

CHARACTERIZATION OF RADIONUCLIDE BEHAVIOR
IN LOW-LEVEL WASTE SITES

A. P. Toste
L. J. Kirby
D. E. Robertson
K. H. Abel
R. W. Perkins

Pacific Northwest Laboratory
Operated by Battelle Memorial Institute
for the U. S. Department of Energy
Richland, Washington 99352

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Kirby, D. E. Robertson, K. H. Abel and R. W. Perkins,
Pacific Northwest Laboratory, Richland, Washington
99352.

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and R. W. Perkins

Battelle, Pacific Northwest Laboratory
Richland, Washington 99352

SUMMARY

Our laboratory is investigating the subsurface migration of radionuclides in groundwater at the Maxey Flats, Kentucky, shallow land burial site and at a low-level aqueous waste disposal facility. At Maxey Flats, radionuclide and tracer data indicate groundwater communication between a waste trench and an adjacent experimental study area. Areal distributions of radionuclides in surface soil confirm that contamination at Maxey Flats has been largely contained on site. Of the radionuclides detected in the surface soil, only ^3H and ^{60}Co concentrations appear to be derived from waste. Plutonium exists in the anoxic subsurface waters at Maxey Flats as a reduced, anionic complex; some of the plutonium appears to be complexed with EDTA, whereas organic acids seem to be associated with ^{137}Cs and ^{90}Sr . At the aqueous waste disposal site, ^3H and mainly anionic species of certain radionuclides, including ^{60}Co , ^{106}Ru , ^{99}Tc , ^{131}I , and traces of ^{238}Pu , ^{239}Pu , ^{240}Pu , appear to migrate from a trench through soil adjacent to the trench. Radionuclides in the particulate and cationic forms appear to be efficiently retained by the soil. In general, observations indicate that the physicochemical form of the radionuclides mediates their subsurface migration in groundwater at both waste disposal sites.

INTRODUCTION

One of the most crucial issues facing the nuclear industry today is the long-term disposal of radioactive waste. The concern is not only about waste being generated now or in the future but also about waste generated in the past which has already been stored under less than ideal conditions. An intensive research effort on waste management technology is currently underway. Research on the long-term storage of high-level radioactive waste appears to be progressing rapidly; several approaches are currently being assessed. However, many uncertainties still exist concerning the long-term fate of low- and high-level waste. Much remains to be understood about the subsurface migration of radionuclides in soil and groundwater. In spite of this, commercial shallow land burial of low-level radioactive waste has existed for over two decades at six sites in the U.S.: West Valley, New York; Maxey Flats, Kentucky; Sheffield, Illinois; Barnwell, South Carolina; Beatty, Nevada; and Hanford, Washington.

Unforeseen problems necessitated the closing of the West Valley and Maxey Flats sites in 1975 and 1977, respectively. Problems included water seepage into waste trenches and soil subsidence, resulting in dissolution by groundwater and subsurface transport of some of the stored radioactive waste at both sites.

The factors governing subsurface migration of radionuclides appear to be extremely complex. Recent research indicates that the mobility of radio-

nuclides depends on their physicochemical form. (1-3) For example, hexavalent plutonium and uranium migrate through soil faster than their corresponding tetravalent species, and technetium migrates faster as pertechnetate ion than it does in the reduced form. Also, decontaminating agents such as EDTA buried with radioactive waste or biodegradation products of organic material in waste or soil may complex with radionuclides, enhancing their mobility.

Research at Pacific Northwest Laboratory is aimed at determining the physicochemical forms of radionuclides in groundwater and their role in the subsurface migration of radionuclides. Two waste disposal sites were selected as field locations for the research: 1) the shallow land burial site at Maxey Flats, Kentucky; and 2) an aqueous low-level waste facility. In this report, we describe the following research at Maxey Flats: 1) a study of groundwater movement and radionuclide migration; 2) the areal distribution of radionuclides in soil in and around the site; and 3) the role of organic chelators in the migration of radionuclides. We also report on research undertaken at the aqueous low-level waste disposal system: 1) a study of the physicochemical forms of radionuclides entering and leaving the system; and 2) the possible role of organics in radionuclide transport at the site.

BACKGROUND

Maxey Flats Shallow-Land Burial Site

Access to the Maxey Flats shallow land burial site provided us with the opportunity to study the subsurface migration of radionuclides at an actual burial site where the groundwater is anoxic. The Maxey Flats site is one of several existing commercial shallow land burial sites in the eastern United States where rainfall is comparatively high. Trench cap subsidence and infiltration of surface water into waste trenches are greater problems at wet eastern sites compared to arid western sites. At Maxey Flats, the resulting seepage of contaminated groundwater and remedial construction efforts have spread the buried radionuclides. Atmospheric transport of radionuclides in the plume from an on site water evaporator and spillage of radioactive solutions have also contributed to the spread of radioactivity.

Aqueous Waste Disposal Site

Access to a low-level aqueous waste disposal facility provided us with the opportunity to study the transport of a large number of radionuclides in oxygenated groundwater under natural conditions. Reactor effluent water containing low levels of radioactivity are discharged into a seepage trench consisting of a rock-lined basin 3300 m² in area. The seepage trench is connected to a narrow, unlined trench 490 m long x 15 m wide, which lies on a bluff overlooking a river. The trench parallels the river. The effluent water percolates through soil between the trench and the river, some of it eventually emerging at seepage springs along the river bank. A large number of the radionuclides in the effluent

water are efficiently sorbed onto the soil during transit, but some migrate with the groundwater, presumably because of their physicochemical forms. The advantages of using the aqueous disposal system to study radionuclide migration in groundwater are: 1) accessibility; 2) measurable concentrations of radionuclides; 3) a well-defined source of aqueous effluents; and 4) short transit distance from the trench to the springs.

EXPERIMENTAL METHODS

In 1979 we constructed an experimental study area adjacent to the waste-filled area at Maxey Flats. A slit trench consisting of five sections was installed adjacent to waste trench 27 to intercept subsurface water flow from the waste-filled area.

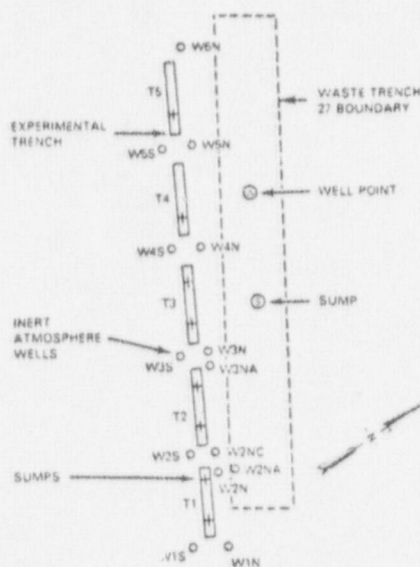


Figure 1. Experimental study area at the Maxey Flats shallow-land burial site. A series of experimental trench sections (T1-T5) and inert atmosphere wells (e.g., W1N) were installed adjacent to waste trench 27 to permit concurrent sampling of leachate from trench 27 (sump or well point) and water from the experimental trenches and wells.

Groundwater was sampled during and after the experimental trench's construction from sumps placed in each trench section. Nonradioactive tracers were added to sections of the experimental trench during its construction to monitor groundwater flow and test various design features of the experimental trench. Sodium bromide was added to trench sections 1, 2, 3 and 5 as a groundwater tracer to test whether groundwater communicates between the experimental study area and the waste-filled area. Pentafluorobenzoic acid was added to the cap of trench section 4 as a cap tracer to test whether water seeps through the experimental trench cap. A series of inert atmosphere sampling wells were subsequently installed around the experimental trench. The experimental wells were filled with argon between the periodic sampling trips.

Groundwater from waste trenches and inert atmosphere sampling wells at Maxey Flats were collected and concentrated prior to chemical analysis. Water samples which underwent detailed chemical speciation analyses for complexes of radionuclides and organic chelators were subsequently fractionated by steric exclusion chromatography prior to chemical analysis.

Trench and spring waters from the aqueous low-level waste site were pumped through a PNL large-volume water sampler, which fractionates chemical species on the basis of their physicochemical form into particulate, cationic, anionic and nonionic fractions.(4) Water was pumped directly from the trench inlet and from the springs through the samplers which consist of assemblies containing membrane filters, ion exchange resins and activated aluminum oxide beds. The filters remove the particulate forms of the radionuclides, the resins remove soluble cationic and anionic species and the activated aluminum oxide removes nonionic species. Five to ten liters of trench water and up to 2000 liters of spring water were pumped through these assemblies on three field trips: March 10, 1981; June 17, 1981; and September 22, 1981.

The analytical procedures used in our research have been described in detail elsewhere.(5) Radionuclides in the surface soil were analyzed by in situ gamma ray spectrometry using an intrinsic germanium detector. Radionuclides in water samples were analyzed using a number of techniques. Tritium analyses were performed by liquid scintillation; ^{90}Sr was chemically separated and counted on a beta proportional counter. Gamma-emitting radionuclides were analyzed by gamma-ray spectrometry using a $\text{Ge}(\text{Li})$ detector. Plutonium and americium were determined using solid state detectors and alpha energy analysis following radiochemical separations. Nonradioactive species were analyzed by neutron activation analysis using the PNL subcritical neutron multiplier,(6) and by chemical and instrumental analysis. Organic analyses were performed by gas chromatography (GC) and combined GC-mass spectrometry (GC-MS) using a procedure specially designed for radioactive water samples.(7)

RESULTS AND DISCUSSION

Studies at Maxey Flats Shallow Land Burial Site

Groundwater Movement and Radionuclide Migration. Initial radionuclide concentrations in water which flowed into the experimental trench during construction were comparable to those in waste trench 27 leachate (Table 1), indicating that subsurface migration of radionuclides has occurred at Maxey Flats. When the experimental trench was resampled later, the radionuclide concentrations were much lower, perhaps because the trench design perturbed the migration of radionuclides. Consequently, we installed and sampled a series of inert atmosphere wells around the experimental trench.

Table 1
Comparison of Radionuclide Concentrations
in Waste Trench 27 Leachate (5-78) and Initial
Experimental Trench Inflow (9-79)

Trench	Concentration, pCi/l				
	H	Total Pu	^{90}Sr	Co	^{137}Cs
Waste:	5.9+05	4.3+03	2.1+05	1.3+03	5.0+03
Experimental:					
Section T1	3.7+08	3.6+01	6.7+03	3.6+01	6.5+01
Section T2E	1.7+09	6.1+01	1.7+05	2.2+02	1.6+03
Section T2W	4.2+09	3.4+03	1.6+05	9.7+03	4.6+03
Section T3E	3.4+09	5.0+03	1.1+05	6.0+03	9.0+01

Evidence of the groundwater tracer, sodium bromide, was found in leachate from waste trench 27 and water from several experimental wells. Changes in bromine concentrations were observed in waste trench 27 leachate and in water from the experimental trench and inert atmosphere wells, indicating that there is some communication by groundwater flow between these locations (Table 2). Bromine concentrations in waste trench 27 leachate were quite low prior to the construction of the experimental trench. Following trench construction, the bromine concentration in waste trench 27 increased from 2.8 to 460 ppm, indicating that some of the NaBr tracer had moved into waste trench 27. Bromine concentrations in inert atmosphere wells W3N, W3NA and W2NA were high indicating strong communication, whereas wells W1N, W2N and W5N do not appear to be connected to the NaBr source. When radionuclide concentrations are compared (see Table 2), only W2NA and W3N appear to be closely related to waste trench 27, possibly connected to it by subsurface fractures.

experimental trench sections T2 and T3 have decreased markedly since the experimental trench was constructed, indicating that there is substantial movement of water away from the experimental trench in this region. The tritium concentration in waste trench 27 leachate has been relatively stable during the same time; 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 only one of the nonradioactive tracers installed in these caps was detected in trace quantity in section 4 of the experimental trench. Both the tritium and bromine data indicate 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.

Table 2
Comparison of Radionuclide and Bromine Concentrations in Waste Trench 27 Leachate (5-78 and 9-80) and Waters from Experimental Wells (9-80)

Waste Trench 27	Radionuclide Concentration (pCi/l)				Bromine ppm
	³ H	²³⁵ Pu	^{239,240} Pu	⁹⁰ Sr	
Sump (5-78)	5.9+08	4.1+03	6.7+02	2.1+05	2.8+00
Sump (9-80)					4.6+02
Well Point (9-80)					2.7+00
Experimental Wells (9-80)					
W1N	4.7+05	3.7+01	6.0+00	1.5+02	1.0+00
W2N	2.9+06	2.1+01	7.7+01	3.3+02	2.0+00
W2NA	7.7+08	2.4+02	4.8+00	1.7+04	1.2+02
W3N	1.2+09	2.4+03	4.4+01	3.4+04	8.2+02
W3NA	2.0+07	2.3+01	5.0+00	2.4+02	5.2+02
W5N	1.0+03	9.4+00	5.0+00	2.1+02	5.0+00

The presence of tritium in leachate from waste trench 27 allowed us to exploit an excellent in situ water tracer to monitor groundwater movement in the vicinity of waste trench 27 and the experimental study area. Tritium concentrations in water from the experimental trench are summarized in Figure 2. The tritium concentrations in samples from

Areal Distribution of Radionuclides in Soil

In mapping the areal distribution of radioactivity in Maxey Flats soil, we grouped the radionuclides we measured into two broad classes: 1) radionuclides endogenous to soil such as ⁴⁰K and ²³⁸Th; and 2) radionuclides exogenous to soil such as ⁶⁰Co and ¹³⁷Cs, which may originate from global fallout or the nuclear fuel process. For example, any ⁶⁰Co and ¹³⁷Cs identified at Maxey Flats could be attributed to global fallout or nuclear waste buried at the site, depending on the levels of radioactivity present. In situ counting measurements and soil analyses performed in the main drainage channels leading from the site indicated the radionuclide concentrations in the soil are very low. Cesium-137 and ⁶⁰Co were the most abundant radionuclides of exogenous origin. The concentrations of these two radionuclides in the surface soil generally ranged from 1-2 pCi/gm of soil but were occasionally slightly greater than 2 pCi/gm. Slightly elevated ⁶⁰Co 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. Cobalt-60 concentrations measured in the west drainage channel were confirmed by soil sample analyses to be surface-deposited. The ⁶⁰Co concentrations on the west side of the site approach global fallout levels about 100 meters from the site. Cesium-137 concentrations measured at all locations by in situ counting and by soil sample analysis were comparable to global fallout levels.

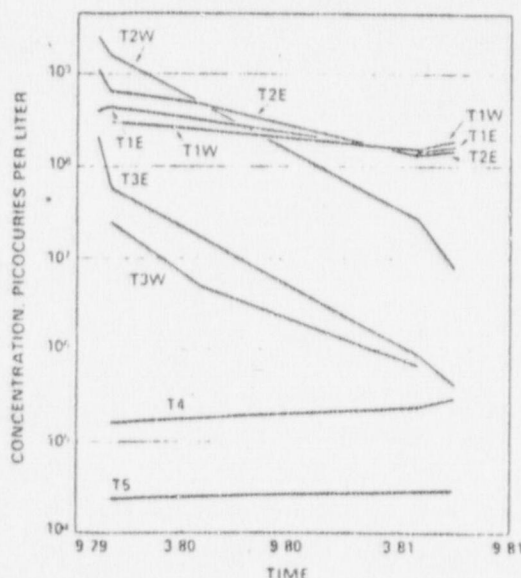


Figure 2. Tritium concentrations in the experimental trench sections during September 1979 through June 1981.

Organics and Radionuclide Migration

Water samples from waste trench 27 and several inert atmosphere sampling wells (W1N, W2NA, W3N, and W3NA) were analyzed for their hydrophilic and hydrophobic organic content. Selected hydrophilic and hydrophobic organics identified to date are listed in Table 3.

whether the organic compounds identified in the survey study are complexed to radionuclides. After steric exclusion fractionation of the waste trench 27 leachate, several peaks appeared in the UV chromatogram, 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 co-eluted with plutonium, suggesting that EDTA

Table 3
Selected Organic Compounds in Water from Experimental Wells
Compared to Waste Trench Leachate

	Waste Trench (ppb) ^b	Inert Atmosphere Wells (ppb) ^b				
	WFT-27	W1N	W2NA ^c	W2NA ^d	W3N	W3NA
<u>Hydrophilic Organics^a</u>						
Dimethyl Phthalate	92	2	115	11	1	2
Hexadecanoic Acid	18	4	48	12	8	4
Pentafluorobenzoic Acid ^e	Trace					
Octadecanoic Acid	17	4	111	9	6	3
Ethylenediaminetetraacetic Acid (EDTA) ^e	990		1330	840		
<u>Hydrophobic Organics</u>						
Methyl Metacrylate	2	9				
Dichloromethylbutane	2	7	38	3	21	36
Dodecane	4				19	19
2(3H)-benzothiazolone	835					
Pentafluorobenzoic Acid	1067					

^a Methyl Ester, ^b $\text{BF}_3/\text{Methanol}$; ^c No entry indicates compound is below detection level; ^d Sampled 5/18/81; ^e Sampled 8/17/81; ^f Methyl Ester

The chelating agent EDTA is the most abundant of the hydrophilic species, appearing in three samples at ppm levels. It was identified in the waste trench 27 leachate (0.99 ppm) and two distinct samples from experimental well W2NA (1.3 ppm and 0.84 ppm, collected on May 18, 1981, and August 17, 1981, respectively). The appearance of EDTA in waste trench 27 and in the vicinity of experimental trench section 2 but not in the vicinity of experimental trench sections 1 and 3 argues that there is a very specific communication between waste trench 27 and the vicinity of experimental well W2NA. The other organic species identified in the hydrophilic organic fraction are generally much less abundant (ppb levels), and consisted mainly of carboxylic acids, potential chelators of radionuclides such as ^{137}Cs and ^{90}Sr . A trace of the nonradioactive tracer pentafluorobenzoic acid (added to the cap of experimental trench section 4) was identified in the leachate, suggesting communication between experimental trench section 4 and waste trench 27.

Most of the hydrophobic organics isolated from the water samples were present at ppb levels. The most interesting exception was pentafluorobenzoic acid (1.1 ppm) which appeared in waste trench 27 leachate, confirming its presence in the vicinity of the waste trench and arguing again for communication between experimental trench section 4 and the waste trench. In general, water samples from the experimental trench area contained only a scattering of hydrophobic organics. Some of the compounds, however (e.g., dichloromethylbutane), are not typically found in uncontaminated soil samples. In contrast, the leachate from waste trench 27 contained quite a diversity of hydrophobic organics, possibly associated with waste dumped into the trench.

The presence of relatively high levels of organic chelators like EDTA in the water samples prompted a second major research effort, a detailed chemical speciation study aimed at determining

is complexed with plutonium in the waste trench leachate. This finding agrees well with an earlier observation that plutonium in leachate from waste trench 27 exists as a strong anionic complex.⁽⁸⁾ Earlier binding studies with solutions of Pu^{3+} , Pu^{4+} and EDTA also revealed that the presence of a strong chelator like EDTA, even at low concentrations comparable to those at Maxey Flats, may actually be more important than the oxidation state of plutonium in determining whether plutonium adsorbs or migrates in soil.⁽⁸⁾

A variety of organic acids and other hydrophilic compounds co-eluted with the ^{90}Sr and ^{137}Cs . It thus appears that chelation of radionuclides by polar organic species such as organic acids may provide a mechanism for the migration of radionuclides at the Maxey Flats commercial shallow land burial site.

Studies at an Aqueous Low-Level Waste Disposal Site

A variety of radionuclides were detected in the trench water including: ^{46}Sc , ^{51}Cr , ^{54}Mn , ^{57}Co , ^{59}Fe , ^{60}Co , ^{65}Zn , ^{95}Zr , ^{95}Nb , ^{103}Ru , ^{106}Ru , ^{124}Sb , ^{125}Sb , ^{131}I , ^{134}Cs , ^{137}Cs , ^{140}Ba , ^{141}Ce , ^{144}Ce and transuranic radionuclides. Most of the radionuclides enter the trench at least partially in the particulate, or insoluble, form (Table 4). The soluble species of each radionuclide in the trench water were either predominantly cationic, anionic, nonionic or a mixture of these forms. Cobalt-60, ^{54}Mn , ^{59}Fe and ^{137}Cs are the predominant gamma-emitters. Occasionally, a number of short- to intermediate-lived radionuclides appeared in the trench water, including the radioiodines (^{131}I , ^{132}I , ^{133}I , ^{135}I), ^{140}Ba - ^{140}La and ^{99}Mo .

In general, the physicochemical forms of the radionuclides in the trench and spring waters dif-

ferred markedly. The radionuclides emerging in the riverbank springs are primarily those which exist in anionic or nonionic species, i.e., ^{51}Cr , ^{60}Co , ^{99}Tc , ^{106}Ru , ^{125}Sb , and ^{131}I . As the trench water percolates through the soil to the riverbank springs the particulate and cationic species appear to be efficiently removed by filtration, sorption, and ion exchange mechanisms by the soil. Unlike

affect their mobility in groundwaters. Therefore, we undertook a comprehensive identification and quantification of the major and minor dissolved organic species in the trench and spring waters. The major organic compound in a sample of trench water has been identified as citrate (0.38 ppm), a chemical used extensively in decontamination solutions. The trench water also contained traces of oxalic acid (another

TABLE 4

Physicochemical Forms of Radionuclides in Trench and Spring Waters
at *an* Aqueous Low-Level Waste Site

Physicochemical Form	% of Total in Various Forms									
	^{60}Co		^{99}Tc		^{103}Ru		^{124}Sb		^{131}I	
	Trench	Springs	Trench	Springs	Trench	Springs	Trench	Springs	Trench	Springs
Particulate	49	0.3	2	0.03	66	0.1	29	0.09	0.4	0.0
Cationic	49	17	3	0.08	16	4	35	0.5	19.6	0.4
Anionic	1	73	93	99	13	91	23	5.4	78	92
Nonionic	1	9	2	1	5	5	13	94.5	2	7.5

the ^{60}Co in the trench water, which was mainly particulate and cationic, the ^{60}Co in the spring water was mainly anionic and cationic. The physicochemical form of ^{106}Ru in the trench water varied considerably. During the March 10, 1981 sampling, the ^{106}Ru was mainly cationic, whereas on June 17, 1981 and September 22, 1981 particulate ^{106}Ru was prevalent, and the anionic and cationic forms were approximately equivalent. However, the ^{106}Ru observed in the spring waters was always mainly anionic. In general, the ^{125}Sb in the trench water was equally particulate, cationic and anionic. In the spring water, however, 80-92% of the ^{125}Sb was nonionic, the remainder being anionic. In contrast, the ^{99}Tc and ^{131}I in both trench and spring waters were mainly anionic as expected.

In general, our experimental data agree well with theoretical predictions. Based on thermodynamic data, Rai and Serne(9) predicted the soil solution species of a number of elements in various weathering environments. The major predicted species of antimony expected under oxic and pH conditions like the trench and spring waters are HSbO_2^0 , $\text{Sb}(\text{OH})_3^0$, and SbO_2^- , which is in good agreement with our analytical results. The predicted ruthenium species would be RuO_4^- , RuO_4^{2-} , and RuO_4^0 , which again is in good agreement with our experimental data. The predicted major species for cobalt would be Co^{+2} , CoOH^+ , and CoSO_4 . These forms could account for the small amounts of cationic ^{60}Co in the spring water. The only anionic species predicted by Rai and Serne of any significance is $\text{Co}(\text{OH})_3^-$, but this was predicted to be present at concentrations many orders of magnitude below the major cation and nonionic cobalt species.

The dissolved organics in the trench and spring waters were also analyzed. The dissolved organic carbon concentrations in the trench and spring waters were typical of natural surface waters, generally ranging between 2 to 3 mg/l. These concentrations are considered sufficiently high to complex metal radionuclide species and thus

decontamination agent), silicone oils (presumably used as lubricants at the reactor) and palmitic and stearic acids. The major organic compounds in the spring water were palmitic and stearic acids (1.8 ppb and 2.3 ppb, respectively), whose origin is biological. These acids could originate as natural decay products of the decomposing biota in the trench or they could be a residue from soaps or detergents used in the reactor. Organic acids have the ability to complex iron and other transition metal radionuclides, and could enhance their groundwater mobility at this site. We are continuing to characterize the organic matter in this system to determine its role in the migration of radioactivity in the groundwater at this site.

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