of the K-65 material being dumped. It is not likely that all the pore space Rn would have been released, because the moisture in the material would hold some of the Rn, and the method of dumping would presumably have exposed to the air only a small fraction of the surface area of the particles of material. The quantity of Rn in pore spaces can be calculated as the ^{226}Ra concentration (we assume the total Rn present is in equilibrium) multiplied by the quantity of material, the material density, and the Rn emanation fraction. The filling of Silo 1 was completed much faster than the filling of Silo 2, and consequently would have had a greater drum dumping rate, so we look first at Silo 1. Later in this Appendix, in our calculations of the volume of the silo air space, the depth of material in Silo 1 is estimated to be about 20 ft (see page J–29). Given the 80-ft diameter of the Silo, this represents a material volume of 100,500 ft^3.

From the characterization data given in Table J–4, the material density is in the range 0.53 to 1.179 g cm^{-3}. For this rough calculation, we assume a density of 0.85 g cm^{-3}. From Table J–5, the average ^{226}Ra concentration in Silo 1 is about 525,000 pCi g^{-1}. Later in this Appendix, we perform an alternative calculation of Rn releases from the K-65 Silos. In that calculation, we conclude that the Rn emanation fraction of the K-65 material would be in the range 0.1 to 0.4 (see page J–76). For this calculation, we assume a value of 0.25. These values result in an estimated upper bound to the quantity of Rn released of 320 Ci. The filling of Silo 1 took about 11 months (Table J–2), so this represents a rate of about 350 Ci y^{-1}. This upper bound is relatively small, compared with the estimated 90% probability range of the Silo Rn release rate for the operational period of Silo 1 (Table J–25, later in this Appendix), 200–4200 Ci y^{-1}. For the dumping of drummed material into Silo 2, the release rate from dumping operations would be even less, because the average ^{226}Ra concentration for Silo 2 material is less than for Silo 1 (Table J–5), and the time required to fill Silo 2 was about 5 years, rather than about one year. We thus consider the K-65 drum dumping operation to be an insignificant source of Rn releases from the FMPC, and no further calculations are performed for this.

Part of the K–65 material in Silo 2 was waste from the onsite (FMPC) processing of high-grade, pitchblende uranium ore from the Belgian Congo (DOE 1990). Since the ore contained ^{226}Ra, the ore processing, in the production area of the site, is another potential source of Rn releases. The processing occurred in the refinery (Plant 2/3), and included digestion of the ore in Nitric acid, followed by two solvent extraction steps (DOE 1990). We estimate an upper bound on Rn releases from this ore processing using the same method used for the drum dumping operations, described above. However, in this case, since the ore was digested, we assume that all of the Rn in the ore could have been released (i.e. not just the Rn in pore spaces).

A contemporary, handwritten spreadsheet (Lynch circa 1958) provides production and ^{226}Ra content information about the ores processed. Table J–8 compiles the applicable data.

Table J–8. Production Information on the FMPC Ore Processing [a]

Processing campaign[b]	Dates	Ore processed (tons)	^{226}Ra assay (mg ton^{-1})	^{226}Ra content (mg)
1	Oct 1955–Jan 1956	418.4	132.5	55,451
2	Aug–Oct 1956	607.5	141.9	86,215
3	Mar–Apr 1957	252.8	129.4	32,707
4	May 1957	124.7	156.7	19,590
4, Australian	May 1957	100.5	154.9	15,818
5	Sep 1957	204.6	133.4	27,547
6	Dec 1957	164.7	nv[c]	27,037
7	Mar 1958	110.8	nv	15,088
7, Australian	Mar 1958	50.8	nv	5,625
8	Jun–Aug 1958	389.1	nv	54,128
Total		2,423.9		339,206

[a] Ref. Lynch circa 1958.
[b] All campaigns were for Belgian Congo ores, except as noted.
[c] "nv" indicates no values were available.

From the data in Table J–8, the total ^{226}Ra content in the ores processed is 339,206 mg. The specific activity of ^{226}Ra is 0.989 Ci g^{-1} (Shleien 1992). Thus, the total activity of ^{226}Ra is 335 Ci. This is then also assumed to be the total quantity of ^{222}Rn that might have been released during the ore processing. The processing occurred from October 1955 through August 1958, a period of 34 months. Thus, the average rate of Rn release could have been up to about 120 Ci y^{-1}. This release rate is very small relative to the estimated 90% probability range of the Silo Rn release rate for the operational period of Silo 2 (Table J–25, later in this Appendix), 3100–7600 Ci y^{-1}. We thus consider the ore processing in Plant 2/3 to be an insignificant source of Rn releases from the FMPC, and no further calculations are performed for this.

Before ores were processed in the refinery, they may have been stored for a short period of time in the Q-11 silos located just south of Plant 1 (Consiglio 1952). The ores obtained from the African Metals Corporation were referred to as Q-11 material. There were six Q-11 silos, each consisting of a cylindrical component, 10 ft in height, and 13 ft in inside diameter; on top of a conical hopper, 12 ft in height (Consiglio 1952). The silos were elevated, with the top about 48 ft above the ground. Design capacity for the silos was 100 tons of ore. Consiglio (1952) also indicates that ore material was stored in the Q-11 silos after being crushed, pulverized, and dried to less than 2% moisture (we assume dry weight percent).

We perform a preliminary estimate of Rn releases from the stored ores, using the method used later in this Appendix to estimate releases from K-65 material stored in drums (see page J–55 for more details). We use the forms of equation J–50, equation J–51, and equation J–52. In this case it is not known if the Q-11 silos were open to the outside air. We assume they were, and thus assume that all Rn released from the ore material is released to the atmosphere. Thus, the ratio $[(\lambda_v + \lambda_d)/\lambda_{eff}]$ is assumed to be one.

From the data in Table J–8, the average ^{226}Ra concentration in the processed ore was about 150,000 pCi g^{-1}. We assume a normal distribution for this parameter, with an assumed relative standard deviation of 20%. Since the stored ore was very dry (2% moisture), the Rn emanation fraction would have been less than for wetter material (Rogers et al. 1984). As for the calculations for the drummed K-65 material, we use emanation fraction data compiled by Rogers et al. (1984) for uranium mill tailings. For material this dry, those data encompass a range of about 0.06–0.35, with a clustering around 0.15. We thus assume the emanation fraction has a triangular distribution, with minimum 0.06, maximum 0.35, and mode 0.15. For the specific gravity of the ore material we assume a range somewhat higher than that for K-65 material, since the ore would contain significantly more uranium. We assume a uniform distribution, with minimum 3.0 and maximum 3.5. Based on the design capacity of the silos (100 tons) and their volume, it seems that the expected material density was about 1.6 g cm^{-3} (remember this was pulverized ore material). We assume a uniform distribution, with minimum 1.4 and maximum 1.8 g cm^{-3}. With a 13 ft diameter, the surface area of ore in the silos would be about 123,000 cm^2. When the silos were full, the average thickness of material would be about 430 cm. From the information in Table J–8, it appears the average processing campaign consumed about 300 tons of ore, or the equivalent capacity of three of the Q-11 silos. We thus assume that, on the average, three silos were used. We further assume, based on the timing of the campaigns, that the silos were used for ore storage about half of the time.

A Monte Carlo simulation was employed for the calculations, using the same methods described later in this Appendix. Parameters not explicitly described above were as used for the calculation of releases from drummed K-65 material (see page J–55). The result is a 90% probability interval (5th to 95th percentiles) of 30–200 Ci y^{-1} released from the Q-11 silos during the ore processing period, October 1955 to August 1958. This release rate is very small relative to the estimated 90% probability range of the Silo Rn release rate for the operational period of Silo 2 (Table J–25, later in this Appendix), 3100–7600 Ci y^{-1}. We thus consider the ore storage in the Q-11 silos to be an insignificant source of Rn releases from the FMPC, and no further calculations are performed for this.

The annual FMPC environmental monitoring report for 1990 (Byrne et al. 1991) indicates that elevated concentrations of ^{226}Ra have been found in the waste pits, in the western area of the site. (We present information about these waste pits in Appendix K of this report.) The waste pits are considered as potential sources of Rn emissions.

A characterization of the waste pits was performed recently (Solow and Phoenix 1987). Solow and Phoenix (1987) describes measured concentrations of radionuclides in boreholes in the waste pits. Table J–9 summarizes results for ^{226}Ra concentrations measured the waste pits. Generally, the measurements were made on composite samples, each of which was formed from material from the complete depth of each borehole. In the case of the clearwell, samples were grab sediment samples, obtained with a dredging sampler.

Table J–9. Summary of Measured ^{226}Ra Concentration ($pCi\ g^{-1}$) in Waste Pit Contents (Solow and Phoenix 1987)

Waste pit	Number of samples[a]	Mean ^{226}Ra concentration	Standard error of the mean[b]
1	5	31	8.4
2	5	120	75
3	7	120	51
4	4	<15[c]	3.4
5	6	550	110
6	4	<22[d]	3.0
Burn pit	6	<2.7[c]	0.27
Clearwell	4	130	110

[a] All samples were composites, except those for the clearwell.
[b] For calculating the standard error, "less than" values were assumed equal to the value.
[c] One of the results was a "less than" value.
[d] All four results were "less than" values.

In Appendix K, the approximate volumes of the contents of the waste pits are provided. With the volumes and the average ^{226}Ra concentrations, the total ^{226}Ra content of the waste pits can be estimated. For this estimation, we assume the pit contents have an average bulk density of 1.6 g cm^{-3}, which is typical for soils. Table J–10 shows the parameters used and the results of these calculations. Also shown, from information in Appendix K, is whether the waste pit was operated as a wet pit, filled with slurried wastes, or as a dry pit, with solid wastes dumped from trucks.

Table J–10. Calculation of ^{226}Ra Quantity in Waste Pits

Waste pit	Type	Volume of contents ($yard^3$)	Concentration ^{226}Ra (pCi g^{-1})	Quantity ^{226}Ra (Ci)
1	dry	40,000	31	1.5
2	dry	13,000	120	1.9
3	wet[a]	227,000	120	34
4	dry	53,000	<15	<0.96
5	wet	102,500	550	69
6	dry	9,000	<22	<0.24
Burn pit	dry	unknown	<2.7	
Clearwell	wet	unknown	130	

[a] This pit was operated in a wet mode from 1959–1968, and in a dry mode for a short time, 1975–1977.

Two of the pits have unknown contents volumes. The quantity of ^{226}Ra in the burn pit is probably insignificant, because the average concentration is very-low. The quantity in the clearwell is estimated to be much less than that in pit 3, because the clearwell is a much smaller (areal extent) pit than pit 3 (see Appendix K), and the concentrations are similar. To summarize the calculations shown in Table J–10, the ^{226}Ra content in dry pits is around 5 Ci. And, the ^{226}Ra quantity in wet pits is probably only slightly greater than 100 Ci.

For comparison, we estimate the total ^{226}Ra content of the K-65 Silos. Later in this Appendix, we estimated the average thickness of the K-65 material in the Silos to be in the range 19.5–23.5 ft (see page J–74). Assuming an average thickness of 21.5 ft, the volume of K-65 material in the two Silos is about 220,000 ft^3. From Table J–5, the average ^{226}Ra concentration in the two Silos is about 417,000 pCi g^{-1}. From the Table J–4, the bulk density of the K-65 material is in the range 0.53–1.179 g cm^{-3}. Assuming an average density of 0.85 g cm^{-3}, the total ^{226}Ra content of the two K-65 Silos is about 2200 Ci.

The material in the K-65 Silos is not covered with water, though there is substantial moisture in the material. The Silos do have covers, which reduce Rn emissions somewhat (though in 1959–1979 the reduction was very slight, based on later calculations in this Appendix). For the wet pits, the ^{226}Ra is much less effective in releasing Rn into the air, because the water cover would significantly reduce the diffusion of Rn out of the waste material. It thus seems reasonable to expect that Rn releases from the waste pits would be less than five percent of Rn releases from the K-65 Silos. We thus consider the waste pits to be an insignificant source of Rn releases from the FMPC, and no further calculations are performed for this.

Recently, the Rn flux from waste pits 1, 2, 3, and 4 have been measured. Tomczak et al. (1992) reports results for pit 4, and summarizes earlier results for pits 1, 2, and 3. The results show total Rn releases from these four pits to be around 5 Ci y^{-1}. This shows that releases for these pits, in recent years (releases may have been higher in earlier years, before covers were applied), are insignificant compared with releases from the K-65 Silos.

In summary, the sources considered for releases of ^{222}Rn and Rn daughters are the K-65 Silos and drummed K-65 material stored on the Plant 1 pad.

General Methodology for Current Estimates of Releases from K-65 Silos

For some other releases at the FMPC, extensive data sets of direct measurements of release quantities are available. However, for radon releases there are no direct measurements of release quantities. In addition, until the 1980s there were very few measurements of parameters that can be used indirectly to calculate radon releases. Because of this limited availability of data, we use models to estimate radon release quantities.

The traditional model used to estimate radon releases from ^{226}Ra-bearing material, such as uranium mill tailings, involves calculations of the quantity of radon formed in the material, and the subsequent diffusion of the radon through the material to the outside air (Rogers et al. 1984). For the K-65 materials, measurements have not been made of the radon diffusion coefficient and radon emanation fraction, which are two key parameters to this traditional calculation. Literature values can be obtained for these parameters, but without site-specific values, the uncertainty ranges are extremely large. To reduce the uncertainties in our results, we have used different models, which we believe make the best use of the limited data that are available.

Earlier in this appendix (see page J–5), the history of structural changes to the silos was discussed. Not all of these changes to the silos would have a significant effect on the release of Rn. The most important change, in terms of Rn emissions, was the sealing of the openings in the silos in 1979. This action would have changed the ventilation rate of the silos, and thus changed the rate of ^{222}Rn release. The addition of the exterior foam layer in 1987 may have further reduced the emission of Rn. This foam layer was found, through laboratory testing, to have a very low Rn diffusion coefficient (Grumski and Shanks 1988). Covering the domes and cracks (and other penetrations) of the domes with this foam would be potentially effective in reducing the emission of Rn. The addition of the earthen berms in 1964 could have slightly decreased any trace releases of Rn through the walls of the silos, although specific information regarding this has not been found. Since the Silos were open to the atmosphere in 1964, with the gooseneck vent, and other unsealed penetrations in the domes, it seems probable that the overwhelming majority of Rn releases would have been through the dome penetrations. We thus think that the construction of the berms around the Silos would have had a negligible impact on Rn releases.

For the current calculations, we assume that a major change to ^{222}Rn releases likely occurred with the sealing of penetrations in 1979. We assume that from mid-September 1958, after Silo 2 was decanted and removed from service, through June 1979, no significant changes in the Rn releases occurred. We also assume that a significant change may have occurred at the end of 1987, when the foam layer was added to the silo domes. We thus separate the calculations into five time periods, as shown in Table J–11. We place primary emphasis on the pre-sealing (mid-September 1958–June 1979) and post-sealing (July 1979–December 1987) periods, because these two encompass most of the FMPC operating history (the time of concern of this Project). We generally refer to these two periods as 1959–1979 and 1980–1987, respectively. Table J–11 also shows the names by which we generally refer to the time periods, and the associated subscripts used for parameter names.

Table J-11. Time Periods for Calculations of Radon Releases from K-65 Silos

Time period	Description	Nominally called	Subscript for variables
mid-July 1952–mid-June 1953	Operational period of Silo 1	1952–1953	52–53
mid-June 1953–mid-September 1958	Operational period of Silo 2	1953–1958	53–58
mid-September 1958–June 1979	Before sealing penetrations in Silo domes (pre-sealing)	1959–1979, or pre	pre
July 1979–December 1987	After sealing penetrations in domes	1980–1987, or post	post
1988	After addition of foam layer	1988	1988

The general methods used to estimate the diffusion and free air exchange releases of Rn in the previous assessments are thought to be adequate. However, there are two significant flaws in the previous estimates of diffusion and free air exchange releases of Rn from the K-65 Silos. First, the sources of many of the parameter values used are not documented in the assessment reports (Borak 1985; Boback et al. 1987; Grumski 1987a; IT 1989). Thus, it is not possible to track the parameter values back to measurements or calculations in a primary reference. Second, the IT calculations assumed that the same release rate existed from 1953 through 1984 (IT 1989). Since it is known that openings in the silos, including the six-inch diameter gooseneck pipe, were sealed in 1979 (Boback 1980a; Boback 1980b; Grumski 1987a), it seems more reasonable to assume that a major change in release rates also occurred when these openings were sealed.

Thus the approach of the current estimates was to use the same basic calculational methods for air exchange and diffusion releases of the previous assessments (Borak 1985; IT 1989), but with changes to best incorporate the additional information located in this study. In fact, the current calculations are quite different, both in terms of the models and the values or distributions of values selected for the parameters. In general, the calculation of ^{222}Rn release rates, Q, is broken into calculations of the releases through air exchange, Q_{exch}, and through diffusion, Q_{diff}. The diffusion release calculation is relatively straightforward, and we use the method used in the previous assessment by Borak (1985), although we use different parameter values. We note here that the results of our calculations indicate that releases through the diffusion pathway are smaller than releases through air exchange, but still contribute a significant fraction of the total releases (see Table J–15 and Table J–16). The calculation of air exchange releases is more complicated, and here we have deviated, in the details, from the previous methods used by Grumski

(1987a). The next subsection of this appendix presents a more detailed description and justification of the basic models used in these current calculations.

For the time periods considered in this current assessment, one could try to estimate releases to a yearly or monthly time resolution. However, essentially all of the parameters used are assumed not to vary significantly from month to month or even from year to year (within the given assessment period). Thus, we feel that any additional resolution gained by estimating releases for shorter time periods would be lost in the uncertainties of the estimates. So, for these current calculations, we will only estimate a release rate for each time period ("1952–1953," "1953–1958," "pre," "post," and "1988"), which is assumed to apply to the entire time period. For convenience, the release rate estimates will be reported in activity released per year.

Because the characteristics of the two K-65 Silos that are important in estimating Rn releases are similar, and in many cases only limited information is available, we use average characteristics to represent both Silos. The models for Rn releases are developed for a single Silo (with the average characteristics), and the results incorporate a factor of 2 to account for the two Silos.

Calculational Strategy for Radon Emissions from the K-65 Silos

We first mention some assumptions made for the air exchange calculations. We assume that the ^{222}Rn concentration in outside air is negligible compared to the silo concentration so that outside air does not provide a source of Rn to the silo air. In our preliminary work on the ^{222}Rn source terms (Voillequé et al. 1991) we estimated that releases by diffusion through the silo domes were insignificant compared to releases by air exchange, and we assumed that the rate of removal of Rn from the silo air space due to diffusion releases was negligible. However, we have made changes since that initial effort, and while the diffusion releases are still estimated to be less than the air exchange releases, we no longer consider them insignificant, and the rate of removal of Rn due to diffusion releases is no longer neglected. Thus, the rate of change in the silo air Rn concentration can be described by an adaptation of a standard equation used for Rn concentration in homes (NCRP 1989):

$$\frac{dC_a}{dt} = \frac{P_{Rn}}{V_0} - C_a \lambda_{eff} \qquad \text{(J–7)}$$

where

C_a = concentration of ^{222}Rn in the silo air,

P_{Rn} = the constrained (by the presence of the silo) rate of release of ^{222}Rn into the silo air (production term) from the K-65 source material (activity per time),

V_0 = volume of the air space in the silo above the K-65 material, and

λ_{eff} = the effective removal rate of ^{222}Rn from the silo air space (fraction per time).

The only mechanisms considered for losses of Rn from the silo air space are releases into the (outside) atmosphere, through air exchange or diffusion through the silo dome, and radioactive decay. Thus,

$$\lambda_{\text{eff}} = \lambda_{\text{Rn}} + \lambda_{\text{v}} + \lambda_{\text{d}} \tag{J–8}$$

where
λ_{Rn} = the radioactive decay constant for ^{222}Rn,

λ_{v} = ventilation rate of the silo, or fraction of the silo air exchanged with the outside per unit time (per day), and

λ_{d} = rate constant for diffusion losses, the fractional rate of Rn loss from the silo air space through diffusion through the silo dome (fraction per time).

It is recognized that the silo ventilation rate (especially during the post-sealing time period), varies with a diurnal cycle component. Thus, to a lesser extent, the ^{222}Rn concentration in the silo air space also varies with a diurnal cycle. However, over a longer period of time the variations in these parameters are expected to be insignificant. Thus, we assume that equilibrium conditions exist, and that the ventilation rate, ^{222}Rn concentration, and ^{222}Rn production rate are constant over the periods of concern. Thus, based on the inputs of Rn to the silo air space equaling the losses, we obtain:

$$P_{\text{Rn}} = C_{\text{a}} \lambda_{\text{eff}} V_0 \tag{J–9}$$

Because the silo air space is a single compartment volume, we assume the contained air to be well mixed. Thus, the air exchange and diffusion release rates can be expressed simply as the activity in the silo air space times the silo ventilation rate or diffusion rate constant, as appropriate:

$$Q_{\text{exch}} = C_{\text{a}} \lambda_{\text{v}} V_0 \tag{J–10}$$

$$Q_{\text{diff}} = C_{\text{a}} \lambda_{\text{d}} V_0 \tag{J–11}$$

where Q_{exch} and Q_{diff} are the rates of release of ^{222}Rn from the silo through air exchange and diffusion through the silo dome, respectively.

These can be summed and rewritten:

$$Q_{\text{exch}} + Q_{\text{diff}} = C_{\text{a}} \lambda_{\text{eff}} V_0 \left(\frac{\lambda_{\text{v}} + \lambda_{\text{d}}}{\lambda_{\text{eff}}} \right) \quad \text{or}$$

$$Q = P_{\text{Rn}} \left(\frac{\lambda_{\text{v}} + \lambda_{\text{d}}}{\lambda_{\text{eff}}} \right) . \tag{J–12}$$

By expanding equation J–9, using equation J–8, and substituting for the products $C_{\text{a}}\lambda_{\text{v}}V_0$ and $C_{\text{a}}\lambda_{\text{d}}V_0$, using equation J–10 and equation J–11, we can also obtain:

$$Q = P_{\text{Rn}} - C_{\text{a}} \lambda_{\text{Rn}} V_0 \tag{J–13}$$

Equations J–10, J–12, and J–13 provide different methods of calculating the air exchange or total Rn release rate, depending on what information is available. We note that equation J–10 is essentially the equation used in the previous assessment (Grumski 1987a).

Strategy for calculation of Q_{post} (1980–1987). For the period 1980–1987, measured concentrations of ^{222}Rn in the silo air (C_a) are available, from a set of samples taken by the FMPC in 1987. In fact, these are the only usable measurements of ^{222}Rn concentrations in the silos that we have located. We would like to use this information, and thus should select either equation J–10 or equation J–13 to calculate $Q_{exch,post}$ or Q_{post}. In addition, data are available on the silo temperature cycling (as discussed earlier, on page J–14) that can be used to calculate λ_v. There are two related problems with the use of equation J–13 for this situation. First, since the major penetrations in the silos have been sealed, we expect the ventilation rate to be very low. The diffusion rate constant is also expected to be very small. In particular, we expect λ_v and λ_d to be significantly less than λ_{Rn}, so that λ_{eff} is only slightly different from λ_{Rn}. If this is the case, then we expect the two terms in equation J–13, P_{Rn} and ($C_a \lambda_{Rn} V_0$), to be approximately equal. For the uncertainty analysis then, we expect the uncertainty in the result, the difference of these two terms, to be large on a relative scale (relative standard deviation).

Second, the traditional method for calculating the release of Rn from a soil-like matrix into the air, for P_{Rn}, requires knowledge of ^{226}Ra concentration in the material, bulk density and porosity of the material, Rn emanation fraction from the material, and Rn diffusion coefficient through the material. No measurements of the Rn emanation fraction have been made, thus one might assume a rather broad range of 0.1 to 0.4 based on typical values for uranium mill tailings (Rogers et al. 1984). The Rn diffusion coefficient for the K-65 material has not been measured either, and the moisture contents of the important upper layers of the K-65 material are also not well characterized, so that diffusion coefficients spanning about two orders of magnitude are conceivable (Rogers et al. 1984). With these uncertainties, the uncertainty in the calculation of P_{Rn} would be very large.

For these reasons, we think that equation J–10, with the calculation of λ_v from the silo temperature cycling data, will make the best use of the available data, and will produce results with less overall uncertainty. Thus, equation J–10 will be used for the calculation of $Q_{exch,post}$. The releases through diffusion, $Q_{diff,post}$, will be calculated separately, using the methods used in the previous assessment. We note that in a later section of this Appendix, we use the standard method of calculating the release of Rn from the K-65 material, using characteristics of the material, as an alternative calculation to compare with our primary methods discussed here (see page J–73).

Strategy for calculation of Q_{pre} (1959–1979). For the period 1959–1979, no direct information is available about the Rn concentration, C_a, or the silo ventilation rate, λ_v. The Rn production rate, P_{Rn}, can be calculated from the release rate for the 1980–1987 period and Rn concentrations from both the pre and post periods. Indirect information about the Rn concentration is available in the exposure rate measurements on the silo domes. The short-lived daughters of ^{222}Rn, which will be present in a significant fraction of their equilibrium concentrations, emit gamma radiation. Thus Rn concentrations can be correlated to gamma exposure rates measured near the Rn source. We acknowledge that this is a rather uncertain way of estimating the Rn concentration, but it is the only approach we know of that uses the available data. Thus we will use equation J–13 for calculating Q_{pre}. This calculation includes releases by both air exchange and diffusion.

Strategy for calculation of Q_{1988} (1988). For 1988, we take two approaches. Preliminary calculations are performed using the same methodologies used for the 1980–1987 period. However, the results seem inconsistent with Rn monitoring data examined. The final approach bases releases for 1988 on releases for 1980–1987 and ratios of the Rn concentrations for the two periods.

Strategy for calculation of $Q_{52\text{-}53}$ and $Q_{53\text{-}58}$ (1952–1958). Very little directly applicable information is available to estimate releases from the K-65 Silos during these operational years. Thus, Rn releases are estimated based on releases for 1959–1979, with factors applied to account for differences due to the operating status.

Implementation of Calculations

As for other calculations in this Task 2/3 Report, the calculations of radon releases from the K-65 Silos and the drummed K-65 material are implemented as Monte Carlo simulations, to account for uncertainties. The Monte Carlo analysis uses distributions of potential values to represent the input parameters. Each distribution is based on the available (often limited) information about the parameter. Then, many iterations of the calculations are performed; each iteration samples from the parameter distributions to obtain parameter values. Thus, the result of the analysis is a distribution of potential values of the release quantities, which can be interpreted with specified percentile ranges (e.g., 5th to 95th percentile).

The Monte Carlo calculations for these analysis were performed using spreadsheet and forecasting software on an IBM-compatible microcomputer. Ten thousand iterations of the calculations were performed. The parameter distributions were generated using Crystal Ball®, version 2.0 for Windows (Decisioneering 1992). In Crystal Ball®, uniform distributions are generated using a multiplicative congruential generator, which has a period of length 2^{29}, and normal and lognormal distributions are generated using the Polar Marsaglia method (Decisioneering 1992).

The following sections describe the models used to perform the current estimates for the various time periods and sources. Annex 2 of this Appendix summarizes the equations and parameter distributions used for these current estimates of releases.

Model for Air Exchange Releases from K-65 Silos for 1980–1987

As discussed above, the air exchange releases for this period after the sealing of the silo penetrations can best be calculated by:

$$Q_{\text{exch,post}} = C_{\text{a,post}} \lambda_{\text{v,post}} V_0 \tag{J–14}$$

During the period from 1980 through 1987, the major penetrations through the silo domes, like the six-inch gooseneck pipe, had already been sealed. However, exchange of air between the silos and the atmosphere continued, through the numerous cracks in the concrete of the domes. Radon releases for this time period are based on measured concentrations of ^{222}Rn in silo air and on a silo ventilation rate calculated from the daily temperature change of silo air.

In this and following sections, we use the subscript "post" to refer to that parameter for the time period 1980 to 1987.

Radon concentration in silos 1980–1987. The silo interior air was sampled on November 4, 1987, prior to the operation of the Radon Treatment System (RTS) and prior to the application of the exterior foam layer to the silo domes (Grumski and Shanks 1988). The RTS is a system that pumps air from the silos through a series of calcium sulfate and charcoal beds, which adsorb ^{222}Rn from the circulating air (Grumski and Shanks 1988). This removes ^{222}Rn, and thus potential daughter products of ^{222}Rn, from the air space of the silos, and reduces the direct radiation exposure rates on the silo domes. The system is used to reduce radiation exposures to personnel involved in work on the silos.

The November 4, 1987, ^{222}Rn samples were analyzed by the FMPC and by Mound Laboratories, also in Ohio (Grumski and Shanks 1988). The results are given in Table J–12. For the sample from Silo 1 that was analyzed by the FMPC (WMCO), a table of detailed counting results is also given in Grumski and Shanks (1988). This table shows that the sample was counted eight times, at times from two days after sampling to 26 days after sampling. Concentration results were decay-corrected to the time of sampling. However, for one of the counts, it appears that the decay time was listed as 19.23 days, while the counting data for this count imply a decay time of 18.23 days. It appears the incorrect decay time then resulted in an incorrect decay correction for the count, with the listed concentration, 2.7×10^7 pCi L^{-1}, erroneously high (this result also was inconsistent with results for the other seven counts). If the decay time is changed to 18.23 days, we calculate that the concentration for that count should have been estimated to be 2.2×10^7 pCi L^{-1}. If this corrected result is used, the average of the eight results is 2.1×10^7 pCi L^{-1}, as presented in Table J–12.

Table J–12. Concentrations of ^{222}Rn (pCi L^{-1}) in K-65 Silo Gas Samples Taken November 4, 1987

Silo	Sample container	WMCO Analysis	Mound Analysis
Silo 1	sampling bag	—	2.3×10^7
Silo 2	sampling bag	—	1.3×10^7
Silo 1	glass flask	2.1×10^7 [a]	2.5×10^7 [b]
Silo 2	glass flask	3.0×10^7	2.9×10^7 [b]

[a] The results for this sample given in the report (Grumski and Shanks 1988), appeared to contain a calculational error. The value presented here is the average of the eight measurements, after the apparent error was corrected (by the authors of this current report).

[b] This value appears to be the average of concentrations measured for two sample flasks.

The significant difference between the sampling bag and glass flask results for Silo 2 was noted in the report (Grumski and Shanks 1988). This report indicated that the difference was "...most likely the result of dilution error associated with the sample bag procedure." Because of this potential error in the results for the samples taken in sampling bags, the two results for sampling bag samples will be ignored for the rest of this analysis.

Thus, the sample estimate of the average concentration of ^{222}Rn in the silos for this measurement episode is 2.62×10^7 pCi L^{-1} and the standard deviation of the four remaining measurements is 4.1×10^6 pCi L^{-1}, or about 16%. The average concentration is assumed to follow a normal distribution.

In addition to the measurement uncertainty, there is also uncertainty in the value of $C_{a,post}$ due to the (presumed) daily variations in the Rn concentration because of daily fluctuations in the silo ventilation rate, $\lambda_{v,post}$. The silo ventilation rate is later estimated to be roughly 0.03 d^{-1}, or about 3% d^{-1}, so even relatively large changes in $\lambda_{v,post}$ would cause only small changes in the Rn concentration in the silo head space. However, only one sampling episode was performed during the period 1980 to 1987, so our data set is very limited.

Typically, the uncertainty of an average value is expressed as the standard error of the mean, which is the standard deviation of the measured values divided by the square root of the sample size. However, because of the additional, unquantified uncertainties, we instead assume that the uncertainty of the average concentration is represented by the standard deviation for the four measured values. Thus, the distribution of values of $C_{a,post}$ is considered to be a normal distribution with mean 2.62×10^7 pCi L^{-1} and standard deviation 4.1×10^6 pCi L^{-1}.

Silo air volume 1980–1987. The volume of air space in the silos, V_0, can be calculated as the sum of the volume of air in the dome part of the silo, V_{dome}, and the volume of air in the cylindrical part of the silo above the K-65 residue material, V_{cyl}. We assume that the silo air space volume does not include the pore spaces of the K-65 material. Because the temperature of the K-65 material, and thus its pore spaces, would only change very slowly over time, the volume expansion in the pore spaces would be insignificant, and this assumption is reasonable. We assume the dome surface is spherically shaped, so these volumes are calculated as:

$$V_{dome} = \frac{\pi h^2}{3}(3R - h) \tag{J–15}$$

where

h = the height of the dome (above the silo walls),

R = $(r^2 + h^2)/2h$, the radius of the "sphere" of which the dome surface is a part, and

r = the radius of the silo. And,

$$V_{cyl} = \pi r^2 H \tag{J–16}$$

where H is the distance from the K-65 residue material to the top of the silo walls, which is the thickness of the cylindrical air layer.

Two drawings by the original designers and builders of the K-65 silos indicate the size of the silos to be 80 ft inside diameter, with a wall height of 26 ft 8 in, and inside dome height of an additional 9 ft 4 in (Preload 1951a; Preload 1951b). The unloading manhole, which is located very close to the center of the dome, is shown as 36 ft above the silo floor. The four influent manholes are shown to be located equally spaced on a circle of 25 ft radius from the center of the dome, and about 32 ft above the silo floor.

From these data, V_{dome} can be directly calculated. First, with $h = 9.33$ ft and $r = 40$ ft, R is determined to be 90.4 ft. Then, V_{dome} is calculated to be 23,900 ft^3.

A small number of documents have been found which can be used to estimate the value of H to calculate V_{cyl}. From a drilling and sampling episode in 1972, the depths of K-65 material were determined to be 20 ft in Silo 1 and 22 ft in Silo 2 (Nelson 1972b). Thus, for Silo 1, H = 26.67 ft – 20 ft = 6.67 ft. And, for Silo 2, H = 26.67 ft – 22 ft = 4.67 ft. With these values of H, V_{cyl} is estimated to be 33,500 ft^3 for Silo 1 and 23,500 ft^3 for Silo 2. The total volume is then estimated to be 57,000 ft^3 for Silo 1 and 47,000 ft^3 for Silo 2.

In 1978, gamma exposure rates were measured in Silo 1, at varying distances above the K-65 residue (Boback 1978). The farthest measurement location was 13 ft above the residue surface, and was also noted to be at the bottom of a manhole opening. It was not noted whether the manhole was one of the influent manholes or the unloading manhole. If the location was one of the influent manholes, the thickness of the K-65 material can be estimated to be 32 ft – 13 ft = 19 ft. Thus, H = 26.67 ft – 19 ft = 7.67 ft, V_{cyl} = 38,500 ft^3 and V_0 = 62,000 ft^3. If the location was the unloading manhole, the K-65 material thickness can be estimated to be 36 ft – 13 ft = 23 ft. Thus, H = 26.67 ft – 23 ft = 3.67 ft, V_{cyl} = 18,400 ft^3, and V_0 = 42,000 ft^3.

In 1958, Silo 2 was decanted and removed from service, with a stated content of 883,400 gallons of residue (Noyes 1958). Since the residue was pumped into the silos as a slurry, we assume the residue occupied a cylindrical shape. Thus, the thickness of residue can be estimated to be 23.5 ft. Thus, H = 26.67 ft – 23.5 ft = 3.17 ft, V_{cyl} = 15,900 ft^3, and V_0 = 40,000 ft^3.

The air volumes of the silos have also been determined by WMCO to be 55,815 ft^3 for Silo 1 and 45,762 ft^3 for Silo 2 (Shanks 1988). These volumes were based on depths of the residue of 20 ft in Silo 1 and 22 ft in Silo 2. These residue depths are the same as those of Nelson (1972b), but were not referenced in the WMCO calculations.

As part of the FMPC Remedial Investigation, the silos were sampled by WMCO in 1989 (DOE 1990). During this sampling episode, the average penetration into the K-65 residue material was 20 ft. No individual values of the penetration were given in the report. The value of 20 ft results in an estimate of V_0 of 57,000 ft^3, as noted earlier.

The range of these estimates of V_0 is from 40,000 ft^3 to 62,000 ft^3. Since we have no information that more definitively determines V_0, we assume the distribution of potential values of V_0 to be uniform, with minimum 40,000 ft^3 and maximum 62,000 ft^3.

Silo ventilation rate 1980–1987. As noted earlier, monitoring of the temperature and pressure differential of the K-65 Silos was performed in 1987 by WMCO (Grumski 1987a). It was concluded that the silos cannot hold any significant pressure and thus that increases in the temperature of the internal silo air resulted in the volumetric expansion of the air and the release of "excess" volume to the atmosphere (Grumski 1987a). Since the silos can not hold any significant pressure, it is certainly plausible that the cracks and other remaining penetrations in the silo domes are large enough and numerous enough that additional ventilation of the silos occurs, due to winds across the silo domes. For the present work, it is assumed that the silo ventilation rate is the sum of a ventilation rate due to the temperature effects and a ventilation rate due to wind effects. That is,

$$\lambda_{v,post} = \lambda_{v,\Delta T} + \lambda_{v,wind} \tag{J–17}$$

We note that the previous assessments did not specifically calculate a silo ventilation rate, although the ventilation rate was implicit in their ^{222}Rn release calculations (Grumski 1987a; IT 1989).

The ventilation rate due to the daily temperature changes, $\lambda_{v,\Delta T}$, is the fraction of the silo air exhaled due to the temperature changes per some unit time period, with units of (air changes) per time. Thus:

$$\lambda_{v,\Delta T} = \Delta V / V_0 \tag{J–18}$$

As discussed earlier in the evaluation of previous estimates (see equation J–5), the ideal gas law gives:

$$\Delta V = (\Delta T / T_0) V_0 \tag{J–19}$$

thus

$$\lambda_{v,\Delta T} = \Delta T / T_0 \tag{J–20}$$

where we define

ΔV = the increase in volume per day,

ΔT = the increase (only) in temperature of silo head space air, per day ($K\ d^{-1}$),

V_0 = the initial silo air volume above the K-65 material, and

T_0 = the initial temperature of the silo air (K).

As noted earlier in the discussion of previous estimates (see page J–14), usable temperature and pressure monitoring data for the two K-65 silos were obtained for 11 complete days (and a few shorter periods also) (Grumski 1987a; Shanks 1991). From this data, which was collected every two hours, the silo daily temperature increase and the initial temperature of the silo air can be determined. However, since only 11 days of data were obtained, the direct use of these data to estimate the annual average value of $\Delta T/T_0$ for the silos could introduce a significant bias. Instead, the daily silo values of $\Delta T/T_0$ can be correlated to daily temperature changes at the Cincinnati airport. Then, the correlations can be used to estimate the average value of $\Delta T/T_0$ for the silos from the Cincinnati temperature data.

Since we are interested only in the increase in silo temperature each day, ideally we would correlate the daily silo values of $\Delta T/T_0$ with the increase (only) in temperature at the Cincinnati airport. However, it is impractical to determine the airport temperature increase for each day of a full year, as would be required. Instead, we determine the difference between the maximum and minimum airport temperature for each day, and then correlate the daily silo values of $\Delta T/T_0$ to this difference. It is recognized that there are uncertainties introduced by performing the correlation in this manner. For example, on many days, the temperature falls during the day so the silo temperature increase, and $\Delta T/T_0$, is zero, but the airport temperature difference (maximum temperature – minimum temperature) is still positive.

The data obtained to perform the correlation are given in Table J–13 and Table J–14. The silo temperature increase and minimum temperature data were obtained from the previous temperature and pressure monitoring of the silos (Shanks 1991). For each silo, the monitoring results included a "bottom" temperature, near the bottom of the air space in the silo, and a "top" temperature, near the top of the silo air space. The average value of $\Delta T/T_0$, given in Table J–13, has been calculated as follows. First, for each silo, the top and bottom temperatures at each measurement time were averaged, as a best estimate of the temperature in the silo air for that point in time. Next, the increase in temperature and the initial temperature were determined for each silo for each of the 11 days, and were converted to the (absolute) Kelvin scale (K). Then, the values of $\Delta T/T_0$ for each silo for each day were calculated. Finally, the average value of $\Delta T/T_0$, for each day, was calculated as the average of the values for the two silos for that day.

Records of the hourly temperature at the Cincinnati airport have been obtained for the period 1948 to 1987, by year (NCDC 1991). The maximum temperature, T_{max}, and the minimum temperature, T_{min}, were extracted for each of the 11 days on which the silo temperature was measured, and are shown in Table J–14. The difference $T_{max} - T_{min}$, in Table J–14, has been calculated for this analysis.

Table J–13. K-65 Silo Values of $\Delta T/T_0$; Data Used for a Linear Correlation[a]

	Silo 1 data[b]			Silo 2 data[b]			Average
Date	T_0 (°F)	ΔT (°F d^{-1})	$\Delta T/T_0$ (d^{-1})	T_0 (°F)	ΔT (°F d^{-1})	$\Delta T/T_0$ (d^{-1})	$\Delta T/T_0$ (d^{-1})
3/27/87	50.65	20.15	0.0395	52.35	22.85	0.0447	0.0421
3/29/87	53.6	21.6	0.0421	56.25	23.65	0.0459	0.0440
3/30/87	42.85	0	0	41.9	0	0	0
3/31/87	40.8	4.6	0.0092	39.1	5.15	0.0103	0.0098
4/01/87	38.85	14.2	0.0285	36.6	16.2	0.0327	0.0306
4/02/87	43.95	12.	0.0238	43.3	14.1	0.0280	0.0259
4/03/87	38.5	11.3	0.0227	37.55	12.6	0.0254	0.0240
4/04/87	40.25	3.15	0.0063	39.	3.6	0.0072	0.0068
5/09/87	61.95	30.9	0.0593	63.1	35.1	0.0672	0.0632
5/10/87	65.25	30.1	0.0574	67.5	34.1	0.0647	0.0610
5/11/87	68.35	27.1	0.0514	71.25	28.8	0.0543	0.0528

[a] The values of ΔT and T_0 must be expressed in absolute temperature units (K) before the ratio is computed.
[b] T_0 and ΔT data obtained from Shanks (1991).

Table J–14. Temperature Difference at Cincinnati Airport; Data Used for a Linear Correlation[a]

Date	Maximum T_{max} (°F)	Minimum T_{min} (°F)	Difference $T_{max} - T_{min}$ (°F)
3/27/87	65	44	21
3/29/87	77	44	33
3/30/87	48	29	19
3/31/87	33	24	9
4/01/87	47	24	23
4/02/87	43	31	12
4/03/87	37	27	10
4/04/87	41	28	13
5/09/87	80	43	37
5/10/87	85	53	32
5/11/87	84	60	24

[a] T_{min} and T_{max} data obtained from NCDC (1991).

We note that some relationship is expected between the minimum temperature and the daily temperature change at the Cincinnati airport. A linear regression of the values of ($T_{max} - T_{min}$) versus the values of T_{min} for 1987 was performed. The regression coefficient was determined to be $R = 0.075$. With this small value of the regression coefficient, we assume the relationship between ($T_{max} - T_{min}$) and T_{min} is weak enough to be considered insignificant for our analysis. Thus, we neglect this possible relationship.

A linear correlation of the average values of $\Delta T/T_0$ (dependent variable) to the Cincinnati airport temperature difference (independent variable), $T_{max} - T_{min}$, was performed using a least squares regression. The regression coefficient is $R = 0.80$. The regression line is given by:

$$\Delta T/T_0\,(\mathrm{d}^{-1}) = \left(0.00179\ {}^\circ\mathrm{F}^{-1}\ \mathrm{d}^{-1}\right)\times\left(T_{max} - T_{min}({}^\circ\mathrm{F})\right) - 0.00516\ \mathrm{d}^{-1} \tag{J–21}$$

For this regression line, the standard error of the estimate, $S_{Y|X}$, is 0.0138 d^{-1}.

From the hourly records of temperature at the airport, additional data were obtained for the complete year 1987 (NCDC 1991). The average of the daily maximum temperatures was determined to be 65.08 °F. The average of the daily minima was determined to be 45.62 °F. The average daily difference (average of ($T_{max} - T_{min}$)) is equal to the difference of the averages of the maxima and minima. Thus, the average daily difference for 1987 is 65.08 – 45.62 = 19.46 °F. This value is also assumed to represent the average daily difference for the assessment period 1980 to 1987.

Thus, the annual average value of $\Delta T/T_0$, and thus $\lambda_{v,\Delta T}$ for the period 1980 to 1987 is estimated from the regression line and the average daily difference as:

$$\begin{aligned}\lambda_{v,\Delta T} &= \Delta T/T_0\\ &= \left(0.00179\ {}^\circ\mathrm{F}^{-1}\ \mathrm{d}^{-1}\right)\times\left(19.46\ {}^\circ\mathrm{F}\right) - 0.00516\ \mathrm{d}^{-1}\\ &= 0.0297\ \mathrm{d}^{-1}\end{aligned} \tag{J–22}$$

It is assumed that the conditional distribution of $\Delta T/T_0$, at the given value of $T_{max} - T_{min}$ = 19.46 °F, is a normal distribution with standard deviation $S_{Y|X}$. Thus, we consider the distribution of potential values of $\Delta T/T_0$ to be a normal distribution with mean 0.0297 d^{-1} and standard deviation 0.0138 d^{-1}. However, with this mean and standard deviation, there is a significant chance that negative values of $\Delta T/T_0$ might be selected from the distribution. Since such negative values are meaningless for the calculation of the ventilation rate, we truncate the distribution at 0, disallowing negative values.

As discussed earlier, it is plausible that the cracks in the silo domes are numerous enough and large enough that the action of winds on the domes could create additional ventilation in the silos, represented by $\lambda_{v,wind}$. However, no data have been found to substantiate an estimate of $\lambda_{v,wind}$. In our preliminary source term work (Voillequé et al. 1991) we arbitrarily assumed that $\lambda_{v,wind}$ ranges from zero to the value of $\lambda_{v,\Delta T}$, with a uniform distribution. However, that assumption introduced a multiplicative factor, with mean 1.5, into the estimate of Q_{exch}, that we now think cannot be substantiated. Since additional information has not been located to substantiate a value for $\lambda_{v,wind}$, we now assume a value of zero.

Results for 1980–1987. Table J–15 summarizes the frequency distribution for the air exchange Rn release rate for 1980–1987. The uncertainty range for this release rate is fairly broad, with a 90% probability interval spanning a factor of seven range.

Table J–15. Summary of Predicted Air Exchange Radon Release Rate (Ci y^{-1}) from the K-65 Silos for the 1980–1987 Period

	Percentiles of distribution				
Period	5th	25th	median	75th	95th
July 1979–December 1987	230	550	810	1100	1600

Model for Diffusion Releases from K-65 Silos for 1980–1987

The diffusion releases for the period 1980 to 1987 are calculated using the same methods used in the previous assessment (IT 1989; Borak 1985). However, since the concentration of ^{222}Rn in the silos has been measured, the calculated releases are based on the measured ^{222}Rn concentration, rather than on a concentration calculated from characteristics of the K-65 waste material. As was described earlier, the flux of ^{222}Rn diffusion through the concrete dome of the silos can be calculated by (Borak 1985; Collé et al. 1981):

$$J = \frac{\varepsilon_c \lambda_{Rn} l_c C_a}{\sinh\left(\frac{l}{l_c}\right)} \qquad \text{(J–23)}$$

where

J = ^{222}Rn flux from the head space through the silo dome surfaces to the surrounding air (pCi m^{-2} d^{-1}, or similar),

ε_c = total porosity of the dome concrete,

λ_{Rn} = decay constant of ^{222}Rn (d^{-1}, or other as appropriate),
l_c = diffusion length of ^{222}Rn in the dome concrete (cm),
C_a = concentration of ^{222}Rn in the silo air (pCi L^{-1}), and
L = thickness of the dome concrete (cm).

The diffusion release rate is then calculated as:

$$Q_{diff,post} = JA_{dome} \tag{J–24}$$

where A_{dome} is the surface area of the silo domes (ft^2, or other). These equations are applied to the 1980–1987 period by using $C_{a,post}$ for the ^{222}Rn concentration in the silo air.

The porosity of the dome concrete, ε_c, and the Rn diffusion length in the dome concrete, l_c, are dependent on the physical characteristics of the dome concrete. A number of documents indicate that the quality of the dome concrete is poor, and apparently has been from the earliest years. Memoranda from the 1950's indicated that there were many cracks in the K-65 Silo walls, and that small quantities of liquid seeped from some of them (Wunder 1954; Martin 1957). In 1984, the Mound Laboratory, a DOE facility, made measurements of the Rn fluence rate (flux) through the domes of the K-65 Silos (Hagee et al. 1985). Mound indicated that the domes had many obvious cracks and fissures, and their measurements showed greatly increased Rn transport through these cracks. Structural studies of the K-65 Silos have been completed more recently. In one study, Pulse-Echo tests were performed on the tanks to determine the concrete quality (Camargo 1986). Results showed general thinning of the domes, with a sharply irregular interior surface, indicating significant deterioration of the domes.

In another study, samples of concrete from Silo 4 (the unused one) were subjected to laboratory analyses (BNI 1990). The samples from Silo 4 were considered to be reasonably representative of the K-65 and Metal Oxide Silos, since they were all built at the same general time, and have been exposed to the same weathering conditions. Results indicated that the concrete was originally placed with medium to high slump. High slump concrete is, in general, less dense, more porous, and less durable overall. Results of the petrographic analysis indicated that reactivity was responsible for expansion of the concrete, which resulted in microcracking, which then allowed ingress of water into the concrete. The reactivity generally occurs within 90 days after the concrete is placed. Freeze-thaw conditions were thought to aggravate these conditions and increase the deterioration of the concrete.

This evidence indicates that the dome concrete has had fairly poor quality since the earliest days, and has deteriorated more over the years. No specific data on the porosity and Rn diffusion length for the K-65 Silos dome concrete have been obtained, so we must rely on literature values. However, we choose values from the literature that are more representative of poor quality concrete.

Concrete porosity. A National Bureau of Standards (NBS) review report cited a measured value of concrete porosity of 0.265 from one study and an assumed range of 0.05 to 0.25 from another study (Collé et al. 1981). The porosity used in the previous assessment was 0.3 (Borak 1985), although the source of the value was not cited. Because of the very limited amount of data found, and the lack of data specific to the FMPC K-65 silos, we assume that the potential values of ε_c follow a uniform distribution. Based on the poor quality of the dome concrete, we assume the upper half of the range cited in the NBS report would apply, as poor concrete quality would be associated with higher porosity. Thus, the porosity, ε_c, is assumed to follow a uniform distribution, with minimum 0.16, and maximum 0.265.

Radon diffusion length in concrete. The NBS review report cited measurements of the ^{222}Rn diffusion coefficient in concrete, that would equate to diffusion lengths from 7.43 cm to 12.7 cm (Collé et al. 1981). Nazaroff and Nero (1988) cite values of the diffusion length in concrete from 6 cm to 20 cm. The diffusion length in concrete used in the previous assessment was 12 cm (Borak 1985). This value was justified by Borak based on a referenced range of values from 6 cm to 23 cm measured for intact concrete (Borak 1986; Jonassen and McLaughlin 1978 [cited by Borak 1986]; Krisiuk et al. 1971 [cited by Borak 1986]). Again, the data found are limited, and are not specific to the K-65 domes. Poor quality concrete would be associated with greater diffusion through it, and thus larger values of the Rn diffusion length. We thus use the upper half of the range of literature values. The potential values of l_c are assumed to be represented by a uniform distribution with minimum 14.5 cm and maximum 23 cm.

Silo dome thickness. A review of the K-65 Silos' history indicated that the thickness of the domes was 4 in (Shanks and Vogel 1988). However, an FMPC report about proposed stabilization activities on the silos indicated that the domes were designed to be 8 in thick at the silo wall, tapering to 4 in thick at the dome center (Grumski 1987a). However, we have reviewed some of the original engineering drawings for the silo construction, and this latter characterization appears to be a misinterpretation. The silo design drawing indicates that the domes would be 8 in thick at the silo wall, but would taper to a thickness of 4 in within about 2 ft from the wall (Preload 1951a). Thus, the thickness of 8 in is only at the very edge of the domes, and is ignored for this analysis.

In addition, a structural assessment of the silo domes was performed in 1985. As part of this assessment, the thicknesses of the domes were measured, and were indicated to be as thin as 3 inches for portions of the centers of the domes, where deterioration has occurred (Camargo 1986). As an estimate of the mean thickness of the domes is not available, we consider the potential values of L to be represented by a uniform distribution with minimum 3 in and maximum 4 in.

Dome surface area. The silo dome surfaces are assumed to be portions of a sphere. As such, their surface area, A_{dome}, can be calculated as:

$$A_{dome} = 2\pi R^2\left(1-\sqrt{1-\frac{r^2}{R^2}}\right) \qquad \text{(J–25)}$$

where

R = $(r^2 + h^2)/2h$, the radius of the "sphere" of which the dome surface is a part,

h = the height of the dome (above the silo walls), and

r = the radius of the silo.

As discussed earlier, $h = 9.33$ ft and $r = 40$ ft, and so R is determined to be 90.4 ft. Thus, $A_{dome} = 5300$ ft^2. For the purposes of these calculations, the uncertainty in A_{dome} is assumed negligible.

Radon decay constant. The half life of ^{222}Rn is 3.8235 d (Walker et al. 1989). Thus, the decay constant for ^{222}Rn, λ_{Rn}, is 0.18129 d^{-1}. For purposes of our calculations, this value is assumed to have negligible uncertainty.

Results for 1980–1987. Table J–16 summarizes the calculated distribution of the predicted diffusion Rn release rate from the K-65 Silos for the 1980–1987 period. As expected, the release rate for diffusion releases is substantially smaller than the release rate for air exchange releases, though not insignificant.

Table J–16. Summary of Predicted Diffusion Radon Release Rate (Ci y^{-1}) from the K-65 Silos for the 1980–1987 Period

	Percentiles of distribution				
Period	5th	25th	median	75th	95th
July 1979–December 1987	72	100	130	170	240

Total Releases from K-65 Silos for 1980–1987

The total Rn release rate from the K-65 Silos for the 1980–1987 period is the sum of the release rates due to air exchange and diffusion:

$$Q_{post} = Q_{exch,post} + Q_{diff,post} \tag{J–26}$$

Table J–17 summarizes the frequency distribution of the calculated total Rn release rate from the K-65 Silos for the 1980–1987 period. Also shown are predictions of the fraction of the total removal of Rn from the Silos that occurs through release to the outside air through air exchange and diffusion (the rest is "removed" by radioactive decay), $[(\lambda_v+\lambda_d)/\lambda_{eff}]_{post}$. The results for this fraction indicate that only a small fraction of the Rn in the Silos is released to the outside air.

Table J–17. Summary of Frequency Distributions of Calculations of Total Radon Release Rates from the K-65 Silos for 1980–1987

		Percentiles of distributions				
Parameter	Units	5th	25th	median	75th	95th
Q_{post}	Ci y^{-1}	360	690	950	1200	1700
$[(\lambda_v+\lambda_d)/\lambda_{eff}]_{post}$		0.071	0.13	0.16	0.20	0.24

Model for K-65 Silo Radon Production Rates

In order to calculate the total Rn releases for the 1959–1979 period, Q_{pre}, using equation J–13, we must first determine the Rn production rate, $P_{Rn,pre}$, for this period. By "Rn production rate" we mean the rate of release of ^{222}Rn from the K-65 material into the silo air space. The production rate is generally constrained by existing Rn in the silo air spaces. For comparison with an alternative calculation of releases from the Silos, discussed later in this Appendix, we also wish to calculate an unconstrained production rate (no Silo and no Rn to inhibit release from the K-65 material), $P_{Rn,0}$, which is equivalent to an unconstrained release rate from the Silos.

The Rn production rate can be determined for the 1980–1987 period based on the Rn concentration and effective removal rate of Rn from the head space. In our preliminary source term work (Voillequé et al. 1991), we assumed that the Rn production rate was the same for the two periods 1959–1979 and 1980–1987, even though the Rn concentrations had changed. It is recognized that the release of Rn from the K-65 material into the silo air space would be higher for a lower silo Rn concentration because the diffusion of Rn out of the K-65 material is a process constrained by the Rn concentration in the silo air. In this current assessment, we account for different Rn production rates for the different time periods. To do this, we use the relationship between the silo Rn concentration and the Rn production rate, and we use the Rn production rate for 1980–1987 as a baseline.

We first calculate the Rn production rate, $P_{Rn,post}$, for 1980–1987. This is calculated based on the concentration of ^{222}Rn measured after the sealing of the silo openings. From an assumption of an equilibrium ^{222}Rn concentration in the silo air, the release rate of ^{222}Rn into the silo air is equal to the rate of loss of ^{222}Rn from the silo air by decay and by release to the atmosphere. As given in equation J–9, this is represented as (see also Collé et al. 1981):

$$P_{Rn,post} = C_{a,post} V_0 \lambda_{eff,post} \tag{J–27}$$

where

P_{Rn} = the constrained (by the presence of the silo) rate of release of ^{222}Rn from the source material into the silo air (Rn production rate) (pCi d^{-1}),

$C_{a,post}$ = the concentration of ^{222}Rn in the silo air (pCi L^{-1}),

V_0 = volume of the silo air space, as used earlier, and

$\lambda_{eff,post}$ = the effective removal rate of ^{222}Rn from the silo air space: the sum of the ^{222}Rn decay constant, λ_{Rn}; the ventilation rate, $\lambda_{v,post}$; and the rate constant for diffusion losses, $\lambda_{d,post}$ (d^{-1} or similar).

The ^{222}Rn concentration, $C_{a,post}$; silo air volume, V_0; and silo ventilation rate, $\lambda_{v,post}$, were discussed previously in this appendix.

The half life of ^{222}Rn is 3.8235 d (Walker et al. 1989). Thus, the decay constant for ^{222}Rn, λ_{Rn}, is 0.18129 d^{-1}. For purposes of our calculations, this value is assumed to have negligible uncertainty.

The rate constant for diffusion Rn losses from the silo air space, $\lambda_{d,post}$, can be calculated from a rearrangement of equation J–11, as follows.

$$\lambda_{d,post} = \frac{Q_{diff,post}}{C_{a,post} V_0} \tag{J–28}$$

The rate of diffusion release, $Q_{diff,\,post}$, was discussed earlier in this Appendix (page J–34).

We next develop the relationship between the Rn production rate and the Rn concentration in the silo air. A National Bureau of Standards (NBS) review report on Rn transport in building materials (Collé et al. 1981) provides useful models for this purpose. For a Rn concentration in air above a Rn source material, the constrained Rn diffusion fluence rate (often simply called Rn flux) from the source material into the air is given by the following equation (Collé et al. 1981). For this relationship, it is assumed that the bottom of the source material is impervious to Rn transport, an assumption that seems reasonable for the K-65 Silos, which have concrete floors.

$$j_D = \sqrt{\frac{D_e \varepsilon_w}{\lambda_{Rn}}} (\phi - C_a \lambda_{Rn}) \tanh\left(\frac{L_w}{l_w}\right) \tag{J–29}$$

where

j_D = constrained (by Rn in silo air) diffusion fluence rate of Rn (Rn flux). The quantity of Rn per unit time per unit area transported by diffusion from the source material (in this case the K-65 material) into the ambient air (silo air space in this case) (pCi m^{-2} s^{-1}, or similar),

D_e = effective diffusion coefficient of Rn through the porous source material (cm^2 s^{-1}, or similar),

ε_w = porosity of the source material,

ϕ = the pore space Rn production rate. Quantity of Rn produced in pore spaces of the source material per unit time per unit volume that is free to migrate through the pores of the material (pCi m^{-3} s^{-1}). Depends on characteristics of the source material, including ^{226}Ra concentration, Rn emanation fraction, bulk density, and porosity, and on the Rn decay constant,

L_w = thickness of the source material (cm, or similar), and

l_w = diffusion length of Rn in the source material (related to D_e) (cm, or similar).

The other parameters have been described earlier. The Rn production rate is then just:

$$P_{Rn} = j_D A_w \tag{J–30}$$

where A_w is the surface area of the source material (the K-65 material) exposed to the (silo) air. With a slight rearrangement we have:

$$P_{Rn} = A_w \sqrt{D_e \varepsilon_w \lambda_{Rn}} (\phi/\lambda_{Rn} - C_a) \tanh\left(\frac{L_w}{l_w}\right) \tag{J–31}$$

Equation J–27 and equation J–28 can be used to calculate $P_{Rn,post}$, but we must also calculate $P_{Rn,pre}$. We assume that the characteristics of the K-65 material have not changed since the Silos were decanted, and thus that D_e, l_w, ε_w, and ϕ are the same for the 1959–

1979 period as they are for the 1980–1987 period. Of the characteristics of the K-65 material that impact these parameters, the one most likely to have changed over this long period is probably the moisture content of the uppermost part of the K-65 material in the silos. The moisture content affects the diffusion coefficient, D_e, and could affect the pore Rn production, ϕ, through the emanation fraction. However, no applicable information has been located that could be used to determine the time history of this moisture content. We thus assume that it has not changed enough to significantly alter D_e or ϕ. The surface area, A_w, and thickness, L_w, of the K-65 material in the Silos also would not have changed over time. With these assumptions of invariant characteristics of the K-65 material, we apply equation J–31 to the two time periods, and ratio the two resultant equations to obtain:

$$P_{Rn,pre} = P_{Rn,post}\left(\frac{\phi/\lambda_{Rn} - C_{a,pre}}{\phi/\lambda_{Rn} - C_{a,post}}\right) \qquad \text{(J–32)}$$

The calculation of $C_{a,pre}$ is discussed later (page J–41).

In order to calculate (ϕ/λ_{Rn}), we make use of the relationship between ϕ, λ_{eff}, and C_a for a contained air space (like the Silos) above the Rn source material (Collé et al. 1981):

$$C_a = \frac{\phi}{\lambda_{eff}}\left(\frac{\varepsilon_w l_w}{\varepsilon_w l_w + h}\right) \qquad \text{(J–33)}$$

where h is the effective height of the contained air space above the source material. For the 1980–1987 period, with rearrangement, we obtain:

$$\frac{\phi}{\lambda_{Rn}} = C_{a,post}\left(\frac{\lambda_{eff,post}}{\lambda_{Rn}}\right)\left(\frac{\varepsilon_w l_w + h}{\varepsilon_w l_w}\right) \qquad \text{(J–34)}$$

In a later section of this Appendix we discuss an alternative calculation which is based on the characteristics of the K-65 material, including ε_w and the diffusion coefficient, which is related to l_w (see page J–73). As part of the alternative analysis, calculations of the quotient $[(\varepsilon_w l_w + h)/\varepsilon_w l_w]$ were performed. The median value of this quotient was determined to be 6.35 (see page J–82). For the calculation of (ϕ/λ_{Rn}) here, we use this median value. Since this quotient is significantly greater than 1, (ϕ/λ_{Rn}), from equation J–34, will be significantly greater than $C_{a,post}$, which in turn is greater than $C_{a,pre}$. Thus, the resultant estimate of $P_{Rn,pre}$, from equation J–32, is not very dependent on the exact value of this quotient. Thus we think the use of the median value of this quotient is adequate for the calculation in equation J–34.

For comparison with the alternative calculation of Rn releases performed later, we additionally calculate the unconstrained Rn production rate, which we call $P_{Rn,0}$. This is done by using equation J–32, and substituting a value of 0 for $C_{a,pre}$. Thus:

$$P_{Rn,0} = P_{Rn,post}\left(\frac{\phi/\lambda_{Rn}}{\phi/\lambda_{Rn} - C_{a,post}}\right) \qquad \text{(J–35)}$$

Table J–18 summarizes the calculated Rn production rate frequency distributions.

Table J–18. Summary of Frequency Distributions of Rn Production Rates from K-65 Material in the K-65 Silos

		Percentiles of distributions				
Parameter	Units	5th	25th	median	75th	95th
$P_{Rn,post}$	pCi d^{-1}	5.6×10^{12}	7.0×10^{12}	8.1×10^{12}	9.3×10^{12}	1.1×10^{13}
$P_{Rn,pre}$	pCi d^{-1}	6.4×10^{12}	8.0×10^{12}	9.2×10^{12}	1.1×10^{13}	1.3×10^{13}
$P_{Rn,0}$	Ci y^{-1}	4700	5900	6800	7900	9400

Model for Total Releases from K-65 Silos for 1959–1979

In 1958 the second silo was decanted, with the excess water removed through the weirs in the sides of the silo. Thus, during the period 1959 to 1979 the K-65 material in the silos should not have been covered with standing water. The six-inch gooseneck pipe vent was open from the silos to the atmosphere. For this time period, the total Rn releases, through air exchange and diffusion, are estimated using equation J–13 with the ^{222}Rn concentration based on exposure rate measurements on the domes of the silos. That is,

$$Q_{pre} = P_{Rn,pre} - C_{a,pre}\lambda_{Rn}V_0 \qquad \text{(J–36)}$$

The calculations of $P_{Rn,pre}$, V_0, and λ_{Rn} were previously discussed. The rest of this section discusses the calculation of the Rn concentration in the silo head space for 1959–1979. Here, and in the following sections, the subscript "pre" refers to that parameter for the time period 1959 to 1979.

No useful measurements of the ^{222}Rn concentration in the silos for the period 1959–1979 have been located. However, an alternative is to make use of the fact that two of the short-lived daughters of ^{222}Rn, ^{214}Pb and ^{214}Bi, emit gamma radiation in significant quantities. Based on our later calculations, the rate constants for losses of ^{222}Rn from the silo air space are relatively small, compared to the decay constants of the Rn daughters. Thus, these daughters would be present essentially in equilibrium with ^{222}Rn, and the high ^{222}Rn concentration in the silo air will have an associated, significant gamma exposure rate.

If measurements of the exposure rate are obtained for a consistent geometry, for a time period when the ^{222}Rn concentration is also known, an exposure rate factor (mR h^{-1} per pCi L^{-1} ^{222}Rn, or similar) can be developed. Then, the ^{222}Rn concentration can be estimated for other time periods when only exposure rate data exist. This is the approach taken. The exposure rate factor (ERF) will be developed based on ^{222}Rn concentration and exposure rate data for the period around 1987. Then, the ^{222}Rn concentration will be estimated for the period 1959 to 1979. That is:

$$ERF = \frac{X_{post} - X_{bkg}}{C_{a,post}} \qquad \text{(J–37)}$$

where

ERF = exposure rate factor (mR h^{-1} per pCi L^{-1}),

X_{post} = the average gross exposure rate on the silo domes during the period 1980–1987 (mR h^{-1}), and

X_{bkg} = the average "background" exposure rate on the silo domes (mR h^{-1}). This exposure rate would include contributions from sources other than the ^{222}Rn daughters in the silo air space. Since this would include contributions from ^{222}Rn daughters in the K-65 residues, this background exposure rate will be much greater than a typical environmental background exposure rate.

Then

$$C_{a,pre} = \frac{X_{pre} - X_{bkg}}{ERF} \tag{J–38}$$

where

$C_{a,pre}$ = concentration of ^{222}Rn in the silo air (pCi L^{-1}), and

X_{pre} = the average gross exposure rate on the silo domes during the period 1959–1979 (mR h^{-1}).

These two equations can be simplified to:

$$C_{a,pre} = C_{a,post}\left(\frac{X_{pre} - X_{bkg}}{X_{post} - X_{bkg}}\right) \tag{J–39}$$

Searches through historical records of the FMPC have located some results of radiation exposure rate measurements on the K-65 Silo domes, which are summarized in Table J–19. The "contact" measurement data will be used in this analysis because the only measurements made after ^{222}Rn had been removed from the silos were made on contact. The measurements made at 4 ft above the surface, and the measurements for which the height was not specified, will not be included in this analysis. Two other measurements will also be disregarded. First, the low value of those made on contact with Silo 2 (85 mR h^{-1}) in November 1980 was made on the edge of the silo dome (Green 1980b), and is thus not considered comparable to the other measurements, which were taken closer to the middle of the domes. Second, the extremely high result of April 1986 was obtained at a crack in the dome surface (Fleming 1986), through which ^{222}Rn was probably moving, and in which decay products had probably plated out. Thus, this measurement is also not considered indicative of the silo ^{222}Rn concentration in the same manner as the other measurements. The contact exposure rates that will be used in this analysis are plotted in Figure J–4.

The data for the period prior to sealing the openings, 1959 to 1979, do not indicate a significant variation in exposure rate. These measurements ranged from 65 to 90 mR h^{-1}. Little information exists about the number and location of measurements made for each measurement episode. Some results were averages, while others were ranges.

Table J-19. Measurements of Exposure Rate (mR h^{-1}) on Domes of K-65 Silos: Before and After Sealing of Dome Penetrations

Date of Measurement	Silo	Height of Measurement	Exposure rate	Comments (reference)
		Prior to Sealing Silo Openings		
April 1964	1	contact	75	Average value, probably silo 1. (Starkey 1964)
March 1972	ns[a]	ns	30	(Levy 1972)
March 1972	ns	contact	75	Maximum reading, assumed to be on contact. (Nelson 1972a)
May 1973	1	contact	65–90	Assumed on contact since other locations were. (Boback 1973)
May 1973	2	contact	70–75	(Boback 1973)
July 1973	2	ns	35	Near center of dome. (Levy 1973)
ns	ns	contact	90	Specified as before sealing of openings in 1979. (Boback 1980a)
		After Sealing Silo Openings		
April 1980	1	contact	250	(Green 1980a)
April 1980	1	4 ft	150	(Green 1980a)
April 1980	2	contact	200–250	(Green 1980a)
April 1980	2	4 ft	150	(Green 1980a)
ns	ns	contact	250	Specified as after sealing of openings in 1979. (Boback 1980a)
November 1980	1	contact	175	(Green 1980b)
November 1980	1	4 ft	140	(Green 1980b)
November 1980	2	contact	85–175	The low value was near edge, rather than center. (Green 1980b)
November 1980	2	4 ft	45–100	The low value was near edge, rather than center. (Green 1980b)
May 1982	1	contact	290	(Grant and Stevens 1982)
May 1982	1	3 ft	18–250	Low value was near edge. (Grant and Stevens 1982)
May 1982	2	contact	400	(Grant and Stevens 1982)
May 1982	2	3 ft	35–280	Low value was near edge. (Grant and Stevens 1982)
April 1986	2	contact	850	Measured at crack in dome; other results not legible. (Fleming 1986)
November 1987	1	contact	168–208	Baseline, average 193. (Grumski and Shanks 1988)
November 1987	1	contact	35.5–68	After operation of RTS[b], average 55. (Grumski and Shanks 1988)
November 1987	2	contact	221–250	Baseline, average 232. (Grumski and Shanks 1988)
November 1987	2	contact	60–76	After RTS, average 68. (Grumski and Shanks 1988)

[a] "ns" indicates that the parameter was not specified in the reference document.
[b] RTS is the acronym for the Radon Treatment System.

For the period after sealing the openings, 1980 to 1987, the data show considerable variation (Figure J–4), but no clear trend is evident. The variation seen is not excessive,

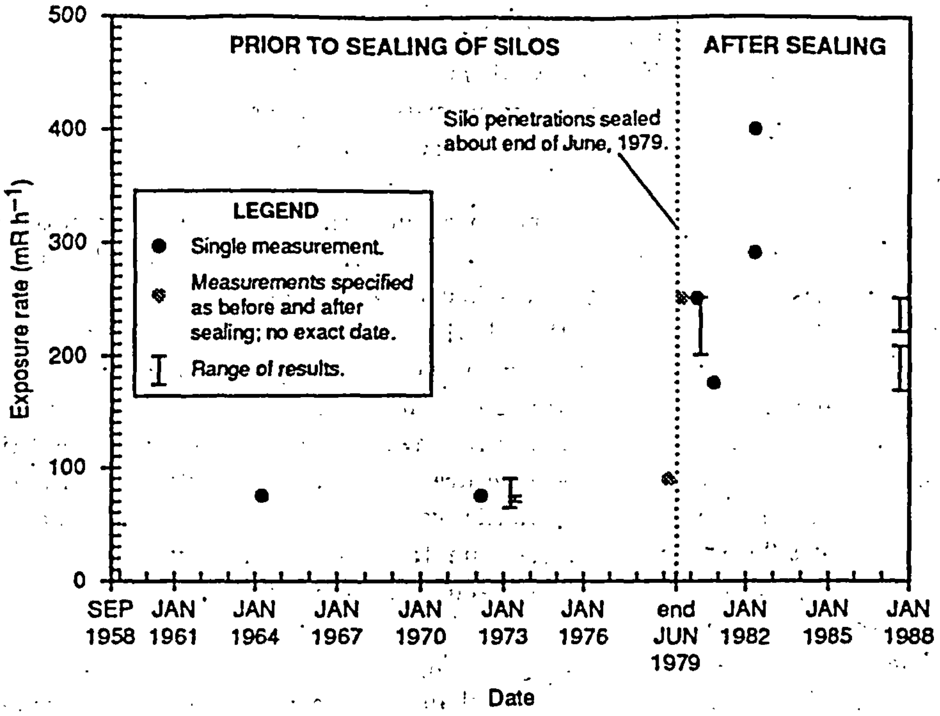

Figure J–4. Contact exposure rate measurements on the K-65 Silo domes prior to and after sealing of Silo penetrations.

considering the uncertainty and response characteristics of typical survey instruments. These measurements ranged from 168 to 400 mR h^{-1} (not including the measurements after operation of the Radon Treatment System (RTS)).

The data taken after operation of the RTS, in November 1987, can be used to estimate the "background" exposure rate due to sources other than the ^{222}Rn in the silo air. As discussed earlier, the RTS is a system to remove Rn and potential daughter products from the silo air space (see page J–28).

The RTS was operated in November 1987, prior to the installation of a foam layer on the silo domes (Grumski and Shanks 1988). The system operated on one silo at a time, with a flow rate of about 1000 ft^3 min^{-1}, and was operated until radiation levels on the dome surface stopped decreasing (Grumski and Shanks 1988). With this flow rate and an average nominal silo volume of 51,000 ft^3, the ventilation rate was (1000 ft^3 min^{-1})/(51000 ft^3) = 0.020 min^{-1}, or 1.2 h^{-1}. The exposure rate measurements were taken during operation of the RTS, but after it had been operating 4.6 h for Silo 1 and after 3.5 h for Silo 2 (Grumski and Shanks 1988). With these flow rate and operating times, and an assumed removal efficiency close to 100%, the ^{222}Rn concentrations in the silo air space should have been reduced to less than 3% of the initial concentrations. Also, in this operating time, any ^{222}Rn daughter radioactivity deposited on surfaces in the silos would have decayed to less than 2% of its original activity.

Thus, for this analysis, the exposure rate measurements made after operation of the RTS are considered to represent the "background" exposure rate, in the absence of ^{222}Rn daughters in the silo air. This background is primarily due to radiation from the radioactivity contained in the K-65 material in the silos (including trapped ^{222}Rn daughters). This set of measurements consists of four measurements, regularly spaced, on each silo dome. The range of the eight measurements was from 35.5 to 76 mR h^{-1} (Grumski and Shanks 1988).

For all of these exposure rate measurements, there is uncertainty in the results due to lack of knowledge about what instruments were used, and how the instruments were calibrated. It is noted that most survey instruments tend to have biases at varying energies, because their response varies with radiation energy. In addition, the measurements of the gross exposure rate before and after the vent sealing have uncertainties due to lack of knowledge about the exact measurement locations. These uncertainties combine to produce uncertainty about the comparability of the measurements made at different times. It seems reasonable that the true average exposure rates would lie within the range of measured values. We thus assume that the potential values of X_{pre}, X_{post}, and X_{bkg} are all represented by uniform distributions, with ranges equal to the observed ranges.

Thus, X_{bkg} is considered to be represented by a uniform distribution with minimum 35.5 mR h^{-1} and maximum 76 mR h^{-1}.

The range to be used for X_{pre} overlaps the range of X_{bkg}, which could result in calculated values of $C_{a,pre}$ that are less than zero. To correct this, the distribution used for X_{pre} is a uniform distribution with a minimum that is the greater of 65 mR h^{-1} and X_{bkg}. This ensures that X_{pre} is always at least as great as X_{bkg}. The maximum value of the distribution is 90 mR h^{-1}.

And, X_{post} is considered to be represented by a uniform distribution with minimum 168 mR h^{-1} and maximum 400 mR h^{-1}.

Table J–20 summarizes the frequency distributions of the calculations associated with predicted total Rn releases for 1959–1979. Included are the calculated Rn concentration in the Silos; the sum of the rate constants for releases by air exchange and diffusion; the fraction of the total removal of Rn from the Silos that occurs through release to the outside air through air exchange and diffusion (the rest is "removed" by radioactive decay), $[(\lambda_v+\lambda_d)/\lambda_{eff}]_{pre}$; and the Rn release rate. The results for the fraction $[(\lambda_v+\lambda_d)/\lambda_{eff}]_{pre}$ indicate that, for this period, almost all of the Rn released into the Silos from the K-65 material is released to the outside air.

Table J–20. Summary of Frequency Distributions of Calculations of Total Radon Release Rates from the K-65 Silos for 1959–1979

Parameter	Units	Percentiles of distributions				
		5th	25th	median	75th	95th
$\lambda_{v,pre}+\lambda_{d,pre}$	d^{-1}	0.83	1.5	2.4	4.3	16
$[(\lambda_v+\lambda_d)/\lambda_{eff}]_{pre}$		0.82	0.89	0.93	0.96	0.99
$C_{a,pre}$	pCi L^{-1}	3.9×10^5	1.4×10^6	2.5×10^6	3.8×10^6	6.3×10^6
Q_{pre}	Ci y^{-1}	4200	5300	6200	7200	8700

Model for 1988 Releases from K-65 Silos

At the end of 1987, the foam layer was applied to the K-65 Silo domes (see page J–5). It is expected that the foam provides significant insulation and therefore reduces the magnitude of the temperature cycling of the head space air. To estimate Rn releases for 1988, we first perform a preliminary calculation, using the same methodology as was used for releases for the 1980–1987 period, but using temperature change data specific to the period after the foam layer was installed. Air exchange releases are calculated based on a head space ventilation rate, which is estimated from head space temperature monitoring data. For the preliminary calculation, diffusion releases are assumed to be equal to diffusion releases for the 1980–1987 period. Thus,

$$Q_{1988,\mathrm{prelim}} = Q_{\mathrm{exch},1988} + Q_{\mathrm{diff}} \tag{J–40}$$

Preliminary calculation of 1988 releases. The calculation of air exchange releases for 1980–1987 are discussed earlier in this Appendix (page J–27). We use the same equation to calculate air exchange releases for 1988:

$$Q_{\mathrm{exch},1988} = C_{\mathrm{a},1988}\lambda_{\mathrm{v},1988}V_0 \tag{J–41}$$

with the same variables as before, except the "1988" subscript indicates parameters for 1988.

In the FMPC environmental restoration work, Operable Unit 4 includes the waste storage silos. Conversations with Operable Unit 4 staff indicated that no measurements were made of the Rn concentration in the K-65 Silo head spaces during the period 1988–1991. At the end of 1991, a layer of bentonite was added on top of the K-65 material inside the Silos (WEMCO 1992). After this addition, Rn monitoring of the head spaces was initiated. However, the bentonite significantly reduces the Rn concentrations so that concentrations after the bentonite was added are not representative of concentrations for the 1988–1991 period.

In the absence of measurements of the Rn concentration for 1988–1991, we assume that the Rn concentration for the 1980–1987 period would not have changed significantly, and can be used as a substitute. For 1980–1987, calculation results (see page J–37) indicated that the rate of Rn release (by air exchange plus diffusion) is small relative to the rate of Rn decay in the head space air. If the releases were smaller, as expected for 1988, the Rn concentration would only increase slightly. Thus, this seems to be a reasonable first approximation. So, we assume the distribution of the Rn concentration in the head space, $C_{\mathrm{a},1988}$, is a normal distribution, with mean 2.62×10^7 pCi L^{-1} and standard deviation 4.1×10^6 pCi L^{-1} (for previous discussion, for 1980–1987, see page J–28).

Since no material was added to the inside of the Silos in 1988, the head space volume is assumed to be the same as that used for the 1980–1987 calculation (see page J–29). Thus, V_0 is considered to have a uniform distribution, with minimum 40,000 ft^3, and maximum 62,000 ft^3.

For the silo ventilation rate, $\lambda_{\mathrm{v},1988}$, we use the same methods as for the period 1980–1987 (see page J–30). Thus, from equation J–17:

$$\lambda_{v,1988} = \lambda_{v,\Delta T,1988} + \lambda_{v,wind,1988} \qquad (J–42)$$

where the variables are as used before, except that the subscript "1988" indicates parameters for 1988. Also, from equation J–20:

$$\lambda_{v,\Delta T,1988} = (\Delta T/T_0)_{1988} \qquad (J–43)$$

where

ΔT = the increase (only) in temperature of silo head space air per day (K d^{-1}), and

T_0 = the initial temperature of the silo air (K).

Monitoring of the temperature of the K-65 Silos' head spaces was not instituted until October 1991. Thus, the data for October 1991 are the only data representative of the 1988–1991 period, since the bentonite layer was added inside the Silos in November 1991. The data from October 1991 were obtained from the FMPC (Byrne 1992c). Since we again have data for only a small part of a year, daily silo values of $\Delta T/T_0$ are correlated to daily temperature changes at the Cincinnati airport. Then, from the average temperature changes at the Cincinnati airport, the correlation can be used to estimate the average value of $\Delta T/T_0$ for the Silos.

The temperature monitoring data obtained to perform the correlation are shown in Table J–21. For some of the days not shown, some temperature data were available, but the data were not complete enough to allow determinations of the daily temperature increase. As shown, values of $\Delta T/T_0$ are first calculated for each Silo for each day, and then an average value is calculated for each day.

Records of the hourly temperature at the Cincinnati airport were obtained as part of the meteorological data set (NCDC 1991). The maximum temperature, T_{max}, and minimum temperature, T_{min}, were extracted for each of the 19 days in October 1991 for which Silo temperature data are available. These data, and the difference, $T_{max} - T_{min}$, calculated for the correlation, are shown in Table J–22.

A linear correlation of the average values of $\Delta T/T_0$ (dependent variable) to the Cincinnati airport temperature difference (independent variable), $T_{max} - T_{min}$, was performed using a least squares regression. The regression coefficient is $R = 0.56$. The regression line is given by:

$$\Delta T/T_0\,(d^{-1}) = \left(5.96\times10^{-5}\ {}^\circ F^{-1}\ d^{-1}\right)\times\left(T_{max} - T_{min}({}^\circ F)\right) - 1.31\times10^{-4}\ d^{-1} \qquad (J–44)$$

For this regression line, the standard error of estimate, $S_{Y|X}$, is 6.17×10^{-4} d^{-1}.

As for the 1980–1987 period, the average daily difference (average of $(T_{max} - T_{min})$) for Cincinnati airport is assumed to be 19.46 °F. Thus, the annual average value of $(\Delta T/T_0)_{1988}$, and thus $\lambda_{v,\Delta T,1988}$ is estimated from the regression line and the average daily difference as:

$$\begin{aligned}\lambda_{v,\Delta T,1988} &= (\Delta T/T_0)_{1988}\\ &= \left(5.96\times10^{-5}\ {}^\circ F^{-1}\ d^{-1}\right)\times(19.46\ {}^\circ F) + 1.31\times10^{-4}\ d^{-1} \qquad (J–45)\\ &= 0.00129\ d^{-1}\end{aligned}$$

Table J-21. K-65 Silo Values of $\Delta T/T_0$ for 1988–1991 Period; Data Used for a Linear Correlation[a]

	Silo 1 data[b]			Silo 2 data[b]			Average
Date	T_0 (°F)	ΔT (°F d^{-1})	$\Delta T/T_0$ (d^{-1})	T_0 (°F)	ΔT (°F d^{-1})	$\Delta T/T_0$ (d^{-1})	$\Delta T/T_0$ (d^{-1})
10/01/91	64.6	0.8	0.00153	64.2	0.7	0.00134	0.00143
10/02/91	64.7	0.7	0.00134	64.3	0.7	0.00134	0.00134
10/03/91	65.0	1.9	0.00362	64.5	0.7	0.00134	0.00248
10/08/91	60.7	0.8	0.00154	61.2	0.7	0.00134	0.00144
10/09/91	60.9	1.1	0.00211	61.1	1.5	0.00288	0.00250
10/10/91	61.9	0.3	0.000575	62.0	0.2	0.000384	0.000480
10/11/91	61.4	0.3	0.000576	61.7	0.2	0.000384	0.000480
10/12/91	60.6	0.4	0.000769	61.2	0.3	0.000576	0.000673
10/13/91	59.9	0.6	0.00116	60.8	0.4	0.000769	0.000962
10/14/91	59.6	0.2	0.000385	59.8	1.0	0.00193	0.00116
10/15/91	59.0	0.4	0.000772	59.3	1.9	0.00366	0.00222
10/16/91	58.2	0.5	0.000966	59.7	0.4	0.000771	0.000868
10/17/91	57.6	0.8	0.00155	59.3	1.9	0.00366	0.00261
10/18/91	57.9	1.2	0.00232	59.6	0.7	0.00135	0.00183
10/22/91	57.0	1.1	0.00213	58.8	0.7	0.00135	0.00174
10/24/91	58.8	0.6	0.00116	59.8	0.3	0.000578	0.000868
10/29/91	60.9	0.7	0.00135	60.9	0.7	0.00135	0.00135
10/30/91	61.5	0.3	0.000576	61.3	0.3	0.000576	0.000576
10/31/91	61.5	0.2	0.000384	61.3	0.2	0.000384	0.000384

[a] The values of ΔT and T_0 must be expressed in absolute temperature units (K) before the ratio is computed.
[b] T_0 and ΔT data obtained from Byrne (1992c).

It is assumed that the conditional distribution of $(\Delta T/T_0)_{1988}$, at the given value of $T_{max} - T_{min}$ = 19.46 °F, is a normal distribution with standard deviation $S_{Y|X}$. Thus, we consider the distribution of potential values of $(\Delta T/T_0)_{1988}$ to be a normal distribution with mean 0.00129 d^{-1} and standard deviation 0.000617 d^{-1}. However, with this mean and standard deviation, there is a significant chance that negative values of $(\Delta T/T_0)_{1988}$ might be selected from the distribution. Since such negative values are meaningless for the calculation of the ventilation rate, we truncate the distribution at 0, disallowing negative values.

As done for the 1980–1987 period, we assume the wind-induced ventilation of the Silo, $\lambda_{v,wind}$, has a value of zero. The addition of the foam layer to the Silo domes would tend to isolate the cracks in the domes, which provides additional support for this assumption.

Comparison of results of preliminary calculation with monitoring data. In the report of Task 5 of this Project (Shleien et al. 1993), we presented results of the FMPC Rn monitoring for locations on the fenceline of the K-65 Area. This monitoring was initiated in March 1987, and has continued to the present. Because the monitoring includes some time before the installation of the foam layer, as well as for the complete 1988–1991 period, it may be useful for comparison with the estimated source terms for 1980–1987 and 1988.

Table J–22. Temperature Difference at Cincinnati Airport for 1988–1991 Period; Data Used for a Linear Correlation[a]

Date	Maximum T_{max} (°F)	Minimum T_{min} (°F)	Difference $T_{max} - T_{min}$ (°F)
10/01/91	80	58	22
10/02/91	80	59	21
10/03/91	76	60	16
10/08/91	68	37	31
10/09/91	74	48	26
10/10/91	63	52	11
10/11/91	63	46	17
10/12/91	66	42	24
10/13/91	60	38	22
10/14/91	64	46	18
10/15/91	57	39	18
10/16/91	57	33	24
10/17/91	65	34	31
10/18/91	75	47	28
10/22/91	75	50	25
10/24/91	71	60	11
10/29/91	80	59	21
10/30/91	72	63	9
10/31/91	67	58	9

[a] T_{min} and T_{max} data obtained from NCDC (1991).

Descriptions of recent Rn monitoring locations (Byrne 1992b) indicate that monitoring has also been performed on or very near the K-65 Silo domes. The incident investigation report for the April 25, 1986, Rn release indicates that Rn monitoring in the area of the Silo domes had been performed in 1986 (DOE 1986). Based on this information and on discussions with FMPC staff, it appears that Rn concentrations in air on the rim of the K-65 Silo domes were measured for at least part of 1987 through 1991. However, we have been unable to obtain such data.

Results for the Rn monitoring on the K-65 Area fenceline are not provided in the annual environmental monitoring reports, but were obtained in computer spreadsheet files directly from the FMPC site (Byrne 1992a). The monitoring locations, called K65 A through K65 P (Byrne 1992b), are shown in Figure J–5, which also shows the approximate locations of "real-time" monitoring, discussed later. The monitoring was performed on a quarterly basis, using two types of alpha-track Rn detectors. As in the Task 5 Report, we only utilize the results from the Type F detectors, of which there were typically two used at each location. Table J1–1, in Annex 1 to this Appendix, provides the average measured Rn concentration for each quarter of monitoring for each location.

We would like to examine the average Rn concentrations for the two periods 1980–1987 (before foam was applied) and 1988–1991 (after foam, but before bentonite added). Since the foam layer was added to the K-65 Silo domes in December 1987, the fourth quarter of 1987 spans the two periods, and we do not consider it representative of the 1980–1987 period.

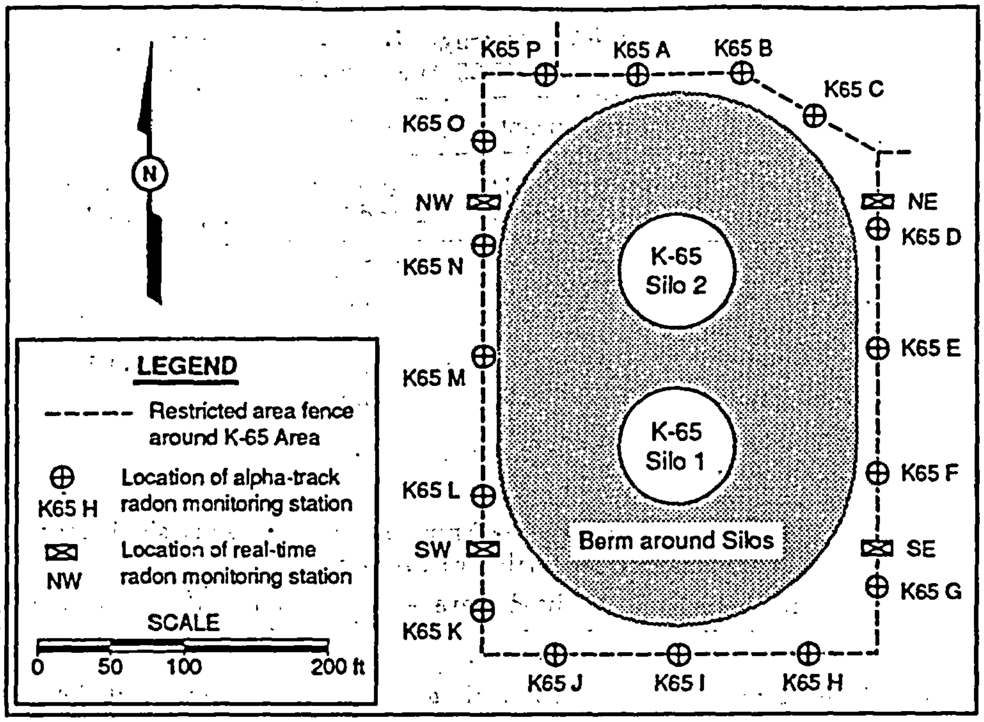

Figure J–5. Locations of FMPC Rn monitoring on the fenceline of the K-65 Area, for 1987–1991. Locations of the routine, alpha-track stations were obtained from Byrne (1992b). The locations of the real-time monitoring instruments are approximate and may have changed (especially the NE and SE locations) over this period (Grumski 1987b; Grumski and Shanks 1988; Byrne 1992b).

This leaves only the second and third quarters of 1987 to represent the first period. Based on the average quarterly measurements, there does not appear to be a consistent annual trend in the concentrations, although there is significant variability from quarter-to-quarter and year-to-year. We calculate the ratios of average concentrations for quarters two and three to average concentrations for the year, for the years 1988–1991, and then use the information to estimate an annual average concentration for 1987 and for 1980–1987.

Table J–23 shows the average concentrations for quarters two and three, the annual average concentrations, and the ratios, based on the data given in Table J1–1 (Annex 1 of this Appendix). The mean of the ratios is 1.21. The annual average for 1987 is thus estimated to be (7.39 pCi L^{-1}) × 1.21 = 8.94 pCi L^{-1}. Since 1987 is the only year of this monitoring during 1980–1987, the average concentration for 1980–1987 is also estimated to be 8.94 pCi L^{-1}. The ratio of the average Rn concentration on the K-65 Area fenceline for 1988–1991 to that for 1980–1987 is thus 5.47/8.94 = 0.61.

Later in this section (see Table J–24), the distributions of calculated results are presented. The distribution of the ratio of $Q_{1988,\mathrm{prelim}}$ to Q_{post} has a median of 0.18, and a 90% probability interval (5th to 95th percentile) of 0.092 to 0.47. This distribution of the ratios of the predicted source terms differs significantly from the estimated ratio of the measured Rn concentrations on the K-65 Area fenceline. The median ratio of source terms is

Table J–23: Average Rn Concentrations on the K-65 Area Fenceline

	Rn concentration (pCi L^{-1})		
Year	Annual average (all four quarters)[b]	Quarters 2 and 3 only	Ratio of annual to quarters 2 and 3
1987		7.39	
1988	6.54	4.67	1.40
1989	5.06	3.92	1.29
1990	2.52	2.36	1.07
1991	7.34	6.82	1.08
mean 1988–1991	5.47		

[a] We do not imply that three figures in the results are significant; they are retained for further calculations.

[b] Values are time-weighted averages, based on the monitoring dates.

a factor of about three less than the ratio of Rn concentrations, and the 90% probability interval of the ratios of source terms does not include the ratio of the Rn concentrations. This seems to indicate that our preliminary calculations significantly underestimate Rn releases for 1988.

There are also some additional data that provide weak evidence that the nature of the Rn releases from the K-65 Silos may have changed with the installation of the foam layer and installation of the Radon Treatment System (RTS), which was installed before the foam layer, to provide the capability for reducing Rn levels in the Silo head spaces. Some "real-time," or "continuous," monitoring of Rn concentrations on the K-65 Area fenceline has been performed by the FMPC. This monitoring uses instruments to continually make short-term measurements of the Rn concentration, with results typically reported as hourly averages. Prior to and during the work associated with the installation of the RTS and the foam layer, real-time measurements were made for a small number of days in November and December 1987 (Grumski 1987b; Grumski and Shanks 1988). Routine real-time measurements were apparently initiated in 1988 (Byrne 1992a), and we have obtained detailed results for October 1991 (Byrne 1992b). The Rn monitoring instruments were located toward the northeast, northwest, southeast, and southwest corners of the K-65 Area fenceline (Figure J–5).

Six days of measurements were made in early November 1987 prior to the installation of the RTS (Grumski 1987b; Grumski and Shanks 1988). These measurements showed prominent peak Rn concentrations, of from 15 to 225 pCi L^{-1}, that occurred during daylight and early evening hours, primarily from 10 am to 8 pm. At other times of the day, concentrations were relatively stable, and were less than 10 pCi L^{-1}. This timing of the peak concentrations is consistent with major Rn releases due to the thermal expansion of head space air during daylight hours, and subsequent air exchange release, as developed for the 1980–1987 period.

The monitoring in October 1991, prior to the addition of the bentonite layer in the Silos, covered the entire month, though data were not always available for all four monitors (Byrne 1992b). Similar to the November 1987 measurements, these data also showed prominent peak Rn concentrations, to as high as 240 pCi L^{-1}. However, these peaks occurred primarily during late night and morning hours, from 9 pm to 9 am. This timing of the peak concentrations is inconsistent with major Rn releases due to thermal expansion of head space air (this is expected, since the temperature increases were significantly reduced in this period). The peak concentrations during the late night and morning hours are significantly higher than the concentrations during the same hours in early November 1987. This seems inconsistent with a continuation or reduction of the same types of releases (air exchange and diffusion) that occurred in 1980–1987, and indicates that perhaps releases after the installation of the RTS and the foam layer are through a different release mechanism. Discussions with the FMPC Operable Unit 4 staff have not resulted in any explanation for this difference in results between early November 1987 and October 1991.

Current estimates of 1988 releases using Rn monitoring data. Because the preliminary method seems to significantly underestimate Rn releases for 1988, and seems inconsistent with the real-time Rn monitoring data, we will instead base current estimates of releases for 1988 on estimated releases for 1980–1987 and the ratio of Rn concentrations on the fenceline of the K-65 Area for the two periods. That is:

$$Q_{1988} = Q_{post} R_{mon} \tag{J–46}$$

where Q_{1988} is the total Rn release rate for 1988, Q_{post} is the total Rn release rate for the 1980–1987 period, and R_{mon} is the estimated ratio of the average long-term Rn concentration on the K-65 Area fenceline for the 1988–1991 period to the average long-term concentration for the 1980–1987 period. We assume that the average values of χ/Q (the ratio of air concentration at a receptor to release rate) on the K-65 Area fenceline are the same for 1988 as for the 1980–1987 period. This seems reasonable since the nature, and the timing, of the releases for 1988 are not understood.

To estimate the ratio, R_{mon}, we use:

$$R_{mon} = \frac{C_{f,88-91}}{C_{f,87} R_{ann:2\&3} R_{long:ann}} \tag{J–47}$$

where

$C_{f,88-91}$ = the average Rn concentration on the K-65 Area fenceline for the period 1988–1991,

$C_{f,87}$ = the average Rn concentration in air on the K-65 Area fenceline for the second and third quarters of 1987,

$R_{ann:2\&3}$ = the average ratio of annual average Rn concentrations on the K-65 Area fenceline to average concentrations for the second and third quarters of the year, and

$R_{long:ann}$ = a factor to incorporate the additional uncertainty in a long-term average Rn concentration on the K-65 Area fenceline based on the average for only one year.

The average Rn concentration for 1988–1991, $C_{f,88-91}$, was shown in Table J–23. We assume this average quantity would have a normal distribution. The uncertainty in this value is estimated based on the year-to-year variability, as seen in the annual averages for 1988 through 1991. When the four individual annual averages are considered, the coefficient of variation is 39.5%. Since there are four years of data, the relative standard error of the mean is estimated to be 19.8%. Thus, $C_{f,88-91}$ is assumed to have a normal distribution, with mean 5.47 pCi L^{-1}, and standard deviation 1.08 pCi L^{-1}.

The average Rn concentration for the second and third quarters of 1987, as seen in Table J–23, is 7.39 pCi L^{-1}. The uncertainty is incorporated through the remaining terms.

Also shown in Table J–23 are the ratios of annual average concentrations of Rn on the K-65 Area fenceline to average concentrations for the second and third quarters only. The mean and standard deviation of these ratios are 1.21 and 0.16, respectively. Since $R_{ann:2\&3}$ is an average quantity, it is assumed to have a normal distribution, with these values of mean and standard deviation.

As discussed above, the year-to-year variability of the average Rn concentrations on the K-65 Area fenceline is described by a coefficient of variation of 39.5%. For the 1980–1987 period, only one year of monitoring, 1987, is available. Thus, $R_{long:ann}$ is assumed to have a normal distribution with mean 1.00 and standard deviation 0.395.

To calculate R_{mon}, we use the standard error propagation formula for products or quotients of independent variables with errors that are small and symmetric about zero. This results in an estimate of R_{mon} with value 0.612 and standard deviation 0.282, with distribution assumed to also be normal.

Calculation results for 1988. Table J–24 summarizes the frequency distributions of the calculations associated with predicted total Rn releases for 1988. The results to be used as our current estimates of releases are the values for Q_{1988}. Because of the added uncertainty in the Rn monitoring results, those results have large associated uncertainties.

Table J–24. Summary of Frequency Distributions of Calculations of Total Radon Release Rates from the K-65 Silos for 1988

		Percentiles of distributions				
Parameter	Units	5th	25th	median	75th	95th
$Q_{exch,1988}$	Ci y^{-1}	9.8	23	35	48	69
$Q_{1988,prelim}$	Ci y^{-1}	98	140	170	210	280
$Q_{1988,prelim}/Q_{post}$		0.092	0.14	0.18	0.26	0.47
Q_{1988}	Ci y^{-1}	120	320	540	810	1300

Models for 1952–1958 Releases from K-65 Silos

The disposal history of the K-65 Silos was discussed earlier in this Appendix (see page J–4). Disposal of K-65 material in Silo 1 began July 19, 1952 (Davis 1952). From information in Strattman (1953) we estimated that Silo 1 was full around the middle of June 1953.

Disposal of K-65 material into Silo 2 was completed and the Silo was decanted in September 1958 (Noyes 1958; NLCO 1962). For our calculations, we assume the three start and completion dates were all in the middle of the month.

Model for releases. For the operational period of the K-65 Silos, mid-July 1952 to mid-September 1958, we have not obtained any contemporary radiological monitoring data that could be used to calculate Rn releases. We base estimates of Rn releases for this period on the estimated releases from the Silos for the 1959–1979 period. After the filling of Silo 1 had been completed, we assume it was decanted. This seems reasonable since some decanting proceeded automatically through the drawoff ports in the walls of the Silos (Dougherty and Jennings circa 1951), and we assume that it would have been desirable to remove the excess slurry liquor from the Silo, to recycle it for other purposes. For this period the piping, including the six-inch diameter gooseneck vent, would have been the same as in the 1959–1979 period. Thus, for the period after the filling of Silo 1 was completed, the Rn releases from Silo 1 can be assumed to be essentially the same as releases for 1959–1979.

For the time when the Silos were being filled, Rn releases are expected to be significantly different from releases after decanting was completed. An operating manual for the K-65 area indicates that the K-65 material was batch transferred to the Silos as a slurry (Dougherty and Jennings circa 1951). As mentioned above, this manual also indicates that some decanting of the Silos occurred automatically, through drawoff ports spaced every six inches up Silo walls, as the liquid level in the Silos passed each port level. While the Silos were operational, it thus appears that part of the time the K-65 material would have been essentially saturated with water, but with no water covering the material. And, part of the time the K-65 material would have been covered with a layer of water a few inches deep. Thus for the operational period the Rn releases would be reduced, relative to those for the 1959–1979 period, due to the quantities of water in and above the K-65 material, which would reduce the diffusion of Rn from the K-65 material.

From the most recent sampling of the Silo contents, in 1991, it appears that the ^{226}Ra concentrations of the two Silos may be significantly different (see Table J–5). The releases for 1959–1979 were based on average characteristics of the two Silos (though not explicitly involving ^{226}Ra concentration). For the time period under consideration here, the difference in concentrations between the two Silos will be accounted for.

Based on the above considerations, we employ the following simple model to estimate releases for these operational periods, based on releases for the 1959–1979 period. Here the Rn release rate, Q_{52-53}, is for the period mid-June 1952 through mid-July 1953, the operational period of Silo 1. The Rn release rate, Q_{53-58}, is for the period mid-July 1953 through mid-September 1958, the operational period for Silo 2.

$$Q_{52-53} = (0.5) Q_{pre} f_{Ra,1} f_{op} \tag{J–48}$$

$$Q_{53-58} = (0.5) Q_{pre} \left(f_{Ra,1} + f_{Ra,2} f_{op} \right) \tag{J–49}$$

where

Q_{pre} = total rate of Rn releases from the two K-65 Silos for the 1959–1979 period, as calculated earlier in this Appendix (see page J–41),

$f_{Ra,1}$ = ratio of the average ^{226}Ra concentration in K-65 material of Silo 1 to the average concentration for the two Silos,

$f_{Ra,2}$ = ratio of the average ^{226}Ra concentration in K-65 material of Silo 2 to the average concentration for the two Silos,

f_{op} = factor to account for the reduced Silo Rn emissions in the operational phase, relative to emissions for the post-operational period (1959–1979), due to the greater quantities of water present, and

(0.5) = factor to convert the Q_{pre} release rate for two Silos to a release rate for a single Silo.

Parameter distributions. For the Rn release rate for 1959–1979, Q_{pre}, we use the exact distribution of values calculated previously.

To calculate the ratios of ^{226}Ra concentrations, we use the measurement results compiled in Table J–5. For Silo 1, the average concentration was 525,000 pCi g^{-1}, with a standard deviation of 158,000 pCi g^{-1}, for a sample size of 12. Thus, the standard error of this mean is 45,600 pCi g^{-1}. For Silo 2, the average was 299,000 pCi g^{-1}, with standard deviation of 119,000 pCi g^{-1}, for a sample size of 11, which results in a standard error of the mean of 35,900 pCi g^{-1}. And, for the two Silos, the average and standard deviation were 417,000 and 179,000 pCi g^{-1}, for a sample size of 23, which gives a standard error of the mean of 37,300 pCi g^{-1}. To calculate the ratios $f_{Ra,1}$ and $f_{Ra,2}$, we use the standard error propagation formula for quotients of independent variables, and the standard errors of the means for the uncertainty terms. It is recognized that the average concentration for both Silos is not independent from the averages for each Silo, but we consider the formula to be an acceptable approximation. The distributions for the ratios are considered to be normal. This results in a distribution for $f_{Ra,1}$ with mean 1.26 and standard deviation (standard error of the mean) 0.157. And, for $f_{Ra,2}$ the mean is 0.717 and the standard deviation is 0.107.

The determination of an appropriate distribution for f_{op} is more difficult. The amount of water that might cover the K-65 material in the Silos is unknown. The operating practices, especially the typical timing of slurrying, flushing, and decanting operations, are also unknown. We assume that a reduction factor (f_{op}) of around 0.5, relative to releases for the decanted, post-operational period, is reasonable. The uncertainty is very large, so we assume that f_{op} is represented by a uniform distribution, with minimum 0 and maximum 1.

Results of calculations. The frequency distributions for calculated values of the Rn release rates Q_{52-53} and Q_{53-58} are summarized in Table J–25. Due to the large uncertainty in the reduction factor f_{op}, the distribution of results for the 1952–1953 period is very broad. For this period, the 90% probability interval has a range of a factor of about 20. For the 1953–1958 period, the releases are dominated by the decanted Silo 1, for which the uncertainty is significantly lower. Thus, for this period, the distribution of results is much tighter than that for the 1952–1953 period.

Model for 1951-1953 Releases from Drummed K-65 Material

In Table J–2 it was shown that the Mallinckrodt Chemical Works (MCW), in St. Louis, began shipping drummed K-65 material to the FMPC in September 1951, about ten months before construction of the K-65 Silos was complete. This material was thus stored onsite

Table J–25. Summary of Predicted Rn Release Rates (Ci y^{-1}) from the K-65 Silos for the Operational Period of the Silos, 1952–1958

Period	Percentiles of distribution				
	5th	25th	median	75th	95th
mid-July 1952–mid-June 1953	200	940	1900	2900	4200
mid-June 1953–mid-September 1958	3100	4100	4900	5900	7600

until it could be placed in the Silos. In this section we evaluate Rn releases from this stored, drummed K-65 material.

Description of drummed K-65 material. Walden (1952) indicates that 12,997 drums of K-65 material were received at the FMPC in the period September 25, 1951, to July 31, 1952. A U.S. Atomic Energy Commission (AEC) letter indicates that the drummed K-65 material was to be temporarily stored on the concrete ore storage pad until the K-65 Silos were completed and ready for operations (Belmore 1951). We understand this pad to be the large concrete pad around, but generally north of, Plant 1. The location of this storage pad is shown in Figure J–6. An original operating manual for the K-65 storage area indicates the K-65 material was to be delivered to the FMPC in 55-gallon drums, each containing about 500 pounds of material (Dougherty and Jennings circa 1951). This manual also indicated the material would have a bulk density of about 90 lb ft^{-3}, and would have moisture content about 40 weight percent. Another procedures manual corroborates the weight of material contained in each drum, and also indicates that the 55-gallon drums were sealed with lids when they arrived at the FMPC (Consiglio 1952).

An internal FMPC memorandum describes the status of the K-65 Silos as of November 1953 (Strattman 1953). At that time Silo 1 was full, and filling of Silo 2 had been proceeding for some time. The rate of receipt of drummed K-65 material from MCW had slowed significantly from the apparent rate in 1951 and the first half of 1952 (from Walden 1952). There was no indication of an onsite (FMPC) inventory of drummed K-65 material. We thus assume that by the time Silo 1 was full, which we estimated occurred in June 1952, drummed K-65 material was placed in the Silos shortly after receipt at the FMPC, and so the quantity of drums stored onsite was negligible after this time. We thus calculate Rn releases from stored, drummed K-65 material for the period September 25, 1951, to about mid-June, 1953. We also assume that all of the stored K-65 material, from this period, was eventually placed into Silo 1.

Model for releases. For Rn releases from the drummed K-65 material, we use the conventional methodology for releases from bulk quantities of ^{226}Ra-bearing material. This methodology is thoroughly described later in this Appendix, in the section regarding the alternative calculation of Rn releases from the K-65 Silos (see page J–73). Because we have no information about the Rn concentration in the air space of the drums, we assume that the concentration is negligible in terms of constraining the release of Rn from the K-65 material into the air space of the drum. This results in a slight upward bias in our estimates of releases, but seems reasonable for the limited data available. We thus use the form of

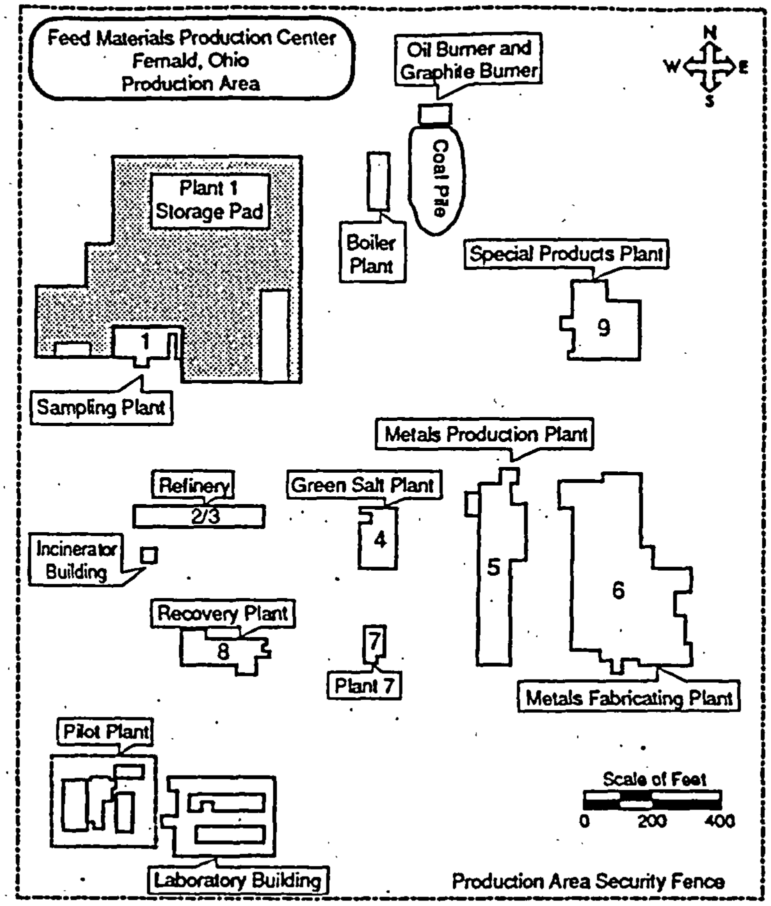

Figure J–6. Location of the storage pad, around Plant 1 in the production area, where drummed K-65 material was stored before placement in the K-65 Silos.

equation J–68 (originally from Collé et al. 1981) to calculate the unconstrained Rn flux from a bare layer of K-65 material, with an impervious layer (the bottom of the drum) below it:

$$j_{D,dr} = \sqrt{\frac{D_{e,dr}\varepsilon_{dr}}{\lambda_{Rn}}}(\phi_{dr})\tanh\left(\frac{L_{dr}}{l_{dr}}\right) \tag{J–50}$$

where the subscript "dr" generally refers to the drummed K-65 material, and:

$j_{D,dr}$ = unconstrained diffusion fluence rate of Rn (Rn flux). The quantity of Rn per unit time per unit area transported by diffusion from the source material (in this case the drummed K-65 material) into the ambient air (pCi m^{-2} s^{-1}, or similar).

$D_{e,dr}$ = effective bulk diffusion coefficient of Rn through the porous source material (cm^2 s^{-1}, or similar).

ε_{dr} = porosity of the source material.

ϕ_{dr} = the pore space Rn production rate. Quantity of Rn produced in pore spaces of the source material per unit time per unit pore volume that is free to migrate through the pores of the material (pCi cm^{-3} s^{-1}). Depends on characteristics of the source material, including ^{226}Ra concentration, Rn emanation fraction, bulk density, and porosity, and on the Rn decay constant.

L_{dr} = thickness of the source material (cm, or similar).

l_{dr} = Rn diffusion length in the source material (related to $D_{e,dr}$) (cm, or similar).

λ_{Rn} = the decay constant for ^{222}Rn.

For this case of the drummed material, the thickness of the source material, L_{dr}, will be less than 100 cm, and the tanh term in the equation will be significantly different from unity. This term will be retained in the equation (unlike was done in the alternative calculation).

As for the alternative calculation of Silo Rn releases, the pore space Rn production rate can be calculated by (see equation J–71):

$$\phi_{dr} = \frac{[\mathrm{Ra}]_{dr} EF_{dr} \rho_{dr} \lambda_{Rn}}{\varepsilon_{dr}} \tag{J–51}$$

where:

$[\mathrm{Ra}]_{dr}$ = concentration of ^{226}Ra in the drummed K-65 material (activity per mass),

EF_{dr} = ^{222}Rn emanation fraction in drummed K-65 material, which is the fraction of the Rn formed (from the ^{226}Ra decay) that is in pore spaces and is free to migrate, and

ρ_{dr} = dry bulk density of drummed K-65 material (g cm^{-3}, or similar).

In this equation, the ^{226}Ra concentration gives the total production rate of ^{222}Rn atoms, per mass of source material. Multiplication by the Rn emanation fraction converts this to the production of Rn in the pore spaces. The factors of ρ_{dr} and ε_{dr} convert the basis from mass of source material to volume of pore space air. Finally, the decay constant converts the quantity of Rn from atoms to activity units.

The Rn release rate from a single drum, Q_{dr}, is then calculated as:

$$Q_{dr} = j_{D,dr} A_{dr} \left(\frac{\lambda_v + \lambda_d}{\lambda_{eff}} \right)_{dr} U_{dr} \tag{J–52}$$

where A_{dr} is the surface area of the K-65 material exposed to the air, and U_{dr} is an uncertainty factor to account for additional uncertainty related to application of this model to the drummed K-65 material. The ratio $[(\lambda_v + \lambda_d)/\lambda_{eff}]_{dr}$ is the fraction of Rn released from the K-65 material that is released (through the drum) into the environment. This ratio was discussed earlier, in relation to releases from the K-65 Silos for the 1959–1979 and 1979–1980 periods (see pages J–37 and J–45).

The total yearly Rn releases from the stored, drummed K-65 material can then be calculated from the release rate per drum and the time-integrated number of drums stored on the site:

$$R_{dr,i} = Q_{dr} N_i \tag{J–53}$$

where $R_{dr,i}$ is the quantity of Rn released (Ci) for the i^{th} year, of 1951–1953, and N_i is the time-integrated number of drums (units of drum months, or similar) stored on the FMPC Plant 1 pad for the i^{th} year.

Parameter distributions. As described above, we assume that essentially all of the stored, drummed K-65 material was eventually placed in Silo 1. Thus, the average ^{226}Ra concentration in the drummed K-65 material is assumed to be the same as the concentration measured (later) in Silo 1. As described earlier in this Appendix, the results from the 1991 sampling of the K-65 Silos are preferred over results from prior sampling episodes. The 1991 results are compiled in Table J–5. For Silo 1, the measured ^{226}Ra concentrations ranged from 306,800 to 890,700 pCi g^{-1}. Because the range of results is so broad, and because there may have been changes in the average concentration in the drummed material as a function of time, we assume that $[Ra]_{dr}$ has a uniform distribution, with minimum 306,800 pCi g^{-1} and maximum 890,700 pCi g^{-1}.

As discussed earlier in this Appendix (see page J–11), measurements of the Rn emanation fraction for the K-65 material have not been performed. In our discussion of the alternative calculation of Rn releases from the K-65 Silos, we conclude that emanation fraction measurements for uranium mill tailings, from the literature, are the best values to use, lacking results specific to the K-65 material (see page J–76). As in the alternative calculation, we assume here that the emanation fraction is within the range compiled by Rogers et al. (1984) for mill tailings. We thus assume that EF_{dr} has a uniform distribution, with minimum 0.1 and maximum 0.4.

As discussed above, an operating manual for the K-65 area indicated the K-65 material would have a bulk density of about 90 lb ft^{-3}, and contained about 40 weight percent moisture (Dougherty and Jennings circa 1951). We assume that the density is a *wet* bulk density, and that the moisture content is percent dry weight (most commonly used for weight percent moisture). This results in a calculated *dry* bulk density of 1.0 g cm^{-3}. This value is within the range seen in measurements of the K-65 material in the Silos (see Table J–4), and thus seems reasonable. However, since the value here was obtained only from an operating manual, and the basis of the value is not known, we assume (arbitrarily) a range of ± 20% about the value. Thus, the bulk density, ρ_{dr}, is assumed to have a uniform distribution, with minimum 0.8 g cm^{-3} and maximum 1.2 g cm^{-3}.

As discussed earlier in this Appendix (see page J–9), no measurements for porosity of the K-65 material have been reported. As in the alternative calculation of Silo Rn releases, we use measured values of the specific gravity, and the relation of porosity to specific gravity and density (see page J–77):

$$\varepsilon_{dr} = \frac{g_{dr} - \rho_{dr}}{g_{dr}} \qquad \text{(J–54)}$$

where g_{dr} is the specific gravity of the K-65 material, and where the density, ρ_{dr}, is expressed here as the numerical value (without units) corresponding to the density given in units of g cm^{-3}. We use the same mean specific gravity used in the alternative calculation (based on measurements reported in DOE 1990), but we double the standard deviation.

Thus, the mean specific gravity of the K-65 material, g_{dr}, is assumed to follow a normal distribution with mean 2.98 and standard deviation 0.24.

As discussed above, the moisture content of the drummed K-65 material was expected to be about 40% dry weight. The moisture content is an important parameter in the determination of the Rn diffusion coefficient, D_{dr} (see below). In our discussion of the moisture content of the K-65 material in the Silos, for the alternative calculation of releases (see page J–78), it appeared that the range of measured moisture contents was 21.8% to 90% dry weight. Based on that large range of values, it seems that the uncertainty in the moisture content for the drummed material might also be large. We thus assume that the range of moisture contents is 20% to 60% dry weight.

As in the alternative calculation, the moisture saturation fraction of the K-65 material, m_{dr}, can be related to the moisture content in dry weight fraction (dry weight percent divided by 100%), M_{dr}, from equation J–73 (Rogers et al. 1984):

$$m_{dr} = \frac{M_{dr}}{\frac{1}{\rho_{dr}} - \frac{1}{g_{dr}}} \qquad \text{(J–55)}$$

where ρ_{dr} and g_{dr} are the bulk density and specific gravity of the K-65 material, as used previously. Again, the density, ρ_{dr}, is expressed as the numerical value (without units) corresponding to the density given in units of g cm^{-3}.

For the distribution of M_{dr}, we assume a uniform distribution over the range 20% to 60% dry weight, or 0.20 to 0.60 dry weight fraction, with one constraint. The saturation fraction must be less than or equal to 1 so M_{dr} is constrained to (from equation J–74):

$$M_{dr} \leq \frac{1}{\rho_{dr}} - \frac{1}{g_{dr}} \qquad \text{(J–56)}$$

We thus consider M_{dr} to have a uniform distribution with minimum 0.20 dry weight fraction, and with maximum 0.60 or $(1/\rho_{dr} - 1/g_{dr})$, whichever is less.

No information was found on the Rn diffusion coefficient for the drummed K-65 material. As discussed earlier in this Appendix (see page J–11), measurements of the Rn diffusion coefficient in the K-65 material in the Silos have also not been made. We use the relationships used in the alternative calculation of Silo Rn releases (see page J–79 for details not repeated here).

Rogers et al. (1984) compile diffusion coefficients from about 200 measurements on various types of soils at various moisture saturations. For cases when little is known about the diffusion coefficient of a soil, they recommend the use of an empirical correlation with The pore space diffusion coefficient is estimated using the following empirical correlation (Rogers et al. 1984), based on the saturation fraction and porosity (from equation J–76):

$$\hat{D}_{dr} = \left(0.07\ \text{cm}^2\ \text{s}^{-1}\right)\exp\left[-4\left(m_{dr} - m_{dr}\varepsilon_{dr}^{\,2} + m_{dr}^{\,5}\right)\right] \qquad \text{(J–57)}$$

where $\hat{D}_{dr}$ is the empirically predicted pore space diffusion coefficient, m_{dr} is the saturation fraction, and ε_{dr} is the porosity.

The uncertainty in estimating the diffusion coefficient for a particular material (the correlation was based on many different soil types) is incorporated (from equation J–77) by:

$$D_{dr} = U_D \hat{D}_{dr} \tag{J–58}$$

where D_{dr} is the adjusted estimate of the pore space diffusion coefficient, that will be used for further calculations, and U_D is an uncertainty factor, represented by a lognormally-distributed random variable with geometric mean 1 and geometric standard deviation 2 (this results in a 95% confidence interval somewhat greater than the one order of magnitude estimated from Rogers et al. 1984). The diffusion coefficient, D_{dr}, is constrained to be less than or equal to 0.11 $cm^2\ s^{-1}$, the coefficient for pure air. The effective diffusion coefficient, $D_{e,dr}$, is then calculated using equation J–59 (from equation J–75).

$$D_{e,dr} = D_{dr}\varepsilon_{dr} \tag{J–59}$$

Based on equation J–79 (Collé et al. 1981), the Rn diffusion length, l_{dr}, is related to the diffusion coefficient by:

$$l_{dr} = \sqrt{\frac{D_{dr}}{\lambda_{Rn}}} \tag{J–60}$$

The surface area of K-65 material exposed to the air in each drum is just the horizontal cross-sectional area of the drum. Thus, for the 55-gallon drums, A_{dr} is about 0.25 m^2, or 2500 cm^2. The nominal weight of material in each drum is about 500 lb, as discussed above. The thickness of the K-65 material in the drum, L_{dr}, can be calculated by:

$$L_{dr} = \frac{W_{dr}}{\rho_{dr}(1+M_{dr})A_{dr}} \tag{J–61}$$

where W_{dr} is the wet weight of material in the drum. For this calculation, we assume that uncertainty in the weight per drum might be ± 20%. Thus, W_{dr} is assumed to have a uniform distribution, with minimum 400 lb and maximum 600 lb.

Information is not available to directly estimate the ratio $[(\lambda_v + \lambda_d)/\lambda_{eff}]_{dr}$. From the current estimates of releases for the period 1980–1987, the ratio $[(\lambda_v + \lambda_d)/\lambda_{eff}]_{post}$ was estimated for the "sealed" K-65 Silos. The sealed Silos for that time period still allowed releases of Rn through cracks and small penetrations in the Silo domes. For the drummed K-65 material, it seems reasonable that the metal, 55-gallon drums would not be airtight. We do not know how lids were installed on the drums, but leakage at the joint between the lid and the drum is expected, especially after transport to the FMPC, movement from railcars to a storage location on the Plant 1 pad, and outdoor storage. We think that fractional leakage from the drums would be less than the fractional leakage from the K-65 Silos. Thus, the ratio $[(\lambda_v + \lambda_d)/\lambda_{eff}]_{dr}$ is calculated by:

$$\left(\frac{\lambda_v + \lambda_d}{\lambda_{eff}}\right)_{dr} = \left(\frac{\lambda_v + \lambda_d}{\lambda_{eff}}\right)_{post} f_{dr} \tag{J–62}$$

where f_{dr} is a reduction factor for leakage from the drums. We think that a reduction factor of about 0.5, with very large associated uncertainty, is appropriate. We thus assume that f_{dr} has a uniform distribution, with minimum 0 and maximum 1.

From the previous calculations (results given in Table J–17), the ratio $[(\lambda_v + \lambda_d)/\lambda_{eff}]_{post}$ had results with fifth and 95th percentiles of 0.071 and 0.24, respectively. The distribution was relatively symmetric. To approximate this distribution, we assume the ratio $[(\lambda_v + \lambda_d)/\lambda_{eff}]_{post}$ has a normal distribution, with mean 0.156 and standard deviation 0.051.

It is felt that the uncertainty in the calculated Rn release rate from the drummed K-65 material has not been totally accounted for in the parameter uncertainties. For all parameters, except perhaps the ^{226}Ra concentration, there is very little directly applicable information to support the choice of parameter distributions. This is the reason for the application of the additional uncertainty factor, U_{dr}. We assume (somewhat arbitrarily) U_{dr} to have a lognormal distribution, with geometric mean 1 and standard deviation 1.4 (for a 95% probability interval, this gives an uncertainty of about $\times/\div$ 2).

The half life of ^{222}Rn is 3.8235 d (Walker et al. 1989). Thus, the decay constant for ^{222}Rn, λ_{Rn}, is 2.098×10^{-6} s^{-1}. For our calculations, this value is assumed to have negligible uncertainty.

To estimate the time-integrated number of stored drums of K-65 material, we create a simple model based on estimated receipt rates and production (dumping into the Silo) rate. Walden (1952) indicates that 12,997 drums of K-65 material were received at the FMPC during the period September 25, 1951–July 31, 1952. For this time period, we assume a constant receipt rate, which would be 41.8 drums d^{-1}. Davis (1952) indicates that dumping of drums, for slurrying to the K-65 Silo 1, began July 19, 1952. Strattman (1953) indicates that about 24,000 drums of K-65 material were placed in Silo 1. From other information in Strattman (1953), we estimated that Silo 1 was full about the middle of June 1952 (assumed June 15, 1952). For the period of filling Silo 1, we assume a constant production rate, which would be 72.3 drums d^{-1} dumped into Silo 1. As discussed earlier in this section, we assume that the onsite K-65 drum inventory had been reduced to negligible levels by the time Silo 1 was full. Thus, for the period August 1, 1952–June 15, 1953, about 11,000 additional drums of K-65 material are assumed to have been received. The receipt rate for this period would have then been 34.5 drums d^{-1}.

From these receipt and production rates, the inventory of drummed K-65 is estimated to be 12,456 drums on July 18, 1952, and 12,060 drums on July 31, 1952. From the constant rates of receipts and production, we estimate the onsite inventory of K-65 drums as a function of time, $n_{dr}(t)$, by the following model:

$$\begin{cases} \text{for } 9/25/51-7/18/52;\ t = \text{ days past } 9/24/51; & n_{dr}(t) = 41.8t \\ \text{for } 7/19/52-7/31/52;\ t = \text{ days past } 7/18/52; & n_{dr}(t) = 12{,}456 - 30.5t \\ \text{for } 8/1/52-6/15/53;\ t = \text{ days past } 7/31/52; & n_{dr}(t) = 12{,}060 - 37.8t \end{cases} \qquad \text{(J–63)}$$

From this model, the average monthly inventory of K-65 drums is calculated. For October 1951 through June 1953, the results are shown in Table J–26. (We assumed that the average inventory for September 1951 was zero.)

Table J–26. Estimated Monthly Average Inventory of Drummed K-65 Material Stored on the Plant 1 Pad

Month	Average number of drums	Month	Average number of drums	Month	Average number of drums
Oct 1951	880	May 1952	9,800	Dec 1952	6,900
Nov 1951	2,200	Jun 1952	11,000	Jan 1953	5,700
Dec 1951	3,400	Jul 1952	12,000	Feb 1953	4,500
Jan 1952	4,700	Aug 1952	11,000	Mar 1953	3,500
Feb 1952	6,000	Sep 1952	10,000	Apr 1953	2,300
Mar 1952	7,200	Oct 1952	9,200	May 1953	1,200
Apr 1952	8,500	Nov 1952	8,000	Jun 1953	130

The time-integrated numbers of drums stored are calculated by summing the average monthly inventories, from Table J–26. Thus, $N_{1951} = 6500$ drum-months, $N_{1952} = 110{,}000$ drum-months, and $N_{1953} = 17{,}000$ drum-months.

Results of calculations. Table J–27 summarizes the frequency distributions for intermediate, calculated parameters. Table J–28 shows the distributions for calculated Rn release quantities for 1951, 1952, and 1953 from drummed K-65 material stored on the Plant 1 pad.

Table J–27. Summary of Frequency Distributions of Intermediate Results: for Rn Releases from Drummed K-65 Material Stored on the Plant 1 Pad

Parameter	Units	Percentiles of distributions				
		5th	25th	median	75th	95th
ε_{dr}		0.58	0.63	0.66	0.70	0.73
m_{dr}	saturation fraction	0.31	0.45	0.59	0.73	0.92
D_{dr}	$cm^2\ s^{-1}$	5.1×10^{-4}	4.7×10^{-3}	1.3×10^{-2}	2.6×10^{-2}	6.0×10^{-2}
$D_{e,dr}$	$cm^2\ s^{-1}$	3.2×10^{-4}	3.0×10^{-3}	8.4×10^{-3}	1.8×10^{-2}	4.2×10^{-2}
l_{dr}	cm	16	47	78	110	170
L_{dr}	cm	49	58	65	74	89
$[(\lambda_v + \lambda_d)/\lambda_{eff}]_{dr}$		0.0064	0.034	0.070	0.11	0.18
ϕ_{dr}	$pCi\ cm^{-3}\ s^{-1}$	0.17	0.30	0.44	0.62	0.94
$j_{D,dr}$	$pCi\ cm^{-2}\ s^{-1}$	3.9	8.7	14	21	33
Q_{dr}	$Ci\ month^{-1}$	4.0×10^{-4}	2.3×10^{-3}	5.5×10^{-3}	1.1×10^{-2}	2.6×10^{-2}

As seen in Table J–28, predicted releases from the drummed K-65 material stored on the Plant 1 pad are much higher in 1952 than in 1951 and 1953. This occurs because the predicted inventory of stored drums peaked in July 1952, and because the inventory is assumed to be zero prior to October 1951 and after June 1953. We also note that the uncertainties in the predicted release quantities are very large. The 90% probability intervals (5th to 95th percentiles) have a range of a factor of about 70. This is not

Table J-28. Summary of Predicted Rn Release Quantities (Ci) from Drummed K-65 Material Stored on the Plant 1 Pad

	Percentiles of distributions				
Year	5th	25th	median	75th	95th
1951[a]	2.6	15	35	73	170
1952	42	240	580	1200	2800
1953[b]	6.9	39	95	190	450

[a] Releases for 1951 are assumed to have occurred in October through December.

[b] Releases for 1953 are assumed to have occurred in January through June.

unexpected, as a number of the parameters had significant associated uncertainties, primarily due to the extremely limited information available to describe the releases.

Model for Radon Daughter Releases

For releases of ^{222}Rn, the short-lived daughters of Rn are primarily responsible for the inhalation doses delivered, because the Rn does not remain in people's lungs for a significant length of time. With fairly short half-lives, the Rn daughters grow in to significant fractions of equilibrium within reasonable distances from the release point. However, for outdoor air at points close to the Rn release point, where the transport time is short enough, Rn daughters will only grow in to very small fractions of equilibrium. At these close-in locations, the direct releases of Rn daughters will be important to outdoor concentrations of Rn daughters, and thus to doses to people. No direct measurements have been made of Rn daughter releases or Rn daughter concentrations in the K-65 Silo air spaces. Rn daughters in the air around the FMPC have not been routinely monitored, though some measurements were made in the late 1970's (see our Task 5 report (Shleien et al. 1993), for more about the historical measurements).

Model for releases. For this assessment, a relatively crude estimate of the releases of the short-lived ^{222}Rn daughters, ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po is developed. We assume that the releases of the daughters are equal to the ^{222}Rn releases times correction factors:

$$Q_{daught} = QF_1F_2 \tag{J–64}$$

where Q_{daught} is the release rate of each of the four short-lived daughters, associated with the Rn release rate Q, the correction factor F_1 is the Rn daughter equilibrium fraction in the head space air, and F_2 is a fractional release factor, to account for deposition of Rn daughters along the release pathway (such as in the cracks or gooseneck vent), before reaching the atmosphere. This equation is applied to Rn daughter releases associated with each of the different Rn releases, both for releases from the K-65 Silos and for releases from the drummed K-65 material stored on the Plant 1 pad. In the case of releases from this drummed material, the same equation is used to calculate the release quantities, $R_{daught,dr}$ (instead of Q_{daught}), from the Rn release quantities R_{dr} (instead of Q).

Parameter distributions. In general, no data have been located that enable the estimation of the parameters F_1 and F_2. One study, conducted in 1993, may provide information to at least qualitatively corroborate the parameter distributions that we choose here. That study is discussed shortly.

No information about the fractional equilibrium of short-lived ^{222}Rn daughters in silo air has been found. The range of possible values of the equilibrium fraction is from 0 to 1. Much of the research into Rn daughters has been focused on homes and occupational environments. The range of equilibrium fractions measured in houses is at least from 0.1 to 0.9 (NCRP 1988). However, houses typically have ventilation rates between about 0.2 and 3 h^{-1} (Nazaroff and Nero 1988), which is much greater than the calculated ventilation rates of the silos ($\lambda_{v,post}$ nominally about 0.002 h^{-1} and $\lambda_{v,pre}$ nominally about 0.05 h^{-1}). It is known that the equilibrium fraction in an enclosed space increases with decreasing ventilation (NCRP 1989). However, at very low ventilation rates, low concentrations of condensation nuclei could lead to significantly increased unattached fractions of Rn daughters, and thus to increased deposition of daughters on surfaces and decreased equilibrium fraction (Nazaroff and Nero 1988). No direct information is available about concentrations of condensation nuclei in the head space air, but this is thought to be a lesser effect. The characteristics of the silo aerosols are not well enough understood to allow a useful model of the airborne concentrations of Rn daughters in the silos. However, it seems reasonable that the equilibrium fraction will be quite close to 1. We thus assume that F_1 follows a uniform distribution, with minimum 0.8 and maximum 1.0.

No information, either from FMPC-specific sources or from other sources, has been located relevant to the fractional release factor, F_2. We thus assume that uniform distributions would apply, and choose ranges for F_2 based on relative differences expected for the different release scenarios. Table J–29 shows the ranges chosen and the justifications.

In the summer of 1993 a pilot study at the FMPC included limited onsite, outdoor measurements of Rn equilibrium ratio (Paine 1994). The preliminary data indicate that the Rn equilibrium ratio may be about 50% or less for meteorological stability classes A, B, D, and E. The samples were apparently collected near the K-65 Silos. A copy of the study report was requested from the FMPC, but has not yet been obtained. If the Rn concentrations were significantly above background, so that essentially all the Rn was from K-65 Silo releases, the measured Rn equilibrium fraction would be representative of the product F_1F_2. Some provisional information was obtained through informal discussions with FMPC staff. It appears that the gross measured Rn concentrations are only somewhat elevated above background concentrations (Tomczak 1994), indicating contributions both from releases from the K-65 Silos and from background concentrations. Since the background concentrations were not measured (Tomczak 1994), the contributions from the Silo releases can not be determined. These measurements were made in 1993 and may be generally representative of conditions that existed in 1988, though the Rn release rate was lower in 1993. Because details (especially about background contributions) are lacking, we can only indicate at this time that the measurements are not inconsistent with our parameter choices, which for 1988 result in a nominal value of F_1F_2 of 0.22 (0.9×0.25).

Table J-29. Ranges of F_2 Chosen for Rn Daughter Calculations

Period	To calculate:	Use F_2:	Range of F_2	Reasoning
1951–1953	$R_{daught,dr,i}$	$F_{2,dr}$	0–0.5	Penetrations in drums, primarily at joint between lid and drum, are assumed to be very small in size, allowing for significant deposition.
1952–1953 and 1953–1958	$Q_{daught,52-53}$ $Q_{daught,53-58}$	$F_{2,52-58}$	0.8–1	Gooseneck vent and other dome penetrations were open to atmosphere, so free exchange means little deposition.
1959–1979	$Q_{daught,pre}$	$F_{2,pre}$	0.8–1	Gooseneck vent and other dome penetrations remained open to atmosphere, so free exchange means little deposition.
1980–1987	$Q_{daught,post,exch}$	$F_{2,post,exch}$	0.5–1	Exchange releases occurred primarily through dome penetrations. Major dome penetrations sealed, but cracks and small penetrations remained. Probably significant, but small amount of deposition.
1980–1987	$Q_{daught,post,diff}$	$F_{2,post,diff}$	0–0.5	Diffusion releases through concrete. Slower transport (versus pressure-driven air exchange) means significantly greater deposition.
1988	$Q_{daught,1988}$	$F_{2,1988}$	0–0.5	Addition of foam layer on domes should cause additional deposition, relative to 1980–1987.

Results of calculations. For the 1980–1987 period, the total Rn daughter release rate is the sum of release rates for air exchange releases and diffusion releases ($Q_{daught,post,exch}$ and $Q_{daught,post,diff}$, respectively). The frequency distributions for these intermediate, calculations are shown in Table J–30.

Table J–30. Summary of Frequency Distributions of Intermediate Calculations; for Current Estimates of Rn Daughter Releases

		Percentiles of distributions				
Parameter	Units	5th	25th	median	75th	95th
$Q_{daught,post,exch}$	Ci y^{-1}	140	350	530	740	1100
$Q_{daught,post,diff}$	Ci y^{-1}	2.7	14	28	45	75

Table J–31 shows the distributions for calculated Rn daughter release quantities for 1951, 1952, and 1953 from drummed K-65 material stored on the Plant 1 pad. The frequency distributions for calculated Rn daughter release rates from the K-65 Silos are summarized in Table J–32.

Table J–31. Summary of Predicted Rn Daughter Release Quantities (Ci)[a] from Drummed K-65 Material Stored on the Plant 1 Pad

	Percentiles of distributions				
Year	5th	25th	median	75th	95th
1951[b]	0.24	2.0	6.2	16	45
1952	3.9	33	100	250	730
1953[c]	0.63	5.4	17	42	120

[a] The release quantities are quantities of each of the short-lived daughters ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po.

[b] Releases for 1951 are assumed to have occurred in October through December.

[c] Releases for 1953 are assumed to have occurred in January through June.

Table J–32. Summary of Predicted Rn Daughter Release Rates (Ci y^{-1})[a] from the K-65 Silos

	Percentiles of distribution				
Period	5th	25th	median	75th	95th
mid-July 1952–mid-June 1953	150	760	1500	2300	3400
mid-June 1953–mid-September 1958	2400	3300	4000	4800	6300
mid-September 1958–June 1979	3300	4200	5000	5900	7200
July 1979–December 1987	170	380	560	770	1200
1988	7.3	41	99	190	390

[a] The release rates are quantities of each of the short-lived daughters ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po.

Total Radon and Radon Daughter Releases for the Operating Period 1951–1988

Total quantities of ^{222}Rn and Rn daughters released from the FMPC during the complete period 1951–1988 can be calculated by summing releases for the individual time periods. We first separately calculate total releases from the K-65 Silos and total releases from the drums of K-65 material stored on the Plant 1 pad. For the Silos:

$$R_{Silos,52-88} = \sum_i Q_i T_i \qquad \text{(J–65)}$$

where $R_{\mathrm{Silos},52\text{–}88}$ is the total quantity of Rn (Ci) released from the K-65 Silos for the complete period of releases from the Silos, 1952–1988; Q_i is the Rn release rate for period i; and T_i is the length (in time) of period i; with i representing each of the periods 1952–1953, 1953–1958, 1959–1979, 1980–1987, and 1988. For the drummed K-65 material:

$$R_{\mathrm{dr},51\text{–}53} = \sum_j R_{\mathrm{dr},j} \tag{J–66}$$

where $R_{\mathrm{dr},51\text{–}53}$ is the total quantity of Rn (Ci) released from the drummed K-65 material for the complete period of such releases, 1951–1953; and $R_{\mathrm{dr},j}$ is the quantity of Rn released for year j; with j representing each of the years 1951, 1952, and 1953.

The total quantity of Rn released from the FMPC for all years (1951–1988) is then:

$$R_{\mathrm{FMPC},51\text{–}88} = R_{\mathrm{Silos},52\text{–}88} + R_{\mathrm{dr},51\text{–}53} \tag{J–67}$$

The same equations are used to calculate total releases of Rn daughters, with the following substitutions: $R_{\mathrm{daught,Silos},52\text{–}88}$ for $R_{\mathrm{Silos},52\text{–}88}$; $Q_{\mathrm{daught},i}$ for Q_i; $R_{\mathrm{daught,dr},51\text{–}53}$ for $R_{\mathrm{dr},51\text{–}53}$; $R_{\mathrm{daught,dr},j}$ for $R_{\mathrm{dr},j}$; and $R_{\mathrm{daught,FMPC},51\text{–}88}$ for $R_{\mathrm{FMPC},51\text{–}88}$.

The values of T_i to be used are easily calculated from the individual periods, and are shown in Table J–33. Results of the calculations are shown in Table J–34.

Table J–33. Lengths of Periods Used in Calculations of Total Rn Releases from K-65 Silos

Period	Period length (years)
mid-July 1952–mid-June 1953	0.917
mid-June 1953–mid-September 1958	5.25
mid-September 1958–June 1979	20.79
July 1979–December 1987	8.50
1988	1.00

Table J–34. Summary of Frequency Distributions of Predicted Total Release Quantities (Ci) of Rn and Rn Daughters from the FMPC for 1951–1988

	Percentiles of distributions				
Parameter	5th	25th	median	75th	95th
$R_{\mathrm{Silos},52\text{–}88}$	110,000	140,000	170,000	190,000	230,000
$R_{\mathrm{dr},51\text{–}53}$	54	300	720	1500	3400
$R_{\mathrm{FMPC},51\text{–}88}$	110,000	140,000	170,000	190,000	230,000
$R_{\mathrm{daught,Silos},52\text{–}88}$ [a]	87,000	110,000	130,000	150,000	190,000
$R_{\mathrm{daught,dr},51\text{–}53}$ [a]	4.5	40.	130	320	880
$R_{\mathrm{daught,FMPC},51\text{–}88}$ [a]	87,000	110,000	130,000	160,000	190,000

[a] The release rates for ^{222}Rn daughters are release rates of each of the short-lived daughters ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po.

Summary of Current Estimates of Radon and Daughter Releases

The estimated Rn and Rn daughter release rates from the K-65 Silos are summarized in Table J–35. The calculated release rates are assumed to be constant over the full time periods assessed. Thus, within a given assessment period, the estimated release quantity for a given length of time is simply the time multiplied by the release rate. This also applies to the various percentiles of the distributions, since the parameter distributions are applied to the full time periods, rather than independently to each year.

Table J–35. Summary of Percentiles of Predicted ^{222}Rn and ^{222}Rn Daughter Release Rates (Ci y^{-1}) from the K-65 Silos

	Rn release rate			Daughter release rate[a]		
Period	5th	median	95th	5th	median	95th
mid-July 1952–mid-June 1953	200	1900	4200	150	1500	3400
mid-June 1953–mid-September 1958	3100	4900	7600	2400	4000	6300
mid-September 1958–June 1979	4200	6200	8700	3300	5000	7200
July 1979–December 1987	360	950	1700	170	560	1200
1988	120	540	1300	7.3	99	390

[a] The release rates for ^{222}Rn daughters are release rates of each of the short-lived daughters ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po.

We also note that for the 1980–1987 period the majority of the Rn released would have been released during daytime hours ($Q_{exch,post}$), when the warming of the silo air caused most of the ventilation of the silos. For the other periods, the majority of the releases would have been caused by other phenomena. For the 1959–1979 period, one driving force was probably wind across the silo penetrations. Thus, for all periods except 1980–1987, the releases are assumed to have occurred continually throughout the day.

The estimated total release rates of ^{222}Rn from the K-65 Silos are also summarized in Figure J–7 and Figure J–8. Figure J–7 is a plot of the distributions of the total release rates. This shows the relative magnitudes of the release rates, and the slopes of the curves indicate the breadth of the uncertainty intervals. Figure J–8 shows the estimated releases versus time. The 1959–1979 period appears, based on release rate and release time, to be the most significant, with a very high release rate for a long period of time.

The estimated Rn and Rn daughter release quantities from the drummed K-65 material stored on the Plant 1 pad are summarized in Table J–36.

The predicted total quantities of Rn released from the FMPC for the entire period of concern for this Project, 1951–1988, are summarized in Table J–37. It can be seen that Rn releases from the drummed K-65 material stored on the Plant 1 pad are relatively insignificant contributors to the total Rn releases for 1951–1988. However, these releases from the drummed K-65 material occurred in 1951–1953, when operations at the FMPC were just beginning. Thus, Rn releases from the drummed K-65 material may be significant contributors to releases of all radionuclides in the earliest years of site operations.

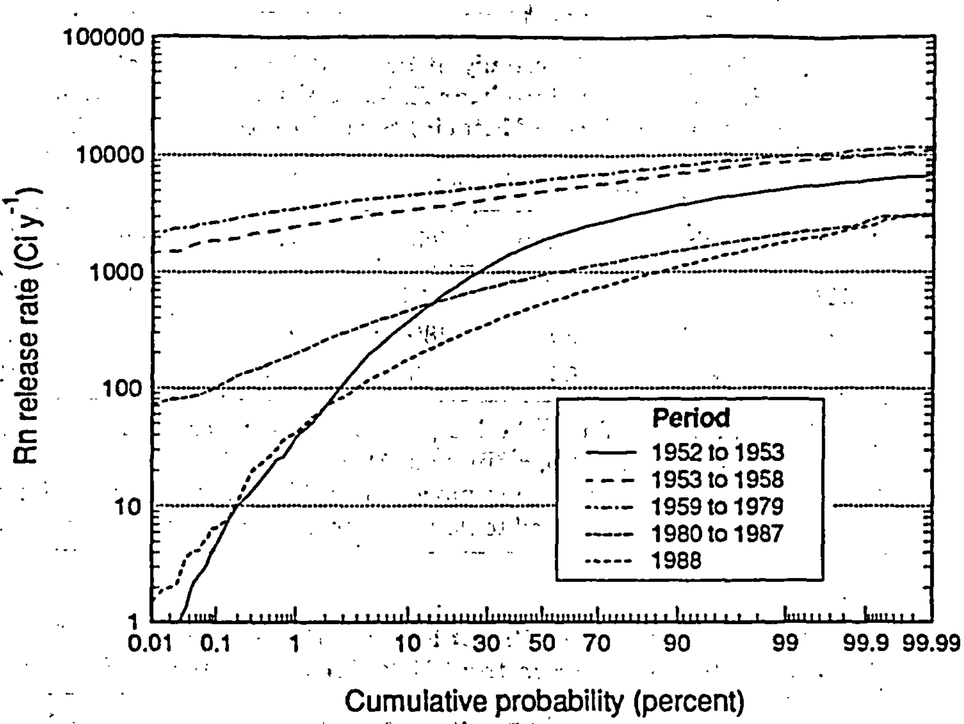

Figure J–7. Probability distributions of the estimates of total ^{222}Rn releases from the K-65 Silos for the separate periods analyzed.

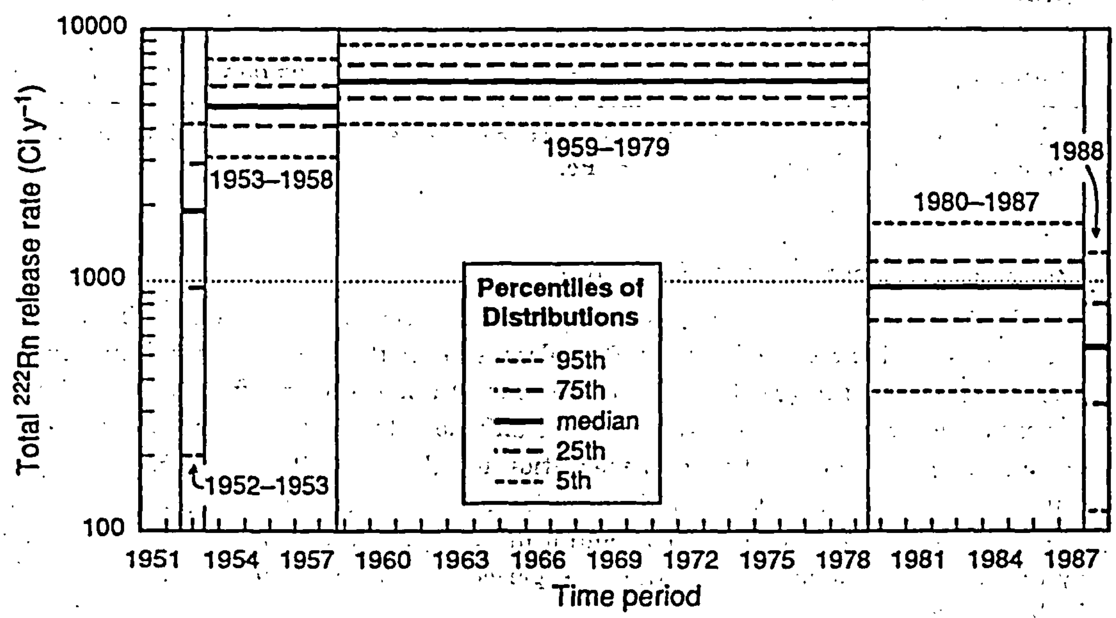

Figure J–8. Estimated ^{222}Rn release rates from the K-65 Silos as a function of time. The periods indicated are only nominal; more precise dates are given in Table J–35.

Table J–36. Summary of Percentiles of Predicted ^{222}Rn and ^{222}Rn Daughter Release Quantities (Ci) from the Drummed K-65 Material Stored on the Plant 1 Pad

	Quantity Rn released			Quantity daughters released[a]		
Year	5th	median	95th	5th	median	95th
1951[b]	2.6	35	170	0.24	6.2	45
1952	42	580	2800	3.9	100	730
1953[c]	6.9	95	450	0.63	17	120

[a] The release quantities for ^{222}Rn daughters are quantities of each of the short-lived daughters ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po.
[b] Releases for 1951 are assumed to have occurred in October through December.
[c] Releases for 1953 are assumed to have occurred in January through June.

Table J–37. Summary of Percentiles of Predicted Total ^{222}Rn and ^{222}Rn Daughter Release Quantities (Ci) from the FMPC for the Entire Period 1951–1988

	Rn release quantity			Daughter release quantity[a]		
Source of releases	5th	median	95th	5th	median	95th
K-65 Silos	110,000	170,000	230,000	87,000	130,000	190,000
Drummed K-65 material stored on Plant 1 Pad	54	720	3,400	4.5	130	880
Both sources	110,000	170,000	230,000	87,000	130,000	190,000

[a] The release quantities for ^{222}Rn daughters are release quantities of each of the short-lived daughters ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po.

Conclusions About Current Estimates

We first make two observations regarding calculated Rn releases. First, recall that equation J–12 indicates that total Rn releases from the K-65 Silos are proportional to $[(\lambda_v+\lambda_d)/\lambda_{eff}]$. This ratio is the fraction of the total removal of Rn from the silo that occurs through release through air exchange and diffusion (the rest is "removed" by radioactive decay). The difference between these ratios for the 1959–1979 and 1980–1987 periods illustrates the reason for the significant difference in total releases for the two periods. For the "post" period (see Table J–17), a significant, but small fraction of the available Rn is lost by release to the outside air. For the "pre" period (see Table J–20), almost all of the Rn lost is through releases.

Second, as expected based on the previous estimates, for the 1980–1987 period, releases from the K-65 Silos through the diffusion pathway are relatively small, but not insignificant, compared to releases through air exchange (see Table J–16 and Table J–17). For the 1980–1987 period the difference of the medians of the distributions is a factor of six.

As shown in Table J–35, Table J–36, Figure J–7, and Figure J–8, the uncertainties in some of the release rates are quite large. In particular, the 90% probability intervals of the ^{222}Rn releases from the drummed K-65 material have ranges of a factor of about 70. This large uncertainty is due to very little direct information pertinent to estimating the releases.

The 90% probability intervals of the Rn releases from the K-65 Silos for 1952–1953, 1980–1987, and 1988 have ranges of factors of about 20, 5, and 10, respectively. For the calculations of Rn releases from the K-65 Silos, the most important contributors to the uncertainties are the lack of direct information about releases during Silo operations, for the 1952–1953 and 1953–1958 periods; the lack of complete information about $\lambda_{v,post}$, for the 1980–1987 period; the limited, indirect information about $C_{a,pre}$, for the 1959–1979 period; and the limited Rn monitoring data, for the 1988 releases.

However, even with the large associated uncertainties, it is clear that the release rate of ^{222}Rn from the K-65 Silos was much greater in the 1959–1979 period than in the 1980–1987 period. This greater release rate for the 1959–1979 period is a very important result, which was not obtained in previous studies.

We acknowledge that the Rn daughter release estimates are extremely uncertain. This is due to the incomplete knowledge about the fractional release of Rn daughters through silo dome penetrations and drum penetrations, and due to the uncertainties in the estimated Rn releases. These releases may be important for estimates of doses to receptors close to the FMPC. At very close distances, exposures of people outdoors to Rn daughters may be due primarily to daughters released from the Silos and drums, since the short travel time would lead to relatively little ingrowth, along the travel path, of daughters from releases of Rn.

Earlier in this Appendix, we discussed previous estimates of the Rn source term from the K-65 Silos (see page J–12). The previous studies estimated Rn releases from information about the Silos and the K-65 material in the Silos. A recent study has instead estimated Rn releases based on a back-calculation from measured Rn concentrations around the FMPC and models of the atmospheric dispersion of the Rn in transport to the monitoring locations (Hamilton et al. 1993). The back-calculation was a linear, least-squares regression on the equation: measured concentration = (source term)×(source coefficient). The regression forced the y-intercept to zero. Here, the measured concentrations were net concentrations measured at the sixteen FMPC boundary fenceline monitoring stations. The source coefficients were the predicted ratios of Rn concentration at the receptor point to the release rate (χ/Q). The result was an estimated release rate of 1150 Ci y^{-1}, for 1989 and 1990. The uncertainty in this estimated release rate was not provided. However, the range of estimated source terms when individual locations were considered (rather than the regression of all locations) was 575 to 4025 Ci y^{-1}. The relative uncertainty in the best estimate (1150 Ci y^{-1}) is thus probably large.

The estimated release rate of Hamilton et al. can be compared to our estimated release rate for 1988, since conditions of the K-65 Silos were unchanged for the period 1988–1991. Our results were a median estimated release rate of 540 Ci y^{-1}, with a 90% probability interval of 120–1300 Ci y^{-1}. The estimated release rate of Hamilton et al. lies within the 90% probability interval of our estimate. This provides some corroboration of the reasonableness of our estimated release rate for 1988.

ALTERNATIVE CALCULATION OF UNCONSTRAINED RADON RELEASES, FOR COMPARISON WITH CURRENT RELEASE ESTIMATES

For assessments of releases of ^{222}Rn from bulk quantities of ^{226}Ra-bearing material, such as uranium mill tailings, one conventional calculational methodology uses models describing the generation of Rn in the material and diffusion through the material into the atmosphere (Rogers et al. 1984). Parameters for the calculations are based on characteristics of the ^{226}Ra-bearing material, including density, moisture content, porosity, Ra concentration, Rn emanation fraction, and Rn diffusion coefficient through the material.

The current estimates of radon releases from the K-65 Silos, discussed earlier in this Appendix, do not use this conventional methodology. For the preliminary source term work (Voillequé et al. 1991), the data on the characteristics of the K-65 material were quite limited, and the current approach was chosen to make better use of the other available data.

Since our preliminary work, some additional data on K-65 characteristics have been obtained. Thus we perform an alternative calculation (alternative to our current estimate) of Rn releases, using the more conventional methodology. To allow comparison with our current estimates, we calculate an unconstrained Rn release rate, that would exist if the Silo domes did not cover the K-65 material. An evaluation of the results of this alternative calculation indicates that the alternative methodology is not as satisfactory as the current methodology. However, the alternative calculation results do provide some corroboration of the reasonableness of the current estimates. The rest of this section describes the model used for the alternative calculation, parameter values chosen, and results of the calculation, and compares the results to those of the current estimates.

Model for Unconstrained Radon Releases

Since we intend to compare the results of this alternative calculation to our current estimates of Rn releases, the calculated end points must be the same. For this purpose, we calculate an unconstrained Rn release rate, which is an estimate of the release rate that would exist if the K-65 Silos were open to the atmosphere, rather than covered (with the silo domes). With Silos covering the K-65 material, Rn concentrations build up in the silo air space, and this Rn constrains the diffusive release of Rn from the K-65 material into the silo air space.

The conventional methodology uses the following equation to calculate the Rn diffusion fluence rate (often simply called Rn flux) from a bare layer of source material, with an impervious layer below it, into air above the source material (Collé et al. 1981).

$$j_{D,0} = \sqrt{\frac{D_e \varepsilon_w}{\lambda_{Rn}}} (\phi) \tanh\left(\frac{L_w}{l_w}\right) \qquad \text{(J–68)}$$

where

$j_{D,0}$ = unconstrained diffusion fluence rate of Rn (Rn flux). The quantity of Rn per unit time per unit area transported by diffusion from the source material (in this case the K-65 material) into the ambient air (pCi m^{-2} s^{-1}, or similar).

D_e = effective bulk diffusion coefficient of Rn through the porous source material (cm^2 s^{-1}, or similar).

ε_w = porosity of the source material.

ϕ = the pore space Rn production rate. Quantity of Rn produced in pore spaces of the source material per unit time per unit pore volume that is free to migrate through the pores of the material (pCi cm^{-3} s^{-1}). Depends on characteristics of the source material, including ^{226}Ra concentration, Rn emanation fraction, bulk density, and porosity, and on the Rn decay constant.

L_w = thickness of the source material (cm, or similar).

l_w = Rn diffusion length in the source material (related to D_e) (cm, or similar).

λ_{Rn} = the decay constant for ^{222}Rn.

We note that equation J–68 is equivalent to equation J–29 for the case when the constraining air concentration, C_a, is equal to zero, which is the case of unconstrained release.

In an earlier section of this Appendix, about current estimates of Rn releases, we estimated that the average volume in the K-65 Silos occupied by head space air (average for the two Silos) was between 40,000 and 62,000 ft^3 (see page J–29). The volume of the dome of each Silo was calculated to be 23,900 ft^3. Thus the head space volume includes an estimated 16,100 to 38,100 ft^3 that is in the cylindrical part of the Silo, which would be the top 3.2 to 7.6 ft of the cylindrical section. With the total height of the cylindrical section of the Silo being 26 ft 8 in, this implies that the average thickness of K-65 material in the Silos would be 19.1 to 23.5 ft. These are thus the potential values for L_w. Later in this section the results of calculations are discussed. The distribution of the calculated Rn diffusion length in the K-65 material, l_w, has fifth and 95th percentiles of 13.6 and 180 cm, respectively. With these values of L_w and l_w, the tanh term in equation J–68 will be very close to unity, being greater than 0.995. This indicates that the K-65 material is essentially infinitely thick in terms of producing Rn flux, in that an increase in thickness would not significantly increase the Rn flux. For simplicity then, we assume the tanh term is equal to one, and equation J–68 is reduced to:

$$j_{D,0} = \sqrt{\frac{D_e \varepsilon_w}{\lambda_{Rn}}}(\phi) \qquad \text{(J–69)}$$

The unconstrained Rn release rate is then just:

$$P_{Rn,0} = j_{D,0} A_w \qquad \text{(J–70)}$$

where A_w is the surface area of the source material (the K-65 material) exposed to the air. We use the P_{Rn} notation because as for the constrained Rn production rates, $P_{Rn,pre}$ and $P_{Rn,post}$, the releases are from the K-65 material to the air above it. The subscript 0 indicates unconstrained releases to ambient air.

The pore space Rn production rate can be calculated by:

$$\phi = \frac{[Ra] EF \rho_w \lambda_{Rn}}{\varepsilon_w} \tag{J–71}$$

where:

[Ra] = concentration of ^{226}Ra in K-65 waste material (activity per mass),

EF = ^{222}Rn emanation fraction in K-65 material, which is the fraction of the Rn formed (from the ^{226}Ra decay) that is in the pore spaces and is free to migrate, and

ρ_w = bulk density of K-65 waste material (g cm^{-3}, or similar).

In equation J–71, the ^{226}Ra concentration gives the total production rate of ^{222}Rn atoms, per mass of source material. Multiplication by the Rn emanation fraction converts this to the production of Rn in the pore spaces. The factors of ρ_w and ε_w convert the basis from mass of source material to volume of pore space air. Finally, the decay constant converts the quantity of Rn from atoms to activity units.

Radium-226 concentration. Concentrations of ^{226}Ra in the K-65 material in the two K-65 Silos were presented earlier in this Appendix (see page J–7). We concluded that the results from the 1991 ASI/IT sampling program are preferred for further use. The depths of these samples are identified only by the zone; either A, B, or C, where zone A is roughly the top third of the K-65 material in the Silo, zone B the middle third, and zone C the bottom third. Based on the overall thickness of the K-65 material, discussed above, the thickness of each zone is roughly 7 ft, or 2 m. If this thickness is used for L_w in equation J–68, with the fifth and 95th percentile values of l_w, as used above, the tanh term is fairly close to one, this time being greater than about 0.8. This indicates that the Rn releases are primarily due to ^{226}Ra in the top zone. We note that from this approach it also follows that the concentrations of ^{226}Ra in the upper parts within zone A (closest to the surface) would be more important to Rn releases than would those in the lower parts of zone A. Computer programs are available to perform Rn diffusion release calculations for multilayer systems (Rogers et al. 1984). However, the available data are not sufficient for such a multilayer calculation, and we continue to use the equation for a single layer of source material (equation J–69).

Thus we would like to use the average ^{226}Ra concentration of zone A of the K-65 material. In some cases there were multiple samples from a given zone and given manhole, though the difference in location within the zone is not known. We thus average the concentrations, from Table J–5, by zone. Table J–38 shows the average concentrations by zone and by manhole. For four of the locations in zone A, no sample results were available. Thus, the available results for zone A may be less than adequate to characterize the average concentration for all of zone A. Instead we use the range of average concentrations, from Table J–38, to represent the average for zone A. The assumed distribution for [Ra] is uniform, with minimum 134,900 pCi g^{-1} and maximum 697,000 pCi g^{-1}. This distribution seems quite broad, but it is justified by the limited characterization data. We also feel confident that it includes the average concentration in zone A material. We note that the implied mean concentration, 416,000 pCi g^{-1}, is very similar to the mean of all samples from both Silos, as given in Table J–5.

Table J–38. Average Concentrations of ^{226}Ra by Zone and Location in the K-65 Material in the K-65 Silos

		^{226}Ra Concentration (pCi g^{-1})	
Zone[a]	Location[a]	Silo 1	Silo 2
A	NE	394,900	134,900
A	SE	367,600	
A	SW		
A	NW	306,800	
B	NE	484,800	179,500
B	SE	697,000	433,300
B	SW		
B	NW	680,900	
C	NE	535,600	386,800
C	SE	601,600	252,600
C	SW		
C	NW	510,400	199,400

[a] Zone A refers to the top one-third of the K-65 material, zone B to the middle third, and zone C to the bottom third. The locations are the manholes, by direction from center, through which samples were obtained.

Radon emanation fraction. As discussed earlier in this Appendix (see page J–11), measurements of the Rn emanation fraction for the K-65 material have not been performed. In the absence of emanation fraction data for the K-65 material, values for similar materials must be used.

DOE (1990) briefly describes the process flow in the refinery (Plant 2/3) at the FMPC, and the processing of the waste products which contributed the K-65 and metal oxide materials. Feed materials (ores or concentrates) were first digested (or leached) with nitric acid. When pitchblende uranium ore was processed, "hot" raffinates (those that contained significant radioactivity, mostly due to ^{230}Th, ^{226}Ra, and daughter products) were produced from the solvent extraction step (using TBP kerosene) in the process. Further processing of the hot raffinates resulted in both K-65 and metal oxide materials. Thus, the K-65 material is only part of the "tailings" from the ore processing at the FMPC. It is assumed that similar processing formed the K-65 material that was brought to the FMPC from the Mallinckrodt Chemical Works in St. Louis.

Conventional methods for milling uranium from ores are described in NRC (1980). The general process in conventional uranium mills is similar to that used at the FMPC, with leaching of the ore (either acid leach or alkaline leach) followed by solvent extraction (using amine, kerosene, or alcohol, for acid leach process) or precipitation (for alkaline leach) to remove the uranium. The solids-containing tailings are removed after the leach process.

Raffinates from the solvent extraction process are generally recycled back to the leach process.

It thus appears that the general processing of ores in the FMPC refinery was similar to conventional acid-leach processing in uranium mills. One difference is that at the FMPC the solid waste materials were separated into two streams, the K-65 and metal oxide materials, while conventional mills produced a single tailings waste.

Radon emanation fraction measurements have been made for samples of tailings from various uranium mills. Since the processing to produce the K-65 material was relatively similar to that to produce uranium mill tailings, we think that emanation fraction measurements for uranium mill tailings are the best substitute for measurements specific to the K-65 material.

Rogers et al. (1984) compiles Rn emanation fraction measurements for tailings from nine different uranium mills in the western U.S. It is well known that the Rn emanation fraction is dependent on moisture content, particularly at very low moisture content (Rogers et al. 1984; Nazaroff and Nero 1988). Since the K-65 material was placed into the Silos as a slurry, and was somewhat protected from drying out, the moisture content in the K-65 material is expected to be relatively high. For moisture saturation fraction above 20%, emanation fractions ranged from about 0.1 to about 0.4, with a relatively uniform distribution of values across the range (Rogers et al. 1984). We thus assume that the emanation fraction for the K-65 material, *EF*, has a uniform distribution with minimum 0.1 and maximum 0.4.

Bulk density of K-65 material. Measurements of bulk density of the K-65 material were discussed earlier in this Appendix (page J–9). The three results, from studies in 1952 and 1972, ranged from 0.53 to 1.179 g cm^{-3} dry density (assumed dry in one case). Although these values seem quite low, no other values were located. Because we feel that knowledge about the bulk density of the K-65 material is incomplete, we assume the bulk density, ρ_w, has a uniform distribution, with minimum 0.53 g cm^{-3} and maximum 1.179 g cm^{-3}.

Specific gravity and porosity of K-65 material. As discussed earlier in this Appendix (page J–9), no measurements of porosity of the K-65 material have been reported. However, the report of the 1989 sampling episode (DOE 1990) presents specific gravity measurements for eight samples of K-65 material. Table J–39 shows these results.

Porosity, dry bulk density, and specific gravity of the K-65 material are related by:

$$\varepsilon_w = \frac{g_w - \rho_w}{g_w} \qquad \text{(J–72)}$$

where g_w is the specific gravity of the K-65 material, and where the density, ρ_w, is expressed here as the numerical value (without units) corresponding to the density given in units of g cm^{-3}. Porosity will be calculated using this equation. For the specific gravity, the six individual samples had a mean of 2.98 and standard deviation of 0.295. The standard error of the mean is thus 0.12. The range of values is from 2.58 to 3.37, which is fairly narrow. We assume the mean specific gravity of the K-65 material, g_w, follows a normal distribution with mean 2.98 and standard deviation 0.12.

Table J–39. Specific Gravity Measurements of K-65 Material from the K-65 Silos

Sample identification	Specific gravity
S1-NE-1A	3.19
S1-NE-1C	2.74
S1-SE-2T	3.37
S1-Compos.[a]	2.58
S2-NW-1A	2.87
S2-NE-2BT	2.59
S2-SW-1A	3.11
S2-Compo[a]	2.78
mean[b]	2.98
standard deviation[b]	0.295

[a] These appear to be composite samples, though there was no indication of what they were composed.

[b] We calculate the mean and standard deviation for the six individual samples only (not composites).

Moisture content of K-65 material. The moisture content of the K-65 material, in units of saturation fraction, is needed for the estimation of the Rn diffusion coefficient through the K-65 material, which will be discussed in the next section. Measurements of moisture content that have been located were summarized earlier, in Table J–4 and subsequent text (see pages J–8 and J–9). As discussed earlier, the top 2 m (or so) of the K-65 material in the Silos will have the most impact on Rn diffusion releases into the ambient air, so we would like to determine moisture content for this top layer of material. As discussed earlier, however, the vertical locations of the moisture content samples are not known, so the profile of moisture content with depth in the K-65 material is unknown. For some of the values given in Table J–4, we made reasonable assumptions about the units of the values as presented in the references. With that caveat, it appears that the range of measured moisture content in the K-65 material is 21.8% to 90% dry weight.

The moisture saturation fraction is the moisture content of a material expressed as the fraction of the maximum moisture content, which occurs when all the pore spaces in the material are filled with water (this condition is saturation). The moisture saturation fraction of the K-65 material, m_w, can be related to the moisture content in dry weight fraction (dry weight percent divided by 100%), M_w, by (Rogers et al. 1984):

$$m_w = \frac{M_w}{\frac{1}{\rho_w} - \frac{1}{g_w}} \qquad \text{(J–73)}$$

where ρ_w and g_w are the bulk density and specific gravity of the K-65 material, as used previously. Again, the density, ρ_w, is expressed as the numerical value (without units) corresponding to the density given in units of g cm^{-3}.

For the distribution of M_w, we assume a uniform distribution over the range of measured values, 21.8% to 90% dry weight, or 0.218 to 0.90 dry weight fraction, with one constraint. The saturation fraction must be less than or equal to 1. Thus, the constraint on M_w is:

$$M_w \le \frac{1}{\rho_w} - \frac{1}{g_w} \tag{J–74}$$

We thus consider M_w to have a uniform distribution with minimum 0.218 dry weight fraction, and with maximum 0.90 or $(1/\rho_w - 1/g_w)$, whichever is less.

Radon diffusion coefficient and diffusion length. As discussed earlier in this Appendix (see page J–11), measurements of the Rn diffusion coefficient in the K-65 material have apparently not been made. We must use values obtained from the literature.

Since there has been confusion in the literature regarding the nomenclature and symbols used for diffusion coefficients, the definitions of diffusion coefficient that we use in this Appendix are reviewed. Per Rogers et al. (1984), the diffusion coefficient for Rn *in the total pore space* of the material is designated by the symbol D. The effective bulk diffusion coefficient *of the material* is designated D_e. The two are related by:

$$D_e = D\varepsilon \tag{J–75}$$

where ε is the porosity of the material. This usage is also consistent with that of Collé et al. (1981).

Rogers et al. (1984) compile diffusion coefficients from about 200 measurements on various types of soils at various moisture saturations. For cases when little is known about the diffusion coefficient of a soil, they recommend the use of an empirical correlation with saturation fraction and porosity, given by:

$$\hat{D} = \left(0.07\ \text{cm}^2\ \text{s}^{-1}\right)\exp\left[-4\left(m - m\varepsilon^2 + m^5\right)\right] \tag{J–76}$$

where $\hat{D}$ is the empirically predicted pore space diffusion coefficient, m is the saturation fraction, and ε is the porosity. The characterization of the K-65 material indicates it is generally similar to soil. Thus, this equation is used to determine the nominal estimate of the pore space diffusion coefficient.

Rogers et al. (1984) indicate that the uncertainty in individual estimates of the diffusion coefficient for a particular soil at a given moisture may be as much as an order of magnitude, especially at higher moisture saturation fractions. We incorporate this uncertainty by:

$$D = U_D \hat{D} \tag{J–77}$$

where D is the adjusted estimate of the pore space diffusion coefficient, that will be used for further calculations, and U_D is an uncertainty factor, represented by a lognormally-distributed random variable with geometric mean 1 and geometric standard deviation 2 (this results in a 95% confidence interval somewhat greater than one order of magnitude).

We think that this degree of uncertainty is reasonable, given that very little is known about the K-65 material and its similarity (or lack of) to the soils measured by Rogers et al. The pore space diffusion coefficient cannot exceed the free space diffusion coefficient. The diffusion coefficient for Rn in pure air is 0.11 $cm^2\ s^{-1}$ (Nielson and Rogers 1982). Thus, D is constrained to be less than or equal to 0.11 $cm^2\ s^{-1}$. The effective diffusion coefficient, D_e, is then calculated using equation J–75.

The Rn diffusion length, l_w, is related to the diffusion coefficient by (Collé et al. 1981):

$$l_w = \sqrt{\frac{D_e}{\varepsilon_w \lambda_{Rn}}} \tag{J–78}$$

which is equivalent to:

$$l_w = \sqrt{\frac{D}{\lambda_{Rn}}} \tag{J–79}$$

The latter equation is used in our calculations.

Surface area of K-65 material in Silos. The physical size of the K-65 Silos was discussed earlier in this Appendix (see page J–3). The inside diameter is 80 ft. Since the K-65 material was placed as a slurry, we assume that the surface is flat. Thus, the surface area of a single silo, A_w, is just the area of a circle of radius 40 ft, which is 4.67×10^6 cm^2. For our calculations, this value is assumed to have negligible uncertainty.

Radon decay constant. The half life of ^{222}Rn is 3.8235 d (Walker et al. 1989). Thus, the decay constant for ^{222}Rn, λ_{Rn}, is 2.098×10^{-6} s^{-1}. For purposes of our calculations, this value is assumed to have negligible uncertainty. (This is the same as was used previously, for the current estimates, but with different units.)

Calculation of quotient needed for current estimate of Rn releases. To calculate the silo Rn production rates, $P_{Rn,pre}$ and $P_{Rn,0}$, discussed earlier (see page J–40), the quotient $[(\varepsilon_w l_w + h)/\varepsilon_w l_w]$ is required. This quotient is calculated here. Here h is the effective height of the contained (by the Silo) air space above the source (K-65) material. In an earlier section of this Appendix, about current estimates of Rn releases, we estimated that the average volume in the K-65 Silos occupied by head space air (average for the two Silos) was between 40,000 and 62,000 ft^3 (see page J–29). For the calculation of h, we assume the mean value of 51,000 ft^3. Since the Silos have an internal radius of 40 ft, h is 10.15 ft = 309 cm.

Implementation and Results of Calculations

The equations used for the calculations are as described above, except that in the case of equation J–70, a units conversion factor is added, as follows.

$$P_{Rn,0} = j_{D,0} A_w CF \tag{J–80}$$

where CF is the units conversion factor. The units desired for the result, $P_{Rn,0}$, is Ci y^{-1}. The units of the parameters are pCi $cm^{-2}\ s^{-1}$ for $j_{D,0}$; and cm^2 (per silo) for A_w. Thus, the units conversion factor is:

$$\begin{aligned} CF &= (2\ \text{silos})\left(3.156 \times 10^{7}\ \text{s y}^{-1}\right)\left(10^{-12}\ \text{Ci pCi}^{-1}\right) \\ &= 6.312 \times 10^{-5}\ \text{s Ci y}^{-1}\ \text{pCi}^{-1} \end{aligned} \qquad \text{(J–81)}$$

As for the current estimates of releases, the Monte Carlo calculations for this analysis were performed using spreadsheet and forecasting software on an IBM-compatible microcomputer. Ten thousand iterations of the calculations were performed. The parameter distributions were generated using Crystal Ball®, version 2.0 for Windows (Decisioneering 1992).

The parameter distributions used in the calculations are summarized in Table J–40.

Table J–40. Parameter Distributions for the Monte Carlo Calculation: Alternative Calculation of Unconstrained K-65 Silo ^{222}Rn Releases

Parameter	Units	Distribution	Descriptive statistics
[Ra]	pCi g^{-1}	uniform	minimum = 134,900; maximum = 697,000.
EF		uniform	minimum = 0.1; maximum = 0.4.
ρ_w	g cm^{-3}	uniform	minimum = 0.53; maximum = 1.179.
g_w		normal	mean = 2.98; standard deviation = 0.12.
M_w	fraction, dry weight	uniform	minimum = 0.218; maximum = 0.90 or $(1/\rho_w - 1/g_w)$, whichever is smaller.
U_D		lognormal	geometric mean = 1; geometric standard deviation = 2.
A_w	cm^2	known[a]	value = 4.67×10^6.
λ_{Rn}	s^{-1}	known[a]	value = 2.098×10^{-6}.
h	cm	known[a]	value = 309.

[a] "known" indicates that a single value is used in the calculations.

Table J–41 presents the results of the calculations, including intermediate results and the alternative estimate of unconstrained Rn releases from the K-65 Silos. Figure J–9 shows the distribution of estimates of the unconstrained Rn release rate, $P_{Rn,0}$.

As seen in Table J–41 and Figure J–9, the distribution of estimates of unconstrained Rn releases from the K-65 Silos is quite broad, indicating substantial uncertainty in these estimates. The 90% probability interval (from the 5th to the 95th percentile) has a range of a factor of about 20. Equation J–69 and equation J–70 are the primary equations for calculating the unconstrained Rn release rate, $P_{Rn,0}$. Of the parameters in these equations, we see in Table J–41 that the effective Rn diffusion coefficient, D_e, and the pore space Rn production rate, ϕ, have very significant uncertainties, with 90% probability intervals having ranges of factors of about 200 and about 9, respectively. These large uncertainties in turn result because of the significant uncertainties in the ^{226}Ra concentration in the K-65 material, [Ra], the Rn emanation fraction, EF, the bulk density of the K-65 material, ρ_w, the moisture content in the K-65 material, M_w, and the uncertainty factor applied to the predicted diffusion coefficient, U_D (see Table J–40).

Table J–41. Summary of Frequency Distributions of Calculated Results: for Alternative Calculation of Unconstrained K-65 Silo ^{222}Rn Releases

Parameter	Units	Percentiles of distributions				
		5th	25th	median	75th	95th
$\bar{\varepsilon}_w$		0.61	0.66	0.71	0.76	0.81
m_w	saturation fraction	0.26	0.43	0.59	0.76	0.95
D	$cm^2\ s^{-1}$	3.9×10^{-4}	4.1×10^{-3}	1.4×10^{-2}	3.1×10^{-2}	6.9×10^{-2}
D_e	$cm^2\ s^{-1}$	2.7×10^{-4}	2.8×10^{-3}	1.0×10^{-2}	2.3×10^{-2}	5.2×10^{-2}
l_w	cm	14	44	82	120	180
$[(\varepsilon_w l_w + h)/\varepsilon_w l_w]$		3.23	4.46	6.35	11.2	35.2
ϕ	$pCi\ cm^{-3}\ s^{-1}$	0.070	0.14	0.23	0.36	0.63
$P_{Rn,0}$	$Ci\ y^{-1}$	580	1800	3500	6100	12,000

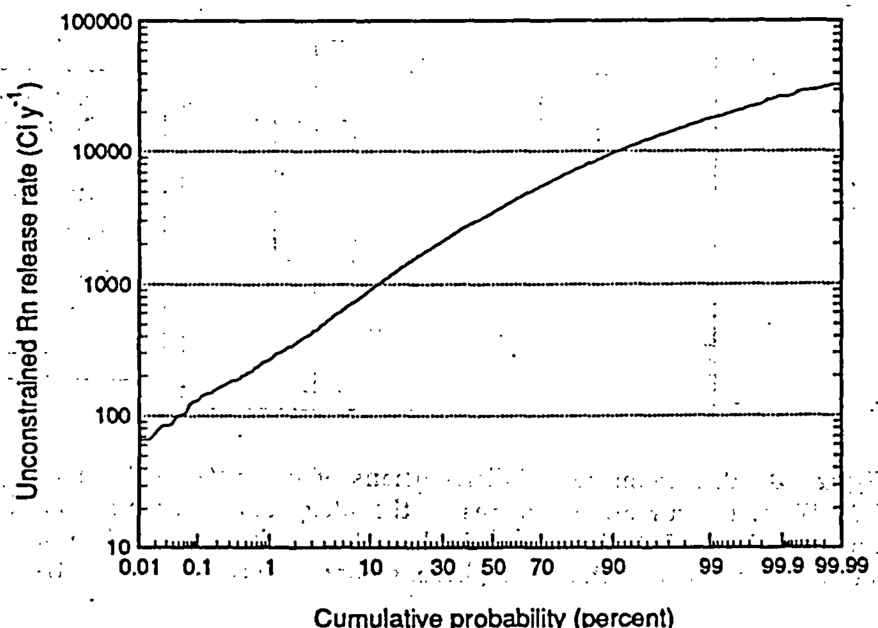

Figure J–9. Probability distribution of alternative estimates of unconstrained Rn releases from the K-65 Silos.

Discussion

Results of the alternative calculation of unconstrained Rn releases from the K-65 Silo are compared to results using the current methodology, in Table J–42 and Figure J–10, which summarize the distributions of results from the two methodologies.

Table J–42. Comparison of Results of Estimates of Unconstrained[a] ^{222}Rn Release Rates (Ci y^{-1}) from K-65 Silos

Method	Percentiles of distributions				
	5th	25th	median	75th	95th
Current estimates	4,700	5,900	6,800	7,900	9,400
Alternative calculation	580	1,800	3,500	6,100	12,000

[a] The "unconstrained" Rn release rate is the release rate estimated to occur if the K-65 material were open to the atmosphere, rather than covered by the K-65 Silos.

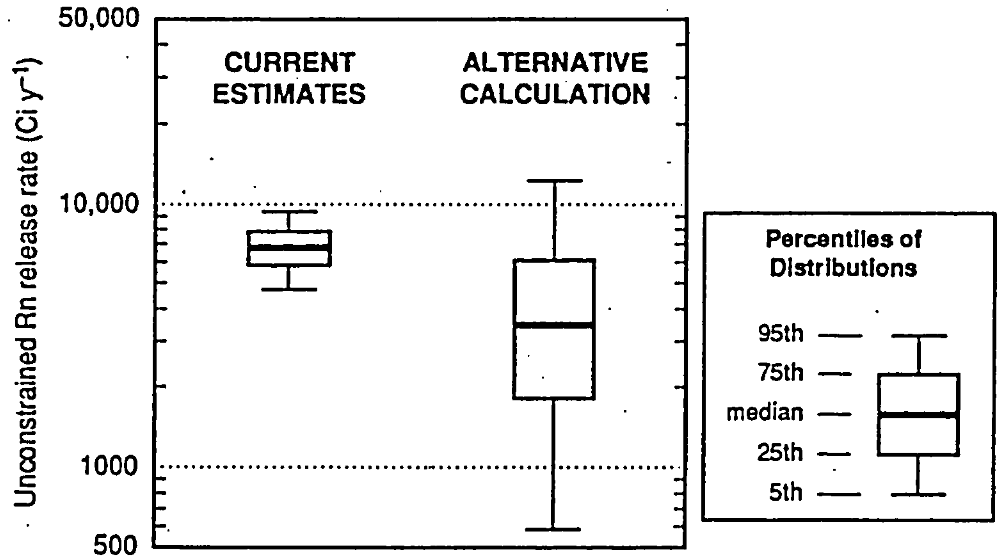

Figure J–10. Summaries of distributions of unconstrained Rn release rate from K-65 Silos, for current estimates methodology and alternative methodology. The "unconstrained" Rn release rate is the release rate estimated to occur if the K-65 material were open to the atmosphere, rather than covered by the K-65 Silos.

Table J–42 and Figure J–10 show that the results of the alternative calculation of unconstrained Rn releases are significantly different from the results using the current methodology. The results of the alternative calculation have a much lower median estimate, and a much broader uncertainty distribution. For the current methodology, the 90% probability interval (5th to 95th percentile) spans a factor of about two, while the same interval for the alternative calculation spans a factor of about 20. The 90% probability interval of the alternative calculation includes the 90% interval of the current estimates.

Knowledge of the Rn concentration in the head space air can be quite important to establishing, either explicitly or implicitly, the lower bound of the distribution of release rate results. The alternative calculation does not make use of the Rn concentration in the Silo head space air (measured in 1987), which has been used in the current estimates for C_a

(see page J–28). As shown in the following paragraphs, this appears to be an important omission of the alternative methodology.

The Rn concentration in head space air can be used to estimate a lower bound for the constrained release of Rn from the K-65 material into the head space of the Silos, as follows. If the Rn concentration in the head space is assumed to be an equilibrium concentration, then we can calculate the equilibrium Rn rate of release of Rn from the K-65 material into the head space required to sustain the Rn concentration. This equilibrium release rate (production term) was previously shown to be:

$$P_{\mathrm{Rn}} = C_{\mathrm{a}} \lambda_{\mathrm{eff}} V_0 \tag{J-9}$$

where
P_{Rn} = the constrained (by the presence of the silo) rate of release of ^{222}Rn into the silo air (production term) from the K-65 source material (activity per time),
C_{a} = concentration of ^{222}Rn in the silo air,
V_0 = volume of the air space in the silo above the K-65 material, and
λ_{eff} = the effective removal rate of ^{222}Rn from the silo air space (fraction per time).

The effective Rn removal rate is:

$$\lambda_{\mathrm{eff}} = \lambda_{\mathrm{Rn}} + \lambda_{\mathrm{v}} + \lambda_{\mathrm{d}} \tag{J-8}$$

where
λ_{v} = ventilation rate of the silo, or fraction of the silo air exchanged with the outside per unit time, and
λ_{d} = rate constant for diffusion losses, the fractional rate of Rn loss from the silo air space through diffusion through the silo dome (fraction per time).

To estimate a lower bound for P_{Rn}, which we call $P_{\mathrm{Rn,min}}$, we use the minimum possible value of λ_{eff}, which is just λ_{Rn}, assuming no releases through ventilation or diffusion. We thus have:

$$P_{\mathrm{Rn,min}} = C_{\mathrm{a,post}} \lambda_{\mathrm{Rn}} V_0 \tag{J-82}$$

where $C_{\mathrm{a,post}}$ is the head space Rn concentration for the 1980–1987 period. Using the mean values of the distributions previously estimated for $C_{\mathrm{a,post}}$ and V_0 (see page J–109), we estimate:

$$\begin{aligned} P_{\mathrm{Rn,min}} &= \left(2.62 \times 10^{7}\ \mathrm{pCi\ L^{-1}}\right)\left(2.098 \times 10^{-6}\ \mathrm{s^{-1}}\right)\left(51{,}000\ \mathrm{ft^3}\ (\text{per silo})\right) \\ &\times \left(10^{-12}\ \mathrm{Ci\ pCi^{-1}}\right)\left(28.317\ \mathrm{L\ ft^{-3}}\right)\left(3.156 \times 10^{7}\ \mathrm{s\ y^{-1}}\right)(2\ \text{silos}) \\ &= 5000\ \mathrm{Ci\ y^{-1}} \end{aligned} \tag{J-83}$$

It is noted that this is only a nominal estimate, and there would be some associated uncertainty, although the uncertainty should be relatively small, since the uncertainties in $C_{\mathrm{a,post}}$ and V_0 are relatively small.

If the distribution of estimates of the unconstrained Rn release rate, based on the alternative calculation (see Table J–41 and Figure J–9), is compared to this nominal estimate of $P_{Rn,min}$, it appears that the alternative calculation significantly underpredicts the unconstrained Rn release rate. It is not feasible to determine which of the parameters are responsible for this underprediction. It is also not apparent how the information on Rn concentration in the silo air space could be incorporated into the calculations. It is not appropriate to simply constrain the results to be greater than $P_{Rn,min}$, because the results then would not follow from the parameter distributions.

Conclusions About the Alternative Calculations

Because of the very large uncertainties and the apparent underprediction of the alternative calculation of the unconstrained Rn releases, we conclude that the alternative calculation methodology is not as satisfactory as the current methodology. Thus, we continue to use the current methodology for calculations of Rn releases, and perform no further calculations using the alternative methodology. The 90% probability interval of the alternative calculation distribution includes the 90% interval of the current estimates distribution, and this does provide some corroboration of the reasonableness of the current methodology.

PARAMETER VALUES TO BE USED FOR BUILDING WAKE EFFECTS MODEL

As discussed in the report of Task 4 of this Project, our model for atmospheric transport of Rn (and particulate releases) from the FMPC site includes a building wake effects model (Killough et al. 1993). For implementing this wake effects model, the cross-sectional area of the FMPC building from which material is emitted is required. For the K-65 Silos, the "building" is considered to be the combination of the K-65 Silos and the surrounding berms.

As described earlier in this Appendix, the berms around the K-65 Silos were constructed in May 1964, and enlarged in June 1983. The monthly project record report for this work (NLCO 1984) indicates that construction was 90% complete on May 1, 1964. It also indicates that the expansion work occurred between May 16 and June 27, 1983 (both construction and expansion were under the same project). For simplicity, we thus assume that on May 1, 1964, conditions changed from no berm to the initial berm, and that on June 1, 1983, conditions changed from the initial berm to the enlarged berm.

Engineering drawings have been obtained, which show dimensions of the berms. A drawing from December 1963 shows the plans for construction of the berms (NLCO 1963). The plans indicate that the earthen embankment (we call it berms) was to be built up to the level of the top of the Silo walls. There was to be a ring, 8 ft wide, with very slight slope (2 inches in 8 ft) next to the Silos, and then a slope of 1½:1 (horizontal to vertical) away from the Silos to the toe of the berms. From two aerial photographs of the K-65 Silos, taken in 1965, it appears that the berms were up to the tops of the Silo walls, and the slope of the berms looks close to 1½:1 (DOE 1965a; DOE 1965b).

A drawing from 1983 shows the plans for expansion of the berms (Geesner 1983). This drawing indicated that the berms were to be expanded in areal extent, to change the slope from 1½:1 to 3:1 (both slopes horizontal to vertical). An aerial photograph of the Silos, from 1987 (DOE 1987), and a topographic map of the site (Woolpert circa 1988) generally corroborate the areal expansion of the berms. Figure J–11 is a drawing of the Silo berms, based on the information in the engineering drawings cited above.

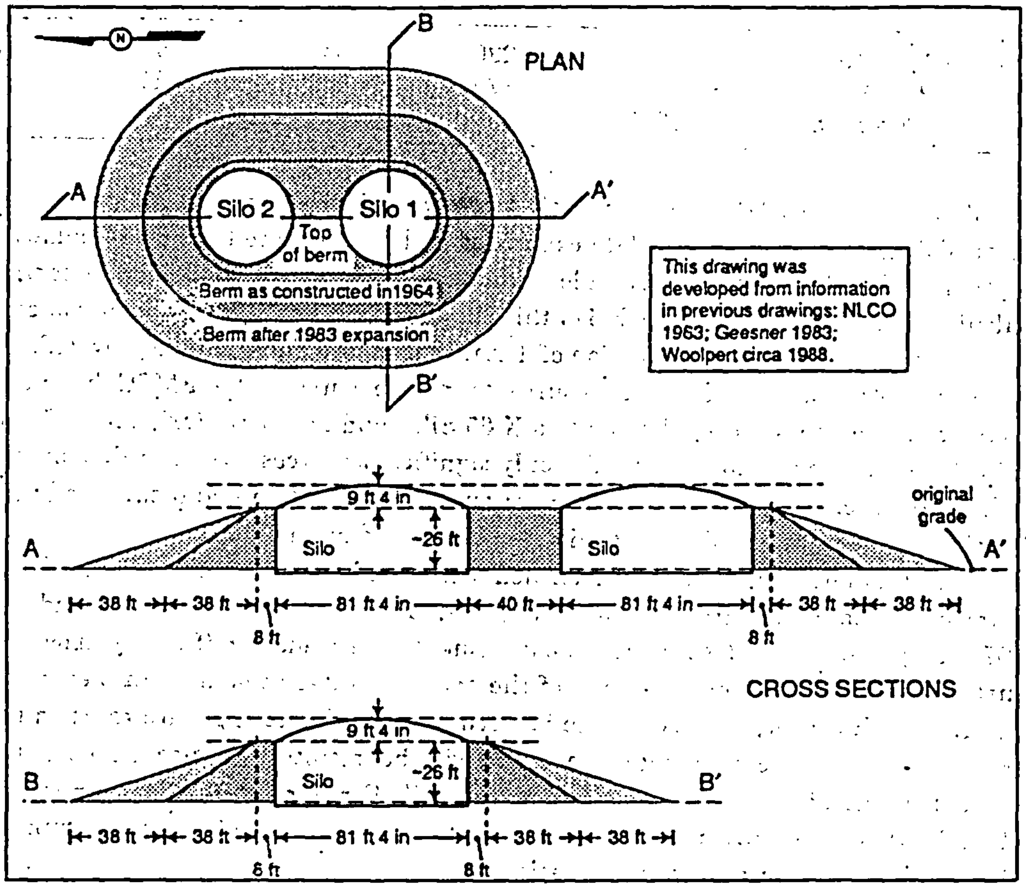

Figure J–11. Plan and cross sections of the K-65 Silo berms.

From the cross sections in Figure J–11, the cross-sectional area of the combined Silos and berms can easily be calculated. The results are shown in Table J–43. For the building wake effects model, the cross-sectional area is represented by an effective height and effective width of the "building." The height of the top of the Silos, above the original ground surface, is about 11 m, so this is used as the effective height. The effective width is then the width that would produce the correct average cross-sectional area. The effective dimensions, to be used in the modeling, are also shown in Table J–43.

PARAMETER VALUES TO BE USED FOR DIRECT EXPOSURE CALCULATIONS

The elevated concentrations of ^{226}Ra and other radionuclides in the K-65 and metal oxide materials produce significant emissions of gamma radiation, which may have exposed

Table J–43. Calculated Cross-Sectional Area and Effective Dimensions of the Combined K-65 Silos and Berms

	Cross-sectional area (m^2)			Modeled dimensions (m)	
Period	North/south	East/west	Average	Height	Width
1952–April 1964	459	230.	344	11	31
May 1964–May 1983	686	360.	523	11	48
June 1983–1988	778	452	615	11	56

people outside the FMPC. In our Task 4 Report (Killough et al. 1993), we described the methodology to be used to calculate exposures and doses due to this direct radiation. In this section of this Appendix, we provide additional information, necessary to complete those calculations. In the report of Task 6 of this Project (work in progress), results of calculations, including exposure rate as a function of distance from the Silos, will be provided.

For assessments of direct exposures to people outside the FMPC boundary due to sources on the site, we consider the two K-65 Silos and the Metal Oxide Silo, Silos 1, 2, and 3 in the waste storage area, to be the only significant sources. We base this on two types of information: results of aerial radiation surveys of the FMPC site and surrounding area, and results of penetrating radiation monitoring performed by the FMPC.

Aerial radiation surveys are performed using thallium-activated sodium iodide radiation detectors, from small airplanes or helicopters, flying at relatively low altitudes (Feimster 1979; Shipman 1985). Because the measurements are made significantly above the ground surface, they are not true measures of the exposure rates at or near (a meter or so above) the ground. However, the results can be used to approximate exposure rates on the ground. When radiation spectrum data are collected, the results can also be used to estimate concentrations of some radionuclides in the soil. For our purposes, the results can help identify where exposure rates outside the site boundary are above background, and help identify the onsite sources of the radiation.

It appears that three aerial radiation surveys have been completed over the FMPC area. An FMPC memorandum (Starkey 1962) indicates that a survey was to be performed at the very end of October, or early in November, 1962. Attached to this memorandum, we found a handwritten drawing of the FMPC area, with count rates (presumably from some type of radiation detector) along what appear to be flight lines east and west across the FMPC area (Anonymous circa 1962). A date on the back of the drawing indicates that it was received by the Atomic Energy Commission in November 1962. The second survey was performed in August 1976 and May to June 1977 (Feimster 1979). The third was performed in April 1985 (Shipman 1985).

Results of the three surveys were qualitatively similar (Anonymous circa 1962; Feimster 1979; Shipman 1979). Over the production area of the FMPC, relatively high exposure rates, to a few hundred $\mu R\ h^{-1}$, were measured. From the production area to the boundaries of the site, exposure rates generally decrease to background levels. Very high exposure rates were also measured above the K-65 and Metal Oxide Silos. Exposure rates around the Silos remained somewhat above background at the western boundary of the site. At the FMPC boundary, exposure rates were not significantly above background, except for near the Silos. These elevated offsite exposure rates are ascribed to radiation from the waste storage Silos. A Department of Energy memorandum provides a figure of results of the 1985 survey with the site boundary added, and this also clearly shows offsite exposure rates around the Silos that are greater than background (Stern 1985). From the aerial surveys, it appears the only FMPC sources of offsite, direct radiation are the K-65 and Metal Oxide storage Silos.

In the report of Task 5 of this Project (Shleien et al. 1993), we compiled exposure rate monitoring data from the FMPC annual environmental monitoring reports. Routine, quarterly exposure rate monitoring had been performed at locations on the FMPC boundary since 1976. Figure J–12 shows the monitoring locations. Based on a review of the exposure rate data from 1976 through 1990, we concluded that exposure rates at locations AMS-1 through AMS-5, and AMS-7, were similar to background exposure rates. Only location AMS-6 showed exposure rates that were clearly elevated above background concentrations. The K-65 and Metal Oxide Silos are the closest substantial radiation source to this monitoring location. It again appears that the K-65 and Metal Oxide Silos are the only FMPC sources of offsite, direct radiation.

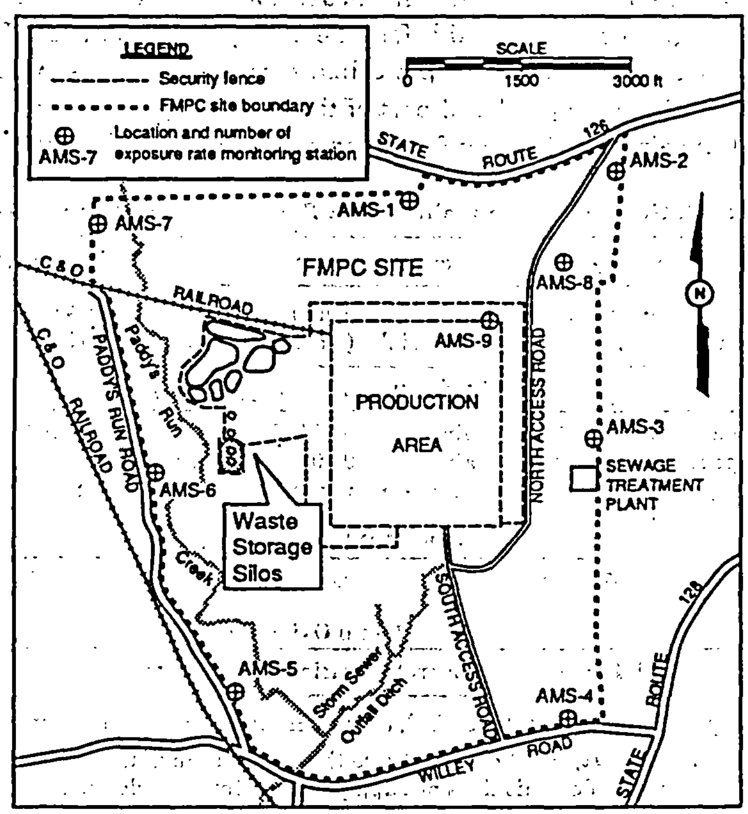

Figure J–12. Onsite locations of FMPC routine exposure rate monitoring, reported in FMPC annual environmental monitoring reports. Through 1985, locations AMS-1 through AMS-7 were called BS-1 through BS-7, respectively.

Calculations of exposure rates to which people were exposed will be performed with the MicroShield 4 computer software (Negin and Worku 1992), using the methodology described in our report of Task 4 of this Project (Killough et al. 1993). In the rest of this section we describe properties of the Silos and the Silo contents, that are required for the MicroShield calculations, including those related to the source and shield geometries, some materials properties for sources and shields, the integration quadrature order (which describes the fineness of the volumetric mesh used for the numerical integrations), material density, moisture content, and radionuclide concentrations. As MicroShield does not support Monte Carlo uncertainty analyses, single values of parameters will be used.

Parameters Applicable to All Calculations

The Task 4 report (Killough et al. 1993) developed some of the generally applicable parameters required for the MicroShield calculations, including those related to the source and shield geometries, some materials properties for sources and shields, and the integration quadrature order. These generally applicable parameters are summarized in Table J–44. The K-65 Silos are generally modeled as a single Silo, with height equal to twice the physical height. For the period before the addition of berms, the K-65 Silos are modeled as three stacked cylinders: the bottom cylinder representing the K-65 waste material, the middle representing the cylindrical part of the Silo air space, and the top representing the dome part of the air space (more information can be found in the Task 4 Report).

Table J–44. Generally Applicable Input Parameters for MicroShield Calculations

Cylinder geometry			Source properties		Shield properties			
designation	height (ft)	radius (ft)	material	density ($g\ cm^{-3}$)	material	thickness (in)	density ($g\ cm^{-3}$)	quadrature order[a]
K-65 Silos before Berms Added								
dome head space	18.67[b]	28.5	air	0.001293	concrete	9.805	2.35	10, 10, 10
cylinder air space	10.8[b]	40	air	0.001293	concrete	8	2.35	10, 10, 10
K-65 waste	42.6[b]	40	concrete	variable	concrete	8	2.35	12, 12, 12
K-65 Silos with Berms								
dome head space	18.67[b]	28.5	air	0.001293	concrete	9.805	2.35	10, 10, 10
Metal Oxide Silo								
waste	31.4	40	concrete	variable	concrete	8	2.35	10, 10, 10

[a] Integration quadrature orders for radial, circumferential, and axial directions.
[b] As mentioned in the text, this height is twice the physical height, to allow the treatment of the two K-65 Silos as a single Silo. This does not apply to the Metal Oxide Silo.

Since the Task 4 work, we have determined that the previously reported quadrature orders for calculations of exposures rates due to the K-65 waste material (Killough et al. 1993; Shleien et al. 1993), are inadequate. The integration quadrature order describes the number of increments into which the source geometry is divided for the numerical integrations. For cylindrical sources, we must specify quadrature order for radial, circumferential, and axial directions. As recommended by the MicroShield manual (Negin and Worku 1992), we have made test calculations using a range of quadrature orders. The tests indicate that for the head space sources and the Metal Oxide Silo source, with quadrature orders of 10 for the three parameters, the exposure rate results obtained were within 1% of the results for higher quadrature order (finer "mesh" size). Thus, for these calculations we will use a quadrature order of 10 for the radial, circumferential, and axial directions. However, for the K-65 material source (in the Silos), similar tests indicate that quadrature orders of 12 for each direction are required to obtain results within 1% of results for higher quadrature order. Thus, for calculations involving the K-65 material source, we will use a quadrature order of 12 for the radial, circumferential, and axial directions. These revised values are shown in Table J–44.

Silo Fill Fraction

The model for calculations of direct radiation exposures due to the K-65 and Metal Oxide Silos was described in the report of Task 4 of this Project (Killough et al. 1993). Some of the important parameters are the heights of the cylinders used to represent the waste material and Rn and Rn daughters (in head space air) of the Silos, and these are generally invariant. However, during the period when the Silos were still being filled with material, these heights were changing. We account for this by incorporating a silo fill fraction, as follows.

The disposal history of the K-65 Silos was discussed earlier in this Appendix (page J–4). The information located indicates that filling of Silo 1 began in July 1952, filling of Silo 1 was completed and filling of Silo 2 began in June 1953, and filling of Silo 2 was completed in September 1958. We make the assumption that each Silo was filled at a uniform rate between these dates. Since the exact dates are not available, we also assume that the beginning and ending dates are represented by the middle of the month in which they occur. With these assumptions, the silo fill fractions, which are the fraction of the eventual total quantity of waste material that has been placed, are calculated. The estimated, annual average silo fill fractions for each of the two Silos are shown in Table J–45.

As described in the Task 4 report (Killough et al. 1993), the two Silos are generally modeled as a single Silo, but with twice the height. (The justification for modeling the two Silos as a single Silo is provided in that report.) As described in that report, for conditions after filling of both Silos was completed (1959 and later), the heights to be used in the model are 42.6 ft for the cylinder representing the K-65 material, 10.8 ft for the cylinder representing the Rn and daughters in the cylindrical part of the head space air, and 18.7 ft for the cylinder representing the Rn and daughters in the dome part of the head space air. For earlier years (1952–1958), silo fill fractions are applied separately to each silo.

For 1954–1958, when both Silos were in use (by "use" we only mean the Silo was storing material, whether or not material was still being added), the heights of K-65 material in each Silo are estimated by multiplying each fill fraction by 21.3 ft (the height when "full"). The two heights are then summed to get the height to be used for the model. The height to be used in the model for the cylinder part of the air space is then 53.4 ft minus the combined K-65 material height. The height to be used for the dome part of the sir space is 18.7 ft.

For 1952, only Silo 1 was in use. The height of K-65 material is calculated as the fill fraction multiplied by 21.3 ft. That result is used for the model. The height to be used in the model for the cylinder part of the air space is then 26.7 ft minus the K-65 material height. The height to be used for the dome part of the sir space is 9.3 ft. These model values are only applied to that part of the year for which exposures could have occurred (July through December).

For 1953, part of the year one silo was in use and part of the year two silos were in use. For each Silo, the model heights are first calculated as done for 1952. Since Silo 2 was only in use for about 7 months of the year, the model heights for Silo 2 are then multiplied by 7/12 to give effective model heights. The (effective) model heights for the two Silos are then summed to obtain the total model heights to be used.

These calculated heights, to be used in the model, are also shown in Table J–45.

Table J–45. Estimated Annual Average Silo Fill Fractions and Cylinder Heights for Modeling K-65 Silos

	Silo fill fraction		Model heights (ft)[a]		
Year	Silo 1	Silo 2	K-65 material	Cylindrical air	Dome air
1952[b]	0.23	0.00	4.9	21.8	9.3
1953	0.89	0.048[c]	19.5[c]	22.8[c]	14.8[c]
1954	1.00	0.20	25.5	27.9	18.7
1955	1.00	0.39	29.6	23.8	18.7
1956	1.00	0.58	33.6	19.8	18.7
1957	1.00	0.77	37.7	15.7	18.7
1958	1.00	0.95	41.6	11.8	18.7
1959 and later	1.00	1.00	42.6	10.8	18.7

[a] The "K-65 material" cylinder represents the K-65 material in the Silos. The "cylindrical air" represents that part of the head space that is in the cylindrical part of the Silos. The "dome air" represents that part of the head space in the dome part of the Silos.

[b] Values for 1952 are not annual averages. They apply only to the period July through December, which is the period of emissions, after filling of the Silo began.

[c] The average fill fraction for Silo 2 for 1953, 0.048, is not an annual average. It applies only to June to December, when Silo 2 was in use. The model heights are calculated to be annual averages (heights for Silo 2 were multiplied by 7/12), to be applied to all of 1953.

As shown in the preliminary calculations in our reports of Tasks 4 and 5 of this Project, the Metal Oxide Silo only contributes a small fraction, relative to that from the K-65 Silos, of the exposure rate at locations outside the FMPC boundary (Killough et al. 1993; Shleien et al. 1993). We have not obtained much production information related to the Metal Oxide Silo. In particular, it is not clear when filling of the Metal Oxide Silo was terminated. A document that appears to be a type of progress report or completion report, from the original site construction contractor, indicates that construction of the Metal Oxide Silo and Silo 4 was completed in July 1953 (Catalytic circa 1950s(b)). Details of the history of filling the

Metal Oxide Silo have not been obtained. Metal oxide materials were generated from the extraction processing (in Plant 2/3) of both uranium ores and other uranium-containing materials (DOE 1990), so metal oxide material could have been placed in Silo 3 earlier than 1955, when ore processing started. Thus, for calculations, we make the assumption that Silo 3 was full in July 1953. This will add a slight positive bias to estimated direct radiation exposure rates. But, because filling of K-65 Silo 1, which contributes more to direct exposures than the Metal Oxide Silo, was complete at this time, the overall impact is thought to be fairly small. If it turns out that this is not the case, additional investigation will be done to better model the filling of the Metal Oxide Silo.

Material Densities and Moisture Contents

Earlier in this Appendix we presented data related to characteristics of the K-65 and metal oxide materials. From those data we choose values to be used for the direct exposure calculations.

For the K-65 Silos, bulk densities were reported in only two studies, from 1952 and 1972 (see Table J–4). Although the results from the two studies appear significantly different, it would be difficult to disregard either data set. The range of results from the two studies was 0.53 g cm^{-3} to 1.179 g cm^{-3}. Since the number of samples involved is quite small, we choose to use the midpoint of this range, 0.85 g cm^{-3}, for the direct exposure calculations. For the Metal Oxide Silo, bulk density was reported in only one study (see page J–12). Two densities were reported, a "free flowing" density and a maximum density. We choose to use the free flowing density, 0.64 g cm^{-3}, for the direct exposure calculations, because we think it is more representative of the *in situ* density.

We note that these densities seem quite low, relative to typical densities of U mill tailings, of around 1.6 g cm^{-3} (Rogers et al. 1984). However, the exact value may not be very important for direct exposure calculations, because of the competing factors of activity and self-shielding. That is, if the density increases, then the total amount of activity emitting radiation increases, but the mass of material shielding that radiation also increases. To evaluate the sensitivity to density, preliminary MicroShield calculations were performed. For the baseline case, we used the radioactivity mass concentrations for Silo 1, the density 0.85 g cm^{-3}, and a dose point at a distance of 1100 ft (the site boundary). For the comparison case, we used a density of 1.6 g cm^{-3}, the same mass activity concentrations (thus the volume concentrations required for MicroShield were adjusted), and the same distance. The results were exposure rates of 3.55 μR h^{-1} for the baseline case, and 3.83 μR h^{-1} for the density of 1.6 g cm^{-3}, a fairly small difference for the large change in density.

As we showed in Table J–4, moisture content in the K-65 material was reported in a few studies. However, the values vary considerably between studies. The range of results for all studies is 21.8% to 90% dry weight. We choose to use the midpoint of this range, 56% dry weight. For the metal oxide material, quantitative results were reported in only one study, although another indicated, qualitatively, that the sample was dry (see page J–11). Since the highest moisture content reported was about 10%, we choose to ignore the moisture content in Silo 3 for the direct exposure calculations.

Radionuclide Concentrations in Silo Contents

Earlier in this appendix we discussed concentrations of radionuclides in the K-65 and metal oxide materials (see page J–7 and page J–11). From the radionuclides that have been detected in the K-65 and metal oxide materials, it is clear that all radionuclides in the naturally-occurring thorium, uranium, and actinium decay series should be assumed present. Of course, this is also expected based on the origins of the materials as waste products from uranium ore processing. Table J–5 and Table J–6 show that in the K-65 and metal oxide materials, the ^{226}Ra concentrations are elevated relative to concentrations of other nuclides with gamma radiation emissions (or gamma-emitting daughter nuclides). Thus, the most important nuclides, in terms of potential direct exposures, are ^{214}Pb and ^{214}Bi, two short-lived daughters of ^{222}Rn.

Of the many nuclides in the three decay series, many contribute negligibly to gamma radiation emissions. We have determined, through MicroShield calculations, that ^{210}Pb, ^{210}Bi, and ^{210}Po, which are later daughters of ^{222}Rn, and the short-lived daughters of ^{227}Ac, contribute significantly less than 1% of the calculated exposure rates. Thallium-206 is a pure beta emitter (HEW 1970) and so does not contribute to the gamma radiation emissions. Astatine-218 and ^{210}Tl have branching ratios of 0.02% (HEW 1970) and are thus not significant relative to other daughters of ^{222}Rn in the uranium series. We have chosen to neglect these insignificant radionuclides. Preliminary calculations have indicated that three nuclides, ^{214}Bi, ^{214}Pb, and ^{208}Tl, account for about 95% of the calculated exposure rates. It is clear that we could eliminate more radionuclides from consideration, but we have chosen to retain in calculations those nuclides for which measurements are available, and the short-lived daughters of ^{220}Rn and ^{222}Rn.

In the three decay series, there are a number of radionuclides with short half lives, relative to the storage time of the Silo materials. For these nuclides we assume concentrations equal to the parent nuclide. Table J–46 lists the (remaining) radionuclides to be considered for the K-65 and metal oxide sources, the method used to determine the concentration (either from the measured concentrations or assumed at equilibrium for short-lived daughters), along with the decay fractions for the daughters (HEW 1970).

A few nuclides require further explanation. For the metal oxide material in Silo 3, there are adequate measurements of ^{231}Pa. However, ^{231}Pa was not detected in any of the measurements of K-65 material. From the metal oxide measurements, it appears that ^{231}Pa is in equilibrium with its daughter, ^{227}Ac. Thus, for the K-65 material we assume that ^{231}Pa is present at the same concentrations as ^{227}Ac.

Similarly, ^{224}Ra was not detected in any of the measurements of K-65 material. In addition, the average concentration of ^{224}Ra measured in the metal oxide material seems anomalously low. Because the half life of ^{224}Ra, 3.66 days (Walker et al. 1989), is short relative to the half life of its parent ^{228}Th, 2.913 y (Walker et al. 1989), ^{224}Ra is expected to be present in equilibrium with ^{228}Th. Thus, for both the metal oxide and K-65 material, we assume that ^{224}Ra is present at the same concentration as ^{228}Th.

Further, the half lives of ^{228}Ra, 5.76 y, ^{228}Ac, 6.15 h, and ^{228}Th are relatively short compared to the storage time of over 30 years, and compared to the half life of ^{232}Th, 1.40×10^{10} y (Walker et al. 1989). Thus, these three nuclides are expected to be present (for

Table J–46. Radionuclides Considered for K-65 and Metal Oxide Material Source Terms for MicroShield Direct Exposure Calculations

Radionuclide	Determination	Decay fraction	Radionuclide	Determination	Decay fraction
^{227}Ac	measurements		^{224}Ra	daughter of ^{228}Th	1.00
^{228}Ac	daughter of ^{232}Th	1.00	^{226}Ra	measurements	
^{212}Bi	daughter of ^{228}Th	1.00	^{228}Ra	daughter of ^{232}Th	1.00
^{214}Bi	daughter of ^{226}Ra	1.00	^{220}Rn	daughter of ^{228}Th	1.00
^{231}Pa	measurements[a] or parent of ^{227}Ac [b]	1.00	^{222}Rn	daughter of ^{226}Ra	1.00
			^{228}Th	daughter of ^{232}Th	1.00
^{234}Pa	daughter of ^{238}U	0.0013	^{230}Th	measurements	
^{234m}Pa	daughter of ^{238}U	1.00	^{231}Th	daughter of ^{235}U	1.00
^{212}Pb	daughter of ^{228}Th	1.00	^{232}Th	measurements	
^{214}Pb	daughter of ^{226}Ra	1.00	^{234}Th	daughter of ^{238}U	1.00
^{212}Po	daughter of ^{228}Th	0.640	^{208}Tl	daughter of ^{228}Th	0.360
^{214}Po	daughter of ^{226}Ra	1.00	^{234}U	measurements	
^{216}Po	daughter of ^{228}Th	1.00	^{235}U	measurements	
^{218}Po	daughter of ^{226}Ra	1.00	^{238}U	measurements	

[a] For the metal oxide material, adequate measurements are available and are used.

[b] For the K-65 material, ^{231}Pa was not detected in any measurements. It is assumed in equilibrium with its daughter, ^{227}Ac.

recent sampling, anyway) at concentrations equal to the concentration of ^{232}Th. In addition, analyses were not performed for ^{228}Ac, ^{228}Ra was not detected in any of the K-65 samples, and ^{228}Th was only detected in a few of the K-65 samples. Metal oxide samples showed that ^{228}Th concentrations were similar to ^{232}Th concentrations, although ^{228}Ra concentrations appear anomalously low. Thus, for both the metal oxide and K-65 materials, we assume that ^{228}Ra, ^{228}Ac, and ^{228}Th are present at the same concentrations as ^{232}Th.

One of the laboratory analyses was for the combination of ^{235}U and ^{236}U. However, ^{236}U is not a naturally occurring isotope of uranium. Since the K-65 and metal oxide materials were derived from natural ores, ^{236}U should not be present in these materials. We assume that measurements of $^{235,236}U$ represent concentrations of ^{235}U.

The form of radionuclide concentrations that MicroShield uses is volume concentrations, in units µCi cm^{-3}. To obtain this form, we multiplied the mean mass concentrations, as shown in Table J–5 and Table J–6, by the bulk densities discussed in the previous section. We used the mean concentrations because they are the most representative of the entire volume of material from which the gamma radiations will be emitted. Of course, emissions from material in the center of the Silos, farthest from any wall, will have less impact on direct exposures due to the shielding of the outer material. However, the existing data do not provide information on the radial distribution of radionuclide concentrations. For nuclides that we base on equilibrium assumptions, the measured concentration of the base radionuclide was also multiplied by the decay fraction. The results of these conversions are shown in Table J–47 and Table J–48 for the K-65 material and metal oxide material, respectively.

Table J–47. Radionuclide Concentrations in K-65 Material: for Use in MicroShield Direct Exposure Calculations

Radionuclide	Concentration ($\mu Ci\ cm^{-3}$)	Basis	Radionuclide	Concentration ($\mu Ci\ cm^{-3}$)	Basis
^{227}Ac	6.51×10^{-3}	measured	^{226}Ra	3.54×10^{-1}	measured
^{228}Ac	9.19×10^{-4}	equilibrium	^{228}Ra	9.19×10^{-4}	equilibrium
^{212}Bi	9.19×10^{-4}	equilibrium	^{220}Rn	9.19×10^{-4}	equilibrium
^{214}Bi	3.54×10^{-1}	equilibrium	^{222}Rn	3.54×10^{-1}	equilibrium
^{231}Pa	6.51×10^{-3}	equilibrium	^{228}Th	9.19×10^{-4}	equilibrium
^{234}Pa	9.52×10^{-7}	equilibrium	^{230}Th	5.73×10^{-2}	measured
^{234m}Pa	7.32×10^{-4}	equilibrium	^{231}Th	5.59×10^{-5}	equilibrium
^{212}Pb	9.19×10^{-4}	equilibrium	^{232}Th	9.19×10^{-4}	measured
^{214}Pb	3.54×10^{-1}	equilibrium	^{234}Th	7.32×10^{-4}	equilibrium
^{212}Po	5.88×10^{-4}	equilibrium	^{208}Tl	3.31×10^{-4}	equilibrium
^{214}Po	3.54×10^{-1}	equilibrium	^{234}U	8.56×10^{-4}	measured
^{216}Po	9.19×10^{-4}	equilibrium	^{235}U	5.59×10^{-5}	measured
^{218}Po	3.54×10^{-1}	equilibrium	^{238}U	7.32×10^{-4}	measured
^{224}Ra	9.19×10^{-4}	equilibrium			

Table J–48. Radionuclide Concentrations in Metal Oxide Material: for Use in MicroShield Direct Exposure Calculations

Radionuclide	Concentration ($\mu Ci\ cm^{-3}$)	Basis	Radionuclide	Concentration ($\mu Ci\ cm^{-3}$)	Basis
^{227}Ac	3.72×10^{-4}	measured	^{226}Ra	1.90×10^{-3}	measured
^{228}Ac	5.01×10^{-4}	equilibrium	^{228}Ra	5.01×10^{-4}	equilibrium
^{212}Bi	5.01×10^{-4}	equilibrium	^{220}Rn	5.01×10^{-4}	equilibrium
^{214}Bi	1.90×10^{-3}	equilibrium	^{222}Rn	1.90×10^{-3}	equilibrium
^{231}Pa	3.57×10^{-4}	measured	^{228}Th	5.01×10^{-4}	equilibrium
^{234}Pa	1.25×10^{-6}	equilibrium	^{230}Th	3.28×10^{-2}	measured
^{234m}Pa	9.60×10^{-4}	equilibrium	^{231}Th	6.39×10^{-5}	equilibrium
^{212}Pb	5.01×10^{-4}	equilibrium	^{232}Th	5.01×10^{-4}	measured
^{214}Pb	1.90×10^{-3}	equilibrium	^{234}Th	9.60×10^{-4}	equilibrium
^{212}Po	3.21×10^{-4}	equilibrium	^{208}Tl	1.80×10^{-4}	equilibrium
^{214}Po	1.90×10^{-3}	equilibrium	^{234}U	9.46×10^{-4}	measured
^{216}Po	5.01×10^{-4}	equilibrium	^{235}U	6.39×10^{-5}	measured
^{218}Po	1.90×10^{-3}	equilibrium	^{238}U	9.60×10^{-4}	measured
^{224}Ra	5.01×10^{-4}	equilibrium			

Radionuclide Concentrations in K-65 Silos Head Spaces

The direct exposures calculations also require information about the radionuclides present in the head space gases of the Silos. Since the Metal Oxide Silo was almost completely filled with waste materials, we consider its head space volume to be negligible.

For the K-65 Silos, the head space volume is significant. The Silos were relatively closed to the environment, even before the dome penetrations were sealed. Thus, there would have been little movement of air in the head space, and thus little resuspension of particulate material from the surface of the K-65 material into the head space. Thus the only radionuclides present in the head spaces of the K-65 Silos would have been ^{222}Rn, which diffused out of the K-65 material, and the short-lived daughters of ^{222}Rn, ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po, which are produced by the decay of the Rn. From work discussed in our Task 4 Report (Killough et al. 1993), we found that the later Rn daughters, ^{210}Pb, ^{210}Bi, and ^{210}Po, do not contribute significantly to direct exposures. These later daughters are not considered for the head space.

Earlier in this Appendix we discussed the ^{222}Rn concentrations in the head spaces of the K-65 Silos. For the period 1980–1987, we determined the mean ^{222}Rn concentrations based on measurements made in 1987 (see page J–29). For 1959–1979, we calculated the median Rn concentrations (see page J–45). For the earliest time period, 1952–1958, the concentrations used are the same as for 1959–1979; but recall that different heights of the K-65 material and head space are applied in the exposure calculations. For 1988, the concentrations used are the same as for 1980–1987. Because the ventilation rates of the Silo head spaces are quite low, we assume that the Rn daughters in the head space and deposited on surfaces in the Silo are in equilibrium with the Rn in the head space. We assume that the daughters are uniformly distributed in the head space gas, although some of the daughters would be plated out on the Silo walls and dome. However, since the source media in this case is air, which will provide little attenuation, the exact distribution of the radionuclides is not important. Thus, the concentrations of Rn daughters are assumed the same as the concentrations of Rn. The Rn and Rn daughter concentrations in head space to be used for direct exposure calculations are shown in Table J–49.

Table J–49. Radionuclide Concentrations in K-65 Silos Head Spaces: for Use in MicroShield Direct Exposure Calculations

	Concentration (μCi cm^{-3})	
Period	^{222}Rn	^{222}Rn daughters[a]
1952–1958	2.5×10^{-3}	2.5×10^{-3}
1959–1979	2.5×10^{-3}	2.5×10^{-3}
1980–1987	2.62×10^{-2}	2.62×10^{-2}
1988	2.62×10^{-2}	2.62×10^{-2}

[a] Concentrations of each of the short-lived daughters ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po.

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ANNEX 1 TO APPENDIX J

MEASURED RADON CONCENTRATIONS AT K-65 AREA FENCELINE

Table J1–1. Average ^{222}Rn Concentrations (pCi L^{-1}) at K-65 Area Fenceline Monitoring Stations; Results from FMPC Routine Monitoring[a]

Nominal period	Monitoring Dates	K65 A	K65 B	K65 C	K65 D	K65 E	K65 F	K65 G	K65 H
2nd qtr 1987	03/04/87–06/10/87	9.6	16.2	14.9	10.6	8.3	11.3	4.2	2.7
3rd qtr 1987	06/11/87–09/05/87	5.4	10.8	11.2	11.5	6.7	11	4.3	2.6
4th qtr 1987	09/06/87–01/07/88	3.2	7.2	6.1	6.8	4.5	3.7	1.8	2.9
1st qtr 1988	01/08/88–03/05/88	4.55	4.75		3.1	5.2	2.1	1.9	4.25
2nd qtr 1988	03/06/88–06/02/88	3.3	3.2	3.8	5.1	3.9	3.3	2.45	3.4
3rd qtr 1988	06/03/88–09/03/88	9.4	9.25	4.2	13.5	4.0	3.55	2.6	4.35
4th qtr 1988	09/04/88–02/01/89	4.25	6.4	9.15	16.95	17.25	12.85		5.8
1st qtr 1989	02/02/89–03/05/89	3.9	3.95	8.3	7.1	9.45	15.2	3.6	3.65
2nd qtr 1989	03/06/89–06/11/89	2.15	2.15	3.0	4.6	3.9	4.35	1.85	2.25
3rd qtr 1989	06/12/89–09/11/89	3.3	2.45	4.7	6.25	5.1	5.05	3.0	7.2
4th qtr 1989	09/12/89–01/24/90	4.0	8.65	7.25	6.35	3.35		7.9	5.4
1st qtr 1990	01/25/90–03/02/90	4.05	11.45	6.0	4.4		2.25	3.3	1.25
2nd qtr 1990	03/03/90–07/04/90	2.95	4.3		3.85	2.75	2.85	2.15	2.55
3rd qtr 1990	07/04/90–10/12/90	2.05	2.5	3.1	3.65	3.75	3.75	2.95	1.65
4th qtr 1990	10/12/90–01/07/91	2.7	4.55	2.75	3.05	2.25	3.5	2.5	1.7
1st qtr 1991	01/08/91–03/08/91	2.8	4.7	4.3	5.4	4.7	5.1	3.9	2.6
2nd qtr 1991	03/09/91–07/02/91	8.7	6.6	12.6	6.8	4.7	5.5	3.5	3.3
3rd qtr 1991	07/02/91–10/01/91	8.7	42.9	32	6.2	8.6	18	1.9	2.2
4th qtr 1991	10/01/91–01/07/92	16.9	12.6	15.1	15.5	15.5	14.5	8.9	3.8

[a] Ref. Byrne 1992a. Concentrations given here are the average of (typically two) results for Type F detectors.

Table J1–1. Average ^{222}Rn Concentrations (pCi L^{-1}) at K-65 Area Fenceline Monitoring Stations; Results from FMPC Routine Monitoring (continued)[a]

Nominal period	Monitoring Dates	K65 I	K65 J	K65 K	K65 L	K65 M	K65 N	K65 O	K65 P	Mean[b]
2nd qtr 1987	03/04/87–06/10/87	3.4	4.7	5.8	12.5	12.1	10.2	7.8	6.8	8.82
3rd qtr 1987	06/11/87–09/05/87	3.7	3.1	3.7	5.0	4.2	3.9	2.9	2.2	5.76
4th qtr 1987	09/06/87–01/07/88	3.6	4.1	.	5.1	4.5	6.3	6.7	5.1	4.77
1st qtr 1988	01/08/88–03/05/88	4.8	5.9	3.85	6.55	4.1	4.45	3.3	2.3	4.07
2nd qtr 1988	03/06/88–06/02/88	5.0	3.6	4.6	6.75	5.4	4.0	3.05	2.95	3.99
3rd qtr 1988	06/03/88–09/03/88	2.55	3.15	4.3	8.6	5.35	3.65	4.0	2.8	5.33
4th qtr 1988	09/04/88–02/01/89	5.65	4.05	3.45	26.5	19	9.55	6.75	4.7	10.15
1st qtr 1989	02/02/89–03/05/89	4.7	4.95	7.3	10.65	10.95	14.2	5.85	3.75	7.34
2nd qtr 1989	03/06/89–06/11/89	2.75	3.0	3.5	7.5	5.25	3.5	2.85	2.8	3.46
3rd qtr 1989	06/12/89–09/11/89		3.0	4.95	9.5	4.35	4.6	2.9	1.45	4.52
4th qtr 1989	09/12/89–01/24/90	5.7	6.55	5.5	10.85	7.5	4.4	5.15	2.05	6.04
1st qtr 1990	01/25/90–03/02/90	1.8	1.5	2.9	2.3	2.9	2.6	2.05	1.8	3.37
2nd qtr 1990	03/03/90–07/04/90	2.2	1.15	1.45	2.25	2.25	2.35	1.8	1.185	2.40
3rd qtr 1990	07/04/90–10/12/90	1.7	1.15	1.25	2.1	1.95	2.2	1.65	0.63	2.25
4th qtr 1990	10/12/90–01/07/91	2.1	1.85	1.6	2.35	2.9	3.7	2.05	1.4	2.56
1st qtr 1991	01/08/91–03/08/91	4.0	1.7	1.9	3.0	4.5	3.0	6.9	2.1	3.79
2nd qtr 1991	03/09/91–07/02/91	2.4	1.9	1.5	10.3	6.7	5.1	3.5	1.5	5.29
3rd qtr 1991	07/02/91–10/01/91	2.0	1.7	2.0	2.2	2.5	4.2	4.1	1.0	8.76
4th qtr 1991	10/01/91–01/07/92	4.4	3.0	3.7	8.2	10.8	15.2	15.3	5.5	10.56

[a] Ref. Byrne 1992a. Concentrations given here are the average of (typically two) results for Type F detectors.
[b] Mean concentration for all locations for the given quarter.

ANNEX 2 TO APPENDIX J

SUMMARY OF CALCULATIONS OF CURRENT ESTIMATES OF RADON AND RADON DAUGHTER RELEASES FROM K-65 SILOS AND K-65 MATERIAL

INTRODUCTION

In this Annex we provide brief, summary information about the calculations of our current estimates of Rn and Rn daughter releases from the K-65 Silos and from K-65 material. Calculational approaches are first summarized. Table J2–1 shows, for each separate time period, the available, useful information for performing the release calculations; the information lacking, that would be useful to improve estimates; and the general approach to the calculations of releases. Table J2–2 summarizes the principal release rates that are calculated, page references for the detailed discussions, and lists of the parameters required for each calculation. Information about the parameters used in the calculations is summarized in Table J2–3. This table lists the parameters used, categorizes the primary types of information available for determining the parameter distributions, provides page references for deatiled information about the parameters, and describes the distributions chosen to represent the parameters, with the parameters shown generally in the order they are discussed in the text of this Appendix.

The remainder of this Annex provides summaries of the primary equations used in the calculations of the current estimates of Rn and Rn daughter releases.

Table J2–1. Data Availability and Calculational Approaches for Radon and Radon Daughter Releases

Period (nominally)	Information available	Information lacking[a]	General approach to release estimates
Releases from K-65 Material Stored in Drums on Plant 1 Pad			
October 1951–June 1953	Quantity of drums received through July 1952; ^{226}Ra concentration in K-65 material (indirect); estimated density and moisture content of K-65 material.	Radon diffusion coefficient, Rn emanation fraction, and porosity of K-65 material; fractional rate of Rn release from the drums.	Radon-222, from decay of ^{226}Ra in K-65 material, diffuses through K-65 material into air space in storage drum. A fraction (an assumed value is used) of the Rn in the air space is released, through penetrations in the drum, into the atmosphere.
Releases from K-65 Material in K-65 Silos			
[1952–1953] = mid-July 1952–mid-June 1953 (operation of Silo 1); [1953–1958] = mid-June 1953–mid-September 1958 (operation of Silo 2)	General information about practices for operating the K-65 Silos; estimated average concentrations of radium-226 for Silo 1 and Silo 2.	Detailed production information for slurrying and decanting operations; all other[b].	Releases based on calculated releases for 1959–1979, with factors (using assumed values) to account for reduced emissions during operations, due to greater moisture content of the K-65 material, and for the differing ^{226}Ra concentrations in the two Silos.
[1959–1979] = mid-September 1958–June 1979	Exposure rates on silo dome surfaces, prior to and after the dome penetrations were sealed (in June 1979); data to calculate volume of silo air space; estimated rate of release of Rn from K-65 material into silo air space, calculated for 1980–1987 period.	Radon concentration in silo air space; ventilation rate of silos.	Radon concentration is estimated based on value for 1980–1987 and ratio of silo dome exposure rate for 1959–1979 to exposure rate for 1980–1987. Radon release rate from K-65 material into silo air space is based on value for 1980–1987, with correction for silo air Rn concentration. Total Rn released is then quantity released from K-65 material into silo air minus quantity that decays while in the silo.
[1980–1987] = July 1979–December 1987	Radon concentration in silo air space; limited data on temperature and pressure changes in silo air; data to calculate volume of silo air space; limited data on thickness of concrete domes of silos; literature values for porosity and Rn diffusion coefficient of concrete of silo domes.	Radon diffusion coefficient, Rn emanation fraction, and porosity of K-65 material; ventilation rate of silo air space due to wind across domes.	Air exchange releases: activity of Rn in silo air is based on concentration and volume; fractional ventilation rate of silo air space due to volume expansion and contraction is based on temperature monitoring; release is activity in air space multiplied by fractional ventilation rate. Diffusion releases: Rn in silo air space diffuses through concrete domes of silos into outside air.
1988	Measurements of Rn concentrations on the K-65 Area fenceline for 1987–1991.	Description of Rn release mechanism; concentration of Rn in silo air.	Total releases are based on releases for 1980–1987 period and ratio of measured Rn concentration for 1988 to concentration for 1980–1987.
Radon Daughter Releases from K-65 Silos and Drummed K-65 Material			
All time periods	Estimated Rn release rates for all periods.	Silo air Rn daughter concentration; Rn daughter release factors.	Releases of Rn daughters are based on Rn releases, with factors to account for Rn equilibrium fraction and daughter deposition during release.

[a] There are no direct data on quantities of Rn released, for any period. There also are no data on Rn diffusion coefficient, Rn emanation fraction, and porosity of the K-65 material, for any period.

[b] For this operational period of the Silos, there are essentially no data of the types found for other years, that can be used to estimate Rn releases.

Table J2–2. General Summary of Calculations of Radon and Radon Daughter Releases, for Current Estimates

To calculate:	Description	Page	Basic parameters required
$Q_{exch,post}$	Air exchange Rn release rate from K-65 Silos after sealing of Silos, 1980–1987.	J–27	$C_{a,post}$; $\Delta T/T_0$; $\lambda_{v,wind}$; V_0.
$Q_{diff,post}$	Diffusion Rn release rate from K-65 Silos after sealing of Silos, 1980–1987.	J–34	$C_{a,post}$; ε_c; λ_{Rn}; l_c; L; A_{dome}.
Q_{pre}	Total Rn release rate from K-65 Silos before sealing of Silos, 1959–1979.	J–38, J–41	$C_{a,post}$; V_0 ; λ_{Rn}; $\Delta T/T_0$; $\lambda_{v,wind}$; $Q_{diff,post}$; $[(\varepsilon_w l_w + h)/\varepsilon_w l_w]$; X_{bkg}; X_{pre}; X_{post}.
Q_{1988}	Total Rn release rate from K-65 Silos, 1988.	J–46	$Q_{exch,post}$; $Q_{diff,post}$; R_{mon}.
Q_{52-53}	Total Rn release rate from K-65 Silos during operation of Silo 1, 1952–1953.	J–53	Q_{pre}; $f_{Ra,1}$; f_{op}.
Q_{53-58}	Total Rn release rate from K-65 Silos during operation of Silo 1, 1953–1958.	J–53	Q_{pre}; $f_{Ra,1}$; $f_{Ra,2}$; f_{op}.
Q_{dr}; $R_{dr,51}$; $R_{dr,52}$; $R_{dr,53}$	Radon release rate per drum of stored K-65 material; annual Rn releases from drummed K-65 material.	J–55	$[Ra]_{dr}$; EF_{dr}; ρ_{dr}; g_{dr}; λ_{Rn}; A_{dr}; N_{1951}; N_{1952}; N_{1953}; M_{dr}; U_D; U_{dr}; W_{dr}; $[(\lambda_v + \lambda_d)/\lambda_{eff}]_{post}$; f_{dr}.
$Q_{daught,i}$	Radon daughter release rates, for the different time periods.	J–64	Radon release rates (the various Q, and R_{dr}); F_1; $F_{2,dr}$; $F_{2,52-58}$; $F_{2,pre}$; $F_{2,post,exch}$; $F_{2,post,diff}$; $F_{2,1988}$.

Table J2–3. Parameter Distributions for Monte Carlo Calculations of Current Estimates of ^{222}Rn and Daughter Releases from K-65 Silos and Drummed K-65 Material

Parameter	Units	Primary basis: Site-specific information	Primary basis: Literature	Primary basis: Scientific judgment	Page	Distribution	Descriptive statistics[a]
$C_{a,post}$	pCi L^{-1}	X			J–28	normal	mean = 2.62×10^7; stdev = 4.1×10^6
$\Delta T/T_0$	d^{-1}	X			J–31	normal[b]	mean = 0.0297; stdev = 0.0138
$\lambda_{v,wind}$				X	J–34	known[c]	value = 0
V_0	ft^3	X			J–29	uniform	min = 40,000; max = 62,000
λ_{Rn}	d^{-1}		X		J–37	known[c]	value = 0.18129
ε_c			X		J–35	uniform	min = 0.16; max = 0.265
l_c	cm		X		J–36	uniform	min = 14.5; max = 23
L	cm	X			J–36	uniform	min = 3; max = 4
A_{dome}	ft^2	X			J–36	known[c]	value = 5300
X_{bkg}	mR h^{-1}	X			J–42	uniform	min = 35.5; max = 76
X_{pre}	mR h^{-1}	X			J–42	uniform	min = 65 or X_{bkg}, whichever is larger; max = 90
X_{post}	mR h^{-1}	X			J–42	uniform	min = 168; max = 400
$\dfrac{(\varepsilon_w l_w + h)}{\varepsilon_w l_w}$				d	J–40	known[c]	value = 6.35
R_{mon}		X			J–52	normal[b]	mean = 0.612; stdev = 0.282
$f_{Ra,1}$		X			J–55	normal	mean = 1.26; stdev = 0.157
$f_{Ra,2}$		X			J–55	normal	mean = 0.717; stdev = 0.107
f_{op}				X	J–55	uniform	min = 0; max = 1.0
$[Ra]_{dr}$	pCi g^{-1}	X			J–59	uniform	min = 306,800; max = 890,700
EF_{dr}			X		J–59	uniform	min = 0.1; max = 0.4
ρ_{dr}	g cm^{-3}	X			J–59	uniform	min = 0.8; max = 1.2
g_{dr}		X			J–59	normal	mean = 2.98; stdev = 0.24
A_{dr}	cm^3	X			J–61	known[c]	value = 2500
M_{dr}	dry weight fraction	X			J–60	uniform	min = 0.2; max = 0.6 or $(1/\rho_{dr} - 1/g_{dr})$, whichever is smaller
f_{dr}				X	J–61	uniform	min = 0; max = 1.0
W_{dr}	lb	X			J–61	uniform	min = 400; max = 600
U_D				X	J–61	lognormal	GM = 1; GSD = 2

[a] Abbreviations: stdev = standard deviation; min = minimum; max = maximum; GM = geometric mean; GSD = geometric standard deviation.
[b] Actually this distribution is truncated on the low side at zero.
[c] "known" indicates that a single value is used in the calculations.
[d] This parameter is based on other parts of our calculations.

Table J2-3. Parameter Distributions for Monte Carlo Calculations of Current Estimates of ^{222}Rn and Daughter Releases from K-65 Silos and Drummed K-65 Material (continued)

		Primary basis					
Parameter	Units	Site-specific information	Literature	Scientific judgment	Page	Distribution	Descriptive statistics[a]
U_{dr}				X	J–62	lognormal	GM = 1; GSD = 1.4
$\left[\frac{\lambda_v + \lambda_d}{\lambda_{eff}}\right]_{post}$				[d]	J–62	normal	mean = 0.156; stdev = 0.051
N_{1951}	drum-mo.	X			J–62	known[c]	value = 6500
N_{1952}	drum-mo.	X			J–62	known[c]	value = 110,000
N_{1953}	drum-mo.	X			J–62	known[c]	value = 17,000
F_1				X	J–65	uniform	min = 0.8; max = 1.0
$F_{2,dr}$				X	J–65	uniform	min = 0; max = 0.5
$F_{2,52-58}$				X	J–65	uniform	min = 0.8; max = 1.0
$F_{2,pre}$				X	J–65	uniform	min = 0.8; max = 1.0
$F_{2,post,exch}$				X	J–65	uniform	min = 0.5; max = 1.0
$F_{2,post,diff}$				X	J–65	uniform	min = 0; max = 0.5
$F_{2,1988}$				X	J–65	uniform	min = 0; max = 0.5

[a] Abbreviations: stdev = standard deviation; min = minimum; max = maximum; GM = geometric mean; GSD = geometric standard deviation.
[c] "known" indicates that a single value is used in the calculations.
[d] This parameter is based on other parts of our calculations.

EQUATIONS FOR 1980–1987 AIR EXCHANGE RELEASES

As was discussed previously, the air exchange releases for the period 1980 to 1987 can be calculated by:

$$Q_{exch,post} = C_{a,post}\lambda_{v,post}V_0 \tag{J–14}$$

where

$$\lambda_{v,post} = \lambda_{v,\Delta T} + \lambda_{v,wind} \tag{J–17}$$

For these calculations, $\lambda_{v,wind}$ is set equal to zero. From equation J–20, we substitute for $\lambda_{v,\Delta T}$, to obtain:

$$\lambda_{v,post} = \Delta T/T_0 \tag{J–84}$$

This last (equation J–84) is the equation used in the spreadsheet calculations, with no units conversion needed.

Then, the release rate is calculated in the spreadsheet as:

$$Q_{\text{exch,post}} = C_{\text{a,post}} \lambda_{\text{v,post}} V_0 \text{CF}_1 \tag{J–85}$$

where CF_1 is the units conversion factor. The units of the result, $Q_{\text{exch,post}}$, are Ci y^{-1}. The units used for the parameters are pCi L^{-1} for $C_{\text{a,post}}$; d^{-1} for $\lambda_{\text{v,post}}$; and ft^3 (per silo) for V_0. Thus,

$$\begin{aligned} \text{CF}_1 &= \left(10^{-12} \text{ Ci pCi}^{-1}\right)\left(365.25 \text{ d y}^{-1}\right)\left(28.317 \text{ L ft}^{-3}\right)(2 \text{ silos}) \\ &= 2.0686 \times 10^{-8} \text{ Ci d L pCi}^{-1} \text{ y}^{-1} \text{ ft}^{-3} \end{aligned} \tag{J–86}$$

EQUATIONS FOR 1980–1987 DIFFUSION AND TOTAL RELEASES

As was previously discussed, the diffusion release rates are calculated as:

$$Q_{\text{diff,post}} = J A_{\text{dome}} \tag{J–24}$$

where

$$J = \frac{\varepsilon_c \lambda_{\text{Rn}} l_c C_{\text{a,post}}}{\sinh\left(\frac{L}{l_c}\right)} \tag{J–23}$$

These two equations are combined to give:

$$Q_{\text{diff,post}} = \frac{A_{\text{dome}} \varepsilon_c \lambda_{\text{Rn}} l_c C_{\text{a,post}} \text{CF}_2}{\sinh\left(\frac{L\left(2.54 \text{ cm in}^{-1}\right)}{l_c}\right)} \tag{J–87}$$

where we have inserted a units conversion factor for L and the units conversion factor, CF_2, for the result. The units of the result, $Q_{\text{diff,post}}$, are Ci y^{-1}. The units of the parameters are ft^2 (per silo) for A; fraction for ε_c; d^{-1} for λ_{Rn}; cm for l_c; pCi L^{-1} for C_a; and inches for L. Thus, the units conversion factor is:

$$\begin{aligned} \text{CF}_2 &= \left(10^{-12} \text{ Ci pCi}^{-1}\right)\left(30.48 \text{ cm ft}^{-1}\right)^2\left(10^{-3} \text{ L cm}^{-3}\right)\left(365.25 \text{ d y}^{-1}\right)(2 \text{ silos}) \\ &= 6.7866 \times 10^{-10} \text{ Ci L d pCi}^{-1} \text{ ft}^{-2} \text{ cm}^{-1} \text{ y}^{-1} \end{aligned} \tag{J–88}$$

The total release rate for the period is calculated as the sum of the air exchange and diffusion releases:

$$Q_{\text{post}} = Q_{\text{exch,post}} + Q_{\text{diff,post}} \tag{J–89}$$

EQUATIONS FOR TOTAL RELEASE RATE FOR 1959–1979 AND UNCONSTRAINED RELEASE RATE

Preliminary calculations are performed first:

$$C_{a,pre} = C_{a,post}\left(\frac{X_{pre} - X_{bkg}}{X_{post} - X_{bkg}}\right) \tag{J–39}$$

$$\lambda_{d,post} = \frac{Q_{diff,post}}{C_{a,post}V_0}\frac{\left(10^{12}\ \text{pCi Ci}^{-1}\right)}{\left(28.317\ \text{L ft}^3\right)\left(365.25\ \text{d y}^{-1}\right)(2\ \text{silos})} \tag{J–28}$$

$$\lambda_{eff,post} = \lambda_{Rn} + \lambda_{v,post} + \lambda_{d,post} \tag{J–90}$$

$$\frac{\phi}{\lambda_{Rn}} = C_{a,post}\left(\frac{\lambda_{eff,post}}{\lambda_{Rn}}\right)\left(\frac{\varepsilon_w l_w + h}{\varepsilon_w l_w}\right) \tag{J–34}$$

where appropriate conversions factors have been added to equation J–28. These equations are used in the spreadsheet to obtain these intermediate results.

The Rn production rates are then calculated:

$$P_{Rn,post} = C_{a,post}V_0\lambda_{eff,post}\left(28.317\ \text{L ft}^{-3}\right). \tag{J–27}$$

$$P_{Rn,pre} = P_{Rn,post}\left(\frac{\phi/\lambda_{Rn} - C_{a,pre}}{\phi/\lambda_{Rn} - C_{a,post}}\right) \tag{J–32}$$

From equation J–36, the Rn releases for the period 1959 to 1979 are then calculated by:

$$Q_{pre} = \left[P_{Rn,pre} - C_{a,pre}\lambda_{Rn}V_0\left(28.317\ \text{L ft}^{-3}\right)\right]\text{CF}_3 \tag{J–91}$$

where CF_3 is a units conversion factor. The units of the result, Q_{pre}, are Ci y^{-1}. The units used for the parameters are pCi d^{-1} for $P_{Rn,pre}$; pCi L^{-1} for $C_{a,pre}$; d^{-1} for λ_{Rn}; and ft^3 (per silo) for V_0. Thus,

$$\begin{aligned}\text{CF}_3 &= \left(10^{-12}\ \text{Ci pCi}^{-1}\right)\left(365.25\ \text{d y}^{-1}\right)(2\ \text{silos}) \\ &= 7.305\times10^{-10}\ \text{Ci d pCi}^{-1}\ \text{y}^{-1}\end{aligned} \tag{J–92}$$

From equation J–35, the unconstrained Rn release rate, $P_{Rn,0}$, is calculated by:

$$P_{\mathrm{Rn},0} = P_{\mathrm{Rn,post}}\left(\frac{\phi/\lambda_{\mathrm{Rn}}}{\phi/\lambda_{\mathrm{Rn}} - C_{\mathrm{a,post}}}\right)CF_2 \tag{J–93}$$

where we have again added a units conversion factor.

EQUATION FOR 1988 RELEASES

The Rn release rate for 1988 is calculated by the following, with no units conversion required:

$$Q_{1988} = Q_{\mathrm{post}}R_{\mathrm{mon}} \tag{J–46}$$

EQUATIONS FOR 1952–1958 RELEASES FROM K-65 SILOS

The release rates of Rn from the K-65 Silos for the operational periods of the Silos are calculated by the following, with no units conversion required:

$$Q_{52-53} = (0.5)Q_{\mathrm{pre}}f_{\mathrm{Ra},1}f_{\mathrm{op}} \tag{J–48}$$

$$Q_{53-58} = (0.5)Q_{\mathrm{pre}}\left(f_{\mathrm{Ra},1} + f_{\mathrm{Ra},2}f_{\mathrm{op}}\right) \tag{J–49}$$

EQUATIONS FOR 1951–1953 RELEASES FROM DRUMMED K-65 MATERIAL

A number of preliminary calculations are first performed, to calculate $\varepsilon_{\mathrm{dr}}$; m_{dr}; D_{dr}; $D_{\mathrm{e,dr}}$; l_{dr}; L_{dr}; and $[(\lambda_v + \lambda_d)/\lambda_{\mathrm{eff}}]_{\mathrm{dr}}$. The equations for these calculations are not repeated here (see pages J–59 through J–61).

The pore space Rn production rate is first calculated by:

$$\phi_{\mathrm{dr}} = \frac{[\mathrm{Ra}]_{\mathrm{dr}}\,EF_{\mathrm{dr}}\rho_{\mathrm{dr}}\lambda_{\mathrm{Rn}}}{\varepsilon_{\mathrm{dr}}} \tag{J–51}$$

The unconstrained Rn flux from the K-65 material in the drums is next calculated:

$$j_{\mathrm{D,dr}} = \sqrt{\frac{D_{\mathrm{e,dr}}\varepsilon_{\mathrm{dr}}}{\lambda_{\mathrm{Rn}}}}(\phi_{\mathrm{dr}})\tanh\left(\frac{L_{\mathrm{dr}}}{l_{\mathrm{dr}}}\right) \tag{J–50}$$

The Rn release rate from a single drum is then calculated as:

$$Q_{\mathrm{dr}} = j_{\mathrm{D,dr}}A_{\mathrm{dr}}\left(\frac{\lambda_v + \lambda_d}{\lambda_{\mathrm{eff}}}\right)_{\mathrm{dr}} U_{\mathrm{dr}}\mathrm{CF}_4 \tag{J–52}$$

where CF_4 is a units conversion factor. Here, the units desired for the result, Q_{dr}, are Ci month^{-1}. The units of the parameters are pCi cm^{-2} s^{-1} for j_{dr}; cm^2 for A_{dr}; and fraction for the ratio $[(\lambda_v + \lambda_d)/\lambda_{eff}]_{dr}$. Thus, the units conversion factor is:

$$\begin{aligned} CF_4 &= \left(2.63 \times 10^{6} \text{ s month}^{-1}\right)\left(10^{-12} \text{ Ci pCi}^{-1}\right) \\ &= 2.63 \times 10^{-6} \text{ s Ci month}^{-1} \text{ pCi}^{-1} \end{aligned} \qquad \text{(J–94)}$$

Finally, the total yearly Rn releases from the stored, drummed K-65 material is calculated by:

$$R_{dr,i} = Q_{dr} N_i \qquad \text{(J–53)}$$

EQUATION FOR RADON DAUGHTER RELEASES

It was shown earlier that the releases of short-lived ^{222}Rn daughters are calculated by:

$$Q_{daught} = Q F_1 F_2 \qquad \text{(J–64)}$$

This equation is applied in the spreadsheet to Q_{52-53}; Q_{53-58}; Q_{pre}; $Q_{exch,post}$; $Q_{diff,post}$; and Q_{1988}; to obtain $Q_{daught,52-53}$; $Q_{daught,53-58}$; $Q_{daught,pre}$; $Q_{daught,exch,post}$; $Q_{daught,diff,post}$; and $Q_{daught,1988}$; respectively. For releases from the drummed K-65 material, daughter releases are calculated by:

$$R_{daught,dr} = R_{dr} F_1 F_2 \qquad \text{(J–95)}$$

EQUATIONS FOR TOTAL RADON AND RADON DAUGHTER RELEASES FOR THE OPERATING PERIOD 1951–1988

The total release quantities of Rn from the FMPC for the site operating period, 1951–1988, are calculated by the following, with no units conversion required:

$$R_{Silos,52-88} = \sum_i Q_i T_i \qquad \text{(J–65)}$$

$$R_{dr,51-53} = \sum_j R_{dr,j} \qquad \text{(J–66)}$$

$$R_{FMPC,51-88} = R_{Silos,52-88} + R_{dr,51-53} \qquad \text{(J–67)}$$

The same equations are used to calculate total releases of Rn daughters, with the following substitutions: $R_{daught,Silos,52-88}$ for $R_{Silos,52-88}$; $Q_{daught,i}$ for Q_i; $R_{daught,dr,51-53}$ for $R_{dr,51-53}$; $R_{daught,dr,j}$ for $R_{dr,j}$; and $R_{daught,FMPC,51-88}$ for $R_{FMPC,51-88}$.

APPENDIX K

OTHER SOURCES AND EPISODIC RELEASES TO THE ATMOSPHERE

INTRODUCTION

A comprehensive review of the History of FMPC Radionuclide Discharges (Boback et al. 1987) was undertaken in the fall of 1988 to assure that all potential sources (current and historical) of airborne uranium and thorium emissions from the Feed Materials Production Center (FMPC) had been adequately assessed (Hill and Dolan 1988; Clark et al. 1989). The study identified six areas where emissions from FMPC to the atmosphere had been potentially underestimated. These included:

- uranium trioxide gulping process in Plant 2/3
- other unmonitored uranium processes
- thorium processes, both monitored and unmonitored
- fugitive emissions from building ventilation
- laboratory hood exhausts
- fugitive emissions from the waste storage area.

Unmonitored uranium emissions from the UO_3 gulping process are addressed in Appendix H. Appendix K of the interim source term report (Voillequé et al. 1991) reviewed and summarized uranium release estimates from the other unmonitored sources for the years 1960–1962. The total uranium release estimates for these sources, which included unmonitored process emissions, building ventilation, laboratory hoods, incinerators, and fugitive emissions from waste pits, were minor compared with other sources such as the Plant 8 and Plant 2/3 scrubbers and the dust collectors from the various FMPC production facilities. Although these sources were believed to be relatively insignificant, the bases for most of the previous estimates were not well documented, and there were no uncertainties associated with them. These shortcomings have been overcome by a thorough review of original sources of information, literature reviews, implementation of alternate models/methods, and reconstruction of releases with uncertainties.

This appendix also includes an assessment of uranium releases from accidents. We separate these into two categories, "non-routine events" and "episodic releases." Clark et al. (1989) estimated hypothetical historic releases of uranium from a variety of non-routine events, such as fires and spills, which did occur rather frequently during the operation of FMPC production facilities. These were not release estimates for actual events, but rather estimated amounts released to the environment based on the frequency of such events and the observed or calculated release quantities for a typical event of that type. Previous estimates of uranium releases for non-routine events during the 1960–1962 period were included in Appendix K of Voillequé et al. (1991), and were comparable to the other unmonitored releases from waste incineration and from fugitive emissions from the FMPC waste pits.

In contrast to the hypothetical non-routine events, "episodic releases" are defined as *actual* historic releases of large enough magnitude and short enough duration to warrant special dose assessment procedures. Several approaches were taken to identifying episodic releases, which include the review of historic documents describing accidental conditions and the examination of environmental monitoring data, particularly air monitoring and gummed-film data. Monitored as well as unmonitored releases may be episodic. Our assessments identified some potential episodic releases not monitored at the source. Identification of episodic releases and estimates of the quantities released are included in this appendix.

The estimates of uranium releases from incinerators and other unmonitored processes will be addressed first, followed by an assessment of fugitive emissions from the waste pits. The last sections of this appendix deal with episodic releases and non-routine events, including a release of radon from the K–65 silos in April 1986.

URANIUM RELEASES FROM WASTE INCINERATION OPERATIONS AT THE FMPC

Boback et al. (1987) list estimated releases of uranium to the atmosphere from five waste incineration systems which have been used at the FMPC. A review of the historic operations of these facilities (as well as an open burn pit) and reconstruction of past releases is presented in the following sections. In addition to estimates of release quantities, other important characteristics of the source terms from incineration, such as the flow rates and temperatures of stack gases and particle sizes, are also addressed here. These parameters were not characterized previously in the interim Task 2/3 report (Voillequé et al. 1991). Releases and release parameters for the earlier incinerators were typically not measured or were measured very infrequently; therefore releases must be estimated indirectly. These indirect methods used included combustion engineering principles, examination of uranium content of incinerator residues and mass balance considerations, published literature on similar processes, and environmental measurements of air, soil, and gummed film in the vicinity of the incinerators.

WASTE INCINERATION IN THE OPEN BURN PIT

The burn pit was located between Pits 3 and 4, west of the production area (Figure K–1). It was constructed in 1957 as a site to excavate clay to line Pits 1 and 2. The burn pit was subsequently used to dispose of laboratory chemicals and to burn combustible materials, including pyrophoric and reactive chemicals, oil, and other low-level contaminated combustible materials (Solow and Phoenix 1987). The boundaries of the burn pit can no longer be distinguished from the covered Pit 4.

Although the solid waste sent to the burn pit was supposed to be uncontaminated or decontaminated, correspondence during 1964–1965 indicates that unexpectedly large amounts of uranium ended up in burn pit residues (Davis and Davies 1964; Audia 1964; Noyes 1965; Starkey 1965a; Klein 1965; Davis 1965a; Davis 1965b).

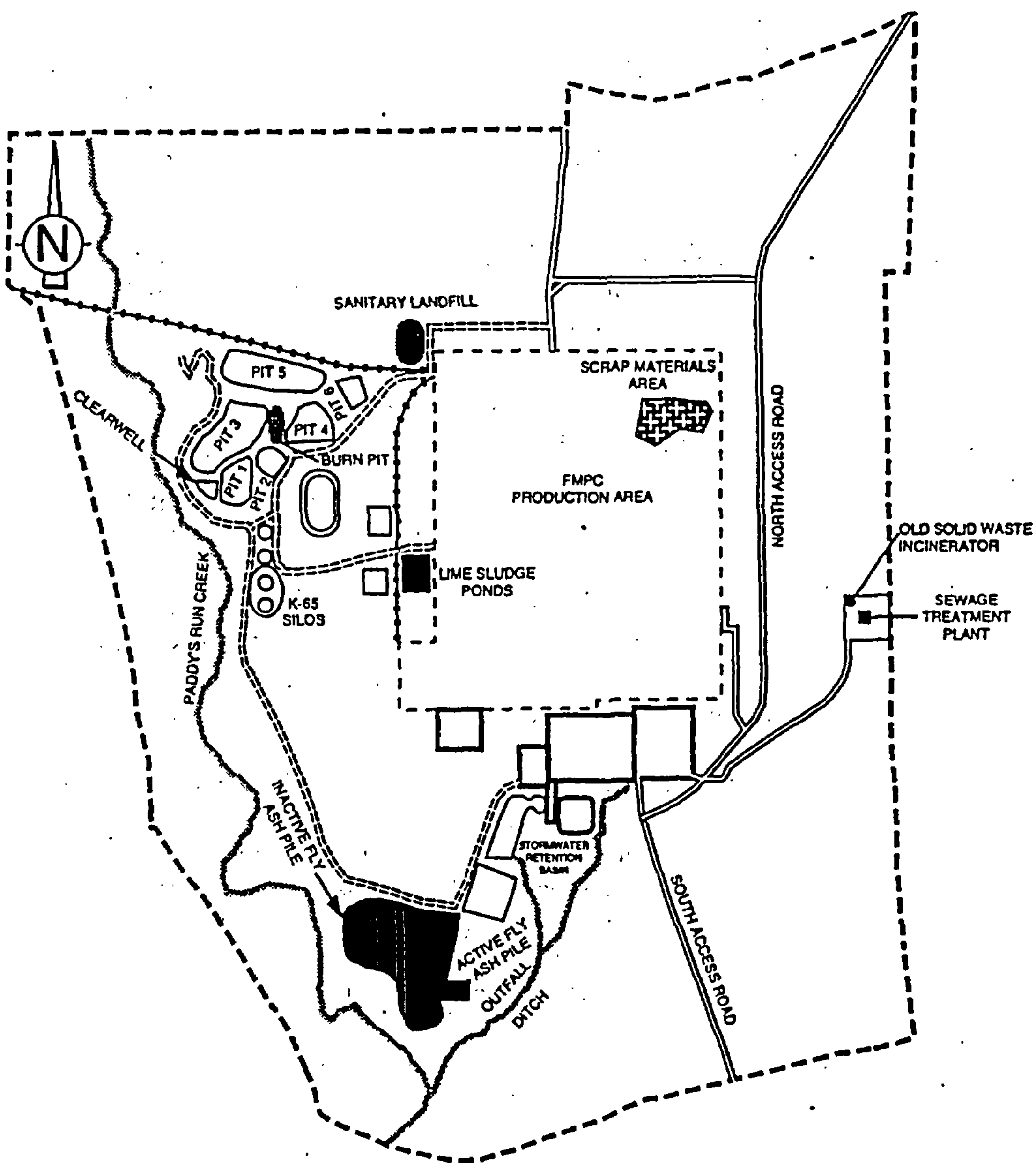

Figure K-1. Location of waste pits, burn pit, and old solid waste incinerator relative to the production area and other landmarks. Incinerators within the production area are shown in a subsequent figure. The fly ash pile shown as "inactive" was used until the mid-1960s.

Audia (1964) indicated a burn pit capacity of about 280,000 cubic feet, with about 240,000 cubic feet present at that time. He argued that a previous estimate (reference not cited) of 104,000 pounds of uranium in the pit was too high but stated that "there is no question that there is a substantial amount." Plans at that time were to move material more toward one end, cover with a layer of clay, and discard trash in the cleared end. Much of the burnable trash then going to the pit was to be routed in the future to the incinerator, where uranium recovery was possible (Noyes 1965). The success of this waste segregation may have been somewhat limited, as evidenced by large scale analyses of ash from burn pit dumpsters and incinerator dumpsters (Davis and Palmer 1968). The uranium content of ash from waste placed in incinerator dumpsters averaged only 2.5 times that of ash from waste placed in burn pit dumpsters.

There has been no previous estimate of uranium releases to air from open burning at the burn pit. This pathway was evaluated by the Fernald Dosimetry Reconstruction Project in conjunction with evaluation of waste burned in the solid waste incinerator.

OLD SOLID WASTE INCINERATOR (1954–1979)

The old solid waste incinerator, which operated from November 16, 1954 to December 31, 1979, was used to dispose of combustible general plant refuse, some of which was likely to contain recoverable uranium, including such items as scrap wood pallets, fuel core shipping containers damaged beyond repair, oily sludges and sanitary sewage sludges. The major portion of these materials was delivered to the incinerator through the use of a "Dempster Dumpster" collection system (Anonymous 1970). Some items, such as broken pallets, shipping boxes, and drums of sludge, were delivered to the incinerator by truck. The incinerator was located east of the production area near the site sewage treatment plant (Figure K–1).

History and Operation of the Old Solid Waste Incinerator

This section gives an overview of the history and operation of the Old Solid Waste Incinerator (OSWI). However, a more detailed chronological history of notes and information, gathered mainly from historic reports of the Industrial Hygiene and Radiation (IH&R) Department, is included as Annex 1 to this appendix.

Grossly contaminated combustibles such as dust collector bags, contaminated gloves, and other items showing high levels of contamination were separated from the general refuse stream at the point of generation (Anonymous 1970). Some attempts were made to burn these grossly contaminated items at the incinerator, but "the activity level of the stack discharge was above an acceptable limit" (Anonymous 1970). In 1962, the types of wastes burned at the incinerator included wood (railroad bracing, broken skids), rags, paper, gloves, clothing, shoes, sanitary sewage sludge, and filter bags and cartridges (Noyes 1962). The ash was processed through the Recovery Plant and the uranium was returned to the production stream.

The following information on the early old solid waste incinerator was obtained from Engineering Drawing 39X–X–00002, "Preliminary Drawing — Incinerator, October 2, 1953," and from NLO memoranda. The old solid waste incinerator was a variation on the Plibrico No. 222 design. In Engineering Drawing 39X–S–00004, "Foundation Plan, Details — Incinerator, February 18, 1954," the incinerator is shown surrounded by chain link fencing on the N, W, and E sides and topped by a slanted, corrugated asbestos roof which was about 20 feet above grade. This roof is visible in an aerial photograph from 1965 (Figure K–2), in which the roof is roughly half as tall as the stack. The physical stack height was 36 feet above grade plus an additional 5 feet of 16 gauge, 4x4 mesh wire cloth, which acted as a spark arrester. Flue gases from the combustion chamber passed through four 90° angles (created by interior baffles) before exiting the stack. These angles served to enhance settling and impingement of entrained ash. There were no secondary burners at this time nor was there filtration of stack gases. A 15 x 24" draft door allowed control of air to the primary combustion chamber. Ashes fell below a grate and were shoveled out into drums via five clean-out doors.

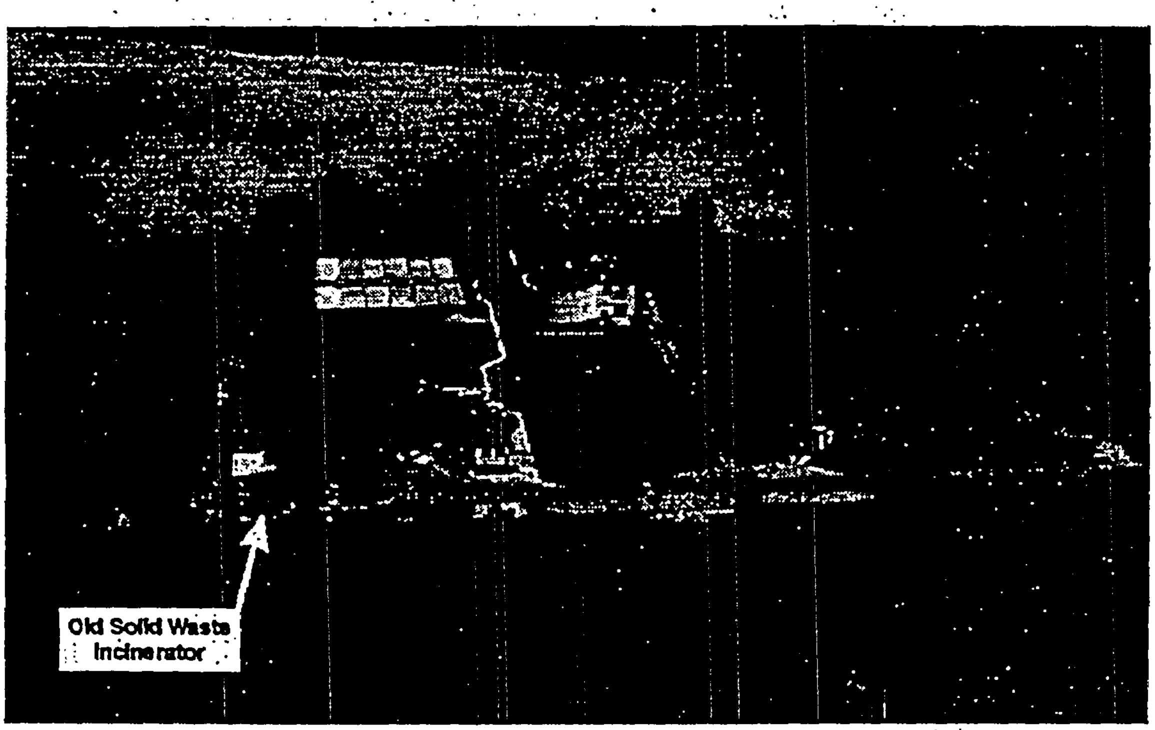

Figure K-2. Photograph of the FMPC sewage treatment plant area in 1965. The old solid waste incinerator is in the NW corner of this area. The dark plume from the stack is blowing approximately W towards the production area. Surrounding land use is grazing and farming.

The OSWI was significantly modified in early 1970 "to improve the performance of the unit in regard to smoke density and particulate discharge" (Anonymous 1970). Noyes (1969) discussed the need to modify the incinerator to comply with Executive Order 11282, "Control of Air Pollution Originating From Federal Installations," because both the visibility standard and the particulate quantity criteria for refuse disposal incinerators were not being met. Plibrico, the manufacturer of the incinerator, proposed the installation of added baffling, additional settling chambers, and secondary combustion as the most economical method of improving the performance of this unit, although 100% compliance with the emission standards in the Executive Order was not guaranteed. They indicated that such a guarantee would be available only with the addition of flue gas scrubbers and/or an assurance that oily wastes would not be charged into the unit.

The construction proposal CP-69-17 (Anonymous 1969), as well as the engineering drawing 39X-G-00009 "Modifications to Plant Incinerator, April 24, 1969," describe the modifications to the plant incinerator. These included:

- movement and replacement of existing stack with one of equal height (36 feet)
- addition of after-burners (one each side @ approximately 2,000,000 BTU h^{-1} each) in a new secondary combustion chamber
- addition of air jets in new secondary combustion chamber
- addition of burn-off compartment for liquid wastes

Nelson (1969) indicated that these modifications to the plant incinerator would necessitate a shutdown of the facility for approximately four weeks (which he estimated would occur in February 1970). During this period, NLO planned to open-burn their refuse at the old burn pit, which was approved by Karl (1969).

After the modifications to the incinerator, it could be described as a multiple chamber in-line incinerator (National Air Pollution Control Administration 1969). The as-built stack height after these modifications was apparently not 36 feet, as the preliminary drawing had indicated. Drawing 39X-M-00012 (September 1972), as well as other sources, shows the incinerator after the modifications had been completed, with a stack height of 45 feet.

In order to evaluate the dispersion of effluents from the OSWI, the stack effluent temperature and flow rate must be estimated. We could locate no original measurements of these parameters in historic documentation, although there apparently were measurements made in the early years (see Annex I). In the application for a permit to operate an air contaminant source, Riestenberg (1978) gave a temperature range of 600–1500 °F for the exit gas from the OSWI. The flow rate is not given on this permit application, but handwritten notes by Grant (1986) give an estimate of 1181 feet per minute for the OSWI. No supporting calculations are shown, although Grant (1993) recalled that his notes were obtained from files which included the measurement data.

We checked the reasonableness of these temperature and flow rate values using standard combustion analysis calculations (National Air Pollution Control Administration 1969). We assumed an incineration rate of 750 lbs h^{-1} (the quoted average for the FMPC incinerator) of a 50%/50% mixture of wood and paper (moisture content 15%) at 200–300% excess air. Radiative heat losses were assumed to be 15% (National Air Pollution Control Administration 1969). The temperature of the exit gas from the OSWI under these conditions was computed to be 1000–1400 °F, which is within the temperature range given on the NLO permit application. The exit velocity of flue gases was estimated to be 780–850 feet per minute, based only on the volume of combustion product gases. Additional excess air would enter the primary combustion chamber by natural draft. Thus the exit velocity in Grant's notes may be considered reasonable. Table K–1 summarizes the physical and operating parameters for the OSWI. Particle size considerations for effluents from all incinerators are discussed later in this appendix.

Table K-1. Summary of Physical and Operating Parameters for the Old Solid Waste Incinerator (operated 11/16/54 to 12/31/79)

Parameter	Value	Reference	Notes
Physical stack height	Through 1969:36 ft After 1970: 45 ft	Engineering Drawings 39X-M-00012 and 39X-X-00002 Grant (1986)	See discussion in text.
Building dimensions	Through 1969: 15.4 x 8.2 x 9 feet tall. After 1970: 39.1 x 8.2 x 9 ft tall	Engineering Drawings 39X-M-00012 and 39X-X-00002	Before 1970 the incinerator building was covered by a sloping, corrugated asbestos roof which was about 20 feet above grade.
Stack inner dimensions	Before 1970: 24 x 70 in., rectangular ($11.7\ ft^2$) After 1970: 27 x 71 in., oval	Engineering Drawings 39X-M-00012 and 39X-X-00002	This lining of the newer stack was oval shaped, with short and long axis dimensions as shown. A 3" width refractory stack is assumed (AEC 1971) for computing the inner dimensions of this stack.
Exhaust gas velocity	780–850 ft min^{-1} 1181 ft min^{-1}	This study Grant (1986)	Based on combustion analysis and stack geometry before 1970.
Exhaust gas temperature	1000–1400 °F 600–1500 °F 1048 °F	This study Riestenburg (1978) Grant (1986)	Based on combustion analysis (see text). No supporting calculations given for other references, but Grant (1993) recalled the value was based on measurements.
Operating schedule	Until 1969: 8 h d^{-1}, 5–6 d wk^{-1}, 49–50 wk y^{-1} (1960–2400 h y^{-1}) 1970–1979: 6 h d^{-1}, 3 d wk^{-1}, 49 wk y^{-1} (882 h y^{-1})	(Karl 1967) (Riestenberg 1978)	The plant-wide waste generation rate in 1967–1968 was 6500 lb d^{-1} (See Table K–4). This corresponds to an operating schedule for the OSWI of 8.7 h d^{-1} at an average firing rate of 750 lb h^{-1}, which is in good agreement with the schedule indicated. When the burning of wooden skids was halted (sometime between 1967 and 1969), the operating schedule was reduced.

Previous Uranium Release Estimates for the Old Solid Waste Incinerator

The previous release estimates (Boback et al. 1987) for the old solid waste incinerator, were 15 kg in 1954, 118 kg y^{-1} for 1955 through 1968, 94 kg y^{-1} for 1969, and 71 kg y^{-1} for 1970 through 1979. The total estimated release for 1954–1979 would thus be ≈2500 kg. The source term estimates were supposedly based on data from several stack emission tests. Investigators from IT Corporation concluded that documentation for estimates of historical releases from non-production sources (including the oil burner and old solid waste incinerator) was limited and included information which conflicted with release estimates reported by Boback et al. (IT 1989).

Reconstruction of Uranium Source Term for the Old Solid Waste Incinerator

A number of original sources of documentation were located and carefully reviewed to permit a reconstruction of the source term for the old solid waste incinerator. During this process, a chronological history of important events and other notes was compiled, which is included as Annex 1 to this appendix. This annex provides a detailed record of relevant changes and correspondence which was used to aid interpretion of the original data. The types of quantitative information we evaluated to estimate the source term from the OSWI fall into the following categories:

- **Category 1.** Stack measurements of uranium and gross alpha activity in stack gases.
- **Category 2.** Uranium content of incinerator residues and mass balance considerations.
- **Category 3.** Environmental measurements of uranium and gross alpha activity in air and deposited on gumpaper around the incinerator.

Although Category 1, effluent monitoring of the stack gases, provides the most relevant data for source term reconstruction, there were very few measurements made. The second category of data was particularly plentiful in the 1960s, when a large emphasis was placed on documenting the uranium content of various plant wastes and residues. The third category (environmental measurements) was most important for evaluating operations in the 1950s, in which a number of tests of burning contaminated materials were undertaken, and measurements of associated environmental contamination were made (see time line in Annex 1). Although a source term is difficult to reconstruct using environmental measurements, they do provide a direct indication of the effect of those early tests on contamination levels in the surrounding environment.

Category 1 information — Stack measurements of uranium and gross alpha in stack gases from the OSWI. Five tests were made of particulate emissions from the incinerator stack in May 1977 (Ross 1977). In all five tests, the measured releases were greater than the Ohio EPA limit of 0.1 lb particulates per 100 lb burned. The uranium content of the waste burned was not documented, but there is no reason to conclude that it was not typical plant waste. The measured loss of uranium during the five tests was 0.06, 0.10, 0.13, 0.15, and 0.17 pounds per hour. Using the Crystal Ball™ uncertainty analysis program (Decisioneering 1992), we defined a custom parameter distribution which described the hourly uranium release rate, using the measured values. This hourly release rate was multiplied by the operating schedule estimate of 882 hours per year for the post-1969 period (Table K–1), which was assumed to be normally distributed with a standard deviation of 10% of the mean. The annual source term forecast based on these stack tests is illustrated in Figure K–3. The median estimate is 52 kg per year with a 5th to 95th percentile range of 20 to 78 kg per year.

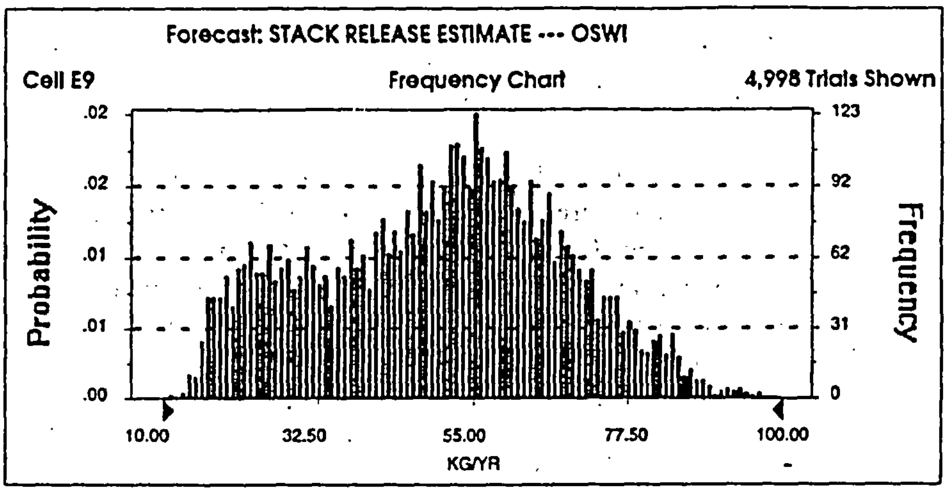

Figure K–3. Revised annual release estimate of uranium from the Old Solid Waste Incinerator in the 1970s, based on stack testing in 1977.

Earlier stack testing at the OSWI tended to be part of operational testing as opposed to compliance testing (see Annex 1). For example, particulate samples were collected from the trash incinerator flue gas in April 1964 to check the air contamination resulting from disposing of various uranium contaminated organic materaials at the incinerator (IH&R monthly report, April 1964; Starkey 1964b). The original analytical data sheets for these tests were located and are presented in Table K–2 below. The average of 18 measurements of uranium in flue gases was 1.7 mg U m^{-3} (range 0.27–5.9).

We computed a source term estimate from the stack testing data in Table K–2 using the Crystal Ball™ uncertainty analysis software (Decisioneering 1992). The concentrations of uranium in the stack gas were defined using a custom distribution which described the actual measurements. In addition to the uranium concentrations in flue gas, the volumetric flow of stack gases and the operating schedule must be estimated. The stack gas velocity was defined by a triangular distribution in which the most probable value (1200 fpm) was documented by Grant (1986), the minimum value (800 fpm) was established by combustion analysis (Table K–1), and the maximum value (1440 fpm) was 20% higher than Grant's estimate. The operating schedule used was 1960–2400 hours per year (Table K–1), with a most probable value at the midpoint of this interval (2180 hours per year).

The estimated uranium source term based on the 1964 stack tests is shown in Table K–3. The median annual source term estimate is 67 kg per year, with a 5th–95th percentile range of 6–250 kg per year.

Table K–2. Stack Sampling of the OSWI in April 1964 to Evaluate the Air Contamination Resulting from Burning Uranium Contaminated Organic Materials

Date	Description of Material Burned[a]	TBP or sludge present?	µg U m^{-3} flue gas: 1st impinger	2nd impinger	Total
4/8	Background; Ad. Blg. trash	No	370	2270	2640
4/9	Background trash	No	1140	590	1730
4/9	Background sample – Shipping and Receiving	No	1060	4850	5910
4/10	Background. N–6 Insp. & WINLO filters	No	830	140	970
4/10	Background. Plt. 9 trash	No	1640	230	1870
4/10	Plant 9	Yes	1910	360	2270
4/10	Plant 2/3 Maint., Boiler Plt	Yes	730	140	870
4/10	Plant 2/3 Maint., Boiler Plt	Yes	360	360	720
4/13	Dumpster E-6	Yes	300	35	335
4/13	Dumpster E-6	Yes	250	20	270
4/13	Dumpster S-9	Yes	325	165	490
4/15	(illegible)	Yes	1200[b]		1200
4/15	(Same as previous sample)	Yes	390[b]		390
4/15	Same as previous two samples, fire stirred	Yes	1500[b]		1500
4/21	Background. Dumpster – Comb. Raff.	No	1320[b]	640[c]	1960
4/21	Background. Dumpster NW4	No	1920[b]	131[c]	2051
4/21	Background	No	4850[b]		4850
4/28	None given	Unknown	1500[b]	160[c]	1660

[a] As recorded on analytical data sheets. "Background" is interpreted to mean typical plant trash, as opposed to the uranium-contaminated organic materials being tested.
[b] Sampled with pleated filter instead of impinger.
[c] Millipore backup filter.

Table K–3. Estimated Uranium Source Term from the OSWI, Based on Stack Testing in 1964

Release Rate	Percentile of Estimated Source Term Distribution: 5%	25%	50%	75%	95%
g U per hour	3	16	31	52	120
kg U per year	6	34	67	110	250

There were only a handful of analytical data sheets located for stack monitoring of the OSWI before 1964, and most were analyzed only for gross alpha, not uranium. Ten samples collected on May 25, 1962 ranged from 61 to 1100 dpm alpha per cubic meter of flue gas. Four samples obtained from the top of the stack on May 11, 1962 ranged from 89 to 240 dpm alpha per cubic meter. If this activity were all due to natural uranium, these samples represent a range of 40–730 μg U per cubic meter of flue gas, which is well within the distribution defined by the 1964 measurements.

Category 2 information — Evaluation of uranium content of incinerator residues and mass balance considerations. In our draft source term report (Voillequé et al. 1991, we used a mass balance approach to provide a basis for the incinerator source term in the 1960s. We chose to use this method in order to take advantage of the large amount of data on uranium in incinerator and burn pit ash residues. The data were obtained by the FMPC to estimate the amount of recoverable uranium in the solid waste streams. The most complete evaluation located was by Stevenson (1968), who tabulated results of a comprehensive program to evaluate the uranium content and production of various plant wastes between October 21, 1967 and April 9, 1968. Production rates of the various types of wastes and the uranium content of the resulting residues after incineration were obtained from Stevenson (1968) and are listed in Table K–4.

In order to apply a mass balance calculation to the residue data, a release fraction (fraction released to air during incineration) is needed. One estimate of the release fraction was reported by Bostick et al. (1991) from emissions testing of uranium-contaminated wastes at the K-1435 Mixed Waste Incinerator in Oak Ridge, TN. These tests indicated that 2.9% of the uranium fed to the incinerator was discharged to stack gases (prior to air cleaning equipment). In addition, Glauberman and Loysen (1964) conducted a survey of AEC contractors to determine the extent of the use of incinerators and their effectiveness for uranium contaminated waste reduction. Two contractors operating incinerators without air cleaning equipment for uranium-contaminated wastes provided estimates of the percent retention of uranium in the ash. One estimate was 99% retention and the other was 95–100% (0–5% release). Four contractors operating incinerators with air cleaning equipment estimated 99% retention of uranium in the ash. For our assessment, we described the release fraction to flue gas as a triangular distribution with a most probable value of 3% and minimum and maximum values of 1 and 5%.

Based on the measured uranium content of incineration residues, plant-wide waste generation rates, and mass balance considerations, an airborne source term estimate for incineration was derived (See Table K–4 for explicit explanation of the calculation.) In addition to the uncertainty in the release fraction, the plant-wide waste generation rate, the %residue, and the %U in residue were also defined as uncertain parameters which were normally distributed with standard deviations of 10% of the mean.

The annual airborne source term calculated using this method is 100 kg U y^{-1}. The distribution of the estimate is roughly normal, with a mean of 102 kg per year. The 5th–95th percentile range is 54–150 kg per year. This source term includes a 7 kg per year atmospheric source term from wastes routed to the burn pit (Table K–4).

Although the late 1960s provided the most data on uranium content of incinerator residues, earlier measurements were compiled from original data sheets and NLO memoranda. Because a number of "special burnings" were known to have occurred, the possible releases of uranium from those tests needed investigation. Figure K–4 illustrates the need for this analysis. The uranium content of incinerator residues from the special burning tests were considerably higher than those obtained for routine burning in the late 1960s and beyond.

Table K–4. Mass Balance Approach to Estimate Uranium Releases to Air from Incineration of Solid Wastes at the FMPC During 1967–1968[a]

	Waste Type						
	Oil Burner Sludge	Skids	Shipping Containers	Sewage Sludge	Incinerator Dumpsters	Burn Pit Dumpsters	Total
Average plant-wide waste generation rate (lb waste d^{-1})[b]	233	2500	210	132	2238	1194	6500
% Residue[b]	35.4	2.7	2.2	35.2	11.6	9.9	—
Residue generation rate (lb residue y^{-1}), based on 240 operating d y^{-1} [b]	19,800	16,200	1100	11,200	62,310	28,370	139,000
% U in residue [b]	9.0	12.21	6.0	"Nil"[c]	4.48	1.66	5.1
U in residue (lb U y^{-1})	1780	1978	66	112	2791	471	7200
U in original waste (lb y^{-1}) [d]	1835	2039	68	115	2877	486	7420
Airborne source term[e] (lb U y^{-1})	55	61	2	3	86	15	220
(kg U y^{-1})	25	28	1	2	39	7	100

[a] Significant figures shown to illustrate calculation, but do not imply this degree of precision.

[b] Based on evaluation of solid waste streams at the FMPC, conducted between October 21, 1967 and April 9, 1968 (Stevenson 1968).

[c] Observed result of "nil" was believed by Stevenson (1968) not to be typical of FMPC sewage sludge, which historically had shown some uranium content. For the calculations in this table, an estimated value of 1% U in sewage sludge residue is used.

[d] From mass balance considerations, *i.e.*, U in original waste = U in residue ÷ $(1-f_a)$, where f_a is the airborne release fraction, with a most probable value of 0.03 (see text).

[e] Airborne source term = U in original waste – U in residue.

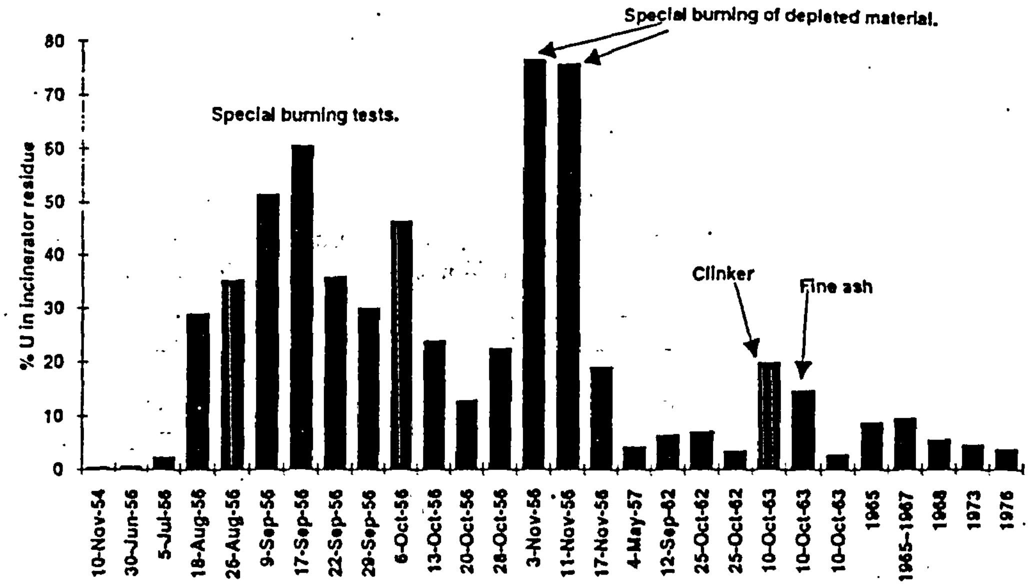

Figure K-4. Uranium content of residues from incineration of solid wastes at the FMPC. With the exception of the Nov 3, 1956 burn, which occured at the burn pit, all residues were collected from the old solid waste incinerator. Data were obtained from analytical data sheets, IH&R department reports, Vath (1967), Audia (1969), Harmon (1973), and Kruezmann and Neblett (1976).

There were 18 days in 1956 when known special burning tests of contaminated solid waste took place at the FMPC. For 16 of those days, data were located on the uranium content of the residue. Using the mass balance approach described above, a source term for each burn was estimated (Table K-5). The footnotes to Table K-5 explain the methodology more explicitly. It was assumed that each special burn lasted for eight hours at the average feed rate of 750 pounds of waste burned per hour (Riestenberg 1978). In addition, the % residue was assumed to be 9%, which is the overall average including all waste types (Table K-4). The total source term for these 18 days of special burning is estimated to be 41 kg (median), which is about 40% of the annual source term from solid waste incineration in the 1960s. Because of the uncertainty in the airborne release fraction, the 5th-95th percentile interval is 22-61 kg.

Category 3 information — Environmental measurements of uranium and gross alpha activity in air and deposited on gumpaper around the incinerator. A number of environmental measurements around the OSWI were made in the 1950s, apparently to evaluate the possibility of incinerating various types of materials as well as to document the uranium losses. The reader is referred to Annex 1 of this Appendix for a chronological history of events relevant to incinerator testing in the 1950s. As mentioned previously, it is difficult to back-calculate a source term from environmental measurements, but they provide an important direct indication of contamination levels during particular events. For example, there were no residue data for two of the 18 known special burns in 1956 (Table K-5), but environmental measurements were made. These environmental data can also indicate the rate of decrease of contamination with distance from the release point. The data presented in this section were obtained from original analytical data sheets, unless noted otherwise, and are discussed in chronological order.

Table K–5. Estimate of Airborne Uranium Source Terms From Known Special Burns of Contaminated Solid Waste at the FMPC in 1956[a]

Date of Special Burning Test	%U in Residue	kg U in Residue[b]	kg U in Original Waste[c]	Airborne Release (kg U)[d]
26 May	ND[e]			
7 June	ND[e]			
23 June	18.1	44.42	45.79	1.37
30-June	0.6	1.59	1.63	0.05
5-July	2.3	5.58	5.76	0.17
18-August	28.9	70.86	73.05	2.19
26-August	35.4	86.89	89.57	2.69
9-September	51.6	126.5	130.4	3.91
17-September	60.78	149.1	153.7	4.61
22-September	36	88.34	91.08	2.73
29-September	30	73.62	75.9	2.28
6-October	46.5	114.1	117.6	3.53
13-October	24	58.9	60.72	1.82
20-October	12.8	31.41	32.38	0.97
28-October	22.4	54.97	56.67	1.70
3-November	76.8	188.5	194.3	5.83
11-November	76	186.5	192.3	5.77
17-November	19	46.63	48.07	1.44
		TOTAL		41

[a] Significant figures shown to illustrate calculation, but do not imply this degree of precision.

[b] kg U in residue = 750 lb waste $h^{-1} \times 8$ h $d^{-1} \times 0.4545$ kg $lb^{-1} \times$ 0.09 kg residue (kg waste)$^{-1} \times$ kg U (kg residue)$^{-1}$ (column 2).

[c] From mass balance considerations, *i.e.*, U in original waste = U in residue ÷ $(1-f_a)$, where f_a is the airborne release fraction, with a most probable value of 0.03 and a range from 0.01 to 0.05.

[d] Airborne source term = U in original waste – U in residue.

[e] No data. However, see environmental measurements in next section.

On May 26, 1956, there was a special burning of contaminated gloves in the OSWI. The wind was from the SW at about 6–8 mph, and there was a drizzle to light rain throughout the day. The measured alpha deposition to gumpaper as a result of this test is illustrated in Figure K–5, and measurements of gross alpha activity in air are tabulated in Table K–6. The gumpaper data suggest a rapid decrease in uranium contamination within 100 m of the incinerator.

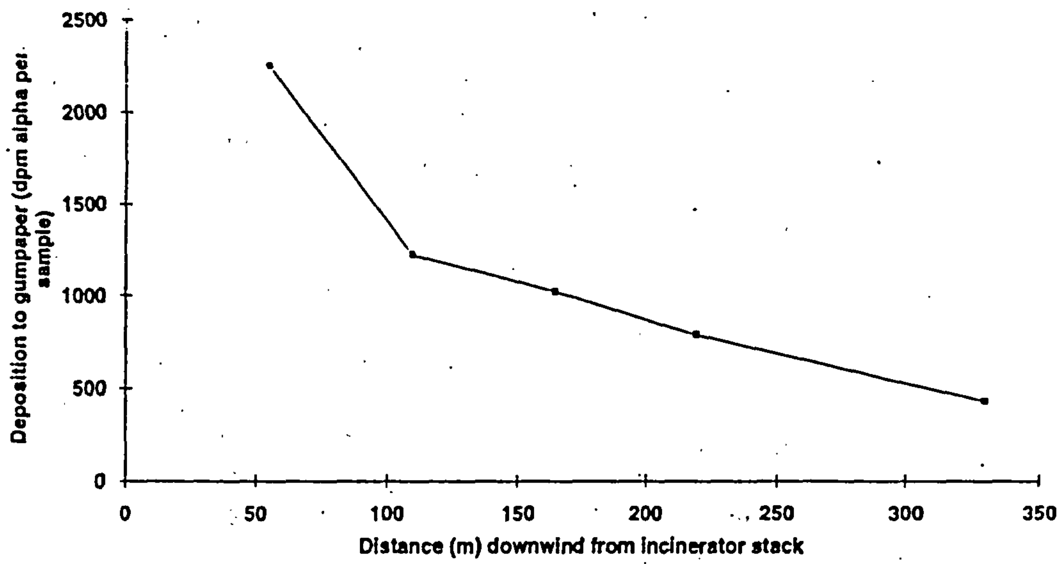

Figure K–5. Measured deposition of fallout to gumpaper during special burning of contaminated gloves in the old solid waste incinerator on May 26, 1956. The sample area of the gumpaper is 1 ft^2 (0.093 m^2) (Shleien et al. 1993).

From Table K–6 and other similar tables to follow, it can be seen that the incineration operation was dusty, with relatively high airborne contamination levels present on the platform where waste was dumped and fired. For some of these special burning tests, the analytical data sheets note that the incinerator operator was wearing respiratory protection. The concentrations in air downwind of the incinerator during the test were quite low, but sampling times were short, and the uncertainties in the measurements would be high. As discussed in Shleien et al. (1993), the conversion of gross alpha concentration measurements in air to uranium concentration is subject to large uncertainty for these short count rates and times. We view the gross alpha activity measurements as a qualitative indication of uranium contamination around the OSWI, and have not used them to reconstruct release estimates.

Table K–6. Air Dust Samples Taken During a Special Burning of Contaminated Gloves in the Old Solid Waste Incinerator on May 26, 1956[a]

Location	Approximate Distance from Stack (m)	Number of Samples	dpm alpha m^{-3} (range of values)
General area sample on incinerator platform	Not Applicable	9	624[b] nd[c] – 1267
Breathing zone sample on incinerator platform	Not Applicable	9	653[b] nd – 1226
5 Stack Lengths in the E, N, and W directions	55	6	4 – 7
10 Stack lengths in the E, N, and W directions	110	6	nd – 3
15 Stack lengths in the E, N, and W directions	165	6	nd – 5
25 Stack lengths in the E, N, and W directions	274	3	nd – 1
100 Stack lengths in the E, N, and W directions	1100	3	nd – 2

[a] In addition to these general air and breathing zone samples, one 230-min stack sample collected during the test was 118 dpm alpha per cubic meter air.
[b] Average
[c] "nd" = Not detectable

On June 7, 1956, a second special burning of contaminated gloves took place at the OSWI. Air dust samples taken during the test are tabulated in Table K–7. The wind was from the west, was very light and somewhat variable. The time of the test burn was 5:00 PM to 10:00 PM. In addition to the environmental measurements shown, one stack sample collected during the duration of the test contained 1080 dpm alpha per cubic meter air.

Table K–7. Air Dust Samples Taken During a Second Special Burning of Contaminated Gloves in the Old Solid Waste Incinerator on June 7, 1956

Location	Approximate Distance from Stack (m)	Number of Samples	dpm alpha m^{-3} (range of values)
General area sample on incinerator platform	Not Applicable	24	131[a] 8 – 662
8 Stack lengths in the NE, E, and SE directions	88	9	nd[b] – 10
10 Stack lengths in the NE, E, and SE directions	110	18	nd – 5

[a] Average. Samples taken while drums were dumped, gloves spread out and burned.
[b] "nd" = Not detectable

On June 27, 1956, 10 air dust samples were taken on the incinerator platform, while dumping and burning of papers was occurring. There was no notation of the type of waste being burned (i.e. contaminated or not). The samples ranged from 3 to 78 dpm alpha per cubic meter.

On June 29, 1956 another set of air dust samples downwind of the incinerator were taken between 8:45 AM and 2:30 PM (Table K–8). There is no notation of the type of waste being burned.

Table K–8. Air Dust Samples Taken During a Burning at the Old Solid Waste Incinerator on June 29, 1956

Location	Approximate Distance from Stack (m)	Number of Samples	dpm alpha m^{-3} (range of values)
5 Stack lengths to the NE (downwind)	55	6	nd[a] –15
20 Stack lengths to the NE (downwind)	220	6	1 –17
25 Stack lengths to the S (downwind)	270	2	2 – 7

[a] "nd" = Not detectable

A load of contaminated shoes was burned at the incinerator on June 30, 1956. The wind during the test was light (4–6 mph) and variable in direction. There were several types of samples taken during that run. The ash which resulted from this test burn was only 0.6% U (Table K–5), indicating that the shoes were not highly contaminated. A 135-min stack sample contained 573 dpm alpha per cubic meter of sampled air. A 154-min high-volume air sample from the base of the incinerator stack (assumed to be outside) measured a concentration of 0.79 dpm alpha per cubic meter air. Twelve air dust samples collected on the incinerator platform, while shoes were burning and were being dumped out of drums and put into the incinerator, ranged from <1 to 224 (average: 54) dpm alpha per cubic meter. A gumpaper sample about 10 stack lengths (110 m) to the E contained roughly twice as much uranium (1 mg per sample) as three other samples placed 10 stack lengths to the N, 25 stack lengths to the NE, and 30 stack lengths to the SW (about 0.4 mg U per sample).

On two separate days in March 1957, air dust samples were taken at varying distances from the incinerator in the downwind direction. The location of the samples was noted on the analytical data sheets as a certain number of paces from the fence. On March 4, the wind was from the NE from 5 to 15 mph and gusty. The first sample was taken at the fence SW of the incinerator. From a recent scale map of the sewage treatment plant area (DOE 1992), we determined that this distance is about 30 feet. The March 4 and March 20 data are plotted together in Figure K–6, assuming a "pace" is equal to 2.5 feet.

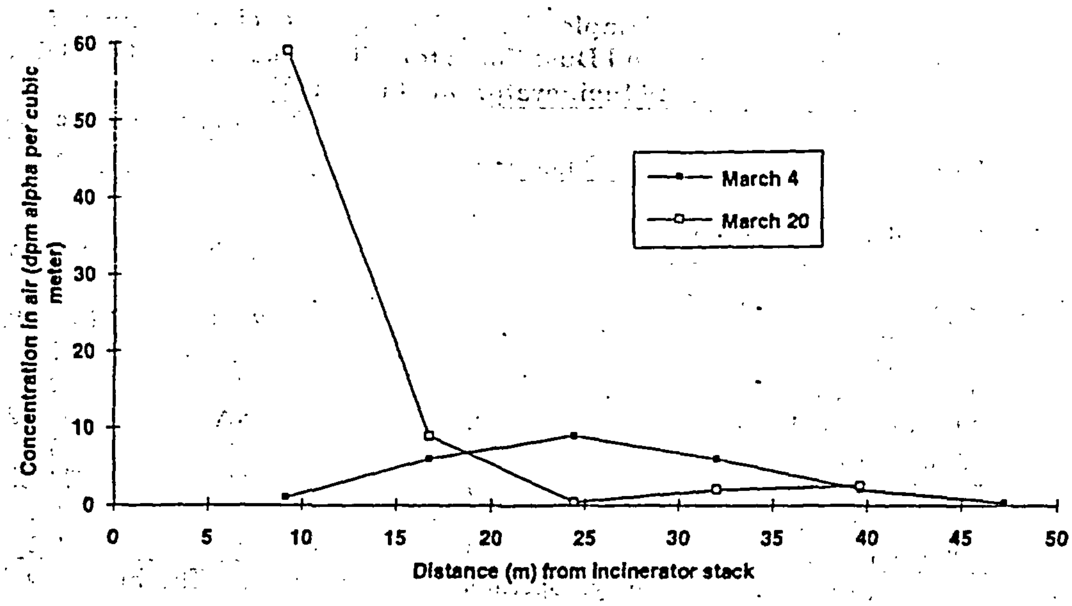

Figure K–6. Measurements of gross alpha in air with distance downwind from the incinerator on March 4 and March 20, 1957. Values plotted for March 20 are the averages of two 10-min replicate samples at each distance.

On May 4, 1957, 110 drums of contaminated clothing and 15 drums of dust collector filter bags were burned in the incinerator. The IH&R monthly report indicated that the air dust and fallout samples were higher than normal. We located the original analytical data sheets for samples taken during this test. Table K–9 shows the results of air dust samples. The results could indicate that greater deposition occurred at farther distances from the incinerator than the March 1957 tests would indicate. However, the uranium content of the waste being burned was very inhomogeneous, and the samples shown in Table K–9 were taken sequentially throughout the day. The close-in sampling began at 9:30 AM and the farthest sampling was finished around 2:00 PM. The general air samples on the incinerator platform show great variability, depending on whether or not filters bags were being dumped and fired. Therefore, we suspect that the effluent from the stack was similarly variable. The magnitude of the contamination measured in air does indicate that this burn was likely to be one of the more important test burns which have lead to local contamination from the old solid waste incinerator.

Table K-9. Air Dust Samples Taken During a Special Burning of Contaminated Clothes and Dust Collector Filter Bags in the Old Solid Waste Incinerator on May 4, 1957

Location	Number of Samples	dpm alpha per cubic meter average	dpm alpha per cubic meter range
General area sample on incinerator platform[a]	5	685[b]	3 – 2692
Breathing zone sample on incinerator platform[a]	7	2201	13 – 7426
11 m downwind	2	4	2–6
28 m downwind	2	3	2–4
33 m downwind	2	NA	nd[b]–2
55 m downwind	4	6	5–8
88 m downwind	4	38	26–50
110 m downwind	4	26	3–43
132 m downwind	4	10	4–19

[a] Operation was very dusty. Operators wore respirators. The highest values were measured during dumping and firing filter bags.
[b] "nd" = Not detectable

The important conclusions which can be drawn from the air and gumpaper sampling data from the 1950s are that operations at the OSWI were dusty, with airborne contamination on the platform requiring respiratory protection, at least during burning of some types of wastes. The contamination levels appear to drop off rapidly within the first few hundred meters of the source. Because of the short sampling times and the fact that gross alpha activity rather than uranium measurements were made, these data were used for qualitative purposes only.

One other set of environmental measurements from the 1960s was examined to see if it would shed light on reconstruction of airborne source terms from the OSWI. Klein (1963, 1964) briefly described the results of a 19-month study of fallout around the OSWI and the oil burner (discussed in the next section of this appendix). During the study, special gumpaper stands were placed downwind (adjacent and NE) from the two incinerators. Uranium deposition at these stations was compared to that measured on gumpaper at other nearby permanent stations in order to determine the extent of fallout due to the incinerators. We determined the locations of the special gumpaper fallout stations from an undated map which indicated their positions. The special station near the OSWI is about 40 m to the NNE. These data are compared to those collected at the permanent station E–2, which is about 200 m to the NNE of the OSWI and directly east of the center of the production area. Klein (1964) concluded that fallout in the area of the OSWI was 2.7 times greater than that at the nearby permanent station.

We located the original analytical data sheets for these measurements to examine the results more closely. First, the original data were corrected for the collection efficiency of gumpaper for particulates, which is 15% for a weekly exposure period and 14% for the monthly exposure period (Shleien et al. 1993). The corrected cumulative deposition to gumpaper for the 19-month period February 1963 through September 1964 was 19.1 g m^{-2} at the OSWI gumpaper station and 6.0 g m^{-2} at the E-2 station. Thus, the net deposition, due to the OSWI airborne source term, would be 690 mg m^{-2} mo^{-1}, or 23 mg m^{-2} d^{-1}.

Evaluation of Airborne Source Term From Ash Handling at the OSWI

As early as 1958, Ross writes: "A ground survey of the area where ashes are shoveled out of the furnace and into drums shows some surprisingly high results. These are caused by spillage from the shovels. It is probable that a wind break or some type of enclosure around this area would lessen the ground contamination by preventing ashes from being blown about while they are being drummed." As late as 1972, a request was made to black top the area around the incinerator, because this crushed stone area had become contaminated due to spills (Farr 1972).

Current soil sampling confirms that the OSWI was a significant source of local environmental contamination. The soil sampling results from the Remedial Investigation and Feasibility Study (RI/FS) indicate that concentrations of radionuclides in the soils adjacent to the solid waste incinerator are well above background levels. The two highest samples, closest to the incinerator showed 25,670 pCi g^{-1} and 2376 pCi g^{-1} of ^{238}U (Anonymous 1990). For perspective with residue values presented earlier in this appendix, ash containing 8% U is equivalent to about 53,000 pCi g^{-1} total U (27,000 pCi g^{-1} ^{238}U). The soil contamination extends toward the ENE from the OSWI (Figure K-7).

Evaluation of airborne releases from non-routine events at the FMPC, including solid spills, is discussed later in this appendix. There were no emission factors located which would apply to this particular situation (hand-shoveling of ash into drums). The amount of ash handled per year is fairly well known (Table K-4), but the fraction which could have been spilled is unknown. In addition, not all of the spilled material becomes airborne; in fact only about 0.5% becomes airborne, according to EPA emission equations (discussed in the non-routine events section). The parameters used in our evaluation of this source term are listed in Table K-10. Spillage of 5% of the total ash handled in a year is equivalent to about 7000 lbs of ash, or 160 kg U. However, the estimated median amount becoming airborne is 2 kg U per year (5–95% interval of 0.4 to 6 kg). This estimated airborne release due to ash handling is <3% of the uranium estimated to be released via the stack during the 1960s.

Table K-10. Assumptions Used in Uncertainty Analysis of Source Term from Resuspension of Spilled Incinerator Ash in the 1960s

Parameter	Minimum	Most Probable[a]	Maximum	Basis
Ash residues handled y^{-1} (metric tons)[b]	62.5	69.5	76.4	Stevenson (1968)
Percent of residue spilled	1%	5%	10%	Assumption
Percent of spilled residue becoming airborne	0.03%	0.5%	3%	See discussion under "Non-routine Releases" section.
% U in residue	.1%	5.1%	12.2%	See Table K-4.

[a] Triangular distribution assumed for uncertainty analysis.
[b] Minimum and maximum values set at 10% of Stevenson's estimate.

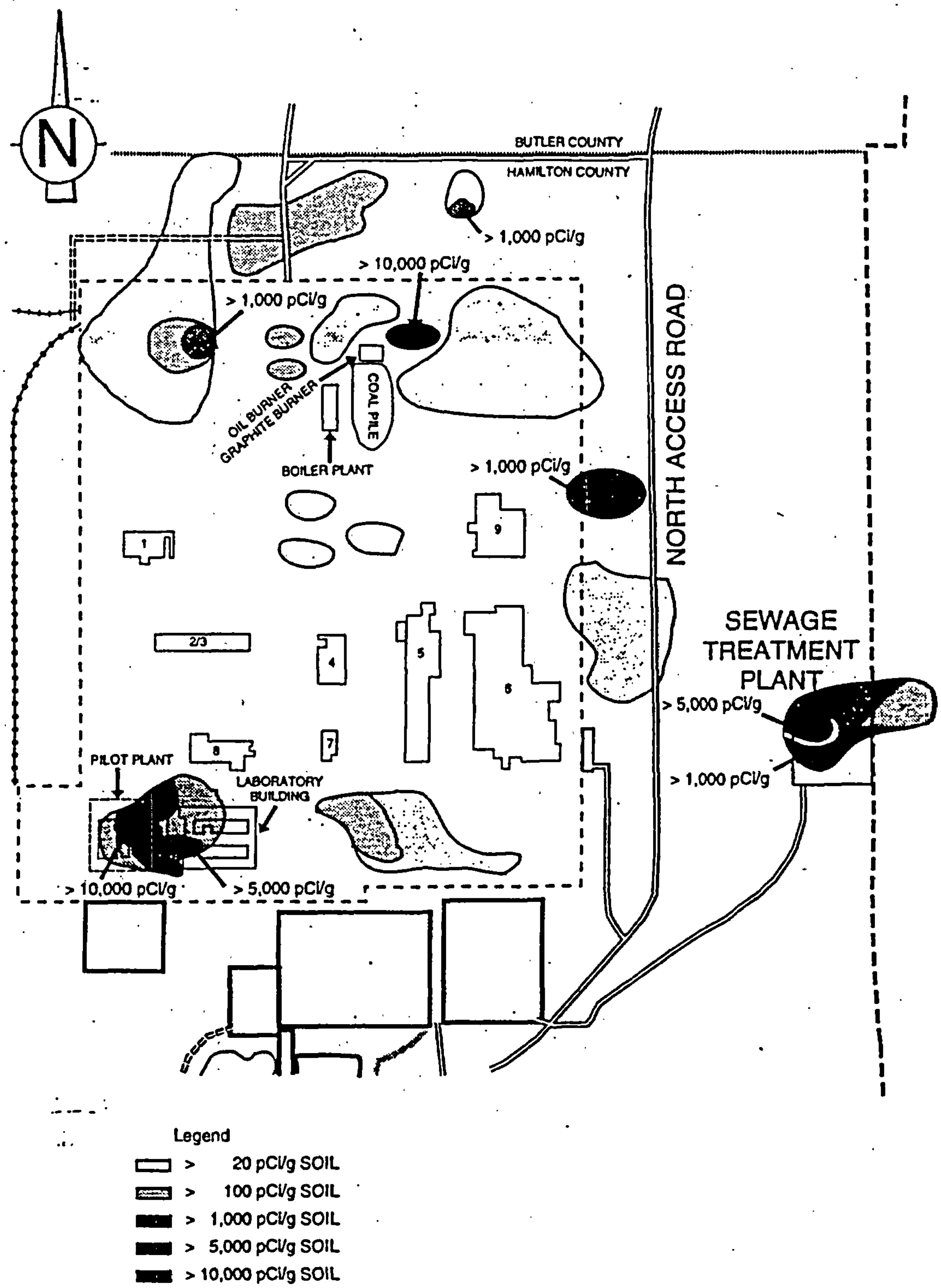

Figure K–7. Regions of higher radioactivity of uranium in the soil at the FMPC, based on plots from RI/FS measurements (Frazier 1990). The regions at the far right of the figure are associated with the old solid waste incinerator.

Summary of Revised Source Term Estimates for the Old Solid Waste Incinerator

Table K-11 summarizes our revised source term estimates for the OSWI and the basis for those estimates. A number of original analytical data sheets, letters, memos, and engineering drawings were studied in developing these estimates, but the primary bases are highlighted here. The estimate for the 1970–1979 period was based on 1977 stack testing and a reduced operating schedule as compared to the previous time periods. The estimate for the 1960s was based on 1964 stack testing as well as a mass balance calculation. The source term estimate for the 1950s was increased over the 1960s by 40 kg per year, based on our analysis of the special burn tests in 1956 (Table K-5). Also, the waste generation rate and the general contamination level of the waste were likely higher in the 1950s than the 1960s, which supports a higher source term for the 1950s, even if special burning did not occur every year. The total estimated release of uranium from the OSWI from 1954–1979 is 2200 kg, with a 5th–95th percentile range of 1600–2900 kg. Over 75% of the total release occurred before 1970. Although handling of incinerator ash resulted in localized contamination, our estimate of airborne release due to these activities is only 2–3 kg per year throughout the 1950s and 1960s.

Table K-11. Revised Source Term Estimates for the Old Solid Waste Incinerator

	g U released per hour of operation	kg U per year released via stack	Basis
1954	NA	20	Based on 1 1/2 months at the 1955 release rate.
1955–1959	NA	140[a] (94–190)[b]	1960s mass balance evaluation + 40 kg y^{-1} additional release for more highly contaminated waste and greater waste generation rate.
1960–1969	31 (3–120)[b]	67[a] (6–250)[b]	1964 stack tests.
	---	100[a] (54–150)[b]	Mass balance calculations.[d]
1970–1979	27–77[c]	52[a] (20–78)[b]	1977 stack tests.

[a]Estimated median. Distribution shape through 1969 is roughly normal.
[b]5th–95th percentile range
[c]Measured range of values
[d]Used in computation of total releases from OSWI.

OIL BURNER (1962–1979)

Two other incinerators, the oil burner and the graphite burner, were located within the production area north of the boiler plant (Figure K–8). Waste oil generated at FMPC was primarily mineral oils and coolant or cutting oils. Some extraction solvents were also mixed in with these liquids. The mineral oils came from garage operations and the changeover of oils in equipment such as gear boxes and hydraulic systems. Most of these oils were described as only slightly contaminated (Boback 1972). Oil was also used to cover the chips packaged in steel drums during shipment from Mallinckrodt Chemical Works (MCW) to FMPC (Mead 1972). At FMPC the oil was drained from the chips and then decanted to remove sludge. The decanted oil was shipped to MCW for reuse, although some excess oil was processed at the FMPC oil burner facility.

In the early years of FMPC operations, waste oil was burned outdoors in the open, in shallow pans which each burned 100–150 gallons per day. Also, oil was dumped over burnable solid trash for combustion in an on-site burn pit (see discussion above). Both techniques were halted because of the heavy smoke produced (Boback 1972). There also were several attempts to adapt the Scrap Recovery Plant to burn contaminated oils with conventional burners or existing equipment (Brandner et al. 1963); however, there were process difficulties and high air dust levels.

Brandner et al. (1963) describe the incineration of waste contaminated oil at the FMPC. They divided the over 100 classes of waste liquid organic material stored at the FMPC into four principal types:

1. **Mineral Oils.** Typically containing 2–4 g L^{-1} uranium, and accompanied by a water phase which is usually lower in uranium content. Sludges which had settled to the bottom of some of these drums ranged from 5–10% uranium.
2. **Emulsions.** Uranium levels of 5–10 g L^{-1} were typical for emulsions and for any accompanying water phase below the emulsion.
3. **Waste Extraction Solvent.** Uranium levels of 2–4 g L^{-1} in the TBP–kerosene solvents were typical.
4. **Wastes Rich in Chlorinated Hydrocarbon Solvents.** Not amenable to incineration since they would require careful blending and decomposition products would be highly toxic.

Only the first two types of liquid organic waste material were being incinerated in 1963 (Brandner et al. 1963).

History and Operation of the Oil Burner

This section gives an overview of the history and operation of the oil burner. In addition, a more detailed chronological history of notes and information, gathered mainly from historic reports of the Industrial Hygiene and Radiation (IH&R) Department, is included as Annex 2 to this appendix.

The oil burner was constructed in early 1962. Boback et al. (1987) give the start-up date as March 31, 1962. Brandner et al. (1963) indicate that other refinements and modifications were made that year, including procedures and equipment for preparing feed and regulating burning conditions. Eventually, the oil burner system consisted of a series of five treatment tanks, an oil pre-heat tank, and a five-foot-square refractory brick enclosure with a 3 x 3 ft square stack which housed the stainless steel burner pot (Boback 1972).

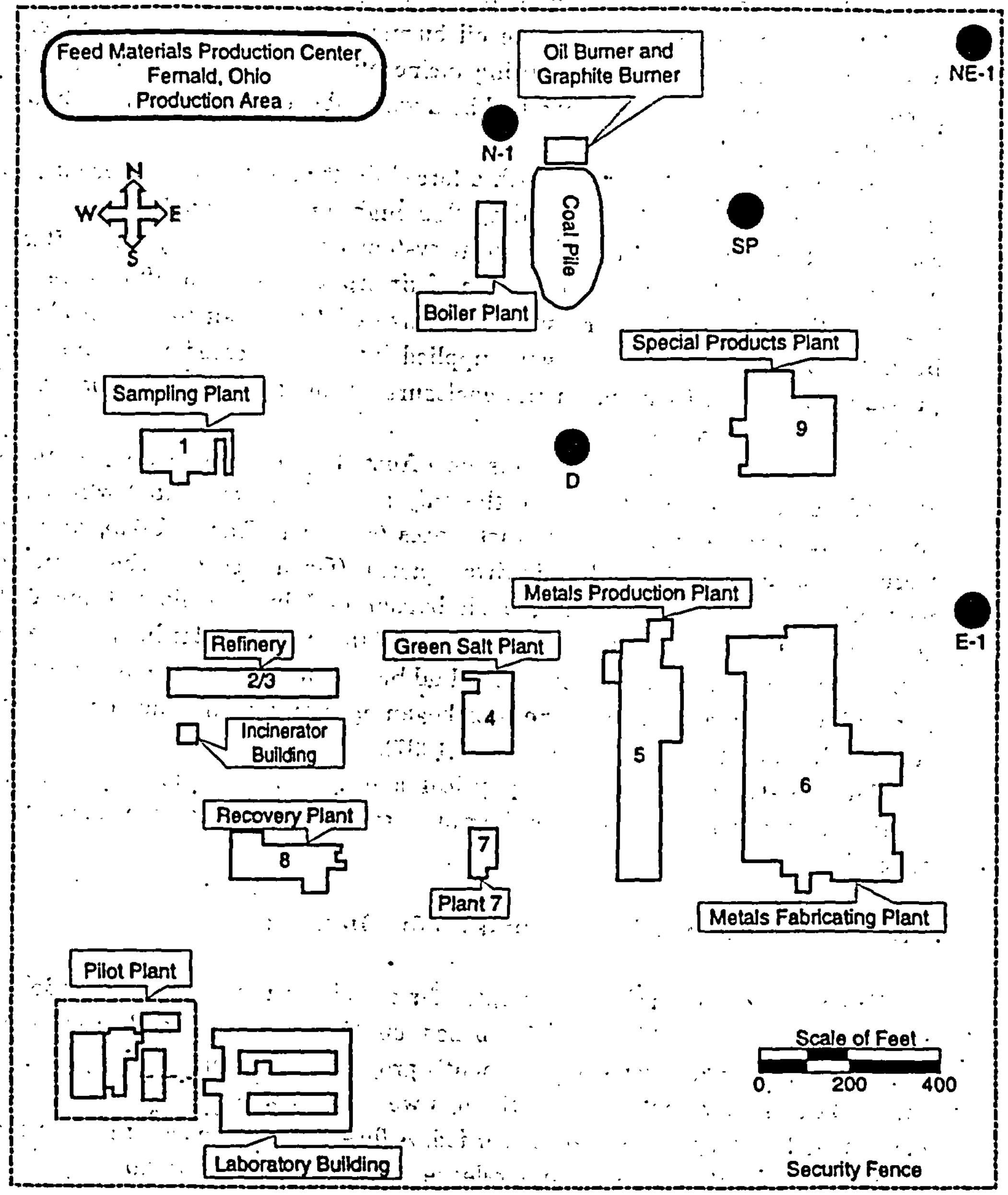

Figure K–8. Map of FMPC production area, showing location of the oil burner (operated 1962–1979), the graphite burner (operated 1965–1984), and the new incinerator building, which housed the new solid and liquid waste incinerators in the 1980s. The circles represent the locations of selected gumpaper fallout stations used to measure deposition of uranium (discussed in text). The location of the "SP" station is very approximate.

The four main operational steps for the oil burning operation, (1) cold or warm feed preparation, (2) oil heating, (3) pot feeding, and (4) oil burning are described in some detail in Brandner et al. (1963). In addition, the standard operating procedure for the burner was located and reviewed (Baer 1966a). The water fraction was routed to an open evaporator where the water was boiled away by a high temperature steam coil. The oily sludge generated during preparation of the oil burner feed was drummed and sent to the solid waste incinerator, where the resulting incinerator ash was routed to the Recovery Plant. Feed rate to the burner was controlled by a valve above the feed pan to about 20–30 gal h^{-1} (Brandner et al. 1963).

The oil burner was operated with a forced draft of excess air, in order to limit smoking. Brandner et al. (1963) indicate that this high pressure draft system was supplied by compressed air and a low pressure draft system was supplied by a fan. Both systems were adjustable both in location and direction of air input to keep the off-gas clean. Boback (1972) indicates that the 1/2" high pressure line delivered 100 psi air to a point about 12" above the burner pot. Low pressure air was supplied by a fan located near the burner enclosure, providing air at six locations in the enclosure, through ducts which had blast gates for air control at each entry point.

Stack testing for particulate emissions from the oil burner were conducted on May 13, 1976 (Ross 1976). These data were the only measurements located which provide data on the velocity and temperature of stack gases (shown in Table K–12), although the values were confirmed by later handwritten notes (Grant 1986). The resulting calculated particulate emission rate from the oil burner (1.8 lb h^{-1}) did not meet the Ohio EPA standard of 0.2 lb per 100 lb of material burned in an incinerator having a capacity less than 100 lb h^{-1} (Ross 1976). However, the old oil burner was not shut down until June 15, 1979. A new liquid organic waste incinerator began operating in the incinerator building (Figure K–8) in the spring of 1983 (Boback et al. 1987).

Table K–12 summarizes the physical and operating parameters for the oil burner. Particle size considerations for effluents from all incinerators are discussed later in this appendix.

Previous Uranium Release Estimates for Oil Burner

Previous uranium release estimates for the oil burner presented by Boback et al. (1987) were supplied by Neblett (1985), based on "knowledge gained from supervising these operations and from waste management's project assignments in the past". Neblett believed that data to substantiate these estimates were not available, and that the records for these operations had long since been discarded. A flow diagram showing his estimate is shown as Figure K–9. In 1962, the annual release was estimated to be 20 kg y^{-1}, rather than 27, because the burner started operation after the first three months of the year. The total estimated release over the entire operating history would be ≈470 kg U.

Table K–12. Summary of Physical and Operating Parameters for the Oil Burner (Operated 3/31/62 to 6/15/79)

Parameter	Value	Reference	Notes
Physical stack height	15 ft	Engineering drawing 10X-M-00322	
Building dimensions	5 x 5 x 5 ft	Engineering drawing 10X-M-00322	Cubic shaped outer structure tapers to 3-foot x 3-foot stack with total height of 15 feet above ground.
Stack inner dimensions[a]	20 x 20 inches (2.78 ft^2)	Engineering drawing 10X-M-00322	Outer dimensions of stack were 3 x 3 ft. Stack composed of 8" thick firebrick.
Exhaust gas velocity (ft min^{-1})[a]	350–400	Ross 1976 Grant 1986	Measured with rotating vane anemometer during particulate emission test.
Exhaust gas temperature	120–200 °F	Ross 1976 Grant 1986	Estimated temperature of stack gases at particulate sampling point and at metering orifice.

[a] The cross sectional area at the point where velocity was measured was about 12 ft^2, for a volumetric flow rate of 350 fpm × 12 ft^2 = 4200 actual cfm (3373 standard cfm) according to Ross (1976). The velocity in the upper part of the stack would have been 4200 cfm ÷ 2.78 ft^2 = 1500 fpm (disregarding cooling of gases) or 1200 scfm.

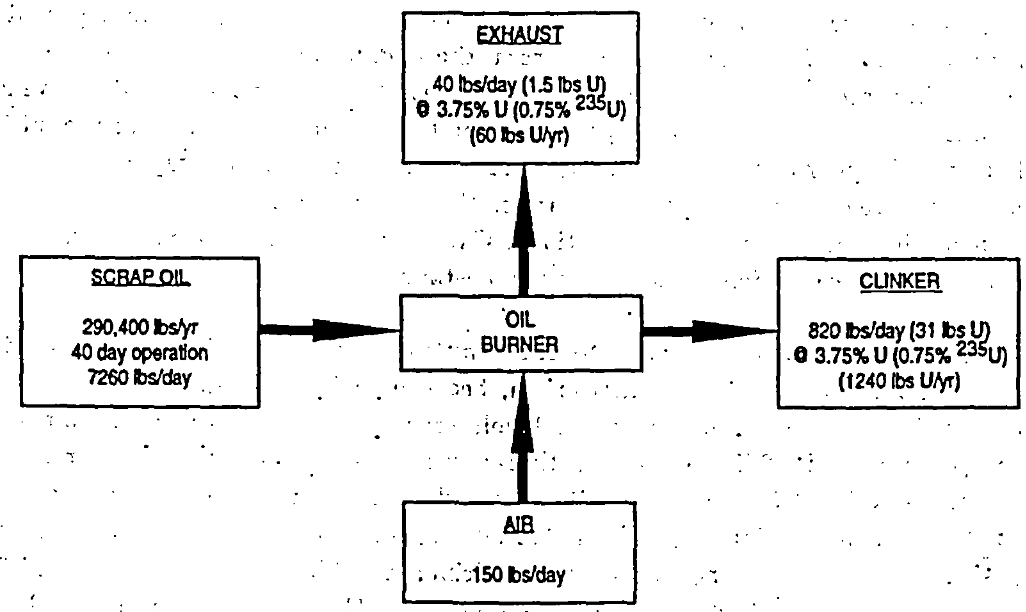

Figure K–9. Basis for previous estimates of releases of uranium from oil burner (from Neblett 1985).

Neblett's exhaust emission rate of 40 lbs of particulate material per day (Figure K–9) is reasonable based on measurements during the emissions test (Ross 1976), which resulted in an estimate of 1.8 lb h^{-1} (43 lb per 24-h day). An evaluation of the other parameters in his estimate requires a more in-depth look at historic records. Our review of historic documentation and revised source term estimates are presented in the following sections.

Evaluation of Historic Documentation to Reconstruct Uranium Source Terms from the Oil Burner

A number of original sources of documentation were located and carefully reviewed to permit a reconstruction of the source terms for the oil burner. During this process, a chronological history of important events and other notes was compiled, which is included as Annex 2 to this appendix. This annex provides a detailed record of relevant changes and correspondence which was used to aid interpretion of the original data. It also includes information on burning of oils before the final oil burner was constructed in 1962. The types of quantitative information which were evaluated to estimate the source term from the Oil Burner fall into the following categories:

- **Category 1.** Processing rate information (operating schedule).
- **Category 2.** Materials balance information.
- **Category 3.** Measurements of uranium and gross alpha activity in stack gases.
- **Category 4.** Environmental measurements of uranium and gross alpha activity in air and deposited on gumpaper around the incinerator.

There were very few category 3 stack measurements which would permit a credible reconstruction of the source term from those data alone. Therefore, we searched for processing rate and materials balance information (categories 1 and 2) to derive source term estimates. In doing so, it was apparent that the operating schedule of the oil burner was heavy in the year or two immediately after construction, and then tapered off. Thus, while it may be a reasonable long-term average, the previous constant source term estimate by Neblett did not reflect these variations. Category 3 and 4 information was collected and reviewed for verification of the source term estimates.

Category 1. Processing rate information. The processing rate was important to examine for source term reconstruction, because in later years the oil burner was only operated whenever the oil inventory levels reached sufficient size to permit a production campaign (Mead 1972). However, there was a large backlog of contaminated liquids accumulated by 1962. Coates (1962) estimated a total inventory of contaminated burnables at 2812 drums (112,480 gal) of burnable oil (includes 237 drums of emulsions), 3599 drums (143,960 gal) of other organic solvents (trichlor, perchlor, TBP-kerosene), and 700 drums (21,000 gal) of "enriched" organics in August 1962. (The non-metric unit of "gallons" is used throughout this section because that is the unit typically used in the original sources.) Coates noted that operation of the oil burner would begin on a 3-shift-per-day basis as soon as lights and other required changes could be installed. The oil burner was started as a production unit operating 24 hours per day, five days per week in January, 1963 (Brandner et al. 1963). Boback (1972) indicated that the oil burner, which consumed about 500 gallons per day on a 3-shift schedule, eliminated the backlog of contaminated liquids by August 1964.

We located handwritten ledgers (Anonymous 1964–1968) titled "Oil Burner and Incinerator Operations" for January 1964 through December 1968, as well as a number of monthly memoranda titled "Resume of Oil Burner and Incinerator Operations During the Month of [Month, Year]," which confirmed the data on the ledgers. In addition, the total waste processing rate for 1969 was given as 32,500 gallons in Anonymous (1970). Figure K–10 illustrates the waste processing and oil burning trends at the FMPC. Because the earliest ledgers were for 1964, values for 1962 and 1963 were estimated based on the following premises:

1. Processing at one shift per day in 1962 and three shifts per day in 1963 (Coates 1962, Brandner et al. 1963);
2. Consumption of the entire backlog of liquids given by Coates (1962) between September 1962 and August 1964 (as indicated by Boback 1972);
3. Continuous receipt of waste at a baseline rate of 7200 gallons per month (based on 1965 monthly average waste processing rate, after backlog was consumed).

The estimated peak processing rate of 271,000 gallons of waste in 1963 is well within the capabilities of the oil burner system, which was stated by Brandner et al. (1963) to be capable of consuming 400–700 55-gal drums of waste per month, which would translate to 264,000–462,000 gallons per year, under continuous processing conditions.

On average, 0.5 gallon of oil was burned per gallon of waste processed (determined from data in Anonymous 1964–68). The majority of the remainder of the waste volume was void space in the drums, water, or sludge, which was processed separately.

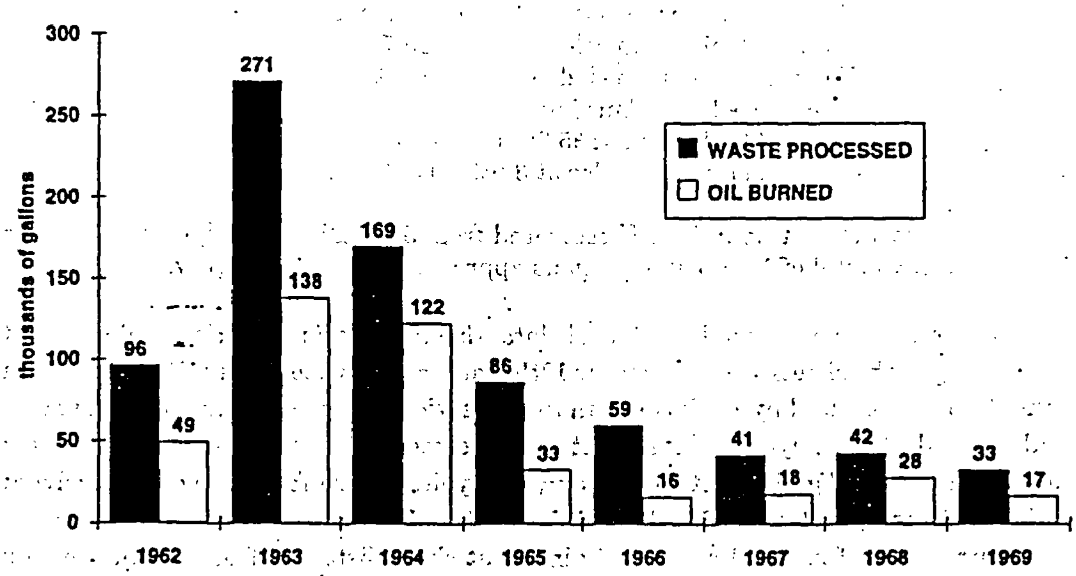

Figure K–10. Amounts of liquid waste processed and oil burned at the FMPC oil burner in the 1960s.

Neblett's estimate of 290,400 pounds of oil burned per year would correspond to about 39,100 gallons of oil burned per year, assuming a specific gravity of 0.89 (ranged from 0.88–0.90 on analytical data sheets). This value is a factor of three less than that actually burned in 1964 (Anonymous 1964-1968), but agrees very well with the six-year average for 1964-1969 (38,900 gallons oil burned per year). In 1975, Stevenson estimated the waste processing rate at the oil burner at 7500 gallons per year of waste lubricating oils and 1200 gallons per year of spent TBP-kerosene solvents. This total of 8700 gallons per year shows a further decrease in the processing rate to 22% of the six-year average for 1964–1969. Perkins (1976) indicated a current operating schedule of 4–6 weeks per year with a throughput of ≈45 kg per hour, which translates to 9000–13,000 gallons per year.

Category 2. Materials balance information. The most relevant original data located from FMPC records was from a materials balance test in October 1961. This test was performed in a prototype oil burner. Approximately 570 gallons of various types of oil were burned, resulting in recovery of about 16.4 pounds of uranium from the waste (DeFazio 1962). A rough draft of DeFazio's letter (dated 2/23/62) gives more information on the results of the mass balance tests than the final letter:

> "On October 9, 1961, a materials balance test was started which lasted for six days. The results are as follows:
>
> | | |
> |---|---|
> | Oil burned | 569 gallons – 4270 pounds |
> | Average U concentration | 3.7 g U/l |
> | Specific gravity of oil | 0.90 |
> | Total U in 569 gallons | 17.6 pounds |
> | Total ash after burning | 45 pounds |
> | Weight of dry ash | 44 pounds |
> | Heavy sludge from bottom of tank | 17.5 pounds |
> | U concentrations in ash | 35.7% |
> | U concentrations in sludge | 3.9% |
> | U recovered from burning | |
>
> (44 lbs. ash at 35.7% U) = 15.7 lbs.
> (17.5 lbs. sludge at 3.9% U) = .7 lbs.
>
> A total of 16.4 pounds of U recovered from 569 gallons of oil, or 16.4 pounds from a total of 17.6 pounds, equals approximately 90% recovery."

We located the original analytical data sheets for this materials balance test, which according to the sheets, began October 13th, rather than October 9th. We were able to verify the data summarized by DeFazio, with the exception of the concentration of uranium in the oil burned. No analytical data sheet was located for this measurement; however, this concentration (3.7 g U per liter) is within the typical range for waste oil and solvents at the FMPC (Brandner et al. 1963). In addition to the data summarized above, there were also measurements of gross alpha activity in air at short distances from the burner during the test. However, these data were were not analyzed further, because they are not as relevant as similar measurements taken in and around the final oil burner, which was constructed the following year.

The 1961 materials balance test provides an estimate of 90% recovery of uranium in the ash and sludge. The release to air, presuming any other losses are negligible, would be 10% of the uranium in the original feed material. Because there was no replication of the test or the measurements, no uncertainty can be assigned to that release fraction estimate. However, other estimates of the release fraction from burning U-contaminated waste, including oil, were located in the scientific literature. Bostick et al. (1991) measured a release fraction of 2.9% to off-gas (prior to air-cleaning equipment) for uranium in contaminated oil fed to the Oak Ridge mixed waste incinerator. AEC contractors operating incinerators for uranium-contaminated materials in the 1960s reported release fractions of 0–5% (Glauberman and Loysen 1964).

A source term estimate can be developed from the materials balance information, using the uranium content of oils and solvents burned, annual processing rates, and an estimate of the airborne release fraction. For our uncertainty analysis, we used a uniform distribution for the airborne release fraction, ranging from 1 to 10%. The uranium concentration of the liquid wastes is shown in Table K–13, along with estimates of the fraction of the FMPC liquid wastes which fall into those categories. For our source term reconstructions, it was reasonable to assume 97% of the liquid wastes processed were mineral oils and extraction solvents, which both contain uranium at an estimated concentration of 2–4 g U L^{-1}. The other 3% of waste was assumed to contain concentrations ranging from 5–10 g U L^{-1} which are more typical of the emulsions. The distribution shape of uranium concentrations in the waste was assumed to be uniform within the values given.

Table K–13. Fraction of Liquid Waste of Different Types and Uranium Concentrations

Liquid Waste Type	U concentration (g U L^{-1})[a]	Fraction of waste inventory backlog in 1962[b]	Fraction of annual volume processed in 1970s[c]
Mineral Oils	2–4	0.36	0.86
Extraction Solvents (e.g. TBP-kerosene)	2–4	0.51	0.14
Emulsions	5–10	0.03	
Other	unknown	0.10	

[a]Brandner et al. (1963).
[b]Coates (1962).
[c]Stevenson (1975).

The airborne source term estimates for the oil burner, derived using this materials balance/processing rate approach, are tabulated in Table K–14 for 1962 through 1979. We did not have the detailed processing record to estimate the source term each year separately for the 1970s. The distribution of each annual source term estimate is approximately normal. Neblett's previous estimate of 60 lbs (27 kg) uranium per year from the oil burner is an underestimate, according to our method, for the years 1962 through 1964. However, our median estimate for the entire operating period (370 kg) is in good agreement with the previous estimate (470 kg).

Table K-14. Revised Source Term Estimates for the Oil Burner (kg U y^{-1}), Derived Using a Materials Balance/Processing Rate Approach

Year	Median	5th percentile	95th percentile
1962	37	18	63
1963	104	50	177
1964	91	44	156
1965	24	12	42
1966	12	6	20
1967	13	7	23
1968	20	10	36
1969	12	6	21
1970–1978	6	3	11
1979	3	2	6
TOTAL (1962–1979)	**370**	**270**	**470**

Category 3 Information. Stack measurements of uranium and gross alpha in stack gases from the oil burner. There were no historic memoranda or reports located which indicated that any direct measurements were made of uranium releases from the oil burner stack. However, a few analytical data sheets were located with this type of information. The analytical data sheet for the 1976 particulate testing (Ross 1976) provided a uranium analysis result as well as the particulate analysis result which had been presented in the memo. This stack air sample was taken on May 13, 1976 on a pleated filter for six hours. The concentration of uranium in this off-gas sample was 45.4 µg m^{-3}. Using the measured volumetric flow rate of stack gases (Table K–12), the emission rate would be 0.3 g U per hour. The paraffin used in the particulate emissions test was apparently an uncontaminated material, as this uranium emission rate would correspond to a release of only 2 kg U per year under continuous processing conditions.

Another analytical data sheet from November 4–5, 1976 contained measurements of particulates, uranium, and water content for air over the oil burner stack, while burning under air pressure and while using steam. The concentration of uranium was 4.3 µg m^{-3} while burning under air pressure and 164 µg m^{-3} while using steam. Again, these concentrations correspond to release rates which are quite low compared to those developed using the materials balance/processing rate approach described above.

Category 4 Information. Environmental measurements of uranium and gross alpha activity in air and deposited on gumpaper around the incinerator. There are two types of environmental measurements which could be used to verify our source term estimates for the oil burner:

- short-term measurements of radioactivity in air a short distance from the stack
- monthly deposition measurements of uranium onto gumpaper near the oil burner

Brandner et al. (1963) and Anonymous (1970) refer to measurements of uranium concentrations in the plume at 5 to 10 feet downwind from the top of the stack (Table K–15).

When burning was properly controlled to prevent smoking, the uranium concentration in air averaged approximately 0.05 mg m^{-3}.

Table K–15. Average Airborne Uranium Concentrations Measured 5–10 feet from Top of Oil Burner Stack under a Variety of Burning Conditions (Brandner et al. 1963)

Burning Condition	mg m^{-3}
Properly controlled burning	0.05
Light smoke being emitted from stack	0.2
Entrained ash visible above pot	0.4
Flame coming out of stack	0.8
Heavy smoke being emitted from stack	1.9

Because of the potential importance of these measurements to verification of source terms from the oil burner, we located the original analytical data sheets for these measurements. In reality, the measurements made near the oil burner were of gross alpha activity, not uranium. We were able to locate data for a total of 54 measurements on nine separate days (Table K–16).

From other FMPC memoranda, we determined that a ratio of 1.5 dpm alpha per microgram U was used by the FMPC in the 1960s to convert gross alpha measurements in air to uranium in air, which was probably what Brandner et al. (1963) had used. In practice, this ratio is subject to some measurement uncertainty. In our conversion of the gross alpha measurements to estimates of uranium concentration, we used an uncertainty distribution for the alpha-to-uranium ratio which was determined from 149 ambient air measurements or uranium and gross alpha during 1957–1959 at the FMPC (see Figure B2–2 of Shleien et al. 1993). The median ratio obtained from this dataset was indeed 1.5, and the 25th–75%tiles were 1.17 to 2.29. The distribution of this ratio as well as the measured distribution of gross alpha in air were used to develop an estimate of the uranium distribution in air close to the oil burner stack (Figure K–11).

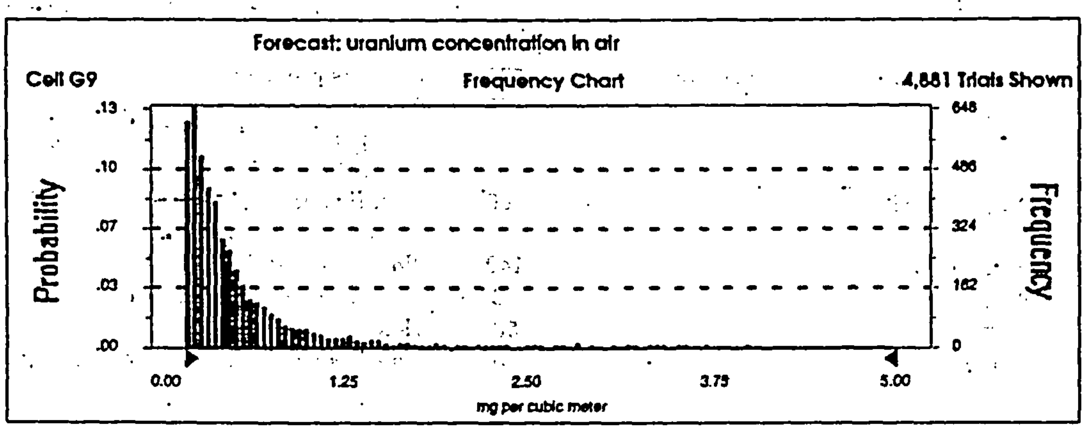

Figure K–11. Distribution of estimated uranium concentrations in air close to the oil burner stack during operation. Data obtained from original analytical sheets from 1962 and 1963.

Table K-16. Measurements of Gross Alpha in Air Near Oil Burner Release Point in 1962–1963

Date	Distance from Burner (ft)	dpm alpha m^{-3}	Notes from original data sheet.
June 4, 1962	7	159	Puffing black smoke occasionally.
June 4, 1962	8	54	Burning rate 13.32 gal in 23 min.
June 4, 1962	6–7	61	
June 4, 1962	6–7	1043	Sample considered most representative of those taken so far.
June 8, 1962	5	506	Burning rate 13.3 gal in 22 min.
	5	345	Little heavier smoke than previous sample.
June 8, 1962	5	514	Light smoke.
June 8, 1962	5	555	Same as above.
June 24, 1962	4	6	Burning very good. No smoke. Burning rate 13.3 gal in 28 min.
June 24, 1962	2	260	No smoke. Burning rate 13.3 gal in 27 min.
June 24, 1962	3	88	Same as previous.
June 24, 1962	2	980	Burning rate 13.3 gal in 18 min. Filter burned slightly. Low pressure air on.
June 24, 1962	3	82	Burning rate 13.3 gal in 25 min. No low pressure air on.
June 24, 1962	2	570	Burning rate 13.3 gal in 18 min. Filter burned.
June 24, 1962	2	95	Burning rate 13.3 gal in 18 min. Smoking slightly.
July 18, 1962	Not given.[a]	320	No baffle over burner pot (applies to all samples this day). Light smoke.
July 18, 1962	Not given.[a]	200	Black smoke.
July 18, 1962	Not given.[a]	460	Heavy smoke.
July 18, 1962	Not given.[a]	220	Smoke clearing up.
July 18, 1962	Not given.[a]	65	No smoke visible.
July 18, 1962	Not given.[a]	790	Smoking a little.
July 18, 1962	Not given.[a]	360	Burner had been operating for over 30 min with no visible smoke. Rate 13.3 gal in 22 min.
July 18, 1962	Not given.[a]	670	Same as previous.
August 9, 1962	4	160	New burning pot and cross vane installed (applies to all samples this day). Light smoke.
August 9, 1962	4[b]	560	Slightly heavier smoke. Filter started to burn.
August 9, 1962	4[b]	140	No smoke visible.
August 9, 1962	4[b]	68	Light smoke. Wind shifting.
August 9, 1962	4[b]	53	Heavy smoke.
August 9, 1962	4[b]	35	No smoke visible. Burning rate 38 gal per hour.
August 9, 1962	4[b]	61	Same as previous, light smoke.
Sept. 6, 1962	3	210	Burning heavy emulsion with large door partially open (applies to all samples this day).

(continued next page)

Table K–16. Measurements of Gross Alpha in Air Near Oil Burner Release Point in 1962–1963 (cont.)

Date	Distance from Burner (ft)	dpm alpha m^{-3}	Notes from original data sheet.
Sept. 6, 1962	3	130	No smoke.
Sept. 6, 1962	3	1400	No smoke.
Sept. 6, 1962	3	1900	Heavy smoke.
October 4, 1962	5	310	No smoke.
October 4, 1962	5	240	Occassional smoke.
October 4, 1962	5	2	No smoke. Flame lowered by increasing air at ring in stack.
October 4, 1962	5	390	Light smoke.
October 4, 1962	5	110	Same as above.
August 21, 1963	5 ft W, 3–4 ft above[c]	34	Wind blowing from east. Normal burning approx. 20–30 gal per hour (applies to all samples this day).
August 21, 1963	5 ft W, 4–5 ft above[c]	21	
August 21, 1963	5 ft W, 3–4 ft above[c]	2	
August 21, 1963	5 ft W, 4–5 ft above[c]	6800	
August 21, 1963	5 ft SW, 3–4 ft above[c]	110	
August 21, 1963	5 ft SW, 4–5 ft above[c]	<0.4	
August 21, 1963	5 ft NW, 3–4 ft above[c]	720	
August 21, 1963	5 ft NW, 4–5 ft above[c]	740	
August 21, 1963	10 ft W, 4–5 ft above[c]	17	
August 21, 1963	10 ft W, 4–5 ft above[c]	2	
August 21, 1963	5 ft downwind	2000	Flames emanating from furnace and light smoke.
August 21, 1963	5 ft downwind	5500	Flames emanating from furnace and light smoke.
August 26, 1963	5 ft W, 4–5 ft above[c]	6300	Normal burning conditions. Rate 20 gal per hour.
August 26, 1963	5 ft NW, 3–4 ft above[c]	1400	As above.
August 26, 1963	5 ft NW, 4–5 ft above[c]	610	As above.

[a] Location of samples not given on analytical data sheet, but IH&R July 1962 monthly reports says samples were taken in off-gas from the oil burner.
[b] Location not given, but assumed to be the same as first sample taken that day.
[c] Above furnace height.

The distribution of uranium concentrations in air close to the burner (Figure K–11) is highly skewed to the lower concentrations. The median concentration is 0.24 mg per cubic meter; the 5th to 95th percentile interval is 0.02 to 2.9, which is in good agreement with the values presented by Brandner et al. (Table K–15). From the data it appears that sometimes the sampling missed the flue gases altogether (concentrations are barely detectable) and other times the concentrations were quite high, indicating that the flue gases were probably captured in that sample.

It might be possible to back-calculate an emission rate (g U h^{-1}) from these air concentrations, but assumptions must be made about the amount of dilution of stack gases between the release point and the measured location (typically 2 to 10 feet downwind). <u>If it is assumed that the dilution is a factor of 10</u>, the median source term based on the air concentration measurements is 17 g U released per hour, or 100 kg per year, assuming continuous processing for 5 d wk^{-1}, 50 wk y^{-1}. This is in reasonable agreement with our source term estimates for the higher processing-rate years (Table K–14).

Fallout measurements. One other set of environmental measurements from the 1960s were examined to see if they would shed light on reconstruction of airborne source terms from the oil burner. Klein (1963, 1964) briefly described the results of a 19-month study of fallout around the OSWI and the oil burner. During the study, special gumpaper stands were placed downwind (adjacent and NE) from the two incinerators to measure local fallout. Uranium deposition at these stations was compared to that measured on gumpaper at other nearby permanent stations. We determined the approximate locations of the special gumpaper fallout stations from an undated map which indicated their positions. The special station near the oil burner was about 400 feet to the ESE, according to this map, as shown in Figure K–8. However, Klein's memos indicate that the special gumpaper stand was to the NE. The data from the special gumpaper stations were compared to those collected at the permanent stations N–1, which is less than 200 feet to the NW and station NE–1 which is about 1000 feet to the NE of the oil burner. Klein (1964) concluded that fallout in the area of the oil burner was 3.6 times greater than at N–1 and 6.8 times greater than at NE–1.

We located the original analytical data sheets for these measurements so they could be examined more closely. We also included the permanent station "D" in our comparisons (location shown on Figure K–8). First, the measured depositions in µg U ft^{-2} were corrected for the collection efficiency of gumpaper for particulates, which had been determined to be 15% for a weekly exposure period and 14% for the monthly exposure period (Shleien et al. 1993). They were then normalized to a daily deposition rate (mg m^{-2} d^{-1}). The data are illustrated in Figure K–12. The higher deposition at the oil burner gumpaper station is readily apparent. Station D, which is closer to major production area sources, is next highest, followed by N–1 and NE–1. The cumulative deposition over the 19-month period February 1963 through September 1964 was 45500 mg U m^{-2} for the oil burner, 20700 for station D, 10500 at station N–1, and 6000 at station NE–1. The graphite burner was not yet operating at this location (see next section), and there is no other known important source of uranium in this area of the production plant. The net deposition, due to the oil burner airborne source term, would be 1960 mg m^{-2} mo^{-1}, or 65 mg m^{-2} d^{-1}, if it is assumed that the average of N–1 and NE–1 is a baseline deposition rate due to other sources. As demonstrated in the previous sections, 1963 was a peak processing year for the oil burner.

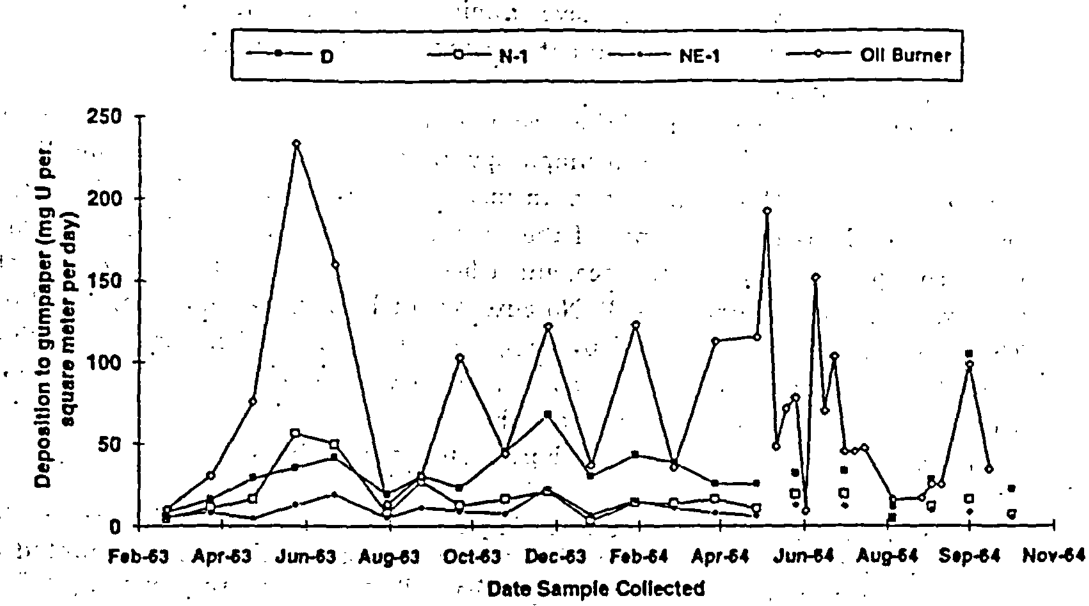

Figure K–12. Deposition to gumpaper (corrected for weathering) at a station near the oil burner compared to stations within the production area but further away (see Figure K–8 for locations). The oil burner was processing at a high rate during this period.

GRAPHITE BURNER (1965–1984)

A substantial quantity of waste graphite was generated at the FMPC. Most of this generation was a result of the uranium metal recast step, and consisted primarily of scrap furnace crucibles and molds (Anonymous 1970). The process application of these graphite materials caused them to be quite contaminated with uranium and daughter products. The uranium content of the crucibles could reach 3% after several uses (Boback 1972); the quantity of uranium contained in this waste stream was sufficient to make recovery of this material economically attractive (Anonymous 1970). In the early years of operation, graphite was burned in a furnace in the Recovery Plant and the residue was leached to recover the uranium. This process was discontinued in 1960 because the carbon content of the product was too high (Mead 1972).

History and Operation of Graphite Burner

In October 1965, an experimental graphite furnace was built, and it was established that graphite could be successfully incinerated with essentially all of the uranium contained in the total feed being recovered in the ash (Anonymous 1970). This substantially reduced the quantity of material that had to be chemically processed for final uranium recovery.

The only pre-treatment given to the graphite waste was to break it into pieces small enough to be fed into the burner. A minimal amount of sorting was done: (1) some types of scrap known, by previous chemical analysis, to contain minimal uranium were discarded directly into the dry residue waste pit, and (2) visible pieces of uranium metal were removed. The removal of this massive uranium from the graphite feed was necessary to "permit the burner to operate without causing a problem from radioactivity in the stack discharge" (Anonymous 1970).

The following description of the graphite burner operation is provided in Anonymous (1970): The graphite burner was a simple apparatus, consisting of a refractory lined cylinder with a steel shell. The cylinder had an inside diameter of 27" and is 60" high. Ports are spaced around the circumference of the cylinder to introduce air. Graphite scrap was fed into a charge port above the furnace, and ashes were raked out of seven ports at the bottom (see example port in Figure K–13). No auxiliary fuel was necessary except to start the fire. This was normally accomplished by using coal or wood and kerosene. After the fire was started and the graphite near the bottom was ignited, the fire was self-sustaining. Fresh scrap was periodically fed into the top of the furnace.

Boback (1972) describes graphite burner operation as follows:

> "The graphite burner is a simple outdoor incinerator. The combustion chamber is a refractory-lined, 1/4"-thick, carbon steel cylinder mounted on four legs and topped with an eight-foot-tall steel stack. An elevated platform permits graphite to be charged into a port near the stack bottom.
>
> For operation, a 3-inch layer of sand is placed in the bottom of the burner shell. A wood fire is started and oil-soaked nugget coal is added until a two-foot-thick bed of red hot coals is obtained. A layer of graphite is placed on the coals and an air lance is inserted through a bottom port. When the graphite becomes orange-red, more graphite is added and the air supply is reduced.
>
> Additional graphite is added only if the previous charge is orange-red. The air lance is removed when the combustion is proceeding at an acceptable rate.
>
> A four-foot-thick bed of burning graphite is maintained. As the charge burns down, ashes are raked out through seven ports into 3-1/2 gallon buckets. Cooled ashes are transferred to 55-gallon drums for storage and later processing to recover the uranium. Ash from this burner is about 60% uranium.
>
> After startup, the burning usually proceeds smoothly, producing only a slightly visible stack discharge. Uranium in the stack effluent has ranged from 0.15 to 8.3 mg/m^3. The average discharge of airborne uranium is estimated at less than 0.5 pounds per 24 hours of operation."

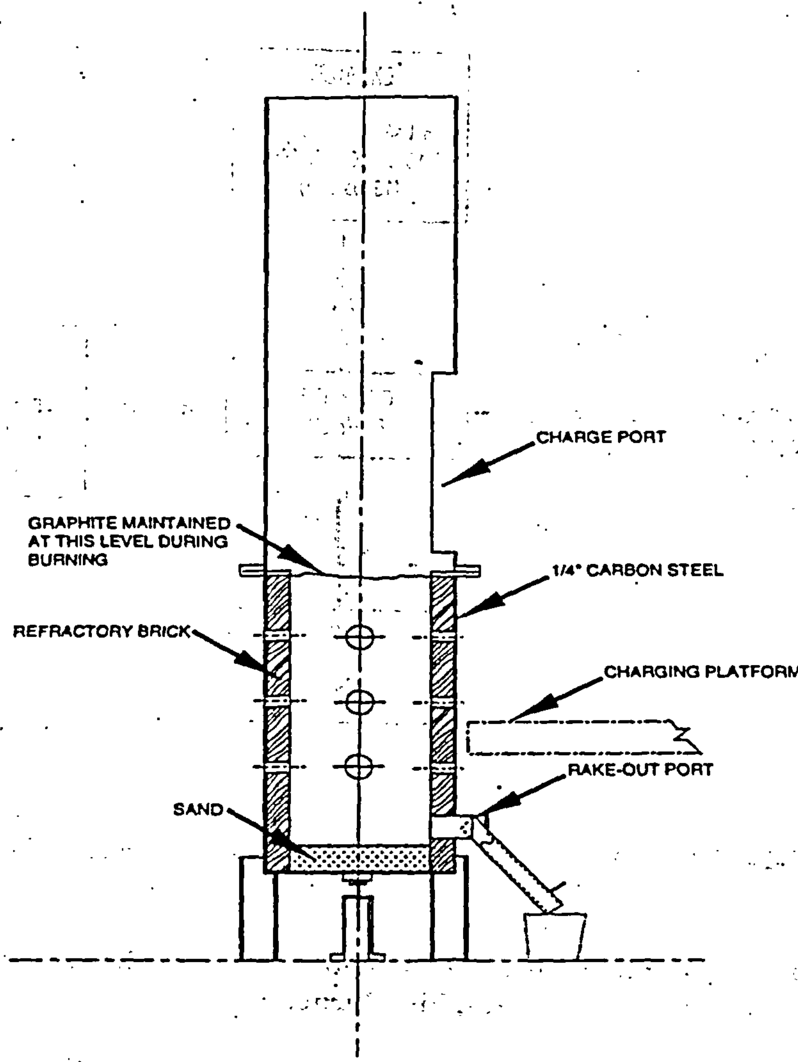

Figure K–13. Schematic of graphite burner; showing example rake-out port (one of seven), charge port, etc. Burner stood on concrete pad northeast of boiler plant; release height is 14.5 ft. above ground level.

Previous Uranium Release Estimates for Graphite Burner

The release estimates presented by Boback et al. (1987) for the graphite burner were provided by Neblett (1985), based on "knowledge gained from supervising these operations and from waste management's project assignments in the past." Neblett believed that data to substantiate these estimates were not available, and that the records for these operations had long since been discarded. A flow diagram showing his estimate is shown as Figure K–14. The total release estimate over the operating history of the graphite burner would be ≈ 130 kg uranium.

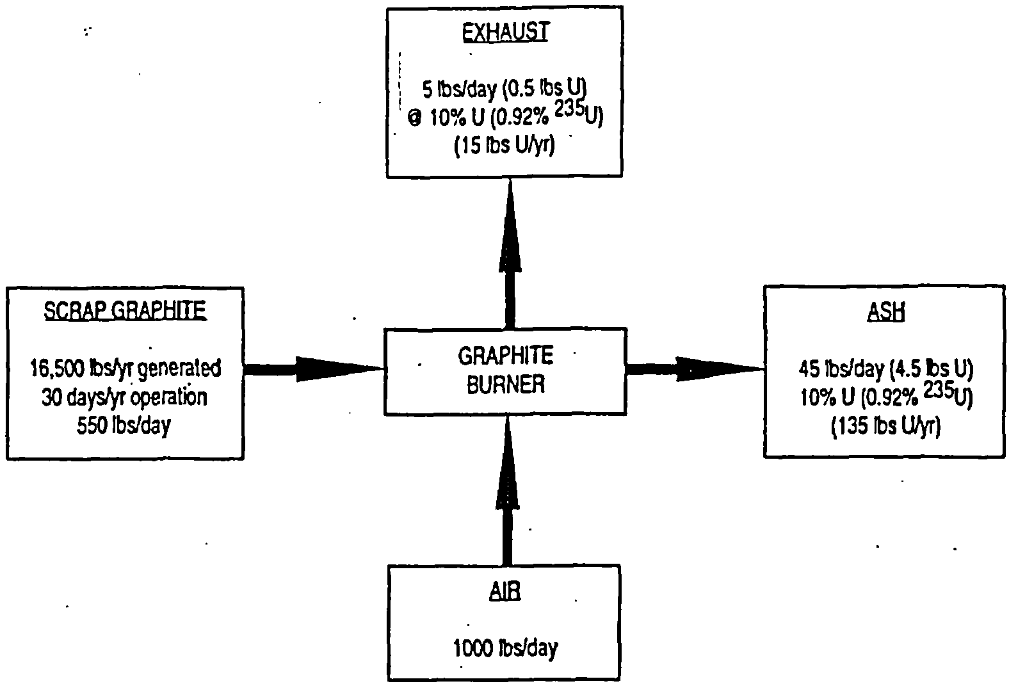

Figure K–14. Neblett (1985) estimate for historic releases of uranium from graphite burner.

Additional Historic Documentation Located to Support Reconstruction of Uranium Source Term from Graphite Burner

All of the original measurements and memoranda which were located for reconstruction of the source term for the graphite burner were generated during 1965, 1966, and 1967. The types of measurements included stack samples taken inside the burner, as well as air samples taken directly downwind of the burner during operation. Table K–17 shows a summary of measurements of uranium in air while burning uranium contaminated graphite.

Measurements of the loss of uranium in the graphite burner stack are summarized in an internal NLO memorandum (Ross 1966). We were able to locate and verify all of these measurements from original analytical data sheets. For this reason, we consider this source particularly reliable and definitive for dose reconstruction purposes. There were no data errors, however, the memo incorrectly identified three stack samples taken on 11/12/65 as "downwind samples."

One set of stack samples from the graphite burner were obtained on Millipore filters through a 1/4" stainless steel tube (Ross 1966). The velocity in the tube was about 2900 ft min^{-1} as opposed to the stack velocity of about 500–600 ft min^{-1}, thus the samples were obtained with higher than isokinetic flows. A total of 22 stack measurements were taken on three days (one each in December 1965, January 1966, and February 1966). The other set were apparently taken inside the burner "at open top of crucible." The method for these samples is unclear; however, in his memo, Ross used the former to estimate the uranium releases to air. The data from the two sets of measurements are shown in Table K–18.

Table K–17. Uranium Measurements in Air While Burning Uranium Contaminated Graphite in the Graphite Burner (Starkey 1965b)

	Uranium ($\mu g\ m^{-3}$)			Alpha [a] ($dpm\ m^{-3}$)
	High	Low	Avg.	Avg.
Process sample at top of burner or inside top of burner	8300	160	2100	3150
Downstream samples 3 ft to 5 ft downwind from burner in burner airstream	930	8	300	450[b]
Downstream samples, 12 ft from burner	13	13	13	20
Downstream samples, 80 ft from burner	3	2	3	5
Downstream samples, 150 ft from burner	12	10	11	15

[a] Calculated value given in Starkey (1965b). Apparently based on a ratio of 1.5 dpm alpha per microgram U.

[b] Additional data for this distance from the burner were located in an analytical data sheet for samples collected October 28, 1965. Four 15-minute samples of gross alpha in air, 4 ft downwind of the graphite burner stack and 18 inches above the horizontal top level of the burner, were: 590, 610, 490, and 185 dpm alpha per cubic meter air (average 470 dpm m^{-3}). The airborne contamination level at the same location when the operator was raking and charging the furnace was about three times higher (1400 dpm m^{-3}).

The burner was operated continuously, being shut down only for an emergency or lack of feed (Baer 1966b). During calendar year 1969, approximately 150,000 pounds of graphite scrap were burned in this facility (Anon 1970). It was not necessary to operate the burner all year; an average production rate was estimated by Anonymous (1970) as 1,000 to 1200 pounds of graphite scrap per 24-hour operating day. This corresponds to about 150 operating days per year, compared to Neblett's nominal estimate of 30 days per year. Perkins (1976) states the normal operation schedule as 4 weeks per year at a graphite throughput rate of 150–175 pounds per hour.

We used the stack measurements, the estimated stack flow rate and the estimated days per year of operation to determine an estimate of the annual airborne source term from operation of the graphite burner. Because the parameters involved in the estimate are all subject to some uncertainty, we used Monte Carlo sampling implemented by the Crystal Ball™ software system (Decisioneering 1992), using the parameter assumptions defined in Table K–19. The mean annual source term estimate was 15 kg per year with a 5th–95th percentile range of 3.1 to 37 kg per year. A summary of the source term characteristics for the graphite burner is compiled in Table K–20.

Table K-18. Uranium Concentration in Air Samples from Graphite Burner Stack During Operation (Ross 1966)[a].

	Uranium Concentration ($\mu g\ m^{-3}$)	
	Sampled at Open Top of Crucible[b]	Sampled Through Hole in Stack[c]
	160	240
	260	180
	290	190
	230	1500
	3510	1500
	600	1600
	480	430
	700	350
	4000	350
	6000	425
	1100	375
	150	300
	330	350
	8300	560
	3500	720
	190	1700
	800	270
	700	130
	470	280
	630	300
	870	270
		160
Number	21	22
Minimum	150	130
Maximum	8300	1700
Average	1584	554
Median	630	350

[a] Data verified by examination of original analytical data sheets.

[b] Location of samples within burner is not clear. Samples taken on six separate days in October and November 1965.

[c] Samples taken on three separate days in December 1965 and January and February 1966.

Table K–19. Assumptions Used in Uncertainty Analysis of Graphite Burner Source Term

	Minimum	Most Probable[a]	Maximum
Uranium concentration in stack gases ($\mu g\ U\ m^{-3}$)	130	554	1700
Exit flow rate ($m^3\ min^{-1}$)[b]	118	168	218
Days per year of operation	10	30[c]	200

[a] Triangular distribution.
[b] Assumed ± 30% of measured flow rate in Ross 1966.
[c] Neblett (1985) estimate. Confirmed in Perkins (1976).

In addition to an airborne source term through the stack, some uranium could have become airborne when ash was spilled during the rake-out process. The burner sat on a 55-ft by 85-ft concrete pad (Baer 1966b; DOE 1992), so gross contamination could have been easily cleaned up. Our assessment of this pathway for the old solid waste incinerator indicated that resuspension of spilled ash was relatively insignificant compared with stack emissions. Airborne resuspension of spills is handled in a general way under "non-routine events," later in this appendix.

Table K–20. Summary of Source Term Characteristics for the Graphite Burner (operated 11/1/65 to 9/14/84)

Parameter	Value	Reference/Basis	Notes
Uranium Source Term ($kg\ y^{-1}$)	15	Uncertainty analysis based on assumptions defined in Table K–19.	5th–95th percentile range is 3.1–37 $kg\ y^{-1}$.
Physical Release Height	14.5 (ft)	Engineering Drawing 10X-M-00324	Eight-ft stack above 5-ft furnace, mounted 1.5 feet above grade.
Building outer dimensions	37" diameter cylinder, 14.5 ft tall	Engineering Drawing 10X-M-00324	
Stack inside diameter	36.5 in.	Engineering Drawing 10X-M-00324; Anon 1970	Inside diameter of furnace area is 27", due to refractory lining.
Exhaust gas velocity	500–600 fpm 168 $m^3\ min^{-1}$	Ross 1966	

KELLEY SOLID WASTE INCINERATOR (OPERATED 11/1/79 TO 4/28/86)

In 1978, it was proposed to move the old solid waste incinerator to the Calciner Building, Plant 3, inside the production area, in part to reduce the ground contamination which was known to be occuring in the area of the sewage treatment plant (Anonymous 1978). On June 18, 1979, the new solid waste incinerator, manufactured by the Kelley Company, Model No. 780/31, was delivered to NLO, and installation into the incinerator building (39A), in the Plant 2/3 area (Figure K–8) was complete by October 31, 1979 (Anonymous 1982).

The incinerator was a controlled air, pyrolytic chamber with a thermal reactor mounted above the main chamber that burned particulate matter which would otherwise escape the stack (Baer 1981). The Standard Operating Procedure (Baer 1981) describes the proper operation of the incinerator. There was no gas cleaning equipment for the effluent air. For proper combustion and efficient operation, the main chamber temperature was kept below 1500 °F. A water spray, regulated by a flowmeter, cooled the incinerator internally to minimize the emission of particulate matter. The rated capacity of the incinerator is 700 lbs h^{-1}. Physical and operating parameters needed to assess dispersion from this release point are given in Table K–21.

Table K-21. Summary of Physical and Operating Parameters for the New Solid Waste Incinerator

Parameter	Value	Reference	Notes
Physical stack height	53 ft	Engineering Drawing 39A-M-00025[a]	
Building dimensions	52 × 54 ft × 37 ft high	DOE 1992	
Stack cross sectional area	2.292 ft^2	Anonymous (1982) Heatherton (1981)	Engineering Drawing 39A-M-00025 gives a stack diameter of 21", which is equivalent to 2.4 ft^2. However, the 2.292 ft^2 value is presented with stack testing information.
Exhaust gas velocity ($ft\ min^{-1}$)	2250 ± 450 1462–2908	Anonymous (1982)	Mean, S.D. and range during stack testing on 10 separate days in 1980–1982
Exhaust gas temperature (°F)	1330 ± 340 726–1669	Anonymous (1982)	Mean, S.D. and range during stack testing on 10 separate days in 1980–1982

[a]Incinerator Bldg. Solid Waste Incinerator. General Arrangement Plan and Elevation. November 13, 1979.

The incinerator was not designed for the disposal of radioactively contamined materials. Contaminated articles to be burned were to be inspected, emptied of all metal chips, turnings, and oxides, and vacuumed or wiped clean of obvious contamination. Contaminated production or process wastes, dust collector bags, contaminated gloves and rags, and other obviously contaminated materials were not to be burned in the incinerator (Baer 1981). However, operations in the new solid waste incinerator were suspended on April 28, 1986 after a series of investigations in the spring of that year (e.g. Huey et al. 1986), which showed that too much contaminated material was being sent to the incinerator, resulting in ash content of 2.5% U by weight. The solid waste incinerator did not resume operation after that time.

Uranium Release Estimates for the Kelley Solid Waste Incinerator

Previous release estimates for the Kelley Solid Waste Incinerator presented by Boback et al. (1987) are tabulated in Table K–22. Handwritten notes (Anonymous 1985) suggest that the value for 1984 is actually a total for the solid waste incinerator and the liquid waste incinerator, which are in the same building (39A). The source term for the solid waste incinerator alone was 2.4 kg and the liquid waste incinerator was 4 kg per year. The basis for the solid waste incinerator estimate appeared to be:

- Average charge rate: 337 lbs per hour
- Uranium in effluent: 0.36 gram U per 100 lb charged
- Operating schedule: 40 hours per week; 50 weeks per year.

In an informational letter to the Ohio EPA, Wing (1980) estimated emissions of 1.5 lb U per year (0.7 kg per year) from the Kelley Solid Waste Incinerator.

Table K-22. Previous Uranium Release Estimates for the Kelley Solid Waste Incinerator (Boback et al. 1987)

Calendar Year	Annual Release (kg)
1984	6.4[a]
1983	2.4
1982	1.8
1981	1.2
1980	0.68

[a] Should have been 2.4, according to handwritten notes illustrating the basis for the 1984 stack loss estimate (Anonymous 1985). The sum of releases for both the solid and liquid waste incinerators was 6.4 kg.

We used this same basic approach to estimating a source term for the solid waste incinerator, but examined the uncertainty in the input parameters more carefully. It appears that the contractor estimate (at least for 1984) was supposed to be an upper bound, because the incinerator typically did not operate 2000 hours per year. A typical operating schedule for the FMPC incinerator was estimated to be 400 to 600 lbs waste burned per hour for 6 h d^{-1}, 2 d wk^{-1}, 98 d y^{-1}, or 1176 hours per year, equally distributed throughout the year (Ostendorf 1979). Wing (1980) estimated a maximum burn rate of 340,000 pounds per year of solid waste, which is equivalent to about 630 hour per year, at the average refuse charge rate (see below). For our analyses, the operating schedule is defined as a triangular distribution with a most likely value of 1176 hours per year, and minimum and maximum values of 600 and 2000 hours per year.

A number of stack tests were conducted at the new solid waste incinerator. The results for 10 separate days of testing in 1980–1982 are summarized in Anonymous (1982). From this summary, the amount of particulate material released was obtained. The actual values ranged from 0.09 to 0.71 pounds particulate per 100 pounds of refuse charged. For our uncertainty analysis, this parameter was described by a custom distribution based on the original measurements. Another parameter obtained from the stack testing summary was the refuse charging rate, which ranged from 172 to 758 pounds per hour and averaged 536 pounds per hour.

The stack testing summary (Anonymous 1982) does not contain any information on uranium emissions, only total particulates. However, we located original analytical data sheets for seven of those ten tests, and two others, which provided data on the uranium content of particulates collected from the effluent air from the incinerator. The measured values ranged from 0.32 to 15.4 mg U per g particulate. For our uncertainty analysis, this parameter was described by a custom distribution based on the original measurements.

The annual uranium release rate for the Kelley Solid Waste Incinerator, based on the operating data and measurements just described, is illustrated by the distribution in Figure K–15. The median estimate of the total release over the 5.5-y operating period is 8 kg.

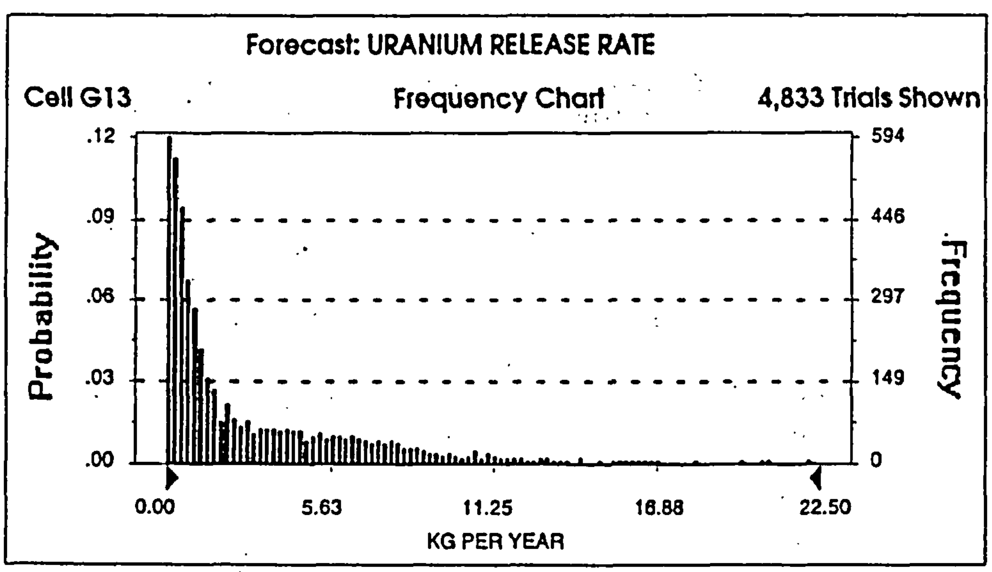

Figure K–15. Source term estimate for the Kelley Solid Waste Incinerator. The median source term estimate is 1.3 kg per year, and the 5th and 95th percentile values are 0.1 kg and 17 kg, respectively.

TRANE THERMAL LIQUID WASTE INCINERATOR (OPERATED 3/28/83 TO 5/7/86)

Within a month after the old oil burner was closed down, a project proposal for a new liquid waste incinerator had been prepared (Anonymous 1979). The proposal indicated that stack gases from the new unit would be cooled with dilution air and passed through a new bag filter collector to ensure compliance with Ohio EPA requirements for particulate emissions from incinerators. The facility was to be capable of processing 20 55-gal drums of waste oil per month when operating on one shift per day.

The incinerator began operating in late March 1983. Physical and operating parameters for the incinerator are given in Table K–23. Operations at the liquid waste incinerator were suspended on May 7, 1986 due to uncertainties associated with the characteristics of the waste oil feed stream as well as the status of permit action on the incinerator. Operations were never resumed.

Table K–23. Summary of Physical and Operating Parameters for the Trane Thermal Liquid Waste Incinerator

Parameter	Value	Reference	Notes
Physical stack height	53 ft	Audia (1980)	
Building dimensions	52 × 54 ft × 37 ft high	DOE 1992	
Stack inside diameter	14"	Audia (1980)	
Exhaust gas flow rate	4500 acfm 4200 fpm	Audia (1980)	
Exhaust gas temperature (°F)	400	Audia (1980)	
Waste processing rate	max: 56 lb (7.5 gal) per hour	Audia (1980)	Typical value was 7 gal per hour.

Boback et al. (1987) estimated releases of 3 and 4 kg from the liquid waste incinerator for 1983 and 1984, respectively. They state that the release estimates for the liquid organic waste incinerator were based on performance criteria and the concentration of uranium in the incinerator feed. Handwriten notes (Anonymous 1985) state that the liquid waste incinerator would emit 4 kg yr^{-1} under the *maximum* permitted rate. These calculations (partly illegible) appear to use a uranium concentration of 8.7 g per L in the waste. This is the concentration given by Wing (1980), who indicated that there was a large backlog of liquid wastes containing about 8.7 g U per L. According to our previous assessments of FMPC liquid wastes, typical oils and solvents would contain U concentrations of 2–4 g U L^{-1} (see Oil Burner section above). For our source term estimates for the new liquid waste incinerator, we used a uniform distribution between 2 and 9 g U L^{-1} to describe the uranium concentration in liquid waste being processed.

The quantity of liquid waste generated at this time was approximately 1000 gal per month. The Ohio EPA permit application for the liquid waste incinerator (Audia 1980) provides the following additional information relevant to estimating releases (see also Table K–23). The operating schedule was to be 8 hours per day, 5 days per week, 49 weeks per year (1960 hours per year). At an average processing rate of 7 gallons per hour, this operating schedule is equivalent to 14,000 gal per year. Additional information in Wing (1982) specifically indicates an intended processing rate of 12,000 gal per year of lubricating and cooling oils and 400 gal per year of kerosene/TBP mixtures, both of which would contain low-level U contamination. Wing (1980) stated that the maximum amount of oil expected to be burned was 145,000 liters (38,300 gal) per year and that a typical year would be about 1/4 of this amount (9600 gal per year). He estimated a first year emissions rate of 8.6 lb U (3.9 kg), based on 1.36 g U released per 100 lb. charged. No reference was given for the basis of this emissions ratio. For our analyses, we used a processing rate of 12,000 gal per year with a standard deviation of 1,000.

As was determined for the old oil burner, we used a uniform airborne release fraction of 1–10% for the amount of uranium released from the waste to the off-gas from the incinerator. The basis for this distribution is discussed in the previous oil burner section. A collection efficiency of 90% for the bag collector was used (CIV 1980). The distribution describing the estimated annual release from the Trane Thermal Liquid Waste Incinerator is shown in Figure K–16. The median estimate of the total release over the 3.1-y operating period is ≈4 kg.

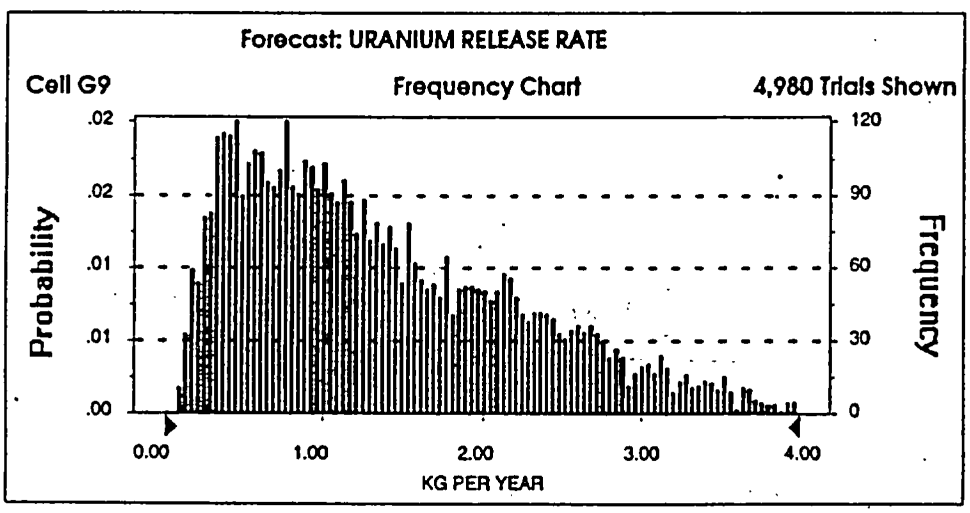

Figure K–16. Source term estimate for the Trane Thermal Liquid Waste Incinerator. The median source term estimate is 1.2 kg per year, and the 5th and 95th percentile values are 0.3 kg and 3 kg, respectively.

PARTICLE SIZE AND DEPOSITION CHARACTERISTICS FOR URANIUM EMISSIONS FROM FMPC INCINERATORS

The solid waste incinerators, the graphite burner, and the oil burner all operated without any emissions control equipment. Thus, any uranium-contaminated particles which were entrained in the exiting air stream were emitted directly to the atmosphere. Although there were some fallout studies of uranium deposition around the oil burner and graphite burner (Klein 1963; 1964), these were not of the quality necessary to quantify the particle size and deposition characteristics. The most applicable experimental data located were generated by scientists from the Health and Safety Laboratory (HASL) in New York (Weinstein and Breslin, unpublished manuscript titled "Environmental Contamination from Burning Uranium Metal, circa 1959). These experiments involved the open burning of uranium metal, in the form of turnings, shavings and chips, in quantities ranging from 20 grams to 900 pounds. Experiments were conducted both in the laboratory and in the field.

Mass median diameters of uranium oxide in stack effluents were 3.2 to 6.5 microns compared to 1.34 to 1.76 microns for oxide in the smoke plume 25 feet downwind at ground level. A value of 0.51 microns was determined in a laboratory experiment.

Weinstein and Breslin present curves of uranium deposition in $\mu g\ ft^{-2}$ with distance from a natural draft field incinerator with a 12-foot stack. Following burning of 160 lbs of uranium, average total deposition decreased by about a factor of 7 (from 10 to 1.5 $\mu g\ ft^{-2}$) between 100 feet and 300 feet from the source. For a 160 lb source, total deposition decreased from about 40 $\mu g\ ft^{-2}$ at 100 feet to about 15 at 300 feet (about a factor of 3). Their computed average deposition velocity to gummed paper was 0.66 $m\ s^{-1}$ for a 100 pound uranium fire, 0.47 $m\ s^{-1}$ for a 160 pound fire and 0.57 $m\ s^{-1}$ for a 900 pound uranium fire. These data support other evidence presented in this report which indicates a rapid drop-off of contamination with distance from the FMPC incinerators.

UNMONITORED RELEASES OF URANIUM FROM FMPC BUILDINGS

This section on unmonitored emissions from FMPC buildings is divided into three categories: unmonitored process emissions, building ventilation, and laboratory hoods. No revised estimates are provided for emissions from unmonitored processes and lab hoods. Some new information uncovered since our interim source term report (Voillequé et al. 1991) did permit a thorough reevaluation and reconstruction of releases from building ventilation.

Unmonitored Process Emissions

An addendum to the estimates of radionuclides released from FMPC during 1951–1984 (Boback et al. 1987) was prepared by Clark et al. (1989) following the determination in June 1988 of higher than expected airborne concentrations of uranium at air monitoring stations in the NE quadrant of the plant. These air concentrations were traced to the operation of the Plant 2/3 denitration gulping system which was a previously unmonitored source of uranium emissions. Uranium releases from the gulping process are presented in Appendix H. Because this system was previously thought to be an insignificant source of airborne emissions, concern was raised about other unmonitored and potentially unreported air emission sources that had or still existed.

All unmonitored radionuclide emission sources were investigated during the plant-wide vacation shutdown in July 1988. Processes associated with monitored stacks were well characterized in terms of emissions and were allowed to restart after the vacation shutdown was completed. However, the investigation found 35 unmonitored process stacks (associated with 26 separate production processes) that were potential sources of significant radionuclide emissions. The majority of sources associated with unmonitored stacks are generally described as wet exhausts. These are scrubbed exhausts, such as Plant 2/3 UO_3 gulping, and exhausts from processes involving acid dissolution of uranium. Previous testing had quantified emissions from the Plant 8 furnace discharges in 1988.

The method used by Clark et al. (1989) to estimate emissions from the current unmonitored uranium processes at FMPC was either engineering calculations or actual stack emission measurements. Processes were not sampled if sufficient data existed to approximate emissions (Hill 1989a). Calculations considered the uranium concentration in each process, the capacity of the stack blower, and other operating parameters. Information from equipment manufacturers and process experience were used in the calculations. Where data were not sufficient to estimate emissions, personnel from Westinghouse Materials Company of Ohio and two environmental emissions testing firms sampled the process exhausts, using EPA Method V for stack sampling.

In the cases of historic processes which were no longer in use, extensive literature searches and interviews were used to re-create the production processes. Based on this process information, emissions were determined based on comparison to current operations or by calculation based on estimated equipment efficiencies and throughput for processes where no current operation was available for comparison (Hill and Dolan 1988).

An emission factor per ton of uranium processed or per hour of operation was developed for each unmonitored process (Hill 1989a). These emission factors per ton (or per hour) were then used together with the plant-by-plant production data to estimate the emissions from the unmonitored processes in each year. Table K–24 summarizes their estimates of unmonitored process emissions for 1953–1988. The year-by-year estimates are presented in Table 3 of Hill (1989a). The total of 324 kg was only <1% of the total atmospheric releases of approximately 135,000 kg U reported by Boback et al. (1987) for 1951–1984.

An unmonitored source that was not included in Hill and Dolan (1988) is a box furnace, which was installed in the pilot plant in 1956 to process U_3O_8, enriched uranium turnings, sawdust and other residues generated in the production of enriched cores (Mead 1972). Hill and Dolan did not estimate the annual emission for this source because no production data were available. However, they state that the exhaust air from the furnace is "a probable source of radionuclide emissions." Starkey (1964a) estimated emissions from the oxidation furnace dust collector at 6 pounds of uranium per month, when the furnace was in use. At that time, it must have been used about two months per year because his average monthly emission estimate is 1 lb mo^{-1} (5 kg y^{-1}). This was 12% of his estimated total of unmonitored emissions from the Pilot Plant.

We have not derived any new source terms for these miscellaneous unmonitored process emissions. From our review of the documentation, the methods used to derive the estimates in Table K–24 appear reasonable. A subjective uncertainty of a factor of three is applied to the previous estimate, resulting in an uncertainty band of 110–970 kg U over the 1953–1988 period.

Table K-24. Estimates of Miscellaneous Unmonitored Process Emissions for 1953–1988

Process	Uranium Release (kg)[a]	Percent of Total
Plant 6 Briquetting	121.0	37.3
Plant 6 Pickling	12.2	3.8
Plant 9 Briquetting	49.1	15.1
Plant 9 Pickling	3.7	1.1
Nitric Acid Recovery	33.4	10.3
Cooling Towers	105.1	32.4
Total	324	100

[a] Values from Table 3 of Hill (1989a).

Building Ventilation

For building ventilation, data collected by continuous air monitors (CAMs) located in each plant in 1987 were used with engineering information concerning the building exhaust fans (Hill and Dolan 1988, Hill 1989c). All radiation measured by the CAMs was assumed to be normal uranium; an assumption that Hill and Dolan felt was reasonable because the plants process large quantities of depleted uranium and lesser amounts of low-level enriched uranium. The average concentrations reported as monthly results from the individual CAM units were averaged to provide a typical building concentration for each plant.

The building exhaust fans, which were the principal source of fugitive emissions, were not used all year in most of the plants; they were used primarily in the summer to lower the temperature in the buildings. Estimates of the actual operating hours for each fan were obtained from Operations personnel, and the assumption was made that exhaust fan use has not varied appreciably during the history of operations (Hill 1989c). Because the CAMs were usually placed on the floor in areas of highest suspected concentrations, the exhaust fans are in the ceiling or high on the walls, and make-up air enters the plant through open doors and windows, the measured concentration was multiplied by a factor of 0.1 to account for dilution (Hill 1989c).

Hill assumed that emissions from building ventilation would be proportional to production rates (Hill 1989c). Historical release estimates were estimated by multiplying the 1987 release estimate for each plant by the ratio of production for that plant in the year in question to production in 1987. Explicit calculations for each plant are provided as an attachment to Hill (1989c).

The resulting uranium release estimates from building ventilation are given in Table 1 of Clark et al. (1989). The total release estimate for 1953–1988 was 389 kg from building ventilation as compared to about 179,000 kg from all sources. The highest annual release estimate was 33 kg in 1960. The most significant contributors to the building ventilation emissions were Plant 6 (39% of total) followed by Plants 2/3 (20% of total) and Plant 4 (29% of total) (Hill and Dolan 1988).

In our view, one of the main weaknesses of Hill's assessment of historic releases in FMPC building ventilation is the assumption that past air dust levels could be scaled from the 1987 measurements according to the ratio of production in an earlier year to that in 1987. This approach may not reflect the increasing emphasis which was placed on contamination control equipment and procedures through the years.

An evaluation of air dust levels in uranium feed materials production facilities for 1948–1956 was located (Breslin 1958), which permitted a forward projection of ventilation releases from historic air measurements. These measurements by the Health and Safety Laboratory for 1954–1956 should be quite typical of the Fernald operations, because in fact the FMPC was one of the two consolidated uranium production facilities in which the measurements were taken during those three years. The surveys were designed to obtain time-weighted average daily exposures to workers. Representative replicate air samples were collected at all the jobs and in all areas to which each employee was assigned during the working day. Breslin (1958) presents the data in a series of plots illustrating the percentage of workers exposed to a certain concentration range in different types of plants in the uranium production facility. Our readings of Breslin's plots (estimated to be accurate within ± 2%, or a fraction of 0.02) are tabulated in Table K–25. One data set, measurements in scrap recovery operations in 1955, did not add up to a total frequency of 1.0, presumably due to an error in the drafting of that figure.

Table K–25. Fraction of Workers Exposed to Various Airborne Contamination Levels in Feed Materials Production Facilities in 1954–1956 (from Breslin 1958)

Type of Plant		dpm per cubic meter air						
		0–55	55–110	110–220	220–440	440–880	880–1800	>1800
All	1954	0.78	0.08	0.06	0.03	0.03	0.00	0.02
	1955	0.57	0.19	0.11	0.05	0.02	0.03	0.03
	1956	0.83	0.09	0.03	0.04	0.01	0.00	0.00
Rolling	1954							
	1955	0.28	0.25	0.24	0.11	0.02	0.10	0.00
	1956							
Reduction & Recasting	1954	0.78	0.10	0.08	0.00	0.04	0.00	0.00
	1955	0.72	0.15	0.04	0.07	0.00	0.00	0.02
	1956	0.78	0.19	0.01	0.01	0.01	0.00	0.00
Refining	1954	0.78	0.08	0.09	0.03	0.00	0.00	0.02
	1955	0.89	0.09	0.02	0.00	0.00	0.00	0.00
	1956	0.82	0.03	0.06	0.09	0.00	0.00	0.00
Scrap Recovery	1954	0.46	0.18	0.00	0.00	0.00	0.00	0.36
	1955	0.35	0.12	0.00	0.00	0.00	0.00	0.34
Sampling	1955	0.29	0.71	0.00	0.00	0.00	0.00	0.00
	1956	0.14	0.56	0.14	0.08	0.00	0.08	0.00
Fuel Fabrication	1954	0.81	0.04	0.03	0.07	0.04	0.01	0.00
	1955	1.00	0.00	0.00	0.00	0.00	0.00	0.00
	1956	0.97	0.03	0.00	0.00	0.00	0.00	0.00

We used the measurements of Breslin and the ventilation characteristics of the Plants as defined by Hill (1989c) to estimate the release of uranium by building ventilation in 1954–1956. See Annex 3 to this appendix for ventilation capacities and fan operating factors. The distributions of airborne contamination levels shown in Table K–25 above were entered as custom distributions in the Crystal Ball™ uncertainty analysis software (see Annex 3). When Breslin's data were not available for a particular year and type of plant, we used the adjacent year's data for the same type of plant. Plant 9 was not operating at this time, but we included Plants 1, 2/3, 4, 5, 6, and 8 in our assessment. Breslin's operating categories were correlated with the FMPC plants as follows: Plant 1: Sampling; Plant 2/3: Refining; Plants 4 and 5: Reduction and Recasting; Plant 6: Rolling; and Plant 8: Scrap Recovery. Because the mid-1950s was a peak operating period, we assumed that operations were underway virtually full-time (3 shifts per day). Because of the 2-week vacation shut-down and other maintenance operations, we used an overall operating fraction of 0.95 for all active plants in 1954–1956.

The measurements of uranium in air in active working areas may not be representative of building exhaust air, although it should be roughly proportional. This issue relates to the dilution factor parameter that Hill used for make-up air. This dilution factor is intended to describe the ratio of the concentration of radioactivity in air measured by the constant air monitors in the working areas to the concentration in air leaving by the exhaust fans. If inlet air entered the building above the working areas, then the exhaust fans would be removing air which was relatively less contaminated than that measured in the working areas. This factor was treated as an uncertain parameter in our analysis which could range from 1.0 (no dilution) to 10, with a most likely value of 3. As discussed previously, Hill had used a factor of 10 dilution, which in our judgement would be a more reasonable upper bound than a central estimate.

The release estimates from building ventilation in 1954, 1955, and 1956 are presented in Table K–26 below. They are considerably higher (medians ranging from 150–220 kg per year) than Hill's previous estimates for these years. We feel they are a better estimate because they are linked to direct measurements of airborne contamination made at the time. However, they are still quite uncertain, for the reasons discussed above.

Table K–26. Median Release Estimates for Uranium in FMPC Building Exhaust Ventilation in 1954–1956, Based on Air Dust Data from Breslin (1958) and Building Ventilation Characteristics Described by Hill (1989c)

Percentile of Estimate	kg U per year		
	1954	1955	1956
5%	35	37	37
25%	68	73	76
50%	150	220	190
75%	400	420	390
95%	730	760	650

In the mid-1960s, Starkey (1964a) provided an estimate of unmeasured dust losses to the atmosphere in building exhaust air, which is shown in Table K–27. He indicated that none of his estimates for the exhaust systems were based on direct measurements. The estimates of releases from building ventilation were minor compared to other process sources which Starkey examined at the same time, so there may have been little incentive to refine his initial estimates for building ventilation. His estimate does lend credence to our considerably higher release estimates.

Table K-27. Historic Estimates of Unmonitored Dust Losses to the Atmosphere via Roof and Wall Exhaust Fans (Starkey 1964a)

Plant	Operating Fraction[a]	Average Monthly Release (lb U mo^{-1})	Annual Release (kg U y^{-1})
1	1	3	16
4	5/13	30	163
5	3/50	3	16
6	1	3	16
8	5/9	5	27
9	1	5	27
Pilot Plant	1	4	22
Total		53	287

[a] The effect of discontinuous operation of exhaust fans was included in the estimate of the average monthly release.

An estimated source term for uranium in building ventilation was projected forward in time from the 1956 release estimate. The projection to future years (through 1970) was made by scaling the 1956 estimated release (by plant) according to plant-specific production rates. The production data used for this assessment are included in Table K3–1 in Annex 3 to this appendix. For some plants, which contained different types of production activities, the key production processes, which were associated with high airborne contamination levels were used for the projection. For example, major activities in Plant 9 included both production of uranium ingots and machining of metal products (Appendix C). Because the casting operation appeared to be the more signficant one in terms of airborne radioactivity levels, we based our projection for Plant 9 only on the uranium ingot production rates (Table K3–1), and we used Breslin's measurements of uranium in air in reduction and recasting operations areas. In addition, we used the production data for rolling operations in Plant 6, since the fabrication operations in that plant are much less significant in terms of contamination levels.

With the exception of the magnitude of the dilution factor, we felt that the method of Hill was reasonable for projecting as far back as 1970. Hill's estimates, adjusted to a dilution factor of 3 rather than 10, are presented in Figure K–17 along with the other estimates. Uncertainty was propagated through the projections. The detailed distributions for the annual estimates are included in Table K3–2 of Annex 3 to this appendix. Roughly 80% of the total releases from building ventilation occurred between 1957 and 1970, inclusive. The other 20% occurred in 1954–1956 (≈14%) and during 1971–1987 (≈6%).

The increase in the estimated release between 1957 and 1960 is primarily due to the increase in scrap recovery operations in Plant 8. These operations produced high airborne activity levels inside the plant (Table K–25), which are reflected in emissions in building ventilation. Plant 8 production was a factor of 2 higher in 1960 as compared to 1956. In our final dose report, the entire building ventilation source term will be modeled as if it had been released from Plant 8. This simplification is warranted by the relatively small releases and the fact that over half of the total uranium released in building ventilation through 1970 is believed to have come from Plant 8.

It is encouraging that Starkey's estimate for 1964 is in very good agreement with our forward projection (Figure K–17). The forward and backward projections were overlapped for three years (1970–1972) to see how they compare (Figure K–17). The forward projection results in median source term estimates which are a factor of 2–3 higher than those obtained using the backward projection for these three years; however, the uncertainty distributions overlap. Considering the range of estimates over the history of plant operations, we consider this agreement good. In addition, both methods produce estimates of releases which are minor relative to other sources after 1970 (see concluding section of this appendix). No further investigation into the differences in the two projection methods was made.

The total estimated release of uranium in FMPC building ventilation during 1954 through 1987 is 4100 kg (median estimate) with a 5th–95th percentile range of 970–15,000 kg. This is about a factor of ten higher than Hill's previous estimate (390 kg). The two main reasons for the large difference are: (1) the use of a lower dilution factor for building make-up air and (2) the use of higher in-plant airborne contamination levels, measured in the 1950s, to make a forward projection through 1970.

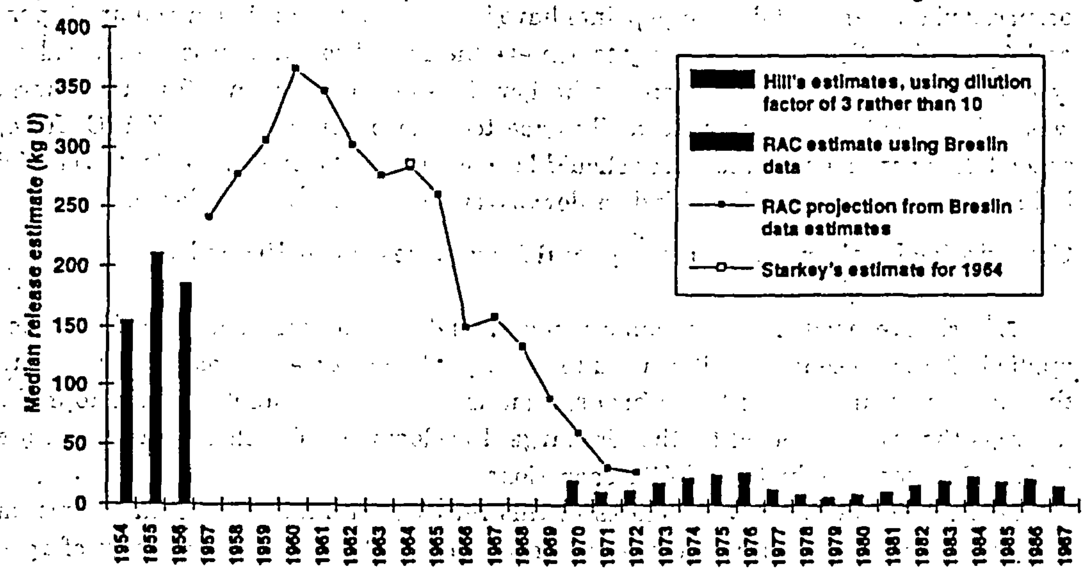

Figure K–17. Summary of release estimates for uranium in building exhaust ventilation.

Laboratory Hoods

Uranium emissions from laboratory hoods were based upon the average number of samples processed per year in each hood, the probable loss per sample and the estimated uranium concentration of the samples (Hill and Dolan 1988). Existing hoods and known former laboratory exhausts were evaluated. The resulting emission estimate was a constant at 1.9 kg y^{-1}, or 66.5 kg over the period 1953–1987. Hill and Dolan (1988) indicate that the estimate for emissions from laboratory hoods is probably high; however, a more accurate value would have required long-duration stack tests for each vent, which were not warranted due to the low emission estimate. From review of the relevant documentation, we agreed that no further assessment was warranted.

EPISODIC RELEASES

Accidental releases are frequently characterized as increases in the effluent discharge rates due to unplanned and non-routine events. Typical events can include spills, fires, and cleanup system failures. However, when the frequency of unusual events is high, one questions whether the adjective "non-routine" is correct. Similarly, when a large release is the result of a conscious operational decision, it hardly qualifies as unplanned. Such situations complicate the definition of the term accidental releases, so the term episodic releases will be defined below for a specific purpose.

Semantics aside, the important concerns about such releases for dose reconstruction are whether they were detected and/or sampled and whether their magnitudes are sufficient to warrant special treatment in the dose estimation process. Radionuclide releases that occurred via unsampled discharge points have been estimated in this appendix, Appendix H, and Appendix I. This was necessary to achieve the goal of completeness for the radionuclide source term. The second question is whether the event caused an effluent discharge that was substantially above that normally expected and observed at the FMPC. If so, then special dose assessment procedures should be employed to document the doses to individuals living in areas that were downwind, or downriver, at the time of the release.

Criteria for Implementation of Special Dose Assessment Procedures

Criteria are needed to determine when special dose assessment procedures should be applied. These procedures will be used to estimate the movement of particular discharges in the local environment and to perform special assessments of radiation doses to individuals in areas that were affected by the discharge. Development of such criteria is not a simple task in the context of historic FMPC operations.

Presentations of release estimates in this report have illustrated both the magnitudes and the variability of effluent discharges from the FMPC. In the early years of operation, large amounts of uranium were frequently discharged to the atmosphere and to the river. As many as ten facilities contributed to the total discharges from the FMPC. The largest discharges were not always from the same facilities, although some facilities were clearly more important sources of effluents than others. This means that a large increase in the effluent from a facility that was a minor contributor to the total discharge could have a negligible effect on the total release rate. Also, the magnitudes of the total discharges have decreased substantially over the years. This has the effect of lowering the threshold above which a particular release of radioactivity would deserve special attention in dose assessment.

These factors require that the criteria for special dose assessment procedures must consider the magnitude of the release in the context of the releases from all of the facilities at the FMPC and the relative importance of the release to the total discharge at the time it occurred. Releases in recent years that were large enough to be significant perturbations to the overall plant effluent and the cause of an inquiry (Investigation Board 1988) would have had a relatively minor effect on the total monthly discharge during early years of operation.

In consideration of these historical facts, the following are criteria that can be used to determine whether special evaluation of a release from a particular event is warranted:

- the event under consideration caused the composite release rate of the FMPC to increase by a factor of ten or more above the value that would otherwise have been observed, and
- the duration of the high release rate caused by the particular event was less than 10 days.

The second criterion takes into consideration the fact that natural dispersion phenomena also play a significant role in the dosimetric analysis. For releases of long duration, the variability in dispersion conditions, including wind direction, will spread the effect over a wider area and reduce the magnitude of the increased dose to individuals in any particular area.

Episodic Releases Identified from Document Review

A review of available incident reports was conducted to identify those which involved potential releases of uranium to the atmosphere (Table K–28). Further study reveals that three short-term releases from the Pilot Plant, two which involved breached UF_6 cylinders and one which resulted from dust collector failure, satisfy the criteria for special environmental dose evaluations. It should be noted that the failure of dust collectors was fairly common and resulted in relatively large quantities of uranium being released from the FMPC. However, these episodes generally occurred over periods of time that resulted in release rates within a factor of ten of that normally observed. A compilation of major dust loss incidents is documented in Adams (1985) and, with the exception of the one described below, are included in the annual source terms presented in Appendix E. Finally, many of the incidents shown in the table were described in the incident reports without estimates of emissions. It was assumed in such cases that the releases were minor because little effort was expended in determining the magnitude of the releases. This assumption is justified in light of calculations made in the section entitled "Non-routine releases," which appears later in this report.

The first episodic release occurred in the Pilot Plant on November 7, 1953, when a cylinder containing uranium hexafluoride (UF_6) was breached releasing approximately 100 pounds (45 kg) of the gas. The cylinder had been heated up, and UF_6 was flowing via a feed line to the reactor (which converts UF_6 to UF_4) when a plug developed in the reactor. After the cylinder was shut off to purge the feed line, leakage of UF_6 was observed around the valve stem and a dry ice fire extinguisher was used to freeze the valve. As a cap was being placed on the valve outlet, the hex plug in the valve broke loose, and UF_6 escaped rapidly from the valve outlet. The Fire Department succeeded in stopping the leak with the use of water spray. Davis (1953) gives few other details about the effluent. However, he does note that the duration of the release was 15 minutes and that there was a strong northerly wind blowing at the time.

The second episodic release occurred in the Pilot Plant sometime between November 12 and November 16, 1960, when slightly enriched uranium was lost from dust collector G20-_0. There was some question as to the amount of uranium lost and when it was lost. An assessment of the potential release is presented in Appendix V of the Task 4 Report (Killough et al. 1993). After evaluating documents pertaining to the release and environmental monitoring records, it was concluded that the release was 300 or 500 kg d^{-1} for the first five or three days, including November 16, and about 200 kg d^{-1} during the last three days of the event.

The third episodic release occurred in the Pilot Plant on February 14, 1966. The following description of the UF_6 release was obtained from NLO (1966) and Boback and Heatherton (1966). On February 14, 1966, at approximately 8:40 AM, about 3800 lbs (1724 kg) of uranium, as UF_6, escaped from a 10-ton cylinder being heated by steam to transfer the gas to the process system in the Pilot Plant. NLO (1966) states that 2150 lbs (975 kg) were accounted for in the waste streams. Thus, as much as 1650 pounds (750 kg) may have been released to the atmosphere.

The sequence of events in the 1966 incident is as follows. The cylinder was cradled in a movable vaporizer chest and connected to the process equipment with copper tubing. The cylinder had been heated up, using steam. An operator accidentally removed the cylinder valve while attempting to open it. Another operator quickly manipulated a water deluge valve, but failed to actuate it. A full stream of UF_6 gas was expelled from the container and vented to the atmosphere through a hood positioned over the vaporizer chest. Pilot Plant personnel began to spray the cylinder with a water hose and were quickly joined by the fire brigade (time = 8:47 AM), who connected fire hoses and began to direct the spray into the cloud near the place where it was leaving the chest. The hood was then raised, and a direct water stream was applied to the end of the chest so that it rebounded against the cylinder at the valve opening. Continued application of water for about one hour finally cooled the cylinder and reduced its pressure sufficiently to permit a wooden plug to be driven in the valve opening.

The escaping gas was carried by wind in a southeasterly direction over a laboratory building and the administration building. Airborne UF_6 hydrolyzes quickly on contact with moisture in air to form UO_2F_2 and HF. Boback and Heatherton (1966) state that there was a "light fog of steam and hydrolyzed UF_6 which drifted near the lab and Administration buildings." Personnel involved in the emergency actions or who had any reason to believe that they may have inhaled some of the material were asked to submit urine samples. During the week following the incident, 280 employees and four visitors submitted 1024 urine samples which were analyzed for uranium. Of these, 115 employees had U concentratrions that exceeded 0.025 mg U L^{-1}. Six employees, who were directly involved in emergency procedures, had uranium concentrations that exceeded 1 mg L^{-1}. Sixty employees had a uranium concentration between 0.1 and 0.9 mg L^{-1}. Some of these were not near the release site, but walked in the foggy area near the lab and administration buildings. After 24 hours, the urinary uranium concentration of most employees had dropped to pre-incident levels.

Table K–28. Summary of Incident Reports Involving Potential Uranium Releases to the Atmosphere

Date reported and description of incident	Duration of release to the atmosphere	Total U released to atmosphere lbs(kg)	Meets episodic release criteria?
11/2/52. Broken crucible in the 3037 area of the Pilot Plant. (Heatherton 1952)	Not reported.	None reported –probably contained.	No
11/7/53. Release of UF_6 from defective cylinder in Pilot Plant. (Davis 1953)	15 min	100(45)	Yes
4/6/54. Metal oxide spill from a cyclone in the Spray Calciner system in the combined raffinate area. The spill was due to removal of an inspection plate from the cyclone. (Turner 1954)	Not reported.	Probably minor – some fine dust was observed to blow onto stone east of the area.	No
6/4/54. Spill of South African concentrate from drums. Drums were in transport to Plant 2 when one of the trailers broke loose from the train and struck a light pole located at the south end of the slope west of the Chemical Warehouse. Sixteen drums spilled—14 filled in the process of cleanup. (Costa 1954)	Not reported (assume 1 hour).	<2(<1) (assuming a max U conc. of 1.25%)	No
7/8/54. Small amount of leakage of UO_3 from juice hoppers stored on storage pad. (Walden 1954)	Not reported.	None reported.	No
12/6/54. Spill of diuranate cake from two drums at the Plant 8 storage pad. (Harrell 1954)	Not reported.	None indicated (all recovered).	No
6/24/55. Fire in feeding tray of oxidation furnace (Plant 8). (Stefanec 1955)	0.5 hr	None reported.	No
1/25/56. Release of metal oxides from storage silos in the Plant 1 area resulting in widespread contamination of ground, buildings and equipment in area extending from "A" to "B" street, and from the North side of the Chemical Warehouse to the North side of the Refinery Building. Area was subsequently decontaminated. (Heatherton 1956; Strattman 1956)	Unknown	Probably insignificant (material deposited on snow, which was then removed).	No
6/29/56. Explosion in Extraction Area of Pilot Plant. (Halcomb 1956)	Instantaneous	Probably minor (appears to have been contained).	No

(continued next page)

Table K-28. Summary of Incident Reports Involving Potential Uranium Releases to the Atmosphere (cont.)

Date reported and description of incident	Duration of release to the atmosphere	Total U released to atmosphere lbs(kg)	Meets episodic release criteria?
1/10/58. Release of UF_6 in Pilot Plant when H_2 line in reactor column broke and UF_6 backed out. (Klein 1958)	0.75 hr	Probably minor (appears to have been contained).	No
5/10/58. Explosion of D43–104 digestor in Plant 8. (Beers 1958)	Instantaneous	Probably minor (appears to have been contained).	No
6/19/59. Release of hot uranyl nitrate solution from vent of the #212 sparge tank onto the denitration pad, the roadway east of the Refinery and the gravel area east to Plant 4. Approximately 1000 lbs. of uranium lost. (Harr 1959)	Not reported.	Probably insignificant– most washed into storm sewer & river (see Appendix L)	No
12/29/59. Explosion in Digestor Tank 101 in Plant 8 due to hydrogen buildup when vent system plugged up. (Noyes 1960; Beers 1960)	Instantaneous	None reported	No
11/15/60. Dust loss from Dust Collector G20-20 in the Pilot Plant. The dust collector bags were found to have been chemically attacked, presumably by hydrofluoric acid. Two separate, sequential releases occurred - the first when the damaged bag was tied off and the second when all bags were replaced. (Starkey et al. 1960; Killough et al. 1993)	a. 5 or 3 days (one of 2 dates possible for last observation of normal collector operation) b. 3 days	a. 3300(1500) b. 1320(600)	Yes. [See Killough et al. (1993)]
3/27/61. Spill of hot black oxide onto graveled area of Plant 9 from 55-gallon drum containing a 10-gallon drum of the oxide. The bottom of the 55-gallon drum had burned through allowing the black oxide to fall to the gravel. (Brevard 1961)	Not reported.	Probably insignificant– immediately cleaned up and monitored.	No
2/21/62. Remelt furnace explosion in Plant 9. (Starkey 1962)	Instantaneous	Probably none– appears to have been contained	No
10/15/62. Fire in drummed chips stored near the southeast corner of Plant 6. Six 30-gallon drums involved. (NLO 1962)	Not reported.	Not reported.	No. (See calculations in Non-routine Releases.)
10/16/62. Fire in drummed chips stored near the southeast corner of Plant 6. Seventeen 30-gallon drums involved. (NLO 1962)	Not reported.	Not reported.	No. (See calculations in Non-routine Releases.)

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Table K–28. Summary of Incident Reports Involving Potential Uranium Releases to the Atmosphere (cont.)

Date reported and description of incident	Duration of release to the atmosphere	Total U released to atmosphere lbs(kg)	Meets episodic release criteria?
10/17/62. Fire in drummed chips stored near the southeast corner of Plant 6. One 55-gallon drum involved. (NLO 1962)	Not reported.	Not reported.	No. (See calculations in Non-routine Releases.)
10/20/62. Fire in drummed chips stored near the southeast corner of Plant 6. Twelve 55-gallon drums and 16 30-gallon drums involved. (NLO 1962)	Not reported.	Not reported.	No. (See calculations in Non-routine Releases.)
5/27/63. Loss of uranium, in the form of black oxide, to atmosphere from Plant 4 due to dust collector bags not being seated correctly in the tube sheet of dust collector G-4-8. (Martin et. al. 1963)	20 days	939(425)	No. (Included in routine source term.)
6/4/63. Fire in Pilot Plant Pangborn Rotoblast equipment. (Vath 1963)	Not reported.	Minor loss reported.	No
4/7/65. Fire in drummed chips stored on the Plant 6 southeast pad. (Ross 1965)	Not reported.	Not reported.	No. (See calculations in Non-routine Releases.)
2/14/66. UF_6 release from a 10-ton cylinder of uranium hexafluoride in the Pilot Plant when an operator accidentally unscrewed the cylinder valve. (NLO 1966; Boback & Heatherton 1966)	1 hr	1650(750)	Yes
7/25/69. Orange oxide (400 lbs) discharged from the Refinery gulping systems onto the roof of the Denitration Area. Release was cleaned up or released to sewer system; material balance indicates that no material was lost to the atmosphere (i.e., all material accounted for). (Adams 1969)	0.5 hr	None indicated.	No
4/10/70. Depleted sludge fire in Pit #4. (Heatherton 1975)	6 hours	17.17(7.8) (See calculations in Non-routine Releases.)	No

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Table K-28. Summary of Incident Reports Involving Potential Uranium Releases to the Atmosphere (cont.)

Date reported and description of incident	Duration of release to the atmosphere	Total U released to atmosphere lbs(kg)	Meets episodic release criteria?
6/14/78. Dust loss in the Plant 9-NI-1039 dust collector servicing the NPR furnace and the crucible burnout area due to damage to the collector bags. (Adams 1978)	Unknown, but at least 20-35 days	153(70)	No
6/15/81. Dust loss from Plant 4 dust collector G4-14. (Nutter 1981)	32 days	33(15)	No
6/19/81. Dust loss from Plant 4 dust collector G4-14 due to bag failure. (Nutter 1981)	4 days	130(59)	No
6/29/81. Dust loss from Plant 4 dust collector G4-14 due to bag failure. (Nutter 1981)	10 days	25(11)	No
9/8/81. Loss of greensalt from Plant 4 dust collector G4-2 due to bag failure. (Nutter 1981)	10 days	440(200)	No
9/10/81. Additional loss from Plant 4 dust collector G4-2 due to hold-up in sampler and residual material in the collector (see previous episode). (Nutter 1981)	1 day	86(39)	No
12/12/84. Loss from the Stacks of Nos. G5-260 and -261 dust collectors. (Martin et al. 1985)	Unknown	33(15)	No
12/14/84. Dust loss from Plant 9 dust collector G9N1-1039 due to bag failure. (Martin et al. 1985; Adams 1985)	Unknown, probably over a few months	273(124)	No
12/18/84. Stack loss from the Plant 9 Machining precipitron. (Nutter 1984)	2 years	5.7(2.6)	No
2/4/85. Uranium chip fire. (NLO 1985a)	Not reported. Assume 30 minutes	<1(<0.45)	No
2/25/85. Magnesium fluoride and depleted U release inside Plant 5. (NLO 1985b)	Minutes	0.08(0.04)	No
4/12/85. Release of uranium oxide during filter change-out in Plant 8 Dust Collector #8035. (NLO 1985c)	Not reported. Assume minutes.	1.1(0.5)	No
11/1/85. Magnesium flash during the reduction of charge #72221. Considered a minor event. (NLO 1985d)	Instantaneous	Probably not significant	No
12/3/85. Smoke from #46 Rockwell Furnace due magnesium flash of charge #73003. Considered a minor event. (NLO 1985e)	Instantaneous	Probably not significant	No
1/19/86. Cracking of reaction vessel #2 at the Pilot Plant, which released UF_6 to atmosphere. (WMCO 1988a)	Unknown	14.5(6.6)	No

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Table K–28. Summary of Incident Reports Involving Potential Uranium Releases to the Atmosphere (cont.)

Date reported and description of incident	Duration of release to the atmosphere	Total U released to atmosphere lbs(kg)	Meets episodic release criteria?
11/11/86. Spill of 300 lbs of UO_3 from Bank 9 fluid bed reactor system in Plant 4. (DOE 1986)	Minutes	Not significant.	No
12/30/86. Derby on fire in East Break Out area of Plant 5. Considered a minor event. (WMCO 1986)	Minutes	Probably not significant.	No
2/23/87. Spill of green salt in Reduction Area of Plant 5. (WMCO 1987c)	Unknown	Probably none.	No
2/27/87. Fire on grizzley conveyer in area of remelt furnace pot No. 10596 (Plant 5 East Breakout) caused by sparks emitted by derby charge No. 31528. (WMCO 1987d)	Unknown	None indicated.	No
1/15/88. Release of UF_4 through dust collector G-2 at Pilot Plant. (Collier 1988)	Unknown	None indicated.	No
1/18/88. Release of uranyl nitrate from Plant 2/3. Incident was discovered when an area of the Plant 2/3 roof and nearby ground within FMPC was found contaminated with uranyl nitrate. (WMCO 1989)	Unknown	40(18)	No. (Release was contained in the production area.)
2/26/88. Dust release from G-2-239 Hoffman high vacuum system in Plant 8. (WMCO 1988b)	Minutes	None indicated.	No
3/14/88. Depleted UF_4 spill in Plant 4, Depleted Packaging Operations. (WMCO 1988c)	Minutes	None indicated.	No
6/30/88. Release of uranium from UO_3 and gulping operations at Plant 2/3 Refinery. (WMCO 1989)	4 weeks	145(66)	No
10/4/88. Uranium chip fire in Plant 6 involving five drums. (WMCO 1989)	Not reported	None indicated.	No

Monitoring of the Pilot Plant following the 1966 episodic release indicates that most of the contamination occurred in the immediate area of the incident, i.e., the Pilot Plant proper and the north pad of the Pilot Plant. Offsite areas just south of the FMPC were monitored for alpha contamination with hand-held instruments and showed no contamination above instrument background. Milk samples collected offsite were analyzed on 2/15/66 and found to have uranium concentrations well below significant levels and consistent with previous samples. Soil, vegetation, and water samples collected within the FMPC at varying distances south of the Production Area, did not contain any significant concentrations of uranium.

Episodic Releases Identified Using Monitoring Data

In addition to reviewing documents, air monitoring and gummed film data obtained from 1958 through 1984 were evaluated to identify potential episodic releases. An initial screening assessment of air monitoring data indicated that 14 undocumented episodic releases may have occurred during this time period (see Appendix B–Part 2 of Shleien et al. 1993). Further analyses of these data were performed to determine if the apparent releases meet the criteria for implementing special dose assessment procedures.

First, a "baseline" concentration of airborne uranium was estimated for each location and time period of concern. The baseline concentration was defined as the average uranium concentration during the 3-week period before and the 3-week period after the elevated concentration was observed. This is illustrated in Figure K–18. If the elevated concentration was determined to be at least 10 times greater than the baseline concentration, the result was considered for further evaluation. Eight results exceeded the baseline value by this amount, as illustrated in Figures K–18 through K–25.

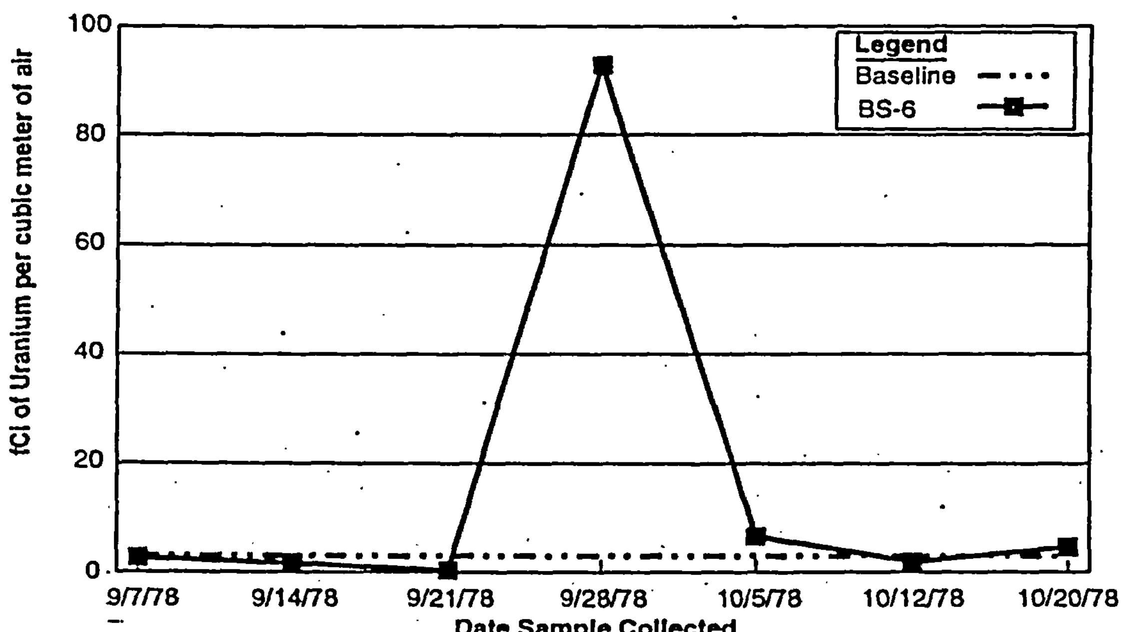

Figure K–18. Uranium in air at BS–6 during the period from 9/7/78 through 10/20/78.

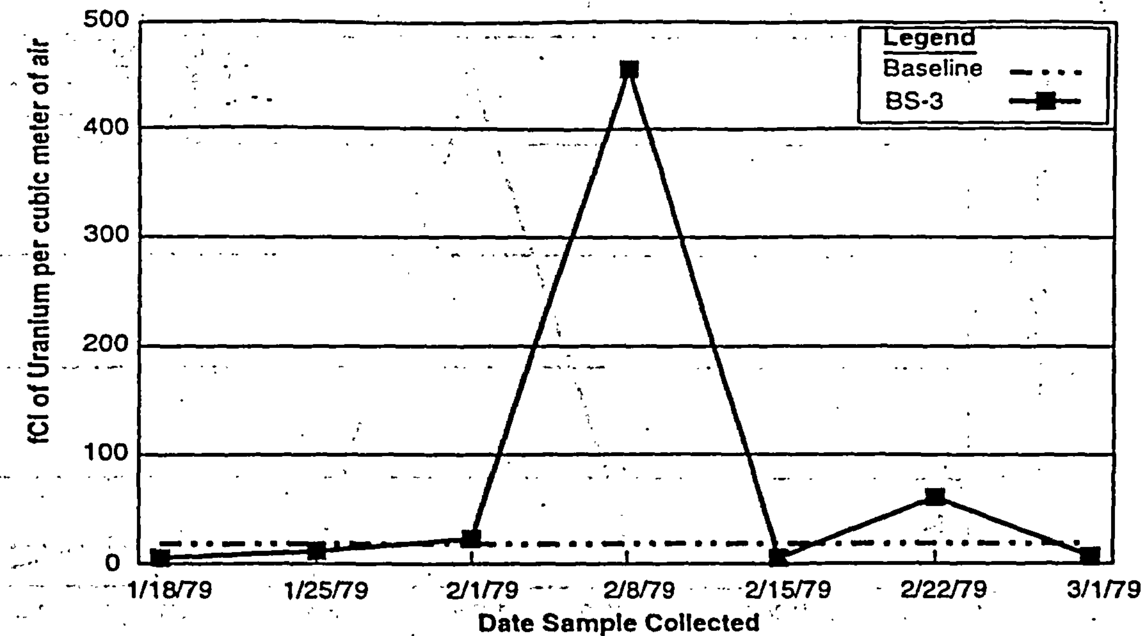

Figure K-19. Uranium in air at BS-3 during the period from 1/18/79 through 3/1/79. (See also Figure B2-20 of the Task 5 report (Shleien et al. 1993).

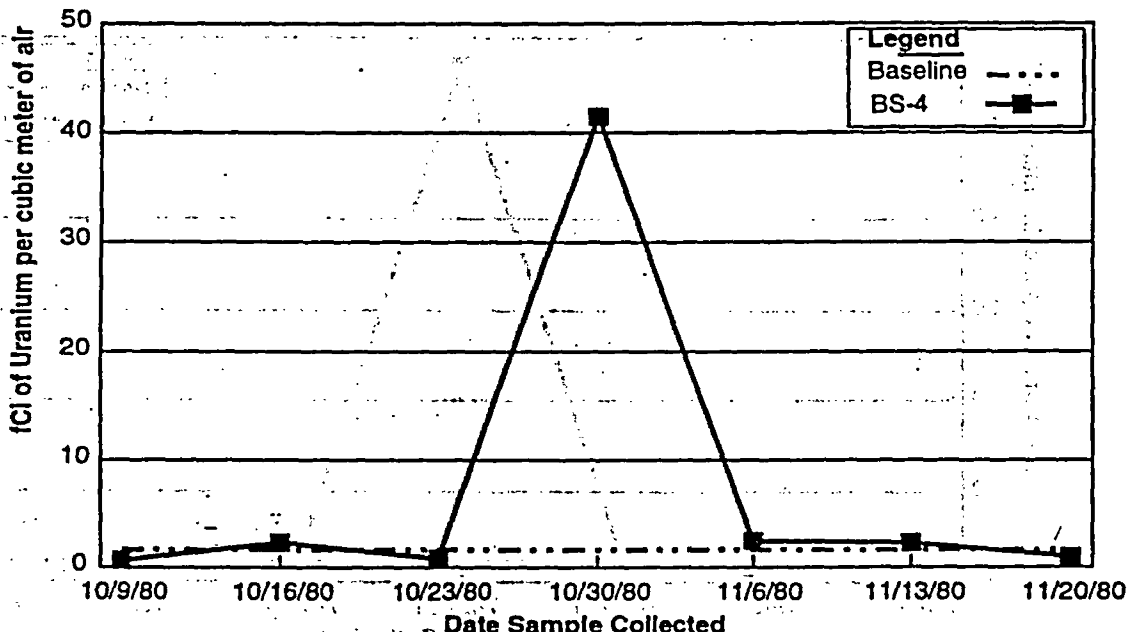

Figure K-20. Uranium in air at BS-4 during the period from 10/9/80 through 11/20/80.

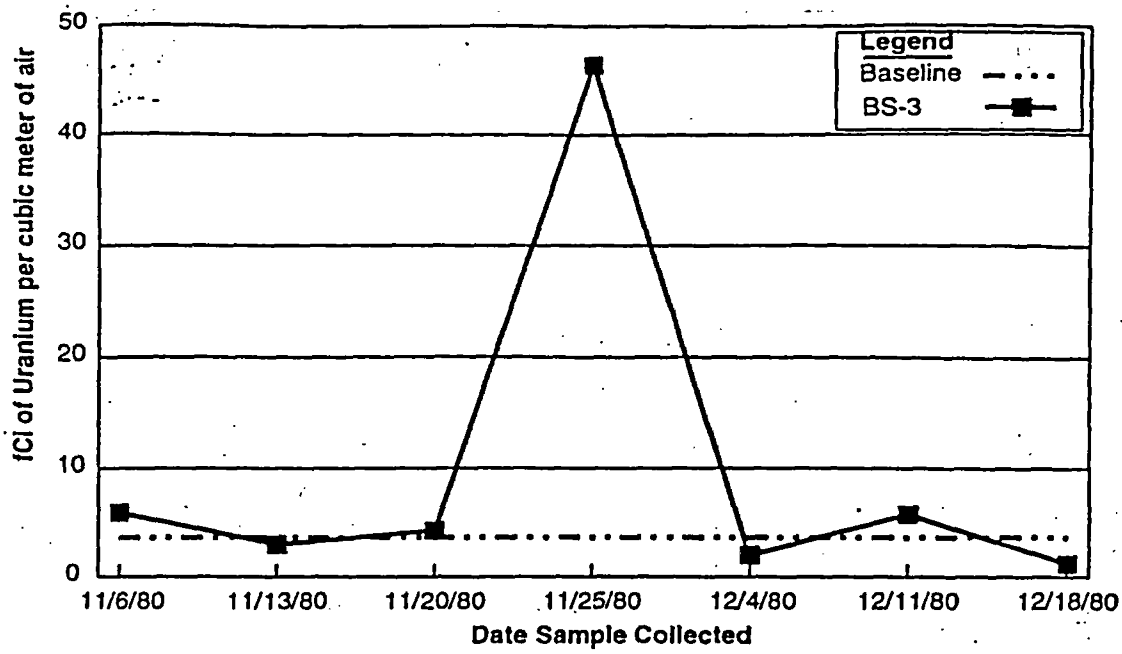

Figure K-21. Uranium in air at BS-3 during the period from 11/6/80 through 12/18/80.

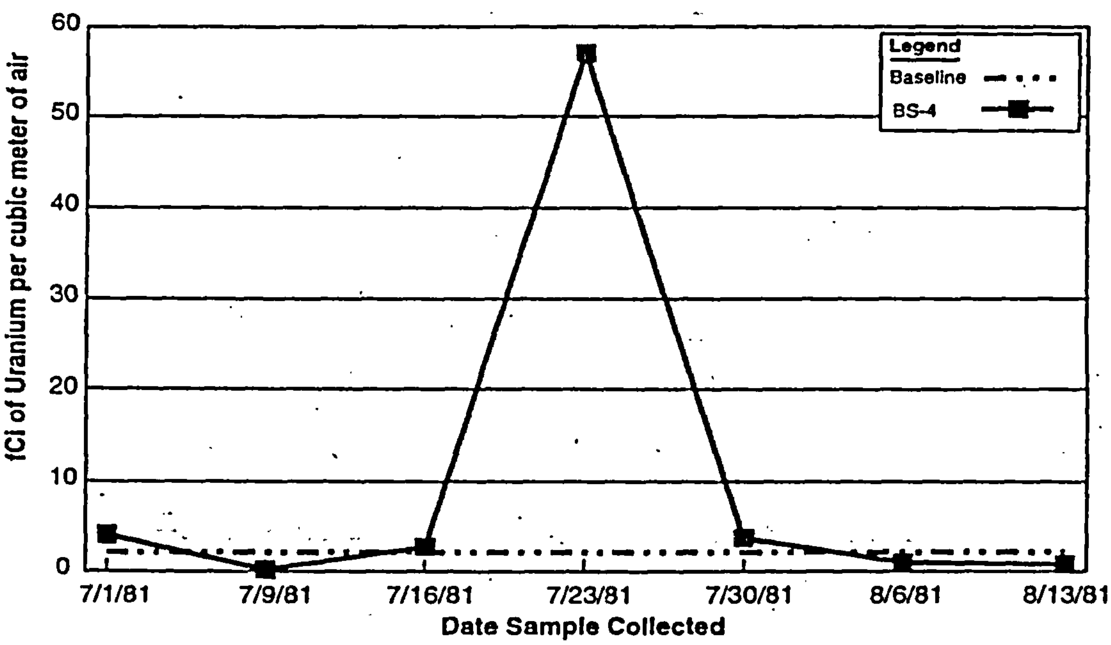

Figure K-22. Uranium in air at BS-4 during the period from 7/1/81 through 8/13/81.

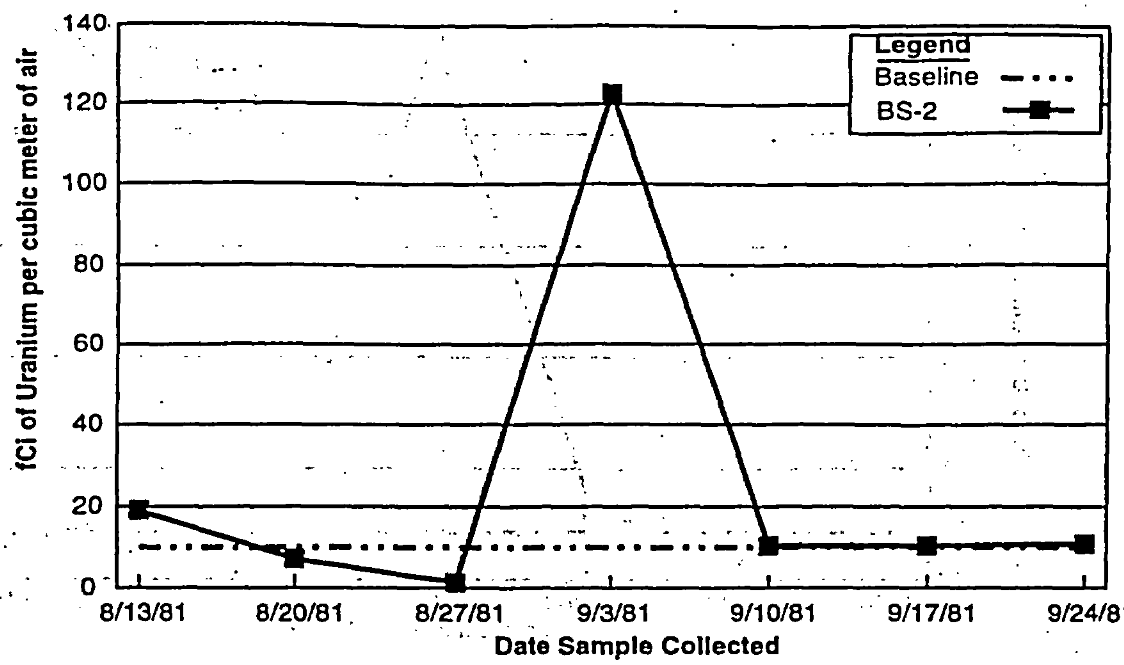

Figure K–23. Uranium in air at BS–2 during the period from 8/13/81 through 9/24/81.

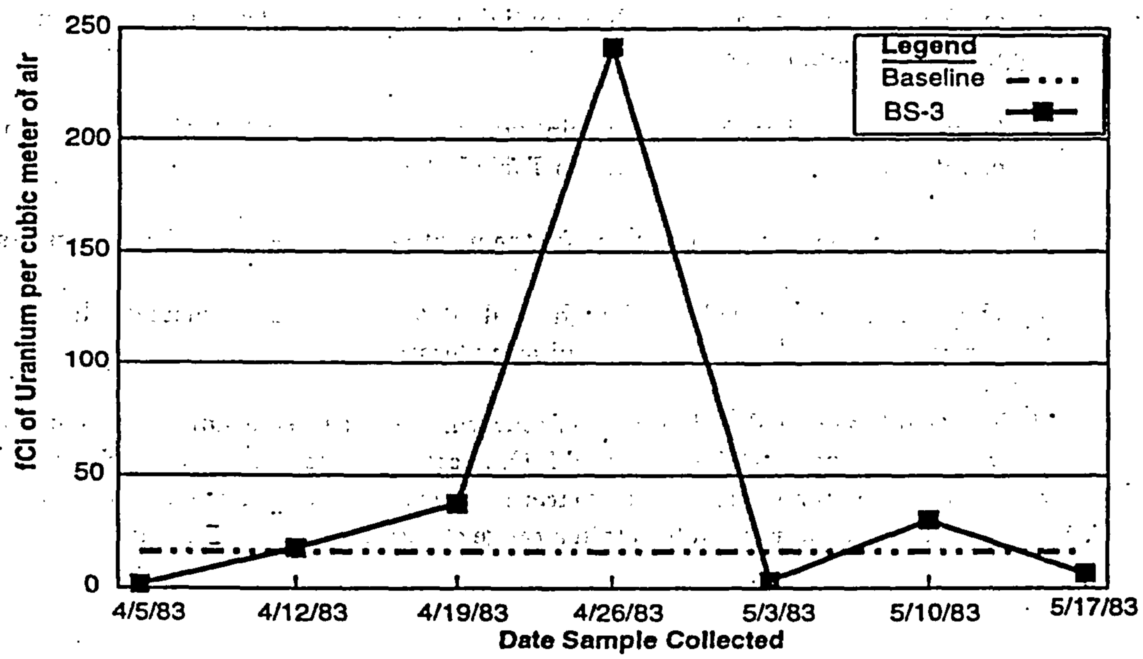

Figure K–24. Uranium in air at BS–3 during the period from 4/5/83 through 5/17/83.

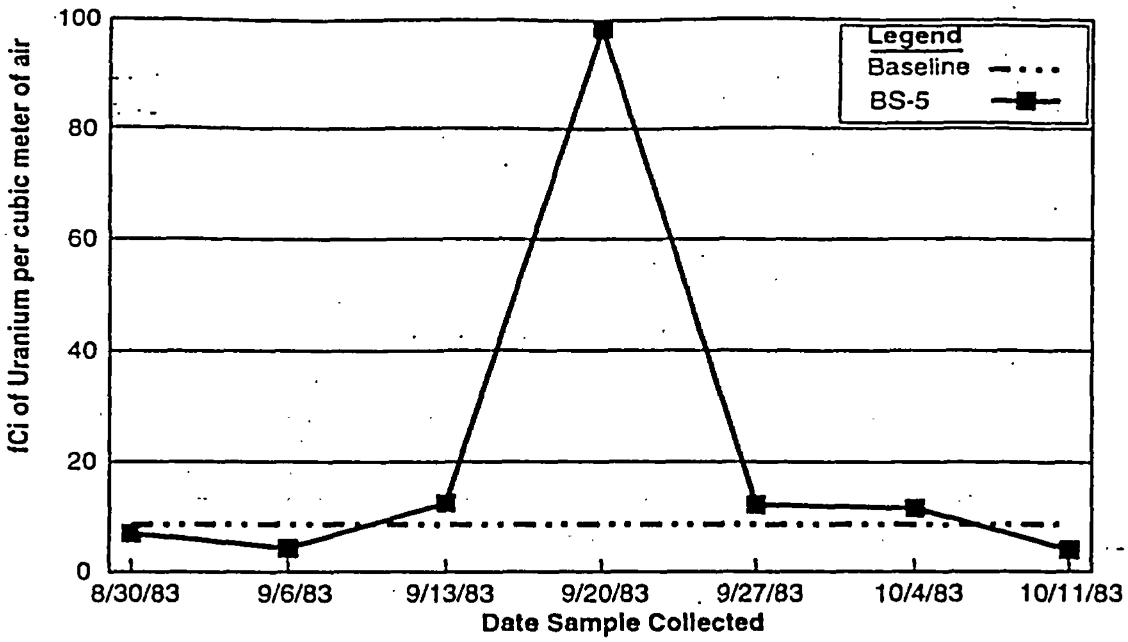

Figure K-25. Uranium in air at BS-5 during the period from 8/30/83 through 10/11/83.

Next, the airborne uranium concentrations were used to estimate the possible source term. The source term for each potential release was calculated using the following approach and assumptions:

1. It was assumed that the release point was the center of the FMPC. Distances to each air monitoring location are presented in Table K-29.

2. Average meteorological conditions for the month of the potential episode were assumed.

3. The building wake model (Killough et al. 1993) was used to calculate dispersion (X/Q) parameters (s m^{-3}) for each air monitoring location.

4. For each time period and location of concern, the measured uranium concentration in air was used, along with the estimated X/Q, to calculate the release rate. (Note: All locations during the time period of concern which showed elevated airborne uranium concentrations were used to obtain a range of possible source term values.)

Results of the calculations are shown in Table K–30. Based on the upper range of estimated source term values, three episodic releases are apparent. They occurred during the weeks ending on September 28, 1978, February 8, 1979, and September 20, 1983. The range of release rate values estimated for each of these episodes are 6–56 kg d^{-1}, 12–100 kg d^{-1}, and 47–57 kg d^{-1}, respectively. The source of these episodic releases is unknown, as supporting documentation could not be found. Gulping of UO_3 in Plant 2/3 is one suspect, as it was identified in 1988 as an unmonitored radioactive emission source (Vaaler and Nuhfer (1989). However, Semones and Sverdrup (1988) indicate that this process was not in operation during the years 1978 and 1979. They estimated a total loss of 130 kg of uranium for the year 1983, which represents an average daily loss over 100 times less than the episodic release calculated here. Other possible unmonitored sources of magnitude observed include the incinerator (through 1979) and Plant 8 scrubbers.

Table K–29. Locations of Air Monitoring Stations

Station	Distance from FMPC center (m)	Direction from FMPC Center
BS–1	1000	N
BS–2	1200	NE
BS–3	730	E
BS–4	1600	SE
BS–5	1200	SW
BS–6	1100	W
BS–7	1600	NW

Non-routine Events

Most of the releases which occurred as a result of accidents at the FMPC did not qualify as episodic releases, using the criteria discussed in the previous section. In order to account for all of these releases, it is necessary to include them in the total annual source term. In addition, providing an estimate and the associated uncertainty for such releases provides limits which ideally encompass all accidental releases, including those for which documentation is no longer available. Clark et al. (1989) included these additional uranium emissions from "non-routine events" over the 37-year history of the FMPC. Details of the analysis can be found in Vaaler and Nuhfer (1989). The investigation involved research of historical site documents to determine the types and frequency of accidents. Estimates of uranium emissions were derived from the historical information and best engineering judgment based on familiarity with plant operations. Four categories of non-routine events were identified: uranium fires, solid spills, UF_6 leaks, and releases of uranyl nitrate. Two non-routine releases (in 1966 and 1984) which were included in Boback et al. (1987) were not duplicated in the addendum report estimates for non-routine events.

Table K–30. Calculation of Episodic Release Rates Using Weekly Air Monitoring Data

Date	Station	Air conc. (fCi m^{-3})	X/Q (10^{-7} s m^{-3})	Release Rate (10^{-7}Ci s^{-1})	Release Rate (kg d^{-1})	Criterion (kg d^{-1})[a]	Criterion exceeded?
9/28/78	BS-2	10	4.6	0.2	5.9	31	No
	BS-3	40	6.4	0.6	16		No
	BS-4	13	3.2	0.4	11		No
	BS-5	47	2.7	1.7	45		Yes
	BS-6	46	2.2	2.1	56		Yes
2/8/79	BS-1	82	2.4	3.5	90	31	Yes
	BS-2	40	8.4	0.5	12		No
	BS-3	228	9.0	2.6	67		Yes
	BS-4	75	4.2	1.8	47		Yes
	BS-5	124	4.8	2.6	68		Yes
	BS-6	81	2.1	3.8	100		Yes
10/30/80	BS-4	21	3.6	0.6	15	33	No
	BS-5	7	0.9	0.8	21		No
11/25/80	BS-1	12	6.6	0.2	4.8	33	No
	BS-3	23	11.0	0.2	5.7		No
	BS-6	7	2.5	0.3	7.2		No
7/23/81	BS-4	29	2.3	1.3	33	54	No
	BS-5	11	1.5	0.7	19		No
9/3/81	BS-1	14	2.8	0.5	13	54	No
	BS-2	61	4.6	1.3	35		No
	BS-7	7	0.8	0.9	24		No
4/26/83	BS-3	121	8.9	1.4	36	50	No
	BS-4	43	4.1	1.1	28		No
	BS-5	14	3.3	0.4	12		No
9/20/83	BS-1	61	2.8	2.2	57	50	Yes
	BS-5	49	2.7	1.8	47		No

[a]Represents ten times the average daily release rate estimated for the year of operation.

The total estimated uranium emissions to the atmosphere from non-routine events between 1952 and 1988 was 2,784 kg (Vaaler and Nuhfer 1989), which is 2% of the approximately 135,000 kg U previously reported by Boback et al. (1987) for 1951–1984. The authors assigned upper limits to each category of non-routine release and state that "the total uncertainty of all the categories results in an additional 60% as an upper limit to the non-routine emission estimate." For the period 1952 through 1988, Vaaler and Nuhfer (1989) estimated releases of 931 kg from uranium fires, 1063 kg from solid spills (outdoor), 518 kg from UF_6 leaks, and 272 kg from liquid uranyl nitrate hexahydrate (UNH) releases, for a total release of 2784 kg over the 37-year period.

A technical review of the Vaaler and Nuhfer (1989) report by the IT Corporation (IT 1989) indicated that many of the calculational methods and assumptions were not adequately documented, and therefore may be inappropriate. In an attempt to derive a range of release rates that encompass the true values and that are defendable, *RAC* evaluated and, if warranted, selected more appropriate models and parameter values. In addition, uncertainty analyses were performed using Monte Carlo simulations with Crystal Ball® software (Decisioneering 1992). The results are discussed in the following sections.

Airborne Emissions from Uranium Fires

Two types of fires, chip fires and a sludge fire, were considered. The chip fire estimate in Vaaler and Nuhfer (1989) involved the use of the following variables: the concentration of uranium in air above the drum containing the burning chips; the affected volume of air; wind speed; and duration of the fire. The calculational methodology is simplistic and very conservative, as it assumed a constant maximum concentration above the fire. IT (1989) observed that only one drum was assumed for each fire, while many of the fires involved multiple drums. In addition, no documentation could be found to support the number of fires used. A random check of five years of fire reports by IT (1989) showed an average underestimate of 15% by Vaaler and Nuhfer. The underestimate could be explained primarily by the fact that events other than uranium fires were included in the fire report tallies. However, the derivation of the number of fires was not documented, and there is thus some uncertainty associated with the values used. Finally, IT (1989) noted that the uranium concentration in air above the burning drum was based on one measurement of >100,000 dpm m^{-3}. The concentration used in the calculation (100,000 dpm m^{-3}) may or may not bound the true value. In addition, *RAC* observed that the mean wind speed (9.1 mph) used was for Cincinnati, not FMPC, and that the duration of the fire (30 minutes) was not necessarily representative of the fires described in the documents reviewed.

The model used to calculate the chip fire emissions was considered to be inappropriate. Fortunately, the results of the U. S. Atomic Energy Commission (AEC) Health and Safety Laboratory (HASL) laboratory and field tests, which involved burning uranium chips, were available (Weinstein and Breslin 1959). Although the results were never published, they are very appropriate to this problem. In the laboratory, chips ranging in quantity from 20 g to 1 kg were burned on a wire screen bed in a combustion chamber. The field experiments conducted included the measurement of emissions from burning natural uranium chips contained in a 30-gallon drum. The 30-gallon drum was housed in a 55-gallon drum covered by a 12-ft stack. Both drums were provided with multiple air holes drilled near the bottom to provide a natural draft. Finally, 900 lbs (408 kg) of depleted uranium were burned in a line array of open drums. The results of the laboratory and field experiments were plotted in Weinstein and Breslin (1959). The original data were not reported in the text; however, they were shown in the plot. Because the curve fitting method was not documented, a linear regression of the experimental results was performed by *RAC*. A correlation coefficient of 0.88 was calculated, indicating a fairly good fit of the line with the data. The original data and the resulting plot are shown in Figure K–26.

The annual release of uranium from chip fires was calculated by taking the product of the mass of uranium in drums, the fraction of uranium released in the fire, the number of drums involved, and the number of fires per year. The simulation was performed using Microsoft Excel and Crystal Ball.

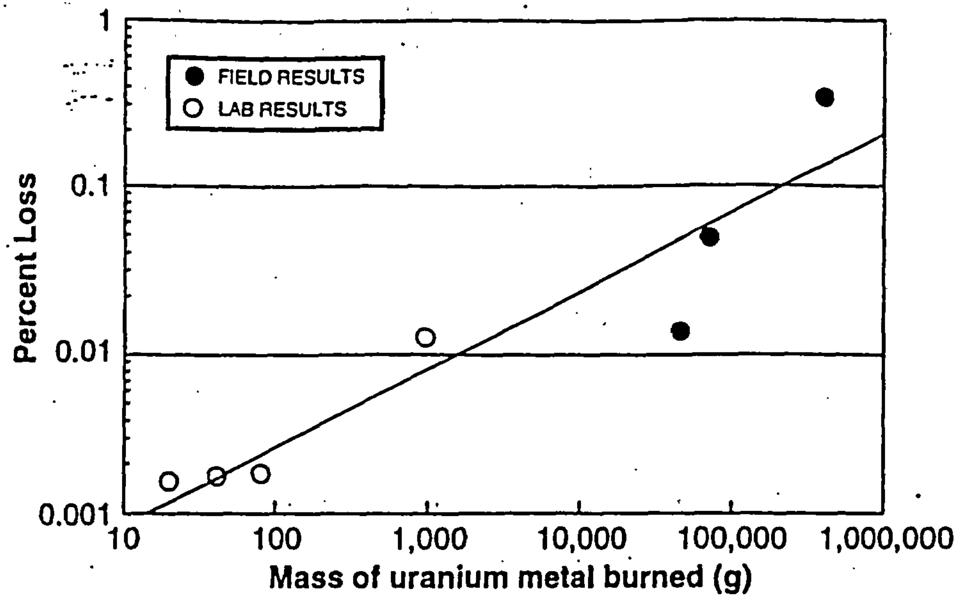

Figure K–26. Fume loss from burning uranium metal [derived from data in Weinstein and Breslin (1959)].

The mass of uranium contained in 30-gallon and 55-gallon drums was assumed to be represented by a range of values (a linear distribution). The experiments conducted by Weinstein and Breslin (1959) involved igniting 100 lbs (45 kg) and 160 lbs (73 kg) of natural uranium chips in a 30-gallon drum. It was thus assumed that the minimum mass that could be involved in a fire was 100 lbs (45 kg). The maximum mass was assumed to be 132 kg, which represents the maximum volume in the 30-gallon drum scaled up to a 55-gallon drum.

The percent loss of uranium from burning uranium metal was assumed to range from 0.05 to 0.15 (a linear distribution). These fractions were derived from Figure K–26 and correspond with the minimum mass (45 kg) of uranium used in the individual drum experiments and the mass (408 kg) of uranium burned in the line array of open drums. It was felt that the latter test best represents the fires involving multiple drums. A simplifying assumption implied by the use of these factors is that fires were allowed to burn to completion, as they were in the experiments. In fact, most fires were extinguished within 30 minutes.

It was assumed that the number of drums involved range from 1 to 56 (the minimum and maximum documented in incident reports). Most of the reports show that 1 to 6 drums were typically involved. (Unusually higher numbers of drums were involved during one 5-day time period in October 1962 when a large number of drums were temporarily stored on a pad near Plant 6.) Thus, a lognormal distribution, with a mean of 3.5 and a maximum value of 56 was constructed to represent the number of drums.

A triangular distribution was used to represent the number of fires occurring per year. The most probable number of fires used were those presented in Table 1 of Vaaler and

Nuhfer (1989). The minimum and maximum values were assigned values equal to the mean value ± 15%.

Using a Monte Carlo simulation, 500 runs were made to determine the median and the 5th to 95th percentile range. The results are shown in Table K–31. The 50th percentile results are approximately equal to those calculated by Vaaler and Nuhfer (1989). However, the upper 95th percentile estimate is about 500% of the median. This greatly exceeds the 60% upper bound estimated by Vaaler and Nuhfer (1989).

Table K–31. Uranium Chip Fire Emissions

Year	Most Probable Number of fires	Uranium released (kg y^{-1}) 50%	5%	95%
1952–1961	100	16	1.9	119
1962	76	13	1.4	94
1963	161	26	3.0	209
1964	135	23	2.6	169
1965	131	22	254	168
1966	102	17	1.9	128
1967	98	16	1.8	119
1968	64	9.5	1.2	65
1969	73	12	1.3	93
1970	68	11	1.2	84
1971	20	3.3	0.4	25
1972	17	2.8	0.3	21
1973	4	0.6	.07	5.0
1974	0	0	0	0
1975	6	1.1	.11	7.3
1976	1	0.2	.02	1.3
1977	3	0.5	.005	3.9
1978	6	1.0	.11	7.5
1979	1	0.2	.02	1.3
1980	4	0.7	.07	5.1
1981	7	1.1	0.1	8.4
1982	18	2.9	0.3	23
1983	17	2.8	0.3	21
1984	16	2.7	0.3	19
1985	20	2.7	0.4	20
1986	16	2.6	0.3	19
1987	16	2.6	0.3	19
1988	3	0.5	.005	3.9
TOTAL		450	92	2260

The metal sludge fire, which occurred in April 1970, was estimated by Vaaler and Nuhfer (1989) to have released 120 kg of uranium. The model used was the one used to calculate emissions from chip fires, which was based on one measurement made above a burning drum of uranium chips. However, the affected volume was assumed to be very large, encompassing an area of 6.5 m^2, and extending 7.6 m vertically above the sludge. The assumption of constant activity within this volume was thought to be very conservative.

Alternatively, we used two measurements obtained during the course of the sludge fire (Heatherton 1970). The measurements were made 2000 ft (610 m) downwind of the fire. The results were 7 $\mu g\ m^{-3}$ and 10 $\mu g\ m^{-3}$.

The building wake model (Killough et al. 1993) was used to estimate the average dispersion coefficient at a distance of 610 m during the month of April. The average dispersion coefficient was calculated to be 1.1×10^{-5} s m^{-3}. A conservative bound was selected by first examining the individual dispersion coefficients calculated for each wind direction. The lowest value, which would result in the highest source term estimate, was 2.0×10^{-7} s m^{-3}. A lognormal distribution was constructed with the geometric average of 1.1×10^{-5} s m^{-3} and a lower limit of 2.0×10^{-7} s m^{-3}. The measured downwind concentration was assumed to be represented by a normal distribution of values, with a mean of 8.4 $\mu g\ m^{-3}$ and a standard deviation of 2.1 $\mu g\ m^{-3}$. The fire was reported to have lasted for six hours. Using Crystal Ball, the median source term value was estimated to be 30 kg, with a lower 5% confidence limit of 4.8 kg and an upper 95% confidence limit of 223 kg. This range encompasses the 120 kg reported in Vaaler and Nuhfer (1989).

Solid Spills

In regard to the solid spill calculations, IT (1989) state that "there is lack of clear support documentation for the assumptions used (the average wind speed used, the assumed silt content of the spilled material, the average mass of material spilled per incident, the moisture of the spilled material)." This is compounded by the fact that Vaaler and Nuhfer (1989) tried to categorize all spilled materials as either those with high uranium content and those with low uranium content. The physical characteristics of the different materials involved most likely vary greatly.

A more serious error was also discovered when examining the model used. The algorithm presented in Vaaler and Nuhfer (1989) is:

$$EF = 0.0018\,(s/5)(u/5) + [(m/2)^2(L/6)] \qquad \text{(K-1)}$$

where:

EF = emission factor (lb material airborne/ton spilled)
s = material silt content (%)
u = mean wind speed (mph)
m = moisture content (%)
L = loader capacity (yd^3)

Upon further examination, it was discovered that this algorithm was derived from the EPA document entitled "Compilation of Air Pollution Emission Factors," AP-42 (EPA 1985). This algorithm originally appeared in the section entitled "Aggregate Handling and Storage Piles" in 1983 and was revised in 1988. The original equation, as it appeared in 1983, was:

$$EF = k(0.0018)(s/5)(u/5)(H/5) + [(m/2)^2(Y/6)^{0.33}] \qquad \text{(K-2)}$$

where:

k = particle size multiplier

H = material drop height (ft)
Y = dumping device capacity (yd^3)

The equation was intended to describe a batch drop operation, where aggregate material is added to or removed from an aggregate storage pile using a truck or front-end loader. The algorithm used in Vaaler and Nuhfer (1989) ignores the particle size multiplier and material drop height, which makes their results more conservative. The exponent associated with the "Y/6" parameter was also ignored. This makes the model extremely conservative. It is also questionable as to whether or not the algorithm, even if properly transcribed, is appropriate to small spills and subsequent cleanup. The user is cautioned in AP–42 that the quality of the model is reduced if the source conditions used in developing the equation are not met. Those conditions include a material silt content of from 1.3–7.3%, moisture content of from 0.25–0.7%, and dumping capacity of from 2.1–7.6%. The silt content of the FMPC material greatly exceeds the range of recommended values and the loading capacities used are far less than the recommended values. The 1988 version of "Aggregate Handling and Storage Piles" no longer includes this equation, but rather has replaced it with a modified version of the continuous drop operation equation found in the 1983 version:

$$EF = k(0.0032)(u/5)^{1.3}+(m/2)^{1.4} \qquad \text{(K–3)}$$

This equation is recommended for both drop batch and continuous drop operations. EPA also assigns a higher quality rating to this equation than it did to the original equations. The quality rating is retained if the following source conditions are met: 1) silt content ranges from 0.44 to 19%; 2) moisture content ranges from 0.25–4.8%; and 3) wind speed ranges from 1.3–15 mph. Again, the FMPC material exceeds the silt content conditions.

Model bias between equations K–2 and K–3 was tested using Microsoft Excel and Crystal Ball. Table K–32 presents the values used in the simulations. For the sake of simplicity, and lacking specific particle size distribution data, the particle size multiplier (k) was not used. This makes the results more conservative by, at most, a factor of two. Similarly, the drop height in equation K–2 was not used, making the results more conservative by, at most, an additional factor of two.

The mean (geometric) emission factor calculated using equation K–2 is 4.6 g kg^{-1}, with a 5th–95th percentile range of from 0.3 to 29 g kg^{-1}. In contrast, the mean (geometric) emission factor calculated using equation K–3 is 0.3 g kg^{-1}, with a 5th–95th percentile range of from 0.03 to 1.5 g kg^{-1}. The bias introduced by the model selected is thus considerable. Although equation K–3 is the most recent model recommended by the EPA, it appears that the original batch drop model is more appropriate to the spill scenario since it includes a factor to account for the size of the spill and cleanup operation. Neither model was intended for small spill scenarios; however, the results of the calculations using the batch drop equation seem reasonable — it estimates that as much as 3% of the material spilled could become airborne.

Using the parameter values shown in Table K–32, and the number of spills found in Vaaler and Nuhfer (1989), it was estimated that a median annual release of 2 kg of uranium occurred during the years from 1953 through 1969. The 5th to 95th percentile range is from 0.3 to 35 kg y^{-1}. This interval does not encompass the calculated emission of 57 kg y^{-1}

reported in Vaaler and Nuhfer (1989). The emissions from spills in later years were estimated to be insignificant (a median value of 0.25 kg y^{-1}).

Table K–32. Parameter Values Used in Estimating Spill Emissions

Parameter	Distribution	Mean/Min	Std. Dev./Max	Comments
u (m/s)	Lognormal	2.14	1.43	FMPC met data
m (%)	Uniform (range)	0.1	1.0	Assume material is very dry.
s (%)	Uniform (range)	50	99	Material varies, but Vaaler and Nuhfer indicate that as little as 35–70% and as much as 99% of the material is silt.
Loader capacity–spill (m^3)	Lognormal	0.2	0.38	Most likely spill is 55-gal drum. Largest spill was equivalent to 18.4 drums.
Loader capacity–cleanup (m^3)	Uniform (range)	0.005	0.01	Smallest cleanup device is a shovel. Largest is that used in Vaaler and Nuhfer (1989).

UF_6 Releases

IT (1989) found little documentation to support the UF_6 emission estimates made in Vaaler and Nuhfer (1989). Most notably, information on the number of releases and the magnitude of the releases is lacking. We could not find documentation to add any new insights to these estimates. For this reason, the methods used by Vaaler and Nuhfer (1989) were considered to be adequate, although probably conservative.

Vaaler and Nuhfer (1989) note that samplers located throughout the UF_6 process area indicate low levels of airborne radioactive material. Based on these data, the emission to atmosphere from UF_6 leaks or releases to the building from 1980–1988 is considered to be very small. The only event found in documentation for which a reasonable quantitative amount (6 kg) was determined was the 1986 vessel crack (WMCO 1988a).

More frequent releases were indicated during the 1950s and 1960s, when a cold trap system did not exist to remove residual UF_6 from the process piping. When connections were broken for maintenance or cylinder changes (pigtail connection), a portable Hoffman vacuum was used to reduce the quantity of UF_6 released.

Vaaler and Nuhfer (1989) estimated the release of UF_6 from pigtail and maintenance operations prior to 1980 to be approximately 0.4 kg y^{-1}. This was estimated assuming an average annual production rate for this period. The number of pigtail connections and subsequent releases to the building are proportional to the production rate. The number of maintenance operations were assumed to be 52 times per year. The amount of release that occurred during pigtail and maintenance operations was related to the pigtail and pipe volumes. All of the UF_6 was assumed to react with moisture in air to form solid UO_2F_2 and HF). The Hoffman vacuum was then assumed to withdraw 70% of the release (of which 99% was assumed to retained.) Ten percent of the amount entering the building was assumed to enter the atmosphere via exhaust systems.

We estimated the UF_6 leaks due to pigtail and maintenance operations using the equations described in Vaaler and Nuhfer (1989), which model the UF_6 leaks as a function of UF_4 production. However, to provide some estimate of uncertainty, parameters were arbitrarily varied according to Table K–33. In addition, UF_4 production data for the years 1953 through 1967 (see Appendix C) were used in place of the average annual values used by Vaaler and Nuhfer (1989). [Note that the former authors indicate no UF_6 releases during the years 1958 through 1961. However, based on production data (Appendix C), we estimated UF_6 releases for all years from 1953 through 1967, with the exception of 1957.]

Table K–33. Parameter Values Used in Estimating UF_6 Releases from Pigtail and Maintenance Operations

Parameter	Distribution	Minimum	Maximum	Comments
Maintenance/y	Uniform	48	52	Plant not always operating
% of release entering Hoffman	Uniform	50	70	Efficiency unknown. Assume value reported in Vahler and Nuhfer (1989) is maximum value.
% leaving building	Uniform	10	20	Unknown. Assume value reported in Vahler and Nuhfer (1989) is minimum value.
% not retained by Hoffman	Uniform	0.01	0.10	Unknown. Assume value reported in Vahler and Nuhfer (1989) is minimum value.
Pipe volume (cc)	Uniform	111	136	Vary value estimated in Vahler and Nuhfer (1989) by ±10%.
Pigtail volume (cc)	Uniform	408	498	Vary value estimated in Vahler and Nuhfer (1989) by ±10%.

Releases from pigtail and maintenance operations were estimated to be minor, amounting to a less than 1 kg of uranium per year during the years prior to 1968. The highest value was estimated for 1964, the highest year for production. A 50th percentile value of 0.8 kg y^{-1} (with a 5th to 95th percentile range of possible values of from 0.6 to 1.2 kg y^{-1}) was estimated for that year. This is a factor of two higher than the Vaaler and Nuhfer (1989) estimate of 0.4 kg y^{-1}.

UF_6 leaks from vessel cracks were also varied to provide bounding ranges for the years 1953 through 1956, and 1958 through 1967. The methods of Vaaler and Nuhfer (1989) were used. It was assumed that the emissions from vessel cracks ranged from 4 to 6 kg, with a maximum value corresponding to that measured during the 1986 vessel crack (WMCO 1988a). It was further assumed that from 2 to 6 cracks occur per year [Vaaler and Nuhfer (1989) assumed an average of 4 cracks per year before 1980]. The results show a median value of 20 kg of U released per year [approximately the value calculated by Vaaler and Nuhfer (1989)], with a 5th to 95th percentile range of from 11 to 31 kg y^{-1} for the years prior to 1968.

No vessel cracks were modelled after 1980, with the exception of the 1986 event. The 6 kg release reported for the 1986 vessel crack was assumed to be a good estimate. To calculate the total UF_6 released during that year from all non-routine events, however, we assumed a triangular distribution with 6 kg as the likliest value, 5.1 kg as the minimum value, and 6.9 as the maximum value (i.e. 6 kg ± 15%).

Miscellaneous UF_6 releases were calculated by Vaaler and Nuhfer (1989) by assuming that an average of 22.5 kg of UF_6 (15.4 kg U) was released per month inside the building, and that 10% of the material exits the building as UO_2F_2. Lacking any other data, it was assumed that from 10 to 30 kg of UF_6 was released per month inside the building. Furthermore, it was assumed that anywhere from 5 to 15% of the material exits the building. The results ranged from 11 to 31 kg U y^{-1} (5% to 95%), with a 50% value of 20 kg U y^{-1}. Miscellaneous releases after 1980 were determined by Vaaler and Nuhfer (1989), based on data from samplers in the UF_6 process area and on incident reports, to be very small. They arbitrarily assigned an annual emission rate of 2 kg per year after 1980. We assumed a uniform distribution of 1 to 2 kg per year.

Vaaler and Nuhfer (1989) also estimated uranyl nitrate (UNH) releases. Although considered to be rare [only two documented cases were found by Vaaler and Nuhfer (1989)], the authors assumed that six incidents occurred over the operating history. One of the documented cases occurred in 1959 and involved a release of 454 kg. The second occurred in 1988 and released 18 kg of UNH. Nevertheless, Vaaler and Nuhfer (1989) assumed that each event released 454 kg of UNH. A careful examination of data from the 1988 incident cleanup led them to assume that only 10% of the UNH becomes airborne. Thus, a total of 272 kg of UNH was estimated to be released to the air during the entire operating period. Rather than assuming the entire release, we used the original data reported in Vaaler and Nuhfer (1989) for the two documented incidents (note we could not locate these reports). We then varied the airborne fraction from 0.1 to 0.25.

Summary of Non-routine Releases

Non-routine releases are summarized in Table K-34. A total release of 1300 kg for the entire operating period was estimated. Leaks of UF_6 account for the majority of the total quantity of estimated non-routine releases. However, this comparatively large value is a function of the lack of information concerning the miscellaneous leaks and subsequent releases from buildings and should be considered to be very conservative.

Table K–34. Estimated Emissions Due to Non-Routine Events (kg)[a]

Year	U Fires	Solid Spills (Outdoors)	UF_6 Leaks[c]	Concentrated Liquid UNH Releases
1952	16		44	
1953	16	2.1	44	
1954	16	2.1	44	
1955	16	2.1	44	
1956	16	2.1	44	
1957	16	2.1	0	
1958	16	2.1	44	
1959	16	2.1	45	81
1960	16	2.1	45	
1961	16	2.1	45	
1962	13	2.1	45	
1963	26	2.1	45	
1964	23	2.1	45	
1965	22	2.1	45	
1966	17	2.1	45	
1967	16	2.1	44	
1968	11	2.1		
1969	12	2.1		
1970[b]	48	0.6		
1971	3.3	0.6		
1972	2.8	0.6		
1973	0.6	0.6		
1974	0	0.6		
1975	1.0	0.6		
1976	0.2	0.6		
1977	0.5	0.6		
1978	1.0	0.6		
1979	0.2	0.6		
1980	0.7	0.6		
1981	1.1	0.6		
1982	2.9	0.6		
1983	2.8	0.6		
1984	2.7	0.6		
1985	3.3	<0.1	1.5	
1986	2.7	<0.1	7.5	
1987	2.6	<0.1	1.5	
1988	0.5	<0.1	1.5	3.2
TOTAL	446	31	676	84[d]
5%	92	5	413	50
95%	2260	635	995	115

[a]Median values.
[b]Includes sludge fire.
[c]Blanks represent years where releases were not expected, as discussed in Vaaler and Nuhfer (1989). Data for years 1985 through 1988 are based on building air sampler data and cold trap incident reported in Vaaler and Nuhfer (1989).
[d] Data from Vaaler and Nuhfer (1989). Original reports could not be found to verify.

EMISSIONS FROM WASTE PITS

A series of waste disposal pits has been used for storage of low-level radioactive wastes during the course of the operations at the FMPC. These pits were located near the western boundary of the site, close to Paddy's Run Creek (Figure K-1). The waste pits consist of waste pit numbers 1 through 6, the burn pit and the clearwell. The waste pits are numbered chronologically in order of construction. The pits also are typically referred to as "wet" if they received waste via pipes in slurry form or "dry" if they received solid waste from trucks. General characteristics of the waste disposal pits are summarized in Table K-35.

Table K-35. Characteristics of FMPC Waste Pits[a]

Pit Number and Type	Lining	Volume (yd^3)	Maximum Depth (ft)	Period of Use	Status
Pit 1 Dry	Clay from burn pit	40,000	17	1952–1959	Retired, covered with topsoil
Pit 2 Dry	Compacted clay	13,000	13	1957–1964	Retired, covered with topsoil
Pit 3 Wet	Compacted clay	227,000	27	1959–1968 (wet mode) 1975–1977 (dry mode)	Retired, covered with topsoil
Pit 4 Dry	Compacted clay	53,000	24	1960–1986	Retired, covered with clay and synthetic cover
Pit 5 Wet	Rubberized elastomeric membrane	102,500	30	1968–1983	Retired
Pit 6 Dry	Elastomeric membrane	9,000	24	1979–1985	Inactive, 75% full
Burn Pit Dry	Natural clay	unknown	unknown	1957–1986	Retired, covered with topsoil
Clearwell Wet	Clay	unknown	unknown	1959–1987	Inactive

[a]Updated from Solow and Phoenix (1987). See Table K-36 for estimated amounts of uranium in waste pits.

Types of waste sent to the dry pits include waste filter cakes, graphite, brick scrap, sump liquor and cakes, depleted slag, process residues, trailer cakes, nonburnable trash, asbestos, barium chloride, slag leach slurry, and lime sludge. Wet pits received lime neutralized raffinate concentrate, slag leach residues, filter cakes, fly ash, depleted slag, scrap green salt, process residues, and filter cakes (Solow and Phoenix 1987). The burn pit is discussed further in the following section on incineration of FMPC wastes. The clearwell received surface runoff from the waste pit area and was used until March 1987 as a final settling basin prior to discharge of liquids to the Great Miami river through manhole 175 (see Appendix L).

Methodology Used to Estimate Releases from Waste Pits in Clark et al. (1989).

Historical emissions of uranium and thorium from fugitive dust from the FMPC waste pits were estimated (Clark et al. 1989) in accordance with methods recommended by the U.S. Environmental Protection Agency (EPA) for hazardous waste sites (EPA 1987) and the Ohio EPA (OEPA 1980). These methods provide equations for estimating fugitive dust releases from waste pits during:

- load-in of material into the area
- wind erosion of the waste material
- load-out of material from the waste pit

In the calculation of fugitive dust emissions from the FMPC waste pits, only the first two categories were judged to contribute significantly to the overall generation of dust emissions, because vehicular movement was minimal in the pits themselves, and no load-out of waste from the pits has occurred (Hill and Dolan 1988).

The load-in contribution to fugitive emissions was calculated only for pits receiving waste in a dry form (Pits 1, 2, 4 and 6), not for those that received waste in a wet form (Pits 3 and 5). Parameters used in the load-in calculation (OEPA 1980) include:

- silt content of the stored waste material
- moisture content of the stored material
- mean wind speed
- effective loader capacity

Data were not available for the amount of material placed in each pit for each year. Therefore, a total emission estimate for the load-in operation for the four dry pits was done over their entire operating history. Parameter values and an example calculation are given in Kispert (1988).

Estimates of the emission rate due to wind erosion depend upon the size of the contaminated area and local meteorological conditions (EPA 1987). The calculation considers the area of the waste pit that is exposed to winds that exceed 12 mph, and the number of days per year when rainfall is $\leq$0.01 inch. The percentage of time that the ground wind speed exceeded 12 mph (9.7%) was determined from FMPC meteorological records for 1987. The number of dry days per year for the Cincinnati area was estimated to be 236, from OEPA (1980). The silt content for all pits except Pit 4 was assumed to be 10% (Kispert 1988). Because of the many massive forms (e.g., drums, concrete, and graphite crucibles) deposited in Pit 4, the silt content was assumed to be much lower (1.5%) (Kispert 1988).

Operational records and historical photographs were used to determine the pit surface area that was exposed and subject to wind erosion each year. In years when the pit surface was covered completely with liquid, the surface area subject to wind erosion was assumed to be zero. When uncertainties arose regarding the amount of waste area exposed to erosion, the larger rather than the smaller area was chosen. The intent of the authors was to ensure that an *underestimate* of fugitive dust emissions would not occur (Hill and Dolan 1988). Parameter values and an example calculation are given in Kispert (1988). Over a 35-year period, the calculated wind erosion component to fugitive dust emissions contributed over 99% of the total estimate (i.e., the dust generated during load-in operations was <1% of the total).

An important simplifying assumption for both the load-in and wind erosion calculations was that the mass concentration of the uranium in the waste material in each pit was uniform and was calculated by the ratio of the total mass of uranium to the total mass of the material placed in each waste pit. Therefore, the concentration of waste material placed in each pit was assumed to be homogeneous and constant over time. The total quantities of uranium and material placed in each pit were obtained from records of the FMPC Nuclear Materials Control and Accountability Group (Kispert 1988). Although only Pit 1 values are given in Kispert (1988), the data from Poff et al. (1985), shown in Table K-36, illustrate the large variation in the calculated average concentration of uranium in the FMPC waste pits.

Table K-36. Variation in Estimate of Uranium and Thorium Concentrations in FMPC Waste Pits[a]

Pit	Waste Quantity (metric tons)	Uranium (kg)	% ^{235}U	Thorium (kg)	Calculated Average Conc. $(g\ U\ g^{-1}$ waste)	$(g\ Th\ g^{-1}$ waste)
1	40,500	52,000	0.71	400	1.3×10^{-3}	7.7×10^{-6} [b]
2	13,000	1,206,000	0.21	Unknown	9.3×10^{-2}	– [b]
3	255,000	129,000	0.78	488	5.1×10^{-4}	1.9×10^{-6}
4	64,967	3,048,087	0.18	61,800	4.7×10^{-2}	9.5×10^{-4}
5	88,213	50,309	0.83	17,000	5.7×10^{-4}	1.9×10^{-4}
6	9,309	843,142	0.21	Unknown	9.1×10^{-2}	–

[a]From Poff et al. 1985, with the exception noted in b. Represents waste stored through 12/31/84.
[b]Data from Rathgens (1974).

Estimated total uranium emissions were 29 kg, 892 kg, 41 kg, 395 kg, 15 kg, and 187 kg, for Pits 1, 2, 3, 4, 5, and 6, respectively. These fugitive emissions (1559 kg over the 36-year period) are approximately 0.03% of the total 5,000 MT discarded to the waste pits during 1953 through 1988.

Thorium emissions from wind erosion were calculated using the same method as described for uranium. A minor amount of thorium (85 kg) was estimated to be released as fugitive emissions from the waste pits during 1953 through 1988.

The uncertainty in the total quantities shown in Table K-36 and the simplifying assumptions about waste homogeneity are important limitations of the analysis of fugitive dust emissions. In addition, the EPA's own analysis of their calculation technique (EPA 1987) indicated that fugitive emission estimates are good only to an order of magnitude. The results presented in Clark et al. (1989) represented their best approximation, given the data available.

Evaluation of Method Used to Estimate Releases from Waste Pits in Clark et al. (1989)

The methods presented by the EPA for estimating fugitive dust releases from hazardous waste sites (EPA 1987) fall into three categories, according to the geometry of the source: line models (e.g. contaminated roads), area models (e.g. dried lagoons, landfills), and pile models (e.g. mine tailings or aggregate piles). For the FMPC analysis, Kispert (1988) chose a pile model, an empirical equation derived from the iron and steel industry. For the same waste and climatic characteristics, pile models predict higher fugitive dust releases than area models. Based on the geometry of the Fernald pits, we believe an area model would have been more appropriate to estimate wind erosion of material.

The load-in equation is the same one used in Vaaler and Nuhfer (1989) to estimate emissions from solid spills (Equation K–1). As discussed previously, that equation was incorrectly derived from the EPA document, AP–42, and produces overly conservative results. Based on the correct equation, K–2, fugitive dust generated by the load-in of material would be insignificant, compared to that caused by wind erosion, and can therefore be dismissed from further consideration.

The parameter values used in the coal pile model were based on little data and may have been used inappropriately. Kispert (1988) used 9.7% as the percentage of time the wind blows >12 mph, based on a partial year (1987) of data from the FMPC meteorological station. The height of the wind speed measurements was not given. Based on five years of wind speed measurements collected at a height of 10 m at the FMPC, we determined the percentage of time the wind blows >12 mph to be 4.5%. The EPA source document is not clear on the height which is to be used in the equation. The empirical equation was derived based on the wind speed at a height of 1 foot (0.3 m) above the ground; however, their example calculations use the wind speed at the mean height of the pile, which was about 12 m. The Fernald waste pits were excavated below grade to maximum depths which ranged from 13 to 30 feet (4 to 9 m) (Table K–35). Because wind speed increases with height above the ground, the fraction of the time the wind speed exceeded 12 mph should have been considerably less than 4.5% in the pits themselves.

The EPA model requires as an input parameter the %silt (actually silt plus clay) of the waste, which is the percentage, by weight, of material which passes through a 200 mesh sieve (<75 μm). Kispert (1988) assumed a silt content of 10% for the dry waste material. Measurements obtained from borings into the waste pits (Solow and Phoenix 1987) indicate that the actual silt content of waste (massive forms were avoided) is considerably higher, ranging from 43% in Pit 1 to 75% in Pit 4. A boring from Pit 2, which contributed most of the fugitive dust during the period analyzed, was 61% silt.

The estimate of U concentration in the waste is subject to considerable uncertainty. Concentrations in five borings from Pit 2 (Solow and Phoenix 1987) ranged from 53 to 17,900 pCi ^{238}U g^{-1}, with a geometric mean of 1100 pCi g^{-1}. The mean concentration used in the erosion calculation (Table K–36) was 0.093 g U g^{-1} waste, or 31,000 pCi ^{238}U g^{-1}.

The choice of a pile model instead of an area source model provides additional bias. This model bias was examined further by conducting benchmark calculations using the coal pile model, the wind erosion equation used by agricultural scientists (Woodruff and Siddoway 1972), and the uranium mill tailings model used in MILDOS (NRC 1981). The latter two models were selected as most appropriate for modeling the pit releases, according to the criteria presented in Smith et al. (1982). Benchmark model comparisons were performed for Pit 1, because an example coal pile model calculation was provided by Kispert (1988) for this pit. The emission rates for particles less than 20 microns from Pit 1 in 1957 were estimated to be 3.2, 1.3, and 0.3 tons/acre-year using comparable data and the coal pile, agriculture erosion, and MILDOS models, respectively. (Note: The coal pile calculations were performed for particles < 20 μm, a silt content of 61%, and frequency of winds > 12 mph = 4.5%.) As expected, the coal pile model was the most conservative; however, the results are all within a factor of 10. Because the wind erosion equation requires the use of nomograms and qualitative data, and is thus not easily adapted to uncertainty analyses, the MILDOS model was selected for use in providing more realistic and site-specific estimates of pit releases.

The MILDOS algorithms for estimating dust emissions are:

$$u^*_t = C_t\,([(\rho_s - \rho)/\rho]\ gd)^{1/2}\,(1.8 + 0.6\log_{10}\omega) \qquad \text{(K–4)}$$

where:

u^*_t = threshold shear velocity (cm s^{-1})
C_t = dimensionless coefficient = 0.1
ρ_s = particle density, g cm^{-3}
ρ = density of air, 1.2×10^{-3} g cm^{-3}
g = gravitational acceleration, cm s^{-2}
d = average diameter of saltating particle, cm
ω = water content expressed in weight percent

$$q_h = C_h\,u^{*2}\,(u^* - u^*_t) \qquad \text{(K–5)}$$

where:

q_h = horizontal flux of particulate matter (<20 μm), g cm^{-2} s^{-1}
u^* = shear (or friction) velocity, cm s^{-1}
C_h = empirical constant relating shear velocity to horizontal flux, 10^{-6} g s^2 cm^{-4}

$$q_v = q_h\,(C_v/C_h)(1/u^{*3}_t)[(u^*/u^*_t)^{p/3} - 1] \qquad \text{(K–6)}$$

where:

q_v = vertical flux of particulate material, g cm^{-2} s^{-1}
Cv = coefficient of proportionality for vertical flux, 2×10^{-10} g cm^{-2} s^{-1}
p = percent of material that has a diameter < 20 μm

Two possible sets of source terms were considered prior to beginning calculations — field measurements and estimates based on waste records. However, first, the radionuclides of concern were selected through a simplistic screening process. Solow and Phoenix (1987) analyzed pit samples for various radionuclides using radiochemistry and gamma spectrometry. The radionuclides that were routinely detected were compared by using a hazard index, calculated by multiplying the average concentration in each pit by the respective dose conversion factor for inhalation. The total hazard for each pit was estimated by summing the values associated with each nuclide. The relative hazard index was then derived by dividing each nuclide hazard value by the sum. The results are shown in Figure K–26. The figure shows that ^{230}Th and ^{238}U could potentially contribute the majority of the dose from inhalation of resuspended particulates. (Note: the relatively significant contribution from ^{234}U in Pit 2 is a function of an apparent outlier in the measured data.) Based on this screening assessment, further analyses focused on ^{230}Th and ^{238}U.

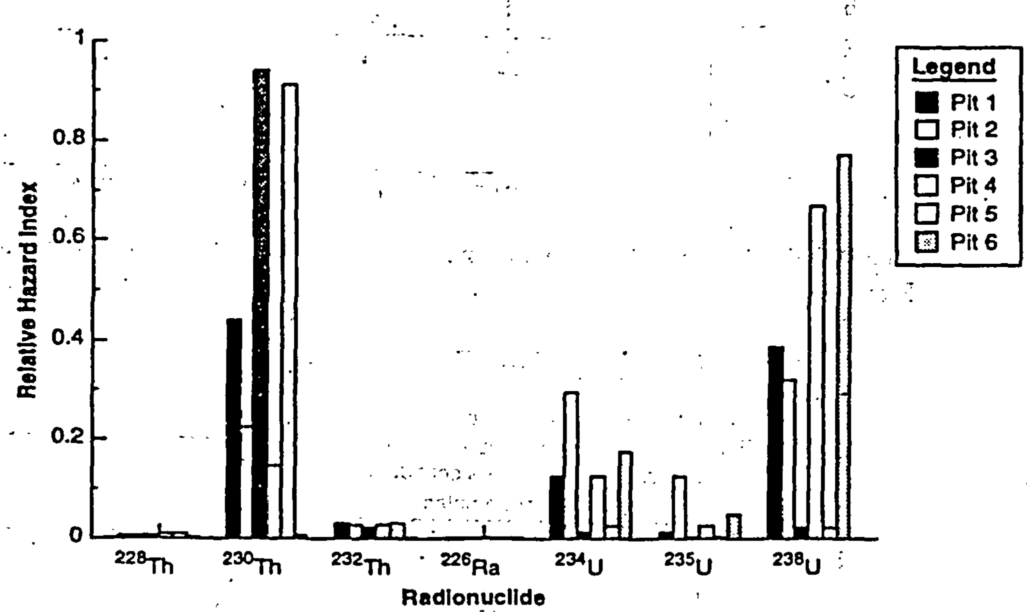

Figure K–26. Relative hazard index of radionuclides in Pits 1–6.

The next step was to select the radionuclide concentrations in the pits. The measured concentrations (Solow and Phoenix 1987) were compared with the values calculated from disposal records (Table K-36) to see if they were representative of those estimates. Figures K-27 and K-28 show the wide disparity between measured and estimated values. The small number of samples (4–7 per pit) contributes to the large uncertainty associated with the measured values. Because we have more confidence in the source terms estimated from disposal records, these values were used in subsequent calculations, with two exceptions. Thorium concentrations for Pits 2 and 6 were not available from disposal records, so the mean measured values reported in Solow and Phoenix (1987) were used in these instances.

Table K-37 presents the parameter value distributions used in the Crystal Ball simulations. The friction velocities used in the simulation, derived using FMPC meteorological data and Equation G–1 in Killough et al. (1993), are reported in Table K-38. The vertical flux calculated for each pit is shown in Table K–39. The flux was then multiplied by the area of pit exposed each year, as presented in Kispert (1988), by the

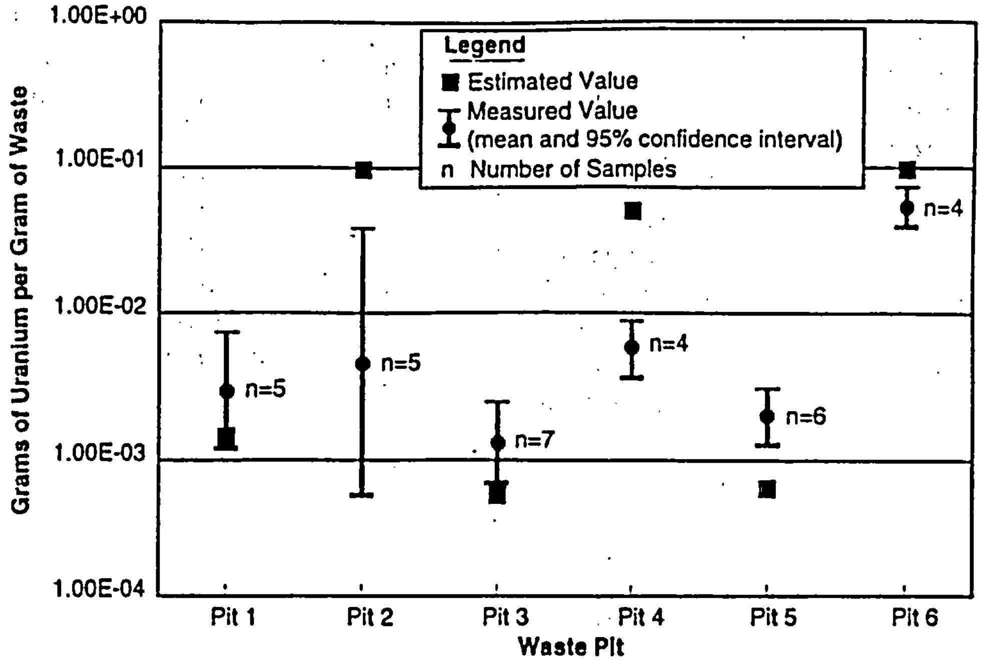

Figure K-27. Comparison of measured and estimated uranium concentrations in Pits 1-6.

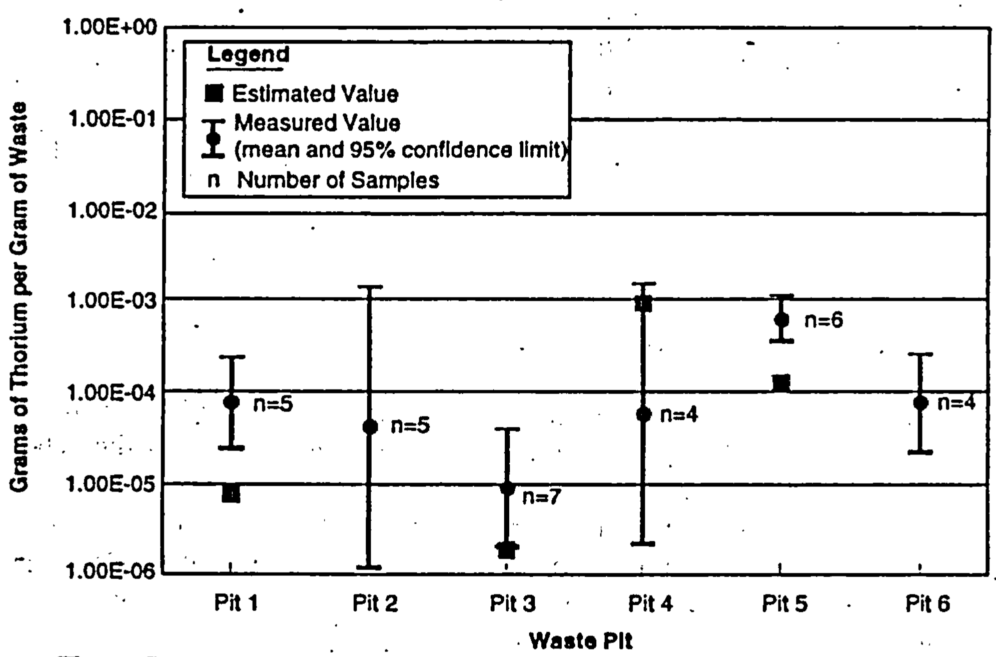

Figure K-28. Comparison of measured and estimated thorium concentrations in Pits 1-6.

radionuclide concentration estimated for each pit, and by the fraction of the year that there is no moisture [0.65 according to Kispert (1988)]. Pit 4 is unique in that it was shown by geophysical survey to contain a very high volume of buried ferrous metal objects (Solow and Phoenix (1987). Approximately 25-30% of the pit area has a high density of solid buried objects. It was thus assumed that an average 25% of the surface area was not erodible. A normal distribution was used, with a 10% standard deviation, to represent the nonerodible fraction in the final release calculation for Pit 4.

The uncertainties associated with the area exposed and the radionuclide concentrations are unknown and are not included in the calculations. However, Kispert (1988) states that the estimated areas are conservatively high. It was thus assumed that any potential underestimates of the radionuclide concentrations are offset by the conservative estimates of pit areas exposed.

Table K–37. Parameter Value Distributions Used in Uncertainty Analyses of Waste Pit Emissions

Pit number	Parameter	Distribution	Mean/Min	Std. Dev./ Max.	Comments
1	ω (%)	uniform	10	17	Maximum value for each pit is an average of measurements from Solow and Phoenix (1989). Minimum value for all dry pits assumed to be 10%.
	ρ (%)	normal	21	2.1	Mean values for each pit derived from Solow and Phoenix (1989). Assume SD = 10% of the mean.
2	ω (%)	uniform	10	32	
	ρ (%)	normal	32	3.2	
3	w(%)	uniform	45	50	Minimum values for all wet pits assumed to be 90% of the value reported in Solow and Phoenix (1987).
	ρ (%)	uniform	26.5	2.5	
4	ω (%)	uniform	10	28	
	ρ (%)	uniform	23	2.3	
5	ω (%)	uniform	170	190	
	ρ (%)	uniform	19	1.9	
6	ω (%)	uniform	10	55	
	ρ (%)	uniform	24	2.4	
All	C_t	nominal value	0.1		NRC (1981)
	ρ_R	nominal value	2.4		NRC (1981)
	g	nominal value	980		NRC (1981)
	d	nominal value	0.03		NRC (1981)
	u^*	varies with wind speed and stability class	See Table K–38 for values		Killough et al. (1993)

Table K-38. Shear Velocities (m s^{-1})

Wind Speed (m s^{-1})	Stability Classes					
	A	B	C	D	E	F
1	0.1	0.1	0.1	0.1	0.1	0.1
2	0.4	0.4	0.3	0.3	0.3	0.2
5	0.7	0.7	0.6	0.5	0.4	0.4
7	1.0	0.9	0.8	0.7	0.6	0.5
9	1.3	1.2	1.0	0.9	0.8	0.7
11	1.6	1.4	1.3	1.2	1.0	0.8

Table K-39. Vertical Flux Rates (10^{-10}g cm^{-2} s^{-1}) from Waste Pits[a]

Pit number	50%	5%	95%
1	2.8	1.0	7.8
2	42	7.3	228
3	3.5	1.0	11
4	3.4	1.0	11
5	0.2	0.15	0.3
6	2.5	0.7	11

[a]Estimated for particles < 20 μm physical dimameter.

Total emissions for all pits, over their operating lifetimes, are presented in Table K-40. As expected, the results for five of the six pits exceed or, in the case of Pit 2, approximate those presented Kispert (1988). However, surprisingly, the results for Pit 2, calculated by MILDOS, exceed the Kispert calculations by a factor of three.

Table K-40. Total Fugitive Emissions (kg) from Waste Pits

Pit Number	^{238}U			^{230}Th		
	50%	5%	95%	50%	5%	95%
1	6	2	8	0.03	0.01	0.08
2	2500	410	12400	1	0.2	6
3	10	3	29	0.07	0.01	0.1
8	410	130	1350	9	3	32
5	0.2	0.15	0.3	0.07	0.05	0.1
6	32	9	110	0.001	0.0004	0.006

The major source of this large difference is the parameter "p" used in Equation K-6 of the MILDOS model. This parameter represents the percentage of material that has a physical diameter less than 20 μm. Because the vertical flux of material is proportional to this parameter raised to the power of three, small changes are amplified. It is thus the most sensitive parameter in the equation. For example, a change in "p" from 21% (the value measured in Pit 1) to 32% (the value measured in Pit 2) results in a 20-fold increase in the vertical flux. For comparison, an increase of 10% in the silt content parameter used in the coal pile model used by Kispert results in only a 3-fold increase in the flux. Because of this sensitive parameter, the uranium emissions are the largest contributor to total unmonitored releases from the FMPC.

The uranium emissions calculated for Pits 2, 4, and 6 are shown in Table K–41. The uranium releases from Pits 1, 3, and 5 were less than 1 kg y^{-1}. The thorium emissions for these pits were neglible (<0.1 kg y^{-1}). Given the low relative hazard index (see Figure K–26) of ^{230}Th calculated for Pits 2, 4, and 6, ^{230}Th disposed in the waste pits will not be included in the final source term. However, daughter radionuclides resulting from the decay of U-238 will be estimated and included in the source term. Because of the close physical proximity of the pits, the fugitive uranium releases from Pits 2, 4, and 6 will be modeled as a single pit for the final dose calculations. The total release from all pits was estimated to be 2961 kg (50th percentile).

Table K–41. Estimated Fugitive Emissions from Pits 2, 4, and 6

PIT 2	Area[a]	^{238}U emission rate (kg y^{-1})		
Year	ft^2	50%	5%	95%
1957	17781	127	27	615
1958	35562	229	51	1308
1959	35562	229	51	1308
1960	40008	289	52	1531
1961	44453	330	55	1711
1962	44453	330	55	1711
1963	44453	330	55	1711
1964	44453	330	55	1711
1965	22227	150	24	846
1966	13336	96	15	501
PIT 4	**Area[a,b]**	**^{238}U emission rate (kg y^{-1})**		
Year	**ft^2**	**50%**	**5%**	**95%**
1960	9583	2.2	0.8	6.8
1961	19166	4.4	1.4	14
1962	38333	8.8	2.6	27
1963	43124	10	2.9	30
1964	47916	11	3.2	30
1965	47916	11	3.2	30
1966	52708	11	3.5	37
1967	52708	11	3.5	37
1968	57499	13	4	39
1969	57499	13	4	39
1970	57499	13	4	39
1971	57499	13	4	39
1972	62291	13	4	40
1973	62291	13	4	40
1974	62291	13	4	40
1975	67082	15	4	49

(continued next page)

Table K-41. Estimated Fugitive Emissions from Pits 2, 4, and 6 (cont.)

PIT 4	Area[a]	^{238}U emission rate (kg y^{-1})		
Year	ft^2	50%	5%	95%
1976	71874	17	5	49
1977	81457	18	5	54
1978	91040	20	6	61
1979	95832	25	9	75
1980	95832	25	9	75
1981	95832	25	9	75
1982	95832	25	9	75
1983	95832	25	9	75
1984	95832	25	9	75
1987	95832	25	9	75
1988	95832	25	9	75
PIT 4	**Area[a]**	**^{238}U emission rate (kg y^{-1})**		
Year	ft^2	50%	5%	95%
1979	3240	1.2	0.4	4.6
1980	4860	2.1	0.6	7.1
1981	8100	3.6	1.1	13
1982	8100	3.6	1.1	13
1983	8100	3.6	1.1	13
1984	8100	3.6	1.1	13
1985	8100	3.6	1.1	13
1986	8100	3.6	1.1	13
1987	8100	3.6	1.1	13
1988	8100	3.6	1.1	13

[a]From Kispert (1988).
[b]An average of 75% (± 7.5%) of the area shown was assumed to be erodible, due to the presence of solid ferrous objects in Pit 4.

RADON RELEASE FROM K-65 SILOS, APRIL 25, 1986

Two projects were undertaken by the FMPC in 1986 to preserve the structural integrity of the K-65 Silo domes (WMCO 1987a). In January, 20-ft diameter, protective covers were placed on the centers of the domes. A subcontractor began applying a weatherproof coating to the domes. Then, on April 25, 1986, the Silos were vented to the atmosphere for several hours. The application of the weatherproofing was not completed.

Description of the April 25, 1986, Episodic Release

Two reports describing the April 25, 1986, Rn release have been located. The first was a preliminary letter report, issued by the DOE (Reafsnyder 1986). The second was the formal report of the DOE Incident Investigation Board (DOE 1986). The following description of the incident and Rn release is taken from DOE (1986).

On April 14, 1986, a subcontractor began applying the weatherproof coating to the K-65 Silo domes. The coating material was a neoprene hapalon, applied as a fluid. This work was stopped on April 17, 1986, due to radiation safety concerns. Work resumed on April 18, 1986, but only for part of the day. On the afternoon of April 18, 1986, work was again stopped, after it was discovered that air from the Silos was escaping through the protective coating and apparently causing high radiation levels above the dome areas. It was then noticed that bubbles were present in the coating on Silo 2, and cracks and holes were also visible on Silo 2.

On Friday, April 25, 1986, the K-65 Silos were vented, without authorization, by FMPC staff. The venting was apparently performed in order to reduce the pressure in the Silos and to reduce the high radiation above the Silo domes, to allow the application of the coating to the domes to proceed. The venting was accomplished by removing one of the blank flange on the Silo domes, and installing a new two inch flange, an elbow, a quick release coupling, and lengths of flexible, schedule 80, plastic pipe. The blank flange was removed from Silo 2 at about 10:15 am, and the blank flange from Silo 1 was removed immediately after. Between 10:30 and 10:40 am, two 50 ft sections of the pipe were attached to the new flange on Silo 2. Between 10:50 and 11:00 am, one 50 ft section of the pipe was attached to the new flange on Silo 1. At 1:00 pm, it was agreed that the blank flanges were to be reinstalled. At 2:00 pm, it was reported that the blank flanges had been reinstalled on the K-65 Silos. Thus, the venting took place from about 10:15 am to 2:00 pm, a total of about 3¾ hours.

Current Estimate of Rn Release

None of the documents obtained include estimates of the quantities of Rn released during the Silo venting. The preliminary report (Reafsnyder 1986) and the incident investigation report (DOE 1986) did report hourly measurements of Rn concentrations in air near the Silos. However, the location of the measurement instrument was not clearly described, and the exact locations of the release points (the ends of the flexible pipe) were not described, although they were within about 35 ft of the Rn monitor (DOE 1986). These data cannot be used to reconstruct the Rn release rate.

Instead, we will estimate an upper-bound release rate that could reasonably have occurred, using the models for Rn releases that are used in Appendix J for ongoing Rn releases from the Silos. It seems likely that attaching a 2-inch vent pipe to the Silo domes would have increased the air exchange ventilation of the Silo air space. So we calculate the excess releases as air exchange releases, using an equation similar to that used for the ongoing air exchange Rn releases for the 1980–1987 period (see Appendix J):

$$Q_{\text{epi,max}} = C_a \lambda_v V_0 \text{CF} U \qquad \text{(K–7)}$$

where

$Q_{\text{epi,max}}$ = the (estimated maximum reasonable) Rn release rate from the K-65 Silos during the April 25, 1986, episodic release,

C_a = the Rn concentration in the Silo head space air,

λ_v = the ventilation rate of the Silo head space air,

V_0 = the volume of the Silo head space air,

CF = a units conversion factor, and

U = an uncertainty factor, the reason for which will be discussed later.

The quantity of Rn released can then be calculated by:

$$R_{epi,max} = Q_{epi,max} t \qquad \text{(K–8)}$$

where t is the length of time the release continued. This approach assumes that the release rate remains constant. However, it actually would decrease during the release time, because the Rn concentration, C_a, would decrease due to the additional ventilation of the air space. Thus, our simplification results in estimates that are biased somewhat high.

As for the ongoing releases for 1980–1987, calculated in Appendix J, we assume the Rn concentration in the head space air is the same at the time of the release as at the time it was measured in November 1987. The concentration is assumed to be represented by a normal distribution, with mean 2.62×10^7 pCi L^{-1}, and with standard deviation 4.1×10^6 pCi L^{-1}.

The head space ventilation rate, λ_v, is very uncertain for this episodic release. For the 1980–1987 period, air exchange releases are presumed to have occurred through the numerous cracks in the Silo domes. For that period, the driving force was thought to be the temperature-induced expansion and contraction of the head space air. For the 1959–1979 period, releases occurred through open penetrations through the domes, including a 6-inch diameter gooseneck vent pipe and many smaller penetrations (Appendix J). For that period, the releases were thought to be caused also by wind across the Silo domes. The ventilation of the Silos with a 2-inch pipe open to the atmosphere is not really similar to either of the previous situations. However, we are trying to place an upper bound on the releases. The ventilation rate during this episodic release would be less than the ventilation rate for the 1959–1979 period, since for the episodic release, the areal extent of openings in the Silo domes is considerably less. Thus, we assume that the ventilation rate for the episodic release is the same as that calculated for the 1959–1979 period.

In the calculations of ongoing releases for the 1959–1979 period (Appendix J), we actually calculated the fractional loss rate for air exchange plus diffusion Rn releases from the Silos, $\lambda_{v+d,pre}$. Because of the way the calculations were performed, the diffusion releases were not separated from the air exchange releases. However, the diffusion releases are probably only a small fraction of the total releases (based on estimated diffusion releases for 1980–1987, the difference in Rn concentrations for 1959–1979 and 1980–1987, and estimated total releases for the two periods). Thus, it seems reasonable to assume that, for the 1959–1979 period, the ventilation rate, $\lambda_{v,pre}$, is equal to the total fractional loss rate, $\lambda_{v+d,pre}$. This is done for our calculations here. From the calculations in Appendix J, $\lambda_{v+d,pre}$ had a broad distribution with median value 2.4 d^{-1}, and 90% probability range (5th to 95th percentiles) of 0.83 to 16 d^{-1}. For the calculations here, we use the exact distribution as calculated in Appendix J.

For the volume of the Silo head space, we use the same values used in the calculations of ongoing releases (Appendix J). Thus, a uniform distribution is assumed, with minimum 40,000 ft^3, and maximum 62,000 ft^3 (per silo).

For the units conversion factor, we desire the Rn release rate, $Q_{epi,max}$, to have units Ci h^{-1}. The component factors have units pCi L^{-1} for C_a, d^{-1} for λ_v, and ft^3 (per silo) for V_0. Thus, the conversion factor is:

$$CF = [(28.317 \text{ L ft}^{-3})(10^{-12} \text{ Ci pCi}^{-1})(2 \text{ silos}) \div (24 \text{ h d}^{-1})] \qquad \text{(K–9)}$$

$$= 2.36 \times 10^{-12} \text{ d L Ci h}^{-1} \text{ ft}^{-3} \text{ pCi}^{-1}$$

It is felt that the uncertainty in the calculation of release rate for this episodic release has not been totally accounted for by the parameter uncertainties. In particular, the previous calculation of $\lambda_{v+d,pre}$, which is now used for λ_v, assumes $\lambda_{v+d,pre}$ to be a long-term average value. In the present calculations, we are concerned with the ventilation rate for a short period of time on a single day. The same criticism applies to the Rn concentration C_a. Because of these additional uncertainties, we apply an additional uncertainty factor, U. In this case, we assume (somewhat arbitrarily) U to have a lognormal distribution, with geometric mean 1, and geometric standard deviation 1.4 (for a 95% confidence interval, this gives an uncertainty of about ×/÷ 2).

Calculations were performed as a Monte Carlo simulation, with 10,000 iterations, using the methods described in Appendix J for the calculations of ongoing Rn releases. The initial results for the upper bound on the Rn release quantity, $R_{epi,max}$, are a median estimate of 29 Ci, with 90% probability range (5th to 95th percentiles) of 8–210 Ci. However, the 95th percentile value is not a realistic estimate, because this quantity is significantly more than the quantity of Rn in the Silo air spaces. The total quantity of Rn in the sir spaces is just the concentration, C_a, times the volume, V_0, times 2 silos. If the uncertainties in these two parameters (described above) are accounted for, the 90% probability range for the quantity of Rn in the air spaces of the two Silos is 52–100 Ci, with median 74 Ci. The unrealistically high value of 210 Ci released is partly due to our simplying assumption that the concentration in the Silos remains constant during the release. The 95th percentile of the upper bound on the Rn release quantity, $R_{epi,max}$, can be assumed to be no greater than 100 Ci. We thus conclude that the upper bound of the excess Rn released during the episodic release is probably within 8–100 Ci, with a best estimate of the upper bound being around 30 Ci.

The median estimate of the upper bound on the Rn release rate, $Q_{epi,max}$, is about 7.6 Ci h^{-1}. From information in Appendix J, the median estimates of diffusion and air exchange releases for the 1980–1987 period correspond to a Rn release rate of about 0.2 Ci h^{-1} during daylight hours. From a comparison of these release rates, it appears that this Rn release of April 25, 1986, could meet the criteria for an episodic release. (Since we have only made upper-bound estimates, we cannot be more definite.)

Supplemental Environmental Measurements

At the time of this episodic release, a few Rn monitoring programs were in place at the FMPC (Reafsnyder 1986; DOE 1986). These included (1) onsite measurements at 17 locations and offsite measurements at three locations within two miles of the site, performed by Mound Laboratories; (2) measurements at the boundary air monitoring stations, onsite locations, and some offsite locations, using alpha-track monitors for three-month-long measurements, performed by the FMPC; and (3) continuous measurements (actually provided hourly results) with a Rn gas monitoring instrument, very near the K-65 Silos.

Regarding results of the Mound Laboratories measurements, the DOE (1986) report indicates that average measured concentrations, for onsite and offsite locations, for the two-week period that included April 25, 1986, were higher than similar averages for the preceding 1½-year period. We have compiled results of the Mound measurements in the Task 4 report of this Project (Killough et al. 1993). Based on our compilation (Table PS–1 of the Task 4 report), the averages for this two-week period were generally higher than the long-term averages, but were not higher than the range of results for other one- or two-week periods measured by Mound.

Regarding the alpha-track measurements performed by the FMPC, due to the Rn release the detectors were retrieved and analyzed earlier than scheduled. For the period March 18–April 29, 1986, concentrations at two offsite locations appeared higher (at 1.29 pCi L^{-1}) than average offsite concentrations for 1985 (at 0.59 and 0.37 pCi L^{-1}) (DOE 1986). From the 1986 annual environmental monitoring report (WMCO 1987a), however, it appears that the two offsite locations, called OS1 and OS2, were in opposite directions from the K-65 Silos. This suggests that the episodic Rn release is not the only reason for the potentially elevated concentrations measured.

The hourly Rn measurements were made within about 35 ft of where the Rn was discharged from the flexible pipes (DOE 1986), although the exact locations of the measurements and the discharge points were not indicated in the report. Measurement results for April 23 and April 24, 1986, were markedly different from those of April 25, 1986. For April 23 and 24, peak concentrations, occurring from mid-morning to mid-afternoon, were about 10 and 40 pCi L^{-1}, respectively. On April 25, the peak concentration was 694 pCi L^{-1}. This significant difference may indicate that Rn releases on April 25 were substantially greater than on April 23 and 24. However, on April 23 and 24, the releases are assumed to be from the domes of the Silos, while on April 25, the releases were from the domes plus from the flexible pipes that had been attached. The exact locations of the release points in relation to the measurement point are not known. In addition, wind directions around the Silos on these days are not known. Thus, we really cannot make a quantitative comparison.

Conclusions — Episodic Radon Release

In conclusion, there was a Rn release on April 25, 1986, that may meet our criteria for an episodic release. The information available for quantitatively estimating the release rate and release quantity is quite limited. Thus, we have only performed upper-bound estimates. Radon monitoring was performed for time periods surrounding the episodic release. Of these data, the hourly measurements, made close to the K-65 Silos, provide the best corroboration (though only qualitative) that an episodic release occurred on April 25, 1986. However, it appears that comparisons of concentrations measured during the release time with concentrations measured at other times will not be useful for quantitative assessments of the release.

SUMMARY AND CONCLUSIONS – OTHER SOURCES AND EPISODIC RELEASES

A variety of historical unmonitored sources of uranium releases to the atmosphere have been evaluated in this appendix. This concluding section will summarize the estimated releases from these miscellaneous sources, compare them with previous estimates, and attempt to place them in perspective with the major atmospheric releases from FMPC operations. Table K–42 presents the total release estimates from the unmonitored sources over their entire period of operation. In addition, the table illustrates the difference between our reconstructed source terms and those previously developed by the FMPC contractor. In

Table K–42. Summary of Total Estimated Releases of Uranium to the Air from Miscellaneous Sources at the FMPC

Source	Inclusive Dates	Total Release Estimate (kg U) Median	5th–95th Percentile Range	Previous Estimate[a]
Old Solid Waste Incinerator	1954–1979	2200	1600–2900	2471
Oil Burner	1962–1979	370	270–470	467
Graphite Burner	1965–1984	230	61–730	129
New Solid Waste Incinerator	1979–1986	8	0.6–90	14
Liquid Waste Incinerator	1983–1986	4	0.9–9	12[d]
Building Ventilation	1954–1987	4100	970–15,000	390
Miscellaneous Process Emissions	1953–1988	[b]	110–970[c]	324
Lab Hoods	1953–1987	[b]	20–200[c]	66.5
Waste Pits	1953–1988	3000	900-12,000	1560
Non-routine Releases	1952–1988	1300	780–2900	2784
Episodic Releases	See Table K–43	See Table K–43		

[a]From FMPC operating contractor. See individual sections of Appendix K for sources of information.
[b]Not reconstructed.
[c]Subjective uncertainty of a factor of 3 applied to previous estimate.
[d]Based on maximum processing rate.

contrast to previous estimates, the reconstructed source terms all carry some estimate of uncertainty and are well documented.

The agreement between past and revised release estimates is good for the incinerators. With the exception of the new liquid waste incinerator, which is a minor source, the 5th–95th percentile range of our estimate encompasses the previous estimate. The reconstructed release estimate from building ventilation is significantly higher than the previous estimate, due to two main reasons: (1) the use of a lower dilution factor for building make-up air and (2) the use of higher in-plant airborne contamination levels, measured in the 1950s, to make a forward projection through 1970.

Median estimates of releases from the waste pits were about two times higher than previous results. This difference is a function of the use of a model which relied on site-specific data, particularly soil characteristics. The model was highly sensitive to particle size, which varied greatly among pits and was highest for pits 2 and 4. These pits also had the highest estimated U concentrations, thus resulting in substantially higher release estimates during time periods when these pits were active.

The median release estimate for non-routine releases is less than that previously calculated by Vaaler and Nuhfer (1988), although the 5th to 95th percentile range encompasses the previous estimate. The lower median estimate reflects the selection of different models and the use of site-specific data for fires and spills. Uranium hexafluoride release models were not revised, although subjective uncertainty limits were assigned to input parameters.

Most of these miscellaneous sources were not releasing uranium to the atmosphere over the entire production history at the FMPC. Figure K–29 illustrates the time-dependent nature of the release estimates for the three most significant of the unmonitored sources. All three sources were most important in the late 1950s and early 1960s. The waste pit source term is strongly influenced by the timing of use of Pit 2, which was closed in the mid-1960s. The building exhaust source term is highly dependent upon the production rate of scrap recovery operations, which peaked in 1960. By 1970, each of these sources contributed less than 100 kg uranium per year to the atmosphere (Figure K–29).

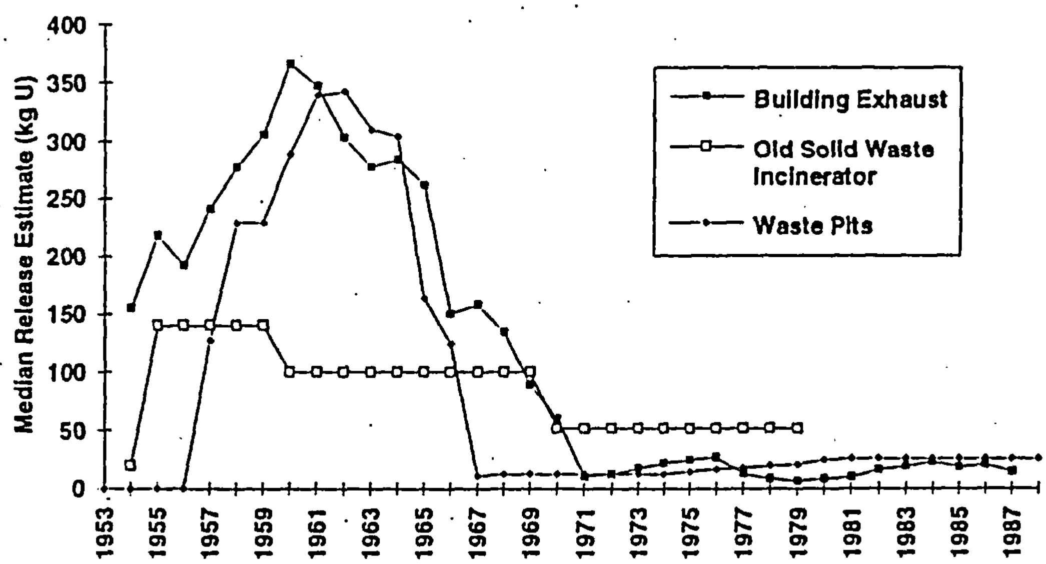

Figure K–29. Time-dependent median source term estimates for the more important sources of unmonitored releases of uranium at the FMPC (excluding the UO_3 gulping process).

Six incidents involving releases of uranium were identified which met our criteria for special treatment as episodic releases. Three of these episodic releases were documented in incident reports and occurred on November 7, 1953, between November 12 and 16, 1960, and on February 14, 1966. The remaining three episodic releases were identified by air monitoring data, although documentation could not be found to identify the sources. These events occurred sometime during the weeks ending on September 28, 1978, February 8, 1979, and September 20, 1983. The six episodic releases are summarized in Table K–43. In terms of total quantity of uranium released, the dust loss which began on 11/12/60 had the most impact. However, the incident on 2/14/66 had the largest rate of release.

A release of about 30 Ci of radon occurred on April 25. 1986, from unauthorized venting of the K-65 silos. This source term also may be treated separately as an episodic release.

Table K-43. Summary of Six Episodic Releases of Uranium Which Were Identified from Incident Reports and Air Monitoring

Start Date	Description	Uranium Released (kg)	Duration of Release
11/7/53	Release of UF_6 from defective cylinder in Pilot Plant	45	15 min
11/12/60	Dust loss from dust collector bags in Pilot Plant	310	1 or 3 days
	Replacement of bags	470	5 days
2/14/66	Release of UF_6 from cylinder in Pilot Plant due to operator error	750	1 hr
9/21/78	Unknown	20–370[a]	7 days
2/1/79	Unknown	60–680[a]	7 days
9/13/83	Unknown	290–360[a]	7 days

[a]Range of values is based on results from several different ambient air monitoring stations. See Table K-30.

After careful examination of many types of information, the conclusion is well supported that the magnitudes of uranium releases from the miscellaneous unmonitored sources are minor relative to the three major sources of emissions from the FMPC, which are: the scrubbers in Plant 8, the scrubbers in Plant 2/3, and the plant-wide dust collectors. Figure K-30 illustrates the relative importance of the various sources — Figure K-30a is plotted on a logarithmic scale, so that the uncertainty distributions can be seen more clearly. Figure K-30b is plotted using a linear scale, which more accurately illustrates the true relative magnitude of these sources.

When all of the sources investigated in Appendix K are combined, using appropriate statistical measures, the grand total of the releases is 16,000 kg (median estimate), with a 5th–95th percentile range of 9,300 to 28,000 kg. This total does not include the November 1960 dust loss from the Pilot Plant, which is included with the total dust collector source term. See the main text of this report for an overview of total source terms from historic atmospheric releases at the FMPC.

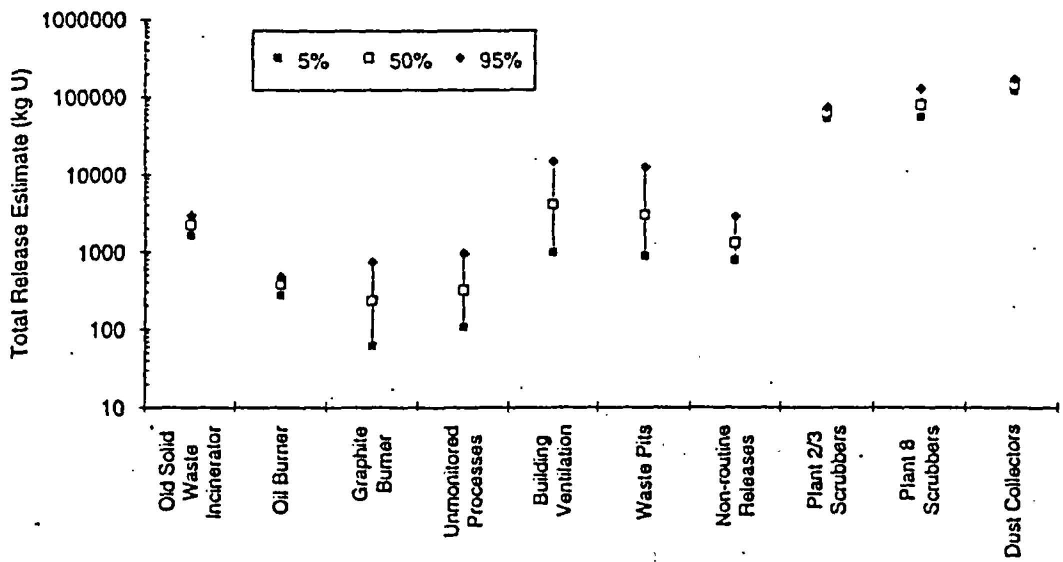

Figure K–30a. *Note logarithmic scale.*

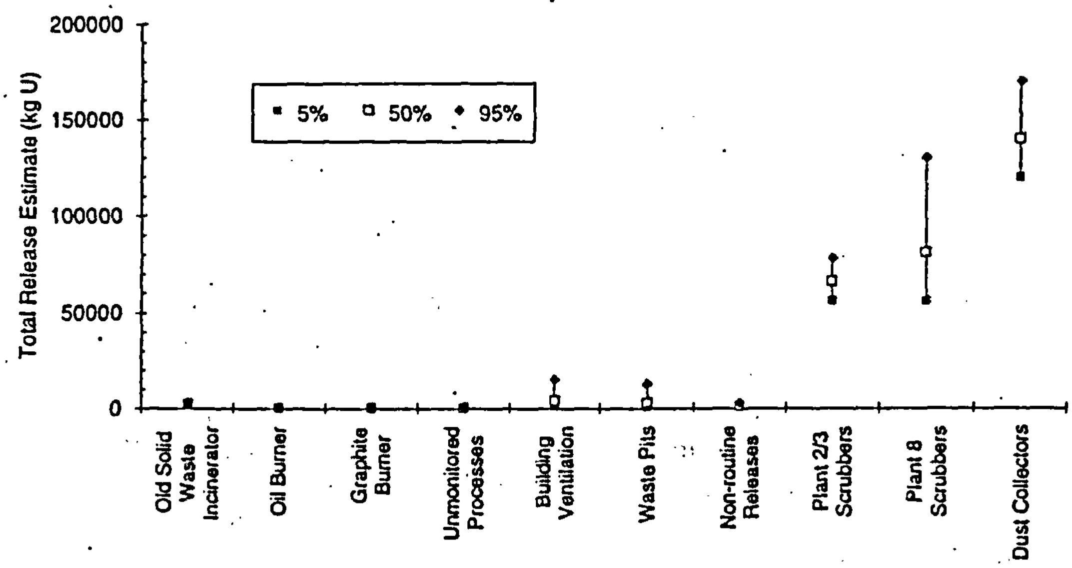

Figure K–30b. *Note linear scale.*

Figure K–30. Relative importance of miscellaneous unmonitored sources of atmospheric releases of uranium compared with releases through scrubbers and dust collectors. The 50% point represents the median (best estimate). The 5% and 95% points encompass a 90% probability range on the total estimates.

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ANNEX 1 TO APPENDIX K

CHRONOLOGICAL HISTORY OF EVENTS AND NOTES RELATED TO RECONSTRUCTION OF SOURCE TERMS FROM THE OLD SOLID WASTE INCINERATOR (OSWI)

Note: The information in this annex was compiled from Industrial Hygiene and Radiation (IH&R) Department Monthly Reports (or reports from Sections within the IH&R Department), unless noted otherwise. Comments by RAC researchers are italicized, to distinguish them from the notes and observations from the FMPC contractor documents.

Date	Event/Notes
Nov 16, 1954	Official start-up date of OSWI given in Boback et al. 1987.
December 1954	Three air dust samples taken at incinerator. *One Analytical Data Sheet (ADS) located.*
Dec 21-28, 1954	Air velocity measurements made on stack of incinerator.
Jan 3-10, 1955	Velocity and temperature measurements were taken in incinerator stack.
April 1955	Sampler installed in incinerator.
May 2-6, 1955	A day was spent at the incinerator working with the stack sampler there. Temperature measurements were taken along with the sample. The sampling probe had deteriorated due to the heat in the chimney, but we were able to get a sample of an afternoon's burning. *ADS located but no volume of air sampled is given. 26.3 mg U collected for afternoon (approximately 3–4 h) would be an emission rate of only 7–9 mg U h^{-1}.* Sampling apparatus has been brought in to wait for the fabrication of a new probe.
May 9, 1955	Stack sampling log book started. Samples collected in the incinerator stack.
June 14, 1955	New probe being fabricated for incinerator stack.
June 1955	New sampling probe for incinerator stack finished.
July 26, 1955	Preparing to get together a sampler impinger in series with a large diameter tube with the first sampler to collect incinerator stack samples
July 1955	Revisions were made in original sampler and procedure for use in incinerator stack. New sampler and procedure have not been tried.
Nov 1955	General air samples taken from downwind of incinerator stack while burning normal loads, so that a background could be obtained. While a load of material from Plant 9 was being burned, fifteen minute samples were taken at 1 and 3 stack lengths downwind. The results were 3 dpm per m^3 and 2 dpm per m^3, respectively. On a load of refuse from the Cafeteria, fifteen minute air samples were taken at 3 and 5 stack lengths downwind. The results were 0.78 and 3 dpm per m^3, respectively.

Nov 14-20, 1955	An effort was made to estimate flow rate in the incinerator by measuring the flow into the feed door, but the temperature was too high to allow use of velometer or anemometer. We were also unable to get a temperature reading at the top of the stack. L. Williams was asked to have an opening made in the stack. It was decided to get general air samples downwind from the incinerator to establish a background before burning of SF material is undertaken. Considerable time was spent in trying to locate a portable generator. By the time the generator was located the weather changed so that sampling could not be done.
Nov 21-27, 1955	Air samples obtained downwind from incinerator while burning thorium contaminated materials. *ADS not located.* Results generally low. Additional samples obtained while burning normal uranium-contaminated material. *ADS located.*
Nov 22,1955	Twenty-one drums of uranium-contaminated wood and paper was burned and general air samples taken downwind from the incinerator. *ADS located.*
Dec 2,1955	Preliminary work begun for air sampling tests in conjunction with burning of contaminated materials at the incinerator.
Dec 6, 1955	Preliminary report submitted on the results of burning normal U contaminated material in the incinerator. Recommendations were made to burn non-contaminated refuse only until further tests can be made. *Implies that results indicated an unacceptable loss of material and/or worker exposure. However, burning of contaminated wastes continued and a different conclusion was reached next year.*
Feb 21, 1956	Accountability requested permission to burn a large number of drums of contaminated material from GE *(General Electric)*. Arrangements will be made ASAP. Stack and air dust samples will be taken during burning.
Mar 2, 1956	Preparations made for stack and downwind air sampling during burning of GE wastes.
Mar 6, 1956	Two attempts at measuring velocity traverse of incinerator stack. Two different Pitot tubes were used with an inclined manometer reading from 0–0.5 " water. At no time was a differential registered. The possibility of using a "Hastings Air Meter" will be investigated. Burning of contaminated materials from GE has not yet started.
Apr 9-15, 1956	Air samples taken in vicinity of the incinerator.
May 1956	Burning of waste from GE was started May 8. *Air dust sample ADS located. Burning contaminated waste paper from GE, as well as paper, wood, and other NLO non-contaminated material.* The drums are burned between loads of refuse coming from the FMPC operation. Air samples were taken downwind when conditions permitted.
May 21, 1956	Five fallout trays placed in vicinity of incinerator for background info prior to special burning test. *ADS located.*
May 26, 1956	On Saturday, May 26, a test burning of contaminated gloves took place at the incinerator. 100 drums of gloves were burned. The net weight for each drum was assumed to be 62 pounds. Report to be submitted. *ADS located.*
May 29, 1956	A stack sampler for the incinerator was designed and constructed. On a trial run, the sampler performed well. Rain occurred throughout test. Stack samples, air dust samples at various stack lengths, breathing zone air dust on the platform, and gummed paper were taken. A report will be prepared.

June 5, 1956	Re: contaminated burnables from GE: Accountability accepts these shipments, because this installation has the facilities for handling scrap materials generated by other AEC sites and is "obligated to accept this type of material."
June 7,1956	Second incinerator test, burning contaminated gloves, run Thursday evening June 7. *ADS located for stack samples.* The wind from W and samples could not be taken downwind at a distance of more than about 30 stack lengths.
June 26, 1956	All results from burning of contaminated gloves returned; report being written. Contaminated shoes to be burned on Saturday June 30. *ADS located – ash 0.6% U.*
July 2, 1956	Incinerator report 90% complete. 18.1% U on analysis of ashes from 96 drums of gloves and 32 drums of GE paper burned Saturday, June 23.
July 3, 1956	Incineration of contaminated burnable items — several tests have been run, burning contaminated gloves, shoes, and paper. Results indicate this can be done without causing excess contamination of the air or ground surfaces in the vicinity. Report being prepared.
August 1956	30 drums of contaminated shoes were checked on the storage pad and okayed to burn in the incinerator
Sept. 6, 1956	Active project: Disposing of contaminated material at the incinerator. Report of burning contaminated materials at the incinerator is being revised.
Sept. 15, 1956	*A special firing of rubber gloves, mop heads and rags resulted in ash which was 61% uranium (Anonymous 1956a)*
Sept. 18,1956	Memo to L. Williams giving permission to burn sewage sludge.
Sept. 22, 1956	*Special firing of rubber gloves and rags resulted in ash which was 36% uranium (Anonymous 1956b).*
March 4, 1957	Evaluation of burning contaminated materials at the incinerator is not complete.
March 1957	Fallout from incinerator: preliminary work was completed using a Bausch & Lomb dust counter to determine roughly where to expect the fallout. This survey showed most of the material was falling out within 1-1/2 stack heights distance downwind. *This is about 50 feet.* More background data is being obtained with fallout trays and it is anticipated the proposed test burning of filter bags will be accomplished this month.
April 30, 1957 (McKelvey 1957)	The size of the drum storage area is limited and we fully understand our responsibility to aid in the elimination of any of this material. We are endeavoring to determine the air, ground, and stream pollution effects which might result from the incineration of relatively highly contaminated material such as dust collector filter bags and work gloves which cannot be cleaned. Previous tests have indicated that slightly contaminated burnables can be burnt at the incinerator without adding appreciably to the air contamination level in the vicinity of the incinerator and at downwind locations. Before proceeding with the burning of large quantities of more highly contaminated material, we would like to test burn a few bags.
May 4, 1957	On May 4, 1957, 110 drums of contaminated clothing and 15 drums of dust collector filter bags were burned in incinerator. This produced about four drums of ashes containing 53% U. *ADS shows only 4.2 % U for 100 drums burned and ash collected 5/4/57.* Air dust and fallout samples higher than normal. Preliminary report complete and will be distributed.

May 1960 (Construction Proposal CP-60-40)	A new incinerator SW of Plant 8 was proposed (CP-60-40). Justification includes a need for disposing of soil residues, such as oily or organic sludges. The amount of contaminated trash that is being burned at the present incinerator is increasing, in spite of supervisory efforts to prevent SS materials from being thrown into the non-contaminated trash containers. Assays of the existing incinerator ash as high as 12.6% U and the 0.6% U assay for flue dust obtained from the ledge at the stack are indications of the amount being burned. The airborne U losses are both an accountability and health and safety problem. Estimate of plant trash generation rate is 35,000 pounds per day, which includes three open truck-loads of shipping dunnage per day and approximately twenty fork-lift skids per week. A preliminary estimate of cost for proposed incinerator is $324,000. *This proposal was not approved.*
November 1961	Tests are now being made for disposing of problem oils and emulsions by allowing them to evaporate and burn in the trash incinerator. This does not seem to be an acceptable solution to the problem since this results in significant air contamination levels. Tests on burning waste extraction solvent are also planned for the near future.
January 1962	Sludge [left after boiler or solvent reclaiming] contains combustible oils and greases as well as some chlorinated hydrocarbons. Tests are being run to see if it's feasible to dispose of this solvent by evaporating off the more volatile solvents in the back of the trash incinerator. After these solvents are evaporated, the oils left will ignite and burn to a residue which may be suitable for Plant 8 feed. No health problem has been observed, as yet, in performing these tests. However, they will continue to be followed closely for the possible discharge of uranium or phosgene from the incinerator stack.
February 1962	The tests to evaporate and burn sludges containing chlorinated hydrocarbons have been continuing at the trash incinerator. It was found that if these organics are placed too near the fire and that if the flames could reach the sludge containers, then phosgene is generated. In one instance 20 ppm of phosgene was measured in the flue gas. In another instance 50 ppm of phosgene was measured in the flue gas. The present tests are being carried out with the waste organic containers back far enough from the fire so that it is unlikely that the flames can reach these containers. No phosgene has been detected under these conditions.
May 1962	Samples have been taken from the plume discharging from the trash incinerator while only trash was being burned and while residues were being evaporated and burned. No unusually high uranium concentrations have been found on background samples when no residue was in the incinerator. The remaining samples have not been analyzed yet.
June 1962	Results from sampling the trash incinerator while burning contaminated materials and processing residues indicate average activity (10 samples) was about 300 dpm alpha per cubic meter. This is about twice the level found in background samples when known contaminated materials or residues were not placed in the incinerator. The high sample of the ten was 1100 dpm alpha per cubic meter. *ADS located. Test run 5/25/62.*
July 18, 1962 (Bipes 1962)	Ten air dust samples were taken in the smoke plume emanating from the trash incinerator stack outlet which contaminated slug boxes were being burned and contaminated solvent residue was being burned or evaporated. Results indicated a maximum of 1100 alpha dpm per cubic meter, and a minimum of 61 alpha dpm per cubic meter (average 295). It was felt that the samples collected were quite representative. Four background air dust samples were collected at the same location while burning regular trash. *This letter is discussing same set of data as presented in June 1962 IH&R report.*

September 1962	Air samples taken in the exhaust stream from a vacuum cleaner being evaluated at the incinerator for simultaneously vacuuming and drumming incinerator ash. The air dust level directly in the exhaust stream was 0.4 MAC. An analysis of the material being vacuumed showed the U content to be 6.4%.
March 1964	The methods used to dispose of both stripped and unstripped extraction solvent (TBP-kerosene) at the trash incinerator were checked. The disposal of stripped solvent offers little potential for air contamination, and this process is continuing. However, the disposal of unstripped extraction solvent has been discontinued until samples can be collected to show the effect of this process on air contamination. Arrangements are being made to allow stack gas samples to be withdrawn from the trash incinerator stack.
April 1964	Particulate samples were collected from the trash incinerator flue gas to check the air contamination resulting from disposing of various uranium contaminated organic materials at the incinerator. *ADS located.* It was reported that air contamination from present procedures is tolerable, but it should not be increased by significant procedure changes or an increase in the load of contaminated materials burned at this facility. *See also Starkey (1964b).*
May 1964	A uranium fallout study conducted for one year in the vicinity of the oil burner and the incinerator indicated that fallout near the disposal equipment was between 3 and 7 times higher than the fallout at the nearby permanent station. The volume reduction processes will be further studied and evaluated in the future in regard to uranium fallout with the stack emissions from these operations. *See Klein (1963; 1964) and analyses in text of Appendix K, this report.*
July/Aug 1964	The first burning of formaldehyde solution at the trash incinerator along with stripped extraction solvent was observed. No irritating gas or vapor could be detected or measured around the incinerator or in the diffused smoke downwind from the incinerator. The simultaneous burning of stripped extraction solvent seems to be effective in preventing accumulations of explosive vapor-air mixtures.
1965 (Vath 1965)	Average uranium assay on incinerator ash for the period May through December 1965 was 8.76 ± 3.62% U. Average isotopic assay for the same period was 0.784 ± 0.039% ^{235}U.
1968	Stevenson evaluated uranium content and production of various plant wastes.
1969	Incinerator was operated 3 days per week in 1969, per verbal communication with L. Pennington, 10/8/85. B. Weisman indicated the 3 day per week began after 1965 and no later than 1970. The incinerator had been operated more frequently but was reduced to a 3 day per week operation when the burning of wooden skids was halted. *This information was recorded in handwritten notes.*
February 1970	Modifications made to incinerator which included addition of secondary chamber with after burners, burn-off pan for liquid wastes, air jets, new stack. *See construction proposal CP-69-17 (Anonymous 1969) and discussion in text of Appendix K, this report.*
February 1970	Report "Incineration of Radioactive Wastes" written, summarizing history of incineration operations at the FMPC. *See Anonymous (1970).*
February 1977 Bioassay Lab Monthly Report	A chromel elumel thermocouple sensor and a Hoskins Pyrometer indicator have been provided for temperature measurements of the incinerator off-gas.

March 1977	The incinerator stack was sampled on 3/21/77 and again on 3/24/77. Tentative results show a particulate loading of about twice the amount allowed. Much difficulty has been encountered in sampling the stack because of the extreme variations in temperature and rate of flow encountered. More work and more sampling needs to be done to get a better sample. Bioassay department analyzed two stack samples for particulates, water, and uranium.
April 1977	The incinerator stack was sampled again on April 12, 1977. results show the emission is greater than the limit, but variations in the stack velocity and temperature prevented isokinetic sampling. It is apparent that the incinerator is emitting a larger amount of particulates than allowed.
May 1977	Sampling of the incinerator stack was completed. Results show a particulate discharge of more than 0.1 lb per 100 lb of trash burned. The results of this latest test and comments were sent to the Engineering Division for justification of an improved, all-purpose incinerator.
May 1977 (Ross 1977)	Five tests were made of particulate emissions from the incinerator stack. In all five tests the results were greater than the Ohio EPA limit of 0.1 lb particulates per 100 lb burned. The average loss of uranium was 0.12 pounds per hour (range 0.06 to 0.17).
June 1978	Based on data obtained during incinerator stack sampling last year, an incinerator which meets the present state limit for particulates would discharge 3-11 pounds of uranium per year. This information was given to the Oak Ridge S&EC Division during a conference phone call with the NLO Engineering Division. On the basis of this data, OR decided to drop their previously imposed requirement for an air-cleaning device on the replacement incinerator.
1978	Proposal to move incinerator to a new location inside the production area.
December 31, 1979	Official shut-down date of OSWI given in Boback et al. 1987.

ANNEX 2 TO APPENDIX K

CHRONOLOGICAL HISTORY OF EVENTS AND NOTES RELATED TO RECONSTRUCTION OF SOURCE TERMS FROM BURNING OF CONTAMINATED OIL

Note: The information in this annex was compiled from Industrial Hygiene and Radiation (IH&R) Department Monthly Reports (or reports from Sections within the IH&R Department), unless noted otherwise. Comments by RAC researchers are italicized, to distinguish them from the notes and observations from the FMPC contractor documents.

Date	Event/Notes
January 1959	Industrial hygiene aspects of burning contaminated oil were investigated during January. The results of air dust samples taken in the plume of the burning contaminated oil varied widely however, when it was possible to get enough samples thought to be adequate, they ran far higher than would be desirable for such an operation. The Engineering Division is presently investigating further combustion techniques on non-contaminated oil and after they have found a suitable method for burning with a minimum of smoke, further air dust samples will be taken by this department and recommendations made at that time. *Analytical Data Sheets located. The location of the tests is given as in the open field SE and in the rear of the boiler plant. Dates measurements were made are: Dec 30, 1958, Jan 6, Jan 13, and Jan 15, 1959. Tests appeared to be short-term (i.e. an hour or less). Air samples analyzed for gross alpha activity only. Samples were taken in air at various distances and ranged from not detectable (<0.3) to 1375 dpm alpha per cubic meter. The maximum concentration was measured on Jan. 6, 20 feet from burning oil and about 15 feet off ground, in the body of smoke.*
February 1959	This department is working with the Engineering Division in attempting to find a satisfactory method for burning uranium contaminated oil. A method has been devised by the Engineering Division which permits the oil to be burned with virtually no smoke being evolved. Air dust samples taken under these conditions indicated that there is a good possibility that this oil might be disposed of in this manner without unduly exposing personnel. Another type of burner is to be tried early in March, after which a complete report will be made of the entire study. *Analytical data sheets located for samples taken during burning on Feb 6, Feb 12, Feb 17, Feb 25, and Feb 26. Uranium and gross alpha activity measurements made on oil and in air during burning. On Feb 17, the oil incinerator is described as "constructed of a 30-gallon drum at the base of a stack made up of three 55-gallon bottomless drums which were placed one atop the other." The maximum concentration measured was 2 ft above this stack, at 8500 dpm alpha per cubic meter. On Feb 26, a note was made that condensate from the boiler plant cooling towers was falling in the area.*

April 1959	The burning of oil at the Boiler Plant is progressing in a slow, but what appears to be satisfactory manner. One new method was tried during the month of April, that of burning atomized oil. This proved to be quite unsatisfactory because of the high air dust levels that resulted. It was recommended that this method be stopped immediately, which was done. *See also Starkey 1959:* Air samples collected in heat waves given off during burning in steam atomizing oil burner ranged from 321 to 2913 dpm alpha per cubic meter (average 1479). Since the results were high, recommendation was made not to burn this type of oil in this fashion. *Analytical data sheets located for burn on April 1. Location is given as north of the boiler plant in a home-made burner burning contaminated oil. Three samples taken 200 feet upwind ranged from 2–8 dpm alpha per cubic meter. Nine samples taken within 10 ft of the flame and smoke averaged 1700 dpm alpha per cubic meter (range 509 to 2913). Three samples taken 35 feet downwind ranged from 321 to 1379 dpm alpha per cubic meter.*
May 1959	This department is working with the Water Treatment Department in developing a satisfactory method for the disposal of contaminated oil. It has been determined that the present equipment, operating at a burning rate of 20 gallons per hour or less, can be operated in such a manner so as not to exceed acceptable air dust levels. However, it has been impossible to determine whether or not the equipment is operating properly after dark; therefore, the Water Treatment Department is installing flood lights in order to provide adequate lighting for a 24-hour operation. If the oil can be burned with no visible smoke, there appears to be no accompanying problem of high air dust levels. As soon as the lights have been installed, this department will run a check on the operation to determine if the operators will be able to tell with the naked eye whether a satisfactory operation is being carried out or not. This will permit much more oil to be disposed of in addition to reducing the quantity of uranium being released to the atmosphere by eliminating the high concentrations caused when the burning is begun and terminated each day.
June 1961	A final report on the field test which was conducted to determine the advisability of disposing of contaminated oil by dumping it onto the fly ash pit was submitted to management. *See DeFazio (1961).* The field tests indicate that stream pollution is quite possible, if not inevitable, if large quantities of oil are disposed of in this manner at the FMPC fly ash pit. It was recommended that no further disposal of oil by this method be carried out, since the proximity of Paddy's Run and the fly ash pile makes this a very precarious method of disposal.
August 1961	Oil burning tests are scheduled to start on 9/5/61 for the purpose of determining whether or not a satisfactory procedure can be developed for the burning of uranium contaminated oil in air. A full month's operation is presently planned with more burning possible as required to secure additional health and safety information.
September 1961	Oil burning tests are still being carried out to determine a suitable method of disposing of the back log of contaminated oil. Numerous revisions are being made to the burning rig to improve its performance. Samples of the oil, the residue after burning, and the off-gases from burning have been analyzed and indicate that very little of the uranium in the oil comes off in the off-gas. It is planned to measure the rate of burning, weigh the residue, and analyze the off-gas by an orstat analyzer and by alpha count to evaluate the performance of the oil burning rig. These tests should be completed during October. *Analytical data sheets located for burning on Sept 8, Sept 12, 18, Sept 22, Sept 26, Sept 27, and Sept 29.*

October 1961	The material balance study for the oil burning operation was completed during October. This special test was conducted on 18 drums of oil drawn from the Plant 8 storage tank. This blend had been centrifuged and was one of the easiest oils to burn encountered to date. Visually it appeared as though a high recovery rate was accomplished; however, we are awaiting analytical results before making any final conclusions. *Analytical data sheets located for burning on Oct 11, 13, 14, 16, 17, 18, and 19. October 13th was the beginning of the special material balance test. See text of appendix K for discussion of results.*
November 1961	A report was written summarizing the oil burning tests which have been performed as a possible means of disposing of the backlog of contaminated oil at the FMPC. *These data were located in a draft version of DeFazio (1962) and confirmed by examination of analytical data sheets. See text of this appendix.* The recovery of uranium which could be expected by burning the oil was estimated (approximately 90%). It was found that airborne uranium could be held to acceptable levels for continued burning of a reasonably good grade of oil. It was recommended that future burning be performed using an improved facility. Tests are now being made for disposing of problem oils and emulsions by allowing them to evaporate and burn in the trash incinerator. This does not seem to be an acceptable solution to the problem since this results in significant air contamination levels. Tests on burning waste extraction solvent are also planned for the near future.
January 1962	A detailed design for a new waste contaminated oil burner has been completed. The Engineering Division will guide the construction of this unit, the Production Division will obtain the approval for its construction, and the unit will be built by the Maintenance Department. The Health and Safety Division is closely following these oil burning tests to see if a burner can be operated without too much uranium in the flue gas.
February 1962	Construction of the final oil burner began around the end of February 1962 (DeFazio 1962).
March 1962	The new waste oil burning facility is almost completely constructed. The initial tests of waste oil burning in this facility will be monitored as was done with the previous facility.
3/31/62	*Official start-up date given for oil burner by Boback et al. 1987.*
May 1962	Sampling has been started on the flue gas from the waste oil burner; however, it is not completed at this time. *Analytical data sheets located for May 22 in smoke downwind of burner, but burner was not operating as expected. Concentrations were low (4 and 7 dpm alpha per cubic meter).*
June 1962	The off-gas from the new oil burner has been sampled with various amounts of smoke being emitted from the burner. Results thus far indicate that the presence of smoke indicates uranium contamination in the off-gas as was found with the old oil burner. Light smoke indicates about 500 α dpm per cubic meter. More samples will be collected to show the conditions on starting up and shutting down the burner. *Analytical data sheets located for sampling on June 4, June 8, and June 27. See table in text of appendix.*

July 1962	Samples were taken in the off-gas from the oil burner while it was being started up and after stable operation had been obtained. The start-up samples ranged from 65 to 800 alpha dpm per cubic meter. This was lower than had been expected since some of these samples were taken in heavy smoke. After the burner had reached a stable operation, the levels in the off-gas ranged from 350 to 700 alpha dpm per cubic meter. These samples were higher than expected since there was no visible smoke from the burner for this set of samples. The results from this sampling contradict our previous conclusion that contamination levels can be estimated from the amount of smoke being discharged from the burner. The levels found are higher than desired from the industrial hygiene and contamination control standpoints, therefore further investigation of the procedure will be carried out. *Analytical data sheets located. See table in text of appendix.*
September 1962	A report that the operator of the waste oil burner might be exposed to organic vapors as well as fumes and dust was investigated. A respirator equipped with organic vapor cartridges has been issued.
May 1963	The operation of the waste oil burner was checked several times in May. There was little evidence of smoke or entrained ash in the stack gas, and the burning seemed to be under good control. As a result, no additional stack gas samples were taken. This operation is being considered as a subject for a paper. Additional work will be performed as required to obtain sufficient data for this paper.
June 1963	Oil feed to the waste oil burner was sampled and analyzed for uranium to evaluate the performance of the burner.
July/Aug 1963	Additional monitoring has been done of the off-gas from the waste oil burner. Results confirm previous conclusions that uranium discharge is not excessive under good operating conditions. *Analytical data sheets located. See table in main text of appendix K, this report.*
September 1963	A paper titled "Burning Waste Contaminated Oil" was prepared for presentation at the Eighth AEC Air Cleaning Seminar to be held at ORNL on October 22-25, 1963. *See Brandner et al. (1963).*
May 1964	A uranium fallout study conducted for one year in the vicinity of the oil burner and the incinerator indicated that fallout near the disposal equipment was between 3 and 7 times higher than the fallout at the nearby permanent station. The volume reduction processes will be further studied and evaluated in the future in regard to uranium fallout with the stack emissions from these operations. *See Klein (1963; 1964) and analyses in text of Appendix K, this report.*
August 1964	Plans underway to burn blends of oils contaminated with normal uranium, enriched uranium, and thorium at the oil burner.
1969	During calendar year 1969, approximately 650 drums of waste were processed at the oil burner (Anonymous 1970).
1972	*Proposal to dispose of contaminated oil by applying to the coal pile and burning in the boiler was criticized by DeFazio (1972). Contaminated oil (two samples) contained 1500 and 490 ppm uranium and was certain to contaminate boiler plant equipment. Recommendation was to burn this oil in existing oil burner.*
February 14, 1974	Air sample taken on coal pile SW of oil burner showed 929 $\mu g\ m^{-3}$ particulates, 56 $\mu g\ m^{-3}$ oil and 23 $\mu g\ m^{-3}$ uranium. *Source: analytical data sheet.*
1975	Processing rate estimated at 7500 gal per year waste lubricating oils, spent coolants, etc, and 1,200 gal per year spent TBP-kerosene solvents . *See Stevenson (1975).*

1976	On 5/13/76 a sample of the emission from the oil burner and measurements of its emission rate were obtained. A sample of the particulate emission was collected on a pleated filter for six hours. The emission velocity was measured with a rotating van anemometer. The area of the smoke column and temperatures in the stack and rotometer were estimated since they continually fluctuated. The results of this test are at best an approximation but the results are great enough (1.8 lb/hour) so that if they are high by 50% they are still above the OEPA Standard of 0.2 lb/100 lb. *See Ross (1976). Analytical data sheet located; sample was also analyzed for uranium. Concentration of uranium in off-gas was 45.4 μ g m^{-3}.*
June 15, 1979	*Official shut-down date for oil burner given in Boback et al. 1987.*

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ANNEX 3 TO APPENDIX K

DETAILED INFORMATION FOR RECONSTRUCTION OF URANIUM RELEASES FROM FMPC BUILDING EXHAUSTS

An estimated source term for uranium in building exhausts was projected forward in time from airborne activity measurements made in 1954, 1955, and 1956. The projection to future years (through 1970) was made by scaling the estimated release to certain key production processes which were associated with high airborne contamination levels. See the main text of appendix K for further explanation of the rationale and method. Table K3–1 includes the production data which were used to make this forward projection. Appendix C contains a comprehensive set of production data tables.

Table K3–1. Uranium Production Data (in metric tons) Used For Projected Source Term for Uranium in Building Exhaust between 1957 and 1970

Year	Plant 2/3	4	5[a]	6[b]	8	9[c]
1956	5329	5029	20596	12470	1764	0
1957	8370	9358	18793	15074	1927	0
1958	10039	12117	19476	13665	2018	732
1959	11540	9454	21124	14033	2568	1251
1960	12187	11388	27294	18532	3188	1388
1961	11039	10642	21161	15370	2902	2364
1962	6288	9468	21428	15430	2820	2663
1963	0	10482	24528	14507	2657	3660
1964	0	7203	19303	11313	3505	5297
1965	543	6797	16666	12310	2134	5361
1966	1347	6174	16405	7683	1617	1197
1967	1835	6263	18141	7576	1837	1258
1968	3251	4809	15483	5029	2222	691
1969	2028	2821	10655	3380	1036	778
1970	880	1923	8310	3309	649	499
1971[d]	809	580	3719	1068	307	422
1972[d]	2761	347	2900	0	111	599

[a]Sum of derby and ingot production. See Appendix C.
[b]Rolling operations only.
[c]Ingot production only.
[d]A projection was made through 1972 in order to permit comparison with the backwards projection from 1987 measurements (see text of appendix K). However, for the reconstructed source terms, the forward projection was used through 1970.

Table K3-2. Estimated Source Term (kg U) in FMPC Building Exhausts, 1954–1987[a]

Year	Percentile of Distribution				
	5%	25%	50%	75%	95%
1954	35	68	155	395	728
1955	37	73	218	416	763
1956	37	76	192	394	650
1957	48	100	241	468	801
1958	63	126	277	511	860
1959	69	129	306	603	1007
1960	81	153	366	735	1239
1961	82	152	347	681	1138
1962	74	139	303	643	1072
1963	64	129	277	607	1017
1964	63	129	284	719	1222
1965	62	127	261	520	871
1966	36	69	150	350	590
1967	39	74	158	386	652
1968	32	60	134	421	721
1969	22	41	89	216	364
1970	15	28	60	141	238
1971	2	5	11	24	41
1972	3	5	12	27	46
1973	4	8	18	40	67
1974	5	10	22	49	84
1975	6	11	25	55	94
1976	6	12	27	60	102
1977	3	6	13	29	49
1978	2	4	9	20	34
1979	1	3	7	15	25
1980	2	4	9	20	34
1981	2	4	10	23	39
1982	4	7	17	37	62
1983	4	9	19	43	74
1984	5	10	23	52	89
1985	4	8	19	42	71
1986	5	9	21	46	79
1987	3	6	15	33	56

[a]See text of Appendix K for explanation of approach used for reconstruction of source terms. These data are plotted in Figure K-17.

Figure K3–1 illustrates the typical distribution shape for the annual source term estimate from building exhausts. Monte Carlo sampling (5000 trials) was performed for each year and for the sum of 1957–1970. Uncertainties in the following parameters were propagated:

- Plant-specific uranium concentration in in-plant air
- Dilution factor for exhaust air vs. working area air

These input parameter distributions are provided in the tables and figures which follow Figure K3–1.

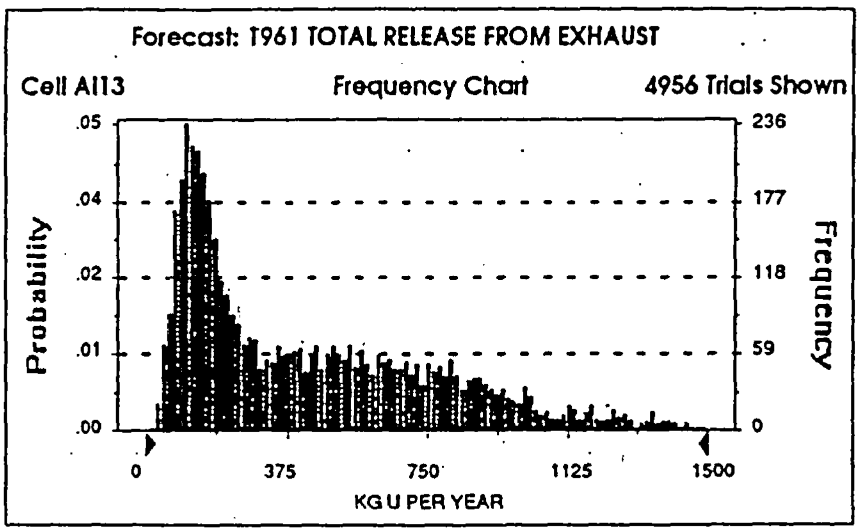


Figure K3–1. Illustration of typical distribution shape for building exhaust source term.

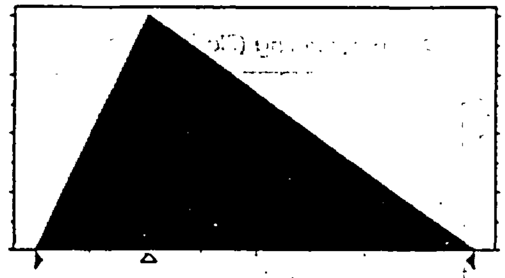
Distributions for Assumptions used in Building Exhaust Uncertainty Analysis

Dilution Factor for Exhaust vs. Working Area Air

Triangular distribution with parameters:

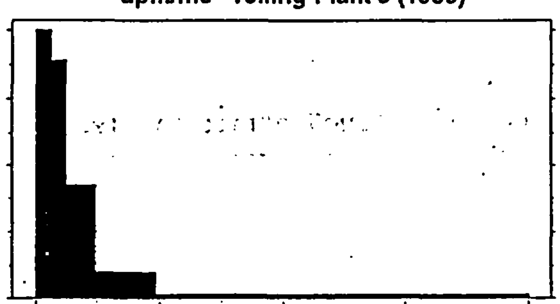
Minimum	0.10
Likeliest	0.33
Maximum	1.00

Selected range is from 0.10 to 1.00



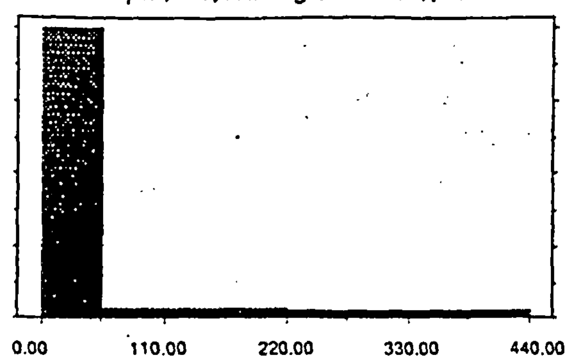
Airborne Uranium Contamination (dpm m^{-3}) Around Rolling Operations (Used for FMPC Plant 6)

Custom distribution with parameters:				Relative Prob.
Continuous range	0.00	to	55.00	0.280000
Continuous range	55.00	to	110.00	0.250000
Continuous range	110.00	to	220.00	0.240000
Continuous range	220.00	to	440.00	0.110000
Continuous range	440.00	to	880.00	0.020000
Continuous range	880.00	to	1,800.00	0.100000
Total Relative Probability				1.000000



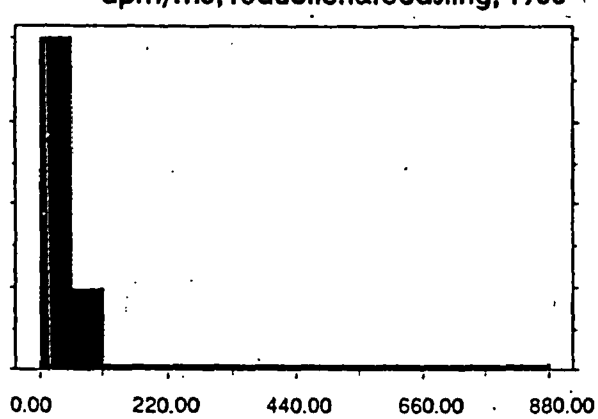
Airborne Uranium Contamination (dpm m^{-3}) Around Refining Operations (Used for FMPC Plant 2/3)

Custom distribution with parameters:				Relative Prob.
Continuous range	0.00	to	55.00	0.820000
Continuous range	55.00	to	110.00	0.030000
Continuous range	110.00	to	220.00	0.060000
Continuous range	220.00	to	440.00	0.090000
Total Relative Probability				1.000000



Airborne Uranium Contamination (dpm m^{-3}) Around Reduction and Recasting Operations (Used for FMPC Plants 4, 5, and 9)

Custom distribution with parameters:				Relative Prob.
Continuous range	0.00	to	55.00	0.780000
Continuous range	55.00	to	110.00	0.190000
Continuous range	110.00	to	220.00	0.010000
Continuous range	220.00	to	440.00	0.010000
Continuous range	440.00	to	880.00	0.010000
Total Relative Probability				1.000000



Airborne Uranium Contamination (dpm m^{-3}) Around Scrap Recovery Operations (Used for FMPC Plant 8)

Custom distribution with parameters:				Relative Prob.
Continuous range	0.00	to	55.00	0.350000
Continuous range	55.00	to	110.00	0.120000
Continuous range	1,800.00	to	3,000.00	0.340000
Total Relative Probability				0.810000

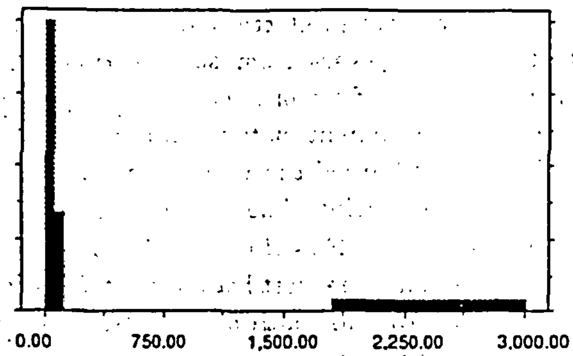


Table K3-3. Ventilation Capacities and Fan Operating Factors used in Building Exhaust Estimates

Plant	Ventilation Capacity (cfm)[a]	Fan Operating Factor[b]
1	62,000	0.08
2/3	262,500	0.33
4	316,050	0.42
5	62,000	0.33
6	126,000	0.36
8	91,000	0.33
9	219,150	0.25

[a]From Hill 1989c. Maximum ventilation capacity of exhaust fans.
[b]From Hill 1989c. Fraction of year in which fans were assumed to operate at maximum capacity.

For each plant, the annual release rate was computed using the following equation:

Annual release (kg U y^{-1}) = Concentration in working areas (dpm U m^{-3}) × Ventilation Capacity (ft^3 m^{-3}) × Dilution factor (unitless) × Fan Operating Factor (unitless) × (2.83×10^{-2} m^3 ft^{-3}) (5.256×10^5 min y^{-1}) (1×10^{-3} kg g^{-1}) ÷ [(2.22×10^{12} dpm Ci^{-1})(6.8×10^{-7} Ci U g^{-1})].

APPENDIX L

SURFACE WATER DISCHARGES

INTRODUCTION

Liquid wastes that are generated at FMPC come from three main sources: process water, sanitary sewage, and storm water. Detailed descriptions and diagrams of some of these processes are available (Pennak 1973). These waste streams from the FMPC facility include sump water from the plant production areas, waters from the waste pit area, and waters flowing into the storm sewers from surface runoff over soil contaminated with uranium from spills or deposition of airborne effluents. Liquid effluent streams from FMPC are released to the offsite environment at two locations. These include: (1) The combined sewer and process effluents discharged through the main effluent pipeline at Manhole 175 into the Great Miami River at a point almost directly east of the plant site. This point is about 3 miles (5 km) upstream from New Baltimore; (2) Paddy's Run Creek, a small stream with intermittent flow, lying along the west boundary of the site that joins the Great Miami River approximately 1.5 miles (3 km) south of the FMPC, which received discharges from the storm sewer outfall ditch, and surface runoff from a portion of the production area. The flow in Paddy's Run Creek generally exists only during the period January to May. For the balance of the year it is considered a dry stream bed with occasional flows of a few hours to a day following heavy rains (Patton 1985). Figure L–1 shows the general features of the liquid waste discharge points from the FMPC site.

Initially, source term estimates and uncertainties for surface water discharges were derived for the 1960 to 1962 period and presented in an interim draft report (Voillequé et al. 1991). Based on the sources of information and data for that time period, we developed methods for estimating uranium releases to the Great Miami River and to Paddy's Run Creek on a monthly basis. In the present report, we use similar methods of investigation to derive source term estimates for uranium and other radionuclides discharged in liquid effluents from the FMPC for all years of operations. These estimates are reported on an annual basis and the data from original analytical data sheets and other records are tabulated in an annex at the end of this appendix. The tables of daily or monthly data, presented as Tables L1–1 through L1–36 in the annex, will be referenced in the appropriate sections of this report. Much of the background information provided in the interim draft report for the early sixties is presented in this report as well.

FACILITIES FOR HANDLING LIQUID EFFLUENTS

General Sump System

Each of the individual production plants at the facility had collection sumps and treatment equipment to remove the uranium and thorium from the process waste water. After sampling and analysis was performed to check that uranium content was within pre-set allowable discard limits (in the sixties, these were pH > 6.3 and uranium concentration < 0.01 g L^{-1} or 0.05 g L^{-1} depending upon the source of effluent) (McCreery 1965), the filtrate was pumped to the General Sump. Thorium wastes were segregated, co-precipitated with barium carbonate and aluminum sulfate to reduce ^{228}Ra activity and then pumped to the wet chemical pit (Pit 3 until 1968, Pit 5 after late 1968) (Keller 1978). From here the water passed to the chemical waste pit where settling occurred, and the liquid was decanted to the clearwell portion of the pit before discharge through Manhole 175 which carried it by pipe to the Great Miami River.

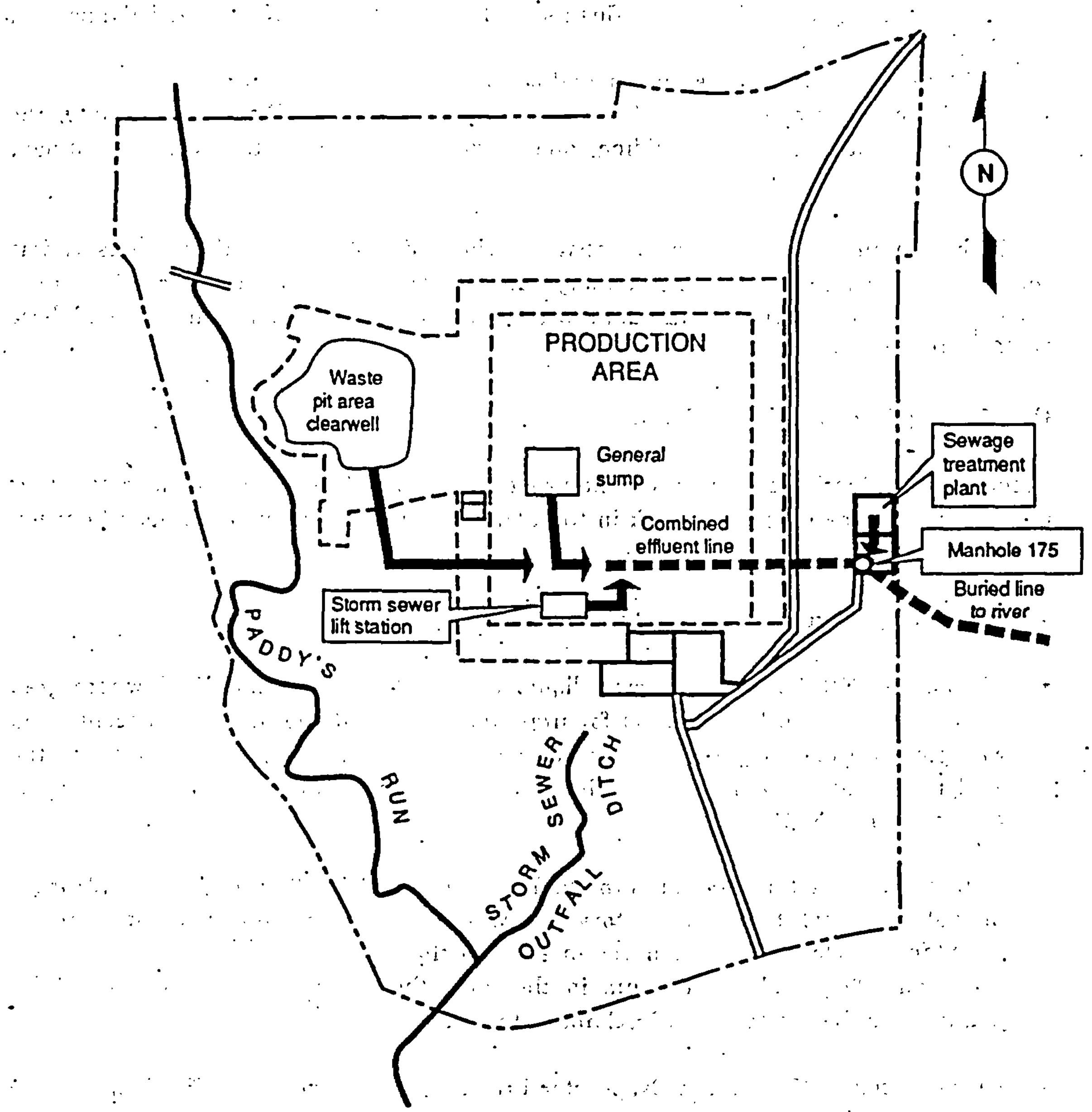


Figure L–1. Liquid effluent flow and discharge points from the FMPC site.

In the early years of facility operation, the General Sump System consisted of three 20,000 gallon receiving tanks (F18-1, F18-2, F18-3), one 5,000 gallon receiving tank (F18-4), and three 50,000 gallon settling tanks (F18E-1, F18E-2, F18E-3) (NLCO 1957). The settling tanks were installed in late 1956 which accounted for the reduction in contaminants released in the river (Starkey 1958a). The functions of the receiving and settling tanks are summarized below.

Three 20,00 Gallon Receiving Tanks:

- F18-1 received effluents from the Refinery sump area, condensate from the digestion area, sampling plant (Plant 1) effluents, and in emergencies, Neutralized Evaporated Product from Plant 2/3.
- F18-2 received Pilot Plant effluents, and when necessary, Plant 8 filtrates.
- F18-3 received waste streams from contaminated sewers of Plants 5, 6 and 9, the Decontamination pad and building, and condensate return to the Water Treatment Plant.

If the uranium concentration was above the limit of 0.01 g L^{-1} in these tanks, it was sent back to Receiver Tanks in the Refinery Sump of Plant 2/3 for further processing. If the waste was within the pH and uranium concentration limits, it was pumped to one of three 50,000 settling tanks.

One 5,000 gallon Receiving Tank:

Received high fluoride content waste liquors from Plant 4. Then the effluent was either pumped back to the neutralizer tank in the Plant 2/3 Refinery Sump, or pumped to one of the settling tanks.

Three 50,000 gallon Settling Tanks:

- F18E-1 and F18E-3 received waste liquid from F18-1, F18-2 and F18-3 where grab samples were taken from the top for uranium analysis. If the uranium concentration was greater than 0.02 g L^{-1} it was designated a "rush" sample, and taken to the analytical laboratory for total soluble and insoluble uranium analysis and pH measurements (NLCO 1957).

  If the estimated total uranium in the tank was greater than 100 pounds (e.g. 0.24 g L^{-1} in 50,000 gallons) it was "mandatory to notify the Plant superintendent" according to the Standard Operating procedures in effect at that time (NLCO 1957). If there were less than 100 pounds of uranium in the tank, the sump supervisor could use his judgment on the possibilities of reclaiming the uranium.

- F18E-2 received Neutralized Evaporator Product (NEP) from Plant 2/3. Samples were taken from a bottom valve. If the concentration was above the limit of 0.01 g U L^{-1}, the effluent was sent back to the Plant 2/3 refinery sump. If below the limit, the effluent was pumped to either of the other two 50,000 gallon tanks (F18E-1 or F18E-3).

In 1968, major improvements were made in the General Sump area for waste effluent processing facilities involving the installation of two new 15,000 gallon sludge settling tanks with hopper bottoms and decanting pipes; a new 50,000 gallon sludge settling and decant tank with a flat bottom; and a new head tank for regulated continuous discharge to the river (OHIO 1968).

Individual Plant Sumps and Normal Operations

The descriptions of the individual plants which follow provide an overview of liquid effluent flow at FMPC. The liquid effluent volume and uranium releases from the various site facilities were provided in monthly loss reports (Yoder 1955, Cuthbert 1960–1961, Marshall 1963, Schwan 1967–1984). Table L–1 provides monthly data on uranium quantities in effluents to the General Sump from the process areas. Although these data are from the early sixties, the relative fraction of uranium discards remained fairly steady over the years.

Plant 1. Due to the infrequency of pumping of liquid effluent from Plant 1, effluent was usually pumped to the Plant 2/3 Refinery Sump Receiver Tank (Fl-608) for recovery of uranium (Cahalane 1961).

Plant 2/3. Three waste streams from Plant 2/3 are important: the sump effluent, the Neutralized Evaporated Product (NEP), and the slag leach slurry from the refinery. While the volume of Neutralized Evaporated Product (NEP) was measured as it was pumped to the General Sump, the Plant 2/3 sump effluent volume was calculated by subtracting the sum of all other individual plant discards into the General Sump from the total volume pumped from the General Sump to the chemical pit. The Plant 2/3 Sump Effluent accounted for roughly 70–80% of the total volume sent to the General Sump, and 25–30% of the uranium in effluents. Table L–1 shows that the NEP waste stream contributed over 60% of the uranium to the General Sump each month, but only 5% of the total volume. The slag leach slurry was pumped directly to the chemical waste pit.

Plant 4. Waste liquors from plant 4 which were high in fluorides but rather low in uranium, were pumped directly to the only 5000 gallon tank in the General Sump (F18-4). Routinely, Plant 4 contributed less than half a percent to either the volume or total uranium quantity each month.

Plant 5. Liquid waste from the remelt or casting area accounted for approximately 1–2% of the volume, and less than 1% of the uranium, sent to the General sump (Tank F18-3).

Plant 6. Contaminated effluents from the machining area were pumped to the General sump (Tank F18-3), contributing on the average 5% of the volume and less than 1% of the uranium to the General Sump. The Heat-Quench Water from the Metal Fabrication Area was pumped directly to the wet chemical pit.

Plant 8. Routinely, effluents were pumped directly to the waste pits from Plant 8, and are not listed in Table L–1. In an emergency when discard limits were exceeded, they were pumped through the General Sump (Tank F18-2) for processing and sampling (Cahalane 1961). Because this was an infrequent occurrence, Plant 8 effluents contributed less than a half percent to the volume and uranium totals of the General Sump. However, records summarized in Appendix M indicate that Plant 8 contributed approximately 1200 kg per month directly to the waste pits during 1960, 1961 and 1962.

Plant 9. Approximately 1–2% of the volume (< 0.1 % of the uranium) to the General sump (Tank F18-3) contained enriched uranium from Plant 9 (Special Products). The waste stream from the Zirnlo Slurry was routed directly to the wet chemical waste pit.

Table L-1. Uranium Discards (kg) to the General Sump From Process Areas[a]

Date	Plant 2/3 Effluent	NEP	Plt 4	Plt 5 Cast.	Plt 6 Mach	Pilot Plant	Anal. Lab	Decon Area	Plt 9 (Enr)	Total
1960	6406	15312	35	152	54	920			24	
1961	5511	17144	54	81	290	2830			20	
1962	3874	4283	32	108	245	560			16	
Total	15791	36739	121	340	590	4310	1105	85	60	59140
% of Total	27	62	<0.2	<0.5	1	7	2	<0.1	<0.1	100

[a] From NLCO 1960–1962.

Pilot Plant. Waste effluents from the Pilot Plant refinery, which contained enriched uranium, were pumped to General Sump (Tank F18-2) before being pumped to the pit. Several different waste solutions from at least seven or eight different areas of the Pilot Plant were discharged into the sump including the tin decladding decantation liquors, 3620 area caustic scrub solutions, Winlo filtrate, extraction area raffinate, open air reduction rotoclone scrubber solution, derby shock wastes, and runoff from outside storage pad areas (Cseplo 1961). Only the first two solutions were neutralized to a pH of 7 or higher before being pumped to the sump. Discards from the Pilot Plant were variable from month to month, contributing from as little as 2% up to 10% of the total volume, and from 2% to 9% of the uranium quantity to the General Sump.

Surface and subsurface drainage in the Pilot Plant Area, however, flowed into a manhole on the warehouse storage pad, and then, by gravity, into an open drainage ditch which discharged into Paddy's Run Creek (DeFazio 1962). Analysis of samples indicated that uranium concentrations varied from 7 to 28 ppm with some flows over 5 gallons per minute to the ditch.

Decontamination Building and Area. Effluents from this area were variable, but usually contributed less than 1% of the volume, and up to 3% of the total uranium quantity to the General Sump in some months.

Analytical Laboratory. Approximately 10% of the volume and 3% of the uranium discharged to the General Sump each month came from the Analytical Laboratory.

There are three process waste streams from the plants which are routed directly to the wet chemical waste pit. They were:

1. Zirnlo Slurry from Plant 9 (Special products)
2. Heat-Treat Quench Water from Plant 6 (Metal Fabrication)
3. Slag Leach Slurry from Plants 2/3 (Refinery).

Chemical Waste Pits

Six chemical waste pits have been constructed since operations began at the FMPC. Pits are identified by number based on chronological sequence of their construction, and by type, "dry" or "wet" pits depending upon the main type of material discarded or discharged. Pits 1 (1,080,000 cubic feet) and 2 (351000 cubic feet) were dry, although some wet materials were added to Pit 2 just prior to completion of Pit 3. Completed in 1959, Pit 3 (6,115,500 cubic feet) was designated a wet chemical pit, and received effluents from the General Sump (Settling Tanks F18E-1, F18E-2, and F18E-3) until it was filled in 1968 (NLCO 1974).

Pit 4 (1,431,000 cubic feet) was built in 1960 as a dry pit. A tabulation of recorded monthly discards of dry and wet wastes to the pits for the time period 1960 to 1962, and annual totals for 1952 to 1974 is located in Appendix M. Characteristics of the waste pits and a description of the methodology used to estimate atmospheric releases from them are given in Appendix K.

In the early years, two overflow lines with valves extended from the "fluoride" pit (Pit 3) to a short tributary of Paddy's Run that lies just west of the pit. In a site review by the US Department of the Interior, Theis (1955) noted that these outlets were apparently not used customarily, and that the tributary and Paddy's Run were usually dry. He did suggest the possibility of groundwater contamination from the waste pits (See Appendix M).

Sanitary Sewage

The sanitary waste collection and treatment system was a completely separate system from the process waste system. The sewage was treated in a recirculating trickling filter facility, originally sized for 750,000 gallons per day (gpd) but by the late 1970s was receiving only about 125,000 gpd (Keller 1978). The sewage sludge was then incinerated onsite (Pennack 1973). Sampling and analysis were performed on the waste stream before it joined the other effluent streams at Manhole 175. Daily records of waste volume discharged, river flow and calculated concentrations of uranium, nitrates, and fluorides added to the river were maintained, and reported monthly to the Ohio Department of Health (Carr 1955, Walden 1957, Flowers 1960–1961, P&G 1985).

The Chemical Feed Sump from the Water Treatment and the Boiler Plant Area was sampled for Nuclear Materials Control (Starkey 1964a). The results routinely indicated that the stream, although high in volume (approximately 90,000 gallons per day), contributed approximately 5 pounds (2.5 kg) uranium per month to the river.

Storm Sewer System

The storm water system consists of a grid work of catch basins and about 70,000 feet of buried pipe lines which drains the surface runoff from the immediate vicinity of the processing areas of the facility, a 5,500,000 square foot area (Nelson 1971). Although it was assumed, when operations began in 1952, that the storm sewer system would handle only water, recommendations to install a storm sewer lift station were frequent when sampling of storm sewer drainage indicated uranium contamination. The initial storm sewer system included a storm water detention basin and sump to handle small quantities of contaminated liquids, but no provisions had been made to empty the sump (Quigley 1952).

The detention sump had not been placed in service by February 1954 (Ross et al. 1954). In late 1955, a Storm Sewer Lift Station, located about 2800 feet south and 4100 feet east of the center of the production site (Theis 1955), near the southern end of the system, was installed (OHIO 1955). It was designed to divert and pump waste water flows in the storm sewer system to the process waste discharge line (Manhole 175) to the Great Miami River. A recording flow meter and continuous proportional sampler monitored the discharges, and provided daily data for uranium and liquid effluents discharged to the Miami River from that point (Pennack 1973).. Since the storm sewer lift station was not connected to any process, all the uranium lost through it was assumed to be from leaks and spills (Ross 1972). The lift station in place in the early years was designed to take only the initial runoff during a heavy rain. The pumping capacity of the system was approximately 500,000 gallons per day or 350 gallons per minute (DeFazio 1960).

Throughout the late 1950s and 1960s, daily storm sewer samples continued to reflect spills or releases of radioactive process effluents and chemical materials (Starkey 1961a). As a consequence, the majority of the uranium and radioactivity in the combined plant effluent originated from the storm sewer. When the capacity of the storm sewer lift station was reached, water overflowed through the storm sewer outfall to Paddy's Run Creek, a small intermittent stream lying along the west boundary of the site that joins the Great Miami River approximately 3 km south of the FMPC. The volume of storm water that overflowed the storm sewer lift station to Paddy's Run was related to rainfall amounts and patterns. Storm water flow lagged the actual precipitation event by several hours, usually showing an increase in flow the next day (Patton 1985).

Memoranda and various reports suggest growing concern about the liquid effluent handling system at the FMPC from the mid–1950s onward. Table L–2 summarizes the major changes that were proposed and undertaken in response to many of the considerations about unmonitored runoff to the storm sewer and to Paddy's Run. By the late 1960s, water at the Storm Sewer Lift Station was sampled by two proportional automatic samplers: one sampled effluents going to Manhole 175, while the other was activated by an overflow of water going to the storm sewer outfall ditch to Paddy's Run Creek(Nelson 1971). Both samplers were equipped with recording flow meters.

DOCUMENTATION OF LIQUID WASTE DISCHARGES FROM FMPC

Appendix A outlines the sources of information and the types of documents that were found in a variety of repositories around the country for use in the completion of this project. A significant number of documents were related to the liquid effluent system onsite and uranium discharges in liquid wastes from the site because these losses were documented rather thoroughly over the years. Specific documentation is referenced throughout the report. In this section, the documentation used in compiling daily or monthly data for liquid effluent discharges for all years of operation are described briefly.

Table L–2. Major Changes in the Liquid Effluent Handling System at the FMPC

Date	Modification to System
Oct 1951	First Operations at the FMPC; Storm Sewer System with detention basin and sump installed, but detention basin sump not yet in service in 1954. • Process Effluents to River–Measured • All Runoff to Paddy's Run–Periodically Measured
Feb 1954	Recommendation to install a continuous sampler at the discharge point to the river (MH 175)
Jul 1955	Storm Sewer Lift Station Installed • Process Effluents & Most Runoff–Measured • Some Runoff & Storm Sewer Overflow–Not Measured
May 1962	Recommendation to install sampler and flow meter in Paddy's Run near Willey Road at southern plant boundary (Jeffers 1962).
Nov 1965	Recommendation to install sampler and flow meter at the storm sewer outfall ditch (Starkey 1965c)
Jan 1966	Installation of pH cell and recorder in Storm Sewer Lift station; alarm sounds in Water Plant when a high or low pH recorded (Riestenberg 1966).
May 1966	Renovations to outfall pipe to the river so that discharge of the FMPC effluent is in deep portion of the stream (Starkey 1966a).
Aug 1968	Storm Sewer Ditch Monitor Installed • Process Effluents, Runoff & Overflow Measured • Some Runoff to Paddy's Run Not Measured
Fall 1968	New tanks installed and key improvements in effluent handling at the General Sump
Jan 1969	Waste Pit 5 opens, replacing Pit 3 which had been at capacity for months
Apr 1973	Renovations to outfall sewer to river (CP-73-8) caused by "wear, tear, decay, and action of the elements".
Aug 1986	Storm Water Retention Basin Installed with capacity of 6 million gallons and emergency spillway overflow at 365 feet.

- Original analytical data sheets from the Health and Safety Division for various times from 1954 through 1974 provided uranium, radium and thorium concentrations on a daily, weekly, biweekly or monthly basis on daily or composite samples taken at the MH 175. Similar data sheets provided concentration results for uranium at the Storm Sewer Lift Station.

- "Discharge of Liquid Wastes into the River" (DLW), was a monthly report listing the daily discharge of liquid wastes from the Sanitary Sewer, Storm Sewer, Manhole 175, and Storm Sewer Outfall. Measured volumes and uranium concentrations were listed on a daily basis for these waste streams.
- "Measured Losses and Removals of SS Material From the Production Stream" (MLR) reports, changed to "Routine Operating Losses" report in 1964, provided a monthly summary of uranium discards to the General Sump and stack losses. Volumes and quantities of normal and enriched uranium discarded as liquid waste from each process area are listed for the month. In addition, the MLR reports give the losses to Paddy's Run, discards to the chemical or wet pit, and effluents pumped from the clearwell of the pit to the river. Many of these reports were located covering all years of operations.
- Descriptive reports on key topics were prepared by different departments on a regular basis. Monthly river and effluent flows, and concentrations of uranium and other contaminants in effluents at Manhole 175, the storm sewer, the waste pits, and Paddy's Run outfall were provided in a monthly report, "Comments on Monthly River and Effluent Flow". The Industrial Hygiene and Radiation Department issued monthly reports describing various radiation and air dust studies, stack losses, environmental sampling activities, liquid effluent measurements in the river, and special investigations of problem areas at the facility. Finally, "Aquifer Contamination Control" Reports to the Manager provided quarterly highlights of contamination problems or action taken to improve the effluent control system at the storm sewer, the General Sump, the pit area. the river and the test wells (Starkey 1965a, 1965b, 1967a, 1967b, 1967c, 1968).
- "Comments on Ground Contamination" biweekly reports described ground contamination areas onsite, results of ground contamination surveys of process areas, and charted estimated uranium losses to the storm sewer and rainfall totals for the month. These latter types of reports, which are more descriptive in nature, have been useful in providing background information for conditions that existed at the site in the early years, and in highlighting unusual events and unplanned releases, and are referenced at appropriate locations within the text.

ESTIMATES OF URANIUM DISCHARGED IN LIQUID EFFLUENTS VIA MH 175 TO THE GREAT MIAMI RIVER

Uranium in liquid effluents leave the FMPC production area by the main effluent line to the Great Miami River or to Paddy's Run Creek via the Storm Sewer Outfall Ditch (SSOD) or runoff from the west side of the production area. Principal contributors to these uranium-bearing effluents included storm sewer runoff, effluent from the clearwell of the liquid waste pit, and treated effluent from the sanitary sewage treatment plant. To calculate the quantity of uranium lost from the FMPC, two key measurements are necessary:

- the concentration of uranium, and
- the volume of effluent to the river (MH 175) or to Paddy's Run.

The total uranium discharged each day via MH 175 to the river was calculated by multiplying the daily uranium concentration (mg L^{-1}) and the volume of water discharged per day (liters). For Paddy's Run Creek discharges, the measured concentration of uranium and the total volume to the creek taken during specific outfall events, i.e., heavy rainfalls, or for a particular month were used to estimate uranium losses. The uncertainty analyses of these computations are discussed in a later section. Figure L–2 shows the annual uranium release estimates to the Great Miami River and to Paddy's Run Creek for all years. This and the next major sections of this appendix describe the documentation, methodology, and uncertainty analyses computations employed to arrive at these estimates. Data on uranium concentration in liquid effluent taken at MH 175 before discharge to the river are shown in Tables L1–1 to L1–13 in the annex for 1954 through 1969. The results of uranium concentration measurements in the storm sewer and storm sewer outfall ditch to Paddy's Run Creek for 1954 to 1966 are displayed in Tables L1–14 to L1–22 in the annex.

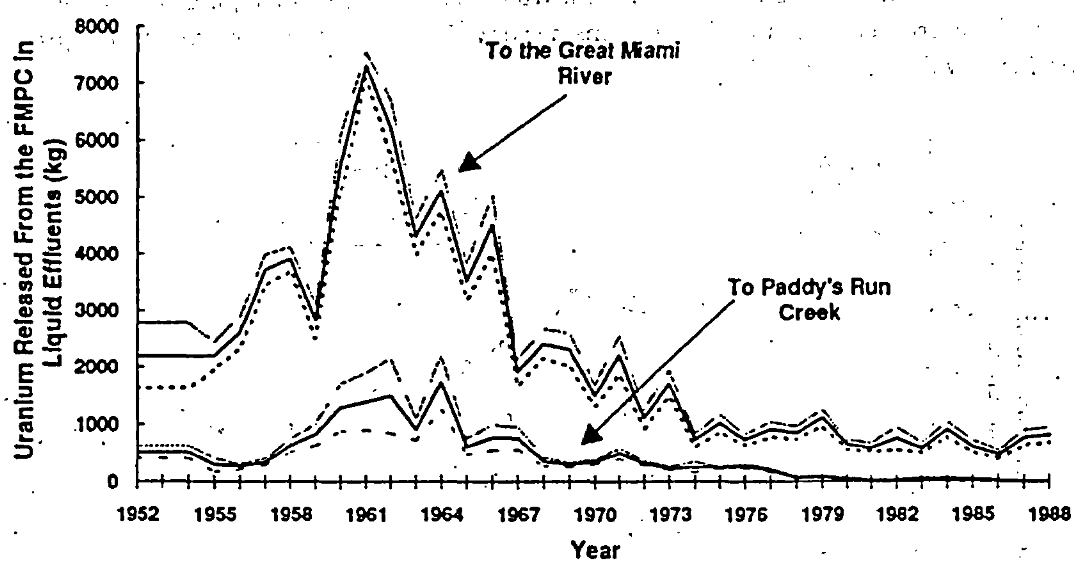


Figure L–2. Uranium losses to the Great Miami River via Manhole 175 and to Paddy's Run Creek from the FMPC for all years of operation. The uncertainty of each estimate is described by the 95th percentile (top, broken line), and the 5th percentile (lower, dotted line).

The magnitude of the uranium releases to the river peaked in 1961 with 7300 ± 140 kg uranium. From 1974 onward, the annual releases were below 1000 kg. The uranium losses to Paddy's Run show much more month to month variation than do the uranium loss estimates to Manhole 175. However, the average quantity of 500 kg uranium discharged through Manhole 175 to the Great Miami River each month during the early 1960s (Table L –3) was roughly five times greater than the average quantity of 100 kg of uranium lost to Paddy's Run during that same time (Table L–6). The volume of effluent to Paddy's Run averaged from 2 to 3 million gallons per month during this time period, while Manhole 175 discharged approximately 30 to 40 million gallons each month during the same period (Figure L–3).

Figure L–3 compares the monthly average liquid effluent flow from the FMPC to the river and to Paddy's Run for all years. The average volume of liquid to the river via MH 175 from the FMPC shows a gradual decrease from 30 to 35 million gallons (110 to 130 million liters) per month in the early sixties to about 15 million gallons (60 million liters) per month in the seventies and eighties. The highest average volume of effluent to the river through the main discharge pipeline (1,400,000 gallons per day) occurred in 1961. Average monthly effluent flow to Paddy's Run is approximately ten times lower than the flow directly to the river, although flow from the site to the storm sewer outfall ditch generally occurs only during heavy rainfall events. The relative difference in flow and variation from month to month can be seen in Tables L1-6 to L1-8, which list the daily and monthly volumes for 1960, 1961 and 1962 to the river, and in Tables L1-18 to L1-22, which list effluent volumes to Paddy's Run for 1960, 1961, 1964 and 1966. These monthly variations in volume are typical of other years as well. Table L1-36 lists the annual effluent volume totals to the river and to Paddy's Run for 1959 to 1984.9

The volume of effluents discharged through Manhole 175 did not show great variation for most months. It was fairly consistent from day to day, showing a gradual decrease over time from greater than a million gallons per day (MGD) in the early sixties to approximately half that volume since 1976.

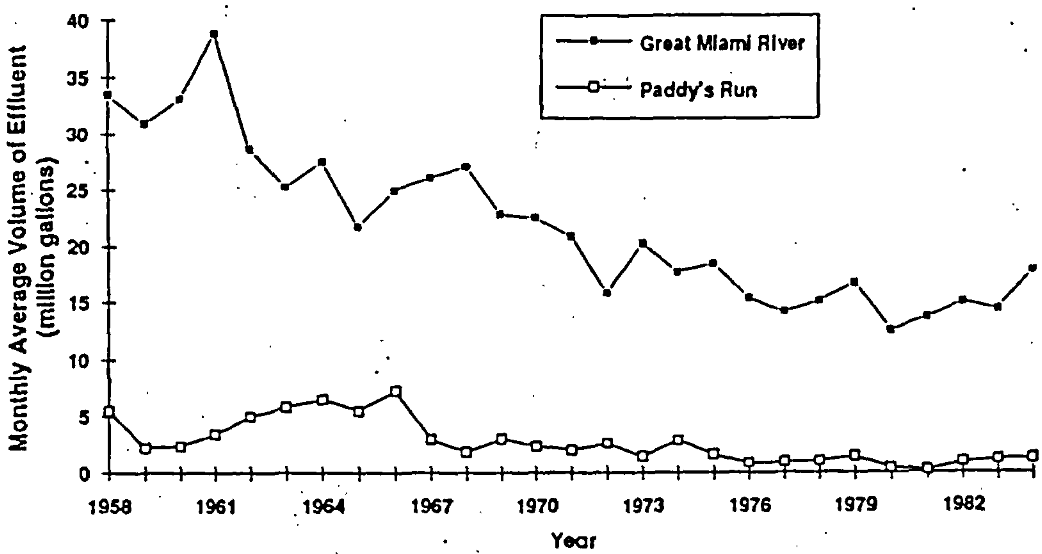


Figure L–3. Comparison of the monthly average volume of effluent to the Great Miami River and to Paddy's Run Creek from 1958 to 1984.

Discharges to the Great Miami River Via Manhole 175

Manhole 175 (MH 175), located on the eastern side of the facility, is the discharge point for waste water leaving the site through the main effluent line to the Great Miami River. MH 175 is the final junction point of the major waste effluent streams from the facility. This station is equipped with a recording pH meter, and a Parshall flume flow station equipped with a recirculating sampling line. The discharge flow to the Miami River was continuously measured and a composite sample collected and analyzed on a daily basis. The total uranium discharged each day was calculated by multiplying the daily uranium concentration (mg L^{-1}) and the volume of water discharged per day (liters). The uncertainty analysis of these computations are discussed in a later section.

For discharges to the river, both of these quantities were known on a daily or monthly basis for most years of operation, except for 1952 to 1954. Daily uranium concentration measurements on 24-hour composite samples from Manhole 175 for 1954 through 1969 were located, and used in the source term derivation. For the occasional day or month when data sources were not located, an average value for that time period was assumed. Uranium concentration measurements from original analytical data sheets from 1954 through 1969 are listed in Tables L1–1 to L1–13 in the Annex. In addition, Tables L1–6 to L1–8 contain the daily volume measurements from MH 175 to the river. For the interim source term derivation for 1960 to 1962 (Voilleque 1991), daily volume measurements were available for most of 1960 and 1961 (February, April, May, July–December 1960 and January–August 1961) in DLW monthly reports, monthly volume measurements were available from MLS reports (Cuthbert 1960–1961), and from monthly ledger tabulations (Rathgens 1974). An equivalent procedure was followed for all years, with MLS reports, routine operating loss reports and analytical data sheets providing the basis for calculating losses to the river and to Paddy's Run.

Figure L–4 shows the daily uranium concentration and volume measurements taken at MH 175 before discharge to the river for July through October 1960 as an example of the type of variation seen in these parameters. Whereas, daily uranium concentrations varied by a factor of 10 during this period, the effluent volume was more constant. Figure L–5 shows that, over time, the uranium concentration at MH 175 decreased gradually with less variation seen on a day to day basis. The concentration of uranium in the liquid effluent is higher, and shows more daily variation in 1957 than in 1967. In 1967 the daily uranium concentration ranged from 1.5 to 6.6 mg L^{-1}, compared to 1957 where concentrations as high as 20 mg L^{-1} were seen (See Tables L1–3 and L1–12 in the annex).

Uncertainties Associated With Discharges to Manhole 175

Sources of uncertainty for the estimates of losses of uranium through Manhole 175 to the Great Miami River come primarily from the analytical errors in measurement of flow, and in sampling and determination of uranium concentration in the water. Generally, there were differences of 10% or less in the unaccounted–for volume going into Manhole 175 from the various areas onsite. It appeared that the effluent volume to the river was monitored reasonably well (Courtney 1965). Estimates of error for the daily uranium concentration measurements, imprecision in sample preparation for the fluorometric uranium analysis, and volume measurements were made regularly (Brown 1967).

Uranium Measurements. For the fluorometric analysis of uranium, the limit of error (LE) at the 95% confidence level was reported as ± 7.1 mg U L^{-1} at the level of 25 mg U L^{-1} (28%) in the mid–1960s (NLCO 1966). Control samples indicated the precision and bias of the method for an individual analysis, and were routinely analyzed in a "manner similar to the US AEC GAE program samples". These control samples had a LE of ±10.3 mg U L^{-1} (bias of +0.2 mg U L^{-1}) at the level of 50 mg U L^{-1} (21%). The minimum detectable level of uranium by fluorometric analysis was approximately 0.5 mg L^{-1}.

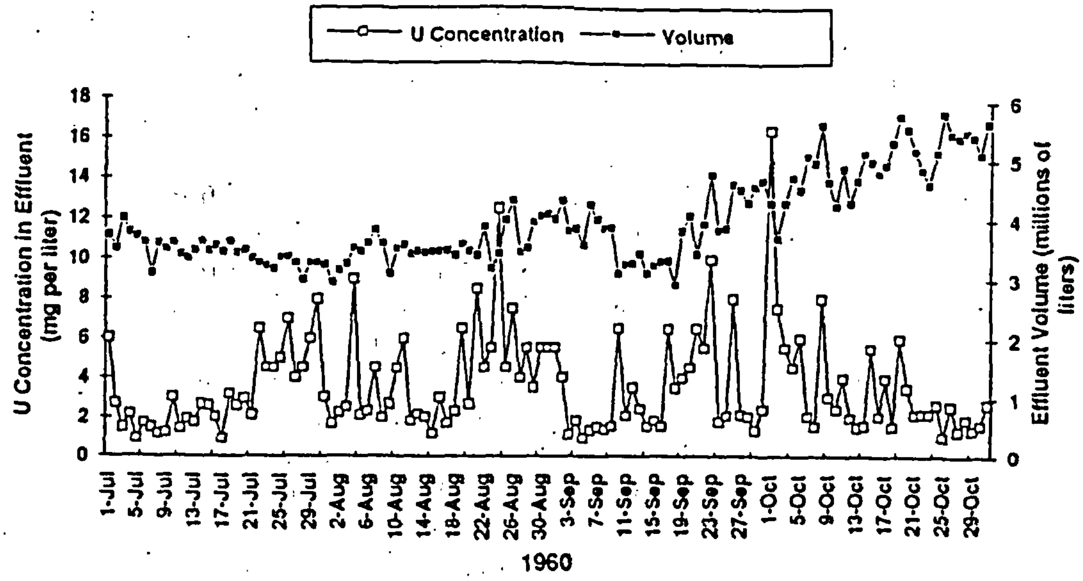


Figure L–4. Daily uranium concentration (left axis) and volume of liquid effluent (right axis) released to the river for four months in 1960. This figure illustrates the difference in variation seen in uranium concentrations and volume of effluent seen in early years. Whereas the concentration varied by a factor of 10, the effluent volume was more uniform, increasing gradually by a factor of 2 during this period.

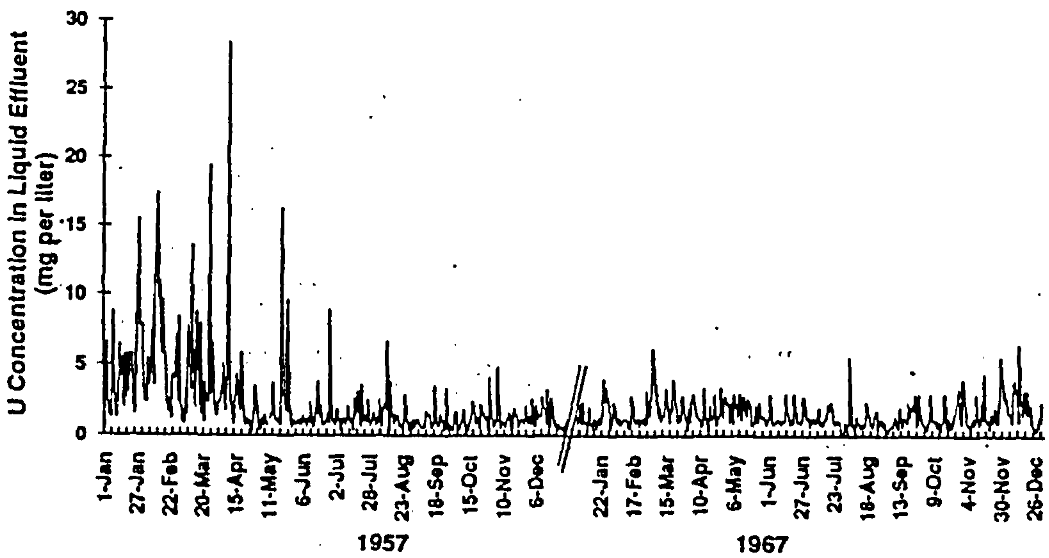


Figure L–5. Comparison of daily uranium concentrations measurements at the discharge point to the river from 1957 and 1967. The annual average concentration in 1957 was 2.5 ± 3.1 mg L^{-1}, compared to that in 1967 of 1.5 ± 1.0 mg L^{-1}. The extremes in concentrations decreased in the 1970s and 1980s.

The uranium concentration of 24-hour composite samples from Manhole 175 generally averaged from 2.5 to 5 mg U L^{-1}, about 5 times lower than measurements used for LE determinations (Tables L1–1 to L1–13, Annex). Consequently, the relative LE for the Manhole 175 uranium concentration measurements would be expected to be higher as a percentage of the uranium concentration. Based on the measured error limits, and on discussions with individuals from the analytical laboratory at FMPC, the errors associated with the daily uranium concentrations was assumed to be 50% at the 95% confidence level for the 1950s and 1960s. We assume that the daily measurement value represents the mean of a normal distribution of values. Thus the relative standard deviation for each daily measurement is assumed to be 50% divided by 1.96, or 25.5%. For the seventies and 1980s, the relative standard deviation was assumed to be 15%, because of improvements at the MH 175 discharge point and in the analytical procedures.

Volume measurements. For flow through Manhole 175, the Limit of Error (LE) for the Parshall Flume flow station was reported as 1.5% of the monthly volume totals in routine quality control reports (NLCO 1966, Brown 1967), although there was no indication whether this was at the 95% confidence limit. Water plant personnel at FMPC generally assumed a variability of about 10% on the daily flow measurements. For these tabulations, a relative standard deviation of 10% on the daily Parshall flume results was assumed to account for measurement error.

For days during a month when daily volume records were not available, the daily average was calculated from the monthly total. The relative standard deviation of daily volume measurements for a month ranged from 6% to 20% for the 18 months in the 1960–1962 period, for which such measurements were available. For those days when an average daily flow was used, a total relative standard deviation of 20% was assumed to account for the normal variation in flow seen throughout the month.

Total uranium determinations. The total uranium discharged each day was calculated by multiplying the daily uranium concentration (mg L^{-1}) and the volume of water discharged per day (liters). A standard deviation for each daily uranium concentration measurement and volume measurement was calculated by multiplying the daily measurement by the assumed relative standard deviation. The product of the variances of the daily uranium concentration and volume measurements were determined. The standard deviation of the monthly uranium totals was determined using a standard error propagation technique. To determine the 90% confidence intervals (i.e., 5% to 95% predictions) surrounding the estimates, the error was multiplied by 1.645. To illustrate the methodology that was developed previously (Voillequé 1991) to calculate losses to the river for all years, monthly estimates of uranium lost to the river for 1960 to 1962 are shown in Table L–3 with the associated standard deviations. The same method was used to compute the uncertainty of the volume measurements, and those for the 1960–1962 period are shown in Table L–4. Using the same methodology, estimates of uranium released by way of the main discharge point (MH 175) for all years of operations were calculated, and are shown in Figure L–2. The annual estimates are compiled in Table L–5, along with the documentation sources for each year.

For 14 of the 37 years, daily measurements of uranium at the discharge point to the river were used to reconstruct the annual losses of uranium to the river. For other years, except for 1952 and 1953, monthly reports were used. Figure L–6 shows very good agreement for monthly uranium losses to the river calculated from daily analytical data sheets (ADS), or tabulated from monthly reports for that same period. Hence, the use of monthly reports to provide the uranium loss estimates for our source term reconstruction appears justified by this agreement.

Table L–3. Monthly Estimates of Uranium Discharged From Manhole 175 to the Great Miami River with Associated Standard Deviations (SD)[a]

	1960		1961		1962	
Month	U (kg)	SD	U (kg)	SD	U (kg)	SD
Jan	290	20	630	35	480	40
Feb	340	25	730	40	540	40
Mar	300	20	730	35	410	30
Apr	540	40	1020	55	570	40
May	630	40	850	45	480	30
Jun	530	35	640	35	325	25
Jul	330	20	530	30	320	25
Aug	470	30	930	70	380	25
Sep	380	25	480	30	1480	240
Oct	530	35	200	20	390	30
Nov	540	35	310	25	370	30
Dec	720	40	300	20	470	50
Annual	5600	300	7300	140	6200	300

[a] From Voillequé 1991; daily measurements for these monthly totals are compiled in Tables L1–6 to L1–8 in the Annex. These tables illustrate the results of the methodology used to determine uranium quantities discharged in liquid wastes to the river for all years.

Table L–4. Monthly Estimates of Effluent Volume (million gallons) Through Manhole 175 to the Great Miami River With Associated Standard Deviations (SD)[a]

	1960		1961		1962	
Month	Volume	SD	Volume	SD	Volume	SD
Jan	35.2	1.2	47.0	0.9	34.2	1.2
Feb	32.3	0.8	41.9	0.8	31.9	1.2
Mar	31.5	1.0	45.9	0.8	31.8	1.1
Apr	28.8	0.5	45.1	0.8	25.2	0.9
May	30.1	0.7	42.0	0.8	24.6	0.9
Jun	31.1	1.1	39.0	0.7	28.5	1.0
Jul	28.0	0.5	47.6	0.9	29.5	1.0
Aug	29.0	0.5	46.0	1.0	31.7	1.1
Sep	30.3	0.6	28.1	1.0	28.4	1.1
Oct	40.7	0.7	24.8	0.9	23.2	0.8
Nov	38.1	0.7	28.3	1.0	23.9	0.9
Dec	42.2	0.8	29.9	1.1	30.1	1.1
Annual	397	2.7	465	3.0	343	3.6

[a] From Voillequé 1991; daily measurements for these monthly totals are compiled in Tables L1–6 to L1–8 in the Annex. These tables illustrate the results of the methodology used to determine the volume of effluent discharged to the river for all years.

Table L–5. Annual Uranium Losses to the Great Miami River By Way of MH 175 With Uncertainty Range (kg)

Year	Total U (kg)	5th %ile	95th %ile	Information Sources
1952	2200	1600	2800	a
1953	2200	1600	2800	a
1954	2200	1600	2800	a, b, Table L1–1
1955	2200	1900	2400	b, Table L1–1
1956	2600	2300	2900	b, Table L1–2
1957	3700	3400	4000	c, Table L1–3
1958	3900	3700	4100	c, Table L1–4
1959	2800	2500	3100	c, Table L1–5
1960	5600	5100	6100	c, Table L1–6
1961	7300	7100	7500	c, Table L1–7
1962	6200	5700	6700	c, Table L1–8
1963	4300	4000	4600	c, Table L1–9
1964	5100	4700	5500	c, Table L1–10
1965	3500	3200	3800	d
1966	4500	4000	5000	c, Table L1–11
1967	1890	1700	2100	c, Table L1–12
1968	2400	2100	2700	d
1969	2300	2000	2600	c, Table L1–13
1970	1500	1300	1700	d
1971	2200	1900	2500	d
1972	1100	940	1300	d
1973	1700	1500	1900	d
1974	720	620	850	d
1975	1010	860	1200	d
1976	730	640	820	d
1977	910	780	1000	d
1978	850	740	960	d
1979	1050	960	1240	d
1980	640	560	720	d
1981	600	530	670	d
1982	750	550	950	d
1983	590	510	670	d
1984	900	770	1000	d
1985	610	510	710	d, e
1986	460	390	550	d, e
1987	770	650	890	d, e
1988	810	680	940	d, e

a Assume annual totals from 1955.

b Some daily measurements at MH 175 available; NLCO 1954, NLCO 1955, NLCO 1956.

c Based on daily measurements at MH 175, and monthly operating loss reports; NLCO 1957 to 1969.

d From Schwan 1967 to 1983.

e. Annual Environmental Monitoring Reports (Aas et al. 1986, Aas et al. 1987, WMCO 1988, WMCO 1989.

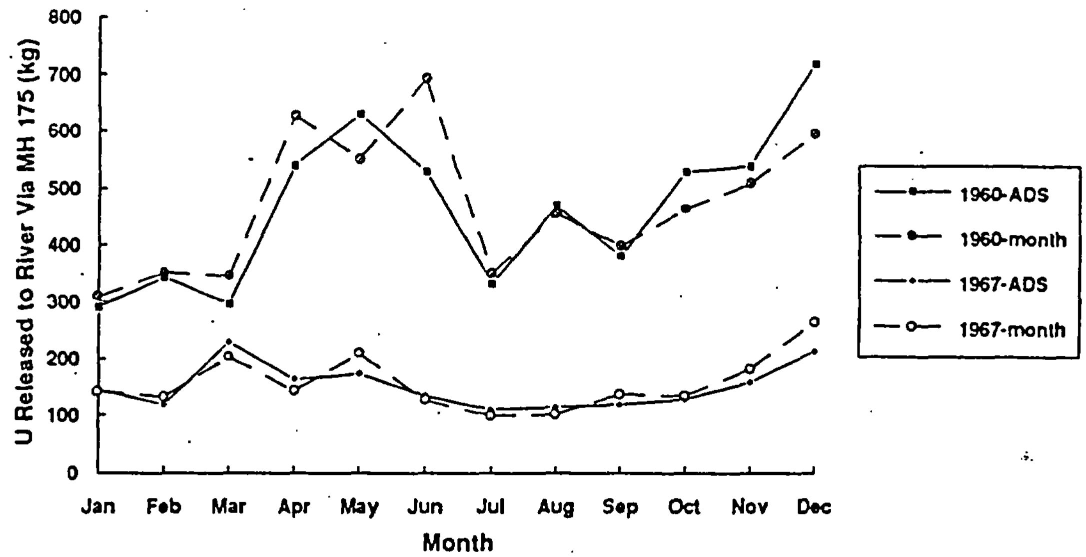


Figure L–6. Comparison of uranium quantities discharged to the river from Manhole 175 for 1960 and 1967, based on daily measurements reported in analytical data sheets from the Bioassay Department (ADS) and from monthly loss reports (month) (Cuthbert 1960–1961, Schwan 1967–1983).

Overall, the quantity of uranium discharged ranged from about 200 kg in October 1961 up to a high of 1480 kg in September 1962. Releases were higher in 1961 than in 1960 or 1962. This is reflected in the annual totals of approximately 5600 kg in 1960, 7300 kg in 1961 and 6200 kg in 1962. These annual totals are 25 to 35% higher than those listed in historic reports from FMPC (Boback et al. 1987). Table L–4 shows the monthly total effluent volumes to the river in 1960, 1961 and 1962. Total flow through MH 175 was higher in 1961, with an average flow rate of 1.3 million gallons per day (MGD), than in either 1960 (average of 1.1 MGD) or 1962 (average of 0.9 MGD).

Uranium releases exceeded 100 kg on at least one day in April 1960 (Table L1-6, annex), August 1961 (Table L1–7, annex), and September and December 1962 (Table L1-8, annex). Losses for the first 9 days of September 1962, which were approximately equal to the total uranium loss for an average month, caused much concern at FMPC (Starkey 1962a). Large releases in 1962 on September 6th (190 kg), 8th (170 kg), and 10th (680 kg), were due to several large accidental releases from Plant 8 during that time. In some months, there was less variation in amounts of uranium discharged per day (for example, December 1960, January 1961), than in other months (for example, September 1960, February 1962). Differences in rainfall patterns and production activities, and the occurrence of spills and unusual releases contribute to the variation. Spills and accidental releases are discussed more thoroughly in an upcoming section.

Enrichment Categories for Uranium in Liquid Releases

The distribution of uranium among the three uranium enrichment categories changed over time at the FMPC. Of the total uranium released to the river, Figure L–7 shows the fraction of the discharges that were normal, enriched and depleted uranium during each year from 1960 to 1984 (Cuthbert 1960–1962, Schwan 1967–1983). Normal uranium represented the greatest fraction of uranium in the releases until 1967, and from 1970 to 1976. Releases of enriched uranium were minor until 1964 when it reached 40% of the total, and fluctuated between 20% and 60% of the total until 1971. Only a small fraction of depleted uranium was released until 1977 when it rose rapidly to 80% to 90% of the total uranium in liquid effluents. No normal uranium was released after 1978. These relationships of the enrichment categories of uranium in liquid effluents released from the site are quite similar to those for uranium receipts and shipments from the site (See Appendix C).

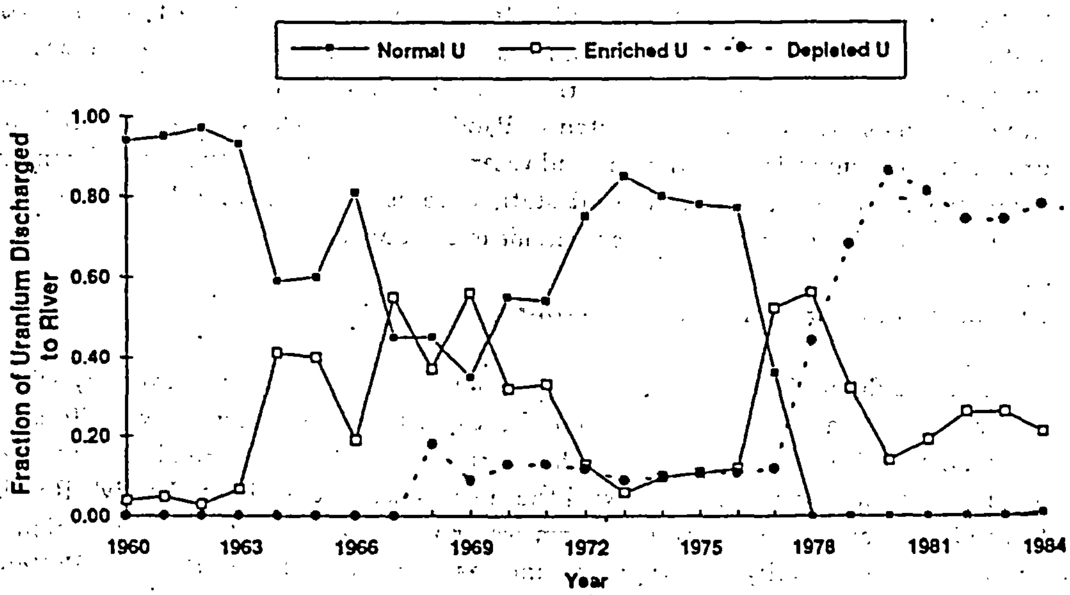


Figure L–7. Relative fraction of normal, enriched and depleted uranium released to the Great Miami River Via Manhole 175 From the FMPC from 1960 to 1984.

ESTIMATES OF URANIUM DISCHARGED TO PADDY'S RUN FROM THE FMPC

Water collected in the storm sewer system and passed through the storm sewer lift station before being discharged through Manhole 175 to the Great Miami River. A flow meter and continuous sampler monitored the discharges. Since the storm sewer lift station is not connected to any process, all the uranium lost through it was assumed to come from leaks and spills (Ross, 1972). Initially, the storm sewer system had only a detention basin and sump for emptying it when necessary. However, the detention basin was not used, and in July 1955 the storm sewer lift station was installed. Prior to that all runoff from the site went directly to Paddy's Run. The lift station in place in the early sixties was designed to take only the initial runoff during a heavy rain. The pumping capacity of the pumps was approximately 500,000 gallons per day or 350 gallons per minute (DeFazio 1960).

Of the total quantity through the Storm Sewer system, most was discharged through the Lift Station while a percentage overflowed and was discharged through the outfall. Figure L-8 shows the magnitude and variability of the uranium discharges to the storm sewer lift station from 1955 to 1968. The major peaks in September 1962, March 1964 and February 1966 coincide with accidental spills to the storm sewer system, or nonroutine releases of materials (Table L-10). Frequently, uranium concentrations measured at the storm sewer lift station were higher in the late winter or early spring following warmer weather when thawed material in the pipes and on the ground could flow freely. Tables L1-14 to L1-21 in the annex contain the uranium concentrations measured at the storm sewer outfall to Paddy's Run and at the storm sewer lift station from 1954 to 1966. Table L1-23 lists the monthly uranium losses and percentage of total storm water flow that discharged through the outfall and to the lift station for 1960, 1961 and 1962. Clearly, flow to the storm sewer system, and, ultimately to Paddy's Run was quite variable, depending upon total rainfall, and rainfall patterns. Generally, from 2 to over 50% of the flow through the lift station was discharged to Paddy's Run. In some instances, where flow was particularly high, there were reports of up to 80% of the flow being lost to Paddy's Run (Starkey 1964c). Runoff to the storm sewer outfall ditch to Paddy's Run Creek is a major contributor to the uranium contamination in the groundwater to the south of the site. Uranium levels measured in the SSOD and at the lift station are used in Appendix M to develop a source term for groundwater contamination outside of the FMPC.

Estimates of Uranium Losses to Paddy's Run

Liquid effluent from the site flowed to Paddy's Run when the capacity of the storm sewer lift station was reached. When the capacity of the storm sewer lift station was reached, water overflowed through the storm sewer outfall to Paddy's Run Creek. The volume of storm water that overflowed the storm sewer lift station to Paddy's Run is related to rainfall amounts and patterns. Storm water flow lags the actual precipitation event by several hours, usually showing an increase in flow the next day (Patton, 1985). Furthermore, contaminants were getting into Paddy's Run from areas other than the storm sewer outfall, perhaps from the vicinity of the Pilot Plant storage pad, from the waste pits, or from the vehicle washing station northwest of Plant 1 (Starkey 1959).

Ground contamination occurred on the west side of the Pilot Plant when the sump overflowed the drain to the southwest corner of the site and into Paddy's Run if the rainfall was sufficient (Flowers 1961, Gessiness 1961). By August 1961, curbing had been installed around the sidewalk between the Pilot Plant Annex and the Pilot Plant to direct some of the contaminated runoff to a catch basin, preventing contamination of the soil (Quigley 1961). Pilot Plant personnel made a survey of the ditches and mud holes west of the Pilot Plant, and made note of several large uranium contaminated ditches running to the southwest, eventually discharging into a large gully due west of the Pilot Plant at the second fence (Shaw 1961). In addition, there was a partially excavated hole on the west side of the Pilot Plant which was usually filled with contaminated water. Memoranda indicate that there were plans to pump out the hole (Shaw 1961, Gessiness 1961). It was reported that surface and subsurface drainage in the Pilot Plant Area flowed into a manhole on the warehouse storage pad, and then, by gravity, into an open drainage ditch which discharged into Paddy's Run Creek (DeFazio 1962). In addition, it was not unusual in the earlier years to drain water from the fluoride pit (Waste Pit 3) directly to Paddy's Run Creek when heavy rains caused high flow in the stream (Starkey 1956).

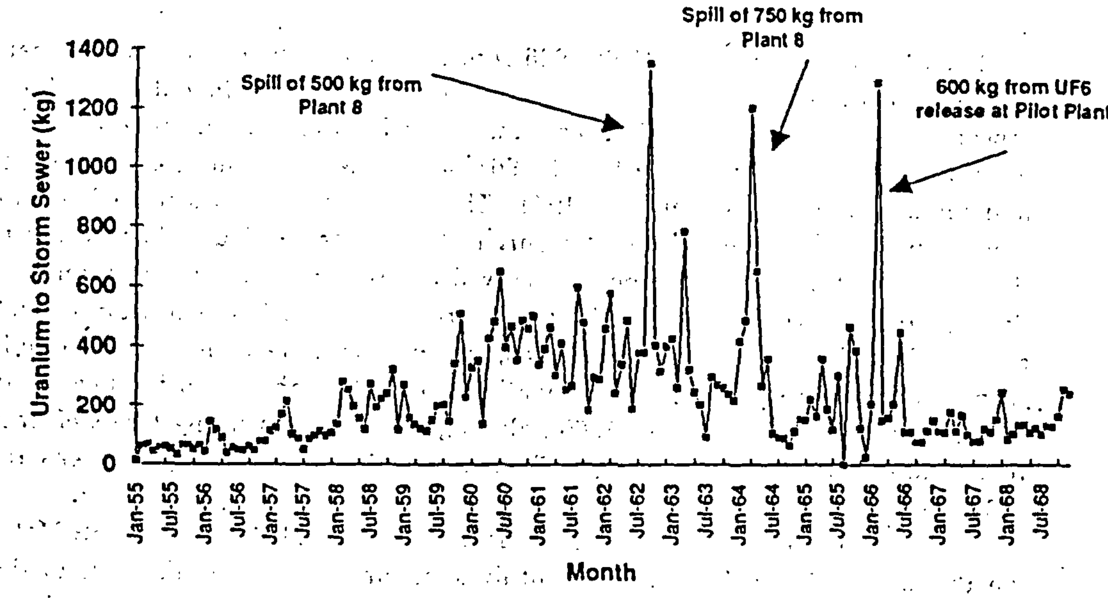


Figure L–8. Monthly quantities of uranium to the storm sewer system from runoff at the FMPC from January 1955 through December 1968. These values were reported in routine operating loss reports from the FMPC. The uranium measured in the storm sewer system comes from leaks, accidental spills and ground contamination events. Nonroutine events involving liquid effluents are recorded in Table L–10.

Prior to the late 1960's, there was no continuous metering of the flow of water through the storm sewer to Paddy's Run Creek (Pennack, 1966), although there was discussion on the continuous measurement of the surface flow in Paddy's Run for some time (Jeffers 1962), and on the purchase of a portable flow meter and sampler (Chapman 1959). In 1966 it was proposed to install a 1,000 gallons per minute (gpm) V–notch weir meter and proportional sampler just downstream from the Storm Sewer Lift Station. Prior to that time, Water Treatment department personnel took grab samples and estimated the flow at the weir notch south of the parking lot (Ross, 1965). Depending upon the duration of the flow, a number of other grab samples would be taken at half hour intervals, and composited. A sample of the composite was then sent to the Bioassay Laboratory for analysis. There continued to be concern regarding the significance of grab samples from the storm sewer outfall in representing uranium quantities lost to Paddy's Run (Quigley 1965). On days when there was a storm sewer outfall flow, the uranium concentration of the outfall sample was usually much higher than the 24-hour composite from the lift station. Analytical results suggested that day-to-day differences in uranium concentrations between the Storm Sewer Outfall grab samples and Storm Sewer Lift Station samples could be significant, but that monthly uranium totals were similar (Ross 1965).

Another source of effluent to Paddy's Run Creek originated as runoff from a portion of the production area near the pilot plant, and as drainage from the waste pit area. In the 1950s, there was a drainage ditch to the south of the waste pits to direct runoff to Paddy's Run (NLCO 1959).

Source of Information for Estimates of Uranium to Paddy's Run

For 72 months during the 1960–1966 period, documentation was available that indicated the dates of outfall flows to Paddy's Run, the volume discharged in gallons, and the uranium concentration for each flow to Paddy's Run. Tables L-17 to L-21 in the annex list the losses for those months in 1960–1964 and 1966 where detailed information was located for individual outfall events (Rathgens 1974). The values in the tables come from two types of reports discussed earlier. The first report is "Discharge of Liquid Wastes into the River" (DLW), a monthly report listing the daily discharge of liquid wastes from the Sanitary Sewer, Storm Sewer, Manhole 175, and Storm Sewer Outfall. The Storm Sewer Outfall category lists the dates, volume in gallons, and measured uranium concentration in ppm for each flow to Paddy's Run. For some months, the total number of outfall flows is not known with certainty (e.g., May – Sep 1960), although records of monthly totals of uranium and volume are available for all months (Chapman 1956, Pennack 1973, Rathgens 1974, Bardo 1985, Patton 1985).

The second type of report is the "Measured Losses and Removals of SS Material From the Production Stream" (MLR), a monthly summary of uranium discards to the General Sump and stack losses. Volumes and quantities of normal and enriched uranium discarded as liquid waste from each process area are listed. In addition, the MLR reports give the losses to Paddy's Run, discards to the chemical pit, and "removals" from the pit to the river.

Uncertainties of Estimating Uranium Losses to Paddy's Run Creek

The uncertainty associated with estimation of uranium losses to Paddy's Run includes three major components. One area of uncertainty involves unmonitored losses from the site above the point where the storm sewer outfall enters Paddy's Run (where the measured losses were recorded). Records of numerous samples obtained from Paddy's Run indicated that the standards were exceeded in various locations north of where the storm sewer outfall enters Paddy's Run Creek (DeFazio 1960). Quantitative information on the amounts of materials discharged to Paddy's Run from drainage north of the storm sewer outfall location is sparse. One report noted that samples of water in the manhole at the Pilot Plant warehouse showed "uranium contamination but not above what would have been expected normally" (Shaw 1961). The concentration of uranium in the water in the gully was highest at the point due west of Plant 2 and 8 and tapered off at the point west of the Pilot Plant (Shaw 1961). One report noted that the analysis of samples from the open drainage ditch west of the Pilot Plant indicated that uranium concentrations varied from 7 to 28 mg U L^{-1} with some flows over 5 gallons per minute (DeFazio 1962).

If these limited data are used to determine whether or not this drainage might be a significant contributor to the total discharges from FMPC to Paddy's Run, then we can calculate the quantity of uranium that would be discharged through this unmonitored drainage ditch if these conditions existed continuously for a month, and compare that value to our monthly estimates. If we assume that a continuous flow of runoff water of 5 gallons per minute (216000 gallons per month) with an average uranium concentration of 28 mg U L^{-1} occurs for an entire month, then we would expect about 20 to 25 kg of uranium per month from this source. This compares to roughly 100 kg of uranium lost to Paddy's Run through the storm sewer outfall ditch each month. Although this rough calculation is conservative, and based on extremely limited data, it represents one source of material loss to Paddy's Run that was not monitored. It may have been the most significant unmonitored source. Consequently, we assume an additional release of 25% above the monthly effluent volume and uranium quantities reported by the FMPC in analytical data sheets and monthly reports.

A second component of uncertainty surrounding the estimation of discharges to Paddy's Run is associated with the collection of grab samples in the storm sewer outfall ditch prior to its convergence with Paddy's Run, and uranium analysis of the grab samples by the fluorometric method. In our interim source term report (Voillequé et al. 1991), data on the number of outfalls to Paddy's Run per month, the volume of water per outfall event, and the uranium concentration of grab samples taken during the overflow event were available for 17 of 36 months in 1960–1962 (See Tables L1–18 and L1–19). Uranium was analyzed by the fluorometric method similar to MH 175 samples. For the individual outfall events in these months, the limit of error (LE) for the uranium concentration measurement at the 95% confidence level was assumed to be 75%, higher than the LE assumed for the uranium determination at the MH 175 discharge point (50%) because the sampling protocol for Paddy's Run involved intermittent grab sampling rather than continuous sampling (Courtney 1965).

Reports indicated that the accuracy of the V–notch Weir flow station ranged from 8% to 15% for normal to flood condition flows, respectively. (Noyes 1966). For this report, the variation is assumed to be 15% for all events. When these errors associated with volume and uranium concentration measurements for individual outfall events are propagated through the month, the LE on the monthly totals range from 4% to 15% of the monthly totals. Consequently, for months when detailed information on number of outfall events was not available, a LE of 15% was assumed for the monthly totals for these 19 months.

A third component of uncertainty for uranium loss to Paddy's Run Creek involves time periods when rainfall, and consequently runoff, were quite high and the capacity of the storm sewer lift station flow meter and V–notch Weir at Paddy's Run may have been exceeded. The water flowing to Paddy's Run occurred when the capacity of the storm sewer lift station was reached. Of the total quantity through the Storm Sewer system, most was discharged through the Lift Station while a percentage overflowed and was discharged through the outfall. Monthly data on measured outfall volume and total uranium to Paddy's Run from the storm sewer overflow indicate that from 2 to 55% of the total flow passed through the outfall to Paddy's Run, with an average of 21 ± 11% (Table L1–23).

The pumping capacity at the lift station was approximately 500,000 gallons per day or about 350 gallons per minute (DeFazio 1960). During this time period (1960–1962), there were an average of 3 to 6 times a month when daily flow through the storm sewer lift station was greater than 600,000 gallons per day, with volumes from 750,000 to 850,000 gallons measured occasionally (Starkey 1960–1961). Without specific rainfall patterns and amounts for those specific days, however, it is difficult to speculate whether the flow was greater than the storm sewer lift station could handle. Based on the occurrence of the storm sewer lift station exceeding its stated capacity roughly 10% to 20% (3–6 times) each month, we assume an additional uncertainty of 20% on the monthly totals of effluent volume and uranium quantity.

These uncertainty estimates for each of the three sources of error that were discussed (unmeasured losses to Paddy's Run, sampling and analytical, and exceeding the capacity of the storm sewer lift station), were incorporated into our final source term estimates for uranium lost to Paddy's Run. Our release estimates, increased by 25% due to unmonitored losses to Paddy's Run, were multiplied by the combined estimates for analytical error and overflow at lift station (15% plus 20%) to provide a bound around each estimate of uranium discharged to Paddy's Run. To determine the 90% confidence intervals surrounding the estimates, the error was multiplied by 1.645. Tables L–6 and L–7 list the monthly quantities of uranium losses and discharge volumes to Paddy's Run for 1960, 1961 and 1962, as an example of the methodology. The uranium concentration data for the storm sewer outfall ditch from original analytical data sheets for 1954 to 1966 are presented in the annex in Tables L1–14 to L1–21.

Table L–6. Monthly Estimates of Uranium Losses to Paddy's Run With Associated Standard Deviations (SD)

	1960		1961		1962	
Month	U(kg)	SD(kg)	U(kg)	SD(kg)	U(kg)	SD(kg)
Jan	160	65	100	40	170	130
Feb	170	70	100	40	160	130
Mar	4	2	230	90	390	310
Apr	40	15	120	50	35	35
May	160	60	120	50	160	130
June	220	130	80	30	90	75
July	170	70	120	45	90	75
Aug	90	10	20	7	60	45
Sep	90	30	330	100	6	5
Oct	110	40	60	90	100	80
Nov	72	30	140	70	75	60
Dec	50	20	30	90	135	110
Annual	1300	200	1400	220	1500	430

[a] From Voillequé 1991; measurements for these monthly totals are compiled in Tables L1–18, L1–19 and L1–22 in the Annex. These tables illustrate the results of the methodology used to determine uranium quantities discharged in liquid wastes to Paddy's Run for all years of operations.

For annual losses in the early sixties, the discharges to Paddy's Run were 1055 ± 201 kg in 1960, 1131 ± 439 kg in 1961, and 1273 ± 272 kg in 1962. Few documents listed uranium losses to Paddy's Run routinely, or summarized these losses on a monthly or annual basis. The latest Remedial Investigation / Feasibility Study Groundwater draft report (RIFS 1990), is one of the few documents that lists losses to Paddy's Run. The RIFS report estimates for losses to Paddy's Run for 1960, 1961 and 1962 are 910, 1180 and 1190 kg, respectively. Our estimates for these years are listed in Table L-8 along with the estimates for all years.

Table L-7. Monthly Estimates of Effluent Volume to Paddy's Run With Associated Standard Deviations (SD)

Month	1960 Volume (million gallons)	1960 SD	1961 Volume (million gallons)	1961 SD	1962 Volume (million gallons)	1962 SD
Jan	0.19	0.05	3.3	0.5	8.9	2.5
Feb	9.5	1.6	3.4	0.5	5.3	1.5
Mar	0.05	0.01	11	1.5	22	6.1
Apr	0.64	0.14	4.1	0.6	1.6	0.44
May	0.8	0.04	4.1	0.6	0.02	0.05
Jun	4.9	1.4	1.7	0.3	1.4	0.4
Jul	4.0	0.65	3.7	0.5	8.4	2.3
Aug	0.8	0.15	0.35	0.05	1.2	0.33
Sep	2.9	0.82	1.9	0.52	0.11	0.03
Oct	1.9	0.31	0.95	0.26	3.3	0.94
Nov	1.4	0.22	3.6	1.0	3.1	0.95
Dec	21.5	0.22	3.1	0.9	4.6	1.3
Annual	28	2.4	42	2.5	60	7.4

[a] From Voillequé 1991; measurements for these monthly totals are compiled in Tables L1-18, L1-19 and L1-22 in the Annex. These tables illustrate the results of the methodology used to determine the volume of effluent discharged to Paddy's Run for all years.

Figure L-9 compares monthly uranium losses to Paddy's Run from the Storm Sewer Outfall Ditch for three time periods: 1959 to 1962, 1969 and 1970, and 1979 and 1980. The data show that the quantity of uranium lost to Paddy's Run varied considerably from month to month in the early years, so that an average value over a short period of time may not adequately have described a particular month, or several month period. The figure also shows the gradual decrease in total quantity and in monthly variability of uranium released to Paddy's Run. The decline reflects a decrease in production in the seventies and eighties, along with some improvements in the effluent handling system onsite.

Annual estimates of uranium released to Paddy's Run are shown in Figure L-2 with those releases directly to the river from the FMPC. In Table L-8, estimates of uranium losses to Paddy's Run are listed for all years of operations, with the associated uncertainties.

NONROUTINE RELEASES TO SURFACE WATER

Releases of contaminated liquids from spills, drum ruptures, and overflow of sump ponds have been considered in determining the total quantity of uranium released in liquid effluents from FMPC. Regular ground contamination reports were issued on a regular basis. As early as September 1953, an investigation of contamination of the storm sewer outfall to Paddy's Run was conducted after local residents reported changes in the stream from the previous year (Blase and Starkey 1953). The investigators at the site concluded that the primary source of contamination to Paddy's Run was iron salts in runoff from the coal pile.

At that time, all surface drainage from the plant site discharged directly to Paddy's Run via the storm sewer system. During the 1950s, brief "Storm Sewer Contamination" memoranda encouraged plant supervisors to minimize the causes of increased ground contamination and spills (Stewart 1957), but generally no quantitative details of incidents were provided.

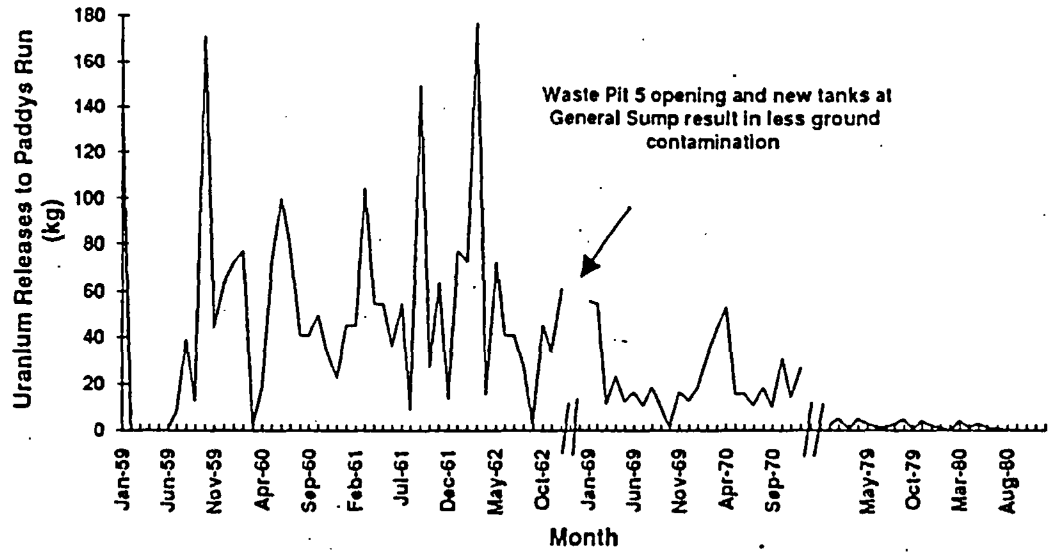


Figure L-9. Monthly uranium losses to Paddy's Run Creek by way of the Storm Sewer Outfall Ditch for three time periods: 1959–1962, 1969–1970 and 1979–1980. The gradual decline in uranium releases over the years coincides with improvements in the liquid effluent handling system, and with a decline in production activities.

On June 1, 1959, an external area ground contamination survey program of all production plant was initiated on a weekly schedule to inform plant supervision of existing major ground contamination areas, their sources, remedies, and the effect of ground contamination on the storm sewer system (Dodd 1959). Frequently, spills of contaminated materials were described by thickness, and area of gravel covered. For example, a "quarter inch thick" spill covering one square yard, occurred on the graveled area near Plant 4 in February 1964 (Starkey 1964b). Initially all major contaminated areas of soil were to be removed to the waste pits. By 1961, however, the excavation activity was viewed as "not only ridiculous but also an expensive" practice, because of recurring contamination in some locations of the process area (Flowers 1961). With the emphasis on ground contamination, however, the number and extent of spills did appear to decrease over time, shown in Table L-9, in which we have compiled information on the monthly frequency and general source of spills affecting the storm sewer system from 1959 to 1969.

Table L-8. Annual Uranium Losses to Paddy's Run With Uncertainty Estimates

Year	Uranium(kg)	5th %ile	95th %ile	Primary Information Sources
1952	522	410	630	a
1953	522	410	630	a
1954	522	410	630	b, d, Table L1-14
1955	300	190	405	b, d, Table L1-15
1956	270	210	320	b, d, Table L1-16
1957	340	280	410	b, d, Table L1-17
1958	630	510	750	b, d
1959	840	640	1000	c
1960	1300	800	1800	e, Table L1-18, L1-19
1961	1400	1000	1600	e, Table L1-19, L1-20
1962	1500	1100	2100	d, Tables L1-17 & 11-18
1963	901	720	1100	b, Table L1-18
1964	1722	1260	2200	d, e, Tables L1-18 & L1-21
1965	622	490	760	b
1966	771	550	1000	d, Table L1-22
1967	753	560	950	e
1968	358	280	430	e
1969	290	250	340	e
1970	349	300	390	e
1971	499	410	590	e
1972	322	280	370	e
1973	231	200	265	e
1974	255	210	300	e
1975	245	180	250	b
1976	272	230	310	e
1977	204	170	230	e
1978	68	60	80	e
1979	84	70	100	e
1980	50	40	60	e
1981	20	18	22	f
1982	20	18	22	f
1983	54	40	70	e
1984	57	50	70	e
1985	39	30	50	f
1986	17	15	20	f
1987	<0.5	<0.5	0.5	f
1988	<0.5	<0.5	0.5	f

a Assume annual totals from 1954; estimates based on uranium measurements at the storm sewer outfall. the storm sewer lift station not installed until August 1955.
b Based on monthly reports of storm sewer losses; assume 20% to storm sewer outfall ditch.
c Routine monthly reports of operating losses for all months.
d Analytical data sheets for daily losses to storm sewer outfall ditch.
e Monthly records of outfall events to Paddy's Run.
f Annual Environmental Monitoring Reports; assumed uncertainty range of 10%.

Table L–9. Monthly Frequency and General Location of Spills Affecting the Storm Sewer System During 1959 Through 1969[a]

Year	Date	Number of Incidents Affecting Storm Sewer	Areas Involved
1959	June	22	All processing areas
1961	April	12	All processing plants
	May	14	All processing plants
	June	13	All processing plants
	July	10	All processing plants
	August	8	All processing plants
	Sep	15	All processing plants
	Oct	10	Plant 2/3, 6, 8, 9, Pilot
1962	Sep	16	Plant 4, 5, 6, 8, 9, Pilot
	Nov	11	Plant 1 pad, 4, 5,6, 8, 9, Pilot
1963	March	16	All processing plants
	June	7	Plant 2/3, Plant 6, Plant 8, Pilot
1964	Feb	18	All processing plants
1965	Mar	4	Plant 8, roads
	Apr	1	Railway
	May	2	Plant 2, 4
1966	Jan	9	Plant 1 pad, 2/3, 8, 9
	Feb	7	Plant 2, 8
	Mar	16	Plant 8, tank farm
	Apr	10	Tank farm, Plant 8, 2/3
	May	5	Plant 8, 2/3, tank farm
	June	4	Plant 2/3, 8
	July	2	Plant 1, roads
	Aug	4	Plant 2/3, Lab Bldg.
	Sep	2	Roads
	Oct	1	Bldg. 64(Th warehouse)
	Nov	2	Plant 9
	Dec	3	Plant 8, 2

(continued on next page)

Table L–9. Monthly Frequency and General Location of Spills Affecting the Storm Sewer System During 1959 Through 1969[a] (continued)

1967	Jan	12	Plant 8, 4
	Feb	11	Plant 8
	Mar	3	Plant 8
	Apr	4	Plant 8, 1
	May	3	Plant 8, tank farm
	June	10	Plant 8, 4, tank farm
	July	9	Plant 8
	Aug	8	Plants 8, 2/3, 4, roads
	Sep	1	Plants 8
	Oct	2	Plant 6, roads
	Nov	4	Plants 2/3, 4, 8, roads
	Dec	4	Plants 2/3, 4, 8
1968	June	4	Plants 8, 2, roads
	July	4	Plants 4, Pilot, roads
	Aug	2	Roads
	Sep	2	Plant 8, roads
	Oct	4	Plants 2, 6, 8, roads
	Nov	1	General Sump Area
	Dec	2	Plant 8, roads
1969	Jan	1	Plant 8
	Feb	3	Plant 8
	Mar	3	Plant 8, roads
	Apr	1	Plant 8
	Nov	1	General Sump
	Dec	2	Plant 8, roads

[a] Data were compiled from the monthly reports, "Comments on Ground Contamination in Process Area"(Flowers 1959–1962; Dodd 1958–1959) and "Incidents Affecting the Storm Sewer System"(Riestenberg 1965–1969) that were available for this time period.

From the review of numerous ground contamination reports since 1954, it becomes clear that several locations in the production area continued to be problem areas. These are:

- Plant 8. Contamination prevalent at the east and west end of the plant. Contamination at the north side was caused by the operation of the box furnace. Some of this contamination was checked with the enlargement of the paved area so that it could be flushed from the pavement to the existing sump and storm sewer system (Chapman 1956). Increase in level of storm sewer losses with initiation of the airport scrap handling operation in April 1960.
- Plant 6. The Machining Area from the east pad near the intersection of First and "E" Streets continued to be contaminated from runoff and underground leakage from acid

lines below floor level (Bussert 1956, Tippenhauer 1957). The east pad serves a dual purpose as a plant entrance and a work area, resulting in contamination being spread routinely by vehicles moving through the area (Smith 1961). Although the east pad proper was designed to drain into a sump, "E" street was not so constructed. The lack of curbing on the south end of the pad allowed contamination to drain to the dirt field (Spenceley 1959).

- Plant 2/3. Ore spills common on the SW side. Orange oxide contamination occurs at the SE corner of Plant 2 at the "gulper" station. This problem arose from the muffler discharge connections and from breakage of filter bags in the gulper system (Chapman 1956). Most contamination was restricted to the concrete pad, although the surrounding gravel was replaced after the scrubber system replaced the dry bag collector in late 1956.
- Plant 1 Storage Pad. The area east of the Drum Reconditioning Building usually contained several hundred empty contaminated drums waiting to be baled. Loose contamination fell from the drums onto the pad which flowed into the storm sewer.
- Pilot Plant. The most contaminated areas around the Pilot Plant generally were near the storage pads to the south and west of the Pilot Plant, where the sump overflowed the drain to the SW corner of the facility to Paddy's Run. The small pad near the fence on the west side of the plant was "badly contaminated with piles of U_3O_8" in the mid-1950s (Chenault 1955). Occasionally, equipment that had been inadequately cleaned was stored on the ground near the SW pad the Pilot Plant (Starkey 1958b). On the west side of the Pilot Plant, the principal contamination was from spills of nitric acid wastes with low uranium concentrations around the nitric acid absorber and storage tank (Davis 1957). In August 1957, a large volume of sump liquor with a low uranium concentration was accidentally spilled while loading the sump truck in that area. This action required "moving a lot of dirt" (Davis 1957). Contaminated soil was removed from near these storage pads periodically, but this area was drained by natural seepage and surface runoff into Paddy's Run Creek.

Over the years, several attempts were made to locate, and thereby eliminate, specific sources of the uranium that were found at the Storm Sewer Lift Station (Chapman 1961, Starkey 1969, Riestenberg 1969, Ross 1972, Lenyk 1977). Generally these surveys indicated that, except for the Boiler Plant area, uranium was entering the storm sewer system plantwide by surface drainage (Lenyk 1977). The main sources of contamination appeared to be the transportation and use of dirty drums, dirty pallets, storage on the ground, and re-drumming operations at some of the storage pads. Furthermore, the use of contaminated oil as dust palliatives on secondary roads and the fly ash pile near the SE corner of the site between the storm sewer outfall ditch and Paddy's Run Creek contributed to storm sewer contamination for years (Karl 1960; Starkey 1960) (See Figure K–1, Appendix K).

For a significant spill into the storm drain, the flow from the lift station could be directed to the General Sump by reversing the flow from the sump, using an emergency gate or diversion valve installed in the early 1970s (Keller 1978). Contamination of this type would usually be washed into the storm sewer system or into Paddy's Run depending upon the location of the contamination. Contamination in Paddy's Run was the primary result of ground spills at the facility (Starkey et al. 1961). The lift station, installed in June 1955, would handle the majority of the flow in the sewers, with the first fifteen minutes of flow going to the river or catch basins, and the rest flowing over to Paddy's Run (Glass 1955a).

To ascertain the significance of contamination incidents and major unplanned releases of liquid on the determination of the surface water source term, we closely examined reports of incidents involving unusual losses of uranium in liquid effluents, and listed them in Table L–10. The data have been taken from various documents to provide as complete a record as possible of the major accidents or unusual events that discharged quantities of uranium and other radionuclides higher than "normally" released on a daily or monthly basis.

"Notice of Contamination Source" forms were prepared for incidents of chemical spills, radioactive spills, and releases of contaminants directly to the storm sewer due to mechanical problems (Flowers 1960a). The most significant incidents that contributed to possible increases in the uranium quantities in liquid effluent were reported in "Comments on Monthly River and Effluent Flow" reports (Fischoff 1960–1962). These events were based on the daily calculated uranium losses in the effluent and on formal incident reports received. As the scope of our investigation expanded for all years of FMPC operation, a somewhat similar procedure was followed with the emphasis on those events which may have caused contamination in the storm sewer greater than would be expected from "routine" operations. Table L–10 summarizes the major unplanned releases and losses of material into the liquid effluent system that were reported or recorded in memoranda, daily log sheets, or various types of reports. It provides a brief description of the event, the date, reference source, and general location of the spill or accidental release. The table includes the detailed summary of events for the 1960–1962 period from the Draft Interim Task 2 and 3 report (Voillequé et al. 1991).

The release points for spills or accidental discharges from the FMPC facility would be the same for unplanned as for "routine" liquid effluent releases, that is, through MH 175 to the Great Miami River, or to Paddy's Run. In many cases, the unplanned releases involved quantities of material that were similar in magnitude to daily discharges through MH 175. For example, the incidents on November 21, 1959 (Beers 1960a), January 28, 1960 (Flowers 1960a), and June 1961 (Cuthbert and Quigley 1961) involved the lost of from 2 to 11 kg U, but the main emphasis of these reports was on equipment failure or the need for better procedures.

Occasionally, unplanned releases involved large quantities that were easily measured at the Storm Sewer Lift Station and Manhole 175 (See Figure L–8). For example, in 1962, the uranium concentration measured at Manhole 175 was 125 mg L^{-1} on September 10 (about 25 times the concentration measured for routine releases), and 15 mg L^{-1} on September 11, reflecting the release of approximately 1000 pounds (450 kg) of uranium to the storm sewer from a digester filter overflow in Plant 8 on September 10. The unplanned releases of September 4 and September 7, 1962 were monitored at Manhole 175 as higher-than-usual concentrations of 10 mg L^{-1} on September 5, 45 mg L^{-1} on September 6, and 45 mg L^{-1} uranium on September 8. This series of losses of materials to the storm sewer system during September 1962 contributed to the highest estimated monthly release of 1500 ± 240 kg (± standard deviation) of uranium via Manhole 175 (Tables L–3 and L–4), compared to the average monthly discharge of about 350 kg in 33 million gallons of effluent.

Table L-10. Major Unplanned Liquid Releases and Spills to the Onsite Liquid Effluent System at the FMPC

Date (reference)	Plant Area	Release Amount	Description of Event or Circumstances
9 June 1954 (Costa 1955)	Roadway storage pad to Plant 2	871 lb. South African Concentrate	Transport trailer broke loose from train, spilling contents of 16 drums; cleaned up and drummed.
6 Dec 1954 (Harrell 1954)	Storage pad	Unknown	Diuranate cake and black oxide in dollies turned over, splitting two drums of diuranate cake
July 1955 (NLCO 1955)	Plant 1 pad	Unknown	Scrap material spilled over pad due to poor stacking of material and burst drums causing greater contamination than normal of ground and storm sewers.
Oct 1955 (Glass 1955a; Stewart 1955)	Plant 2/3	Varies from 2 to 26 x maximum allowable conc. (MAC) of 0.22 dpm mL^{-1}.	NW corner of acid recovery contamination by raffinate dumping station to storm sewer; ruptured drums on pad lost to Paddy's Run at the scrap pit.
1 Nov 1955 (Chapman 1955)	Plant 2/3	26 lb. of U in 195,000 gallons	Loss due to removal and cleaning of vapor lines between denitration and acid recovery
2 Nov 1955 (Glass 1955b)	Plant 6 General sump	40 lb. from general sump to river in 20,000 gallons.	Refinery sump surge capacity reached so no reprocessing could occur when high levels detected in Tank F18-1. Cause traced to filter problem in Plant 6.
17 Nov 1955 (Chapman 1955)	General Sump	19 lb.	Spill of 2000 gallons of calciner feed in Combined Raffinate Area.
23 Nov 1955 (Stewart 1955)	Plant 2/3	28.9 lb. U in 341,000 gallons	Condensate from denitration vapor line went to general sump prior to analysis; after analysis (10 g L^{-1}), material drummed and returned to refinery.
25 January 1956 (Strattman 1956)	K-65 Silo Area	Estimated 1000 lb. of 2700 lb. insoluble metal oxide that was sent to the silo.	Metal oxide dust blew out between the top and sidewalk of the first silo, covering several hundred feet around silo; removed with snow layer to concrete trench between Plant 1 and Refinery.
7, 19 Mar 1957 (Stewart 1957)	Storm Sewer	53 lb./day; 10 mg L^{-1} at lift station	Unknown cause; high "U" stream flushed into storm sewer system.

(continued on next page)

Table L–10. Major Unplanned Liquid Releases and Spills to the Onsite Liquid Effluent System at the FMPC (continued)

Date (reference)	Plant Area	Release Amount	Description of Event or Circumstances
3 April 1957 (DeFazio 1957)	Roadway	Spill material 12,000 mg U L^{-1}	Barrel of material spilled on road at "B" and 2nd Street; material pushed into Storm Sewer manhole.
22 July 1958 (Noyes 1958)	Drainage system at NE corner		Proposal to modify and repair drainage system surrounding Production Area to eliminate recurrence of flood condition.
16 Sep 1958 (Ross 1958)	Refinery Area	8.32 mg L^{-1}	Spill of raffinate in refinery area showed a U concentration of 4100 mg L^{-1}; rain washed spill to storm sewer and Paddy's Run.
23 July 1959 (Harr 1959)	Plant 2/3	1000 lb. U; about 400 lb. to storm sewer	Release of hot uranyl nitrate solution from the 8" vent of the #212 sparge tank on to the denitration pad, the roadway east of the Refinery and the gravel area east to Plant 4. Gravel excavated to pit.
21 Nov 1959 (Beers 1960a)	Plant 8 Storm Sewer	500–750 gallons of 1800 mg L^{-1} U; 12 lb.	Digestion filter pump failure
5 Jan 1960 (Flowers 1960a)	Source unknown	46 kg (101 lb.)	Detected in storm sewer and MH 175 samples; concentration 12 mg U L^{-1}.
28 Jan 1960 (Flowers 1960a)	Plant 8	11 kg (24 lb.)	Not given
18 Feb 1960 (Flowers 1960b)	Plant 8 to Pit 3	"Unknown" (MAC not exceeded in Paddy's Run)	Effluent line from Plant 8 broke near entry to Pit 3; flow to Paddy's Run via drainage ditches
29 Aug 1960 (Harr 1960)	General Sump	111 lb. U to waste pit	One of tanks (F18E-3) was pumped too pit before analysis.
1 Oct 1960 (Beers 1960b)	Plant 8 Storm Sewer	70 kg (155 lb. UO_3)	Not clear; 16.5 mg U L^{-1} detected in storm sewer and MH 175 samples.
20 Feb 1961 (Starkey 1961a)	Pilot Plant, west side	Not given in report	Process and contaminated water pumped onto ground; area "cleaned up".
20 Mar 1961 (Bravard 1961a)	Sump Area, Plant 9 "D" Street	Spill material had 1 g U L^{-1}; "2–3 mR h^{-1}"	Overflow of sump pit that empties filtrate hold tank diked area to graveled area covering 10' by 40'.

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Table L-10. Major Unplanned Liquid Releases and Spills to the Onsite Liquid Effluent System at the FMPC (continued)

Date (reference)	Plant Area	Release Amount	Description of Event or Circumstances
27 Mar 1961 (Bravard 1961b)	Plant 9, So. gravel area	150 kg (330 lb.); 10 gal drum black oxide	55-gal drum with 10-gal drum inside failed when burning briquettes added; area cleaned up.
28 Apr 1961 (Beers 1961)	Plant 8 UAP Acid Filtrate	158 kg (347 lb.) U; 830 gal of 50 g U L^{-1}.	Spill contaminated 40–50 yards of gravel; storm sewer was closed and material was drummed
Jun 1961 (Cuthbert & Quigley 1961)	Pilot Plant, outside	1.5 kg (3 lb.) U	Area SW of Pilot Plant; material removed to waste pit.
4 Sep 1962 (Gessiness 1962)	Plant 1 storage pad	91 SS kg U (200 SS lb.)	Leakage from drums of contaminated solvent being transported to digestion.
7 Sep 1962 (Gessiness 1962)	Plant 1 Storage Pad	307 SS kg (675 SS lb.)	Leakage from drums of contaminated solvent being transported to digestion.
10 Sep 1962 (Noyes 1962a; Strattman '62;)	Plant 8 Storm Sewer	455 kg (1000 lb.) U in 1820 gallons	Winlo digestion filter overflow of liquid containing UO_2Cl_2.
13 Dec 1962 (Beers 1962; Noyes 1962b & 1962c)	Plant 8 Storm Sewer	70 SS kg (153 lb.) enrich U, 92 SS kg (203 lb.) normal U.	Calculated release based on storm sewer sample from MH 23 and digester sample in Plant 8; due to plugged filtrate line to precipitator.
1–10 Mar 1964 (Starkey 1964b)		1640 lb. U to Paddy's Run	Not clear; probably involved Plant 8.
14 Feb 1966 (Starkey 1966a)	Pilot Plant	1230 lb. U	UF_6 release.
6 Jun 1966 (Nelson 1966)	Plant 2	900 lb.	Process "slop" liquor leaked from diked area beneath the NE and SE hold tanks on N side of refinery.
2 Aug 1966 (Noyes 1966)	Plant 2	600 lb. of U at 1.12% ^{235}U	Open nitric acid valve to NE hold tank allowed overflow of materials with U concentration of 50–70 g L^{-1} to storm sewer.
12 Oct 1966 (Starkey 1966b)	Plant 3	100 lb. U onto graveled area	Leaking overhead line near the SE corner of the plant; some gravel was removed for reprocessing.

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Table L–10. Major Unplanned Liquid Releases and Spills to the Onsite Liquid Effluent System at the FMPC (continued)

Date (reference)	Plant Area	Release Amount	Description of Event or Circumstances
January 1967 Riestenberg 1967)	Plant 8	Various	UAP filter, filtrate receiver problems resulted in 8 contamination notices.
March 1967 (Riestenberg 1967)	Plant 8	Various	Sump filter problems; frozen discharge line.
28 Jun 1967 (Levy 1967)	Plant 2/3	41 to 100 lb. U in 450,000 gal. (17.6 g L^{-1}).	Slop Tank F1-25A, located in diked area N of plant, overflowed; most contained, some leaked via trenches to storm sewer.
10 Oct 1968 (Starkey 1969)	Plant 8	1–2 mR hr^{-1} reading	Liquid material coming from UAP scrubber stack covered ground area of 26' by 15'; no action taken.
14 Oct 1968 (Starkey 1969)	Plant 8	5 mR hr^{-1} reading	Spill covering 4' by 4' area at edge of pad near Bldg. 72 scale area; area cleaned.
Feb 1969 (Riestenberg 1969a)	Plant 8	About 500 lb. in two weeks	Trouble with acid filtrate pumps causing low pH readings at lift station; two rebuilt centrifugal pumps installed.
March 1969 (Riestenberg 1969b)	Plant 2/3	100 mg L^{-1} at 55 –65 gpm in storm sewer	Flushing pad area on west end of Refinery
Dec 1971 (Ross 1972)	Storm Sewer	Several hundred lb./mo.	Should investigate
27 Apr, 3 and 8 May 1978 (Riestenberg 1978)	Plant 6	Up to 11 mg L^{-1} U at storm sewer; 50 mg L^{-1} at MH near Plt 6.	Briquette processing floor leak, and broken storm sewer line; operations stopped until floor repairs completed; flow to MH 175 was diverted to General Sump.
18 January 1988 (WMCO 1989)	Plant 2/3	40 lb. (19 kg) uranium	Plant 2/3 roof and ground area NE of plant contaminated with uranyl nitrate vented through stack with water vapor.
Spring 1989 (Dugan et al. 1990)	Gravel area S of Plant 7	1356 lb. (615 kg) black material; U conc. of 1.0%	Black material (fly ash) fell from a dump truck in the spring; in July, the material was drummed.

Because very little rainfall fell during September 1962 (Table L1-23), the loss of uranium to Paddy's Run Creek was only 6 ± 3 kg (± standard deviation) with an estimated monthly volume loss of 110,000 ± 21,000 gallons (± standard deviation). Although highly dependent on rainfall, the average discharge per month to Paddy's Run Creek during this period was roughly 140 kg in 3 to 5 million gallons of water (Tables L-6 and L-7).

In 1955, daily measurements from September through December indicate quite high uranium concentration measurements at MH 175 on November 2 (7.6 mg L^{-1}), and November 30 (6.2 mg L^{-1}), compared to an average 4 mg U L^{-1}. These events were related to filtration problems in Plant 6, and to cleaning the denitration vapor lines when condensate from the line was sent to the General Sump without analysis; respectively (Chapman 1955). The material was drummed and returned to refinery for further processing.

In February, March, and April 1964, more uranium was lost to the storm sewer (over 5000 lb.) than in any other three-month period of operations (Fischoff 1964a, 1964b, 1964c). Although no single cause was given for this high loss of materials, varying factors apparently contributed to it. There were extreme weather conditions over the previous eight months with higher than average rainfall. During this time, additional storage pads were being constructed to prevent further spills onto dirt and graveled areas, and this activity may have loosened dirt as a source of contamination in runoff. Finally, work began on repairing the Plant 8 roof where a chronic ground and storm sewer contamination problem existed. During this repair in February and March, all loose contamination was to be removed from the roof before resurfacing and gutter replacement. This loose contamination may have been a source of storm sewer contamination, although it is not clear from the available documentation how the material was handled. This work was completed by April 1964, when a significant portion of the Plant 8 roof area was connected to down spouts directly to the plant sump system (Starkey 1964c). Interestingly, K.N. Ross, of the Industrial Hygiene and Radiation Department who noted contamination problems in memoranda and reports, was on leave from the site at the Nuclear Metals Division in Albany, NY from January 13, 1964 to May 18, 1964.

What seemed to be more common was the situation where a higher than average uranium concentration was noted at MH 175 alerting personnel that an unplanned release or spill of materials containing uranium had occurred. The origin of these higher releases could not always be traced to a definite source or particular location within the facility. For example, in 1960 higher uranium concentrations were measured on January 5 (12 mg L^{-1}), February 9 (10 mg L^{-1}), February 18 (13 mg L^{-1}), April 11 (30 mg L^{-1}), and May 15 (21 mg L^{-1}) than the average range of 2 to 6 mg L^{-1} uranium (See Table L1-6, annex). Based on these concentration measurements and the corresponding volumes for that day, the probable size of the release or discharge would be calculated (Flowers, 1960a; Beers, 1960b).

On other occasions, situations occurred which did not seem to produce an effect upon the uranium concentration in the effluent at MH 175, such as those in March and April 1961 when an overflow in a sump pit occurred, and Plant 8 UAP acid filtrate spilled and contaminated 40-50 yards of gravel (Table L-10). Furthermore, the addition of contaminated water from extinguishing radioactive fires, or flushing of spill areas into manholes, which were not infrequent events, were not always seen at MH 175 (Fischoff 1961). Such conditions may have been due to closing the storm sewer near the spill until it was cleaned up, or to an insufficient volume of the effluent for proper flow in the lines caused by low rainfall. Another possibility is the occurrence of extreme freezing temperatures during a particular month which would cause accumulation in the lines (Fischoff 1961). Generally, these latter incidents were noted when melting snow or excess rainfall increased the effluent flow through the lines causing a higher-than-usual flow and greater quantities of uranium at MH 175, such as during periods in February and April 1961 when the average volume and uranium concentration were about twice as high as normal (Table L1–7, annex).

Clearly, Manhole 175 sampling results often did not correlate well with known abnormal releases in the process area. The reverse was also true. In many cases the magnitude of "routine" releases masked the unplanned discharges of some material. On some occasions, excess uranium was noted on the day of an unusual or unplanned occurrence, while other events occurred which did not seem to produce an effect at the sampling location (Fischoff 1961b). It does appear, however, that the major unplanned releases were detected (e.g. September 10, 1962) at the discharge points from the site. The fact that the large increases in uranium concentration in effluent discharged to the storm sewer system (Figure L–8) correspond to documented accidental spills bears this out. Thus, it is probable that unplanned or accidental liquid releases or spills were detected and have been accounted for as additions to the "usual" or "routine" discharges of uranium measured at Manhole 175 and Paddy's Run Creek. The review of incident reports covering all years of operations suggests that major incidents were not missed, and information regarding major and minor incidents of all kinds were communicated rather frequently by memo, report or letter.

CHEMICAL AND PHYSICAL FORM OF URANIUM IN LIQUID EFFLUENTS

Several uranium species of both the +4 and +6 oxidation states may have been present in solution in liquid waste streams flowing from the FMPC. The species containing uranium of the +6 oxidation state would probably predominate because most of the uranium discards to the General Sump came from Plant 2/3 (Table L–1), in which the liquid digested material was composed of hexavalent uranium compounds almost exclusively. Uranium in the +4 oxidation state in the form of green salt (UF_4) was also discharged from some of the other plants. In addition, some uranium–containing solids which have not been identified specifically were carried in suspension in the liquid waste streams (Alpaugh 1956). There may also have been very small particulates of the insoluble compounds U_3O_8 and UO_2 among the suspended solids.

The species of uranium in the +6 state would include the well–known uranyl ion, UO_2^{++}, and hydrolytic products such as $UO_2(OH)^+$, $(UO_2)_2(OH)_2^{++}$, $(UO_2)_3(OH)_4^{++}$, and others. The very complex hydrolytic reactions involving these species have been described in the literature (Gmelin 1984). The ratios of these various ionic species in waste streams, Paddy's Run Creek, or the Great Miami River would be a function of the pH of the water. Based on the volume of liquid effluent discharged to the river (Tables L–4 and L–7, Figure L–3), most of the UF_4 releases from the plants would have dissolved in the waste streams even though it is not very soluble in water (about 30 mg L^{-1}). Hydrolytic reactions of UF_4 probably occurred. Some of the unidentified suspended solids containing uranium that were released in the waste streams might have dissolved during the continued dilution downstream.

The presence of suspended solids in liquid process waste discharged to the Miami River is considered in assessing the relative solubility of uranium in liquid releases. General concern about the level of total suspended solids (TSS) or filterable materials in the liquid effluents to the river was a long-standing issue at the site (Starkey et al. 1962). The primary problem was that "settleability and radioactivity of the solids are such that the State of Ohio pollution standards cannot always be met without serious curtailment of production processes" (Boback and Heatherton 1958). Daily measurements of TSS were made on 24-hour composite effluent samples at MH 175 beginning in 1956 (NLCO 1956). Table L1-24 in the annex lists the daily measurements of TSS to the river in 1957, and shows the extreme fluctuation for that year because no settling occurred before discharge to the river. The annual average value in 1957 was 400 mg L^{-1}, with a maximum of 4600 mg L^{-1} measured on October 12, 1957. After April 1958, all solids from the General Sump were sent to Pit 3 for settling, and the liquor pumped to the river via MH 175. This improvement was reflected in the decline of average TSS at MH 175 to less than 100 mg L^{-1} in the 1960s and early 1970s (Figure L-10). The decline continued to less than 25 mg L^{-1} since 1975. Table L1-25 summarizes the monthly average TSS concentrations in liquid effluents for 1957 to 1966.

Various chemicals and coagulants were tested to assess their effectiveness in removing these solids. In a series of twelve tests in 1958 on effluent samples from around the site, Separan 210, a Dow Chemical Company flocculating agent, reduced the TSS by approximately 70%, beta activity by 90% and alpha activity by 74%. Based on these tests, a TSS concentration of 25 parts per million (ppm) was suggested as a design criterion for wastes released to the river (Boback & Heatherton 1958). After 1958, the TSS in effluents dropped significantly with the transfer of material to the General Sump for settling before release to the river. In the seventies, the U.S. EPA National Pollutant Discharge Elimination System (NPDES) permit for TSS was set at 100 mg L^{-1} (Boback et al. 1977).

Similarly, methods were assessed for their usefulness in removing soluble uranium from the liquid effluent (Dugan 1971). In 1971, tests results showed that the addition of lime slurry decreased the soluble uranium concentration of storm sewer effluent. However, the addition of lime to the storm sewer to neutralize acid spills and to prevent corrosion at the lift station was usually associated with higher TSS levels in effluents to the river (Boback 1971b). Other causes of TSS exceeding the limit were related to runoff from the coal pile (Starkey 1968b) and variable pH of the effluent (Boback 1971c).

In summary, the ratios of various ionic species of uranium compounds in waste streams, Paddy's Run Creek, or the Great Miami River is a function of the pH of the water. Based on the high volume of liquid effluent released, many of the uranium species released from the plants would have dissolved in the waste streams, although suspended solids were prevalent in the effluent. Among the suspended solids may have been very small particulates of the insoluble compounds U_3O_8 and UO_2. It is clear that not all the suspended solids measured on a daily basis were uranium, but the average monthly values may provide an upper bound for the amount of insoluble uranium released in liquid effluent.

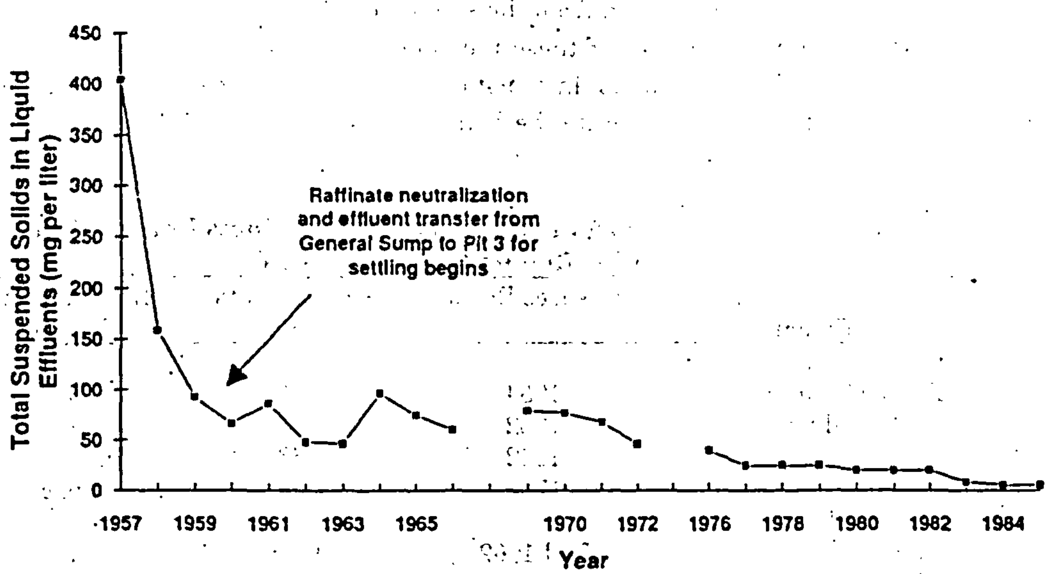

Figure L–10. Annual average concentration of total suspended solids in liquid effluents released at MH 175 to the river. Daily measurements were made beginning in 1956 at MH 175. Major improvements in the liquid effluent treatment in 1958 lead to a significant decrease in TSS.

RADIONUCLIDES OTHER THAN URANIUM

Uranium was the primary material processed at the FMPC with some thorium processing occurring at various times. Most of the feed material had previously been separated chemically from the naturally occurring daughter radionuclides. Consequently, most effluents from the facility consisted primarily of uranium and, when it was being processed, thorium. Beginning in 1953, thorium operations were performed in the Metals Fabrication Plant (Plant 6), Recovery Plant (Plant 8), Special Products Plant (Plant 9) and the Pilot Plant. Thorium oxide for thorium metal conversion was made during the period of 1954 to 1956 by aqueous precipitation of thorium fluoride from an aqueous hydrofluoric acid solution (Jester 1964). Severe corrosion problems, caused by hot nitric-hydrofluoric acid mixtures, forced a change to the more expensive oxalate process in Plant 9. Appendix D in this report, and Appendix C in the Task 4 report, Environmental Pathways — Models and Validation, describe the products of radioactive decay of uranium and thorium. Four isotopes of radium naturally occur as decay products of thorium and uranium. Two of these, ^{228}Ra and ^{224}Ra, are decay products of thorium. Radium-223 is a decay product of ^{235}U, and ^{226}Ra is a decay product of ^{238}U. When the relative importance of releases of these radionuclides to water was assessed for the 1960 to 1962 period, it was found that the radium isotopes were of primary importance (Appendix C, Killough et al. 1993).

Appendix D also describes other radionuclides that were released during FMPC operations from the processing of recycled uranium, that is, uranium that was not completely separated from fission and activation products before it was returned to the FMPC as feed material. Recycled uranium was processed at the FMPC beginning in the fall of 1962 (Voillequé et al. 1991). When recycled uranium was processed, some fission and activation products were discharged from the site in both liquid and airborne effluents. This section provides annual estimates of these radionuclides released in liquid waste from the site. Table L–11 lists these products, information on release and measurement periods, and sources of information used to generate the source terms.

Table L–11. Decay, Fission and Activation Products Released in Liquid Effluents From the FMPC

Materials in Liquid Effluents	Releases Began	Measurements Began	Information Source
Decay Products			
Total Thorium	1954	1956	a, b, c
Total Radium	1952	1955	a
^{226}Ra	1952	1968	a, b, c
^{228}Ra	1954	1968	a, b, c
Fission Products			
^{90}Sr	Fall 1962	1976	d
^{99}Tc	Fall 1962	1969	b, d
^{106}Ru	Fall 1962	1976	b, d
^{137}Cs	Fall 1962	1976	d
Activation Products			
^{237}Np	Fall 1962	1976	d
^{238}Pu	Fall 1962	1976	d
$^{239,240}Pu$	Fall 1962	1976	d

[a] Original analytical data sheets for some periods; NLCO 1955b, NLCO 1956, NLCO 1957, NLCO 1969, NLCO 1970, NLCO 1971, NLCO 1972, NLCO 1973, NLCO 1974.
[b] Various monthly reports including routine operating loss reports, Industrial Hygiene and Radiation Department reports and Aquifer Contamination Reports.
[c] Based on correlations between releases of uranium and other radionuclides when measurements were made; see Table L–13.
[d] Based on correlations between releases of uranium and other radionuclides when measurements were made; see Table L–12.

Thorium and radium were measured in liquid effluents beginning in the early 1950s, and original analytical data sheets for radium measurements were located for 1955, 1956, 1957, 1969 and 1970–1974 (Tables L1–25 to L1–32), and for thorium for 1956 and 1957 (Tables L1–33 and L1–34). Measurements were made on weekly or biweekly composites for radium, and monthly composites for thorium. A regular sampling program for ^{226}Ra and ^{228}Ra was begun in 1968, for ^{99}Tc in 1969, and for all other radionuclides of interest in 1976 (Boback et al. 1987, NLCO 1975). Periodic monthly composite samples from MH 175 were analyzed for ^{99}Tc (technetium) and ^{106}Ru-^{106}Rh (ruthenium-rhodium) activity beginning in the late 1960s when higher levels of beta activity were measured in effluents sent to Waste Pit 3 (Starkey 1968a, NLCO 1971, NLCO 1974). However, the bioassay lab procedure for ^{106}Ru was not documented for those years (Berger et al. 1985). Routinely, monthly composites of the daily samples from MH 175 were analyzed for ^{226}Ra, ^{228}Ra, ^{106}Ru and thorium with annual composites analyzed for the other radionuclides through the mid-1980s. Analysis of ^{232}Th in liquid wastes to the river replaced total thorium measurements in 1984 (Facemire et al. 1985).

Release estimates of these other radionuclides are based on correlations between the total annual releases of uranium and those of the other radionuclides. These ratios of releases, computed for years when measurements were made, provide a basis for estimating the release of the other radionuclides for years when they were not measured. This methodology is described in Appendix D in the present report, and in Appendix C of Task 4 (Killough et al. 1993). Ratios of the annual average activity of a radionuclide (or, quantity of thorium) to the annual uranium quantity were calculated for years when data were available. The measured concentrations at MH 175 reported in analytical data sheets were used to calculate the ratio for some years (NLCO 1955b, NLCO 1956, NLCO 1957, NLCO 1969, NLCO 1970, NLCO 1971, NLCO 1972, NLCO 1973, NLCO 1974). Annual average concentrations of radium, thorium and the fission and activation products in liquid effluents were reported by the FMPC in historic release reports (Boback et al. 1987), and in annual environmental monitoring reports (Boback et al. 1977, Boback et al. 1978, Boback & Ross 1979, Boback & Ross 1980, Boback & Ross 1981, Fleming et al. 1982, Fleming & Ross 1983, Fleming & Ross 1984, Facemire et al. 1985, Aas et al. 1986, Aas et al. 1987, WMCO 1988, WMCO 1989). The annual average uranium concentration at MH 175, or total quantity of uranium to the river was used for these correlations depending upon the source of data (analytical data sheets or total release estimates, respectively). The variability of the release ratio from year to year was considered in deriving the uncertainty associated with the estimated releases of these other radionuclides. The release estimates and uncertainty analysis were computed using Monte Carlo techniques in the Crystal Ball® program (Decisioneering 1993), assuming a lognormal distribution for the ratios of the radionuclide of interest to uranium.

Table L–12 shows the relative concentrations of activation and fission products relative to uranium, μCi (kg U)$^{-1}$, based on thirteen years of measurements. For radium and thorium, the ratios are based on a somewhat longer measurement history. Table L–13 shows that the ratios of releases are based on measurements as early as 1956 for thorium, and 1968 for ^{226}Ra and ^{228}Ra. Measurements of total radium, made in the early 1950s (NLCO 1955b, NLCO 1956, NLCO 1957), were used to calculate a ratio of ^{226}Ra activity (assuming a specific activity of 0.99 μCi per μg Ra) to uranium, which was used to estimate ^{226}Ra releases in the 1950s. During the 1950s, the ^{226}Ra concentrations are higher than in later years because, from October 1955 to August 1958, some of the uranium ore processed was pitchblende, which had very high uranium (and thus decay product) concentrations (See Appendix J). For later years, a second ^{226}Ra ratio (50 $\pm$ 80 μCi (kg U)$^{-1}$) based on measurements made from 1968–1988, was used to calculate releases estimates. A single ratio for ^{228}Ra to uranium (90 $\pm$ 80 μCi (kg U)$^{-1}$), based on measurements made from 1968–1988, was used to calculate ^{228}Ra releases. These estimates were calculated for years when thorium processing occurred, because ^{228}Ra is a decay product of thorium (See Appendix D).

Relative concentrations of thorium with respect to uranium are reported as kilograms of thorium per kilogram of uranium, (kg Th) (kg U)$^{-1}$. Because thorium processing occurred only during specific years, release estimates are calculated for 1954 to 1957, and for 1968–1988. Ratios of thorium to uranium quantities were calculated for two periods: the 1950s and 1964–1988. The ratio for the early time [0.41 $\pm$ 0.04 kg Th (kg U)$^{-1}$] is based on concentrations of thorium to uranium measured at MH 175 in 1956 and 1957 (NLCO 1956, NLCO 1957). For later years, the ratio [0.013 $\pm$ 0.015 kg Th (kg U)$^{-1}$] is based on measurements from 1967–1988.

Table L–12. Relative Concentrations of Activation (Pu, Np) and Fission Products (^{90}Sr, ^{99}Tc, ^{106}Ru, ^{137}Cs) Measured in Liquid Waste Discharges, μCi (kg U)$^{-1}$ [a]

Year	$^{239,240}Pu$	^{238}Pu	^{237}Np	^{137}Cs	^{106}Ru	^{99}Tc	^{90}Sr
1976	0.00024	0.00049	0.00024	24	3.7	1.1×10^4	no data
1977	<0.053	<0.024	<0.48	80	7.8	9.5×10^1	71
1978	<0.038	<0.027	0.036	17	1.2	1.1×10^2	7.8
1979	0.024	0.0082	0.16	5	1.5	2.8×10^3	2.6
1980	2.2	0.006	<0.16	16	1.4	1.4×10^3	4.1
1981	0.05	0.0088	<0.24	4	1.2	7.3×10^3	4.3
1982	0.02	0.0065	0.4	3.7	0.045	1.3×10^4	4.2
1983	0.13	0.0085	<0.30	9.3	0.51	3.5×10^4	9.8
1984	0.049	0.029	0.20	17	0.49	1.9×10^4	12
1985	0.024	0.012	<0.27	16	<0.68	1.3×10^4	8.5
1986	<0.022	<0.022	<0.022	<2.2	<22	3.3×10^3	2
1987	<0.073	<0.072	<0.31	<9.7	<43	3.5×10^3	2.9
1988	<0.028	<0.02	<0.04	<6	<39	7.3×10^3	1.5
Mean	0.31	0.01	0.16	19	2.0	8.9×10^3	11
StdDev	0.76	0.01	0.16	22	2.4	9.7×10^3	19

[a] Data for these ratios of activity (μCi) to quantity (kg) of uranium are taken from Annual Environmental Monitoring Reports (Boback et al. 1977, Boback et al. 1978, Boback & Ross 1979, Boback & Ross 1980, Boback & Ross 1981, Fleming et al. 1982, Fleming & Ross 1983, Fleming & Ross 1984, Facemire et al. 1985, Aas et al. 1986, Aas et al. 1987, WMCO 1988, WMCO 1989).

The result of these computations for thorium are shown in Figure L–11, where the relative quantities of total thorium are compared to the total quantity of uranium discharged in liquid effluents for those years when thorium was processed. The higher thorium releases in the 1950s were related to the fact that thorium oxide for thorium metal conversion was made during the period of 1954 to 1956 by aqueous precipitation of thorium fluoride from an aqueous hydrofluoric acid solution (Jester 1964). This process caused severe corrosion problems, caused by hot nitric-hydrofluoric acid mixtures. For later thorium operations, a change to the more expensive oxalate process in Plant 9 occurred. After 1964, the quantities of thorium discharged to the river were approximately two orders of magnitude less than the quantities of uranium. The thorium releases in the mid-1950s were substantially higher. Similarly, the relative changes in activity of ^{228}Ra and ^{226}Ra in liquid effluents from the FMPC with time, shown in Figure L–12, are similar to the pattern of thorium releases. The highest releases occurred during the 1950s and 1960s, with a gradual decrease in activity in the 1970s and 1980s. Tables L–14 and L–15 show the annual estimates for thorium, ^{228}Ra, and ^{226}Ra, discharged in liquid effluents from the FMPC, along with the uncertainty estimates for each measurement.

Figure L–13 displays the total release estimates for the radionuclides, $^{239,240}Pu$, ^{238}Pu, ^{237}Np, ^{137}Cs, ^{106}Ru, ^{99}Tc, and ^{90}Sr, for all years of operations. Table L–16 provides the annual estimates of fission and activation products discharged in liquid effluents from the FMPC for each year from 1962 to 1988. Because the processing of recycled uranium at the

FMPC did not begin until October 1962, values for 1962 are based on only three months of operation. Since 1962, total releases of ^{99}Tc were approximately 300,000 mCi (300 Ci), with an uncertainty range of 100,000 to 800,000 mCi (100 to 800 Ci). The best estimate for releases of $^{239,240}Pu$ since 1962 is 8.8 mCi, with an uncertainty range of 1.9 to about 30 mCi.

Table L–13. Relative Concentrations of Radium and Thorium to Uranium Measured in Liquid Waste Discharges[a]

Year	^{226}Ra μCi (kg U)$^{-1}$	^{228}Ra μCi (kg U)$^{-1}$ [a]	Thorium kg Th (kg U)$^{-1}$
1955	1600[b]		
1956	220[b]		0.44
1957	530[b]		0.37
Mean (1950s)	780		0.41
Stdev (1950s)	590		0.04
1967			0.012
1968	270	590	0.069
1969	250	390	0.028
1970	104	260	0.015
1971	61	24	0.018
1972	48	13	0.016
1973	21	5.30	0.008
1974	7.50	5.60	0.017
1975	7.02	8.60	0.0035
1976	9.72	11	0.0076
1977	8.00	77	0.0057
1978	3.81	5.10	0.0065
1979	0.68	8.20	0.0061
1980	0.56	5.20	0.0033
1981	19	12	0.0053
1982	4.03	17	0.0052
1983	2.40	11	0.0035
1984	<17	<14	0.0044
Mean	50	90	0.013
Stdev	80	170	0.015

[a] Values are derived from the following sources: routine analytical data sheets for uranium, ^{226}Ra and thorium in the 1950s (see Tables L1-1 to L1-13, L1-26 to L1-28 and L1-34 and L1-36), and ^{228}Ra in 1969, 1967–1975, Boback et al. 1987; 1976–1988, Annual Environmental Monitoring Reports (Boback et al. 1977, Boback et al. 1978, Boback & Ross 1979, Boback & Ross 1980, Boback & Ross 1981, Fleming et al. 1982, Fleming & Ross 1983, Fleming & Ross 1984, Facemire et al. 1985, Aas et al. 1986, Aas et al. 1987, WMCO 1988, WMCO 1989).

[b] For 1955, 1956 and 1957, the ratio is derived from total radium measurements of μμg per mL (Tables L1-26 to L1-28), assuming a specific activity of 0.99 μCi per μg Ra.

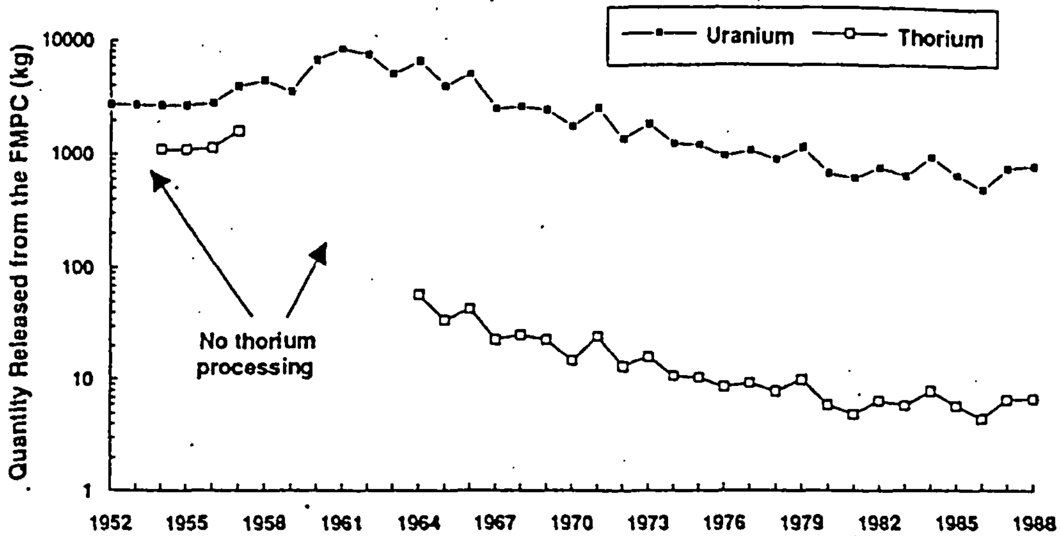

Figure L–11. Relative annual estimates for uranium and thorium released in liquid effluents from the FMPC. Thorium processing occurred from 1954 to 1957 and from 1964 through 1988. The uranium values represent total uranium quantities released to both the Great Miami River and to Paddy's Run Creek. Figure L–2 shows the uranium releases individually to the river and to Paddy's Run.

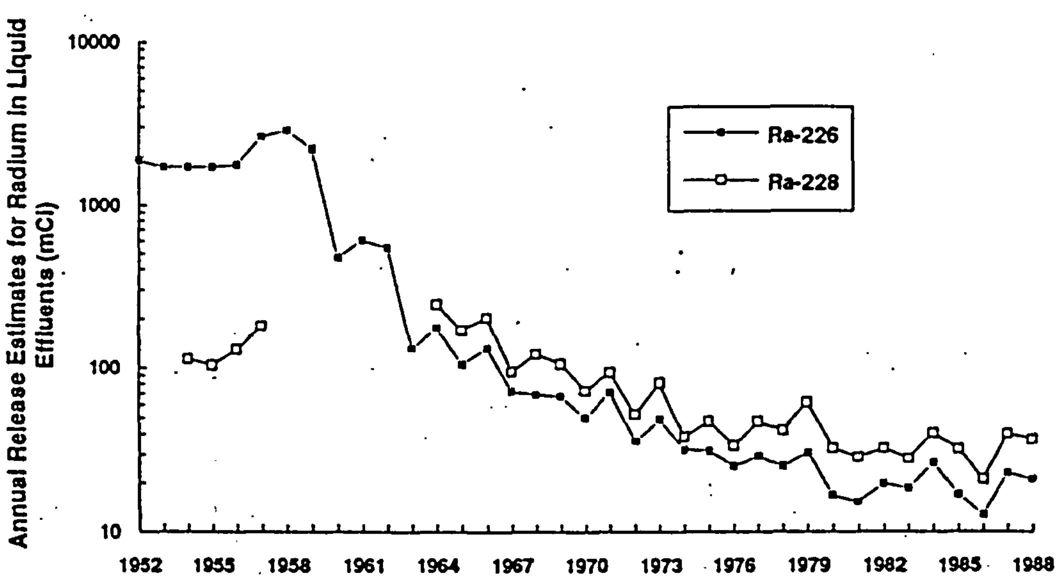

Figure L–12. Annual estimates of ^{226}Ra and ^{228}Ra releases in liquid effluents from the FMPC. Release estimates for ^{228}Ra, a decay product of thorium, are given for 1954–1957, and 1964–1988, the years when thorium processing occurred (see Table L–15).

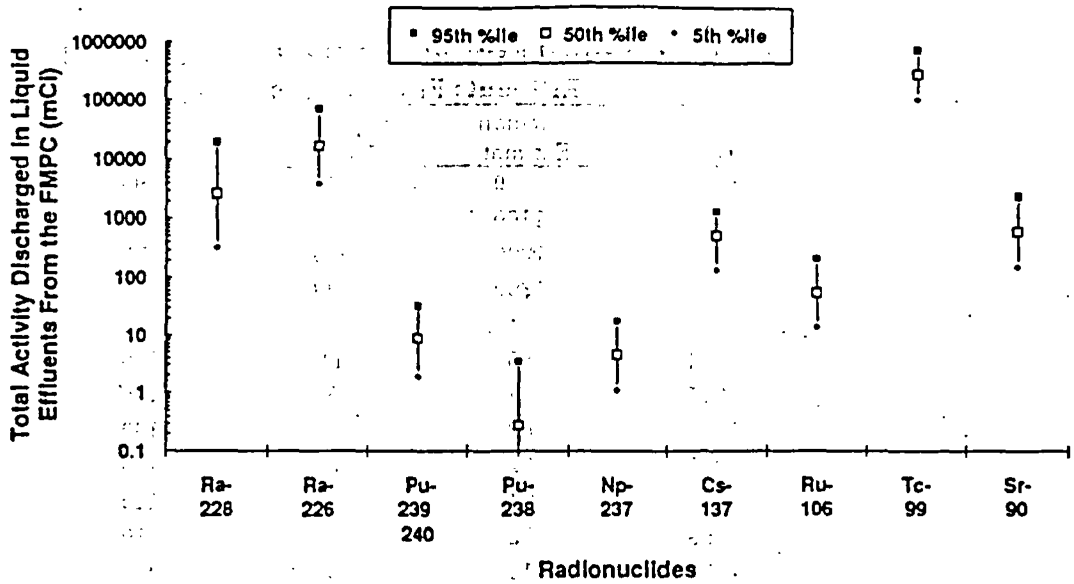

Figure L–13. Estimates of total activity of radium, fission and activation products released from the FMPC in liquid effluents. For radium, the values represent releases from 1952 to 1988; for the other radionuclides, releases occurred from 1962 onward. The uncertainty of each estimate is shown as the 95th and 5th percentiles.

Tables L–14 to L–16 also show the gradual decrease in release estimates in the 1970s and 1980s. These decreases over time for all radioactive materials are related to a general reduction in production activities from the higher levels observed in the fifties and sixties, as well as to a number of changes in liquid effluent handling and treatment at the site, including

- major improvements in the General Sump area for waste effluent processing in 1968, and the
- construction of new wet chemical Waste Pit 5 by 1969.

By 1967, Waste Pit 3 was nearly at its capacity. At the same time, the General Sump was processing large volumes of soluble high beta activity material from a variety of processing campaigns. However, the General Sump was in more frequent need of repairs by the mid–1960s. When holding tanks in the General Sump were being repaired, virtually all effluent from the General Sump was pumped to Waste Pit 3 before proper precipitation and settling could occur. To make more room in the pit, pumping from the waste pit clearwell was increased prior to complete settling of the material. A consequence of this was higher discharges of radionuclides to the river during the sixties.

In 1969, two new 15,000 gallon and a new 50,000 gallon sludge settling tanks in the General Sump area were installed, and a new head tank for regulating continuous discharge to the river was operational (OHIO 1968). Most importantly, the construction of the new wet chemical pit began on July 15, 1968, and was receiving material by the end of that year (Starkey 1968c). The first effluent from the new pit was pumped to the river on January 6, 1969 (Starkey 1969a).

Table L–14. Annual Estimates of Thorium Discharged in Liquid Effluents From the FMPC (kg) [a]

Year	Median Estimate	5th %ile	95th %ile
1954	1100	800	1500
1955	1100	830	1400
1956	1200	910	1500
1957	1600	1300	2100
1964	58	11	280
1965	34	8	150
1966	43	9	190
1967	22	5	100
1968	24	5	110
1969	22	5	110
1970	14	3	63
1971	24	5	110
1972	13	3	50
1973	16	3	67
1974	11	2	48
1975	10	2	48
1976	9	2	43
1977	9	2	44
1978	8	2	41
1979	10	2	51
1980	6	1	28
1981	5	1	24
1982	7	1	30
1983	6	1	26
1984	8	2	37
1985	6	1	25
1986	4	1	20
1987	7	1	33
1988	7	1	35
Total all years	5800	3800	9400

[a] Estimates and uncertainties were calculated with CrystalBall Version 3.0 (Decisioneering 1993). No thorium processing occurred in 1952, 1953, or 1958–1963 (see Appendix C).

In the sixties, unusually high soluble beta activity, measured in the General Sump and the waste pits, was attributed to ^{106}Ru and ^{99}Tc from various processing campaigns such as the processing of NFS feed material which contained ^{106}Ru , or to high ^{236}U refinery runs (Starkey 1967b). In the oxidized state, both are soluble in basic and acidic solutions, so that they were not effectively removed by passage through the General Sump. By 1970, the beta activity attributed to soluble ^{106}Ru and ^{99}Tc had gradually decreased from the levels seen previously (Boback 1969).

Table L–15. Annual Estimates of ^{228}Ra and ^{226}Ra Discharged in Liquid Effluents From the FMPC (mCi)[a]

	Radium-228			Radium-226		
Year	Median Estimate	5th %ile	95th %ile	Median Estimate	5th %ile	95th %ile
1952				1900	616	5300
1953				1700	535	4800
1954	110	14	930	1700	584	5300
1955	100	12	710	1700	622	5200
1956	130	17	1200	1800	623	5400
1957	180	25	1300	2600	907	7700
1958				2900	1105	8500
1959				2200	822	6400
1960				480	46	3300
1961				600	54	6300
1962				540	52	5400
1963				130	21	870
1964	250	36	2000	180	27	1100
1965	170	23	1400	110	17	680
1966	200	25	1600	130	21	830
1967	96	13	820	72	10	460
1968	120	15	1050	69	13	460
1969	110	11	880	68	11	490
1970	74	9	670	50	7	320
1971	95	13	800	72	12	500
1972	53	7	450	36	5	240
1973	82	10	690	49	8	360
1974	38	5	340	32	5	200
1975	48	6	440	31	5	200
1976	34	4	320	25	4	170
1977	48	6	370	29	5	179
1978	42	5	300	25	4	150
1979	63	6	500	31	4	200
1980	33	4	250	17	3	120
1981	29	4	280	15	2	100
1982	33	5	260	20	3	120
1983	28	4	250	18	2	110
1984	41	5	370	27	4	180
1985	32	4	280	17	3	100
1986	21	3	180	13	2	83
1987	40	5	300	23	3	130
1988	37	4	260	21	3	140
Totals	2700	330	20000	18000	15000	22000

[a] Estimates and uncertainties were calculated with CrystalBall Version 3.0 (Decisioneering 1993).

[b] Radium-228 is a decay product of thorium; estimates of ^{228}Ra releases are given for 1954–1957, and 1964–1988, the years when thorium processing occurred.

Table L–16. Annual Activity Estimates of Fission and Activation Products Discharged in Liquid Effluents From the FMPC (mCi)[a]

Year[b]	$^{239,240}Pu$	^{238}Pu	^{237}Np	^{137}Cs	^{106}Ru	^{99}Tc	^{90}Sr
1962	0.39	0.01	0.21	25	2.6	11000	270
1963	1.29	0.04	0.69	82	8.6	38000	900
1964	1.23	0.04	0.66	78	8.2	36000	860
1965	0.87	0.03	0.46	55	5.8	26000	610
1966	1.35	0.05	0.72	86	9.0	39600	940
1967	0.57	0.02	0.30	36	3.8	16600	390
1968	0.72	0.02	0.38	46	4.8	21000	500
1969	0.69	0.02	0.37	44	4.6	20200	480
1970	0.45	0.02	0.24	29	3.0	13000	310
1971	0.66	0.02	0.35	42	4.4	19000	460
1972	0.33	0.01	0.18	21	2.2	9700	230
1973	0.51	0.02	0.27	32	3.4	15000	360
1974	0.22	0.01	0.12	14	1.4	6300	150
1975	0.30	0.01	0.16	19	2.0	8900	210
1976	0.22	0.01	0.12	14	1.5	6400	150
1977	0.27	0.01	0.15	17	1.8	8000	190
1978	0.26	0.01	0.14	16	1.7	7500.	180
1979	0.32	0.01	0.17	20	2.1	9200	220
1980	0.19	0.01	0.10	12	1.3	5600	130
1981	0.18	0.01	0.10	11	1.2	5300	130
1982	0.23	0.01	0.12	14	1.5	6600	160
1983	0.18	0.01	0.09	11	1.2	5200	120
1984	0.27	0.01	0.14	17	1.8	7900	190
1985	0.17	0.01	0.09	10	1.1	4800	120
1986	0.13	0.00	0.07	8	0.84	3700	88
1987	0.21	0.01	0.11	13	1.4	6200	150
1988	0.22	0.01	0.12	14	1.5	6500	160
Total: all years	8.8	0.28	4.4	540	56	300000	600
(5th–95th %ile)	(1.9–33)	(0.16–3.4)	(1.1–18)	(140–1900)	(14–220)	(110000–800000)	(1500–24000)

[a] The median estimates are based on the average ratio of measured activity of these radionuclides to the quantity of uranium released in liquid effluent from 1976 onward. The values are reported in millicuries (mCi); one mCi is equal to 1000 microcuries (µCi) or 0.001 curie (Ci).

[b] Processing of recycled uranium at the FMPC did not begin until October 1962. Consequently, values for 1962 are based on only three months of processing.

By 1969, when the average concentration of ^{228}Ra in the plant effluent was about 1.8 disintegrations per minute per milliliter (d/m/mL) (Table L1–29, annex), the Oak Ridge Operations Atomic Energy Commission requested the concentration of ^{228}Ra in the wastes discharged to the river be reduced (Boback 1969). At that time, the Pilot Plant thorium extraction process was the major source of this radionuclide. A barium sulfate precipitation at the Pilot Plant and additional treatment at the General Sump were intended to reduce the ^{228}Ra in the extraction waste stream before being pumped to Waste Pit 5. Beginning the oxalate process for thorium recovery in Plant 8 in 1969, however, prevented lowering the concentrations quickly. By mid–1970, work at the General Sump had increased as a result of processing thorium scrap in Plant 8. The clear liquid from this process was pumped to the Chemical Waste Pit 5 and the solids were reprocessed through Plant 8. The reduction in average ^{228}Ra concentration at MH 175 from 3.2 d/m/mL in December 1969 to 1.6 d/m/mL in March 1970 (Table L1–29) occurred when there were no thorium extraction operations in the Pilot Plant during that period. Even though all thorium effluent from both Plant 8 and the Pilot Plant was pumped to Pit 5, ^{228}Ra in the effluent from the General Sump to the river still averaged 5.0 d/m/mL in August 1970, and was attributed to incoming effluents from various plants (Boback 1970). By the end of 1970, the concentration of ^{228}Ra had declined. In 1971, the General Sump began solidifying certain ^{228}Ra–bearing wastes from Plant 8 for shipment and burial offsite (Pennack 1971).

SUMMARY AND CONCLUSIONS

Liquid wastes at the FMPC came from three main sources: (1) process water from the production area via the General Sump and clearwell portion of the waste pits, (2) from the sanitary sewer treatment plant, and (3) from the storm sewer system. The facilities for handling liquid wastes from the process areas included collection sumps and treatment equipment at each plant to remove uranium from process waste water before it was pumped to the General Sump. From the General Sump, the effluent was pumped to the waste pits where settling occurred, after which the liquid was decanted to the clearwell portion of the pit. Key improvements in the liquid handling system at the FMPC, especially in 1958, 1968 and 1985, were reflected in noticeable declines in concentrations of uranium, thorium and other radionuclides, as well as in total suspended solids measured at the discharge point to the river.

Liquid effluent left the FMPC site at two locations. The main pipeline exited via Manhole 175 (MH 175) into the Great Miami River at a point almost directly east of the plant site. Liquid waste water also left the site via the storm sewer outfall ditch and runoff into Paddy's Run Creek, when the storm sewer lift station could not handle the runoff volume. Effluent volume and total uranium concentration were measured routinely at both locations (MH 175 and the storm sewer outfall ditch). Daily analytical data sheets, and monthly reports of effluent volume and uranium discharged form the basis of our source term computations.

Table L–17 summarizes our estimates for releases of materials in liquid effluents from the FMPC for all years of operation. Our best estimate of uranium released to the Great Miami River for all years is 82,000 kg. The 5th to 95th percentile uncertainty range is 71,000 to 94,000 kg of uranium. The sources of uncertainty for losses through MH 175 to the Great Miami River come primarily from the analytical errors in the daily measurements of water flow, and in sampling and determination of uranium concentration in the water. Some estimates of uranium in liquid wastes have been made by others on an annual basis (Boback 1971a), or in summary reports evaluating the past discharge history of the facility (Rathgens 1977, Boback et al., 1985). These estimates of uranium to surface water from 1951 through 1984 range from 74,000 to 77,000 kg (Boback et al. 1987, Galper 1988) and fall within the uncertainty range of our estimates. Revisions to historic discharge reports generally focused on amending estimates of uranium loss to airborne effluents, and did not include updated figures for liquid effluents (Boback et al. 1985, Boback et al. 1987).

Table L–17. Summary of Total Estimates of Radioactive Materials Released From the FMPC in Liquid Effluents For All Years of Operation

Material Released to Great Miami River	Median Value	Uncertainty Range (5th %ile to 95th %ile)
	Quantity (kg)	Quantity (kg)
Uranium	82,000	71,000 to 94,000
Uranium (To Paddy's Run)	17,000	14,000 to 20,000
Thorium	5,800	3800 to 9400
	Activity (Ci)	Activity (Ci)
^{228}Ra	2.7	0.33 to 20
^{226}Ra	18	15 to 22
$^{239,240}Pu$	0.0088	0.0019 to 0.033
^{238}Pu	0.00028	0.00016 to 0.0034
^{237}Np	0.0044	0.0011 to 0.018
^{137}Cs	0.54	0.14 to 1.9
^{106}Ru	0.056	0.014 to 0.22
^{99}Tc	300	110 to 800
^{90}Sr	6.0	1.5 to 24

The total release estimate for uranium to Paddy's Run via the storm sewer outfall ditch and runoff is 17,000 kg of uranium. The 5th to 95th percentile uncertainty range is 14,000 to 20,000 kg of uranium. In addition to analytical errors, sources of uncertainty included overflow at the flow meter stations when rainfall, and consequently runoff, were quite high and unmeasured uranium losses through runoff from the west side of the facility directly into Paddy's Run. These latter two, undocumented sources of uranium to Paddy's Run are incorporated into our final release estimates.

Losses to Paddy's Run show much more month to month variation than do the uranium loss estimates to the Great Miami River. The highest annual releases of uranium occurred from 1960 to 1964, when the average quantity of uranium discharged through MH 175 to the river was approximately 500 kg each month, about 3 to 4 times greater than the average quantity of uranium lost to Paddy's Run each month.

The other materials released at various times over the years include decay, fission and activation products of uranium, thorium and recycled uranium. Recycled uranium was not processed until late 1962, so releases of fission and activation products began at that time. Releases of thorium, and one of its decay products, ^{228}Ra, occurred when thorium was processed at the site: 1954–1957, and 1964–1988. Releases of ^{226}Ra occurred throughout the history of the site, and the total release is estimated at 18,000 mCi or 18 Ci, with uncertainty range of 15 to 22 Ci. These values will be used to calculate radiation doses to the population in the vicinity of the FMPC, which will be reported in our final task report.

The chemical form of uranium in liquid effluents is not known with certainty, but several uranium species of both the +4 and +6 oxidation states may have been present in solution in liquid waste streams during this period. The ratios of these various ionic species in the process waste streams, in Paddy's Run Creek, or in the main effluent pipeline to the river, would be a function of the pH of the water. Some uranium-containing suspended solids that were released into the waste streams might have dissolved during dilution downstream from the FMPC.

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ANNEX

DATA TABULATIONS

MANHOLE 175 VOLUME AND URANIUM CONCENTRATION

Tables L1-1 to L1-13 contain daily uranium concentration measurements at Manhole 175 for 1954–1964, 1966, 1967 and 1969, with effluent volume data for 1960, 1961 and 1962 in Tables L1-6, L1-7, and L1-8. The uranium concentration (mg L^{-1}) was determined on 24-hour composite samples. From these measurements, daily and monthly estimates of uranium lost to the river were calculated.

OUTFALL TO PADDY'S RUN

Tables L1-14 to L1-22 contain data on the overflow of effluent to the Storm Sewer Outfall Ditch and Paddy's Run for 1954–1957, 1960–1964, and 1966. The tables list the uranium concentrations measured in grab samples taken periodically during the outfall events, and, for some years, the dates of the overflow (outfall) of the storm sewer lift station to Paddy's Run and the effluent volume measured at the V-notch weir station.

Table L1-23 reports the quantities of uranium in the storm sewer system that are released to the river via the storm sewer lift station, or through the storm sewer outfall ditch to Paddy's Run. Monthly rainfall amounts are also given.

TOTAL SUSPENDED SOLIDS

Table L1-24 lists the daily measurements of total suspended solids (TSS) in liquid effluents discharged to the river in 1957. Table L1-25 provides monthly averages for TSS measured at MH 175 before discharge to the river. The monthly averages are based on daily measurements reported in analytical data sheets from the Bioassay Department at FMPC.

RELEASE OF OTHER RADIONUCLIDES

Tables L1-26 to L1-33 present data on the concentrations of total radium (Tables L1-26 to L1-28), ^{228}Ra and ^{226}Ra (Tables L1-29 to L1-33) released in liquid effluents from the FMPC for various times from 1955 to 1974. Tables L1-34 to L1-35 show the concentration of thorium measured at MH 175 in 1956 and 1957.

VOLUME OF EFFLUENT

Table L1-36 lists the annual volume of effluent discharged to the Great Miami River Via Manhole 175, and to Paddy's Run through the storm sewer outfall ditch.

Table L1–1. Measured Concentrations and Calculated Quantities of Uranium at Manhole 175 in 1954 and 1955

Date 1954	Uranium[a] ($mg\ L^{-1}$)	Total U (kg)
10-Aug	5.35	22
2-Sep	1.41	6
9-Sep	3.10	13
9-Sep	1.95	8
Average	2.95	12

Date 1955	Uranium[a] ($mg\ L^{-1}$)	Total U (kg)
21-Sep	0.31	1
24-Sep	2.27	9
27-Sep	0.87	4
3-Oct	0.82	3
6-Oct	2.16	9
9-Oct	0.41	2
12-Oct	2.37	10
18-Oct	0.82	3
21-Oct	1.13	5
24-Oct	1.24	5
27-Oct	0.89	4
30-Oct	1.34	6
2-Nov	7.62	32
5-Nov	0.81	3
9-Nov	2.06	9
12-Nov	0.83	3
15-Nov	0.82	3
18-Nov	0.68	3
21-Nov	1.20	5
24-Nov	0.37	2
27-Nov	0.81	3
30-Nov	6.18	26
3-Dec	1.79	7
6-Dec	0.67	3
9-Dec	0.60	2
12-Dec	0.80	3
15-Dec	0.60	2
18-Dec	1.00	4
21-Dec	2.13	9
24-Dec	0.77	3
28-Dec	0.87	4
Totals		190
Average	1.4	5.9
StdDev	1.6	6.5
Max	7.6	32
Min	0.3	1

[a] From NLCO 1955; original analytical data sheets from the Bioassay Department at FMPC.

Table L1-2. Measured Concentrations and Calculated Quantities of Uranium at Manhole 175 in 1956

1956 Date	U[a] (mg L^{-1})	Total U (kg)	1956 Date	U[a] (mg L^{-1})	Total U (kg)	1956 Date	U[a] (mg L^{-1})	Total U (kg)
1-Jan	1.26	1	16-May	0.70	3	26-Jun	0.61	3
7-Jan	0.25	2	17-May	0.78	2	27-Jun	0.82	9
10-Jan	0.58	9	18-May	0.59	27	28-Jun	2.24	3
13-Jan	2.13	1	19-May	6.44	6	29-Jun	0.61	2
16-Jan	0.23	3	20-May	1.37	9	30-Jun	0.41	4
19-Jan	0.77	1	21-May	2.15	4	1-Jul	1.02	5
22-Jan	0.30	2	22-May	1.01	5	2-Jul	1.22	7
25-Jan	0.58	7	23-May	1.17	10	3-Jul	1.63	6
28-Jan	1.74	7	24-May	2.44	5	4-Jul	1.43	4
1-Feb	1.74	6	25-May	1.27	9	5-Jul	1.02	6
4-Feb	1.55	13	26-May	2.15	4	6-Jul	1.33	3
15-Apr	3.16	4	27-May	0.98	5	7-Jul	0.82	4
17-Apr	0.99	4	28-May	1.17	12	8-Jul	1.02	6
18-Apr	0.99	9	29-May	2.93	9	9-Jul	1.47	5
19-Apr	2.17	8	30-May	2.15	3	10-Jul	1.26	1
20-Apr	1.95	6	31-May	0.78	44	11-Jul	0.35	3
21-Apr	1.37	3	1-Jun	10.54	6	12-Jul	0.84	3
22-Apr	0.78	10	2-Jun	1.37	6	13-Jul	0.84	2
23-Apr	2.34	8	3-Jun	1.56	8	14-Jul	0.36	5
24-Apr	1.95	39	4-Jun	2.04	10	15-Jul	1.26	3
25-Apr	9.47	10	5-Jun	2.45	4	16-Jul	0.84	9
26-Apr	2.44	23	6-Jun	0.92	3	17-Jul	2.10	2
27-Apr	5.47	3	7-Jun	0.82	8	18-Jul	0.48	8
28-Apr	0.68	8	8-Jun	1.84	3	19-Jul	1.89	5
29-Apr	1.95	3	9-Jun	0.61	2	20-Jul	1.26	2
30-Apr	0.78	4	10-Jun	0.41	2	21-Jul	0.55	3
1-May	1.01	18	11-Jun	0.48	3	22-Jul	0.63	6
2-May	4.29	4	12-Jun	0.70	5	23-Jul	1.47	3
3-May	0.97	15	13-Jun	1.22	8	24-Jul	0.74	3
4-May	3.51	11	14-Jun	1.94	3	25-Jul	0.84	3
5-May	2.54	6	15-Jun	0.82	3	26-Jul	0.84	3
6-May	1.36	4	16-Jun	0.79	3	27-Jul	0.74	4
7-May	0.98	3	17-Jun	0.82	1	28-Jul	0.94	8
8-May	0.73	8	18-Jun	0.30	3	29-Jul	1.89	6
9-May	1.95	6	19-Jun	0.61	5	30-Jul	1.47	8
10-May	1.37	4	20-Jun	1.12	5	31-Jul	1.89	7
11-May	1.07	5	21-Jun	1.22	16			
12-May	1.17	3	22-Jun	3.88	8	Totals		770
13-May	0.78	12	23-Jun	2.04	3	Average	1.57	7
14-May	2.93	22	24-Jun	0.82	3	StdDev	1.52	6
15-May	5.37	3	25-Jun	0.72	3	Max	10.54	44
						Min	0.23	1

[a] From NLCO 1956; original analytical data sheets from the Bioassay Department at FMPC.

Table L1–3A. Measured Concentrations and Calculated Quantities of Uranium at Manhole 175 in 1957

1957 Date	U[a] (mg L^{-1})	Total U (kg)	1957 Date	U[a] (mg L^{-1})	Total U (kg)	1957 Date	U[a] (mg L^{-1})	Total U (kg)
1-Jan	3.42	14	11-Feb	11.64	48	23-Mar	2.72	11
2-Jan	1.90	8	12-Feb	15.52	65	24-Mar	2.38	10
3-Jan	3.99	17	13-Feb	17.46	73	25-Mar	2.38	10
4-Jan	6.65	28	14-Feb	9.70	40	26-Mar	19.40	81
5-Jan	1.71	7	15-Feb	10.96	46	27-Mar	2.94	12
6-Jan	1.33	6	16-Feb	5.82	24	28-Mar	6.62	28
7-Jan	1.33	6	17-Feb	9.70	40	29-Mar	4.24	18
8-Jan	3.61	15	18-Feb	5.96	25	30-Mar	1.48	6
9-Jan	8.74	36	19-Feb	5.62	23	31-Mar	1.48	6
10-Jan	3.61	15	20-Feb	1.70	7	1-Apr	2.20	9
11-Jan	1.33	6	21-Feb	2.56	11	2-Apr	2.40	10
12-Jan	2.76	11	22-Feb	1.88	8	3-Apr	2.38	10
13-Jan	2.28	9	23-Feb	1.20	5	4-Apr	2.94	12
14-Jan	6.46	27	24-Feb	4.26	18	5-Apr	3.68	15
15-Jan	4.18	17	25-Feb	4.26	18	6-Apr	5.16	21
16-Jan	5.51	23	26-Feb	3.92	16	7-Apr	1.84	8
17-Jan	2.09	9	27-Feb	5.28	22	8-Apr	3.50	15
18-Jan	5.78	24	28-Feb	7.14	30	9-Apr	4.42	18
19-Jan	2.66	11	1-Mar	2.38	10	10-Apr	28.40	118
20-Jan	5.82	24	2-Mar	8.34	35	11-Apr	2.58	11
21-Jan	3.10	13	3-Mar	1.02	4	12-Apr	2.02	8
22-Jan	5.82	24	4-Mar	0.86	4	13-Apr	0.84	3
23-Jan	5.28	22	5-Mar	0.80	3	14-Apr	2.02	8
24-Jan	5.82	24	6-Mar	2.04	8	15-Apr	2.94	12
25-Jan	1.54	6	7-Mar	1.54	6	16-Apr	4.42	18
26-Jan	2.12	9	8-Mar	4.08	17	17-Apr	3.50	15
27-Jan	4.42	18	9-Mar	7.76	32	18-Apr	1.84	8
28-Jan	9.18	38	10-Mar	5.78	24	19-Apr	4.04	17
29-Jan	15.52	65	11-Mar	3.24	13	20-Apr	5.88	24
30-Jan	7.76	32	12-Mar	13.58	56	21-Apr	1.10	5
31-Jan	7.76	32	13-Mar	5.82	24	22-Apr	0.74	3
1-Feb	7.82	0	14-Mar	1.96	8	23-Apr	1.24	5
2-Feb	2.56	33	15-Mar	3.92	16	24-Apr	0.76	3
3-Feb	2.38	11	16-Mar	8.74	36	25-Apr	1.10	5
4-Feb	2.38	10	17-Mar	6.46	27	26-Apr	0.54	2
5-Feb	5.52	10	18-Mar	1.54	6	27-Apr	0.24	1
6-Feb	3.58	23	19-Mar	7.98	33	28-Apr	1.26	5
7-Feb	4.42	15	20-Mar	1.36	6	29-Apr	2.02	8
8-Feb	5.62	18	21-Mar	1.02	4	30-Apr	3.50	15
9-Feb	7.74	23	22-Mar	3.82	16	1-May	2.20	9
10-Feb	3.58	32	23-Mar	2.72	11	2-May	1.10	5

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Table L1-3B. Measured Concentrations and Calculated Quantities of Uranium at Manhole 175 in 1957

1957 Date	U[a] (mg L^{-1})	Total U (kg)	1957 Date	U[a] (mg L^{-1})	Total U (kg)	1957 Date	U[a] (mg L^{-1})	Total U (kg)
3-May	0.36	1	14-Jun	0.54	2	25-Jul	0.94	4
4-May	0.44	2	15-Jun	0.98	4	26-Jul	1.02	4
5-May	1.20	5	16-Jun	2.00	8	27-Jul	2.52	10
6-May	0.70	3	17-Jun	1.54	6	28-Jul	0.70	3
7-May	1.48	6	18-Jun	3.82	16	29-Jul	0.80	3
8-May	0.92	4	19-Jun	1.28	5	30-Jul	1.08	4
9-May	0.74	3	20-Jun	2.18	9	31-Jul	1.68	7
11-May	1.20	5	21-Jun	0.72	3	1-Aug	1.28	5
12-May	1.60	7	22-Jun	0.72	3	2-Aug	0.68	3
13-May	1.48	6	23-Jun	0.86	4	3-Aug	0.86	4
14-May	3.72	15	24-Jun	1.10	5	4-Aug	1.48	6
15-May	1.08	4	25-Jun	1.60	7	5-Aug	2.52	10
16-May	1.10	5	26-Jun	0.86	4	6-Aug	0.60	2
17-May	0.94	4	27-Jun	8.94	37	7-Aug	1.78	7
18-May	1.20	5	28-Jun	1.54	6	8-Aug	2.10	9
19-May	0.72	3	29-Jun	0.90	4	9-Aug	2.32	10
20-May	2.54	11	30-Jun	1.14	5	10-Aug	1.90	8
21-May	16.32	68	1-Jul	0.62	3	11-Aug	6.72	28
22-May	8.20	34	2-Jul	1.90	8	12-Aug	0.24	1
23-May	1.82	8	3-Jul	1.06	4	13-Aug	3.84	16
24-May	1.82	8	4-Jul	1.14	5	14-Aug	1.34	6
25-May	1.70	7	5-Jul	0.34	1	15-Aug	1.54	6
26-May	9.64	40	6-Jul	0.94	4	16-Aug	0.80	3
27-May	0.92	4	7-Jul	1.22	5	17-Aug	1.48	6
28-May	0.54	2	8-Jul	0.82	3	18-Aug	1.26	5
29-May	1.10	5	9-Jul	0.72	3	19-Aug	1.44	6
30-May	0.72	3	10-Jul	1.26	5	20-Aug	0.66	3
31-May	0.72	3	11-Jul	2.10	9	21-Aug	0.78	3
1-Jun	0.82	3	12-Jul	1.18	5	22-Aug	0.60	2
2-Jun	1.10	5	13-Jul	0.86	4	23-Aug	0.44	2
3-Jun	0.72	3	14-Jul	1.14	5	24-Aug	1.34	6
4-Jun	1.10	5	15-Jul	0.34	1	25-Aug	2.88	12
5-Jun	0.74	3	16-Jul	2.66	11	26-Aug	0.96	4
6-Jun	1.44	6	17-Jul	1.42	6	27-Aug	0.96	4
7-Jun	0.90	4	18-Jul	2.28	9	28-Aug	0.36	1
8-Jun	1.14	5	19-Jul	3.16	13	29-Aug	0.96	4
9-Jun	0.54	2	20-Jul	1.26	5	30-Aug	0.42	2
10-Jun	1.26	5	21-Jul	0.64	3	31-Aug	1.16	5
11-Jun	0.92	4	22-Jul	3.58	15	1-Sep	0.54	2
12-Jun	2.36	10	23-Jul	0.94	4	2-Sep	1.02	4
13-Jun	0.88	4	24-Jul	1.48	6	3-Sep	1.16	5

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Table L1–3C. Measured Concentrations and Calculated Quantities of Uranium at Manhole 175 in 1957 (cont'd)

1957 Date	U[a] (mg L^{-1})	Total U (kg)	1957 Date	U[a] (mg L^{-1})	Total U (kg)	1957 Date	U[a] (mg L^{-1})	Total U (kg)
4-Sep	0.96	4	16-Oct	1.70	7	26-Nov	0.76	3
5-Sep	0.75	3	17-Oct	2.44	10	27-Nov	1.24	5
6-Sep	0.60	2	18-Oct	2.06	9	28-Nov	2.14	9
7-Sep	0.48	2	19-Oct	0.82	3	29-Nov	0.90	4
8-Sep	0.58	2	20-Oct	1.32	5	30-Nov	0.86	4
9-Sep	0.60	2	21-Oct	1.12	5	1-Dec	1.48	6
10-Sep	1.76	7	22-Oct	0.56	2	2-Dec	0.82	3
11-Sep	1.56	6	23-Oct	2.26	9	3-Dec	2.62	11
12-Sep	1.38	6	24-Oct	1.50	6	4-Dec	0.98	4
13-Sep	1.52	6	25-Oct	1.64	7	5-Dec	0.66	3
14-Sep	0.52	2	26-Oct	1.12	5	6-Dec	2.14	9
15-Sep	0.60	2	27-Oct	1.38	6	7-Dec	1.14	5
16-Sep	0.92	4	28-Oct	0.84	3	8-Dec	1.48	6
17-Sep	3.52	15	29-Oct	0.36	1	9-Dec	1.48	6
18-Sep	0.96	4	30-Oct	4.14	17	10-Dec	1.32	5
19-Sep	0.66	3	31-Oct	1.50	6	11-Dec	2.78	12
20-Sep	0.58	2	1-Nov	1.08	4	12-Dec	1.96	8
21-Sep	1.58	7	2-Nov	0.60	2	13-Dec	1.92	8
22-Sep	0.98	4	3-Nov	1.02	4	14-Dec	0.76	3
23-Sep	1.10	5	4-Nov	0.96	4	15-Dec	3.26	14
24-Sep	0.38	2	5-Nov	4.88	20	16-Dec	0.56	2
25-Sep	0.52	2	6-Nov	2.64	11	17-Dec	0.58	2
26-Sep	3.34	14	7-Nov	0.44	2	18-Dec	2.68	11
27-Sep	0.42	2	8-Nov	0.98	4	19-Dec	1.92	8
28-Sep	0.82	3	9-Nov	0.98	4	20-Dec	0.96	4
29-Sep	0.56	2	10-Nov	0.84	3	21-Dec	0.62	3
30-Sep	0.50	2	11-Nov	0.56	2	22-Dec	0.76	3
1-Oct	0.34	1	12-Nov	1.16	5	23-Dec	0.34	1
2-Oct	1.56	6	13-Nov	1.36	6	24-Dec	0.76	3
3-Oct	1.76	7	14-Nov	1.64	7	25-Dec	0.52	2
4-Oct	0.70	3	15-Nov	0.80	3	26-Dec	0.58	2
5-Oct	0.40	2	16-Nov	0.46	2	27-Dec	0.38	2
6-Oct	0.50	2	17-Nov	0.82	3	28-Dec	0.46	2
7-Oct	0.86	4	18-Nov	1.96	8	29-Dec	0.66	3
9-Oct	1.32	5	19-Nov	1.54	6	30-Dec	0.90	4
10-Oct	1.88	8	20-Nov	1.46	6	31-Dec	1.08	4
11-Oct	0.76	3	21-Nov	1.24	5	Total		3700
12-Oct	0.18	1	22-Nov	0.66	3	Average	2.5	10
13-Oct	0.38	2	23-Nov	0.98	4	StdDev	3.1	13
14-Oct	0.86	4	24-Nov	1.18	5	Max	28	118
15-Oct	1.00	4	25-Nov	1.10	5	Min	0.18	0.75

[a] From NLCO 1957; original analytical data sheets from the Bioassay Department at FMPC.

Table L1-4A. Measured and Calculated Quantities of U at MH 175 in 1958

1958 Date	U[a] (mg L^{-1})	Total U (kg)	1958 Date	U[a] (mg L^{-1})	Total U (kg)	1958 Date	U[a] (mg L^{-1})	Total U (kg)
1-Jan	1.46	6	16-Feb	0.88	4	4-Jul	0.94	5
2-Jan	0.94	4	17-Feb	3.78	16	5-Jul	1.4	6
3-Jan	0.96	4	18-Feb	3.22	13	6-Jul	1.72	7
4-Jan	1.92	8	19-Feb	1.20	5	7-Jul	2.1	8
5-Jan	0.76	3	20-Feb	0.96	4	8-Jul	1.72	7
6-Jan	0.84	3	21-Feb	3.86	16	9-Jul	4.6	18
7-Jan	0.58	2	22-Feb	3.42	14	10-Jul	7.2	27
8-Jan	0.68	3	23-Feb	1.08	4	11-Jul	1.92	7
9-Jan	0.76	3	24-Feb	0.66	2	12-Jul	0.86	3
10-Jan	0.56	2	25-Feb	0.92	3	13-Jul	0.94	4
11-Jan	0.32	1	26-Feb	1.14	4	14-Jul	0.86	3
12-Jan	1.18	4	27-Feb	2.78	11	15-Jul	1.44	5
13-Jan	0.76	3	28-Feb	2.78	11	16-Jul	4.9	20
14-Jan	2.5	9	1-Mar	2.88	11	17-Jul	3.6	14
15-Jan	2.68	10	2-Mar	0.72	3	18-Jul	2.9	11
16-Jan	0.74	3	3-Mar	1.20	5	19-Jul	2.6	10
17-Jan	0.92	3	4-Mar	0.68	3	20-Jul	1.66	6
18-Jan	0.8	3	5-Mar	0.66	2	21-Jul	1.53	7
19-Jan	1.04	4	6-Mar	2.68	10	22-Jul	2.2	8
20-Jan	0.84	3	7-Mar	1.86	7	23-Jul	3.4	14
21-Jan	1.5	6	8-Mar	1.86	7	24-Jul	1.3	5
22-Jan	5.78	22	9-Mar	1.86	7	25-Jul	1.22	5
23-Jan	0.86	3	10-Mar	7.00	27	26-Jul	0.74	3
24-Jan	1.5	6	11-Mar	3.50	14	27-Jul	2.2	8
25-Jan	1.92	7	12-Mar	1.24	5	28-Jul	2.28	9
26-Jan	1.5	6	13-Mar	4.32	17	29-Jul	3.22	12
27-Jan	2.56	10	14-Mar	2.88	12	30-Jul	3.56	13
28-Jan	2.56	10	15-Mar	1.64	6	31-Jul	6.7	25
29-Jan	8.02	30	16-Mar	1.24	5	1-Aug	12.3	59
30-Jan	1.4	5	17-Mar	1.86	7	2-Aug	1.62	7
31-Jan	1.08	4	18-Mar	0.47	2	4-Aug	3.36	15
1-Feb	0.72	3	19-Mar	0.86	3	5-Aug	2.72	14
2-Feb	1.28	5	20-Mar	0.54	2	7-Aug	4.28	19
3-Feb	0.86	3	21-Mar	1.72	7	8-Aug	1.22	5
4-Feb	0.74	3	22-Mar	1.36	5	9-Aug	2.10	8
5-Feb	1.46	6	23-Mar	1.44	5	10-Aug	1.22	5
6-Feb	1.5	6	24-Mar	2.7	10	11-Aug	1.30	6
8-Feb	0.9	3	25-Mar	2.08	8	12-Aug	2.26	11
9-Feb	0.82	3	26-Mar	0.68	3	13-Aug	1.70	7
10-Feb	0.56	2	28-Mar	0.68	3	14-Aug	3.50	16
11-Feb	2.04	8	29-Mar	0.98	4	15-Aug	2.34	11
12-Feb	1.50	6	30-Mar	1.2	5	16-Aug	2.34	10
13-Feb	1.40	5	1-Jul	2.6	10	17-Aug	1.70	7
14-Feb	1.08	4	2-Jul	2.3	9	18-Aug	1.62	7
15-Feb	1.40	5	3-Jul	1.28	5	19-Aug	0.82	4

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Table L1–4B. Measured Concentrations and Calculated Quantities of Uranium at Manhole 175 in 1958 (cont'd)

1958 Date	U[a] (mg L^{-1})	Total U (kg)	1958 Date	U[a] (mg L^{-1})	Total U (kg)	1958 Date	U[a] (mg L^{-1})	Total U (kg)
20-Aug	0.66	2	5-Oct	2.58	10	21-Nov	0.47	2
21-Aug	1.14	0	6-Oct	2.4	10	22-Nov	0.47	2
22-Aug	1.30	5	7-Oct	2.02	8	23-Nov	0.56	2
23-Aug	0.98	4	8-Oct	6.06	23	24-Nov	1.03	4
24-Aug	2.10	10	9-Oct	4.04	15	25-Nov	1.6	6
25-Aug	2.04	7	10-Oct	2.4	9	26-Nov	1.5	6
26-Aug	3.34	15	11-Oct	2.58	9	27-Nov	0.66	2
27-Aug	2.04	9	12-Oct	1.48	5	28-Nov	2.44	9
28-Aug	0.74	3	13-Oct	1.48	5	29-Nov	3.01	12
29-Aug	2.80	13	14-Oct	3.86	15	30-Nov	2.07	9
30-Aug	1.48	7	15-Oct	1.84	6	1-Dec	1.29	5
31-Aug	5.20	25	16-Oct	1.1	4	2-Dec	1.97	8
1-Sep	3.54	16	17-Oct	3.12	12	3-Dec	1.97	7
2-Sep	1.30	6	18-Oct	1.54	5	4-Dec	2.44	9
3-Sep	3.16	15	19-Oct	1.1	4	6-Dec	2.54	10
4-Sep	1.86	9	20-Oct	0.92	3	7-Dec	2.16	8
5-Sep	1.68	9	21-Oct	1.5	5	8-Dec	2.63	10
7-Sep	0.94	4	23-Oct	5.35	19	10-Dec	1.22	5
8-Sep	3.34	15	24-Oct	4.28	15	11-Dec	1.03	4
9-Sep	6.24	31	25-Oct	1.93	7	12-Dec	0.94	4
10-Sep	4.46	20	26-Oct	5.78	13	13-Dec	1.41	5
11-Sep	2.80	13	28-Oct	2.16	9	14-Dec	1.5	6
12-Sep	3.54	16	29-Oct	0.93	4	15-Dec	2.2	8
13-Sep	4.10	18	30-Oct	1.13	5	16-Dec	0.83	3
14-Sep	1.30	5	31-Oct	0.93	6	17-Dec	6.44	24
14-Sep	1.3	5	1-Nov	16.48	108	18-Dec	1.47	6
15-Sep	1.3	6	2-Nov	3.07	11	19-Dec	1.38	5
16-Sep	4.46	18	3-Nov	1.34	5	20-Dec	0.64	2
17-Sep	8.32	38	4-Nov	1.34	5	21-Dec	0.92	4
18-Sep	2.98	12	5-Nov	5.15	22	22-Dec	0.39	2
19-Sep	4.24	33	6-Nov	8.24	38	23-Dec	1.66	6
20-Sep	5.52	22	7-Nov	14.42	64	24-Dec	1.84	7
21-Sep	1.86	7	8-Nov	5.36	25	25-Dec	0.92	3
22-Sep	1.48	7	9-Nov	4.94	22	26-Dec	0.67	2
23-Sep	2.76	10	10-Nov	1.88	8	27-Dec	0.58	2
24-Sep	1.3	5	11-Nov	0.94	5	28-Dec	3.68	12
25-Sep	1.68	7	12-Nov	1.13	4	29-Dec	7.91	28
26-Sep	3.21	14	13-Nov	0.85	3	30-Dec	2.21	10
27-Sep	2.58	10	14-Nov	0.75	3	31-Dec	1.1	4
28-Sep	1.48	5	15-Nov	1.79	8			
30-Sep	2.51	9	16-Nov	1.6	6	Total		2600
1-Oct	3.5	14	17-Nov	1.79	6	Average	2.3	9.4
2-Oct	1.28	6	18-Nov	6.79	33	StdDev	2.0	8.4
3-Oct	4.78	19	19-Nov	0.94	6	Max	17	69
4-Oct	2.94	11	20-Nov	0.52	3	Min	0.32	1.3

[a] From NLCO 1958.

Table L1-5A. Measured Concentrations and Calculated Quantities of Uranium at Manhole 175 in 1959

1959 Date	U[a] (mg L^{-1})	U (kg)	1959 Date	U[a] (mg L^{-1})	U (kg)	1959 Date	U[a] (mg L^{-1})	U (kg)
1-Jan	1.20	5	12-Feb	0.68	3	30-Mar	3.92	16
2-Jan	1.01	4	13-Feb	2.43	10	31-Mar	1.21	5
3-Jan	1.10	5	14-Feb	2.81	12	1-Apr	1.67	7
4-Jan	2.94	12	15-Feb	1.26	5	2-Apr	1.02	4
5-Jan	2.30	10	16-Feb	0.78	3	6-Apr	1.95	8
6-Jan	1.47	6	17-Feb	1.07	4	7-Apr	1.77	7
7-Jan	1.10	5	18-Feb	1.65	7	8-Apr	0.93	4
8-Jan	1.47	6	19-Feb	0.78	3	9-Apr	1.40	6
9-Jan	0.50	2	24-Feb	0.59	2	10-Apr	0.56	2
10-Jan	0.56	2	25-Feb	0.78	3	11-Apr	0.93	4
11-Jan	0.43	2	26-Feb	0.68	3	12-Apr	1.40	6
12-Jan	0.50	2	27-Feb	0.49	2	13-Apr	0.88	4
13-Jan	2.91	12	28-Feb	0.78	3	14-Apr	0.56	2
14-Jan	3.68	15	1-Mar	1.07	4	15-Apr	0.84	3
15-Jan	1.16	5	2-Mar	0.58	2	16-Apr	0.74	3
16-Jan	1.07	4	3-Mar	0.67	3	17-Apr	0.93	4
17-Jan	0.97	4	4-Mar	2.85	12	18-Apr	0.84	3
18-Jan	0.58	2	5-Mar	1.52	6	19-Apr	2.94	12
19-Jan	0.50	2	6-Mar	1.33	6	20-Apr	1.30	5
20-Jan	1.94	8	7-Mar	1.24	5	21-Apr	0.78	3
21-Jan	3.01	13	8-Mar	0.86	4	22-Apr	0.68	3
22-Jan	2.52	10	9-Mar	0.86	4	23-Apr	0.68	3
23-Jan	0.68	3	10-Mar	1.71	7	24-Apr	0.49	2
24-Jan	0.87	4	11-Mar	1.81	8	25-Apr	1.46	6
25-Jan	0.78	3	12-Mar	1.52	6	26-Apr	2.85	12
26-Jan	1.65	7	13-Mar	1.81	8	27-Apr	1.84	8
27-Jan	2.04	8	14-Mar	1.24	5	28-Apr	2.62	11
28-Jan	2.04	8	15-Mar	1.33	6	29-Apr	1.16	5
29-Jan	1.75	7	16-Mar	2.76	11	30-Apr	0.87	4
30-Jan	1.46	6	17-Mar	1.24	5	1-May	0.58	2
31-Jan	1.26	5	18-Mar	3.88	16	2-May	0.49	2
1-Feb	0.97	4	19-Mar	0.76	3	3-May	0.49	2
2-Feb	0.87	4	20-Mar	1.24	5	4-May	1.07	4
3-Feb	1.84	8	21-Mar	0.95	4	5-May	0.52	2
4-Feb	1.94	8	22-Mar	0.76	3	6-May	0.34	1
5-Feb	1.65	7	23-Mar	1.62	7	7-May	0.87	4
6-Feb	1.55	6	24-Mar	2.57	11	9-May	0.97	4
7-Feb	1.84	8	25-Mar	0.84	3	10-May	3.80	16
8-Feb	1.16	5	26-Mar	1.21	5	11-May	1.84	8
9-Feb	1.56	6	27-Mar	0.74	3	12-May	1.82	8
10-Feb	2.23	9	28-Mar	0.84	3	13-May	1.75	7
11-Feb	1.07	4	29-Mar	0.74	3	14-May	0.67	3

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Table L1–5B. Measured Concentrations and Calculated Quantities of Uranium at Manhole 175 in 1959 (cont'd)

1959 Date	U[a] (mg L^{-1})	U (kg)	1959 Date	U[a] (mg L^{-1})	U (kg)	1959 Date	U[a] (mg L^{-1})	U (kg)
15-May	0.95	4	27-Jun	0.97	4	8-Aug	0.86	4
16-May	0.67	3	28-Jun	2.96	12	9-Aug	1.33	6
17-May	0.53	2	29-Jun	5.94	25	10-Aug	0.48	2
18-May	1.71	7	30-Jun	0.97	4	11-Aug	1.81	8
19-May	1.14	5	1-Jul	0.78	3	12-Aug	3.92	16
20-May	1.52	6	2-Jul	0.78	3	13-Aug	1.62	7
21-May	1.05	4	3-Jul	1.84	8	14-Aug	0.54	2
22-May	13.72	57	4-Jul	0.39	2	15-Aug	1.71	7
23-May	1.05	4	5-Jul	2.23	9	16-Aug	2.00	8
24-May	0.38	2	6-Jul	3.49	15	17-Aug	3.92	16
25-May	1.05	4	7-Jul	1.07	4	18-Aug	2.02	8
26-May	1.05	4	8-Jul	0.97	4	19-Aug	1.25	5
28-May	0.95	4	9-Jul	0.87	4	20-Aug	1.15	5
29-May	1.71	7	10-Jul	0.97	4	21-Aug	1.15	5
30-May	0.76	3	11-Jul	1.65	7	22-Aug	0.67	3
31-May	0.57	2	12-Jul	1.26	5	23-Aug	0.48	2
1-Jun	0.57	2	13-Jul	1.75	7	24-Aug	2.40	10
2-Jun	2.00	8	14-Jul	1.07	4	25-Aug	1.96	8
3-Jun	0.86	4	15-Jul	1.52	6	26-Aug	1.05	4
4-Jun	2.38	10	16-Jul	0.95	4	27-Aug	2.11	9
5-Jun	0.95	4	17-Jul	1.14	5	28-Aug	1.82	8
6-Jun	0.71	3	18-Jul	3.52	15	29-Aug	1.54	6
7-Jun	0.38	2	19-Jul	2.19	9	30-Aug	1.06	4
8-Jun	0.57	2	20-Jul	1.43	6	31-Aug	0.86	4
9-Jun	0.67	3	21-Jul	0.67	3	1-Sep	3.36	14
10-Jun	1.14	5	22-Jul	0.95	4	2-Sep	2.21	9
11-Jun	2.72	11	23-Jul	3.92	16	3-Sep	1.34	6
12-Jun	1.75	7	24-Jul	2.38	10	4-Sep	0.86	4
13-Jun	1.07	4	25-Jul	0.29	1	5-Sep	0.67	3
14-Jun	0.49	2	26-Jul	0.37	2	6-Sep	1.06	4
15-Jun	3.40	14	27-Jul	1.14	5	7-Sep	0.67	3
16-Jun	0.68	3	28-Jul	2.09	9	8-Sep	1.15	5
17-Jun	0.68	3	29-Jul	1.52	6	9-Sep	2.50	10
18-Jun	0.58	2	30-Jul	0.56	2	10-Sep	2.98	12
19-Jun	0.39	2	31-Jul	0.86	4	11-Sep	0.86	4
20-Jun	0.68	3	1-Aug	1.62	7	12-Sep	1.25	5
21-Jun	1.55	6	2-Aug	0.86	4	13-Sep	1.06	4
22-Jun	0.87	4	3-Aug	0.67	3	14-Sep	1.73	7
23-Jun	2.04	8	4-Aug	2.94	12	15-Sep	0.86	4
24-Jun	1.26	5	5-Aug	1.62	7	16-Sep	3.26	14
25-Jun	2.97	12	6-Aug	1.14	5	17-Sep	1.92	8
26-Jun	2.43	10	7-Aug	1.14	5	18-Sep	0.96	4

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Table L1-5C. Measured Concentrations and Calculated Quantities of Uranium at Manhole 175 in 1959 (cont'd)

1959 Date	U[a] (mg L^{-1})	U (kg)	1959 Date	U[a] (mg L^{-1})	U (kg)	1959 Date	U[a] (mg L^{-1})	U (kg)
19-Sep	0.48	2	31-Oct	3.03	13	12-Dec	3.23	13
20-Sep	0.37	2	1-Nov	6.98	29	13-Dec	1.81	8
21-Sep	0.76	3	2-Nov	2.77	12	14-Dec	1.94	8
22-Sep	1.62	7	3-Nov	2.90	12	15-Dec	1.55	6
23-Sep	0.57	2	4-Nov	3.83	16	16-Dec	1.63	7
24-Sep	0.67	3	5-Nov	2.77	12	17-Dec	1.48	6
25-Sep	1.62	7	6-Nov	2.51	10	18-Dec	2.96	12
26-Sep	1.52	6	7-Nov	8.37	35	19-Dec	2.07	9
27-Sep	5.70	24	8-Nov	1.98	8	20-Dec	1.78	7
28-Sep	1.62	7	9-Nov	11.16	46	21-Dec	1.78	7
29-Sep	0.95	4	10-Nov	2.51	10	22-Dec	2.07	9
30-Sep	0.95	4	11-Nov	2.90	12	23-Dec	1.48	6
1-Oct	1.43	6	12-Nov	3.43	14	24-Dec	5.33	22
2-Oct	1.24	5	13-Nov	4.62	19	25-Dec	2.96	12
3-Oct	2.66	11	14-Nov	3.96	16	26-Dec	6.66	28
4-Oct	3.14	13	15-Nov	1.85	8	27-Dec	6.07	25
5-Oct	1.52	6	16-Nov	1.98	8	28-Dec	3.55	15
6-Oct	2.28	9	17-Nov	1.58	7	29-Dec	2.52	10
7-Oct	1.52	6	18-Nov	1.40	6	30-Dec	2.66	11
8-Oct	2.09	9	19-Nov	3.32	14	31-Dec	1.78	7
9-Oct	0.76	3	20-Nov	2.21	9			
10-Oct	0.95	4	21-Nov	43.20	180	Ann Total		2800
11-Oct	2.09	9	22-Nov	3.47	14	Average	1.9	8
12-Oct	1.33	6	23-Nov	6.40	27	StdDev	2.6	11
13-Oct	2.38	10	24-Nov	2.68	11	Max	43	180
14-Oct	1.71	7	25-Nov	1.42	6	Min	0.29	1

[a] From NLCO 1959; original analytical data sheets from the Bioassay Department at FMPC.

Table L1–6A. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1960

	January 1960			February 1960		
Date	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)
1	1.63	1180400	7	0.95	1090600	4
2	2.37	1180400	11	1.5	1030750	6
3	2.52	1180400	11	1.2	1030750	5
4	0.89	1180400	4	2.54	1047375	10
5	11.83	1180400	46	2.83	984200	11
6	1.63	1180400	7	3.64	1083950	15
7	1.92	1180400	9	1.6	1010800	6
8	1.78	1180400	8	1.75	1044050	7
9	1.63	1180400	7	9.9	1067325	40
10	2.81	1180400	13	3.65	1825000	25
11	2.97	1180400	13	1.95	1057350	8
12	2.97	1180400	13	3.1	1014125	12
13	2.97	1180400	13	2.25	1044050	9
14	1.8	1180400	8	1.85	1040725	7
15	2.01	1180400	9	1.75	1093925	7
16	1.17	1180400	5	2.55	1024125	10
17	0.42	1180400	2	2.6	1056325	10
18	2.33	1180400	10	13.4	1052450	53
19	0.95	1180400	4	2.3	1062765	9
20	1	1180400	4	2.3	1120525	10
21	1	1180400	4	2.4	1047375	10
22	1.8	1180400	8	2.15	1049880	9
23	1	1180400	4	2.5	1050700	10
24	1.35	1180400	6	1.75	1117200	7
25	1.75	964250	6	2.8	1250203	13
26	2.8	874499	9	1.95	1250203	9
27	3.3	1070660	13	1.65	1250203	8
28	2.8	1024100	11	1.75	1250203	8
29	1.7	970900	6	1.25	1250203	6
30	1.85	877800	6			
31	1.25	1050700	5			
Totals	68.2	35162509	286	81.8	32297335	343
StdDev		1200000	21		780000	23
Average	2.2	1134274	9	2.8	1113701	12
Max	11.8	1180400	46	13.4	1825000	53
Min	0.4	874499	2	1	984200	4

[a] From NLCO 1960–1962; 24 hour composite samples.

[b] From Cuthbert 1960–1962 and Fischoff 1960–1962; a monthly average value was used when daily measurments were not located.

Table L1-6B. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1960

Date	March 1960 Uranium[a] (mg L^{-1})	March 1960 Volume[b] (gal)	March 1960 Uranium (kg)	April 1960 Uranium[a] (mg L^{-1})	April 1960 Volume[b] (gal)	April 1960 Uranium (kg)
1	1.45	1028500	6	4.4	997500	17
2	1.6	1028500	6	2.8	1010800	11
3	1.9	1028500	7	6.35	1050700	25
4	2.5	1028500	10	3.9	1123850	17
5	1.45	1028500	6	3.5	1037400	14
6	0.9	1028500	3	2.3	984200	9
7	1.2	1028500	5	3.5	950925	13
8	1.35	1028500	5	2.15	937650	8
9	2.2	1028500	9	1.8	931000	6
10	2.95	1028500	11	2.65	970900	10
11	2.46	1028500	10	29.5	931000	104
12	2.6	1028500	10	4.5	990850	17
13	1.95	1028500	8	4.5	964250	16
14	1.65	1028500	6	2.5	927675	9
15	1.25	1028500	5	4.5	940975	16
16	3.9	1028500	15	8.5	924350	30
17	5.45	1028500	21	4.5	997500	17
18	2.85	1028500	11	8	924350	28
19	2.4	1028500	9	3.5	970900	13
20	1.85	1028500	7	3.65	931000	13
21	2.35	1028500	9	5.5	944300	20
22	1.65	1028500	6	5.5	944300	20
23	1.75	1028500	7	5	1024100	19
24	1.4	1028500	5	3.5	957600	13
25	2.3	860520	7	4	984200	15
26	4.9	1049880	19	5.5	824450	17
27	2.15	1030560	8	2.9	856650	9
28	5.9	936520	21	2.9	924350	10
29	5.4	1010800	21	2.2	933950	8
30	3	997500	11	7	908150	24
31	2.55	931000	9			
Totals	77.2	31500780	295	151	28799825	544
StdDev		1040000	18		527000	37
Average	2.5	1016154	10	5	959994	18
Max	5.9	1049880	21	29.5	1123850	104
Min	0.9	860520	3	1.8	824450	6

[a] From NLCO 1960–1962; 24 hour composite samples
[b] From Cuthbert 1960–1962 and Fischoff 1960–1962; a monthly average was used when daily measurments were not located.

Table L1–6C. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1960

Date	May 1960			June 1960		
	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)
1	3.1	914625	11	4.5	1056400	18
2	4.5	995125	17	5	1056400	20
3	4.5	1017450	17	3.5	1056400	14
4	2.75	964250	10	4.5	1056400	18
5	2.75	984200	10	3.5	1056400	14
6	2.4	950950	9	3.5	1056400	14
7	7.5	844550	24	3	1056400	12
8	5.5	957600	20	5.5	1056400	22
9	4.1	944300	15	3.5	1056400	14
10	7	1000825	26	1.95	1056400	8
11	3.05	997500	12	3.5	1056400	14
12	6	1057350	24	5.5	1056400	22
13	4.5	977550	17	8	1056400	32
14	3	970900	11	9	1056400	36
15	20.5	957600	74	7.5	1056400	30
16	9	897750	31	9.5	1056400	38
17	4	934325	14	8.5	1056400	34
18	3	917700	10	2.9	1056400	12
19	3.2	877800	11	2.3	1056400	9
20	7.5	834575	24	1.9	1056400	8
21	5.5	970900	20	4	1056400	16
22	6.5	867825	21	4.5	1056400	18
23	5	934325	18	5	1056400	20
24	9.5	960925	35	5.5	1056400	22
25	3.5	1056400	14	2.5	950950	9
26	7.5	1056400	30	4	950950	14
27	6	1056400	24	2.4	894425	8
28	5.5	1056400	22	4.2	981550	16
29	5.5	1056400	22	2.8	980875	10
30	5.5	1056400	22	2.05	964250	7
31	4	1056400	16			
Totals	172	30125700	629	134	31076600	528
StdDev		1430800	37		1060000	34
Average	5.5	971797	20	4.5	953833	11
Max	20.5	1057350	74	9.5	1056400	38
Min	2.4	834575	9	1.9	894425	7

[a] From NLCO 1960–1962; 24 hour composite samples.

[b] From Cuthbert 1960–1962 and Fischoff 1960–1962; a monthly average was used when daily measurments were not located.

Table L1-6D. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1960

	July 1960			August 1960		
Date	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)
1	6	984200	22	1.65	778050	5
2	2.7	924350	9	2.25	831250	7
3	1.45	1054025	6	2.5	861175	8
4	2.15	994175	8	9	927675	32
5	0.9	977550	3	2.1	914375	7
6	1.7	950700	6	2.3	947625	8
7	1.45	815400	4	4.5	1007475	17
8	1.12	946800	4	1.95	950950	7
9	1.2	921100	4	2.7	818000	8
10	3	950700	11	4.5	927500	16
11	1.4	899200	5	6	940975	21
12	1.9	884450	6	1.8	901740	6
13	1.75	917700	6	2.15	914622	7
14	2.65	954300	10	2	908181	7
15	2.6	917700	9	1.15	914622	4
16	2	937650	7	3	917700	10
17	0.9	911050	3	1.7	917700	6
18	3.18	957600	12	2.3	897750	8
19	2.6	907725	9	6.5	944300	23
20	2.95	917750	10	2.65	914375	9
21	2.1	884450	7	8.5	899164	29
22	6.5	864500	21	4.5	1020775	17
23	4.5	851200	14	5.5	837900	17
24	4.5	837900	14	12.5	904400	43
25	5	891000	17	4.5	1055500	18
26	7	891000	24	7.5	1138900	32
27	4	864500	13	4	913800	14
28	4.5	791350	13	5.5	929900	19
29	6	864500	20	3.5	1044700	14
30	8	864500	26	5.5	1072200	22
31	3	854500	10	5.5	1080000	22
Totals	98.7	28183525	334	129.7	29033279	466
StdDev		1254400	19		1019200	27
Average	3.2	909146	11	4.2	936557	15
Max	8	1054025	26	12.5	1138900	43
Min	1	791350	3	1.2	778050	4

[a] NLCO 1960–1962; 24 hour composite samples
[b] From Cuthbert 1960–1962 and Fischoff 1960–1962; a monthly average was used when daily measurments were not located.

Table L1–6E. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1960

	September 1960			October 1960		
Date	Uranium[a] (mg L⁻¹)	Volume[b] (gal)	Uranium (kg)	Uranium[a] (mg L⁻¹)	Volume[b] (gal)	Uranium (kg)
1	5.5	1058000	22	16.5	1128500	70
2	4	1135000	17	7.5	971400	28
3	1.05	1004000	4	5.5	1128500	23
4	1.8	1018000	7	4.5	1242800	21
5	0.9	942000	3	6	1189200	27
6	1.3	1121000	6	2.05	1335600	10
7	1.45	1058000	6	1.5	1307100	7
8	1.35	1019660	5	8	1478500	45
9	1.5	1021490	6	3	1221400	14
10	6.5	821900	20	2.4	1114200	10
11	2.05	861690	7	4	1282100	19
12	3.5	867630	11	1.95	1128500	8
13	2.4	907500	8	1.45	1228400	7
14	1.5	823400	5	1.55	1349900	8
15	1.8	857400	6	5.5	1314200	27
16	1.5	876200	5	2.05	1264200	10
17	6.5	877200	22	4	1299900	20
18	3.5	770000	10	1.5	1399900	8
19	4	1008800	15	6	1518500	34
20	4.5	1077300	18	3.5	1461350	19
21	6.5	906000	22	2.15	1364200	11
22	5.5	1037200	22	2.2	1278500	11
23	10	1257100	48	2.2	1211400	10
24	1.75	1010600	7	2.65	1354200	14
25	2.05	1017800	8	1	1532100	6
26	8	1210650	37	2.6	1439200	14
27	2.1	1189200	9	1.3	1424900	7
28	2	1132100	9	1.9	1449900	10
29	1.3	1199900	6	1.35	1428500	7
30	2.4	1228600	11	1.6	1349900	8
31				2.7	1492800	15
Totals	98.2	30315320	381	110.1	40689750	530
StdDev		1097600	24		1430800	35
Average	3.3	1010511	13	3.6	1312573	17
Max	10	1257100	48	16.5	1532100	70
Min	0.9	770000	3	1	971400	6

[a] NLCO 1960–1962; 24 hour composite samples

[b] From Cuthbert 1960–1962 and Fischoff 1960–1962; a monthly average was used when daily measurments were not located.

Table L1-6F. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1960

	November 1960			December 1960		
Date	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)
1	4.5	1514200	26	13.5	1178500	60
2	1.5	1628500	9	6	1314200	30
3	2.5	1471400	14	4.25	1149900	18
4	1.7	1428500	9	4	1150000	17
5	1.7	1171400	8	4.5	1178500	20
6	1.25	1214200	6	8	1207100	37
7	4.5	1214200	21	2.6	1357100	13
8	2.35	1263500	11	1.85	1342800	9
9	8	1177100	36	4.5	1378500	23
10	4.5	1429750	24	2.5	1371360	13
11	2.25	1197250	10	5	1542780	29
12	4	1173700	18	2	1271400	10
13	3.5	1332100	18	3.2	1474900	18
14	1.85	1324900	9	4.5	1343900	23
15	1.7	1371360	9	6.5	1549900	38
16	5	1260650	24	5.5	1460600	30
17	4	1182100	18	5	1482800	28
18	1.6	1249900	8	3.15	1342800	16
19	4.5	1171370	20	4.5	1442800	25
20	2.75	1092800	11	1.65	1557100	10
21	2.5	1121400	11	4	1507100	23
22	7	1314200	35	3.1	1535600	18
23	15.5	1207100	71	5	1535600	29
24	5.5	999950	21	2.5	1471400	14
25	2.45	1364200	13	3.15	1517800	18
26	2	1213700	9	7	1478500	39
27	1.85	1242800	9	3.2	1292800	16
28	6	1342800	30	11	1107100	46
29	4	1199900	18	4	1214200	18
30	4.5	1200000	20	3.35	1114200	14
31				3.55	1309900	18
Totals	115	38074930	544	143	42181140	720
StdDev		1391600	33		1489600	39
Average	3.8	1269164	18	4.6	1360682	23
Max	15.5	1628500	71	13.5	1557100	60
Min	1.3	999950	6	1.7	1107100	9

[a] NLCO 1960–1962; 24 hour composite samples
[b] From Cuthbert 1960–1962 and Fischoff 1960–1962; a monthly average was used when daily measurments were not located.

Table L1–7A. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1961

Date	January 1961			February 1961		
	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)
1	4.5	1192800	20	4.5	1542800	26
2	5.5	1359900	28	2.2	1521400	13
3	5.5	1499900	31	1.55	1464200	9
4	3.45	1342800	18	2.15	1407100	11
5	6.5	1378500	34	1.85	1592800	11
6	3.3	1399900	17	3	1621300	18
7	3.75	1642775	23	2.6	1571300	15
8	3.95	1499900	22	4	1635600	25
9	6.5	1564200	38	5	1671300	32
10	4.5	1589200	27	6.5	1671300	41
11	2.7	1485600	15	5.5	1678500	35
12	3.5	1507100	20	8	1671300	51
13	2.5	1499900	14	11.5	1492800	65
14	4	1507500	23	8	900000	27
15	5	1200700	23	5.5	1578500	33
16	3.15	1578500	19	4	1642800	25
17	2.25	1478500	13	5.5	1585600	33
18	4.5	1478500	25	6.5	1385600	34
19	2.2	1499900	12	2.45	1261400	12
20	1.55	1642775	10	2.2	1242800	10
21	1.7	1557100	10	2.15	1367800	11
22	2.1	1521350	12	6.5	1289200	32
23	3.5	1542800	20	2.75	1428500	15
24	2.05	1556100	12	2.45	1383200	13
25	1.7	1692800	11	6	1549900	35
26	3.5	1671300	22	6.5	1564200	38
27	2.05	1692800	13	4.5	1549900	26
28	3.5	1664200	22	5	1635600	31
29	5	1657100	31			
30	3.5	1514200	20			
31	3.5	1628500	22			
Totals	110	47047100	627	130	41906700	727
Average	3.6	1517648	20	4.6	1500000	26
Stdev	1.3	125004	7	2.4	171000	42
Max	6.5	1692800	38	11	1680000	65
Min	1.6	1192800	10	1.6	900000	9

[a] NLCO 1960–1962; 24 hour composite samples
[b] Cuthbert 1960–1962 and Fischoff 1960–1962

Table L1-7B. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1961

Date	March 1961			April 1961		
	Uranium[a] (mg L⁻¹)	Volume[b] (gal)	Uranium (kg)	Uranium[a] (mg L⁻¹)	Volume[b] (gal)	Uranium (kg)
1	2.4	1649900	15	3.1	1307100	15
2	4.5	1635600	28	4.5	1321400	22
3	2.45	1564200	14	5.5	1464200	30
4	4.5	1485600	25	4	1571300	24
5	4	1421400	21	3.2	1374900	17
6	6.5	1324900	33	2.8	1367800	14
7	6	1267100	29	2.15	1421400	12
8	5.5	1432800	30	2.75	1339200	14
9	2.05	1521400	12	6.5	1507100	37
10	1.6	1499900	9	7.5	1414200	40
11	4	1428500	22	4	1514200	23
12	2.6	1442800	14	6	1567100	36
13	3.5	1438200	19	10.5	1557100	62
14	2.6	1410600	14	4.5	1614200	27
15	2.45	1428500	13	7.5	1514400	43
16	3.5	1428500	19	7	1364200	36
17	5.5	1457100	30	8.5	1485600	48
18	4.5	1442800	25	4.5	1546500	26
19	5.5	1307100	27	5	1614200	31
20	4.5	1257100	21	5	1585600	30
21	6.5	1485600	37	5	1564200	30
22	5	1571700	30	6.5	1439200	35
23	5.5	1571400	33	6	1617800	37
24	5	1514200	29	5.5	1714200	36
25	4.5	1557100	26	7.5	1682100	48
26	4.5	1557100	26	7.5	1514200	43
27	5.5	1621300	34	8.5	1524900	49
28	5.5	1635600	34	7.5	1471400	42
29	3.6	1599900	22	13	1499900	74
30	2.3	1435600	12	6.5	1571300	39
31	4.5	1507000	26			
Totals	130	45900500	728	180	4.50E+07	1018
Average	4.2	1480000	23	5.9	1501697	75
Stdev	1.4	103000	39	2.3	102926	55
Max	6.5	1650000	37	13	1714200	163
Min	1.6	1260000	9	2.15	1307100	25

[a] NLCO 1960–1962; 24 hour composite samples
[b] Cuthbert 1960–1962 and Fischoff 1960–1962

Table L1–7C. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1961

Date	May 1961 Uranium[a] (mg L^{-1})	May 1961 Volume[b] (gal)	May 1961 Uranium (kg)	June 1961 Uranium[a] (mg L^{-1})	June 1961 Volume[b] (gal)	June 1961 Uranium (kg)
1	6.5	1514200	37	4.5	1057100	18
2	5	1542800	29	4.5	1157100	20
3	5.5	1635600	34	4.5	1085700	18
4	6	1471400	33	2.1	1028500	8
5	6.5	1592800	39	5.5	1142800	24
6	7.5	1528500	43	4.5	999950	17
7	7.5	1549900	44	4.5	1185500	20
8	9	1518500	52	5.5	1278600	27
9	6.5	1564200	38	6.5	1230750	30
10	4.5	1692800	29	4.5	1053350	18
11	3.5	1642800	22	4.5	1049300	18
12	3.5	1485000	20	2.8	1029600	11
13	4.5	1628500	28	3.5	1024100	14
14	4	1471400	22	6	1321100	30
15	5.5	1492800	31	5.5	1098000	23
16	5.5	1499900	31	2.8	1678500	18
17	4.5	1289200	22	2.3	1349900	12
18	7.5	1292800	37	3.5	1671300	22
19	4	1310600	20	4.5	1135700	19
20	4	1499400	23	8	1449900	44
21	5.5	1185650	25	7.5	1528500	43
22	6.5	1142800	28	4.5	1521300	26
23	2.5	1189200	11	5.5	1399900	29
24	5.5	1178500	25	2.65	1528500	15
25	5	1174900	22	4.15	1460600	23
26	5	1139200	22	4.5	1560600	27
27	2.5	1244200	12	3.5	1571400	21
28	3.5	1162400	15	4.5	1542800	26
29	3.65	1228500	17	2.5	1485600	14
30	3.5	1171400	15	1.8	1476500	10
31	4.5	1128500	19			
Totals	160	43168350	845	131.1	39102450	645
Average	5.1	1392527	27	4.4	1303415	22
Stdev	1.5	183101	40	1.5	218659	35
Max	9	1692800	52	8	1678500	44
Min	2.5	1128500	11	1.8	999950	8

[a] NLCO 1960–1962; 24 hour composite samples
[b] Cuthbert 1960–1962 and Fischoff 1960–1962

Table L1-7D. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1961

Date	July 1961 Uranium[a] (mg L⁻¹)	July 1961 Volume[b] (gal)	July 1961 Uranium (kg)	August 1961 Uranium[a] (mg L⁻¹)	August 1961 Volume[b] (gal)	August 1961 Uranium (kg)
1	3.3	1328500	17	2.25	1571400	13
2	2.3	1421400	12	5.5	1685600	35
3	2.35	1371400	12	3	1678500	19
4	7	1414200	37	7.5	1699900	48
5	3.7	1521400	21	14	1664200	88
6	4.5	1407100	24	21.5	1699900	138
7	2.4	1617100	15	25	1685600	159
8	2.1	1582800	13	6	1692800	38
9	2.55	1642800	16	5	1685600	32
10	1.8	1578500	11	2.45	1642800	15
11	1.7	1417800	9	4.5	1657100	28
12	5.5	1592800	33	2.85	1549900	17
13	4.5	1621300	28	1.1	1457100	6
14	1	1257100	5	1.7	1607100	10
15	3.5	1221400	16	1.4	1564200	8
16	3.5	1602100	21	1.75	1578500	10
17	1.6	1407100	9	1.45	1635600	9
18	1.45	1632100	9	4.5	1539200	26
19	4	1642900	25	1.45	1614200	9
20	1.8	1630600	11	1.75	1599900	11
21	3.5	1649900	22	2.25	1607100	14
22	2.9	1528500	17	4	1692800	26
23	4.5	1653500	28	9.5	1657100	60
24	1.5	1621300	9	5.5	1635600	34
25	1.85	1599900	11	4.5	983160	17
26	1.3	1564200	8	1.75	983160	7
27	0.9	1578500	5	1.8	983160	7
28	2.1	1599900	13	1.55	983160	6
29	1.45	1664200	9	2.15	983160	8
30	6.5	1628500	40	4.5	983160	17
31	4.5	1642800	28	3.5	983160	13
Totals	90	47641600	534	155.7	45983820	928
Average	3	1536826	17	5	1483349	70
Stdev	1.6	123133	30	5.5	275414	36
Max	7	1664200	40	25	1699900	159
Min	0.9	1221400	5	1.1	983160	6

[a] NLCO 1960–1962; 24 hour composite samples
[b] Cuthbert 1960–1962 and Fischoff 1960–1962

Table L1-7E. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1961

	September 1961			October 1961		
Date	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)
1	7	983160	26	11.5	746000	32
2	4.5	983160	17	3.8	746000	11
3	2.65	983160	10	2.1	746000	6
4	2.45	983160	9	2.15	746000	6
5	2.5	983160	9	1.75	746000	5
6	3.5	983160	13	6.5	746000	18
7	1.7	983160	6	1.8	746000	5
8	1.8	983160	7	4	746000	11
9	1.75	983160	7	2.1	746000	6
10	1.5	983160	6	1.9	746000	5
11	3	983160	11	1.55	746000	4
12	5	983160	19	1.65	746000	5
13	10	983160	37	1.35	746000	4
14	6	983160	22	1.05	746000	3
15	5.5	983160	20	1.55	746000	4
16	5	983160	19	0.95	746000	3
17	5.5	983160	20	1.15	746000	3
18	4.5	983160	17	1.25	746000	4
19	3.65	983160	14	4.5	746000	13
20	6.5	983160	24	1.7	746000	5
21	5.5	983160	20	1.1	746000	3
22	5.5	983160	20	1.5	746000	4
23	6.5	983160	24	1.4	746000	4
24	7.5	983160	28	1.4	746000	4
25	4.5	746225	13	3.1	746000	9
26	4.5	746225	13	1.1	942200	4
27	4.5	746225	13	1.6	942200	6
28	4.5	746225	13	1.4	942200	5
29	4.5	746225	13	0.6	942200	2
30	4.5	746225	13	1.7	942200	6
31				1.5	942200	5
Totals		28073190	481		24303200	205
Stddev		1030000	31		880000	16
Average	4.5		16	2.3		7
Max	10		37	11.5		32
Min	1.5		6	0.6		2

[a] NLCO 1960–1962; 24 hour composite samples

[b] From Cuthbert 1960–1962 and Fischoff 1960–1962; a monthly average was used when daily measurments were not located.

Table L1-7F. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1961

Date	November 1961 Uranium[a] (mg L⁻¹)	Volume[b] (gal)	Uranium (kg)	December 1961 Uranium[a] (mg L⁻¹)	Volume[b] (gal)	Uranium (kg)
1	1.75	942200	6	1.85	944300	7
2	1.6	942200	6	2.5	944300	9
3	7.5	942200	27	1.55	944300	6
4	2.45	942200	9	2.8	944300	10
5	0.95	942200	3	6	944300	21
6	1.55	942200	6	2.95	944300	11
7	1	942200	4	1.95	944300	7
8	1.2	942200	4	2.15	944300	8
9	2.12	942200	8	2.6	944300	9
10	7	942200	25	8	944300	29
11	1.65	942200	6	3.05	944300	11
12	2.15	942200	8	2.2	944300	8
13	3.1	942200	11	1.35	944300	5
14	10	942200	36	2.2	944300	8
15	2.4	942200	9	1.2	944300	4
16	7.5	942200	27	2.3	944300	8
17	2.5	942200	9	3	944300	11
18	1.1	942200	4	5.5	944300	20
19	1.9	942200	7	5.5	944300	20
20	2.5	942200	9	1.95	944300	7
21	2.75	942200	10	1.15	944300	4
22	2.15	942200	8	1.4	944300	5
23	6	942200	21	3.55	944300	13
24	3	942200	11	1.25	944300	4
25	1.4	942200	5	1.45	944300	5
26	2.85	944300	10	2	1056250	8
27	1.6	944300	6	2.55	1056250	10
28	1.45	944300	5	1.8	1056250	7
29	1.3	944300	5	2.25	1056250	9
30	1.75	944300	6	1.7	1056250	7
31				2.05	1056250	8
Totals		28276500	307		29945000	297
StdDev		1030000	23		1100000	20
Average	2.9		10	2.6		10
Max	10		36	8		29
Min	1		3	1.2		4

[a] NLCO 1960–1962; 24 hour composite samples
[b] From Cuthbert 1960–1962 and Fischoff 1960–1962; a monthly average was used when daily measurments were not located.

Table L1–8A. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1962

	January 1962			February 1962		
Date	Uranium[a] (mg L⁻¹)	Volume[b] (gal)	Uranium (kg)	Uranium[a] (mg L⁻¹)	Volume[b] (gal)	Uranium (kg)
1	9.00	1090000	37	3.05	1144000	13
2	1.20	1090000	5	8.50	1144000	37
3	1.75	1090000	7	6.50	1144000	28
4	6.00	1090000	25	2.70	1144000	12
5	7.00	1090000	29	4.50	1144000	19
6	5.50	1090000	23	2.80	1144000	12
7	3.50	1090000	14	6.50	1144000	28
8	1.60	1090000	7	1.60	1144000	7
9	1.85	1090000	8	6.50	1144000	28
10	2.15	1090000	9	1.75	1144000	8
11	1.30	1090000	5	1.30	1144000	6
12	1.60	1090000	7	2.25	1144000	10
13	2.55	1090000	11	2.25	1144000	10
14	3.35	1090000	14	2.35	1144000	10
15	4.50	1090000	19	8.00	1144000	35
16	1.60	1090000	7	13.50	1144000	58
17	2.05	1090000	8	2.10	1144000	9
18	2.30	1090000	9	1.90	1144000	8
19	1.65	1090000	7	6.50	1144000	28
20	2.40	1090000	10	4.50	1144000	19
21	4.50	1090000	19	5.50	1144000	24
22	3.60	1090000	15	5.00	1144000	22
23	1.55	1090000	6	4.50	1144000	19
24	1.60	1090000	7	1.70	1144000	7
25	1.55	1090000	6	4.00	1144000	17
26	6.50	1259860	31	6.00	1144000	26
27	2.45	1144000	11	4.00	1058600	16
28	1.35	1144000	6	4.50	1058600	18
29	4.50	1144000	19			
30	20.50	1144000	89			
31	3.05	1144000	13			
Totals		34229860	480		31861200	535
StdDev		1230000	40		1200000	38
Average	3.7	1104189	15	4.4	1137900	19
Max	20.5	1259860	89	13.5	1144000	58
Min	1.2	1090000	5	1.3	1058600	6

[a] NLCO 1960–1962; 24 hour composite samples

[b] From Cuthbert 1960–1962 and Fischoff 1960–1962; a monthly average was used when daily measurments were not located.

Table L1-8B. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1962

	March 1962			April 1962		
Date	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)
1	2.00	1058600	8	14.00	853528	45
2	13.50	1058600	54	9.00	853528	29
3	4.50	1058600	18	12.00	853528	39
4	4.00	1058600	16	8.50	853528	27
5	2.45	1058600	10	9.00	853528	29
6	1.60	1058600	6	6.50	853528	21
7	1.75	1058600	7	7.00	853528	23
8	1.90	1058600	8	7.00	853528	23
9	3.05	1058600	12	5.50	853528	18
10	1.30	1058600	5	5.00	853528	16
11	2.90	1058600	12	4.00	853528	13
12	1.90	1058600	8	11.00	853528	35
13	2.05	1058600	8	3.70	853528	12
14	3.15	1058600	13	3.45	853528	11
15	1.30	1058600	5	2.15	853528	7
16	1.55	1058600	6	2.90	853528	9
17	1.60	1058600	6	5.50	853528	18
18	1.70	1058600	7	11.00	853528	35
19	2.30	1058600	9	4.50	853528	15
20	5.00	1058600	20	2.20	853528	7
21	7.00	1058600	28	5.50	853528	18
22	5.00	1058600	20	7.50	853528	24
23	3.10	1058600	12	4.00	853528	13
24	4.50	1058600	18	2.05	853528	7
25	1.55	1058600	6	2.20	853528	7
26	1.95	1058600	8	1.50	853528	5
27	2.55	853528	8	8.00	754300	23
28	4.50	853528	15	5.00	754300	14
29	5.00	853528	16	4.00	754300	11
30	6.50	853528	21	6.50	754300	19
31	5.00	853528	16			
Totals		31791240	407		2520000	572
Stdev			30			38
Average	3.4		13	6.0		19
Max	13.5		54	14.0		45
Min	1.3		5	1.5		5

[a] NLCO 1960–1962; 24 hour composite samples
[b] From Cuthbert 1960–1962 and Fischoff 1960–1962; a monthly average was used when daily measurments were not located.

Table L1–8C. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1962

Date	May 1962			June 1962		
	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)
1	6.00	754300	17	2.05	959400	7
2	9.00	754300	26	2.25	959400	8
3	2.80	754300	8	1.45	959400	5
4	5.50	754300	16	1.90	959400	7
5	2.55	754300	7	4.50	959400	16
6	6.00	754300	17	1.50	959400	5
7	6.50	754300	19	1.85	959400	7
8	5.50	754300	16	2.55	959400	9
9	1.95	754300	6	6.50	959400	24
10	6.00	754300	17	5.00	959400	18
11	2.50	754300	7	5.00	959400	18
12	3.75	754300	11	1.75	959400	6
13	3.95	754300	11	1.45	959400	5
14	3.50	754300	10	1.45	959400	5
15	2.50	754300	7	2.25	959400	8
16	4.50	754300	13	5.00	959400	18
17	6.00	754300	17	4.00	959400	15
18	3.50	754300	10	5.50	959400	20
19	2.25	754300	6	10.50	959400	38
20	7.00	754300	20	1.65	959400	6
21	5.50	754300	16	2.85	959400	10
22	6.50	754300	19	1.75	959400	6
23	11.50	754300	33	1.15	959400	4
24	2.70	754300	8	5.00	959400	18
25	7.50	754300	21	2.45	959400	9
26	6.50	959400	24	2.25	898600	8
27	5.50	959400	20	2.15	898600	7
28	5.50	959400	20	1.95	898600	7
29	5.00	959400	18	1.50	898600	5
30	5.50	959400	20	1.00	898600	3
31	5.50	959400	20			
Totals		24613900	478		28478000	325
Average	5.1		15	3.0		11
Stdev			30			20
Max	11.5		33	10.5		38
Min	2.0		6	1.0		3

[a] NLCO 1960–1962; 24 hour composite samples

[b] From Cuthbert 1960–1962 and Fischoff 1960–1962; a monthly average was used when daily measurments were not located.

Table L1–8D. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1962

Date	July 1962			August 1962		
	Uranium[a] (mg L⁻¹)	Volume[b] (gal)	Uranium (kg)	Uranium[a] (mg L⁻¹)	Volume[b] (gal)	Uranium (kg)
1	1.35	928560	5	5.00	1040800	20
2	0.95	928560	3	5.50	1040800	22
3	8.50	928560	30	2.10	1040800	8
4	6.50	928560	23	1.75	1040800	7
5	11.50	928560	40	2.35	1040800	9
6	8.50	928560	30	6.00	1040800	24
7	1.80	928560	6	3.00	1040800	12
8	3.50	928560	12	5.00	1040800	20
9	1.15	928560	4	2.95	1040800	12
10	1.40	928560	5	1.40	1040800	6
11	2.80	928560	10	1.40	1040800	6
12	1.80	928560	6	2.00	1040800	8
13	2.70	928560	9	2.00	1040800	8
14	4.50	928560	16	1.65	1040800	6
15	5.50	928560	19	1.55	1040800	6
16	4.50	928560	16	1.30	1040800	5
17	3.45	928560	12	2.45	1040800	10
18	1.70	928560	6	1.90	1040800	7
19	1.20	928560	4	1.85	1040800	7
20	1.15	928560	4	2.15	1040800	8
21	1.90	928560	7	2.75	1040800	11
22	1.65	928560	6	5.50	1040800	22
23	2.65	928560	9	2.60	1040800	10
24	1.20	928560	4	3.10	1040800	12
25	0.85	928560	3	5.50	1040800	22
26	0.65	1040800	3	7.50	1040800	30
27	0.70	1040800	3	4.00	930400	14
28	1.10	1040800	4	2.40	930400	8
29	1.35	1040800	5	5.50	930400	19
30	1.45	1040800	6	3.50	930400	12
31	1.00	1040800	4	5.00	930400	18
Totals		29458800	315		31712800	387
Average	2.9		10	3.2		12
Stdev	2.6		24	1.7		25
Max	11.5		40	7.5		30
Min	0.7		3	1.3		5

[a] NLCO 1960–1962; 24 hour composite samples
[b] From Cuthbert 1960–1962 and Fischoff 1960–1962; a monthly average was used when daily measurments were not located.

Table L1-8E. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1962

	September 1962			October 1962		
Date	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)	Uranium[a] (mg L^{-1})	Volume[b] (gal)	Uranium (kg)
1	4.00	930400	14	2.30	736900	6
2	2.85	930400	10	6.00	736900	17
3	2.00	930400	7	6.50	736900	18
4	9.50	930400	33	2.05	736900	6
5	10.50	930400	37	1.70	736900	5
6	45.00	1100000	187	6.50	736900	18
7	8.50	1200000	39	2.55	736900	7
8	45.00	1000000	170	5.50	736900	15
9	6.50	930400	23	2.10	736900	6
10	125.00	1440000	680	7.50	736900	21
11	15.00	1130000	64	2.60	736900	7
12	5.00	930400	18	2.35	736900	7
13	5.00	930400	18	10.00	736900	28
14	4.50	930400	16	12.00	736900	33
15	5.50	930400	19	8.50	736900	24
16	3.50	930400	12	4.50	736900	13
17	2.70	930400	9	10.50	736900	29
18	4.00	930400	14	7.00	736900	19
19	2.95	930400	10	2.05	736900	6
20	4.00	930400	14	2.30	736900	6
21	2.60	930400	9	1.45	736900	4
22	2.05	930400	7	1.45	736900	4
23	1.85	930400	7	1.25	736900	3
24	1.80	930400	6	1.30	736900	4
25	5.00	930400	18	2.20	736900	6
26	3.50	930400	12	7.50	796231	23
27	2.35	736900	7	2.60	796231	8
28	4.00	736900	11	7.12	796231	21
29	1.75	736900	5	2.95	796231	9
30	1.60	736900	4	2.10	796231	6
31				3.38	796231	10
Totals		28356000	1481		23199860	390
StdDev		1050000	238		834000	28
Average	11.3		49	4.4		13
Max	125.0		680	12.0		33
Min	1.6		4	1.3		3

[a] NLCO 1960–1962; 24 hour composite samples

[b] From Cuthbert 1960–1962 and Fischoff 1960–1962; a monthly average was used when daily measurments were not located.

Table L1-8F. Uranium Quantities and Effluent Volume Measured at Manhole 175 in 1962

Date	November 1962 Uranium[a] (mg L⁻¹)	Volume[b] (gal)	Uranium (kg)	December 1962 Uranium[a] (mg L⁻¹)	Volume[b] (gal)	Uranium (kg)
1	5.62	796231	17	1.90	790030	6
2	2.85	796231	9	2.30	790030	7
3	2.75	796231	8	2.00	790030	6
4	8.00	796231	24	0.95	790030	3
5	3.25	796231	10	8.00	790030	24
6	4.00	796231	12	4.50	790030	13
7	2.35	796231	7	1.25	790030	4
8	1.70	796231	5	0.65	790030	2
9	5.00	796231	15	0.65	790030	2
10	4.50	796231	14	1.35	790030	4
11	1.90	796231	6	1.40	790030	4
12	8.50	796231	26	1.65	790030	5
13	2.45	796231	7	36.00	946500	129
14	1.70	796231	5	8.50	1093000	35
15	2.25	796231	7	7.50	1093000	31
16	2.20	796231	7	3.20	1093000	13
17	3.25	796231	10	1.00	1093000	4
18	3.55	796231	11	3.90	1093000	16
19	3.05	796231	9	3.25	1093000	13
20	12.00	796231	36	4.00	1093000	17
21	2.40	796231	7	2.65	1093000	11
22	2.25	796231	7	2.25	1093000	9
23	2.05	796231	6	1.65	1093000	7
24	2.30	796231	7	5.00	1093000	21
25	2.10	796231	6	1.50	1093000	6
26	15.50	790030	46	1.25	1093000	5
27	2.75	790030	8	4.50	1093000	19
28	6.50	790030	19	6.50	1093000	27
29	2.75	790030	8	2.30	1093000	10
30	2.15	790030	6	1.35	1093000	6
31				1.85	1093000	8
Totals		23855925	365		30100860	465
Average	4.1		12	4.0		15
Stdev		871000	28		1090000	50
Max	15.5		46	36.0		129
Min	1.7		5	1.0		1

[a] NLCO 1960–1962; 24 hour composite samples

[b] From Cuthbert 1960–1962 and Fischoff 1960–1962; a monthly average was used when daily measurments were not located.

Table L1–9. Measured Concentrations and Calculated Quantities of Uranium at Manhole 175 in 1963

1963 Date	U[a] (mg L^{-1})	Total U kg U	1963 Date	U[a] (mg L^{-1})	Total U kg U	1963 Date	U[a] (mg L^{-1})	Total U kg U
1-Apr	2.50	10	13-May	0.98	4	25-Nov	2.20	9
2-Apr	2.45	10	14-May	2.00	8	27-Nov	2.20	9
3-Apr	3.15	13	15-May	2.00	8	28-Nov	2.60	11
4-Apr	2.65	11	16-May	1.12	5	29-Nov	5.20	22
5-Apr	2.35	10	17-May	2.00	8	30-Nov	4.40	18
6-Apr	5.50	23	18-May	1.10	5	1-Dec	4.20	17
7-Apr	2.65	11	19-May	1.14	5	2-Dec	2.80	12
8-Apr	2.34	10	20-May	2.00	8	3-Dec	2.00	8
9-Apr	2.40	10	21-May	1.16	5	4-Dec	2.80	12
10-Apr	2.75	11	22-May	0.72	3	5-Dec	3.00	12
11-Apr	1.80	7	23-May	0.74	3	6-Dec	3.00	12
12-Apr	1.55	6	24-May	1.80	7	7-Dec	3.00	12
13-Apr	1.60	7	25-May	2.20	9	8-Dec	5.80	24
14-Apr	7.50	31	26-May	1.24	5	9-Dec	2.80	12
15-Apr	2.75	11	27-May	2.20	9	10-Dec	3.80	16
17-Apr	7.00	29	28-May	1.34	6	12-Dec	10.40	43
18-Apr	6.00	25	30-May	0.98	4	13-Dec	4.80	20
19-Apr	3.05	13	31-May	1.02	4	14-Dec	2.60	11
20-Apr	2.75	11	2-Nov	2.80	12	15-Dec	3.00	12
21-Apr	2.15	9	3-Nov	1.00	4	16-Dec	4.00	17
22-Apr	5.50	23	4-Nov	4.00	17	17-Dec	2.60	11
23-Apr	1.90	8	5-Nov	4.20	17	18-Dec	4.40	18
24-Apr	1.45	6	6-Nov	2.20	9	19-Dec	3.40	14
25-Apr	2.25	9	7-Nov	2.80	12	20-Dec	4.20	17
26-Apr	1.75	7	8-Nov	5.00	21	21-Dec	1.60	7
27-Apr	1.85	8	9-Nov	5.20	22	22-Dec	2.00	8
28-Apr	1.50	6	10-Nov	3.80	16	23-Dec	2.20	9
29-Apr	2.20	9	11-Nov	3.00	12	24-Dec	1.00	4
30-Apr	3.72	15	12-Nov	4.00	17	25-Dec	5.60	23
1-May	1.60	7	13-Nov	4.00	17	26-Dec	5.80	24
2-May	1.75	7	14-Nov	6.60	27	27-Dec	3.00	12
3-May	1.10	5	15-Nov	6.80	28	28-Dec	2.80	12
4-May	2.05	9	16-Nov	4.40	18	29-Dec	0.80	3
5-May	1.55	6	17-Nov	2.60	11	30-Dec	0.80	3
6-May	1.25	5	18-Nov	1.32	5	31-Dec	0.80	3
7-May	1.28	5	19-Nov	1.52	6			
8-May	0.92	4	20-Nov	4.00	17	AnnTotal		
9-May	2.20	9	21-Nov	0.72	3	Average	2.86	12
10-May	0.90	4	22-Nov	3.20	13	StdDev	1.81	8
11-May	0.68	3	23-Nov	8.20	34	Max	10.40	43
12-May	0.72	3	24-Nov	4.80	20	Min	0.68	3

[a] From NLCO 1963, analytical data sheets from the Bioassay Department at the FMPC.

Table L1–10A. Measured Concentrations and Calculated Quantities of Uranium at Manhole 175 in 1964

1964 Date	U[a] (mg L^{-1})	Total U (kg)	1964 Date	U[a] (mg L^{-1})	Total U (kg)	1964 Date	U[a] (mg L^{-1})	Total U (kg)
1-Jan	3.00	12	14-Feb	2.60	11	29-Mar	3.40	14
2-Jan	2.40	10	15-Feb	6.00	25	30-Mar	3.00	12
3-Jan	4.00	17	16-Feb	3.00	12	31-Mar	2.20	9
4-Jan	5.20	22	17-Feb	2.40	10	1-Apr	3.00	12
5-Jan	3.00	12	18-Feb	5.60	23	2-Apr	3.80	16
6-Jan	4.60	19	19-Feb	6.20	26	3-Apr	3.40	14
7-Jan	4.80	20	20-Feb	4.60	19	4-Apr	2.80	12
8-Jan	4.60	19	21-Feb	4.20	17	5-Apr	3.00	12
9-Jan	4.40	18	22-Feb	5.20	22	6-Apr	2.60	11
10-Jan	4.40	18	23-Feb	3.20	13	7-Apr	3.60	15
12-Jan	2.20	9	25-Feb	15.40	64	9-Apr	3.60	15
13-Jan	2.00	8	26-Feb	10.60	44	10-Apr	2.60	11
14-Jan	2.20	9	27-Feb	7.40	31	11-Apr	2.60	11
15-Jan	5.40	22	28-Feb	7.00	29	12-Apr	2.60	11
16-Jan	7.40	31	29-Feb	7.40	31	13-Apr	2.20	9
17-Jan	2.40	10	1-Mar	4.20	17	14-Apr	2.20	9
18-Jan	2.00	8	2-Mar	6.80	28	15-Apr	2.80	12
19-Jan	4.00	17	3-Mar	7.60	32	16-Apr	3.20	13
20-Jan	5.20	22	4-Mar	5.80	24	17-Apr	3.00	12
21-Jan	3.00	12	5-Mar	6.40	27	18-Apr	4.80	20
22-Jan	2.40	10	6-Mar	5.60	23	19-Apr	3.60	15
23-Jan	2.60	11	7-Mar	6.00	25	20-Apr	4.80	20
24-Jan	3.80	16	8-Mar	7.00	29	21-Apr	4.40	18
25-Jan	3.80	16	9-Mar	3.80	16	22-Apr	3.40	14
26-Jan	2.00	8	10-Mar	5.00	21	23-Apr	4.20	17
27-Jan	2.40	10	11-Mar	3.40	14	24-Apr	2.60	11
28-Jan	2.00	8	12-Mar	3.20	13	25-Apr	3.80	16
29-Jan	2.20	9	13-Mar	6.20	26	26-Apr	8.60	36
30-Jan	2.20	9	14-Mar	10.00	42	27-Apr	4.00	17
31-Jan	2.60	11	15-Mar	6.20	26	28-Apr	3.00	12
1-Feb	3.00	12	16-Mar	4.80	20	29-Apr	2.20	9
2-Feb	2.60	11	17-Mar	4.20	17	30-Apr	4.40	18
3-Feb	1.34	6	18-Mar	3.00	12	1-May	5.00	21
4-Feb	2.40	10	19-Mar	2.60	11	2-May	2.80	12
5-Feb	2.80	12	20-Mar	5.00	21	3-May	2.80	12
6-Feb	4.20	17	21-Mar	4.60	19	4-May	4.40	18
7-Feb	3.20	13	22-Mar	4.60	19	5-May	6.60	27
8-Feb	2.40	10	23-Mar	3.00	12	6-May	3.40	14
9-Feb	2.00	8	24-Mar	4.00	17	7-May	1.28	5
10-Feb	2.80	12	25-Mar	5.00	21	8-May	2.40	10
11-Feb	4.60	19	26-Mar	3.80	16	9-May	3.20	13
12-Feb	3.80	16	27-Mar	2.00	8	10-May	2.40	10

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Table L1–10B. Measured Concentrations and Calculated Quantities of Uranium at Manhole 175 in 1964 (cont'd)

1964 Date	U[a] (mg L^{-1})	Total U (kg)	1964 Date	U[a] (mg L^{-1})	Total U (kg)	1964 Date	U[a] (mg L^{-1})	Total U (kg)
12-May	4.20	17	25-Jun	3.00	12	8-Aug	0.96	4
13-May	6.60	27	26-Jun	1.42	6	9-Aug	2.20	9
14-May	3.60	15	27-Jun	4.40	18	10-Aug	1.36	6
15-May	4.60	19	28-Jun	1.46	6	11-Aug	2.00	8
16-May	2.60	11	29-Jun	1.28	5	12-Aug	2.20	9
17-May	1.20	5	30-Jun	5.80	24	13-Aug	2.20	9
18-May	4.20	17	1-Jul	2.20	9	14-Aug	1.10	5
19-May	4.00	17	2-Jul	2.40	10	15-Aug	0.80	3
20-May	5.60	23	3-Jul	1.28	5	16-Aug	0.52	2
21-May	9.20	38	4-Jul	2.20	9	17-Aug	3.40	14
22-May	2.60	11	5-Jul	1.06	4	18-Aug	1.08	4
23-May	14.60	61	6-Jul	3.60	15	19-Aug	2.00	8
24-May	8.00	33	7-Jul	3.80	16	20-Aug	1.08	4
25-May	3.00	12	8-Jul	2.00	8	21-Aug	2.00	8
26-May	2.60	11	9-Jul	1.12	5	22-Aug	1.38	6
27-May	2.20	9	10-Jul	0.84	3	23-Aug	3.40	14
28-May	3.00	12	11-Jul	0.88	4	24-Aug	0.86	4
29-May	2.20	9	12-Jul	3.60	15	25-Aug	0.90	4
31-May	0.72	3	14-Jul	1.34	6	27-Aug	1.30	5
1-Jun	3.60	15	15-Jul	3.20	13	28-Aug	2.00	8
2-Jun	7.20	30	16-Jul	1.06	4	29-Aug	1.32	5
3-Jun	3.00	12	17-Jul	1.80	7	1-Sep	0.92	4
4-Jun	3.80	16	18-Jul	3.80	16	2-Sep	0.88	4
5-Jun	4.40	18	19-Jul	2.20	9	3-Sep	0.72	3
6-Jun	5.40	22	20-Jul	1.80	7	4-Sep	1.10	5
7-Jun	2.80	12	21-Jul	1.24	5	5-Sep	0.74	3
8-Jun	2.40	10	22-Jul	0.94	4	6-Sep	0.80	3
9-Jun	2.80	12	23-Jul	2.60	11	7-Sep	0.76	3
10-Jun	2.20	9	24-Jul	1.50	6	8-Sep	0.62	3
11-Jun	2.20	9	25-Jul	1.10	5	9-Sep	2.00	8
12-Jun	2.80	12	26-Jul	1.24	5	10-Sep	0.74	3
13-Jun	3.20	13	27-Jul	0.94	4	11-Sep	0.70	3
14-Jun	2.20	9	28-Jul	2.40	10	12-Sep	0.52	2
15-Jun	1.20	5	29-Jul	3.40	14	13-Sep	0.36	1
16-Jun	1.04	4	30-Jul	2.40	10	14-Sep	0.52	2
17-Jun	1.46	6	31-Jul	4.40	18	15-Sep	0.70	3
18-Jun	3.60	15	1-Aug	3.80	16	16-Sep	0.82	3
19-Jun	1.50	6	2-Aug	3.60	15	17-Sep	0.72	3
21-Jun	2.80	12	4-Aug	2.60	11	19-Sep	2.40	10
22-Jun	2.20	9	5-Aug	2.40	10	20-Sep	1.14	5
23-Jun	1.54	6	6-Aug	2.20	9	21-Sep	1.24	5
24-Jun	1.16	5	7-Aug	1.32	5	22-Sep	2.40	10

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Table L1-10C. Measured Concentrations and Calculated Quantities of Uranium at Manhole 175 in 1964 (cont'd)

1964 Date	U[a] (mg L⁻¹)	Total U (kg)	1964 Date	U[a] (mg L⁻¹)	Total U (kg)	1964 Date	U[a] (mg L⁻¹)	Total U (kg)
23-Sep	2.40	10	2-Nov	1.80	7	12-Dec	2.00	8
24-Sep	2.80	12	3-Nov	0.64	3	13-Dec	1.32	5
25-Sep	0.98	4	4-Nov	0.68	3	14-Dec	0.90	4
26-Sep	0.76	3	5-Nov	1.26	5	15-Dec	0.82	3
27-Sep	2.40	10	6-Nov	0.78	3	16-Dec	1.40	6
28-Sep	1.56	6	7-Nov	1.60	7	17-Dec	1.26	5
29-Sep	1.52	6	8-Nov	1.20	5	18-Dec	0.70	3
30-Sep	1.18	5	9-Nov	0.94	4	19-Dec	1.10	5
1-Oct	1.04	4	10-Nov	0.72	3	20-Dec	0.52	2
2-Oct	3.00	12	11-Nov	1.28	5	21-Dec	0.50	2
3-Oct	1.39	6	12-Nov	1.28	5	22-Dec	0.66	3
4-Oct	0.72	3	13-Nov	1.80	7	23-Dec	0.76	3
5-Oct	0.60	2	14-Nov	0.92	4	24-Dec	2.00	8
6-Oct	0.68	3	15-Nov	3.80	16	25-Dec	2.00	8
7-Oct	0.82	3	16-Nov	2.20	9	26-Dec	2.20	9
8-Oct	0.72	3	17-Nov	0.98	4	27-Dec	1.28	5
9-Oct	0.60	2	18-Nov	0.98	4	28-Dec	1.38	6
10-Oct	0.64	3	19-Nov	2.20	9	29-Dec	1.20	5
11-Oct	0.90	4	20-Nov	1.12	5	30-Dec	1.26	5
12-Oct	0.56	2	21-Nov	2.20	9	31-Dec	0.88	4
13-Oct	1.00	4	22-Nov	0.74	3			
14-Oct	1.04	4	23-Nov	1.60	7			
15-Oct	0.86	4	24-Nov	1.60	7			
16-Oct	0.58	2	25-Nov	2.40	10			
17-Oct	0.60	2	26-Nov	1.16	5			
18-Oct	2.60	11	27-Nov	0.50	2			
19-Oct	2.60	11	28-Nov	2.60	11			
20-Oct	1.23	5	29-Nov	0.82	3			
21-Oct	1.20	5	30-Nov	1.12	5			
22-Oct	2.40	10	1-Dec	0.64	3			
23-Oct	0.92	4	2-Dec	2.20	9			
24-Oct	0.52	2	3-Dec	3.00	12			
25-Oct	0.66	3	4-Dec	1.80	7			
26-Oct	1.60	7	5-Dec	1.16	5			
27-Oct	1.40	6	6-Dec	1.06	4			
28-Oct	2.60	11	7-Dec	1.50	6	Total		5100
29-Oct	4.20	17	8-Dec	1.08	4	Average	3.3	14
30-Oct	3.00	12	9-Dec	1.02	4	StdDev	1.3	8
31-Oct	1.20	5	10-Dec	1.34	6	Max	7.4	64
1-Nov	0.64	3	11-Dec	2.40	10	Min	1.3	1.5

[a] From NLCO 1964, original analytical data sheets from the Bioassay Department at the FMPC.

Table L1-11A. Uranium Concentrations Measured at Manhole 175 in 1966

1966 Date	U[a] (mg L^{-1})	1966 Date	U[a] (mg L^{-1})	1966 Date	U[a] (mg L^{-1})	1966 Date	U[a] (mg L^{-1})	1966 Date	U[a] (mg L^{-1})
1-Jan	2.4	10-Feb	3.6	22-Mar	0.98	1-May	1.36	10-Jun	1.58
2-Jan	1.46	11-Feb	3.6	23-Mar	2.2	2-May	1.18	11-Jun	1.46
3-Jan	1.02	12-Feb	4.2	24-Mar	1.34	3-May	0.84	12-Jun	0.7
4-Jan	1.1	13-Feb	4.2	25-Mar	1.12	4-May	0.66	13-Jun	2.4
5-Jan	1.12	14-Feb	22	26-Mar	1.04	5-May	0.68	14-Jun	1.72
6-Jan	1.16	15-Feb	8	27-Mar	0.72	6-May	0.82	15-Jun	1.14
7-Jan	1.42	16-Feb	3.1	28-Mar	0.82	7-May	0.78	16-Jun	2.6
8-Jan	0.98	17-Feb	3	29-Mar	1.22	8-May	0.88	17-Jun	1.28
9-Jan	0.92	18-Feb	1.26	30-Mar	2.4	9-May	2	18-Jun	2.8
10-Jan	0.82	19-Feb	1.22	31-Mar	1.6	10-May	2	19-Jun	0.74
11-Jan	0.78	20-Feb	0.8	1-Apr	1.5	11-May	2.2	20-Jun	1.06
12-Jan	1.28	21-Feb	1.14	2-Apr	0.88	12-May	5	21-Jun	1.14
13-Jan	1.08	22-Feb	1.02	3-Apr	2	13-May	2.2	22-Jun	2.2
14-Jan	0.78	23-Feb	2.6	4-Apr	1.2	14-May	1	23-Jun	1.8
15-Jan	1.08	24-Feb	1.36	5-Apr	1.08	15-May	0.88	24-Jun	1.02
16-Jan	0.68	25-Feb	2	6-Apr	0.9	16-May	1.3	25-Jun	0.76
17-Jan	2	26-Feb	0.96	7-Apr	1.24	17-May	0.82	26-Jun	0.78
18-Jan	0.94	27-Feb	1.38	8-Apr	1.8	18-May	2.2	27-Jun	2
19-Jan	1.1	28-Feb	3.8	9-Apr	1	19-May	2.4	28-Jun	2.2
20-Jan	0.48	1-Mar	2.2	10-Apr	1.28	20-May	0.96	29-Jun	1.06
21-Jan	1.42	2-Mar	1.34	11-Apr	3.8	21-May	1.2	30-Jun	3
22-Jan	2.6	3-Mar	3.2	12-Apr	4.6	22-May	3	1-Jul	1.24
23-Jan	0.9	4-Mar	2.2	13-Apr	6.4	23-May	1.38	2-Jul	0.9
24-Jan	1.08	5-Mar	1.06	14-Apr	4.4	24-May	2	3-Jul	0.6
25-Jan	0.74	6-Mar	1.14	15-Apr	2	25-May	3	4-Jul	2
26-Jan	0.9	7-Mar	0.84	16-Apr	2.2	26-May	1.24	5-Jul	1
27-Jan	0.82	8-Mar	0.66	17-Apr	0.78	27-May	1.6	6-Jul	1.62
28-Jan	2.8	9-Mar	0.98	18-Apr	2.2	28-May	2.2	7-Jul	0.8
29-Jan	0.92	10-Mar	0.76	19-Apr	2.4	29-May	1.2	8-Jul	0.94
30-Jan	0.6	11-Mar	0.8	20-Apr	16	30-May	0.76	9-Jul	0.7
31-Jan	0.66	12-Mar	4	21-Apr	3.8	31-May	2	10-Jul	0.68
1-Feb	4	13-Mar	1.28	22-Apr	2.4	1-Jun	2.2	11-Jul	0.82
2-Feb	3	14-Mar	1.06	23-Apr	1.54	2-Jun	1.04	12-Jul	0.88
3-Feb	3.2	15-Mar	1.04	24-Apr	3.6	3-Jun	1.46	13-Jul	2.4
4-Feb	3.8	16-Mar	0.88	25-Apr	2	4-Jun	0.86	14-Jul	0.96
5-Feb	1	17-Mar	1.46	26-Apr	2.8	5-Jun	0.84	15-Jul	0.58
6-Feb	2.2	18-Mar	2	27-Apr	3	6-Jun	136	16-Jul	0.76
7-Feb	3.8	19-Mar	1.6	28-Apr	5.8	7-Jun	10.2	17-Jul	0.76
8-Feb	4.6	20-Mar	1.7	29-Apr	1.32	8-Jun	3.6	18-Jul	0.52
9-Feb	3	21-Mar	2.2	30-Apr	1.2	9-Jun	2	19-Jul	0.82

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Table L1-11B. Uranium Concentrations Measured at Manhole 175 in 1966 (cont'd)

1966 Date	U[a] (mg L^{-1})	1966 Date	U[a] (mg L^{-1})	1966 Date	U[a] (mg L^{-1})	1966 Date	U[a] (mg L^{-1})
20-Jul	1.18	29-Aug	0.78	8-Nov	2	17-Dec	0.52
21-Jul	1.1	30-Aug	1	9-Nov	1.26	18-Dec	0.58
22-Jul	0.78	31-Aug	0.68	10-Nov	0.88	19-Dec	0.6
23-Jul	4.8	1-Sep	0.68	11-Nov	1.22	20-Dec	0.5
24-Jul	2	2-Sep	0.68	12-Nov	0.74	21-Dec	0.82
25-Jul	0.7	3-Sep	2.2	13-Nov	0.4	22-Dec	0.94
26-Jul	0.64	4-Sep	1.4	14-Nov	0.92	23-Dec	0.88
27-Jul	0.54	5-Sep	0.72	15-Nov	1.24	24-Dec	0.76
28-Jul	0.8	6-Sep	1.8	16-Nov	1.06	25-Dec	0.74
29-Jul	0.54	7-Sep	0.92	17-Nov	0.98	26-Dec	3.2
30-Jul	0.56	8-Sep	0.7	18-Nov	0.9	27-Dec	2.2
31-Jul	0.58	9-Sep	1.12	19-Nov	1.34	28-Dec	5.2
1-Aug	0.72	10-Sep	1.04	20-Nov	0.86	29-Dec	5.6
2-Aug	80	11-Sep	3.4	21-Nov	0.9	30-Dec	3
3-Aug	10	12-Sep	0.72	22-Nov	0.88	31-Dec	2.2
4-Aug	5.6	13-Sep	0.84	23-Nov	2.4		
5-Aug	5.2	14-Sep	1.06	24-Nov	0.86		
6-Aug	3	15-Sep	2.4	25-Nov	2.8		
7-Aug	2.8	16-Sep	1.26	26-Nov	2.6		
8-Aug	2.6	17-Sep	1.6	27-Nov	2		
9-Aug	1.48	18-Sep	1.46	28-Nov	2.4		
10-Aug	2.2	19-Sep	2.8	29-Nov	2		
11-Aug	5.8	20-Sep	2.8	30-Nov	22		
12-Aug	4.4	21-Sep	2.2	1-Dec	2.8		
13-Aug	3	22-Sep	1.04	2-Dec	1.38		
14-Aug	2.8	23-Sep	1.08	3-Dec	1.14		
15-Aug	2.4	24-Sep	1.36	4-Dec	0.78		
16-Aug	2.6	25-Sep	3.6	5-Dec	2.4		
17-Aug	0.9	26-Sep	2.2	6-Dec	0.82		
18-Aug	2.2	27-Sep	2.6	7-Dec	3.2		
19-Aug	1.36	28-Sep	2.2	8-Dec	2.4		
20-Aug	1.34	29-Sep	1.34	9-Dec	0.96		
21-Aug	2.2	30-Sep		10-Dec	0.64		
22-Aug	2.2	1-Nov	4.6	11-Dec	1.16		
23-Aug	2	2-Nov	3.6	12-Dec	0.44		
24-Aug	1.4	3-Nov	2.6	13-Dec	0.74		
25-Aug	2	4-Nov	3.2	14-Dec	0.84	Average	2.6
26-Aug	1.56	5-Nov	3.8	15-Dec	0.56	StdDev	8.8
27-Aug	0.9	6-Nov	2.2	16-Dec	0.58	Max	136
28-Aug	0.62	7-Nov	2.2	17-Dec	0.52	Min	0.4

[a] From NLCO 1966, analytical data sheets from the Bioassay Department at the FMPC.

Table L1–12A. Uranium Concentrations Measured at Manhole 175 in 1967

Date 1967	U[a] (mg L^{-1})	Date 1967	U[a] (mg L^{-1})	Date 1967	U[a] (mg L^{-1})	Date 1967	U[a] (mg L^{-1})	Date 1967	U[a] (mg L^{-1})
1-Jan	1.2	11-Feb	1.2	24-Mar	1.52	4-May	2.4	14-Jun	0.94
2-Jan	1.38	12-Feb	0.6	25-Mar	1	5-May	0.86	15-Jun	3
3-Jan	0.84	13-Feb	0.52	26-Mar	1.14	6-May	3	16-Jun	1.6
4-Jan	1.08	14-Feb	0.66	27-Mar	2.8	7-May	2.6	17-Jun	1.4
5-Jan	2.2	15-Feb	2.8	28-Mar	2.8	8-May	2.2	18-Jun	0.74
6-Jan	0.74	16-Feb	2.2	29-Mar	1.24	9-May	1.32	19-Jun	1.14
7-Jan	2.4	17-Feb	1.06	30-Mar	1.22	10-May	2	20-Jun	0.84
8-Jan	0.82	18-Feb	0.88	31-Mar	0.42	11-May	3	21-Jun	3
9-Jan	0.98	19-Feb	0.9	1-Apr	2	12-May	1.1	22-Jun	2.2
10-Jan	0.8	20-Feb	1.26	2-Apr	0.96	13-May	2	23-Jun	0.98
11-Jan	0.32	21-Feb	0.62	3-Apr	2.4	14-May	2.8	24-Jun	0.98
12-Jan	2	22-Feb	1.2	4-Apr	2.6	15-May	1.76	25-Jun	1.02
13-Jan	0.74	23-Feb	0.94	5-Apr	3	16-May	1.36	26-Jun	0.86
14-Jan	0.76	24-Feb	1.26	6-Apr	2.2	17-May	2.6	27-Jun	1.18
15-Jan	0.5	25-Feb	0.78	7-Apr	1.48	18-May	2.4	28-Jun	2.8
16-Jan	1.2	26-Feb	0.76	8-Apr	1.24	19-May	2.2	29-Jun	2.6
17-Jan	1.08	27-Feb	3.2	9-Apr	1.3	20-May	0.62	30-Jun	1.2
18-Jan	0.44	28-Feb	1.38	10-Apr	1.04	21-May	0.76	1-Jul	1.36
19-Jan	0.98	1-Mar	1.52	11-Apr	0.58	22-May	0.98	2-Jul	1.22
20-Jan	1.22	2-Mar	2.2	12-Apr	0.88	23-May	2.2	3-Jul	0.66
21-Jan	0.76	3-Mar	3.2	13-Apr	3.4	24-May	1.02	4-Jul	1.24
22-Jan	1.24	4-Mar	6.2	14-Apr	1.22	25-May	1.02	5-Jul	0.92
23-Jan	1.8	5-Mar	5.6	15-Apr	1.08	26-May	2.4	6-Jul	1.34
24-Jan	4	6-Mar	3.4	16-Apr	0.34	27-May	1.42	7-Jul	0.94
25-Jan	2.4	7-Mar	3	17-Apr	2.2	28-May	1.24	8-Jul	0.88
26-Jan	3.4	8-Mar	1.22	18-Apr	1.28	29-May	1.42	9-Jul	1.4
27-Jan	2.6	9-Mar	2	19-Apr	1.1	30-May	1.12	10-Jul	2
28-Jan	2.4	10-Mar	1	20-Apr	1.42	31-May	0.74	11-Jul	0.74
29-Jan	0.5	11-Mar	1.4	21-Apr	3	1-Jun	1.08	12-Jul	0.94
30-Jan	0.76	12-Mar	1.32	22-Apr	0.74	2-Jun	1.36	13-Jul	0.62
31-Jan	1.3	13-Mar	1.8	23-Apr	1.12	3-Jun	3	14-Jul	1
1-Feb	2.4	14-Mar	2.6	24-Apr	1.36	3-Jan	1.14	15-Jul	1.2
2-Feb	1.72	15-Mar	3.4	25-Apr	1.62	5-Jun	0.78	16-Jul	1.2
3-Feb	0.96	16-Mar	1.6	26-Apr	3.4	6-Jun	0.9	17-Jul	2.2
4-Feb	0.84	17-Mar	0.98	27-Apr	1.64	7-Jun	0.88	18-Jul	1.8
5-Feb	1.22	18-Mar	2	28-Apr	1.3	8-Jun	1.14	19-Jul	2.2
6-Feb	0.68	19-Mar	1.8	29-Apr	2.4	9-Jun	1.12	20-Jul	2.4
7-Feb	0.82	20-Mar	4	30-Apr	2.8	10-Jun	1.04	21-Jul	1.3
8-Feb	1.04	21-Mar	3.4	1-May	2.2	11-Jun	0.76	22-Jul	0.84
9-Feb	1.12	22-Mar	2.6	2-May	2.4	12-Jun	1.04	23-Jul	0.78
10-Feb	1.08	23-Mar	0.92	3-May	0.9	13-Jun	1.08	24-Jul	1.1

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Table L1-12B. Uranium Concentrations Measured at Manhole 175 in 1967 (cont'd)

Date 1967	U[a] (mg L^{-1})	Date 1967	U[a] (mg L^{-1})	Date 1967	U[a] (mg L^{-1})	Date 1967	U[a] (mg L^{-1})
25-Jul	1	4-Sep	0.52	15-Oct	0.9	25-Nov	2
26-Jul	1	5-Sep	0.58	16-Oct	0.92	26-Nov	0.9
27-Jul	0.7	6-Sep	0.74	17-Oct	3	27-Nov	0.92
28-Jul	0.02	7-Sep	0.64	18-Oct	0.76	28-Nov	2.2
29-Jul	0.08	8-Sep	1.4	19-Oct	0.48	29-Nov	2.4
30-Jul	0.1	9-Sep	1.34	20-Oct	1.26	30-Nov	5.6
31-Jul	0.98	10-Sep	0.54	21-Oct	0.8	1-Dec	4
1-Aug	0.48	11-Sep	0.52	22-Oct	0.52	2-Dec	3.6
2-Aug	0.74	12-Sep	2	23-Oct	0.76	3-Dec	2.6
3-Aug	0.8	13-Sep	1.2	24-Oct	1.24	4-Dec	2.8
4-Aug	5.6	14-Sep	0.58	25-Oct	1.6	5-Dec	1.6
5-Aug	0.7	15-Sep	1.28	26-Oct	2.2	6-Dec	1.6
6-Aug	0.88	16-Sep	1.12	27-Oct	2.4	7-Dec	1.56
7-Aug	0.46	17-Sep	1.16	28-Oct	3.4	8-Dec	0.94
8-Aug	0.64	18-Sep	0.88	29-Oct	1.02	9-Dec	2.8
9-Aug	1.4	19-Sep	2.4	30-Oct	0.84	10-Dec	4
10-Aug	0.78	20-Sep	1.34	31-Oct	4	11-Dec	3.6
11-Aug	0.86	21-Sep	2	1-Nov	3.4	12-Dec	3
12-Aug	0.52	22-Sep	2.2	2-Nov	2.2	13-Dec	1.02
13-Aug	0.58	23-Sep	1.8	3-Nov	0.82	14-Dec	6.6
14-Aug	0.8	24-Sep	3	4-Nov	0.72	15-Dec	2.8
15-Aug	0.6	25-Sep	0.68	5-Nov	0.76	16-Dec	1.44
16-Aug	0.92	26-Sep	1.22	6-Nov	0.52	17-Dec	2.2
17-Aug	2.4	27-Sep	3	7-Nov	0.88	18-Dec	3.2
18-Aug	1.42	28-Sep	1.38	8-Nov	1.26	19-Dec	1.56
19-Aug	1.54	29-Sep	1.06	9-Nov	0.74	20-Dec	3.2
20-Aug	0.84	30-Sep	0.78	10-Nov	1.02	21-Dec	1.58
21-Aug	0.66	1-Oct	0.74	11-Nov	3	22-Dec	2.2
22-Aug	0.6	2-Oct	0.58	12-Nov	1.4	23-Dec	1.02
23-Aug	1.42	3-Oct	0.42	13-Nov	0.76	24-Dec	0.58
24-Aug	1.38	4-Oct	0.9	14-Nov	0.84	25-Dec	0.58
25-Aug	1.8	5-Oct	1.04	15-Nov	2.4	26-Dec	0.32
26-Aug	0.48	6-Oct	3	16-Nov	1.12	27-Dec	0.52
27-Aug	1.1	7-Oct	1.14	17-Nov	4.4	28-Dec	0.62
28-Aug	0.84	8-Oct	1.06	18-Nov	1.08	29-Dec	1.32
29-Aug	1.12	9-Oct	0.94	19-Nov	0.9	30-Dec	1.02
30-Aug	0.64	10-Oct	1.16	20-Nov	1.14	31-Dec	2.4
31-Aug	0.9	11-Oct	0.54	21-Nov	0.46	Average	1.5
1-Sep	0.54	12-Oct	0.98	22-Nov	1.6	StdDev	1.0
2-Sep	0.24	13-Oct	1	23-Nov	0.9	Max	6.6
3-Sep	0.2	14-Oct	0.38	24-Nov	1.4	Min	0.02

[a] From NLCO 1967, analytical data sheets from the Bioassay Department at the FMPC.

Table L1–13A. Uranium Concentrations Measured at Manhole 175 in 1969

1969 Date	U[a] ($mg\ L^{-1}$)	1969 Date	U[a] ($mg\ L^{-1}$)	1969 Date	U[a] ($mg\ L^{-1}$)	1969 Date	U[a] ($mg\ L^{-1}$)	1969 Date	U[a] ($mg\ L^{-1}$)
1-Jan	18	11-Feb	2.8	23-Mar	1.28	6-May	0.94	18-Jun	0.96
2-Jan	4	12-Feb	2.2	24-Mar	4.2	7-May	3.8	19-Jun	0.8
3-Jan	2.6	13-Feb	2.2	25-Mar	2.8	8-May	4.4	20-Jun	0.98
4-Jan	2.4	14-Feb	1.04	26-Mar	2	9-May	2.8	21-Jun	1.1
5-Jan	3	15-Feb	3	28-Mar	2.6	10-May	2.2	22-Jun	1.5
6-Jan	2.4	15-Feb	3	29-Mar	3	11-May	1.02	23-Jun	2.2
7-Jan	2	16-Feb	2.8	30-Mar	2.2	12-May	1.34	24-Jun	1.6
8-Jan	2.2	17-Feb	1.54	31-Mar	1.1	13-May	0.92	25-Jun	2.8
9-Jan	2.4	18-Feb	2.8	1-Apr	1.1	14-May	0.62	26-Jun	2.2
10-Jan	2.2	19-Feb	5.2	2-Apr	2.8	15-May	0.9	27-Jun	1.18
11-Jan	2.8	20-Feb	5.6	3-Apr	1.32	16-May	1.38	28-Jun	1.38
12-Jan	2.2	21-Feb	8.2	4-Apr	1.44	17-May	1.2	29-Jun	1.16
13-Jan	2.2	22-Feb	5	5-Apr	3	18-May	2.2	30-Jun	1.28
14-Jan	1.68	23-Feb	3.2	6-Apr	2.8	19-May	0.84	1-Jul	2.2
15-Jan	1.4	24-Feb	5.8	7-Apr	2.6	20-May	0.98	2-Jul	1.8
16-Jan	3.4	25-Feb	12.6	8-Apr	2.2	21-May	1.06	3-Jul	2
17-Jan	3.8	26-Feb	11	9-Apr	3	22-May	0.98	4-Jul	2
18-Jan	2.8	27-Feb	10.2	10-Apr	2.2	23-May	1.24	5-Jul	2
19-Jan	1.34	28-Feb	6.6	11-Apr	1.2	24-May	0.96	6-Jul	2.2
20-Jan	2.4	1-Mar	5.4	12-Apr	1.12	25-May	0.42	7-Jul	2.4
21-Jan	2.4	2-Mar	2.4	13-Apr	0.78	26-May	1.2	8-Jul	4.4
22-Jan	2.2	3-Mar	3.4	14-Apr	0.7	27-May	1.16	9-Jul	3.2
23-Jan	3.8	4-Mar	3	15-Apr	2.4	28-May	1.6	10-Jul	5
24-Jan	2.6	5-Mar	1.54	16-Apr	1.1	29-May	1.8	11-Jul	3.6
25-Jan	2.6	6-Mar	7	17-Apr	1.42	30-May	2.2	12-Jul	2.4
26-Jan	2.2	7-Mar	5.6	18-Apr	2.2	31-May	1.6	13-Jul	3
27-Jan	1.48	8-Mar	4.6	19-Apr	3	2-Jun	1.8	14-Jul	2.2
28-Jan	3.2	9-Mar	2.6	20-Apr	2.2	3-Jun	1.04	15-Jul	2
29-Jan	2.2	10-Mar	3.8	22-Apr	1.42	4-Jun	2.8	16-Jul	2.2
30-Jan	2	11-Mar	4	23-Apr	1.3	5-Jun	0.88	18-Jul	1.44
31-Jan	3.2	12-Mar	4	24-Apr	1.1	6-Jun	1.42	19-Jul	3.2
1-Feb	2.8	13-Mar	4	25-Apr	2	7-Jun	1.16	20-Jul	3.4
2-Feb	2.8	14-Mar	2.6	26-Apr	1.14	8-Jun	0.74	21-Jul	3.2
3-Feb	2.4	15-Mar	2	27-Apr	1.22	9-Jun	0.86	22-Jul	2
4-Feb	2.4	16-Mar	2.4	28-Apr	0.8	10-Jun	0.72	23-Jul	2.2
5-Feb	1.04	17-Mar	3.8	29-Apr	0.44	12-Jun	5.2	24-Jul	2.2
6-Feb	2.4	18-Mar	3.2	1-May	2.8	13-Jun	2.8	25-Jul	8.2
7-Feb	2.2	19-Mar	3	2-May	1	14-Jun	3.2	26-Jul	8.4
8-Feb	1.28	20-Mar	2.2	3-May	0.7	15-Jun	2.6	27-Jul	4
9-Feb	2.6	21-Mar	1.58	4-May	0.54	16-Jun	2	28-Jul	5.2
10-Feb	2.6	22-Mar	2.6	5-May	0.9	17-Jun	1.16	29-Jul	2.4

(Continued on next page)

Table L1-13B. Uranium Concentrations Measured at Manhole 175 in 1969 (cont'd)

1969 Date	U[a] (mg L⁻¹)	1969 Date	U[a] (mg L⁻¹)	1969 Date	U[a] (mg L⁻¹)	1969 Date	U[a] (mg L⁻¹)
30-Jul	2.2	14-Sep	0.38	27-Oct	1.42	7-Dec	2.6
31-Jul	1.16	15-Sep	0.94	28-Oct	1.42	8-Dec	3.4
1-Aug	2	16-Sep	1.06	29-Oct	1.38	9-Dec	4.6
2-Aug	2.2	17-Sep	1.42	30-Oct	1.24	10-Dec	3
3-Aug	3	18-Sep	0.78	31-Oct	1.36	11-Dec	2
4-Aug	2.8	19-Sep	0.78	1-Nov	2.6	12-Dec	0.9
5-Aug	3.2	20-Sep	2.2	2-Nov	2.2	13-Dec	0.72
6-Aug	1.54	21-Sep	0.76	3-Nov	1.24	14-Dec	1.24
7-Aug	2.2	22-Sep	0.5	4-Nov	3	15-Dec	1.4
8-Aug	3.2	23-Sep	0.9	5-Nov	1.36	16-Dec	1.36
9-Aug	3.8	24-Sep	0.88	6-Nov	2.2	17-Dec	2
10-Aug	2	25-Sep	0.74	7-Nov	3.6	18-Dec	3.2
11-Aug	1.32	27-Sep	2.2	8-Nov	3	19-Dec	2.8
12-Aug	3	28-Sep	1.16	9-Nov	2.2	20-Dec	2.4
13-Aug	4.4	29-Sep	0.64	10-Nov	0.72	21-Dec	4.6
14-Aug	1	30-Sep	0.58	11-Nov	0.98	22-Dec	3
15-Aug	1.08	1-Oct	1	12-Nov	2.4	23-Dec	2.4
16-Aug	1.32	2-Oct	0.92	13-Nov	4	24-Dec	3.4
17-Aug	1.26	3-Oct	1.24	14-Nov	1.3	25-Dec	2.2
18-Aug	2.2	4-Oct	1.24	15-Nov	0.8	26-Dec	1.34
19-Aug	3.2	6-Oct	1.06	16-Nov	0.9	27-Dec	2.2
20-Aug	0.7	7-Oct	1.04	17-Nov	3	28-Dec	1.16
21-Aug	0.7	8-Oct	0.76	18-Nov	3.4	29-Dec	3.6
22-Aug	0.72	9-Oct	1.02	19-Nov	1.12	30-Dec	
23-Aug	1.16	10-Oct	1.4	20-Nov	1	31-Dec	3.6
24-Aug	0.8	11-Oct	1.8	21-Nov	1.34		
27-Aug	2	12-Oct	0.98	22-Nov	1.06		
28-Aug	1.44	13-Oct	1.12	23-Nov	2		
29-Aug	0.8	14-Oct	1.32	24-Nov	0.72		
30-Aug	0.64	15-Oct	2.2	25-Nov	2.2		
31-Aug	0.84	16-Oct	1.3	26-Nov	2.4		
2-Sep	3.4	17-Oct	1.02	27-Nov	2.6		
3-Sep	1.28	18-Oct	0.68	28-Nov	2.8		
5-Sep	1.04	19-Oct	1.02	29-Nov	0.9		
6-Sep	1.5	20-Oct	0.84	30-Nov	0.48		
7-Sep	1.58	21-Oct	0.92	1-Dec	0.44		
9-Sep	1.28	22-Oct	0.68	2-Dec	0.92		
10-Sep	1.26	23-Oct	0.58	3-Dec	2.2	Average	2.21
11-Sep	0.7	24-Oct	1.2	4-Dec	1.22	StdDev	1.76
12-Sep	0.78	25-Oct	0.86	5-Dec	1	Max	18
13-Sep	1.1	26-Oct	1.2	6-Dec	1.26	Min	0.38

[a] From NLCO 1969, analytical data sheets from the Bioassay Department at the FMPC

Table L1–14. Uranium Quantities Measured at the Storm Sewer Outfall in 1954 [a]

Date	U(mg L^{-1})	Date	U(mg L^{-1})
6-Jun	0.892	15-Aug	0.08
6-Jul	0.064	16-Aug	0.15
9-Jul	0.112	17-Aug	0.037
16-Jul	0.086	22-Aug	0.107
17-Jul	0.491	25-Aug	0.029
18-Jul	2.52	28-Aug	0.284
19-Jul	0.54	11-Sep	0.214
22-Jul	0.1338	14-Sep	0.086
25-Jul	0.406	18-Sep	0.026
27-Jul	0.216	22-Sep	0.083
grab	4.15	26-Sep	0.107
31-Jul	0.11	2-Oct	0.064
1-Aug	0.026	5-Oct	0.091
2-Aug	0.134	6-Oct	0.112
3-Aug	0.139	7-Oct	0.299
4-Aug	0.1498	10-Oct	0.067
5-Aug	0.061	14-Oct	0.164
6-Aug	0.112	18-Oct	0.244
7-Aug	0.4815	24-Oct	0.564
9-Aug	0.075		
11-Aug	0.067	Avg	0.49
12-Aug	0.051	StdDev	0.93
13-Aug	0.396	Max	4.15
14-Aug	0.321	Min	0.026

[a] From NLCO 1954; all samples were taken at the storm sewer outfall because the storm sewer lift station was not operational until August 17, 1955.

Table L1–15. Uranium Quantities Measured at the Storm Sewer Outfall and Lift Station in 1955

1955 Date	U (mg L^{-1})	Sample Location[a]	1955 Date	U (mg L^{-1})	Sample Location[a]	1955 Date	U (mg L^{-1})	Sample Location[a]
1-Jan	0.887		25-May	0.07		24-Sep	0.773	Lift
15-Jan	0.374		30-May	0.284		27-Sep	0.567	Lift
18-Jan	0.927		3-Jun	0.586		30-Sep	0.567	Lift
21-Jan	0.25		6-Jun	0.634		6-Oct	1.1133	Lift
24-Jan	0.015		10-Jun	0.516		9-Oct	0.206	Lift
28-Jan	0.148		13-Jun	0.554		12-Oct	2.782	Lift
30-Jan	0.176		16-Jun	0.157		15-Oct	0.618	b
2-Feb	0.515		19-Jun	0.342		21-Oct	0.31	b
5-Feb	0.438		24-Jun	0.351		24-Oct	0.516	Lift
9-Feb	0.309		27-Jun	0.115		27-Oct	0.512	Lift
12-Feb	0.297		30-Jun	0.166		30-Oct	0.824	Lift
8-Mar	0.399		3-Jul	0.39		2-Nov	1.236	Outfall
13-Mar	0.204		7-Jul	0.293		5-Nov	0.348	Lift
16-Mar	0.375		10-Jul	0.412		9-Nov	0.359	Lift
19-Mar	0.361		13-Jul	0.29		12-Nov	0.464	Lift
24-Mar	0.121		16-Jul	0.251		15-Nov	0.876	Lift
27-Mar	0.158		19-Jul	0.193		18-Nov	0.3	b
3-Apr	0.5		22-Jul	0.068		24-Nov	0.506	Lift
6-Apr	0.927		25-Jul	0.513		27-Nov	0.282	Lift
9-Apr	0.148		28-Jul	0.261		30-Nov	1.893	Lift
12-Apr	1.334		1-Aug	0.361		6-Dec	0.328	b
15-Apr	0.287		4-Aug	0.135		12-Dec	0.366	b
18-Apr	0.148		8-Aug	0.406		15-Dec	0.318	Lift
21-Apr	0.115		11-Aug	0.3		18-Dec	0.194	Lift
24-Apr	0.234		14-Aug	0.218		21-Dec	0.316	Lift
27-Apr	0.144		17-Aug	0.126	Lift[a]	24-Dec	0.176	Lift
1-May	0.172		20-Aug	0.361	Lift	28-Dec	0.168	Lift
7-May	0.379		24-Aug	0.198	Outfall			
9-May	0.168		27-Aug	0.329	Outfall			
12-May	1.327		31-Aug	0.242	Lift	Average	0.43	
15-May	0.107		7-Sep	0.184	Lift	StdDev	0.42	
18-May	0.078		10-Sep	1.634	Lift	Max	2.782	
21-May	0.095		21-Sep	0.444	b	Min	0.015	

[a] NLCO 1955; all samples taken at outfall until August 17 when lift station opened.
[b] Location not specified on the analytical data sheets.

Table L1–16A. Uranium Quantities Measured at the Storm Sewer Lift Station in 1956

Date Collected	U (mg L^{-1})	Date Collected	U (mg L^{-1})	Date Collected	U (mg L^{-1})	Date Collected	U (mg L^{-1})
1-Jan	0.072	16-May	0.522	26-Jun	0.638	6-Aug	0.312
4-Jan	0.264	17-May	1.01	27-Jun	0.612	7-Aug	0.176
7-Jan	0.104	18-May	0.366	28-Jun	0.612	8-Aug	0.722
10-Jan	0.081	19-May	0.592	29-Jun	0.29	9-Aug	0.274
13-Jan	0.176	20-May	0.244	30-Jun	0.612	10-Aug	0.946
16-Jan	0.282	21-May	0.244	1-Jul	0.522	11-Aug	0.214
19-Jan	0.678	22-May	0.522	2-Jul	0.6	12-Aug	0.234
22-Jan	0.405	23-May	0.586	3-Jul	1.632	13-Aug	0.566
25-Jan	0.564	24-May	0.14	4-Jul	2.04	14-Aug	0.63
28-Jan	2.71	25-May	0.156	5-Jul	1.428	15-Aug	0.63
1-Feb	0.58	26-May	0.976	6-Jul	0.426	16-Aug	0.352
4-Feb	1.742	27-May	0.976	7-Jul	0.232	17-Aug	0.174
15-Apr	1.58	28-May	0.592	8-Jul	0.194	18-Aug	0.202
16-Apr	0.572	29-May	0.586	9-Jul	0.254	19-Aug	0.036
17-Apr	1.186	30-May	0.504	10-Jul	0.312	20-Aug	0.698
18-Apr	0.71	31-May	0.586	11-Jul	0.78	21-Aug	0.166
19-Apr	1.36	1-Jun	0.574	12-Jul	2.1	22-Aug	1.188
20-Apr	0.314	2-Jun	0.382	13-Jul	0.63	23-Aug	0.718
21-Apr	0.226	3-Jun	2.928	14-Jul	0.526	24-Aug	0.792
22-Apr	0.586	4-Jun	1.624	15-Jul	0.37	25-Aug	0.404
23-Apr	0.488	5-Jun	0.714	16-Jul	0.84	26-Aug	0.694
24-Apr	0.488	6-Jun	0.658	17-Jul	1.786	27-Aug	7.92
25-Apr	2.538	7-Jun	0.58	18-Jul	0.488	28-Aug	1.584
26-Apr	0.586	8-Jun	0.406	19-Jul	0.84	29-Aug	0.792
27-Apr	0.488	9-Jun	0.426	20-Jul	1.26	30-Aug	0.99
28-Apr	1.756	10-Jun	0.136	21-Jul	0.739	31-Aug	1.188
29-Apr	1.756	11-Jun	0.816	22-Jul	0.63	1-Sep	0.718
30-Apr	0.592	12-Jun	0.368	23-Jul	0.946	2-Sep	0.396
1-May	0.644	13-Jun	0.562	24-Jul	0.912	3-Sep	0.478
2-May	0.78	14-Jun	0.272	25-Jul	0.946	4-Sep	0.304
3-May	0.696	15-Jun	1.364	26-Jul	0.254	5-Sep	0.138
4-May	0.54	16-Jun	0.368	27-Jul	0.214	6-Sep	0.594
5-May	0.974	17-Jun	0.388	28-Jul	1.47	7-Sep	0.184
6-May	0.626	18-Jun	0.29	29-Jul	2.1	8-Sep	0.202
7-May	0.436	19-Jun	1.16	30-Jul	0.74	9-Sep	1.98
8-May	0.192	20-Jun	1.224	31-Jul	2.72	10-Sep	0.792
9-May	0.436	21-Jun	0.658	1-Aug	0.37	11-Sep	1.09
10-May	0.452	22-Jun	0.816	2-Aug	0.39	12-Sep	0.46
11-May	0.244	23-Jun	0.638	3-Aug	0.39	13-Sep	0.35
12-May	0.296	24-Jun	0.348	4-Aug	0.118	14-Sep	1.782
13-May	0.104	25-Jun	0.446	5-Aug	0.214	15-Sep	2.178

[a] From NLCO 1956; All samples taken at lift station.

Table L1–16B. Uranium Quantities Measured at the Storm Sewer Lift Station in 1956 [a] (cont'd)

1956 Date	U (mg L⁻¹)	1956 Date	U (mg L⁻¹)	1956 Date	U (mg L⁻¹)
16-Sep	1.08	24-Oct	0.542	1-Dec	5.89
17-Sep	0.392	25-Oct	0.342	2-Dec	0.672
18-Sep	0.426	26-Oct	1.728	3-Dec	0.57
19-Sep	0.63	27-Oct	0.362	4-Dec	0.576
20-Sep	0.306	28-Oct	0.476	5-Dec	0.23
21-Sep	0.222	29-Oct	0.304	6-Dec	0.614
22-Sep	0.698	30-Oct	0.532	7-Dec	3.23
23-Sep	0.358	31-Oct	0.552	8-Dec	3.9
24-Sep	0.324	1-Nov	0.96	9-Dec	1.996
25-Sep	0.612	2-Nov	0.704	10-Dec	0.57
26-Sep	0.238	3-Nov	1.536	11-Dec	0.442
27-Sep	0.17	4-Nov	0.276	12-Dec	0.556
28-Sep	0.204	5-Nov	0.59	13-Dec	0.26
29-Sep	0.34	6-Nov	0.384	14-Dec	0.476
30-Sep	0.34	7-Nov	4.37	15-Dec	1.236
1-Oct	0.408	8-Nov	4.75	16-Dec	0.856
2-Oct	0.358	9-Nov	3.072	17-Dec	0.666
3-Oct	2.88	10-Nov	0.556	18-Dec	1.14
4-Oct	1.98	11-Nov	0.384	19-Dec	0.308
5-Oct	0.476	12-Nov	0.57	20-Dec	2.09
6-Oct	0.72	13-Nov	0.48	21-Dec	3.23
7-Oct	0.442	14-Nov	0.346	22-Dec	1.33
8-Oct	0.712	15-Nov	1.52	23-Dec	2.28
9-Oct	0.204	16-Nov	0.95	24-Dec	2.28
10-Oct	0.392	17-Nov	0.308	25-Dec	0.76
11-Oct	0.494	18-Nov	0.556	26-Dec	0.596
12-Oct	2.8	19-Nov	0.326	27-Dec	0.384
13-Oct	0.426	20-Nov	1.14	28-Dec	1.632
14-Oct	0.442	21-Nov	1.9	29-Dec	0.96
15-Oct	0.374	22-Nov	0.556	31-Dec	5.51
16-Oct	0.426	23-Nov	0.556		
17-Oct	0.324	24-Nov	0.288		
18-Oct	0.19	25-Nov	1.9		
19-Oct	0.4	26-Nov	1.71		
20-Oct	0.742	27-Nov	0.538	Avg	0.86
21-Oct	0.456	28-Nov	0.346	StdDev	0.95
22-Oct	1.9	29-Nov	0.76	Max	7.92
23-Oct	0.576	30-Nov	1.248	Min	0.036

[a] From NLCO 1956; all samples taken at lift station.

Table L1–17. Uranium Quantities Measured at the Storm Sewer Outfall and Lift Station in 1957[a]

1957 Date	OutFall U (mg L^{-1})	Lift Station U (mg L^{-1})
1-Feb	0.76	1.06
9-Feb	2.56	4.6
3-Apr	3.86	2.02
22-May	1.326	1.1
25-May	1.428	7.46
31-May	2	0.96
27-Jun	1.28	1.34
28-Jun	0.76	1.54
27-Jul	0.28	3.16
8-Nov	1.64	3.22
14-Nov	1.24	2.3
18-Nov	1.14	1.8
7-Dec	1.52	1.96
8-Dec	0.16	0.78
Average	1.43	2.38
Stdev	0.94	1.81
Max	3.86	7.46
Min	0.16	0.78

[a] From NLCO 1957.

Table L1-18. Uranium Concentrations Measured at the Storm Sewer Outfall in 1962, 1963 and 1964 [a]

U Conc. (mg L^{-1})			Summary	U Conc. (mg L^{-1})		
1962	1963	1964		1962	1963	1964
35	41	16.4		6	3.4	4.8
29.5	28	16.2		5.5	3.4	4.8
18.5	16	15.6		5.5	3.3	4.8
17.5	15.2	13.4		5.5	3.2	4.6
15.5	14.5	13.2		5.5	3	4.6
15	8	12.6		5.5	3	4.6
12.5	7.5	12.2		5.5	3	4.4
11	7.5	12		5.5	2.8	4.4
10.5	7.5	11.8		5.5	2.8	4.4
10.5	6	10		5.5	2.2	4.2
10	6	10		5.5	2	4
9.5	6	9		5	2	4
9	6	8.6		5		4
5	6.2	8.4		5		3.8
9.5	5.5	8.2		4.5		3.8
8.5	5.5	7.6		4.5		3.8
8.5	5.4	7.4		4.5		3.8
8.5	5.2	7		4.5		3.8
8	5	6.8		4.5		3.6
8	5	6.8		4		3.1
7.5	4.8	6.6		4		3
7	4.6	6.6		2.3		2.8
7	4.6	6.4		1.9		2.6
7	4.5	6.4		0.8		2.4
7	4.5	6				2.4
6.5	4.5	5.8				2
6.5	4.4	5.8				1.4
6.5	4.4	5.6				0.3
6.5	4.2	5.4				
6.5	4	5.4	Average	8.06	6.65	6.39
6	3.6	5	StdDev	5.86	7.02	3.73
6	3.4	4.8	Max	35	41	16
6	3.4	4.8	Min	0.80	2.00	0.30

[a] From Rathgens, 1965; handwritten ledger sheets summarizing uranium measurements taken throughout the year for frequency distribution. The data for 1962 and 1963 were used to calculate total uranium to Paddy's Run from storm sewer outfall assuming 250, 000 gallons per day (See Figure L–3). For 1964, data from analytical data sheets are given in Table L1–21 in the annex.

Table L1–19. Uranium Concentrations and Volume of Effluent to Paddy's Run Via the Storm Sewer Outfall Ditch in 1960

1960 Month	# Outfall Events [a]	Date	U (mg L^{-1})	Volume (gal)	Uranium (kg)	DLW[b] (kg)	MLR[c] (kg)
Jan-60	?	27	6.0	44400	1	1	128
Feb-60	7	4	6.0	72000	2	45	
		5	5.0	5124000	96		
		6	1.8	79800	1		
		9	6.0	98160	2		
		10	4.5	1770750	30		
		11	0.6	30000	0.1		
		25	1.5	400420	2		
Mar-60	3	14	2.3	24000	0.2	3	
		16	2.4	3150	<0.1		
		17	6.4	12000	0.3		
Apr-60	3	3	9.8	269800	10	10	10
		26	9.5	264000	9		
		30	19.5	109560	8		
May-60	3	12	8.5	18500	1	125	125
		16	48.0	100500	18		
		20	14.5	64800	4		
Jun-60	?	27	4.1	66730	1	64	176
Jul-60	?	3	13.5	2174000	111	137	138
		13	8.5	500000	16		
		18	6.0	447900	10		
Aug-60	?	29	18.5	42000	3	73	
		30	30.0	621000	70		
Sep-60	0					0	73
Oct-60	4	5	18.0	198000	13	87	87
		8	15.0	210000	12		
		15	35.0	48000	6		
		19	14.0	1050000	56		
Nov-60	5	9	14.0	546000	29	50	
		16	0.9	31200	<0.1		
		22	13.5	402720	21		
		23	11.5	24480	1		
		28	16.0	109600	7		
Dec-60	3	6	10.5	580000	23	40	40
		11	9.0	316300	11		
		26	5.5	259200	5		
Totals				16112970	579	635	770
Avg/Event				460371	17		

[a] Records were not complete to verify number of events for all months. These events refer only to material lost to Paddy's Run through the storm sewer outfall ditch. Additional quantities were lost to Paddy's Run through runoff from the west side of the facility. These additional quantities are included in our final source term estimates reported in Table L–8.
[b] From Starkey 1960-1961.
[c] From Cuthbert 1960, 1961.

Table L1-20. U Concentrations and Effluent Volume to Paddy' Run Via in 1961

Month	# Events [a]	Date	U (mg L^{-1})	Vol. (gal)	U (kg)	DLW (kg)[b]	MLR (kg)[b]
Jan-61	2	6	6.5	1632000	40	81	86
		15	11.5	948000	41		
Feb-61	7	10	13.0	240000	12	82	41
		17	17.0	126000	8		
		18	13.5	333000	17		
		22	7.0	157200	4		
		25	7.0	1026000	27		
		26	4.5	168000	3		
		28	4.5	639600	11		
Mar-61	9	4	6.5	472500	12	179	219
		5	5.0	2382000	45		
		6	6.5	478500	12		
		8	6.5	1452100	36		
		12	4.5	2054200	35		
		13	4.5	1141100	19		
		19	7.5	264000	7		
		21	6.0	336000	8		
		22	5.5	252000	5		
Apr-61	8	9	8.5	534000	17	94	75
		10	2.6	36000	1		
		12	7.0	786200	21		
		13	5.5	600000	12		
		15	9.0	567000	19		
		16	6.0	216000	5		
		25	10.5	433000	17		
		28	6.5	84000	2		
May-61	6	5	8.5	94500	3	89	108
		6	11.5	126000	5		
		7	6.5	1238400	30		
		8	8.5	1368000	44		
		9	4.5	336000	6		
		18	8.0	96000	3		
Jun-61	4	2	9.5	66000	2	64	64
		8	21.5	300000	24		
		9	25.0	132000	12		
		14	7.5	876000	25		
Jul-61	7	4	7.5	547800	16	91	89
		5	10.5	564600	22		
		15	5.5	739200	15		
		20	10.5	363000	14		
		21	9.5	528000	19		
		22	4.0	125200	2		
		30	15.5	42000	2		
Aug-61	2	1	17.5	144000	10	13	15
		24	9.0	90000	3		
Totals						693	697
Avg/Event			8.7	560,000	15.4		
Max			25	2,380,000	45		
Min			2.6	36,000	0.5		

[a] Records for individual outfall events located for January to August 1961.
[b] From Starkey 1960–1961 (DLW) and Cuthbert 1960,1961 (MLR).

Table L1–21. Uranium Concentrations and Volume of Effluent to Paddy's Run Via the Storm Sewer Outfall Ditch in 1964[a]

1964 Mon	# Outfall Events	Date	Volume (gallon)	U Conc. (mg L^{-1})	U/Event (kg)	U/Month (kg)
Jan-64	9	2	214500	4.4	4	58
		3	288000	10.0	11	
		5	336000	7.4	9	
		9	132000	8.4	4	
		15	8400	1.4	0.04	
		19	168000	10.0	6	
		20	564000	9.0	19	
		24	12000	15.6	1	
		25	132000	6.4	3	
Feb-64	9	5	1374000	16.4	85	98
		15	312000	8.2	10	
		16	64800	13.4	3	
Mar-64	10	2	66000	16.2	4	506
		4	7152000	6.8	184	
		5	150000	4.8	3	
		8	823000	11.8	37	
		9	9288000	3.8	133	
		10	1764000	4.4	29	
		12	350640	6.6	9	
		14	1560000	12.6	74	
		21	112000	6.0	3	
		25	660000	12.0	30	
Apr-64	12	2	1992000	6.4	48	338
		3	3702000	3.8	53	
		5	1200000	4.2	19	
		6	2148000	3.8	31	
		13	3120	3.8	0.05	
		19	3144000	4.0	48	
		20	360000	4.8	7	
		21	7800000	3.6	106	
		22	792000	3.0	9	
		24	325700	4.0	5	
		26	378000	7.6	11	
		27	90000	4.6	2	
May-64	1	11	5200	0.3	0.01	0.01
Jun-64	5	2	144000	4.8	3	190
		6	306000	6.6	8	
		12	13000000	2.6	128	
		13	1920000	2.0	15	
		18	2256000	4.4	38	
Jul-64	4	6	66000	2.4	1	36
		7	540000	5.0	10	
		12	960000	6.8	25	
		18	3500	4.8	0	

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Table L1-21. Uranium Concentrations and Volume of Effluent to Paddy's Run Via the Storm Sewer Outfall Ditch in 1964[a] (cont'd)

1964 Mon	# Outfall Events	Date	Volume (gallon)	U Conc. (mg L^{-1})	U/Event (kg)	U/Month (kg)
Aug-64	3	11	312000	5.6	7	30
		21	360000	13.2	18	
		22	216000	7.0	6	
Sep-64	2	18	720000	5.8	16	37
		19	960000	5.8	21	
Oct-64	1	18	492000	8.6	16	16
Nov-64	4	18	511200	12.2	24	73
		19	1320000	5.4	27	
		25	210000	4.6	4	
		28	1080000	4.6	19	
Dec-64	6	2	248400	3.1	3	78
		3	1134000	5.8	25	
		4	1302000	4.0	20	
		11	2600000	2.4	24	
		16	216000	2.9	2	
		24	204000	5.4	4	
Totals			70,000,000		1458	1458
Average			1,300,000	6	24	122
Max			13,000,000	16.4	180	506
Min			3100	0.3	0.01	0.01

[a] These events refer only to material to Paddy's Run through the storm sewer outfall ditch. Additional quantities were lost to Paddy's Run through runoff from the west side of the facility. These additional quantities are included in our final source term estimates that are reported in Table L–8.

Table L1–22. Uranium Concentrations and Volume Effluent to Paddy's Run Via the Storm Sewer Outfall Ditch in 1966 [a]

1966 Date	Outfall U Conc. (mg L^{-1})	Lift Station U Conc. (mg L^{-1})	Outfall Vol.(gal)	1966 Date	Outfall U Conc. (mg L^{-1})	Lift Station U Conc. (mg L^{-1})	Outfall Vol. (gal)
1-Jan	4.2	3.4	546000	13-Jul	5	1.24	2808000
2-Jan	2.4	2	3105000	23-Jul	8.8	6.4	
5-Jan	0.96	2.8		26-Jul	0.3	0.54	
6-Jan	2.4	3		28-Jul	14	1.6	
7-Feb	3.8	5.8	92400	7-Aug	7.2	2.6	
8-Feb	3.6	7.2		8-Aug	3.8	2.8	
10-Feb	5.6	8.2	4200000	9-Aug	1.14	11	
11-Feb	4.8	14.8		10-Aug	4.2	2.4	423000
13-Feb	3.2	5.4	708000	11-Aug	5	4	
14-Feb		220		13-Aug	6.2	3.4	
8:00 AM	1.8		13 gpm	14-Aug	4.8	3.4	
10:00 AM	1		200 gpm	15-Aug	3	2.6	
10:15 AM	1600		200 gpm	20-Aug	3.8	2.4	
11:15 AM	490		75 gpm	3-Sep	3.6	2.4	
1:15 PM	43	11	130 gpm	15-Sep	5.2	5.4	
2:30 PM	16		116 gpm	19-Sep	4	6.4	
16-Feb	6.5	7.5		20-Sep	4.8	3.8	756000
28-Feb	8.2	6.2		25-Sep	7.6	6.6	327600
12-Mar	4	3.4		15-Oct	3	3.6	1120000
21-Mar	16	2.8		2-Nov	5	4.6	1264500
23-Mar	2.6	3.6		3-Nov	4.8	4.4	320400
3-Apr	0.22	3.8		4-Nov	3.8	3.8	108000
4-Apr	0.42	1.72		5-Nov	3.8	4	247440
11-Apr	4.2	7.4		8-Nov	4	3.6	
12-Apr	4.4	4.6		10-Nov	2.4	3	
24-Apr	6	4	765,000	25-Nov	8.2	3	
27-Apr	5	4		6-Dec	5.4	4.4	
28-Apr	8	9		7-Dec	5.8	3.2	
30-Apr	3.6	2.6		8-Dec		2.8	
9-May	3.4	3		9-Dec	3.4	4	
11-May	4.2	4		10-Dec	2.6	2.6	
28-May	7.6	2.6					
6-Jun	162	400		Avg/event	7.31	10.78	1052583.8
9-Jun	2.6	5.2		Stdev	20.67	51.16	1230753.3
6-Jul	2.4	1.04		Max	1600	400	4200000
10-Jul	4.4	2		Min	0.22	0.54	50000

[a] From NLCO 1966, analytical data sheets; total volume was not given for all outfall events. an average value was used to calculate the total quantity of uranium to Paddy's Run. These events refer only to material to Paddy's Run through the storm sewer outfall ditch. Additional quantities were lost to Paddy's Run through runoff from the west side of the facility. These additional quantities are included in our final source term estimates that are reported in Table L–8.

Table L1–23. Reported Quantities of Uranium Discharged to the Storm Sewer System at the FMPC[a]

Date	Rain[b] (inches)	Total U to Storm Sewer (kg)	Storm Sewer Lift Station (kg)	Storm Sewer Lift Station %	Paddy's Run (kg)	Paddy's Run %
Jan-60	2.5	330	200	61	130	39
Feb-60	3	340[b]	210	61	130	39
Mar-60	0.5	180[b]	180	99	3	2
Apr-60	1	425	400	93	10	7
May-60	3.5	510	385	75	125	25
Jun-60	5.5	650	470	73	175	27
Jul-60	4.5	395	260	65	137	35
Aug-60	1.5	490[b]	415	85	73	15
Sep-60	1	355	280		0	d
Oct-60	2	480	395	82	90	18
Nov-60	2	475[b]	420	88	50	12
Dec-60	1.5	500	460	92	40	8
1960 Totals		5200	4100		1000	
Jan-61	1	422	341	81	80	19
Feb-61	3.5	475	393	83	82	17
Mar-61	4.5	642	463	72	179	28
Apr-61	3.5	396	302	76	94	24
May-61	6	500	408	82	90	18
Jun-61	3.5	319	255	80	65	20
Jul-61	8.5	359	268	75	90	25
Aug-61	2	609	596	98	15	2
Sep-61	3.2	740[c]	479	65	261	35
Oct-61	1.5	230[b]	184	80	46	20
Nov-61	3.5	400[b]	294	73	106	27
Dec-61	3	310[b]	288	93	22	7
1961 Totals		5400	4300		1100	
Jan-62	3.5	590[b]	457	77	135	23
Feb-62	4.5	700	574	82	126	18
Mar-62	3	550[b]	243	44	310	56
Apr-62	0.5	425[b]	340	80	85	20
May-62	4	615[b]	486		129	c
Jun-62	1	320[b]	190	59	130	41
Jul-62	6.5	450[b]	378	84	72	16
Aug-62	2	425[b]	380	89	45	11
Sep-62	0.5	1383[c]	1378	99	5	b
Oct-62	3	480[b]	404	84	76	16
Nov-62	1.5	376[c]	317	84	59	b
Dec-62	1	505[b]	399	b	106	c
1962 Totals		6800	5500		1300	

[a] From Cuthbert 1960–1962 and Starkey 1960–1961 unless otherwise noted. For our source term estimates, additional material was assumed to be lost to Paddy's Run through runoff from the west side of the site.
[b] From Fischoff 1960–1962.
[c] Uranium loss was calculated as 21% of the loss to the storm sewer system.
[d] No losses were reported to Paddy's Run through the storm sewer outfall ditch.

Table L1–24A. Daily Measurements of Total Suspended Solids (TSS) in Liquid Effluents Discharged to the River in 1957 (mg L^{-1})[a]

Date	TSS	Date	TSS	Date	TSS	Date	TSS	Date	TSS
1-Jan	982	12-Feb	1029	26-Mar	1427	7-May	88	19-Jun	121
2-Jan	1105	13-Feb	1375	27-Mar	177	8-May	242	20-Jun	159
3-Jan	982	14-Feb	1374	28-Mar	731	9-May	47	21-Jun	150
4-Jan	1490	15-Feb	1055	29-Mar	726	11-May	191	22-Jun	122
5-Jan	762	16-Feb	253	30-Mar	392	12-May	159	23-Jun	87
6-Jan	651	17-Feb	1524	31-Mar	548	13-May	129	24-Jun	242
7-Jan	517	18-Feb	1016	1-Apr	130	14-May	192	25-Jun	225
8-Jan	1271	19-Feb	177	2-Apr	110	15-May	86	26-Jun	46
9-Jan	904	20-Feb	76	3-Apr	89	16-May	45	27-Jun	139
10-Jan	1093	21-Feb	214	4-Apr	299	17-May	139	28-Jun	45
11-Jan	1049	22-Feb	117	5-Apr	439	18-May	86	29-Jun	141
12-Jan	2374	23-Feb	75	6-Apr	2318	19-May	162	30-Jun	48
13-Jan	1608	24-Feb	306	7-Apr	1209	20-May	816	1-Jul	539
14-Jan	1283	25-Feb	598	8-Apr	1230	21-May	1621	2-Jul	539
15-Jan	2021	26-Feb	413	9-Apr	261	22-May	195	3-Jul	111
16-Jan	1403	27-Feb	407	10-Apr	926	23-May	742	4-Jul	76
17-Jan	1241	28-Feb	466	11-Apr	919	24-May	274	5-Jul	524
18-Jan	746	1-Mar	95	12-Apr	300	25-May	458	6-Jul	413
19-Jan	1261	2-Mar	163	13-Apr	120	26-May	2609	7-Jul	142
20-Jan	881	3-Mar	85	14-Apr	412	27-May	1235	8-Jul	103
21-Jan	37	4-Mar	71	15-Apr	379	28-May	357	9-Jul	438
22-Jan	476	5-Mar	98	16-Apr	483	29-May	172	10-Jul	271
23-Jan	253	6-Mar	101	17-Apr	165	30-May	426	11-Jul	150
24-Jan	537	7-Mar	262	18-Apr	653	31-May	306	12-Jul	41
25-Jan	281	8-Mar	261	19-Apr	837	1-Jun	50	13-Jul	411
26-Jan	631	9-Mar	121	20-Apr	1649	2-Jun	57	14-Jul	598
27-Jan	1144	10-Mar	395	21-Apr	162	3-Jun	52	15-Jul	442
28-Jan	1167	11-Mar	109	22-Apr	73	4-Jun	583	16-Jul	607
29-Jan	1293	12-Mar	803	23-Apr	99	5-Jun	132	17-Jul	92
30-Jan	854	13-Mar	509	24-Apr	76	6-Jun	526	18-Jul	397
31-Jan	1120	14-Mar	470	25-Apr	110	7-Jun	99	19-Jul	328
1-Feb	1000	15-Mar	315	26-Apr	156	8-Jun	130	20-Jul	483
2-Feb	1491	16-Mar	1286	27-Apr	260	9-Jun	233	21-Jul	48
3-Feb	965	17-Mar	509	28-Apr	78	10-Jun	166	22-Jul	953
4-Feb	1027	18-Mar	1263	29-Apr	106	11-Jun	1197	23-Jul	110
5-Feb	1192	19-Mar	366	30-Apr	709	12-Jun	456	24-Jul	381
6-Feb	1394	20-Mar	206	1-May	463	13-Jun	240	25-Jul	272
7-Feb	1154	21-Mar	532	2-May	176	14-Jun	32	26-Jul	84
8-Feb	702	22-Mar	404	3-May	110	15-Jun	153	27-Jul	83
9-Feb	829	23-Mar	253	4-May	115	16-Jun	577	28-Jul	57
10-Feb	262	24-Mar	539	5-May	658	17-Jun	95	29-Jul	182
11-Feb	354	25-Mar	205	6-May	140	18-Jun	610	30-Jul	195

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Table L1-24B. Daily Measurements of Total Suspended Solids (TSS) in Liquid Effluents Discharged to the River in 1957 (mg L^{-1})[a] (cont'd)

Date	TSS	Date	TSS	Date	TSS	Date	TSS
31-Jul	333	11-Sep	221	22-Oct	105	1-Dec	148
1-Aug	221	12-Sep	618	23-Oct	124	2-Dec	58
2-Aug	373	13-Sep	116	22-Oct	105	3-Dec	299
3-Aug	272	14-Sep	86	23-Oct	124	4-Dec	121
4-Aug	756	15-Sep	108	26-Oct	69	5-Dec	52
5-Aug	788	16-Sep	87	27-Oct	60	6-Dec	157
6-Aug	227	17-Sep	73	28-Oct	33	7-Dec	87
7-Aug	424	18-Sep	247	29-Oct	51	8-Dec	183
8-Aug	194	19-Sep	58	30-Oct	792	9-Dec	525
9-Aug	963	20-Sep	87	31-Oct	387	10-Dec	251
10-Aug	854	21-Sep	89	1-Nov	34	11-Dec	158
11-Aug	806	22-Sep	54	2-Nov	62	12-Dec	539
12-Aug	665	23-Sep	109	3-Nov	65	13-Dec	85
13-Aug	1213	24-Sep	50	4-Nov	104	14-Dec	238
14-Aug	288	25-Sep	62	5-Nov	2154	15-Dec	698
15-Aug	471	26-Sep	46	6-Nov	1102	16-Dec	94
16-Aug	391	27-Sep	49	7-Nov	324	17-Dec	117
17-Aug	165	28-Sep	148	8-Nov	278	18-Dec	79
18-Aug	455	29-Sep	71	9-Nov	338	19-Dec	298
19-Aug	294	30-Sep	58	10-Nov	53	20-Dec	88
20-Aug	74	1-Oct	54	11-Nov	43	21-Dec	62
21-Aug	330	2-Oct	264	12-Nov	40	22-Dec	173
22-Aug	590	3-Oct	317	13-Nov	95	23-Dec	142
23-Aug	214	4-Oct	73	14-Nov	101	24-Dec	53
24-Aug	342	5-Oct	53	15-Nov	92	25-Dec	127
25-Aug	1222	6-Oct	41	16-Nov	145	26-Dec	81
26-Aug	155	7-Oct	82	17-Nov	82	27-Dec	92
27-Aug	137	9-Oct	68	18-Nov	272	28-Dec	120
28-Aug	166	10-Oct	497	19-Nov	121	29-Dec	89
29-Aug	546	11-Oct	85	20-Nov	147	30-Dec	127
30-Aug	66	12-Oct	4591	21-Nov	94	31-Dec	11
31-Aug	280	13-Oct	194	22-Nov	110		
1-Sep	76	14-Oct	208	23-Nov	258		
2-Sep	57	15-Oct	76	24-Nov	185		
3-Sep	57	16-Oct	45	25-Nov	106		
4-Sep	45	17-Oct	165	26-Nov	102		
5-Sep	65	18-Oct	146	27-Nov	81		
6-Sep	92	19-Oct	42	28-Nov	137	Average	400
7-Sep	335	20-Oct	32	29-Nov	66	Max	4600
8-Sep	69	21-Oct	43	30-Nov	103	Min	11

[a] From NLCO 1957; original analytical data sheets from the Bioassay Department at the FMPC.

Table L1-25. Monthly Average Total Suspended Solids (TSS) in Liquid Effluents Discharged to the River ($mg\ L^{-1}$)[a]

	1957	1958	1959	1960	1961	1964	1966
Jan	1010	140	140	79	110	270	78
Feb	740	145	105	74	110	270	76
Mar	430	100	63	93	72	150	57
Apr	490	180	65	55	55	180	55
May	400	180	170	58	89	140	31
Jun	230	170	140	45	64	96	40
Jul	290	190	93	72	65	72	150
Aug	450	320	51	58	50	59	61
Sep	110	76	57	44	57	38	28
Oct	290	59	60	54	63	33	
Nov	230	150	91	60	140	55	49
Dec	175	190	72	98	150	63	42
Average	404	158	92	66	85	119	61
StdDev	255	67	39	18	34	84	34
Max	1010	320	170	98	150	270	150
Min	110	59	51	44	50	33	28

[a] From NLCO 1957, NLCO 1958, NLCO 1959, NLCO 1960, NLCO 1961, NLCO 1964, NLCO 1966; daily measurements of total suspended solids made on 24 hour composite samples from the discharge point at MH 175 to the river.

Table L1–26. Radium Quantities Measured at Manhole 175 in 1955

1955 Date	Total Radium µµg mL⁻¹ [a]	Total Radium mg [b]
16-Sep	0.79435	3.30
24-Sep	1.4522	6.04
3-Oct	0.781	3.25
6-Oct	0.0875	0.36
30-Oct	3.141	13.06
2-Nov	0.7114	2.96
12-Nov	3.093	12.86
15-Nov	0.8728	3.63
3-Dec	6.0765	25.27
6-Dec	6.4279	26.73
9-Dec	2.15	8.94
Mean	2.33	9.67
StdDev	2.18	9.05
Max	6.43	26.73
Min	0.09	0.36

[a] NLCO 1955b, analytical data sheets; *uug* is an outdated unit, equivalent 0.000000000001 g.
[b] Calculated quantities using same methods for U quantities, assuming effluent flow of 1 million gallons per day to river via Manhole 175, see Figure L–3.

Table L1–27. Measured Concentrations and Calculated Quantities of Radium at Manhole 175 in 1956

1956	Total Radium		1956	Total Radium		1956	Total Radium	
Date	µµg mL⁻¹ [a]	mg [b]	Date	µµg mL⁻¹ [a]	mg [b]	Date	µµg mL⁻¹ [a]	mg [b]
7-Jan	0.8528	3.55	21-May	0.737	3.06	29-Jun	0.1429	0.59
19-Jan	0.9254	3.85	22-May	0.2447	1.02	30-Jun	0.2608	1.08
22-Jan	0.1153	0.48	23-May	0.4164	1.73	1-Jul	0.387	1.61
15-Apr	0.22	0.91	24-May	0.0209	0.09	2-Jul	0.9033	3.76
16-Apr	0.3905	1.62	25-May	0.1474	0.61	3-Jul	0.3218	1.34
17-Apr	0.27731	1.15	26-May	0.1262	0.52	4-Jul	0.3609	1.50
18-Apr	0.2751	1.14	27-May	0.2336	0.97	5-Jul	0.5036	2.09
19-Apr	0.5894	2.45	28-May	0.2031	0.84	6-Jul	0.155	0.64
20-Apr	0.1836	0.76	29-May	0.0811	0.34	7-Jul	0.1532	0.64
21-Apr	0.1535	0.64	30-May	0.0999	0.42	8-Jul	0.7199	2.99
22-Apr	0.3286	1.37	31-May	0.0658	0.27	9-Jul	0.3747	1.56
23-Apr	0.8205	3.41	1-Jun	0.1051	0.44	10-Jul	1.568	6.52
24-Apr	0.5295	2.20	2-Jun	0.0126	0.05	11-Jul	0.0409	0.17
25-Apr	0.08	0.33	3-Jun	0.3367	1.40	12-Jul	0.1318	0.55
26-Apr	1.26	5.24	4-Jun	0.1625	0.68	13-Jul	0.79	3.28
27-Apr	0.048	0.20	5-Jun	0.4047	1.68	14-Jul	0.047	0.20
28-Apr	0.0975	0.41	6-Jun	0.4254	1.77	15-Jul	1.1217	4.66
29-Apr	0.221	0.92	7-Jun	0.5095	2.12	16-Jul	0.4374	1.82
30-Apr	0.03	0.12	8-Jun	0.2462	1.02	17-Jul	0.9541	3.97
1-May	0.154	0.64	9-Jun	0.5136	2.14	18-Jul	0.1292	0.54
2-May	0.2237	0.93	10-Jun	0.3551	1.48	19-Jul	0.996	4.14
3-May	0.0938	0.39	11-Jun	0.1242	0.52	20-Jul	0.5295	2.20
4-May	0.079	0.33	12-Jun	0.0049	0.02	21-Jul	0.6789	2.82
5-May	0.0854	0.36	13-Jun	0.0249	0.10	22-Jul	0.9204	3.83
6-May	0.0602	0.25	14-Jun	0.0205	0.09	23-Jul	0.298	1.24
7-May	0.0729	0.30	15-Jun	0.0481	0.20	24-Jul	0.109	0.45
8-May	0.0175	0.07	16-Jun	0.0146	0.06	25-Jul	0.159	0.66
9-May	0.0153	0.06	17-Jun	0.0258	0.11	26-Jul	0.1015	0.42
10-May	0.241	1.00	18-Jun	0.1534	0.64	27-Jul	0.0858	0.36
11-May	0.23	0.96	19-Jun	0.1307	0.54	28-Jul	0.0737	0.31
12-May	0.2131	0.89	20-Jun	0.2636	1.10	29-Jul	0.5831	2.42
13-May	0.3951	1.64	21-Jun	0.2782	1.16	30-Jul	2.5701	10.69
14-May	0.0555	0.23	22-Jun	0.4937	2.05	31-Jul	2.9258	12.17
15-May	0.0082	0.03	23-Jun	0.2965	1.23			
16-May	0.0063	0.03	24-Jun	0.1615	0.67			
17-May	0.075	0.31	25-Jun	0.2475	1.03	Mean	0.35	1.44
18-May	0.1775	0.74	26-Jun	0.172	0.72	StdDev	0.44	1.84
19-May	0.4683	1.95	27-Jun	0.1076	0.45	Max	2.93	12.17
20-May	0.2295	0.95	28-Jun	0.5067	2.11	Min	0.0049	0.02

[a] NLCO 1956. Values from original analytical data sheets; *uug* is an outdated unit, equivalent to 0.000000000001 gram.

[b] Calculated quantities using same methods for U quantities, assuming effluent flow of 1 million gallons per day to river via Manhole 175, see Figure L–3.

Table L1-28. Measured Concentrations and Calculated Quantities of Radium at Manhole 175 in 1957

1957 Date	Total Radium µµg mL⁻¹ [a]	Total Radium mg [b]	1957 Date	Total Radium µµg mL⁻¹ [a]	Total Radium mg [b]
1-Jan	0.3873	2	4-Feb	2.043	8.49
2-Jan	1.0107	4	5-Feb	1.0436	4.34
3-Jan	0.9892	4	6-Feb	0.9294	3.86
4-Jan	5.1073	21	7-Feb	1.7832	7.41
5-Jan	0.9014	4	8-Feb	1.8905	7.86
6-Jan	0.309	1	9-Feb	2.1296	8.85
7-Jan	0.4164	2	10-Feb	1.9407	8.07
8-Jan	0.5862	2	11-Feb	0.3313	1.38
9-Jan	0.3549	1	12-Feb	2.0535	8.54
10-Jan	0.5732	2	13-Feb	2.6339	10.95
11-Jan	0.3	1	14-Feb	0.2927	1.22
12-Jan	0.8751	4	15-Feb	1.8532	7.71
13-Jan	0.6343	3	16-Feb	1.6025	6.66
14-Jan	0.2397	1	17-Feb	0.1939	0.81
15-Jan	0.344	1	18-Feb	1.4565	6.06
16-Jan	1.1787	5	19-Feb	1.3825	5.75
17-Jan	0.7535	3	20-Feb	2.0049	8.34
18-Jan	0.4709	2	21-Feb	0.3253	1.35
19-Jan	0.8058	3	22-Feb	0.0961	0.40
20-Jan	0.5284	2	23-Feb	0.1069	0.44
21-Jan	0.0064	0	24-Feb	0.0845	0.35
22-Jan	0.3082	1	25-Feb	1.515	6.30
23-Jan	0.0221	0	26-Feb	1.6653	6.92
24-Jan	0.1167	0	27-Feb	2.2093	9.19
25-Jan	0.1868	1	28-Feb	0.5988	2.49
26-Jan	0.8341	3	1-Mar	1.6952	7.05
27-Jan	0.7516	3	3-Mar	0.4304	1.79
28-Jan	1.4341	6			
29-Jan	2.5368	11			
30-Jan	1.269	5	Average	1.1	4.90
31-Jan	0.9171	4	StdDev	1.1	5.10
2-Feb	6.4124	26.66	Max	6.41	26.66
3-Feb	4.8745	20.27	Min	0.01	0.03

[a] NLCO 1957, original analytical data sheets; *uug* is an outdated unit, equivalent to 0.000000000001 gram.
[b] Calculated quantities using same methods for U quantities, assuming effluent flow of 1 million gallons per day to river via Manhole 175, see Figure L–3.

Table L1–29. Concentration of ^{228}Ra Measured at Manhole 175 and Calculated Activities to the River in 1969 and 1970

1969 [a]	dpm mL^{-1} [b]	pCi L^{-1}	uCi [c]	1970 [b]	dpm mL^{-1} [b]	pCi L^{-1}	uCi [c]
13-Apr	4.95	2200	44000	5-Jan	1.45	653	2832
20-Apr	2.01	900	18000	12-Jan	1.06	477	2070
4-May	4.18	1900	37000	19-Jan	1.39	626	2715
11-May	3.32	1500	30000	26-Jan	1.23	554	2402
18-May	2.58	1200	23000	Feb	1.16	522	9709
25-May	0.70	320	6300	Mar	1.52	684	12722
8-Jun	0.64	290	5700	Apr	2.82	1269	23603
15-Jun	1.16	520	10000	May	3.3	1485	27621
22-Jun	2.09	940	18700	June	0.11	50	921
29-Jun	0.33	150	2900	July	0.19	86	1590
6-Jul	0.18	83	1600	3-Aug	0.048	22	187
13-Jul	0.53	240	4700	17-Aug	0.054	24	211
20-Jul	0.03	13	250	31-Aug	0.069	31	270
27-Jul	0.19	87	1700	7-Sep	0.08	36	312
3-Aug	0.05	23	460	14-Sep	0.018	8	70
10-Aug	0.18	80	1600	28-Sep	0.03	14	117
17-Aug	2.08	940	18600	12-Oct	<0.01	<5	<40
24-Aug	3.08	1400	28000	30-Nov	0.029	13	113
31-Aug	3.32	1500	30000	14-Dec	0.02	9	78
12-Oct	4.86	2200	43000				
19-Oct	2.84	1300	25000				
Nov	2.56	1200	98000				
Dec	3.2	1400	122000				
Total			660,000	Total			88,000
Average	1.90	870	20387	Average	0.81	364	4864
StdDev	1.61	726	22449	StdDev	1.01	456	8330
Max	4.95	2200	98000	Max	3.30	1485	27621
Min	0.03	13	250	Min	0.02	8	70

[a] NLCO 1969. In 1969, the date represents the beginning of a two–week composite sampling period; Nov and Dec represent monthly composites. All values were taken from original analytical data sheets.

[b] NLCO 1970. In 1970, composite samples were taken weekly in January, biweekly from Aug to Dec, and monthly from Feb to July. All values are from original analytical data sheets.

[c] Calculated quantities using same methods for U quantities, assuming effluent flow of 750,000 gallons per day to river via Manhole 175, see Figure L–3.

Table L1-30. Concentration of ^{226}Ra and ^{228}Ra Measured at Manhole 175 and Calculated Activities to the River in 1971

1971	^{226}Ra			^{228}Ra		
Date [a]	dpm mL^{-1} [b]	pCi L^{-1}	uCi [c]	dpm mL^{-1} [b]	pCi L^{-1}	uCi [c]
17-May	0.6	270	8900	0.06	27	890
31-May	0.3	135	4400	0.12	54	1800
14-Jun	0.75	338	11000	0.23	104	3400
28-Jun	0.4	180	5900	0.07	32	1040
12-Jul	0.28	126	4100	0.01	5	150
26-Jul	0.14	63	2100	0.03	14	440
16-Aug	0.42	189	6200	0.08	36	1200
30-Aug	0.41	185	62100	0.14	63	2100
13-Sep	0.09	41	1300	0.01	5	150
27-Sep	0.02	9	300	0.03	14	440
11-Oct	0.02	9	300	0.03	14	440
1-Nov	0.06	27	890	0.04	18	590
15-Nov	0.02	9	300	0.05	23	740
29-Nov	0.06	27	890	0.02	9	300
13-Dec	0.46	207	6800	<0.01	<4.5	<10
Average	0.27	121	4000	0.07	30	970
StdDev	0.23	105	3400	0.06	28	910
Max	0.75	338	11,000	0.23	104	3400
Min	0.02	9	300	0.01	5	150

[a] Beginning date of sampling period; two-week composites. Average, Max and Min values represent a two-week period.

[b] NLCO 1971. Values from original analytical data sheets; reported as *disintegrations per minute per milliliter, dpm/mL.*

[c] Calculated quantities using same methods for U quantities, assuming effluent flow of 620,000 gallons per day to river via Manhole 175, see Figure L–3.

Table L1-31. Concentration of ^{226}Ra and ^{228}Ra Measured at Manhole 175 and Calculated Activities to the River in 1972

1972	^{226}Ra			^{228}Ra		
Date [a]	dpm mL^{-1} [b]	pCi L^{-1}	uCi [c]	dpm mL^{-1} [b]	pCi L^{-1}	uCi [c]
3-Jan	0.51	230	7500	<0.01	<4.5	
17-Jan	0.44	198	6500	<0.01	<4.5	
31-Jan	0.35	158	5200	0.06	27	890
14-Feb	0.4	180	5900	0.12	54	1800
28-Feb	0.12	54	1800	0.02	9	300
13-Mar	0.08	36	1200	<0.01	<4.5	
3-Apr	0.05	23	740	0.02	9	300
17-Apr	0.04	18	590	0.02	9	300
1-May	0.09	41	1300	0.02	9	300
15-May	0.15	68	2200	0.07	32	1040
29-May	0.13	59	1900	0.13	59	1900
12-Jun	0.11	50	1600	0.15	68	2200
3-Jul	0.01	5	150	0.02	9	300
17-Jul	0.06	27	890	0.02	9	300
31-Jul	0.04	18	590	0.02	9	300
14-Aug	0.08	36	1200	0.01	5	150
4-Sep	0.05	23	740	0.01	5	150
18-Sep	0.02	9	300	0.01	5	150
2-Oct	0.02	9	300	0.01	5	150
16-Oct	0.02	9	300	<0.01	<4.5	<10
30-Oct	0.07	32	1000	0.01	5	150
13-Nov	0.08	36	1200	0.01	5	150
27-Nov	0.1	45	1500	<0.01	<4.5	<10
11-Dec	0.08	36	1200	<0.01	<4.5	<10
Average	0.13	58	1900	0.04	19	600
StdDev	0.14	64	2100	0.05	21	680
Max	0.51	230	7500	0.15	68	2200
Min	0.01	5	150	0.01	5	150

[a] Beginning date of sampling period; two-week composite. Average, Max and Min values represent a two-week period.

[b] NLCO 1972. Values from original analytical data sheets; reported as *disintegrations per minute per milliliter, dpm per mL.*

[c] Calculated quantities using same methods for U quantities, assuming effluent flow of 620,000 gallons per day to river via Manhole 175, see Figure L–3.

Table L1-32. Concentration of ^{226}Ra and ^{228}Ra Measured at Manhole 175 and Calculated Activities to the River in 1973

1973	^{226}Ra			^{228}Ra			Total Radium	
Date [a]	dpm mL^{-1} [b]	pCi L^{-1}	uCi [c]	dpm mL^{-1} [b]	pCi L^{-1}	uCi [c]	dpm mL^{-1} [b]	pCi L^{-1}
1-Jan	0.03	14	440	0.03	14	440	0.06	27
15-Jan	0.05	23	740	0.01	5	150	0.06	27
29-Jan	0.06	27	890	0.01	5	150	0.07	32
12-Feb	0.04	18	590	0.02	9	300	0.06	27
26-Feb	0.02	9	300	0.01	5	150	0.03	14
19-Mar	0.01	5	150	0.02	9	300	0.03	14
2-Apr	0.01	5	150	0.02	9	300	0.03	14
16-Apr	0.01	5	150	0.03	14	440	0.04	18
30-Apr	0.01	5	150	0.01	5	150	0.02	9
14-May	0.01	5	150	0.01	5	150	0.02	9
28-May	0.01	5	150	0.03	14	440	0.04	18
11-Jun	0.01	5	150	0.02	9	300	0.03	14
2-Jul	0.02	9	300	0.02	9	300	0.04	18
16-Jul	0.11	50	1600	0.01	5	150	0.12	54
30-Jul	0.49	221	7200	<0.01	<4.5		0.5	225
13-Aug	0.18	81	2700	<0.01	<4.5		0.19	86
3-Sep	0.07	32	1000	<0.01	<4.5		0.08	36
17-Sep	0.04	18	590	<0.01	<4.5		0.05	23
1-Oct	0.03	14	440	0.02	9	300	0.05	23
15-Oct	0.03	14	440	<0.01	<4.5		0.04	18
29-Oct	0.02	9	300	<0.01	<4.5		0.03	14
12-Nov	0.02	9	300	0.01	5	150	0.03	14
3-Dec	0.04	18	590	0.01	5	150	0.05	23
17-Dec	0.04	18	590	<0.01	<4.5		0.05	23
Average	0.06	26	840	0.02	8	262	0.07	32
StdDev	0.10	45	1500	0.01	3	8	0.10	44
Max	0.49	221	7200	0.03	14	31	0.5	225
Min	0.01	5	150	0.01	5	10	0.02	9

[a] Beginning date of sampling period; two-week composite. Average, Max and Min values represent a two-week period.

[b] NLCO 1973. Values from original analytical data sheets; reported as *disintegrations per minute per milliliter, dpm per mL.*

[c] Calculated quantities using same methods for U quantities, assuming effluent flow of 620,000 gallons per day to river via Manhole 175, see Figure L–3.

Table L1-33. Concentration of ^{226}Ra and ^{228}Ra Measured at Manhole 175 and Calculated Activities to the River in 1974

1974	^{226}Ra			^{228}Ra		
Date [a]	dpm mL^{-1} [b]	pCi L^{-1}	uCi [c]	dpm mL^{-1} [b]	pCi L^{-1}	uCi [c]
13-Jan	0.04	18	590	0.01	5	150
27-Jan	0.04	18	590	0.01	5	150
10-Feb	0.02	9	300	0.01	5	150
24-Feb	0.04	18	590	0.02	9	300
10-Mar	0.04	18	590	0.03	14	440
28-Mar	0.04	18	590	0.06	27	890
14-Apr	0.02	9	300	0.04	18	590
28-Apr	0.02	9	300	0.02	9	300
12-May	0.02	9	300	0.01	5	150
2-Jun	0.02	9	300	0.01	5	150
16-Jun	0.02	9	300	0.01	5	150
30-Jun	0.02	9	300	0.01	5	150
14-Jul	0.02	9	300	0.02	9	20
28-Jul	0.02	9	300	0.01	5	150
11-Aug	0.02	9	300	0.01	5	150
1-Sep	0.01	5	150	0.01	5	150
15-Sep	0.02	9	300	0.01	5	150
29-Sep	0.02	9	300	0.01	5	150
13-Oct	0.02	9	300	0.01	5	150
28-Oct	0.01	5	150	0.01	5	150
11-Nov	0.03	14	440	0.04	18	590
25-Nov	0.01	5	150	0.02	9	300
9-Dec	0.01	5	150	0.02	9	300
30-Dec	0.02	9	300	0.02	9	300
Average	0.02	10	340	0.02	8	260
StdDev	0.01	5	150	0.01	6	190
Max	0.04	18	590	0.06	27	890
Min	0.01	5	150	0.01	5	150

[a] Beginning date of sampling period for two-week composite. Average, Max and Min values represent a two-week period.

[b] NLCO 1974, original analytical data sheets; reported as *disintegrations per minute per milliliter, dpm per mL.*

[c] Calculated quantities using same methods for U quantities, assuming effluent flow of 620,000 gallons per day to river via Manhole 175, see Figure L-3.

Table L1-34. Quantity of Thorium (mg L^{-1}) Measured at Manhole 175 in 1956[a]

Date	Th	Date	Th	Date	Th	Date	Th	Date	Th	Date	Th
18-Apr	0.4	9-Jun	0.22	21-Jul	0.82	3-Sep	0.54	15-Oct	0.06	26-Nov	2.82
19-Apr	0.4	10-Jun	0.52	22-Jul	0.76	4-Sep	0.1	16-Oct	0.23	27-Nov	0.87
20-Apr	0.45	11-Jun	0.51	23-Jul	0.64	5-Sep	0.14	17-Oct	0.76	28-Nov	2.06
21-Apr	0.46	12-Jun	0.48	24-Jul	0.5	6-Sep	0.19	18-Oct	0.97	29-Nov	0.34
22-Apr	0.27	13-Jun	0.23	25-Jul	0.38	7-Sep	nd	19-Oct	0.27	30-Nov	1.94
23-Apr	0.32	14-Jun	0.19	26-Jul	0.4	8-Sep	nd	20-Oct	0.61	1-Dec	1.58
24-Apr	0.34	15-Jun	0.58	27-Jul	nd	9-Sep	0.008	21-Oct	1.2	2-Dec	0.8
25-Apr	0.24	16-Jun	0.34	28-Jul	nd	10-Sep	nd	22-Oct	0.11	3-Dec	1.07
26-Apr	0.3	17-Jun	0.27	29-Jul	0.16	11-Sep	0.003	23-Oct	0.34	4-Dec	nd
27-Apr	0.64	18-Jun	0.5	30-Jul	0.61	12-Sep	0.61	24-Oct	0.4	5-Dec	0.02
28-Apr	0.32	19-Jun	0.66	31-Jul	0.28	13-Sep	1.16	25-Oct	nd	6-Dec	0.36
29-Apr	0.34	20-Jun	0.1	2-Aug	0.21	14-Sep	0.36	26-Oct	0.15	7-Dec	0.12
30-Apr	0.52	21-Jun	0.32	3-Aug	0.74	15-Sep	0.32	27-Oct	0.34	8-Dec	nd
1-May	0.18	22-Jun	0.52	4-Aug	0.58	16-Sep	0.94	28-Oct	0.47	9-Dec	1.03
5-May	0.23	23-Jun	0.42	5-Aug	0.3	17-Sep	0.66	29-Oct	0.54	10-Dec	1.03
6-May	0.42	24-Jun	0.72	6-Aug	0.34	18-Sep	0.2	30-Oct	0.1	11-Dec	0.74
7-May	0.32	25-Jun	0.79	7-Aug	0.51	19-Sep	0.59	31-Oct	0.1	12-Dec	nd
8-May	0.15	26-Jun	0.51	8-Aug	0.2	20-Sep	0.85	1-Nov	0.85	13-Dec	nd
9-May	0.44	27-Jun	0.11	9-Aug	0.5	21-Sep	1.6	2-Nov	0.49	14-Dec	0.85
10-May	nd	28-Jun	0.74	10-Aug	0.22	22-Sep	0.46	3-Nov	0.71	15-Dec	5.72
11-May	0.27	29-Jun	1.3	11-Aug	0.19	23-Sep	0.54	4-Nov	0.79	16-Dec	4.12
12-May	0.47	30-Jun	0.51	12-Aug	0.25	24-Sep	0.2	5-Nov	0.88	17-Dec	4.76
13-May	0.3	1-Jul	0.9	13-Aug	0.4	25-Sep	0.67	6-Nov	0.61	18-Dec	1.8
14-May	0.64	2-Jul	0.61	14-Aug	nd	26-Sep	0.74	7-Nov	1.1	19-Dec	3.66
15-May	0.72	3-Jul	1.1	15-Aug	0.09	27-Sep	1	8-Nov	0.79	20-Dec	3.8
16-May	0.24	4-Jul	0.64	16-Aug	0.2	28-Sep	0.7	9-Nov	0.29	21-Dec	2.92
17-May	0.04	5-Jul	0.72	17-Aug	nd	29-Sep	1.15	10-Nov	0.84	22-Dec	1.92
18-May	0.38	6-Jul	0.47	18-Aug	0.27	30-Sep	0.54	11-Nov	0.59	23-Dec	1.8
19-May	0.48	7-Jul	0.12	19-Aug	0.51	1-Oct	0.49	12-Nov	0.25	24-Dec	1.54
20-May	0.58	8-Jul	0.34	20-Aug	0.09	2-Oct	0.14	13-Nov	nd	25-Dec	1.09
21-May	1.06	9-Jul	0.4	21-Aug	0.4	3-Oct	0.49	14-Nov	1.6	26-Dec	0.8
22-May	0.92	10-Jul	0.9	22-Aug	0.09	4-Oct	0.58	15-Nov	0.71	27-Dec	0.91
23-May	0.84	11-Jul	1.4	23-Aug	0.1	5-Oct	0.38	16-Nov	1.3	28-Dec	1.92
24-May	0.48	12-Jul	0.27	24-Aug	0.44	6-Oct	nd	17-Nov	1.28	29-Dec	2.18
25-May	0.38	13-Jul	0.36	25-Aug	nd	7-Oct	0.016	18-Nov	1.32	30-Dec	2.51
26-May	0.14	14-Jul	0.76	26-Aug	0.42	8-Oct	nd	19-Nov	0.61		
27-May	0.24	15-Jul	0.2	27-Aug	0.36	9-Oct	0.36	20-Nov	1.56		
28-May	0.6	16-Jul	0.64	28-Aug	nd	10-Oct	0.27	21-Nov	0.56		
29-May	0.34	17-Jul	0.44	29-Aug	0.18	11-Oct	0.31	22-Nov	0.71	Avg	0.70
30-May	0.56	18-Jul	0.36	30-Aug	0.2	12-Oct	0.1	23-Nov	2.22	StdDev	0.77
7-Jun	0.74	19-Jul	0.31	31-Aug	0.1	13-Oct	0.57	24-Nov	0.81	Max	5.7
8-Jun	0.34	20-Jul	1.14	1-Sep	0.2	14-Oct	0.32	25-Nov	3.3	Min	0.003

[a] From NLCO 1956.
nd none detected

Table L1-35. Quantity of Thorium (mg L^{-1}) Measured at Manhole 175 in 1957[a]

Date	Thorium	Date	Thorium
1-Jan	2.75	3-Feb	0.66
2-Jan	2.07	4-Feb	0.3
3-Jan	3.08	5-Feb	0.56
4-Jan	2.46	6-Feb	0.42
5-Jan	2.73	7-Feb	0.7
6-Jan	2.28	8-Feb	0.74
7-Jan	1.48	9-Feb	0.84
8-Jan	2.28	10-Feb	0.26
9-Jan	1.76	11-Feb	1.76
10-Jan	1.12	12-Feb	1.22
11-Jan	1.42	13-Feb	1.65
12-Jan	2.6	14-Feb	1.34
13-Jan	1.96	15-Feb	0.88
14-Jan	0.48	16-Feb	0.18
15-Jan	1.24	17-Feb	1.16
16-Jan	0.62	18-Feb	1.02
17-Jan	0.26	19-Feb	0.34
18-Jan	0.28	20-Feb	0.24
19-Jan	0.14	21-Feb	1.78
20-Jan	0.16	22-Feb	0.02
21-Jan	0.14	23-Feb	0.07
22-Jan	0.48	24-Feb	0.21
23-Jan	0.06	25-Feb	0.95
24-Jan	0.2	26-Feb	0.28
25-Jan	0.14	27-Feb	0.21
26-Jan	0.16	28-Feb	nd
27-Jan	0.42	1-Mar	0.12
28-Jan	0.42		
29-Jan	1		
30-Jan	0.88	Average	0.60
31-Jan	0.7	StdDev	0.42
1-Feb	0.88	Max	1.78
2-Feb	0.86	Min	0.02

[a] From NLCO 1957.
nd - none detected

Table L1–36. Annual Effluent Volume Discharged to the Great Miami River and to Paddy's Run [a]

Year	Effluent Volume (million gallons)	
	Great Miami River	Paddy's Run[b]
1959	371	26
1960	397	28
1961	465	41
1962	343	60
1963	303	70
1964	330	78
1965	261	66
1966	299	87
1967	313	35
1968	324	22
1969	274	36
1970	270	28
1971	249	24
1972	189	31
1973	242	17
1974	212	34
1975	221	19
1976	183	10
1977	169	11
1978	180	12
1979	199	17
1980	149	5
1981	163	2
1982	179	11
1983	172	14
1984	213	15

[a] From Cuthbert 1960–1961, Flowers 1959–1962, Marshall 1963, Rathgens 1974, Ross 1958, Shwan 1967–1983, Starkey 1958b, Starkey 1960–1961, WMCO 1989.
[b] Through the Storm Sewer Outfall Ditch.

APPENDIX M

GROUNDWATER CONTAMINATION OUTSIDE THE FMPC

INTRODUCTION

Byrne et al. (1991) provides a brief history of the measurement of offsite uranium contamination in groundwater around the FMPC. Sampling by the State of Ohio in late 1981 indicated elevated levels of gross beta radioactivity in three wells south of the FMPC. Subsequent sampling by the FMPC showed that the activity was due to naturally-occurring ^{40}K, and thus not associated with the FMPC. However, the FMPC sampling showed significantly elevated concentrations of uranium in other wells near the site. Because of the elevated uranium concentrations, the FMPC groundwater monitoring program was expanded in 1982 to include many private wells around the site. Since then the private well monitoring program has continued, with frequent expansions to include other wells.

The significant offsite uranium contamination in groundwater is south of the site, and is now called the "South Plume." Uranium concentrations in wells in the South Plume remain elevated. There are additional known areas of groundwater contamination on the FMPC site, but only the South Plume area extends outside the site boundary at this time (Byrne et al. 1991; FERMCO 1993). Since this dose reconstruction project is concerned with past doses to people around the site, the groundwater contamination to be considered in this Project is limited to the South Plume. Figure M–1 shows the estimated areal extent of the South Plume uranium contamination as of the end of 1991, as well as the locations of the private wells monitored (discussed later). The area of the South Plume has been estimated by the FMPC (Schwarzman 1992b), based on monitoring results from the private wells and from other monitoring wells, not shown in Figure M–1. The area shown includes the area where uranium concentrations are estimated to be greater than 20 $\mu g\ L^{-1}$ (or about 13.5 pCi L^{-1}). Of the private wells monitored by the FMPC, only three, numbers 12, 15, and 17, have shown uranium concentrations above the range of background (Fleming and Ross 1983; Fleming and Ross 1984; Facemire et al. 1985; Aas et al. 1986; WMCO 1987; WMCO 1988; WMCO 1989; Dugan et al. 1990; Byrne et al. 1991). Although well 26 is within the area of groundwater contamination, it is screened deeper in the aquifer, and the uranium concentrations are at background levels.

In our report of Task 4 of this Project (Killough et al. 1993), we concluded that because of the limited area of the South Plume, only a small number of people would have potentially received radiation doses from contaminated groundwater. Toward the main objective of this Project, the determination of the feasibility of an epidemiological study, doses to these people would be less important than doses through other pathways. For this reason, we further concluded that a detailed assessment of the groundwater transport of radionuclides and detailed assessments of doses to individuals potentially exposed through groundwater pathways, are not warranted. For other project objectives, it is still important to estimate potential doses through the groundwater pathway, so instead we use simple

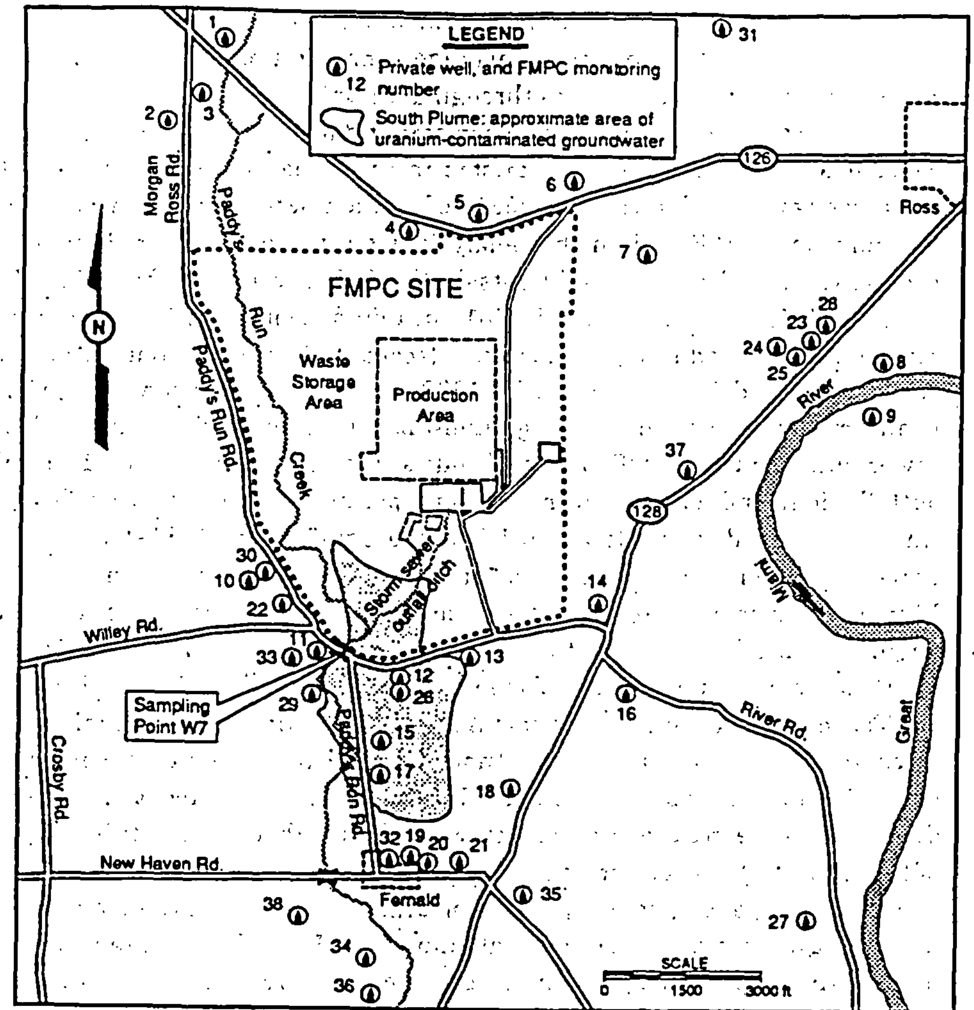

Figure M-1. Approximate area of uranium-contaminated groundwater in the South Plume, as of the end of 1991, as estimated by the FMPC (Schwarzman 1992b). The area shown includes the area where uranium concentrations are estimated to be greater than 20 µg L^{-1} (or about 13.5 pCi L^{-1}). Locations of the private wells around the FMPC sampled in the FMPC routine monitoring program are also shown, based on annual environmental monitoring reports (Fleming and Ross 1983; Fleming and Ross 1984; Facemire et al. 1985; Aas et al. 1986; WMCO 1987; WMCO 1988; WMCO 1989; Dugan et al. 1990; Byrne et al. 1991). Although well 26 is within the area of groundwater contamination, it is screened deeper in the aquifer, and the uranium concentrations are at background levels. Sampling point W7 is a location for sampling the surface water in Paddy's Run, at the Willey Road bridge.

methods to estimate concentrations of uranium in the three contaminated wells. For the small group of people exposed to the contaminated groundwater, doses will be calculated later, in Task 6 of this Project.

For those years for which groundwater uranium monitoring data are available, the measured concentrations in private wells around the FMPC will be used directly in exposure assessments. Measurements have been obtained for late 1981 through 1992, and will be used to estimate annual average concentrations for 1982–1988 (the scope of this Project includes assessing doses through 1988 only).

For years when groundwater monitoring data are not available, we estimate uranium concentrations that might have existed in the three contaminated wells. In this Appendix, we investigate the lateral movement of contaminated groundwater to offsite locations of groundwater use. Based on that analysis, we conclude that uranium contamination in the groundwater had not migrated outside the FMPC boundary by 1962. However, sometime between 1962 and the end of 1981, uranium contamination in the South Plume had migrated offsite and into private wells. Recent studies of the groundwater around the FMPC site (Dames and Moore 1985; DOE 1990a) have concluded that the primary source of the uranium contamination in the groundwater is uranium in waters released to the storm sewer outfall ditch (SSOD) and to Paddy's Run (see Figure M–1). The soils in parts of the outfall ditch and Paddy's Run are very permeable, and apparently allow contaminated water to move directly downward into the aquifer. For 1963–1981, we first calculate an upper bound on uranium concentrations in the three contaminated wells in the South Plume. For a more realistic assessment, uranium concentrations are calculated using an empirical model, that uses the available measured concentrations in the wells and the quantities of uranium released from the FMPC into Paddy's Run and the SSOD.

There are three annexes to this Appendix. The first includes tables of data that support calculations described in the text. Annex 2 includes information about quantities of uranium discarded to the waste pits on the FMPC. Information about the transport of uranium deposited on the ground surface may be useful to other parts of this Project. A special study of this transport was performed for this Project, and is summarized in Annex 3.

In this Appendix, concentrations of uranium in water are generally presented as total uranium concentrations using activity units (pCi L^{-1}). However, in some cases we retain the units of the original information source. To convert from activity units to mass units (or vice versa), the specific activity of natural uranium has been assumed to apply. The value of 6.75×10^{-7} Ci g^{-1} (Rich et al. 1988) has been used.

POTENTIAL SOURCES OF GROUNDWATER CONTAMINATION FROM FMPC

The status of groundwater contamination in the vicinity of the FMPC has been investigated (Dames and Moore 1985; Solow and Phoenix 1987; DOE 1990a; WMCO 1990). These studies indicate there are two potential sources of groundwater contamination originating on the FMPC site (see Figure M–1): (1) historical releases of uranium-contaminated water to Paddy's Run and to the Storm Sewer Outfall Ditch (SSOD), and (2) possible releases from the solid and liquid waste pits in the waste storage area.

Paddy's Run is a small stream which generally flows only during January to May (Pennak 1973). The flow rate ranges from 0.2 to 4.0 $ft^3\ s^{-1}$. For the balance of the year it may be considered a dry bed stream with occasional flash flows of a few hours duration following heavy rains. The bottom sediments of Paddy's Run and the SSOD are very permeable in the area north and west of the South Plume, so these areas are recharge areas for the regional aquifer (DOE 1990a). Thus, uranium contamination in Paddy's Run and the SSOD percolates downward through the permeable sediments to ultimately reach the groundwater. As discussed in Appendix L of this report, releases of uranium directly to Paddy's Run were due to storm water runoff across contaminated ground on the FMPC, primarily areas on the western part of the site. Releases to the SSOD were primarily from overflows of the site storm sewer system, when heavy rains exceeded the storm sewer lift station capacity.

Of the potential sources, the principal source of uranium contamination in the South Plume has been determined to be the historical releases to Paddy's Run and the SSOD (DOE 1990a). The waste pits probably have not been significant direct contributors to the uranium contamination outside the site boundary. Runoff from contamination in and around the waste pits, however, probably contributed to releases to Paddy's Run. While it appears the waste pits are not of great concern relative to the groundwater contamination, Annex 2 of this Appendix provides some information about materials disposed of in the waste pits. Additional discussion regarding the waste pits can be found in Appendix K.

RADIONUCLIDES OF CONCERN IN THE SOUTH PLUME GROUNDWATER

The radionuclide of primary concern in the South Plume has been uranium (see, for example, DOE 1990a; Dames and Moore 1985; Byrne et al. 1991). However, other radionuclides were also released into surface waters, so the potential exists for other radionuclides to be present in the South Plume groundwater. Appendix D of this report discusses releases of radionuclides other than uranium from the FMPC to air and to surface waters. In Appendix D we performed a screening-level assessment of the relative importance of radionuclide releases to surface waters, based primarily on information about releases to the Great Miami River. From this assessment, the radium isotopes ^{226}Ra and ^{228}Ra were determined to be of primary importance (see Appendix D).

However, this determination only applies to radionuclides in surface waters. For radionuclides in groundwater, the environmental transport is different, and different exposure pathways may be pertinent. Since large quantities of these two radium isotopes were released to surface waters, they would be of concern in the groundwater if they migrated into the South Plume along with the uranium.

From recent environmental monitoring reports for the FMPC, it appears that the private, offsite wells have not been routinely monitored for radionuclides other than uranium (Dugan et al. 1990; Byrne et al. 1991; WEMCO 1992; FERMCO 1993). In the 1989 report, radionuclides other than uranium were not discussed (Dugan et al. 1990). The 1990 report summarized results from the comprehensive groundwater program (which involves wells in addition to the routine monitoring wells), which monitored for ^{90}Sr, ^{99}Tc, ^{226}Ra, ^{228}Ra, and ^{232}Th (Byrne et al. 1991). This report only provided those results that were above the Department of Energy (DOE) concentration guidelines for drinking water. For these other radionuclides, none of the wells in the South Plume area exceeded the guidelines. In the 1991 and 1992 reports, no results for other radionuclides were presented, due to problems with data validation (WEMCO 1992; FERMCO 1993).

As part of the Remedial Investigation/Feasibility Study (RI/FS) of the environmental restoration work at the FMPC, a study of the South Plume groundwater included installation of additional groundwater monitoring wells, followed by sampling and analysis of these new wells and the existing monitoring wells in the late 1980s and 1990 (DOE 1990a). The radionuclide analyses of the well samples included total uranium, total thorium, isotopic uranium, isotopic thorium, isotopic plutonium, ^{226}Ra, ^{228}Ra, ^{237}Np, ^{99}Tc, ^{90}Sr, and gamma-emitting radionuclides by gamma spectroscopy. Except for uranium, none of these radionuclides were found at concentrations above natural background (DOE 1990a).

Another report prepared for the RI/FS work included a comprehensive compilation of groundwater contamination monitoring data from August 1987 through April 1990, as well as descriptions of the nature and extent of groundwater contamination at the FMPC (DOE 1990b). Some results from that report are summarized here. Outside the FMPC boundary, the only recurrent detections of radionuclides other than uranium were for ^{226}Ra and ^{228}Ra, both at a monitoring well (not one of the private wells) near the town of Fernald, south of New Haven Road (see Figure M-1 for general location). These measured concentrations were low and were spatially isolated. It was concluded that the presence of radium in this well was probably not due to the South Plume. No other radionuclides (other than uranium and radium) were recurrently detected in the wells of the South Plume area. Thus, for all radionuclides other than uranium, it was concluded that the FMPC was not contributing significant quantities to the South Plume.

Based on these studies of other radionuclides, there are no indications that other radionuclides are present in offsite groundwater in concentrations above background. It appears that radium and other radionuclides have not migrated into the South Plume groundwater in significant quantities. We note that the sampling for other radionuclides was limited to a few, recent years, with no results for private wells. We conclude that uranium is the primary radionuclide of concern in the South Plume groundwater. Thus, uranium is the only radionuclide considered in the remainder of this Appendix.

PRELIMINARY INVESTIGATION OF LATERAL MOVEMENT OF CONTAMINATED GROUNDWATER TO OFFSITE LOCATIONS

The migration of contamination in the aquifer to offsite locations is a critical factor in the assessment of potential doses from groundwater. The time of arrival of the contaminated plume at wells, primarily located south of the FMPC, determines the time of first potential exposure of individuals using well water for various purposes. A special study, that is summarized below, was conducted to determine whether the contaminated water had migrated to offsite locations from the FMPC prior to 1962 (Ichimura 1991a). That date was chosen because the period of interest for the preliminary report of Tasks 2 and 3 of this Project was 1960–1962 (Voillequé et al. 1991).

The purpose of the analysis was to estimate the time of possible exposure to contaminated groundwater using published information. To accomplish this objective, it was necessary to estimate horizontal movement of the plume and its position relative to the southern boundary of the FMPC. We use "groundwater velocity" to mean the horizontal rate of movement of the groundwater, and "solute velocity" or "plume velocity" for the effective rate of movement of the contaminant (or the contaminant plume front) in the aquifer.

The published information used for this analysis of groundwater movement comes from reports by Advanced Sciences, Inc., and International Technology Corp. (ASI-IT 1990), and GeoTrans (1985). These reports assimilate large quantities of geologic, hydrologic, chemical, and source term data to construct a model of the south groundwater plume.

Analysis of the ASI-IT Model

Groundwater travel time estimates can be based on the provisionally calibrated groundwater flow and transport model described in the report "Engineering Evaluation/Cost Analysis South Plume" that was prepared by ASI-IT (1990). Appendix A of that report describes a three-dimensional solute transport model. Results of calculations made using this model show the projected plume would carry uranium beneath New Haven Road near Highway 128 and eventually to the Great Miami River just upstream of the confluence with Paddy's Run. The velocity of groundwater along the plume is about 1,300 ft y^{-1}.

At the present time (actually, for 1989), plume geometry, as calculated by the computer model, is an elongated ellipse oriented in the northwest-southeast direction (Figure M–2). Analytical data from the Remedial Investigation/Feasibility Study (RI/FS) program, available as of September 15, 1989, were utilized for the evaluation of the South Plume by ASI-IT (1990). This orientation is due to the channeling of groundwater through a narrow north-south buried aquifer. Areas of maximum predicted uranium concentrations, for 1989, are located approximately 800 ft south of Willey Road. The maximum uranium concentration predicted by the model is 600 $\mu g\ L^{-1}$. The area occupied by the plume exceeding 30 $\mu g\ L^{-1}$ is approximately 100 acres.

The northern boundary of the plume is defined by a curved line, through the storm sewer outfall ditch and Paddy's Run, in which uranium loading occurs. The groundwater recharge rate along this boundary is 32 in y^{-1}. The loading concentration is variable and unspecified in the ASI-IT (1990) report.

Another parameter assumed in the model is a distribution coefficient of 0.016 $ft^3\ lb^{-1}$ (1.0 mL g^{-1}). According to the ASI-IT report, the corresponding retardation factor is 9. The best model calibration for a retardation factor of 9 was attained using a longitudinal dispersivity of 50 ft and a transverse dispersivity of 1 ft.

According to the ASI-IT report, the model of the South Plume gives an approximate plume length of 4,200 ft at 1990 conditions. Furthermore, the groundwater solute transport model indicates that the horizontal plume velocity is 220 ft y^{-1}. Therefore, the estimated horizontal spread of the plume has occurred for the time: (4,200 ft)/(220 ft y^{-1}) = 19 y. This number means that the plume would have begun migrating laterally away from the point where the contaminants entered the aquifer roughly 19 years ago. Assuming that current conditions refer to January, 1990, the initial entry of uranium into the sand and gravel aquifer must have occurred by 1971. Prior to that time, the contaminants would have been moving downward from the surface to the aquifer. According to this model, no contaminants would have reached the aquifer or been transported laterally away from the point of entry in the early sixties.

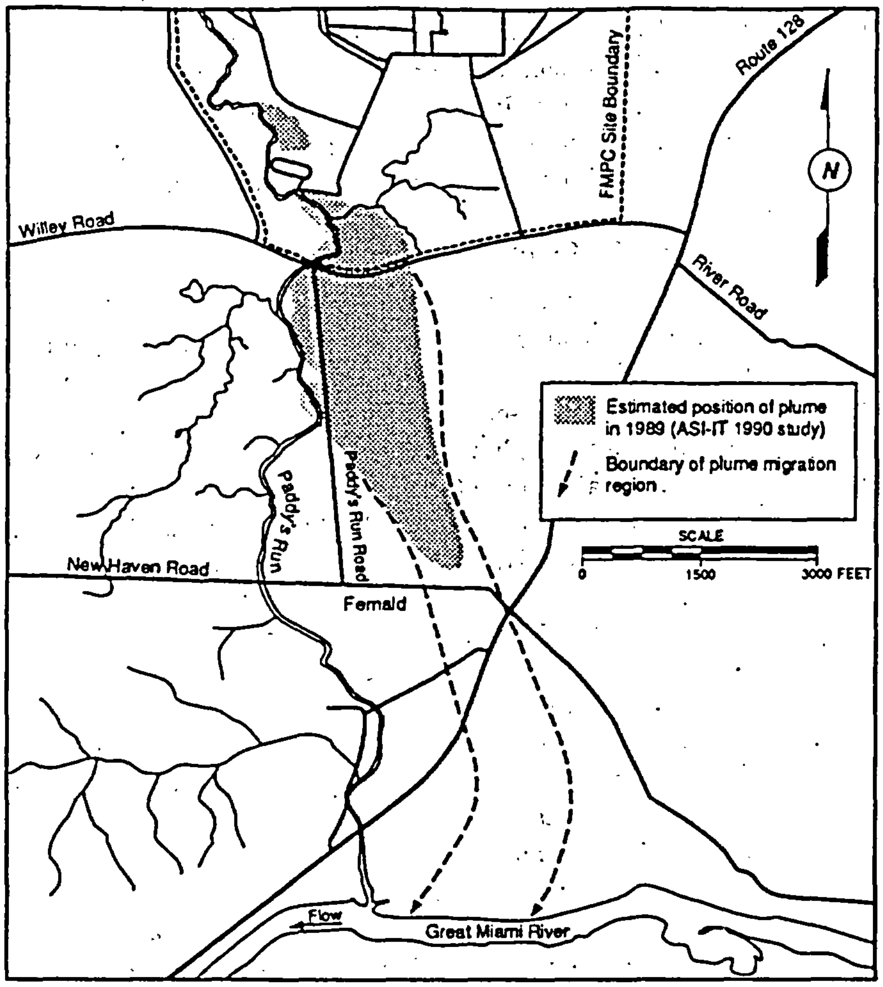

Figure M-2. Model-calculated estimate of the extent of the groundwater plume as of 1989. The boundary shown represents a calculated concentration of 30 $\mu g\ L^{-1}$ (about 20 pCi L^{-1}). Redrawn from ASI-IT (1990).

The foregoing analysis is based on information reported for the ASI-IT model of the South Plume. According to the ASI-IT report, the groundwater flow model is considered to be well calibrated because the model has successfully reproduced flow conditions throughout the South Plume area. However, the calibration of the solute transport is considered to be provisional because of uncertainty with calibration of retardation and dispersivity. Decreasing the retardation factor will increase the rate of movement of the plume. Similarly, increasing the dispersivity will increase the rate of plume spreading.

To estimate the plume boundary using the solute transport model, ASI-IT had to estimate the contaminant source term loading rates. Contaminant loading time periods and source areas were fixed. However, the time rate of contaminant release was varied as part of the model calibration and was originally derived from literature reviews. Finally, ASI-IT did not provide the source term data in its report.

The model produces a horizontal projection of the South Plume. Because the distance traveled by the plume is measured in the horizontal plane, the travel time analysis does not include time required for transport in the vertical direction. This component of the total travel time can be significant because low permeability materials underlie areas around the FMPC. Therefore, only a minimum travel time can be determined using this analysis.

GeoTrans Groundwater Modeling Analysis

Other estimates of groundwater velocities were reported as part of an earlier groundwater modeling study (GeoTrans 1985). In that study, the reported groundwater velocity was 800 ft y^{-1}. Assuming that the plume velocity has the same relationship to the groundwater velocity as in the ASI-IT (1990) report, the velocity of the plume is estimated to be (800/1300)×(220), or about 135 ft y^{-1}. Using this estimate for the plume velocity and the current plume length of 4200 ft, the initial release of uranium to the aquifer is estimated to have occurred 31 years before 1990, or prior to 1959.

Groundwater particle tracing studies conducted by GeoTrans (1985) showed a groundwater travel time of 3.4 years to the FMPC southern boundary. The uranium migration rate is expected to be less than that of the groundwater because of retardation effects. If the GeoTrans estimate is accurate, the South Plume would have been about 400 ft in length, or just at the southern border of the FMPC at year 1962. Therefore, it is unlikely that the significant part of the South Plume was offsite and impacting wells in 1962 based on this model.

Conclusions About Lateral Movement of Groundwater Contamination

Because the recent ASI-IT (1990) study uses more current data and additional model refinements, its results should be more reliable and are preferred for that reason. The width of the aquifer varies near the FMPC, and consequently the groundwater flow velocity also varies. The ASI-IT study shows the slower horizontal flow near the FMPC (the aquifer is relatively wide there), in the recharge area, and the faster flow near the village of Fernald, south of the site (the aquifer narrows between two outcrops of bedrock).

Based on results provided by ASI-IT (1990), the estimated initial release of uranium into the sand and gravel aquifer occurred by 1971. However, earlier more conservative estimates, based on a report by GeoTrans (1985) indicate that the horizontal South Plume development occurred by 1959 and may have barely reached the southern border of the FMPC in 1962. These preliminary estimates indicate that the plume of uranium contaminated water did not impact offsite groundwater users until after 1962.

URANIUM CONCENTRATIONS IN PRIVATE WELL WATER FOR 1982–1988

As discussed in our Task 5 Report (Shleien et al. 1993), the most important program for monitoring uranium concentrations in private well water around the FMPC has been the FMPC monitoring program. The FMPC began its routine monitoring of private wells around the site in early 1982 (Byrne et al. 1991), although results were not reported in the annual environmental report for 1982 (Fleming and Ross 1983). Since the wells sampled were not under the control of the FMPC, inclusion in the program was based on the well owner's request. Samples were generally taken on a monthly frequency, although a few of the wells were sampled less frequently. The annual environmental reports for 1983–1990 (Fleming and Ross 1983; Fleming and Ross 1984; Facemire et al. 1985; Aas et al. 1986; WMCO 1987; WMCO 1988; WMCO 1989; Dugan et al. 1990; Byrne et al. 1991) generally provide the minimum, maximum, and annual average uranium concentrations for each well in the monitoring program.

The locations of the private wells sampled in the FMPC program are shown in Figure M–1. The well locations were obtained from the annual environmental reports and from a detailed drawing obtained from the FMPC (Schwarzman 1992a). In our Task 5 Report (Shleien et al. 1993), we presented annual average concentrations for wells 1 through 38 for years 1983 through 1990, obtained from the annual environmental monitoring reports. We also presented monthly measurement results for the three contaminated wells, wells 12, 15, and 17 for November 1981 through February 1985, obtained from a compilation by the groundwater study of Dames and Moore (1985).

Also in our Task 5 Report, we examined the annual average concentrations for the 1983–1990 period to estimate a range of background concentrations. We concluded that a reasonable estimate of the range of long-term average, background concentrations of uranium in private well water around the FMPC, for individual wells, is 0.09 to 1.3 pCi L^{-1} (total uranium). This range compares well with background concentrations estimated by the FMPC (Byrne et al. 1991) to be 0.068–2.2 pCi L^{-1}. These background concentrations can be used for comparisons to the concentrations in the contaminated wells.

Additional, detailed results of the FMPC monitoring have been obtained. Kraps (1993) provides results of the monthly results for the private well sampling, for 1984 through 1992. The monthly results for the three contaminated wells, wells 12, 15, and 17, are compiled in Table M1–1, in Annex 1 of this Appendix. In Table M1–1 we also repeat the monthly results for these three wells for 1981 through 1983, from a compilation in Dames and Moore (1985).

From the monthly concentrations, annual average concentrations for 1982–1992 for the three contaminated wells are calculated, with the results shown in Table M–1. Here we convert the mass concentrations to radioactivity concentrations, with units of pCi L^{-1}. Since only one or two results were available in 1981 for these wells, we do not calculate annual averages for 1981. The annual average concentrations are also plotted in Figure M–3. These annual average concentrations, for 1982–1988, will be used as the basis of dosimetry calculations for these years, in Task 6 of this Project (the scope of this Project includes calculating doses through only 1988).

ESTIMATED UPPER BOUND ON URANIUM CONCENTRATIONS IN PRIVATE WELL WATER IN THE SOUTH PLUME FOR 1963–1981

In this section we describe preliminary calculations, to develop an upper bound estimate of the concentrations of uranium that might have existed for 1963–1981 in the South Plume groundwater, prior to the start of monitoring.

Table M–1. Annual Average Measured Concentrations of Uranium (pCi L^{-1}) in the Three Contaminated Wells

Year	Well 12	Well 15	Well 17
1982	170	320	45
1983	180	290	39
1984	170	220	36
1985	140	200	31
1986	150	190	31
1987	200	200	40
1988	170	190	38
1989	170	190	27
1990	130	180	30
1991	100	170	27
1992	100	150	25

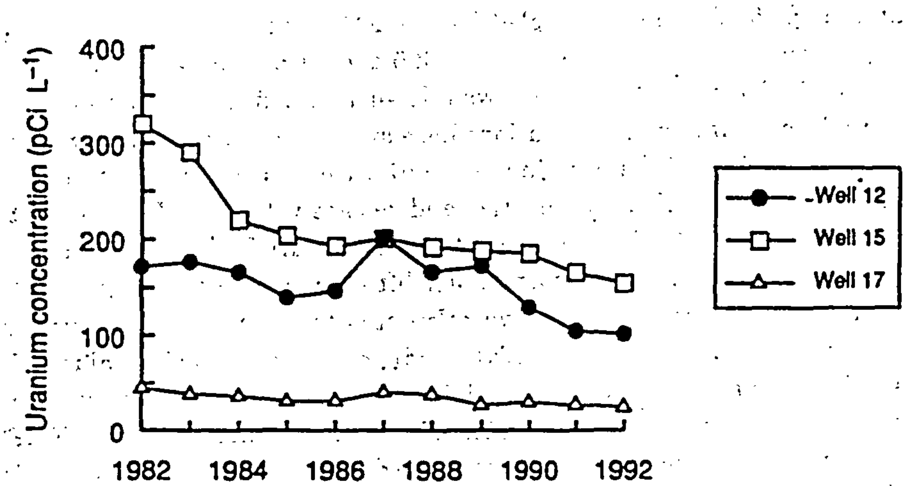

Figure M–3. Annual average concentrations of uranium in well water for the three contaminated private wells, for 1982 through 1992.

Basis for Upper Bound Estimate

As discussed earlier in this Appendix, it has been determined that the principal source of uranium contamination in the South Plume is the historical releases to Paddy's Run and the SSOD. Because of the finite velocity of the uranium plume in the groundwater, there would be a time lag between the release of uranium to Paddy's Run and the SSOD, and the appearance of the contamination in downgradient wells in the aquifer, such as wells 12, 15, and 17 in the South Plume. Uranium concentrations at these downgradient wells due to surface water releases from a particular time should not exceed concentrations measured in surface water runoff from Paddy's Run and the SSOD. Therefore, we use surface water concentrations as an upper bound on uranium concentrations that might have existed in wells 12, 15, and 17 for the period 1963–1981.

This use of surface water concentrations to bound groundwater concentrations is applicable for the following reasons. Sources of groundwater that eventually flow into the South Plume area of the aquifer include groundwater flowing from the west and the north, in two branches of the same aquifer, water from bedrock, and recharge sources (DOE 1990a). Recharge sources in the area include precipitation recharge and recharge by stream infiltration through Paddy's Run, the SSOD, and other streams (DOE 1990a). Surface waters which recharge from Paddy's Run and the SSOD will be diluted by groundwater from the other sources. Therefore, groundwater concentrations of uranium could be as high as surface water concentrations only when a large quantity of recharge from infiltration in Paddy's Run and the SSOD occur, and when there is no mixing with existing groundwater from other sources.

Applicable Uranium Concentration Data for Paddy's Run and the SSOD

In our Task 5 Report (Shleien et al. 1993), we compiled measured concentrations of uranium in Paddy's Run, sampled by the FMPC at sampling point W7, the Willey Road bridge (Figure M-1). Results for 1955–1965 and 1975–1991 were obtained, and are shown in Table M-2. Because this location was downstream from the confluence of the SSOD and Paddy's Run, uranium concentrations here are diluted by clean water from north of the site.

Concentrations of uranium in the SSOD are obtained both from measurements and from estimates based on release quantities and release volumes. Measurement results for 1954–1957, 1960–1964, and 1966 are presented in Appendix L. These measurements were made at the overflow outfall from the storm sewer lift station. When sufficient measurement data for effluent volumes were available, we calculated the annual average uranium concentration as the volume-weighted average of the individual concentration values. For other cases, an unweighted average was used. Additional measurement results, for 1975–1984, are compiled in Dames and Moore (1985). All of these results were from FMPC measurements. The annual average concentrations measured in the SSOD are shown in Table M-2.

In Appendix L of this report, estimates of the quantities of uranium released to Paddy's Run were developed. The majority of uranium released was to the SSOD, with a smaller quantity going directly into Paddy's Run. In Appendix L we also compiled data on the water volumes released in the SSOD. The uranium release quantities and release volumes can be used to estimate the concentrations released to the SSOD. These estimates will generally be biased somewhat high, since not all of the uranium released was through the SSOD. However, the estimates should be adequate for our purposes. Table M1-2 shows the release quantities; release volumes, for years for which complete data were obtained; and estimated release concentrations of uranium. The estimated release concentrations (in the SSOD) are also shown in Table M-2. For 1963, 1966, and 1982, the estimated concentrations are significantly less than the measured concentrations, by factors of about 2 to 3. These discrepancies are not readily explained, though for 1963 and 1966 the effluent volume data were incomplete, thus not allowing a volume-weighted average of the measured values.

For comparison, Table M-2 also shows the annual average uranium concentrations in well 15, from Table M-1. We use well 15 for comparison purposes because it has had the highest uranium concentrations of the contaminated wells, for the periods measured.

Table M–2. Comparison of Uranium Concentrations in Well 15 with Those in Paddy's Run and the SSOD

	Uranium concentrations in water (pCi L^{-1})			
Year	Paddy's Run at Willey Road	Measured SSOD	Estimated SSOD	Well 15
1952				
1953				
1954		230		
1955	100	290		
1956	240	580		
1957	100	970		
1958	480			
1959	780		5800	
1960	1100	6400	8300	
1961	470	4900	6100	
1962	367	5400	4500	
1963	690	4500	2300	
1964	720	3700	3900	
1965	580		1700	
1966		4900	1600	
1967			3800	
1968			2900	
1969			1400	
1970			2200	
1971			3700	
1972			1900	
1973			2400	
1974			1300	
1975	92	525	2300	
1976	160	1800	4800	
1977	20	2[illegible]0	3300	
1978	63	550	1000	
1979	11	720	880	
1980	19	500	1800	
1981	21	230	1800	
1982	12	560	320	320
1983	76	320	690	290
1984	15	420	680	220
1985	43			200
1986	49			190
1987	5.8			200
1988	7			190
1989	6.4			190
1990	6.5			180
1991	5			170
1992				150

Estimated Upper Bound Uranium Concentration

For the short length of time that monitoring of private wells has been performed, the concentrations of uranium in well 15 are seen to be generally lower than concentrations in the SSOD, but higher than concentrations in Paddy's Run at the Willey Road bridge (Table M–2). This relationship is consistent with (though it does not prove) waters from both Paddy's Run and the SSOD being the sources of the groundwater contamination. It seems reasonable that this relationship would also have generally existed in earlier years (before well measurements were made, starting in late 1981). However, when the plume first reached the well, concentrations in the well water may have initially been much lower.

We thus conclude that an upper bound on the annual average concentrations of uranium in the three contaminated wells for 1963–1981 is the maximum annual average concentration in the SSOD for earlier years. Based on the data in Table M–2, this upper bound is 8300 pCi L^{-1}, for releases in 1960. For this upper bound we use the estimated concentration, because the measurement data were available only for part of the year (see Appendix L), and because the estimated value was similar to the average of the available measured results.

We recognize that this upper bound on uranium concentrations that might have existed in private wells in the South Plume area for 1963–1981 is an extremely conservative (i.e. the estimated value is too high) estimate of uranium concentrations in the wells. The upper bound is conservative for at least three important reasons. First, of the measured and estimated uranium concentrations in the SSOD, for 1952–1988, we used the highest annual average value, and assumed it applied to the whole period 1963–1981. Second, the uranium that infiltrated into the groundwater came from both the SSOD and Paddy's Run. In Table M–2, uranium concentrations in the SSOD and in Paddy's Run are shown, with concentrations in Paddy's Run lower than in the SSOD. The combined source would have had uranium concentrations between those of the two sources, which would have been lower than concentrations in the SSOD. And third, dilution of the uranium infiltrating into the groundwater from the SSOD and Paddy's Run was not considered. As discussed earlier (see page M–11), sources of groundwater that eventually flow into the South Plume area of the aquifer include groundwater flowing into the area from the west and the north, in two branches of the same aquifer, water from bedrock, and recharge sources (DOE 1990a). Surface water recharge from Paddy's Run and the SSOD would be diluted by uncontaminated groundwater from the other sources, thus reducing the concentrations of uranium in the aquifer.

ESTIMATED URANIUM CONCENTRATIONS IN PRIVATE WELL WATER IN THE SOUTH PLUME FOR 1963–1988, BASED ON AN EMPIRICAL MODEL

We believe the use of the upper bound uranium concentration of 8300 pCi L^{-1}, to represent uranium concentrations in water at private wells in the South Plume for the complete period 1963–1981, is unrealistically conservative. In this section we describe a ratio model used to determine a more realistic, though still somewhat conservative, estimate of uranium concentrations in the private wells during 1963–1981.

Linear, Empirical Model with Travel Time

Historical releases of uranium from the FMPC into Paddy's Run or the SSOD did not immediately move into the offsite part of the South Plume groundwater. The uranium had to move downstream, to the area where the aquifer outcrops in the stream bottoms, infiltrate vertically through the stream bed and underlying soil to reach the aquifer, and then migrate horizontally, in the aquifer, southward into the South Plume area. We use a model to describe this movement of uranium from historical releases into the South Plume groundwater.

General model. Figure M–4 shows the steps involved in the transport of uranium from FMPC releases to wells in the South Plume. The general model we use is based on an assumption that uranium concentrations in the water at each point in the transport are directly related to concentrations (or releases quantities) at the previous point, and that this relationship is multiplicative, by a constant factor. For each of the three transport steps, this assumption seems a reasonable first approximation. The solubility of the uranium is important in determining how much uranium migrates into the aquifer. Our assumption of a constant multiplicative factor implicitly includes an assumption of a constant ratio of soluble uranium to insoluble uranium in the surface water releases, for all years of releases.

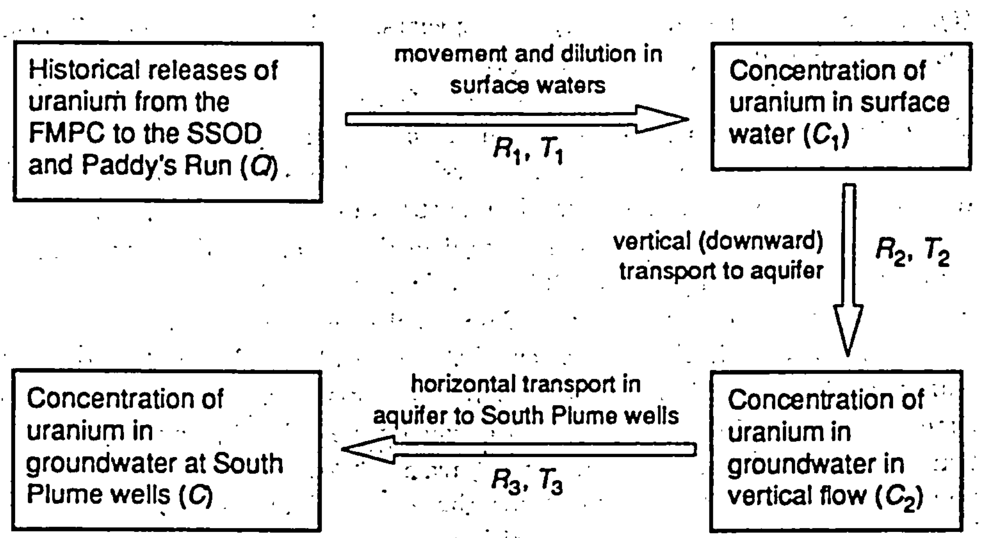

Figure M–4. Generalized description of the transport of uranium from historical FMPC releases into the SSOD and Paddy's Run to groundwater wells in the South Plume area. The arrows represent the three steps involved in the transport. The boxes show the different endpoints of each step.

Given that each step of the transport can be represented by such constant, multiplicative factors, the model is described by the following set of equations.

$$C_1 = R_1 Q$$
$$C_2 = R_2 C_1 \qquad \text{(M–1)}$$
$$C = R_3 C_2$$

where C_1 is the concentration of uranium in surface water, C_2 is the concentration in groundwater in vertical flow, C is the concentration at wells in the South Plume, Q is the annual quantity of uranium released from the FMPC to the SSOD and Paddy's Run, and R_1, R_2, and R_3 are constant ratio factors.

These equations can be rewritten as:

$$C = RQ \qquad \text{(M–2)}$$

where the overall ratio factor is

$$R = R_1 R_2 R_3 \qquad \text{(M–3)}$$

Measured uranium concentrations (C) in South Plume wells are available for some years, and quantities of uranium released from the FMPC into the SSOD and Paddy's Run (Q) have been estimated for all operating years. These data can be used to estimate the ratio factor R by calculating values of C/Q.

The general model given by equation M–2 will be developed further for application to the available data. To summarize, this model assumes that there is a linear relationship between annual quantities of uranium released (Q) and annual average concentrations of uranium in the wells in the South Plume (C), and the relationship can be represented by a constant factor R. The model is empirical in that available data for release quantities and measured well concentrations will be used to estimate the ratio factor R, which will then be used to estimate the concentrations C for which measurements do not exist.

Specific model. The discussion of the general model neglected the time required for transport of the uranium through each step. This transport time is referred to as travel time. In Figure M–4 we have shown travel times T_1, T_2, and T_3, associated with each transport step. The total travel time T, from the stream water to the groundwater in the South Plume area, is the sum of the three individual travel times. From studies of the lateral movement of uranium contamination in groundwater (summarized on page M–8), we concluded that by 1962 the uranium contamination had not extended outside the FMPC boundary. Since uranium releases to Paddy's Run and the SSOD began in 1952, the travel time for uranium to move from these surface waters to reach the nearest private wells in the South Plume area is many years. Thus, travel time is accounted for in the specific implementation of the empirical model.

The travel times for the vertical transport into the aquifer (T_2) and horizontal transport in the aquifer (T_3) are at least a few years (as discussed later). However, the travel time in surface waters (T_1) is very much shorter, because the distance from the uranium release points to the area where infiltration into the aquifer occurred is very short. We thus ignore the travel time T_1.

To account for travel time between uranium releases and arrival of contamination at wells, we use the following empirical model. Here we have added the subscript i to indicate that calculations are performed on an annual basis.

$$C_i = R_{i,T} Q_{i-T} \qquad \text{(M–4)}$$

where

C_i = estimated annual average concentration of uranium in groundwater at private wells in the South Plume area, for year i (pCi L^{-1}).

$R_{i,T}$ = ratio factor, to relate uranium release quantities to estimated concentrations in the South Plume groundwater (pCi L^{-1} per kg). The subscript T indicates that the ratio factors may be different for different travel times. For a given travel time, T, a single distribution of ratio factors will be applied to each year i. The subscript i is used because the uncertainties in the ratio factors for individual years are considered independent.

Q_{i-T} = the quantity of uranium (kg) released from the FMPC to Paddy's Run and the SSOD in year $i–T$. Here we use the uranium quantity for year $i–T$ so that the concentration for a given year (i) is based on the release T years earlier (hence $i–T$). This accounts for the travel time T.

T = the total travel time (lag time) (y) for uranium to move from surface waters of Paddy's Run and the SSOD into groundwater in the South Plume area. T is the sum of the vertical travel time T_2 and the horizontal travel time T_3.

Groundwater and solute transport modeling involve large uncertainties, some of which may not be adequately accounted for in our parameter uncertainty analysis. Because of this, and because only a small number of people were potentially exposed to the contaminated groundwater, we make some conservative assumptions for the model calculations. For estimating the travel time, we use as receptor point the offsite private well that is closest to the source of the groundwater contamination. For determining the ratio factors, we use the uranium concentrations from the contaminated well with the highest concentrations.

Parameters and Calculations

As for the majority of the source term calculations presented in this report, for these calculations of uranium concentrations in groundwater we use Monte Carlo simulations, for a concurrent parameter uncertainty analysis. The rest of this section describes the parameter distributions used and the implementation of the calculations.

Uranium released and measured concentrations in groundwater. The quantities of uranium released from the FMPC into Paddy's Run and the SSOD are calculated in Appendix L of this report. The values used here are the best estimates (medians of the distributions), from that Appendix, of the annual total mass of uranium released into Paddy's Run and the SSOD. Table M–3 shows these release quantities.

For the development of the ratio factors, $R_{i,T}$, annual average measured concentrations of uranium in the groundwater of the South Plume are also required. Table M–1 shows these concentrations, for the three contaminated wells in the South Plume area. For developing the ratios, we use data from the well with the highest concentrations, well 15.

Table M–3. Median Estimated Annual Quantities of Uranium Released from the FMPC into Paddy's Run and the SSOD

Year	Uranium released (kg)	Year	Uranium released (kg)	Year	Uranium released (kg)
1952	522	1965	622	1978	68
1953	522	1966	771	1979	84
1954	522	1967	753	1980	50
1955	300	1968	358	1981	20
1956	270	1969	290	1982	20
1957	340	1970	349	1983	54
1958	630	1971	499	1984	57
1959	840	1972	322	1985	39
1960	1300	1973	231	1986	17
1961	1400	1974	255	1987	<0.5
1962	1500	1975	245	1988	<0.5
1963	901	1976	272		
1964	1722	1977	204		

Travel time. The travel time T is estimated based on the preliminary investigation of lateral movement of contaminated groundwater to offsite locations (Ichimura 1991a), which was summarized earlier in this Appendix (see page M–5). That investigation reviewed two main studies of the groundwater around the FMPC: (1) a study by GeoTrans, Inc. (GeoTrans 1985), and (2) a study by Advanced Sciences, Inc. and International Technology, Inc. (ASI-IT 1990). The two groundwater studies include information about the plume velocity, which is the velocity at which the uranium contamination moves horizontally in the aquifer. The plume velocity can be used to help estimate travel time.

From the GeoTrans study, we estimated the uranium contamination entered the aquifer in about 1959 (see page M–8), at which time horizontal spreading of the plume would have begun. Since uranium was first released into Paddy's Run and the SSOD in 1952, the vertical travel time (time for the uranium to infiltrate into the aquifer) is estimated to be about 7 y. The location where the uranium is thought to have infiltrated from surface waters into the aquifer is Paddy's Run and the SSOD, in the area near their confluence. The distance from this area to the closest offsite well, well 12, is about 500 ft (see Figure M–1). The uranium plume velocity was estimated to be 135 ft y^{-1} (see page M–8). So, the horizontal travel time to well 12 is estimated to be (500 ft/135 ft y^{-1}) = 3.7 y.

From the ASI-IT study, we estimated that the uranium contamination entered the aquifer in about 1971 (see page M–6). Since uranium was first released into Paddy's Run and the SSOD in 1952, the vertical travel time is estimated to be about 19 y. For this study, the plume velocity was estimated to be 220 ft y^{-1}. Thus, the horizontal travel time to well 12 is estimated to be (500 ft/220 ft y^{-1}) = 2.3 y.

The plume velocities from the two studies result in different estimates of vertical and horizontal travel times to the nearest offsite well. The total travel time of uranium from surface waters of Paddy's Run and the SSOD to groundwater at well 12 is the sum of the vertical and horizontal travel times. The travel times are summarized in Table M–4.

Table M–4. Summary of Uranium Travel Time from Surface Waters of Paddy's Run and the SSOD to Well 12 in the South Plume

		Estimated travel time (y)		
Study	Plume velocity ($ft\ y^{-1}$)	Vertical to aquifer	Horizontal to well 12	Total
GeoTrans	135	7	3.7	11
ASI-IT	220	19	2.3	21

From Table M–4, estimated total travel times to well 12, based on the two studies, are 11 and 21 y. From our reviews of the two basis reports (GeoTrans 1985 and ASI-IT 1990), the uncertainties in the uranium plume velocities are large. In general terms, we expect the ASI-IT report to be of higher quality, because it was based on additional and more recent data. But, we have no strong evidence to indicate this, and no justifiable way to quantify any difference in the data quality from the two reports (GeoTrans and ASI-IT). Thus, with only two estimates of travel time, we assume that all travel times within the range 11–21 y are equally likely. Since T is a whole number, we assume that T has a discrete probability distribution over the range 11–21 y, inclusive, with each value having equal probability. (This is similar to a uniform distribution, but is discrete, rather than continuous.)

Ratio factors. We determine distributions of $R_{i,T}$ from the measured uranium concentrations in well water and the estimated uranium quantities released to Paddy's Run and the SSOD. For each T, in the range 11–21, we calculate, for all possible years j, the ratio $C_{j,15}/Q_{j-T}$, where Q_{j-T} is as defined earlier, and $C_{j,15}$ is the measured uranium concentration in well 15 for year j. The subscript j has the same meaning as the index i, but is used here for calculations to determine the ratios $R_{i,T}$, while i is used in equation M–4 to determine C_i. Here we use well 15 concentrations because they are the highest of concentrations from the three contaminated wells. Because only eleven years of uranium concentration data are available, we can form only eleven of these ratios for each travel time T. Thus, for each travel time T, a distribution of ratios is developed.

To illustrate the construction of the distribution of ratios for a particular travel time, we use the example of travel time 13 y. Table M–5 shows the concentrations and release quantities used for $T = 13$ y, and the ratios calculated, for the available data. The ratios range from 0.44 to 2.5 pCi L^{-1}, with geometric mean 0.908 and geometric standard deviation 1.62. The cumulative distribution of calculated ratios for travel time 13 y is plotted in Figure M–5. As seen in Figure M–5, the distribution of ratios, for 13-year travel time, is reasonably well represented by a lognormal distribution. This lognormal representation is superior to that of a normal distribution (though the normal distribution is not shown here).

This same procedure was used to develop distributions of the ratios for each travel time. Eleven distributions are developed (one for each travel time, of 11–21 y). Upon examination, the resulting eleven distributions of these ratios are adequately represented by lognormal distributions, and, overall, the distributions appear more lognormal in shape than normal. (We note that many of the other distributions appear closer to a lognormal distribution than does the distribution for 13-y travel time, and a few appear farther from a lognormal distribution.) We thus assume that all $R_{i,T}$ will be represented by lognormal distributions, described by a geometric mean and geometric standard deviation. Table M–6 summarizes information about the eleven distributions of ratios, showing the minimum and maximum ratios computed for each travel time, and the geometric means and geometric standard deviations which will be used to represent the distributions of $R_{i,T}$.

Table M–5. Construction of the Frequency Distribution of Ratio Factors for Travel Time 13 Years [a]

Year j	C_j (pCi L^{-1})	Year $j-T$	Q_{j-T} (kg)	Ratio C_j/Q_{j-T} (pCi L^{-1} per kg)
1982	320	1969	290	1.10
1983	290	1970	349	0.831
1984	220	1971	499	0.441
1985	200	1972	322	0.621
1986	190	1973	231	0.823
1987	200	1974	255	0.784
1988	190	1975	245	0.776
1989	190	1976	272	0.699
1990	180	1977	204	0.882
1991	170	1978	68	2.50
1992	150	1979	84	1.79

[a] The subscript j has the same meaning as the index i, but is used for calculating the ratios $R_{i,T}$, while i is used in equation M–4 to determine C_i.

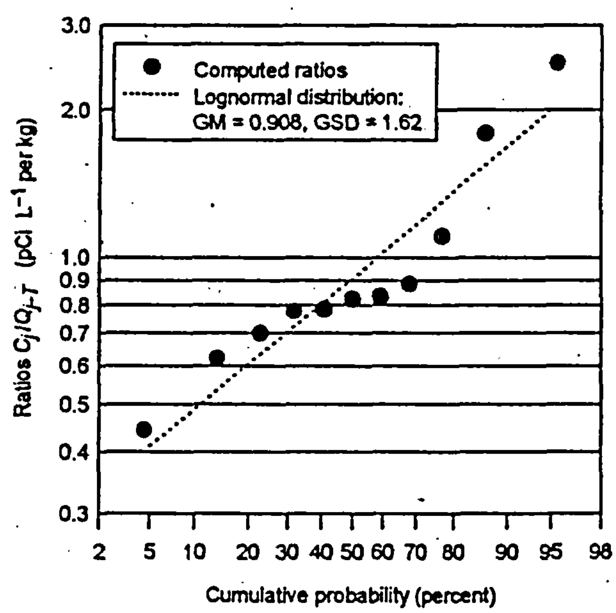

Figure M–5. Log-probability plot of the cumulative distribution of calculated ratios for travel time 13 y. The line indicates the lognormal distribution chosen to represent the distribution.

Table M–6. Development of Ratio Factors $R_{i,T}$ (pCi L^{-1} per kg): Range of Computed Ratios, and Geometric Mean (GM) and Geometric Standard Deviation (GSD) of Distributions Used to Represent Ratio Factors

Travel time T	Minimum ratio	Maximum ratio	GM	GSD
11	0.641	7.50	1.38	2.24
12	0.581	3.00	1.07	1.78
13	0.441	2.50	0.908	1.62
14	0.401	2.21	0.796	1.52
15	0.381	0.823	0.640	1.29
16	0.385	0.823	0.567	1.29
17	0.292	0.779	0.526	1.34
18	0.186	0.736	0.440	1.55
19	0.168	0.655	0.393	1.55
20	0.128	0.655	0.331	1.61
21	0.116	0.321	0.290	1.62

For a given T, $R_{i,T}$ is represented by a single distribution that is dependent only on T. We retain the index i only to indicate that, for a given T, we will consider the $R_{i,T}$ for the different years i to be independent. This is explained further below.

Implementation of calculations. As indicated earlier, the calculations are performed as a Monte Carlo simulation. The Monte Carlo calculations for this analysis were performed using spreadsheet and forecasting software on an IBM-compatible microcomputer. Ten thousand iterations of the calculations were performed. The parameter distributions were generated using Crystal Ball®, version 3.0 for Windows (Decisioneering 1993). In Crystal Ball®, uniform distributions are generated using a multiplicative congruential generator which has a period of length 2^{31}–2, and lognormal distributions are generated using the Polar Marsaglia method (Decisioneering 1993).

For each iteration of the calculations, a value of T is first chosen at random from the discrete distribution described above. Then, for each year i, a value of $R_{i,T}$ is independently chosen from the distribution for travel time T, as described by parameters in Table M–6. The values of $R_{i,T}$ for different years (different i) would presumably not actually be independent. However, for these calculations we assume they are independent so that uncertainties are conservatively estimated. Then, for each year i, C_i is calculated using equation M–4. Because the assumed minimum travel time is 11 y, and the first releases of uranium from the FMPC to Paddy's Run and the SSOD occurred in 1952, equation M–4 can only be used to calculate concentrations for years starting in 1963, at the earliest (depending on T, the earliest possible year could be later). Thus, calculations of C_i are performed only for years 1963–1988. This procedure is repeated for each iteration.

Results

The predicted annual average concentrations of uranium in private well water in the South Plume area, C_i, are summarized in Table M–7, in terms of percentiles of the distributions of results. In Figure M–6 the predicted uranium concentrations in the South

Plume are compared with measured concentrations in well 15, measured concentrations in Paddy's Run, measured and estimated concentrations in the SSOD, and estimated quantities of uranium released to the SSOD and Paddy's Run. The shapes of all the curves are generally similar, with an increase followed by a gradual decrease over time, but the estimated well concentrations curve is shifted later in time (as expected).

Table M-7. Summary of Frequency Distributions of Predicted Uranium Concentration, C_i, (pCi L^{-1}) in Private Well Water in the South Plume Area

Year (i)	5th percentile	median	95th percentile
1963	0	0	640
1964	0	0	930
1965	0	0	1000
1966	0	0	870
1967	0	0	730
1968	0	180	680
1969	0	230	890
1970	0	230	1400
1971	0	230	2000
1972	0	240	2400
1973	93	290	2800
1974	83	370	2400
1975	73	490	2700
1976	64	580	2300
1977	73	620	2100
1978	100	620	1900
1979	160	570	1400
1980	180	510	1200
1981	180	460	1100
1982	170	410	1100
1983	150	360	990
1984	130	300	810
1985	120	260	740
1986	100	230	560
1987	90	210	550
1988	78	190	490

The median prediction of the uranium concentration is zero prior to 1968 (Table M-7). This indicates that, given the assumptions that have been made about travel times, it is likely that uranium contamination in the groundwater would not have reached offsite wells prior to 1968.

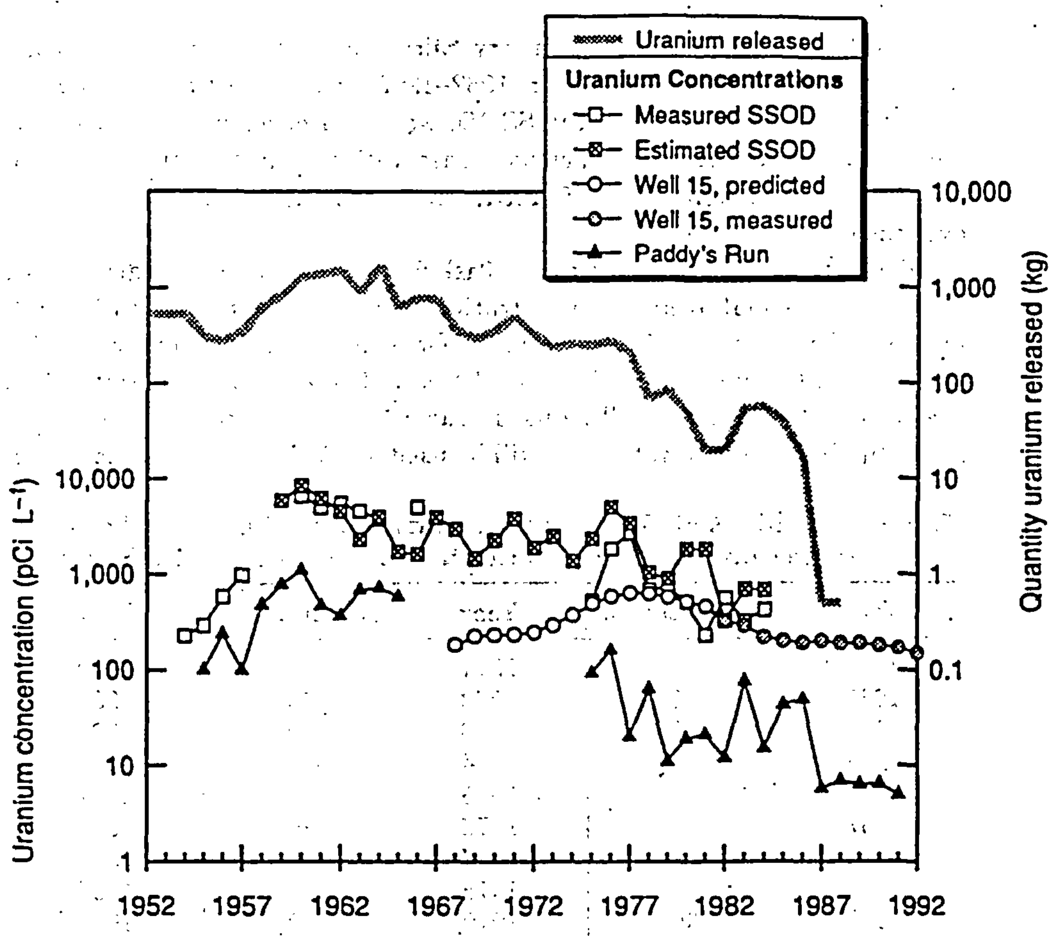

Figure M-6. Comparison of the predicted and measured uranium concentrations in the South Plume (well 15) with measured concentrations in Paddy's Run, and measured and estimated concentrations in the SSOD. The median predicted well concentrations are zero prior to 1968; these zero values are not plotted. The Paddy's Run concentrations were measured at the Willey Road bridge, sampling point W7. All concentrations are annual averages. For comparison, the annual quantities of uranium released to the SSOD and Paddy's Run are shown (note the separate scale).

SUMMARY AND CONCLUSIONS

Routine monitoring of the three contaminated wells in the South Plume has been performed by the FMPC since 1982. The annual average measured concentrations are suitable for use in the dosimetry calculations for 1982–1988. Our evaluation of the lateral movement of uranium in the groundwater indicated that the uranium plume had not reached the FMPC boundary by 1962. Thus, uranium concentrations in the South Plume wells were estimated for 1963–1981. We first estimated an upper bound on uranium concentrations that might have existed in these wells for this period, based on the maximum concentration in the source surface waters. But, we believe the upper bound is too conservative to use for the dosimetry calculations. An empirical model, which uses the measured well concentrations for 1982–1992 and the estimated quantities of uranium released to Paddy's Run and the SSOD, was then developed. The empirical model allows more realistic, though still somewhat conservative, estimates of uranium concentrations in the South Plume wells for 1963–1981. These estimated concentrations are thus preferred over the upper bound estimate.

For the dosimetry calculations of Task 6, we will employ a single best estimate, for each year, of the annual average concentration of uranium in the South Plume to which people may have been exposed. For those years for which measurements are available, 1982–1988, the measured values from well 15 will be used. For years prior to 1982 the median values of predicted concentrations will be used. Table M–8 summarizes the uranium concentrations in South Plume groundwater that will be used for Task 6 dosimetry calculations.

Table M–8. Values of Uranium Concentration (pCi L^{-1}) Used to Represent Annual Average Concentrations in Contaminated Wells of the South Plume Area

Year	Concentration	Year	Concentration	Year	Concentration
1951–1962[a]	0	1971	230	1980	510
1963	0	1972	240	1981	460
1964	0	1973	290	1982	320
1965	0	1974	370	1983	290
1966	0	1975	490	1984	220
1967	0	1976	580	1985	200
1968	180	1977	620	1986	190
1969	230	1978	620	1987	200
1970	230	1979	570	1988	190

[a] The concentration listed is applied to each year in this range.

Insoluble chemical forms of uranium would be adsorbed readily on soils in the aquifer, while soluble forms would be adsorbed to lesser degree. It is thus reasonable that the uranium which has reached offsite wells in the South Plume would be in a soluble form. Thus, for the dosimetry calculations (in Task 6), all the uranium in the contaminated groundwater source is assumed in soluble chemical form.

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ANNEX 1 TO APPENDIX M

DATA TABLES

Table M1-1. Monthly FMPC Measurements of Uranium Concentrations (mg L^{-1}) in Wells 12, 15, and 17[a]

Date	Well 12	Well 15	Well 17	Date	Well 12	Well 15	Well 17	Date	Well 12	Well 15	Well 17
Nov-81	0.190			Aug-85	0.189	0.224	0.053	May-89	0.31	0.24	0.034
Dec-81	0.160	0.320	0.054	Sep-85	0.198	0.274	0.044	Jun-89	0.25	0.23	0.030
Jan-82				Oct-85	0.220	0.338	0.0322	Jul-89	0.30	0.31	0.038
Feb-82	0.240	0.520	0.050	Nov-85	0.243	0.352	0.0518	Aug-89	0.22	0.28	0.038
Mar-82	0.240	0.410	0.069	Dec-85	0.214	0.352	0.0461	Sep-89	0.25	0.30	0.036
Apr-82	0.280	0.450	0.071	Jan-86	0.155	0.232	0.039	Oct-89	0.23	0.31	0.036
May-82	0.310	0.450	0.075	Feb-86	0.105	0.208	0.034	Nov-89	0.21	0.26	0.039
Jun-82	0.250	0.440	0.078	Mar-86	0.201	0.378	0.053	Dec-89	0.19	0.32	
Jul-82	0.270	0.440	0.099	Apr-86	0.286	0.341	0.055	Jan-90	0.21	0.27	
Aug-82	0.234	0.470	0.046	May-86	0.226	0.337	0.061	Feb-90	0.21	0.26	
Sep-82	0.238	0.480	0.061	Jun-86	0.223	0.280	0.042	Mar-90	0.21	0.28	0.038
Oct-82	0.280	0.490		Jul-86	0.222	0.286	0.040	Apr-90	0.19	0.25	0.039
Nov-82	0.220	0.502	0.054	Aug-86	0.245	0.233	0.043	May-90	0.19	0.24	0.040
Dec-82	0.230	0.554	0.066	Sep-86	0.222	0.278	0.045	Jun-90	0.18	0.27	0.044
Jan-83	0.255	0.539	0.065	Oct-86	0.227	0.271	0.049	Jul-90		0.27	0.040
Feb-83	0.306	0.578	0.055	Nov-86	0.332	0.301		Aug-90		0.27	0.044
Mar-83	0.239	0.483	0.045	Dec-86	0.160	0.280		Sep-90	0.19	0.30	0.056
Apr-83	0.225	0.460	0.060	Jan-87	0.17	0.28	0.077	Oct-90	0.18	0.33	0.041
May-83	0.249	0.419	0.057	Feb-87	0.13	0.25	0.115	Nov-90	0.16	0.28	0.052
Jun-83	0.287	0.416	0.066	Mar-87	0.32	0.31	0.064	Dec-90		0.26	
Jul-83	0.275	0.370	0.056	Apr-87	0.41	0.32	0.047	Jan-91		0.31	0.047
Aug-83	0.287	0.376	0.059	May-87	0.33	0.30	0.056	Feb-91	0.18	0.29	0.041
Sep-83	0.274	0.379	0.068	Jun-87	0.36	0.33	0.056	Mar-91	0.18	0.25	0.045
Oct-83	0.260	0.390	0.062	Jul-87	0.36	0.27	0.049	Apr-91	0.1546	0.2398	0.0536
Nov-83	0.252	0.393	0.053	Aug-87	0.30	0.33	0.049	May-91	0.19	0.22	0.043
Dec-83	0.230	0.363	0.041	Sep-87	0.33	0.29	0.052	Jun-91	0.16	0.23	0.028
Jan-84	0.245	0.365	0.053	Oct-87	0.30	0.28	0.043	Jul-91	0.17	0.22	0.047
Feb-84	0.235	0.358	0.045	Nov-87	0.29	0.30	0.050	Aug-91	0.13	0.20	0.031
Mar-84	0.256	0.355	0.052	Dec-87	0.27	0.30		Sep-91	0.12	0.22	0.034
Apr-84	0.270	0.348	0.050	Jan-88	0.30	0.27	0.073	Oct-91	0.12	0.25	0.036
May-84	0.266	0.318	0.051	Feb-88	0.24	0.26	0.069	Nov-91	0.17	0.26	0.037
Jun-84	0.270	0.311	0.059	Mar-88	0.28	0.31		Dec-91	0.13	0.25	
Jul-84	0.255	0.298	0.055	Apr-88	0.24	0.25	0.047	Jan-92	0.06	0.20	0.028
Aug-84	0.236	0.286	0.048	May-88	0.26	0.31	0.053	Feb-92	0.185	0.2027	0.040
Sep-84	0.257	0.292	0.054	Jun-88	0.24	0.30	0.054	Mar-92	0.19	0.24	0.031
Oct-84	0.222	0.312	0.068	Jul-88	0.23	0.27	0.047	Apr-92	0.10	0.21	0.032
Nov-84	0.240	0.355	0.058	Aug-88	0.23	0.31	0.059	May-92	0.18	0.23	0.041
Dec-84	0.190	0.304	0.054	Sep-88	0.23	0.30	0.048	Jun-92	0.16	0.26	0.050
Jan-85	0.189	0.360	0.048	Oct-88	0.23	0.26		Jul-92	0.307	0.24	0.043
Feb-85	0.240	0.290	0.041	Nov-88	0.26	0.28		Aug-92	0.18	0.24	0.031
Mar-85	0.201	0.297	0.054	Dec-88	0.21	0.28		Sep-92	0.12	0.21	0.033
Apr-85				Jan-89	0.23	0.29	0.054	Oct-92	0.05	0.24	
May-85		0.348	0.043	Feb-89		0.27	0.051	Nov-92	0.15	0.22	
Jun-85	0.169	0.236	0.055	Mar-89	0.26	0.26	0.044	Dec-92	0.13	0.25	
Jul-85	0.205	0.248	0.038	Apr-89	0.35	0.26	0.039				

[a] All results are from FMPC sampling. Results for November 1981 through 1983 were obtained from Dames and Moore (1985). Results for 1984 through 1992 were obtained from Kraps (1993). Blanks indicate no value was available, which apparently means no sample was taken for that month. For conversion of concentrations to units of pCi L^{-1}, multiply by 675.

Table M1–2. Calculation of Estimated Concentrations of Uranium in Storm Sewer Outfall Ditch Releases[a]

Year	Release quantity (kg)	Release volume (gal)	Concentration estimate (pCi L^{-1})
1952	520		
1953	520		
1954	520		
1955	300		
1956	270		
1957	340		
1958	630		
1959	840	26,000,000	5800
1960	1300	28,000,000	8300
1961	1400	41,000,000	6100
1962	1500	60,000,000	4500
1963	900	70,000,000	2300
1964	1700	78,000,000	3900
1965	620	66,000,000	1700
1966	770	87,000,000	1600
1967	750	35,000,000	3800
1968	360	22,000,000	2900
1969	290	36,000,000	1400
1970	350	28,000,000	2200
1971	500	24,000,000	3700
1972	320	31,000,000	1900
1973	230	17,000,000	2400
1974	255	34,000,000	1300
1975	245	19,000,000	2300
1976	270	10,000,000	4800
1977	200	11,000,000	3300
1978	68	12,000,000	1000
1979	84	17,000,000	880
1980	50	5,000,000	1800
1981	20	2,000,000	1800
1982	20	11,000,000	320
1983	54	14,000,000	690
1984	57	15,000,000	680
1985	39		
1986	17		
1987	<0.5		
1988	<0.5		

[a] Data for release quantities to Paddy's Run and release volumes to SSOD are from Appendix L. We note that release quantities are total to Paddy's Run, which includes some material not released through the SSOD. This means the concentration estimates are biased somewhat high.

ANNEX 2 TO APPENDIX M

INFORMATION REGARDING DISCARDS OF MATERIAL TO WASTE PITS

Two potential sources of offsite groundwater contamination originating on the FMPC site are (1) historical releases of uranium-contaminated water to Paddy's Run and to the Storm Sewer Outfall Ditch (SSOD), and (2) possible releases from the solid and liquid waste pits. Of these, the principal source of uranium contamination in the South Plume has been determined to be historical releases to Paddy's Run and the SSOD (DOE 1990a). The waste pits have not been significant direct contributors (by "direct" we mean through infiltration of contaminants through the bottoms of the pits) to the uranium contamination outside the site boundary. They are an indirect source, however, because runoff from contamination in and around the waste pits probably contributed to the historical releases to Paddy's Run. In our interim report (Voillequé et al. 1991), we evaluated the importance of the waste pits as a source of ground water contamination, and as a source of fugitive dust emissions (Appendix K). We studied the 1960 to 1962 period in some detail to improve our understanding of the general movement of materials around the site from the receipt of feed material, to transport and processing through the production areas, to waste disposal or shipment of product offsite. As part of this process, we compiled monthly quantities of uranium discarded to the pits for 1960–1962 in our consideration of the waste pits as a potential direct source of groundwater contamination. This annex is comprised of much of that data.

A series of waste disposal pits have been used for storage of low-level radioactive wastes during the course of the operations at the FMPC. These pits were located near the western boundary of the site, close to Paddy's Run. The waste pit area consists of waste pits numbered 1 through 6, the burn pit, and the clear well (Figure M2–1). The waste pits are typically referred to as "wet" if they received waste via pipes in slurry form or "dry" if they received solid waste from trucks. General characteristics of the waste disposal pits are summarized in Table K–35.

FMPC ANNUAL ESTIMATES OF URANIUM DISCARDED TO THE WASTE PITS

The total quantities of uranium discarded to the pits have been reported over the years in a number of records. In 1974, Gessiness reported a total of 195,000 kg of uranium to the wet pits and 2,500,000 kg to the dry pits from 1952 to 1974 (Table M2–1). In general, the quantity of uranium discarded to the dry waste pits is much higher than that in wet discards. Figure M2–2 shows that annual discards to the dry pits exceeded 100,000 kg U from 1955 to 1957 and from 1968 through 1974. In 1956, over 500,000 kg of uranium were discarded to the dry waste pit 1. For other years, from 5,000 to 50,000 kg of uranium were discarded to the dry pits. Uranium in slurries sent to the wet pits gradually increased from 43 kg in 1952 up to 22,000 kg in 1959, with levels varying from 2,000 to 10,000 kg from 1960 to 1974. The highest levels of uranium discarded to the wet pits were reported in 1963 and 1964 (about 30,000 kg), due in part to a Nuclear Materials Accounting adjustment made retrospectively for FY 1963 and 1964 (Table M2–1).

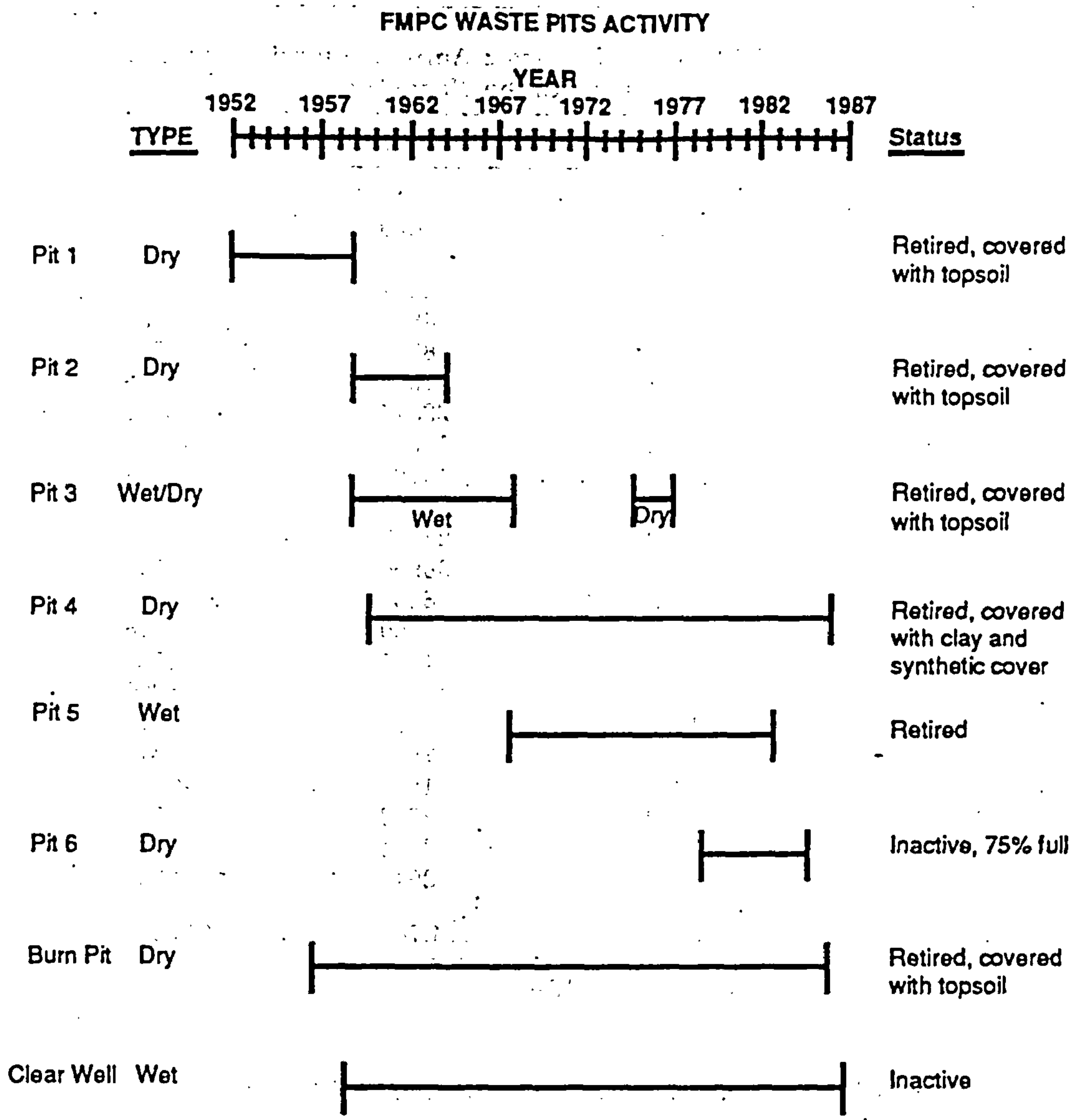

Figure M2–1. Time line of FMPC waste pits activity. The dry pits received material by truck while the wet pits usually received material by pipe in slurry form.

Thus, by mid-1974, approximately 2.7 million kg of uranium had been discarded to the waste pits (Gessiness 1974). In 1985, it was estimated that 5.3 million kg of uranium had been discarded to the waste pits from facility startup through December 1984 (Poff et al. 1985). By 1974, Pits 1 and 2 were filled, while Pits 3, 4, and 5 were still receiving waste materials (See Appendix K). By 1985, pit 1 to 5 had been "retired", while Pit 6 was 75% full (Figure M2–1).

Table M2-1. Reported Annual Quantities of Uranium (kg) Discarded to the Waste Pits From 1952 to 1974 [a]

Fiscal year	Wet Pits	Dry Pits
1952	43	0
1953	409	0
1954	863	325
1955	2107	170076
1956	4809	506235
1957	8743	361769
1958	12575	5789
1959	22315	8882
1960	11089	11971
1961	13782	49229
1962	7182	8887
1963	28147	45872
1964	30960	48112
1965	6857	3649
1966	6200	1226
1967	4555	34520
1968	10129	195309
1969	7181	160972
1970	7552	163471
1971	1557	170394
1972	2958	126311
1973	2041	283822
1974	2646	129322
Total	194,700	2,486,143

[a] From Gessiness 1974.

DISCARDS TO THE WASTE PITS FROM 1960 TO 1962

As part of our investigation of the waste pits as a source of ground water contamination, we compiled information for the early sixties on various types of discards to the waste pits. Much of these data were included in our interim source term report for the 1960 to 1962 period (Voillequé 1991). These data provide an understanding of the types, quantities and methods for quantifying these discards. In the early sixties, liquid and solid wastes were discarded to waste pits 3 and 4, respectively. Monthly totals of volume (or net weight) and quantity of uranium were reported in routine monthly loss reports (Cuthbert 1960–1961), in monthly ledger sheets (Courtney 1969), in monthly land burial reports (Noyes 1961), and in General Sump Effluent Control Logbooks for 1960, 1961 and 1962 which listed daily discards from the plants to the General Sump and from the General Sump to the waste pits (NLCO 1960; NLCO 1961; NLCO 1962).

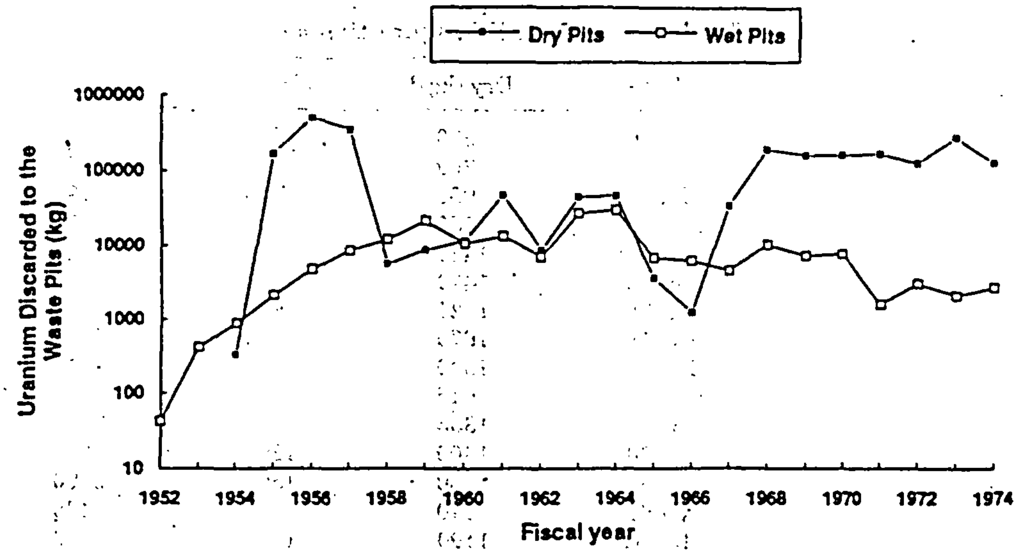

Figure M2–2. Reported quantities of uranium discarded to the wet and dry waste pits from 1952 to 1974 (Gessiness 1974). Approximately 2.7 million kg of uranium had been discarded by 1974.

Figure M2–3 shows the monthly quantities of uranium in liquid and dry wastes deposited in the waste pits from January 1960 to December 1962. For this time period, total uranium discards to the pit were nearly 1,000 kg or greater for all months except for January 1960, and July–December 1962. From May 1960 to June 1961, the totals exceeded 2,000 kg per month, with the highest quantity discarded in April 1961 (3,600 kg). The total uranium discarded to the pit in 1960 and 1961 was approximately 26,000 kg, and in 1962 approximately 12,000 kg. Uranium data for dry and wet discards are given in Table M2–2.

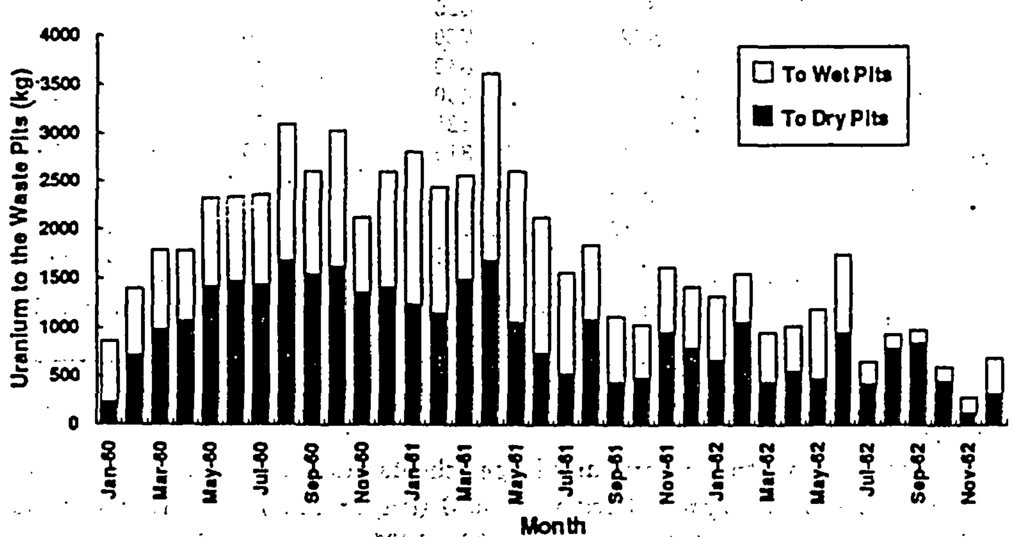

Figure M2–3. Reported quantities of uranium in materials discarded to the waste pits.

Table M2–2. Monthly Quantities of U to the Waste Pits

Date	Dry (kg)[a]	Wet (kg)	Total (kg)
Jan-60	230	630	860
Feb-60	709	685	1394
Mar-60	980	815	1795
Apr-60	1060	730	1790
May-60	1415	910	2325
Jun-60	1470	870	2340
Jul-60	1425	936	2361
Aug-60	1675	1420	3095
Sep-60	1540	1070	2610
Oct-60	1618	1418	3036
Nov-60	1355	780	2135
Dec-60	1400	1200	2600
Jan-61	1230	1590	2820
Feb-61	1140	1295	2435
Mar-61	1490	1070	2560
Apr-61	1680	1945	3625
May-61	1050	1555	2605
Jun-61	740	1390	2130
Jul-61	520	1040	1560
Aug-61	1075	770	1845
Sep-61	440	675	1115
Oct-61	480	560	1040
Nov-61	950	665	1615
Dec-61	790	625	1415
Jan-62	660	660	1320
Feb-62	1050	495	1545
Mar-62	435	515	950
Apr-62	550	470	1020
May-62	470	715	1185
Jun-62	945	800	1745
Jul-62	420	230	650
Aug-62	780	150	930
Sep-62	840	135	975
Oct-62	450	150	600
Nov-62	125	160	285
Dec-62	315	375	690
Total 1960	14880	11470	26350
Total 1961	11590	14385	25975
Total 1962	7040	5350	12390

[a] Discards include trailer cake from Plant 8, and ceramics and graphite from Plant 5.

Dry Discards

Routine monthly reports and ledger sheets tabulated solid or dry waste deposited in the waste pits that originated from Plant 8 (trailer cake) and Plant 5 (graphite or ceramic material) (Courtney 1969; Cuthbert 1960–1961). Table M2–3 contains the data on uranium discarded to the pit from Plant 8 and Plant 5 in dry material as an example of one source of material to the dry pits. The data show that monthly discards to the waste pits were dominated by the Plant 8 trailer cake residues, with Plant 5 contributing a small fraction during this time period. Discards of uranium in trailer cake from Plant 8 approached or exceeded 1,000 kg per month during much of 1960 and 1961 (Figure M2–4).

Table M2–3. Quantities of Uranium Transferred to the Waste Pits: from Plant 8, Normal and Enriched U in Trailer Cake, and from Plant 5, Total U in Graphite or Ceramics (kg) [a]

	Plant 8		Plant 5		Plant 8		Plant 5		Plant 8		Plant 5
Date	Normal	Enriched	Total	Date	Normal	Enriched	Total	Date	Normal	Enriched	Total
Jan-59	805		9	Jan-62	590		65	Jan-65	169		39
Feb-59	777		10	Feb-62	987		66	Feb-65	25	280	24
Mar-59	657		8	Mar-62	368		66	Mar-65		243	50
Apr-59	1089		4	Apr-62	490		65	Apr-65		13	26
May-59	982			May-62	409		62	May-65		30	8
Jun-59	858			Jun-62	889		58	Jun-65		92	45
Jul-59	949			Jul-62	397		26	Jul-65		52	
Aug-59	868			Aug-62	754		27	Aug-65		52	46
Sep-59	899			Sep-62	804		42	Sep-65		60	115
Oct-59	991			Oct-62	400		54	Oct-65		20	51
Nov-59	1100			Nov-62		70	55	Nov-65		51	31
Dec-59	545			Dec-62		265	50	Dec-65		66	2
Totals	10520	0	31	Totals	6088	335	636	Totals	194	959	437
Jan-60	215		17	Jan-63		480	40	Jan-66		41	4
Feb-60	710		0	Feb-63		80	53	Feb-66		94	5
Mar-60	954		29	Mar-63	399		61	Mar-66		89	10
Apr-60	1051		13	Apr-63	1428		73	Apr-66		54	4
May-60	1397		20	May-63	1440		73	May-66		32	
Jun-60	1466		9	Jun-63	1621		95	Jun-66	51		13
Jul-60	1418		12	Jul-63	883		41	Jul-66	17		
Aug-60	1674		6	Aug-63	1023		100	Aug-66	36		32
Sep-60	1502		38	Sep-63	317	473	103	Sep-66	11		32
Oct-60	1557		64	Oct-63		1535	69	Oct-66	6		
Nov-60	1315		45	Nov-63		1545	180	Nov-66	31		
Dec-60	1375		28	Dec-63		425	50	Dec-66	23		6
Totals	14634	0	281	Totals	7111	4538	938	Totals	175	310	106
Jan-61	1218		14	Jan-64		809	354	Jan-67	38		8
Feb-61	1130		12	Feb-64		556	33	Feb-67	45		
Mar-61	1488		5	Mar-64	834	87	48	Mar-67	54		14
Apr-61	1675		8	Apr-64	570		21	Apr-67	62		
May-61	1038		19	May-64	341		45	May-67	85		5
Jun-61	730		12	Jun-64	476			Jun-67	53		5
Jul-61	101	392	29	Jul-64	141		22	Jul-67	72		
Aug-61	1062		16	Aug-64	628		51	Aug-67	42		5
Sep-61	372	44	21	Sep-64	359		42	Sep-67	48		
Oct-61		474	10	Oct-64	221		26	Oct-67	46		
Nov-61	896		55	Nov-64	225		58	Nov-67	16		
Dec-61	743		53	Dec-64	125		32	Dec-67		19	4
Totals	10453	910	254	Totals	3920	1452	732	Totals	561	19	41

[a] From Courtney 1969.

There was concern about Plant 8 trailer cake discard measurements. In 1964, Nuclear Materials Control personnel pipe-sampled seven lots of trailer cake as it was being discarded into the chemical waste pit (Vath 1964b). Percent loss at 110°C, as moisture, and percent uranium were requested for these samples and the original Plant 8 samples. On a dry weight basis, the Plant 8 samples had 0.306 ± 0.247% uranium while the pit samples had 0.406 ± 0.181 % uranium. The report concluded that the comparison indicated a significant bias of 0.10% uranium or 33% of the original plant sample value (Vath 1964b). These limited data indicate that more uranium may have been discarded from the Plant 8 process than was detected by their sampling program. A limit of error estimate of 50% was assumed for trailer cake losses to the pit (NLCO 1966).

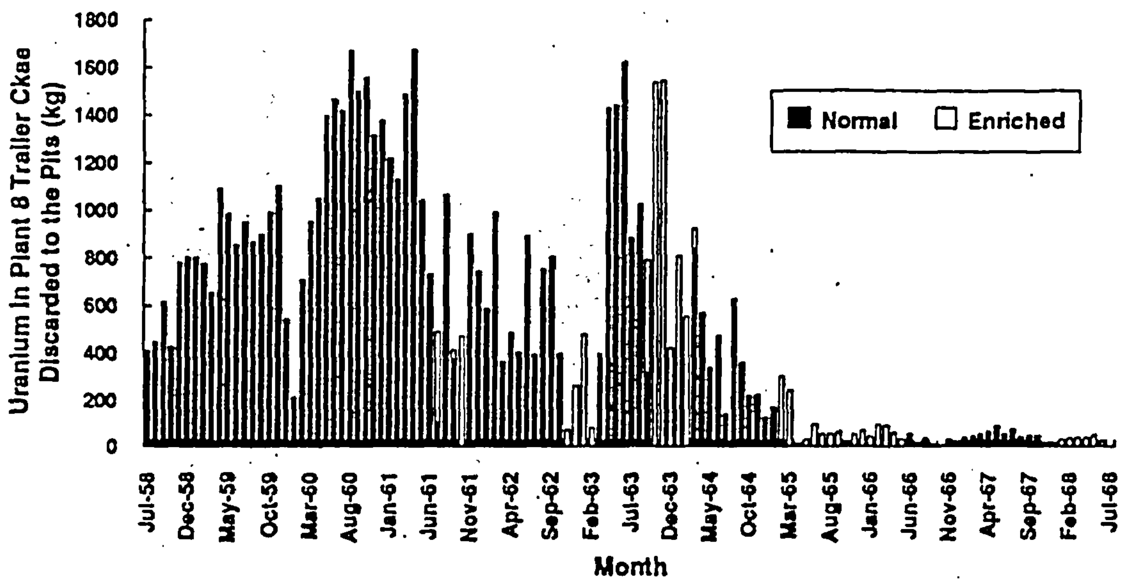

Figure M2–4. Monthly quantities of normal and enriched uranium in trailer cake from Plant 8 Discarded to the Waste Pits from July 1958 through June 1968.

Wet Discards

In the early sixties, liquid discharges were pumped to Waste Pit 3 directly from Plant 6, Plant 8, Plant 2/3 Refinery, and from the General Sump which processed waste from Plants 2/3, 4, 5, 6, 9, the analytical laboratory, and the decontamination area (Appendix L). Table M2–4 lists quantities of uranium in liquid effluents pumped to the waste pits from these sources, for 1960–1962. Figure M2–5 shows that the greatest contributors of uranium in liquid effluent to the waste pits were the Plant 2/3 Refinery, and the General Sump. Uranium discharges to the wet pits exceeded 500 kg per month except for June to October 1962. Discards during this three year period were highest from July 1960 to July 1961 when uranium discharged to the pits generally exceed 1,000 kg per month.

General sump logbooks recorded daily volume and uranium concentration measurements of liquid discards to the pit, and these data are tabulated for 1960, 1961, and 1962 in Tables M2–5, M2–6, and M2–7, at the end of this Annex (NLCO 1960; NLCO 1961; NLCO 1962). Figure M2–6 compares the volume and uranium quantities on a monthly basis for transfer of material from the General Sump to Waste Pit 3. This figure shows that effluent volume to the pits averaged approximately 250,000 gallons per day until June 1962 when it dropped three-fold to about 80,000 gallons per day. The uranium quantities were highest from July 1960 to July 1961 with a gradual decrease in quantities in 1962. The tables illustrate the detailed records that were maintained on the transfer of materials sent to the waste pits from the production buildings by way of the General Sump. Similar records were maintained for other years.

Table M2–4. Monthly Quantities of Uranium (kg) in Liquid Effluent Pumped to the Waste Pits From 1960 to 1962 [a]

Month	General Sump	Plant 2/3	Plant 8	Plant 6
Jan-60	298	298	34	1
Feb-60	320	295	68	1
Mar-60	240	511	62	2
Apr-60	270	394	67	[b]
May-60	240	602	74	[b]
Jun-60	270	512	90	5
Jul-60	220	662	51	4
Aug-60	370	955	100	[c]
Sep-60	240	738	92	1
Oct-60	210	1111	96	[b]
Nov-60	330	351	99	2
Dec-60	590	517	89	3
Jan-61	510	979	101	2
Feb-61	440	765	93	1
Mar-61	210	658	102	3
Apr-61	710	1137	95	5
May-61	300	1160	90	5
Jun-61	400	893	93	7
Jul-61	240	667	113	16
Aug-61	315	322	95	40
Sep-61	270	312	68	23
Oct-61	190	256	104	6
Nov-61	270	305	86	3
Dec-61	250	249	100	22
Jan-62	230	316	91	19
Feb-62	180	207	100	11
Mar-62	220	204	84	8
Apr-62	170	191	68	40
May-62	250	362	94	12
Jun-62	140	567	88	10
Jul-62	86	93	52	2
Aug-62	66	[b]	77	6
Sep-62	56	[b]	82	[b]
Oct-62	68	[b]	95	[c]
Nov-62	250	[b]	93	<0.5
Dec-62	550	[b]	125	1
Total 1960	3600	6900	920	19
Total 1961	4800	8200	1200	140
Total 1962	2300	1900	1100	110

[a] From Cuthbert 1960–1961, NLCO 1960, NLCO 1961, NLCO 1962.
[b] Records indicate none pumped.
[c] No records located.

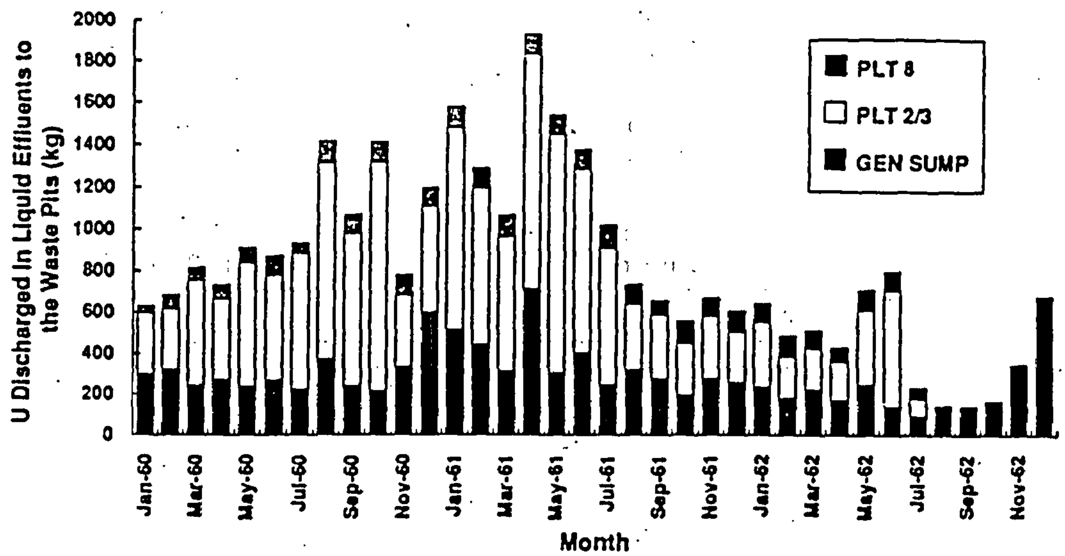

Figure M2–5. Monthly quantities of uranium in liquid effluents pumped to Waste Pit during 1960, 1961 and 1962.

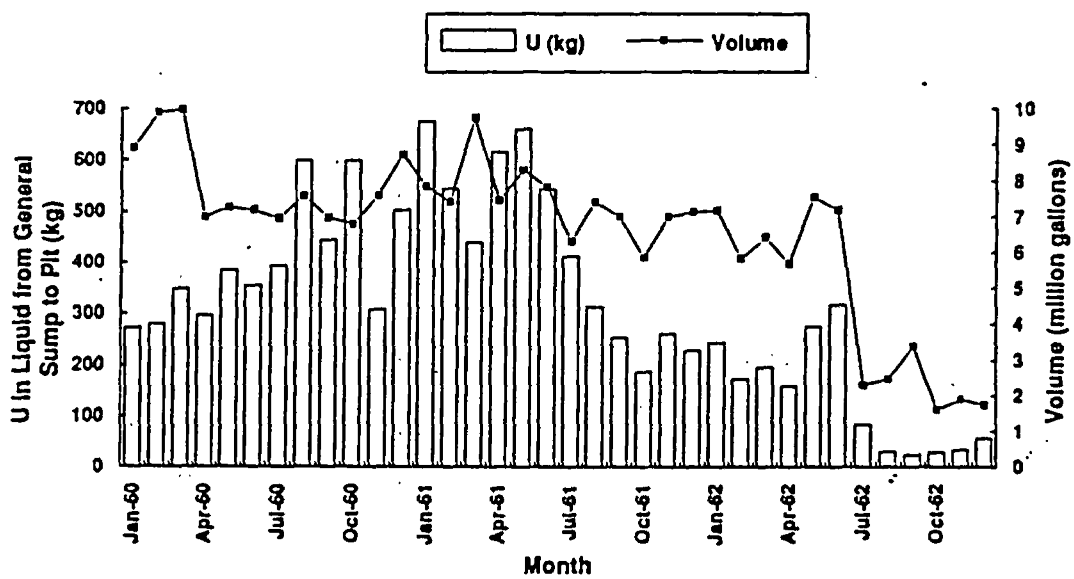

Figure M2–6. Monthly summary of effluent volume and uranium quantities discharged from the General Sump to Waste Pit 3 in 1960, 1961 and 1962. The daily discharge quantities are tabulated in Tables M2–5, M2–6, and M2–7.

Frequently, higher than usual quantities of uranium were discarded to the wet pit (Pit 3) without detection by the General Sump safeguards. This was attributed to wet weather, and to the fact that while the uranium was insoluble, the spot checks only detected dissolved uranium (Harr 1960). Plant 8 pumped its liquid waste (UAP effluent) directly to the waste pit, but its contribution was small. There was concern, however, that the Plant 8 sump effluent were not being measured accurately. A 10-day test from May 12 to 22, 1964, in which Plant 8 pumped to the General Sump rather than directly to the pit, revealed that the volume pumped as shown in Plant 8 records (144,000 gallons) was only one-fourth of the actual volume (593,000 gallons) measured by General Sump personnel (Vath 1964a). The SS uranium discarded was 11 times greater (1,044 lb.) than originally reported by Plant 8 records (94 lb.) for this 10-day period.

CONCLUSIONS

For 1960, 1961, and 1962, the reported annual totals were about 23,000 kg, 63,000 kg, and 16,000 kg, respectively. There is general agreement between the discard totals taken from the annual (Gessiness 1974) and monthly (Courtney 1969) reports for 1960 and 1962 for both wet and dry discards to the pits. For 1961, however, the reported annual total (63,000 kg) is significantly higher than the total tabulated from monthly reports. This is due to differences in quantities of dry materials. However, it should be noted that the monthly reports only list dry discards from Plant 5 and Plant 8. Contaminated residues, General Sump sludge, turnings and solid metal scrap were put into the pit at various times, but the exact quantities were not always documented.

Table M2–5. Daily Record of General Sump Effluent Pumped to Pit in 1960[a]

1960	January		February		March		April		May		June	
Date	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)
1			372932	17	441420	25	255304	28	140756	11	308168	44
2			393456	58	473760	34			332024	27	285040	9
3	512796	8	397432	40	395276	17	329084	12	310520	202	305424	46
4	420500	42	373324	25	404488	33	327898	5	307916	95		
5	419328	27	490828	57			329476	7	355096	55	109940	29
6	394492	32			232568	12	305620	25	302820	34	237272	5
7	419524	14	430892	20	390572	41	305424	24			305956	52
8	389928	27	418348	17	373520	37	276180	13	143708	2	308952	88
9			439516	30	349468	33			352744	48	284844	9
10	419916	5	438928	21	372736	10	284648	8	305394	49	306992	23
11	373912	6	373716	9	373716	4	381378	8	312480	37		
12	442456	37	353584	26			311248	32	302092	21	189896	7
13	400528	35			345448	26	304052	34	306012	22	331632	11
14	449044	37	279160	3	326928	3	285628	12			284844	3
15	394436	25	394808	19	397432	33	124252	72	141540	2	278236	71
16			395864	22	491792	42			285040	6	267064	50
17	327516	5	392336	22	325948	6	140560	3	319340	31	361620	41
18	326536	7	398355	44	396452	44	237860	15	310520	26		
19	379680	32	327124	32			309484	46	306992	19	142716	3
20	395668	30			325752	3	261128	37	354564	22	237468	7
21	373128	6	322224	5	440832	45	259204	54			309484	50
22	399632	31	326928	9	395472	50	237272	11	142520	6	262360	45
23			401156	30	394100	44			259224		331828	9
24	324576	3	401352	30	348784	35	189504	4	284648	18	306012	47
25	403861	50	399000	34	457940	60	259812	49	303212	5		
26	394688	27	451416	32			355740	79	303660	26	237468	13
27	350644	30			186760	9	313068	29	349664	40	237860	12
28	351524	39	511728	6	235900	12	305032	18		32	306193	35
29	402724	37	420700	6	351036	36	311108	29	188524		331828	29
30					348096	35				6	304640	42
31	375872	7			396060	40			237272	7		
Totals												
Vol	8909613		9905107		9972256		6999964		7258282		7173737	
U (kg)		600		610		770		650		850		780

[a] From NLCO 1960. Values include quantities of neutralized evaporative product (NEP) from Plant 2/3 and effluent from General Sump pumped to pit (see Table M2–4). Gaps in data indicate either that no pumping occurred on that day, or that the day's pumping is accounted for in an adjacent day's measurement; all material pumped is accounted for.

Table M2–5. Daily Record of General Sump Effluent Pumped to Pit in 1960 (cont'd.) [a]

1960	July		August		September		October		November		December	
Date	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)
1	310128	55	260884	86	264264	93			334180	10	405076	67
2			190092	10	301448	40	189896	4	307776	53	300328	55
3			236488	13			237468	9	329222	71		
4	284648	17	316890	134			275065	28	257872	34	233940	10
5	250388	50	260932	102	236292	5	262108	29			332612	21
6	320544	41			237272	5	316792	668	240736	7	353724	62
7	378666	33	141932	10	259756	54	306064	51	237272	6	332808	17
8	222350	18	284452	6	189896	13			379204	10	401688	73
9			306404	71	264124	91	142520	5	332808	11	349368	26
10	189112	17	309876	89			237076	9	380380	5		
11	337720	40	285236	7	190278	10	284648	14	313992	55	279748	15
12	337305	55	353188	160	189896	12	285040	10			322024	11
13	317464	44			237076	8	310660	83	187644	9	332024	12
14	342760	45	237664	7	261772	112	262360	55	284648	11	332024	17
15	327584	54	285040	8	302848	88			300328	60	381164	14
16			285236	17	284452	9	190092	5	332024	12	375340	98
17	142912	5	306796	81			189700	6	332808	8		
18	252134	21	304238	64	142324	4	284452	12	306600	77	283472	20
19	260870	48	305002	44	284256	14	358624	80		0	331632	30
20	278640	64			350448	54	306936	38	187740	8	359464	126
21	240708	45	238056	7	361544	52	284648	5	380576	40	379546	41
22	343772	47	237272	40	433440	108			332612	18	379008	31
23			283276	15	356416	48	189700	9	353192	117	378224	17
24	110544	2	344764	40			284648	6				
25	280268	44	304620	39	236684	3	284452	9	236292	-8		
26	292368	41	265104	83	332612	11	315420	77			237076	16
27	289872	43			272856	25	312932	70	235116	13	365344	159
28	353032	28	237860	46	310520	34	303072	33	285040	9	282688	78
29	285236	6	282100	47	336924	53			380380	17	331828	54
30			353584	48	324772	31	141148	4	332220	10	330848	30
31	190092	4	357300	47			237272	5			330848	10
Totals												
Vol	6939117		7574286		6962170		6792793		7580662		8721846	
U (kg)		870		1300		980		1300		680		1110

[a] From NLCO 1960. Values include quantities of neutralized evaporative product (NEP) from Plant 2/3 and effluent from General Sump pumped to pit (see Table M2–4). Gaps in data indicate either that no pumping occurred on that day, or that the day's pumping is accounted for in an adjacent day's measurement; all material pumped is accounted for.

Table M2–6. Daily Record of General Sump Effluent Pumped to Pit in 1961 [a]

1961	January		February		March		April		May		June	
Date	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)
1			7574286	105	388394	52			252560	11	347416	61
2			310128	152	382876	56	144170	4	390216	127	401117	68
3	332220	20	353780	88	322272	49	282884	58	389844	79		
4	333396	35		0			350644	12	352604	11	299054	9
5	407624	153	280924	78	341774	22	292440	82	352800	14	329290	59
6	414448	220	284452	23	333360	18	326396	54			354564	13
7			353388	104	424286	67	346138	58	352747	14	352619	62
8	189504	21	332416	27	339082	6			363680	19	395630	54
9	284452	20	312676	94	457996	95	196756	11	436404	71	246538	54
10	310324	129	245700	10	251852	11	251876	14	330900	37		
11	366660	118		0			282548	96	313068	9	200866	8
12	284648	17	179896	13	338688	11	139892	30	332056	70	235698	15
13	310912	88	283276	15	335064	37	333860	133			348467	41
14			378310	60	418796	46	344568	162	147560	6	329052	25
15	332416	10	259148	67	330800	63			351622	6	403594	132
16	284648	34	291560	9	351232	10	251972	10	303660	7	350114	96
17	351036	93	311048	93	291856	63	382592	110	332256	136		
18	284648	14					383180	94	389608	49	249026	23
19	297780	81	230802	18	248538	15	308880	61	283136	45	250982	10
20	372596	161	286594	10	298764	18	405504	100			393200	83
21			356720	38	379252	45	321000	15	236168	117	251580	11
22	234332	10	139046	3	379036	64			389846	67	328354	107
23	238252	14	336346	24	384408	71	105924	55	295580	92	200872	8
24	362740	50	374402	33	270392	32	202072	16	348136	85		
25	308168	47					455784	23	290844	144	99652	4
26	332220	12	368102	15	197593	7	328652	14	343452	67	253344	11
27	307720	51	388922	60	252560	10	389608	58			354956	10
28			439161	59	302295	10	422700	80	282408	129	252364	9
29	281512	19			341648	50					287656	213
30	293956	51			344276	41	195493	7	183872	39	303856	11
31	331828	23			50512	2			247944	5		
Totals												
Vol	7848040		7398413		8757602		7445533		8292971		7819861	
U (kg)		1500		1200		970		1400		1500		1200

[a] From NLCO 1961. Values include quantities of neutralized evaporative product (NEP) from Plant 2/3 and effluent from General Sump pumped to pit (see Table M2–4). Gaps in data indicate either that no pumping occurred on that day, or that the day's pumping is accounted for in an adjacent day's measurement; all material pumped is accounted for.

Table M2–6. Daily Record of General Sump Effluent Pumped to Pit in 1961 (cont'd.) [a]

1961	July		August		September		October		November		December	
Date	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)
1			298998	33	380270	28	48904	1	302680	14	303992	23
2	101416	4	238308	8			50216	2	343554	32		
3			290388	20			50708	1	252168	13	242436	11
4	200872	10	292388	15	389460	19					289815	13
5	302876	9			200872	14	201656	5	266676	10	289304	26
6	300840	168	150752	3	287977	8	290312	25	252168	11	267550	17
7	303060	148	350252	8	441704	39			295092	201	349048	23
8			291760	30	303856	14	100044	3	251902	10	302284	18
9	105336	4	252168	9			245364	5	268168	15		
10	252756	7	301596	15	241816	39	201656	3	293364	15	157536	9
11	252168	8	352700	13	200053	8	245896	21			251188	34
12	251580	11			396939	47	292152	22	200872	9	258168	9
13	251776	8	213200	8	291564	34	339525	30	256844	10	373422	14
14	338136	170	204600	33	301896	6			293080	12	303268	9
15			202046	19	173480	22	100828	3	339922	32	355664	15
16	148988	7	294105	16			202048	17	332000	12		
17	252364	24	304000	10	303056	13	242564	20	303464	8	201264	4
18	302680	14	202057	35	201460	12	252148	10			202244	11
19	285459	72			251186	9	296494	15	199500	5	343784	43
20	302288	9	201068	10	238276	7	315856	11	252560	10	329194	39
21	303660	9	201852	27	364250	24			341122	45	348930	22
22			293308	53	324604	46	152124	4	308444	13	292488	20
23	144284	7	343992	20			247197	18	167340	4		
24	252365	6	344423	36	151152	13	242600	50	107240	2	201068	7
25	341488	81	303072	15	290983	40	290976	36			100632	4
26	296564	39			301879	14	293524	62	165336	2	151732	4
27	290780	32	285196	69	338958	24	247226	9	344640	7	326780	31
28	240876	45	202048	6	275230	43			266364	7	323456	37
29			303072	150	239442	32	301896	8	286914	35	352996	57
30	250594	8	392000	14			302680	5	308204	44		
31	201164	9	290976	17	96672	5	291297	28			202440	5
Totals												
Vol	6274370		7400325		6987035		5845891		6999618		7120683	
U (kg)		910		690		560		410		580		500

[a] From NLCO 1961. Values include quantities of neutralized evaporative product (NEP) from Plant 2/3 and effluent from General Sump pumped to pit (see Table M2–4). Gaps in data indicate either that no pumping occurred on that day, or that the day's pumping is accounted for in an adjacent day's measurement; all material pumped is accounted for.

Table M2–7. Daily Record of General Sump Effluent Pumped to Pit in 1962 [a]

1962 Date	January Vol(gal)	January U(kg)	February Vol(gal)	February U(kg)	March Vol(gal)	March U(kg)	April Vol(gal)	April U(kg)	May Vol(gal)	May U(kg)	June Vol(gal)	June U(kg)
1	50120		252364	13	344176	20			346344	29	287056	88
2	260045	30	302876	8	258972	10	193720	37	352388	10	151732	5
3	217460	2	99064	3	100632	1	308464	13	291116	46		
4	378332	74					265756	8	353584	8	280784	60
5	366613	35	253424	5	251574	18	313072	10	98666	1	286860	16
6	252554	8	251972	9	247632	22	179508	28			347704	10
7			397432	39	239775	38	99842	2	343993	34	285824	17
8	246876	49	397236	28	298684	39			303464	6	247716	7
9	303740	53	303464	7	262250	38	245112	37	344176	56	201852	2
10	152121	4	152904	3	155008	5	254128	8	304052	13		
11	202432	3					252952	5	347162	49	313750	24
12	338884	20	208568	4	259916	17	252952	6	101024	3	350036	39
13	202038	5	346724	15	302876	14	303856	6			340928	40
14			288784	22	306312	13	151928	2	283276	5	302720	27
15	253148	14	201656	6	262364	12			286272	43	348684	13
16	253148	8	252560	55	262560	15	297851	12	353388	11	201656	1
17	252364	8	151536	3	99446	5	252952	4	282940	36		
18	303660	13					308282	37	303464	7	317500	9
19	305684	60	250012	46	252756	7	252560	9	202440	5	299355	26
20	251590	6	313072	7	279125	37	101024	3			302680	38
21			197932	24	278432	37	150752	1	244132	66	346584	20
22	317052	8	92952	3	232648	27			252364	4	353780	19
23	369172	13	92005	16	257952	5	237468	36	340332	38	104613	6
24	373780	20	100828	1	100436	4	252365	10	257756	4		
25	202648	6					252756	11	298348	38	353388	12
26	251972	8	253932	6	202832	6	304052	5	248952	9	352604	37
27	99676	3	360820	32	300708	19	300328	46			402724	103
28			300132	23	236092	7	149925	4	303660	10	302484	64
29	202636	5			262344	10			288764	44		
30	353780	24			253344	5	252756	14	99848	8	99652	15
31	403744	59			96712	2			302680	23		
Totals												
Vol	7165269		5822249		6405558		5680233		7534585		7182666	
U (kg)		540		380		430		350		610		700

[a] From NLCO 1962. Values include quantities of neutralized evaporative product (NEP) from Plant 2/3 and effluent from General Sump pumped to pit (see Table M2–4). Gaps in data indicate either that no pumping occurred on that day, or that the day's pumping is accounted for in an adjacent day's measurement; all material pumped is accounted for.

Table M2–7. Daily Record of General Sump Effluent Pumped to Pit in 1962 (cont'd) [a]

1962	July		August		September		October		November		December	
Date	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)	Vol(gal)	U(kg)
1	252756	7	148008	5			50316	1	99848	4		
2			101217	5			150556	6	99652	5		
3			241584	9			47964	3				
4					1899892	4	49924	2			150752	4
5	252952	37			50944	1	92792	3	49728	1	97104	2
6			50120	1	49336	1			99848	5	99652	5
7			147616	4	99456	4			49140	3	49140	2
8			96908	1			47964	1	101220	3		
9			96516	2			96320	5	99848	1		
10	254116	10	95340	2	98476	4	47180	3			49456	3
11					50120	1	47964	2			48552	2
12					99848	1	100808	2	50120	4	148988	7
13			100240	2	100236	4			99652	4	148008	14
14	49336	22	100044	1	100440	2			148988	10	145656	11
15			101808	1			49728	3	49728	1		
16			93380	2			48748	1	100820	7		
17	144068	20	99652	2	49727		46592	1			49728	1
18	143304	22			50316	1	96908	4			49728	3
19	197932	16			150164	2	50708		49140	1	99652	3
20	98672	6	99456	2	48944				99652	4	99652	31
21	95732	3	99260	3	94948	4			149380	5	50120	6
22			50905	3			100436	2				
23	100828	3	96712	5					49924			
24	50904	3	149962	4	95536	5	100044	5			150556	13
25	149772	13			99260	3	94164	3				
26	99848	3			100632	4	101132	3	96516	2		
27	200284	8	93968	2	46590	4			100828	2	50120	1
28			50904	2	100436	6			99652	3	150556	9
29	101808	5	98868	2			41888	2	99652	3		
30			98944	1			98868	8	99848	4		
31	101612	5	148008	4			49336	2			99456	5
Totals												
Vol	2293924		2459420		3385301		1610340		1893184		1736876	
U (kg)		180		66		50		63		72		120

[a] From NLCO 1962. Values include quantities of neutralized evaporative product (NEP) from Plant 2/3 and effluent from General Sump pumped to pit (see Table M2–4). Gaps in data indicate either that no pumping occurred on that day, or that the day's pumping is accounted for in an adjacent day's measurement; all material pumped is accounted for.

ANNEX 3 TO APPENDIX M

FATE OF URANIUM DEPOSITED ON THE GROUND SURFACE

INTRODUCTION

A source of ground contamination outside the facility boundaries was the deposition of uranium that had been discharged to the atmosphere from the variety of atmospheric sources at FMPC described in Appendices E, H, and I. The behavior of uranium deposited on the soil surface is of interest for determining contributions to alternate human exposure pathways. The physical size and relatively high density of uranium particles released to the atmosphere led to deposition of these particles on surfaces at ground level. Unless the vegetation cover is very dense most of the deposition would be from air to soil. Deposition on vegetation is gradually lost by weathering and is ultimately a source of contamination of the top layer of the soil.

Uptake of uranium from soil by vegetation is not an important transport mechanism. The maximum value reported (Peterson 1983) for the plant/soil concentration factor was that for grasses, 5×10^{-3} (dry weight basis). For a uniform concentration of 30 pCi g^{-1} in the soil, the grasses growing in that soil would be expected to reach 0.15 pCi g^{-1} at equilibrium. To estimate the maximum removal from soil by plant uptake, one can consider the removal by a very dense crop of vegetation. If the dry matter yield is assumed to be 0.4 kg m^{-2}, which is quite high, and it is assumed that multiple cuttings of the vegetation occur over an extended growing season, the removal rate of uranium from the soil is less than 0.1 pCi cm^{-2} y^{-1}.

A special study that was conducted to determine the primary transport pathway for uranium deposited on soil around the FMPC is summarized below. Calculations addressed in this study compare uranium migration due to infiltration, surface soil erosion, and surface water runoff (Ichimura 1991b).

GENERAL METHOD

There are numerous transport models available in the literature; however, to simplify this evaluation, the calculations were based on simple algebraic and "handbook" models. The goal of this study was to estimate the annual quantities and average concentrations of uranium that would be transported through each of the above pathways from a unit area of soil having homogeneous uranium concentration in soil.

Uranium migration estimates within this report only apply to the flat lying, poorly drained soils around the FMPC which are covered with pasture grass. These soils include the Fincastle silt loam and the Henshaw silt loam (Lerch et al. 1982). For these calculations it was assumed that the land slope is less than 2% and the typical drainage length before encountering ravines is 660 ft. Also, these soils are poorly drained and require artificial drainage to help maintain crop productivity. Where artificial drainage is not installed, the water content in these soils is high during extended periods in the winter and spring. For this reason, the infiltration calculation assumes saturated groundwater flow in the vertical direction.

WATER BALANCE

The fate of adsorbed uranium in soil can be estimated if the rates of soil and water movement are well known. Therefore, the hydrologic water balance must be estimated to quantify water movement. Water balance considers the rates of rainfall, runoff, infiltration, and evapotranspiration. The infiltration component of the water balance moves uranium through the soil column. The runoff waters carry uranium by desorption and ion exchange from contaminated soil and transport surface soil in the form of suspended sediments away from contaminated areas. To compare these transport mechanisms, all models assume that the source of uranium is the soil, and that the soil is infinitely replenished with uranium. For comparison purposes, the concentration of uranium in the soil is assumed to be:

$$C_s = 30 \text{ pCi g}^{-1}$$

This concentration is typical of ^{238}U concentration found in soils on the FMPC site (Solow and Phoenix 1987). Note, however, that the magnitude of soil uranium concentration used for these comparisons is not critical, as we are most interested in the relative transport rates for the different mechanisms. Furthermore, if the soil concentration is C_s, then the associated soil-water concentration, C, can be obtained from the distribution coefficient (K_d) relationship:

$$K_d = \frac{C_s}{C} \quad \left(\frac{\text{pCi g}^{-1}}{\text{pCi mL}^{-1}}\right)$$

A distribution coefficient of 9 mL g^{-1} has been estimated for the sand-clay aquifer which underlies the FMPC (ASI-IT 1990). At present, the distribution coefficient of the surface soils is not known.

Movement of uranium is dependent upon the various components of the water balance. This section describes the models, data, and results of the water balance calculations which require estimating annual surface runoff, annual evapotranspiration, and annual water infiltration. In summary, the water balance for a watershed can be described as follows:

$$\begin{aligned} \text{Annual Precipitation} = {} & \text{Annual Runoff} \\ & + \text{Annual Evapotranspiration} \\ & + \text{Annual Infiltration} \end{aligned}$$

First, the annual runoff from the area was determined. The U.S. Soil Conservation Service (SCS) has developed a number of models for estimating runoff from ungauged watersheds. The model used is described in the reference (SCS 1969). It requires estimates of watershed size, annual precipitation, annual temperature, and land condition. For this calculation, the size of the watershed was assumed to be about 40,000 m^2 (10 acres). The average annual precipitation in the vicinity of the FMPC is 97 cm y^{-1} (38 in y^{-1}) (ASI-IT 1990). The average annual temperature is 12°C (54°F) (Spieker 1968). The land condition was assumed to be a "Good Pasture." Applying the above parameters yields an estimated runoff of 5.1 cm y^{-1} (2.0 in y^{-1}).

Next, the evapotranspiration was estimated using pan evaporation data. For the FMPC area, the average annual pan evaporation is 111 cm y^{-1} (44 in y^{-1}) (Veihmeyer 1964). By applying the method described by Jensen (1980), the estimated annual evapotranspiration was 79 cm y^{-1} (31 in y^{-1}).

Using these values calculated above, the annual infiltration rate is estimated to be 13 cm y^{-1} (5 in y^{-1}). This annual infiltration rate compares well with the estimated groundwater recharge rate of 15 cm y^{-1} (6 in y^{-1}) in the glacial till which surrounds the FMPC (GeoTrans 1985). Figure M3–1 summarizes the results of the water balance calculations for flat lying areas underlain by low permeability soils around the FMPC.

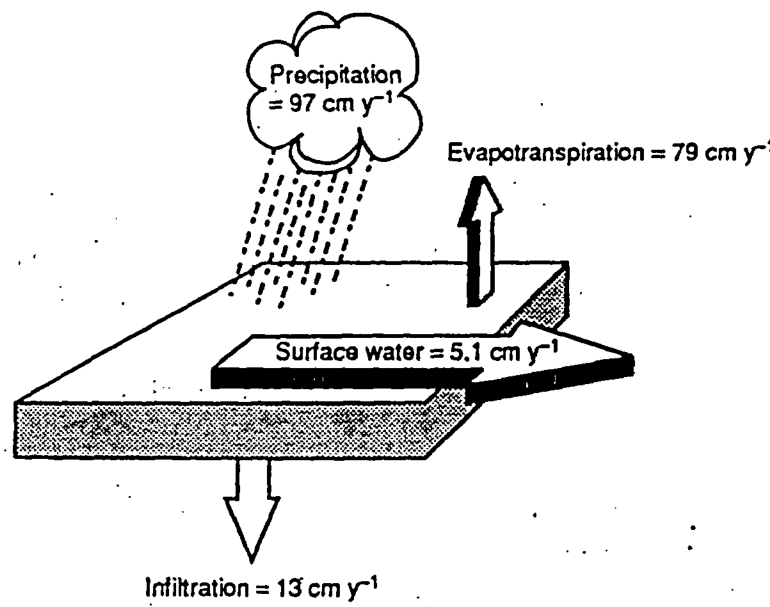

Figure M3–1. Summary of annual water balance summary for flat lying, low-permeability pasture land around the FMPC.

ANNUAL SHEET SOIL EROSION

In this section, the annual sheet erosion rate around the FMPC is described using the Universal Soil Loss Equation (USLE) (Wischmeier and Smith 1978). The USLE is designed to estimate average soil loss due to sheet and rill erosion. The USLE uses rainfall pattern, soil type, slope, cover, and land management practice to estimate the annual erosion rate. Since deposited uranium is attached to soil particles, the amount of uranium in motion is partially dependent on soil particle transport. Soil particle transport is estimated by using the soil erosion rates.

Due to the low slope angle, lack of ravines, and ground cover management practices around the FMPC, the estimated annual erosion rate is 45 g m^{-2} y^{-1} (0.2 tons $acre^{-1}$ y^{-1}). For an assumed soil density of 1.6 g cm^{-3} (100 lb ft^{-3}), the associated soil loss thickness is 3×10^{-3} cm y^{-1} (1×10^{-4} ft y^{-1}). Soil loss estimates for other erosion rates are shown in Table M3–1. It can be seen from the table that, even at the high erosion rate of 4500 g m^{-2} y^{-1} (20 tons $acre^{-1}$ y^{-1}), the thickness of soil lost due to erosion is negligible.

Table M3–1. Estimates of Soil Loss for Various Erosion Rates

Erosion rate (tons $acre^{-1}$ y^{-1})	Thickness of soil lost (cm y^{-1})[a]
0.1	0.002
0.2	0.003
5.0	0.06
10.0	0.2
20.0	0.3

[a] Assumed soil unit weight = 1.6 g cm^{-3}.

ANNUAL URANIUM TRANSPORT BY SOIL LOSS

The model of uranium transport due to erosion is shown in Figure M3–2. As shown in this figure, the quantity of uranium leaving a unit area is the sum of all uranium in the soil removed from the surface in a given year.

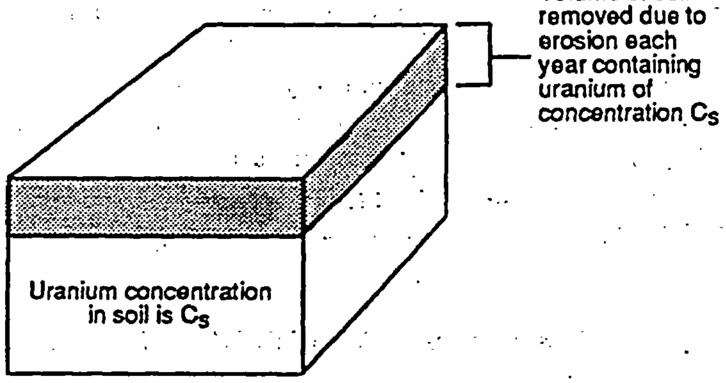

Figure M3–2. Uranium transport by soil erosion.

The amount of uranium leaving a square centimeter of area is calculated from the soil erosion losses each year. If the annual soil loss is 3×10^{-3} cm y^{-1} for each square centimeter, erosion will move about 5×10^{-3} g y^{-1} of soil. Assuming a uranium concentration in soil of 30 pCi g^{-1}, the quantity of uranium leaving each square centimeter by sheet erosion is estimated to be 0.14 pCi y^{-1}.

ANNUAL URANIUM TRANSPORT BY INFILTRATION

Figure M3–3 shows the uranium transport model for infiltration that is described below. Annual movement of uranium through the soil column assumes a continuous supply of uranium is being added to the soil column. This assumption is consistent with continuous airborne deposition of uranium during early FMPC operations. The calculations have assumed that the deposition resulted in a homogeneous uranium concentration in soil of 30 pCi g^{-1}. In the model, flow through porous media is assumed to be steady and saturated, and dispersion and diffusion are considered negligible. The ratio of uranium in water and soil is related by the distribution coefficient (K_d).

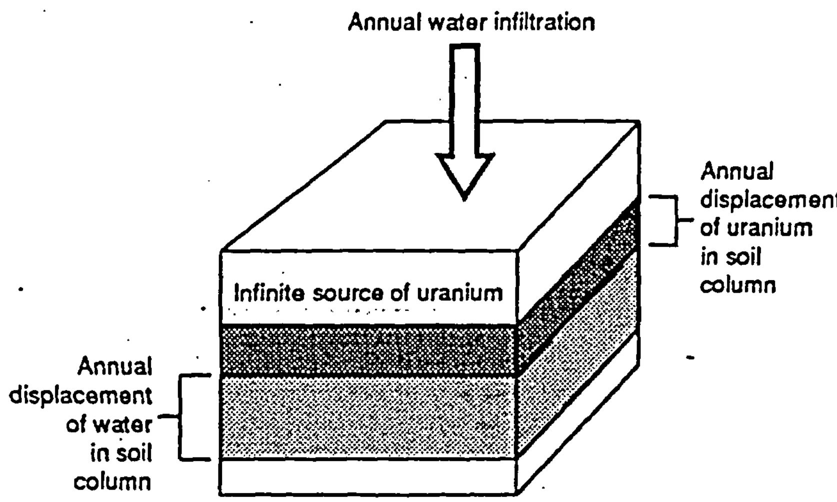

Figure M3–3. Annual uranium transport by infiltrating water.

According to this model, the quantity of 15 cm (6 in) of infiltrating groundwater will move 50 pCi of uranium in the soil column each year. The concentration of uranium in the water is 3.3 pCi mL^{-1} and the distribution coefficient is 9 mL g^{-1}. However, due to uranium adsorption, the uranium will only move 1.0 cm into the soil column each year while the water will move a distance of 61 cm (2 ft). This calculation assumes that the soil effective porosity is 0.25.

ANNUAL URANIUM TRANSPORT BY SURFACE WATER RUNOFF

The model of uranium migration by surface water runoff is illustrated in Figure M3–4. Movement of uranium by surface water assumes that a continuous supply of uranium is available from the soil and the concentration of uranium in the soil is constant. Uranium from contaminated soil exchanges with the runoff component of precipitation, and the exchange ratio is dependent upon the distribution coefficient (K_d).

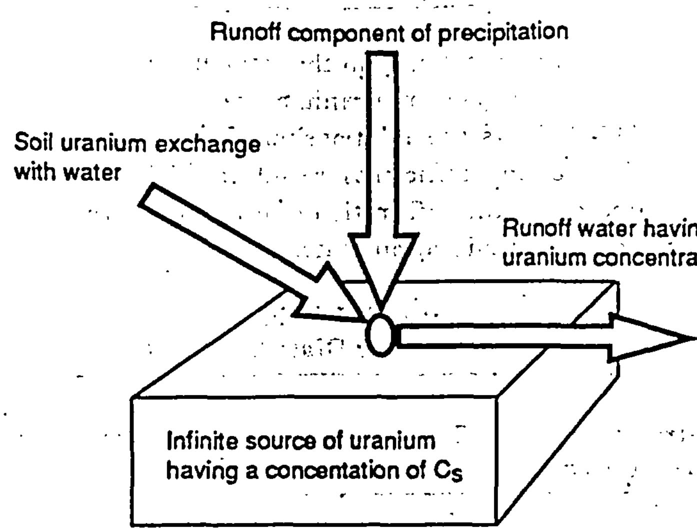

Figure M3–4. Uranium transport by runoff water.

For comparison purposes, the soil concentration is assumed to be 30 pCi g^{-1} and the distribution coefficient is 9 mL g^{-1}. According to the hydrologic water balance calculations, runoff is 5.1 cm y^{-1} (2.0 in y^{-1}). Therefore, the quantity of uranium moved by surface water runoff is 17 pCi y^{-1}, and the concentration of uranium in the water is 3.3 pCi mL^{-1}.

URANIUM TRANSPORT SUMMARY

Table M3–2 shows the rate of uranium migration from a square centimeter of soil having a concentration of 30 pCi g^{-1} and a distribution coefficient of 9 mL g^{-1}. The results show that uranium deposited on pastured soils is primarily transported by infiltration and that soil erosion transports the least amount of uranium.

Table M3–2. Comparison of Uranium Transport Mechanisms[a]

Transport mechanism	Medium	Transport rate (pCi y^{-1})	Uranium concentration in medium
Soil sheet erosion	Soil	0.14	30 pCi g^{-1}
Infiltration	Soil	na	30 pCi g^{-1}
	Water	50	3.3 pCi mL^{-1}
Surface runoff	Water	17	3.3 pCi mL^{-1}

[a] Per cm^2 soil having a uranium concentration of 30 pCi g^{-1}.

It should be noted that the distribution coefficient of the surface soil is unknown. The distribution coefficient of 9 mL g^{-1} used in this study was obtained from a provisional calibration of the ASI-IT (1990) South Plume model. Therefore, this distribution coefficient may not be applicable because the groundwater model does not predict uranium transport at the surface. Because of the uncertainty in the estimated value of the distribution coefficient for the surface soil, an analysis of uranium transport for different values of K_d was performed. Table M3-3 shows the relationship of the distribution coefficient to uranium mobility. Lower distribution coefficients result in higher rates of uranium migration. However, in all three cases, the infiltration of uranium down the soil column towards the groundwater is the dominant migration route.

Table M3-3. Sensitivity of Uranium Transport Calculations to the Value of the Distribution Coefficient

Distribution coefficient (mL g^{-1})	Transport mechanism	Transport rate (pCi cm^{-2} y^{-1})	Uranium concentration in water (pCi mL^{-1})
5	Infiltration	91	6.0
	Surface runoff	31	6.0
9	Infiltration	50	3.3
	Surface runoff	17	3.3
20	Infiltration	23	1.5
	Surface runoff	7.6	1.5