
A Review of Research Conducted by Los Alamos National Laboratory for the NRC with Emphasis on the Maxey Flats, KY, Shallow Waste Burial Site

Prepared by E.B. Fowler, W.L. Polzer

Los Alamos National Laboratory

Prepared for
U.S. Nuclear Regulatory
Commission

8809220040 880831
PDR NUREG
CR-5170 R PDR

NOTICE

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, or any of their employees, makes any warranty, expressed or implied, or assumes any legal liability of responsibility for any third party's use, or the results of such use, of any information, apparatus, product or process disclosed in this report, or represents that its use by such third party would not infringe privately owned rights.

NOTICE

Availability of Reference Materials Cited in NRC Publications

Most documents cited in NRC publications will be available from one of the following sources:

1. The NRC Public Document Room, 1717 H Street, N.W.
Washington, DC 20555
2. The Superintendent of Documents, U.S. Government Printing Office, Post Office Box 37082,
Washington, DC 20013-7082
3. The National Technical Information Service, Springfield, VA 22161

Although the listing that follows represents the majority of documents cited in NRC publications, it is not intended to be exhaustive.

Referenced documents available for inspection and copying for a fee from the NRC Public Document Room include NRC correspondence and internal NRC memoranda; NRC Office of Inspection and Enforcement bulletins, circulars, information notices, inspection and investigation notices; Licensee Event Reports; vendor reports and correspondence; Commission papers; and applicant and licensee documents and correspondence.

The following documents in the NUREG series are available for purchase from the GPO Sales Program: formal NRC staff and contractor reports, NRC-sponsored conference proceedings, and NRC booklets and brochures. Also available are Regulatory Guides, NRC regulations in the *Code of Federal Regulations*, and *Nuclear Regulatory Commission Issuances*.

Documents available from the National Technical Information Service include NUREG series reports and technical reports prepared by other federal agencies and reports prepared by the Atomic Energy Commission, forerunner agency to the Nuclear Regulatory Commission.

Documents available from public and special technical libraries include all open literature items, such as books, journal and periodical articles, and transactions. *Federal Register* notices, federal and state legislation, and congressional reports can usually be obtained from these libraries.

Documents such as theses, dissertations, foreign reports and translations, and non-NRC conference proceedings are available for purchase from the organization sponsoring the publication cited.

Single copies of NRC draft reports are available free, to the extent of supply, upon written request to the Division of Information Support Services, Distribution Section, U.S. Nuclear Regulatory Commission, Washington, DC 20555.

Copies of industry codes and standards used in a substantive manner in the NRC regulatory process are maintained at the NRC Library, 7920 Norfolk Avenue, Bethesda, Maryland, and are available there for reference use by the public. Codes and standards are usually copyrighted and may be purchased from the originating organization or, if they are American National Standards, from the American National Standards Institute, 1430 Broadway, New York, NY 10018.

NUREG/CR-5170
LA-11354-MS
RW

A Review of Research Conducted by Los Alamos National Laboratory for the NRC with Emphasis on the Maxey Flats, KY, Shallow Land Burial Site

Manuscript Completed: May 1988
Date Published: August 1988

Prepared by
E.B. Fowler, W.L. Polzer

Los Alamos National Laboratory
Los Alamos, NM 87545

Prepared for
Division of Engineering
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, DC 20555
NRC FIN A7040

ABSTRACT

Studies to determine the impact of the Maxey Flats low-level waste burial site on the environment have been conducted since 1963. Neither the migration or lack of migration of waste radionuclide to offsite by subsurface flow has been unequivocally proved; the migration of tritium may be an exception. Some radionuclides, e.g. plutonium, did migrate short distances from a burial trench. The movement of "only a short distance" was attributed to the importance of a biopopulation in the soil and to the importance of soluble iron as a competitor for the chelate of the potentially mobile plutonium/chelate system. In both cases a soluble plutonium complex is degraded and the plutonium is released in a form that is sorbed by the soil. Soil moisture data in conjunction with tritium data indicate that infiltration into the trench was predominantly through the trench cap. Tritium data also indicate subsurface migration of tritium.

CONTENTS

ABSTRACT	111
I. INTRODUCTION.	1
II. STUDIES BEFORE 1978	2
III. STUDIES BEYOND 1978	4
A. Laboratory Studies-Sorption Related to Soil Type	4
B. Preliminary Sorption Experiment.	4
C. Electrical Charge Relationships.	8
D. Laboratory Studies on Maxcy Flats Waste and Soil	15
E. Fate of Complexed Radionuclides.	15
F. Role of Iron in Chelate/Pu System.	24
G. Tritium.	40
H. Soil Moisture Studies.	42
I. Deep Well Studies.	48
J. Radionuclides Offsite.	51
K. Experimental Trenches.	51
IV. SUMMARY	55
V. IN RETROSPECT	57
A. Establishment of Waste Burial Sites.	58
B. Existing Waste Burial Sites.	60
REFERENCES	61

A REVIEW OF RESEARCH CONDUCTED BY LOS ALAMOS NATIONAL LABORATORY
FOR THE NRC WITH EMPHASIS ON THE MAXEY FLATS, KY, SHALLOW
LAND WASTE BURIAL SITE

by

E. B. Fowler and W. L. Polzer

I. INTRODUCTION

A legacy from the birth and growth of the Atomic Age, one by-product, radioactive waste, continues to challenge a wide variety of disciplines--both scientific and nonscientific--with regard to its handling, treatment, and disposal. After more than four decades of effort, much is left to be learned. The apparent slow progress cannot be related to lack of interest, but rather to the nature of the material to be disposed. Whereas other industrial wastes may be oxidized or reduced or incinerated or even recovered as useful materials, radioactive waste remains radioactive whatever approach to treatment or storage or disposal is employed. Thus, it cannot be released to the environment and in its case, dilution is not the solution to pollution. Containment is the only plausible option and containment implies a knowledge of material/environment interaction in some cases for thousands of years---hence, an apparently impossible task.

For those radioactive wastes disposed over the past forty years, the problems take on a different hue. Much of the radioactive waste generated in the Manhattan Project and early AEC days was stored "on site." High level liquid wastes at the Hanford works were stored at tank farms and some special wastes at Los Alamos were stored in the "General's Tanks."

Contaminated solid material was buried in special areas dedicated to receive such debris. As the volume of radioactive waste generated in the nuclear communities increased, the lack of space "on site" dictated the establishment of other (low-level) burial grounds dedicated to the containment of such wastes.

Shallow-land sites for the burial of low-level radioactive wastes were selected with a view to serving production areas; they should be removed from population centers, should not be subject to flooding, should be divorced from usable aquifers, and should not be subject to erosion; stimulation of a local economy was also considered important. One site selected was at Maxey Flats, KY, about 9 miles northwest of Morehead. The burial site proper covers about 45 acres; burial of waste was initiated in 1963.

High level wastes were excluded from Maxey Flats and liquid wastes were excluded after 1972. Wastes were to be shipped to the site in containers suitable for handling. Steel drums, wooden boxes, cardboard boxes, and plastic wrapped debris formed the bulk of the waste received. It is probable that some liquid chemicals in containers (glass?) were received as part of the trash in barrels.

Wastes were disposed in excavated pits and covered with the soil/rock debris that had been removed at the time of pit excavation. The fill was compacted and caps of soil mounded over the closed pit.

Water accumulates in the trenches either from percolation of precipitation (annual rainfall about 43 inches) and/or from subsurface flow of groundwater. The trenches are pumped and the removed liquid passed through a gas fired evaporator. Vapor is disposed to the atmosphere; it contains tritium and traces of other radionuclides.

Radionuclide contaminants reported in off-site water and sediments as well as other considerations led to the closure of the site (1977) to the receipt of off-site waste for on-site burial. Pumping of the pit waters and evaporation continue. The evaporator heel is buried onsite. Other current activities include attempts to reduce infiltration through the installation of plastic covers on the surface at the site.

This document presents a brief review of research conducted at Los Alamos National Laboratory pertinent to the Maxey Flats, KY, burial site. It is not intended that an all inclusive compilation be presented. An attempt has been made to present laboratory observations, to carry those forward to field application, and to draw conclusions that most reasonably (as interpreted by these authors) are supported by the data presented. The conclusions are not presented as "Scripture"; in an area of study where there are as many concrete and absolute unknowns as there are associated with subsurface migration of radionuclides from burial pits, there can be a number of interpretations of data. The lack of extensive detail is intentional; it is the contribution of each referenced work to an integrated scheme that is important. For those who wish to review detail, references are listed.

The charter, which we were given about 1977, requested answers to: (1) are radionuclides migrating from the trenches at Maxey Flats, (2) if so are they migrating off site, (3) if so, what is migrating and how, and (4) can the rate of migration be determined? In retrospect, each of those questions should be asked prior to the establishment of a low-level waste burial site; the appurtenances required to obtain data necessary to acceptable answers should be installed at the beginning. It is gratifying that after all the effort at Maxey Flats starting in 1963, proof positive of migration to off-site from the burial pits has not been found. Tritium may be an exception as shown by Rickard and Kirby¹. The evidence presented supports their conclusion that subsurface migration of tritium has occurred, however, within the site boundary not off site. New approaches, new techniques, may demonstrate otherwise. Samples will be available to many future generations.

II. STUDIES BEFORE 1978

Two of the more important efforts at the Maxey Flats site prior to 1978 related to monitoring and to a search for possible migration of waste liquids from the burial trenches to depths well beyond the floor of those trenches. The latter effort was undertaken by the USGS; drilling rigs explored to depths ranging from about 28 ft to 310 ft.

In an extensive report by Zehner² published 1983, the USGS efforts through 1979 are detailed. Data are presented relative to possible rates of flow through the soil, migration of some radionuclides, and the importance of fissures to flow patterns. However, the reader is left with continuing uncertainty relative to transport of radionuclides off site. The author deduces a minimum transport velocity of 50 ft/yr for cobalt-60 and manganese-54 by fissure flow. On the other hand, the final conclusion reached is that "Over-land runoff is the most likely means of transport . . ." (page 131, paragraph 5).

With respect to radiochemical analyses of samples from wells drilled by the USGS, the authors state "samples from most wells are of limited value in determining radiochemical quality of ground water at the burial site" (page 109, paragraph 4). The fact that specific isotopes (³H, ⁹⁰Sr, ⁶⁰Co, ²³⁹Pu, ²³⁸Pu) had been detected was not considered strong evidence for subsurface migration; several mechanisms are discussed other than subsurface migration, which could account for their presence.

In a 1975 report, Meyer³ reviewed again the possible pathways for migration of radionuclides to uncontrolled environment. The author points out ". . . there is insufficient radiological and hydrological information to estimate in what form, in what quantity, or at what rate Pu is migrating." Perhaps a more important conclusion related to the inability of containers and trenches in humid areas (of that time period) to retain radioactive waste. The paper has value as a presentation of earlier waste management practices.

A third study relative to the referenced time period was that of Montgomery et al.⁴ The report details an evaporator study, an environmental study and a test well study. Certain recommendations were made for future research, some of those recommendations may later have been considered. Conclusions related to the evaporator stack effluent as an important source of tritium at the environs. Further ". . . the transport of radionuclides from the site surface by precipitation run-off was a major source of the radioactivity detected in the drainage pathway" (page 73, paragraph 1). The radionuclides referenced were ⁵⁴Mn, ⁶⁰Co, ⁹⁰Sr, ¹³⁷Cs, ²³⁸Pu, and ²³⁹Pu.

Relative to the presence of radionuclides in test well samples (only ³H, ⁶⁰Co, and ⁹⁰Sr in the soluble fraction), the authors state that even though subsurface transport might explain the occurrence of radionuclides in the test wells, their presence off site appears to be primarily from surface water run-off and evaporator plume depletion. Thus, at the time of closure of the Maxey Flats site late in 1977, the following questions remained:

1. Does subsurface migration of radionuclides occur?
2. If there is migration, what migrates and in what quantity and what direction?

3. If certain radionuclides do not migrate, what is the inhibitory mechanism?
4. What soil chemistry retards or enhances migration?

The remainder of this report reviews the research after 1978, which addressed the above questions.

III. STUDIES BEYOND 1978

A. Laboratory Studies - Sorption Related to Soil Type

An initial assignment from the Nuclear Regulatory Commission (NRC) to the Los Alamos National Laboratory related to a study of different soil types with respect to their chemical and physical characteristics and their ability to sorb different radionuclides commonly found in the leachate obtained at waste burial sites. Those results are reported here to introduce the research approach employed later throughout the Maxey Flats study.

Investigations concomitant with the field studies at Maxey Flats were conducted in the laboratory on a variety of soils collected within the continental United States. Soils had been collected in 1/2 to 1 ton amounts each from the A, B, and C horizons of the respective soil profiles. The soils collected and their characteristics have been reported.⁵

B. Preliminary Sorption Experiments

Data relative to the differential sorption of various radionuclides from waste by two different soils and by different horizons of the same soil are presented in Table I. Results are presented for isotopes of plutonium, americium, and uranium with respect to their sorption by Fayette soil and Fuquay soil. Comparisons among the horizons as well as between the soil types can be made. The two soils differ in degrees of sorption for all radionuclides as specific horizon is compared with respective horizon. Further, for a specific radionuclide and a specific soil, the degree of sorption varies among horizons. Those observations are neither surprising nor earth shaking; they provide documentation.

The degree of sorption of a waste radionuclide by the soil as waste solution percolates from the burial pit into soil environment influences the extent of migration. The chemical and physicochemical nature of the soil solution (containing waste) as well as the nature of the soil (or soil horizon) determine the degree of sorption. Relative data obtained by analyses of waste, soil and soil/waste solution for the two soils are presented in Table II. A definite relationship between degrees of sorption (Table I) and a chemical entity or combination of entities (Table II) was not evident for the two soils under study. The pH value of 8.01 (A2 horizon) of the waste/Fuquay soil system (Table II) is probably the result of elutriation from the Ap horizon as the soil has developed. The pH of the Ap, B2t and C horizon soil/waste solutions decreases from 7.55 to 7.26 to 7.12 respectively. The soils are acid (pH = 6.8); the solution is slightly basic due to the addition

TABLE I
RADIONUCLIDE CONCENTRATIONS IN TWO RAW WASTES AND DEGREES OF SORPTION IN WASTE/SOIL SYSTEMS

System	^{238}Pu	$^{239,240}\text{Pu}$	^{241}Am	^{238}U	^{235}U	$^{233,234}\text{U}$	pH
<u>Fayette Soi</u>							
<u>Raw Waste (dpm/ml x 10²)</u>							
Total	11300±400	98±3	113±5	9±1	4.1±0.1	20±1	
Soluble	30±10	3±1	3±1	10.7±0.5	4.4±0.4	21.8±0.5	
<u>Degrees of Sorption (%)</u>							
<u>Raw Waste/Fayette Soil</u>							
Ap	83.3	80.0	85.0	72.9	80.9	71.1	8.03
B	88.0	88.3	83.3	96.3	95.5	90.8	7.20
C	87.7	86.7	86.7	92.5	96.6	86.2	7.60
<u>Fuquay Soil</u>							
<u>Raw Waste (dpm/ml x 10²)</u>							
Total	11300±400	98±3	113±5	9±1	4±1	20±1	
Soluble	21±3	2.0±0.4	1.8±0.4	10.4±0.1	3.7±0.1	22.5±0.4	
<u>Degrees of Sorption (%)</u>							
<u>Raw Waste/Fuquay Soil</u>							
Ap	-47.6	-40.0	38.9	95.7	99.2	97.8	7.55
A2	-33.3	-15.0	-16.7	62.5	64.7	64.4	8.01
B2 _r	98.6	97.0	90.0	99.2	99.7	99.1	7.26
C	99.0	91.0	79.4	99.4	99.5	99.9	7.12

The degree of sorption is the ratio between the concentration in the raw waste/soil system (i.e., soluble fraction) divided by the concentration in the soluble fraction of raw waste for the respective radionuclide times 100. In the case of Raw waste/Fuquay soil, the five negative numbers indicate negative sorption, i.e., solubilization from the respective insoluble fractions in the raw waste.

TABLE II
CHEMICAL AND PHYSICAL PROPERTIES OF RAW WASTE AND RAW WASTE/SOIL SYSTEMS

System	EC mmhos	Na mx10 ²	Ca mx10 ⁴	CO ₃ ⁻ mx10 ⁴	HCO ₃ ⁻ mx10 ³	Al mx10 ⁶	Fe mx10 ⁶	COD mx10 ³	PO ₄ ⁼ mx10 ⁶	pH	Eh mV
<u>Fayette Soil</u>											
Raw Waste											
total	1.12	1.4	17.5	8.7	9.0	40.4	272	6.18	370	8.71	384
Soluble	1.07	1.3	0.21	5.9	7.2	3.7	2	1.56	10.7	8.74	399
Raw Waste/Fayette Soil											
Ap	1.03	1.12	11.9	0.52	8.5	3.7	2	4.8	10.9	8.03	416
B	0.72	0.80	4.4	0.01	3.7	3.7	1	2.2	5.2	7.20	411
C	0.83	0.93	7.9	0.14	5.8	3.7	0.5	2.2	8.8	7.60	412
<u>Fuquay Soil</u>											
Raw Waste											
Total	1.12	1.38	17.5	8.7	9.0	40.4	273	6.18	370	8.71	8.84
Soluble	1.18	1.29	5.3	9.9	7.8	3.7	3.8	1.6	9.8	8.57	3.97
Raw Waste/Fuquay Soil											
Ap	0.97	1.2	1.7	0.16	6.78	10.0	6.8	4.7	9.5	7.55	411
A2	1.08	1.24	4.7	0.51	8.03	14.8	10.0	3.8	8.0	8.01	379
B2†	0.94	0.95	0.75	0.04	3.47	3.7	2.0	0.97	0.28	7.26	432
C	0.84	0.95	0.31	0.03	3.40	3.7	2.0	0.98	0.4	7.12	440

A correlation between any of the properties and the degree of sorption (Table II) is not apparent. An exception may be the inverse relationship between pH and degree of sorption for uranium and americium by the Fuquay A2 horizon soil and for uranium by the Fayette Ap horizon soil.

(4:1) of waste (pH 8.7) to the soil in the batch method employed in the experiment. Again in the case of the Fuquay system (Table I) and for americium and uranium species, the lowest degree of sorption was noted for the A2 horizon where the pH was highest (8.01). For the Ap, B2t, and C horizons, degrees of sorption increased with a decrease in pH. However, for the two plutonium species, ^{238}Pu and ^{239}Pu , ($^{239,240}\text{Pu}$ are considered together), the lowest degree of sorption was noted for the Ap horizon. In point of fact, the concentration of the two species in the supernate after reaction of waste with Ap horizon soil was greater than their respective concentrations in the soluble fraction of the waste per se; results for plutonium species and americium species in the A2 horizon were similar. The conclusion was reached that the Ap and A2 horizons contain in their soluble organic fraction a chelate or complexing entity that solubilizes plutonium from the insoluble fraction of the waste and that the resulting complex is not sorbable. In the strict sense of ion exchange the complex possesses the wrong charge or no charge.

In the case of Fuquay Ap horizon soil, the degrees of sorption for ^{238}Pu and $^{239,240}\text{Pu}$ were negative, i.e., plutonium was brought into solution from the insoluble fraction. On the other hand, the degrees of sorption for those species by the Fayette Ap horizon soil were positive. The difference is attributed to the presence of chelates (Fuquay) as opposed to their absence or, if present, in lesser amounts (Fayette). Other experiments have produced similar results.

The results from the initial investigations can be summarized as:

1. soils differ markedly with respect to their degree of sorption of different waste radionuclides,
2. the horizons within a specific soil differ with respect to their degree of sorption of different waste radionuclides,
3. in general, the degree of sorption increases with the depth of the profile,
4. some soils, notably the Ap horizon of the South Carolina Fuquay soil, possess entities that solubilize plutonium from the insoluble fraction of the waste,
5. the solubilized plutonium (4 above) is apparently converted to a form (a chelated form has been suggested), which does not sorb to the soil,
6. a relationship between degree of sorbability and chemical constituents of the soil/waste solution is not readily apparent, and,
7. in the case of the Fuquay soil, the pH of the solution and thus the charge on the respective radionuclides is related to the degree of sorption by the soil.

Item 7 above dictated the following experimental approach, i.e., determine the charge characteristics of the various radionuclides in the waste in the absence and presence of soils and relate the charge characteristics to the degree of sorption.

C. Electrical Charge Relationships

Two possible modes of removal of radionuclides as related to the hydrogen ion concentration of the solution involve precipitation (or coprecipitation) and ion exchange. In the majority of soils, cation exchange will be predominant.

The physical nature of the insoluble fraction of radioactive waste was investigated using autoradiographic techniques.⁶ The association of alpha emitting material with a crystalline precipitate is shown in Fig. 1. The crystalline material was shown by X-ray diffraction to be more than 60% calcium carbonate.

The unassociated radioactive entities may be similar to those shown in Fig. 2. That material remained in solution (soluble fraction) after the waste had been reacted with soil and the mixture centrifuged to remove particles greater than 0.05 μm nominal diameter. The lack of association of some radioactive material with waste debris is attributed to complexation of that material probably by an organic chelate. It is that fraction that will migrate through the soil provided the complex is not degraded.

The carbonate/alpha emitter association will be readily changed as the pH of the soil environment changes. The pH of typical soils ranges from about 6 to about 8; the pH of the waste was adjusted over a range from 6.3 to 8.6 and the soluble calcium determined. The relative concentrations of calcium increased from 5.3 at pH 8.6 to 14.3 at pH 6.3. Thus, as waste passes from an acid soil to a more nearly basic soil, carbonates may precipitate and remove some radionuclides as associates with the precipitate. In the reverse solubilization, alpha emitters are released and thus are free to react with soil by cation exchange and be immobilized or to react with chelate ligands and be free to move with the soil solution. Certainly other chemical and physical reactions can and do occur.

Changes in the hydrogen ion concentration in the waste are reflected in changes in the species of those radionuclides that are multivalent. The field stability diagram, Fig. 3, shows the relationships among pH, Eh, and plutonium species. It follows that the characteristics of the soil, which influence the characteristics of the soil solution, will determine the charge on the ion, in this case plutonium, and its degrees of sorption through cation, or anion, exchange reactions.

By employing data from field stability diagrams and the pertinent analytical data, the net charge distribution of a soluble constituent can be calculated from Eq. (1) below.

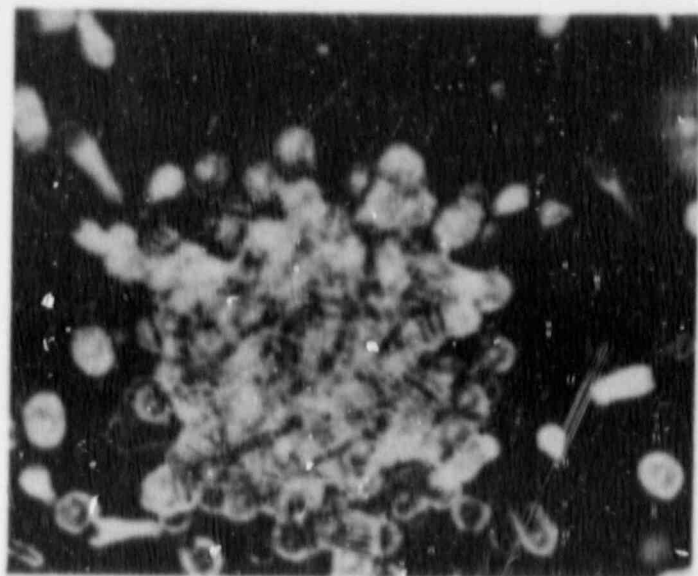


Figure 1. Alpha emitting material associated with crystalline material from the insoluble fraction (less than $0.05 \mu\text{m}$) of a waste. The insoluble fraction was shown to be more than 60% calcium carbonate by x-ray diffraction. The formation of such precipitates is in part responsible for the removal of plutonium in waste treatment plants and their formation, as waste percolates through soil, would inhibit migration of that radionuclide.



Figure 2. Autoradiograph of waste supernate after centrifugation to remove particles larger than $0.05 \mu\text{m}$. The tracks are caused by alpha emitters unattached to visible particles. The emitters are probably plutonium and are probably chelated, thus they do not sorb and remain in solution---designated soluble fraction.

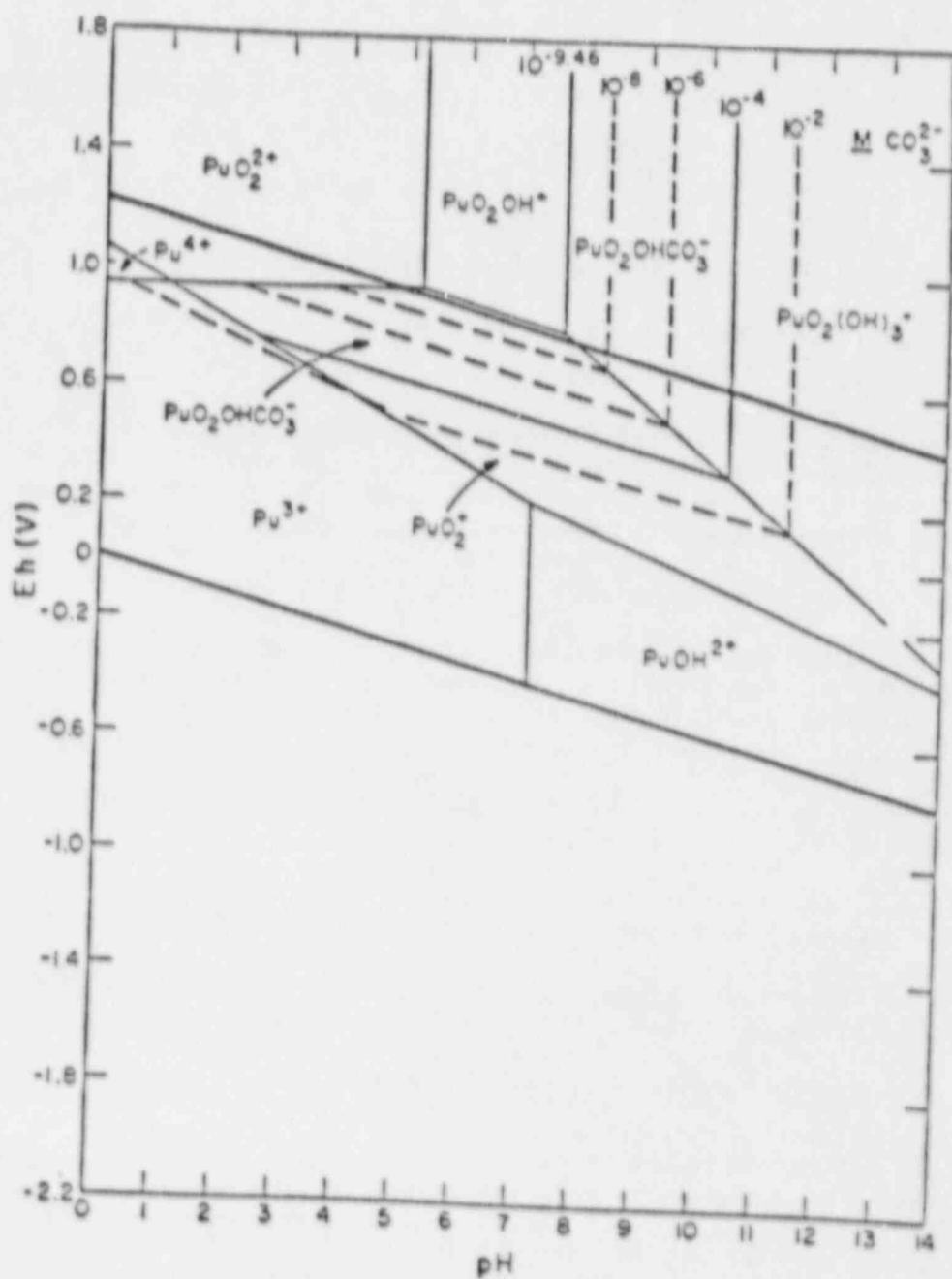


Figure 3. Field stability diagram relating the relationships among pH, Eh, and plutonium speciation. According to Nelson et al.(7) the Pu (III, IV) species most readily sorb to soil. Soils with a pH below 7 would be most effective in removal of plutonium by cation exchange.

$$\frac{[\text{PuO}_2\text{OHCO}_3^-]}{[\text{PuO}_2^+]} = 10^{18.2} [\text{OH}^-] [\text{CO}_3^{2-}] 10^{\left[\frac{-0.94 + \text{Eh}}{0.0592} \right]} \quad (1)$$

For the predominant species the charge distribution is expressed as the

numerical value of the ratio $\frac{[\text{PuO}_2\text{OHCO}_3^-]}{[\text{PuO}_2^+]}$, that value decreases as the concentration of PuO_2^+ increases relative to the concentration of $\text{PuO}_2\text{OHCO}_3^-$.

As the pH value of the soil decreases that relative value of PuO_2^+ concentration increases (Table III).

From Table III and the theory of cation exchange, Fuquay soil should show a high degree of sorption for plutonium. However, it has been pointed out (Table I) that plutonium was not removed from solution by Fuquay Ap soil, rather it was brought into solution from the insoluble fraction. That result, contrary to that expected from calculation, again suggests the presence of complexer or chelator, possibly of biological origin, in the Ap soil.

To determine whether the phenomenon could be related to different charge distributions in the deeper horizons, charge distribution was calculated for other soil solutions generated by each of four horizons; results are presented in Table IV. Soils from horizons B2t and C should exhibit a high degree of sorption; referring to Table I (Fuquay system) it will be seen that B2t and C do; overriding solubilization of the insoluble radionuclide is not a characteristic of the lower soil horizons.

Experiments reported in this section were designed to answer questions posed in the preceding section. The results may be summarized as follows:

1. autoradiographs of the insoluble fraction of waste demonstrated an association of alpha emitting material with crystals.
2. the insoluble fraction of waste was shown to be more than 60% CaCO_3 .
3. the calcium is not tightly bound in the precipitate but is readily released to the waste solution as the pH is lowered from 8.6 to 6.3. Those pH values are not unknown in soil; thus, as waste migrates from a more nearly acid to a more nearly basic soil, the calcium would precipitate and "occlude" alpha emitters. Such a mechanism could inhibit migration of certain radionuclides.
4. a theoretical calculation of the charge distribution for the species ^{238}Pu demonstrated an inverse relationship between the concentration $[\text{PuO}_2^+]$ and the pH of the soil solution, i.e., the more acidic the soil, the higher the preponderance of PuO_2^+ ions and the greater the enhancement of cation exchange. The above was confirmed when

TABLE III
 INFLUENCE OF FOUR DIFFERENT SOILS
 ON THE NET CHARGE OF SOLUBLE ^{238}Pu IN RAW WASTE

System	Charge ^a	
	Distribution	pH
Waste	$1.1 \pm 0.03 \times 10^{-1}$	8.5
Waste/Puye	$5.1 \pm 0.5 \times 10^{-3}$	7.1
Waste/Carjo	$4.8 \pm 0.5 \times 10^{-2}$	6.4
Waste/Fayette	$6 \pm 1 \times 10^{-4}$	6.6
Waste/Fuquay	$7 \pm 2 \times 10^{-4}$	5.5

$$^a \frac{[\text{PuO}_2\text{OHCO}_3^-]}{\text{PuO}_2^+} = 10^{18.2} [\text{OH}^-] [\text{CO}_3^{2-}] 10^{\left[\frac{-0.94 + \text{Eh}}{0.0592} \right]}$$

The ion PuO_2^+ would be sorbed to the negatively charged soil micelle by cation exchange. Cation exchange removals of plutonium by Fayette and Fuquay soils would be expected; the pH of those soils would enhance production of PuO_2^+ (see Fig. 5).

TABLE IV
 INFLUENCE OF FUQUAY SOIL HORIZONS ON THE CHARGE
 DISTRIBUTION OF SOLUBLE ²³⁸Pu IN RAW WASTE

System	Charge Distribution
Raw Waste	2.6±0.6
Raw Waste/Soil	
Horizon	
Ap	8±2 x 10 ⁻³
A ₂	1.9±0.3 x 10 ⁻²
B2t	3±1 x 10 ⁻³
C	1.7±0.3 x 10 ⁻³

Horizons Ap, B2t, and C exhibit comparable charge distributions for ²³⁸Pu. Solubilization of plutonium by Ap horizon solution is not a result of charge distribution.

degrees of sorption, as related to pH, for the C horizons of Fuquay (pH = 7.1) and Fayette (pH = 7.6) were compared. The C horizons were chosen for comparison because C was the best defined horizon, and

5. the Ap horizon of the Fuquay soil did not exhibit the degree of sorption to be expected from a theoretical calculation of charge distribution. The apparent anomaly was attributed to the possible presence in the Ap horizon of a chelate (biologically generated?). Such material would have an undesirable, positive effect on migration of certain radionuclides from the burial trench.

Thus, association of waste radionuclides (e.g., plutonium) with precipitates as well as cation exchange reactions are pH dependent and act to inhibit migration, albeit at different values. A first assumption would suggest that proper choice of soil and pH adjustment would solve some problems associated with shallow land burial; however, the results obtained using the Fuquay Ap soil demonstrate that under certain conditions complexed (chelated) radionuclides will neither associate nor exchange, and thus remain free to migrate with the soil solution.

D. Laboratory Studies on Maxey Flats Waste and Soil

About 1977 members of the Environmental Science Group at the Los Alamos National Laboratory designed experiments applicable to the Maxey Flats burial site to address the questions posed in section I.

In general, samples of soil and soil solution from various locations at the site were obtained by implanting porous cups at various depths and employing vacuum for the removal of the sample. The samples of soil and soil solution were analyzed for physical, chemical, and radiochemical entities using standard methods and in the case of radiochemical analyses, the methods developed for the analysis of soil and vegetation relative to the Palomares, Spain incident.⁸ Details relative to sampling of soil, and soil solution have been reported.^{6,5} The area surrounding trench 19-S was chosen as an initial study site. An experimental trench was later constructed for more nearly controlled studies. Location of the two experimental areas is shown in Fig. 4. Physical and chemical characteristics of the 19-S trench water are presented in Table V. The grid for the initial and second series of sampling holes is presented in Fig. 5. The holes for the first installation were hand-drilled, were of various depths, and were dictated in location and depth by rock debris or by inherent sandstone bedding. The sampling holes of the second series were drilled by drilling rig to predetermined depths.

E. Fate of Complexed Radionuclides

The early work of the Brookhaven National Laboratory conducted by Weiss and others at Maxey Flats centered around an inventory of radionuclides in the burial trenches. A related effort led to the identification of some exotic organic compounds and to the identification of some bacterial species that were found to be associated with trench wastes.¹⁰ Certain of the organic compounds were believed to act as chelates.

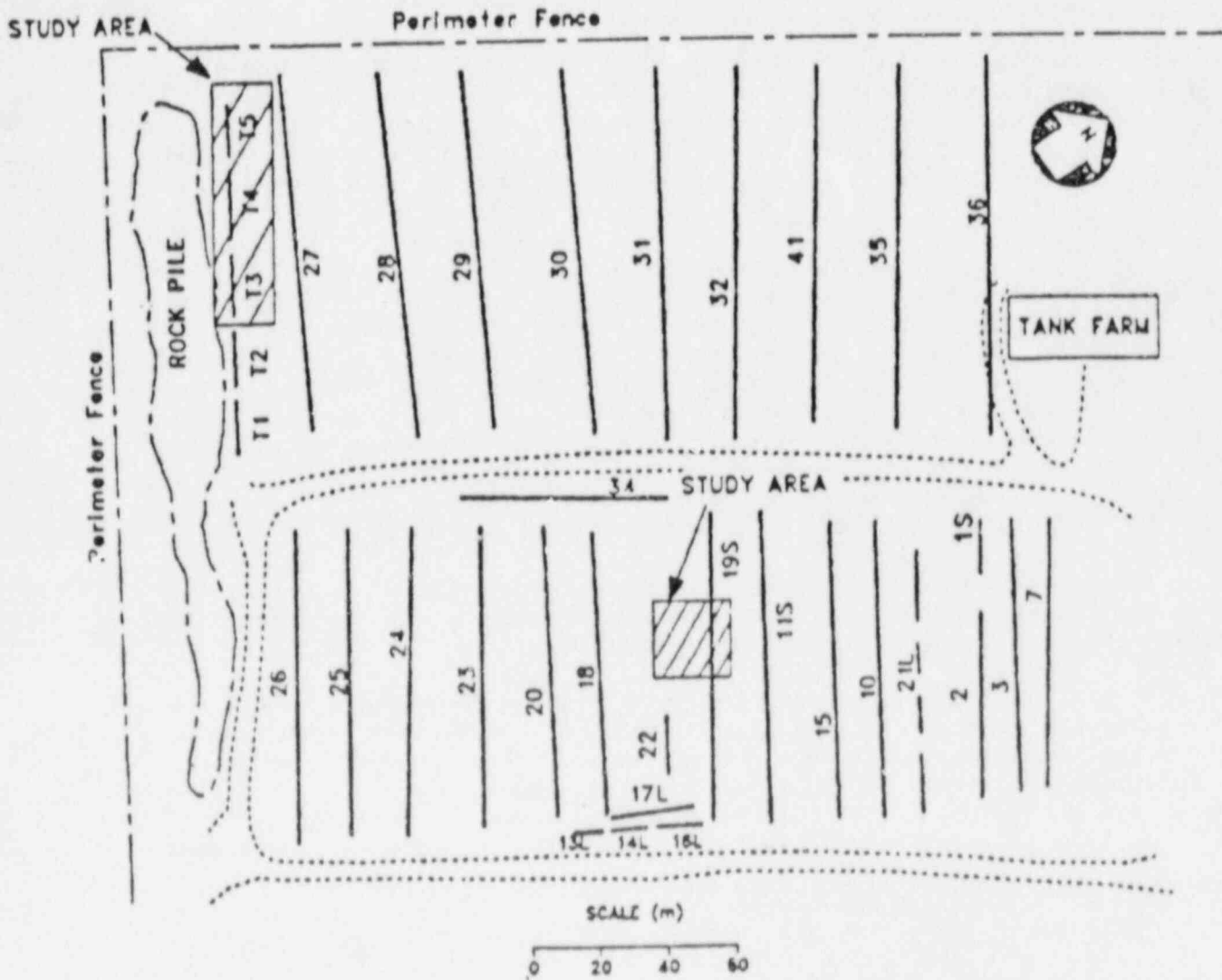


Figure 4. Location of two study areas at the Maxey Flats Shallow Land Burial Site. The area associated with Trench 19-S was established by the Los Alamos/University of California Berkeley team in 1978/1979. Samples of soil solution were obtained from implanted porous cups. The T series study area was established at a later date.

TABLE V
CHARACTERISTICS OF TRENCH 19-S SUMP WATER

pH	6.6
Temp., °C	21.0
Specific Cond. ($\mu\text{mhos/cm @ } 25^{\circ}\text{C}$)	2.34×10^3
Ca(ppm)	58.
Fe(ppm)	150.
Li(ppm)	0.23
Mg(ppm)	130.
Mn(ppm)	0.76
K(ppm)	25.
Na(ppm)	100.
Total Alkalinity(ppm as CaCO_3)	980.
HCO_3^- (ppm)	1200.
CO_3^{2-} (ppm)	0.
Cl(Ppm)	680.
^3H pCi/l	6.9×10^7
^{137}Cs pCi/l	3.24×10^3
^{60}Co pCi/l	1.32×10^3
^{90}Sr pCi/l	2.56×10^5
^{241}Am pCi/l	7.67×10^2
^{238}Pu pCi/l	1.72×10^5
$^{239,240}\text{Pu}$ pCi/l	2.1×10^4
$^{238}\text{Pu}/^{239}\text{Pu}$	8.2

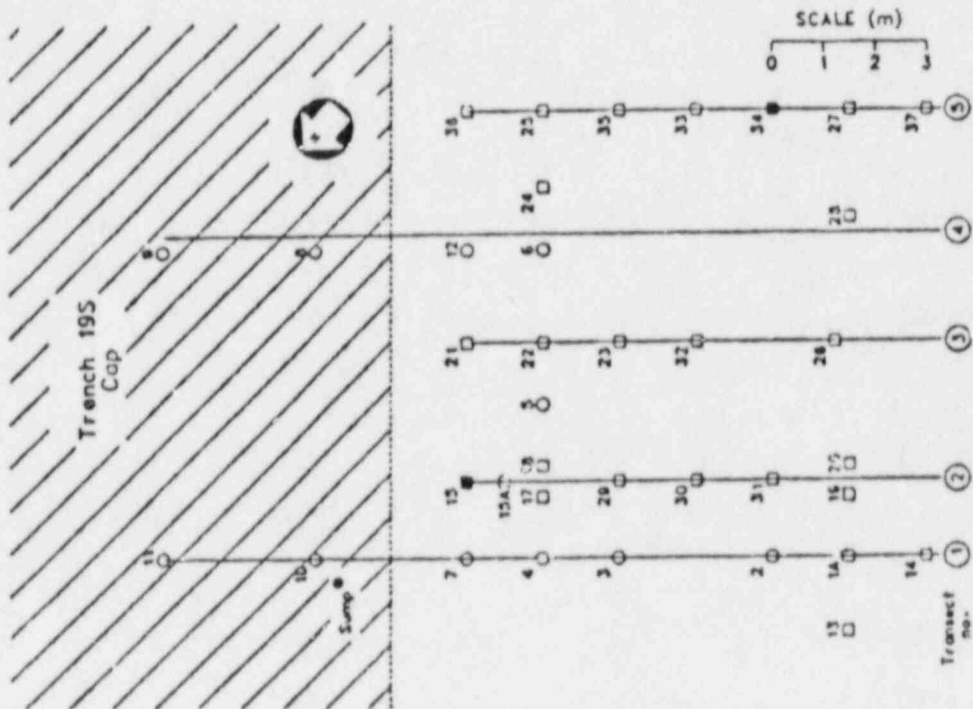


Figure 5. Plan view of the array of samplers at the 19-S area. The circles relate the location of the drill rig second series (1980). The black squares relate the location of holes that were aborted because high level activity was encountered; open squares relate to hand drilled holes. Hole 34 encountered an undocumented burial.

In an extensive report by Zehner² published 1983, the USGS efforts through 1979 are detailed. Data are presented relative to possible rates of flow through the soil, migration of some radionuclides, and the importance of fissures to flow patterns. However, the reader is left with continuing uncertainty relative to transport of radionuclides off site. The author deduces a minimum transport velocity of 50 ft/yr for cobalt-60 and manganese-54 by fissure flow. On the other hand, the final conclusion reached is that "Over-land runoff is the most likely means of transport . . ." (page 131, paragraph 5).

With respect to radiochemical analyses of samples from wells drilled by the USGS, the authors state "samples from most wells are of limited value in determining radiochemical quality of ground water at the burial site" (page 109, paragraph 4). The fact that specific isotopes (³H, ⁹⁰Sr, ⁶⁰Co, ²³⁹Pu, ²³⁸Pu) had been detected was not considered strong evidence for subsurface migration; several mechanisms are discussed other than subsurface migration, which could account for their presence.

In a 1975 report, Meyer³ reviewed again the possible pathways for migration of radionuclides to uncontrolled environment. The author points out ". . . there is insufficient radiological and hydrological information to estimate in what form, in what quantity, or at what rate Pu is migrating." Perhaps a more important conclusion related to the inability of containers and trenches in humid areas (of that time period) to retain radioactive waste. The paper has value as a presentation of earlier waste management practices.

A third study relative to the referenced time period was that of Montgomery et al.⁴ The report details an evaporator study, an environmental study and a test well study. Certain recommendations were made for future research, some of those recommendations may later have been considered. Conclusions related to the evaporator stack effluent as an important source of tritium at the environs. Further ". . . the transport of radionuclides from the site surface by precipitation run-off was a major source of the radioactivity detected in the drainage pathway" (page 73, paragraph 1). The radionuclides referenced were ⁵⁴Mn, ⁶⁰Co, ⁹⁰Sr, ¹³⁷Cs, ²³⁸Pu, and ²³⁹Pu.

Relative to the presence of radionuclides in test well samples (only ³H, ⁶⁰Co, and ⁹⁰Sr in the soluble fraction), the authors state that even though subsurface transport might explain the occurrence of radionuclides in the test wells, their presence off site appears to be primarily from surface water run-off and evaporator plume depletion. Thus, at the time of closure of the Maxey Flats site late in 1977, the following questions remained:

1. Does subsurface migration of radionuclides occur?
2. If there is migration, what migrates and in what quantity and what direction?

3. If certain radionuclides do not migrate, what is the inhibitory mechanism?
4. What soil chemistry retards or enhances migration?

The remainder of this report reviews the research after 1978, which addressed the above questions.

III. STUDIES BEYOND 1978

A. Laboratory Studies - Sorption Related to Soil Type

An initial assignment from the Nuclear Regulatory Commission (NRC) to the Los Alamos National Laboratory related to a study of different soil types with respect to their chemical and physical characteristics and their ability to sorb different radionuclides commonly found in the leachate obtained at waste burial sites. Those results are reported here to introduce the research approach employed later throughout the Maxey Flats study.

Investigations concomitant with the field studies at Maxey Flats were conducted in the laboratory on a variety of soils collected within the continental United States. Soils had been collected in 1/2 to 1 ton amounts each from the A, B, and C horizons of the respective soil profiles. The soils collected and their characteristics have been reported.⁵

B. Preliminary Sorption Experiments

Data relative to the differential sorption of various radionuclides from waste by two different soils and by different horizons of the same soil are presented in Table I. Results are presented for isotopes of plutonium, americium, and uranium with respect to their sorption by Fayette soil and Fuquay soil. Comparisons among the horizons as well as between the soil types can be made. The two soils differ in degrees of sorption for all radionuclides as specific horizon is compared with respective horizon. Further, for a specific radionuclide and a specific soil, the degree of sorption varies among horizons. Those observations are neither surprising nor earth shaking; they provide documentation.

The degree of sorption of a waste radionuclide by the soil as waste solution percolates from the burial pit into soil environment influences the extent of migration. The chemical and physicochemical nature of the soil solution (containing waste) as well as the nature of the soil (or soil horizon) determine the degree of sorption. Relative data obtained by analyses of waste, soil and soil/waste solution for the two soils are presented in Table II. A definite relationship between degrees of sorption (Table I) and a chemical entity or combination of entities (Table II) was not evident for the two soils under study. The pH value of 8.01 (A2 horizon) of the waste/Fuquay soil system (Table I) is probably the result of elutriation from the Ap horizon as the soil has developed. The pH of the Ap, B2t and C horizon soil/waste solutions decreases from 7.55 to 7.26 to 7.12 respectively. The soils are acid (pH = 6.8); the solution is slightly basic due to the addition

TABLE I
RADIONUCLIDE CONCENTRATIONS IN TWO RAW WASTES AND DEGREES OF SORPTION IN WASTE/SOIL SYSTEMS

System	^{238}Pu	$^{239,240}\text{Pu}$	^{241}Am	^{238}U	^{235}U	$^{233,234}\text{U}$	pH
<u>Fayette Soi</u>							
<u>Raw Waste (dpm/ml x 10²)</u>							
Total	11300±400	98±3	113±5	9±1	4.1±0.1	20±1	
Soluble	30±10	3±1	3±1	10.7±0.5	4.4±0.4	21.8±0.5	
<u>Degrees of Sorption (%)</u>							
<u>Raw Waste/Fayette Soil</u>							
Ap	83.3	80.0	85.0	72.9	80.9	71.1	8.03
B	88.0	88.3	83.3	96.3	95.5	90.8	7.20
C	87.7	86.7	86.7	92.5	96.6	86.2	7.60
<u>Fuquay Soil</u>							
<u>Raw Waste (dpm/ml x 10²)</u>							
Total	11300±400	98±3	113±5	9±1	4±1	20±1	
Soluble	21±3	2.0±0.4	1.8±0.4	10.4±0.1	3.7±0.1	22.5±0.4	
<u>Degrees of Sorption (%)</u>							
<u>Raw Waste/Fuquay Soil</u>							
Ap	-47.6	-40.0	38.9	95.7	99.2	97.8	7.55
A2	-33.3	-15.0	-16.7	62.5	64.7	64.4	8.01
B2y	98.6	97.0	90.0	99.2	99.7	99.1	7.26
C	99.0	91.0	79.4	99.4	99.5	99.9	7.12

The degree of sorption is the ratio between the concentration in the raw waste/soil system (i.e., soluble fraction) divided by the concentration in the soluble fraction of raw waste for the respective radionuclide times 100. In the case of Raw waste/Fuquay soil, the five negative numbers indicate negative sorption, i.e., solubilization from the respective insoluble fractions in the raw waste.

TABLE II
CHEMICAL AND PHYSICAL PROPERTIES OF RAW WASTE AND RAW WASTE/SOIL SYSTEMS

System	EC mmhos	Na mx10 ²	Ca mx10 ⁴	CO ₃ ⁻ mx10 ⁴	HCO ₃ ⁻ mx10 ³	Al mx10 ⁶	Fe mx10 ⁶	COD mx10 ³	PO ₄ ⁼ mx10 ⁶	pH	Eh mV
<u>Fayette Soil</u>											
Raw Waste											
total	1.12	1.4	17.5	8.7	9.0	40.4	272	6.18	370	8.71	384
Soluble	1.07	1.3	0.21	5.9	7.2	3.7	2	1.56	10.7	8.74	399
Raw Waste/Fayette Soil											
Ap	1.03	1.12	11.9	0.52	8.5	3.7	2	4.8	10.9	8.03	416
B	0.72	0.80	4.4	0.01	3.7	3.7	1	2.2	5.2	7.20	411
C	0.83	0.93	7.9	0.14	5.8	3.7	0.5	2.2	8.8	7.60	412
<u>Fuquay Soil</u>											
Raw Waste											
Total	1.12	1.38	17.5	8.7	9.0	40.4	273	6.18	370	8.71	8.84
Soluble	1.18	1.29	5.3	9.9	7.8	3.7	3.8	1.6	9.8	8.57	3.97
Raw Waste/Fuquay Soil											
Ap	0.97	1.2	1.7	0.16	6.78	10.0	6.8	4.7	9.5	7.55	411
A2	1.08	1.24	4.7	0.51	8.03	14.8	10.0	3.8	8.0	8.01	379
B2†	0.94	0.95	0.75	0.04	3.47	3.7	2.0	0.97	0.28	7.26	432
C	0.84	0.95	0.31	0.03	3.40	3.7	2.0	0.98	0.4	7.12	440

A correlation between any of the properties and the degree of sorption (Table II) is not apparent. An exception may be the inverse relationship between pH and degree of sorption for uranium and americium by the Fuquay A2 horizon soil and for uranium by the Fayette Ap horizon soil.

(4:1) of waste (pH 8.7) to the soil in the batch method employed in the experiment. Again in the case of the Fuquay system (Table I) and for americium and uranium species, the lowest degree of sorption was noted for the A2 horizon where the pH was highest (8.01). For the Ap, B2t, and C horizons, degrees of sorption increased with a decrease in pH. However, for the two plutonium species, ^{238}Pu and ^{239}Pu , ($^{239,240}\text{Pu}$ are considered together), the lowest degree of sorption was noted for the Ap horizon. In point of fact, the concentration of the two species in the supernate after reaction of waste with Ap horizon soil was greater than their respective concentrations in the soluble fraction of the waste per se; results for plutonium species and americium species in the A2 horizon were similar. The conclusion was reached that the Ap and A2 horizons contain in their soluble organic fraction a chelate or complexing entity that solubilizes plutonium from the insoluble fraction of the waste and that the resulting complex is not sorbable. In the strict sense of ion exchange the complex possesses the wrong charge or no charge.

In the case of Fuquay Ap horizon soil, the degrees of sorption for ^{238}Pu and $^{239,240}\text{Pu}$ were negative, i.e., plutonium was brought into solution from the insoluble fraction. On the other hand, the degrees of sorption for those species by the Fayette Ap horizon soil were positive. The difference is attributed to the presence of chelates (Fuquay) as opposed to their absence or, if present, in lesser amounts (Fayette). Other experiments have produced similar results.

The results from the initial investigations can be summarized as:

1. soils differ markedly with respect to their degree of sorption of different waste radionuclides,
2. the horizons within a specific soil differ with respect to their degree of sorption of different waste radionuclides,
3. in general, the degree of sorption increases with the depth of the profile,
4. some soils, notably the Ap horizon of the South Carolina Fuquay soil, possess entities that solubilize plutonium from the insoluble fraction of the waste,
5. the solubilized plutonium (4 above) is apparently converted to a form (a chelated form has been suggested), which does not sorb to the soil,
6. a relationship between degree of sorbability and chemical constituents of the soil/waste solution is not readily apparent, and,
7. in the case of the Fuquay soil, the pH of the solution and thus the charge on the respective radionuclides is related to the degree of sorption by the soil.

Item 7 above dictated the following experimental approach, i.e., determine the charge characteristics of the various radionuclides in the waste in the absence and presence of soils and relate the charge characteristics to the degree of sorption.

C. Electrical Charge Relationships

Two possible modes of removal of radionuclides as related to the hydrogen ion concentration of the solution involve precipitation (or coprecipitation) and ion exchange. In the majority of soils, cation exchange will be predominant.

The physical nature of the insoluble fraction of radioactive waste was investigated using autoradiographic techniques.⁶ The association of alpha emitting material with a crystalline precipitate is shown in Fig. 1. The crystalline material was shown by X-ray diffraction to be more than 60% calcium carbonate.

The unassociated radioactive entities may be similar to those shown in Fig. 2. That material remained in solution (soluble fraction) after the waste had been reacted with soil and the mixture centrifuged to remove particles greater than 0.05 μm nominal diameter. The lack of association of some radioactive material with waste debris is attributed to complexation of that material probably by an organic chelate. It is that fraction that will migrate through the soil provided the complex is not degraded.

The carbonate/alpha emitter association will be readily changed as the pH of the soil environment changes. The pH of typical soils ranges from about 6 to about 8; the pH of the waste was adjusted over a range from 6.3 to 8.6 and the soluble calcium determined. The relative concentrations of calcium increased from 5.3 at pH 8.6 to 14.3 at pH 6.3. Thus, as waste passes from an acid soil to a more nearly basic soil, carbonates may precipitate and remove some radionuclides as associates with the precipitate. In the reverse solubilization, alpha emitters are released and thus are free to react with soil by cation exchange and be immobilized or to react with chelate ligands and be free to move with the soil solution. Certainly other chemical and physical reactions can and do occur.

Changes in the hydrogen ion concentration in the waste are reflected in changes in the species of those radionuclides that are multivalent. The field stability diagram, Fig. 3, shows the relationships among pH, Eh, and plutonium species. It follows that the characteristics of the soil, which influence the characteristics of the soil solution, will determine the charge on the ion, in this case plutonium, and its degrees of sorption through cation, or anion, exchange reactions.

By employing data from field stability diagrams and the pertinent analytical data, the net charge distribution of a soluble constituent can be calculated from Eq. (1) below.

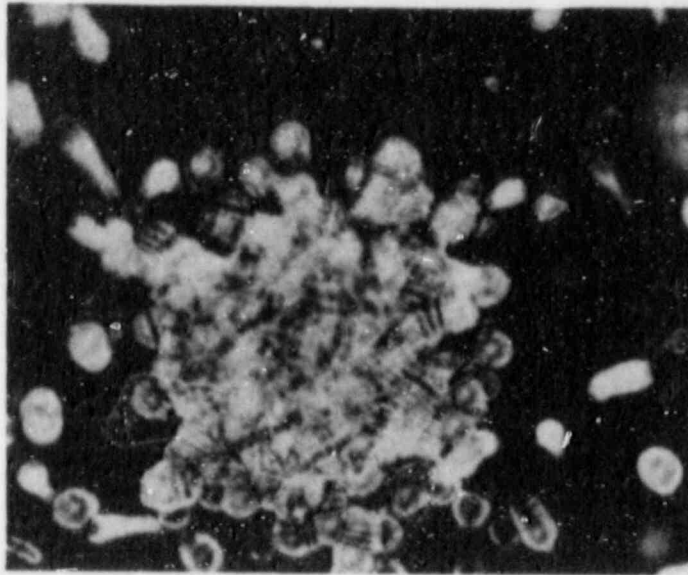


Figure 1. Alpha emitting material associated with crystalline material from the insoluble fraction (less than $0.05 \mu\text{m}$) of a waste. The insoluble fraction was shown to be more than 60% calcium carbonate by x-ray diffraction. The formation of such precipitates is in part responsible for the removal of plutonium in waste treatment plants and their formation, as waste percolates through soil, would inhibit migration of that radionuclide.



Figure 2. Autoradiograph of waste supernate after centrifugation to remove particles larger than $0.05 \mu\text{m}$. The tracks are caused by alpha emitters unattached to visible particles. The emitters are probably plutonium and are probably chelated, thus they do not sorb and remain in solution---designated soluble fraction.

TABLE X
 SELECTED CHEMICAL AND PHYSICAL CHARACTERISTICS OF SPIKED
 MAXEY FLATS TRENCH 19-S WASTE-TILSIT SOIL SYSTEMS

	<u>Tilsit Ap Soil</u>	<u>Tilsit C Soil</u>
pH	6.71±0.02	6.11±0.02
Eh (mV)	492±5	430±16
EC (dS/m)	1.51±0.01	0.86±0.01
Total Alkalinity as CaCO ₃ (ppm)	3.78±1	86±7
DOC (ppm)	475±9	400±10
Ca (ppm)	33±1	1.3±0.3
Mg (ppm)	110±20	7±1
Fe (moles/l)	(1.1±0.4)×10 ⁻⁵	(4.8±0.3)×10 ⁻⁶
% Cationic	3: 9	47±8
% Anionic	43: 10	35±7
% Neutral	20±	19±2
Pu (moles/l)	(2.1±0.4)×10 ⁻⁹	(3.1±1.5)×10 ⁻¹⁰
% Cationic	3±2	31±11
% Anionic	71±18	58±24
% Neutral	28±17	12±7

Analytical measurements were performed on the centrifuged solution to define the characteristics of the soluble fraction (less than 0.05 μm diameter) of the waste. The charge characteristics of plutonium and iron were determined using cation, anion, and mixed-bed exchange resins. The waste was spiked at a level of 2.1×10⁻⁶ moles.

TABLE XI
IN SITU CHEMICAL AND PHYSICAL CHARACTERISTICS OF TRENCH
 WASTE AND SOIL SOLUTIONS FROM THE TRENCH 19-S
 STUDY AREA OF THE MAXEY FLATS BURIAL FACILITY

	Trench Waste	Soil Solutions		
		Approximate Distance From Trench (m)		
		1.5	3.0	6.0
^3H (pCi/l)	7.8×10^7	2.3×10^7	7.3×10^6	2.0×10^4
Waste Dilution Factor	--	3.0	10.8	4.1×10^3
pH	7.25	6.80	6.30	5.60
Eh (mV)	-78	72	330	240
EC (dS/m)	1.55	1.41	3.1	3.3
DOC (ppm)	106	57	15	18
Alkalinity as CaCO_3 (ppm)	585	575	278	78
Fe (moles/l)	6.45×10^{-4}	2.54×10^{-4}	2.33×10^{-6}	5.39×10^{-5}
% Cationic	67	68	22	81
% Anionic	29	28	26	7
% Neutral	4	4	52	12
Pu (mole/l)	1.53×10^{-11}	4.75×10^{-13}	$< 6.14 \times 10^{-14}$	$< 6.14 \times 10^{-14}$
			$\geq 7.6 \times 10^{-10}$	$\geq 2.2 \times 10^{-15}$
% Cationic	10	2	70	0
% Anionic	89	97	30	100
% Neutral	1	1	0	0

On-site field measurements of pH, Eh, alkalinity, and EC were made at the Trench 19-S study area. Also, aliquots of samples were passed through cation, anion, and mixed-bed ion exchange resins at that time. Analyses of plutonium, iron, and DOC were performed in the laboratory on the above samples; plutonium and iron were also performed on the aliquots passed through ion exchange resins.

plutonium.¹² Therefore, distribution coefficients based on the concentration of reduced inorganic species in solution should provide more meaningful information in interpreting the mobility of plutonium even though that concentration is much less than the oxidized inorganic species or the reduced organic complexed species of plutonium; hence, thermodynamic relations were used to determine concentrations of the reduced inorganic species. Also, the concentrations of the PuEDTA species in solution are controlled by the relative concentrations of inorganic plutonium species and by the concentration of noncomplexed EDTA ligand. The thermodynamic approach provided a means of evaluating those relative concentrations.

The basic assumptions used in the thermodynamic considerations include the following:

1. The two metal constituents of importance in controlling the concentration of PuEDTA are plutonium and iron. Iron competes with plutonium for the EDTA ligand. Therefore, the concentration of the PuEDTA species is dependent not only on the concentration of inorganic plutonium species but also on the concentration of inorganic iron species.
2. The inorganic iron species are controlled by a specific iron mineral. Ferrosic oxide $[\text{Fe}(\text{OH})_8]$ is probably that mineral under conditions of reduction-oxidation cycles or under conditions of partial oxidation when highly reduced systems are exposed to more oxidized systems. The presence of silica or organic anions may inhibit the transformation of the metastable $\text{Fe}_3(\text{OH})_8$ to more crystalline, less soluble oxides.¹⁰ However, soil iron hydroxide $[\text{Fe}(\text{OH})_3]$ and magnetite (Fe_3O_4) are much less soluble and should eventually precipitate, causing $\text{Fe}_3(\text{OH})_8$ to dissolve. Magnetite is more stable at $\log (\text{Fe}^{2+}) + (2 \times \text{pH})$ greater than 4 and $\text{Fe}(\text{OH})_3$ (soil) is more stable at $(\text{Fe}^{2+}) + (2 \times \text{pH})$ less than about 4. Goethite ($\alpha\text{-FeOOH}$) and lepidocrocite ($\gamma\text{-FeOOH}$) crystallize rapidly at pH less than 6.¹⁰
3. Quantitative values of Eh can best be estimated from the concentrations of iron species in solution and from the stability of the iron solid controlling those concentrations.¹⁴ Eh measurements using a platinum electrode are of value in showing trends only.
4. For short term equilibration times, as in the case of laboratory experiments, the Eh for the plutonium system may be different from that for the iron system.¹⁴ In such instances, two Eh values must be evaluated.

Variable parameters are soluble plutonium, soluble EDTA, Eh, pH, and EC. The various plutonium species can be expressed relative to one another in terms of pH, Eh, noncomplexed EDTA concentration, and stability constants; the

concentration of soluble plutonium permits the calculation of absolute concentrations of the various plutonium species. In a similar manner, the concentration of soluble EDTA permits the calculation of absolute concentration of the various EDTA species. Electrical conductivity is used to estimate ionic strength,¹⁵ which in turn is used to estimate the activity coefficients of specific species; activity coefficients relate thermodynamic concentrations to measured concentrations. The derivations may be expressed in a number of ways, depending on the information required.

As outlined above, in the first of the two laboratory experiments, three aliquots of anoxic Trench 19-S waste were spiked with 2.1×10^{-6} moles/l plutonium and then allowed to equilibrate under oxic laboratory conditions: waste 1 was equilibrated for approximately 1 hour, waste 2 for 5 days, and waste 3 for 6 days. At the end of each equilibration period a waste aliquot was centrifuged and the soluble characteristics of the waste determined (Table IX). In each case a substantial amount of plutonium had been removed by centrifugation indicating precipitation of plutonium during equilibration under oxic laboratory conditions. An ion activity product (IAP) for the precipitate, $\text{Pu}(\text{OH})_4$, was evaluated and compared with the K_{sp} of $\text{Pu}(\text{OH})_4$ found in the literature. The IAP $\text{Pu}(\text{OH})_4$ is equal to $(\text{Pu}^{4+})(\text{OH}^-)_4$ and is the same as the K_{sp} $\text{Pu}(\text{OH})_4$ in the presence of the precipitate under equilibrium conditions. The OH^- concentration was determined from the measurement of the pH, and the Pu^{4+} concentration was calculated from estimated chemical parameters derived from analyses given in Table IX and from thermodynamic relations. The estimated chemical parameters and the calculated IAP $\text{Pu}(\text{OH})_4$ are given in Table XII. A comparison of the IAP $\text{Pu}(\text{OH})_4$ with the K_{sp} $\text{Pu}(\text{OH})_4$ indicates good agreement for 2 of the 3 waste aliquots (waste 2 and waste 3). In those two systems, the IAP $\text{Pu}(\text{OH})_4$ was within a factor of 3 of the K_{sp} $\text{Pu}(\text{OH})_4$ ¹¹ in the third system (waste 1), they differed by an order of magnitude. That difference is not excessive and is attributed to a general phenomenon usually observed during precipitation of solids; initially the solubility of the precipitate is usually less than that observed under equilibrium conditions. Waste 1 was equilibrated for only about 1 hour before the soluble characteristics were determined. The above results suggest that the use of thermodynamic relations in estimating concentrations of analytically nondetectable species of plutonium yields relatively good estimates of such concentrations.

In the second laboratory experiment, aliquots of the spiked waste were added to the Ap and C soils of the Tilsit series and equilibrated for 5 and 6 days, respectively. After equilibration, the systems were centrifuged and the characteristics of the soluble fraction determined (Table X). In the case of the spiked-waste/soil systems, the concentration of soluble plutonium was considerably less than that in the presence of the $\text{Pu}(\text{OH})_4$ precipitate; the probable mechanism of plutonium removal from solution is by ion exchange. According to Nelson et al.,¹² the adsorption of inorganic $\text{Pu}(\text{III,IV})$ exceeds the adsorption of inorganic species of oxidized plutonium [$\text{Pu}(\text{V,VI})$]. For

TABLE XII
ESTIMATED CHEMICAL PARAMETERS OF SPIKED MAXEY FLATS
TRENCH 19-S WASTE EQUILIBRATED UNDER OXIC LABORATORY CONDITIONS

	Waste 1	Waste 2	Waste 3
PuEDTA	3.4×10^{-9}	1.4×10^{-8}	2.1×10^{-8}
Inorganic Pu	1.1×10^{-10}	6.0×10^{-10}	6.6×10^{-10}
FeEDTA	1.3×10^{-5}	1.2×10^{-5}	1.4×10^{-5}
Inorganic Fe	3.9×10^{-6}	5.1×10^{-6}	6.0×10^{-6}
[Fe ²⁺]	4.1×10^{-7}	4.3×10^{-7}	3.4×10^{-7}
Eh Fe(mV)	77	44	4
[Fe ⁺]	7.4×10^{-19}	2.2×10^{-19}	3.5×10^{-20}
[FeEDTA ⁻]	2.5×10^{-6}	9.5×10^{-7}	1.6×10^{-7}
[PuEDTA ⁻]/[Pu ³⁺]	4.3×10^5	5.5×10^5	5.8×10^5
Eh Pu(mV)	290	290	260
[Pu ⁴⁺]	5.4×10^{-26}	2.0×10^{-25}	5.2×10^{-26}
IAP Pu(OH) ₄	7.8×10^{-52}	9.8×10^{-51}	1.7×10^{-50}
K _{sp} Pu(OH) ₄	6.3×10^{-51}	6.3×10^{-51}	6.3×10^{-51}

The chemical parameters were estimated from analyses given in Table IX and from thermodynamic relations and were used to calculate the IAP_{Pu(OH)₄}. The differences in the three wastes are attributed to differences in time of equilibration: waste 1 was equilibrated for approximately one hour, waste 2 for five days and waste 3 for six days. Those wastes had been spiked with 2.1×10^{-6} moles/l plutonium before equilibration.

example, the K_d values of inorganic Pu(III,IV) are about 10^6 ml/g for a wide range of environmental waters; the K_d values of inorganic Pu(V,VI) are less than 10^3 for similar systems. The K_d of Pu(III,IV) will be lower in the presence of organic complexers because the organically complexed species of plutonium do not sorb. Under those conditions, as in the case of the Trench 19-S waste, the inorganic Pu(III,IV) usually cannot be analytically measured because the concentration of the organic Pu(III,IV) greatly exceeds that of the inorganic Pu(III,IV); the two species are not differentiated in the analyses. However, inorganic Pu(III,IV) concentrations can be evaluated through thermodynamic analyses.

In the laboratory spiked-waste/soil experiment, the concentration of the inorganic Pu(III,IV) was estimated from the analytical measurements given in Table X and from thermodynamic relations. The K_d values for the inorganic Pu(III,IV) were calculated and compared with those quoted by Nelson et al.,¹² and with those calculated from field experiments at the Maxey Flats burial site. Table XIII lists the chemical parameters estimated from analytical measurements and from thermodynamic relations. The PuEDTA concentration is estimated from the anionic and neutral fractions of measured plutonium; the FeEDTA concentration is estimated from the cationic and neutral fractions of measured iron. Also, the inorganic concentration of plutonium is estimated from the cationic fraction of measured plutonium and the inorganic concentration of iron is estimated from the cationic and neutral fractions of measured iron. Other parameters are based on the above estimated organic and inorganic concentrations and thermodynamic relations. The concentration of adsorbed plutonium is based on the concentration of plutonium added to the system and the concentration of measured soluble plutonium.

The results of the above calculations indicate a K_d for inorganic Pu(III,IV) of 3×10^6 for the Ap soil and 6×10^4 for the C soil when each was interacted with the spiked Trench 19-S waste. The K_d for the Ap soil compares favorably with those previously quoted by Nelson et al.¹² The K_d for the C soil is about an order of magnitude lower. Even though the K_d for the C soil is somewhat lower than for other quoted K_d values, it does not appear to be unrealistic. The C soil should be comparable to the deeper soil at the Trench 19-S study area at the Maxey Flats site. Distribution coefficients for the inorganic Pu(III,IV) are evaluated for the field experiment in the next section. Similar K_d values calculated under different conditions, as in field experiments, will lend support to thermodynamic evaluations.

In the field experiment, porous ceramic cup samplers were implanted in the soil near Trench 19-S in a horizontal and vertical array. Soil solutions were collected through a porous cup sampler and analyzed in a similar manner. Analytical measurements of waste and soil characteristics (Table XI) and thermodynamic relations were used to calculate the inorganic Pu(III,IV). The soils collected during the implantation of the porous cup samplers were also analyzed for plutonium. Based on those analyses and the calculated inorganic

TABLE XIII
ESTIMATED CHEMICAL PARAMETERS OF SPIKED MAXEY FLATS
TRENCH 19-S WASTE-TILSIT SOIL SYSTEMS

	<u>Tilsit Ap</u>	<u>Tilsit C</u>
PuEDTA	$(2.1 \pm 0.5) \times 10^{-9}$	$(2.2 \pm 1.1) \times 10^{-10}$
Inorganic Pu	$(6.3 \pm 2.1) \times 10^{-11}$	$(9.6 \pm 4.8) \times 10^{-11}$
FeEDTA	6.9×10^{-6}	2.59×10^{-6}
Inorganic Fe	4.0×10^{-6}	2.26×10^{-6}
[Fe ²⁺]	1.4×10^{-6}	1.3×10^{-6}
[e ⁻] (V)	1.6×10^{-4}	5.1×10^{-7}
Eh Fe(V)	0.23	0.37
[Fe ³⁺]	8.3×10^{-16}	2.2×10^{-13}
[FeEDTA ⁻]	5.5×10^{-6}	2.2×10^{-6}
[PuEDTA ⁻]/[Pu ³⁺]	8.3×10^2	1.3
Eh Pu (V)	0.31	0.33
[Pu ⁴⁺]	4.6×10^{-23}	6.0×10^{-21}
[Pu ³⁺]	2.0×10^{-12}	1.2×10^{-10}
[PuOH ²⁺]	8.1×10^{-13}	1.3×10^{-11}
[PuO ₂ ⁺]	6.5×10^{-11}	7.2×10^{-11}
Inorganic Pu (calculated)	6.8×10^{-11}	2.1×10^{-10}
PuEDTA (calculated)		
% Anionic	90	80
% Neutral	10	20
Pu sorbed (moles/g)	8.56×10^{-9}	8.56×10^{-9}
K _d Pu(III,IV) (ml/g)	3.1×10^6	6.1×10^4

Concentrations expressed as moles/l unless noted.

The chemical parameters were estimated from analyses given in Table X and from thermodynamic relations and were used to calculate K_d Pu(III,IV). The

Waste-Tilsit Ap soil system was equilibrated for five days and the Waste-Tilsit C soil system for six days. The waste had been spiked with 2.1×10^{-6} moles/l plutonium before being mixed with the soil.

Pu(III,IV) concentrations, K_d values were calculated for each of the three soil locations. Those results are given in Table XIV; the iron minerals that were assumed to control the inorganic concentration of iron are also given in Table XIV. Two values of plutonium concentration (less than and greater than or equal to) are given for the two locations farthest from the trench. Both ^{238}Pu and $^{239,240}\text{Pu}$ were present in the waste and soil solutions. However, at those two locations, the concentration of $^{239,240}\text{Pu}$ was below analytical detection; the concentration of ^{238}Pu was analytically identified and measured. Thus, the less than value represents the detection limit for $^{239,240}\text{Pu}$ and the greater than or equal to concentration represents the measured concentration of ^{238}Pu . Similarly, greater than and less than or equal to values are presented for the calculated K_d values at those two locations for the same reasons.

The calculated K_d values for the three soil locations appear to be comparable. The K_d for the soil location 1.5 m from the trench may be in error because at the time the porous cup sampler was implanted, the soil was collected before an apparent fracture was breached. As a result, the soil solution collected by the porous cup may contain more plutonium than the solution in contact with the soil before the soil sample was collected. If in error, the actual K_d is probably greater than that calculated. The K_d values in Table XIV are also comparable to the K_d values calculated for the Tilsit C soil in the laboratory experiment. The similarities in those calculated K_d values suggest that the method of evaluation of inorganic Pu(III,IV) is realistic.

The results of the evaluation of an IAP for $\text{Pu}(\text{OH})_4$ in the waste system and the evaluation of K_d values for inorganic Pu(III,IV) sorbed to soil suggest that the use of thermodynamic relations for modeling the influence of soil chemistry on the mobility of PuEDTA is realistic and feasible. Thus, the thermodynamic relations and the analytically determined characteristics of the waste and soil solutions are used to evaluate the differences in the solution chemistry between the waste in the trench and the solution in the nearby soil that affect the mobility of PuEDTA. Chemical parameters estimated for that evaluation are given in Table XV. With the exception of the oxidized species of inorganic plutonium and iron, concentrations are lower in the soil solutions than in the waste. That reduction, in part, may be attributed to the dilution of the waste with soil water as the waste moves into the nearby environment. Thus, in order to compare data from the trench waste with those in the soil solution, the concentrations of interest are normalized with respect to EDTA, FeEDTA, or PuEDTA. Those results are given in Table XVI.

The concentration of PuEDTA relative to the total EDTA concentration is less in the soil solutions than in the trench waste and that difference indicates that factors other than waste dilution by soil water play an important role in reducing the concentration of the plutonium complex (see Subsection E). The concentration of inorganic Fe(II) in the soil solutions tends to be about the same or slightly higher than that in the trench waste;

J. Radionuclides Off Site

Based on the physical nature of tritium and the supposed underground drainage gradient, northwest to southeast, the concentrations of tritium and certain other radionuclides were determined on soil and soil solutions outside the fence perimeter, but on site. Nine, 8.3 cm diameter, holes were hand augered at distances of 5 to 10 meters outside the project fence line. The holes were drilled to various depths as dictated by rocks; maximum depth reached was about 110 cm. Location of the holes, A through I is shown in Fig. 11. The analytical results for ^3H , ^{137}Cs , and ^{60}Co in the bore-hole soils are presented in Table XX.

Hole E is the only location where the three radionuclides were found at both depths sampled. Hole E is at the exit of the east main drain. The concentration of each of the three radionuclides decreases with depth suggesting surface contamination carried by run-off.

Analytical results obtained from Hole H soils showed ^{137}Cs (0-20 cm depth), ^{60}Co (0-36 cm depth) and the highest levels of soil tritium (0-36 cm depth). Hole H is less than 30 meters from well 11E. Water from that well contained tritium concentrations several orders of magnitude above those in waters from the other E series wells (Table XXI).

Trench #31 is in close proximity to both Hole H and well 11E. One is tempted to conclude that trench #31 is the source of tritium in Hole H and in well 11E. If the evaporator plume were the primary source, well 12E tritium content should be comparable to that in well 11E. It is not. Well 12E water had a tritium concentration about 300 times lower than that at well 11E. The results presented indicate a possible connection between well 11E and a source such as trench #31.

The elevated levels found at Hole E probably relate to surface run-off along the main east drain. In no case was evidence obtained for extensive underground migration of the radionuclides studied, including tritium.

Rickard et al.¹ studied the accumulation of radionuclides in forest litter and new leaves of trees in the vicinity of Hole H and well 11E. The least tentative of their conclusions states ". . . hickory leaves showed elevated levels of ^{60}Co indicating that the source of ^{60}Co contributing to the tree leaves is probably through local contamination of the rooting substrate . . ."

K. Experimental Trenches

The attempts to characterize the underground hydrology at the Maxey Flats site began with the S hole traverse in 1963. The E series holes were drilled in 1973 and the UB series continued the effort in 1977. The conclusions from those efforts were to the effect that the hydrology beneath the site was so complex as to make understanding "difficult."

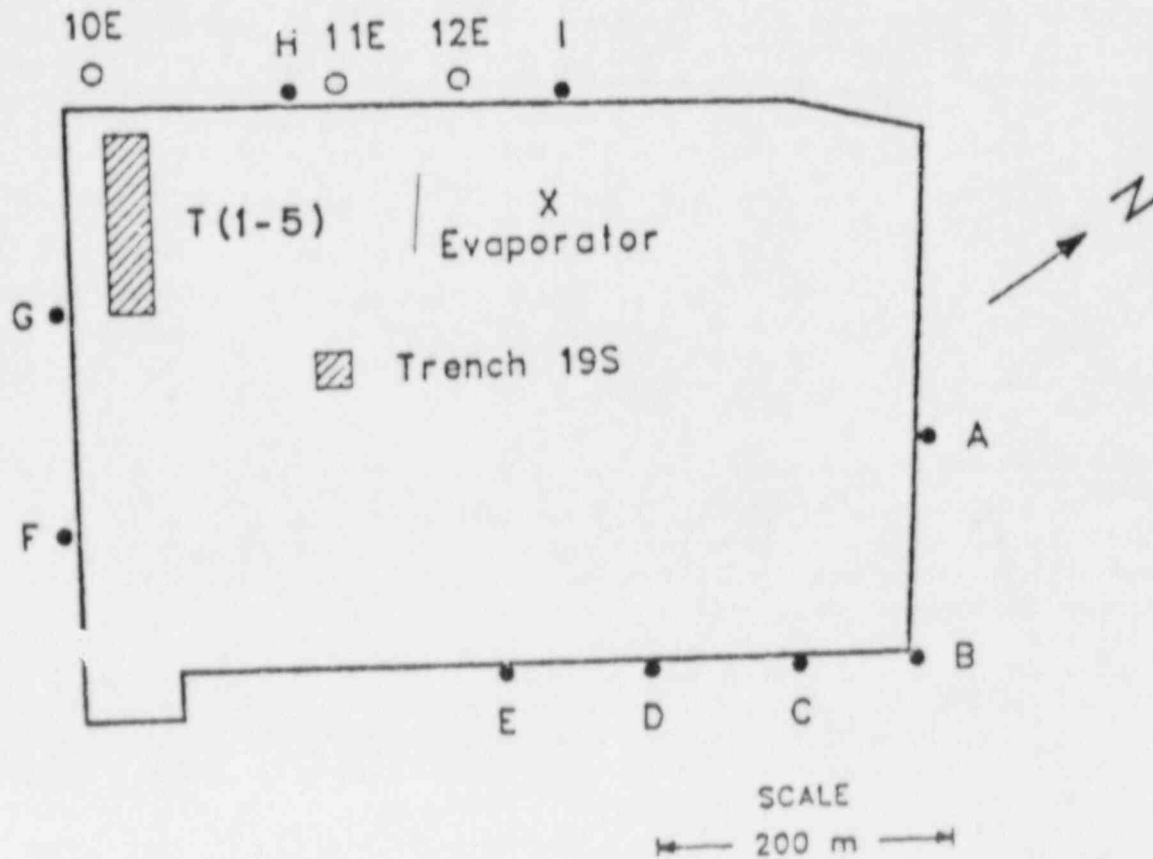


Figure 11. Approximate location of perimeter sampling stations. The approximate locations of the sampling stations (holes A through I) are presented relative to the boundary fence and to the experimental study areas (T4 and Trench 19S). Each sampling location is between 5 and 10 m outside the fence. At each location an 8.3-cm-diam soil auger was used to obtain soil samples in approximately 20-cm increments to depth dictated by impenetrable rocks. The open circles indicate the locations of U.S. Geological Survey (USGS) wells 10E, 11E, and 12E.

TABLE XX
TRITIUM, CESIUM, AND COBALT LEVELS IN SOILS COLLECTED
OUTSIDE THE WASTE BURIAL SITE BOUNDARY

Hole	Depth (cm)	Activity		
		^3H ($\times 10^3$ pCi/l)	^{137}Cs (pCi/g)	^{60}Cs (pCi/g)
A	0-23	48	0.34s	ND ^a
	23-41	38	0.18	ND
	41-61	34	ND	0.14
	61-76	28	ND	ND
	76-91	22	ND	ND
	91-109	16	ND	ND
B	0-18	52	0.68	ND
	18-20	55	ND	ND
C	0-23	81	0.52	0.25
	23-41	59	ND	ND
	41-58	34	ND	ND
D	0-28	52	ND	ND
	28-48	32	ND	ND
	48-66	25	ND	ND
	66-89	15	ND	ND
	89-99	12	0.14	ND
E	0-23	100	0.84	0.34
	23-33	23	0.32	0.20
F	0-20	24	0.23	ND
	20-41	24	0.14	ND
G	0-20	10	ND	ND
	20-36	9.9	ND	ND
	36-48	8.1	ND	ND
	48-61	8.1	ND	ND
H	0-20	230	0.53	0.58
	20-36	300	ND	0.33
I	0-28	84	0.32	ND
	28-41	94	ND	ND
	41-61	72	ND	ND

^aBelow detection.

The concentrations in hole "E" are probably related to wash from the east main drain; those at hole "H" are related to trench 31 and previous waste storage areas. The decreases in concentration with depth in six of the nine holes strongly suggest surface contamination.

TABLE XXI
 TRITIUM LEVELS IN WELL 11E AND
 THE TWO ADJACENT WELLS

Well	^3H ($\times 10^3$ pCi/l)
10E	0.7
11E	3480
12E	11.9

The relatively high level of tritium in well 11E may argue for subsurface flow to off-site in this case up gradient. The radionuclide could originate from trench #31, which has the highest recorded concentrations of buried tritium more than 484,000 curies.

The need for definitive information relating to subsurface migration led to a further search for a productive experimental design, a task approaching the impossible when the numerous interdependent unknowns are considered. However, a decision was made to construct a slit trench at some location on the Maxey Flats site to study subsurface flow and the influence of cap structure on percolation of rainwater. The final trench was a series of five trenches separated by soil columns left in place at time of excavation (Fig. 4).

The Los Alamos team was to study fracture flow from the standpoint of physical and chemical characteristics of the soil solution. However, the trench assigned to Los Alamos did not have fissures in its walls, so the trench was instrumented with a vertical stack of porous cups; Schulz installed a vertical stack of soil moisture sensors and mini-porous cups so as to document wetting of the cap and the trench wall.^{17,18}

The trenches were backfilled with crushed limestone. The influence of the crushed limestone on the physical and chemical analyses of trench sump solutions has not been documented.

At the time of construction of sub-trenches 1 and 2 a seepage of soil water into those trenches occurred. Plutonium was identified in the water, probably soluble (chelated) as well as associated with suspended material. It was suspected that a small perched aquifer, perhaps produced by liquid from the adjacent burial trench #27, had been tapped.

The Los Alamos team had suggested, based on its previous work at trench 19-S, that if migration of plutonium were to occur, it would probably migrate as a chelated or complexed species. The above observation and the following reinforce that conclusion.

A large percentage of the effort of Pacific Northwest Laboratory (PNL) at Maxey Flats since the institution of the slit trench project relates to solution chemistry. EDTA has been identified as the most abundant and HEDTA has been identified as the second most abundant chelate in the trenches; the PNL statements with respect to the presence of those two agents implicates trench #27. By this circuitous route, it can be concluded that plutonium as one radionuclide is found dissolved in soil solution and that chelates may account for that location of the radionuclide.

Answers to the questions, what is migrating, at what rate and in what direction have not been put forth for the short term. Long-term results may provide answers.

IV. SUMMARY

The effort at Maxey Flats to determine its impact on the environment has continued since 1963. The results of the many studies have been given many interpretations. In the optimistic case and most probable to date, radionuclides are not leaving the site via subsurface flow; in a worse case, radionuclides as well as other toxic substances may be leaving the site. In

all fairness, it must be concluded that neither the migration or lack of migration of waste radionuclides by subsurface flow at Maxey Flats has been unequivocally proved; the migration of tritium may be an exception.

The sampling and analyses of soil and soil solution at trench 19-S (closed in 1965) led to the conclusion that some radionuclides, e.g., plutonium, did migrate short distances from the burial trench.

A theoretical tour de force to answer the question "why only a short distance?" indicated the importance of a biopopulation in the soil as well as the importance of Fe(II) as a competitor for the chelate of the potentially mobile plutonium/chelate system, a concomitant release of plutonium to the soil and immobilization of plutonium by sorption.

To the question "what happens if chelated waste from a trench contaminates the near-surface soil -- or if a storage tank or pond leaks?" the answer was reassuring. Certain experimental results indicated that there were in the waste examined two different chelates; one was biodegradable and thermolabile and the other was not biodegradable and was thermostable. The biodegradable complex was degraded by the soil biopopulation and the released plutonium (in this case) was sorbed by the soil. However, the nondegradable fraction would be free to migrate; its fate was not determined. In the experiments reported using Tilsit soil from Maxey Flats and waste from trench 19-S, the soluble, nondegradable fraction of complexed waste was only 14% of the soluble fraction removed by soil/waste interaction.

The refractory chelates include EDTA. However, it has been shown that EDTA will biodegrade with time.¹⁹ Thus, the 14% plutonium associated with the refractory chelate will be slowly released to sorption by soil.

An important reaction that would explain in part the lack of demonstratable soluble plutonium at distances greater than 3 meters from the trench involves the Fe(II), Fe(III),/chelate system. The constants for solubility favor the iron/chelate system over the plutonium/chelate system. Thus, there is a competition for the EDTA chelate in the iron bearing soil; as a result, iron scavenges the chelate from plutonium and plutonium is released to be sorbed by the soil.

The result of the above reaction as well as that discussed in the preceding paragraph is a degradation of the soluble migrating form of plutonium complex and the immobilization of plutonium on the soil matrix.

Although plutonium has been the prime target of investigation at Maxey Flats, other radionuclides have been the subject of study. Tritium, normally considered a good tracer, is so widely dispersed at the site that its value in that respect is minimized. However, the relatively high concentrations of tritium at varied locations on- and off-site are of value as indicators of "hot spots."

A survey of the trench 19-S area using porous cups to collect liquid samples revealed an undocumented burial¹. It is suspected that other such areas exist; their presence perturbs interpretation of data and could negate conclusions relative to migration.

Schulz has used soil moisture data in conjunction with tritium data as indicators of direction of flow of subsurface and near-surface liquid. In the two cases studied, Schulz concluded that infiltration at the trench was predominantly through the cap (old trench 19-S) or by subsurface flow (new trench #4).

At one location, the northwest boundary of the site, soil water, monitor-well water, and vegetation showed elevated levels of tritium. Although the evaporator plume may have contributed tritium to the area, it is more probable that a burial trench (#31) and a storage area for waste acted as prime sources. Indications are that subsurface migration of tritium does occur and in this case upgrade from the source. The high concentration of tritium in deep wells at the northwest boundary supports the above conclusions.

Where ^{60}Co and ^{137}Cs have been detected outside the perimeter fence, their presence can be attributed to surface drainage from the site, e.g., at the fence line in the main east drain.

The five experimental slit trenches constructed in 1979 have indicated the probable presence of small perched aquifers that contain radionuclides -- in one case plutonium. There has been a dearth of information relative to migration per se, direction and especially rates. Solution chemistry has been performed on sump waters. Data are not too different from those previously obtained using porous cups.

It is gratifying to note that the effort at Maxey Flats by a number of different groups with different approaches and over a period of some fifteen years has not detected and proved the subsurface migration of radionuclides from the site; tritium may be an exception. One must conclude, until proved otherwise, that off-site radionuclides arose from on-site surface activities.

V. IN RETROSPECT

The historical experience available relative to burial and containment of radioactive waste at the time of establishment of the Maxey Flats, Kentucky, burial site was minimal if indeed such existed.

The tools necessary to assess reactions accurately, reaction rates -- physical, chemical, and biological--were not incorporated into site or trench design. In addition, documentation relative to what, when, and where was often missing. In some cases undocumented burials were discovered; the perturbation of those burials on analytical results and conclusions remains unknown but could be and probably was so extensive as to negate otherwise acceptable results.

The location of trenches was inexact and samples collected at depth could not be defined as "in trench" or "distance from edge of trench"; thus to define subsurface transport relative to any one trench or to define leachate from specific wastes is highly questionable.

Documentation of trench contents with respect to radionuclides and/or chemical species was lacking; possible interactions among contents such as

solubilization, sorption or desorption, etc., could not be theorized with any degree of certainty. Olsen et al.²⁰ have recently reported results obtained from a study of an Oak Ridge burial trench which had received some 10^6 curies of waste radionuclides. Attempts to control the migration of ^{90}Sr by elevating the pH into the carbonate region with sodium hydroxide increased the mobility of ^{233}U . Those results again reinforce the admonition relative to separation of materials which may interact adversely.

In the earlier days of burial, spills of liquid and solid wastes contaminated surface soil; rainfall in the humid climate (about 43 inches per year) spread contamination with run-off and as a consequence the origins of radionuclides found in off-site samples are open to question. The location of burial trenches (sites) in "humid" areas compounds problems for both present and future control and containment.

Little reliance can be placed on "trench floors" of so called impervious clay, siltstone, limestone, or other such material. Those natural materials do have fissures and do provide drainage planes which may outcrop off site.

Site preparation at Maxey Flats involved "cut and fill" in an attempt to provide a level surface into which trenches were excavated. Fill had been placed over undisturbed vegetation, sod, and surface soil; thus providing a ready conduit from the area of intersection with the trench to offsite where placement of fill had been discontinued. The formation of a layer high in organic matter, decomposing under anerobic conditions, and covered by a relatively loose packed fill is an invitation to disaster.

A. Establishment of Waste Burial Sites

Based on the above listed observations, the following recommendations are made with respect to the establishment of burial sites; recommendations include those intended to facilitate control, monitoring and research.

A site or reserve should be located in an arid or semiarid region, should be removed from populated areas, should have controlled access and should incorporate an extended buffer zone. Air monitoring and deep well monitoring capability should be provided at the perimeter. The area should be fenced against cattle and human intrusion; on-site rodent control should be practiced at all times.

Absolute control, strictly supervised, at each and every burial is a must.

As a minimum all wastes should be segregated as to organic or inorganic; those two classes should not be intermixed at the source and should be labeled at the place where they were generated.

A further separation into Level 1 ("high" -- to be defined) and Level 2 ("low" -- to be defined) would be desirable.

TABLE XVIII
TRITIUM IN SOIL SOLUTION FROM TWO DEPTHS AT TWO
LOCATIONS AT EXPERIMENTAL TRENCH #4

<u>Depth</u>	<u>Location 1</u>	<u>Location 2</u>
Upper (0-3 meter)	150±60	70 ± 10
Lower (3-7 meter)	10 ± 5	5 ± 1

The higher concentrations of tritium in soil solutions from the 0 - 3 meter depth probably results from major flow through the more nearly porous structure of the "old fill" and the inhibition of downward percolation through the less porous "original soil."

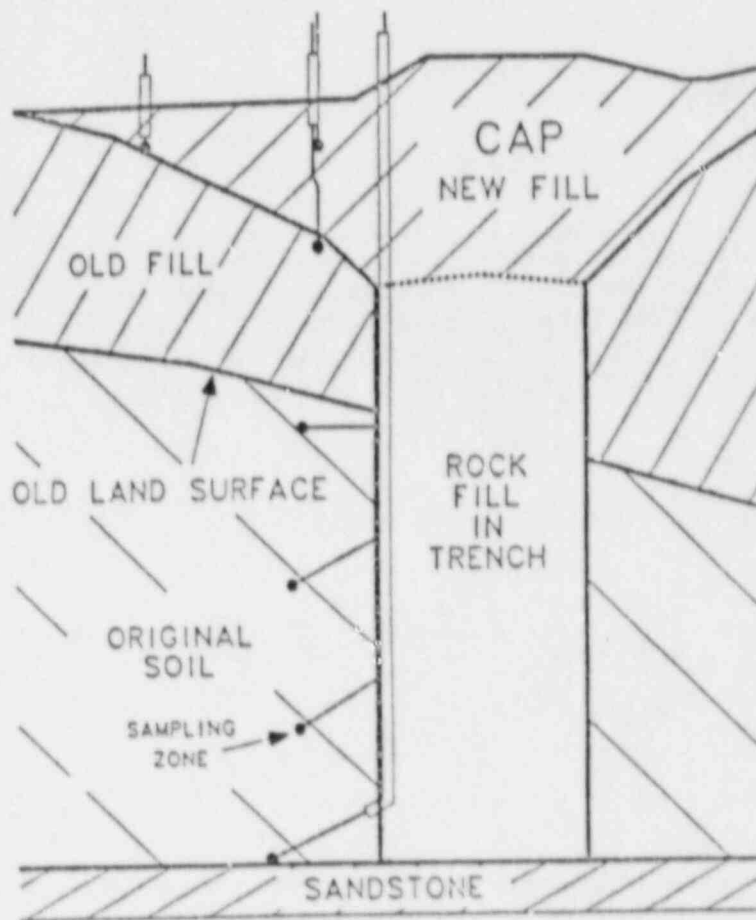


Figure 7. Cross section of the installation at T4. The cross section of porous cup installations at T4 is shown schematically relative to the old landfill--original soil profile.

evaporator; thus, the water level in the trench fluctuates. Based on the premise that soil moisture data could, in a broad sense, indicate changes in direction of flow, moisture sensors were implanted at trench 19-S and about one year later at trench #4.¹⁶

In Fig. 8 the relative soil moisture at three dates is plotted against depth for an extra-trench location about 10 meters from the probable wall of trench 19-S.

At the time of first reading, 10/30/79, the soil from 1.5 to 4.0 meters was very dry -- below the 15 atmosphere permanent wilting point; the moisture content of the soil to a depth of about 1 meter was higher by at least an order of magnitude. By early January, 1980, soil moisture had increased at all sampling points (except the 3 meter point) to a depth of 3 meters and by late June (6/26/80) the soil profile was wet to a depth of 4 meters although not saturated. The water in the trench could not be derived from the soil water because the trench had a free water table at about 2.4 meters; less moisture at the distance of 10 meters, could not move against that gradient. Samples of soil solution were obtained from mini-porous cups implanted adjacent to the moisture sensors. Samples were collected at each moisture tension reading date and analyzed for tritium. The tritium content of the soil solution was essentially constant with depth to 4 meters at about 10^4 pCi/l. That concentration was comparable to the concentration in surface soils at the site and 4 orders of magnitude below that in trench 19-S.

Schulz concluded from the above observation that waste was not moving from the trench to the 10 meter location (tritium data) nor was soil water moving from the 10 meter location to the trench (moisture gradient data). Further, it is implied that changes in soil moisture content, in locus, relate to vegetative cover and evapotranspiration.

Meyer had stated in 1975 that a 1972 program to control water in trenches was designed on the assumption that trench water originated from rain percolating through the cap rather than from ground water. The work of Schulz at trench 19-S would appear to support Meyer's statement.

Schulz then instrumented the cap of trench 19-S and that of experimental trench #4. Results are presented graphically in Fig. 9.

At the end of October (1979) the trench cap at 19-S was wet to a depth of 2 1/2 meters although not saturated. Four months later (2/21/80) the cap was at or near saturation below 1 1/2 meters. The observation is consistent with the conclusion above that water was entering trench 19-S by percolating through the cap. The grass cover on the 19-S cap probably enhances percolation.

Contrary to the results at 19-S are those obtained at trench #4. The cap at #4 was very dry (moisture content below the 15 atmosphere wilting point) to a depth of 2 meters on 10/30/79. The cap was relatively new, perhaps 2 months old, compacted and lacking in vegetative cover. Seven months later, some moisture had percolated downward. The upper 1 1/2 meters were relatively wet.

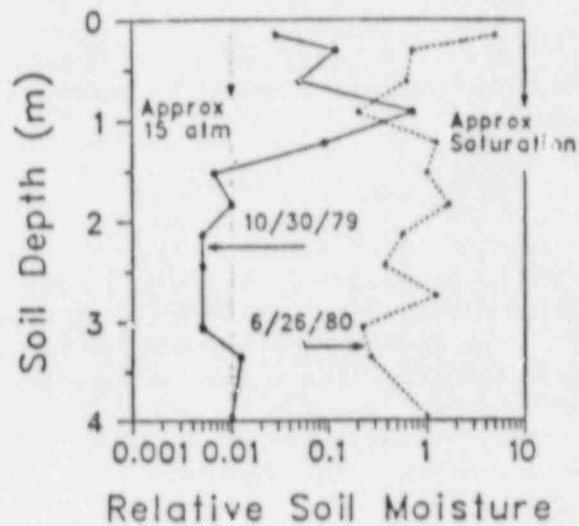
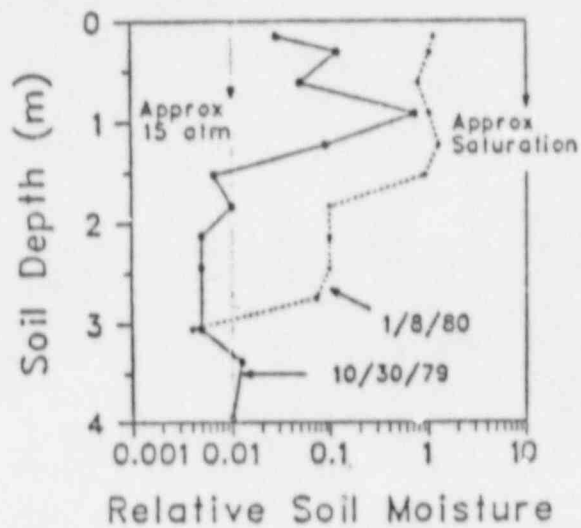


Figure 8. Comparison of relative soil moisture adjacent to Trench 19S in October 1979, January 1980, and June 1980. The relative moisture at a distance of about 10 m from Trench 19S is shown for October 1979 and January 1980 and for October 1979 and June 1980. Relative soil moisture is the reciprocal of resistance of the soil moisture sensor; the lower the relative soil moisture, the lower the moisture content. The soil was quite dry when the first measurements were made in October; as the fall and winter progressed, the soil became wetter with depth. By late June the soil was wet, though unsaturated, down to 4 m.

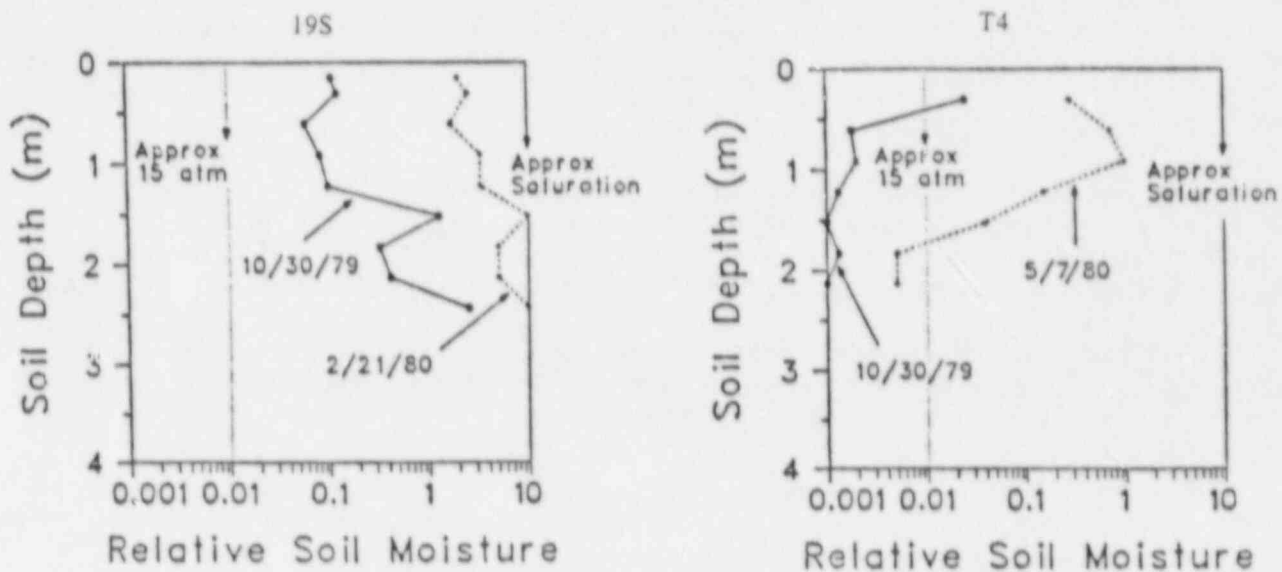


Figure 9. Comparison of relative soil moisture in the caps in Trench 19S and T4. The relative soil moisture in the caps of Trench 19S and T4 is shown for October 1979 and February 1980 and for October 1979 and May 1980, respectively. Relative soil moisture is the reciprocal of resistance of the soil moisture sensor; the lower the relative soil moisture, the lower the moisture content. The rate of moisture movement was much slower in the cap of T4 than in the cap of Trench 19S. In October 1979 the Trench 19S cap was moist; the moisture increased with depth to near saturation at 2.4 m. During the course of the winter, the entire cap became progressively wetter. On the other hand, the cap of T4 was very dry in October 1979. Subsequently, water percolated into the cap, but only to a depth of 1.5 m.

However, the region at 2 meters depth was about as dry as it was on 10/30/79. On the February date, trench #4 contained water about 1 meter deep. That water did not enter the trench through the cap. The conclusion must be made that percolation through the cap and underground flow both add liquid to a trench. The relative importance of the two has not been determined. Protecting trenches against infiltration must consider both sources of liquid.

I. Deep Well Studies

Concomitant with site development at Maxey Flats, eight wells were drilled from southerly to northerly across the site (Fig. 10, #1-8). Radionuclide data from that series of wells are considered of little value because the wells were not cemented and were subject to surface water leakage.

In 1973 the E series of 13 wells, 1E through 14E (no 7E), was drilled (Fig. 10). In general, those wells surround the burial site proper. Data relative to the concentration of radionuclides in samples from the E series wells are limited to tritium. Illustrative data taken from Zehner¹ are presented in Table XIX. The high concentrations of tritium (wells 11E and 13E) are attributed by that author to recharge from a sandstone marker bed. However, the proximity of those wells to the one time waste holding ponds A and C should be considered. Pond liners are not infallible and overflow caused by rainfall does occur. The adjacent position of trench #31 with its extreme concentration of tritium is highly suspect also as a source for the tritium in wells 11E and 13E.

One observation relative to well 14E is of interest. That well has a water level at about 35 feet from the bottom. There is a fissure at 5 feet and one at 15 feet from the bottom. However, the tritium concentration is about equal to the average for the seven lowest concentrations reported in Table XIX. It is recognized that the observation is a single case; however, the data do not support the case that has been argued for fracture flow. Zehner's statement that "Accurate description of the ground-water system . . . is difficult" is certainly not an overstatement of the case.

Other concentrations listed in Table XIX are less than median concentration in rainfall collected at the site and are therefore, of no value in relation to underground flow interpretation.

The UB wells were drilled centrally on site in 1977 (Fig. 10). The high concentrations of tritium found in the UB samples probably reflect that central position. Further, trench #31 is within 150 feet up gradient of the UB series. Trench #31 contains the highest concentration of tritium waste on the site -- more than 484,000 curies.

The UB series of holes was surveyed by down-hole logging. Of the gamma-emitting radionuclides, only ⁶⁰Co was detected - that at UB 1 and in the lower sandstone marker bed. At UB 3, ⁶⁰Co was detected at the 5.4 feet depth but was believed to be associated with operations that contaminated the sample. Cobalt-60 was also observed by the Los Alamos team, at distances from trench 19-S.

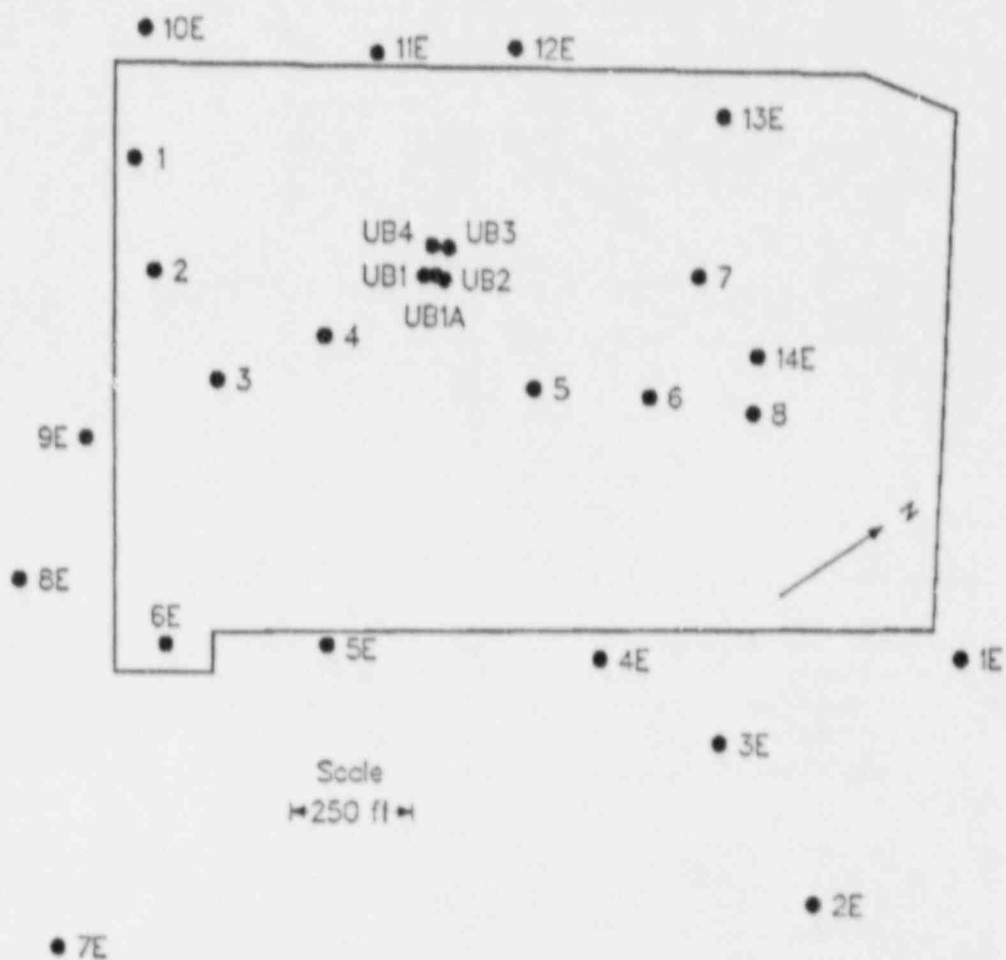


Figure 10. The three series of monitor wells drilled at Maxey Flats. Series I, 1, 2, 3, etc., was drilled north west/south east through the site in 1963. Series II, 1E, 2E, 3E, etc. was drilled so as to surround the site in 1973. Series III, UB4, UB1, etc., was drilled centrally on site in 1977. Some wells were "completed" and provide usable samples.

TABLE XIX
RANGES IN TRITIUM CONCENTRATIONS IN THE E AND UB
SERIES WELLS ON THE MAXEY FLATS SHALLOW LAND BURIAL SITE

Well	Ranges in tritium concentrations (pCi/L)			
1E	5,130	(280) --	5,340	(280)
3E	380	(190) --	7,500	(330)
5E	235	(190) --	3,160	(210)
6E	375	(180) --	18,840	(450)
8E	<200	--	12,410	(380)
10E	660	(200) --	40,140	(620)
11E	730,000	(2,620) --	6,813,130	(7,940)
12E	990	(200) --	27,050	(620)
13E	163,290	(1,250) --	4,592,740	(6,520)
14E	620	(180) --	19,210	(456)
UB1	4,120,000	--	4,180,000	
UB1-A	5,800,000			
UB3	292,000,000			
UB4	7,960,000			

Numbers in parentheses are possible errors due to counting uncertainties; blank if unknown.

The elevated levels in wells 11E and 13E can be explained by their proximity to high level sources. The UB series of wells is located centrally on the site; that location accounts for the elevated levels of tritium. The elevated level in well 1E is not explained by available data. Other concentrations do not differ significantly from those in surface soil and rainfall.

J. Radionuclides Off Site

Based on the physical nature of tritium and the supposed underground drainage gradient, northwest to southeast, the concentrations of tritium and certain other radionuclides were determined on soil and soil solutions outside the fence perimeter, but on site. Nine, 8.3 cm diameter, holes were hand augered at distances of 5 to 10 meters outside the project fence line. The holes were drilled to various depths as dictated by rocks; maximum depth reached was about 110 cm. Location of the holes, A through I is shown in Fig. 11. The analytical results for ^3H , ^{137}Cs , and ^{60}Co in the bore-hole soils are presented in Table XX.

Hole E is the only location where the three radionuclides were found at both depths sampled. Hole E is at the exit of the east main drain. The concentration of each of the three radionuclides decreases with depth suggesting surface contamination carried by run-off.

Analytical results obtained from Hole H soils showed ^{137}Cs (0-20 cm depth), ^{60}Co (0-36 cm depth) and the highest levels of soil tritium (0-36 cm depth). Hole H is less than 30 meters from well 11E. Water from that well contained tritium concentrations several orders of magnitude above those in waters from the other E series wells (Table XXI).

Trench #31 is in close proximity to both Hole H and well 11E. One is tempted to conclude that trench #31 is the source of tritium in Hole H and in well 11E. If the evaporator plume were the primary source, well 12E tritium content should be comparable to that in well 11E. It is not. Well 12E water had a tritium concentration about 300 times lower than that at well 11E. The results presented indicate a possible connection between well 11E and a source such as trench #31.

The elevated levels found at Hole E probably relate to surface run-off along the main east drain. In no case was evidence obtained for extensive underground migration of the radionuclides studied, including tritium.

Rickard et al.¹ studied the accumulation of radionuclides in forest litter and new leaves of trees in the vicinity of Hole H and well 11E. The least tentative of their conclusions states ". . . hickory leaves showed elevated levels of ^{60}Co indicating that the source of ^{60}Co contributing to the tree leaves is probably through local contamination of the rooting substrate . . ."

K. Experimental Trenches

The attempts to characterize the underground hydrology at the Maxey Flats site began with the S hole traverse in 1963. The E series holes were drilled in 1973 and the UB series continued the effort in 1977. The conclusions from those efforts were to the effect that the hydrology beneath the site was so complex as to make understanding "difficult."

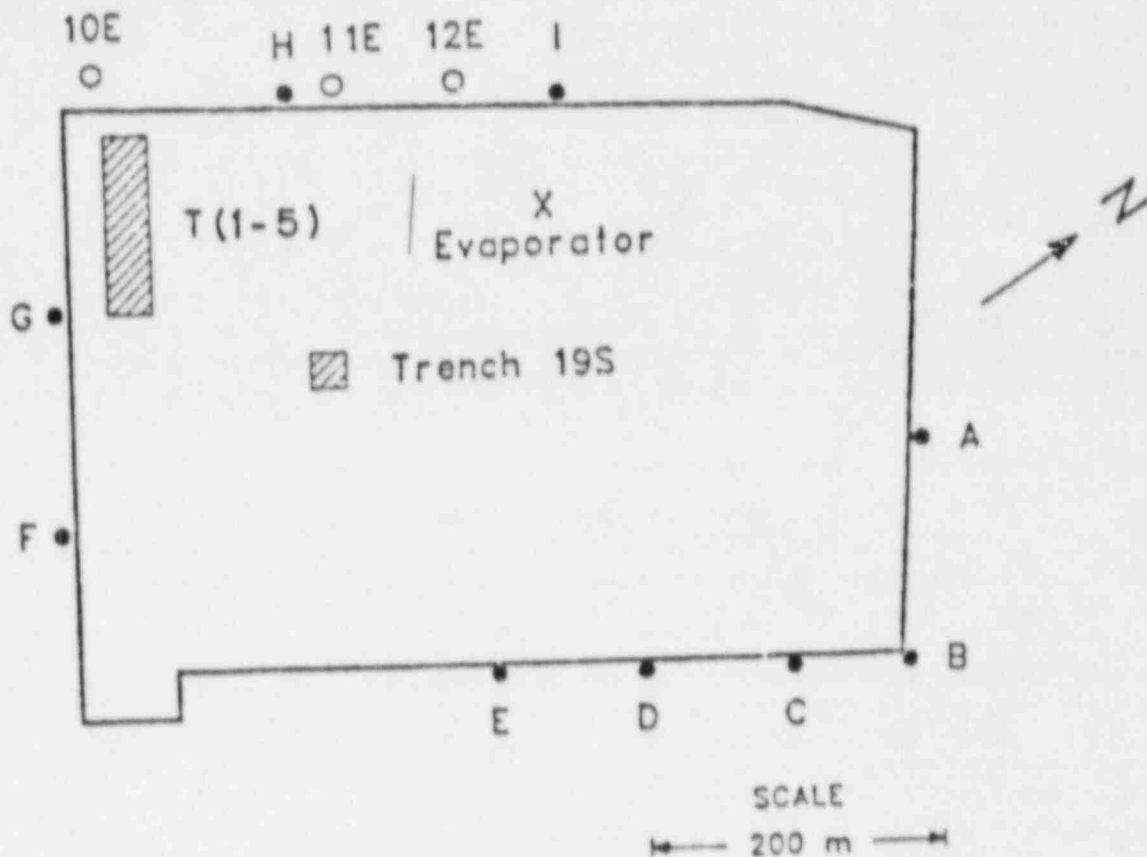


Figure 11. Approximate location of perimeter sampling stations. The approximate locations of the sampling stations (holes A through I) are presented relative to the boundary fence and to the experimental study areas (T4 and Trench 19S). Each sampling location is between 5 and 10 m outside the fence. At each location an 8.3-cm-diam soil auger was used to obtain soil samples in approximately 20-cm increments to depth dictated by impenetrable rocks. The open circles indicate the locations of U.S. Geological Survey (USGS) wells 10E, 11E, and 12E.

TABLE XX
TRITIUM, CESIUM, AND COBALT LEVELS IN SOILS COLLECTED
OUTSIDE THE WASTE BURIAL SITE BOUNDARY

Hole	Depth (cm)	Activity		
		³ H (x 10 ³ pCi/l)	¹³⁷ Cs (pCi/g)	⁶⁰ Cs (pCi/g)
A	0-23	48	0.34s	ND ^a
	23-41	38	0.18	ND
	41-61	34	ND	0.14
	61-76	28	ND	ND
	76-91	22	ND	ND
	91-109	16	ND	ND
B	0-18	52	0.68	ND
	18-20	55	ND	ND
C	0-23	81	0.52	0.25
	23-41	59	ND	ND
	41-58	34	ND	ND
D	0-28	52	ND	ND
	28-48	32	ND	ND
	48-66	25	ND	ND
	66-89	15	ND	ND
	89-99	12	0.14	ND
E	0-23	100	0.84	0.34
	23-33	23	0.32	0.20
F	0-20	24	0.23	ND
	20-41	24	0.14	ND
G	0-20	10	ND	ND
	20-36	9.9	ND	ND
	36-48	8.1	ND	ND
	48-61	8.1	ND	ND
H	0-20	230	0.53	0.58
	20-36	300	ND	0.33
I	0-28	84	0.32	ND
	28-41	94	ND	ND
	41-61	72	ND	ND

^aBelow detection.

The concentrations in hole "E" are probably related to wash from the east main drain; those at hole "H" are related to trench 31 and previous waste storage areas. The decreases in concentration with depth in six of the nine holes strongly suggest surface contamination.

TABLE XXI
 TRITIUM LEVELS IN WELL 11E AND
 THE TWO ADJACENT WELLS

Well	^3H ($\times 10^3$ pCi/l)
10E	0.7
11E	3480
12E	11.9

The relatively high level of tritium in well 11E may argue for subsurface flow to off-site in this case up gradient. The radionuclide could originate from trench #31, which has the highest recorded concentrations of buried tritium more than 484,000 curies.

The need for definitive information relating to subsurface migration led to a further search for a productive experimental design, a task approaching the impossible when the numerous interdependent unknowns are considered. However, a decision was made to construct a slit trench at some location on the Maxey Flats site to study subsurface flow and the influence of cap structure on percolation of rainwater. The final trench was a series of five trenches separated by soil columns left in place at time of excavation (Fig. 4).

The Los Alamos team was to study fracture flow from the standpoint of physical and chemical characteristics of the soil solution. However, the trench assigned to Los Alamos did not have fissures in its walls, so the trench was instrumented with a vertical stack of porous cups; Schulz installed a vertical stack of soil moisture sensors and mini-porous cups so as to document wetting of the cap and the trench wall.^{17,18}

The trenches were backfilled with crushed limestone. The influence of the crushed limestone on the physical and chemical analyses of trench sump solutions has not been documented.

At the time of construction of sub-trenches 1 and 2 a seepage of soil water into those trenches occurred. Plutonium was identified in the water, probably soluble (chelated) as well as associated with suspended material. It was suspected that a small perched aquifer, perhaps produced by liquid from the adjacent burial trench #27, had been tapped.

The Los Alamos team had suggested, based on its previous work at trench 19-S, that if migration of plutonium were to occur, it would probably migrate as a chelated or complexed species. The above observation and the following reinforce that conclusion.

A large percentage of the effort of Pacific Northwest Laboratory (PNL) at Maxey Flats since the institution of the slit trench project relates to solution chemistry. EDTA has been identified as the most abundant and HEDTA has been identified as the second most abundant chelate in the trenches; the PNL statements with respect to the presence of those two agents implicates trench #27. By this circuitous route, it can be concluded that plutonium as one radionuclide is found dissolved in soil solution and that chelates may account for that location of the radionuclide.

Answers to the questions, what is migrating, at what rate and in what direction have not been put forth for the short term. Long-term results may provide answers.

IV. SUMMARY

The effort at Maxey Flats to determine its impact on the environment has continued since 1963. The results of the many studies have been given many interpretations. In the optimistic case and most probable to date, radionuclides are not leaving the site via subsurface flow; in a worse case, radionuclides as well as other toxic substances may be leaving the site. In

all fairness, it must be concluded that neither the migration or lack of migration of waste radionuclides by subsurface flow at Maxey Flats has been unequivocally proved; the migration of tritium may be an exception.

The sampling and analyses of soil and soil solution at trench 19-S (closed in 1965) led to the conclusion that some radionuclides, e.g., plutonium, did migrate short distances from the burial trench.

A theoretical tour de force to answer the question "why only a short distance?" indicated the importance of a biopopulation in the soil as well as the importance of Fe(II) as a competitor for the chelate of the potentially mobile plutonium/chelate system, a concomitant release of plutonium to the soil and immobilization of plutonium by sorption.

To the question "what happens if chelated waste from a trench contaminates the near-surface soil -- or if a storage tank or pond leaks?" the answer was reassuring. Certain experimental results indicated that there were in the waste examined two different chelates; one was biodegradable and thermolabile and the other was not biodegradable and was thermostable. The biodegradable complex was degraded by the soil biopopulation and the released plutonium (in this case) was sorbed by the soil. However, the nondegradable fraction would be free to migrate; its fate was not determined. In the experiments reported using Tilsit soil from Maxey Flats and waste from trench 19-S, the soluble, nondegradable fraction of complexed waste was only 14% of the soluble fraction removed by soil/waste interaction.

The refractory chelates include EDTA. However, it has been shown that EDTA will biodegrade with time.¹⁹ Thus, the 14% plutonium associated with the refractory chelate will be slowly released to sorption by soil.

An important reaction that would explain in part the lack of demonstratable soluble plutonium at distances greater than 3 meters from the trench involves the Fe(II), Fe(III),/chelate system. The constants for solubility favor the iron/chelate system over the plutonium/chelate system. Thus, there is a competition for the EDTA chelate in the iron bearing soil; as a result, iron scavenges the chelate from plutonium and plutonium is released to be sorbed by the soil.

The result of the above reaction as well as that discussed in the preceding paragraph is a degradation of the soluble migrating form of plutonium complex and the immobilization of plutonium on the soil matrix.

Although plutonium has been the prime target of investigation at Maxey Flats, other radionuclides have been the subject of study. Tritium, normally considered a good tracer, is so widely dispersed at the site that its value in that respect is minimized. However, the relatively high concentrations of tritium at varied locations on- and off-site are of value as indicators of "hot spots."

A survey of the trench 19-S area using porous cups to collect liquid samples revealed an undocumented burial. It is suspected that other such areas exist; their presence perturbs interpretation of data and could negate conclusions relative to migration.

Schulz has used soil moisture data in conjunction with tritium data as indicators of direction of flow of subsurface and near-surface liquid. In the two cases studied, Schulz concluded that infiltration at the trench was predominantly through the cap (old trench 19-S) or by subsurface flow (new trench #4).

At one location, the northwest boundary of the site, soil water, monitor-well water, and vegetation showed elevated levels of tritium. Although the evaporator plume may have contributed tritium to the area, it is more probable that a burial trench (#31) and a storage area for waste acted as prime sources. Indications are that subsurface migration of tritium does occur and in this case upgrade from the source. The high concentration of tritium in deep wells at the northwest boundary supports the above conclusions.

Where ^{60}Co and ^{137}Cs have been detected outside the perimeter fence, their presence can be attributed to surface drainage from the site, e.g., at the fence line in the main east drain.

The five experimental slit trenches constructed in 1979 have indicated the probable presence of small perched aquifers that contain radionuclides -- in one case plutonium. There has been a dearth of information relative to migration per se, direction and especially rates. Solution chemistry has been performed on sump waters. Data are not too different from those previously obtained using porous cups.

It is gratifying to note that the effort at Maxey Flats by a number of different groups with different approaches and over a period of some fifteen years has not detected and proved the subsurface migration of radionuclides from the site; tritium may be an exception. One must conclude, until proved otherwise, that off-site radionuclides arose from on-site surface activities.

V. IN RETROSPECT

The historical experience available relative to burial and containment of radioactive waste at the time of establishment of the Maxey Flats, Kentucky, burial site was minimal if indeed such existed.

The tools necessary to assess reactions accurately, reaction rates -- physical, chemical, and biological--were not incorporated into site or trench design. In addition, documentation relative to what, when, and where was often missing. In some cases undocumented burials were discovered; the perturbation of those burials on analytical results and conclusions remains unknown but could be and probably was so extensive as to negate otherwise acceptable results.

The location of trenches was inexact and samples collected at depth could not be defined as "in trench" or "distance from edge of trench"; thus to define subsurface transport relative to any one trench or to define leachate from specific wastes is highly questionable.

Documentation of trench contents with respect to radionuclides and/or chemical species was lacking; possible interactions among contents such as

solubilization, sorption or desorption, etc., could not be theorized with any degree of certainty. Olsen et al.²⁰ have recently reported results obtained from a study of an Oak Ridge burial trench which had received some 10^6 curies of waste radionuclides. Attempts to control the migration of ^{90}Sr by elevating the pH into the carbonate region with sodium hydroxide increased the mobility of ^{233}U . Those results again reinforce the admonition relative to separation of materials which may interact adversely.

In the earlier days of burial, spills of liquid and solid wastes contaminated surface soil; rainfall in the humid climate (about .13 inches per year) spread contamination with run-off and as a consequence the origins of radionuclides found in off-site samples are open to question. The location of burial trenches (sites) in "humid" areas compounds problems for both present and future control and containment.

Little reliance can be placed on "trench floors" of so called impervious clay, siltstone, limestone, or other such material. Those natural materials do have fissures and do provide drainage planes which may outcrop off site.

Site preparation at Maxey Flats involved "cut and fill" in an attempt to provide a level surface into which trenches were excavated. Fill had been placed over undisturbed vegetation, sod, and surface soil; thus providing a ready conduit from the area of intersection with the trench to offsite where placement of fill had been discontinued. The formation of a layer high in organic matter, decomposing under anerobic conditions, and covered by a relatively loose packed fill is an invitation to disaster.

A. Establishment of Waste Burial Sites

Based on the above listed observations, the following recommendations are made with respect to the establishment of burial sites; recommendations include those intended to facilitate control, monitoring and research.

A site or reserve should be located in an arid or semiarid region, should be removed from populated areas, should have controlled access and should incorporate an extended buffer zone. Air monitoring and deep well monitoring capability should be provided at the perimeter. The area should be fenced against cattle and human intrusion; on-site rodent control should be practiced at all times.

Absolute control, strictly supervised, at each and every burial is a must.

As a minimum all wastes should be segregated as to organic or inorganic; those two classes should not be intermixed at the source and should be labeled at the place where they were generated.

A further separation into Level 1 ("high" -- to be defined) and Level 2 ("low" -- to be defined) would be desirable.

A third class designated "trash" should include building and equipment components, glassware, etc. To avoid extensive and expensive segregation, organic trash as dead animals, rubber gloves, clothing, etc., could be buried with other organics. Valid arguments can be made for more extensive segregation, e.g. separation of chemicals and further separation of corrosive from noncorrosive. Regulations as to container material, shape and size, placement in trenches -- loose dump or stack -- are also important facets of waste management. However, it cannot be overemphasized that no regulation is effective without the proper education of the producer and the waste management handlers.

It is possible that a single load may contain waste for as many as five burial pits. Only one load should be placed for burial at any one time; before, during and after burial, monitoring is a must for every load; contents of every load must be documented. No burial of waste can be allowed to proceed until complete and satisfactory documentation of each step has been made and approved.

A cover of soil should be placed over all exposed material and should be compacted at the end of each day. Markers are to be placed which define each day's burial and each laboratory's contribution. The markers are to define the absolute edges of a pit. A permanent concrete curb would be of further assistance to maintain the definition of the pit's edge.

Burial shall be in pits which have been exactly located on an exact topographical map of the site. Exact location shall be given; large monuments shall be placed at the four corners and shall designate period of filling and date of close with log reference.

At the time the pit is excavated, soil samples shall be taken in a grid pattern at each three feet of depth. Percolation rates shall be determined in situ on the floor of the pit following the same grid pattern as above. Sorption rates for the various constituents from an aqueous solution of mixed radionuclides should be determined for the grid soil samples. All soil samples are to be catalogued in a prominent library for future reference or use.

Each pit is to be provided with a perimeter drainage system at floor level so as to conduct collected seepage to a central cistern system. Each pit system should be provided with means for obtaining liquid samples; chemical/radiochemical analyses performed on those samples will provide one monitoring parameter.

Unsophisticated analyses shall be performed at an on-site laboratory to provide early indication of changes in chemical/radiochemical characteristics of trench seepage.

Some of these recommendations have been made over the past 8 to 10 years; they are repeated here for emphasis.

B. Existing Waste Burial Sites

The Maxey Flats burial site had been in operation for more than 10 years when the 1974 NRC research project at the site was initiated.

The above section relative to observations and recommendations recites the problems which became evident as research at the site developed. Those problems were recognized through "retrospective vision".

Once a waste has been placed in a pit and subjected to leaching by ground water for extended periods of time it will enter the environment; an economic means for complete recovery is not within the grasp of our current technology.

The most important lesson to be learned is that preparation of a burial site before the fact is a must.

Although the outlook for environmental control at established burial grounds is dim, there are approaches which could help minimize further negative impacts of those sites on the immediate area. One such approach was recommended to the NRC several years ago in a memo addressing the possibility of subsurface transport at the Maxey Flats site and an attempt at control.

In brief the proposal involved the construction of infiltration galleries at the down-flow side of the trench or site. Collected ground water and such waste as associated would be pumped to storage reservoirs. Those collected wastes would be pumped to a small chemical treatment plant where they would be processed as dictated by analytical monitoring results.

At Maxey Flats the infiltration gallery could be located at the site perimeter; collection reservoirs could be located downstream and treatment plant below; pumping costs would be minimal.

Tritiated water is not removed by routine treatment. At present it is released to the atmosphere. Controlled release and atmospheric dilution could continue.

Such a collection and treatment scheme could be installed at established sites; it would not assure against subsurface migration but would serve to remove a high percentage of possibly migrating solution and thus reduce its impact on the environment.

It is important to note that estimated migration rates based on laboratory results and on theory for the majority of radionuclides tested are low and time and adequate buffer zones are positive factors. Our technology of today does not allow us to "uncreate" what has been created; we must learn to live with it and control it as best we can; at present, that involves collection, removal and treatment of the largest volume of the possible soil/waste solution.

Four questions were posed in the preface, those four served to guide the various investigations at the Maxey Flats site. The questions and related conclusions resulting from the study reported here follow:

1. Are radionuclides migrating from trenches?
Data obtained at trench 19-S and at the experimental trenches provide evidence for positive migration from the trench.
2. If so are they migrating off-site?
Data do not provide evidence of positive migration off-site; migration of tritium may be an exception; that the off-site radionuclides detected early in the history of Maxey Flats did not arise from surface spillage has not been proved or disproved.
3. If so, what is migrating and how?
Tritium has been detected off-site; however, in the evaporator stack effluent the concentration of tritium is high and its deposition is widespread (the qualification in #2 also applies).
4. Can the rate of migration be determined?
The rate of migration has been theorized. It has not been determined because a point source of a specific radionuclide does not exist at the site. The experiment proposed to provide the necessary conditions for rate determination was not conducted.

REFERENCES

1. Kirby, L. J., W. H. Rickard and A. P. Toste, "Chemical Species of Migrating Radionuclides at Commercial Shallow Land Burial Sites," Quarterly Progress Report, 1983, PNL-4432-6.
2. Zehner, H. H., "Hydrogeologic Investigation of the Maxey Flats Radioactive Burial Site, Fleming County, Kentucky," U.S. Geological Survey Open-File Report 83-113, 1983.
3. Meyer, G. Lewis, "Preliminary Data on the Occurrence of Transuranium Nuclides in the Environment at the Radioactive Waste Burial Site Maxey Flats, Kentucky," 1975, Office of Radiation Programs, U. S. Environmental Protection Agency, Washington, DC.
4. Montgomery, Daniel M., Kolde, H. E., and Balnchand, Richard L., "Radiological Measurements at the Maxey Flats Radioactive Waste Burial Site," 1977, EPA-520/5-76/020.
5. Fowler, E. B., Essington, E. H., and Polzer, W. L., "Interactions of Radioactive Wastes with Soils - A Review," 1979, NUREG/CR-1155, LA-UR-79-2910.
6. Fowler, Eric B., Polzer, Wilfred L., and Essington, Edward H., 1981, "Characteristics of Wastes and Soils Affecting Transport of Radionuclides Through the Soil and Their Relationship to Waste Management," NUREG/CR-1763, LA-8563-PR.
7. Nelson, D. M., J. O. Karttunen, K. A. Orlandini, and R. P. Larson, "Influence of Dissolved Organic Carbon on the Sorption of Plutonium in

DISTRIBUTION

	<u>Copies</u>
Nuclear Regulatory Commission, RW, Laurel, Maryland	303
Technical Information Center, Oak Ridge, Tennessee	2
Los Alamos National Laboratory, Los Alamos, New Mexico	<u>50</u>
	355

NRC FORM 328 12-84 NRCM 1102 3201, 3202	U.S. NUCLEAR REGULATORY COMMISSION	REPORT NUMBER (Assigned by TDC add Vol. No., if any)
BIBLIOGRAPHIC DATA SHEET		NUREG/CR-5170 LA-11354-MS
SEE INSTRUCTIONS ON THE REVERSE		
2. TITLE AND SUBTITLE	3. LEAVE BLANK	
A Review of Research Conducted by Los Alamos National Laboratory for the NRC with Emphasis on the Maxey Flats, KY, Shallow Land Waste Burial Site	* DATE REPORT COMPLETED	
	MONTH	YEAR
	May	1988
4. AUTHOR(S)	5. DATE REPORT ISSUED	
E. B. Fowler W. L. Polzer	MONTH	YEAR
	August	1988
7. PERFORMING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)	8. PROJECT/TASK WORK UNIT NUMBER	
Los Alamos National Laboratory Los Alamos, NM 87545		
	9. PIN OR GRANT NUMBER	
	A7040	
10. SPONSORING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)	11a. TYPE OF REPORT	
Division of Engineering Office of Nuclear Regulatory Research U.S. Nuclear Regulatory Commission Washington, DC 20555	Technical	
	b. PERIOD COVERED (Include Dates)	
12. SUPPLEMENTARY NOTES		
13. ABSTRACT (200 words or less)		
<p> Studies to determine the impact of the Maxey Flats low-level waste burial site on the environment have been conducted since 1963. Neither the migration or lack of migration of waste radionuclides to the off site by subsurface flow has been unequivocally proved; the migration of tritium may be an exception. Some radionuclides, e.g. plutonium, did migrate short distances from a burial trench. The movement of "only a short distance" was attributed to the importance of a biopopulation in the soil and to the importance of soluble iron as a competitor for the chelate of the potentially mobile plutonium/chelate system. In both cases a soluble plutonium complex is degraded and the plutonium is released in a form that is sorbed by the soil. Soil moisture data in conjunction with tritium data indicate that infiltration into the trench was predominately through the trench cap. Tritium data also indicate subsurface migration of tritium. </p>		
14. DOCUMENT ANALYSIS - KEY WORDS/DESCRIPTORS	15. AVAILABILITY STATEMENT	
waste burial site Maxey Flats, KY radionuclides	tritium plutonium	Unlimited
16. IDENTIFIERS OPEN ENDED TERMS	17. SECURITY CLASSIFICATION	
	(If applicable) Unclassified	
	(If applicable) Unclassified	
	18. NUMBER OF PAGES	
	19. PRICE	

UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D.C. 20555

OFFICIAL BUSINESS
PENALTY FOR PRIVATE USE, \$300

SPECIAL FOURTH-CLASS RATE
POSTAGE & FEES PAID
USNRC
PERMIT No. G-87

130555139217 1 1A11RW
USNRC - GARM-ADM
DATA & PUBLICATIONS SVCS
OR NUREG
WASHINGTON DC 20555

NUREG/CR-5170

A REVIEW OF RESEARCH CONDUCTED BY LOS ALAMOS NATIONAL LABORATORY FOR THE NRC
WITH EMPHASIS ON THE MAYEX FLATS, KY, SHALLOW LAND BURIAL SITE

AUGUST 1988