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SOURCE TERMS OF RADIONUCLIDES IN SHALLOW-LAND-BURIAL SITES*

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ABSTRACT

A research program is under way to provide source term data on the chemical and radionuclide contents of trench waters at low level radioactive waste disposal sites and to provide information on the geochemical behavior of radionuclides under simulated field conditions. Trench waters were sampled at four commercial waste burial sites and analyzed for chemical and radionuclide constituents. The sorption, diffusion and precipitation behavior of radionuclides were studied using site specific sediments and trench waters. Radionuclide leach rates from solidified waste, under simulated trench conditions, were also determined.

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INTRODUCTION

A field and laboratory program was designed to study the existing commercial low level radioactive waste disposal sites. The prime consideration for continued use of shallow land burial practices for the disposal of low level radioactive waste is the containment of radionuclides from the environment. This investigation provides source term data for radionuclides and other solutes in trench waters at the sites and will describe physical and chemical properties of the geochemical system that control the movement of radionuclides. These objectives were pursued in four task areas: (1) Trench water chemistry; (2) Characteristics of sediments from representative lithostratigraphic units at disposal sites; (3) Radionuclide migration studies; (4) Waste form leaching under simulated trench environments.

A summary of the past and present activites of the Source Term of Radionuclides project is shown in Table 1 and described in earlier reports.[1-6]

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Table 1

Past and Present Activites of the Source Term of Radionuclides Project

Disposal Sites	Trench Water Chemistry	Sediment Character-ization	Radio-nuclide Migration	Leaching in Simulated Trench Environment
Maxey Flats	1976-1979, 1981	IP	Sorption Precipitation	
West Valley	1977, 1978,	IP	Sorption Precipitation	
Barnwell	1977-1979, 1980	IP	Sorption Diffusion	IP
Sheffield	1979, 1982	IP	Sorption Diffusion	

IP = in progress.

TRENCH WATER CHEMISTRY

Since 1976, water samples have been obtained periodically from trenches and wells at the commercial waste disposal sites located in Maxey Flats, KY; West Valley, NY; Barnwell, SC; and Sheffield, IL. Sampling procedures were developed to maintain inert conditions while measuring pH, Eh, conductance, dissolved oxygen and temperature in the field. Figure 1 shows the equipment used to sample trench water at the disposal sites. In the laboratory, the trench waters were analyzed for dissolved radionuclides (^{60}Co, ^{137}Cs, ^{3}H, ^{90}Sr, and Pu isotopes) and major inorganic constituents (Na^+, K^+, NH_4^+, Ca^{+2}, Mg^{+2}, Ba^{+2}, total Fe, Cl^-, SO_4^{-2}, HCO_3^-). In addition, selected trench water samples are being analyzed for chelating agents and carrier cations such as Sr^{+2}, Cs^+, and Co^{+2}.

Tritium is the dominant radionuclide in the trench waters at all the sites. The concentrations of ^{90}Sr, $^{238,239,240}Pu$ and ^{3}H at Maxey Flats and West Valley exceed those at Barnwell and Sheffield by several orders of magnitude (Table 2).

As an example, ternary plots displaying the major cation and anion compositions of Maxey Flats trench waters are shown in Figures 2 and 3.

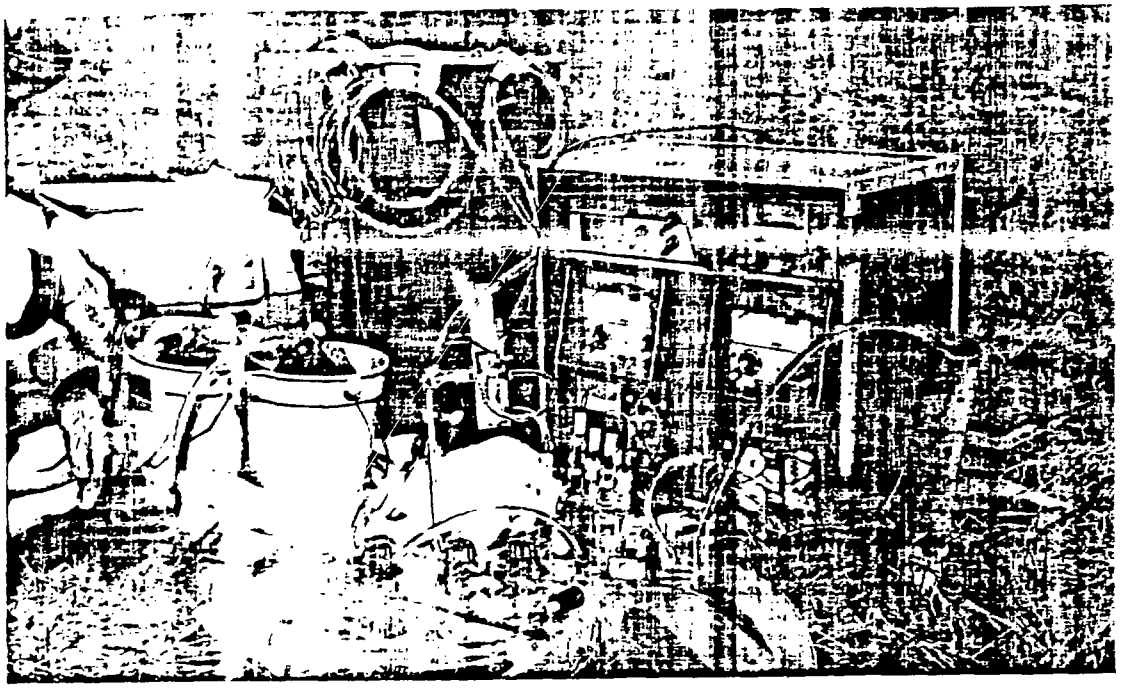

Figure 1. Portable anoxic collection system used to sample trench water from low level radioactive waste disposal sites.

Table 2

Maximum Observed Radionuclide Concentrations in Trench Waters From Low Level Waste Disposal Sites

	Concentration (pCi/L)			
	Maxey Flats	West Valley	Barnwell	Sheffield
Gross Alpha	5.9×10^{5}	1.3×10^{5}	1.6×10^{1}	8.6×10^{2}
Gross Beta	5.3×10^{6}	3.8×10^{7}	9.6×10^{2}	5.6×10^{4}
Tritium	5.1×10^{9}	3.7×10^{9}	7.8×10^{8}	6.2×10^{5}
^{238}Pu	2.1×10^{5}	1.6×10^{5}	4.6×10^{0}	1.4×10^{0}
$^{239/240}Pu$	2.1×10^{4}	3.4×10^{2}	9.3×10^{-1}	9.3×10^{-1}
^{90}Sr	5.4×10^{5}	2.4×10^{7}	4.8×10^{1}	2.2×10^{3}
^{134}Cs	1.7×10^{3}	5.0×10^{3}	3.4×10^{1}	1.4×10^{4}
^{137}Cs	2.3×10^{4}	1.0×10^{6}	5.1×10^{2}	3.0×10^{4}
^{60}Co	5.0×10^{4}	1.8×10^{3}	3.7×10^{2}	2.4×10^{4}
^{241}Am	4.5×10^{4}	3.6×10^{2}	---	---

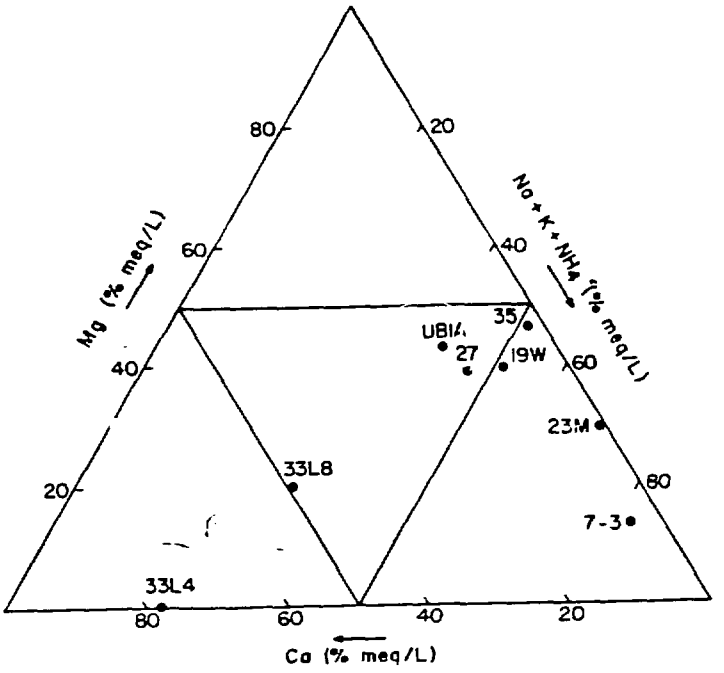

Figure 2. Trilinear diagram displaying major cation compostion of Maxey Flats trench waters. Data for well water UB1A are also shown.

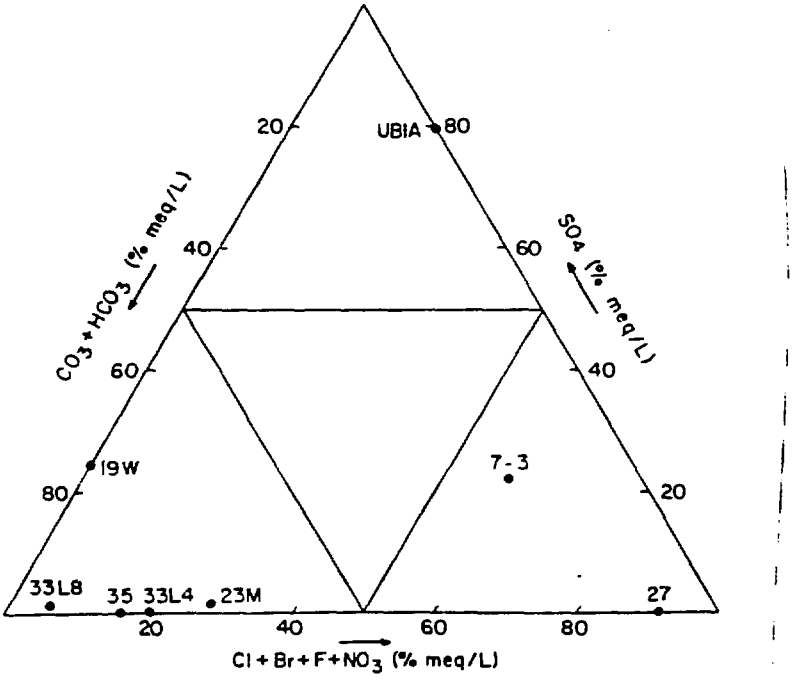

Figure 3. Trilinear diagram displaying major anion composition of Maxey Flats trench waters. Data for well water UB1A are also shown.

SEDIMENT CHARACTERISTICS

Basic information on the physical and chemical characteristics of site specific sediments is essential to the evaluation of their radionuclide retention properties. The characterization included particle size distribution, cation exchange capacity, surface area, organic matter, extractable iron and total carbonate. In addition, parameters relevant to radionuclide migration studies, such as bulk density, porosity and hydraulic conductivity, were determined on repacked sediment cores from the Hawthorne formation at Barnwell.

RADIONUCLIDE MIGRATION TESTS

The principal processes which control radionuclide migration are transport processes and retardation. Transport processes include advection, mechanical dispersion and diffusion. Some reactions such as complex formation with inorganic ligands and with chelating agents can enhance radionuclide mobility. The processes which retard radionuclide migration consist of a large group of reactions that occur between dissolved radionuclides and the sediments and are collectively known as sorption processes. Sorption is the primary process by which radionuclide transport is retarded. Precipitation-coprecipitation reactions and radionuclide scavenging by hydrous oxide/hydroxide precipitates can also retard radionuclide migration.

In this program, we have considered diffusion and sorption as the principal radionuclide migration processes. For this purpose, quantitative measurements of radionuclide diffusion coefficients (D) and sorption coefficients (K_d) using site specific sediments and trench waters are being made. In addition, radionuclide retardation factors and the extent of radionuclide scavenging by iron oxide/hydroxide precipitation upon oxidation of anoxic trench waters are being determined.

BATCH SORPTION

Mathematical models used to calculate radionuclide transport by groundwater generally assume ideal chromatographic behavior for radionuclide sorption. The distribution coefficient, K_d, must be experimentally determined to account for the partitioning of each radionuclide between the solution and the solid phases. Sorption isotherms for selected radionuclides were determined using site specific sediments and radionuclide spiked trench waters.

DIFFUSION

The diffusion behavior of selected radionuclides is being studied by direct measurement of radionuclide penetration into a sediment plug. The diffusion of tritium through clay rich sediments is of particular interest to low level disposal sites because of the abundance of tritium in the trench waters. In addition, experiments are in progress to determine apparent diffusion coefficients for reactive tracers such as ^{137}Cs, ^{60}Co, ^{85}Sr, etc. The diffusion measurements for reactive tracers represent both diffusion and sorption. Tritium migration, on the other hand, reflects sorption-free diffusion.

RETARDATION

Experiments were initiated to measure radionuclide retardation using site specific sediments and simulated trench water. The retardation of a radionuclide is defined conceptually as the ratio of the velocity (V) at which the radionuclide moves through the porous medium divided by the linear velocity.

$$R_f = V/V_l$$

The linear groundwater velocity, V_l, is equal to the velocity at which a nonsorbed tracer (tritiated water) passes through a porous sediment. A schematic for the apparatus for radionuclide retardation testing is shown in Figure 4.

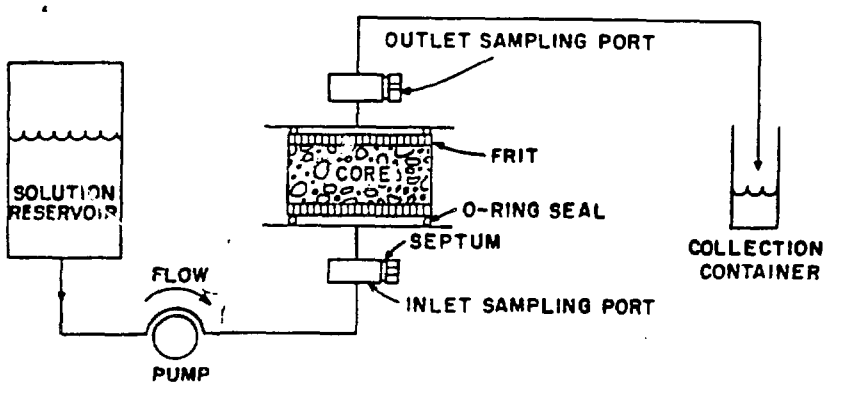

Figure 4 A schematic of the apparatus for radionuclide retardation testing by continuous water flow through sediment core.

PRECIPITATION-COPRECIPITATION

Precipitation-coprecipitation reactions and radionuclide scavenging by precipitates can result in significant retardation of radionuclide migration in both unsaturated and saturated zones. Tests to measure the extent of coprecipitation of radionuclides by iron oxyhydroxide as a result of the oxidation of anoxic trench waters are under way. Scoping experiments to study the scavenging of radionuclides by metal sulfide and carbonate precipitates in anoxic trench waters have been initiated.

LEACHING OF WASTE FORM IN SIMULATED TRENCH ENVIRONMENTS

Data on the leaching behavior of ion exchange resins and boric acid waste streams solidified in cement have been generated using a modified IAEA leach test at BNL.[7] Such tests, although useful in making relative comparisons of waste forms, represent a worst case leaching condition in a water saturated environment. In a realistic situation, the trench environments are only partially saturated and the water moves away from the waste at a rate controlled by the groundwater infiltration rate and the permeability of the sediment. Experiments are being conducted to leach solidified waste surrounded by site-specific soil in a flowing, partially water saturated environment (Figure 5). The waste form consists of Na^+ form resin beads, loaded with ^{137}Cs and ^{85}Sr, and solidified in Portland I cement. Simulated trench water is used as a leachant. For sake of comparison, parallel experiments are being performed using high density polyethylene beads as an inert, porous medium to determine leachability of the waste forms with minimal sorption of the radionuclides onto the surrounding medium.

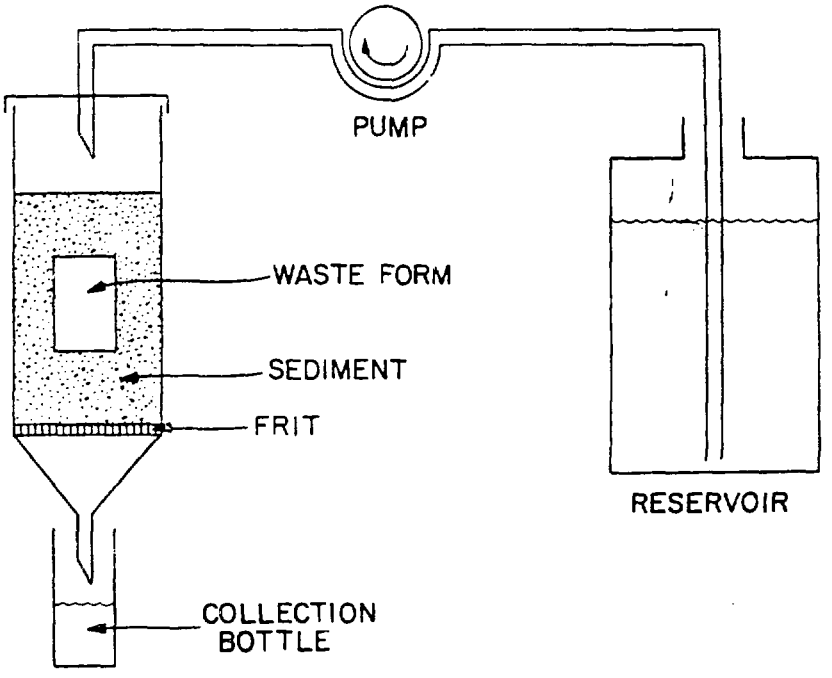

Figure 5 A schematic of the apparatus to measure leaching of solidified waste in a porous solid medium with continuous unsaturated leachant solution flow.

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