Interim Report

**Chemical Species of Migrating
Radionuclides at Commercial
Shallow Land Burial Sites**

**Quarterly Progress Report
October-December, 1983**

**L. J. Kirby
W. H. Rickard
A. P. Toste**

February 1984

**Prepared for the
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**Pacific Northwest Laboratory
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PACIFIC NORTHWEST LABORATORY
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PREFACE

This is the seventh quarterly report for the project "Chemical Species of Migrating Radionuclides at Commercial Shallow Land Burial Sites" (NRC FIN 82291) under a reporting schedule initiated in mid-1982. The next report is scheduled in April, 1984.

The project task organization has undergone several revisions since this reporting schedule was established. To assist the reader to follow the progress of the research, as each new report is prepared we are summarizing previously reported research under a section titled "Summary of Prior Efforts."

Detailed reporting of research findings will continue to be published in quarterly, topical and annual reports and in the open literature.

EXECUTIVE SUMMARY

The primary purpose of this project is to develop an understanding of chemical processes that significantly influence the migration of radionuclides at commercial low-level waste (LLW) burial sites. Chemical measurements of waste trench leachate and identification of chemical changes in leachate during migration will provide a basis for geochemical waste transport models. This project will produce for the U.S. Nuclear Regulatory Commission (NRC) information to support guidance for implementation of 10 CFR 61, particularly in the development of criteria for low level waste disposal site selection, management, permanent closure and monitoring. This project will also produce information needed by the Commonwealth of Kentucky as they finalize plans to stabilize, close and monitor the Maxey Flats site.

- Chemical Forms: Inorganic and Organic Radionuclide Species

Comprehensive organic analyses performed on Maxey Flats waste trench leachate, water from the experimental trench and water from the inert atmosphere wells have determined a large number of hydrophilic and hydrophobic organic compounds in these waters. Analysis of the field-separated solutions from chromatographically fractionated water samples has revealed a complex pattern of association between radionuclides and organic compounds in these waters.

Ethylenediaminetetraacetic acid (EDTA) is a major organic compound in water from waste trench leachates and in water from inert atmosphere well W2NA and evidence points to the migration of plutonium short distances within the site as an EDTA complex. Polar organic compounds (e.g. palmitic and stearic acids) may influence the migration of ^{90}Sr and ^{137}Cs .

Six classes of hydrophilic organic compounds are present in the waste trench leachates: synthetic chelating agents containing ethylenediamine as a core; dicarboxylic acids; monocarboxylic acids; oxygenated carboxylic acids; aromatic carboxylic acids; and phthalic acid esters. The chelating agents are typically used in decontamination operations at nuclear facilities. The four classes of carboxylic

acids are common bioorganic compounds or their microbial diagenesis products. Phthalates are used heavily in industry as plasticizers and may also be microbial diagenesis products.

The most abundant hydrophilic organic compounds identified in the waste leachates are the chelating agents, ranging from 22.9 ppm in trench 19W down to 0.08 ppm in waste trench 33L-8. Three chelating agents were identified in the leachates: ethylenediaminetetraacetic acid (EDTA), N-hydroxyethylethylenediaminetriacetic acid (HEDTA), and ethylenediaminetriacetic acid (ED3A). EDTA is common to all of the leachates over a wide range of concentrations, ranging from 12.4 ppm to trench 19W down to 0.08 ppm in trench 33L-8. EDTA is the most abundant chelating agent in the leachates, with the exception of trench 23M where HEDTA is most abundant. The relative abundance of EDTA in the leachates varies considerably: it constitutes 81% of the chelating agent pool in trench 7-3; 54% in trench 19W; 71% in trench 19S; a low 17.3% in trench 23M; 82% in trench 27; and 100% in trenches 33L-4, 33L-8 and 35. These differences undoubtedly reflect differences in the wastes originally buried, as well as differences in the microbial and/or chemical environments of the waste trenches.

Overall, HEDTA is the second most abundant chelating agent in the waste trenches. In trench 23M it is the most abundant hydrophilic organic compound at 19.5 ppm. This value is actually the highest for any of the chelating agents. An unusually high concentration of ED3A is also present in waste trench 23M and may be derived from the degradation of HEDTA.

Analysis of the hydrophobic organic compounds that were isolated to facilitate the chromatographic analysis of hydrophilic organic species has also permitted us to obtain information on toxic organic compounds that are present in these waters. The hydrophobic organic compounds identified in the waste leachates from Maxey Flats are generally at much lower concentrations than those of the hydrophilic organic compounds (ppb vs ppm). A number of the compounds, e.g., the halogenated hydrocarbons, are toxic and, therefore, of potential

environmental concern. Their concentrations are quite low, however. From a research point of view, some of the hydrophobic organic compounds, e.g., the barbiturates, have been exploited as in-situ, waste-derived tracers of groundwater flow.

- Subsurface Migration and Infiltration Studies

Well-logging measurements of inert atmosphere wells, experimental trench sumps, and three four-inch diameter E-series wells have confirmed that waste radionuclide migration along the sandstone marker bed occurred only within the restricted area at Maxey Flats. Cesium-137 (0.84 pCi/gm) and ^{60}Co (0.54 pCi/gm) were measured in well 14E on the Maxey Flats site at the top of the sandstone marker bed, or about 6.1 meters (20 feet) below the ground surface. Strontium-90 is also present at or below the detection limit (10 pCi/gm or less).

Analysis of composited water samples from selected E-series wells around the Maxey Flats site has revealed that only tritium is present in significant quantities; ^{60}Co , ^{90}Sr , ^{137}Cs , ^{238}Pu , ^{239}Pu and ^{241}Am , as well as other radionuclides specifically mentioned in 10 CFR 61 (e.g., ^{14}C , ^{59}Ni , ^{63}Ni , ^{99}Tc , ^{129}I), were not detectable in these samples. These measurements verify that subsurface migration of radionuclides has occurred over limited distances within the Maxey Flats restricted area, but with the possible exception of tritium, subsurface migration to points outside the restricted area has not been a significant source of contamination by radionuclides of the environs adjacent to the Maxey Flats site.

During the reporting period work on this task work was concentrated on the analysis of samples and data from the Chalk River, Canada site. The work at Chalk River was initiated to allow field sampling to be conducted before winter weather conditions would prohibit field sampling. The sampling program used existing wells and sampling points established at Chalk River and was developed taking into consideration the hydrogeology and stratigraphy of the site. It also considers the application of statistics in siting and managing low-level radioactive waste disposal facilities. This work was undertaken at NRC's request, to initiate a new program.

- Ecological Monitoring at Commercial Shallow Land Burial Sites

Ecological field sampling performed at Maxey Flats has evaluated a broad spectrum of radionuclides in leaves, forest floor litter and surface soil but only ^3H and ^{60}Co appear to be derived from the disposal site. Even these radionuclides are at only slightly elevated levels, well below the MPC's for drinking water. These surveys indicate that leaf fall can be a cost-effective, efficient way to estimate the annual contribution of radionuclides to the forest floor of oak-hickory forests. Tree sap and water from transpiring leaves have also been collected and analyzed for tritium, and indicate that tritium uptake by deeprooted trees has potential to serve as a biomonitor of subsurface movement of water from burial sites. These surveys have indicated that biomonitors may be useful as indicators of radionuclide migration from shallow land burial sites. Intensive tree leaf sampling around the Maxey Flats site has also demonstrated how the data may be used to establish locations for sampling wells in optimum positions for the detection of potential radionuclide migration.

Plans have been formulated for additional sap sampling and leaf water sampling during calendar year 1984. The samplings will be coordinated with the scheduled restart of the evaporator at Maxey Flats, now set for April, 1984. Sampling will include samples taken before evaporator operation, during the initial stages of evaporator operation, and about six months after the start of evaporator operation.

- Technical Program Coordination

The report PNL-4432-6, "Chemical Species of Migrating Radionuclides at Commercial Shallow Land Burial Sites, Quarterly Progress Report, July-September, 1983" was published and distributed in November, 1983.

The report NUREG/CR-3607, "Radionuclide Distributions and Migration Mechanisms at Shallow Land Burial Sites: 1982 Annual Report of Research Investigations on the Distribution, Migration and Containment of Radionuclides at Maxey Flats, Kentucky", was submitted to NRC for publication(1).

TABLE OF CONTENTS

PREFACE	iii
EXECUTIVE SUMMARY	v
INTRODUCTION	1
OBJECTIVES	2
SUMMARY OF PRIOR EFFORTS	3
SIGNIFICANT CURRENT RESEARCH RESULTS	7
Task A - Chemical Forms: Inorganic and Organic Radionuclide Species	7
Task B - Subsurface Migration and Infiltration Studies	11
Task C - Specific Radionuclide Mapping at Maxey Flats and Other Commercial Shallow Land Burial Sites	13
Task D - Ecological Monitoring at Commercial Shallow Land Burial Sites	14
Task E - Technical Program Coordination for Low-Level Waste Research	17
FIGURES	18
TABLES	22
REFERENCES	24

INTRODUCTION

One of the most important factors to be considered during the long-term isolation of radioactive wastes at commercial low level shallow land burial sites is the subsurface migration of radionuclides, which depends, among other things, on the chemical forms of the radionuclides. The oxidation state, ionic form, and the formation of complexes or chelates with organic ligands can markedly influence radionuclide migration. Therefore, determination of the precise chemical species of radionuclides that have migrated is essential for our accurate evaluation of the long-term performance of shallow land burial sites. It is also essential to identify the organic and inorganic compounds and ionic species which are present in buried waste or can be generated in situ by chemical reactions and subsequently participate in the formation of the chemical species that may be shown to migrate.

Pacific Northwest Laboratory (PNL) has assisted in coordinating the NRC research program at Maxey Flats since 1979 to establish and carry out a unified and comprehensive study of the problems specific to that site and to obtain generic information that may be useful in setting criteria for decommissioning of existing commercial sites and for licensing of future commercial LLW burial sites. The objectives of this broad, multidisciplinary, multilaboratory program include determining:

- Radiochemical and chemical composition of leachates in waste burial trenches;
- Subsurface migration rates of radionuclides and the chemical, physical, biological, and hydrogeological factors which affect this migration;
- Areal distribution of radionuclides at commercial shallow land burial sites and the factors responsible for this distribution;
- Concentrations of radionuclides in vegetation both on and off site and the uptake of radionuclides by representative agricultural crops;
- Atmospheric pathways for radionuclide transport and the mechanisms involved; and
- Engineering practices which influence the seepage of surface waters into the burial trenches.

OBJECTIVES

The primary purpose of this project is to develop an understanding of the chemical processes that influence radionuclide migration at commercial LLW burial sites. Chemical measurements of waste trench leachate and identification of chemical changes in chemical forms that occur during migration are being made and will provide a basis for geochemical waste transport models. This project will produce for NRC information to support guidance for implementation of 10 CFR 61, particularly in the development of criteria for commercial LLW disposal site selection, management, permanent closure and monitoring.

Specific objectives of this project with respect to measurements to be made at existing sites are:

- Define the actual chemical species of the radionuclides which are migrating by subsurface routes;
- Identify the chemical characteristics of the trench leachate which retard or enhance radionuclide mobility;
- Determine the chemical changes in leachate properties which result from installation of infiltration barriers such as the flexible vinyl covers at Maxey Flats;
- Provide the chemical and radiochemical input parameters essential for hydrological/geochemical modeling of subsurface radionuclide transport;
- Determine the factors responsible for the areal distributions of surface radionuclides and develop improved procedures for monitoring;
- Develop and test post-closure monitoring protocols that can be used at Maxey Flats and test the applicability of these protocols to operational and post-operational monitoring at other commercial sites; and
- Assist in coordinating LLW research projects for NRC.

Field measurements at shallow land burial sites provide unique opportunities to identify in actual waste burial situations the chemical and geochemical parameters which retard or enhance radionuclide mobility at LLW burial sites.

The work at Maxey Flats also responds to the need for information on the areal distribution of radionuclides and the extent of subsurface transport of radionuclides at that site. This data base is needed by the Commonwealth of Kentucky as they finalize plans to stabilize, close and monitor the Maxey Flats site.

SUMMARY OF PRIOR EFFORTS

Under this research program, PNL assists in coordinating NRC-sponsored LLW research, which has included PNL studies on chemical species and areal distributions of radionuclides, work on trench water chemistry by Brookhaven National Laboratory (BNL), plant uptake studies 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.

To conduct experiments for determining the subsurface transport of radionuclides from existing waste burial trenches at a commercial LLW site, two experimental areas were established within the Maxey Flats, Kentucky, LLW disposal facility. This required the cooperation of several national and state agencies, together with the NRC contractors, for successful implementation. These experimental areas included:

- An experimental slit trench installed during the last quarter of FY 1979 and first quarter of FY 1980; and
- Fourteen inert atmosphere sampling wells installed in undisturbed soil adjacent to the experimental slit trench during the last quarter of FY 1980. A comprehensive array of porous cups and soil moisture cells was also installed for the LANL and UCB programs.

The experimental areas have been described in NUREG/CR-1832, Section IV and NUREG/CR-2383, Section I and are illustrated in Figures 1 and 2 of this report. These experimental areas have supported research studies by PNL, LANL, UCB and BNL.

The two experimental areas have enabled 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 total water balance by the laboratories participating in research at Maxey Flats. These experiments have provided needed information on rates and mechanisms of subsurface transport of radionuclides from waste burial trenches by both saturated and unsaturated flow at the Maxey Flats site.

Field measurements have been performed by PNL at the Beatty, Nevada⁽⁹⁾, and Maxey Flats, Kentucky, (NUREG/CR-2383, Section II and NUREG/CR-1832, Section IV) 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 adjacent to the Maxey Flats facility, and confirmed 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 site operation and maintenance activities, surface contamination and runoff, deposition from the evaporator plume, and the actions of burrowing animals or deep-rooted trees. Vegetational and surface contamination on site and near site are quite low; of the gamma-ray-emitting radionuclides measured, only ⁶⁰Co 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. Tritium concentrations in water near the site have been influenced by surface runoff and by condensation from the evaporator plume.

Following analysis of trench leachates sampled in October, 1978 and November, 1979, Cleveland and Rees⁽⁸⁾ concluded that except for leachate from waste trench 27 most of the plutonium in trench water is in true solution, in the tetravalent state, and exists in complexes with strong organic ligands. PNL research conducted in 1979 and 1980 (NUREG/CR-1832, Section IV) demonstrated that plutonium in water from the experimental trench and in waste trench 27 leachate was present in highly complexed, anionic, reduced (trivalent and tetravalent) species. Ethylenediaminetetraacetic acid (EDTA) is the major organic complexing component in waste trench 27 leachate, but other polar, water-soluble organic compounds are also present (NUREG/CR-2383, Section I).

Plutonium and EDTA co-elute during steric exclusion chromatography of Maxey Flats trench and well waters, and evidence points to the migration of plutonium within the site as an EDTA complex. Polar organic compounds (e.g. palmitic and stearic acids) may influence the migration of ^{90}Sr and ^{137}Cs . These studies have demonstrated the need to determine the stability of the various chelates and complexes under the conditions existing in waste trench water at Maxey Flats, the conditions under which complexes of plutonium, americium, cobalt and strontium can be decomposed and/or sorbed on the geologic media, and the need to separate and measure the concentrations of hydrophilic organic complexing agents that may affect the migration of radionuclides present in waste trench leachate.

Comprehensive organic analyses performed on Maxey Flats waste trench leachate, water from the experimental trench and water from the inert atmosphere wells have determined a large number of hydrophobic organic compounds in these waters(4). Analysis of the hydrophobic organic compounds that were isolated to facilitate the chromatographic analysis of hydrophilic organic species has also permitted us to obtain information on toxic organic compounds that are present in these waters(3). Most of the hydrophobic organic compounds are present at parts-per-billion levels and only three of these compounds -pentafluorobenzoic acid (a non-radioactive tracer added to evaluate trench caps on the experimental trench), 2 (3H)-benzothiazolone (present in waste trench 27 leachate) and a group of alkylphenoxy oligomers (present in inert atmosphere well W2NA) - are present at parts-per-million levels. Two barbiturates, barbitol and pentobarbital, were also identified in water samples from waste trench 27 and inert atmosphere well W2NA.

Changes in tritium concentrations in water from the experimental trench and in inert tracers (e.g., sodium bromide) which were added to the trench (NUREG/CR-1832, pages IV-10 and IX-6) confirm that subsurface flow does occur rather rapidly within the limited area of these experimental facilities. While increases in water level have occurred in the experimental trench, the absence of certain tracers in the water indicates that these increases have occurred without movement through the different trench caps installed on the five sections of the experimental trench (NUREG/CR-2383, Section I).

Ecological field sampling performed at Maxey Flats (NUREG/CR-1832, Section V, and NUREG/CR-2383, Section III) has evaluated a broad spectrum of radionuclides in leaves, forest floor litter and surface soil, but only ^3H and ^{60}Co appear to be derived from the disposal site. Even these radionuclides are at only slightly elevated levels and are comparable to ambient fallout levels. Collections of freshly fallen tree leaves indicate that leaf fall collection is a cost-effective, nondestructive sampling procedure that can be incorporated into ecological monitoring programs to indicate migration or lack of migration from shallow land disposal sites. Tree sap and water from transpiring leaves have been collected and analyzed for tritium, indicating that tritium uptake by deeprooted trees has potential to serve as a biomonitor of subsurface movement of water from burial sites. The field surveys have indicated that biomonitors may be useful as indicators of radionuclide migration from shallow land burial sites.

Carbon-14 analyses of wood from selected trees adjacent to the Maxey Flats restricted area were compared with analyses from tree specimens at a location 12 miles from the Maxey Flats Facility. These limited data suggested that the site may have influenced ^{14}C levels very slightly at a few locations near the perimeter fence. The source of ^{14}C in the wood specimens is from carbon dioxide that is assimilated from air in the photosynthesis process(3).

Remaining important areas of research include the fate of mobile (possibly complexed) radionuclide species after leaving the trench environment, the control of these species within the commercial site boundaries, remedial action that can be taken to halt this mobility, and whether complexing agents such as EDTA can be effectively destroyed in an aerobic environment.

Important observations from research conducted by PNL at Maxey Flats include:

- Identification of the magnitude of transport over short distances within the burial site by subsurface flow;
- Characterization of some of the important chemical species including plutonium-EDTA complexes which are responsible for subsurface migration;

- Identification of surface runoff from the site as a principal source for off-site radionuclide accumulation in vegetation;
- Determination of the off-site distribution of radionuclides at Beatty, Nevada, and Maxey Flats, Kentucky; and
- Establishment of the complexity and variability of waste trench leachates as source terms for the migration studies. (PNL research complements the comprehensive analyses performed on waste trench leachate by BNL. See, for example, NUREG/CR-2383, Section IV, and NUREG/CR-1832, Section III.)

These research observations and related observations from the research performed at Maxey Flats by BNL, LANL, UCB, the University of Arizona and the University of California-Los Angeles are described in NUREG/CR-3607(1), NUREG/CR-2383(6) and NUREG/CR-1832(10).

SIGNIFICANT CURRENT RESEARCH RESULTS

This is the seventh quarterly report for the project "Chemical Species of Migrating Radionuclides at Commercial Shallow Land Burial Sites." The organization of these reports has undergone substantial change since the first report was issued in order to conform to revised research proposal formats requested by NRC. We will continue to report and summarize research results by subject and task.

Task A - Chemical Forms: Inorganic and Organic Radionuclide Species

The migration rates of radionuclides depend to a major degree on their chemical species. It is therefore extremely important that we determine chemical species in the waste burial trenches and in any water which has carried these radionuclides to our sampling points in the experimental trench and inert atmosphere sampling wells, or to springs some distances from the waste-filled trenches. In this task we are determining as nearly as practicable the precise chemical forms for those radionuclides which are of greatest concern from an environmental and health standpoint. For example, plutonium is present at Maxey Flats in waste trench leachate as a reduced

(trivalent and tetravalent), anionic, highly complexed species. Similar identification will be made for other prevalent radionuclides. These include ^{241}Am , ^{60}Co , ^{137}Cs and ^{90}Sr , as well as other radionuclides specifically mentioned in 10 CFR 61 (e.g., ^{14}C , ^{59}Ni , ^{61}Ni , ^{99}Tc , ^{129}I). The precise radiochemical and chemical parameters obtained from these studies will provide important basic information on the chemical species that develop under various burial conditions. In these studies we will continue to supplement our laboratory work with on-site chemical separations employing tracers and organic ligand separations. The on-site separations minimize solution degradation and thereby duplicate as closely as possible actual field conditions.

During the previous quarter we performed a further analysis of the EDTA-like species that were measured in waste trench leachates and inert atmosphere well water at Maxey Flats(3,4,5). We had earlier identified three EDTA-like species of molecular weights 219, 244 and 288 with structures similar to EDTA and related chelating agents (e.g. NTA, DTPA and HEDTA). We suggested that these EDTA-like species might be derived from the degradation of EDTA and assigned tentative structures to the EDTA-like species. Following additional detailed study, we concluded that the molecular weight 288 species is derived from the chelating agent HEDTA and not from EDTA as previously assumed.

The detailed chemical speciation studies have demonstrated that EDTA is co-eluting with plutonium and ^{60}Co during steric exclusion chromatography of water from waste trenches 27 and 19S and inert atmosphere well W2NA. Strontium-90 and ^{137}Cs also appear to be associated with polar organic compounds, notably palmitic (hexadecanoic) and stearic (octadecanoic) acids. Quantitation of EDTA and of radionuclides in the concentrated and fractionated samples has demonstrated that the soluble plutonium is associated with EDTA.

Some of the plutonium originally present in the water samples is removed from the water during concentration of the samples by co-precipitation with iron. This occurs due to exposure to traces of oxygen or because the solubility limits for iron have been exceeded in the concentrated samples. This observation suggests that uncomplexed plutonium will be fixed to the soil with the iron hydroxide precipitate when the anoxic groundwater is exposed to oxidizing conditions.

During this quarter we have extended our analysis of the chelating agents reported previously(1-5). The most abundant hydrophilic organic compounds measured in the waste trench leachates were the chelating agents, which ranged in total concentrations from 22.9 ppm in trench 19W to 0.08 ppm in trench 33L-8. See Table 1.

Three chelating agents were identified in the leachates: ethylenediaminetetraacetic acid (EDTA), N-hydroxyethylethylenediaminetriacetic acid (HEDTA), and ethylenediaminetriacetic acid (ED3A). EDTA is common to all of the leachates over a wide range of concentrations, ranging from 12.4 ppm in trench 19W down to 0.08 ppm in trench 33L-8. EDTA is the most abundant chelating agent in the leachates, with the exception of trench 23M where HEDTA is most abundant. The relative abundance of EDTA in the leachates varies considerably: it constitutes 81% of the chelating agent pool in trench 7-3 and only 17.3% in trench 23M. The differences undoubtedly reflect differences in the wastes originally buried, as well as differences in the microbial and/or chemical environments of the waste trenches.

Overall, HEDTA is the second most abundant chelating agent in the waste trenches. In trench 23M it is the most abundant hydrophilic organic compound at 19.5 ppm. This value is actually the highest for any of the chelating agents. This overabundance of HEDTA, coupled with the unusually high concentration of ED3A, suggests that the waste buried in trench 23M is different than that of most of the waste trenches.

Analytically speaking, detection of HEDTA has presented a challenge. In early analyses of the waste trench leachates, we detected an unknown species of molecular weight (MW) 288 in the methylated hydrophilic organic fraction. On the basis of GC-MS analysis, it clearly has an ethylenediamine core, but its MW did not correspond to any known chelating agent. Methylated HEDTA has a MW of 320. Careful interpretation of the GC-MS fragmentation pattern of the MW 288 species revealed the presence of a lactone ring suggesting that a hydroxyethyl group like that present in HEDTA might be responsible for the lactone formation. GC-MS and MS analysis of methylated HEDTA subsequently revealed that indeed this compound gas chromatographs only as the MW 288 lactone species.

The third chelating agent identified in the waste leachates is ED3A. It is the least abundant of the three chelating agents and is presumably a degradation product of HEDTA or EDTA, since EDTA has been reported to undergo environmental degradation to ED3A(11). ED3A does not appear to be commercially available as a chelating agent but it could be present as a synthetic by-product of the commercial synthesis of HEDTA or EDTA.

The identification of ED3A also posed an analytical challenge. In the first round of analyses of the waste leachates, we identified an unknown species of MW 244 which we assumed to be derived from degradation of EDTA. However, with the aid of Fourier transform infrared analysis using a computerized infrared spectrophotometer interfaced with a gas chromatograph, we observed that the MW 244 species has a strong IR absorbance at 1690 cm^{-1} , indicating the presence of a lactam ring. The structure that emerged is that of ED3A with one of its acetate ligands attached to a nitrogen of the ethylene-diamine core to form a lactam ring. This ring closure is probably a derivatization artifact caused by the heat (100°C) used in the methylation procedure. The species which exists in the aqueous leachates is undoubtedly ED3A in the open, non-lactam form unless the waste had been heated, etc., prior to burial. See Figure 3.

The absence of some of the chelating agents in certain leachates suggests differences in the waste originally buried or, perhaps, in the environment of the trenches. Leachate from trenches 7-3, 27, 33L-4, 33L-8, and 35 lack HEDTA, presumably reflecting differences in the waste. Trenches 33L-4, 33L-8, and 35 also lack ED3A. If ED3A is a degradation product of HEDTA or EDTA, our results indicate that decomposition is not a simple process. The leachates that contain the most ED3A (trenches 19W, 19S, and 23M) also contain both HEDTA and EDTA. However, two other samples which contain ED3A (trenches 7-3 and 27) contain only EDTA in addition. In three of the leachates (trenches 33L-4, 33L-8, and 35) EDTA is present without any ED3A.

The other hydrophilic organic compounds consist of a variety of carboxylic acids. Collectively, the carboxylic acids range in concentration from 8.8 ppm in trench 7-3 down to 0.7 ppm in waste trench 27. The dicarboxylic acids are the most abundant acids. One of these acids, oxalic acid, is used in nuclear

decontamination operations but it is present only in leachate from waste trench 19S at a relatively low concentration of 0.1 ppm. The monocarboxylic acids are poorly represented in the waste leachates. Only two monocarboxylic acids were identified - hexadecanoic acid, commonly known as palmitic acid and octadecanoic acid, commonly known as stearic acid. Groundwater samples typically contain these two compounds plus a variety of other monocarboxylic acids. The lack of monocarboxylic acids in the leachate samples undoubtedly reflects the fact that the water samples are relatively stagnant leachates of buried waste.

As a whole, the carboxylic acids are potential complexing agents of radionuclides, but any such association would be expected to be much weaker than the association of radionuclides and the chelating agents. However, these bioorganic acids could well constitute the precursors of fulvic and humic acids, which often comprise most of the bulk of the dissolved organic carbon (DOC) in groundwater and may be responsible for complexing radionuclides. The importance of such species to the complexation of radionuclides in groundwater is uncertain at this time, but should not be overlooked. Considerable research needs to be carried out on the role of the so-called "natural" organic compounds such as fulvic and humic acids in mobilizing or retarding radionuclide transport in soil solutions.

The hydrophobic organic compounds identified in waste trench leachates are summarized in Table 2. The concentrations of these compounds are generally much lower than those of the hydrophilic organic compounds (ppb vs ppm). A number of the compounds, e.g., the halogenated hydrocarbons, are toxic and of potential environmental concern. Their concentrations are quite low, however. From a research point of view, some of the hydrophobic organic compounds (e.g., the barbiturates) could be exploited as "in-situ," waste-derived tracers of groundwater flow and the amount of information potentially available justifies the research effort in analyzing the hydrophobic organic compounds.

Task B - Subsurface Migration and Infiltration Studies

The experimental trench and wells study was undertaken to determine if subsurface migration of radionuclides had occurred at Maxey Flats, to measure the extent of any such migration, and to define the processes by which movement

occurs. The work has utilized the experimental installations we established on the Maxey Flats site for defining migration rates and specific chemical species of long-lived fission products and the transuranium elements. It is also allowing the determination of the chemical species of migrating nonradioactive elements associated with waste and utilizes nonradioactive tracer movement to evaluate the effects of engineered trench caps and drainage systems on water infiltration rates.

Most samples from the porous cups, experimental trench sumps and inert atmosphere wells (Figure 2) have been collected on a monthly frequency, weather and water levels permitting. The water samples have been analyzed for ^3H , gamma-ray emitting radionuclides (e.g., ^{60}Co , ^{137}Cs), alpha-emitting radionuclides (e.g., ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Am), hydrophilic and hydrophobic organic species, and nonradioactive elements that can be determined by neutron activation (e.g., Br). These measurements verify that within the Maxey Flats restricted area subsurface migration of radionuclides has occurred over limited distances (NUREG/CR-2383, page I-13). Except for tritium, migration to points outside the restricted area has not been a significant source of contamination by radionuclides. During the past quarter analysis of the water samples was suspended to permit the analysis of similar samples from Chalk River, Canada to proceed.

Well-logging of the inert atmosphere wells and experimental trench sumps (Figure 2) and of selected E-series wells (Figure 4) at Maxey Flats has confirmed that waste radionuclide migration along the sandstone marker bed occurred only within the restricted area. Cesium-137 (0.84 pCi/gm of soil) and ^{60}Co (0.54 pCi/gm of soil) were measured in well 14E on the Maxey Flats site at the top of the sandstone marker bed, or about 6.1 meters (20 feet) below the ground surface. The concentrations observed are below the concentrations measured in surface distribution studies (NUREG/CR-2383, Section II and NUREG/CR-1832, Section IV). Naturally-occurring radionuclides quantified included ^{226}Ra (0.99 pCi/gm), ^{232}Th (1.2 pCi/gm) and ^{40}K (10 pCi/gm). Strontium-90 is also present at or below the detection limit by well-logging techniques (≤ 10 pCi/gm of soil). We are continuing our efforts to quantify the ^{90}Sr concentration at this location.

During the past quarter work on this task was concentrated on the analysis of samples and data from the Chalk River, Canada site. The work being conducted

at Chalk River is part of a new proposal submitted to NRC entitled "Chemical Species of Migrating Radionuclides from Low Level Waste Disposal Sites," FIN B2862. A portion of this work is included herein at the request of NRC to implement FIN B2862 to allow field work to be conducted at Chalk River before winter weather conditions prohibited field sampling. The field sampling program utilizes existing wells and sampling points at Chalk River. The sampling program was developed taking into consideration the hydrogeology and stratigraphy of the site and in consideration of the application of statistics in siting and managing low-level radioactive waste disposal facilities.

Task C - Specific Radionuclide Mapping at Maxey Flats and Other Commercial Shallow Land Burial Sites

The Maxey Flats burial site has been contaminated by several sources. These include the overflow of water from the burial trenches as rainwater infiltrated and filled them, spillage during pumping of excess water from the trenches, fallout from entrained radionuclides in the evaporator plume, and surface runoff of radioactive solutions from the site. To obtain a better understanding of the actual sources of radioactivity in surface soils and vegetation on the burial sites and adjacent areas, a detailed study of the levels of radioactivity has been underway at Maxey Flats and related studies should also be useful at other commercial shallow land burial sites.

In situ gamma ray surveys conducted at the Beatty, Nevada⁽⁹⁾ and Maxey Flats, Kentucky (NUREG/CR-2383, Section II and NUREG/CR-1832, Section IV) shallow land burial sites confirmed that most of the waste radionuclides have been retained within the boundaries of those sites. The in situ measurements were correlated with and augmented by analysis of samples taken from each of the counting locations, since only the more energetic gamma ray emitting radionuclides are measured by the in situ procedures.

Slightly elevated tritium concentrations in water samples from springs and seeps outside the Maxey Flats restricted area suggested that other radionuclides might also be slightly elevated at these locations. However, all gamma-ray measurements have indicated that ^{60}Co and ^{137}Cs are present at ambient fallout levels. Strontium-90 and ^{238}Pu , ^{239}Pu , ^{240}Pu were also at ambient fallout levels, as determined from radiochemical separation and analysis of aliquots

of soil and water. The measurements indicate that radionuclide migration to these springs and seeps is not occurring, except possibly for tritium.

These studies have demonstrated that surface contamination by radionuclides has been retained mainly within the restricted area at Maxey Flats. Surface sediments in areas adjacent to the site, primarily on the west side, contain ^{60}Co in concentrations greater than fallout. The origin of the ^{60}Co is from surface runoff into the adjacent woods. Measurements made to date indicate that offsite contamination has not resulted from subsurface transport, except possibly for tritium. The relative contributions of tritium from air (evaporator output), surface and subsurface pathways are being further evaluated during the second six-month curtailment of evaporator use by the Commonwealth of Kentucky prior to submission of a topical report summarizing the area distribution studies.

Task D - Ecological Monitoring at Commercial Shallow Land Burial Sites

In this task we are developing efficient and statistically valid ecological field sampling procedures and methods for post-closure monitoring at shallow land burial sites. Kinds and amounts of radionuclides and trace elements in environmental samples are being measured with emphasis on biotic uptake, bioaccumulation, biotic transport, and ecological pathways in semi-wild ecosystems. We are emphasizing the sampling for pattern approach to provide three dimensional data that are amenable to rigid statistical analysis.

Forest sampling at Maxey Flats indicates the presence of a broad spectrum of naturally occurring and fallout-derived radionuclides in the litter layers, surface soil, and in freshly fallen autumn leaves⁽⁷⁾. Radiochemical analyses of gamma-ray emitting radionuclides show very low levels of all radionuclides and only ^{60}Co was elevated above ambient fallout levels and is most likely of disposal site origin. Collections of freshly fallen tree leaves indicate that leaf fall collection is a cost-effective, nondestructive sampling procedure that can be incorporated into ecological monitoring programs to indicate migration or lack of migration from shallow land disposal sites.

Tree sap and water from transpiring leaves have also been collected and analyzed for tritium; the results indicate that tritium uptake by deep-rooted trees likewise has potential to serve as a biomonitor of subsurface movement of water from burial sites. Tritium is environmentally one of the most mobile of radionuclides and it can move as tritiated water, as vapor, or in surface or groundwater flows. Trees obtain water from their rooting substrate and move it from roots to stems to leaves, and release water to air as vapor. Water can be extracted from fresh tree leaves and analyzed for tritium. If tritium is present in the root zone, it appears in the extracted water. Because trees have deeply penetrating roots, as compared to crop plants, they can be used to monitor subsurface water flows. Leaf water data indicate that tritium in above ambient fallout levels is present within the edge of the forest at Maxey Flats(2, 5). Maple trees, Acer sacharum, have the unusual ability to move water through their trunk before new leaves emerge in the spring. If maple tree roots have access to tritiated water, tritium will appear in the sap stream. Experience at Maxey Flats indicates that sap samples for tritium analyses can be obtained by tapping tree trunks in a cost-effective and a biologically nondestructive fashion useful for post closure ecological monitoring programs.

Autumn leaf-fall samples were collected from the 0.5 m² rectangular wire baskets placed beneath the permanently tagged trees adjacent to the Maxey Flats restricted area. These samples have been dried and shipped to our PNL facilities at Richland, Washington, for analysis by gamma-ray spectrometry. We have determined the exogenous radionuclides (e.g., ⁶⁰Co, ¹³⁷Cs) and obtained information on endogeneous (e.g., ⁷Be, ⁴⁰K, ²²⁶Ra, ²²⁸Th) radionuclides in some of the samples. Comparison of their radionuclide concentration with values reported from earlier sampling runs (NUREG/CR-2383, Section III, and NUREG/CR-1832, Section V) indicates again that buried waste radionuclides have not migrated into the forest adjacent to the Maxey Flats site.

Maple trees have now been tapped at more than 50 locations around the Maxey Flats disposal site and two trees have been tapped at a control location in a similar forest type near Cave Run Lake, about 12 miles south of Maxey Flats. Tritium concentrations in tree sap ranged from below detection level

to 290,000 pCi per liter of tree sap. The highest values occurred in two trees located along the western side of the disposal site and the lowest values were measured near Cave Run Lake, which represented fallout (background) tritium levels. Trees sampled in the vicinity of the disposal site generally had tritium values higher than fallout levels, with the highest concentrations being measured on the steep slope bordering the western side of the disposal site. In general, tritium values declined with increasing distance from the disposal site.

Several sampling trips have been made to evaluate the feasibility of using extractable leaf water from tree species other than maple as an indicator of tritium migration. In addition to the trees sampled around the fence, transects have also been made from the fence westerly into Drip Springs Hollow. Background samples were taken near Cave Run Lake, about 12 miles south of the Maxey Flats facility. Tritium concentrations in leaf water tended to decline as the growing season progressed⁽²⁾. The decline may be due to the additional time elapsing between samplings, extending the time since evaporator shutdown. Further dilution of the tritium contained in the groundwater that the trees were utilizing could have occurred during this time. The decline might also be an artifact of the unseasonably dry weather that prevailed at the Maxey Flats site during the most recent sampling periods.

Two trees sampled along the transect into Drip Springs Hollow contained much higher than normal concentrations of tritium (up to 2.7×10^7 pCi/l). Other trees near this location 50 meters downslope from the fenced area contained substantially lower tritium concentrations, indicating that the source of tritium tapped by these trees is highly localized. These trees may pinpoint an ideal location to establish a monitoring well outside the restricted area.

Plans have been formulated for additional sap sampling and leaf water sampling during calendar year 1984. The samplings will be coordinated with the scheduled restart of the evaporator at Maxey Flats, now set for April, 1984. Sampling will include samples taken before evaporator operation, during the initial stages of evaporator operation, and about six months after the start of evaporator operation.

Carbon-14 is present in the waste trenches at Maxey Flats and is one of the radionuclides of concern for ecological monitoring following site closure (10 CFR 61). Although ^{14}C has been widely used as a tracer in biological experiments, comparatively little is known about the root uptake of ^{14}C from soil or soil water. Carbon-14 analyses on selected samples of wood obtained from trees adjacent to the Maxey Flats restricted area were compared with analyses from trees located about 12 miles south of Maxey Flats facility, in the Daniel Boone National Forest. These limited data suggest that the site may have influenced ^{14}C levels very slightly in a few locations near the perimeter fence, where ^{14}C concentrations are slightly elevated above the "modern value," which is in the range of 8-8.5 pCi/gm. The source of ^{14}C in wood is from carbon dioxide assimilated from air in the photosynthesis process.

Task E - Technical Program Coordination for Low-Level Waste Research

The report PNL-4432-6, "Chemical Species of Migrating Radionuclides at Commercial Shallow Land Burial Sites, Quarterly Progress Report, July-September, 1983" was published and distributed in November, 1983.

The report NUREG/CR-3607, "Radionuclide Distributions and Migration Mechanisms at Shallow Land Burial Sites: 1982 Annual Report of Research Investigations on the Distribution, Migration and Containment of Radionuclides at Maxey Flats, Kentucky", was submitted to NRC for publication(1).

FIGURE 1. Maxey Flats waste trenches and experimental areas. The numbered lines show the locations of selected waste trenches and numbers and letters in parentheses indicate the locations of sumps. Experimental area number 1 includes the experimental trench and inert atmosphere wells. Experimental area number 2 includes the porous cup and tensiometer installations for studying water movement in the burial area. Experimental area number 3 includes the UCB lysimeter installations for water balance studies. These experimental areas support research studies by PNL, LANL, UCB and BNL.

FIGURE 2. Maxey Flats Experimental Area No. 1, containing the 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 PNL groundwater migration and chemical species studies. The experimental trench is also used by LANL, UCB, and BNL in research studies at the Maxey Flats Shallow Land Burial Site.

*DETECTED BY GC-MS

FIGURE 3. Formation of the molecular weight (MW) 244 species during analysis of Maxey Flats waste trench leachates. Ethylenediaminetriacetic acid (ED3A) is formed in the trenches by degradation of HEDTA or EDTA. Ring closure to form the MW 244 species containing the lactam ring occurs during methylation of the ED3A.

FIGURE 4. Locations of the E-series wells around the Maxey Flats restricted area. Three four-inch diameter wells - 13E, 14E and 3E - were logged and only 14E showed evidence of the presence of waste-derived radionuclides, e.g. ^{137}Cs , ^{60}Co and ^{90}Sr . These radionuclides had migrated short distances within the restricted area along the sandstone marker bed.

TABLE 1
Hydrophilic Organic Compounds^a in Waste Leachates from Maxey Flats

	Waste Trenches (ppb) ^{b,c}							
	7-3	19W	19S	23M	27	33L-4	33L-8	35
<u>Chelating Agents</u>								
Ethylenediaminetetraacetic Acid (EDTA)	5,016	12,364	4,261	4,895	962	402	78	868
N-Hydroxyethylethylenediaminetriacetic Acid (HEDTA)		8,350	888	19,511				
Ethylenediaminetriacetic Acid (ED3A) ^d	1,195	2,190	823	3,884	212			
<u>Dicarboxylic Acids</u>								
Oxalic Acid			111					
Succinic Acid	345		92		26	630	39	80
Methylsuccinic Acid	191				21		21	231
Pentanedioic Acid	248				41		54	406
Hexanedioic Acid	1,569	223	226	244	167	451	104	834
Methylhexanedioic Acid								305
Octanedioic Acid						781	57	
Nonanedioic Acid	455	338	610	210	96	1,394	111	314
<u>Monocarboxylic Acids</u>								
Hexadecanoic Acid							61	
Octadecanoic Acid						31	111	
<u>Oxygenated Acids</u>								
4-Oxo-Pentanoic Acid	2,165		241		29		53	185
<u>Aromatic Acids</u>								
Benzoic Acid			635					
2-Hydroxybenzoic Acid							27	
1,3-Benzenedicarboxylic Acid	381	276		440	77			877
1,4-Benzenedicarboxylic Acid	1,181	162		457				1,139
1,2,4-Benzenetricarboxylic Acid	311							
<u>Phthalates</u>								
Dimethyl Phthalate	8,817	2,836	1,772	1,673	391		98	13,565
Methylpropyl Phthalate		213		108	12			874
Diethyl Phthalate					851			

a) Methylated (BF₃/Methanol), acids identified as methyl esters;

b) No entry indicates compound is below detection levels;

c) Samples collected by BNL in 10/81, except for Trench 27 (10/79); 19S collected by PNL on 7/22/82;

d) Identified as Lactam (MW 244).

TABLE 2

Hydrophobic Organic Compounds in Waste Leachates from Maxey Flats

	Waste Trenches (ppb) ^{a,b}						
	7-3	19W	19S	23M	27	33L-4	33L-8
Barbital	48	11		9	35		
Pentobarbital	8	51		8	8		3
Bromodichloromethane			0.2				
Dichloriodomethane	11	11		1		11	2
Bis(2-Methoxyethyl)Ether				12			
Piperidinone			0.5				
Caffeine	4						
Vanillin						38	26
Vanillone						3	1
4-Hydroxybenzaldehyde						1	
Benzotriazole							trace
2(3H)-Benzothiazolone	78	177		53	40		34
Phthalic Anhydride	9	28					trace
Benzoic Acid	3			1	5		
Benzene Acetic Acid	7	6		4	7		6
Methylbenzoic Acid				2			
Benzene Propanoic Acid	9	2		7	51	1	2
2,5-Dimethyl Benzene Butanoic Acid		39					6
N-Butylbenzene Sulfonamide							40
C ₂ -Toluenesulfonamide			1				4
Polyethylene Glycol Oligomers	337						

a) No entry indicates compound is below detection level;

b) Samples collected by BNL in 10/81, except for Trench 27 (10/79); 19S collected by PNL on 7/22/82.

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