
Methods for Assessing Background Levels of Radiation and Radioactive Materials in the Environment Around Uranium Mills

Manuscript Submitted — July 1979
Date Published — April 1980

Prepared for
the U.S. Nuclear Regulatory Commission

Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute



PNL-3256

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Under Contract DE-AC06-76RLO 1830

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GPO Sales Program
Division of Technical Information and Document Control
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555
and
National Technical Information Service
United States Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151

Price: Printed Copy \$ _____*; Microfiche \$3.00

*Pages	NTIS Selling Price
001-025	\$4.00
026-050	\$4.50
051-075	\$5.25
076-100	\$6.00
101-125	\$6.50
126-150	\$7.25
151-175	\$8.00
176-200	\$9.00
201-225	\$9.25
226-250	\$9.50
251-275	\$10.75
276-300	\$11.00

METHODS FOR ASSESSING
BACKGROUND LEVELS OF
RADIATION AND RADIOACTIVE
MATERIALS IN THE ENVIRONMENT
AROUND URANIUM MILLS

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Manuscript Submitted July 1979
Date Published April 1980

Prepared for
the U.S. Nuclear Regulatory Commission
under a Related Services Agreement
with the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830
Fin. No. B2121-8

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Richland, Washington 99352

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1.0 EXECUTIVE SUMMARY

A major concern regarding both existing and proposed sitings of uranium processing mills is their radiological impact on the surrounding unrestricted environment. In order to assess an incremental increase in radioactive background of any property due to mill operations, it is mandatory that natural or background levels of that property be established. This report assesses techniques and costs for determining background levels and mill contributions to the environment above these levels. In this executive summary, three specific programs are identified for the determination of natural background and mill contributions to that background. Since the most significant radiological impact to man within 10 km of mill tailings occurs through airborne ^{222}Rn and its daughters, their measurement is emphasized in the suggested procedures. The next major radiological impact from the mills occurs through airborne movement of particulates from the mill and its tailings piles. Thus, the more sophisticated measurement technologies presented include measurements of airborne radionuclide particulates, as well as methods to measure the dose from ^{222}Rn and its daughters. The most expensive methods for assessing background levels of radioactive materials around uranium mills allow a determination of uranium, thorium, and radium in water, soil, and vegetation, as well as in air. The methodologies are organized by their increasing capital and operating costs. The more expensive techniques provide a better evaluation of the mill contribution to the environment. There is no single universal technique that is applicable to all mills. Although we believe that the following procedures can be used at many mills, each might require modification at specific sites. Many alternatives to these procedures are discussed within the body of this report to allow modifications to satisfy site specific criteria. Although preoperational survey methodologies are discussed in a specific section of the text, the three methodologies discussed below can also be used to measure the extent of the natural background for a preoperational survey.

TABLE 1. Basic Strategies for Assessing Radiation Around Uranium Mills

Strategies	Measurement Technology	Quantity Measured	Estimated Capital Cost (In \$1,000)
A.	Thermoluminescence Dosimetry	Dose	25
B.	Strategy A + Meteorology Air Sampling	Dose + Specific Radionuclides on Airborne Particulates	80
C.	Strategy B + Radiochemical Analysis	Dose + Measurement of Specific Radionuclides Distribution in Airborne Particulates, Soil, Water, and Biota	115
D.	Advanced Concepts		>500

The least expensive methodology, using thermoluminescence dosimetry (TLD) as the monitoring technique, involves capital costs of \$25,000 and

a 0.5 man-year per year operating effort. Thermoluminescent dosimeters measure the gross beta and alpha radiations from atmospheric and soil-originated ^{222}Rn and its daughters at specific sampling locations relative to the mill. There should be dosimetry extending radially from the mill on lines separated 60° from one another. The primary line should be placed on the major wind rose. Dosimeters should be located at the mill site boundary, and 0.5, 1, 2, 3, 4, and 5 km from the boundary. Dosimeter duplication at eight locations is required to ascertain statistical validity of the dosimetry. Each dosimeter station must be comprised of a lead shielded and unshielded TLD chip mounted 0.5 m above ground. In this fashion, the unshielded TLDs receive radiation from both ground and air sources, whereas the shielded TLD is exposed only to the atmospheric component. The TLDs should be changed monthly. Specific procedures and shield designs are discussed in the body of the text. The TLD is not isotope specific nor does it provide the sensitivity of alternate methods; however, it does provide a precision of $\leq \pm 10\%$ at the 10 mr per month level.

The advantages of a TLD system are that it is simple, cost effective, and reliable. It requires no power in the field and has no problems from a variety of weather conditions. The TLD system, in its simplified form, does not require an on-off mechanism controlled by meteorological conditions.

The major disadvantage of the method is that it measures only gross radiation--that from beta and gamma contributions. It does require a meteorology station to provide wind rose data for its initial set-up and then later for interpretation of the data which are obtained. The capability of the TLD methodology will not allow natural background to be differentiated from sources resulting from earlier mill operation or that delivered to the sampler location from sources outside the one being studied. It capably determines the current mill additions to the environment. Although it allows measurement of those materials currently contributed by sources other than the mill in question, it does not allow them to be individually quantified.

The TLD method requires an effort to maintain the quality of the TLD chips and their dose response. Through calibration of the TLD chips in

radon chambers, the technique provides the dose to man from ^{222}Rn and its daughters to be determined directly.

Another methodology involving meteorology-controlled air sampling followed by photon analysis of the sampled particulate material costs \$55,000 in capital and requires 0.75 man-years per year of operating expense. The air sampling methodology and its associated costs should be added to that from the TLD technology for the next level of sophistication in assessing the background. In this fashion, not only is the airborne ^{222}Rn and its daughters measured but also included are the radionuclides transported by airborne particulates from the mill. The air samplers should be located radially on lines separated by 60° from one another extending from the mill boundary, as previously discussed. There would be three high volume air samplers on each line, one omnidirectional and one directional sampler at the boundary of the mill and a second directional sampler 1 km from the boundary in the upwind direction. A central meteorology station controls the air sampler operation based on the wind vector. The samplers collect the airborne particulates on filters which should be changed on a monthly basis. The samplers would be placed 1 m above ground and would be enclosed against weather and animal penetration. For analysis, the air filters are pressed into a defined geometry, typically 5-cm diameter discs, and are analyzed for their gamma-ray emitting radionuclides using a planar intrinsic germanium detector coupled to a 2048 channel analyzer with a hard-copy readout. The costs for this spectrometer are included in this technology's \$55,000 capital. The sensitivity for air particulate analysis with such a system, defined as the minimal detection activity at the 95% confidence level, is 1.4 dpm ^{210}Pb , 1.1 dpm ^{214}Pb , 18 dpm ^{234}U , 2.2 dpm ^{238}U , 14 dpm ^{230}Th , 0.6 dpm ^{235}U , and 8 dpm ^{226}Ra . The use of the TLD technology provides the dose from airborne ^{222}Rn and its daughters. System and technique modification for site specific criteria are discussed in detail within the text.

The advantages of such a system are that the radioisotopes in question can be measured at their environmental disintegration rates. In most cases, lower limits of detection, as designated by the Nuclear Regulatory Commission, can be achieved for air particulates. The measurement of the concentration of specific radioisotopes in airborne particulates,

as a function of sampling location and meteorology, allows an assay of the current mill contribution to the environment versus airborne material resuspended from previous mill deposition or that originating from sources external to the mill under study. Data analysis can be easily computerized for interpretation at any later date through statistical criteria. All advantages/disadvantages of the TLD method previously discussed apply within this technique.

The primary disadvantages of airborne particulate sampling are that the system requires electrical power, mechanical and electrical maintenance, and a meteorology controlled device. The technology measures specifically only radionuclides associated with air particulates. It requires radioactive standards to be used in the laboratory, as well as a license to handle radioactive material. The detector, an intrinsic germanium photon spectrometer, requires a source of liquid nitrogen and a dedicated counting room. As discussed in the text, air samplers may require additional capital modifications to handle excessive dust loads and other weather conditions. Topography effects must be evaluated for some sampler sites.

Further level of sophistication involves the measurement of air samples, biota, soil, and water. The methodology will require a total of \$115,000 in capital equipment (\$25,000 - TLD, \$55,000 - air sampling, \$35,000 - additional equipment as detailed below), a minimum of 2 man-years per year or dedicated time, in addition to dedicated chemistry and counting laboratories.

As in the second suggested technique, a complete TLD, meteorology/atmospheric measurement program similar in all respects to that previously discussed would be employed. In addition, soil, water, and biota would be sampled on the six lines on which the air sampling system has been assembled. Soil samples would be collected at the boundary, 0.5, 1, 2, 3, 4, and 5 km from the boundary. Samples should be taken using a coring device to obtain aliquots at 1/2, 1, 2, 5, and 10-cm depths from a 10 cm x 20 cm surface area. Deeper cores are obtained by digging a trench and inserting a stainless steel flat scoop with 5-cm high walls.

Biological material should be collected from a 1 m² area at the sampling sites. It should be dried, homogenized, and duplicate fractions taken for radionuclide analysis. Water should be collected from sources within the sampling grid in sufficient size to achieve the necessary lower limit of detection as required by the NRC. The specific limits are given in the body of the text. The soil, water, and biota sampling should occur once per year with the air sampling occurring monthly. Photon spectroscopy, using the equipment suggested for the second technique, should be used to analyze all samples. Samples which require radiochemical separations to attain NRC's lower limit of detection would be identified by gamma-ray spectrometry. This will minimize the number of samples requiring radiochemical procedures. The chemical procedures which should be used on the soil, biota, and water samples to isolate the small quantities of radionuclides present from the bulk material are presented in Section 9.1 of this report. A beta counter and a spectrometry system must be used for the analysis of the specific radionuclides isolated with these chemical procedures. In this fashion, all radionuclides such as natural uranium, ²³⁰Th, ²²⁶Ra, ²¹⁰Pb, ²²²Rn, and ²¹⁰Po, in the various environmental materials can be measured at their environmental concentrations. It should be noted that the suggested NRC lower limits of detection are in some cases below that found in the environment for specific radionuclides; these cases are discussed in the text.

The advantages of using this technology are that it allows the measurement of all of the necessary radionuclides in all types of environmental materials at environmental levels. It allows the determination of the background and the effect the mill has had on the background in each component of the mill environment. All advantages and disadvantages previously discussed for TLD measurements and air sampling apply to this technology. Specific disadvantages of this methodology are the additional manpower requirements, including a trained radiochemist, and additional laboratory and counting room facilities.

Throughout the discussion in this report, research areas are identified where required to fulfill the needs of each suggested measurement technique or method. This research arises from the fact that although much equipment is available, it has not been evaluated for the specific

job requirement. Limited research is required to demonstrate the validity of the methodologies suggested here.

Other potential methods for determining the contribution of a mill to the local background include the continuous analysis of ^{222}Rn gas through sampling and the use of lasers and trace element ratios in the ores to differentiate fugitive mill material versus that in the environment. A significant basic research effort would be required to develop the feasibility of these approaches. It is estimated that the laser and trace element ratio technology could be developed with approximately one million dollars of research. It would require approximately \$150,000 in capital equipment to implement at the mill site and a minimum of 2 man-years per year of effort involving personnel trained at the doctoral level in nuclear chemistry or physics.

As discussed in the body of this report, there are many sophistications which can be applied to the aforementioned strategies. In some instances, the increased costs for these modifications could rise as high as \$300,000 for capital and require 4 man-years per year of effort by trained personnel.

Depending on the method chosen, it is suggested that the uranium industry buy the equipment as a consortium, determine the degree of prior contamination of the environment to provide a baseline, and then use the inexpensive technique to monitor their individual emissions to their environment.

2.0 INTRODUCTION

A major concern regarding both existing and proposed sitings of uranium processing mills is their radiological impact on the surrounding unrestricted environment. In order to assess an incremental increase in radioactive background of any property due to mill operation, it is mandatory that natural or background levels of that property be established. In this report, techniques and costs for assessing background levels and mill contributions above these levels will be presented.

Since the most significant radiological impact to man within ten kilometers of mill tailings occurs through airborne ^{222}Rn and its

daughters, their measurement is emphasized in the suggested procedures. The next major radiological impact from the mills occurs through airborne measurement of particulates from the mill and its tailings pile. Thus, the more sophisticated measurement technologies presented include measurements of airborne radionuclide particulates, as well as methods to measure the dose from ^{222}Rn and its daughters. The most expensive methods for assessing background levels of radioactive materials around a uranium mill allow a determination of uranium, thorium, and radium in water, soil, and vegetation, as well as air.

The discussion provides methods for formulating long-term routine sampling or dose measurement programs to assess the background and the contribution of a uranium mill complex to background radiation. The sampling programs provide data on the background and mill-originated contribution to certain airborne, water, soil, and biota contaminants in the vicinity or within 5 km of the mill boundaries pursuant to NRC guidelines.¹ Detailed descriptions of the various effluents from uranium mills and their associated radionuclide burdens have been well documented² and will only be summarized below for clarity.

2.1 Gaseous Effluents

Radon is continually released^{2,3} during all mill process operations at rates that vary considerably depending on the operation. Relatively low radon emission rates are associated with ore storage. During crushing and milling, radon emission is enhanced due to the reduction of particle size and the resultant increased surface area of the feed stock. Radon, reduced in concentration by previous losses, will be fractionally dissolved during leaching and will escape the leachate as its solubility is exceeded; it then transfers to the overlying atmosphere. Radon and its radium parent are depleted in the uranium recovery line. Essentially all of the radium follows the waste stream and is disposed to the tailings pond. Radioactive build-up of radon occurs within the tailings and escapes to the atmosphere at a rate that is governed by diffusion, tailings composition, moisture content, and the prevailing atmospheric conditions.

Radon-222 and its short-lived daughters ^{218}Po , ^{214}Pb , and ^{214}Bi can be assayed to determine the ongoing radon release rate from a milling

complex. Background concentrations arise from the radium naturally present in the environment, previously translocated tailings, and contributions from other mining and milling operations in the area. Lead-210, ^{210}Bi , and ^{210}Po , the longer-lived daughters of ^{222}Rn , associate with atmospheric aerosols and are deposited on the earth's surface. The concentration of these radionuclides represents the long-term cumulative contribution of milling activity to the environment. Background levels of these two radionuclides result from decay of uranium present in the soil and from deposition of radon daughters that are derived from the ever present atmospheric inventory of radon. For all practical purposes, the background levels of ^{210}Pb and ^{210}Bi should be at steady state, with the possible exception of contributions from other local mining and milling activities to the environment of the mill under study.

2.2 Particulate Effluents

The potential for particulate release exists in nearly all phases of the milling operation; however, remedial measures are generally employed to reduce the quantities of material escaping from the plant. Ore particles, containing near equilibrium concentrations of radionuclides in the uranium decay chain, can originate during transportation, ore storage, milling, and transfer operations. The final uranium product can become airborne during drying and packaging steps. Loss of product is perhaps the most minimal of any particulate source as this is profit and every effort is made to minimize its unnecessary departure. The most contributory source of particulate release is the wind erosion and transport of material from the dry beach and sides of the tailings pile. This material is, of course, altered in content, both chemically and radiochemically, by the particular extraction and recovery process employed.

It does contain the isotopes of thorium, radium, and lead present in the original ore, being essentially depleted in only uranium. Thus, the tailings material will contain daughter products of these parents at concentrations dictated by the time elapsed since separation of the particular decay chain.

2.3 Liquid Effluents

With only minor exceptions, liquid wastes are disposed to the tailings pile. Much of the liquid is recovered by decantation and recycled

to process, some percolates into the ground, and the remainder leaves the tailings impoundment by evaporation. Impurities could enter the environment through percolation, potentially reach a subterranean aquifer and be transported with the flow of groundwater. The radionuclides occurring in liquid wastes and their relative concentrations will be governed by the chemistry of the particular process involved. The fact that the radionuclides are in a dissolved state subjects them and their radioactive daughters to fixation processes such as precipitation, ion exchange, and surface adsorption that will dictate their mobility and the rates at which they could migrate throughout the environment. The combined factors must be considered in any evaluation of the immediate or future impact of a uranium mill on its surroundings.

2.4 Nonradioactive Components

The preceding discussion has emphasized the radioactive components of mill waters. The same sources and considerations also govern non-radioactive components of the ore, such as selenium, molybdenum, or vanadium, which could be used as tracers of translocation of mill wastes from the mill site. Effluent streams and sources from a uranium mill thus contain essentially two properties that can be used to monitor the amount and extent of mill-derived impurities added to the environment surrounding the mill: 1) the radioactive isotopes of the uranium decay chain; and 2) elements of the periodic table that occur at elevated concentrations within the uranium ore.

The following sections discuss techniques allowing the natural background to be discerned in various sample types and locations surrounding an operating mill. Methods are also briefly discussed by which the background concentrations of mill-contributed material can be determined prior to start of operation.

3.0 EFFLUENTS

With regard to measuring the background and contaminants around a uranium mill, the objectives were viewed as follows:

- Determine the ground level concentrations of contaminants whose origins were not due to the presence of the uranium mill but due to natural background and other neighboring sources.

- Determine the contribution of the uranium mill to ground level concentrations of the same contaminant species.

Most of the discussion in this section concerns an existing mill. The methodologies that are discussed and recommended use presently available conventional equipment. Although the simplest method of measuring background and mill contributed material to that background uses thermoluminescent dosimeters (TLD), airborne sampling and its associated meteorology will be discussed first. This is due to the fact that the simple TLD method requires some meteorology criteria for its installation and the fact that it measures primarily airborne ^{222}Rn and its daughters. As will be seen, much of the strategy has already been used successfully in other studies of radon emissions.

In this section, the uranium mill site is seen as an area source of fugitive emissions. As previously discussed, principal sources of ground level airborne contaminants near the mill site boundaries are anticipated to be: 1) emanation of ^{222}Rn and the wind and mechanical resuspension of particulates from tailings piles, ore stockpiles, and contaminated roads and soils; and 2) fugitive emissions from milling operations. The recommended strategies are seen as being applicable to an area source allowing long-term measurements during all meteorological conditions. These strategies allow determination of contaminant concentration generated by the mill operation in a given direction from the mill. Missing the plumes' passage in one direction does not invalidate the measurements made in other directions; thus, the strategy's purpose is not to measure total mass flux from the mill but to measure the ground level contaminant concentration in a given location relative to the mill, corrected for contributions from unrelated sources.

The strategies developed in this report are not formulated to provide data required for determining:

- Vertical concentration profiles
- Specific respirable particulate concentrations
- Total source strength
- Total mass contribution to the universal atmospheric burden
- Quantity of stack emissions

- Downwind dispersion from fugitive or stack emissions
- Ground level concentration in all directions

Moreover, the emphasis is not on detailed sampling procedures applicable to every mill but rather on sampling strategy. This approach has been employed since the mills using the sampling strategy are not all alike in character or location, and the strategy for a particular mill must be site specific.

3.1 Measurement Approaches

Three general approaches can be considered in formulating a methodology for sampling airborne contaminants. The first approach samples and measures contaminant concentrations at locations around the mill site while collecting wind speed, wind direction, and atmospheric stability data. Using the meteorological data and an assumed normalized source strength, the ground level concentrations at the sampling locations can be predicted using models. The predicted concentrations can then be compared to the measured values, adjustments can be made to the initial source strength estimate and downwind predictions, and then the residuals can be attributed to background. The weakness of this approach is the reliance on models which have large uncertainties in the predictions and are only suited for flat terrain. In fact, most of these models presently provide only an order of magnitude estimation of material transported to an environmental location. This would make resolution of background from the measured values very difficult.

The second approach measures background contaminant concentration at a site remote from the influence of the mill operation. The remote background concentration is then subtracted from concentrations measured at locations around the mill boundary. The difficulty with this approach is in finding a remote area where the background is the same as at the mill location with respect to meteorology and proximity to other sources of contaminants. A valid application of this approach is where a pre-operational survey can be conducted at a future mill site. Then a remote background sampling location can be found that correlates well with the background at the future site. The validity of the remote station may be subject to change with time as the region becomes more

developed. As will be discussed later in a separate section, one can either measure the mill-contributed radionuclides as a function of soil depth or use ^{210}Pb concentrations in ratio with specific effluent radionuclides or trace elements to provide a measure of the pre-mill background. The procedure does have some disadvantages as will be discussed later.

The third, and recommended, approach for sampling airborne particulates, ^{222}Rn and its daughters measures background upwind of the mill site and mill-originated contaminants downwind of the site. The upwind/downwind approach is most often applied to short-term sampling in one wind direction. The upwind/downwind method for measuring source strength of fugitive emissions has been thoroughly documented by the Environmental Protection Agency (EPA).⁴ The short-term sampling application of the method has been used previously around tailings piles at uranium mills by Bresling and Glauberman⁵ and more recently by Sehmel⁶. The upwind/downwind approach becomes more complicated when all wind directions and long-term sampling are to be considered. The upwind/downwind strategy proposed in the next section is versatile enough for long-term sampling around uranium mills and can be easily modified as needs change.

3.2 Sampling Strategy

Ideally, background atmospheric sampling in the environs of a uranium mill would never entail sampling air that had traversed over the area designated as, or affected by, the mill. From a practical standpoint, this ideal cannot be achieved. First, even though a background sampling station is operated only when it is "upwind" of the mill, there is no assurance that winds in the area have not followed a trajectory which has passed over the mill site. For instance, a sampling station directly downwind of a source can, following a 180° shift in wind direction, be defined as an upwind or background sampling station; yet contaminated air sampled only moments before will be recirculated across that sampler.* Secondly, at least for particulates, the edge of a mill-affected

*For simplicity in this discussion, the term "background sampler" or "contaminant sampler" is generally used in the singular. However, since sampling for trace elements, dose, and gaseous and particulate radioactive material implies different samplers, more than one type of sampler is intended.

area is not a sharp boundary. In winds high enough to cause resuspension of material previously deposited, an upwind background sample can include particulates whose origin was the mill site.

If one attempts to minimize the recirculation and resuspension difficulties by taking background samples at greater distances from the mill site, other questions arise. Is the distant background pertinent to the mill site? Is there another mill or geologic formation in the area which contributes to the measured "background"? Furthermore, sampling far from the mill offers more logistic, electric power, and security problems.

Operation of a large number of wind-direction controlled samplers (for instance one hundred) would offer a means of investigating background airborne concentrations. If each sampler were activated only when it was upwind of the mill, it would include primarily a background effect. The samplers near the mill would undoubtedly include some mill-generated concentrations. The samplers kilometers from the mill would be less affected by the mill sources under study but would embrace the problems mentioned in the previous paragraph. A careful assessment of the concentrations from all these samplers would yield a meaningful background; but the cost of obtaining, installing, operating, and maintaining such an ambitious sampling grid at each mill site would be prohibitive, probably exceeding \$250,000 in capital and two man years per year in maintenance. (Itemized costs are shown in later sections.)

Despite the recirculation and resuspension difficulties mentioned earlier, a sampling strategy can be proposed for near mill sampling which minimizes these difficulties and permits reasonable computations of atmospheric radionuclide concentrations due to the mill. It also allows data obtained from soil, sediment, and plant sampling to be evaluated with respect to its source, mill, or background.

The philosophy proposed for sampling background differs from that felt most appropriate for sampling background plus mill-related contaminants. For background sampling, it is better to miss sampling some valid background atmosphere than it is to sample longer or sample a greater area at greater risk of inadvertently sampling some mill-related contaminants. Conversely, for sampling mill plus background, it is more

prudent to sample continuously and collect a fraction of all mill-originated contaminants regardless of how they arrived at the sampler rather than to sample selectively (such as limited wind directions) at the risk of missing some mill-related contaminants. In view of these philosophies, some possible strategies will be assessed in the next subsection.

3.3 Upwind/Downwind Strategies

Several strategies are shown schematically in Figure 1 for near-mill background and contaminant air sampling. A contaminant (mill and background contributions) sample will be designated by a "C" and a background sample by a "B". The number of C and B samplers are not the total number recommended but are those needed to depict the strategy.

Strategy A is a scheme that is proposed for first consideration. The procedure is to operate an omnidirectional sampler, C_1 at all times, B_1 only when the wind is blowing with a northerly component, and B_2 only when the wind has a southerly component. The sum of operating times of B_1 and B_2 should equal the operating time of C_1 . The background seen by the omnidirectional sampler C_1 in terms of concentration units is calculated from the samples taken by the background samplers B_j . If all three samplers operate at the same flow rate, the total background is calculated by Equation 1.

$$\text{Background concentration} = \frac{\text{Quantity } B_1 + B_2}{\text{Volume Sampled } B_1 + B_2} \quad (\text{Eq. 1})$$

The sampled volume for each sampler is calculated by multiplying sampling flow rate by sampler operating time. If dose is the measured quantity, the background seen by the omnidirectional sampler is again found by summing the dose measured at B_1 and B_2 .

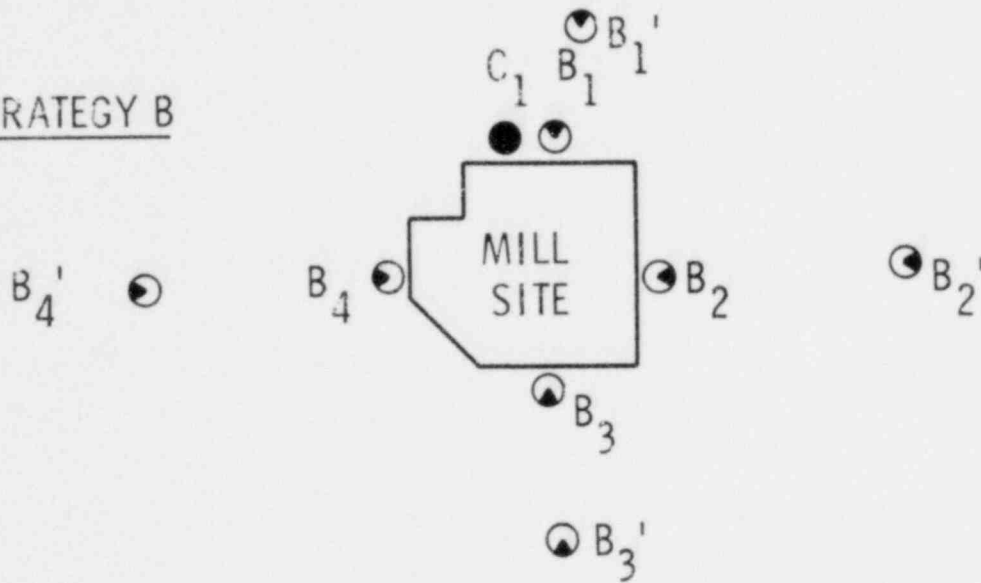
$$\text{Background dose} = \sum_{j=1,2} \text{Dose, } B_j \quad (\text{Eq. 2})$$

The background dose or concentration is then subtracted from the omnidirectionally measured dose or concentration at C_1 . The dose would be measured by a meteorology-controlled TLD system. The TLD would be exposed to airborne ^{222}Rn and its daughters only when the wind was blowing in the directions as discussed for air sampling.

STRATEGY A



STRATEGY B



STRATEGY C



C — OMNIDIRECTIONAL SAMPLER

B — DIRECTIONAL BACKGROUND SAMPLER

FIGURE 1. A schematic of three possible strategies for placement of near-mill background and contaminant sampling

The first weakness of this inexpensive scheme is that prior emissions from the mill contribute to the background at samplers B_1 and B_2 , unless they are located at least 5 km from the mill. A second weakness of Strategy A is that the directional background samplers operating over 180° of wind direction are more likely to sample mill-contaminated atmosphere than they would be if the sampling angles were restricted to, say, 90° . The net contribution of the mill to sampler C_1 would be underestimated when B_1 and B_2 overestimate background. In fact, since C_1 is omnidirectional, background samplers to the north and south of the mill site are no more pertinent than would be background samplers to the east or west of the mill. Use of a number of directional background samplers surrounding the mill would give a more representative value of background than that based on only the two samplers, B_1 and B_2 . This leads to a more expensive Strategy B as shown in Figure 1.

In Strategy B the background is determined from several directional samples, B_{1-4} and B'_{1-4} , arranged around the mill site. The operation of the B samplers is restricted to narrow vectors of wind direction in order to lessen the impact of inadvertent sampling of mill-contaminated air. The omnidirectional samplers, C_j , which sample both background and mill-contaminated atmosphere, can be arranged as desired in directions of concern from the mill site. When the omnidirectional sampler is placed at the mill boundary, it samples mill-contaminated atmosphere over roughly half the compass. A comparison of the B_j - B'_j samplers will quantify that amount of material the B_j sampler receives which resulted from old contamination of the area. In this fashion, the original background, the added mill background, and the material presently leaving the mill can be calculated. The B' samplers are not required if one wants to measure only the present mill contribution to the background. This strategy better satisfies the philosophy expressed above concerning background and contamination sampling than Strategy A and is the recommend strategy for attempting to use air samplers to determine the present and past mill contribution to background. The data reduction is essentially the same as for Strategy A. More discussion in implementing this strategy comes later.

Strategy C shown in Figure 1 is one that intuitively seems most appropriate. In this strategy, both the C and B type samplers are all

activated by selected wind directions. This approach presumes that wind direction is unchanging as air flows over the mill site. Thus, C_1 and B_2 are activated for the same time increment within a given wind direction sector, and other sampler pairs such as C_2 and B_1 are similarly activated for other wind direction ranges. The contaminant and background samplers comprising a pair have the same operating time. Subtraction of background dose or concentration from the downwind C sampler is then simplified. This approach has its greatest merit when a large directionally-dependent variation in background or source exists. If the "straight-line-wind" assumption is true, then the contamination to background ratio is maximized. Strategy C may be appropriate if experience with Strategy B has shown that mill-originated contaminants at the site boundary are not distinguishable from background. This strategy may be appropriate because the directional downwind C_1 sampler would collect material from a smaller range of wind direction than would an omnidirectional sampler.

Strategy C has some drawbacks. As shown in Figure 1, it may be desirable to locate the samplers at a distance outside the boundary so the downwind sampler "sees" a response from the entire mill with a reasonably small acceptance angle (range of wind direction) rather than a part of it using the same acceptance angle. Placing the samplers farther from the mill will increase installation and maintenance costs. Also locating samplers farther from the site boundary (for reasons to be explained) may defeat the purpose of increased selectivity because of greater dispersion of contaminants. Controlling additional samplers by wind direction will also make this strategy more costly than the other two. Another drawback to Strategy C is that flow across the sampler network is not necessarily in a straight line, and equal sampler operating time for samplers C_1 and B_2 will not be precisely true. Weighting on the basis of operating time or sampled volume would be needed in the real situation. Finally, Strategy C is in conflict with the philosophy that sampling restricted to specific wind direction is most effective for background monitoring, while continuous omnidirectional sampling is the safest for determining background and contaminant concentrations.

Strategy B seems to be the most reasonable first approach for routine long-term site boundary sampling by a mill operator for either particulate or gaseous escapement and for interpretation of methods which measure dose. Interpretation of data obtained by sampling soil, water, and biota for mill-contributed contaminants also requires sampling and meteorological strategy of this type to properly exclude contributions from other facilities from the responsibility of the mill under study.

3.4 Implementation of Recommended Strategy B

3.4.1 Sampler Siting

Each mill has a slightly different topography and sampler location is often site specific. Some factors affecting the siting of sampler stations are:

- Location of sources
- Nature of sources
- Location of sensitive receptors
- Meteorology
- Topography
- Land use
- Other contaminant sources in the region, i.e., other mills

How some of these influences come into play will be addressed in the following paragraphs.

The sampler layout tacitly implies that the mill operator takes responsibility for all contaminant sources within the "source area" and assumes that all enclosed sources are mill-originated. Any sources within the "source area" for which the operator does not assume responsibility must either be removed from the source area or quantified by a pre-operational survey and independently monitored for changes, i.e., B versus B' data; otherwise their contribution will be counted as mill-originated by this strategy. Careful selection of sampling sites is important because the mill operator does not generally want to include in his reported release background omissions for which he is not responsible. Thus, the locations of the omnidirectional contaminant samplers and the background samplers are determined by the source area.

A reasonable first place to consider sampling sites is the legal boundary of the mill operation; however, in some cases, offsite contamination originated by the mill may be considered the mill operator's monitoring responsibility. In such cases, the boundary of the source area is moved to enclose such contaminated areas. If that results in moving the source area boundary out more than, say, 0.5 km, then a third sampling array surrounding the extended source area is a better alternative, i.e., B' type samplers.

Another consideration in siting samplers is the location of neighboring contaminant sources of significance such as another mill or a mine. Generally, a directional background sampler should be located along the pathway contaminants would follow from the offsite source to the site. Thus, the contribution from the offsite source will be included in the overall background and will be differentiated from the site emission.

Meteorology is a factor in the choice of sampling sites. Courtney⁷ suggested that sampling be performed in the direction of the prevailing wind and in the wind direction corresponding to stable conditions during which atmospheric dispersion is least effective. Generally, for point sources such as stacks, there is an optimum distance from the stack at which the pollutant concentration is at a maximum for a given type of atmospheric stability. The optimum distances can be calculated from the source characteristics and meteorology.⁸⁻⁹ These estimates are not required, however, for implementing a strategy for the ground level resuspension or emanation sources which are of primary importance in this study. For ground level releases, the ground level concentration is generally highest closest to the source.

The distances from the source where the contaminant is at its lower limit of detection can be estimated for various stability conditions. A method for computing these maximum distances from point sources is given in Reference 4. Such a method applicable to the uranium mill resuspension and gas emanation problem is not currently available but may be a simple derivation based on using a line or area source dispersion model. The experience of Sehmel⁶ near a mill tailings pile indicates that at a downwind distance of 4 to 5 km the concentration of total suspended particulates (TSP) approached background.⁴ Although similar detailed information¹⁰ for radionuclides is just now becoming available,

Breslin and Glauberman⁵ demonstrated the concentration decrease with distance from tailings at inactive mills

Topography plays a role in sampler siting, as well as influencing meteorology. Generally, air samplers should be located on flat open ground and some distance away from low structures. Sehmel's experience⁶ indicates that particulate samplers should be located at least 200 m away from a tailings pile. In his experiments, large quantities of re-suspended material from a tailings pile often overloaded or otherwise rendered inoperable sampling equipment operated at 40 cfm within about 100 m. This 200 m rule is probably applicable to siting samplers near areas where the soil is as readily resuspendible as the dry tailings.

In addition to the above siting criteria, there are some other guidelines pertinent to the omnidirectional samplers. The omnidirectional samplers should be located in the direction of sensitive receptors such as housing, farming, water supplies, business districts, and other areas of non-occupational exposure. A minimum number of four omnidirectional samplers seems reasonable.

There are some guidelines pertinent to the siting of background samplers in addition to those given above. In terms of installation costs, it is more economical to place a background sampler adjacent to an omnidirectional sampler. With this arrangement, the measured background component is also more representative of that seen by the omnidirectional samplers. It seems appropriate that there be a minimum of four background samplers. The wind directions during which they operate (i.e., acceptance angle) should total the 360° of the compass. Overlapping acceptance angles should be avoided unless all 360° of the compass are sampled the same multiple of times. The background samplers should be located such that they collect material external to the source area.

If there is some difference in background between B' and B arcs (or B'' - B' arcs) indicating some source in the intervening distance, it may be difficult to show whether the contamination is mill originated.

If it is determined that the contamination between the B, B', B'' arcs is mill originated, then a decision must be made regarding the mill operation's responsibility for further monitoring of that contaminating source. The following are two resulting alternatives:

- A. The mill operator is not to assume further responsibility for that source.
- B. The mill operator is to assume areal monitoring of the source as part of his overall program.

If (A) is chosen, then there is justification for discontinuing operating the B' and B" arcs as an unnecessary expense. If (B) is chosen, then at the very least the mill operator is required to operate C' samplers along roughly the same arc as the outermost B samplers. Operating C or omnidirectional samplers on the outer B arc is the only way for the mill operator to credit and monitor that intervening contamination as part of his source area. Furthermore, if (B) is chosen there is justification for the operator to discontinue the inner arc of C samplers (except those near sensitive receptors) in favor of just operating the C' arc. Eliminating the inner arc of B and C samplers effectively results in moving the single arc monitor concept farther out from the mill to encompass a larger source area.

The mill operator might then ask, "Now do I have to erect other B' arcs to probe farther outward to see if my source area is even more extended? How far do I have to go?" These are valid questions and can only be answered by future research results.

If by luck the mill operator is only required by law to monitor to some distance limit (such as a property line) thus absolving him of responsibility for a source of airborne contamination outside those limits, there seems to be little reason for the operator to set up B' and C' arcs for a short time period to explore an extended source area to cover himself if the rulings change.

3.4.2 Sampler System Operation

Some of the basic equipment required to implement the recommended sampling strategy are:

Omnidirectional samplers

Directional samplers

Wind direction control system

Elapsed time indicators

This equipment is necessary for measurements of airborne particulate, radon, or dose (thermoluminescent dosimetry--TLD) types of samplers. More details on the equipment, their commercial availability, and approximate costs will be given later.

In general, the sampling array will be continuously activated. All omnidirectional samplers and one directional background sampler(s) in an array will operate at all times. If samples are taken for at least a week before changing collection media (filters, TLDs, etc.) all background samplers will probably have been activated. Although the recommendation is to change sample collection media on a monthly basis, the filters may have to be changed more often due to their dust loading.

A control system is required to activate the appropriate background sampler according to the wind direction. An elapsed time indicator should be included to indicate the amount of time the wind was in each sector and hence the time the matching background sampler operated. A time delay will be required in the direction control circuitry so a new background sampler is not activated until the wind is definitely within its sector instead of switching background samplers on and off due to the oscillation of a wind vane at the border of two sectors.

Wind vanes have a threshold response expressed as a distance constant or a speed (usually <0.45 m/sec). (A good discussion of wind instruments is given in Reference 9.) Wind vane oscillations are also damped to varying degrees depending on the instrument's application. For use in locations where resuspended sand and soil can foul bearings, fairly rugged wind instrumentation is required.

A "calm" occurs when the wind speed is below the threshold of instrument response. During a "calm" the sampling array will not respond to subtle wind direction changes and the last operating background sampler will remain operating. Fortunately, such conditions are not favorable to the transport and resuspension of airborne pollutants; however, radon gas will still emanate from the soil and will tend to diffuse toward some sampling locations. The effect of a "calm" on sampling error has not been assessed and certainly would depend on the frequency of that condi-

tion. Some data on its local effect would arise from differences detected between B_j and B_j' .

Precipitation is another atmospheric condition that is not favorable to the resuspension of particulates and tends to retard radon emanation as well. Precipitation periods are not favorable for operating many types of air samplers. Experience shows that even in the common sampler shelters, moisture can collect on sampling filters and reduce air flow through the filter. Extensive filter dampness can cause loss of the sample, filter breakage, or chemical changes in the collected species. When the air sampler pump depends on the air passing through the filter to cool the motor, wet filters can cause burned-out motors. If this is a possibility, a moisture sensor and relay may prudently be used to turn off air samplers. An elapsed time indicator showing the length of time the array components were inactive due to precipitation may be desirable.

Some duplication in sampling is desirable to estimate the experimental sampling error.* Although several samplers are operated in the basic array, they all ostensibly sample different backgrounds or different combinations of background and source contribution--each background or combination being sampled only once. It is not expected that background and source conditions will be identical from one sampling interval to the next. Therefore, some duplication of sampling is recommended, for the first year of operation at least, in order to estimate the magnitude of sampling error. It would be better from a statistical point of view to have several duplicates of one sample rather than one duplicate each of several samples. Thus, it would seem prudent to operate two or three duplicate samplers along with either one background or omnidirectional sampler. The choice of sampler being duplicated can be changed from one sampling period to the next. Several months of such duplicate sampling may yield sufficient data to estimate sampling error.

3.4.3 Data Reduction

The aim of data reduction is to calculate the background and the mill contribution to the air concentration of specific radionuclides or dose at the location of the omnidirectional sampler. The main task is

*The sampling error is estimated by calculating the standard deviation of duplicate samples.

to estimate the pre-mill background, $B_j - B_j'$, and background B_j seen by the omnidirectional sampler. Allowing for the possibility of operating duplicate samplers and using different flow rates, the volume of air sampled and mass collected should be corrected by the ratio of omnidirectional sampler flow rate to the total background sampler flow rate for each sector. The computational scheme shown in Table 1 will aid in the calculation of background concentration applicable to a given omnidirectional sampler flow rate. If all the air samplers in the array operate at the same flow rate and if there are no duplicated background samplers, then the composition of equivalent background is simplified.

The data reduction method for dosimeters is slightly different. If more than one background dosimeter samples from a given wind sector, the dose readings are averaged for the wind sector. The average doses for all sectors are then added to calculate the equivalent omnidirectional background dose as seen by an omnidirectional dosimeter. Table 2 shows a suggested format for organizing the background dosimeter data.

Once the equivalent background concentration or dose is calculated, it is simply subtracted from that measured by the omnidirectional sampler to yield the present mill contribution. If the total operating time of the background air samplers does not equal that of the contaminant samplers, correction is required. Operating time differences may arise from equipment malfunctions or the lag time of the wind direction control system.

TABLE 2. Computational Table to Estimate the Background Concentration as Seen by a Given Omnidirectional Sampler

<u>Wind Sector</u>	<u>Operating Time, Hrs.</u>	<u>Number of Samplers</u>	<u>Sampler Flow Rate, F_B m^3/hr</u>	<u>Volume Sampled, m^3</u>	<u>Grams or Picocuries Collected</u>	<u>Corrected by Ratio of Omni. Flow Rate to Background Sampler Flow Rate, x^{F_0/F_B}</u>	
						<u>Volume m^3</u>	<u>Mass or Activity</u>
						Total Equivalent Volume	Total Equivalent Mass or Activity

Equivalent Background Concentration = Total Equivalent Mass ÷ Total Equivalent Volume

F_0 - omnidirectional sampler flow rate, $m^3/\Delta T$

F_B - directional background sampler flow rate, $m^3/\Delta T$

TABLE 3. Computational Table to Estimate Background Dose as Seen by a Given Omnidirectional Dosimeter

<u>Wind Sector</u>	<u>Operating Time, hr.</u>	<u>Dosimeters</u>	<u>Dose</u>	<u>Average Dose from Wind Sector</u>
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Sum = Estimated Background Dose

In this data reduction scheme, we have assumed that background concentrations from a particular offsite source remain constant during the traverse from the upwind background samplers to the omnidirectional samplers. Because of atmospheric dispersion, one might expect this not to be the case, particularly when the offsite source is a nearby point source. A method could be devised to factor this phenomenon into the data reduction by modifying the background components that are measured upwind from the omnidirectional sampler in computing the equivalent background. To implement this method, wind speed and stability class data must be continuously recorded. The calculations could be based on models such as those given by Turner.⁸ We assume for this report that these complex calculations are of small significance to the final result; however, the magnitude of any possible errors because of this assumption have not been assessed.

3.4.4 Simple Air Sampling Methodology

The former discussions have covered a variety of information leading toward air sampler siting. In summary, the recommended simple methodology involves meteorology-controlled air sampling followed by photon analysis of the sampled particulate material, costs \$55,000 in capital, and requires 0.75 man-years of operating expense. Photon

analysis is discussed in later sections. The air samplers should be located radially on lines extending from the mill boundary, as previously discussed. (One scheme might separate them by 60° from one another.) There would be three high volume air samplers on each line, one omnidirectional and one directional sampler at the boundary of the mill and a second directional sampler 1 km from the boundary in the upwind direction. A central meteorology station controls the air sampler operation based on the wind vector. The samplers collect the airborne particulates and will be changed on a monthly basis. The samplers would be placed approximately 1 m above ground and enclosed against weather and animal penetration. Air filters which had collected the airborne particulates are pressed into a defined geometry, typically small 5-cm diameter pellets, and are analyzed for their gamma-ray emitting radionuclides using a planar intrinsic germanium detector coupled to a 2048 channel analyzer with a hard-copy readout. The sensitivity for such a system defined as the minimal detection activity at the 95% confidence level is 1.4 dpm ^{210}Pb , 1.1 dpm ^{214}Pb , 18 dpm ^{234}U , 14 dpm ^{230}Th , 0.6 dpm ^{235}U , and 8 dpm ^{226}Ra .

The advantages of such a system are that the radioisotopes in question can be measured to their environmental levels. In most cases, the LLDs, lower limit of detection as designated by the NRC, can be achieved for particulates in air. The measurement of specific radioisotopes as a function of meteorology allows an assay of the current mill contribution to the environment versus that which has been contributed through past activities. The data allow a differentiation between mill-contributed material and that contributed from off-site sources. Data analysis can be easily computerized for interpretations at any later date through statistical criteria.

The primary disadvantages are that the system requires electrical power, mechanical and electrical maintenance, and a meteorology control device. The technology measures only radionuclides associated with air particulates. It requires radioactive standards to be used in the laboratory, as well as a license to handle radioactive material. The detector, an intrinsic germanium photon spectrometer, requires a source of liquid nitrogen and a dedicated counting room. As discussed previously in the text, air samplers may require additional capital modifications

to handle excessive dust loads and other weather conditions. Topography effects must be evaluated for some sampler sites.

A further level of sophistication involves the measurement of air samples, biota, soil, and water. The methodology will require \$90,000 in capital equipment and a minimum of two man-years of dedicated time, in addition to dedicated chemistry and counting laboratories.

The following section gives examples of the application of the simple air sampling methodology and its application within siting criteria and data reduction methods.

3.5 Example of Strategy for Airborne Contaminants

The hypothetical mill site used for this example is sketched in Figure 2. No topography is shown but the locale shown is intended to be gently sloping arid land.

The locations of omnidirectional and background samplers are shown symbolically in the figure. The samplers could be dosimeters, air samplers, or other gas collecting media. A specific discussion of dosimetry is found in Section 4.0. The samplers shown in pairs are located adjacent to each other. The second ring of background samplers is also shown. Samplers C₁ and C₇ are located to measure mill-originated airborne contaminants dispersed in the direction of the company housing. Sampler B₁ samples background from the winds out of 0-60° and is located near C₁ for convenience. C₂ is located to sample contaminants dispersed in the direction of the small farming village by the prevailing wind. C₂, C₃, and C₄ are located outside the region of tails which have been deposited outside the property line by wind and water erosion. Thus, mill-originated contaminants determined from C₂ through C₄ will include the contribution of this offsite source of resuspendable contamination because this hypothetical mill operator assumed responsibility for it as his source. C₃, C₄, and C₅ are sited to allow an estimate of the concentration of mill-originated contaminants dispersed into the range area. Samplers C₂₋₅ and B₂₋₅ are sited between 200 and 400 meters from the tailings or similarly resuspendable soil. Samplers B₆ and B₇ are located to sample incoming offsite contamination from the abandoned mill and to account for it as background. C₅ is sited to distinguish between

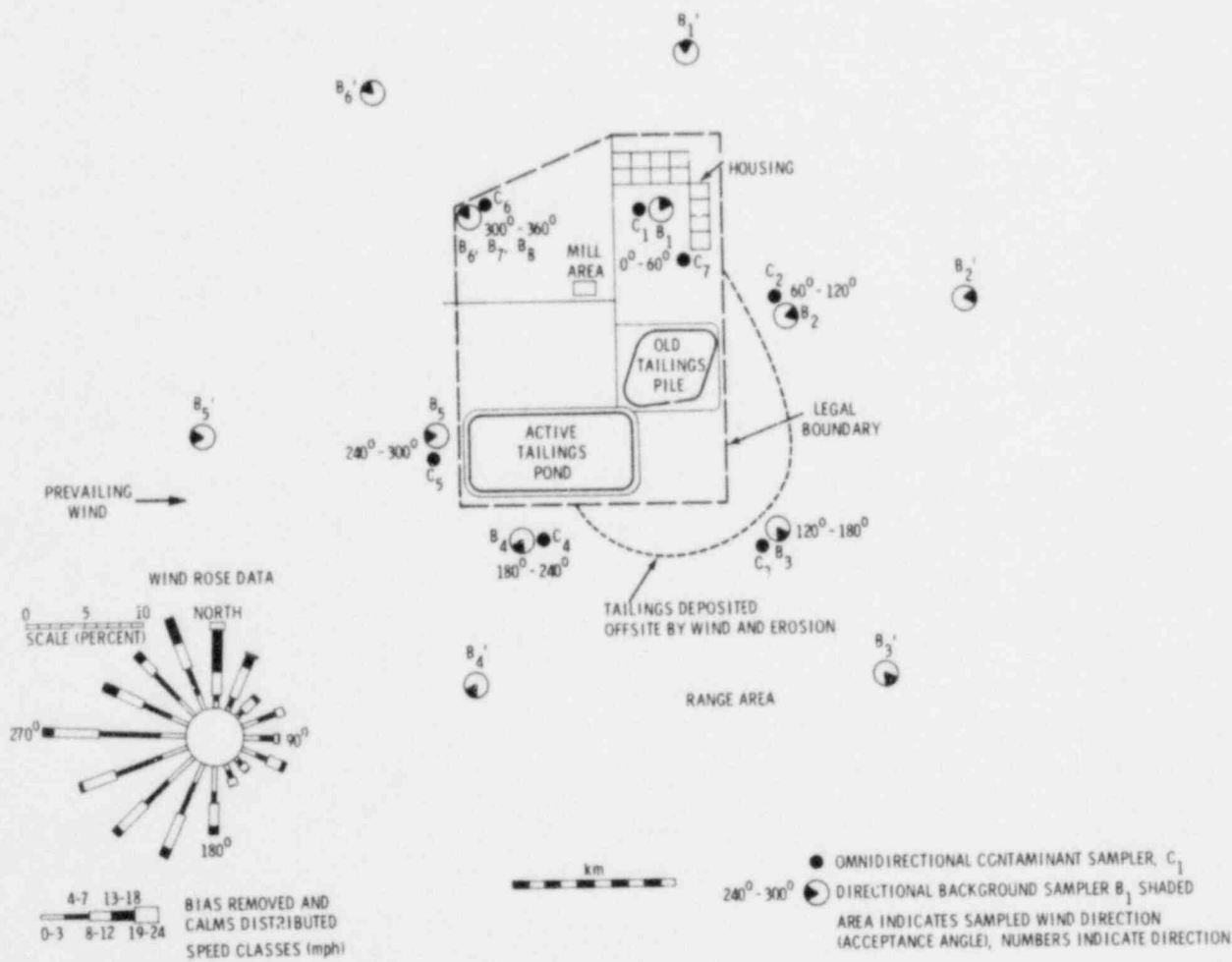
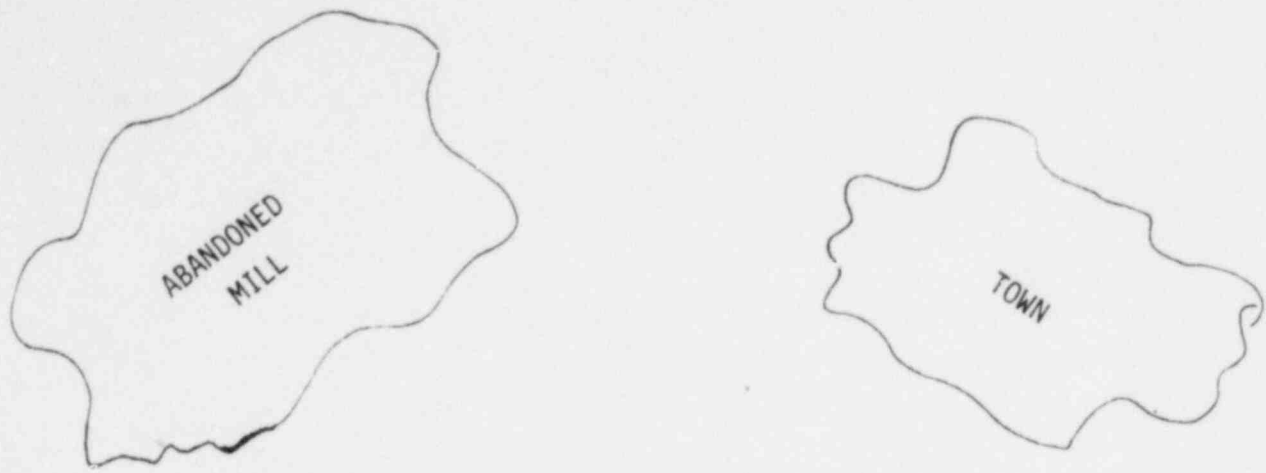


Figure 2. Hypothetical uranium mill site
 Omnidirectional contaminant Sampler C₁
 Directional background Sampler B_i shaded area indicates
 sampled wind direction (acceptance angle)
 Numbers indicate direction

mill-originated contaminant concentration and the offsite-contributed concentration from the abandoned mill which is thought to be significant. Samplers B₇ and B₈ are duplicates of B₆ and operate from the same wind direction sector as B₆. Samplers B₁₋₆ and B'₁₋₆ operate from individual wind direction sensors. The acceptance angles of the background samplers were determined by dividing the 360° evenly into six 60° sectors. The background samplers were located around the source areas so their acceptance angles faced away from the source area and they shared electrical service with contaminant samplers. An attempt has been made to locate the samplers on the major wind rose.

The hypothetical background sampler data from the operation of this sampling array are summarized in Tables 3 and 4 and the effective backgrounds applicable to Sampler C₃ (which operated at 60 m³/hr) in terms of dose and concentration are calculated. Thus, Table 3 shows the calculations of the background ²³⁰Th concentration of 6.1 x 10⁻³ pCi/m³ that should be subtracted from the concentration measured by C₃ (and any other 60 m³/hr sampler) to yield the concentration resulting from mill-originated ²³⁰Th for this sampling time. Differences between B and B' data detail the background material added to that area from the mill since its start of operation. For air sampling and dosimetry by TLD, it is felt this latter quantity may not be measurable by this technique. The values used for calculating sampling errors are also included in Tables 3 and 4.

TABLE 4. Computation of Equivalent Background Concentration for Air Sampler C₃

Flow rate of C₃ = 60 m³/hr - F₀

Corrected by Flow Rate Ratio F₀/F_B

Wind Sector, Degrees	Operating Time, hr.	Samplers	Sampler Flow Rate, F _B m ³ /hr	Volume Sampled, m ³	Volume Sampled, m ³	Collected, Picocuries ²³⁰ Th	Collected Picocuries ²³⁰ Th		
0-60	100	B ₁	60	6,000	6,000	5	5		
60-120	75	B ₂	60	4,500	4,500	7	7		
120-180	60	B ₃	60	3,600	3,600	10	10		
180-240	125	B ₄	60	7,500	7,500	13	13		
240-300	235	B ₅	40	9,400	14,100	21	31		
300-360	105 ea.	B ₆	60	6,300	16,800	6,300 Ave.	195	505	189 Ave.
		B ₇	40				100		
		B ₈	60				210		
	700				42,000		255		

Equivalent Background Conc. = 6.1×10^{-3} pCi/m³ (6.1×10^{-15} μCi/ml)

NOTE: The sample standard deviation is calculated from the measured concentrations of B₆₋₈ (0.031, 0.024, 0.033, respectively) and their average (0.029). The resulting sample standard deviation is 0.0047 or 16% of the mean. The population standard deviation is 0.0038.

Equation:
$$S_x \left[\frac{1}{N-1} \sum_{i=1}^n (X_i - \bar{X})^2 \right]^{1/2} ; \sigma = S \frac{\eta - 1}{\eta}$$

TABLE 5. Computation of Equivalent Background Dose

<u>Wind Sector, Degrees</u>	<u>Operating Time, hr.</u>	<u>Dosimeters</u>	<u>Dose mRad</u>	<u>Average Dose from Wind Sector, mRad</u>
0-60	100	B ₁	27	27
60-120	75	B ₂	35	35
120-180	60	B ₃	50	50
180-240	125	B ₄	100	100
240-300	235	B ₅	120	120
300-360	105 ea.	B ₆	150	
		B ₇	120	150 Ave.
		B ₈	180	—
Equivalent Background Dose				482 mRad

Note: Sampling standard deviations calculated from B₆, B₇, B₈ equals 30 mRad or 20%. The population standard deviation σ is 25 mRad using equation in Table 3.

3.6 Equipment

The basic types of sampling equipment that are used with the latter background measurement strategy are particulate samplers, dosimeters, radon samplers, and meteorological instruments. Some estimates of the equipment costs for setting up a sampling array like that used in the example will be given. It is expected that installation (labor + materials) costs will be a significant fraction of the overall cost of the proposed sampling method; however, these costs have not been estimated at this time since they (labor costs) vary significantly by area.

3.6.1 Particulate Samplers*

Without the benefit of an analysis of how sample flow rate will be determined by the requirements of the lower limit of detection (LLD) as defined by the NRC--see Section 6.1) and analytical methods (as in the previous section), only equipment which will be suitable for many cases will be discussed. The high volume air sampling equipment commercially available for use as specified by the EPA method for ambient particulates¹¹ will serve as the basic piece of equipment. The commercially available high volume samplers (Hi-Vols) operate in the range of 25-100 m³/hr (15-60 cfm) and are built for continuous long-term sampling. The typical Hi-Vol consists of a blower mounted beneath a holder for an 8 in. x 10 in. filter (see Figure 3). The filter is mounted "open face" and horizontally. This sampling unit is then mounted in a sheet-metal shelter. Flow rate and motor speed control are accomplished with a variable transformer and flow rate is measured using a rotameter through which a fraction of the motor exhaust air passes. The rotameter requires periodic calibration using a calibrated orifice placed over the filter. Under typical ambient dust loadings, the flow rate requires at least a daily adjustment. Reliable automatic flow-rate controllers are commercially available for Hi-Vols, and, for long-term sampling, are usually well worth the extra cost in maintenance time saved. The controllers maintain constant airflow in terms of some standard conditions (usually 25°C, 760 mm Hg) regardless of ambient temperature and pressure fluctuations. The current average cost for this type of sampler (purchased in lots of 20) is \$650 with the automatic flow controller and \$300 without.

*All cost data in this report is referenced to January 1979.

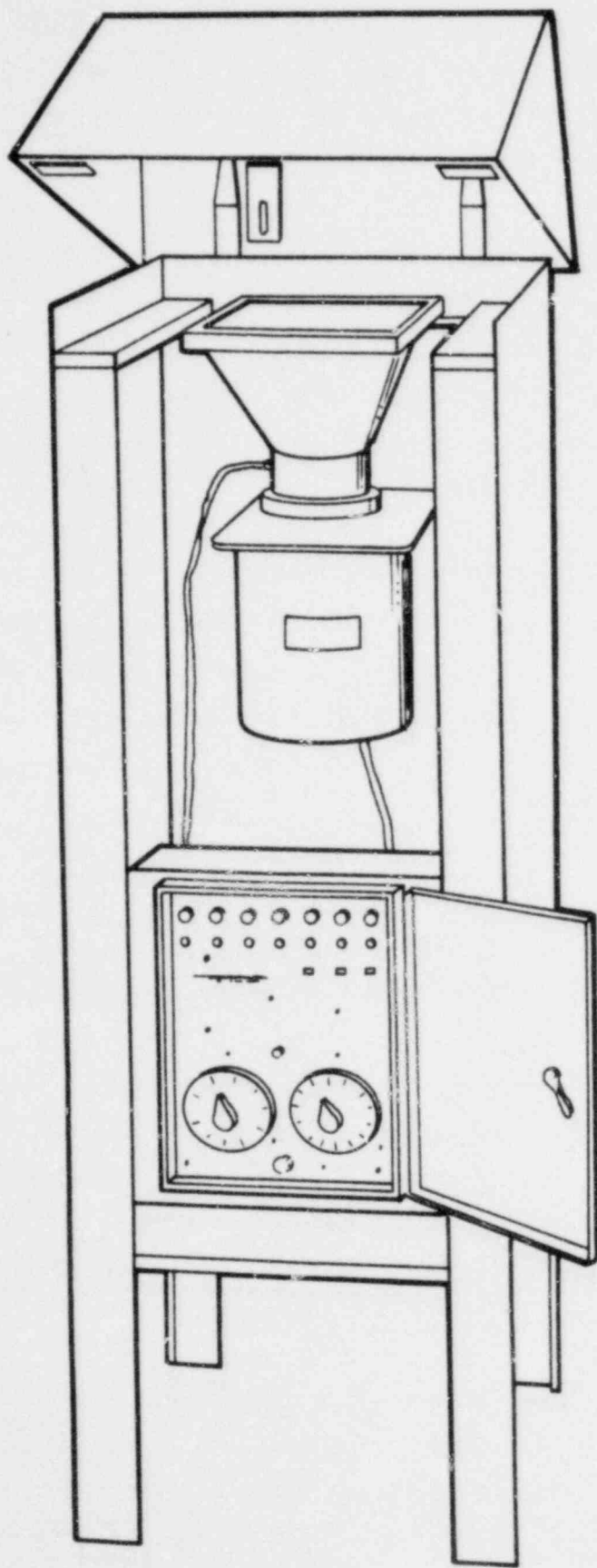


FIGURE 3. Hi-Vol Sampler Shelter with Front Removed to Show Main Components

Another type of sampler similar to the Hi-Vols that may be considered used a 10-cm diameter filter and a flow rate of 8-20 m³/hr (5-12 cfm). This sampler is mounted in a shelter similar to the Hi-Vols and features automatic flow control. There are a variety of other air samplers that are designed for portability but would need to be enclosed in shelters (see Table 5).

The standard Hi-Vol requires some special modifications for use as a directional background sampler. The recommended strategy requires that the sampler be frequently turned on and off, and that the sampler should be sealed to prevent the collection of unwanted wind-blown dust during "off" periods. There is only one domestic manufacturer (General Metal Works) currently producing a device claiming to prevent the collection of unwanted particulates. The device is basically a shield that covers the collection filter during "off" periods. This device is available as an accessory at a current cost of \$195.

For the sampling array given in the example, seven omnidirectional and fourteen directional samplers were required for an approximate cost of \$14,700 including automatic flow controllers.

In instances where dust loading is so severe that filters would have to be changed more frequently than desired, the location of the sampling station should be examined to see if it is too close to a major resuspension source. If the problem can be corrected by moving the samplers 100-200 meters, they should be moved. If the problem is unavoidable, then some modification of the sampling method is in order. One modification commercially available is to pass the incoming sampled air through a cyclone pre-separator with an appropriate particle size cutoff to reduce the total loading on the filter. The coarse particulates extracted by the cyclone are saved and can be analyzed in addition to the filter sample. A commercially available device mounts over the 8 in. x 10 in. filter holder and replaces the shelter roof (see Figure 4). A rotating cylindrical turret has a horizontal inlet nozzle pipe on one side and a wind vane on the other so the nozzle points into the wind. Air is sucked through the nozzle into the cylindrical turret which is a cyclone separator. Air and particulates not settled in the cyclone separator pass down through the 8 in. x 10 in. filter of the Hi-Vol. The aerodynamic equivalent 50% cutoff particle diameter for the cyclone is 5.5 microns

TABLE 6
COMPONENT VENDORS

Supplier	Hi-Vol sampler	Flow controller	Flow calibrator	Fugitive dust cover	Cycle Pre-separator	Hi-Vol filters	Wind direction activated controller	Precipitation activated controller	Wind spread activated controller	One-stage cascade impactor	Particulate Monitor	Radon monitor/sampler
BGI, Inc.	X	X	X			X				X		
Sierra Instruments	X	X	X		X	X				X		
General Metal Works	X	X	X	X		X				X		
Research Appliance Corp.	X	X	X			X				X	X	
Microchemical Specialties (MISCO)	X	X	X			X	X	X	X	X		
Anderson 2000 Inc.	X	X				X				X		
Kurz Instruments		X	X									
Science Associates							X	X	X			
Weather Measure							X	X	X			
Met One							X		X			
Climatronics							X		X			
Staplex	X	X										
Thermosystems, Inc.											X	
Extranuclear Labs, Inc.											X	
GCA											X	
Precision Scientific	X											
Phillips											X	
Wallace Fisher											X	
Lear Siegler											X	
RADeCO	X	X									X	
Victoreen											X	
Eberline											X	X
Nuclear Measurements, Inc.											X	
Gelman						X						
Millipore						X						
Schleicher & Schuell						X						
Reeve Angel						X						
Nuclepore						X						
Environmental Measurements, Inc.												X

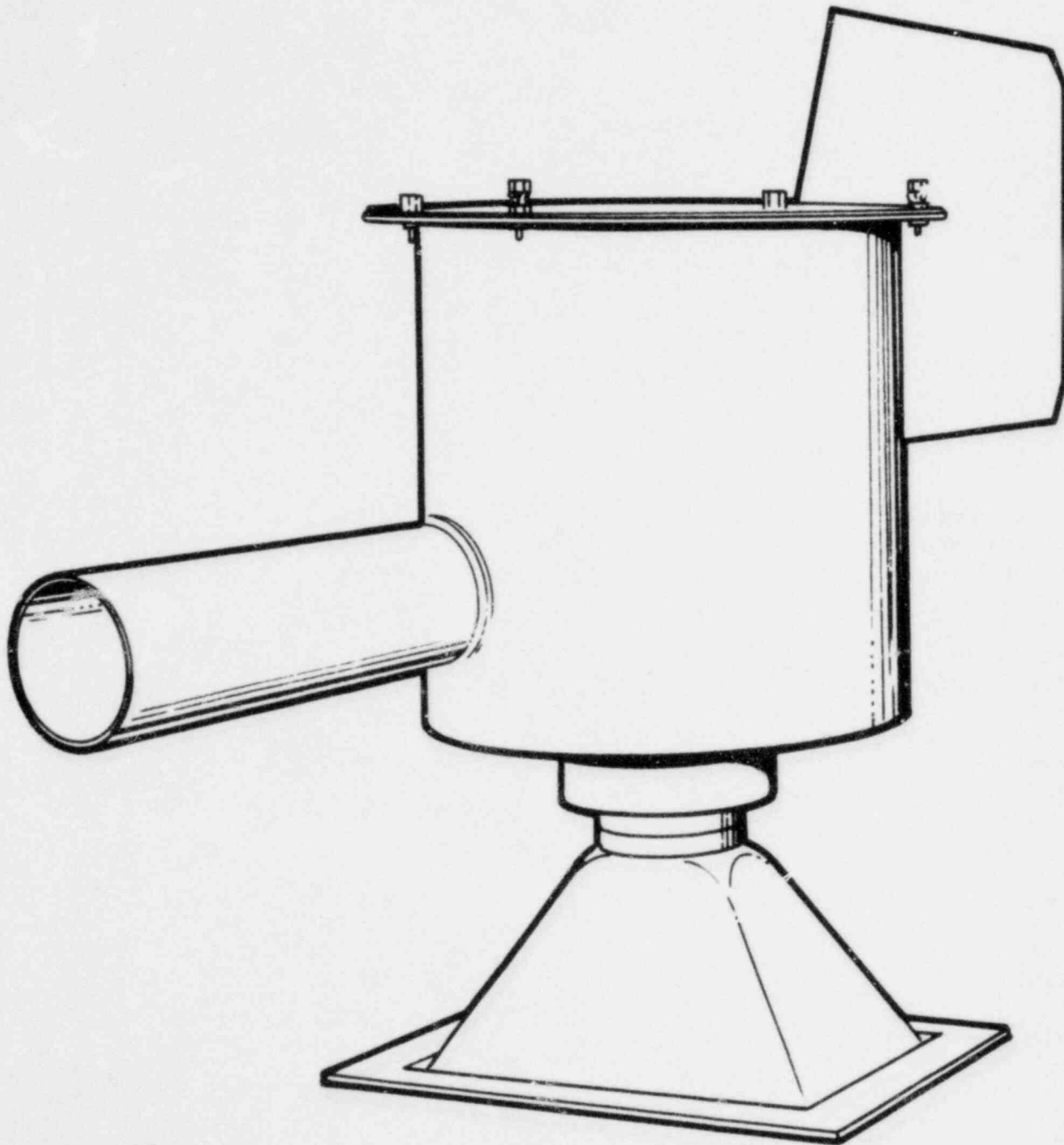


FIGURE 4. Cyclone Pre-separator

at 40 scfm and 11 microns at 20 scfm. One disadvantage of the unit is that no automatic closure is available on the inlet, and experience shows that wind-blown dust will pass through the separator onto the filter when the sampler is turned off. Any closure would have to be shop built. The unit currently costs about \$1,000, so if the sample overloading can be easily cured by moving the sampler, additional expense can be avoided.

The filters used with the particulate samplers should have a high efficiency (>99%) for fine particulates (0.3 micrometer diameter). The common filter media are made of cellulose, glass, or quartz fibers of varying purity. The type of filter should be selected with sampling efficiency and compatibility with analytical technique in mind. The cost of filters will range from 25¢ to 50¢ each, which is nominal compared to the equipment cost.

3.6.2 Radon Samplers

For the measurement of radon, a continuous sampler would be the instrument of choice rather than trying to use a series of grab samples followed by an analysis. Although present-day equipment is still in a state of development, there are commercial instruments available.^{12,13} One system¹² (Model RGM-1) samples air through a scintillation cell, a pump, and flow meter. It is available from Eberline Instruments for about \$6,000. The alpha particles from atmospheric ²²²Rn and its progeny strike a ZnS(Ag) phosphor. The light is detected, processed, and the data supplied to a digital recorder. The entire unit is installed in a field package the size of a suitcase. The second system¹³ (Model RE 350, Bendix Corp.) operates in much the same fashion allowing radon measurements from soil, air, or liquid media. It allows radon concentrations from 1-40,000 pCi/l to be measured. If monitoring of just radon and its daughters were demanded by NRC, at least six instruments would be required (total cost = \$36,000 capital). The vast amount of data supplied by the instruments would require 0.5 man-years per year of effort to interpret for background assessment.

3.7 Measurement Accuracy

A few comments on how the various factors and decisions affecting the overall accuracy of measurements are needed here as they relate to the selection of sampling equipment. The first decision affecting equipment selection is the lowest concentration that is to be measurable. Another important decision is defining "measurable" in terms of statistically meaningful parameters such as standard deviation and confidence level. The NRC¹ has recommended "Lower Limits of Detection" (LLD) in concentrations for uranium mill site boundary measurements of ²³⁰Th, ²²⁶Ra, ²²²Rn, ²¹⁰Pb, and natural uranium, and ask that the actual LLDs be calculated for the procedures in use at each mill. Concurrently, NRC recommends some limits on the accuracy of sample analyses and requests error estimates be routinely made. The amount of sample required to satisfy the analytical accuracy desired is a function of the analytical method employed. In general, the accuracy of a radiochemical analysis by counting disintegrations is a function of the sample counting time, the background, and the gross counting rates measured. The 95% confidence intervals, when there are greater than 50 total counts, can be estimated by:¹⁴

$$\text{Interval limit} = 1.96 \sqrt{\frac{(\text{net} + \text{background}) \text{ c/min}}{\text{sample counting time}} + \frac{\text{background c/min}}{\text{background counting time}}} \quad (\text{Eq. 4})$$

This equation arises from:

$$S_{\text{net rate}} = \frac{R_B}{t_B} + \frac{R_G}{t_G}$$

where R_B = background counting rate

R_G = gross counting rate (background + net)

t_B = counting time for background

t_G = counting time for gross

S = standard deviation of net count rate

According to Student's T, the 95% confidence interval limits for the net count rate are a function of the total counts; i.e.,

$$95\% \text{ conf. interval} = t_{\alpha} \times S_{\text{net rate}}$$

when the number of counts is infinite, $t_{.025} = 1.96$. For example, when the counts = 29, $t_{.025} = 2.045$.

Depending on the counting method, confidence level, and interval limits desired, the minimum amount of material required in a sample for acceptable analysis can be calculated. For example, let's assume 30 dpm of a radionuclide is the minimum necessary for our analytical scheme. Let's further assume that $2 \times 10^{-15} \mu\text{Ci/ml}$ is our desired LLD. The minimum required volume sampled is related to the LLD and the minimum amount of material needed for analysis. In this instance, about 6800 m³ of air must be sampled.* Say the sampling interval is to be 30 days (720 hrs), then air sampler minimum flow is 9.4 m³/hr (5.6 cfm). If the sampling interval were a week, then 40.5 m³/hr (24 cfm) is required. It is not our intent to state here the minimum flow rate required for samplers or LLDs, but rather to point out their relationship.

3.8 Advanced Air Sampling Strategy

There are three approaches that can be taken toward more advanced air sampling programs at uranium mills. The first two are developmental and the third is research oriented. One approach is to improve the quantity or quality of the data arising from the present strategy. Another approach is to perform new tasks to collect different types of data. Research into the sources of error inherent in the recommended strategy is a third approach.

3.8.1 Data Quantity and Quality

One obvious way to improve an existing sampling program is to take more samples. In terms of the recommended strategy an increased number of vectors from the mill could be sampled by the omnidirectional contaminant samplers. Additional sampling arcs could also be added at increasing distances from the mill. Three possible benefits of these additions would be to collect data near more (or new) sensitive receptors, to shed light on how the contaminants have dispersed with distance from the mill, and to improve the quality of estimates of

$$* \text{Volume} = 30 \text{ dpm} \times \frac{\text{pCi}}{2.22 \text{ dpm}} \times \frac{\text{ml}}{2 \times 10^{-15} \mu\text{Ci}} \times \frac{\mu\text{Ci}}{10^6 \text{ pCi}} \times \frac{\text{m}^3}{10^6 \text{ ml}} = 6757 \text{ m}^3$$

environmental impact. There is no one unique expanded program of this type that could be suggested with a unique price affixed; rather, the cost would increase incrementally with the number of samples, station controllers, and associated equipment.

Another way to improve the quality of the data resulting from the recommended strategy is to implement isokinetic sampling. Wedding, et al.,¹⁵ reported significant sampling biases when using Hi-Vols for particulates over 5 μm aerodynamic equivalent diameter (AED). The amount of bias increased with increasing particle size and varied with the shelter's orientation into the wind. The superiority of a Sehmel Rotating Cowl¹⁶ design [the design that the aforementioned cyclone separator (pg. 36) is partly patterned after] was also demonstrated in the same experiments. An isokinetic sampler (see Figure 5) using a basic Hi-Vol filter holder and blower mounted horizontally behind a rectangular nozzle has been designed by Sehmel.¹⁷ The nozzle opening was designed for near isokinetic sampling within a wind speed range and incorporated a positive closure to prevent dust from entering the sampler. Three such isokinetic samplers are required to just cover the wind speed range of 3-11 m/s. A wind speed controller was required to activate the appropriate sampler. Similar samplers would be required for wind velocities exceeding 11 m/s. An isokinetic Hi-Vol with a nozzle automatically adjustable for all wind speeds incorporating a turning vane and mounted on a bearing so as to face into the wind could be built.

Of the above-mentioned samplers, the rotating cowl in the form of the cyclone is commercially available (\$1,100 each), while the others would have to be shop built. The shop-built isokinetic sampler (one wind speed range and no wind vane) with the blower, filter holder, and controller (same type as in Hi-Vol) costs about \$700 each. A central wind speed and direction controller that will activate samplers and record operating times would be required for implementing a sampling array with isokinetic samplers built for set wind speed ranges. Such a controller would have to be flexible enough to allow the user to adjust the wind speed ranges, wind sectors, and the number of samplers controlled. An estimate of the meteorological equipment cost would be

ISOKINETIC SAMPLER - INLET OPEN

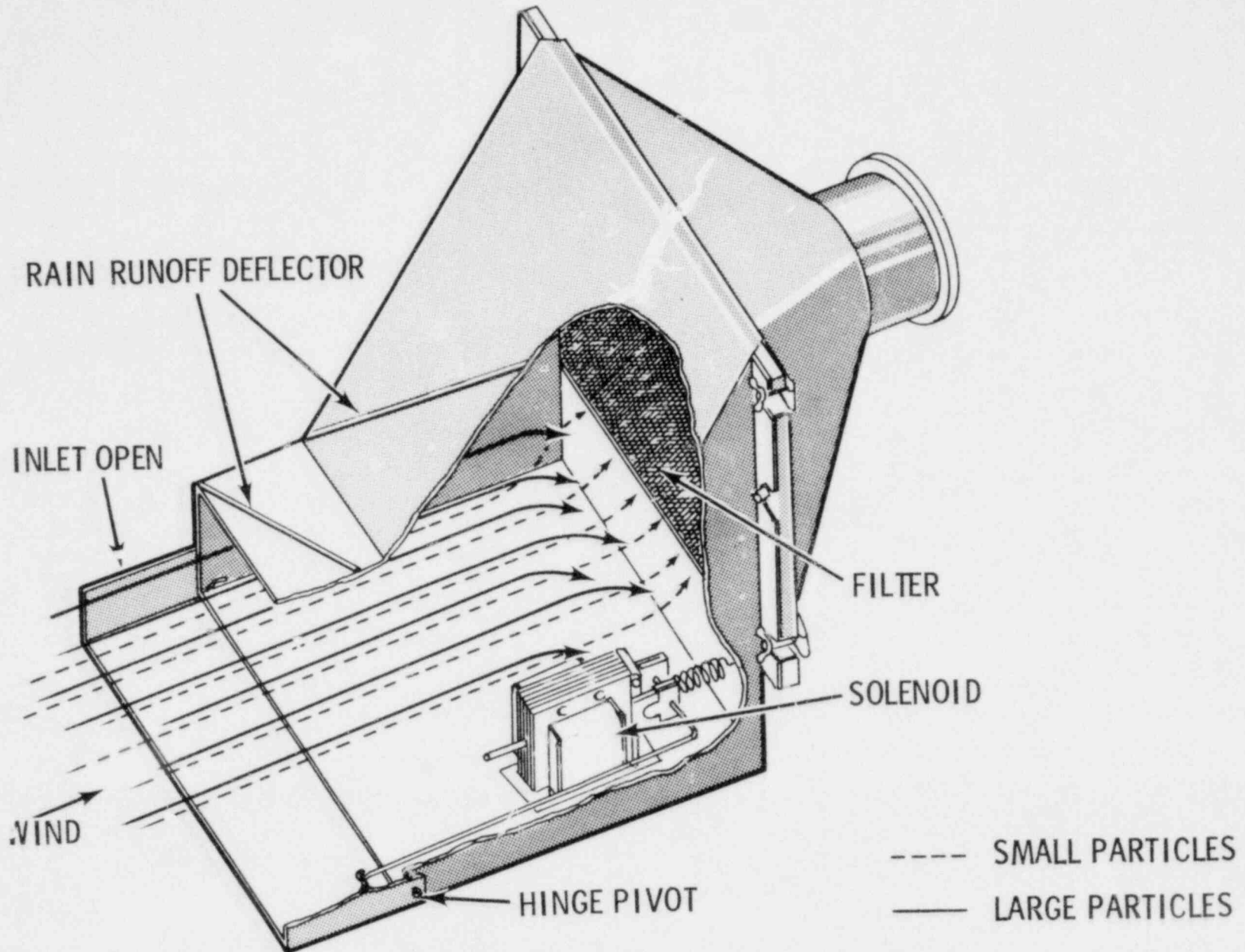


FIGURE 5. Sketch of Sehmel Isokinetic Sampler

\$2,000 + \$250/sampler, exclusive of installation. The installation would involve the purchase of a meteorological tower on which to mount the sensors (\$300 for a 10 m tower), shelter for instrumentation, signal transmission, and a stable electrical supply. For the sampling array shown in the example (see Figure 2) and covering three wind speed ranges, 45 isokinetic samplers would be required for an estimated cost of \$31,500. (An isokinetic sampler that adjusted for wind speed automatically would obviously reduce the number of samplers required, but perhaps not the cost.) With the cost of the wind controllers, the entire system could cost about \$45,000 just for equipment. Sensors for basic meteorology would add about \$3,000.

3.8.2 New Types of Information

The addition of new capabilities to the recommended sampling strategy may yield information useful to uranium mill operators. Some capabilities worth considering may be real-time contaminant concentration monitoring, measurement of the fraction of contaminant particulates that are respirable, and the ability to predict downwind concentrations under various meteorological conditions.

The usefulness of a real-time concentration monitoring capability hinges upon the LLD and speed of response desired for ground-level releases of airborne contamination. Commercial real-time particulate monitors are available for measuring total particulate, respirable particulates, and radioactive particulates and gases. The recommended sampling strategy could be employed in setting up the continuous monitors, and a central processor could be used to interpret the instrument responses. However, real-time monitors generally operate at flow rates in the 2 m³/hr range, which is considerably lower than the Hi-Vols. Also, for some of the monitors the sample collection interval is on the order of minutes rather than weeks. The combination of low flow rate and short sample collection times significantly raises the LLD. Continuous monitoring is usually employed where certain pollution control measures can be taken when releases exceed given action levels (concentrations) for given periods of times. An assessment of action levels, response times, and control measures should be made to indicate whether

a continuous monitoring network is worthy of consideration for site boundary (or beyond) sampling.

Perhaps a more useful addition to the recommended air sampling strategy would be the measurement of the respirable fraction of contaminant particulates. This would involve the collection of particulate samples fractionated on the basis of aerodynamic size. The collected samples would then be analyzed for the contaminants of interest to determine those contained by the respirable fraction. A cascade impactor is commonly used for collecting such samples. A one-stage cascade impactor with an aerodynamic equivalent cut-off particle diameter of about 3.5 microns is sufficient to determine the respirable fraction. Cascade impactors with multiple stages can give more detailed information on the particulate size distribution, but at extra cost. The one-stage Hi-Vol cascade impactors generally operate with flow rates in the 34-68 m³/hr (20-40 scfm) range and are readily adaptable to the conventional Hi-Vol samplers discussed earlier in this report. It is not recommended to supplant the regular Hi-Vol total particulate samplers with size fractionating samplers because the commercially available sampler cover (discussed on page 36) will not sit over the impactor, which makes directional sampling more difficult. Furthermore, it is not recommended to mix results from cascade impactors and regular Hi-Vols in the calculation of background and mill-originated contaminants to a specific location. The size fractionating samplers should be operated as an adjunct to the basic strategy and it would be reasonable to locate them adjacent to the omnidirectional samplers for convenience. The one-stage Hi-Vol impactor with shelter, blower, flow controllers, and collection substrates costs about \$1,050 each.*

It may be desirable to add the capability of predicting downwind concentrations and source terms to the basic sampling program. This would include measuring, recording, and integrating measurements of wind

*Types of ambient cascade impactors other than the Hi-Vols are available with the capability to make respirable fraction or entire size distribution measurements; however, they operate at much lower flow rates (< 2 m³/hr) which result in higher LLDs. They also cost 2-4 times as much as the one-stage Hi-Vols impactors. Their application will not be considered further here.

speed, direction, and atmospheric stability. An offsite arc of Hi-Vol samplers would serve as data points in addition to the site boundary samplers. The choice of the predictive model would depend on the distance to the receptor relative to the length to the site boundary. The cost of such a capability will depend on the complexity of the prediction to be made. Manpower costs could be significant compared to equipment costs. The requirements for predictions would have to be specified before any cost estimates can be made.

3.8.3 Error Assessment

There are some facets of the recommended sampling strategy that merit some research, and they deal with error analysis. An error analysis for the basic strategy needs to be made, including such sources of error as flow rate, time, wind direction measurements, and sample analysis. This level of error analysis would be relatively simple. More difficult sources of error needing addressing are the effects of calms (winds <1 mph), sampling bias (like those investigated by Wedding¹⁵), and the neglecting of dispersion of mill contaminants across the sampler network. The importance of these error sources probably varies depending on whether particulate or gaseous contaminants are being measured and on their mode of generation. Experiments in the field would be required to determine the relative importance of the individual source terms at the mill site and how they fit together to yield the eventual annual transport of contamination.

3.9 Meteorological Instrumentation

The basic sampling strategy requires sampling controllers activated by wind direction. Commercially available wind direction control systems are designed to sense when the wind direction is within a single pre-selected acceptance angle and to activate sampling equipment accordingly. They are equipped with elapsed sampling indicators, 5-20 second delay circuits, and receptacles rated at 10-15 amps (110-115 V AC). Currently they cost from \$350 to \$7,500. With this type of instrument, one is required at each sampler location to operate the directional sampler. For the example given earlier, six controllers are needed at a cost of about \$2,400.

A centrally located wind direction sensor may be a reasonable alternative if wind shear across the source area is negligible. Such a multidirectional controller is not available as a unit but as a custom application using available components. A central controller would use a processor to transmit a signal to each sampler site and compute elapsed times for each sampler. The cost of such a system is estimated to be \$15,000. A precipitation sensor of some type would be useful for turning particulate samplers off and on according to whether there is precipitation or not. Sensors used to trigger relays currently cost \$200-\$300. A single sensor would be adequate if the signal is transmitted to all sampling stations automatically or manually. The cost of signal transmission should be weighed against the cost of a sensor at each station. The circuitry should include some kind of override of the sampler's elapsed time indicators.

For the seven sampling stations shown in the example (Figure 2), the wind controllers (one per station) would cost from \$2,450 to \$10,500. Precipitation sensors for each station would add to the total cost \$1,400 to \$2,100. For the entire particulate sampling array shown in Figure 2, with individual meteorological instruments at each of the seven sampling stations and with automatic flow control, the total cost may be around \$15,200, plus installation. Maintaining the system may cost up to one-half man-year annually.

4.0 DOSIMETRY

A number of different techniques have been used in the past for measurement of background radiation dose in the environment. Among these techniques are photographic film, ion chamber pencils, ion sensitive instruments, and thermoluminescent dosimeters (TLDs). Film dosimeters suffer from a number of shortcomings which make them impractical for use near uranium mills. Film, in many cases, gives erroneous readings after subjection to high temperature and humidity; it cannot be reused; and it requires laboratory chemical processing and special handling. Its response to photon radiation is much different than that of tissue. Film cannot be sensitive to alpha radiation since it must be kept covered during field exposure to prevent damage.

Ion chamber pencils, while being easier to handle in the field, are only rough estimations of photon and some beta radiation and thus do not fulfill the need for radiation monitoring around uranium mills.

Sensitive instruments and air samplers can be used to satisfy radiation measuring requirements; however, they are expensive and require a greater manpower commitment than the TLD-type measurement.

The thermoluminescent dosimeter (TLD) is recommended for dose samplers. In terms of cost, the use of TLDs with the simple meteorology previously discussed is the least expensive method to determine background. However, it is not specific to one radionuclide. Mounting the omnidirectional dosimeter is fairly trivial; however, a directionally-activated TLD is now commercially available. A device activated by the wind direction controller might consist of a TLD mounted on an arm. During the sampling, the TLD is exposed to the atmosphere, and after sampling the TLD can be retracted into a shielded enclosure. During retraction, the TLD can be moved through a series of brushes and/or an air jet to clean any loose radioactive dust from the TLD to prevent continued exposure during periods when it is supposed to be shielded. Housing the TLD in a shielded enclosure with a port that opened when the TLD was to be exposed would be another method. The development of such prototypes might cost \$50,000, but a production version should not cost more than \$200. These system types will not be considered further here.

Thermoluminescent dosimeters (TLDs) can be used for a wide range of measurements of exposure and adsorbed dose related to the environment^{18,19} and thus around uranium mills. There are many useful TLD materials available for measuring exposure and dose at the low levels expected. These materials have varying responses to the alpha, beta, and gamma radiation emitted by the isotopes in the ^{238}U and ^{235}U series decay chains and most of the activity in and around uranium mills is expected to be due to these isotopes.

For most cases, when relating exposure to absorbed dose in tissue, the TLD material, lithium fluoride (LiF), is most convenient for accurate measurement. Lithium fluoride is inexpensive, readily available, and has a response very nearly like that of tissue for photon radiation.

Lithium fluoride is also sensitive to the alpha and beta radiations emitted by the ^{238}U and ^{235}U decay series isotopes. These series are shown in Table 6.²⁰ The energies of the major emitted radiations and the relative abundances are shown in Tables 7 and 8 from the Handbook of Radiological Health.²¹ Radiation energies range from about 50 keV to 1.7 MeV for photons from isotopes in the two decay chains. These photon energies are well within the capabilities of lithium fluoride measurement. Beta radiations vary in energy from 16 keV (^{210}Pb) to 3.26 MeV (^{214}Bi). The lower energy beta radiations are below the useful dosimetry range for lithium fluoride but they constitute a small percentage of the total beta emission. Alpha particle radiation energies vary from about 4.0 MeV (^{238}U) to about 8.0 MeV (^{215}At). TLD materials are also sensitive to these radiations but the dosimeter must be placed in close proximity to the alpha source.

The problem of distinguishing between the "local" background and additional radiation produced by mill operations requires the design of special dosimeter shielding geometry. As noted earlier, some research to complete the design and operational protocol for a TLD system would be required; however, all components are commercially available. Since the true difference between the background and the additional radiation is unknown, it is necessary to assume that the difference in many locations is small. For this situation, a TLD system which includes a number of presently available TL dosimeters or sensors should be used. At least two separate TLD systems are considered in this text. One system would be totally passive, i.e., no moving parts (Section 4.1). The other would involve the use of a small TLD-air sampler system in which TLD dosimeters are placed in close proximity to a filter. The system would use a small battery-powered pump to move air through the filter. Such a system is described by Breslin, et al.²² A different design is also described by K. Schaiger.²³

For both types of systems, studies would be required to optimize the exposure times and number of TLD sensors used. A typical set of data from a calibration exposure to photons is shown in Table 9 for ten TLDs that have been carefully selected and annealed. The readings show a good precision with a standard deviation of about $\pm 3\%$ or ± 30 mR (See Table 9)

Table 7. PRINCIPAL RADIONUCLIDES IN ORE AND TAILINGS

Nuclide*	Half-life	Primary Decay Modes		
		α	β	γ
^{238}U Series				
^{238}U	4.5×10^9 years	x		
^{234}Th	24 days		x	x
$^{234\text{m}}\text{Pa}$	1.17 min		x	x
^{234}U	2.5×10^5 years	x		
^{230}Th	8×10^4 years	x		
^{226}Ra	1.6×10^3 years	x		x
^{222}Rn	3.8 days	x		
^{218}Po	3.1 min	x		
^{214}Pb	26.8 min		x	x
^{214}Bi	19.7 min		x	x
^{214}Po	160 μsec	x		
^{210}Pb	21 years		x	x
^{210}Bi	5 days		x	
^{210}Po	138 days	x		
^{206}Pb	Stable			
^{235}U Series				
^{235}U	7×10^8 years	x		x
^{231}Th	26 hr		x	x
^{231}Pa	3.3×10^4 years	x		x
^{227}Ac	21.6 years		x	
^{227}Th	18 days	x		x
^{223}Ra	11.4 days	x		x
^{219}Rn	4 sec	x		x
^{215}Po	1.8 msec	x		
^{211}Pb	36 min		x	x
^{211}Bi	2.2 min	x		x
^{207}Tl	4.8 min		x	
^{207}Pb	Stable			

*Minor branches and decay modes are not listed.

TABLE 8
The Uranium Series Nuclides and Their Radioactivity Properties

Nuclide	Historical name	Half-life	Major radiation energies (MeV) and intensities†		
			α	β	γ
$^{238}_{92}\text{U}$	Uranium I	4.51×10^9 y	4.15 (25%) 4.20 (75%)	---	---
$^{234}_{90}\text{Th}$	Uranium X ₁	24.1d	---	0.103 (21%) 0.193 (79%)	0.063c‡ (3.5%) 0.03c (4%)
$^{234}_{91}\text{Pa}^m$	Uranium X ₂	1.17m	---	2.29 (98%)	0.765 (0.30%) 1.001 (0.60%)
$^{234}_{91}\text{Pa}$ (99.87% from $^{234}_{91}\text{Pa}^m$, 0.13% from $^{238}_{92}\text{U}$)	Uranium Z	6.75h	---	0.53 (66%) 1.13 (13%)	0.100 (50%) 0.70 (24%) 0.90 (70%)
$^{234}_{92}\text{U}$	Uranium II	2.47×10^5 y	4.72 (28%) 4.77 (72%)	---	0.053 (0.2%)
$^{230}_{90}\text{Th}$	Ionium	8.0×10^4 y	4.62 (24%) 4.68 (76%)	---	0.068 (0.6%) 0.142 (0.07%)
$^{226}_{88}\text{Ra}$	Radium	1602y	4.60 (6%) 4.78 (95%)	---	0.186 (4%)
$^{222}_{86}\text{Rn}$	Emanation Radon (Rn)	3.823d	5.49 (100%)	---	0.510 (0.07%)
$^{218}_{84}\text{Po}$	Radium A	3.05m	6.00 (~100%)	0.33 (~0.019%)	---
$^{214}_{82}\text{Pb}$ (99.98% from $^{218}_{84}\text{Po}$, 0.02% from $^{218}_{84}\text{Po}$)	Radium B	26.8m	---	0.65 (50%) 0.71 (40%) 0.98 (6%)	0.295 (19%) 0.352 (36%)
$^{218}_{85}\text{At}$	Astatine	~2s	6.65 (6%) 6.70 (94%)	? (~0.1%)	---
$^{214}_{83}\text{Bi}$	Radium C	19.7m	5.45 (0.012%) 5.51 (0.008%)	1.0 (23%) 1.51 (40%) 3.26 (19%)	0.609 (47%) 1.120 (17%) 1.764 (17%)
$^{214}_{84}\text{Po}$ (99.98% from $^{214}_{83}\text{Bi}$, 0.02% from $^{214}_{83}\text{Bi}$)	Radium C'	164μs	7.69 (100%)	---	0.799 (0.014%)
$^{210}_{81}\text{Tl}$ (from $^{214}_{83}\text{Bi}$)	Radium C''	1.3m	---	1.3 (25%) 1.9 (56%) 2.3 (19%)	0.296 (80%) 0.795 (100%) 1.31 (21%)
$^{210}_{82}\text{Pb}$	Radium D	21y	3.72 (0.00002%)	0.016 (85%) 0.061 (15%)	0.047 (4%)
$^{210}_{83}\text{Bi}$	Radium E	5.01d	4.05 (0.0007%) 4.69 (0.0005%)	2.161 (~100%)	---
$^{210}_{84}\text{Po}$ (~100% from $^{210}_{83}\text{Bi}$)	Radium F	138.4d	5.305 (100%)	---	0.803 (0.0011%)
$^{206}_{81}\text{Tl}$ (from $^{210}_{83}\text{Bi}$)	Radium E''	4.15m	---	1.571 (100%)	---
$^{206}_{82}\text{Pb}$	Radium G	Stable	---	---	---

†Intensities refer to percentage of disintegrations of the nuclide itself, not to original parent of series.
‡Complex energy peak which would be incompletely resolved by instruments of moderately low resolving power such as scintillators.

THE ACTINIUM SERIES NUCLIDES AND THEIR RADIOACTIVE PROPERTIES

Nuclide	Historical name	Half-life	Major radiation energies (MeV) and intensities†		
			α	β	γ
$^{235}_{92}\text{U}$ ↓ $^{231}_{90}\text{Th}$ ↓ $^{231}_{89}\text{Pa}$ ↓ $^{227}_{89}\text{Ac}$	Actinouranium	$7.1 \times 10^8 \text{y}$	4.37 (18%) 4.40 (57%) 4.58c‡ (8%)	---	0.143 (11%) 0.185 (54%) 0.204 (5%)
	Uranium Y	25.5h	---	0.140 (45%) 0.220 (15%) 0.305 (40%)	0.026 (2%) 0.084c (10%)
	Protoactinium	$3.25 \times 10^4 \text{y}$	4.95 (22%) 5.01 (24%) 5.02 (23%)	---	0.027 (6%) 0.29c (6%)
$^{227}_{89}\text{Ac}$ <div style="display: flex; justify-content: space-around; width: 100%;"> 98.6% ↓ 1.4% ↓ </div> $^{227}_{90}\text{Th}$	Actinium	21.6y	4.86c (0.18%) 4.95c (1.2%)	0.043 (~99%)	0.070 (0.0%)
	Radioactinium	18.2d	5.76 (21%) 5.98 (24%) 6.04 (23%)	---	0.050 (8%) 0.237c (15%) 0.31c (8%)
$^{227}_{90}\text{Th}$ → $^{223}_{87}\text{Fr}$	Actinium K	22m	5.44 (~0.005%)	1.15 (~100%)	0.050 (40%) 0.080 (13%) 0.234 (4%)
$^{223}_{87}\text{Fr}$ → $^{223}_{86}\text{Rn}$	Actinium X	11.43d	5.61 (26%) 5.71 (54%) 5.75 (9%)	---	0.149c (10%) 0.270 (10%) 0.33c (6%)
$^{223}_{86}\text{Rn}$ → $^{219}_{86}\text{Rn}$	Emanation Actinon (An)	4.0s	6.42 (8%) 6.55 (11%) 6.82 (81%)	---	0.272 (9%) 0.401 (5%)
$^{219}_{86}\text{Rn}$ → $^{215}_{84}\text{Po}$	Actinium A	1.78ms	7.38 (~100%)	0.74 (~0.0023%)	---
$^{215}_{84}\text{Po}$ <div style="display: flex; justify-content: space-around; width: 100%;"> ~100% ↓ .00023% ↓ </div> $^{211}_{82}\text{Pb}$	Actinium B	36.1m	---	0.29 (1.4%) 0.56 (9.4%) 1.39 (87.5%)	0.405 (3.4%) 0.427 (1.8%) 0.832 (3.4%)
	Astatine	~0.1ms	8.01 (~100%)	---	---
$^{211}_{82}\text{Pb}$ → $^{211}_{83}\text{Bi}$	Actinium C	2.14s	6.28 (16%) 6.62 (84%)	0.60 (0.28%)	0.351 (14%)
$^{211}_{83}\text{Bi}$ <div style="display: flex; justify-content: space-around; width: 100%;"> 0.28% ↓ 99.7% ↓ </div> $^{211}_{84}\text{Po}$	Actinium C'	0.52s	7.45 (99%)	---	0.570 (7.5%) 0.90 (11.5%)
	Actinium C''	4.79m	---	1.44 (99.8%)	0.897 (0.16%)
$^{211}_{84}\text{Po}$ → $^{207}_{82}\text{Pb}$	Actinium D	Stable	---	---	---

†Intensities refer to percentage of disintegrations of the nuclide itself, not to original parent of series.

‡Complex energy peak which would be incompletely resolved by instruments of moderately low resolving power such as scintillators.

TABLE 9

TABLE 10. Calibration Exposure

<u>TLD</u>	<u>Reading</u>	<u>High Sensitivity Reading</u>
1	10.6	514.7
2	10.5	505.4
3	10.2	506.2
4	10.5	492.6
5	10.1	539.8
6	10.4	526.8
7	10.0	496.1
8	10.0	502.9
9	10.5	506.8
10	10.0	535.2
	<hr/>	<hr/>
	Average 10.3	533.8
	Standard Deviation ± 0.2	514.6
		± 16.6 or 3.32%

for 1000 mR exposure. Precision at field exposures of 10 to 20 mR per month will be on the order of ± 5 to $\pm 10\%$, as has been regularly achieved in the NRC-sponsored Dose to Construction Workers Program.²⁴

The passive TLD system would be in two sections: one shielded from direct radiation emitted from the nearby ground and the other section would be unshielded. Each section will consist of a "bare" set of TLDs and a shielded set of TLDs.

Although the TLD locations would be similar to those illustrated by Strategy B in Figures 1 and 2, many more locations could be used due to the low inherent cost of maintenance of the TLD method.

4.1 TLD Capsule and Shield Design

The use of two TLD capsules at each monitoring location is recommended. The simplest sampler would be assembled from one TLD capsule shielded by lead so as to be sensitive only to radiation from the air in the local area, while a second TLD would not be shielded. The difference between the two sets of dosimeters is related to the amount of airborne ^{222}Rn and its daughters at the specific measurement site.

Only one shielded capsule is suggested because: 1) precision should be better for the unshielded TLDs; and 2) the lead shields add expense. TLD capsules should be made of plastic and metal combinations to provide 1-cm depth dose for photons in one section. (0.025" Al plus 0.015" plastic is one possibility.) The other section would be designed of plastic for light shielding but allow most beta radiation to penetrate (0.010" is suggested). A lead shield made of a standard lead brick 2" x 4" x 4" with a slot for the TLD capsule is appropriate.

4.1.1 Simple TLD Methodology

The least expensive methodology which is recommended for assessing the background in the vicinity of a mill uses thermoluminescence dosimetry (TLD) as the monitoring technique. It involves capital costs of \$25,000 and a 0.5 man-year operating effort. As previously discussed, there should be dosimetry extending radially from the mill on lines separated 60° from one another. The primary line should be placed on the major wind rose. Dosimeters should be located at the mill site boundary, and 0.5, 1, 2, 3, 4,

and 5 km from the boundary. Dosimeter duplication at eight locations is required to ascertain statistical validity of the dosimetry. Each dosimeter station must be composed of a lead shielded and unshielded TLD chip mounted 0.5 m above ground. In this fashion, the unshielded TLDs receive radiation from both ground and air sources, whereas the shielded TLD is exposed only to the atmospheric component. The TLDs should be changed monthly. Specific procedures and shield designs have been discussed above in Section 4.1. The TLD is not isotope specific, nor does it provide the sensitivity of alternate methods; however, it does provide a precision of $\pm 10\%$ at the 10 mR per month level.

The advantages of a TLD system are that it is simple, cost effective, and reliable. It requires no power in the field and has no problems from a variety of weather conditions. The TLD system, in its simplified form, does not require an on-off mechanism controlled by meteorological conditions.

The major disadvantage of the method is that it measures only gross radiation--that from beta and gamma contributions. It does require a meteorology station to provide wind rose data for its initial set-up and then later for interpretation of the data which are obtained. The TLD methodology will not allow natural background to be differentiated from radioactive material contributed to the environment from prior mill operation. It capably determines the current mill additions to the environment. Although it allows measurement of those materials contributed by sources other than the mill in question, it does not allow them to be individually quantified.

The TLD method requires an effort to maintain the quality of the TLD chips and their dose response to a radon chamber.

4.2 Calibration

Calibration exposures should cover the range of expected results. If total exposure is expected to be 10 or 20 mR in one monitoring period, the calibration exposures should cover that range and above. The calibration facilities could use: 1) ^{137}Cs to simulate penetration photons; 2) a uranium slab for beta exposure; and 3) radon exposure chambers with controlled working levels.

Calibrated radon chambers already exist at DOE and the United States Geological Survey (USGS) facilities to allow cross calibration of industrial chambers to known sources. A complete set of TLD operating procedures can be found in Section 9.0.

4.3 TLD Operational Costs

The TLD costs summarized in Table 10 are based on the assumption that each mill site will have a TLD reader. Some companies may want to use one reader to cover more than one site which will reduce overall costs.

TABLE 11
Itemized TLD Operational Costs

<u>Laboratory Equipment</u>	<u>Costs</u>
1) Manual TLD reader (such as Harshaw 2000 system with new charge integrator)	\$8500
2) Annealing ovens	
High temperature - 400°C	350
Low temperature - 100°C	350
3) Shielded storage (4" lead on all sides)	4000
4) Vicor glass dishes for cleaning and annealing	200
5) Nitrogen gas supply. Liquid nitrogen offgas is preferable, if available. Otherwise, laboratory-grade nitrogen gas.	20/wk plus fittings
6) Vacuum probe for handling TLDs (2 each)	100
7) Laboratory storage oscilloscope	1200
8) BCD printer for TLD reader output	750
 <u>Dosimeter Costs</u>	
1) 3000 TLD-700 chips (LiF)	\$4500
2) Dosimeter capsule	
Aluminum and plastic parts	500
50 each per site	250
Fabrication costs - estimate \$5 each	250
3) Lead for shields	400
4) Assembly, distribution, and fabrication of shield	1000
 <u>Meteorology System</u>	 \$4000
Total	\$25850

Labor

If bi-monthly readout is chosen, a technician should be able to accomplish the TLD readout, analysis, and preparation in ten working days or on the average of five working days per month. If more than 15 monitoring stations are selected, technician time would be increased accordingly. A computer-controlled dosimetry readout system²⁵ (Model 2276) is available from Harshaw Chemical Co., Salem, Ohio. A complete meteorology system would be required at each mill site.

5.0 SOIL, WATER, AND BIOTA SAMPLING SITE SELECTION

The topic of sampling soil, water, and biota is addressed only superficially in this report and almost entirely by reference since a wide variety of methods already exist. Biota sampling around uranium mills (and other uranium-contaminated areas) is rarely discussed by itself but in combination with discussions of sampling soil and water where the biota happen to be found. Techniques for soil sampling and sampling site layout are amply discussed in References 26-32.

Typical 5-cm core samples are taken by using a specially designed coring device. This device is constructed of stainless steel and contains four tracks positioned 1/2 cm, 1 cm, 2 cm, and 5 cm from the top of the device (see Figure 6). By driving the coring device into the ground and digging away at each end, a stainless steel blade can be forced along each track to reach the desired sampling depth. Deeper cores are obtained by digging a trench and inserting a stainless steel flat scoop with 5-cm high walls. Large area samples are obtained by scraping a 1-m² section to a depth of 1 cm.

The measurement of uranium and daughter radionuclides in each soil sample is obtained by first preparing a perfectly flat homogeneously mixed pellet. Fifteen grams of the sample matrix is homogenized with two grams of cellulose binder and formed under 30 tons of pressure into a 1/4-in x 2-in pellet in a pressing chamber. The pellet is then placed in Saran wrap and analyzed with a high resolution intrinsic germanium diode that measures the concentration of ²³⁸U, ²³⁵U, and uranium daughters. (Uranium-238 is actually determined from analysis of its ²³⁴Th daughter; therefore, one must be certain that the parent-daughter equilibrium exists in making this measurement.)

The photon detection systems are discussed in Section 6.3.

Table 11 lists the radionuclides, photon energies, and the abundance of each of their photons.

TABLE 12

Radionuclides, Photon Energies, and Abundance of Photons

<u>Radionuclide</u>	<u>Photon Energies (keV)</u>	<u>% Abundance</u>
^{226}Ra	186	4.0
^{214}Po	295	18.9
^{235}U	205	5.0
^{210}Pb	47	4.0
^{230}Th	67	0.4
^{227}Th	236	10.4
^{234}Th (^{238}U)	63	5.7

The rationale, guidelines, and sampling methods for water are well developed.^{26, 32-36} In most industries, water sampling is performed at the point of liquid effluent discharge; however, uranium mills discharge liquid wastes to the tailings pond where they dissipate by seepage and evaporation. Runoff or seepage from tailings ponds, the mill site, and from effluent piping can contribute somewhat to the spread of contaminants into surface or groundwaters. Sampling should be performed from wells and surface water upstream and downstream of the mill vicinity and at the point of the first downstream use of the water. Examples illustrating some soil, water, and biota sampling surveys in the vicinity of a uranium-contaminated area and inactive uranium mill tailings are given in References 37-40. Specific sampling techniques for all types of terrain, as well as double sampling methods, are discussed by Cochran.⁴⁰ Applications of double sampling to field measurement problems in the nuclear industry are illustrated by Delifiner⁴¹ and Gilbert⁴². Before double sampling techniques are requested in the uranium industry, they must be evaluated through additional research efforts to prove their worth to that industry.

5.1 Simple Air, Water, Soil, and Biota Sampling Methodology

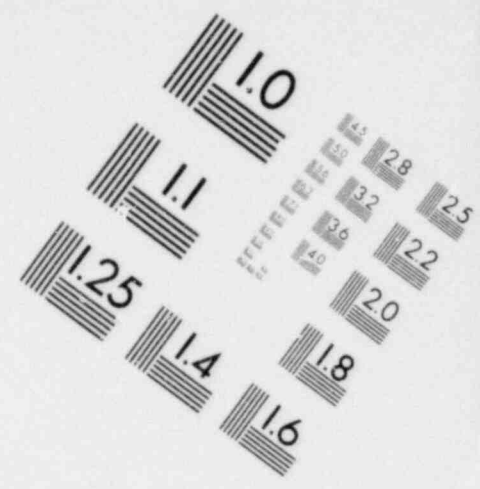
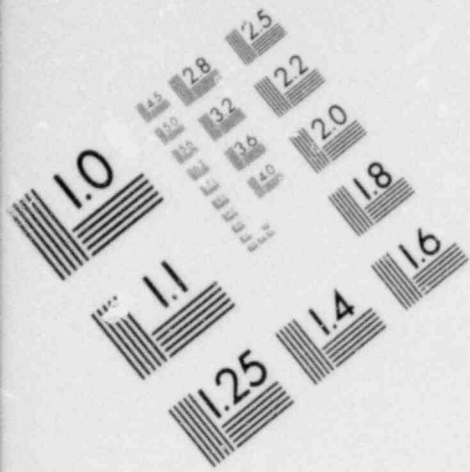
The most costly and sophisticated method to assess the background level of radiation in the environment around a uranium mill involves the sampling of air particulates (as in Section 3.4.4), the determination of airborne ^{222}Rn and its daughters (as in Section 4.1.1), and the sampling of soil, water, and biota.

As in methodologies 3.4.4 and 4.1.1, a complete meteorology/atmospheric sampling program similar in all respects to that previously discussed would be employed. In addition, soil, water, and biota would be sampled on the lines on which the air sampling system has been assembled. Soil samples would be collected at the boundary, 0.5, 1, 2, 3, 4, 5, and 10 km from the boundary. Samples would be taken using the coring device (Figure 6) to obtain aliquots at 1/2, 1, 2, 5, and 10 cm depths from a 10 cm x 20 cm surface area. Deeper cores are obtained by digging a trench and inserting a stainless steel flat scoop with 5-cm high walls.

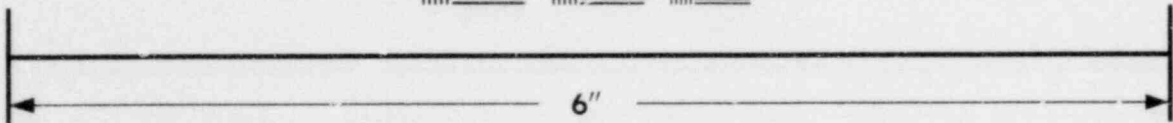
Biological material should be collected from a 1 m² area at the sampling site. It should be dried, homogenized, and duplicate fractions taken for radionuclide analysis. Water should be collected from sources within the sampling grid in sufficient size to achieve the necessary lower limit of detection as designated by the NRC. The specific limits are given in the body of the text. The soil, water, and biota sampling should occur once per year with the air sampling occurring monthly. Photon spectroscopy, using the equipment suggested in Section 6.3, should be used to analyze all samples prior to chemistry. Samples which require radiochemical separations to attain LLD should be identified by gamma-ray spectrometry. This will minimize the number of samples subjected to radiochemical procedures. The chemical procedures which should be used on the soil, biota, and water samples to isolate the small quantities of radionuclides present from the bulk material are presented in Section 9.0 of this report. A beta detector and an alpha spectrometry system (Section 6.2) must be assembled for the analysis of the specific radionuclides isolated with these chemical procedures. In this fashion, an operating mill can capably measure all radionuclide components such as natural uranium, ^{230}Th , ^{226}Ra , ^{210}Pb , ^{222}Rn , and ^{210}Po in the various environmental materials at their environmental levels. It should be noted that the suggested NRC lower limits of detection are in some cases below that found in the environment for specific radionuclides.



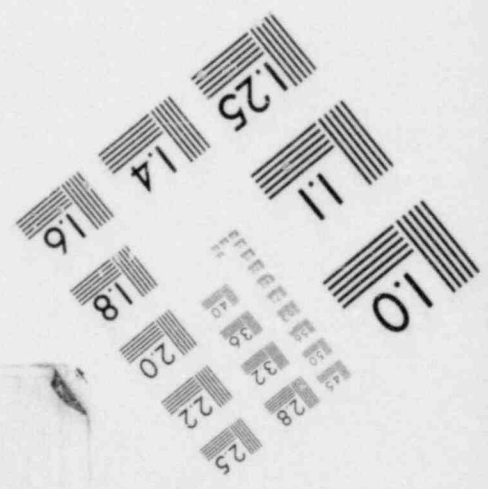
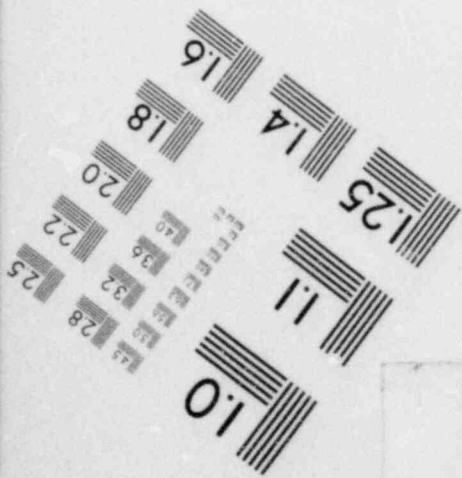
Figure 6. Coring device for sampling soil near uranium mills



**IMAGE EVALUATION
TEST TARGET (MT-3)**



MICROCOPY RESOLUTION TEST CHART



The advantages of using this technology are that it allows the measurement of all of the necessary radionuclides in all types of environmental materials at environmental levels. It allows the determination of the effect the mill has had on the background in each mill environment. All advantages and disadvantages which were discussed in the air sampling technology section apply to this technology. Specific disadvantages of this methodology are the additional manpower requirements including a trained radiochemist and additional laboratory and counting room facilities.

6.0 RADIOCHEMICAL ANALYSIS

6.1 Radiochemistry

Through the use of the air samplers and soil/water/biota sampling programs described in the earlier sections, materials will have been obtained which require analysis for the list of radionuclides in Table 12.

TABLE 13

Radionuclides to Be Measured in the Uranium Mill Environment and Their Associated Limits of Detection

<u>Nuclide</u>	<u>NRC LLD</u>
<u>Air-inhaled</u>	
Natural uranium	1×10^{-4} pCi/m ³
²³⁰ Th	1×10^{-4}
²²⁶ Ra	1×10^{-4}
²¹⁰ Pb	2×10^{-3}
²²² Rn	200
<u>Water-ingested</u>	
Natural uranium	0.2 pCi/l
²³⁰ Th	0.2
²²⁶ Ra	0.05
²¹⁰ Pb	1.0
²¹⁰ Po	1.0
<u>Vegetation-ingested</u>	
Natural uranium	0.2 pCi/kg
²³⁰ Th	0.2
²²⁶ Ra	0.05
²¹⁰ Pb	1.0
²¹⁰ Po	1.0
<u>Meat-ingested (LLD is for forage eaten by cattle.)</u>	
Natural uranium	0.2
²³⁰ Th	0.2
²²⁶ Ra	0.05
²¹⁰ Pb	1.0
²¹⁰ Po	1.0

The list and the LLDs are those being considered by the NRC as of August 1979.⁴³ It has been assumed that the LLD for inhaled and ingested materials is that which must be achieved by the measurement program for specific radionuclides. To achieve all of these LLDs requires a combination of radiochemical procedures and a variety of α , β and γ counting techniques. The literature contains many radiochemical procedures for these isotopes; thus they will not be repeated here. For clarity, however, one full procedure is written in Section 9.

6.2 Alpha/Beta Detection Systems

Following the use of a chemical procedure, the alpha or beta radioactivity can be quantitatively measured with commercially available detector systems. For example, ORTEC provides solid state surface barrier alpha detectors with backgrounds <3 c/day for a $2000 \text{ mm}^2 \times 100 \mu$ thick system for \$2,500. For beta detection, they also sell a $450 \text{ mm}^2 \times 1000 \mu$ thick surface barrier detector for \$1,800. These detectors can be operated with the electronics components shown in Table 13 to provide a commercially available alpha and beta detection system.

TABLE 14

Electronic Components and Their Costs
for an Alpha or Beta Detector System

Detector bias supply, Model 428	\$ 535
Detector preamplifier, Model 142B	495
Detector vacuum system, Model 807	240
Detector amplifier, Model 570	595
Single channel analyzer, Model 550	275
Timer, Model 719	260
Scaler (non-printing), Model 775	445
N.I.M. bin/power supply, Model 401A) Model 402A)	695
Cables	70
	<u>\$3,610</u>

6.3 Photon Detection Systems

A variety of photon measurement techniques can also be used to measure daughter isotopes as well as their parent in the uranium chain. Uranium-238, ^{234}U , ^{214}Pb , ^{210}Pb , ^{230}Th , ^{226}Ra , and ^{235}U can be analyzed using low-energy photons.^{44,45} An analysis of this type normally uses an intrinsic germanium planar detector and requires no prior chemistry to separate the isotopes of interest in the sample. These detectors range in size from 100 mm² to 10 cm². Typically, a 0.5 x 5-cm pellet is prepared by homogenizing 15 grams of the sample matrix with 2 grams of cellulose binder. This is mixed and transferred to a pressing chamber. After applying up to 30 tons of pressure for this unit area, the sample is placed in Saran[®] wrap. The sample can then be analyzed in the low-energy photon spectrometer. The disintegrations per minute per gram of sample that can be easily obtained is illustrated in Table 14. The typical spectral response is shown in Figure 7. The entire analysis uses a small hard-wired analyzer or a mini-computer and printer. The detector costs are on the order of \$10,000, the hard-wired analyzer costs are about \$10,000, and a small computer is about \$20,000. The printer costs can vary from \$1,000 to \$10,000.

A second type of analysis using the coincidence photons of the uranium daughter products has also been developed.⁴⁶ The system employs two large area, typically 15 to 30 cm diameter by approximately 10 cm thick, NaI(Tl) activated crystals. Each detector is normally viewed through a 7.5 cm pure NaI light pipe by photomultiplier tubes and is surrounded by an anticoincidence shield of either NaI(Tl) or plastic phosphor. The detector system is coupled to at least a 4096 word computer memory and uses the gamma-ray decay characteristics of each radionuclide for its identification and measurement. Coincidence counts are stored in an energy-energy plane of the memory according to their photon energies, while single gamma rays which interact with either crystal are stored in a normal manner on the X or Y axis.

The background response of this detector, as well as its high coincidence efficiency, allows isotopes such as ^{214}Bi , ^{214}Pb from the uranium chain, as well as ^{208}Tl from the thorium chain, to be measured. Typical efficiencies and backgrounds are to be found in Table 15. In general, this type of analysis has a detection limit of between 1 and 5 disintegrations per minute per sample. Of course, if one allows the confidence level to

drop to 67%, lower detection limits are achievable for both the coincidence and low energy photon analysis methods. Sample sizes in these instruments range from the small pellets previously described to 10 cm x 30 cm diameter samples. Normally 2 cm thick x 15 cm diameter samples are pressed and analyzed. In both the low-energy photon spectrometer and the coincidence spectrometer, final analysis is done by matching sample photon count rates with radionuclide reference material in standard geometries. The matching can be done by computer or by hand. If the latter is chosen, it will require one man year of effort. These analyzers can be used to analyze air filters, soils, biota which is typically dried, and water which has been passed through cation/anion exchangers to remove the isotopes of interest. To measure isotopes such as ^{210}Pb , ^{210}Po , ^{230}Th , and uranium at disintegration rates below the 1 dpm level, chemistry such as that found in Section 9 must be used.

TABLE 15

Detection Sensitivity for Direct Photon Analysis
Of Samples by a 19 cm² Planar Intrinsic Ge Detector
Using a 1000 Min Count

Isotope	keV	Background (C/M)	D/C*	A _d , d/m	A _Q
^{210}Pb	46.5	.0221	164.40	4.0	21.9
^{214}Pb	352.0	.0415	91.50	3.0	4.1
^{234}U	53.1	<.0161	4511.00	<96.	<567.
^{238}U	63.3	.0481	171.70	6.0	27.5
^{230}Th	67.8	<.0158	1524.00	<32.	<191.
^{235}U	163.4	<.0177	207.50	< 4.6	< 26.5
^{226}Ra	186.0	.0904	415.60	8.0	80.4

$$A_d = \frac{2.71 + 4.65 \sqrt{\text{Background}}}{(\text{eff}) (\Delta T)}$$

$$A_Q = \frac{50 \left\{ 1 + \sqrt{1 + \frac{\text{Background}}{12.5}} \right\}}{(\text{eff}) (\Delta t)}$$

where

A_d = the minimum detectable activity (95% confidence level),⁴⁷

A_Q = the activity required to obtain a precision of 10%,⁴⁷

and

Background = total background in a counting period, ΔT, in a system with efficiency, eff.

* D/C is the reciprocal of the efficiency.

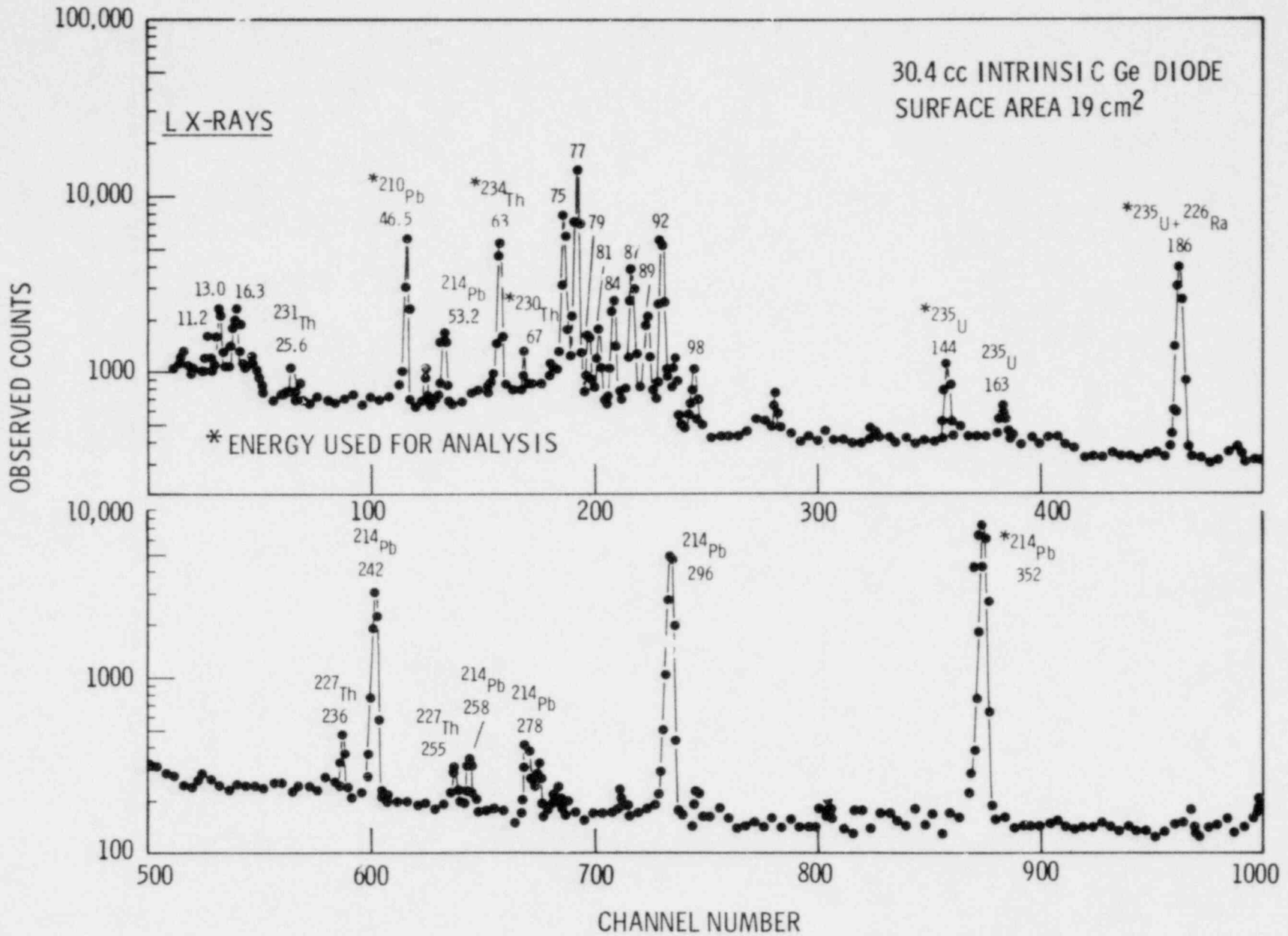


FIGURE 7. A low energy gamma-ray spectrum of soil containing uranium ore

TABLE 16

DETECTION SENSITIVITY FOR 600 gm SAMPLE
USING A 1000 min COUNT WITH MULTIDIMENSIONAL GAMMA-RAY SPECTROMETER

Radionuclide	D/C*	Background (C/M)	A _d , d/m	A _Q
²¹⁴ Bi	25.4	0.56	2.8	9.9
²¹⁴ Pb	2.2	247.0	5.1	15.6
²⁰⁸ Tl	15.0	0.15	0.9	3.5

$$A_d = \frac{2.71 + 4.65 \sqrt{\text{Background}}}{(\text{eff.}) (\Delta T)}$$

$$A_Q = \frac{50}{(\text{eff.}) (\Delta t)} \left(1 + \sqrt{1 + \frac{\text{Background}}{12.5}} \right)$$

where

A_d = the minimum detectable activity (95% confidence level),⁴⁷

A_Q = the activity required to obtain a precision of 10%,⁴⁷

and

Background = total background in a counting period, ΔT, in a system with efficiency, eff.

* D/C is the reciprocal of the efficiency

Instrumentation for α, β, or γ measurement systems can be obtained from Princeton Gamma Tech, ORTEC, Tennelec, Canberra, Beckman, Tracor Northern, and others.

It should be noted here that the equations of Currie⁴⁷ provide a valid method to determine the minimum detectable activity at the 95% confidence level or the activity required to obtain a precision of 10%. The Currie equations are also used as the basis for those listed in HASL-300³².

6.4 Additional Methods

Evaluation of existing or potential environmental contamination resulting from uranium mills is dependent on determining background levels of various radioactive materials and assessing incremental additions to these background levels. In most cases, these levels^{48,49} are above the lower limits of detection shown in Table 12. For example, the LLD for ²²⁶Ra in the

table for ingested vegetation is 0.05 pCi/kg, yet the total estimated dietary intake/day varies from 0.7 to 1.8 pCi. The mean dietary levels in pCi/d are 0.9 - U, 1.4 - ^{226}Ra , 1.4 - ^{210}Pb , and 1.8 - ^{210}Po as obtained from the NCRP⁴⁸. LLD values and NCRP dietary levels for ^{210}Po and ^{210}Pb are of the same magnitude. The LLD values being considered by NRC (Table 12) are unrealistically low--often below natural levels for other environmental species.

The ^{210}Pb values required as LLDs by the NRC are also below those found in air and precipitation in nature⁵⁰. Lead-210 in ground-level air is typically 25 to 35 pCi/m, and it is 1 to 10 pCi/l in rain water. The rain water value of 1 pCi/l is similar to that of Table 12 but the air concentrations of the world often are 10 greater than that shown in Table 12. Any assessment of mill contribution above natural levels will involve quantities of radioactivity in excess of the required sensitivity.

If one assumes that the methodology for the assessment of background around a uranium mill does not have to meet the presently suggested NRC lower limits of detection, then new assessment techniques can be considered.

6.4.1 Radon-222

The first radionuclide that comes to mind for use as a monitor is ^{222}Rn which escapes during essentially all phases of the milling operation. By far, the tailings disposal site represents the greatest source of radon escapement. Use of radon or its short-lived daughters as an indicator of mill activities is complicated by a number of factors:

(1) Radon, as a member of the uranium decay chain, occurs as a natural component of soil and thus is continually diffusing into the atmosphere. This contribution to atmospheric background levels varies due to differences in soil concentrations on both a local and regional scale. Variations also are influenced by many other factors such as barometric pressure, wind speed, temperature, and precipitation.

(2) Most uranium mills are located in proximity to areas of uranium mineralization, which means additional sources of radon. These can take the form of both mining and milling operations in the proximity of the site under evaluation. In addition, outcroppings of uraniferous ore bodies are common in some localities which can also introduce considerable radon to the atmosphere.

(3) When considering the impact of a uranium mill that has operated for a period of time, one must include in the source term the radon that is derived from tailings material which has been removed from the tailings pile by wind erosion and has been deposited in the surrounding environs. This material is deposited as a surface layer of varying depths on the ground surface over rather extensive areas, in some cases five or more kilometers from the tailings. By the very nature of the translocation, this material is enriched in the smaller particle size components of the tailings which in turn are generally enriched in radium concentration. This material will increase the quantities of radon emanated from the soil surface and is a potential indicator of the environmental contribution of post-uranium milling activity.

(4) Atmospheric transport and diffusion of gaseous radon and the aerosols to which radon daughters attach introduce many variables that must be clearly understood in order to properly assess the relationship between the measured air concentration of these radionuclides at a specific network of sites and the source term at the mill. These variables are discussed in detail in the section on atmospheric effects and are mentioned here only to re-emphasize their importance in dispersion of radon and its daughters.

Thus, although there is presently no good way to identify radon sources from radon measurement data, ^{222}Rn could be used as a tracer if all variables involving its production, source, and transport were understood.

6.4.2 Radon Daughters

In essence, the same situations and conditions that were discussed for radon govern the behavior of the short-lived radon daughters. One factor that plays a significant role in the distribution of radon daughters is their rapid attachment after formation to aerosols present in the atmosphere, which subjects them to gravitational and scavenging processes. This process enhances their ground deposition versus that of their gaseous radon parent. This factor will not create a significant increase to the background concentrations of the short-lived daughters but will indeed manifest itself in an increasing concentration of ^{210}Pb and ^{210}Po due to the 22-year half-life of ^{210}Pb . From each curie of ^{222}Rn escaping to the atmosphere, 4.76×10^{-4} Ci of ^{210}Pb is formed, all of which is ultimately deposited on the earth's surface. The distributional pattern of this deposition is governed by many

factors, including the advective transport and dilution of both the radon gas and the atmospheric aerosols containing attached radon daughter products, the 3.824-day half-life of ^{222}Rn , which allows great distances of transport from point of injection to point of decay, and the various processes involved in removal of the aerosols from the atmosphere and their deposition on the earth's surface. All of these factors dilute the concentrations of ^{210}Pb that are available for deposition to the degree that the ^{210}Pb from a particular source becomes indistinguishable from both other point sources and that which is derived on a meso- and macro-scale from the earth's crust.

Particulate transport from the tailings pile and from other sources of the milling operation distributes many radionuclide daughter products of the uranium decay chains that are generally present in equilibrium concentrations representative of the ore being processed. The relatively large size of this particulate material results in rapid gravitational settling and ground disposition within short distances of the source and thus provides an indicator of environmental contamination in the immediate vicinity of a uranium mill. Because of rapid gravitational settling, contributions from other local mining and milling operations in the vicinity are minimized. A natural background of these materials exists because of the presence of uranium occurring naturally in the soil and, in the case of ^{210}Pb and its successors, due to continual deposition of radon daughters produced in the atmosphere. Soils typically contain 1 to 2 pCi/gm U, and 0.5 to 25 pCi/gm ^{226}Ra .^{48, 51} With the exception of the occurrence of nonuniform uranium distribution in the soils surrounding a particular mill site, the surface soil concentrations of the daughter products from either decay of soil-bound uranium or atmospheric radon daughters should be quite uniform and would certainly be at equilibrium. Measurements by Thomas¹⁰ have shown that ^{210}Pb background levels of 2 to 3 dpm/g occur in surface soils in the vicinity of uranium mills at Grants, New Mexico. Windborne transport and redistribution of tailings material at one mill results in a decrease of ^{210}Pb concentrations from the tailings level of 500 dpm/g to background levels over a distance of 8 km or less depending on the direction from the pile. This particular tailings pile is an above-ground disposal site and is conducive to the effects of wind erosion and suspension of tailings material. Downwind deposition of tailings from another pile, while it is a fill-type operation, is of a lesser magnitude, but still significant and definable, exhibiting concentrations that are three to five times higher than background at distances

3 to 5 km downwind of the pile. These effects are long-term in nature and represent the accumulated redeposition of tailings material over two decades of operation. Distribution of tailings material from a new disposal site should be qualitatively similar to those observed for long-term operations. The actual concentrations would, of course, be much lower, which would necessitate sampling from a network more closely centered around the tailings disposal area.

The translocated tailings material is superficially deposited and, in the absence of disturbing factors such as preferential leaching or agricultural activity, remains predominantly on the soil surface. The concentration gradient of ^{210}Pb appears to have a half-thickness of 1 to 2 cm (half-thickness being the depth in which the ^{210}Pb concentration is one-half of the value of the surface). The existing or pre-existing background levels can be determined by analyzing samples obtained at depths below the deposition layer. Thus if one knows how ^{210}Pb relates to the background and how it may be transported, he could use it as a measure of the impact of a mill on its environment and also possibly assess the original pre-mill background radiation levels.

The preceding arguments relating to ^{210}Pb could also be applied to other long-lived members of the uranium decay chain, notably ^{230}Th , ^{226}Ra , and ^{210}Po . These radionuclides are not notably depleted during the milling process and occur at their equilibrium values in the tailings material. These radionuclides undergo alpha decay which requires their isolation from the sample matrix by chemical methods prior to their assay by alpha counting methods. Radium assay can also be accomplished by measuring the shorter-lived radium daughters by various means, but this too can involve chemical treatment of the sample. The 186 keV gamma ray from ^{226}Ra can also be used for analysis, but photons contributed by decay of ^{235}U and other radionuclides necessitate accommodation of their interference in order to quantify the radium concentration present. Lead-210 undergoes beta decay, 4% of which passes through a 46.5 keV gamma transition. This photon is easily measured directly from a sample by gamma-ray spectrometry with a thin window intrinsic germanium diode detector as shown in the gamma-ray spectrum of uranium ore given in Figure 7. The 46.5 keV gamma ray of ^{210}Pb is measured without interference from other radionuclides present in the uranium decay chain. Another feature shown in this figure, which will be discussed subsequently, is the

occurrence of the 63 keV photopeak of ^{234}Th , the first daughter of ^{238}U , which occurs without interference from photons from other radionuclides.

The gamma-ray spectrum of uranium ore tailings given in Figure 8 shows uranium depletion by the great reduction of ^{234}Th present, while most of the other uranium daughter activities originally present in the ore remain undepleted.

The relative concentrations of ^{210}Pb and ^{234}Th present in a soil sample can be used to provide a more sensitive indicator of tailings material than can be done by just determining the distribution of ^{210}Pb around a milling site. In this strategy, if one considers that uranium is in equilibrium with its daughters in normal soil (disequilibrium can exist due to radon loss), the background ratio of ^{210}Pb to ^{234}Th will be $\sim 1:1$. In tailings material, this ratio will be 20-100:1 depending on the efficiency of uranium recovery of the particular mill. Thus, at the point that the ^{210}Pb contribution from tailings equals the natural level, a signal-noise ratio of 1:1 for ^{210}Pb assay, the ratio of $^{210}\text{Pb}:^{234}\text{Th}$ will be between 10-50:1. By employing the $^{210}\text{Pb}:^{234}\text{Th}$ ratio method, determination of surficial contamination from a milling operation should be possible over at least twice the distance as would be possible if just ^{210}Pb were used as the index.

7.0 PREOPERATIONAL SURVEYS

The preoperational survey at a uranium mill site should include sampling air, water, soil, and biota to assess the background levels of contaminants. The procedures to be used are those listed in Section 3.4.4, Section 4.1.1, and Section 5.1. A preoperational survey should be conducted at each uranium mill before or concurrent with the establishment of any monitoring program. These surveys should include measurements of the photon spectrum and exposure rates at locations chosen for routine monitoring. Air samplers and alpha monitor TLD systems, as described by Breslin⁵, could be used to monitor for alpha activity. If correlation of data from the air samplers and the passive TLDs show a direct relationship, the air sampling systems would be unnecessary for a routine monitoring program. As previously discussed, design of the passive TLD system shields should be analyzed for its applicability for this type of measurement.

U ORE TAILINGS, ACID LEACH PROCESS

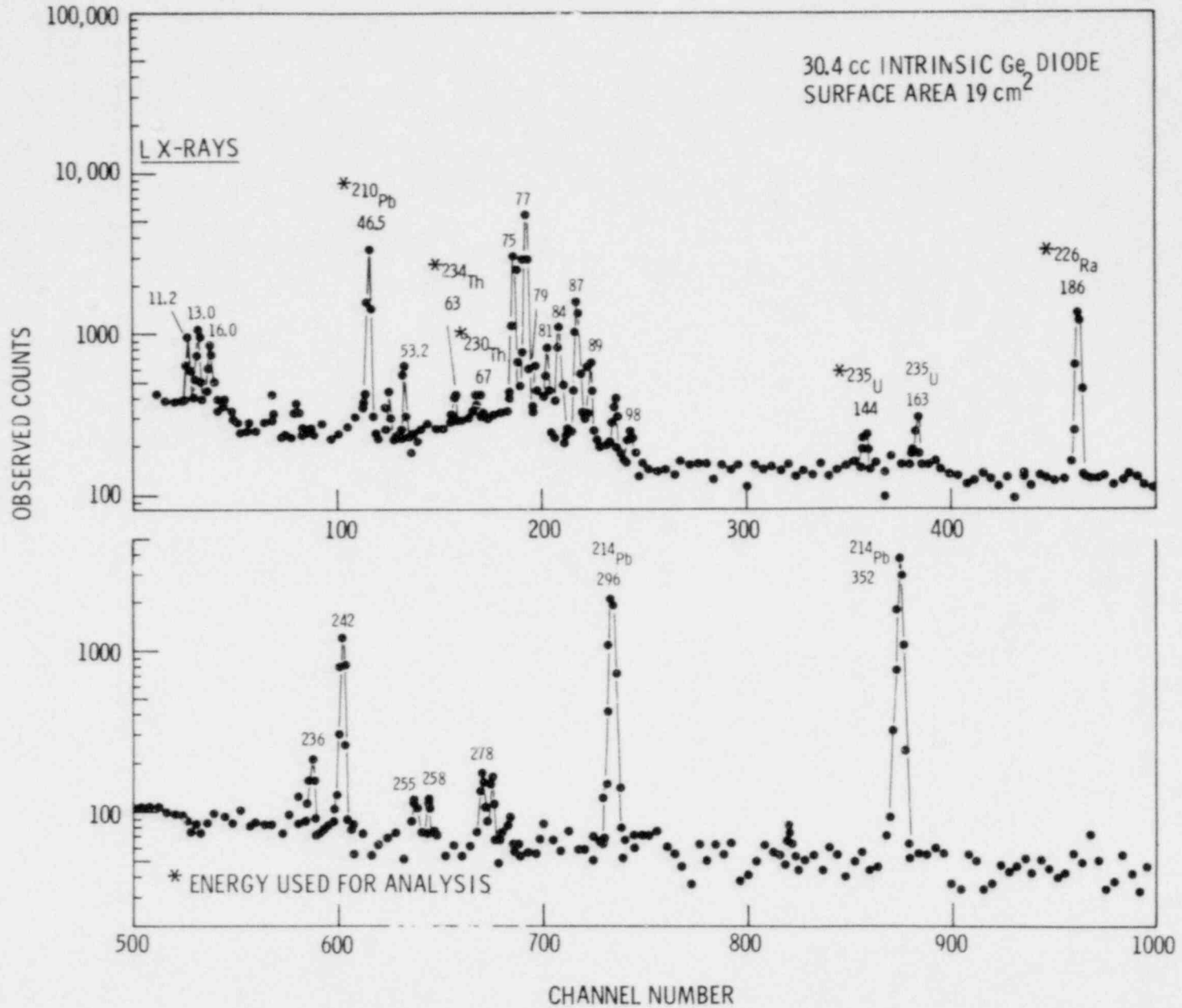


FIGURE 8. The gamma-ray spectrum of uranium ore tailings

Results of these measurements should be correlated with meteorology at each mill site. The number of routine monitoring stations needed will depend on the meteorology. Complete wind rose, topographic, hydrologic data, and environmental descriptions should be obtained to identify sensitive pollutant receptors and likely pathways. The type of meteorological data useful in planning a subsequent monitoring strategy is pointed out by Courtney.⁷ The instrumentation and methodology to obtain the pertinent meteorological data are outlined in Noll and Miller.⁹ Topographic and hydrologic information will be useful in planning soil, water (surface and underground), biota, and air sampling surveys. The various types of sampling should be performed in locations where future samplings during mill operation will occur both onsite and offsite. Samples should be taken and archived for subsequent analysis of background levels of contaminant species that may not have been identified prior to mill operation. Directional air sampling will be needed to identify significant highly directional sources that need to be quantified. The preoperational survey should include identification of remote background sites that may be useful for monitoring purposes. Of course, the preoperational survey absolutely assesses the background levels of radiation and radioactive materials in the environment where a mill is to be constructed.

8.0 BASIC RESEARCH

Throughout the discussion of this paper, research areas have been shown where they are required to fulfill the needs of each measurement technique or method. In general, this research is only necessary for the more sophisticated methodologies. This arises from the fact that although much equipment is available, it has not been evaluated for the specific job requirement.

Potential methods for determination of the contribution of a mill to the local background include continuous analysis of ^{222}Rn by use of lasers, and the use of trace element ratios in the ores to differentiate fugitive mill material versus that in the environment. A significant basic research effort will be necessary to develop the capability and feasibility of these approaches. At the present time, laser analysis is being used for determination of xenon and other specific gases. It is quite feasible for radon analysis. A system could be assembled for ground use and perhaps for air-

borne use at an estimated development cost of \$1 million. To use trace elements as tracers of mill material requires a comprehensive study of trace elements associated with all uranium ores and uranium mill sites. The costs of research would be about \$500 K. To implement a routine program, items like reactor time instruments for neutron activation analysis, atomic absorption instrumentation, mass spectrometers, and X-ray fluorescence systems must be obtained which would cost an additional \$500 K. It would require approximately \$150 K in capital equipment to implement following the basic research effort and a minimum of two man-years per year of expense involving personnel trained at the doctoral level in nuclear chemistry or physics.

As discussed in the body of this report, there are many sophistications which can be applied to the aforementioned strategies. In some instances, the increased costs could rise as high as \$300 K for capital and require four man-years of effort by trained personnel to achieve a given level of sophisticated effort.

9.0 APPENDIX

The procedures described below in this appendix are an attempt to provide a feeling for the detail which must be used in selected TLD and chemistry procedures. Personnel using these procedures must have scientific training to assure their effectiveness. Sampling detail has not been included since it is site specific and has already been discussed in the body of the text.

9.1 Dosimeter Handling Procedures⁵²

1. Inspect the TLD chips, discard any that are chipped, cracked, damaged.
2. Clean TLD chips, if visibly dirty, with trichloroethane. Then rinse with ethyl alcohol and air dry.
3. Place TLD chips in clean annealing dishes, making sure the chips are not living one on top of the other.
4. "Pre-exposure" anneal TLD chips at 400°C for one (1) hour in a furnace equipped with a nitrogen gas atmosphere.
5. Remove TLD chips from 400°C oven and place directly into an oven pre-heated to 100°C and hold for two (2) hours.

6. Remove TLD chips from 100°C oven and place in a shielded container. Cool to room temperature for several hours (usually overnight).
7. Annealed TLD chips are placed in dosimeters using clean forceps.
NOTE: Work under subdued light.
8. Store dosimeters in a shielded container or lead cave until ready to expose.
9. If dosimeters are to be shipped, controls must be kept at the processing lab in a shield.
10. Upon receipt of the dosimeters on site, three environmental dosimeters are placed in a shield, three others are placed in a selected background location off site.
11. The remaining dosimeters are individually exchanged with corresponding dosimeters in the environs in accordance with the site monitoring program.
12. Environmental dosimeters are placed at predesignated locations with little or no shielding. They should be placed between three and six feet above the ground or floor.
13. Appropriate numbers of environmental dosimeters are logged into notebooks. (Ten to twelve dosimeters of the same batch are retained in the lab for calibration and background determinations.)
14. After exchange, place exposed TLDs in a shielded enclosure and hold for 24 hours to allow low energy peak fading to occur.

9.2 TLD Reader Test Procedures

This technique is designed around the use of the "Harshaw" TLD system using the Model 2000A and 2000B detectors and integrating units and a glow curve storage oscilloscope. The TLD "chips" (3 mm x 3 mm x 0.89 mm) used in this program are also developed by the Harshaw Company. TLD reader systems built by other manufacturers have very similar operating procedures.

A. Reader Adjustment

1. Set range selector to X0.1 (Unit B) in automatic position. Meter reading should be 3-5%. If high or low, readjust current suppression on back of Unit B as required.
2. Set range selector to X1 position.
3. Set time period to 60 sec.
4. With drawer closed (Unit A)(heater on), push read button (Unit B). Observe temperature meter, set heater temperature control (Unit A) to hold temperature at 300°C.
5. Check controls T1 and T2. For TLD-700 LiF and 600 LiF, T1 should be at 110°C and T2 should be at 260°C. TLD-400, T1 should be at 110°C and T2 should be at 360°C. For TLD-300, T1 should be at 100°C and T2 at 310°C.
6. Repeat Step 4. When temperature is adjusted and heater shuts off, scale display should read 0.004 to 0.008 nCi.
7. Pull drawer on Unit A all the way open (heater off).
8. Push read switch (Unit B), after 60 sec time period; scale display should read the desired light source number. If not, readjust high voltage (Unit B) to obtain correct light source number.
9. After correct light source reading is obtained, set time period (Unit B) to 30 sec.
10. Close drawer (Unit A) heater is on, push read button (Unit B). Let run through a heat cycle.
11. Turn power on to storage monitor scope, store and erase buttons out. Now push STORE button in; scope will light. Next push ERASE button; scope will turn dark.
12. Push read button again (Unit B). A green dot and tracing should appear on the scope showing the temperature and read-out response of the TLD reader.

13. The system should now be calibrated and ready to start reading TLD "chips."

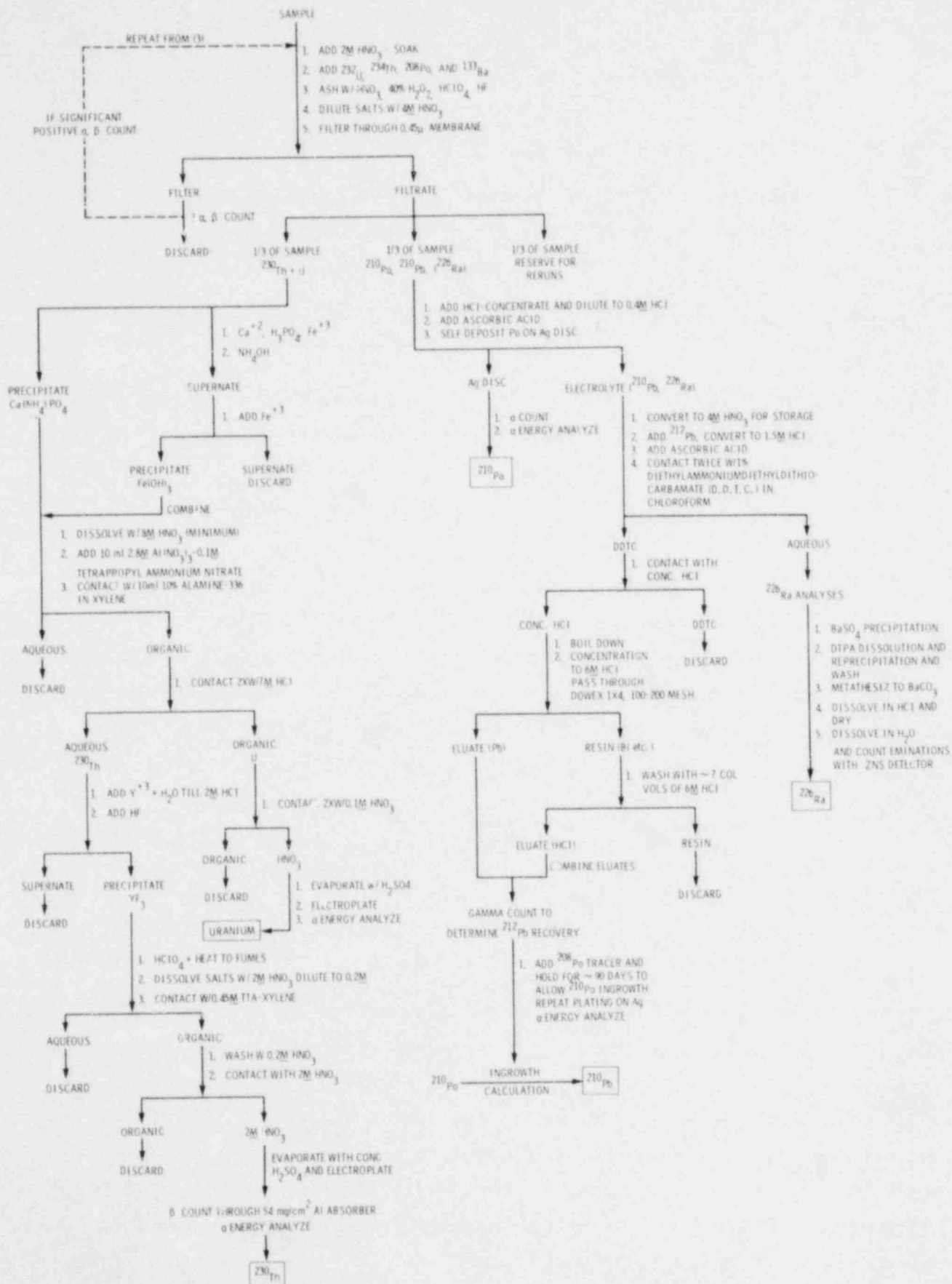
B. TLD Chip Readout

1. With drawer closed, time set at 30 sec, push read button; this is to heat planchet (heating tray). After unit turns off, open drawer.
2. When temperature drops to less than 100°C, place TLD chip (using plastic forceps or vacuum probe) on planchet in the middle of square pocket. NOTE: Work under subdued light (incandescent light).
3. Close drawer (softly so as to not dislodge TLD chip from placement).
4. When temperature drops to 70°C, push read switch. Temperature should rise; observe glow curve monitor scope and temperature meter. As TLD is read out, the response will be shown as a curve tracing on the scope and the numerical integration of light output will be recorded on the integrator.
5. Record all pertinent information of the TLD chip and the total integrated number from the display; record as nCi or μ Ci. The scope display is for verification of complete and proper readout. Pertinent information includes TLD batch number, type, position in dosimeter, identification of dosimeter, dates of exposure, date of reading, and comments about dosimeter condition or history.
6. After readout, open drawer and repeat Steps 2 through 5 for each TLD chip to be read.
7. Read out background TLD chips (TLDs from the same anneal and batch but not exposed).
8. Read out calibration chips for that batch.

9.3 Radiochemical Separation Procedures⁵⁴ for Radioisotopes Associated with Uranium

There are volumes^{55 64} detailing radiochemical procedures which can be used to isolate the radionuclides of uranium and their daughters. For simplicity only one series is shown schematically in Figure 9. Specific radiochemical procedures for various types of environmental samples are discussed in the following sections.

FIGURE 9. Flow diagram of radiochemical procedures for the analysis of ^{226}Ra , ^{210}Po , ^{210}Pb , ^{230}Th and U in environmental samples.



9.3.1 Wet Ashing Samples

Wet ashing with perchloric acid is a potentially dangerous procedure. Extreme caution and attention to detail must be followed. Never leave samples ashing in perchloric acid unattended. Have a wash bottle of distilled H₂O and one of 8N HNO₃ ready to add to samples. All ashing procedures are done wearing rubber gloves, in a fume hood, and using an explosion shield.

1. Weigh samples.
2. Transfer samples to vycor beakers, 400 ml for samples larger than 15 g; 250 ml for smaller samples.
3. Add ²⁰⁸Po, ²³²U, ¹³³Ba, ²³⁴Th spikes to each sample. Use appropriate amount based on sample weight and composition.
4. Cover samples with 4N HNO₃, put watch glasses on and heat until sample is well dissolved. Heat slowly (avoid spattering--don't boil vigorously) until the volume is reduced by about a factor of 4, which will give a nearly concentrated HNO₃. Add additional (~50 ml) concentrated HNO₃ to wash down walls and watch glass. Continue heating until solution is clear and light yellow. A residue (black or white) in the bottom of the beaker is permissible. For samples containing floating fat or oil, at this point heat to near boiling and add small squirts of 30% H₂O₂ (~250 μ). This will whiten the fat and speed ashing. Never add more H₂O₂ than nitric. Always add a small squirt of concentrated HNO₃ after the H₂O₂. (H₂O₂ without HNO₃ occasionally reacts explosively with samples.) When fats are nearly gone, the final ashing may be done using HClO₄.
5. Add concentrated HNO₃ and HClO₄ to samples--20 ml HClO₄ to all small samples and 50 ml HClO₄ to be es, livers, and large tissues. Wet ash one sample at a time in this manner.

Increase the heat setting of the hot plate gradually, keeping the solution just below the boiling point. Watch it closely--as the nitric acid evaporates, the boiling point increases, the color should lighten and perchloric acid will start to react when the nitric acid is nearly gone. When perchloric ashing takes place,

a vigorous boiling and foaming will occur. Do not let this get too vigorous. Keep a watch glass over the top and add $8N$ HNO_3 at the spout edge to cool the solution as needed. When the reaction is very vigorous, it will often continue to boil even if the sample is removed from the heat. At this point, the perchloric acid will occasionally react violently if the rate is not controlled. However, it is necessary to keep the reaction from stopping altogether, which will occur if too much $8N$ HNO_3 is added at one time. If the reaction stops, the sample will have to be heated until it starts. The sure sign of a runaway reaction with perchloric acid is a darkening color, either brown or green. If the solution starts to darken, immediately add a small (1 ml) squirt of $8N$ HNO_3 . It should lighten. If it does not, add more immediately. If a very dark color starts to form, add copious quantities of distilled H_2O using a coarse-tipped wash bottle to squirt it in vigorously. Turn off heat. These precautions will permit safe ashing of samples. After the vigorous reactions have subsided, remove the watch glass, rinse with concentrated HNO_3 into the beaker and put an air condenser on the sample. Boil off the concentrated HNO_3 and raise the temperature until the $HClO_4$ fumes copiously. Cool.

6. Transfer sample with $8N$ HNO_3 to a 250 or 400 ml teflon beaker, depending on size of sample. Add 10 ml concentrated HF. Place beaker in sand bath on a hot plate. Evaporate to incipient fumes of $HClO_4$ with heat lamps on. Remove from sand bath. Cool. Repeat this step twice.
7. Dilute with 25 to 200 ml $2N$ HNO_3 to dissolve all salts. Warm in sand bath. Cool. Assemble plastic filter holder with 0.22 membrane filter. Filter sample and rinse beaker and filter three times with a minimum of $4N$ HNO_3 and once with H_2O . Police beaker during rinses.
8. Transfer liquid to a volumetric flask for dilution--100 ml for filters, lungs, lymphs, and other small samples; 250 ml for bones and kidneys; 500 ml for small livers; and 1000 for large livers.

Use 4N HNO_3 for wash. Add concentrated HNO_3 as needed to bring average concentration to 4M. Bring to volume.

9. Put filter in wide metal dish, attaching with rubber cement. After standing 24 hours, count on low-background beta and alpha counters. Count each sample 1000 minutes.
10. If beta count rate exceeds 2 c/m or alpha count exceeds 0.5 c/m, remove the filter from the dish, place it in the sample beaker and wet ash with HNO_3 and HClO_4 , boil and redilute with the sample. Boil total volume down until it is one-half of original and refilter. Dilute to volume in volumetric flask.
11. Repeat Steps 9 and 10 until no significant activity is present on the filter.

9.3.2 Uranium-Thorium Separation for Samples Other Than Bone

1. To sample aliquot in 4M HNO_3 , add 4 ml of 10 mg/ml Ca carrier, 1 drop of Fe^{+3} (10 mg/ml), 1 ml of 1M H_3PO_4 . Mix in 100 ml lusteroid centrifuge tube.
2. Precipitate by slowly adding concentrated NH_4OH . Swirl. Add small excess. Let stand 5 minutes.
3. Centrifuge. Before decanting, add 3 drops Fe^{+3} while stirring the supernate rapidly. Recentrifuge. Decant supernate to waste.
4. Wash precipitate with 20 ml H_2O . Centrifuge. Decant supernate to waste.
5. Dissolve in 2 ml 8M HNO_3 . Add 100 mg ascorbic acid. Centrifuge to get droplets in one place and transfer to 60 ml separatory funnel.
6. Wash lusteroid with 10 ml 2.8M $\text{Al}(\text{NO}_3)_3$ -0.1M TPAH (TPAH = tetra-n-propylammonium hydroxide) and add to funnel. Take $\text{Al}(\text{NO}_3)_3$ in separate funnel. Add TPAH and Alamine 336 (a 10 volume % solution in xylene) - 10 parts aqueous to 1 part organic. Shake, allow to separate, drain aqueous; repeat.
7. Add 10 ml of 10% Alamine 336 in xylene (pre-washed with 0.5M HNO_3 ; then 7M HCl).

8. Contact 10 minutes. Centrifuge. Discard aqueous.
9. Add 5 ml 2.8M $\text{Al}(\text{NO}_3)_3$ -0.1M TPAH. Shake, centrifuge, and discard aqueous. Repeat.
10. Add 10 ml 7M HCl. Shake, centrifuge, and drain aqueous into polyethylene tube for Th analysis.
11. Repeat Step 10.
12. Add 10 ml of 0.125M HNO_3 . Shake, centrifuge, and drain aqueous into clean 30 ml beaker.
13. Repeat Step 12 and take to dryness under heat lamp.
14. If a residue is present, proceed to procedure for U purification using acetate-anion exchange column. Otherwise proceed directly to electroplating procedure.

9.3.3 Uranium Procedure

1. Dissolve residue in beaker in 1M acetic acid with heat; cool.
2. Prepare a column with Dowex 1 x 4 100-200 mesh Cl form resin (column ID - 9 mm; column volume - ~7 ml). Wash column with 50 ml ammonium acetate solution (60 g to 200 ml), then with 50 ml H_2O , and finally with 50 ml 1M acetic acid.
3. Pass sample in 1M acetic acid through column. Wash column with 50 ml 1M acetic acid. Discard eluate and wash.
4. Elute U from column with 50 ml of 4M HNO_3 followed by 20 ml of 0.125M HNO_3 . If a residue remains, repeat column procedure, boil down and electroplate.

9.3.4 Thorium Procedure

1. To Th fraction add 1 ml 10 mg/ml Nd carrier which is in a separatory funnel in 2M HNO_3 with 0.6M HDEHP. (Shake and drain some Nd from separatory funnel through filter paper in order to prevent HDEHP getting in sample.)
2. Add 15 ml concentrated HF to sample; vortex while adding water to bring volume to 50 ml. Let stand 30 minutes. Centrifuge; decant supernate to waste.

3. Wash precipitate two times with 15-20 ml of 1M HF-0.5M HNO₃.
4. Dissolve precipitate in 2M HNO₃. Add 40 ml 2M HNO₃ while vortexing; still vortexing add 5 ml concentrated HF. Continue vortexing while adding 2M HNO₃ until volume is 60 ml. Vortex longer if possible. Let stand at least 10 minutes. Check for complete precipitation. Centrifuge and decant supernate to waste.
5. Transfer precipitate to small teflon beaker with 8M HNO₃. Add 2-3 ml concentrated HClO₄ and take to near dryness under heat lamp. If sample volume is more than 50λ liquid HClO₄, Th will not extract well.
6. Dissolve residue in 3 ml of 1M HNO₃ and transfer to a 60 ml separatory funnel which contains 10 ml of 0.5M TTA in xylene.
7. Wash the beaker with exactly three 1 ml portions of 0.125M HNO₃ and add to separatory funnel. Contact 10 minutes; add 15 ml H₂O; shake 15 minutes; centrifuge; discard aqueous. All aqueous volumes must be measured carefully.
8. Wash organic with three 5 ml portions of 0.125M HNO₃. Discard washes.
9. Add 10 ml of 2M HNO₃; shake 15 minutes; centrifuge and drain aqueous to a clean 20 ml beaker.
10. Add 7 ml of 2M HNO₃ to funnel, drain 1-2 ml into beaker. Shake, centrifuge, and drain remainder into beaker.
11. Evaporate to dryness. Electroplate.

9.3.5 Uranium-Thorium Electroplating

1. Add 250 λ concentrated H₂SO₄ to sample and rinse beaker walls with 8N HNO₃ (minimum rinse).
2. Evaporate to just fumes of H₂SO₄. After HNO₃ is gone, no further vapor will be seen unless the temperature is raised.
3. Cool and add 1.5 ml H₂O, 2 drops .02% thymol blue indicator, and swirl. Then adjust pH using NH₃ gas (concentrated NH₄OH in special generator consisting of a polyethylene wash bottle with stem cut off inside). Color at end point matches pH 2.30 buffer with indicator. If overshoot, add a tiny drop of concentrated H₄SO₄, and redo.

4. Take a clean prepared cell, add 2 to 3 ml ethanol and let stand for 5 minutes to check for leaks.
 - (a) Prepare a cell by cleaning all parts with 8N HNO_3 , then water and alcohol. Then leak test. Discard ethanol.
 - (b) Transfer pH adjusted electrolyte with a transfer pipette.
 - (c) Put 5 ml of 1/100 H_2SO_4 (pH 2.3) in a 5 ml graduate. Add about 1/2 of it to the sample beaker and rinse the walls well using the pipette. Transfer the wash to the cell and repeat with the second half of the H_2SO_4 (total volume is $250 \lambda + 1500 \lambda + 5 \text{ ml} = 6.75 \text{ ml}$).
5. Place cell on electroplater such that the center electrode is about 3 mm below the surface of the solution.
6. Plug in motor speed control. Adjust speed of rotation to about 1 per second.
7. Clamp electrode.
8. Turn on.
9. Adjust the cell rheostat to give a current of 900 milliamperes.
10. Allow to plate for 40 minutes checking regularly to see that electrolyte solution does not boil. Also, do not permit it to evaporate below the center electrode. Adjust to lower current if boiling starts. (No lower than 800 milliamps.) Push cell upwards to keep center electrode immersed.
11. With cell running, rinse walls and electrode with a few drops of distilled H_2O (up to 2 ml). Use transfer pipette.
12. Allow to electroplate for 20 more minutes.
13. Add 5 ml concentrated NH_4OH . (Keep current on.) Let rotate for 20 seconds.
14. Lower the cell and dump the electrolyte back into its beaker. Rinse cell with distilled H_2O . Turn off current.
15. Disconnect cell. Rinse with distilled H_2O . Disassemble.
16. Use forceps to hold disc and rinse with distilled H_2O and alcohol. Drain on paper towel.

17. Place disc in 1" SS dish; using forceps, flame until dish turns color. Have no more color on edge of disc than pale yellow (about 5 seconds).
18. Cool and place in disc holder. Label with sample number, aliquot, amount of tracer added, date of calibration, and separation date.
19. Count Th fractions (using a 54.2 mg/cm aluminum absorber) on low background beta proportional counter. (Absorber stops soft ^{234}Th betas while passing most $^{234\text{m}}\text{Pa}$ betas.)
20. Count as soon as possible on alpha energy analyzer.

9.3.6 Polonium Analysis

1. Transfer sample aliquot to a 400 ml leached beaker. Evaporate to dryness. Do not bake.
2. Add 25 ml concentrated HCl and evaporate slowly to just dryness.
3. Add an additional 25 ml of concentrated HCl and repeat evaporation. Do not heat excessively.
4. Dissolve in 350 ml of 0.4M HCl and add 100 mg of ascorbic acid. Add teflon-covered magnetic stir bar.
5. Prepare a 2.2 cm silver disc by spraying one side with flat black Krylon, then with clear Krylon. Remove any Krylon from other side with a Q-tip slightly dampened with acetone. Then polish with metallographic Al_2O_3 and rinse with distilled H_2O . Suspend this disc in the solution by hanging from a pyrex hood fixed to a 75 mm watch glass.
6. Heat solution to 90°C while stirring and deposit onto disc for a period of 4-6 hours.
7. Remove disc from solution. Rinse with distilled H_2O and dry. Place disc in folder and count alpha particles with alpha energy analyzer.
8. If ^{210}Pb is to be run, boil down 0.4M HCl with 25 ml concentrated HNO_3 . Take to 25 ml; add 25 ml more concentrated HNO_3 . Take to near dryness and pick up in 4N HNO_3 .

9.3.7 Lead Analysis

1. Take samples to dryness or HClO_4 fumes, and dissolve in 20 ml 1.5M HCl.
2. Transfer sample to a 60 ml separatory funnel. Add ~ 1000 c/m of ^{212}Pb tracer, [NaI(Tl) well counter], and 500 mg of ascorbic acid.
3. Add 20 ml 1% DDTc in chloroform; shake 10 minutes. Centrifuge and drain lower (organic) phase into a clean beaker. (DDTC = diethylammonium diethyldithiocarbamate).
4. Add an additional 20 ml 1% DDTc; shake 5 minutes. Centrifuge and combine organic in first beaker.
5. Drain aqueous to 100 ml polyethylene bottle and save for ^{226}Ra analysis. Rinse separatory funnel with H_2O ; save rinse also.
6. Return the 40 ml of organic to the separatory funnel.
7. Add 10 ml concentrated HCl, shake momentarily, vent, shake for 10 minutes. Centrifuge and drain organic (lower) phase to organic waste container.
8. Add 10 ml of CCl_4 ; shake briefly to remove any traces of DDTc from aqueous phase. Drain lower phase to waste and repeat wash with an additional 5 ml of CCl_4 .
9. Drain aqueous to beaker and evaporate to half volume, or less. Cool.
10. Prepare a 5 ml column of Dowex 1 x 4 100-200 mesh resin. Rinse column with five 5 ml washes of 6M HCl before using. Discard rinses.
11. Pass sample through column. Rinse column with 7 column volumes or ~ 35 ml of 6M HCl. Note time as separation time.
12. If sample is spiked with ^{212}Pb tracer, catch elutes in vial and dilute to a volume for calibration.
13. If measuring radiochemical yield with ^{212}Pb , add ~ 1000 c/m each to two vials and dilute to volume. Count all samples from Step 12

and the two vials on standard NaI(Tl) crystal until 10 counts are recorded in the peak area.

14. Transfer samples to 150 ml beakers. Add ^{208}Po tracer. Take sample to near dryness and wet ash with concentrated HNO_3 and H_2O_2 until organic is mostly destroyed.
15. Take samples to incipient dryness. Dissolve in about 10 ml of 4N HNO_3 and transfer to small glass vials for storage. Rinse as needed using 4N HNO_3 . Store for ^{210}Po ingrowth for about 138 days.
16. Transfer to 400 ml vycor beakers and proceed with Po separation on silver disc. Do not add additional ^{208}Po tracer.

9.3.8 Radium Analysis

1. Transfer sample (~ml in 1.5M HCl) to a 100 ml glass centrifuge tube. Add 1 ml Ba carrier and 1/2 g KCl and vortex. If sample is not dissolved completely at this point, heat to a boil. If solids remain, transfer to a 250 ml beaker and dilute with .125M HNO_3 to 190 ml; heat. Increase KCl to 1 g.
2. Add 10 ml 6M H_2SO_4 ; heat to a boil; cool; and centrifuge. Discard supernate. (Use 20 ml H_2SO_4 if using a larger volume in a beaker.)
3. Wash ppt with .01M H_2SO_4 ; heat to a boil; cool; centrifuge. Discard supernate. Repeat. (Use 10-50 ml wash and wash up to three times to get rid of salts.)
4. Add 10 ml DTPA (diethylene triamine penta acetic acid) solution [5 g DTPA, 2 ml acetic acid and 9.6 ml piperidine per 100 ml (pH ~10)].
5. Add 2 drops H_2O_2 and heat in water bath until clear.
6. Add 12 drops acetic acid and 12 drops 10% Na_2SO_4 (pH 5.3) while stirring. (Precipitate re-forms)
7. Heat to a boil; cool; centrifuge; and discard supernate.
8. Wash precipitate with 10 ml of "DTPA wash" solution (containing 40 ml H_2O , 5 ml DTPA solution, 6 drops acetic acid, and 6 drops

10% Na_2SO_4 . Centrifuge. Discard supernate.

9. Add 20 ml H_2O and 20 ml Na_2CO_3 (saturated). Heat to boil. Cool; centrifuge; discard supernate. Add 5 ml Na_2CO_3 and vortex. Transfer slurry to a 15 ml glass centrifuge tube. Rinse large cone with 1% saturated Na_2CO_3 in H_2O . Discard supernate. Wash precipitate one time with 1% saturated Na_2CO_3 in H_2O ; centrifuge; and discard wash. Then wash one time with 0.1% aerosol and two times with isopropanol. Rinse precipitants of Na_2CO_3 with isopropanol without stirring the first time to get rid of all water. Vortex the second rinse.
10. Transfer to a weighed 1 ml volumetric flask with transfer pipette using small isopropanol washes. Wash any residue from sides of cone by scraping and washing with pipette until sides are clean. Centrifuge and use supernate as wash to clean cone. Recovery depends on thoroughness of cleansing cones when transferring. Centrifuge and discard isopropanol.
11. Place in vacuum desiccator and evacuate. Allow to remain until totally dry.
12. Remove from desiccator; weigh; record weight. Dissolve precipitate in 200 μ 2M HCl. Vortex. Centrifuge. Replace in desiccator until dry.
13. Remove from desiccator and add 200 μ of distilled water. Vortex to dissolve precipitate and centrifuge to get all droplets to bottom.
14. Remove cotton from end of small-tip cotton applicator and push to the bottom of flask to absorb sample, wiping down walls of flask with cotton ball. (Use applicator stick.) Place a glass ring over cotton. Evacuate for 5 minutes in desiccator and take vials to be counted in NaI(Tl) counter to measure ^{133}Ba recovery. Count within 2 hours.
15. Paint the inside of flask with 150 \pm 5 mg of ZnS mixture using end of applicator. Clean all paint from top of flask with acetone on an applicator. If paint remains, stopper will not seal.

16. Make up epoxy, small amounts at a time. With end of applicator stick paint a bead of epoxy around flask top. Then coat sides of stopper. Place in bell jar and evacuate ~5 minutes. Record time. NOTE: When put under vacuum, stoppers should rise. When vacuum is released, they should go back down.
17. Store for ~30 days before counting in radium coincidence scintillation counter.

PREPARATION OF ZnS MIXTURE

In a 100 ml beaker mix: 3 g ZnS (General Electric Type 118-2-11 RETMA Type P-11)
2.25 g white petroleum jelly
0.75 g white mineral oil

Stir until well blended.

RADIUM-226 STANDARD

Make standard by pipetting 1 ml Ba carrier into 15 ml cone. Add Na_2CO_3 until a permanent ppt forms. Transfer to 1 ml volumetric flask. Dissolve in 200 μ of 2N HCl. Add ^{226}Ra tracer. Dissolve in 200 μ distilled H_2O . Add cotton scintillator and seal as per sample procedure.

9.3.9 ^{234}Th Tracer Preparation

1. Put 1 ml purified Nd carrier in each of 2 polypropylene cones. Add ~40 ml uranium nitrate in 0.1 HF stock to each while swirling. Allow to stand 1/2 hour for precipitate to coagulate.
2. Centrifuge fast for 5 minutes and decant the stock U back to bottle for storage.
3. Wash precipitate two times with ~50 ml 1M HF + 0.5M HNO_3 . Centrifuge after each wash. Combine precipitates.
4. Dissolve precipitate in 60 ml of 2N HNO_3 . Add 5 ml concentrated HF. (While vortexing, add about 1/2 of the 2N HNO_3 , then add the HF. Add remainder of 2N HNO_3 .) Continue vortexing for 3 minutes.
5. Centrifuge. Check activity and discard supernate.

6. Transfer precipitate to teflon beaker with minimum 2N HNO_3 (use pipette). Add 2-3 ml HClO_4 and heat on hot plate in sand bath to HClO fumes.
7. Transfer to pyrex beaker and fume to dryness. Do not bake.
8. Dissolve solids in 5 ml of 7M HCl and transfer to a clean 60 ml separatory funnel. Rinse beaker with 5 ml 7M HCl and add to funnel.
9. Add 10 ml 10% alamine-336 in xylene (prewashed with 7M HCl) and shake for 10 minutes.
10. Drain aqueous back to beaker and evaporate to dryness. Add 1 ml concentrated HClO_4 and 1 ml concentrated HNO_3 and take nearly dry.
11. Pick up in 3 ml 1M HNO_3 . Transfer to separatory funnel which has been rinsed clean. Rinse beaker three times with 1 ml H_2O . Add to separatory funnel. Add 10 ml 0.5M TTA (thenozltrifluoroacetone) in xylene. Shake thoroughly.
12. Add 9 ml H_2O and shake 15 minutes.
13. Discard aqueous and wash organic phase with three 5 ml portions of 0.2M HNO_3 . Shake 5 minutes each and discard. Wash.
14. Add 10 ml 2M HNO_3 . Shake 15 minutes, centrifuge, and drain aqueous to a 20 ml beaker.
15. Evaporate to just dry and fume to dryness with 1 ml HClO_4 to destroy any organic.
16. Pick up spike with 20-25 ml 2N HNO_3 .
17. Assay beta activity of 1 ml of tracer mounted on 2.5 cm stainless steel counting dish. Use 54.2 mg/cm^2 absorber when counting. Count 1000 minutes on alpha scintillation counter to check for contamination.

9.3.10 Uranium-232 Tracer Purification

Prepare a column of about 5 ml dowex 1 x 4 100-200 resin. Wash with 50 ml 7M HCl. Take spike to dryness. Pick up in same 7M HCl. Put on column. Wash with 50 ml 7M HCl. Elute ^{232}U with 20 ml .125M HNO_3 in a clean beaker. Evaporate to dryness. Pick up in 10 ml 2M HNO_3 . Prepare six discs to calibrate tracer.

9.3.11 Lead-212 Tracer Purification

1. Put 70 ml of 0.3M HDEHP (diethylammonium diethyldithiocarbamate) which contains 12 mg of extracted thorium nitrate for milking of ^{212}Pb tracer in separatory funnel.
2. Add 5 ml 6M HCl to the separatory funnel and shake for 4 minutes. Centrifuge to separate phases.
3. Carefully drain lower phase into a clean 60 ml funnel.
4. Add 20 ml of 0.075M HDEHP in isooctane to the funnel. Shake 4 minutes and centrifuge to separate phases.
5. Transfer lower phase to a column made from 3 ml 100-200 mesh resin dowex 1 x 4 CL form. The resin should be prewashed with ~20 ml of 6M HCl. Discard upper (organic) phase into organic waste.
6. Allow spike solution to drip through column. Discard first 4 ml. Wash column with an additional 6 or 7 ml 6M HCl. Catch elutes in a graduated test tube. Dilute solution in test tube with H_2O until volume is four times the original. Transfer to a separatory funnel containing 20 ml 1% DDTc in chloroform (make fresh is more than one week old). Shake 10 minutes, centrifuge, or use phase separating filter paper and drain lower (organic) phase to a clean beaker. Discard upper phase to contaminated aqueous waste.
7. Return organic to separatory funnel (after cleaning funnel with acetone and H_2O). Add 10 ml concentrated HCl. Shake 5 minutes, centrifuge, and discard lower phase.
8. Add 10 ml carbon tetrachloride to separatory funnel and shake 1 minute. Discard lower phase.

9. Drain upper phase through pleated filter paper into a beaker, rinse filter with a minimum of concentrated HCl and evaporate to near dryness. Wet ash with H_2O_2 and HNO_3 to destroy organic. If organic persists, add $\sim 1/2$ ml $HClO_4$ and 2 ml HNO_3 and take to dryness.
10. Pick up in 1.5M HCl. Dilute to ~ 10 ml in a small bottle.
11. Take two 1 ml aliquots of the tracer and dilute to volume in a polyethylene scintillator vial to count in a NaI(Tl) counter for assay of tracer.

10. ACKNOWLEDGMENT

The authors would like to thank Dr. George Sehmel, C. W. Thomas, M. R. Petersen, and Paul Nickola of Battelle-Northwest for their helpful discussions. Special thanks are given to R. W. Perkins for his critical review of the text and helpful comments.

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