NUREG/CR-2383
PNL-4067

Radionuclide Distributions and Migration Mechanisms at Shallow Land Burial Sites

Annual Report of Research Investigations on the Distribution, Migration and Containment of Radionuclides at Maxey Flats, Kentucky

Prepared by L. J. Kirby, Editor

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Prepared for
U.S. Nuclear Regulatory Commission

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Manuscript Completed: October 1981
Date Published: July 1982

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NRC FIN B2291

PREFACE

The research covered by this report was performed by Pacific Northwest Laboratory, Brookhaven National Laboratory, Los Alamos National Laboratory, and the University of California-Berkeley for the U.S. Nuclear Regulatory Commission (NRC). Research projects at the Maxey Flats, KY shallow land burial site are reported through June, 1981. Project monitors for NRC Maxey Flats research are Dr. Edward O'Donnell and Dr. George Birchard of the Waste Management Branch, Office of Nuclear Regulatory Research. Chief of the Waste Management Branch is Jared J. Davis.

The report was compiled and edited by Dr. Lynn J. Kirby of Pacific Northwest Laboratory and includes research papers prepared by each of the participating laboratories listed above. The editor is indebted to W. H. Rickard, C. W. Thomas, A. P. Toste, C. L. Wilkerson and M. C. McShane of Pacific Northwest Laboratory; A. J. Weiss, K. S. Czyscinski and R. F. Pietrzak of Brookhaven National Laboratory; W. L. Polzer, E. B. Fowler and E. H. Essington of Los Alamos National Laboratory; and R. K. Schulz of the University of California-Berkeley for preparing the technical papers contained herein. Thanks are also extended to the many reviewers who read the first draft of this report and offered valuable suggestions and criticisms.

ABSTRACT

Subsurface waters at Maxey Flats are anoxic systems with high alkalinity and high concentrations of dissolved ferrous ion. Americium and cobalt in these trench waters are made more soluble by the presence of EDTA, while strontium and cesium are unaffected under the same conditions. EDTA is the major organic complexing component in waste trench 27 leachate, but other polar, water-soluble organics are also present. Evidence points to the migration of plutonium between waste trench 27 and inert atmosphere wells as an EDTA complex. Polar organic compounds may influence the migration of ^{90}Sr and ^{137}Cs. The primary pathway of water entry into the waste burial trenches is through the trench caps, but major increases in water level have occurred in an experimental trench by subsurface flow. The areal distribution of radionuclides at Maxey Flats has been influenced by surface runoff, deposition from the evaporator plume, subsurface flow and the actions of burrowing animals or deep-rooted trees. Vegetal and surface contamination on site and near site are quite low, and only ^{60}Co exceeds commonly observed fallout levels. Radionuclide concentrations in surface soil at Maxey Flats are comparable to concentrations resulting from normal fallout in other areas of high rainfall.

SUMMARY

Background

The Maxey Flats, Kentucky, shallow land burial site is one of several commercial shallow land burial sites that were established in the eastern U.S. where rainfall is comparatively high. Greater problems are experienced with trench cap subsidence and infiltration of surface water in these areas than in the arid western sites. In addition, at Maxey Flats the interpretation of radionuclide movement is complicated by the fact that radioactivity could have spread by processes other than groundwater transport. For example, because of subsidence or trench cap design, some of the trenches have accumulated excess amounts of water which has infiltrated through the trench caps. Seepage has occurred from these trenches, releasing small amounts of the buried waste radionuclides. In addition, site repair work has resulted in some general low-level contamination of the area. Other known sources of contamination include atmospheric transport from the plume of an evaporator being used to remove water from the trenches, and the the spillage of radioactive liquids during the active operation of Maxey Flats as a burial ground. Consequently, it is very difficult to differentiate between environmental contamination from these sources and radionuclide migration by groundwater transport.

A concise history and description of the Maxey Flats site from its opening in 1963 to cessation of disposal operations in 1977 is summarized by Clancy and others (1981)[1]. In October, 1974, the Commonwealth of Kentucky informed NRC of the results of their special six-month environmental study at Maxey Flats. The study, published in December, 1974, concluded that the disposal site was contributing radioactivity to the local environment, but at levels which did not present a public health hazard. The study identified ^{3}H, ^{60}Co, ^{89}Sr, ^{90}Sr, ^{134}Cs, ^{137}Cs,

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[1] J. J. Clancy, D. F. Gray, O. I. Oztunali, "Data Base for Radioactive Waste Management," NUREG/CR-1759, Vol. 1, pp. 3.1-3.20.

^{238}Pu, and ^{239}Pu in individual samples in the unrestricted environment. The radionuclide concentrations considered significant for purposes of this study ranged from slightly to several orders of magnitude (for certain individual samples) above concentrations that were defined as ambient. The Commonwealth of Kentucky then recommended further studies at the site to assess the long range health and safety significance of their findings. Since that time, numerous studies have been carried out by the Commonwealth, their contractors, the U. S. Geological Survey, and contractors for the Nuclear Regulatory Commission (NRC) and the Department of Energy (see Zehner, 1979[2]; Kirby and others, 1981[3]; and Clancy and others, 1981[1]).

To determine the nature and extent of any radionuclide migration from the site, a precise definition of the source term (radiochemical and chemical composition of waste trench leachates) is required. Data are needed on the chemical species of radionuclides in waste trench leachates in the groundwater outside the trenches and in springs, seeps, and streams adjacent to Maxey Flats. Factors that determine the roles of saturated flow, unsaturated flow, and evapotranspiration must be determined. The importance of the atmospheric pathway for radionuclide transport must be determined, and engineering practices must be established to control water seepage into the waste trenches. A program encompassing all of these factors was initiated in 1979 with Pacific Northwest Laboratory (PNL) as coordinator for the research performed at Maxey Flats by Pacific Northwest Laboratory, Brookhaven National Laboratory (BNL), Dames & Moore, Los Alamos National Laboratory (LANL), University of Arizona, University of California-Berkeley (UCB), University of California-Los Angeles and the United States Geological Survey.

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2. H. H. Zehner, "Preliminary Hydrogeologic Investigation of the Maxey Flats Radioactive Waste Burial Site, Fleming County, Kentucky," U. S. Geological Survey Open File Report 79-1329 (1979).

3. L. J. Kirby, Ed., "Research Program at Maxey Flats and Consideration of Other Shallow Land Burial Sites," NUREG/CR-1832 (1981).

The first research review meeting was held at NRC headquarters in July, 1980, to discuss the research findings. Technical papers from this meeting are contained in NUREG/CR-1832[3].

A second research review meeting was held in Frankfort, Kentucky, July 22 and 23, 1981, to discuss the current status and findings of each of the research studies, and the course that future research should take at Maxey Flats. The meeting was initiated with discussions of ongoing and planned site remedial work, followed by technical presentations by the various research laboratories. Significant research observations, unresolved issues and future research directions were developed during this meeting and are included in this summary. Technical papers prepared by the participating laboratories for presentation at this meeting may be found in this report following the summary.

Objectives

The broad objectives of this research program are:

A. To assess the effectiveness of existing shallow land burial sites. This involves determining what has migrated, environmental impacts, and knowing what will improve siting and operating practices.

B. To develop a data base to assist the Commonwealth of Kentucky in the stabilization and decommissioning of Maxey Flats.

To accomplish these broad objectives, a multidisciplinary and multilaboratory research program was established with the objectives of determining:

- the radiochemical and chemical composition of leachates in the burial trenches
- the areal distribution of radionuclides on the site and the factors responsible for this distribution
- the concentrations of radionuclides in vegetation both on and offsite

- the uptake of radionuclides by representative agricultural crops
- the atmospheric pathways for radionuclide transport and the mechanisms involved
- the subsurface migration rates of radionuclides
- the chemical, physical, biological, and hydrogeological factors which affect radionuclide migration
- the engineering and management practices which influence the seepage of surface waters into the burial trenches.

The broad research program to accomplish these objectives is coordinated by PNL and will help to meet information needs in several critical areas. The relevance of the research projects to each other and to these information needs is summarized in the paragraphs below, along with the primary laboratories involved in each aspect of the research. Where more than one laboratory is listed for an item, separate aspects of that need are pursued within the individual research projects.

For performance assessment, information needs include (1) the definition of leachate source terms (PNL, BNL); (2) the identification of retardation mechanisms in the unsaturated zone (LANL); (3) the comparison of laboratory- and field-derived K_d values (LANL); (4) the comparison of K_d values for various soil/groundwater systems (BNL); and (5) quantification of soil moisture changes, flood potentials and groundwater movement in the unsaturated zone (UCB).

For in situ characterization and monitoring, information needs include (1) determination of the amounts and timing of moisture movement through trench caps (PNL, UCB); (2) the mechanism of soil moisture movement in the unsaturated zone (LANL, UCB); (3) definition of retardation processes in the unsaturated zone (LANL); (4) methodology for field determinations of radionuclide movement (PNL, LANL); (5) state-of-the-art methods for radioactivity surveys to characterize radionuclide distributions at shallow land burial sites (PNL); and (6) methodology for monitoring systems to assure waste containment and site stability (PNL, LANL).

For chemical species determination, one must define (1) chemical changes during transport in the saturated and unsaturated zones (PNL, LANL); (2) complexing and/or chelation processes (PNL); (3) identification of the most mobile radiochemicals (PNL); and (4) identification of types of radiochemicals and areas of contamination (PNL).

These research studies complement the work of the Commonwealth of Kentucky to determine the mass balance of precipitation, surface waters, suspended sediments and bottom sediments in streams and selected radionuclides associated with the surface waters and the sediments at Maxey Flats.

Experimental Facilities

Two experimental areas have been established on the Maxey Flats shallow land burial site (see Section I, Figure 1, page I-19). One has an experimental trench that was especially constructed for this program. It was instrumented and allows the study of actual groundwater transport of radionuclides on the site, and of the behavior of new trenches which employ a variety of capping materials. The second experimental area is near an older waste-filled trench. Both installations permit sampling for comprehensive evaluation of the factors involved in the stated research objectives and are located to permit the evaluation of trench caps. The experimental areas provide a physical bond between the research projects of the various participating laboratories and enable a close integration of research results to satisfy overall program objectives.

The experimental installations were initiated in late 1979 with the construction of the experimental trench near waste trench 27 and the installation of porous cups and soil moisture cells near waste trench 19S. Details of these installations were reported in NUREG/CR-1832 and are reviewed in Section I of this report. These facilities have been used to provide samples for radionuclide and groundwater studies of regions of saturated and unsaturated flow, and soil moisture data. The data obtained are providing information on radionuclide migration, chemical species, subsurface flow by saturated and unsaturated flow, trench cap design and surface drainage to control erosion.

A second series of sampling stations was installed late in 1980. Inert atmosphere wells were installed near the experimental trench to help maintain the initial oxidation state of the dissolved radionuclides in groundwater samples. At the same time, additional porous cups and soil moisture cells were installed near the experimental trench, and a large number of additional porous cups and soil moisture cells were installed in the study area near waste trench 19S. These installations are permitting a more complete determination of the influence of the trench face on unsaturated flow. The installations accomplished in late 1980 should assist in the development of a more comprehensive picture of the hydrology of the zone extending through the depth of adjacent trenches than was permitted with the initial drilling efforts which permitted access to half this depth. Details of these installations are presented in Sections I, V and VI of this report.

These experimental facilities support the research of the laboratories involved in the efforts at Maxey Flats. The research is providing a basis for guidelines on the operation of existing shallow land low-level radioactive waste burial sites and guidelines on remedial action and decommissioning. In addition, it is identifying long-term surveillance and maintenance requirements and site selection criteria for future sites.

Research Observations

Research observations, unresolved issues and future research directions summarized in this section are derived from the presentations made at the July, 1981 research review meeting. They are discussed in greater detail in the body of this report.

Subsurface waters at Maxey Flats are anoxic systems; they have low Eh and dissolved oxygen and generate ammonium and sulfide ions. These waters have high alkalinity and high concentrations of dissolved ferrous ion which when exposed to air will oxidize and precipitate as ferric hydroxide. Scavenging resulting from reactions with the sulfide and ferrous ions will affect the potential subsurface migration of radionuclides through fixation of the radionuclides within a solid matrix. Americium and ^{60}Co in Maxey Flats trench waters are made more soluble by the presence of EDTA, while ^{90}Sr and ^{137}Cs are unaffected under the same conditions. Complexes of ^{60}Co with EDTA and NTA are stable in these waters.

EDTA is the major organic complexing component in waste trench 27 leachate, but other polar, water-soluble organics are also present. Plutonium and EDTA coelute during steric exclusion chromatography of Maxey Flats trench and well waters. This correlation offers strong evidence to support the conclusion that plutonium probably migrates as an EDTA complex. Polar organic compounds may influence the migration of ^{90}Sr and ^{137}Cs. Tritium and inert tracer concentrations in water from the experimental trench area confirm that subsurface flow has occurred from the experimental trench and toward the waste burial trenches during the time span of the measurements. Continuing measurements in this complicated hydrologic system will help determine if this flow direction is controlled by the water level existing in the adjacent waste filled trench (trench 27). Major increases in water level have occurred in the experimental trench without movement through trench caps, as confirmed by nonradioactive tracer studies, demonstrating the importance of subsurface flow as a source of water infiltration.

The work to date indicates that the primary pathway of water entry into the waste burial trenches is by entry through the trench caps. Although a significant amount of the soil water probably moves by interstitial flow, there is some evidence that preferential flow may occur within the site boundaries along a sandstone layer and along a landfill-old soil profile interface. Waste movement from trench 19S was indicated by the detection of tritium in the soil solution collected from the adjacent soils. Tritium has moved the greatest distance from trench 19S; ^{137}Cs was not detected in soil solutions from this area. The relative movements of ^{238}Pu and ^{60}Co are greater than ^{137}Cs, but less than ^{3}H. The sorption of ^{137}Cs and ^{238}Pu in laboratory studies agrees with their observed distribution in the trench 19S area. These and other research studies emphasize the importance of understanding the mode of entry of excess water into the waste burial trenches.

The areal distribution of radionuclides at Maxey Flats has been influenced by surface runoff, deposition from the evaporator plume, subsurface flow and the actions of burrowing animals or deep-rooted trees. Vegetal and surface contamination on site and near site are quite low, however, and only ^{60}Co exceeds commonly observed fallout levels. The average concentrations in surface soil at Maxey Flats are comparable to concentrations resulting from normal fallout in other areas of high rainfall.

Unresolved Issues

Issues which must be satisfactorily solved prior to decommissioning the Maxey Flats site include: (1) relative and absolute transport rates of radionuclides via subsurface routes, (2) relative importance of unsaturated flow in radionuclide transport, (3) chemical species and physical forms of migrating radionuclides, (4) chemical forms of dissolved and suspended radionuclides in trench waters of different pH and EH, (5) source term definition based on observed solubilization rates of radionuclides in waste trenches, (6) accurate determination of trench boundaries, (7) chemical toxicity of dissolved material from buried wastes, (8) methods for retarding dissolution and/or movement of toxic wastes; (9) methods of relating areal distributions of radionuclides to the resulting concentrations in surface runoff, (10) methods of minimizing water infiltration into the waste burial trenches and early detection of subsidence, and (12) changes that might be induced in the chemical behavior of radionuclides on leaving the anoxic environment and entering an oxic environment offsite.

The behavior of mobile, possibly complexed, radionuclide species on leaving the trench environment is of primary concern and forms an important basis for these research studies. To control movement within site boundaries may require remedial actions to retard mobility. One needs to determine whether complexing agents such as EDTA can be rendered ineffective prior to leaving the anoxic trench environment or if their mobilization abilities substantially change on entering the aerobic environment at or near the site surface. The effects of these changes on radionuclide mobility need to be established.

Determination of the primary avenue for water entry into the waste trenches will help to define what countermeasures might be taken to minimize water entry. Some of this information has been obtained from the experimental trench study, but additional information is needed from studies during high rainfall periods. Procedures are needed to determine a complete water balance and how it influences direction and rate of radionuclide movement at Maxey Flats.

From studies of unsaturated flow, one could establish whether significant waste movement occurs by liquid flow from the waste trenches into the soil along a landfill-old soil profile interface. One could also determine whether dilution of the soil solution or dispersion and retardation due to chemical reactions with the soil are the main factors determining radionuclide flow toward the site boundary. These studies will help determine the influence of the chemical and physical conditions of the soil and interstitial water on the movement of waste radionuclides through the soil. They will also help establish whether the mechanism of tritium migration through soil involves mainly liquid or vapor flow.

Management protocol for the decommissioning of a shallow land burial facility must assure waste containment and site stability. The prediction of site behavior is essential for the establishment of this protocol.

At Maxey Flats, water flows from wet weather springs and seeps adjacent to the site following periods of heavy rainfall. Establishment of management practices which assure retention of radionuclides within the site requires a knowledge of whether the water flow results primarily from saturation of the upper few meters of soil, or via groundwater discharge from an aquifer that intercepts waste trench leachate and other radionuclide sources.

Future Research Direction

Some of the studies planned for the immediate future are summaried in the following paragraphs.

- Determine the strength of the plutonium-EDTA complex in waste trench leachate at Maxey Flats and the conditions under which complexes of plutonium, americium and cobalt can be decomposed and/or sorbed on the natural materials present.
- Separate and quantify the hydrophyllic organic complexing agents that may affect the migration of radionuclides from waste trench leachate.
- Determine the effect of Eh and pH on the stability of migrating radionuclide chemical species.
- Determine the extent to which there is preferential water flow from waste burial trenches along the landfill-old soil profile interface.
- Determine the effect of changes in soil moisture and chemical and physical properties of the soil solution on the movement of waste radionuclides through soil.
- Determine countermeasures that can be taken to minimize water entry into the waste burial trenches.
- Through lysimeter studies, predict the effects of soil bulk density, surface slope, and crop type and management on run-off and water burden in the burial trenches and determine the

efficiency of plant transpiration on tritium movement from trench waters to the atmosphere.

- Determine the contribution of wet weather springs or seeps to the total water and radioactivity balance at Maxey Flats.
- Complete the areal distribution studies and relate measured radionuclide distributions in soil to radionuclide concentrations in surface runoff water.
- Determine the influence of rain on fallout from the evaporator plume and subsequent influence on radionuclide distributions.
- Complete the identification of specific radionuclide chemical species in experimental trenches, wells and other points on site, and in water sources off site at Maxey Flats.
- Continue to follow radionuclide concentrations and water levels in experimental trenches and wells for groundwater movement and chemical species studies and analyze these data to determine rates and mechanism of subsurface radionuclide transport.

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SECTION I

CHEMICAL SPECIES OF RADIONUCLIDES AT MAXEY FLATS

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CHEMICAL SPECIES OF RADIONUCLIDES AT MAXEY FLATS

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SUMMARY

This report was prepared for presentation at the second annual Maxey Flats research meeting, at which the participating laboratories reviewed the status of each of the research projects at the site and discussed plans for future research at Maxey Flats. Pacific Northwest Laboratory (PNL) coordinates this research, which has included PNL studies on chemical species and areal distributions, work on the trench chemistry by Brookhaven National Laboratory (BNL), plant uptake by the University of California-Los Angeles, engineering studies by the University of Arizona, unsaturated flow studies by Los Alamos National Laboratory (LANL) and the University of California-Berkeley (UCB), and water and mass balance studies by the Commonwealth of Kentucky, utilizing groundwater hydrology studies conducted by the United States Geological Survey.(1) Currently, four laboratories (PNL, BNL, LANL and UCB) are conducting field projects at Maxey Flats with the cooperation of the Department for Natural Resources of the Commonwealth of Kentucky.

To conduct experiments for determining the subsurface transport of radionuclides from existing waste burial trenches, two significant engineering efforts have been required: (1) the installation and instrumentation of an experimental slit trench and (2) the installation of inert atmosphere sampling wells. This has required the cooperation of several national and state agencies, together with the NRC contractors, for successful implementation. The slit trench experiment was initiated during the last quarter of FY 1979 and inert atmosphere sampling wells were installed during the last quarter of FY 1980 in undisturbed soil adjacent to the

experimental slit trench. These two installations are enabling samples to be taken for the study of chemical species of radionuclides, saturated and unsaturated flow, nonradioactive tracer movement, trench cap engineering and drainage, plant uptake, and water mass balance by the laboratories participating in research at Maxey Flats. These experiments have provided badly needed information on rates and mechanisms of subsurface transport of radionuclides from waste burial trenches by both saturated and unsaturated flow. Sample analyses have also helped define the chemical parameters that affect transport.

Field measurements have been completed at the Beatty, NV, and Maxey Flats, KY, burial sites to help determine the areal distribution of radionuclides at these sites and the factors responsible for this distribution. Field measurements were also performed in the drainages adjacent to the Maxey Flats site, and indicate that contamination is largely contained within the restricted area at this burial site. The areal distribution of radionuclides at Maxey Flats has been influenced by surface contamination and runoff, deposition from the evaporator plume, subsurface-to-surface flow and the actions of burrowing animals or deep-rooted trees. Vegetal and surface contamination on site and near site are quite low, however, and only ^{60}Co exceeds commonly observed fallout levels. The average concentrations in surface soil at Maxey Flats are comparable to concentrations resulting from normal fallout in other areas of high rainfall.

PNL research has demonstrated that EDTA is the major organic complexing component in waste trench 27 leachate, but other polar, water-soluble organic compounds are also present. Plutonium and EDTA coelute during steric exclusion chromatography of Maxey Flats trench and well waters, and further evidence points to the migration of plutonium as an EDTA complex. Polar organic compounds may influence the migration of ^{90}Sr and ^{137}Cs. These studies have demonstrated the need to determine the strength of the plutonium-EDTA complex in waste trench leachate at Maxey Flats, the conditions under which complexes of plutonium, americium, cobalt and strontium can be decomposed and/or sorbed on natural materials, and to separate and quantify the hydrophilic organic complexing agents that may affect the migration of radionuclides from waste trench leachate.

Changes in tritium concentrations and nonradioactive tracer concentrations in water from the experimental trench confirm that subsurface flow does occur rather rapidly. Major increases in water level have occurred in the experimental trench without movement through trench caps. These and other findings have helped clarify our understanding of groundwater flow and indicate the direction of further research at this site. Remaining important areas include the fate of mobile (possibly complexed) radionuclide species after leaving the trench environment, the control of these species within the site boundaries, remedial action that can be taken to halt this mobility, and whether the complexing abilities of compounds such as EDTA can be effectively reduced or destroyed in an aerobic environment.

Important research observations include:

- Establishment of the magnitude of transport over short distances by subsurface flow;
- Characterization of some of the important chemical species including plutonium-EDTA complexes which are responsible for subsurface migration;
- Identification of surface transport as a principal means of off-site radionuclide accumulation in vegetation;
- Identification of waste management practices as a principal source of surface contamination versus actual seepage from buried wastes;
- Determination of the offsite distribution of radionuclides at Beatty, Nevada, and Maxey Flats, Kentucky;
- Establishment of the complexity and variability of waste trench leachates as source terms for the migration studies.

These research observations are described in several publications and presentations based on this program(2,3,4,5,6,7).

Two annual research review meetings have been held to report current research findings for each of the research projects at Maxey Flats, and summarize significant research accomplishments and unresolved issues pertinent to decommissioning the site. The most recent meeting also provided an opportunity for the Commonwealth of Kentucky to express their political and social needs relative to decommissioning of the Maxey Flats site. Technical papers from the presentations at these meetings have been and are being combined to form the NRC documents covering the overall research program.

INTRODUCTION

Since commercial waste disposal was started at Beatty, NV, in 1962, six sites have been licensed in the United States for the commercial shallow land burial of low-level radioactive wastes. Two of the sites, West Valley, NY, and Maxey Flats, KY, were closed due to difficulties which included seepage of water into and from the burial trenches. The other sites in the eastern United States have experienced related difficulties, and the Barnwell, SC, site is now the only eastern site that still accepts some radioactive waste for burial.

From the operating experience at the various sites, those located in the eastern half of the country where rainfall rates are relatively high experience considerably greater problems from subsidence and water infiltration than those located in the more arid regions, mainly Nevada and eastern Washington. In addition, interpretation is complicated by the fact that there are other processes for radionuclide transport besides subsurface transport. For example, at both Maxey Flats and West Valley, reworking of subsidence areas to form new trench caps has resulted in localized spread of contamination. At Maxey Flats, additional known sources of contamination include atmospheric transport from the plume of an evaporator used to concentrate trench waters and spillage of radioactive liquids during the time the site was operating as a burial area. Consequently, it was very difficult from available information to differentiate between environmental contamination from these sources and radionuclide migration by subsurface aquifer transport.

One of the potential problems from the long-term isolation of wastes at Maxey Flats, as well as other shallow land burial sites, is the subsurface migration of radionuclides. The oxidation state and ionic form will influence migration, and the formation of complexes or chelates with organic ligands may greatly facilitate the migration of radionuclides. It is therefore extremely important to determine the amounts, chemical species and migration rates of radionuclides which are moved by subsurface routes to obtain an accurate evaluation of the long-term stability of shallow land burial sites. It is also essential to determine those compounds and ionic species which are present or can be generated in situ and subsequently participate in the formation of the chemical species that are shown to migrate. These data must be related to the areal distribution of radionuclides on and adjacent to the site to determine the effects the factors above will have on the retention of radioactive material at the shallow land burial sites.

With this background, the shallow land low-level waste disposal research program was initiated at Maxey Flats, Kentucky, late in 1979. It is a multidisciplinary, multilaboratory program with the broad objectives to:

A. assess the effectiveness of existing shallow land burial sites, determining what has migrated, environmental impacts, and knowing what will improve siting and operating practices;

B. develop a data base to assist the Commonwealth of Kentucky in the stabilization and decommissioning of Maxey Flats.

To help accomplish these objectives, the PNL project has been developed with the technical objectives to:

- determine the amounts and chemical species of radionuclides which are moved by subsurface routes;
- compare the chemical species of radionuclides in burial trenches with species which are observed in an experimental slit trench, inert atmosphere sampling wells, in springs or seeps which flow after heavy rains, and in surface flow from the site;
- determine the areal distribution of radionuclides over waste burial and adjacent areas and compare radionuclide concentrations in surface run-off waters with soil concentrations and MPC values;
- determine the organic compounds that are present or can be produced in situ by chemical reactions and subsequently influence the migration of radionuclides;
- develop efficient and statistically valid field sampling procedures and methods for post-closure monitoring at shallow land burial sites;
- coordinate research at Maxey Flats and other shallow land burial sites.

This project complements the research conducted by BNL, LANL and UCB on source term definition, retardation mechanisms, laboratory- and field-derived K_d values, soil moisture changes, groundwater movement in the unsaturated zone and chemical changes during transport. These projects are related to the broad problem of shallow land burial which is currently under investigation by the Department of Energy and the Nuclear Regulatory Commission (NRC). The research will provide a basis for policy guidance and the setting of criteria for decommissioning of existing sites and for the licensing of future sites for low-level waste burial.

EXPERIMENTAL METHODS

Experimental Slit Trench

An experimental slit trench was constructed adjacent to an existing waste-filled trench at Maxey Flats in 1979. The trench was designed to accommodate a variety of experiments by the laboratories participating in research at Maxey Flats. Sumps were installed to study saturated flow. Porous cups and soil moisture cells were installed in the trench wall, cap and adjacent area to study unsaturated flow near the trench. Nonradioactive tracer studies were included to monitor groundwater movement to and from the experimental trench and to study the permeability of the trench caps to water movement and to root penetration. Several varieties of vegetation were planted to study the agricultural aspects of trench capping. Engineering studies of trench capping and surface drainage were included because of the importance of erosion and rainwater penetration into the trenches. Trench construction is described in an earlier report(3). The experimental trench summary is reviewed in Table I.

Inert Atmosphere Wells

In a second and related construction effort in 1980, inert atmosphere sampling wells were installed in undisturbed ground adjacent to the experimental trench to study subsurface water that would not be influenced by the trench construction materials. The well caps were sealed, permanent sampling and gas lines installed, and the wells purged with argon. During and between sampling activities, the wells are maintained in an inert argon atmosphere to permit chemical separations to be made on water samples from the wells without the adverse effects of traces of oxygen on the chemical equilibria. The well design permits the determination of chemical species of dissolved radionuclides with minimal disturbance of the chemical balance between soil and groundwater.

Fourteen inert atmosphere wells were installed as part of an effort which also included the installation of sampling stations to study unsaturated flow and soil solution near the experimental trench and in an undisturbed region of the Maxey Flats site between two old waste trench caps. Figure 1 shows the locations of these experimental installations relative to the existing waste-filled trenches. The experimental trench and inert atmosphere wells are shown in greater detail in Figure 2. Details of

TABLE I

Maxey Flats Experimental Trench Summary

	Trench Section				
	T-1	T-2	T-3	T-4	T-5
Gamma-Ray Logging	Partial	Yes	-	Yes	-
Geologic Logging*	-	Partial	-	-	-
Water In-Flow	Yes	Yes	Yes	-	-
Sumps	2	2	2	1	1
Porous Cups	3	3	3	3	3
Porous Cups in Trench Wall	-	-	-	Three Vertical Traverses + Cap	-
Soil Moisture Cells in Trench Wall	-	-	-	One Vertical Traverse + Cap	-
Cap Design	Local Soil/ Clay	Local Soil/ Clay	Local Soil/ Mirafi-140/ Clay	Local Soil/ Clay	Clay/ Hypalon/ Clay
Groundwater	NaBr	NaBr	NaBr	-	NaBr
Cap Tracers	Na Benzoate $BaCl_2$ $NiSO_4$	o-Fluorobenzoic Acid $Sr(NO_3)_2$ Li_2CO_3	Na Benzoate	Pentafluorobenzoic Acid	Na Benzoate
Drainage Ditches	Drain Trench Cap for Tracer Studies and Erosion Control				

*Limited by Safety considerations

sampling stations in the other experimental area at Maxey Flats are presented in Section V, "Potential for Migration of Waste Radionuclides at the Maxey Flats, KY Shallow-Land Burial Site," later in this document.

Drilling of the wells and holes for the porous cup and tensiometer emplacements was performed in two phases to maintain maximum control over the potential spread of radioactive material should any be removed from the drill holes. The first location, near the experimental trench, was considered to have the least potential for removal of radioactive material from the ground. In this location (Phase I), fourteen wells were drilled, cased, sealed and capped for inert atmosphere sampling. The well locations relative to the experimental trench and waste trench 27 are shown in Figure 2 and the well design is shown in Figure 3.

Also in Phase I, holes were drilled for additional porous cup and tensiometer installations near sections T3, T4, and T5 of the experimental trench. The drilling results provided a much better definition of the south boundary of waste trench 27, and confirmed that the experimental trench is about 4 meters at the east end and 7 meters at the west end from waste trench 27. In phase II, numerous porous cup and tensiometer installations were made between waste trenches 18 and 19S. See Section V of this document for details of these emplacements. The drilling was complicated by the lack of sufficient documentation of the waste trench boundaries and the presence of unrecorded buried material in this location. Each drill hole was therefore thoroughly examined for radioactive contamination and the presence of wood or metal objects before continuing on the next hole. These precuations were taken to preclude the generation and spread of radioactive contamination during the drilling and subsequent sampling station installations in this location and near the experimental trench.

Sample Collection and Analysis

Since their installation, the inert atmosphere wells that contained enough water to permit pumping have been sampled regularly. Initially, six of the wells contained sufficient water to permit sampling for radionuclide analysis and chemical species determinations. Eight of the wells are now productive sampling stations, but some wells provide water only on an intermittent basis. Several of the wells, mostly on the western end of the experimental trench near sections 4 and 5, have remained dry since installation. After sampling, the water samples are shipped to PNL for analysis for ^{3}H, ^{60}Co, ^{90}Sr, ^{137}Cs, $^{238,239,240}Pu$, ^{241}Am and organic ligands.

Tritium analyses were performed by liquid scintillation spectrometry and ^{90}Sr was chemically separated and counted using a beta proportional counter. Gamma-emitting radionuclides were analyzed by gamma-ray spectrometry using a Ge(Li) detector. Plutonium and americium were determined using solid state detectors and alpha energy analysis.

Ion Exchange Experiments

For laboratory ion exchange experiments with plutonium and several complexing or chelating agents known or thought to be present in the trenches at Maxey Flats, (Dowex-1 x 8, Cl^-, 50-100 mesh) and (Dowex-50 x 8, H^+, 100-200 mesh) ion exchange columns were equilibrated to the desired pH by passing the appropriate buffer solutions through the columns until the effluent pH stabilized. Stock solutions of ethylenediaminetetraacetic acid (EDTA), tributylphosphate (TBP) and triocytlamine (TOA) were prepared and diluted in the appropriate buffer to prepare 10^{-3}, 10^{-6}, and 10^{-7} $\underline{M}$ solutions of the reagents at pH 4, 5, 6 and 7. Plutonium (+3) and plutonium (+4) tracers were prepared and stabilized in hydrochloric acid. Each buffered reagent solution was spiked with plutonium (+3) or plutonium (+4) tracer prior to passage through the ion exchange column. The total plutonium (+3 or +4) activity not absorbed on the column at each pH was then measured by alpha spectroscopy following electrodeposition. Solutions containing ^{85}Sr were similarly prepared, and the effluent activity measured by gamma-ray spectroscopy.

Radionuclide and Organic Ligand Analysis

A water analysis sequence is shown in Figure 4. The water sample is concentrated by rotary evaporation and the pH adjusted to that of the original water (usually 6.5-7.0). Following passage through a 0.45 filter, the concentrate is loaded on a Sephadex G-15 column (2.5 cm i.d. x 31 cm) and chromatographed with specially purified water at a flow rate of 28 ml/hr. In a separate measurement, blue dextran is used as a void volume marker. The column is monitored by following the ultraviolet absorbance at 254 nm and the effluent collected in 4-ml fractions using a fraction collector. Each column fraction is acidified with concentrated HCl and separated into halves. Half of each fraction is destructively analyzed for ^{90}Sr, ^{238,239,240}Pu, and ^{241}Am; the other half is non-destructively analyzed for ^{137}Cs, ^{60}Co, and other gamma emitters using a Ge(Li) detector, and then analyzed for its organic content.

For organic analysis, each subfraction is extracted with chloroform to remove hydrophobic ligands. The chloroform extract is set aside for gas chromatographic (GC) analysis. The aqueous fraction containing water-soluble polar organic compounds is evaporated to dryness under nitrogen at 100°C. The dry residue is then methylated by adding 1 ml of BF_3/methanol (14% w/v) and incubating at 100°C for 40 minutes in a sealed vial. After cooling, 1 ml of chloroform is added and the mixture transferred to a test tube containing 3 ml of 1 $\underline{M}$ KH_2PO_4 at pH 7. The mixture is vortexed one minute, centrifuged, and part of the chloroform layer (0.4-0.7 ml) is transferred to a glass vial. The chloroform is evaporated under nitrogen at room temperature, the residue redissolved in chloroform (0.1-0.2 ml) and a 1-2 l aliquot analyzed by GC and GC-mass spectrometry (GC-MS).

The GC analysis is performed on a Hewlett-Packard 5880 gas chromatograph with an SE 52 column (0.242 ml i.d. x 20 m, fused silica). From an initial value of 40°C, the column temperature is programmed at 20°C per minute for three minutes to 100°C, followed by 8°C per minute for 25 minutes to 300°C, and finally maintained isothermally at 300°C for ten minutes. Tetramethyl-EDTA is used as an internal and external standard to permit quantitative determination of the various organic constituents in the GC samples. The GC identification of the methylated organic species is confirmed by GC-MS using a Hewlett-Packard 5985 GC-MS instrument with an SE 52 column in the GC, an electron impact ionization source (70 eV) in the MS, and a data system with an extensive library of organic standards.

RESULTS AND DISCUSSION

Water flowed into six of the inert atmosphere sampling wells (W1N, W2N, W2NA, W3N, W3NA, W5N) during the post-construction period. The well locations are shown in Figure 2. Samples were taken and analyzed by radiochemical procedures to obtain radionuclide concentrations. Nonradioactive species were determined by neutron activation analysis using the PNL subcritical neutron multiplier(8). The analytical results are summarized in Table 2 and compared with similar data from waste trench 27 leachate and experimental trench water samples. Data presented in Table 2 will serve as a reference basis for future interpretations of sample measurements from these same locations.

Changes in bromine and chlorine concentrations that have been observed in waste trench 27 leachate since 1978 and in water from the experimental trench and inert atmosphere wells indicate that there is some communication by groundwater flow between these locations. These comparisons are made possible in part by the presence of sodium bromide (NaBr) tracer added to sections 1, 2, 3 and 5 of the experimental trench during its construction in 1979. Comparisons can also be made from analytical data for radionuclides in the same samples.

The first indication of communication by groundwater flow is obtained by comparing bromine concentrations in samples taken since 1978. Bromine concentrations in waste trench 27 leachate were quite low prior to the construction of the experimental trench, when NaBr was added as a tracer.

TABLE II

Inert Atmosphere Wells (9-80), Waste Trench 27 (WFT27) Well Point and Sump (9-80) Compared to Waste Trench 27 Sump (5-78) and Experimental Trench (6-80). These data serve as a reference basis for interpretation of sample analyses from the same locations. For example, tritium and inert tracer concentrations in water from the experimental trench area confirm that subsurface flow has occurred from the experimental trench and toward the waste burial trenches during the time period following trench construction.

	Radionuclides, pCi/ℓ				Nonradioactive Species, ppm						
Location	^{3}H	^{90}Sr	^{238}Pu	$^{239\ 240}$Pu	Br	Cl	Al	Mn	Cu	Na	U
WELL W1N	4.7 + 05	1.5 + 02	37.3	6.04	1	45	<0.4	<0.1	<1.2	123	0.02
WELL W2N	2.9 + 06	<3.3 + 02	20.5	77.0	2	110	0.6	<0.1	<1.4	189	<0.01
WELL W2NA	7.7 + 08	1.7 + 04	235	4.82	188	1340	<1.6	23	<4.2	445	<0.09
WELL W3N	1.2 + 09	3.4 + 04	2409	44.3	815	1200	193	55	<6.6	407	0.3
WELL W3NA	2.0 + 07	<2.4 + 02	23.4	<5	517	517	<1.1	7	<2.4	260	<0.03
WELL W5N	1.0 + 03	2.1 + 02	9.41	<5	5	440	1.6	1.4	<2.3	311	0.04
WFT 27 Well Point (9-80)					2.7	7060	6	3.1	<8	387	118
WFT 27 Sump (9-80)					459	2900	<2.4	19	<6	438	<2
WFT 27 Sump (5-78)					2.8	3300	<3	77	<6	344	1.7
Exp. Trench TIE (6-80)					3500	480	<70	2.9	<50	1100	1.4
Exp. Trench TIW (6-80)					3300	650	<80	11	<50	1100	1.2

Following trench construction, the bromine concentration had increased from 2.8 to 460 ppm, indicating that some of the NaBr tracer had moved into waste trench 27. The well point samples had low concentrations of bromine, probably because the well point was installed opposite section 4 of the experimental trench, which contains no added NaBr. Similar conclusions can be drawn by comparing the chlorine data for these same samples.

An additional indication of communication is obtained by comparing bromine concentrations in the inert atmosphere well samples. They could also be influenced by the presence of NaBr in sections 1, 2 and 3 of the experimental trench. Wells W3N and W3NA show this influence even more markedly than the comparison with waste trench 27, since the wells are much closer to the NaBr source. Well W2NA also indicates a communication with the NaBr source while wells W1N, W2N and W5N seem not to be connected to the NaBr source.

To obtain additional verification that such groundwater flow exists, we compared radionuclide concentrations in water samples taken from two regions of the well construction area. Only W2NA and W3N appeared to be closely related to waste trench 27, possibly connecting to it through subsurface fractures. Radionuclide concentrations were markedly different between wells W2N, W2NA and W2NC and also between W3N and W3NA, yet the W2N region wells and the W3N region wells were only separated from others in their regions by a few meters.

The tritium concentration in waste trench 27 leachate is high due to the large volume of tritium-containing waste buried there and in adjacent waste trenches. Consequently, a useful radioactive tracer has been in place for detecting groundwater movement in the area adjacent to waste trench 27. Tritium concentrations in water from the experimental trench and from the inert atmosphere sampling wells are summarized in Figure 5. The tritium concentrations in samples from trench sections T2 and T3 have shown very marked decreases since the experimental trench was constructed, indicating that there is substantial movement of water away from the trench in this region. The tritium concentration in waste trench 27 leachate has been high and relatively stable during this same time, but the waste trench leachate apparently did not flow toward the experimental trench to maintain the tritium concentrations near the levels that were originally observed in sections T2 and T3. It is unlikely that this degree of dilution could have come from water filtering through the trench caps, since none of the nonradioactive tracers installed in these caps were detected in the experimental trench water. The tritium data and the bromine data support the view that some water flow has occurred away from the experimental trench and toward waste trench 27 during the study period. Water from sections T2 and T3 would therefore be diluted by subsurface lateral flow of low-tritium water moving from the south toward the experimental trench.

Plutonium in waste trench 27 leachate is present in the reduced form as a strongly complexed anionic species. To obtain evidence on the chemical behavior of plutonium complexes with complexing agents likely to be present in Maxey Flats trench waters, laboratory ion exchange experiments were performed with plutonium (+3) and plutonium (+4) and several candidate complexing agents (EDTA, TBP, TOA). The results are summarized in Figures 6 and 7. Among the solutions studied, $10^{-6}\underline{M}$ EDTA had the greatest effect on plutonium absorption, and higher concentrations ($10^{-3}\underline{M}$) of TBP or TOA were needed to produce the same relative effects in anion exchange studies. In cation exchange studies with TBP and TOA, the more dilute solutions produced the greater effect. The EDTA concentration having the greatest effect on plutonium absorption in these experiments is about the concentration measured in waste trench 27 leachatge. The presence of a strong complexing agent like EDTA, even at these low concentrations, may actually be more important than the oxidation state of plutonium in determining whether it absorbs on a material with a high exchange capacity. These experiments support the view that plutonium is present in waste trench leachate as a strong complex, probably with EDTA.

Similar ion exchange experiments were conducted with ^{85}Sr to determine what role complexing agents play in the subsurface migration of ^{90}Sr at Maxey Flats. In these experiments, the effect on anion exchange followed the order EDTA > TBP > TOA > tapwater. Strontium-85 was totally absorbed from all solutions studied on cation exchange columns.

Radiochemical and organic ligand analyses were performed concurrently on concentrated water samples using the analytical sequence summarized in Figure 4 to obtain supporting evidence for the probable chemical species of various radionuclides. Figure 8 presents the data from a typical run, in which the void volume and EDTA distribution curves are superimposed on the corresponding steric exclusion chromatogram. Radiochemical analyses are summarized in the histograms at the base of the figure.

After steric exclusion chromatography with Sephadex G-15, several UV-absorbing peaks appeared, corresponding to column-included, organic-containing species. Plutonium eluted in one region of the UV chromatogram, just prior to ^{90}Sr and ^{137}Cs. EDTA coeluted with plutonium, suggesting that EDTA is complexed with plutonium in the waste trench leachate. On the GC scan, EDTA is easily the major component, accounting for about 37% of the organic material, while NTA and DTPA were not observed.

Preliminary results indicate that a variety of organic acids and other water-soluble, polar compounds co-eluted with ^{90}Sr and ^{137}Cs. Under the large peak around fraction 25 there are two major components, an n-C_{16} acid and an n-C_{18} acid, accompanied by small amounts of dicarboxylic acids. It thus appears that polar organic species may be involved in the mechanism for migration of some radionuclides at the site.

The analytical observations summarized above were performed first on waste trench 27 leachate, and later on water samples from well W2NA. Similar results were obtained from the well samples. Plutonium and EDTA co-eluted, while ^{90}Sr and ^{137}Cs were collected in fractions containing the water-soluble, polar organic compounds. Although the well samples contained additional organic species not found in the waste trench leachate, it is significant that the same radionuclide-organic species associations were found in both types of samples. The results confirm the importance of complexing agents to the mechanism for subsurface migration of radionuclides at Maxey Flats.

The Commonwealth of Kentucky has undertaken a program to stabilize the trench caps at Maxey Flats in order to prevent the infiltration of surface runoff water into the waste-filled trenches. As part of this stabilization program, a significant portion of the site has been covered by flexible plastic material which will ultimately be part of new trench caps designed to eliminate direct infiltration into the trenches. A plastic cover over most of the waste trench adjacent to our experimental trench and inert atmosphere wells may benefit our experiments by severely limiting the direct infiltration of surface water, thereby permitting the extent of subsurface migration to be quantified.

At present, discernible effects of the plastic covers on water levels in the waste-filled trenches have not been observed. We expect a reduction in the volume of trench leachate pumped to the evaporator as the infiltration of surface water is limited by the plastic covers. This should allow a comparison between existing data that were influenced by both vertical and horizontal water movement with future data that will be influenced by predominantly horizontal flow and the experiments will better define the effects of these processes on radionuclide migration.

CONCLUSIONS

EDTA is the major organic complexing component in waste trench 27 leachate, but other polar, water-soluble organics are also present. Plutonium and EDTA coelute during steric exclusion chromatography of Maxey Flats trench and well waters. This correlation offers strong evidence to support the conclusion that plutonium probably migrates as an EDTA complex. Polar organic compounds may influence the migration of

^{90}Sr and ^{137}Cs. Tritium and inert tracer concentrations in water from the experimental trench area confirm that subsurface flow has occurred from the experimental trench and toward the waste burial trenches during the time period of the measurements. Additional measurements in this complicated hydrologic system will help determine if this is the standard flow direction, or an artifact of relatively low rainfall conditions that followed the trench installation. Major increases in water level had occurred in the experimental trench without movement through trench caps, as confirmed by nonradioactive tracer studies.

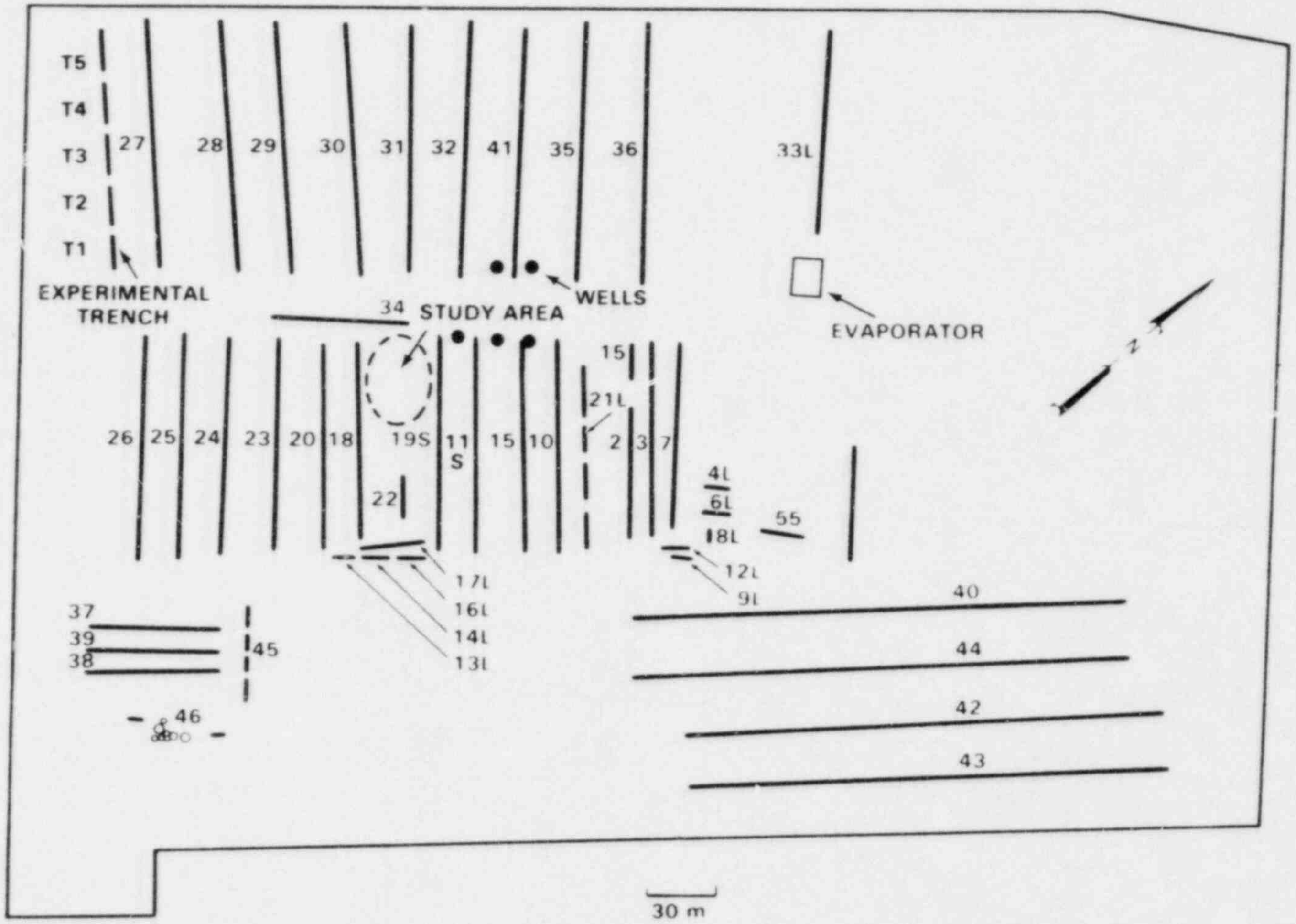


Figure 1. Maxey Flats Waste Trenches and Experimental Areas. The numbered lines show the approximate locations of the waste trenches. The experimental trench area (upper left-hand corner) is shown in greater detail in Figure 2. The study area by waste trench 19S is described in greater detail in Section V of this report. These experimental areas are used by PNL, BNL, LANL and UC-B in research studies at the Maxey Flats shallow land burial site.

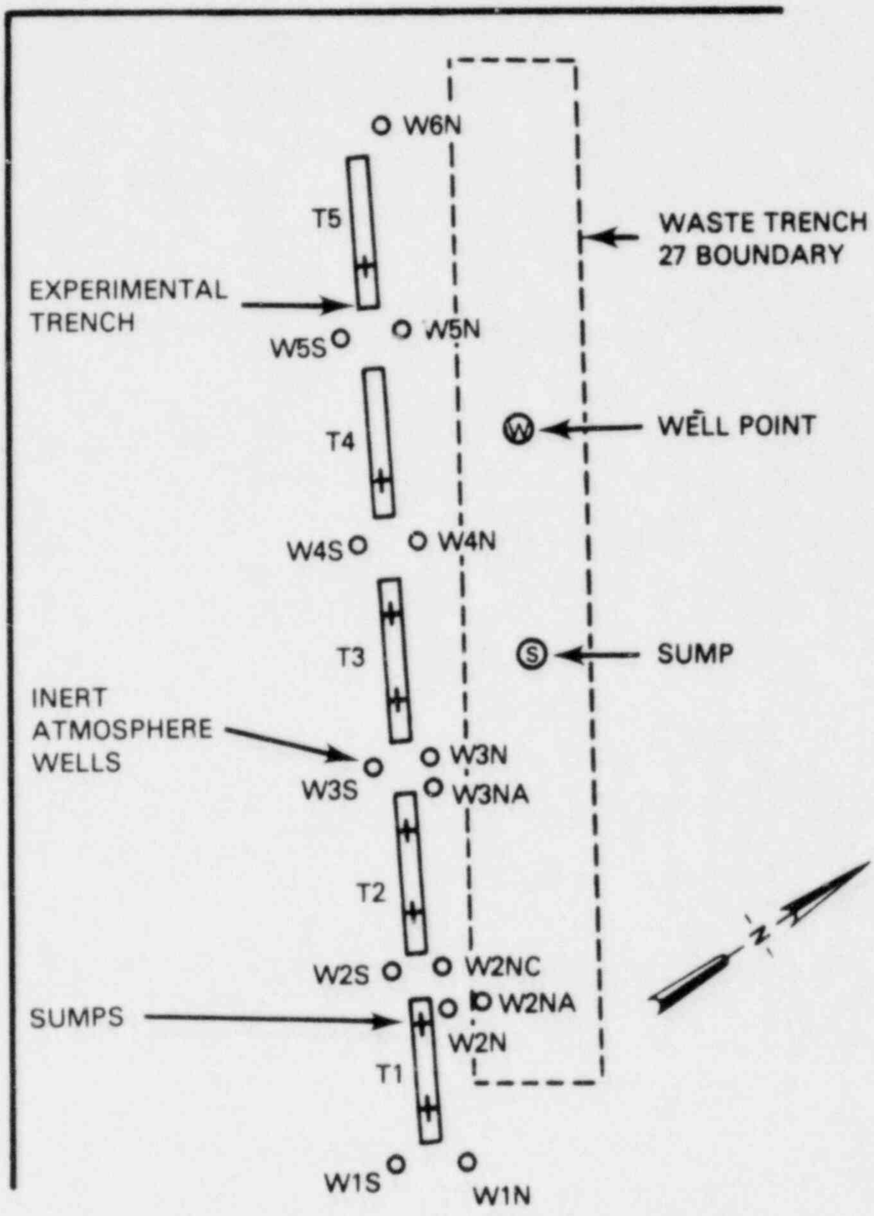


Figure 2. Maxey Flats Experimental Trench and Wells near Waste Trench 27. These facilities allow concurrent sampling of the experimental trench, inert atmosphere wells and adjoining waste trench for groundwater migration and chemical species studies. The experimental trench is also used by BNL, LANL and UC-B in research studies at the Maxey Flats Shallow Land Burial Site.

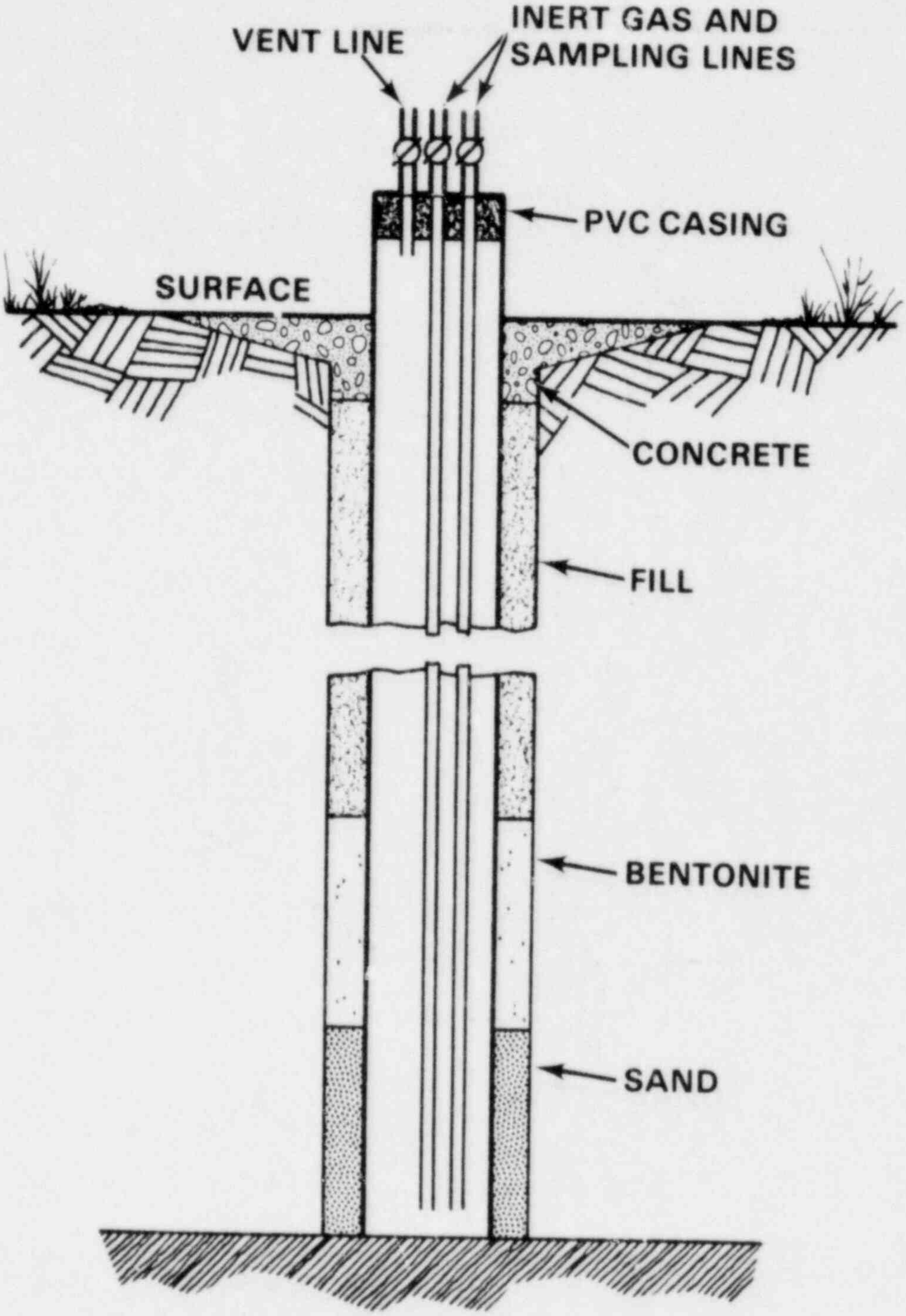
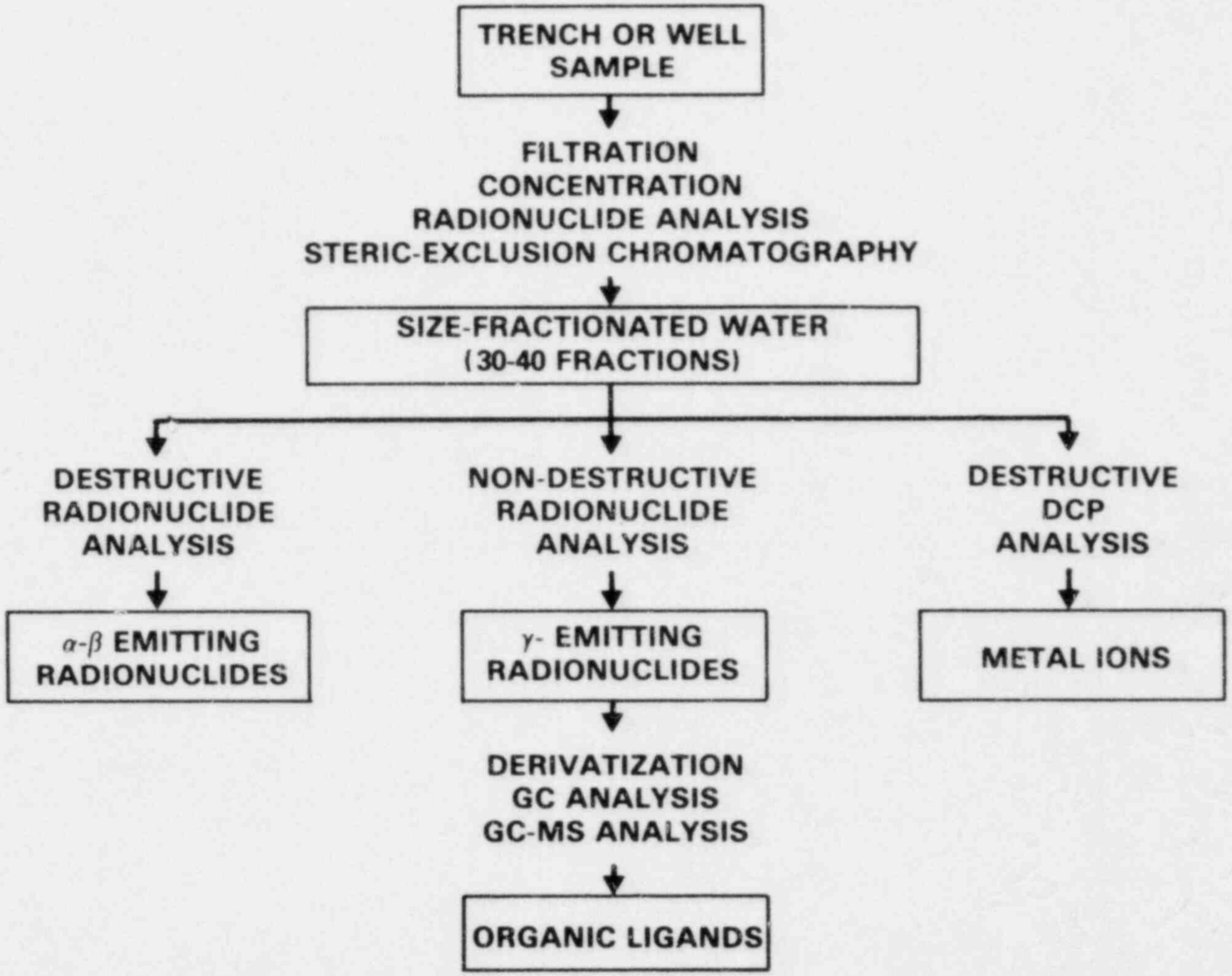


Figure 3. Inert Atmosphere Sampling Well Design. The wells are maintained under an argon atmosphere to assure samples drawn will represent the existing anoxic conditions of the groundwater.

Figure 4.

TRENCH AND WELL WATER ANALYSIS SEQUENCE



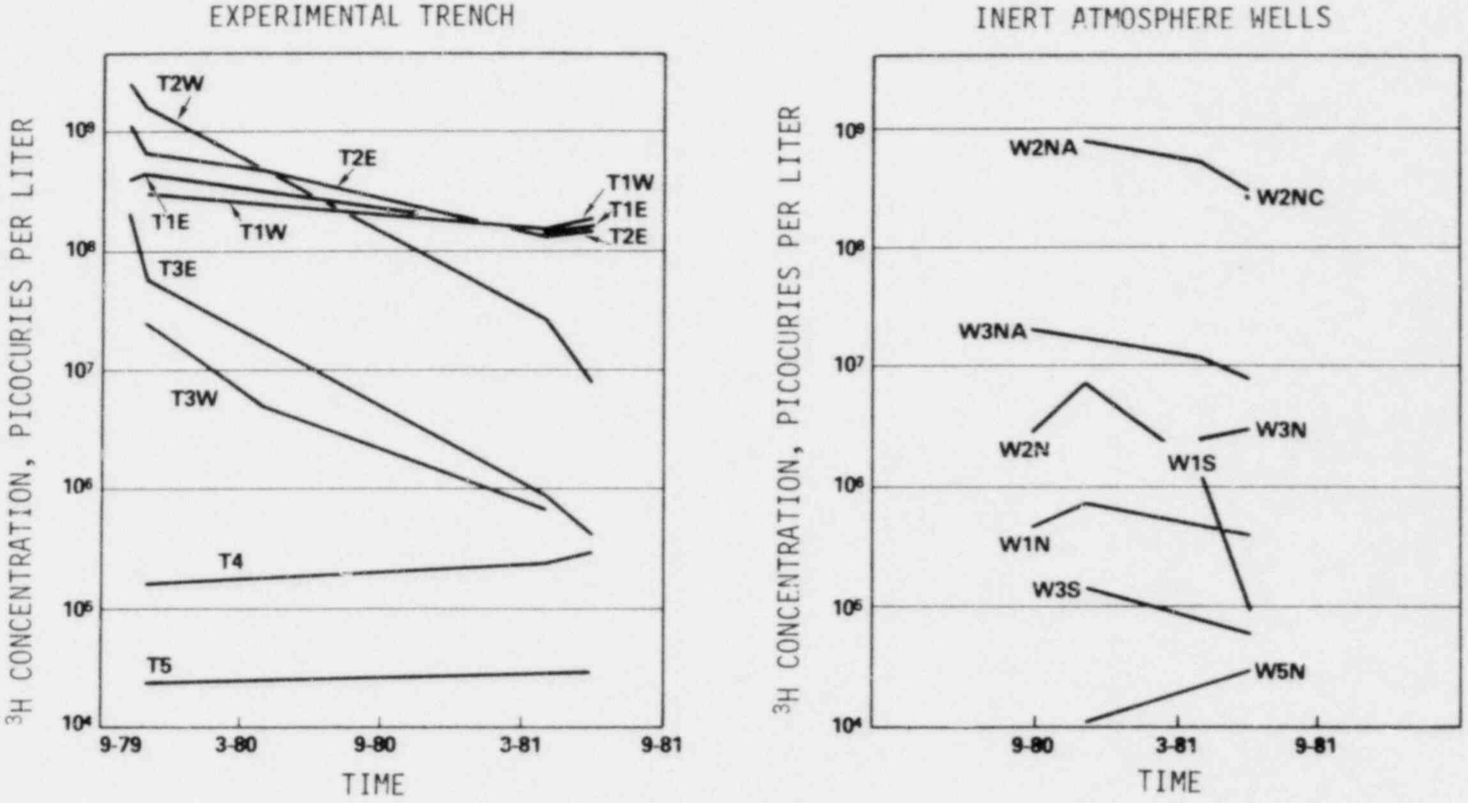


Figure 5. Tritium concentrations in the Experimental Trench and inert atmosphere wells showing changes from 9-79 through 6-81 and 9-80 through 6-81, respectively. Major changes in tritium concentrations in experimental trench sumps T2W, T3E and T3W, when correlated with nonradioactive tracer data on the same samples, indicate that groundwater flow was from the experimental trench and toward waste trench 27 during the indicated time span.

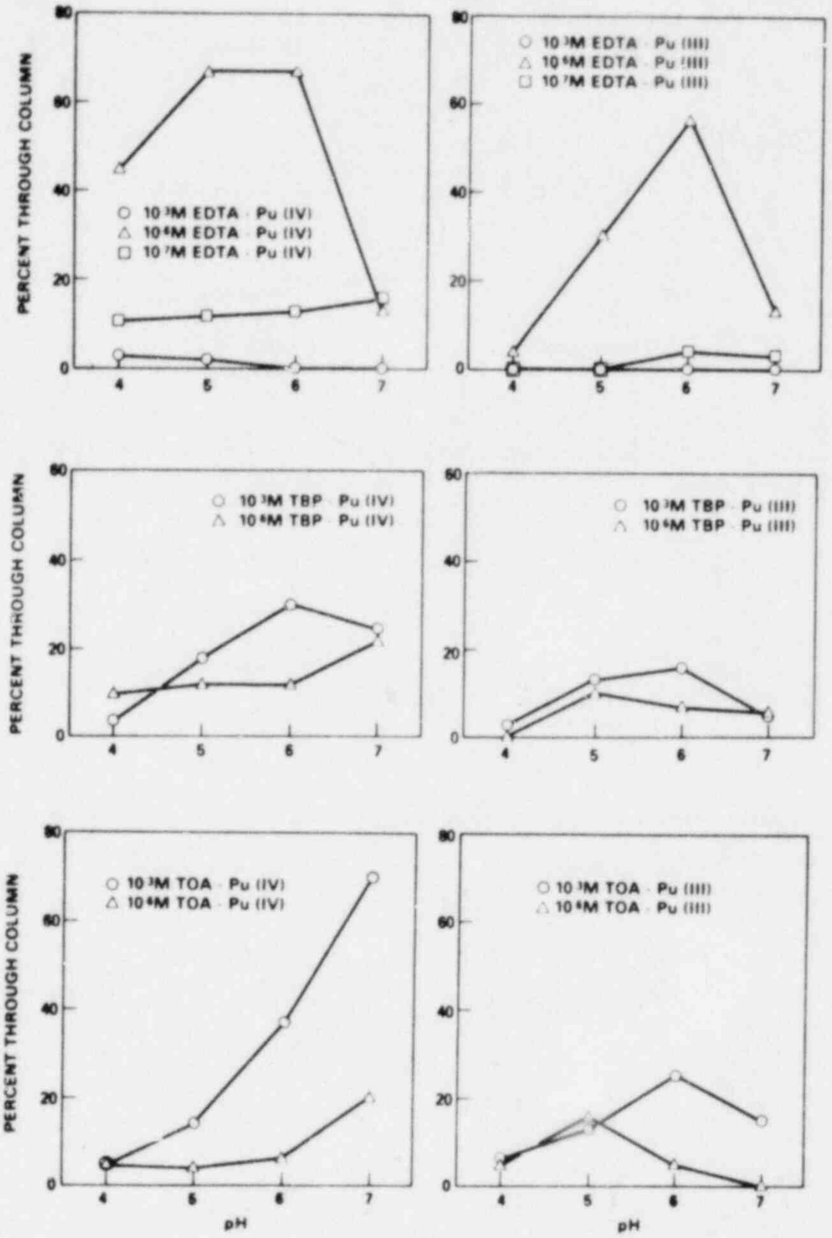


Figure 6. Anion Exchange of Plutonium Solutions with Various Complexing Agents. Laboratory experiments were made to determine the behavior of plutonium complexes with complexing agents likely to be present in Maxey Flats trench waters. The EDTA concentration having the greatest effect on plutonium absorption is approximately the concentration of EDTA in Waste Trench 27 leachate. The experiments show that the presence of a strong complexing agent like EDTA, even at these low concentrations, may actually be more important than the oxidation state of plutonium in determining whether plutonium absorbs on a material with a high exchange capacity.

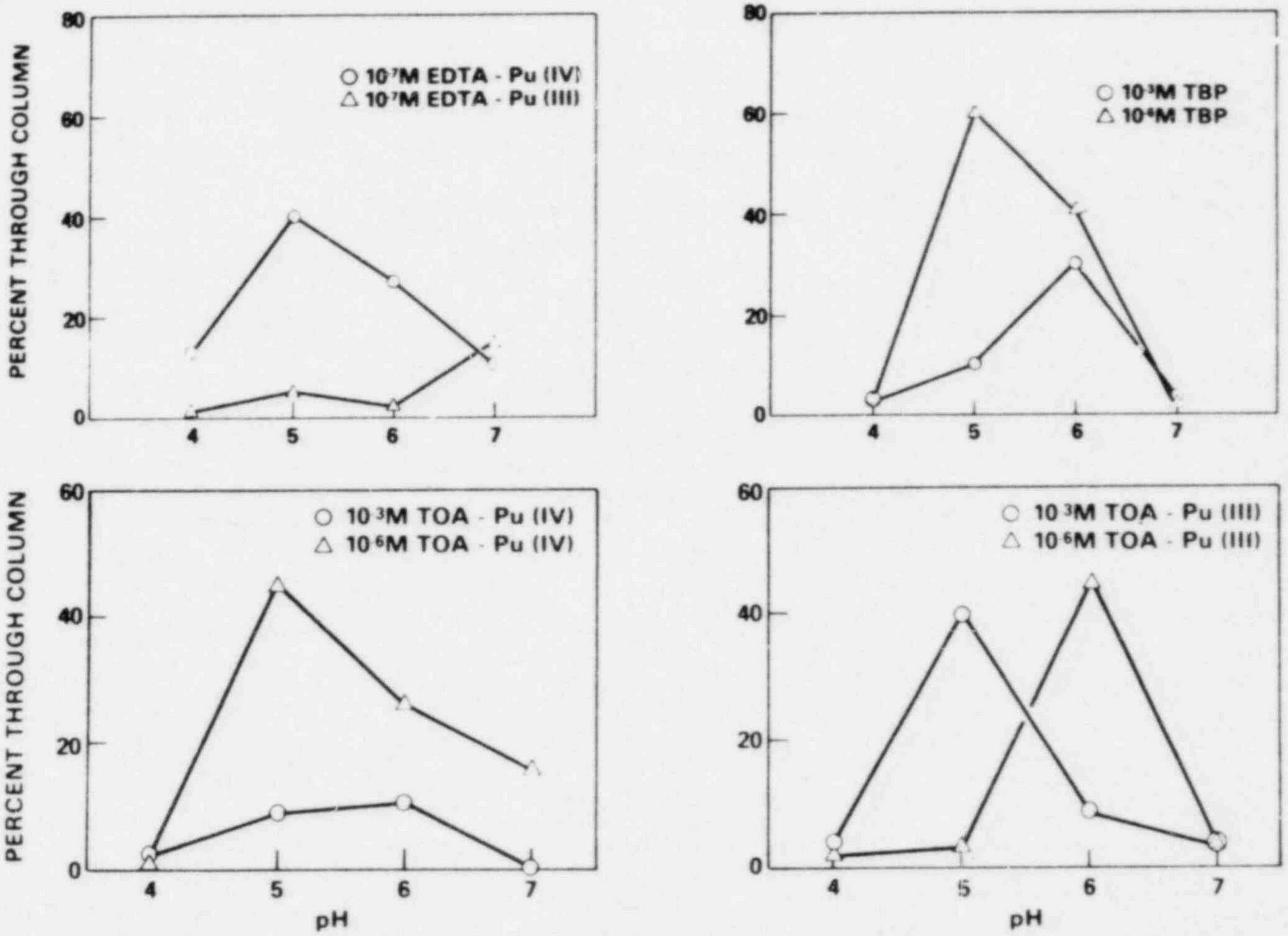
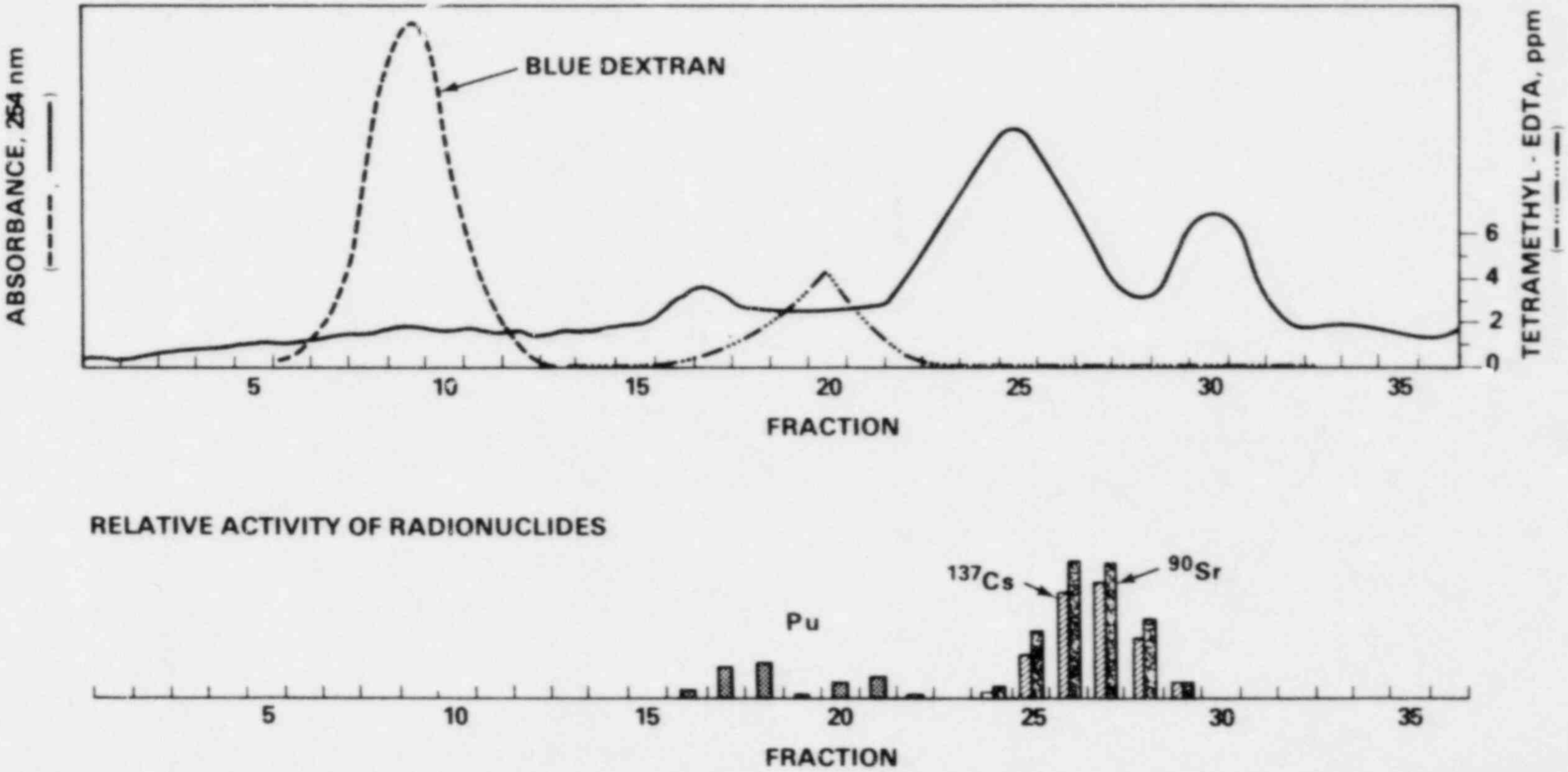


Figure 7. Cation Exchange of Plutonium Solutions with Various Complexing Agents. These laboratory studies together with the anion exchange studies summarized in Figure 6 show that the presence of a strong complexing agent like EDTA, even at these low concentrations, may actually be more important than the oxidation state of plutonium in determining whether plutonium absorbs on a material with a high exchange capacity.

Figure 8.

STERIC EXCLUSION CHROMATOGRAPHY (SEPHADEX G-15) FRACTIONATION OF MAXEY FLATS WASTE TRENCH LEACHATE (10-FOLD CONCENTRATE)



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SECTION II

AREAL DISTRIBUTION OF RADIONUCLIDES AT MAXEY FLATS

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AREAL DISTRIBUTION OF RADIONUCLIDES AT MAXEY FLATS

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SUMMARY

High resolution gamma-ray spectrometry was used by PNL to determine radioactivity levels in surface soil at the Maxey Flats, Kentucky shallow land burial site. In situ gamma-ray measurements made within the restricted area and in the drainages adjoining the site were compared with soil sample analyses and confirm that surface contamination has been retained largely within the restricted area. Cobalt-60 concentrations within the west drainage channel, in the first 100 meters west of the fence, are above the fallout levels measured farther below the site. In the south and east drainage channels, ^{60}Co concentrations in surface soil exceed fallout levels at a few locations adjacent to the fence. Soil sample analyses confirm that this radioactivity is contained in the top few centimeters of soil. Cesium-137 concentrations measured in the drainage channels are comparable to fallout levels in other high rainfall areas of the United States.

INTRODUCTION

The Maxey Flats site has been contaminated by several sources and the potential exists for a general low-level contamination of the restricted area and main drainage channels adjacent to the site. Possible sources of contamination include the upward seepage of water from the burial trenches as rainwater infiltrated and filled them, spillage during pumping of excess water from the trenches, fallout and washout of entrained radionuclides in the evaporator plume, and the malicious release of radioactive solutions from a containment vessel.

Movement of radionuclides within or from the site may result from surface water runoff, dispersal from the evaporator plume, vertical and lateral movements through the soil or movement through cracks or joints in the underlying rock strata, and biotic pathways through vegetation growing on and adjacent to the trenches. Such movements would produce a radionuclide distribution which is unique to the site and its surrounding terrain.

Radiation mapping studies conducted by Dames and Moore for the Commonwealth of Kentucky(1) determined the approximate extent of areas exceeding 25 μR/hr exposure rates. Selected soil samples from these areas were analyzed for ^{60}Co, ^{90}Sr, ^{137}Cs and ^{238,239}Pu. Radiation mapping was shown to be an effective screening procedure to locate areas of soil contamination. The measurements demonstrated that significant soil contamination was confined to the location immediately adjacent to the restricted area, but emphasized the relative importance of contaminated runoff to offsite releases of radionuclides.

High resolution gamma-ray spectrometry was used by PNL to determine radioactivity levels in surface soil at the Beatty, Nevada shallow land burial site as part of an effort to confirm the boundaries of existing waste burial trenches, locate any additional radioactive wastes beyond the established burial area, characterize the distribution of radionuclides around the waste burial site, and determine whether movement of radioactivity from unearthed waste drums had occurred(2). The in situ counting data were compared with analyses for soil samples taken at each counting location. Up to twenty-four gamma-emitting radionuclides were measured. From these measurements we determined where soil mixing had occurred and where surface depositions of waste radionuclides had taken place. These techniques proved to be very valuable in confirming the integrity of the site while also pointing out a few specific potential problem areas.

To obtain a better understanding of the actual distribution of radioactivity in surface soils and vegetation on the Maxey Flats shallow land burial site and adjacent areas, one task of the PNL research project is to determine the actual spectrum of radionuclides on and adjacent to the site. In situ gamma-ray spectrometry, combined with soil sample analysis by Ge(Li) coincidence/anticoincidence counting systems for gamma-emitting radionuclides and destructive analysis to determine ^{90}Sr, ^{238,239,240}Pu and ^{241}Am, will provide detailed information that is not obtainable by other techniques. The soil radionuclide concentrations will be related to the concentrations of these radionuclides in runoff water and to the MPC's for those radionuclides. The results of this study will help determine whether subsurface migration, as well as surface runoff, is responsible for radionuclide distributions at the site. Possible seepage locations in the drainages will be located to help determine any influence that the Maxey Flats site has had on radioactivity levels in the surrounding countryside.

EXPERIMENTAL METHODS

In Situ Gamma-Ray Spectrometry

The in situ gamma-ray spectrometry system, which is similar to systems used in previous research by Pacific Northwest Laboratory(2,3,4), is illustrated in Figure 1. It consists of a Princeton Gamma-Tech coaxial intrinsic germanium diode coupled to a Canberra Series 80 multichannel analyzer and a cassette data recorder. A portable 1.5 kW Honda generator supplies the electrical power. The detector is attached to a tripod stand and can be positioned from ground level to a height of 1 meter. All field measurements at the Maxey Flats site were made with the detector 1 meter above ground level. Signal and power cables 30 meters long connect the detector to the analyzer, which is transported in a vehicle. The liquid nitrogen reservoir for the detector system has sufficient capacity to permit at least 12 hours of continuous operation.

For the measurements in the drainages adjacent to the restricted area, a portable backpack gamma-ray spectrometer system was used. The system consists of a Princeton Gamma-Tech coaxial intrinsic germanium diode coupled to a Davidson Model 4106A multichannel analyzer. Data acquisition is completed by memory transfer to a magnetic tape with a portable cassette recorder. This electronic package, together with a 140 volt d.c. Globe gel/cell battery pack, is mounted on a specially adapted backpack frame and the detector is suspended from a modified photographer's tripod.

The gamma-ray spectrometry systems are capable of measuring a wide variety of gamma-emitting radionuclides. The radionuclides measured include the naturally-occurring radionuclides ^{40}K, ^{208}Tl, ^{212}Pb, ^{214}Pb, ^{212}Bi, ^{214}Bi, and ^{228}Ac, as well as ^{60}Co and ^{137}Cs. Thorium-228 is determined from its daughter concentrations. Several factors determine the length of the counts, including the concentrations and decay schemes of the radionuclides present and the desired counting statistics. A major factor affecting the field measurements is the time that can be allotted for data acquisition, which is dependent on restrictions imposed by site operating schedules and certain types of activities being conducted on site. At Maxey Flats, limits imposed by operating schedules were not at all restrictive. Weather, on the other hand, frequently terminated counting acativities because of the need to operate the electronic systems in clean, dry air.

Calibration of the counting systems was made by comparing the disintegration rates of the naturally-occurring radionuclides in the soil with the count rates for the same radionuclides at each in situ counting location. By plotting the ratios of the in situ count rates to soil disintegration rates versus gamma-ray energy, calibration factors were derived for ^{137}Cs and ^{60}Co. The calibration factors so derived depend on the assumption that all the radionuclides are uniformly dispersed throughout the soil. All data reduction was accomplished in the laboratory using a PDP 11/44 computer and software developed for these analyses.

In situ gamma-ray spectrometric measurements were conducted within and adjacent to the restricted area at Maxey Flats in May, 1980. The counting locations are summarized in Figure 2. Wherever possible a twenty-meter grid spacing was used within the site and along the fence and a few additional measurements were made at possible seepage areas on the west, south and east sides of the site. The blank spaces shown on Figure 2 indicate that no measurements were made due to the presence of buildings or roads, or because some trench cap stabilization work was being performed at the time. In situ gamma-ray spectrometric measurements were performed outside the restricted area and in the main drainage channels leading from the site in May, 1981. The counting locations are summarized in Figure 3.

Soil Sample Analyses

Soil samples were collected from each location where a count was made and returned to the laboratory for analysis. Samples were taken at the site surface at counting locations within the restricted area. Two soil samples were taken wherever possible at each counting location in the main drainage channels leading from the site, one at the surface and a second sample 15 cm below the surface. Analysis of the soil samples for up to twenty-four gamma-emitting radionuclides (e.g., ^{40}K, ^{208}Tl, ^{212}Pb, ^{214}Pb, ^{212}Bi, ^{214}Bi, ^{228}Ac, ^{228}Th, ^{60}Co and ^{137}Cs) was performed with a Ge(Li) coincidence/anticoincidence counting system at PNL facilities in Richland, Washington. Destructive analysis is continuing to determine the levels of ^{90}Sr, $^{238,239,240}Pu$ and ^{241}Am in selected soil samples using laboratory procedures similar to those developed for studies conducted for the Electric Power Research Institute(5).

RESULTS AND DISCUSSION

Two general kinds of radionuclides were determined by these measurements. They are endogenous (natural) radionuclides originating from the parent soil material and exogenous (man-induced) radionuclides originating from the nuclear fuel process. Cobalt-60 and ^{137}Cs are exogenous radionuclides and both are constituents of global fallout as well as being components of low level radioactive wastes stored at Maxey Flats. Potassium-40 and ^{228}Th are endogenous radionuclides originating from the underlying rock strata and soil. A typical gamma-ray spectrum recorded during the gamma-ray spectrometric measurements at Maxey Flats is shown in Figure 4. Most of the peaks shown in the spectrum are due to endogenous radionuclides, chiefly natural uranium, thorium, and their daughters, plus ^{40}K. Very low levels of radionuclides were measured in most soil samples. Cesium-137 and ^{60}Co were the most abundant radionuclides of exogenous origin and these have their origin in global fallout, which is ubiquitously distributed over all land surfaces. In the spectrum illustrated, ^{137}Cs (0.662 MeV) and ^{60}Co (1.174 and 1.332 MeV) are present in relatively low concentrations.

The distribution of ^{60}Co in surface soils within and adjacent to the restricted area at Maxey Flats in May, 1980 is plotted in Figure 5. The data have been separated into three levels of ^{60}Co soil concentrations-greater than 2 pCi ^{60}Co per gram of soil, 1-2 pCi ^{60}Co per gram of soil, and less than 1 pCi ^{60}Co per gram of soil. A similar plot for ^{137}Cs distribution in surface soils within and adjacent to the restricted area at Maxey Flats is shown in Figure 6.

The above average ^{60}Co and ^{137}Cs concentrations measured at a few locations within the restricted area are probably due to operating and maintenance activities, but a clear pattern has not emerged from which to predict potentially contaminated surface areas. In general, a distribution pattern for any one radionuclide may not be used to predict a distribution pattern for another radionuclide, except in those instances relating to the purely endogenous radionuclides originating from the parent soil material. Even these latter distributions may be skewed by the presence of specific processing wastes from thorium or uranium purification processes because of the introduction of non-equilibrium quantities of their daughters from the decay chains.

The ^{60}Co concentrations measured by in situ counting procedures near the southeast corner of the site (within the solid contours on Figure 5) do not correlate with the ^{60}Co concentrations in soil samples from these counting locations. Concentrations indicated by in situ counting were higher than those measured in the soil samples, possibly because the trench cap does not shield all of the radiation from the waste buried there. The highly sensitive gamma-ray spectrometers will respond to this condition. Soil samples from other counting locations within the restricted area showed a much closer correlation with the in situ measurements. These results underscore the need to secure soil samples concurrently with the in situ counts, especially where the possibility exists for a large external (in this case, buried) radiation source to influence the count rates.

The soil in the west drainage channel leading from the restricted area contains surface concentrations of ^{60}Co that exceed the background fallout levels away from the site. Figure 7 shows an expanded view of the top 100 meters of this drainage channel with isopleths indicating the concentration ranges. All counts made downhill from the coordinates N330, W540 showed the ^{60}Co concentration to be at the fallout level measured at background locations away from the site (less than 0.1 pCi ^{60}Co per gram of soil). In the south and east drainage channels ^{60}Co concentrations in surface soil exceed these fallout levels in a few locations adjacent to the fence, but concentrations within the drainages are at the background levels.

Soil sample analyses correlated very well with the in situ measurements at all locations in the drainage channels. Virtually all of the ^{60}Co is contained in the first few centimeters of soil. This is true of the purely fallout material measured in background locations away from the site and at all locations where ^{60}Co concentrations exceeded fallout levels within the drainage channels. A release of radioactive solutions over the hill on the west side of the site is said to have occurred during the time the site was operating as a burial ground, and this could account for the ^{60}Co concentrations in the west channel. Surface runoff can also contribute to these concentrations, and surface runoff is probably the cause of the ^{60}Co distribution near the fence in the south and east drainage channels.

Cesium-137 concentrations in surface soil within the restricted area and in the main drainage channels leading from the site are similar to concentrations found in other locations of the United States that have rainfall patterns like those of eastern Kentucky. Although it is apparent that surface runoff from the site could contribute ^{137}Cs to the adjacent drainages, in situ measurements and soil sample analyses indicate that ^{137}Cs is not derived from the Maxey Flats site. The final report will address this issue, as well as the influence of the Maxey Flats site on ^{3}H, ^{90}Sr, ^{238,239,240}Pu and ^{241}Am areal distributions.

CONCLUSIONS

In situ counting measurements and soil sampling performed in the main drainage channels leading from the site confirm that contamination is slight, and only ^{60}Co seems to have been derived from the site. Elevated ^{60}Co levels were measured at the tops of the east and south drainage channels next to the fenced area, but concentrations were comparable to global fallout levels in the hollows below. Elevated ^{60}Co concentrations were measured in the west drainage channel well down the hill, and the soil sample analyses confirmed that this activity is surface-deposited. The ^{60}Co concentrations on the west side of the site approach global fallout levels about 100 meters from the site, toward Drip Springs Creek. Cesium-137 concentrations measured at all locations by in situ counting and by soil sample analysis were comparable to global fallout levels.

Results of the areal distribution measurements thus far confirm that contamination has been largely contained within the restricted area at

Maxey Flats, in agreement with earlier observations by Dames and Moore(1). Site remedial and repair work have resulted in a general redistribution of surface material on some portions of the restricted area, with the corresponding decrease or outright removal of previously existing locations of greater than average radioactive content. Consequently, the trend has been toward lower radionuclide surface distributions and diffuse patterns of distribution.

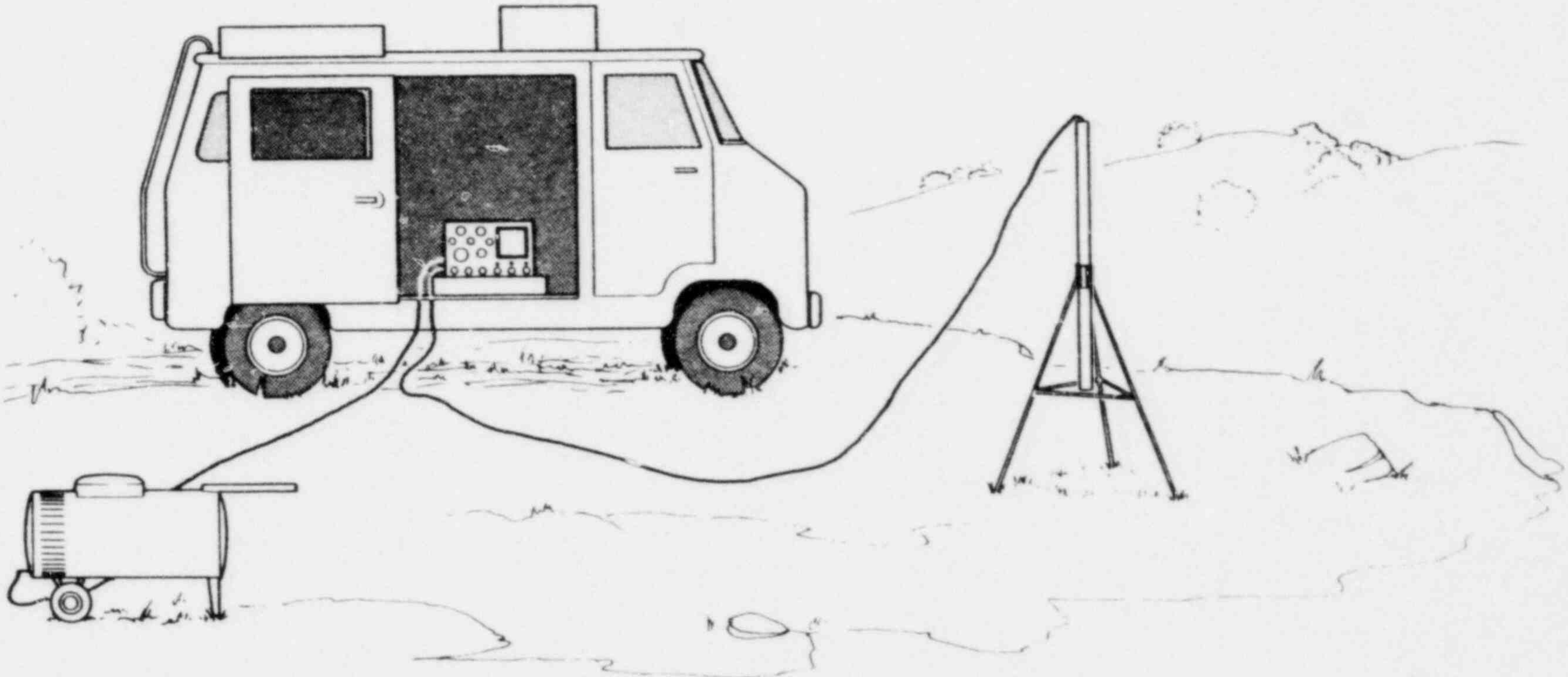

Figure 1. Artist's Concept of High Resolution Gamma-Ray Spectrometer for Field Measurements. An intrinsic germanium diode is coupled to a multichannel analyzer and a cassette data recorder. A portable 1.5 kw generator supplies the electrical power. The detector can be positioned from ground level to a height of 1 meter. The entire system and liquid nitrogen supply are transported in a vehicle.

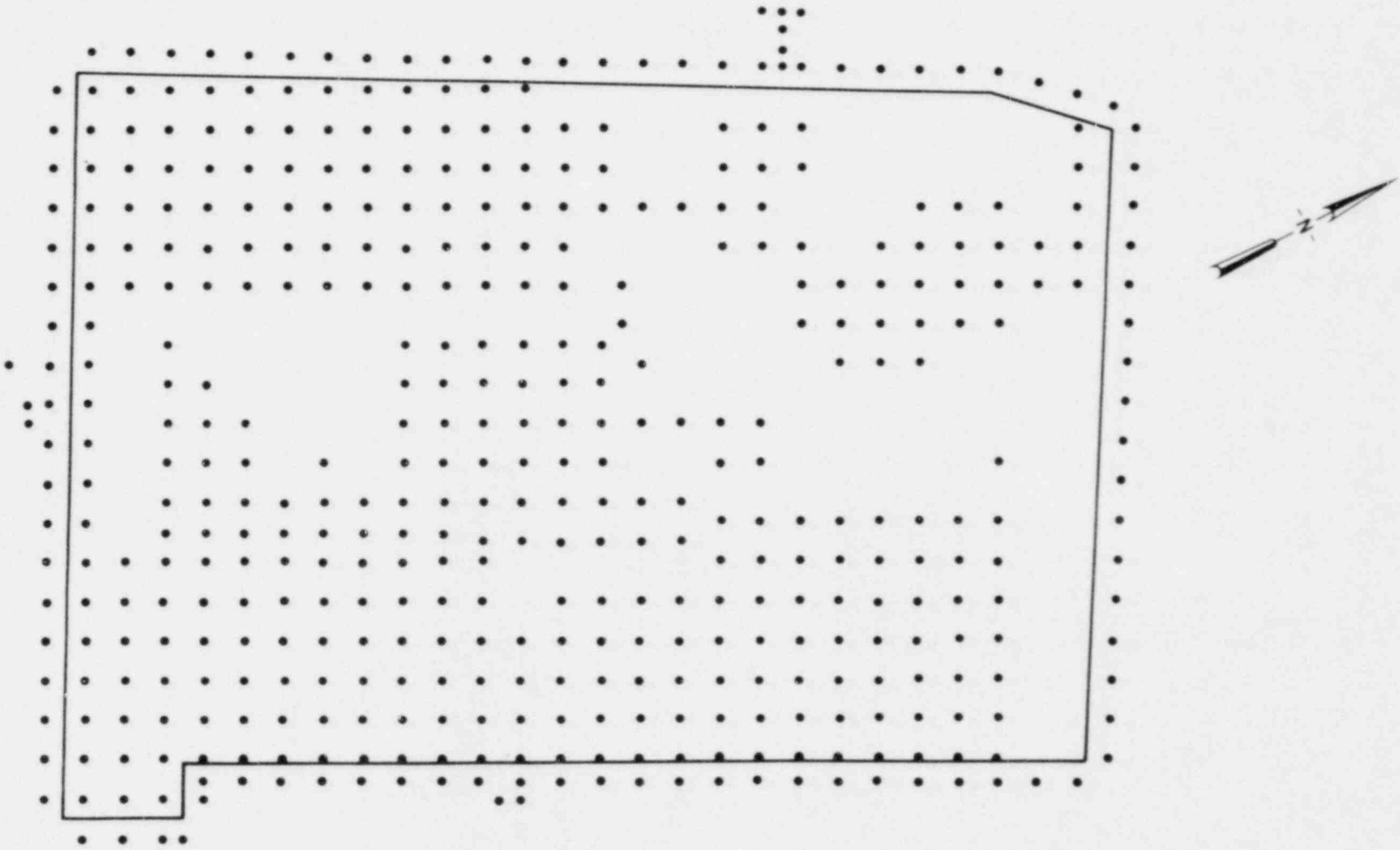

Figure 2. In Situ Gamma-Ray Counting Locations Within and Adjacent to the Restricted Area at Maxey Flats. Wherever possible, a twenty meter grid spacing was used and some additional measurements were made at possible seepage areas.

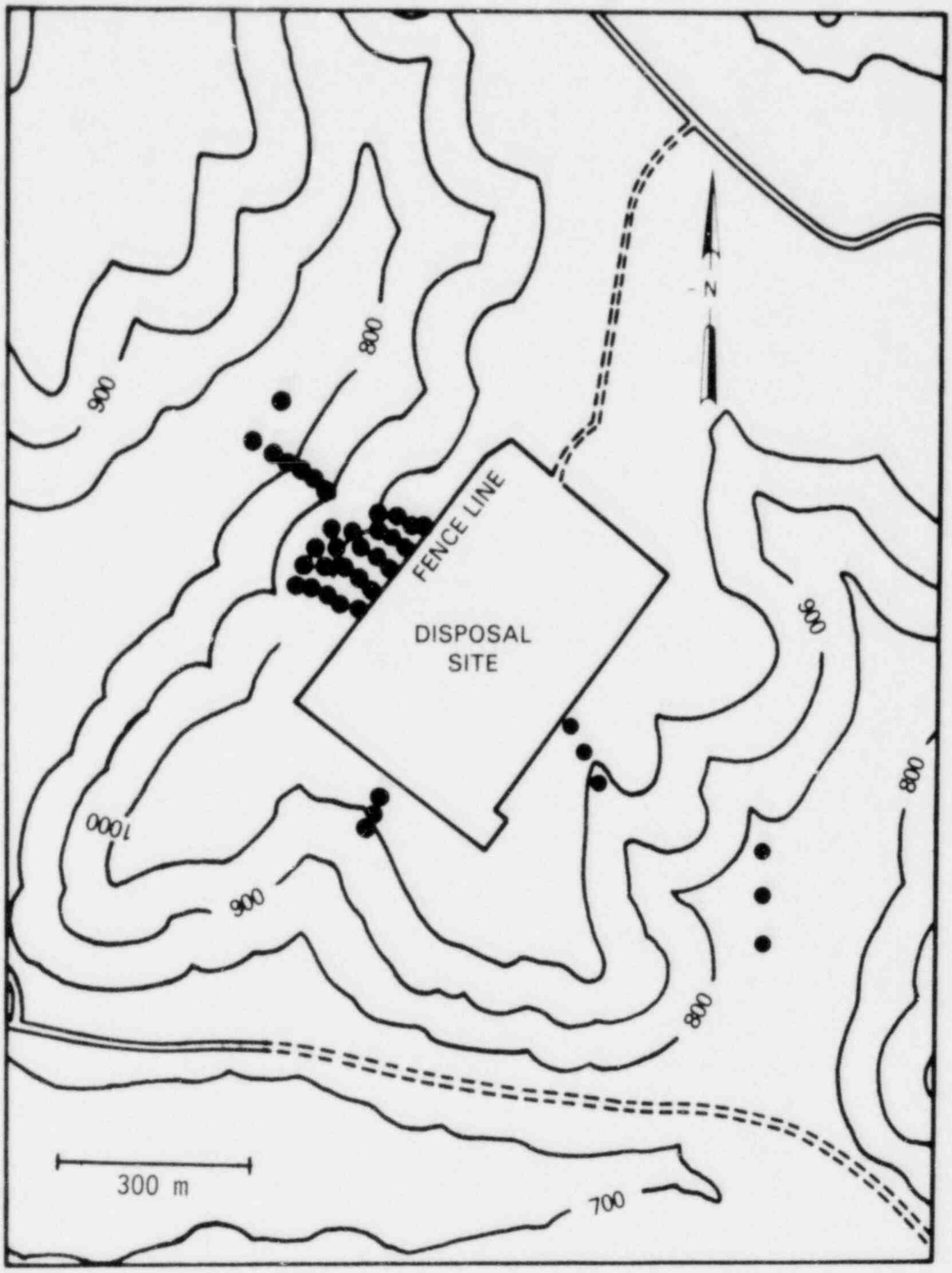

Figure 3. In Situ Gamma-Ray Counting Locations in the Main Drainage Channels Adjacent to the Restricted Area at Maxey Flats. Elevations indicated are in feet.

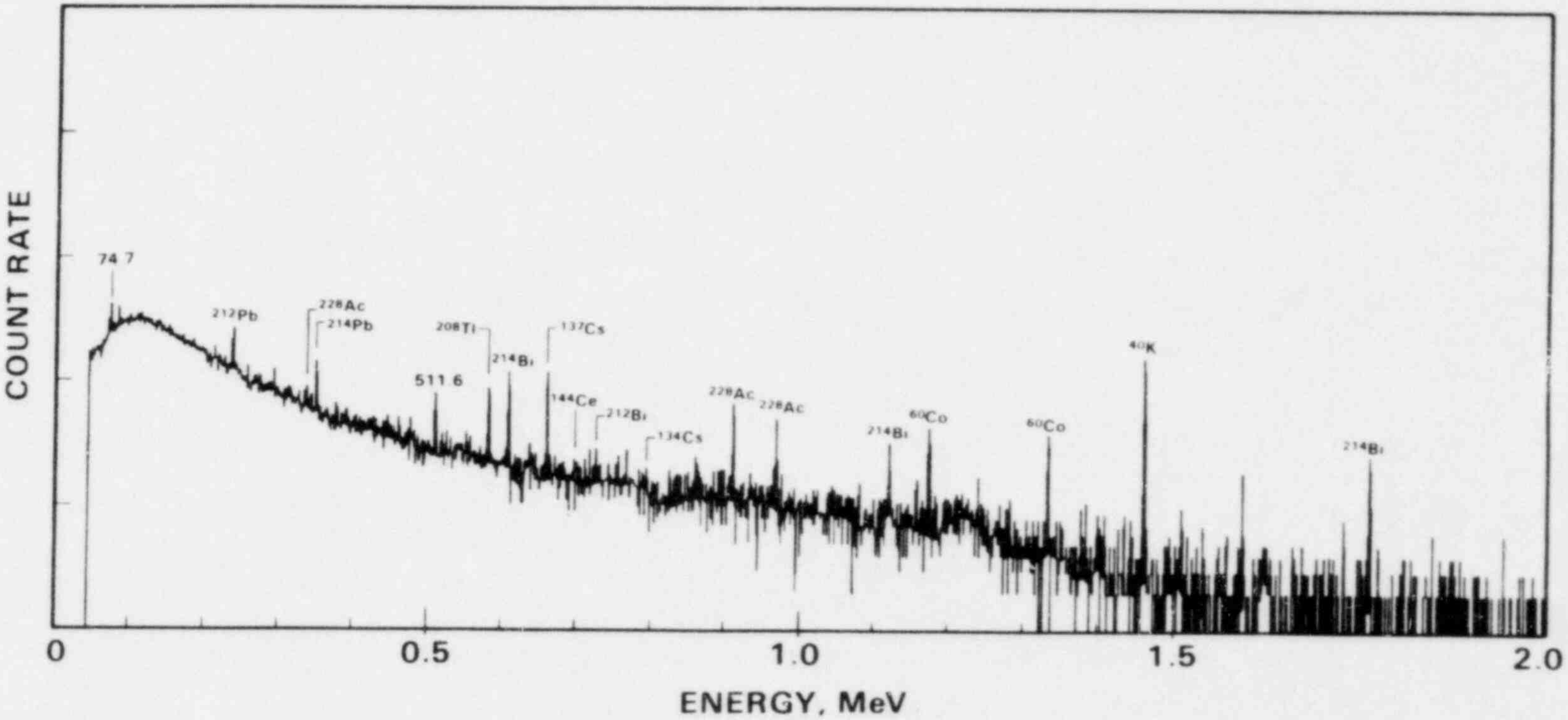

Figure 4. In Situ Gamma-Ray Spectrum, Maxey Flats, KY. Most of the gamma-ray peaks observed are due to endogenous radionuclides originating from the parent soil material. In the example above, the exogenous radionuclides ^{60}Co and ^{137}Cs are also present in low concentrations.

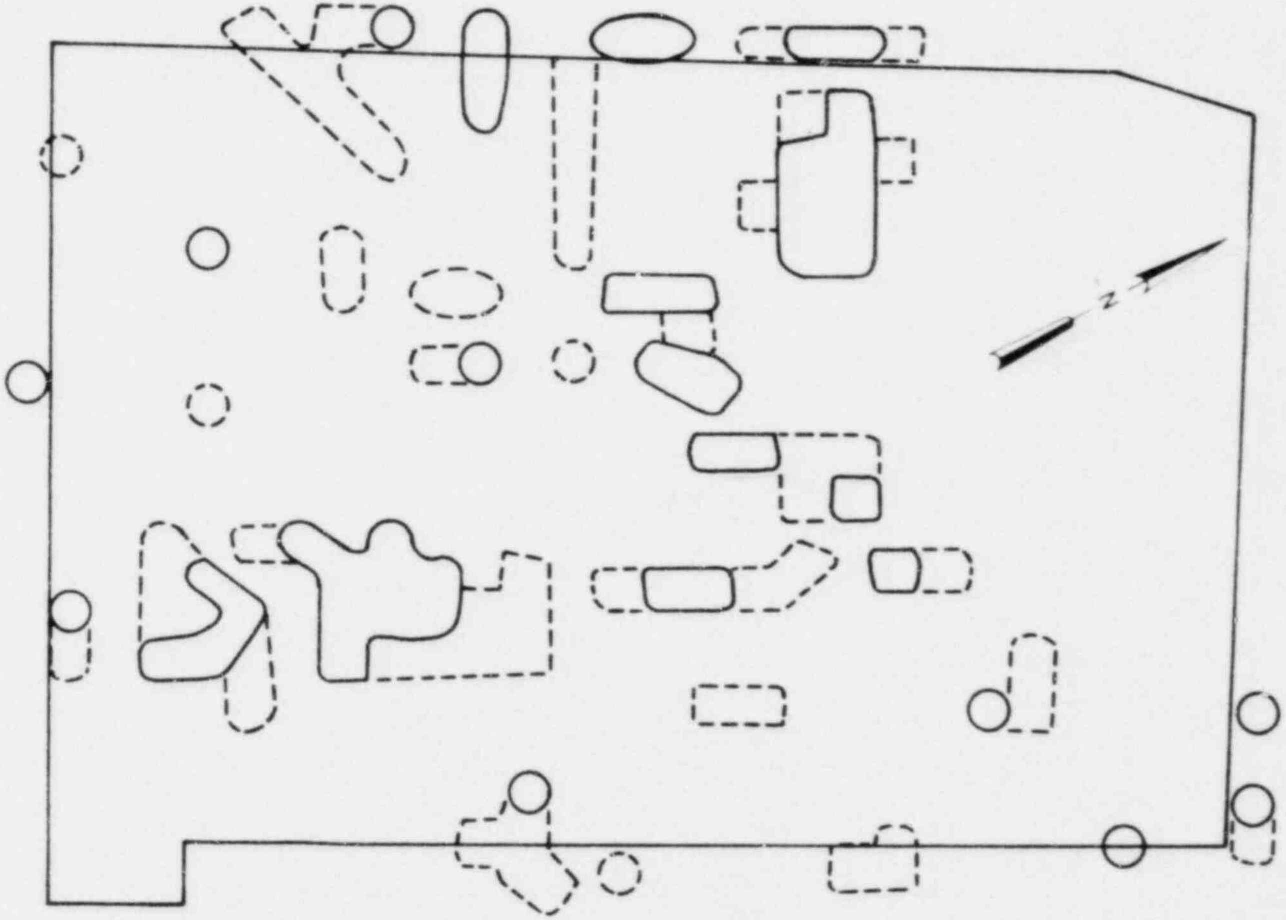

Figure 5. Cobalt-60 Distribution in Surface Soil Within and Adjacent to the Restricted Area at Maxey Flats (May, 1980). Areas within the solid isopleths have ^{60}Co concentrations > 2 pCi per gram of soil. Areas within the dotted isopleths have ^{60}Co concentrations 1-2 pCi per gram of soil. All other areas on site have ^{60}Co concentrations < 1 pCi per gram of soil.

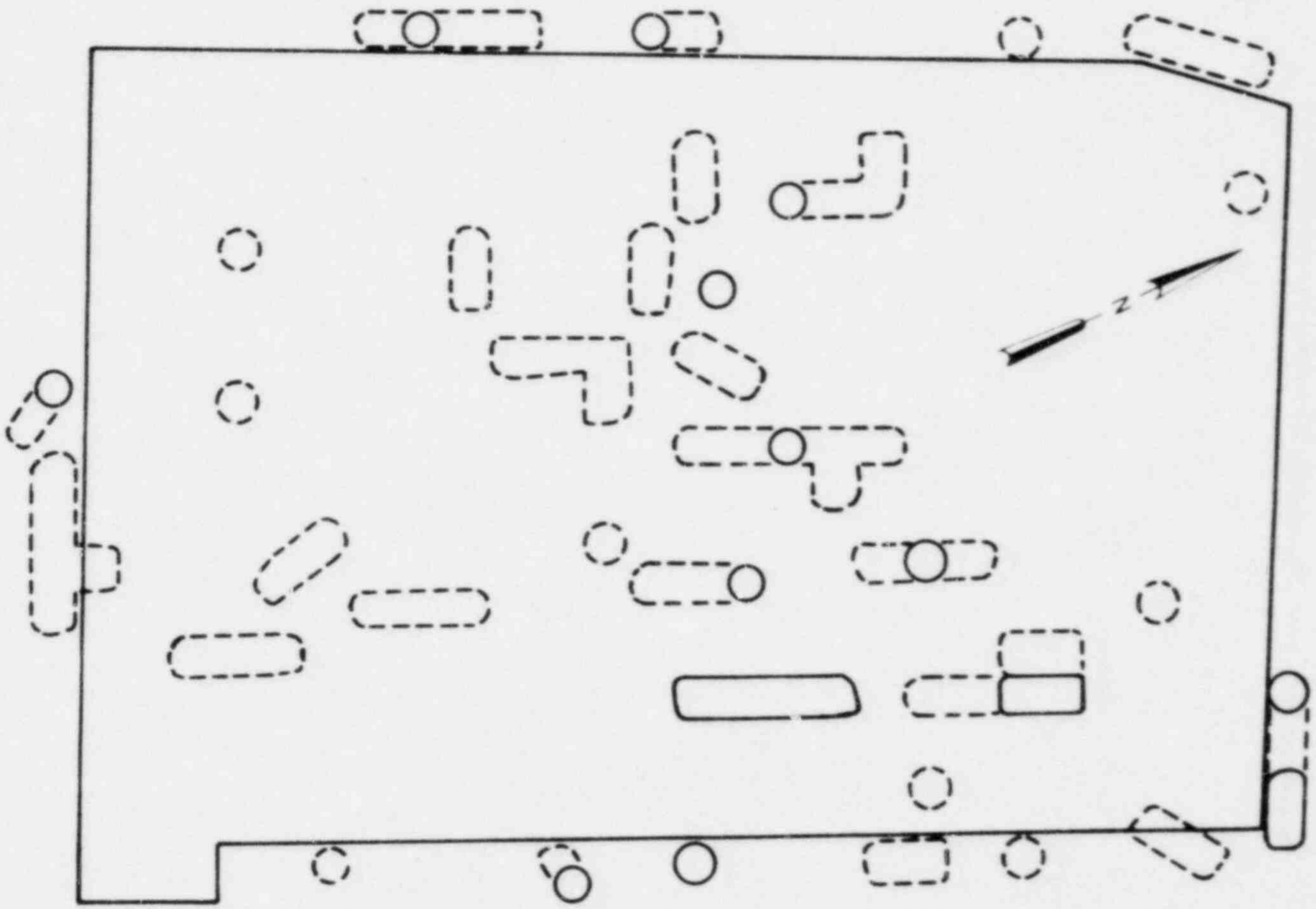

Figure 6. Cesium-137 Distribution in Surface Soil Within and Adjacent to the Restricted Area at Maxey Flats (May, 1980). Areas within the solid isopleths have ^{137}Cs concentrations > 2 pCi per gram of soil. Areas within the dotted isopleths have ^{137}Cs concentrations 1-2 pCi per gram of soil. All other areas on site have ^{137}Cs concentrations < 1 pCi per gram of soil.

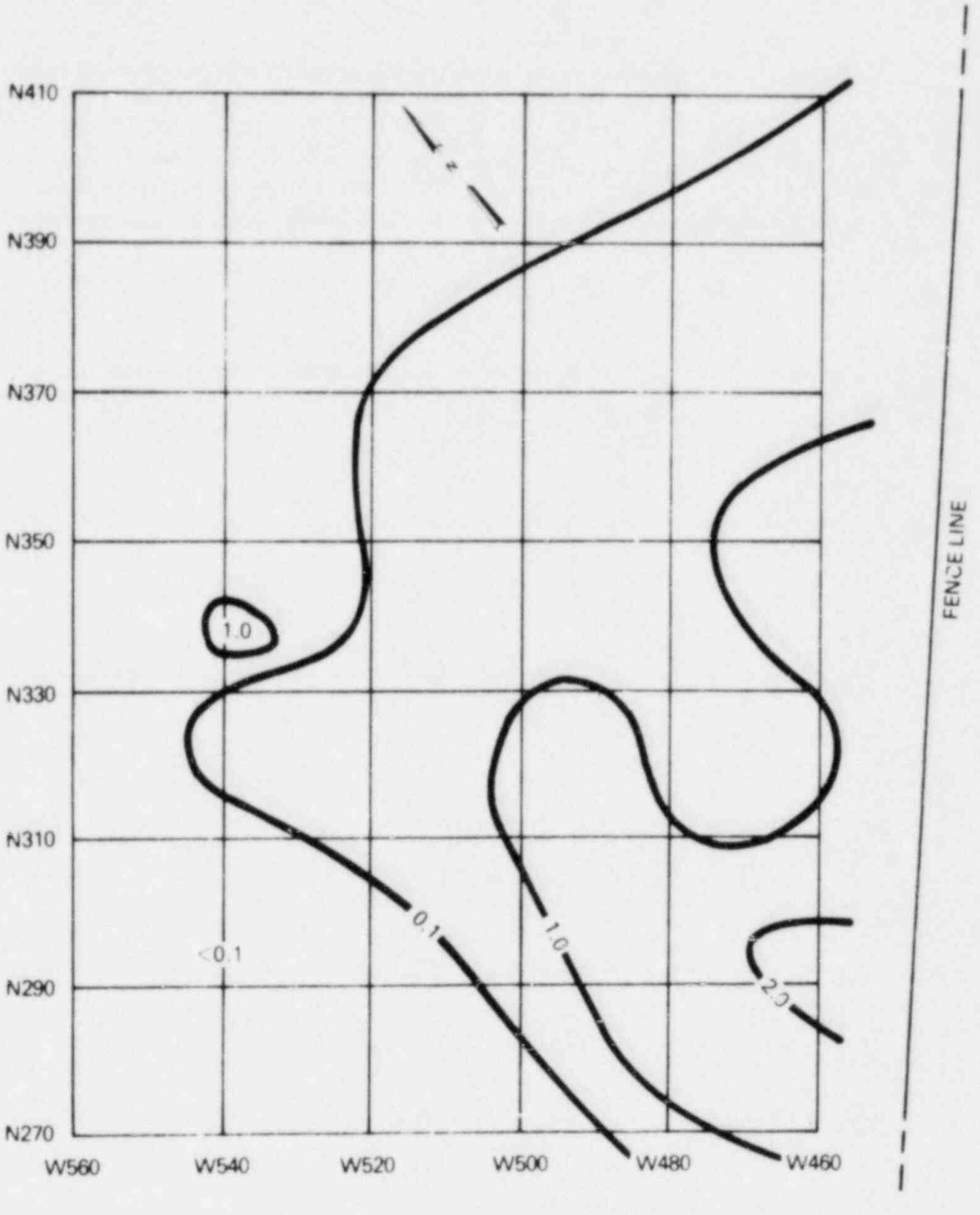

Figure 7. Cobalt-60 distribution in surface soils within the west drainage channel adjacent to the restricted area at Maxey Flats (May, 1981). The main channel follows approximately along the coordinate N330. The heavy isopleths indicate the approximate extent of the ^{60}Co surface concentrations indicated in pCi per gram of soil. Except for one localized area having ^{60}Co soil concentrations 1-2 pCi per gram of soil at about N335, W540, the ^{60}Co concentrations have decreased to background levels about 100 meters from the site boundary. The evaporator location is about 110 meters inside, to the right of the fence line, at the approximate coordinates N360, W330.

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SECTION III

RADIOECOLOGY OF THE MAXEY FLATS SITE

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RADIOECOLOGY OF THE MAXEY FLATS SITE

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SUMMARY

Several possible migration pathways exist for radionuclides buried at the Maxey Flats shallow land burial site in eastern Kentucky. To evaluate potential biotic pathways, forest floor litter, newly fallen leaves, newly opened leaves and surface soil were sampled in the forest surrounding the restricted area and analyzed by high resolution gamma-ray spectroscopy. Most of the radionuclides in the forest were associated with the soil, with relatively small amounts of radionuclides associated with the litter and leaves. The endogenous radionuclides ^{40}K and ^{228}Th dominated soil distributions, but the exogenous radionuclides ^{60}Co and ^{137}Cs were relatively more abundant in the litter layer. The ^{60}Co content of newly fallen leaves was more variable than the other radionuclides, suggesting that the burial site has influenced the ^{60}Co concentrations at a few sampling locations and newly opened leaves from one hickory tree suggested that the source of the elevated ^{60}Co concentrations was the rooting substrate. The other radionuclide concentrations exhibited little variation between tree species from location to location.

INTRODUCTION

Information concerning the migration of radionuclides in the environment is needed for assessing the possible health and biological effects of radionuclides produced in the nuclear fuel cycle. A terminal point in the fuel cycle is the disposal of radioactive waste materials to a permanent repository. Ideally, such a repository would prevent the interred radioactive materials from contacting the biological components of the terrestrial and aquatic environments over a period of time long enough for physical decay to reduce radioactivity to negligible levels. Because of long physical half-lives, some radionuclides will persist for decades, others for centuries and a few for millenia. Land burial is an effective way to dispose of low level wastes because of the shielding properties that the earth provides against external ionizing radiation. Land burial is also relatively inexpensive and if buried deep enough the materials would not be contacted by plant roots or by burrowing animals. If appropriately covered, burial trenches would not become deflated by wind and water action nor would rain water percolate through the trench covering and provide opportunities to move radionuclides downward to groundwater for possible transport beyond the burial site boundaries.

This study was initiated to determine if radioactive materials buried in trenches at the Maxey Flats Waste Disposal Site have migrated into the surrounding oak-hickory forest. The burial ground is located at Maxey Flats, a 600 m wide flat-topped ridge in rural Fleming County, Kentucky(1). The elevation of the ridge crest is about 310 m above mean sea level, rising about 100 m above the surrounding valleys. The valley lands are under various kinds of crop management, i.e., row crops or pasture grass, but the adjacent steep slopes and ridge crests are covered by deciduous trees. The level portions of Maxey Flats have been deforested. The burial ground itself occupies about 40 acres and is completely enclosed by a chain link fence. Most of the ground inside the fence has been planted with perennial pasture to help reduce soil erosion. The grass is periodically mowed, fertilized and seeded to discourage the self-establishment of volunteer tree seedlings and also for esthetic purposes.

The first radioactive waste material was buried at Maxey Flats in May, 1963 and encapsulation forms later included steel drums and wood or cardboard boxes. Some liquid wastes were solidified by mixing with cement and paper and the mixture poured into polyethylene-lined trenches. With few exceptions the wastes are buried in trenches 76-110 m in length, 6 m wide and 6 m deep. After filling, the trenches were covered with a minimum of 1 m of a mixture of native soil and rock and compacted and shaped to encourage surface water runoff. Because the annual precipitation at Maxey Flats averages 110 cm per year, water percolating through the trench cover is one of the important ways to move buried radionuclides away from the disposal site and possibly into the surrounding forest. By 1972 some of the completed trenches became filled or partially filled with rain water that had percolated into the trenches through the earth cover. A water management program was initiated to dewater the trenches. This consisted of pumping the water, storing it and then reducing the water volume by heat evaporation using buildings and equipment located on the burial ground property.

The movement of radionuclides from the burial trenches at Maxey Flats has several possible pathways. These include surface water runoff (spills), air dispersal of the steam plume originating from the operating evaporator facility and vertical and lateral water movements through the soil or movement through cracks or joints in the underlying rock strata. Biotic pathways could also provide a migration route, especially through deep-rooted plants growing on and adjacent to trenches or by the activities of animals digging into the earth and exhuming buried wastes. Deeply rooted trees located downslope from the trenches might also tap contaminated subterranean water and bring radionuclides from below ground during the mineral uptake processes conducted by living trees.

EXPERIMENTAL STUDIES IN THE FOREST ADJOINING THE RESTRICTED AREA

The forest at Maxey Flats joins the burial ground boundaries on three sides. As a preliminary survey to determine if radionuclides associated with the burial ground were present in the forest, the forest floor litter was sampled at eighteen locations around the burial ground, as shown in Figure 1. Circular plots 0.32 m^2 in area were located on the ground between trees and all decaying leaves and humus were plucked by hand. Collecting was done in September prior to the onset of autumnal leaf fall. The litter was placed into plastic bags for shipment to Pacific Northwest Laboratory for ovendrying at 50-60°C, grinding and analysis. One hundred gram samples of milled, dry litter were counted for gamma-emitting radionuclides using instrumentation similar to that described by Wogman et al.(2). Two general kinds of radionuclides were associated with the forest floor litter-endogenous (natural) radionuclides originating from the parent soil material and exogenous (man-induced) radionuclides originating from the nuclear fuel process. Cesium-137 and ^{60}Co are exogenous radionuclides and both are constituents of global fallout as well as being components of low level radioactive wastes stored at Maxey Flats. Potassium-40 and ^{228}Th are endogenous radionuclides originating from the underlying rock strata and soil.

Very low levels of radionuclides were measured in the forest floor litter, as shown in Figure 2. Cesium-137 and ^{60}Co were the most abundant radionuclides of exogenous origin and these have their origin in global fallout, which is ubiquitously distributed over all land surfaces. The variation among ^{228}Th, ^{40}K and ^{137}Cs radionuclides in the samples shows about the same pattern, but ^{60}Co content of the litter was much more variable, suggesting that the source of ^{60}Co measured in some of the forest floor litter samples is the burial ground itself rather than global fallout. See Figure 2 and Table 1. The ^{60}Co data are summarized on maps in Figure 3, showing concentrations measured for the samples collected near the burial site in 1979.

Table 1. Mean values and coefficient of variation (Cν) of radionuclides (d/m/g) in soil, litter and newly fallen leaves at Maxey Flats, Kentucky, September-December, 1979. The variability among the litter and leaf samples suggests that the source of ^{60}Co measured in some of the samples is the burial ground itself rather than global fallout.

	^{40}K	^{137}Cs	^{228}Th	^{60}Co
Soil (mean)	21.96	3.14	1.95	0.458
Cν	0.22	0.52	0.077	0.92
Litter (mean)	4.36	3.46	0.45	0.70
Cν	0.66	0.38	0.36	1.68
Leaves (mean)	4.00	0.61	0.26	0.27
Cν	0.32	1.19	0.27	1.63

After autumn leaf fall in early December, 18 locations were sampled on the forest floor near the same locations from which litter had been collected earlier. Newly fallen leaves were collected by hand-picking leaves from inside 0.66 m^2 circular plots. These samples were treated in the same manner as litter and the results of radiochemical analyses are shown in Figure 2. The newly fallen leaves had about the same amount of ^{40}K and ^{228}Th as did litter samples. However, ^{60}Co content of newly fallen leaves was very low as compared to litter, with some samples having ^{60}Co levels below counting detection limits.

Because ^{60}Co could have appeared in newly fallen tree leaves by external absorption of airborne particles, it was decided to collect newly opened tree leaves in the spring to determine if ^{60}Co was present within the physiologically active leaves. The range of values expressed as d/m/g dry tissue from three maple and three hickory trees collected in the vicinity of the evaporator facility in April, 1980 is shown in Table 2.

Table 2. Radionuclides in Tree Tissues, Maxey Flats, Kentucky, April, 1980. Hickory leaves appeared to accumulate more ^{137}Cs, ^{228}Th and ^{60}Co than did maple leaves. The source of ^{60}Co is probably the rooting substrate, rather than fallout from the evaporator plume.

	Maple (d/m/g)	Hickory (d/m/g)
^{60}Co	<0.03-0.07	<0.05-4.0
^{137}Cs	<0.12-0.24	0.43-0.50
^{228}Th	<0.04	0.15-0.19
^{40}K	37-38	36-52

The leaves of one hickory tree had 4.0 d/m/g of ^{60}Co, suggesting that the source of ^{60}Co was the rooting substrate rather than airborne external contamination delivered by the steam plume from the evaporator facility. Hickory leaves also appeared to accumulate more ^{137}Cs and ^{228}Th than maple leaves.

To determine if individual trees had enhanced levels of ^{60}Co, leaves from separate trees were collected. Trees were marked with a metal tag nailed to the bole and three wire baskets, each 0.5 m^2, were placed around thre trunk of each tree to collect the leaves as they fell during the period of leaf fall in the autumn of 1980. Baskets were set out in late September and emptied in early December. The average amount of leaf fall collected in the baskets was 364 g/m^2.

Mineral soil was collected beneath each tree after scraping away dead leaves and humus layers to expose the mineral soil. Potassium-40 was the most abundant radionuclide in the mineral soil, as shown in Figure 4. Cesium-137 and ^{228}Th were present in intermediate amounts and ^{60}Co was the least abundant. The ^{60}Co content of newly fallen leaves was also more variable than the other radionuclides, suggesting again that ^{60}Co was enhanced at only a few of the locations. This suggested that ^{60}Co was derived through migration from the burial ground. Most other trees were at background (global fallout) levels.

Potassium-40 was the most abundant radionuclide measured in newly fallen leaves, and there was relatively little variation between tree species or from location to location (Figure 4). Cobalt-60 and ^{137}Cs contents of newly fallen leaves were much lower than ^{40}K. Cobalt-60 was present in measurable amounts in some leaf samples but not in others, again suggesting that the origin of some of the ^{60}Co was the burial ground. The ^{60}Co data are summarized on maps in Figure 5, showing concentrations measured for the samples collected near the burial site in 1980.

An inventory of radionuclides in the soil, forest floor litter and newly fallen leaves in 1979 is shown in Table 3 on the following page. These data indicated that on an areal basis (m^2) most of the radionuclides in the forest were associated with the surface soil, with relatively small amounts of radionuclides associated with the litter and the newly fallen leaves. The endogenous radionuclides

Table 3. Radionuclides in Soil, Forest Floor Litter and Newly Fallen Leaves, Maxey Flats, Kentucky, 1979. Most of the radionuclides in the forest are associated with the surface soil.

Sample Type	^{40}K	^{137}Cs	^{228}Th	^{60}Co
		$(d/m/m^2)$		
Newly fallen leaves	1,200	177	75	78
Forest floor litter	4,800	3,800	500	700
Soil (upper dm)	2,800,000	426,000	250,000	52,000

^{40}K and ^{228}Th dominated the radionuclides in the soil. Cesium-137 and ^{60}Co, exogenous radionuclides, were more abundant in litter than was ^{228}Th. Cesium-137 appeared to be retained in the litter layer, probably reflecting its airborne origin (fallout). Cobalt-60 has a relatively short half-life of about 5 years as compared to about 30 years for ^{137}Cs and thus canot be expected to persist in the forest floor for as long a time as ^{137}Cs. The reason for the migration of ^{60}Co is not entirely clear at this time but may be due to organic complexation with chelating compounds buried along with the radioactive materials, permitting ^{60}Co to be more mobile than other gamma-emitting radionuclides. Tritium and ^{14}C analyses are being conducted in plant sap and wood for presentation at a future time.

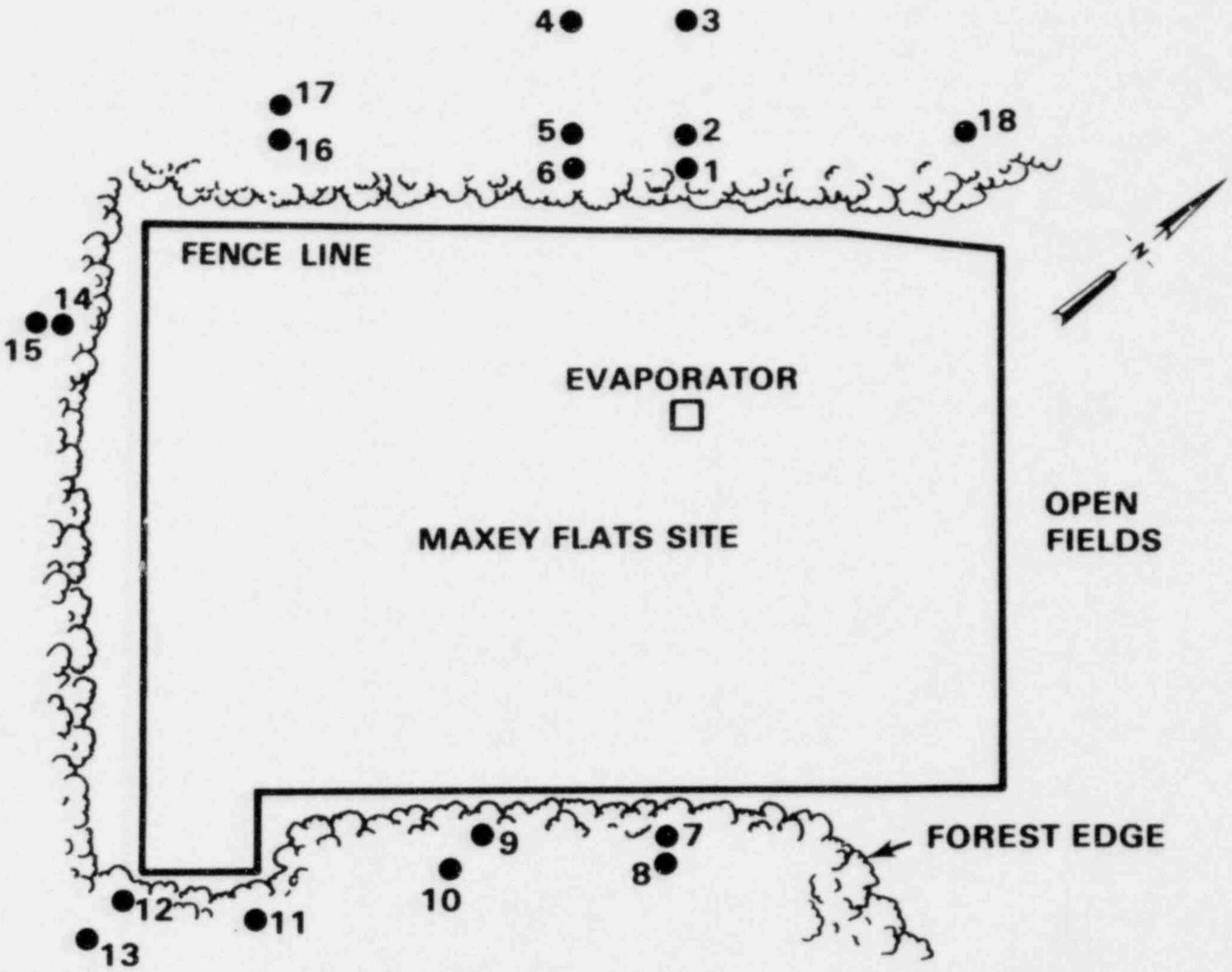


Figure 1. Map Showing the Boundary of the Oak-Hickory Forest in Relation to the Burial Site at Maxey Flats. The bold-faced numbers shown outside the fence line are the approximate locations of the sampling stations within the adjacent forest.

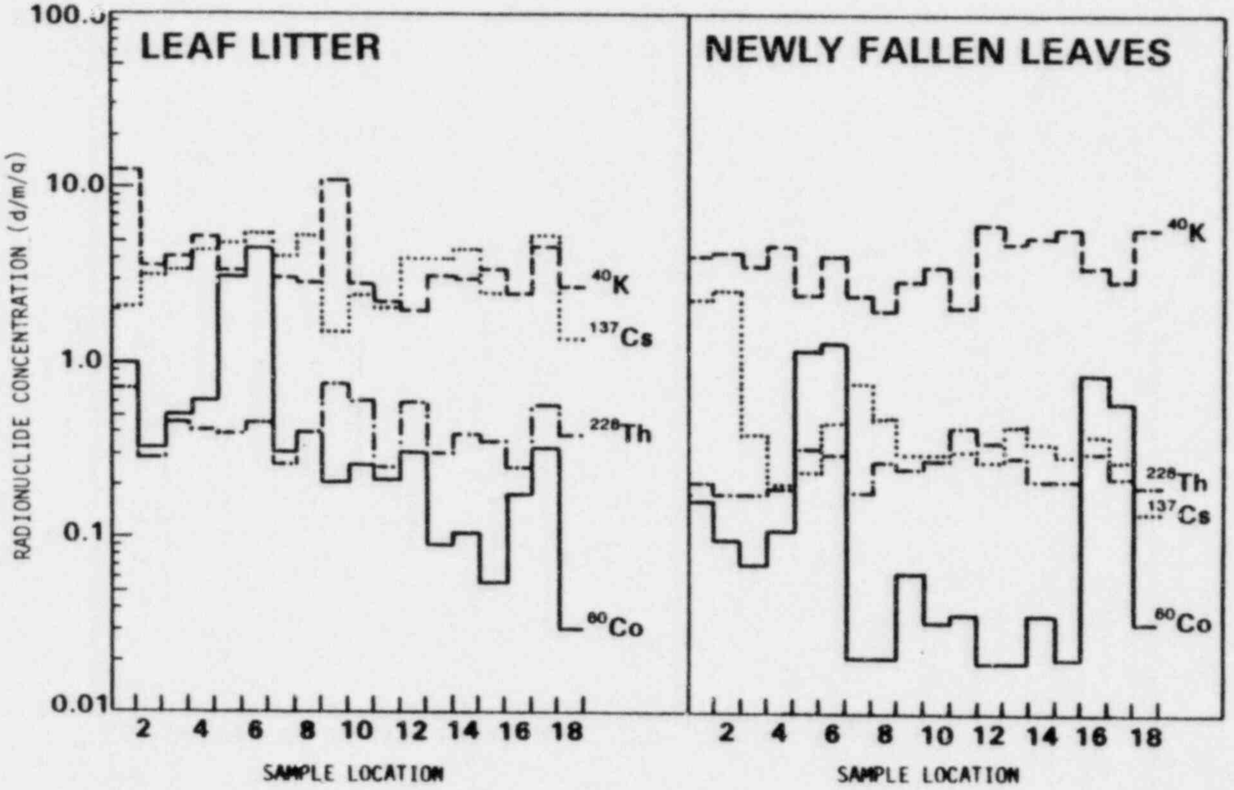
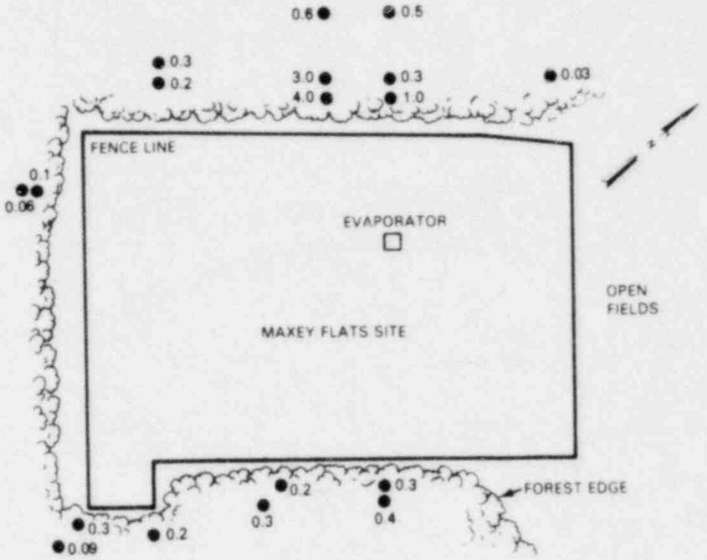


Figure 2. Radionuclide Content of Leaf Litter and Newly Fallen Leaves at Maxey Flats in 1979. Very low levels of radionuclides were measured. Cesium-137 and ^{60}Co were the must abundant radionuclides of exogenous origin and have their origin in global fallout. The variation among ^{228}Th, ^{40}K and ^{137}Cs radionuclides in the samples shows about the same pattern, but the ^{60}Co content was much more variable, suggesting that the source of ^{60}Co measured in some of the samples is the burial ground itself rather than global fallout. The ^{60}Co concentrations were highest at locations 5 and 6 in the west drainage channel.

^{60}Co Concentrations (d/m/g) in Leaf Litter, 1979



^{60}Co Concentrations (d/m/g) in Newly Fallen Leaves, 1979

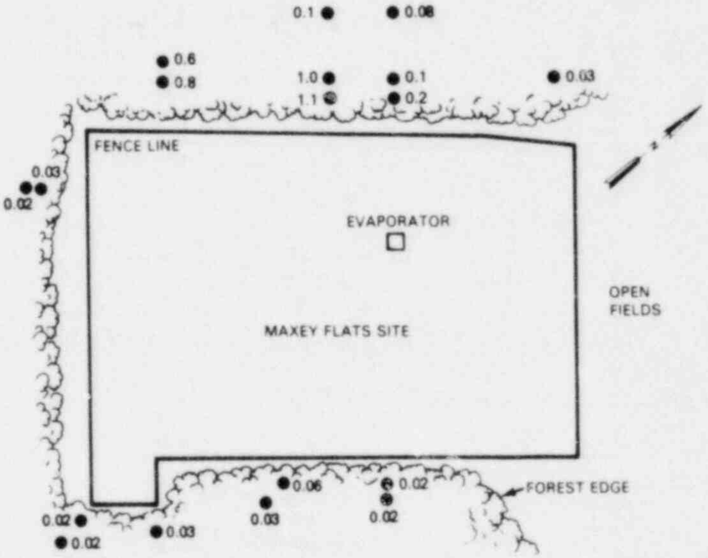


Figure 3. Maps showing ^{60}Co concentrations in leaf litter and newly fallen leaves at Maxey Flats in 1979. The dots represent the sampling locations summarized in Figure 1. Cobalt-60 concentrations were more variable than the other radionuclides measured, suggesting that the source of ^{60}Co measured in some of the samples is the burial ground itself rather than global fallout. This variation is particularly noticeable in the west drainage channel near the site boundary.

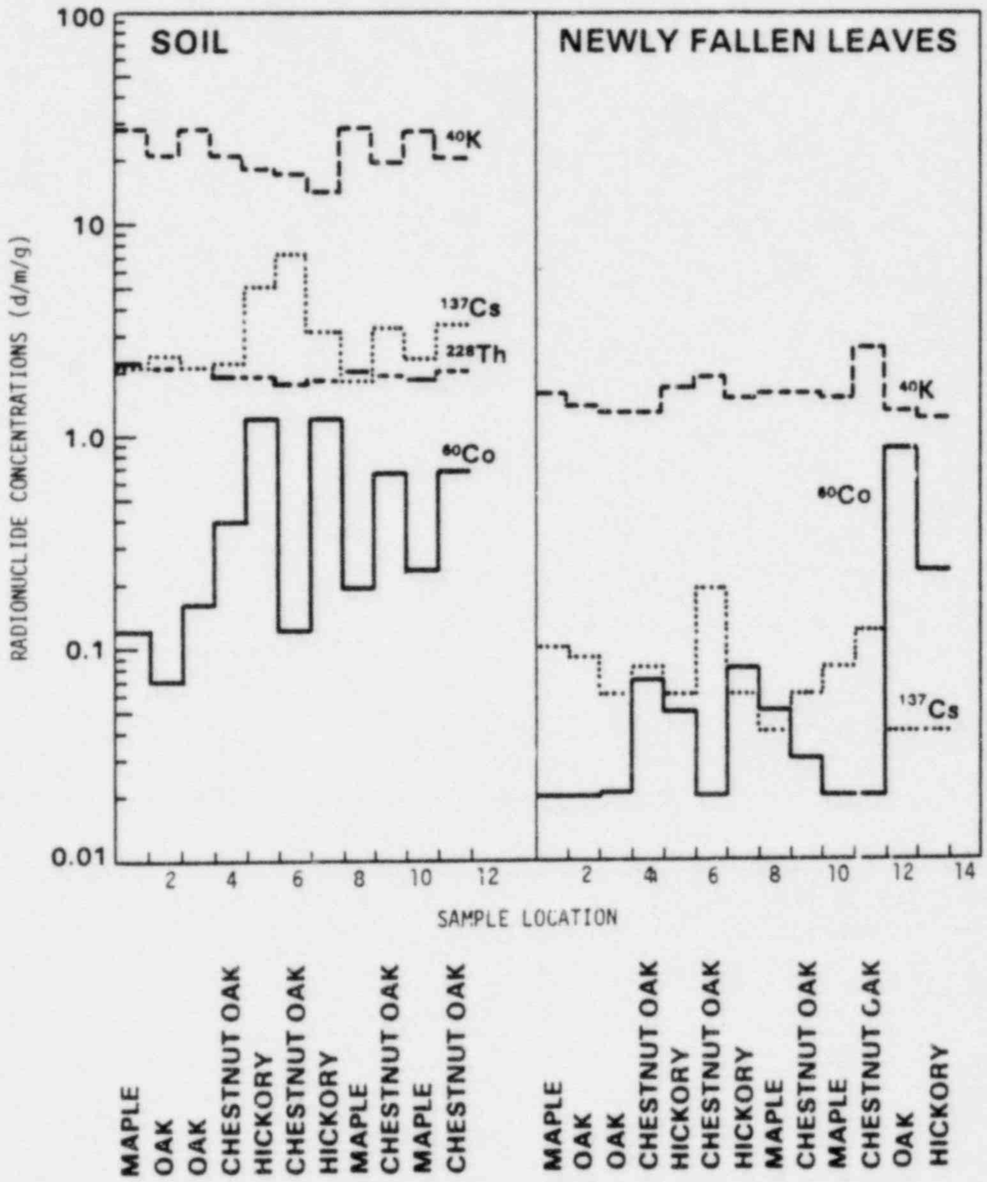
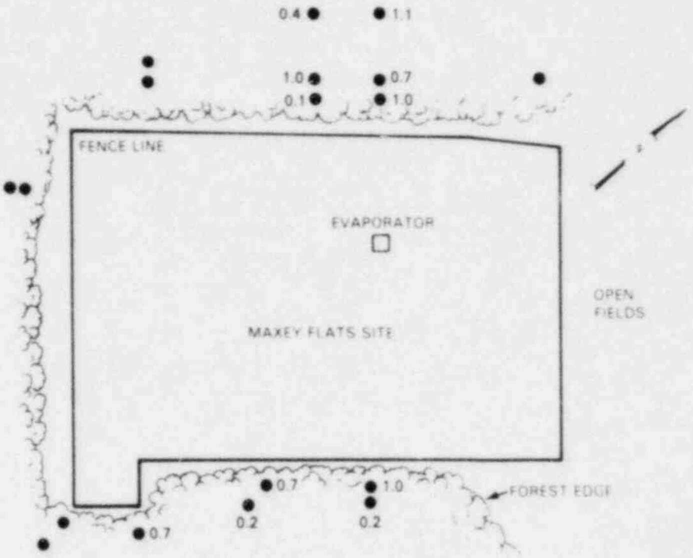


Figure 4. Radionuclide Content of Soil and Newly Fallen Leaves Beneath Individual Trees at Maxey Flats in 1980. Cobalt-60 is the least abundant radionuclide measured in the soil samples. The ^{60}Co concentrations in newly fallen leaves were more variable than the other radionuclides, and ^{60}Co was enhanced at only a few of the locations.

^{60}Co Concentrations (d/m/g) in Soil, 1980



^{60}Co Concentrations (d/m/g) in Newly Fallen Leaves, 1980

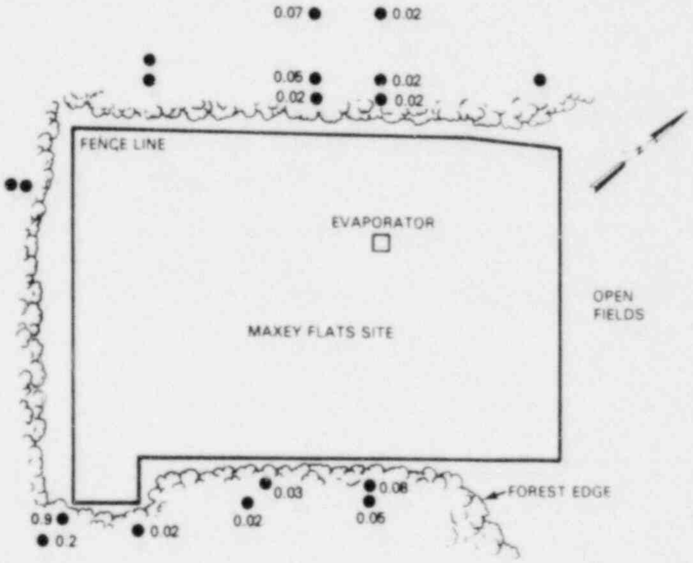


Figure 5. Maps showing ^{60}Co concentrations in soil and newly fallen leaves at Maxey Flats in 1980. The dots represent the sampling locations summarized in Figure 1. Cobalt-60 was the least abundant of the radionuclides measured in the soil samples. The ^{60}Co concentrations in newly fallen leaves were more variable than the other radionuclides and ^{60}Co was enhanced at only a few locations.

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SECTION IV

TRENCH WATER-SOIL CHEMISTRY AND INTERACTIONS AT THE MAXEY FLATS SITE-II

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TRENCH WATER-SOIL CHEMISTRY AND INTERACTIONS AT THE MAXEY FLATS SITE-II

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August 1981

SUMMARY

The chemistry of leachates in the Maxey Flats trenches is an important factor in evaluating the site's effectiveness for retaining radionuclides. Trench water composition reflects interactions of the buried waste with infiltrating groundwater and the effects of bacterial decomposition of organic material. Aerobic, anaerobic, sulfate reducing, denitrifying and methanogenic bacteria are present in the leachates. Bacterial processes cause decreases in redox potential (Eh), sulfate and nitrate concentrations, as well as increases in alkalinity and ammonia relative to local groundwater compositions. The major radionuclides of concern present in trench waters include: ^{3}H, ^{60}Co, ^{90}Sr, ^{134,137}Cs, ^{241}Am, and ^{238,239,240}Pu.

Several additional laboratory and field analyses were performed on the fourth sampling of trench waters to better characterize the waters and to assure the reliability of some procedures. New procedures include: sulfide ion measurement in the field; laboratory ion chromatographic determinations of sulfate, chloride, and bromide; Eh and pH measurements in the laboratory; and laboratory measurement of ferrous-ferric concentration ratios by a colorimetric procedure. No overall systematic change in any disposal trench was observed. Analysis of water collected from the experimental interceptor trenches at Maxey Flats shows a chemical composition intermediate between disposal trench water and local groundwater.

The existence of organo-radionuclide complexes that may enhance radionuclide mobility is a subject of much concern in low-level waste management. Low concentrations of free EDTA, NTA, and DTPA* are present in some leachates. Gel filtration experiments were inconclusive in demonstrating radionuclide-chelate associations. A laboratory controlled environmental chamber that maintained anoxic water conditions was used to demonstrate the persistence of Co-EDTA and Co-NTA complexes in solution, contrary to results for Zn-EDTA and Cu-EDTA reported in the literature.

Sorption experiments demonstrated the ability of EDTA to lower the apparent sorption capacity of shale from Maxey Flats. Experiments to measure

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*EDTA-ethylenediaminetetraacetic acid; NTA-nitrilotriacetic acid; DTPA-diethylenetriamine pentaacetic acid.

steady-state sorption behavior with site-specific materials are continuing, using an improved apparatus that maintains the anoxic character of the trench water.

1. INTRODUCTION

This report on the geochemistry of the Maxey Flats, Kentucky, site is part of a larger research program for the U.S. Nuclear Regulatory Commission (USNRC). The containment of radionuclides at shallow land burial sites is the prime factor in the evaluation of this method of disposal of low-level radioactive waste. This study is an attempt to monitor the behavior of existing low-level disposal sites, provide an understanding of significant factors which affect prediction of radionuclide movement along the groundwater flow paths, and assist in the development of criteria for the selection and licensing of future low-level disposal sites.

Brookhaven National Laboratory (BNL) began a field and laboratory testing program in 1976 sponsored by the USNRC, in cooperation with the U.S. Geological Survey (USGS), to study the existing commercial low-level radioactive waste disposal sites. This investigation measures the source terms (concentrations in solution) of radionuclides and other solutes in the disposal trench waters, and describes the physical and chemical properties of the site geochemistry that control the movement of radionuclides. Procedures used to collect and analyze trench waters and source term data obtained previously were described in earlier reports.(1,2)

Three studies are described in this report: (a) trench water analyses, (b) radionuclide complexes, and (c) sorption experiments.

1.1 Trench Water Analysis

Water samples from selected disposal trenches, experimental research trenches, and nearby wells were analyzed for inorganic, organic, and radiochemical constituents. The results of these analyses are part of the source term data base for the commercial disposal sites. The well and experimental trench waters were sampled to detect and identify radionuclides that might be migrating along groundwater flow paths, and to compare local groundwater compositions to those of the disposal trenches.

Four of the experimental trenches (T-2 to T-5) were sampled aerobically during the sampling trip in October-November 1979; experimental trench T-2 was also sampled anaerobically. An additional analysis for bromide ion was included because NaBr was used as a tracer in the interceptor trench experiment.

Anaerobic procedures for the collection, preservation, and analysis of anoxic trench waters have been developed and are continually updated to supplement standard methods as necessary.

1.2 Radionuclide Complexes

The existence of organo-radionuclide complexes in disposal trench waters has been a subject of much concern since the reported enhancement of ^{60}Co migration was attributed to complexing with EDTA.(3) Plutonium may exist as complexes of organic chleating agents in the disposal trench waters at Maxey Flats, KY.(4) Tests to detect the existence of free ogranic chelating agents and radionuclide complexes in selected trench waters were performed.

1.3 Sorption Geochemistry

Laboratory batch sorption studies were performed with anoxic trench waters collected from the Maxey Flats, KY, site. Anaerobic techniques and equipment were developed to maintain anoxic water conditions during the sorption tests. Conclusions derived from these studies are expected to have direct application to the evaluation of radionuclide migration along groundwater flow paths as well as help in developing siting criteria for future sites.

2. EXPERIMENTAL APPROACH

The objective of the analytical program is to measure the concentration of radionuclides and other chemical species present in trench waters. The anaerobic technique allowed samples to be successfully stored for months without visible signs of the reddish brown ferric hydroxide that forms after exposure of most trench waters to air.(2) In addition to analyses reported earlier,(1) several additional inorganic analyses were performed to more thoroughly characterize these waters. During the last field sampling, dissolved sulfide was measured with an ion selective electrode.(5) Sulfate analyses were made using an ion-chromatographic (IC) technique.(6) Bromide and chloride separations and analyses were also made using the ion chromatographic technique.(6) A colorimetric technique was used to determine total iron and ferrous iron concentrations.(7) Laboratory measurements of pH and Eh were made in a glove box filled with argon (99.999% Ar) to evaluate the extent of shifts in the chemical equilibria present in these waters, which were anticipated to occur during the sample collection, transportation, and filtering processes prior to analyses. These chemical changes occur because the waters have been removed from contact with the soil and have experienced temperature and gas partial pressure changes. Hopefully, these alterations of the sample environment are not drastic, and the inorganic analyses are still representative of in situ field conditions.

Trench waters were analyzed for major inorganic constituents using ion specific electrodes, colorimetric and atomic adsorption methods. Organic compounds were determined by methylene chloride solvent extraction followed by gas chromatographic separations and mass spectrometric analyses. Radiochemical analyses were performed by gas flow proportional counting (alpha and beta gross activity), liquid scintillation counting (tritium), Ge-(Li) gamma-ray spectroscopy, and radiochemical separations (^{90}Sr, ^{238,239,240}Pu).

Samples from selected disposal trenches, as well as the experimental interceptor trenches at Maxey Flats were analyzed for uncomplexed EDTA, DTPA, and NTA. These particular compounds were selected for analysis because they are among the most frequently encountered complexing agents used in decontamination solutions and laboratory procedures. A series of gel filtration experiments were performed with trench waters in an effort to detect the existence of organo-radionuclide complexes. Gel filtration chromatography is a method of separating materials based on their molecular sizes. Details of the experimental method are reported elsewhere.(8)

The stability of cobalt-EDTA and cobalt-NTA complexes was investigated in a controlled laboratory environment chamber by using solid and liquid phases designed to simulate the trench environment at Maxey Flats. Anoxic water conditions are developed in the chamber, indicated by a low redox potential, and the presence of sulfide in the water is determined by measurements with a Ag/AgS electrode. The amount of ^{60}Co in solution is measured by periodically removing an aliquot and measuring its ^{60}Co content by gamma-ray spectroscopy. Experimental details are described elsewhere.(9)

Sorption studies of radionuclides are continuing using site specific soil and trench waters from Maxey Flats. The anaerobic technique was improved to assure the anoxic character of the trench water, since oxygen contamination would cause the precipitation of ferric hydroxide and coprecipitation of radionuclides. The sorption measurement involves adding unfiltered trench water to radionuclide chloride salts. A series of preparations are performed using a manifold and a glove box flushed with ultra high purity argon. The radionuclides: ^{241}Am, ^{85}Sr, ^{134}Cs, ^{137}Cs, ^{60}Co, and ^{152}Eu were used in these studies. Details of the procedure are reported elsewhere.(8)

3. RESULTS AND DISCUSSION

3.1 Trench Water Inorganic Analyses

Four disposal trenches were sampled in October 1979 along with one observation well and four experimental trenches. Anaerobic sampling procedures(2) were used for the disposal trenches and one of the experimental trenches (T-2E). The other experimental trenches were collected aerobically since they did not contain sufficient water for anaerobic sampling. The bulk of the routine analyses of these waters were reported earlier.(7) The completion of the water analyses and results of additional testing are reported here (see Table 1).

Results for the disposal trench water analyses are similar to those reported for waters collected during earlier sampling trips.(2) The waters frequently show low sulfate and nitrate content, high titration alkalinity, the presence of ammonia and sulfide, and low Eh values. These are characteristics of chemically reducing, anoxic water regimes frequently found in stagnant waters in natural environments, and are typical of organic-laden waste waters in which bacterial activity has consumed dissolved oxygen. The observed trench water compositions result from a combination of two processes; (1) leaching of the waste by groundwaters percolating through the buried material and, (2) bacterial degradation of organic matter leached from the waste. The extent to which the local groundwater is changed by these two processes is a function of how long water accumulations persist in the trenches. A lengthy stay increases leaching, as well as the extent to which bacterial degradation processes modify the leachate composition. Sodium, potassium,

Table 1

Analysis of Trench Water and Well Water Samples Taken From
Maxey Flats, Kentucky (October 30, 1979-November 2, 1979)
(mg/L)[a]

Component	Disposal Trenches 19S	 27	 30	 32	Well UB1-A	Experimental Trenches T-2E	 T-3W[f]	 T-4E[f]	 T-5[f]
Sulfide (mV)	430	-490	b	-540	b	-79	b	b	b
Barium	<2	17	<2	19	<2	<2	<2	<2	<2
Cesium	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Iron[c]	60	1400	10	0.4	<0.1	1.3	1.3	0.5	0.2
Iron[d]	58	1400	10	<0.5	<0.5	2.0	1.2	<0.5	<0.5
Fe+2/Fe+3 [d]	30/1	39/1	25/1	1	b	4/1	0.07/1e	b	b
Lithium									
Total alkalinity (as $CaCO_3$)	900	300	6400	1700	30	300	450	300	320
Hardness (Mg+Ca) (as $CaCO_3$)	650	4800	5400	1400	1700	1600	1500	1900	2000
Inorganic Carbon	170	74	1500	350	15	68	120	50	86
D.O.C.	300	500	260	670	5.1	120	15	29	14
Nitrogen (N) (NH_3-Probe)	30	60	50	90	<1	20	g	5	<1
Nitrogen (N) (NH_4 -Color)	34	75	110	88	<1	13	1.0	<1	<1
Chloride-I.C.[h]	190	6100	200	330	350	1200	160	140	84
Bromide-I.C.	1.3	180	3.8	3.3	8.2	180	2200	2.4	4100
Nitrogen (NO_2-Color)	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
Nitrogen (NO_2+NO_3)	0.12	0.75	0.11	0.8	0.50	0.51	0.39	0.05	1.4
Phosphate	<1	<1	2.0	1.1	<1	<1	<1	<1	<1
Silica	15	19	42	29	19	7.7	8.7	7.5	0.74
Sulfate-Color	23	1100	>60	>60	1700	530	980	1500	1300
Sulfate-I.C.	<0.5	2.2	j	9.6	1700	530	980	1500	1300
Sulfate-I.C. (Acidified)	k	62	85	64	1900	560	1100	1500	1400
pH[l]	7.1	5.6	7.9	7.5	6.4	7.0	6.8	7.2	7.5
Tritium pCi/L	8.7 E7 (<1)	5.1 E9 (<1)	1.8 E10 (<1)	2.1 E9 (<1)	4.9 E6 (<1)	6.7 E8 (<1)	3.7 E7 (<1)	1.5 E5 (1.1)	1.7 E4 (3.1)
^{241}Am pCi/L	1.3 E3 (7.6)	4.7 E3 (3.0)	1.2E2 (2.2)	6.0 E3 (3.0)	b	n	n	n	n
^{60}Co pCi/L	3.3 E3 (1.5)	4.0 E3 (2.6)	3.6 E4 (<1)	2.3 E3 (3.6)	1.1 E2 (1.7)	n	n	n	n
^{134}Cs pCi/L	b	b	4.3 E2 (9.6)	1.1 E2 (23)	b	n	n	n	n
^{137}Cs pCi/L	2.7 E3 (1.4)	2.2 E3 (3.2)	2.9 E4 (<1)	3.2 E3 (2.4)	b	n	n	n	n
^{22}Na pCi/L	n	n	n	n	n	4.9 E1(19)	n	n	n

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[a] Measurement given in mg/L, unless otherwise noted.
[b] Analysis not performed.
[c] Atomic absorption method.
[d] Colorimetric method.
[e] Sample was not filtered anaerobically.
[f] Trench was not sampled anaerobically.
[g] Unstable system.
[h] Ion chromatography
[i] Analytical limitation due to low iron concentration.
[j] Insufficient sample.
[k] Analytical limitation
[l] pH after anoxic filtration.
[m] Number in ( ) = 2 σ% counting error.
[n] Less than detection limit.

and chloride concentrations in the trench waters are good indicators of the extent of leaching, since they are conservative relative to ensuing bacterial processes. Bacterial decomposition of organic material modifies the trench water composition in the following manner. Dissolved oxygen in the water is consumed rapidly, followed by nitrate and sulfate reduction. Carbon dioxide and ammonia are generated continually during these stages, resulting in increased alkalinity and ammonia levels in the waters and decreased concentrations of sulfate and nitrate relative to uncontaminated groundwaters. In the absence of heavy metals, such as iron, sulfide concentrations can build to detectable levels. Concentrations of other ions such as calcium, magnesium, and strontium are controlled by the changes in carbonate alkalinity developed from bacterial action. As reported previously,(2) titration alkalinity curves show an irregular behavior reflecting the complex nature and high buffering capacity typical of these anoxic trench waters. Presumably, the highly irregular shape of the titration curves is due to the presence of titratable components leached from the wastes.

The ferrous iron determination is useful in evaluating redox equilibria of trench waters. Laboratory pH and Eh measurements are also required for this evaluation. Sulfide was detected in field measurements during collection of these waters (high negative mV values in Table 1 indicate measurable sulfide concentrations) as would be expected in anoxic waters. Quantification of this analysis is in process. The ion chromatographic analyses were performed to check the reliability of the colorimetric sulfate procedure. Bromide interferes with chloride determinations when analyzed by the indirect colorimetric technique.(2) To circumvent this difficulty, an ion chromatographic technique was employed for both halogen determinations. Bromide salts were added as tracers to experimental trenches T-1, T-2, T-3, and T-5.(1)

The presence of high iron and sulfide ion concentrations in trench 27 is puzzling in that a high iron concentration should keep free sulfide ion to extremely small levels when equilibrium exist. Conflicting results were also obtained for sulfate concentrations (colorimetric vs ion chromatographic procedures) in trench 27 (Table 1). If the high sulfate concentration is correct (1100 mg/L), this would also conflict with the observation of detectable sulfide found in the field measurements. We believe that the high sulfide field measurement (-490 mV) may be in error due to equipment malfunctions but disequilibrium may exist. This particular trench requires more detailed investigation to resolve these apparently contradictory results.

For the multiply sampled trenches, no consistent pattern of change with time is evident. However, detecting an evolutionary change in trench water composition is complicated by two factors. First, the four year monitoring interval is short relative to the lifetime of the disposal site. Second, water in the trenches is pumped out periodically to prevent overflows when the accumulation rate exceeds the rate at which the water migrates through the groundwater flow paths. After each pumping, the trench water-soil system may require time to establish conditions as they existed prior to the pumpout.

Preservation of the anoxic nature of the water sample was a prime consideration in developing sampling and analytical procedures. Some shifts in the numerous chemical equilibria present in these complex waters are to be anticipated during the sample collection, transportation, and filtering processes prior to analysis. One method of evaluating the extent of these changes is to examine Eh-pH relationships measured in the field, and later in the laboratory during subsequent sample manipulations.

Figure 1 presents an Eh-pH stability diagram for iron species frequently found in both oxidized and reduced natural environments. Measurements of trench water Eh and pH in the field correlate well with those expected for equilibria between aqueous ferrous iron, limonitic iron oxide (a common soil component), and iron sulfides (commonly formed in anoxic sediment-water systems). After sampling, transporation and laboratory filtration, the Eh-pH values for the various trenches have shifted, but are still in good agreement with the equilibria lines for these systems. This indicates that the water systems have not been drastically altered during sample handling. Waters from well UB1-A differ from the trench waters in that the redox environment in the well is more oxidizing and not poised by iron equilibria.

A more detailed picture of the chemical systems for individual trenches can be seen in Figure 1. Field data for trench 32 indicates an equilibrium with iron sulfide. Field measurements for sulfide ion(8) showed the highest readings for trench 32, and the color of the water drawn from the trench was black. Precipitated iron sulfide is black and is frequently observed in natural anoxic waters but so is organic debris. The remaining waters appear to be controlled by ferrous iron and iron oxides commonly present in soils. In the trench 27 waters, total dissolved iron is in the range of 10^3 mg/L. Chloride content in this water is also high so that the actual concentration of free ferrous iron is in the proper range (for this diagram) due to extensive iron-chloride pairing at these relatively high iron and chloride concentrations. Experimental interceptor trench T-2E also shows an equilibrium of ferrous iron and iron oxide, although the redox state of this water is more oxidizing than the disposal trenches.

Comparison of the disposal trench waters with the more typical groundwaters (well UB1-A) shows the contrast between ground waters and the anoxic trench waters. The experimental trench waters show a chemistry intermediate between the well and disposal trench waters. Results of dissolved organic carbon (DOC) (Table 1) and extractable organic material (Table 2) also show an intermediate composition between the complex disposal trenches and the well waters. This may reflect an increased influence of disposal trench waters on the experimental trenches relative to their influence on the well waters.

Relatively low sulfate concentrations, high alkalinity, and predominance of ferrous iron in solution indicate an reducing water chemistry in the experimental trenches.

3.2 Trench Water Organic Analysis

Results of the identification and quantification of organic compounds are given in Table 2. The suite of organic compounds present is similar to those

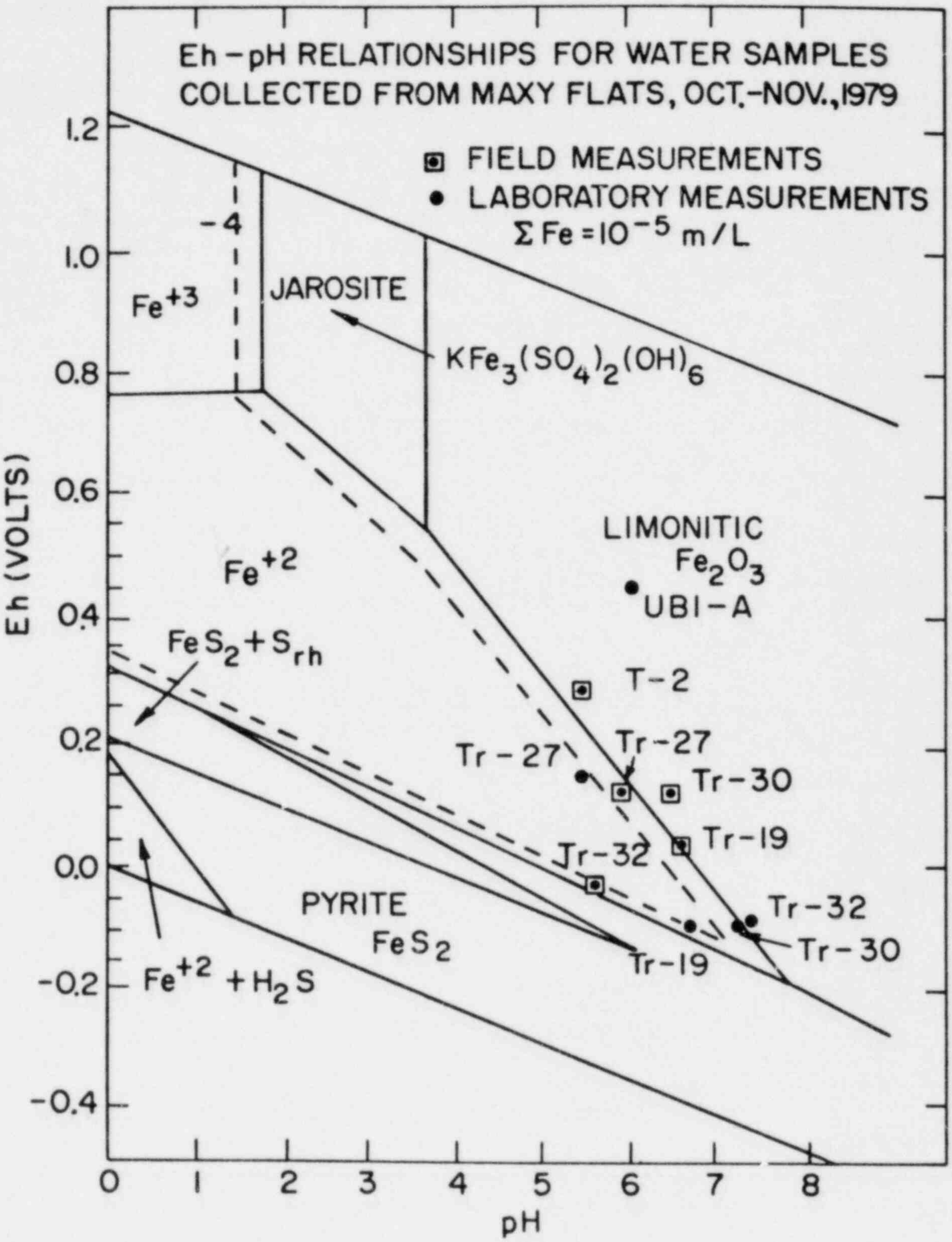

Figure 1 Eh-pH relationships for water samples collected from Maxey Flats, October-November 1979. Diagram modified after Garrels and Christ.(10)

Table 2

Organic Compounds Identified in Trench and Well Waters
Maxey Flats, Kentucky, Disposal Site (October-November 1979)
(mg/L)

Compound	Disposal Trenches				Well	Experimental Trenches			
	19S	27	30	32	UB1A	T-2E	T-3W	T-4E	T-5
Acidic Fraction:									
2-Methylpropionic acid	0.12	0.24	d						
2-Methylbutanoic acid	0.52	0.60							
3-Methylbutanoic acid	0.80	0.76							
Valeric acid		0.28							
Ethylene glycol	N.Q.[a]	N.Q.	N.Q.	N.Q.	N.Q.		N.Q.		
Diethylene glycol	N.Q.				N.Q.				
3-Methylpentanoic acid	0.67	0.45		0.98					
C6 acid [b]	0.26	0.16							
Phenol	0.04	0.23		0.08		0.06			
Hexanoic acid	0.13	0.64							
2-Methylhexanoic		0.24		0.57					
C8 acid		N.Q.	N.Q.			N.Q.			
Cresol (isomers)	1.2	0.56		0.28					
2-Ethylhexanoic acid	1.9	9.7	1.1	3.2		0.44			
C8 acid [c]	0.96	0.08	1.8	0.80					
C8 acid [c]	1.1	0.06							
Benzoic acid		0.24							
Octanoic acid		0.10		0.20					
Phenylacetic acid	0.34	0.16							
Nonanoic acid	0.08								
Phenylbutyric acid				N.Q.					
Phenylpropionic acid		0.50							
Polyethylene glycol	N.Q.	N.Q.			N.Q.				
Phenylhexanoic acid	N.Q.	N.Q.		N.Q.					
Toluic acid			2.7	2.1					
dioctyl adipate							N.Q.		
di-isooctyl pthalate							N.Q.		
Neutral Fraction:									
p-Dioxane	N.Q.	N.Q.		N.Q.	N.Q.	N.Q.	N.Q.		
Methylisobutyl ketone	0.20	1.8							
Toluene	9.5	0.56		2.4					
Xylene	0.03			0.13					
Cyclohexanol	0.10								
Dibutyl ketone	N.Q.								
2-Ethyl 1-hexanol		0.17							
Diethylene glycol diethyl ether		N.Q.							
Fenchone		N.Q.	0.08						
Triethyl phosphate	0.28	0.29							
Camphor	N.Q.	N.Q.							
Naphthalene	0.03	0.06		0.07					
Tributyl phosphate	0.17			0.30					
Phthalate ester	N.Q.		N.Q.	N.Q.					
2-butanone								N.Q.	
tetrahydrofuran						N.Q.		N.Q.	N.Q.
cyclohexanone						N.Q.		N.Q.	

[a] Not quantified.

[b] Quantified using 3-Methylpentanoic acid standard.

[c] Quantified using 2-Ethylhexanoic acid standard.

[d] A blank space was left in the table where the compound was not detected.

found in previous water samples taken from the Maxey Flats disposal trenches.(2) Major components in these waters are also similar to those found in West Valley trench water samples.(2) The results for the interceptor trenches and the well water reflect the influence of nearby trenches 27 and 32, respectively. The organic compounds: 2-butanone, tetrahydrofuran, cyclohexanone, dioctyl adipate, and diisooctyl phthalate were found in the experimental trench water, but were not found in disposal trench waters. The possibility exists that the organics found in the experimental trenches may be derived from plastics used in the trench construction, particularly since some are not in disposal trenches.

3.3 Tritium in Rainfall

Rainfall collectors were placed in the vicinity of wells 11E, 12E, the UA wells and UB wells by USGS personnel. Rain samples collected by USGS between March 1977 and September 1979 were shipped to BNL for tritium analyses. The tritium content in these samples are given in Table 3. In general, a relative increase or decrease in the tritium content from one sampling period to the next is consistent at all the sampling locations. This could indicate that the relative changes are controlled by meteorological variations. A more quantitative evaluation of this data would require information concerning the amounts of rainfall during these time intervals, along with the quantities and tritium concentration of trench waters processed through the evaporator.

3.4 Chelating Agents in Trench Waters

There is considerable concern about the presence of organic chelating agents in low-level waste disposal trenches. Radionuclides complexed with chelating agents are not likely to be removed by the sediment surrounding the disposal trenches, thereby resulting in considerably enhanced migration potential. Strong organic chelating agents are used in decontaminating solutions in academic, medical, and industrial situations. The waste products are disposed of by shallow land burial, and it is probable that some radionuclides enter the trench environment already in complexed form. In one documented case, complexing with an organic chelating agent has been linked to enhanced radionuclide migration.(3) Plutonium in the trench waters at the Maxey Flats, KY, disposal site may exist primarly as complexes of the tetravalent ion with strong organic chleating agents, such as ethylenediaminetetraacetic acid (EDTA).(4)

Samples of water from selected disposal trenches at Maxey Flats, as well as the experimental trenches, were analyzed for uncomplexed EDTA, DTPA, and NTA. Analyses of trench water samples from the West Valley, NY, disposal facility containing high DOC were also performed. Water samples from the Barnwell, SC, and Sheffield, IL, sites, selected on the basis of the presence of activity in these waters, were also analyzed.

Results show (Table 4) the contents of free chelating agents to be quite low, except for two trenches (Maxey Flats 19S and West Valley 3). These data represent the amounts of free chelating agents, rather than the total amount

Table 3

Tritium Content in Rainfall Samples Collected at the Maxey Flats, Kentucky, Disposal Site
[pCi/L ($\pm 2\sigma$%)][a]

Date	11E	13E	UA	UB
3/9/77-4/20/77	3.87 E3 (4.2%)	1.53 E5 (1.3%)	<7 E2	-----
4/13/77-5/17/77	6.36 E3 (12%)	2.62 E4 (19%)	2.01 E3 (35%)	-----
5/77-6/21/77	2.75 E4 (2.3%)	2.85 E4 (2.1%)	4.27 E4 (1.8%)	-----
6/21/77-7/20/77	1.08 E4 (4.1%)	1.65 E4 (3.2%)	1.08 E4 (4.2%)	-----
7/20/77-8/25/77	2.99 E3 (13%)	4.96 E4 (1.8%)	1.47 E3 (20%)	-----
8/25/77-9/21/77	6.29 E3 (5.8%)	7.75 E3 (4.9%)	3.46 E3 (10%)	-----
9/21/77-10/14/77	7.28 E3 (5.3%)	4.36 E3 (9.0%)	<7 E2	-----
10/14/77-11/22/77	1.30 E4 (3.9%)	7.90 E3 (5.1%)	1.53 E4 (3.3%)	-----
11/22/77-12/20/77	1.04 E5 (<1)	4.68 E4 (1.7%)	4.91 E3 (8.1%)	-----
12/20/78-3/20/78	1.72 E4 (3.5%)	1.05 E5 (<1)	-----	4.6 E4 (2.0%)
3/22/78-4/19/78	4.28 E4 (1.9%)	5.55 E4 (1.6%)	1.65 E4 (3.0%)	6.96 E4 (1.4%)
4/19/78-5/15/78	8.66 E4 (1.2%)	4.93 E4 (1.8%)	1.70 E3 (40%)	2.74 E5 (<1%)
5/15/78-6/16/78	2.90 E4 (2.1%)	1.83 E5 (1.1%)	8.81 E4 (2.8%)	3.63 E4 (1.2%)
6/16/78-7/12/78	4.11 E3 (7.5%)	2.20 E4 (2.7%)	3.71 E3	7.41 E3 (6.0%)
7/12/78-8/9/78	4.25 E3 (3.8%)	3.38 E3 (10.1%)	5.91 E2 (38.0%)	-------
8/9/78-9/5/78	5.71 E3 (6.0%)	6.50 E3 (5.5%)	2.63 E2 (86%)	3.12 E3 (9.9%)
9/5/78-10/5/78	3.82 E3 (10.2%)	5.28 E3 (6.3%)	<2.4 E2	2.92 E3 (10.6%)
10/5/78-11/9/78	8.70 E3 (4.5%)	7.50 E3 (5.2%)	6.54 E3 (6.1%)	2.72 E4 (2.6%)
11/8/78-12/5/78	1.03 E5 (<1%)	1.73 E4 (2.9%)	2.69 E4 (2.2%)	4.73 E4 (1.7%)
12/5/78-2/28/79	2.2 E4	6.3 E4	1.5 E4	1.6 E4
2/28/79-3/28/79	6.25 E3 (5.6%)	1.57 E5 (1.1%)	9.07 E3 (4.7%)	2.58 E3 (7.7%)
3/28/79-4/23/79	4.27 E4 (2.2%)	4.84 E4 (2.0%)	1.79 E4 (3.4%)	7.16 E3 (5.3%)
4/23/79-5/22/79	1.02 E4 (8.5%)	7.14 E3 (11.6%)	2.92 E3 (27%)	5.00 E3 (14%)
5/22/79-6/19/79	3.82 E4 (2.7%)	2.86 E4 (3.2%)	3.46 E3 (14%)	1.14 E4 (5.8%)
6/19/79--8/2/79	<6.2 E2	3.13 E3 (19%)	4.50 E3 (16%)	<6.2 E2
8/2/79-8/28/79	3.40 E3 (19%)	8.16 E2 (73%)	<6 E2	7.93 E2 (75%)
8/29/79-9/28/79	5.57 E3 (12%)	8.85 E4 (1.6%)	1.81 E5 (1.1%)	2.87 E4 (3.3%)

[a] Number in ( ) = 2σ% counting uncertainty.

of chelating agents present in the sample. The feasibility of analyzing these waters for total amounts of chelating agents present is being investigated. Such information is necessary before the extent of radionuclide complexing in the trench waters can be assessed.

It is difficult to form substantial conclusions about the existence of organo-radionuclide complexes on the results for the reported gel filtration experiments.(8) An apparent cesium-EDTA association is purely circumstantial. The gel columns are designed to separate components based on size alone, and since ion exchange reactions occur with $^{60}Co^{+2}$, an unequivocal interpretation of the result is not possible.

Because the trench waters used in the gel filtration experiments were oxidized samples rather than the anoxic chemical regimes present in the trench waters in situ, the observed results may be controlled by the chemical regime of the oxidized water and not representative of the anoxic systems. This possibility cannot be discounted because air exposure does produce changes in the trench water chemistry.

Table 4

Concentration of Free Chelating Agents in Selected Disposal Trenches

Disposal Site	Date Sampled	Trench	Free Chelating Agent[a] (microgram/gram)		
			DTPA	EDTA	NTA
Maxey Flats, KY	10-79	19S	12	1	<0.01
Maxey Flats, KY	10-79	27	0.20	<0.01	<0.01
Maxey Flats, KY	5-78	27	2.0	0.06	<0.01
Maxey Flats, KY	10-79	32	0.10	0.25	0.03
Maxey Flats, KY	10-79	30	<0.02	<0.01	<0.01
Maxey Flats, KY	10-79	T-5	<0.02	<0.01	<0.01
Maxey Flats, KY	10-79	T-4E	0.20	<0.01	<0.01
Maxey Flats, KY	10-79	T-3W	0.10	<0.01	<0.01
Maxey Flats, KY	10-79	T-2E	0.12	<0.01	<0.01
Maxey Flats, KY	10-79	T-2E (anoxic)[b]	0.12	<0.01	<0.01
West Valley, NY	10-78	3	28	1.6	0.08
West Valley, NY	10-78	5	1.2	0.10	<0.01
West Valley, NY	10-78	8	0.40	0.10	0.06
Barnwell, SC	3-79	25/21D1	<0.02	<0.01	0.03
Sheffield, IL	4-79	18A	<0.02	<0.01	<0.01

[a] Analyses performed by LFE Laboratories, Richmond, CA.

[b] All experimental trench waters were collected under oxic conditions except where noted.

Figures 2 and 3 illustrate the stability of Co-EDTA and Co-NTA complexes in the controlled environment chamber using a synthetic water of a composition similar to Maxey Flats trench 32 and ultrasonically disaggregated Maxey Flats shale. Experimental details are reported elsewhere.(9) Both systems show a rapid decrease in metal ion concentration in solution followed by an asymptotic approach to steady state conditions. The decreases may simply be due to sorption of the complex onto the shale. When a trace metal is in complexed form its potential for sorption is considerably reduced, however, a small amount of sorptive capacity is still possible.(9) The decrease in cobalt concentration for the NTA system was greater than that for the EDTA system. This may be due to the fact that NTA is much more biodegradable.(12) Neither of these experiments showed the precipitous drop in trace metal concentration under strongly reducing conditions shown by Zn and Cu in the work of Reddy and Patrick.(11) After apparent steady state conditions were established in the chamber containing the Co-EDTA complex, an aqueous solution of manganese chloride was added, such that the cobalt and manganese were in equimolar concentrations. This was done to test the displacement chelation effect proposed by Lindsay(13) to explain the results of

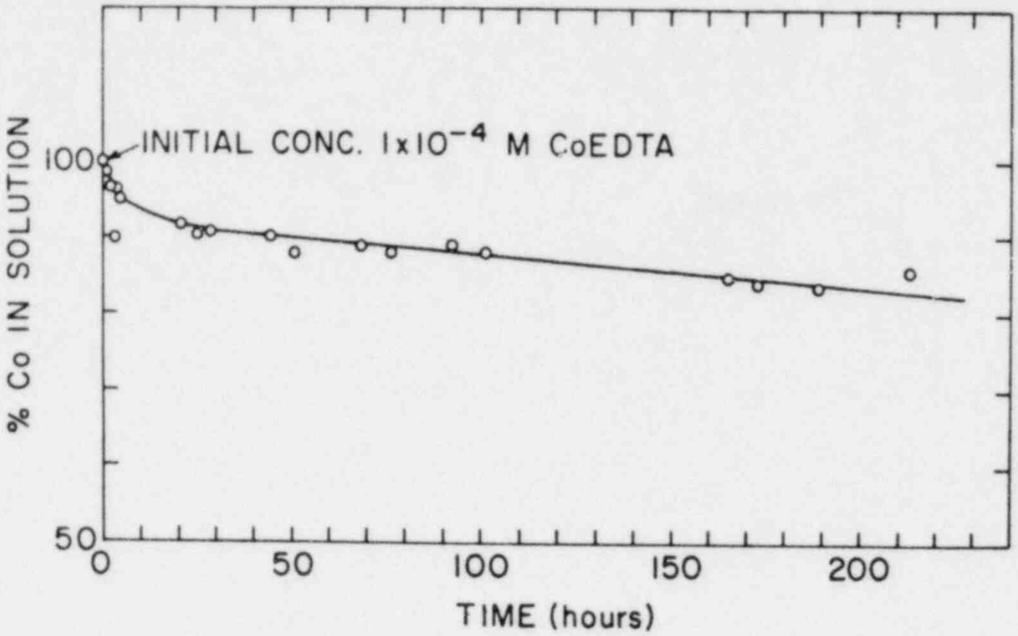

Figure 2 Controlled environment Co-EDTA stability under reducing conditions. Maxey Flats shale and synthetic trench 32 water.

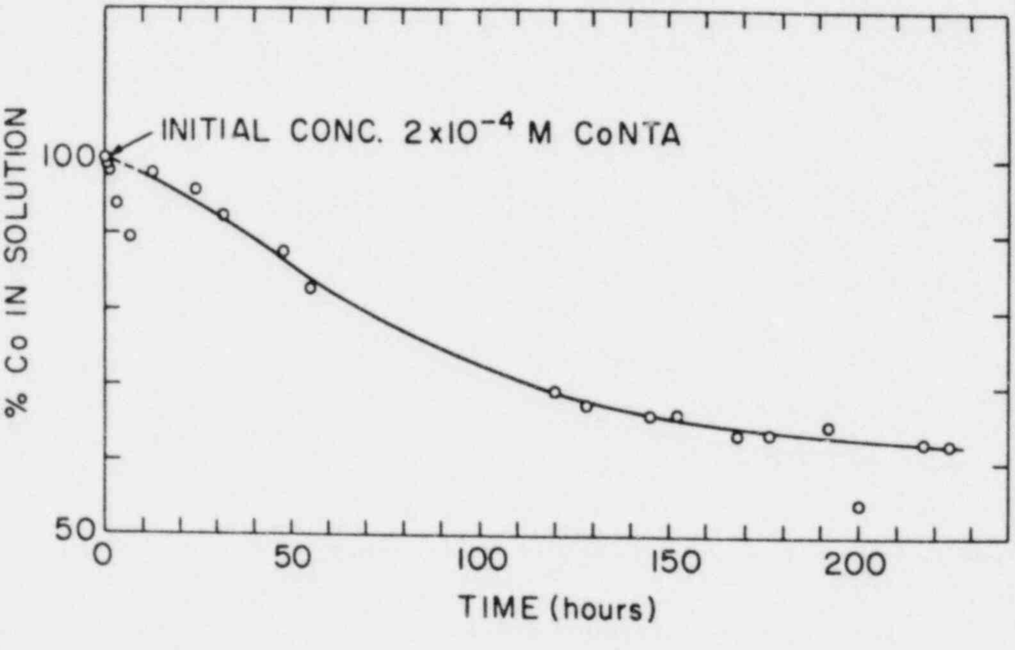

Figure 3 Controlled environment Co-NTA stability under reducing conditions. Maxey Flats Shale and synthetic trench 32 water.

Reddy and Patrick.(11) Results are shown in Figure 4. Here again only a small (approx. 15%) decrease in Co concentration was observed rather than a precipitous one.

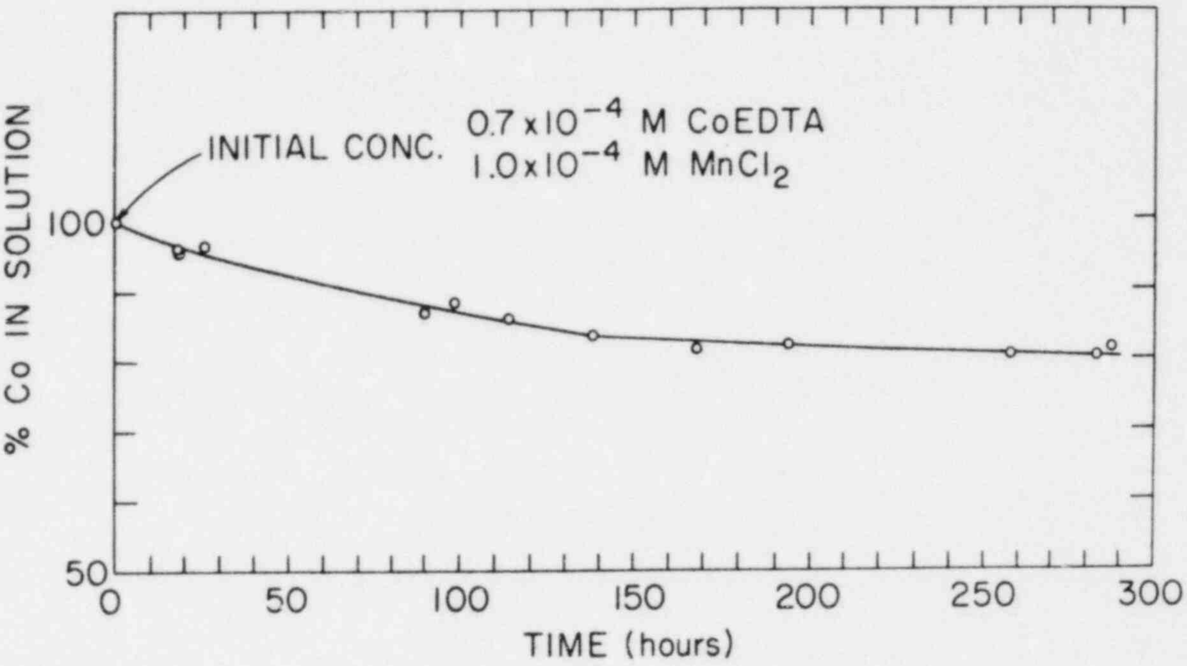

Figure 4 Controlled environment Co-EDTA stabililty (after addition of $MnCl_2$) under reducing conditions. Maxey Flats shale and synthetic trench 32 water.

These results surprise the authors in that cobalt was anticipated to show behavior similar to that of copper, because both are transition metals. To determine if the solubility of CoS was not exceeded in the experiment, the stability constants of Co-EDTA and Co-NTA complexes(13,14) were used to determine the amount of uncomplexed metal ion in solution. This value was then used in the solubility product for CoS, where the sulfide ion ($S^=$) concentration was calculated from the sulfide electrode calibration curve given by Berner.(15) The calibration curve had been verified in previous laboratory work with these electrodes. Results showed that the system in the controlled environment chamber was not over saturated with respect to CoS. This is in contrast to the calculations referred to by Reddy and Patrick for the systems containing Cu and Zn. On the basis of our calculations, a precipitous decrease in trace metal concentration would not be expected in contrast to the Zn and Cu results. However, these theoretical calculations are limited by the reliability of stability constants and the complexity of the real system. The calculations used here ignore the effect of other complexes in solution, which would raise the amount of free metal in solution.

The results shown here indicate that cobalt complexes with EDTA and NTA persist in strongly reducing anaerobic water environments typical of burial trenches, where anoxic water conditions develop. The geochemical system has only a relatively minor ability to remove the cobalt from solution in contrast to the effect reported by Reddy and Patrick for Zn and Cu in similar environments. The effect of added manganese appears to be insignificant in contrast to the displacement chelation argument proposed by Lindsay.(13) Therefore, the experiment described by Reddy abd Patrick is being repeated using a spiked Zn-EDTA complex under conditions analogous to those used in their study. A sharp drop in Zn concentration should be observed if there is a fundamental difference in the nature of cobalt complexation compared with that of Zn and Cu. If not, there is a significant difference in the geochemical environments of the experiments.

3.5 Radionuclide Sorption

3.5.1 Effects of Complexing Agents

Organic chleating agents, such as EDTA, NTA, and DTPA are known to increase the solubility of many metals due to the formation of stable metal chelate complexes. In solution, complexing markedly affects the extent of metal adsorption. A series of scoping experiments were performed to ascertain the effect of EDTA on radionuclide sorption. Other organic substances included in this study, which could act as complexing agents, were a decontamining agent, oxalic acid, and solvents used as extractants, which had been identified in trench waters. The effect of these organic substances on the sorption of radionuclides could not be anticipated. Water from Maxey Flats Well UB1-A, a groundwater relatively free of the organic substances found in the disposal trench waters, was used in these experiments. Results indicate that organic complexing agents exert different effects for specific radionuclides (Table 5). All the organics decreased the ^{241}Am sorption; EDTA produced the most drastic change.

Strontium and cobalt showed little decrease in K_d, except in the EDTA system. A decrease of an order of magnitude was observed for ^{60}Co in the EDTA system. Migration of cobalt away from waste disposal pits at Oak Ridge(3) has been attributed to increased mobility due to EDTA complexing. Complexing by EDTA appears to be a potentially important mechanism for increased mobility of some radionuclides in the systems investigated.

Cesium sorption appears unaffected by the added organics. Alkali metals show relatively little affinity towards the formation of complexes,(16-18) and the cesium results are consistent with this pattern.

A preliminary study was performed to determine the effect of EDTA on retention of radionuclides by various commonly encountered soil forming minerals. Clay minerals, particularly the expandable 2:1 layer silicates, exhibit the ability to adsorb organic molecules.(19) Samples of the major classes of clay minerals were used along with limonite, a common iron bearing soil component. Distribution coefficients were measured with and without the addition of EDTA to the well water-mineral system. Results of this study are

Table 5

Effect of Organic Substances on Radionuclide Sorption Coefficients
(Water and Shale Collected at the Maxey Flats, Kentucky, Disposal Site)

		K_d(mL/g)[a]				
Isotope	Conc. (mg/L)	Oxalic Acid	TBP[b]	THF	MIBK	EDTA
^{241}Am	0	1.2 ± 1.5 E6	1.2 ± 1.5 E6	1.2 ± 1.5 E6	1.2 ± 1.5 E6	1.2 ± 1.5 E6
	2	1.6 E5	----	----	8.0 ± 3.0 E4	3.0 ± 4.0 E5
	10	2.9 ± 1.5 E5	----	1.9 ± 1.0 E5	----	3.0 ± 2.0 E4
	100	----	8.3 E4	----	1.6 E5	3.0 ± 3.0 E3
^{85}Sr	0	3.9 ± 0.2 E0	3.9 ± 0.2 E0	3.9 ± 0.2 E0	3.9 ± 0.2 E0	3.9 ± 0.2 E0
	2	2.2 ± 0.1 E0	----	----	3.1 ± 0.3 E0	5.2 ± 0.0 E0
	10	1.2 E0	----	1.6 ± 1.0 E0	4.4 ± 0.4 E0	2.4 ± 1.3 E0
	100	4.1 ± 0.2 E0	2.1 ± 0.1 E0	3.2 ± 1.4 E0	2.4 ± 0.3 E0	1.7 E0
^{134}Cs	0	5.2 ± 0.3 E3	5.2 ± 0.3 E3	5.2 ± 0.3 E3	5.2 ± 0.3 E3	5.2 ± 0.3 E3
	2	4.8 ± 0.7 E3	----	----	5.2 ± 0.7 E3	5.5 ± 0.9 E3
	10	4.5 ± 0.1 E3	----	5.1 ± 0.4 E3	----	5.1 ± 0.6 E3
	100	4.9 ± 0.1 E3	4.9 ± 0.2 E3	5.3 ± 0.6 E3	5.8 ± 0.5 E3	4.7 ± 1.4 E3
^{137}Cs	0	5.3 ± 0.2 E3	5.3 ± 0.2 E3	5.3 ± 0.2 E3	5.3 ± 0.2 E3	5.3 ± 0.2 E3
	2	4.4 ± 0.3 E3	----	----	5.1 ± 0.1 E3	5.4 ± 0.2 E3
	10	4.3 ± 0.1 E3	----	5.0 ± 0.2 E3	----	4.6 ± 0.3 E3
	100	5.3 ± 0.1 E3	5.0 ± 0.1 E3	5.3 ± 0.1 E3	4.6 ± 0.3 E3	4.7 ± 0.1 E3
^{60}Co	0	4.1 ± 0.7 E2	4.1 ± 0.7 E2	4.1 ± 0.7 E2	4.1 ± 0.7 E2	4.1 ± 0.7 E2
	2	4.7 E2	----	----	3.4 ± 0.1 E2	1.9 ± 0.1 E2
	10	5.7 ± 0.4 E2	----	4.2 ± 0.2 E2	----	2.2 ± 0.2 E1
	100	3.1 ± 0.1 E2	5.0 ± 2.0 E2	4.3 ± 0.4 E2	4.8 ± 0.1 E2	1.5 ± 0.9 E0

Water: Well UB1-A collected at the Maxey Flats, Kentucky, disposal site, May 1978.
Shale: Nancy Shale, Maxey Flats, Kentucky, disposal site, trench 46 at 20 foot depth.
TBP: Tributyl phosphate.
THF: Tetrahydrofuran.
MIBK: Methyl-isobutyl ketone.
EDTA: Ethylenediaminetetraacetic acid tetrasodium salt dihydrate.
Reaction containers: Polystyrene test tubes.
Solution/soil ratio: 20 mL/g.
Equilibration time : 250 hours.

[a] K_d is reported as an average ± one standard deviation of observed results on a 10-mL aliquot of the liquid phase: $K_d \pm$ s.d. $= \bar{a} \times 10^b \pm c \times 10^b = a \pm c$ Eb.

[b] A saturated solution.

presented in Table 6. When EDTA was added to the trench water, the observed K_d generally decreased for all the minerals, with the notable exception of the montmorillonite (nontronite) clay. An increase in K_d was actually observed for americium in the montmorillonite system. Adsorption of organic molecules (particularly polar molecules) by montmorillonite type clays is well known.(19) These results suggest that montmorillonite clays in the soil around disposal trenches would tend to counteract the increased migration potential of organo-radionuclide complexes. Montmorillonite type clays could also be used as disposal trench linings to enhance the retention capacity of the geologic media. Additional studies of this type would be necessary to determine which members of the montmorillonite family of clay minerals are most effective in this regard. These results can be used in formulating siting criteria, based on the mineralogy of a potential burial site, and improving burial procedures.

Table 6

Effect of EDTA on Radionuclide Sorption Coefficients
(Water Collected at the Maxey Flats, Kentucky, Disposal Site, and Soil Forming Minerals)

Isotope	EDTA (mg/L)	K_d(mL/g)[a]					
		Shale	Moncmorillonite (Nontronite)	Kaolinite	Illite	Limonite	Vermiculite
^{241}Am	0	2.2 E6	5.6 ± 0.4 E1	1 E 7	8.3 ± 3.8 E 4	1 E 7	6.0 ± 0.3 E2
	100	3.3 ± 3.4 E3	8.4 ± 0.1 E1	7.0 ± 0.4 E 0	4.4 ± 0.6 E 2	1.2 ± 0.7 E 3	9.1 ± 0.9 E1
^{85}Sr	0	3.9 ± 0.2 E0	1.8 ± 0.2 E1	1.3 E 0	3.9 ± 0.4 E 0	1.1 ± 0.1 E 1	9.0 ± 2.0 E0
	100	1.7 E0	1.9 ± 0.1 E1	4.0 E-1	3.0 ± 2.0 E 0	8.1 ± 0.2 E 0	2.6 ± 0.8 E0
^{134}Cs	0	5.2 ± 0.3 E3	1.7 ± 0.2 E2	1.1 ± 0.1 E 1	4.6 ± 0.7 E 3	1.3 ± 0.1 E 3	1.1 ± 0.1 E3
	100	4.7 ± 1.4 E3	1.4 ± 0.1 E2	1.0 ± 0.1 E 1	4.4 ± 1.3 E 3	1.2 ± 0.1 E 3	1.0 ± 0.1 E3
^{137}Cs	0	5.3 ± 0.2 E3	1.6 ± 0.2 E2	1.1 ± 0.1 E 1	4.6 ± 0.3 E 3	1.3 ± 0.1 E 3	1.1 ± 0.1 E3
	100	4.7 ± 1.4 E3	1.4 ± 0.1 E2	1.1 ± 0.1 E 1	4.9 ± 0.2 E 3	1.2 ± 0.1 E 3	9.8 ± 0.3 E2
^{60}Co	0	4.1 ± 0.7 E2	9.7 ± 0.5 E0	2.0 ± 0.1 E 1	6.8 ± 1.9 E 2	2.8 ± 0.6 E 2	3.2 ± 0.1 E2
	100	1.5 ± 0.9 E0	1.1 ± 0.1 E1	2 E-1	5.0 ± 7.0 E-1	2.0 ± 2.0 E-1	1 E0

Water: Well UB1-A collected at the Maxey Flats, Kentucky, disposal site, May 1978.
Shale: Nancy Shale, Maxey Flats, Kentucky, disposal site, trench 46, at 20 foot depth.
Equilibration time: 250 hours.
Solution/Soil ratio: 20 mL/g.
Reaction container: Polystyrene test tube.

[a] K_d is reported as an average ± one standard deviation of observed results on a 10-mL aliquot of the liquid phase.

$K_d \pm s.d. = a \times 10^b \pm c \times 10^b = a \pm c$ Eb.

3.5.2 Effect of Radionuclide Concentration

Knowledge of the mobility of radionuclides from disposal trenches along the groundwater flow path is a prime concern in the evaluation of a waste burial site performance. Correlation between batch and column methods, commonly used to measure the degree of radionuclide retardation, is desirable but unfortunately not straightforward. Several chemical and physical phenomena could strongly affect the K_d results, e.g., precipitation, colloid formation, wall adsorption, and reaction kinetics. Very low concentrations of radionuclides would be anticipated as mobile species along groundwater flow paths from the disposal sites. However, much higher concentrations of radionuclides are used in the laboratory to facilitate experimental work. Consequently, it is necessary to determine K_d at several radionuclide concentrations, ideally differing by orders of magnitude, to minimize systematic errors. This was accomplished indirectly by using different solution to soil ratios.

Sorption experiments with materials from Maxey Flats wsere not completed in time for this report. However, similar sorption experiments completed with materials from the Barnwell, SC, site are presented here. Results for batch K_d with materials from Barnwell appear to be dependent on the ratio of soil and solution for some isotopes. The observed K_d behavior for ^{85}Sr is linear and nearly constant (Figure 5). However, the results for ^{241}Am are clearly nonlinear (Figure 6). In the range of soil/solution ratios appropriate for an intact core of Barnwell, South Carolina, Hawthorne sediment saturated with solution, the data is nearly linear but sharply dependent on the soil/solution ratio. The observation for ^{241}Am could be an artifact of the experimental method and needs to be further investigated.

3.5.3 Sorption Isotherms

Mathematical models for the prediction of radionuclide migration incorporate the concentration of radionuclide adsorbed by soil or rock.(20) The functional relationship between the concentration of radionuclide in solution and the amount adsorbed is the adsorption isotherm.(21)

The Langmuir adsorption isotherm is:

$$\frac{C_{eq}}{x/m} = \frac{C_{eq}}{S} + \frac{1}{k\,S}$$

C_{eq} = activity in solution per unit volume
x/m = activity on soil per unit mass
S = constant
k = constant

but

$$K_d = \frac{x/m}{C_{eq}}$$

Consequently, a plot of $1/K_d$ vs C_{eq} is a plot equivalent to the Langmuir adsorption isotherm (Figure 7, for the ^{85}Sr experiment with Barnwell materials).

Sorption experiments using anoxic trench waters and shale from the Maxey Flats site for several solution/soil ratios are in progress. The radionuclide sorption from solution appears to have reached a steady state (Figure 8). The samples will be disassembled in a glove box containing ultra-pure argon; the pH and Eh of each sample will be measured. A final K_d will be determined from a filtered aliquot of solution. Sorption isotherms for the radionuclides will then be evaluated.

4. GENERAL CONCLUSIONS

During the early stages of this investigation, the objective of the project was simply to gather descriptive data concerning disposal trench water compositions. This information was to be used as source term data for input

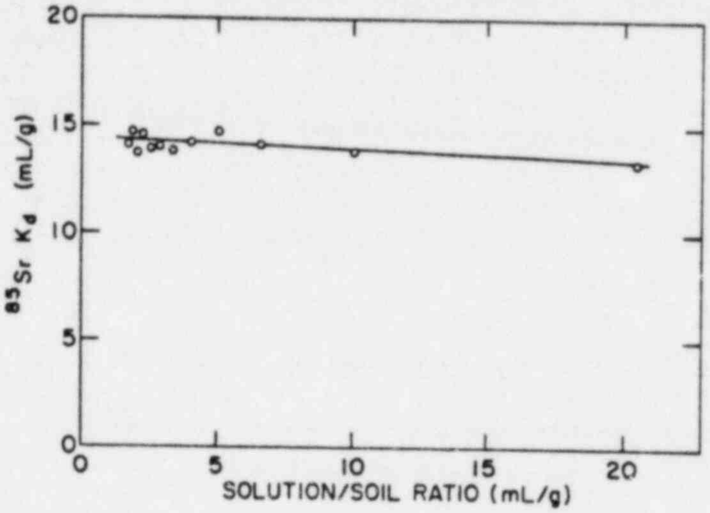

Figure 5 ^{85}Sr K_d vs solution/soil ratio-Barnwell sediment sediment and trench water-6D1 (contact time 119 days).

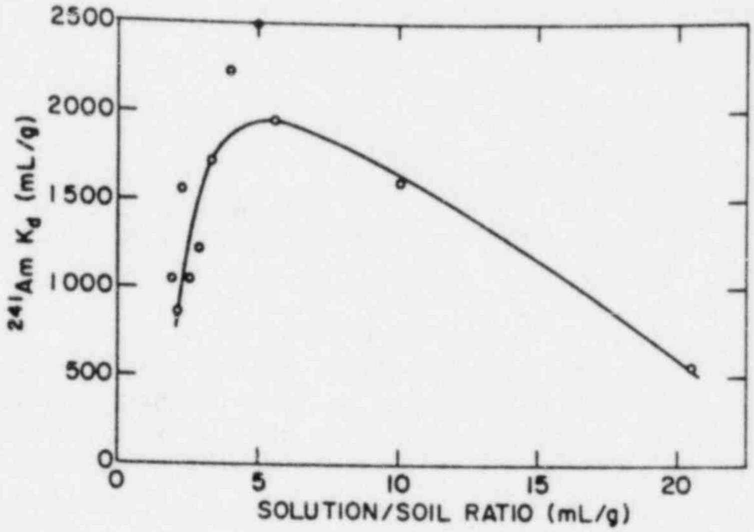

Figure 6 ^{241}Am K_d vs solution/soil ratio-Barnwell sediment ratio-Barnwell sediment and trench water-6D1 (contact time 119 days).

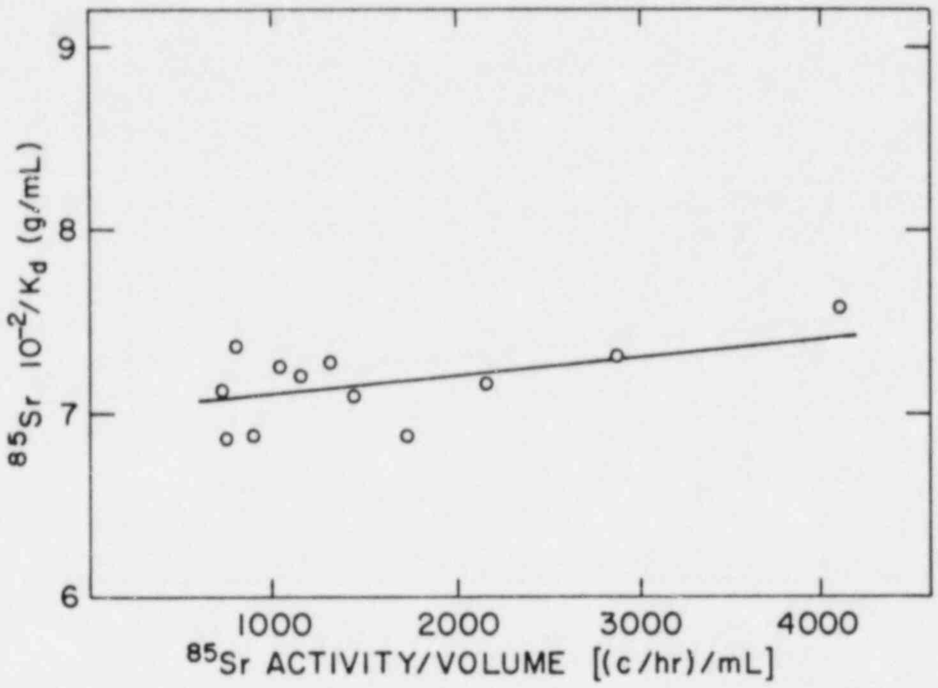

Figure 7 Langmuir adsorption isotherm for ^{85}Sr sorption on Hawthorne sediment from Barnwell trench 6D1 water.

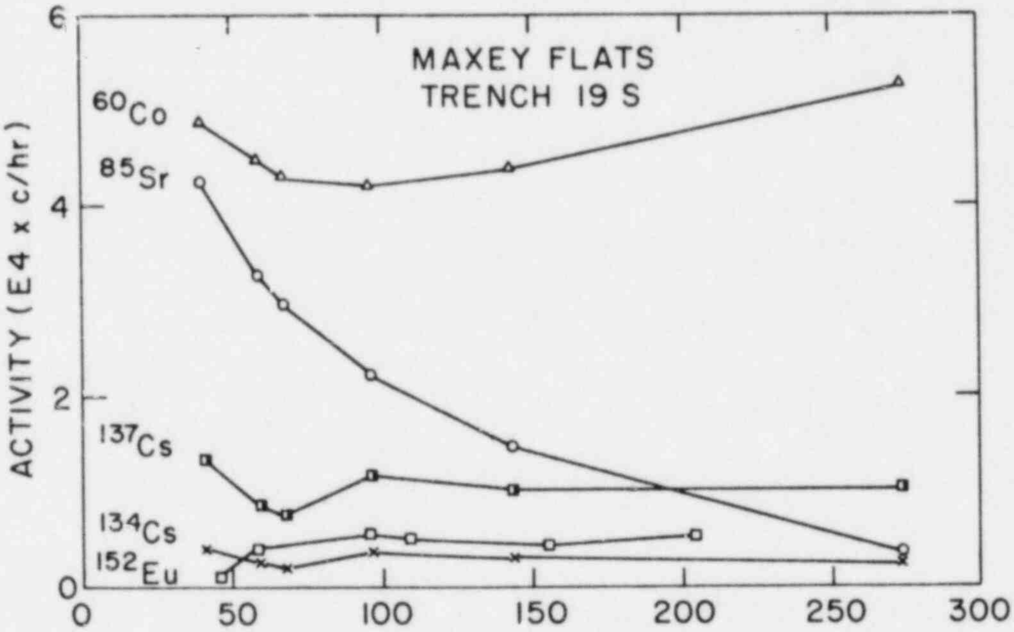

Figure 8 Activity remaining in solution in trench 19S water vs contact time with shale from Maxey Flats.

into modeling efforts. Routine sampling and analysis of disposal trench waters appear to have generated sufficient information to characterize the geochemical regimes at the disposal sites studied. At this point, emphasis should be directed towards evaluating the role of water chemistry and sorption processes on the retention and migration of radionuclides in the geochemical environments typified by the anoxic trench waters from Maxey Flats and West Valley, as well as the relatively more oxidized waters at Barnwell. This approach serves two purposes; (1) by understanding the behavior of existing sites, appropriate remedial actions can be initiated to modify the geochemical environment to improve radionuclide retention where needed and, (2) when effective retention mechanisms are identified in the existing sites, these geochemical mineral-radionuclide associations can be used as siting criteria for the selection of future disposal sites.

The total retention capacity of a particular trench geochemical environment is the sum of all the mechanisms which can prevent radionuclide migration from the immediate trench vicinity. Sorption processes (based mainly on K_d studies) have been largely credited as the total retention mechanism. However, this is not correct, particularly for the anoxic, chemically reducing, geochemical environments typified by Maxey Flats and West Valley. From information collected in this program, two other mechanisms appear to have an effect on radionuclide retention; (1) precipitation of iron monosulfides (FeS) in the anoxic trench environment and (2) precipitation of iron oxyhydroxides as trench waters migrate away from the immediate trench vicinity and mix with more oxidized groundwaters. Field measurements have shown the presence of free sulfide ion in solution and black particulate matter in the Maxey Flats trench waters.(8) Active sulfate reducing bacteria have also been identifed in trench waters.(2) Exposure of iron bearing trench waters to air results in the precipitation of ferric oxyhydroxide and causes drastic changes in the water chemistry. Coprecipitation of radionuclides during both sulfide and hydroxide precipitations is expected. In fact, the ferric oxyhydroxide precipitation reaction is a well known water purification technique. Both mechanisms act as in situ scavenging mechanisms and increase the <u>total</u> retention capacity of the anoxic, chemically reducing geochemical environments. This is an important point to be kept in mind when considering these environments for future disposal sites. Results of sorption studies involving these systems showed that anoxic water regimes exhibited lower sorption capacities than more oxidized systems. However, the retention in such a situation would be the sum of that due to sorption processes and co-precipitation reactions. To date, no quantitative data are available from this program or other studies to evaluate the relative importance of these mechanisms. However, they should play major roles based on general chemical principles.

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SECTION V

POTENTIAL FOR MIGRATION OF WASTE RADIONUCLIDES AT THE MAXEY FLATS, KY SHALLOW-LAND BURIAL SITE

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Potential for Migration of Waste Radionuclide at the Maxey Flats, KY Shallow-Land Burial Site[1]

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Summary

The Los Alamos National Laboratory (Los Alamos) and the University of California (UCB) in a cooperative project are studying the movement of water into trenches and water and radionuclide movement out of trenches at the Maxey Flats, KY shallow-land burial site. Those trenches include a waste trench and an experimental trench. In addition to the field studies, Los Alamos is conducting laboratory investigations using Maxey Flats waste and soils. The results from those investigations are being compared with the results of the field investigations to determine whether laboratory results can be used to predict radionuclide migration at existing as well as future burial sites of low-level radioactive waste.

Porous cups were implanted in the soil near the Waste Trench 19S and between Trench 27 and the experimental trench. Those porous cups allow the in situ sampling on a seasonal basis of soil solutions at various depths and distances from those trenches.

The distribution of ^{3}H in the soil solutions collected from the porous cups suggests several pathways by which soil water moves at the Maxey Flats burial site; a) interstitial flow, b) preferential flow along a sandstone layer, and c) preferential flow along a landfill - original soil interface. Liquid waste movement from Waste Trench 19S into the adjacent soil occurs by the first and second pathways.

The distribution of radionuclides in soil solutions indicates that waste ^{3}H

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[1] Work performed for the Nuclear Regulatory Commission under Research Order No. 60-81-171.

moved the greatest distance away from Waste Trench 19S and waste ^{137}Cs the least distance; the relative movement of waste ^{238}Pu and ^{60}Co lies between that of ^{137}Cs and ^{3}H. Waste ^{3}H was detected along a sandstone layer to about 9 meters from the trench, the extent of the porous cup sampling at that depth; no ^{137}Cs was detected in soil solutions from the same area. Plutonium-238 and ^{60}Co were detected at about 2 meters from the trench.

The sorption of ^{137}Cs and ^{238}Pu as indicated by laboratory studies agrees well with their observed distribution in the Waste Trench 19S area; the sorption of ^{60}Co does not agree as well as the sorption of ^{238}Pu.

The collection and analysis of soil solutions will continue in order to evaluate the effects of changes in soil moisture and changes in the chemical and physical properties of those solutions on the movement of waste radionuclides through soil. Onsite lysimeter studies will be conducted in cooperation with UCB. Radionuclides from a single liquid waste source (Waste Trench 19S sump) will be used in the lysimeters. The single source will remove the uncertainies of interpretation caused by multiple sources occurring in the trench area. Also, the significance of preferential flow of liquid waste from burial trenches through soil along the interface of landfill and the underlying original soil profile will be explored.

Introduction

Solid low-level radioactive waste buried at shallow-land burial sites comes into contact with water either from precipitation which has infiltrated the soil or from decomposition of buried waste. At least a portion of the waste radionuclides will dissolve, thus creating a potential for migration of those radionuclides into the nearby soil and shallow ground water and perhaps ultimately into adjacent streams.

The Los Alamos National Laboratory (Los Alamos) and the University of California, Berkeley (UCB) are studying the movement of water into trenches and water and radionuclide movement out of trenches at the Maxey Flats, KY shallow-land burial site. An understanding of the pathways of water and radionuclide migration should aid in determining optimum ways of minimizing waste radionuclide transport. The results of this study are applicable not only to existing burial sites, but also to designing and locating new burial sites. The cooperative project is a part of the NRC sponsored research program at Maxey Flats.

In addition to the field studies, Los Alamos is conducting laboratory investigations using Maxey Flats waste and soils to study the influence of waste-soil interactions on the mobility of waste radionuclides. The results from those investigations are being compared with the results of the field investigations to determine whether laboratory results can be used to predict radionuclide migration at existing as well as future burial sites of low-level radioactive waste.

The purpose of this report is to present data and their interpretation relative to water and radionuclide migration at Maxey Flats and is a continuation and expansion of earlier interpretations that were presented in 1980 (Polzer et al, 1981). For continuity some data presented in 1980 also are presented in this report.

Materials and Methods

The field studies are being conducted at an experimental trench area and at the Waste Trench 19S area as shown in Figure 1. Porous cups were implanted in the soil for sampling of soil solutions under both saturated and unsaturated conditions. This method of sampling allows the collection of soil solution under in situ conditions, e.g. anoxic conditions. In 1979 porous cups were implanted in the northeast wall of section four (T4) of the experimental trench prior to backfilling and capping of that trench. In 1980 additional porous cups were implanted between Waste Trench 27 and sections T3, T4 and T5. Sampler locations are shown in Figure 2. A total of 35 porous cups was implanted at the experimental trench area. At the Waste Trench 19S area an array of 11 holes was instrumented in 1979 with 27 porous cups. An additional 24 holes were instrumented in 1980 with 62 porous cups. The locations of the holes at the Waste Trench 19S area are shown

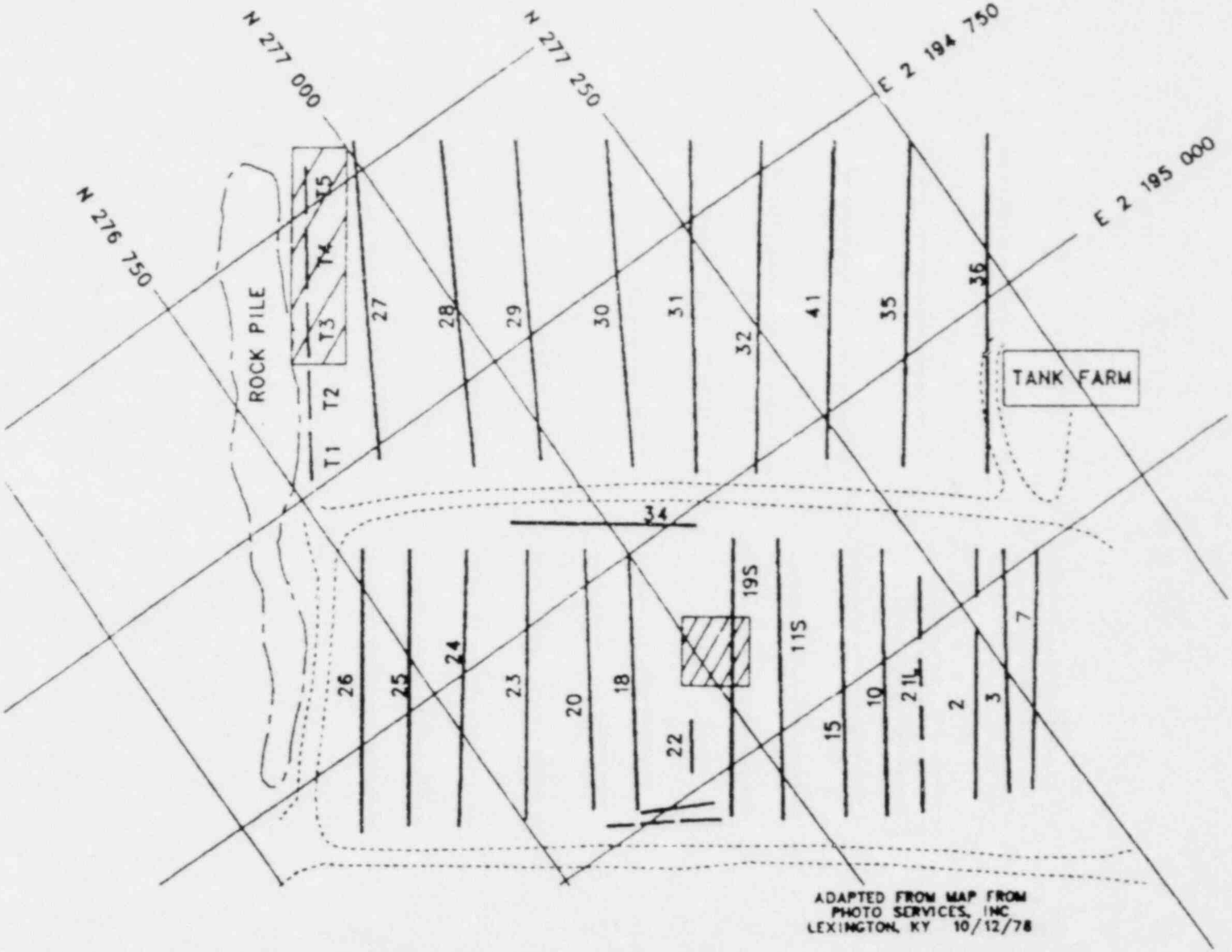

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Fig. 1. The approximate locations of the burial trenches (heavy lines) are shown relative to the study areas at Waste Trench 19S and the experimental trench (T1 through T5) adjacent to Waste Trench 27. Los Alamos and UCB sample arrays are located within the hatched areas.

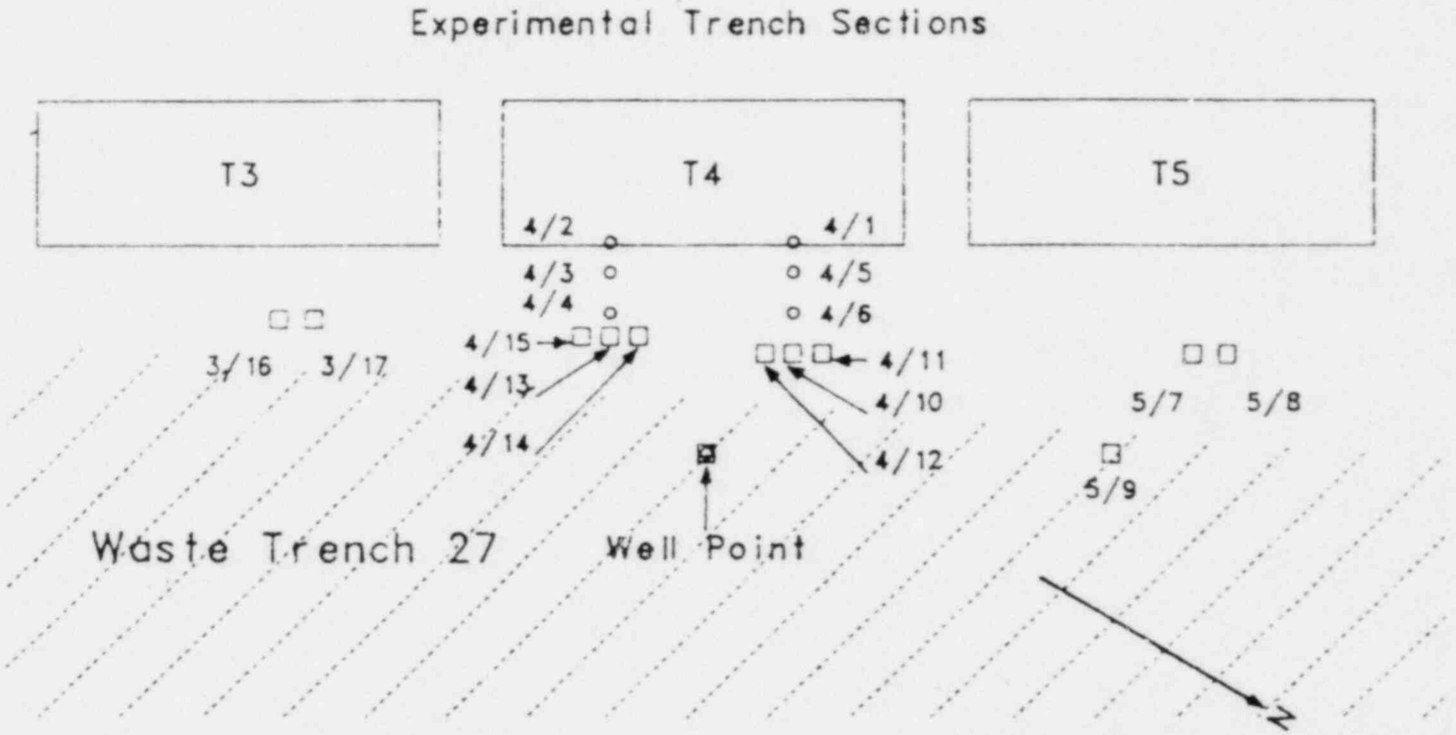

Fig. 2. A plan view of sampling locations near sections 3, 4 and 5 of the experimental trench is shown schematically. The locations of porous cup samplers installed in 1979 are coded by open circles; locations of those installed in 1980 are coded by open squares. The depths at which the samplers were installed are given in Table I. The hatched area in Fig. 1 represents this portion of the experimental trench area.

in Figure 3. The trench edge is not well defined; therefore, the location of holes relative to the trench is only approximate. The depths at which the cups were installed ranged from 0.5 to 8.1 meters at both the experimental trench area and at the Waste Trench 19S study area. The depths at which the cups were installed are given in Tables I and II.

The porous cups are sampled for soil solutions approximately four times a year to coincide, in general, with specific seasons. The soil solutions are analyzed for physical and chemical characteristics, e.g. pH, electrical conductivity and alkalinity. Those characteristics will be published at a later date. Analysis of radionuclides include ^{3}H, ^{238}Pu, $^{239,240}Pu$, $^{233,234}U$, ^{238}U, ^{137}Cs and ^{60}Co. Details of the sampling and analytical procedures are given by Fowler, et al (1981).

Results and Discussion

Potential Water Movements

The following tritium data reflect possible water movements at the burial site and are representative of the data collected in our field studies. Levels of tritium in soil moisture at the experimental trench area are presented in Table III for two depths, 0-3 meters and 3-7 meters. The upper three meters is disturbed soil from onsite trench excavations, the 3-7 meter depth is the original undisturbed soil profile. The low levels of tritium in the lower zone suggest limited moisture movement from the old landfill into the original soil profile. One explanation may be that percolating soil moisture moves predominately along the landfill/soil interface rather than penetrating the original soil profile as depicted in Figure 4. If such an interface were connected to a trench and if the water level were above the interface, then such an interface would provide a favorable pathway for waste radionuclides to migrate from the trench area. The same zone may also act as a preferential means of transporting surface water into trenches.

In general the tritium levels in soil solutions from porous cups located at

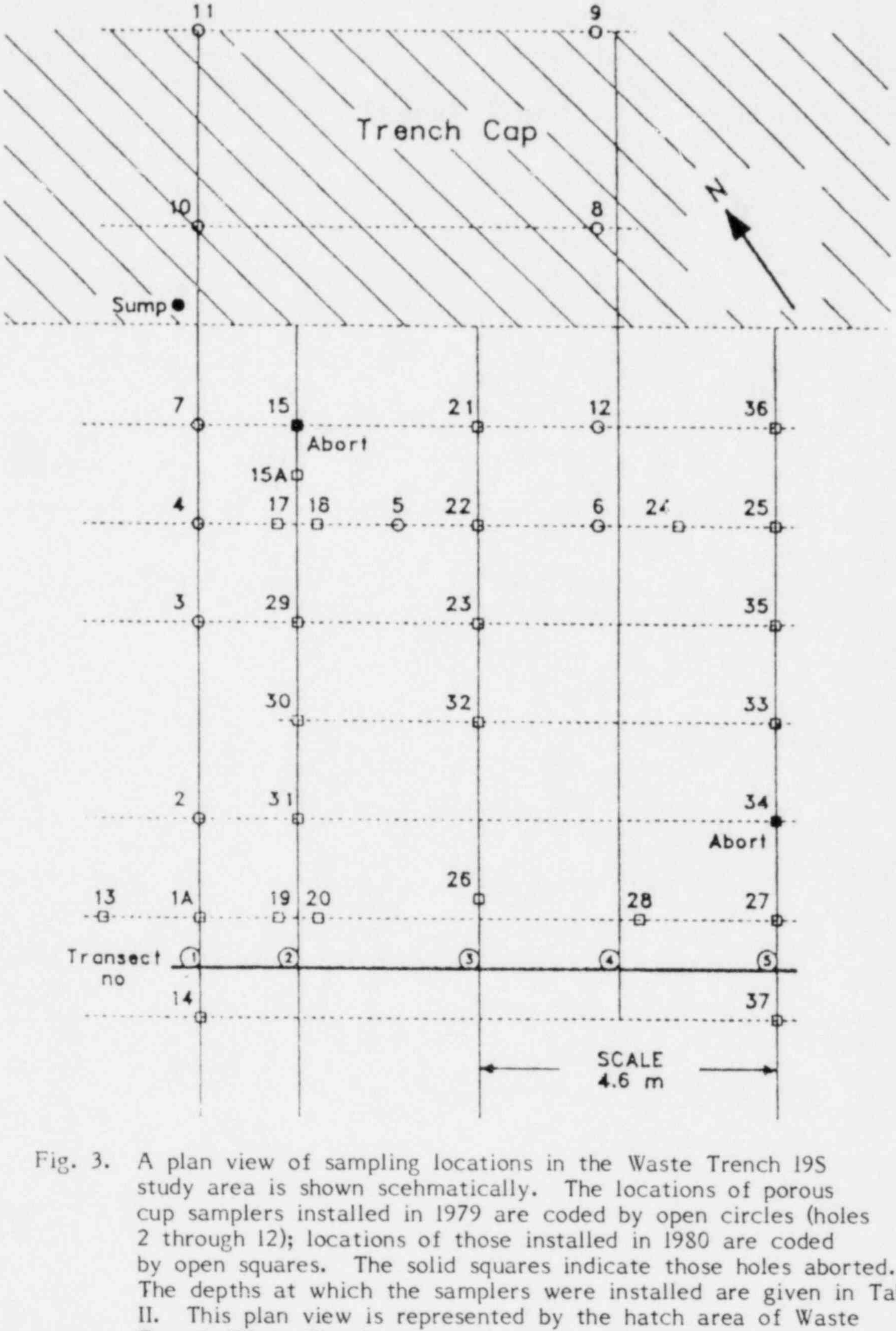

Fig. 3. A plan view of sampling locations in the Waste Trench 19S study area is shown scehmatically. The locations of porous cup samplers installed in 1979 are coded by open circles (holes 2 through 12); locations of those installed in 1980 are coded by open squares. The solid squares indicate those holes aborted. The depths at which the samplers were installed are given in Table II. This plan view is represented by the hatch area of Waste Trench 19S in Fig. 1.

Table I

The number of porous cup samplers and the depth of their installation at the experimental trench area are presented with respect to the trench section and the hole designation. A plan view of the hole location is given in Fig. 2.

Trench Section	Hole Designation	Depth of Drilled Hole (m)	Number of Cups	Depth of Cups (m)
5	5/7	7.6	2	2.5, 6.7
	5/8	4.6	2	1.2, 4.6
	5/9	2.7	2	1.2, 2.7
4	4/1	7.6	3	4.0, 5.9, 7.6
	4/2	7.6	4	3.8, 5.3, 6.7, 7.6
	4/3	1.9	2	1.3, 1.9
	4/4	0.8	1	0.8
	4/5	1.7	2	1.2, 1.7
	4/6	0.8	1	0.8
	4/10	7.0	2	3.0, 7.0
	4/11	5.5	2	1.8, 5.5
	4/12	4.0	2	0.6, 4.0
	4/13	7.0	2	3.0, 7.0
	4/14	5.5	2	1.8, 5.5
	4/15	4.0	2	0.6, 4.0
3	3/16	6.9	2	2.9, 6.9
	3/17	4.6	2	1.8, 4.6

Table II

The number of porous cup samplers and the depth of their installation at Waste Trench 19S study area are presented with respect to the sampling transect and the hole designation. A plan view of the hole locations is given in Fig. 3.

Transect	Hole Designation	Depth of Drilled Hole (m)	Number of Cups	Depth of Cups (m)
1	11 (Trench Cap)	1.1	2	0.6, 1.1
	10 (Trench Cap)	1.2	2	0.6, 1.2
	7	3.1	2	1.3, 3.1
	4	2.2	3	0.6, 1.2, 2.2
	3	3.7	3	1.1, 2.5, 3.7
	2	3.7	3	1.2, 2.5, 3.7
	1A	2.1	2	1.2, 2.1
	14	1.6	1	1.2
2	15A	3.5	2	1.2, 2.4
	17	8.0	3	3.4, 5.5, 7.9
	18	1.2	1	1.2
	29	8.0	4	1.2, 3.4, 5.5, 8.0
	30	8.1	4	1.2, 3.4, 5.5, 8.1
	31	8.1	4	1.2, 3.4, 5.5, 8.1
	19	8.0	2	5.5, 7.9
	20	4.0	2	1.2, 4.0
3	21	3.0	2	1.2, 2.7
	22	8.1	4	1.2, 3.4, 5.5, 7.3
	23	7.9	4	1.2, 3.4, 5.5, 7.3
	32	1.6	1	1.2
	26	2.1	1	1.2
4	9 (Trench Cap)	1.2	2	0.6, 1.2
	8 (Trench Cap)	1.2	2	0.5, 1.2
	12	2.7	2	1.2, 2.7
	6	3.7	3	0.6, 2.5, 3.7
	28	3.7	2	1.2, 3.7
5	36	1.8	1	1.2
	25	7.3	4	1.2, 3.4, 5.5, 7.3
	35	7.3	4	1.2, 3.4, 5.5, 7.3
	33	7.3	4	1.2, 3.4, 5.5, 7.3
	27	7.3	4	1.2, 3.4, 5.5, 7.3
	37	3.7	2	1.2, 3.7
Other	5	2.1	3	0.6, 1.2, 2.1
	13	3.8	2	1.2, 3.8
	24	3.5	2	1.2, 3.4

Table III

The tritium level in water extracted from the shallow landfill near T4 of the experimental trench is compared with that from the undisturbed soil beneath the landfill.

	Mean concentration ± SE (x10^3 pCi/l)	
Depth (m)	Sampler Locations 4/1, 4/5, 4/6	Sampler Locations 4/2, 4/3, 4/4
0-3 (landfill)	150 ± 60	70 ± 10
3-7 (original soil)	10 ± 5	5 ± 1

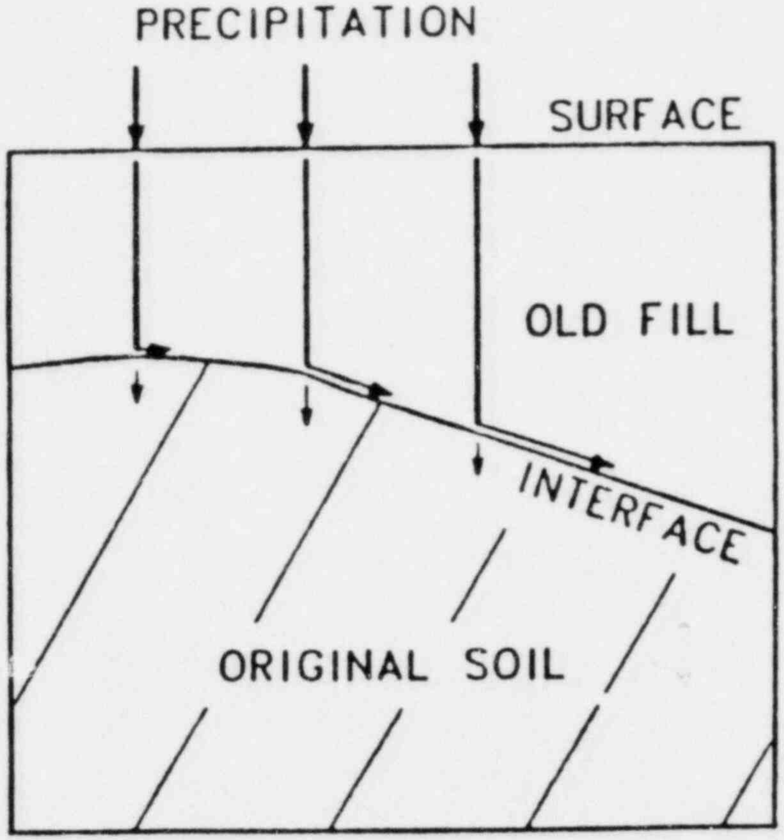

Fig. 4. A possible influence of a landfill - original soil interface on moisture movement in the experimental trench area is shown schematically. Precipitation infiltrates the soil, percolates to the interface and then flows preferentially along the interface to a discharge point. Only a small amount of the precipitation enters the original soil.

the experimental trench area were low (less than 10^5 pCi/ℓ). However, one exception to the low levels of tritium was observed. The soil solution collected from a depth of 6.9 meters at location 3/16 (see Fig. 2) had a tritium level of about 10^7 pCi/ℓ. That level of tritium is comparable to the level observed in T3W, the sump at the west end of section 3 of the experimental trench (Kirby, 1981, p. IV-11). Location 3/16 is approximately 3.5 meters from T3W. The comparable tritium levels suggests that the source of tritium is the same for T3W and 3/16.

At the Waste Trench 19S study area tritium data for two transects at right angles to the trench are presented as indicators of water movement in the area. Figure 5 shows tritium in soil solutions obtained from one of the transects where samples were installed at the 0- to 4-meter depth in 1979. Several trends are indicated. Levels of tritium decreased to a minimum, then increased at greater distances from the trench. In addition to the lateral change, a vertical change was observed; tritium levels increased with depth. The above trends suggest that water moves away from the trench and downward. At a distance of about 11 meters from the trench an apparent second source of tritiated water appears to be moving from the near surface down and towards the trench. The minimum in the curves could indicate that the waters from the two sources may have intermixed at a distance of approximately five meters from the trench. The greater lateral distribution and higher concentration of tritium at the 3- to 4-meter depth for each sampling location suggests that the movement of water away from the trench is much greater at the 3- to 4-meter depth than at the shallower depths and that the net contribution of infiltrating water to soil water is much less at the greater depths. The increase in hydrostatic head of the water in the trench at the greater depths could account for the increased lateral distribution of tritiated water away from the trench in the deeper samples. A perched zone or a zone of preferential flow resulting from a sandstone bed at the 4-meter depth also could explain the increased tritium level

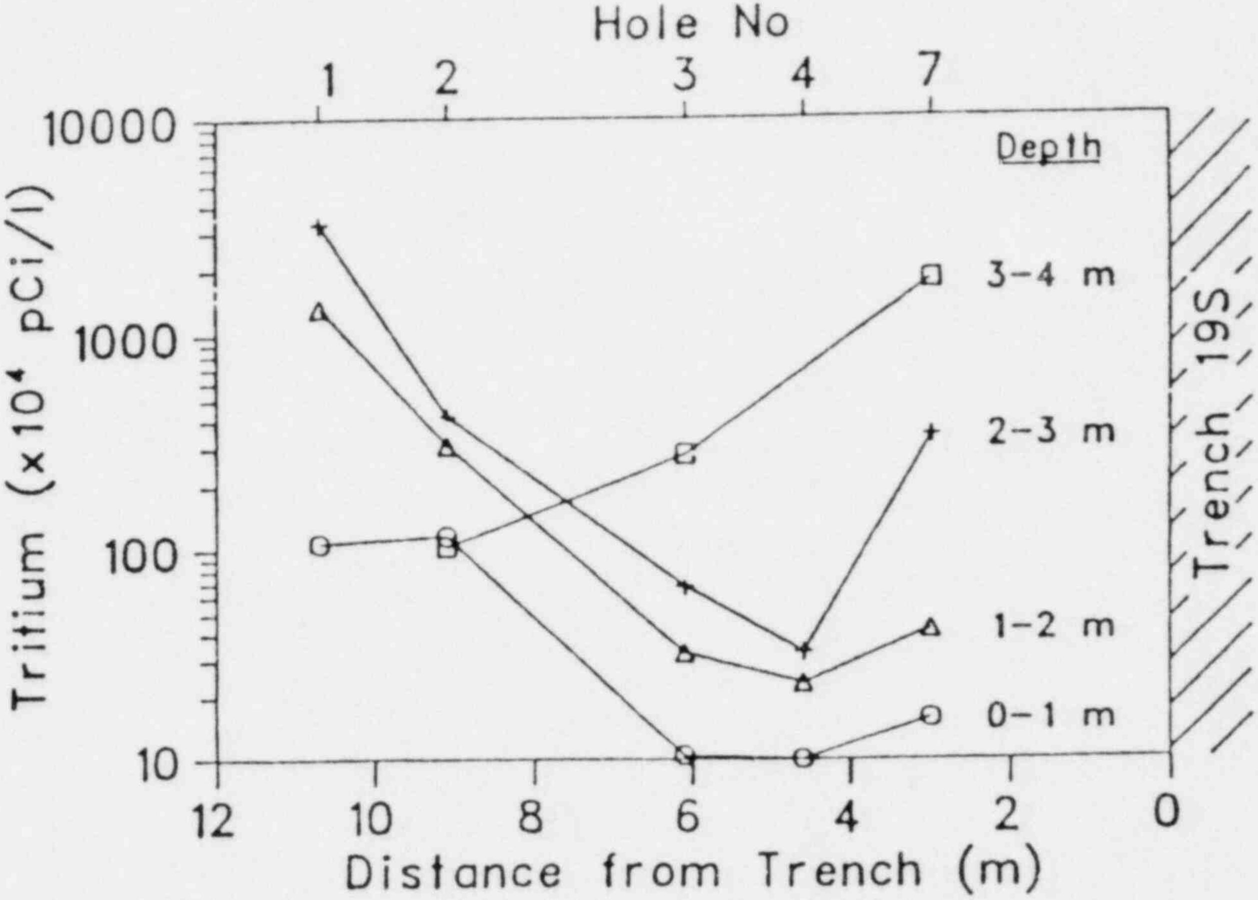

Fig. 5. The distribution of tritium is shown for the top 4 meters of soil along Transect 1 (Fig. 3) in the Waste Trench 19S study area. That distribution suggests that tritiated water moves away from the trench and downward, the distance of movement is greatest at the 3- to 4- meter depth. Also a second source of tritiated water above the 3- meter depth at hole 1 appears to be moving down and towards the trench.

at the 3- to 4-meter depth. That bed is relatively impermeable to vertical flow of trench liquid. The sandstone bed was encountered for all holes augered to the 4-meter depth at the Waste Trench 19S study area with the exception of those along the trench edge. Its thickness varied from 20 to 30 cm for those holes augered to a depth greater than the 4 meters.

Figure 6 shows the distribution of tritium in soil for Transect 2 which is parallel to and about 1.5 meters from transect one (see Fig. 3). The distribution is for depths of 3.4 meters and greater. The trends are similar to those of Figure 5 in that the tritium concentration decreased with distance from the trench and reached a minimum at approximately 7 to 9 meters from the trench. Nearer to the trench the tritium level is greater at 3.4-meter depth than at the 5.5-meter depth. The higher tritium level at the 3.4-meter depth could reflect a perched zone or a zone of preferential flow of water; that depth is approximately the depth of the sandstone layer.

The interpretation of the tritium distribution in the Waste Trench 19S study area is summarized in Figure 7. That figure shows a schematic of the possible water movements in the Waste Trench 19S study area. Water from the trench moves down and away from the trench apparently as interstitial flow. The sandstone layer at about the 4-meter depth probably acts as a barrier to the downward movement of water moving out of the trench above that level and provides for some lateral displacement or movement of that water. A second source of tritiated water apparently moves down and towards the trench, mixing with the water from the trench at a distance of about 5 meters from the trench.

Radionuclide Distribution

In general the distribution of radionuclides in soil solutions collected from Waste Trench 19S suggest that migration of radionuclides from the trench differs with the radionuclide. Tritium, as tritiated water, moves as water moves and

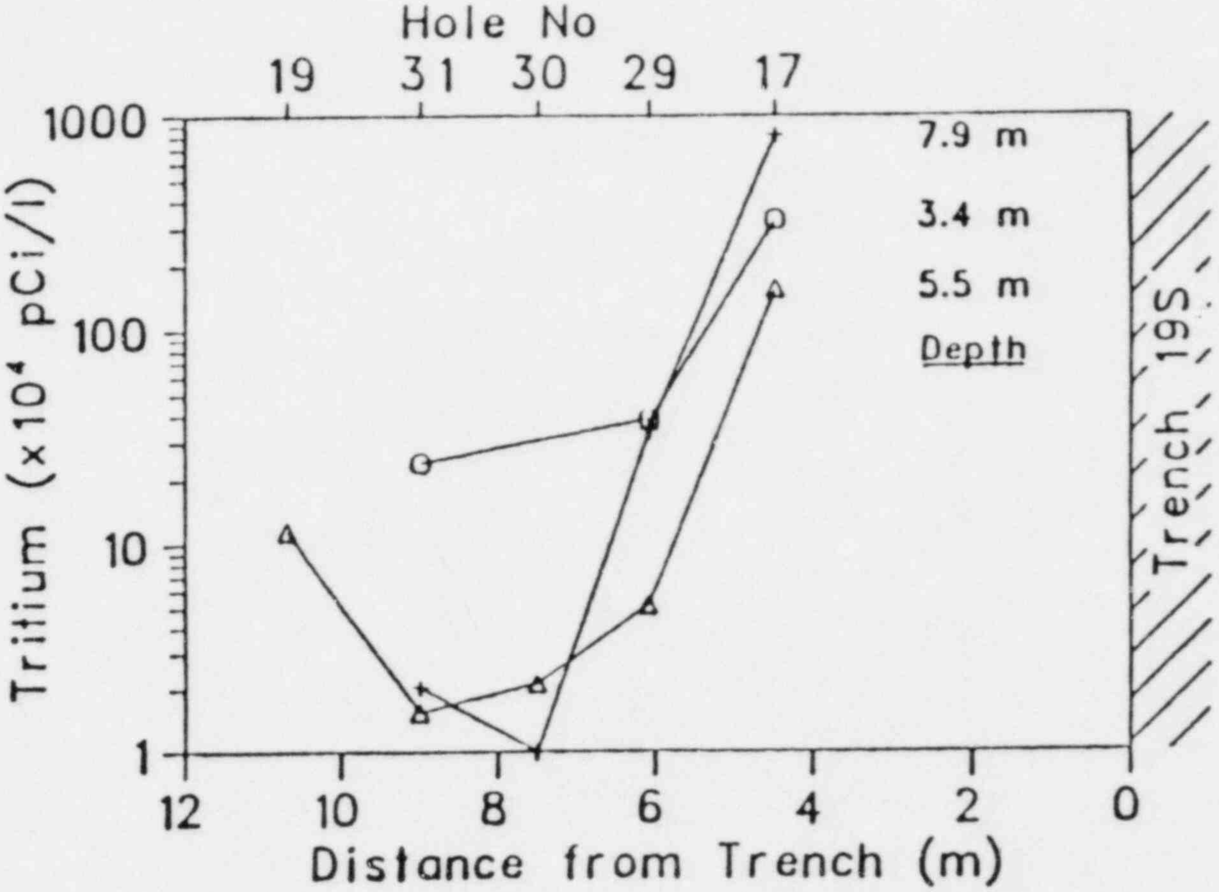

Fig. 6. The distribution of tritium is shown for the depths between 3.4 and 7.9 meters along Transect 2 (Fig. 3) in the Waste Trench 19S study area. That distribution suggests tritiated water moves away from the trench and downward reaching a minimum at approximately 7 to 9 meters from the trench. The level of tritium tends to be higher at the 3.4-meter depth than either above or below it and probably reflects a perched zone or a zone of preferential flow of water. That zone is associated with a relatively impermeable sandstone layer at the 3.4-meter depth.

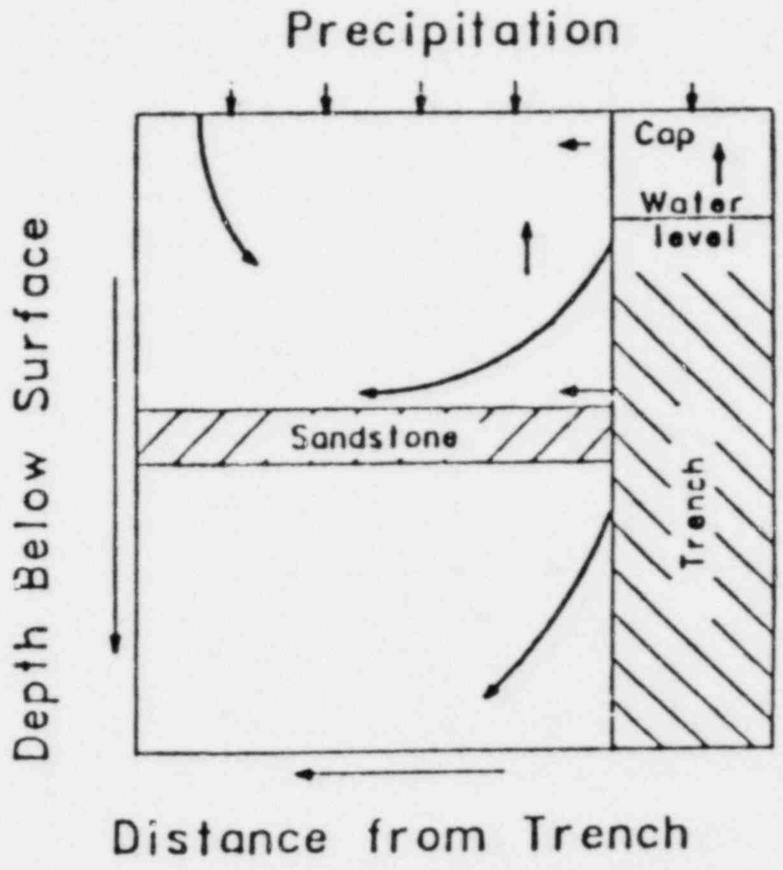

Fig. 7. The schematically shown water movements near Waste Trench 19S are based on tritium distributions. The flow of liquid waste into the surrounding medium is probably caused by a hydraulic head which is a result of the free water in the Trench. The impermeable sandstone layer diverts the shallow flow along that layer. The waste liquid becomes diluted with infiltrating precipitation.

otherwise interacts little with the soil. Decreased levels with distance from the trench are indicative of the dilution of the trench (tritiated) water with water from other sources, e.g., percolating rainfall. On the other hand, ^{137}Cs present in the trench liquid was not observed in any of the soil solutions. Plutonium-238 and ^{60}Co from the trench liquid were detected in soil solutions collected in hole 7 at a depth of 3.1 meters approximately 2 meters from Waste Trench 19S and were not detected in solutions collected farther away from the trench (Fowler et al, 1981).

A comparison of laboratory results with those obtained in the field may provide an explanation for the observed field results and also may give some indication of the value of laboratory studies in predicting radionuclide migration in the field.

A laboratory experiment was designed to evaluate the influence of Maxey Flats Tilsit Ap and Tilsit C soils on the mobility of radionuclides from Waste Trench 19S liquid. The Tilsit Ap soil was interacted with the waste under aerobic conditions and the Tilsit C was interacted with the waste under anaerobic conditions. Also the experiment was conducted under conditions where biological activity was either maximized or minified. Sorption of the waste radionuclides was determined at intervals of 0 (1 hour), 3, 9 and 53 days. The details of the experiment are presented by Fowler et al, 1981.

The results of the above laboratory experiments indicate sorption of essentially all of the ^{137}Cs by those two soils. That sorption is attributed to a cation exchange type reaction. Those results explain the lack of detectable ^{137}Cs in the soil solutions from near Waste Trench 19S.

In the case of plutonium the laboratory experiments indicate that a small fraction of ^{238}Pu in the Waste Trench 19S liquid was not sorbed by the Maxey Flats soils as shown in Table IV. We hypothesized that the nonsorbed ^{238}Pu was in the form of organic complexes. For example, ^{238}Pu could be complexed with ethylenediaminetetraacetric acid (EDTA). Czyscinski and Weiss (1980) have reported the

Table IV

The level of nonsorbable ^{238}Pu in Waste Trench 19S liquid judged to be in the form of nonbiodegradable organic complexes is estimated for anaerobic and aerobic systems. Total concentration of ^{238}Pu in the waste trench liquid was approximately 4000 pCi/ℓ.

Characteristic of Complexes	Approx. Level of ^{238}Pu (pCi/ℓ)
Nonbiodegradable in anerobic system	1800
Nonbiodegradable in aerobic system	500

presence of EDTA in Waste Trench 19S. Under anerobic conditions approximately 1800 pCi/ℓ at ^{238}Pu should remain nonsorbed in the soil solutions collected from near Waste Trench 19S if dilution of the trench waste is negligible (see Table IV). Although that level may be less than the complexed levels suggested by Cleveland and Rees (1981), it is within a factor of four of the concentrations reported by them.

In Table V those laboratory results are compared to ^{238}Pu results obtained from soil solutions near Waste Trench 19S. The source of a portion of the soil solution is liquid waste and is assumed to be from Waste Trench 19S. The fraction of the liquid waste in the soil solutions is estimated on the basis of the difference in tritium level in the soil solutions and that in the trench liquid. The estimated level may then be compared with the nonsorbable level observed in laboratory studies. The above comparison assumes that the ^{238}Pu complex is extremely stable and does not dissociate readily. Laboratory results suggest that the waste from Waste Trench 19S does contain very stable complexes. The data obtained from samples of soil solution collected for three samplings between October 1979 and June 1980 agree quite well with the data obtained in laboratory studies. The laboratory studies suggest that about 1800 pCi/ℓ of ^{238}Pu in the trench water is nonsorbable, whereas field results indicate nonsorbable levels of between 780 and 4000 pCi/ℓ. Plutonium-238 was not detected in soil solutions collected farther then approximately 2 meters from the trench edge (see Fig. 3) even though ^{3}H was detected. One explanation might relate to an increased dilution of the waste water with soil water of another source (e.g. rainwater); that dilution may decrease the level of nonsorbable ^{238}Pu to levels below the limit of detection.

In the case of ^{60}Co the laboratory results indicate that between 20 and 230 pCi/ℓ may be nonsorbable on interaction of the Waste Trench 19S liquid with the Tilsit C soil. The concentration of the nonsorbable fraction is dependent on the

Table V

The levels of nonsorbable ^{238}Pu in Waste Trench 19S liquid are estimated based on levels in soil solutions collected from the Waste Trench 19S study area (hole 7) and from laboratory studies using liquid from that waste trench. Only a fraction of the soil solution at that field study area can be attributed to the waste liquid; that fraction is estimated from the ratios of ^{3}H in soil solutions to that in waste liquid.

			^{238}Pu (pCi/ℓ)	
Type of Sample	Soil Depth (m)	$\frac{^{3}\text{H in Soil Solution}}{^{3}\text{H in Waste Liquid}}$	Soil Solutions (observed)	Waste Trench 19S Liquid (Estimated)
Field				
7-1	3.1	1.7-2.1	1800-2300	3700-4000
7-2	1.3	43-210	3-77	780-3300
Lab	-	1.0	1800	1800

Table VI

The levels of nonsorbable ^{60}Co in Waste Trench 19S liquid are estimated based on levels in soil solutions collected from the Waste Trench 19S study area (hole 7) and from laboratory studies using liquid from that waste trench. Only a fraction of the soil solution at that field study area can be attributed to the waste liquid; that fraction is estimated from the ratios of ^{3}H in soil solutions to that in waste liquid.

			^{60}Co (pCi/ℓ)	
Type of Sample	Soil Depth (m)	$\frac{^{3}\text{H in Soil Solution}}{^{3}\text{H in Waste Liquid}}$	Soil Solutions (observed)	Waste Trench 19S Liquid (Estimated)
Field				
7-1	3.1	1.7-2.1	110-150	190-300
7-2	1.3	43-260	23-24	1000-7300
Lab	-	1.0	20-230	20-230

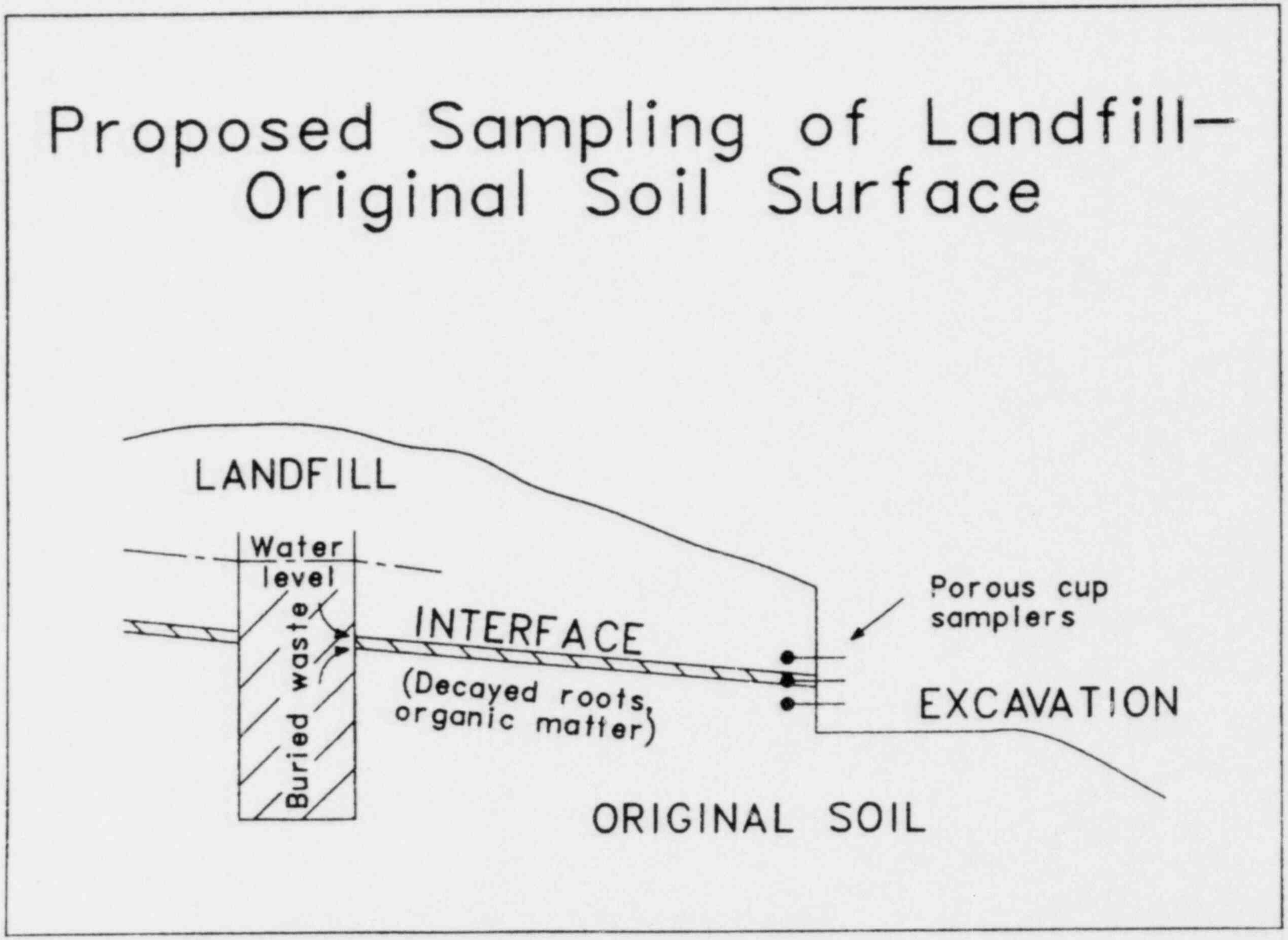

Fig. 8. The proposed sampling of the interface between landfill and the original soil profile is shown schematically.

interaction time; the higher concentrations are associated with the shorter period of interaction. The levels observed in soil solutions collected near Waste Trench 19S were between 23 and 150 pCi/ℓ as shown in Table IV. A level of nonsorbable ^{60}Co for trench water was estimated in a manner similar to that for ^{238}Pu assuming that the source of the ^{60}Co was the trench liquid. A comparison of the estimated values with those observed in laboratory studies shows good agreement for solutions collected from sampler 7-1 which is located at the 3.1-meter depth. Lack of good agreement is shown for solutions collected from sampler 7-2 which is located at the 1.3-meter depth. Apparently ^{60}Co from another source is also present in the solution collected from the 3.1-meter depth.

Future Plans

The information obtained thus far in the Maxey Flats studies suggest several pathways by which waste water and radionuclides may move from burial trenches. One such pathway could be along an interface between landfill and the original soil profile. We propose to excavate the soil along such an interface as shown by the schematic in Figure 8. The soil solutions from the walls of the excavation would then be sampled above, below, and at the interface in order to evaluate preferential movement of waste water and radionuclides. Documentation of preferential movement of waste water and radionuclides along such an interface could explain the presence of some radionuclides at the boundary of the burial site. Also such a study could provide information pertinent to the design of future burial sites.

The collection and analyses of soil solutions will continue in order to evaluate the effects of changes in soil moisture and changes in the chemical and physical properties of those solutions on the movement of waste radionuclides though soil.

Lysimeter experiments will be initiated to study radionuclides migration from a single waste source rather than the unknown multiple sources that exist in the trench area. Those experiments are planned in cooperation with UCB. Our part of that study will pertain to the movement of radionuclides relative to changes in soil moisture. We propose to spike a band of soil with liquid waste inside several of the lysimeters and then observe movement of tritium and other radionuclides away from the spiked zone. Porous cups will be used to sample the soil solutions inside of the lysimeters. Also we propose to differentiate between vapor phase and liquid phase movement of tritiated water. Such a differentiation is important in evaluating the pathways of waste radionuclide migration. Results from field, laboratory and lysimeter studies will be compared and their relative value for prediction of radionuclide movement will be evaluated.

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SECTION VI

STUDY OF UNSATURATED ZONE HYDROLOGY AT MAXEY FLATS

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STUDY OF UNSATURATED ZONE HYDROLOGY AT MAXEY FLATS

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OVERVIEW AND SUMMARY

The problem of serious concern at Maxey Flats is the intrusion of excessive water into the waste burial trenches and the exit of that water while carrying radionuclides. The work reported here is a continuation and expansion of the work reported by Schulz (1980) at a research meeting held at the Nuclear Regulatory Commission Headquarters on July 16, 1980. At the time of the 1980 report, the primary focus of the work was to determine the pathways of water entry into the waste burial trenches. In the fall of 1979, trench caps and space between trenches were instrumented to study water movement through the caps and the soil profile adjacent to trenches. The initial results indicated that deeper instrumentation was desirable; that instrumentation was installed in the fall of 1980. The first set of instrumentation has gone through a full seasonal cycle and is reported on in the body of this paper; the fall 1980 instrumentation has only gone through the winter and spring of 1981 and is briefly reported upon. The work to date indicates that the primary pathway of water entry into the waste burial trenches is by entry through the trench caps. The initial studies on pathways of water entry into the trenches has now led into a study of some possible countermeasures for minimizing the entry of water into the trenches. The essence of the approach is to employ simple lysimeters to measure water run-off, water use by plant cover, and percolation through the soil. Fifteen lysimeters are currently being installed at Maxey Flats to carry out the study. Each lysimeter is essentially a cylindrical tank 1.5 m (5 ft) in diameter and 3.0 m (10 ft) deep installed in the soil so the open end extends 15 cm (6 in) above the ground surface. The tanks are filled with soil and are instrumented to accomplish water balance measurements. By that procedure, effects of such parameters as soil bulk density, surface slope, and crop type and management on water burden in the trenches will be determined to aid in management and closing of the site. In addition, it is intended that information obtained will also be useful in formulation of guidelines for the selection of future sites, best management

procedures for operations of sites as related to water problems, and to provide part of the information needed for eventual closing of the sites. The above work is being carried out in close cooperation with Drs. W. L. Polzer and E. H. Essington of the Los Alamos National Laboratory and the Los Alamos Group will focus on the study of radionuclide movement in association with the hydrology studies.

SCOPE OF WORK

The scope of the work is twofold: 1) To determine the most important pathways of water entry in the waste burial trenches at Maxey Flats, and then, 2) To formulate some effective countermeasures for minimizing entry of water into the trenches. In addition, the transport of radionuclides from the trenches by water movement out of the trenches is being studied in a cooperative project with personnel from the Los Alamos Scientific Laboratory.

EXPERIMENTAL APPROACH

In the fall of 1979, a transect across trench 19S and the adjacent area was instrumented as shown in Fig. 1. Two types of instrumentation were installed in the soil profile: (1) soil moisture cells (Soiltest Co.), and (2) mini-porous cups. The soil moisture cells were installed to allow measurements of relative soil moisture content (soil moisture tension gradients). The mini-porous cups were implanted very close (5 cm) to the soil moisture cells to allow extraction of soil water in the region of the soil moisture cells. Mini-porous cups were used instead of full-sized cups so as not to influence subsequent measurements made with the soil moisture cells. The mini-cups have a volume of approximately 2 ml and the soil solution samples are taken after the soil moisture cell readings. The soil moisture cells are then not read again until at least a two-week period has passed. The porous cups allow extraction of soil under unsaturated conditions down to about 70 centibars of soil water suction (tension); therefore, they can be used for sampling saturated or moist unsaturated soils.

It was originally intended to instrument the transect to a depth of 7 m (22 ft) into the soil profile. However, it was not possible to bore holes to this depth with drilling equipment available at the time the transect was set up. Frequently, rock was encountered forcing abandonment of even shallow holes. The mini-porous cups and the soil moisture cells were installed side by side in the holes as depicted in Fig. 1. Electrical leads from each of the moisture sensors and fine tubing (bore - 1 mm) from the mini-porous cups terminated in an instrument box mounted above the soil surface and to one side of the instrument stack or "station" located vertically in the soil profile.

After the instruments were installed and the holes refilled with the soil removed from the holes, a tracer was sprayed on a swath 3 m (10 ft) wide by 23 m (75 ft) long with the swath being centered over the instrument stations. The tracer consisted of 250 g of pentaflourobenzoic acid dissolved in 8 liters of water. The purpose of the tracer was to augment information on movement of the soil water. In a cooperative effort, Dr. Glenn Thompson of the University of Arizona furnished the tracer and performed the analyses of the soil solutions for benzoic acid content.

The experimental trench, section T4, was instrumented in a similar fashion (Fig. 2) with the exception that it was not possible to bore holes in the area away from the trench due to rocks. The instrumentations of both trenches were completed October 26, 1979.

In the fall of 1980, new transects were instrumented across both trench 19S and the experimental trench, section T4, to a depth of 8 m (25 ft). The transects were instrumented in a manner similar to that described above. Detailed description of the fall 1980 transects will not be given in this report as the data obtained to date was for only one half of a full season wetting and drying cycle, i.e. winter and spring of 1981. A report of results from that instrumentation will be included in the next report of this series.

RESULTS AND DISCUSSION

Soil moisture cell measurements at three selected depths at station no. 1, and with the passage of time, are given in Fig. 3. Station no. 1 was located approximately 10 m away from the edge of trench 19S and in the undisturbed area between the trenches. The moisture sensor located at the 0.9 m depth indicated a rather strong seasonal cycling with an overdraft of water use by the Kentucky fescue crop during the late summer and fall and a replenishment of soil moisture at that depth during winter and spring by infiltration of water in excess of plant water use. At no time did the soil become saturated at that depth. At the 2.4 m depth, there was little seasonal cycling, suggesting little plant root uptake of water from that depth in the soil profile. During the entire period, the soil at this depth remained unsaturated.

Fig. 4 depicts relative soil moisture at four depths in the trench caps on trench 19S with the passage of time. At the 0.9 m depth, a very marked seasonal wetting and drying cycle was observed. During the winter and spring, the trench cap was essentially saturated but was dried out substantially by plant extraction of water during the summer and fall period. A similar moisture status was observed at the 1.8 m depth. At 2.4 m, little or no seasonal cycling was observed and the soil remained approximately saturated during the entire period of measurements.

From the observation given above, it is evident that no water moved from station no. 1 (located in the soil profile) to station no. 4 (located in the trench cap) as to do so, the water would have to move against a concentration gradient.

In Tables 1 and 2, the ^{3}H concentrations in the soil solutions in stations no.1 and no. 4 are given. The concentration of ^{3}H in the trench cap increased markedly with increasing depth, indicating the waste in the trench below the cap was the ^{3}H source and that ^{3}H was migrating upward into the cap by being carried in the upward movement of water during the summer and fall periods of water depletion by the fescue crop. At station no. 1, the ^{3}H was relatively low, and uniform with depth in the soil profile, indicating the evaporator was the source of the ^{3}H at this location, and not the waste in the trench located 10 m away. This condition would evidence no or little movement of water from the trench to the location of station no. 1, 10 m outside the trench.

Table 3 gives the distribution of pentaflourobenzoic acid in the trench cap on January 1980. The tracer had been sprayed on the soil surface in October 1979 and as this tracer is readily water soluble and not absorbed by soil, it will tend to trace movement of "new" (rain) water into the trench cap. The data indicate that the rain water infiltrated and percolated readily through the trench cap.

More evidence of the lack of substantial lateral water movement in the top few meters of the soil profile can be seen by comparing Fig. 4 with Fig. 5. The data of Fig. 5, relative moisture as a function of depth, measured on June 3, 1981, station 2A, was instrumented to a depth of 8 m in the fall of 1980 and was directly adjacent to station 2, shown in Fig. 1. At this location and on June 3, 1981, the profile was saturated in the top 0.7 m (following a very wet month of May), then an increasingly dry profile to 3 m, then increasing moisture content to saturation at a depth of 7 m. It is interesting to note that merely 1 m away and at a depth of 2.4 m, the trench was constantly saturated.

A schematic cross section of the experimental trench, section T4, is shown in Fig. 2 along with stations 6 and 8. Figure 6 shows the change of relative moisture content in the center of the trench cap from February 1980 to June 1981. At the 0.3 m depth, the moisture content remains essentially constant for the entire period with perhaps a small seasonal cycling, but always in the unsaturated condition. At the 1.2 m depth, the moisture remains relatively constant and unsaturated for the entire period. At the 1.8 m depth, a much different pattern is shown. The trench cap was constructed of dry surface soil material in October of 1979 and the wetting front just reached the 1.8 m depth of the trench cap by April of 1980; then the soil moisture content continued to increase at that depth with the passage of time and leveled off about September 1980, and remained unsaturated until the date of the last reading, June 1981. The wetting front did not reach the 2.1 m depth until June of 1980, then slowly the moisture increased but the soil remained unsaturated during the period under observation. The important feature is that at no time during this period did the soil material of the trench cap become close to saturated, therefore no water could exit the unsaturated soil and flow into the cavity formed by the crushed rock below the experimental trench cap. The striking difference in performance of the cap on the experimental trench compared to that on trench 19S is probably due to age differences--a fresh trench cap as compared to an aged one with 15 years of fescue cropping.

WORK UNDERWAY

The work described above was primarily obtained from the instruments installed in 1979 in trench caps and the soil profiles between trenches.

Those essentially gave information down to about 3 meters depth. In the fall of 1980, instrument transects were installed to 8 m, about the depth of the bottom of waste burial trenches. Information has been collected from these deep transects during the past winter and spring and is continuing. After a full seasonal cycle is complete, a picture of the unsaturated (and in some cases saturated) hydrology to a depth of about the bottom of the trenches will be obtained.

Lysimeters have been installed to enable predictions of site functions as affected by various types of site management, and possible procedures to be followed in site closing. The lysimeters will allow complete water balance studies, therefore the effects of crop types and management of the crops, surface slope, and bulk density of the trench caps on increasing or decreasing water entry into waste burial trenches can be evaluated. It is also expected that lysimeter experiments will be useful in quantifying radionuclide movement and especially ^3H release to the atmosphere by evaporation of ^3H_2O and plant transpiration of ^3H_2O. A schematic of a lysimeter is shown in Fig. 7.

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Schulz, R. K. Study of Unsaturated Zone Hydrology. In Research Program at Maxey Flats and Consideration of Other Shallow Land Burial Sites. NUREG/CR-1832 PNL-3510 (1980).

Table 1

^3H concentration in soil solution, station no. 1, adjacent to trench 19S. Sampled 1/15/80. The tritium concentration is essentially constant with depth in the soil profile and much lower than that in the trench cap.

Soil depth m	^3H
	pCi/ℓ
0.15	0.09×10^6
0.30	0.08 "
0.60	0.09 "
0.90	0.08 "
1.50	0.11 "
2.10	0.11 "
3.90	0.08 "

Table 2

^3H concentration in soil solution of the trench cap, station no. 4, trench 19S. The tritium concentration in the trench cap is much higher than that found in the soil profile (Table 1); therefore the primary source of the tritium is the trench water, not the evaporator. The changes in ^3H concentration are probably due to the high mobility of the water in the trench cap. During rainfall, dilution occurs and during dry periods tritium will move up from the trench. Also, surface deposits from the evaporator will have a small effect.

Soil depth m	^3H	
	Sampling date	
	1/15/80	2/22/80
	pCi/ℓ	
0.15	0.15×10^6	0.28×10^6
0.30	1.06 "	0.65 "
1.50	—	1.07 "
2.10	1.12 "	2.07 "
2.40	15.41 "	7.93 "

Table 3

Pentafluorobenzoic acid tracer concentration in soil solution of trench cap. The tracer was applied to the trench cap surface and the data indicate that rain water infiltrated and percolated readily through the trench cap.

Soil depth m	Pentafluoro-benzoic acid
	ppm
0.15	14
0.30	17
0.90	67
1.20	30
2.10	161
2.40	175

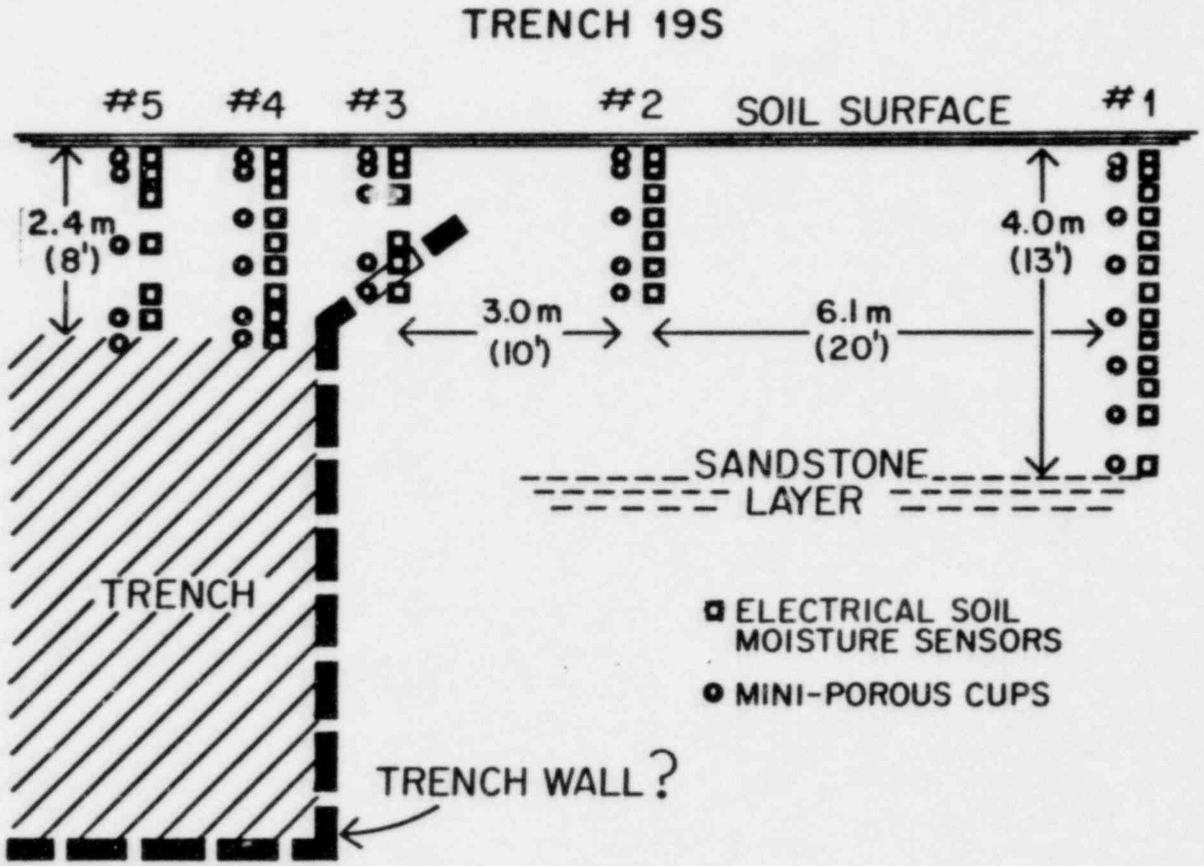

Figure 1 - Profile view of instrumental transect across trench 19S and adjacent soil.

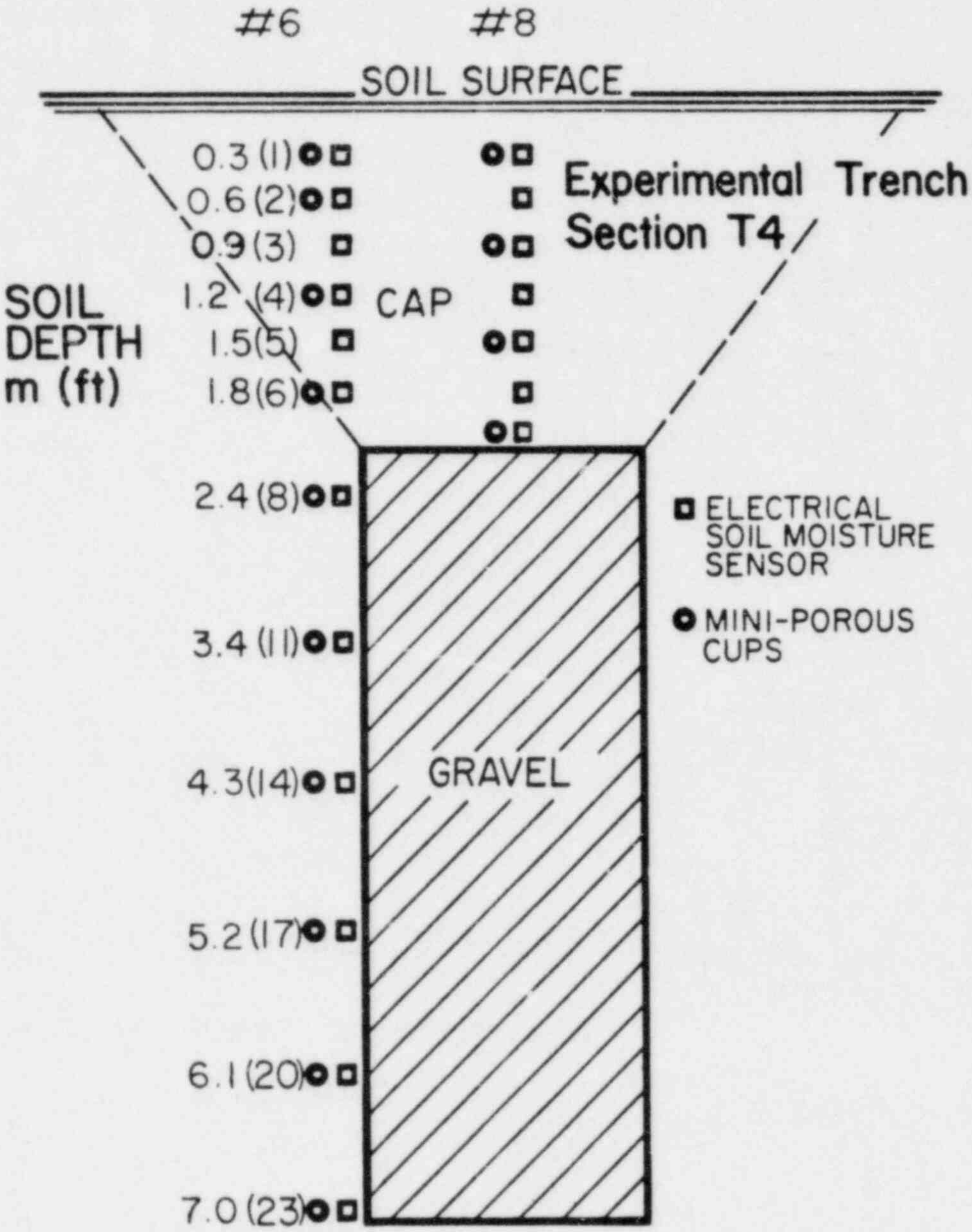

Figure 2 - Profile view of instrumentation of the experimental trench, section T4.

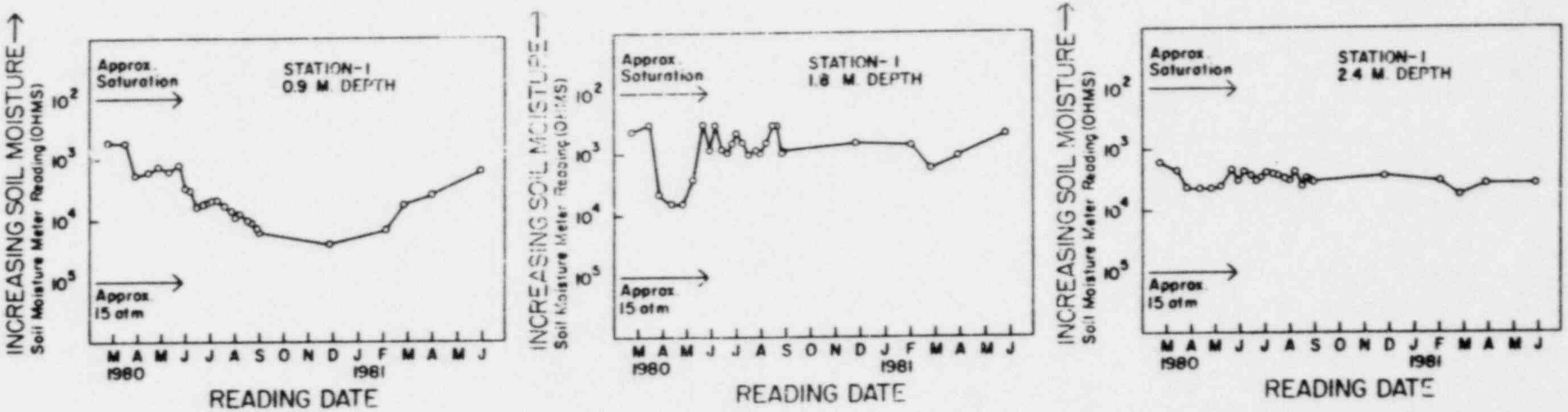

Figure 3 - Relative soil moisture content at 3 depths in soil profile, station no. 1, 10 m away from trench 19S, during the period Feb. 1980-June 1981. Strong seasonal cycling is shown at the 0.9 m depth and little seasonal cycling at the 2.4 m depth. Soil profile remained unsaturated at all times.

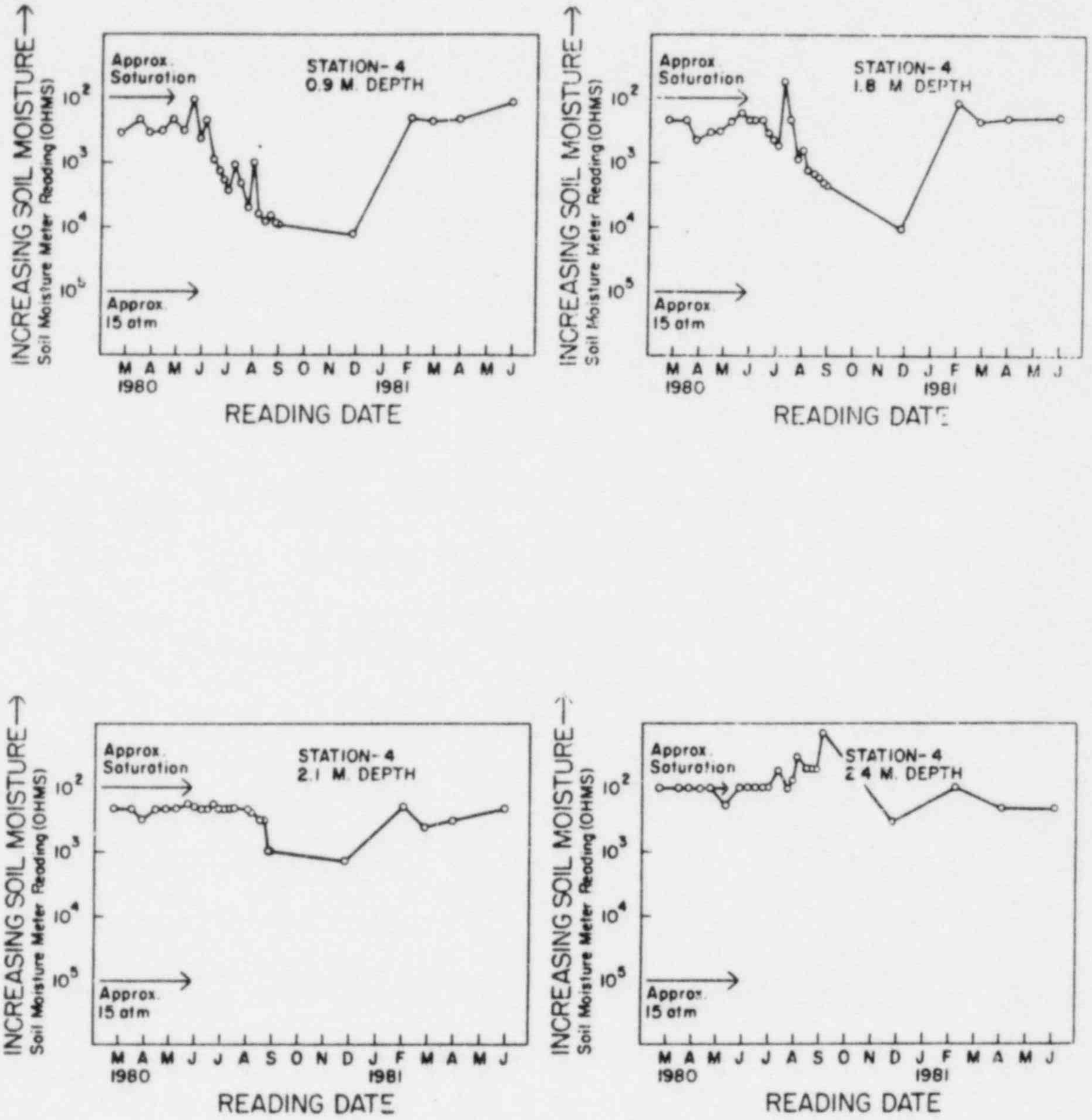

Fig. 4 - Relative soil moisture contents at 4 depths in trench cap of trench 19S, during the period Feb. 1980-June 1981. A marked seasonal cycling was observed at the 0.9 m depth and the soil was essentially saturated in the winter and spring at that depth.. It was substantially dried out during the summer and fall by overdraft water extraction by the Kentucky fescue. A similar moisture pattern was observed at the 1.8 m depth. At 2.4 m into the trench cap, little or no seasonal cycling was observed and the soil remained approximately saturated during the entire period of measurements.

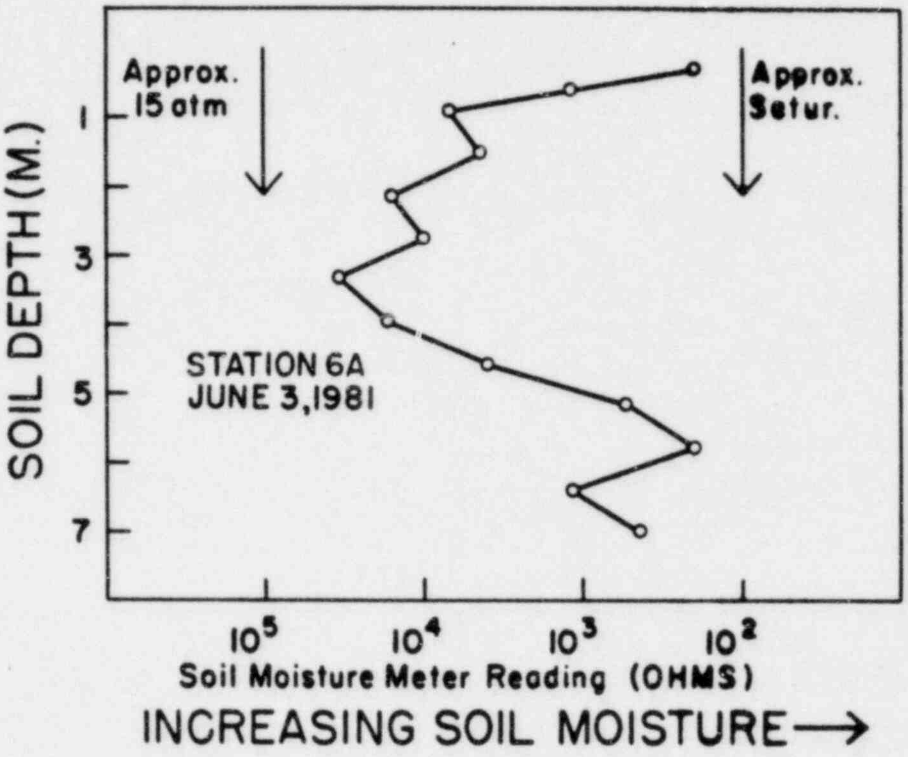

Figure 5 - Relative soil moisture content of soil profile at station 2 A, 3 m away from trench 19S, on June 3, 1981. It is of interest to note that the soil profile was quite dry at the 3-4 m depth, whereas the trench was constantly saturated at the 2.1 m depth and the trench wall was only 3 m from station 2 A.

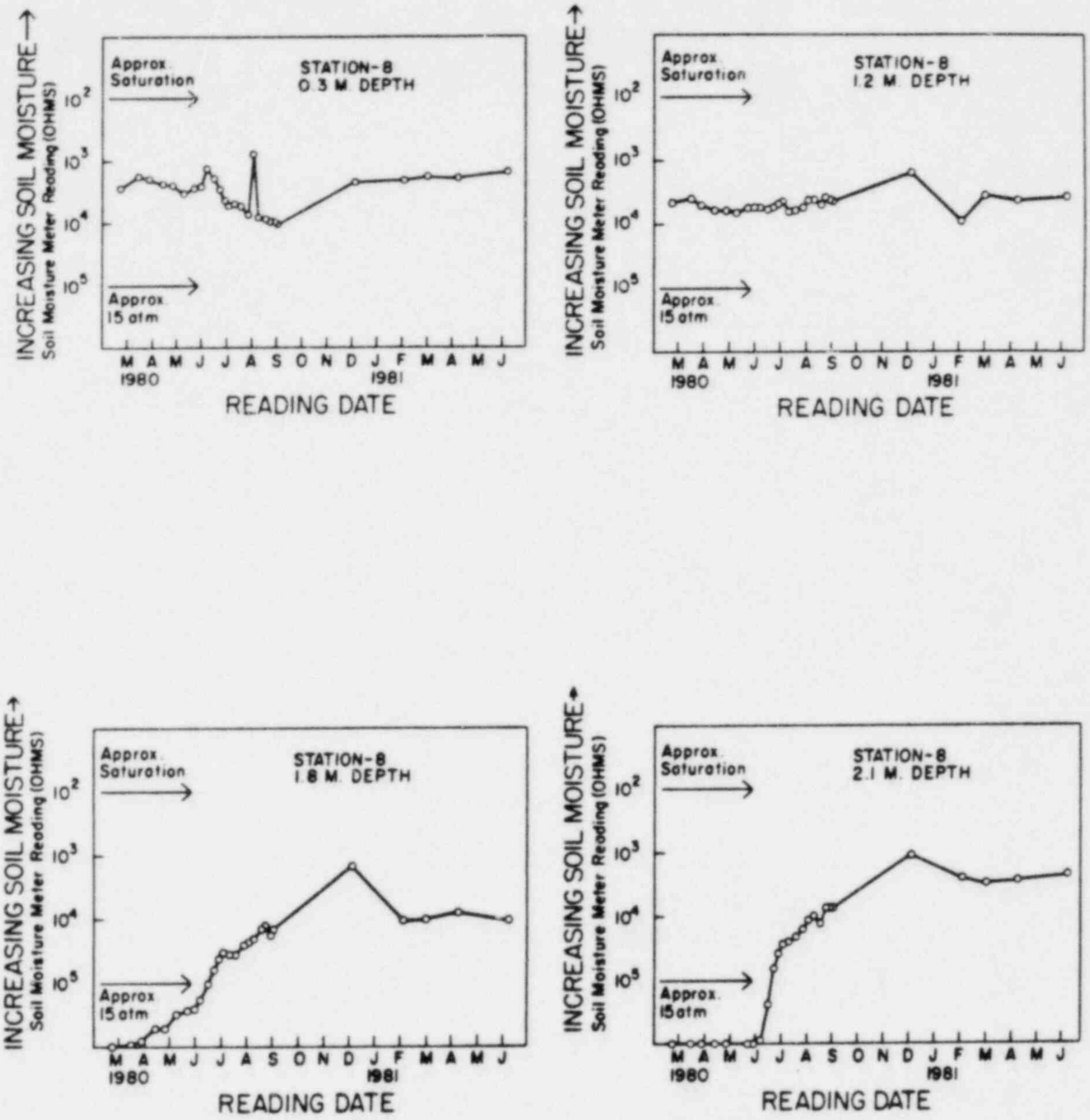

Figure 6 - Relative soil moisture contents at 4 depths in center of trench cap, experimental trench, section T4, during the period Feb. 1980-June 1981. The trench cap was constructed using dry soil in late summer of 1979. By the time observations began on March 1980, the top 1.2 m of soil was moist. The wetting front reached the 2.1 m depth, just above the crushed rock, by June 1980, and the soil became moist but not near saturation by November 1981. This indicates passage of no water into the rock below up to this date. The cap on the experimental trench is a new cap, while that on 19S is an old cap and has had fescue growing on it for many years. This probably would explain the differences in the performance of the trench caps.

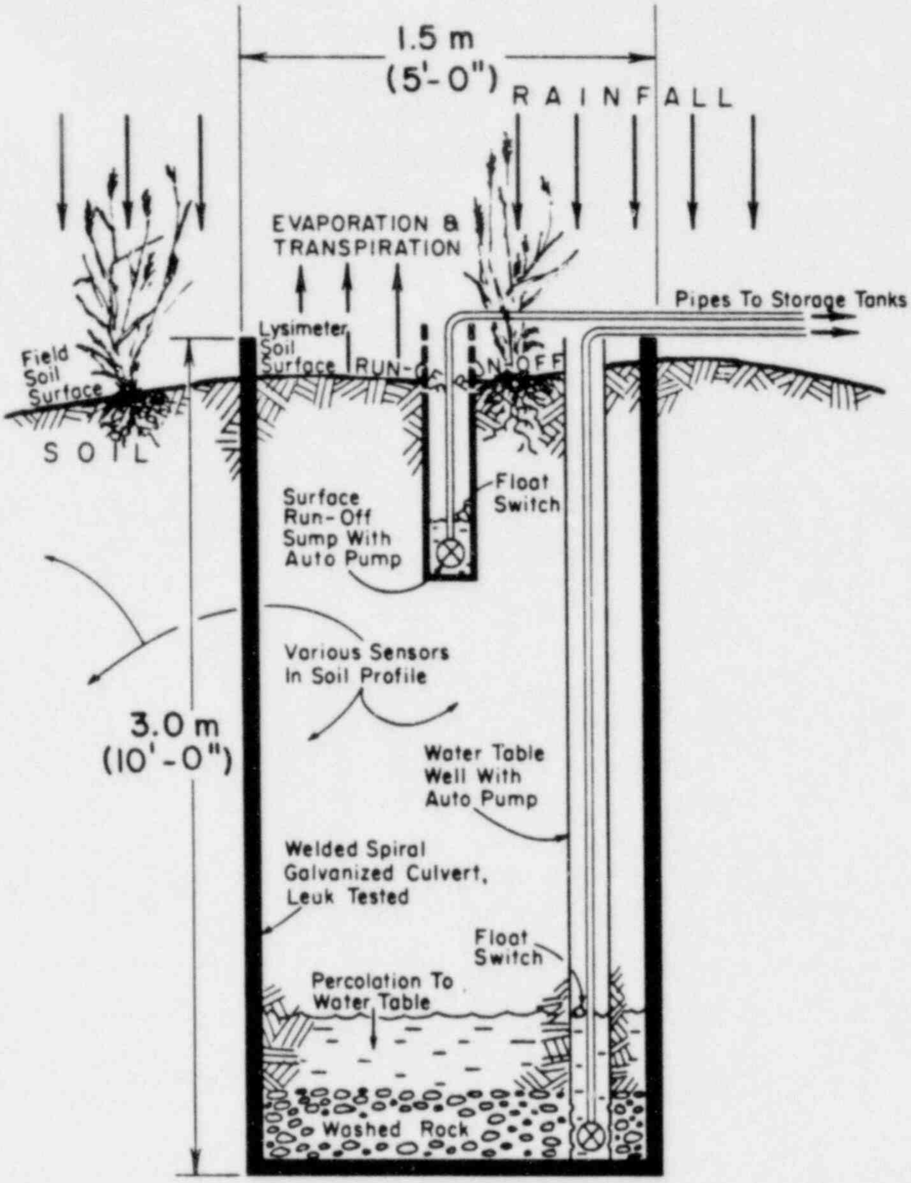

Figure 7 - Cross section of an installed lysimeter. System gives complete accountability of all rain water falling on soil surface. Amount of water striking soil surface is known from rainfall measurements. Over a sufficiently long time period, the water striking the surface has three fates: I. Surface run-off. Measured by collection in surface run-off sump and pumping to collection tank. II. Deep percolation to the water table. Measured by maintaining constant water table level by pumping to storage tank. III. Surface evaporation and plant transpiration. Surface evaporation and plant transpiration = rainfall - (surface run-off + deep percolation). Lysimeters to be used to evaluate effects of crop type, management, compaction and surface slope on percolation. Lysimeters also to be used for radionuclide migration studies, especially transport of ^3H from water table to the atmosphere by plant uptake and transpiration.

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BIBLIOGRAPHIC DATA SHEET

1. REPORT NUMBER (Assigned by DDC)
NUREG/CR-2383
PNL-4067

4. TITLE AND SUBTITLE (Add Volume No., if appropriate)
Radionuclide Distributions and Migration Mechanisms at Shallow Land Burial Sites: Annual Report of Research Investigations on the Distribution, Migration and Containment of Radionuclides at Maxey Flats, Kentucky

2. (Leave blank)

3. RECIPIENT'S ACCESSION NO.

7. AUTHOR(S)
L. J. Kirby, Editor

5. DATE REPORT COMPLETED
MONTH October | YEAR 1981

9. PERFORMING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)
Pacific Northwest Laboratory
Richland, WA 99352

DATE REPORT ISSUED
MONTH July | YEAR 1982

6. (Leave blank)

8. (Leave blank)

12. SPONSORING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)
Division of Health, Siting and Waste Management
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, DC 20555

10. PROJECT/TASK/WORK UNIT NO.

11. CONTRACT NO.
FIN B2291

13. TYPE OF REPORT
Contractor Report

PERIOD COVERED (Inclusive dates)
July 1980 - July 1981

15. SUPPLEMENTARY NOTES

14. (Leave blank)

16. ABSTRACT (200 words or less)

Subsurface waters at Maxey Flats are anoxic systems with high alkalinity and high concentrations of dissolved ferrous ion. Americium and cobalt in these trench waters are made more soluble by the presence of EDTA, while strontium and cesium are unaffected under the same conditions. EDTA is the major organic complexing component in waste trench 27 leachate, but other polar, water-soluble organics are also present. Evidence points to the migration of plutonium between waste trench 27 and inert atmosphere wells as an EDTA complex. Polar organic compounds may influence the migration of ^{90}Sr and ^{137}Cs. The primary pathway of water entry into the waste burial trenches is through the trench caps, but major increases in water level have occurred in an experimental trench by subsurface flow. The areal distribution of radionuclides at Maxey Flats has been influenced by surface runoff, deposition from the evaporator plume, subsurface flow and the actions of burrowing animals or deep-rooted trees. Vegetal and surface contamination on site and near site are quite low, and only ^{60}Co exceeds commonly observed fallout levels. Radionuclide concentrations in surface soil at Maxey Flats are comparable to concentrations resulting from normal fallout in other areas of high rainfall.

17. KEY WORDS AND DOCUMENT ANALYSIS

17a. DESCRIPTORS

17b. IDENTIFIERS/OPEN-ENDED TERMS

18. AVAILABILITY STATEMENT
Unlimited

19. SECURITY CLASS (This report)
Unclassified

20. SECURITY CLASS (This page)
Unclassified

21. NO. OF PAGES

22. PRICE
$

*U.S. GOVERNMENT PRINTING OFFICE : 1982 0-361-297/2305

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

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