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EVALUATION OF ISOTOPE MIGRATION-LAND BURIAL

WATER CHEMISTRY AT COMMERCIALLY OPERATED LOW-LEVEL RADIOACTIVE WASTE DISPOSAL SITES

QUARTERLY PROGRESS REPORT
JULY - SEPTEMBER 1982

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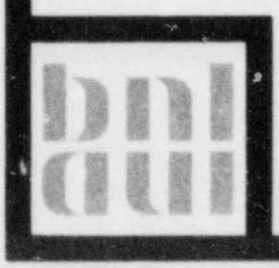
NUCLEAR WASTE MANAGEMENT DIVISION

DEPARTMENT OF NUCLEAR ENERGY BROOKHAVEN NATIONAL LABORATORY
UPTON, NEW YORK 11973



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SUMMARY

This report presents the progress made in the following areas in the Source Term of Radionuclides (Shallow Land Burial) program during the fourth quarter of fiscal year 1982:

- Trench water from the Sheffield diposal site was collected and analyzed for the major cations and gamma emitting radionuclides.
- Important properties of soils from the Sheffield disposal site were determined.
- Diffusion coefficients were calculated for ^{3}H, ^{85}Sr, ^{134}Cs, ^{137}Cs and ^{60}Co from radionuclide penetration depth profiles in repacked sediment from the Hawthorne formation at Barnwell.
- Experiments to study leaching of cement-resin composite waste forms in an environment simulating burial conditions are under way.

ACKNOWLEDGEMENTS

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The cooperation and assistance of R. Moore of U. S. Ecology for his assistance during our sampling trip to the Sheffield disposal site is gratefully acknowledged.

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EVALUATION OF ISOTOPE MIGRATION - LAND BURIAL WATER CHEMISTRY AT COMMERCIALLY OPERATED LOW-LEVEL RADIOACTIVE WASTE DISPOSAL SITES JULY-SEPTEMBER 1982

1. TRENCH WATER SOURCE TERM INVESTIGATION AT THE SHEFFIELD DISPOSAL SITE

1.1 Introduction

Disposal site source term investigations involve characterization of trench waters and evaluation of the effectiveness of disposal sites for retaining radionuclides introduced from the buried radioactive waste. Specifically, source term investigations include field measurements of pH, Eh, dissolved oxygen and specific conductance, and laboratory analyses for inorganic and radionuclide constituents in trench waters.

In July 1982, water samples were collected from trench 18 at the Sheffield disposal site. A trench location map is shown in Figure 1.1. Trench 18 was sampled because that was the only trench containing water at that time. In this report, we present the trench water chemistry in terms of the field parameters, major cations and dissolved radionuclides.

1.2 Experimental

The trench water, sampled using an anaerobic procedure, was stored at 4°C in an inert atmosphere to prevent oxidation. In the laboratory, an aliquot of the sample was filtered anaerobically through a 0.45 μm membrane filter and analyzed immediately for alkalinity (titration), ferrous and total iron (colorimetric), ammonia (probe), and pH (probe). A second aliquot of trench water was filtered; the filtrate was acidified with ultrapure nitric acid and subsequently analyzed for dissolved radionuclides and metals. Detailed descriptions of field sampling, filtration, and analytical procedures are given elsewhere (Czyscinski et al., 1981).

1.3 Results

Results of field measurements of pH, Eh, dissolved oxygen, specific conductance, and temperature are compiled in Table 1.1. The major cation composition of the trench water is presented in Table 1.2; analysis of the anion composition is in progress. The concentrations of the gamma emitting radionuclides identified are shown in Table 1.3. Radiochemical analyses of gross alpha, gross beta, tritium, ^{90}Sr and plutonium isotopes are in progress.

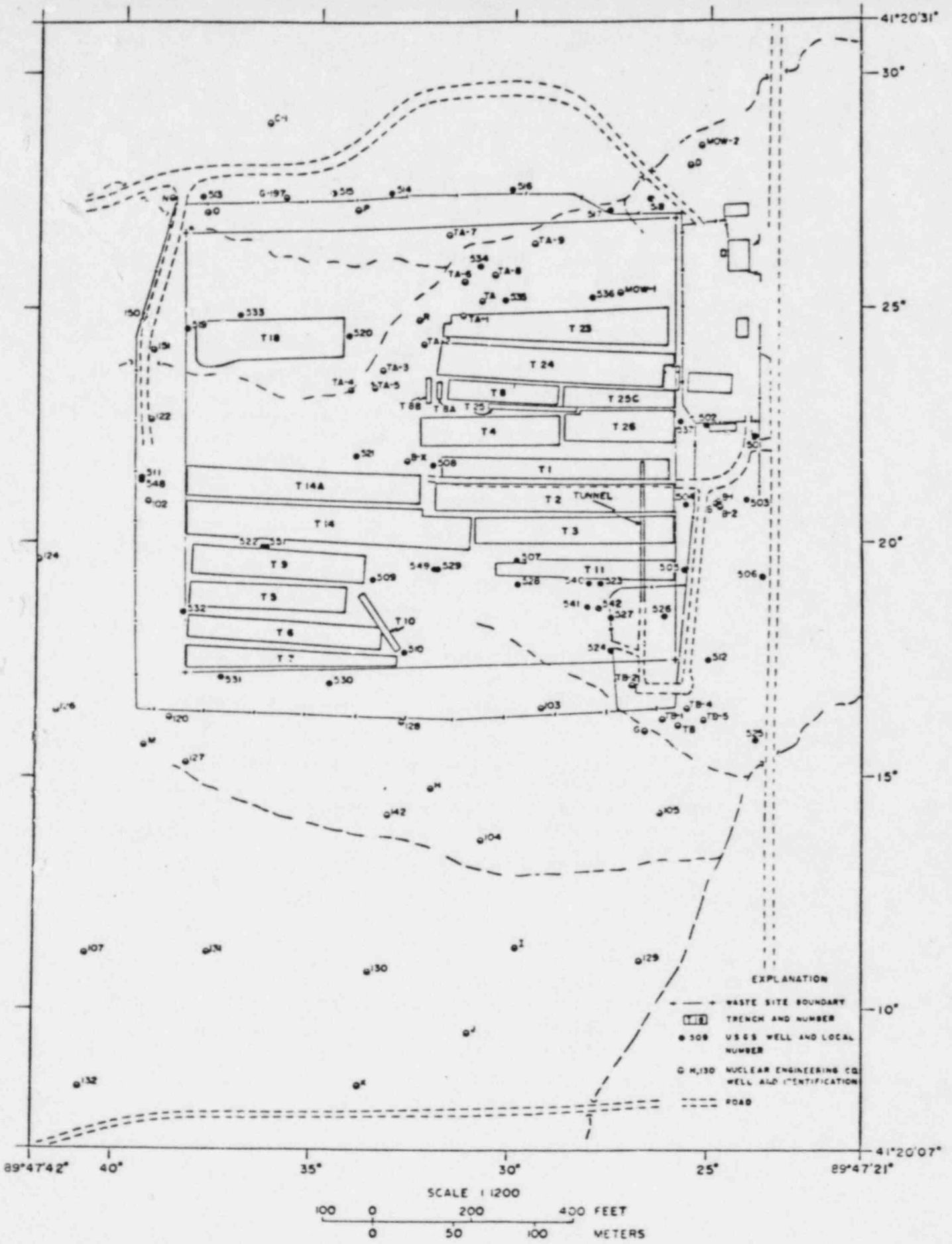

Figure 1.1 Map of the low-level waste disposal facility at Sheffield, IL., operated by U. S. Ecology (Foster and Erickson, 1980).

Table 1.1

Field Measurements of Trench 18 Water From the Sheffield Disposal Site[a]

Parameter	Observed Value
Temperature	10.5°C
Specific Conductance	2000 µmho/cm
Dissolved Oxygen	<0.1 mg/L
Eh	256 mV
pH	7.1

[a]Sampled July 1982.

Table 1.2

Cation Concentrations in Trench 18 Water From the Sheffield Disposal Site[a]

Cation	Concentration (mg/L)
Barium	<1
Calcium	219
Cesium	<0.1
Iron-total[b]	<0.1
Iron-Ferrous[b]	<0.1
Magnesium	126
Potassium	73
Sodium	76
Strontium	0.6
Ammonium[c] (as N)	8.5

[a]Sampled July 1982.
[b]Colorimetric analysis.
[c]Probe analysis.

Table 1.3

Dissolved Radionuclides in Trench 18 Water From the Sheffield Disposal Site[a]

Radionuclides	Activity pCi/L[b]
^{60}Co	2.7×10^{2} (5)
^{137}Cs	4.8×10^{1} (34)
^{134}Cs	Not detected
^{90}Sr	In progress
^{3}H	"
Gross alpha	"
Gross beta	"
$^{238,239}Pu$	"

[a]Sampled July 1982.
[b]The number in ( ) represents 2σ percent counting uncertainty.

2. CHARACTERIZATION OF SOIL SAMPLES FROM THE SHEFFIELD DISPOSAL SITE

2.1 Sample Collection

In May 1982, a core containing units of Peoria Loess, Radnor Till, Roxana Silt, and Hulick Till was collected by P. Piciulo (Brookhaven National Laboratory) at the Sheffield disposal site. The core was taken at a location outside the boundary of the disposal site, about 100 feet east of boring 504 (Figure 1.1). In addition, a sample of the Toulon member pebbly sand was obtained from the tunnel located at a depth of ~12 meters beneath trenches 1, 2, 3, and 11. The soil types sampled and their corresponding depths in core are given in Table 2.1.

Table 2.1

Soil Types Sampled at the Sheffield Disposal Site

Soil Type	Depth in Core (meters)
Peoria Loess	0.86-1.68
Roxana Silt	3.94-4.75
Radnor Till	7.34-8.30
Toulon (Pebbly sand)	~12 (sampled from tunnel)
Hulick Till	14.0-14.7

2.2 Soil Characterization

The procedures used to characterize the soil samples are described elsewhere (Pietrzak and Dayal, 1982a). Briefly, the samples were air dried, crushed and sieved to exclude gravel and pebbles (>2 mm). Soil characterization included determination of the following properties: particle size distribution, surface area, extractable iron, organic matter, calcium carbonate, and cation exchange capacity.

2.3 Results

The soil characterization results are given in Table 2.2. A ternary plot displaying sand, silt, and clay compositions of the Sheffield soils is shown in Figure 2.1.

Table 2.2

Characteristics of Soils at the Sheffield Waste Disposal Site

Parameter[a]	Peoria Loess	Roxana Silt	Radnor Till	Hulick Till	Pebbly Sand
Sand (%)	4.2 ± 0.1	2.4 ± 0.6	30 ± 7	41 ± 1	98.6 ± 0.1
Silt (%)	77.5 ± 0.3	88 ± 2	30 ± 2	39 ± 1	0.2 ± 0.1
Clay (%)	18.3 ± 0.4	10 ± 2	41 ± 8	20.2 ± 0.4	1.23 ± 0.03
Cation Exchange Capacity (meq/100g)	9.4 ± 0.1	17.4 ± 0.7	16.8 ± 0.4	10.21 ± 0.03	2.3 ± 0.2
Surface Area[b] (m^2/g)	I.P.[c]	I.P.	I.P.	I.P.	I.P.
Organic Matter (%)	2.1 ± 0.3	0.12 ± 0.07	0.5 ± 0.9	1.92 + 0.08	0.28 ± 0.04
Iron (%)	3.19 ± 0.05	3.30 ± 0.06	8.3 ± 0.4	1.4 ± 0.1	0.41 ± 0.04
Carbonate as $CaCO_3$ (%)	25.5 ± 0.6	6.4 ± 0.3	2.7 ± 0.2	18.3 ± 0.8	7.8 ± 0.8

[a]Analyses performed on air-dried sieved soil (<2 mm).
[b]Ethylene glycol absorption.
[c]I.P. = in progress.

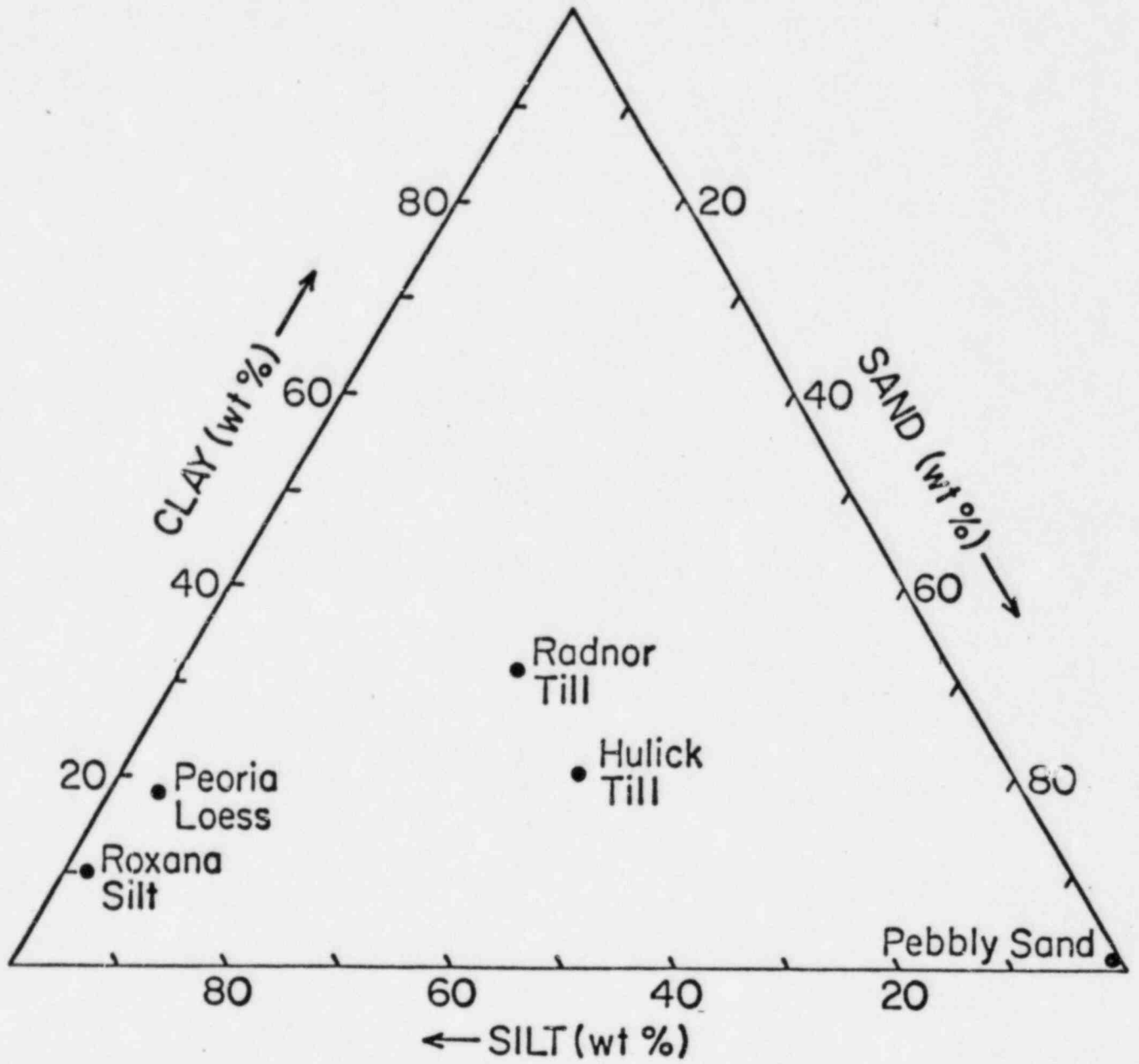

Figure 2.1 Trilinear diagram showing sand, silt and clay compositions of Sheffield soils.

3. RADIONUCLIDE MIGRATION STUDIES

3.1 Diffusion Coefficients of Radionuclides

3.1.1 Introduction

Radionuclide penetration experiments to determine the concentration profiles of radionuclides in water-saturated sediment from the Barnwell disposal site were reported in the last quarterly report (Pietrzak and Dayal, 1982a) based on a method employed by Fried et al. (1980). The concentration profiles have been used to calculate the effective diffusion coefficients for ^{3}H, ^{85}Sr, ^{134}Cs, ^{137}Cs and ^{60}Co as described below in 3.1.2.

Briefly, the experiment consisted of measuring the radionuclide concentrations in sections of a wet sediment plug that had been left in contact with radionuclide spiked simulated Barnwell trench 6D1 water. The diffusion experiments were allowed to continue for periods of 2.2 days and 17.2 days. The data obtained from the long-term experiments were used in the diffusion calculation.

3.1.2 Calculation of the Diffusion Coefficient

The radionuclide depth profiles observed in the sediment plugs (Pietrzak and Dayal, 1982a) were represented by curves calculated as solutions to the linear diffusion equation

$$\frac{\delta C}{\delta t} = D. \frac{\delta^2 C}{\delta x^2} \qquad (1)$$

C = bulk radionuclide concentration
D = effective diffusion coefficient
x = radionuclide penetration depth into the sediment
t = duration of the experimental period.

The solution to the planar diffusion equation was used to calculate the diffusion coefficients as outlined by Crank (1975). This calculation provides an estimate of the upper limit of the effective diffusion coefficients.

3.1.3 Results

The observed diffusion profiles of ^{85}Sr, ^{134}Cs, ^{137}Cs, ^{60}Co, and tritium are shown in Figures 3.1 to 3.5. The distance from the sediment/water interface represents the extent of radionuclide penetration into the sediment. C/Co corresponds to the ratio of the radionuclide concentration in the topmost sediment section to that in the section at depth. Continuous curves represent theoretical diffusion profiles based on plane-sheet diffusion model. The effective diffusion coefficients estimated from these data are given in Table 3.1. In the calculation, some data points representing deeper sections of the core were not considered because of poor counting statistics and high background.

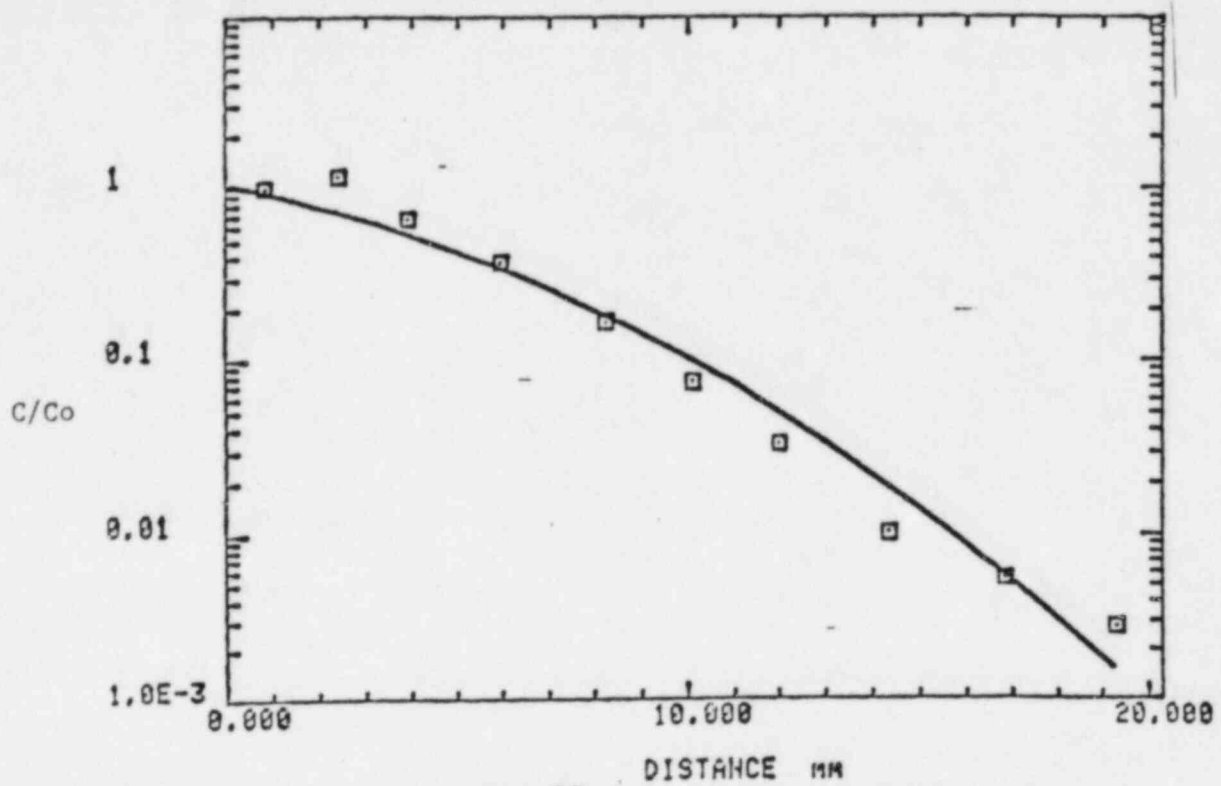

Figure 3.1 Diffusion profile of ^{85}Sr in a repacked sediment core.

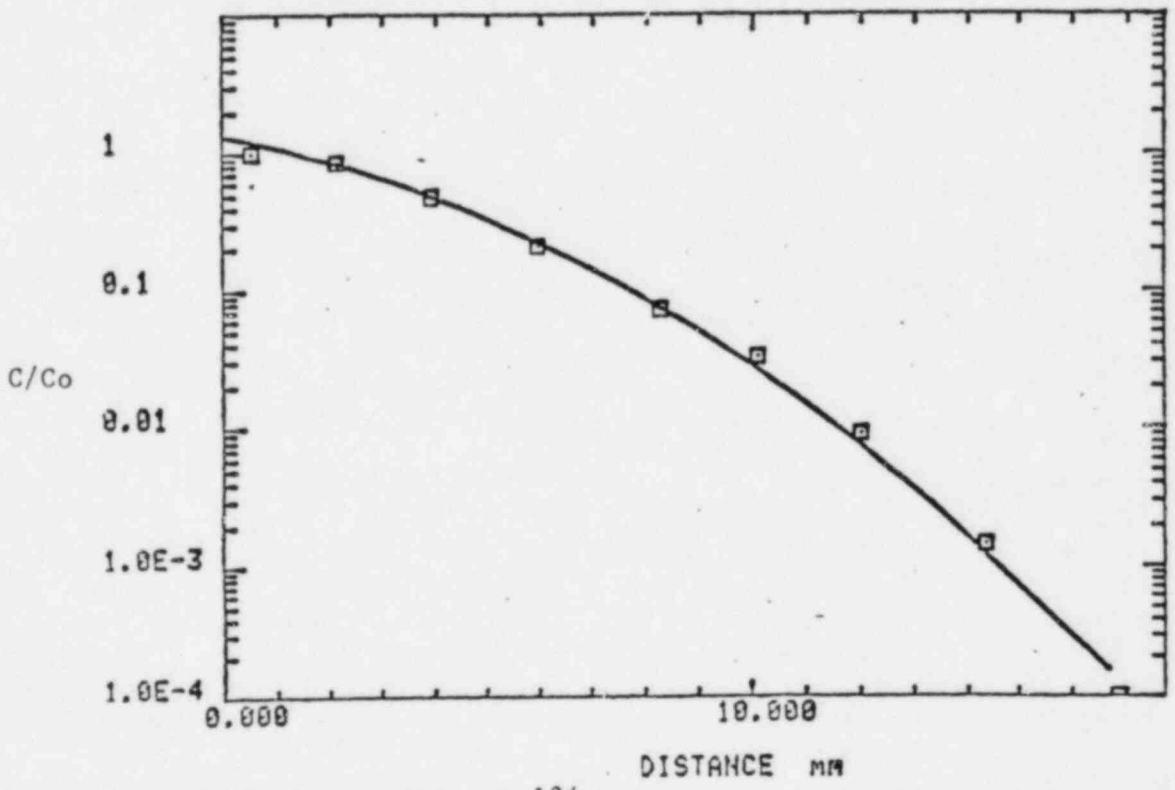

Figure 3.2 Diffusion profile of ^{134}Cs in a repacked sediment core.

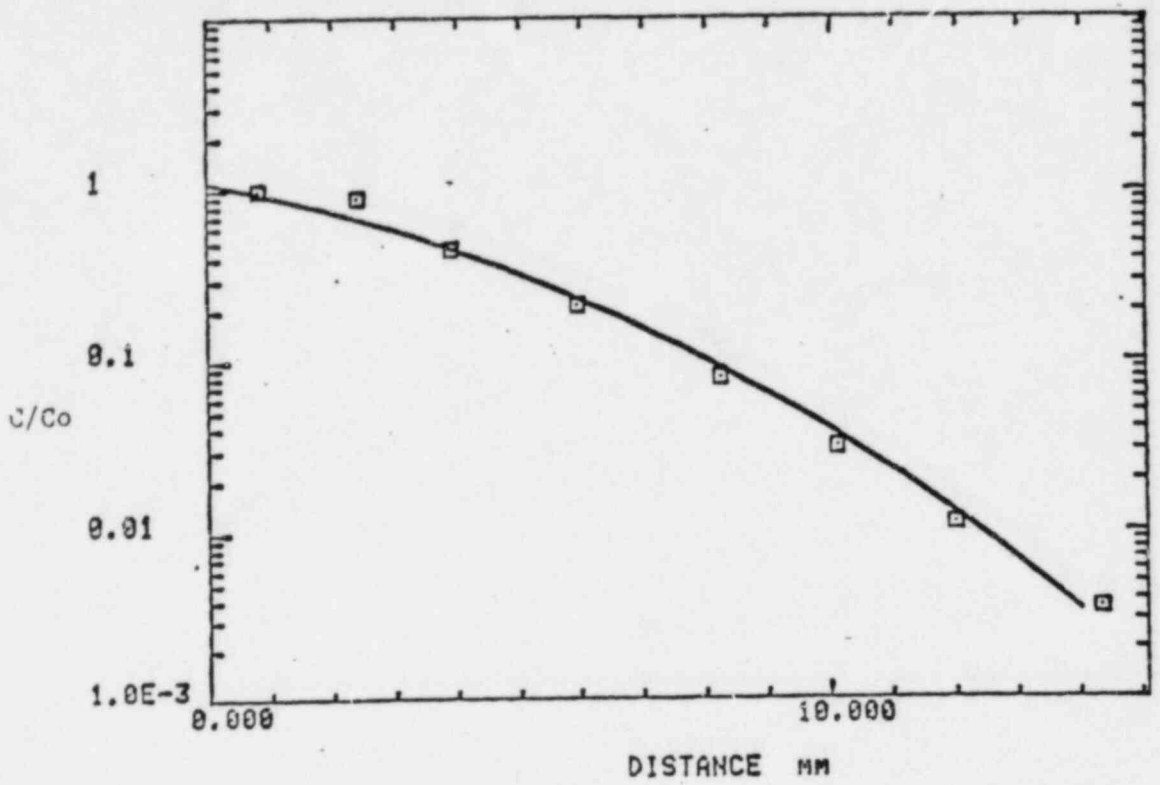

Figure 3.3 Diffusion profile of ^{137}Cs in a repacked sediment core.

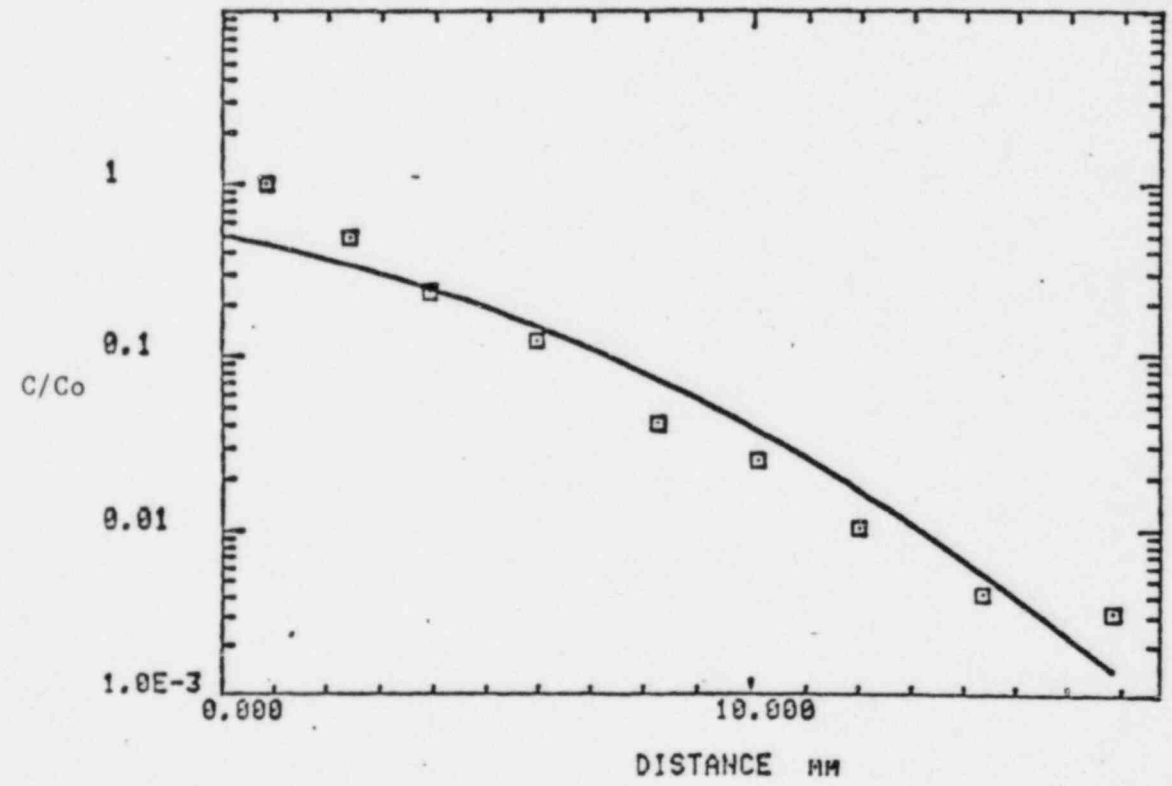

Figure 3.4 Diffusion profile of ^{60}Co in a repacked sediment core.

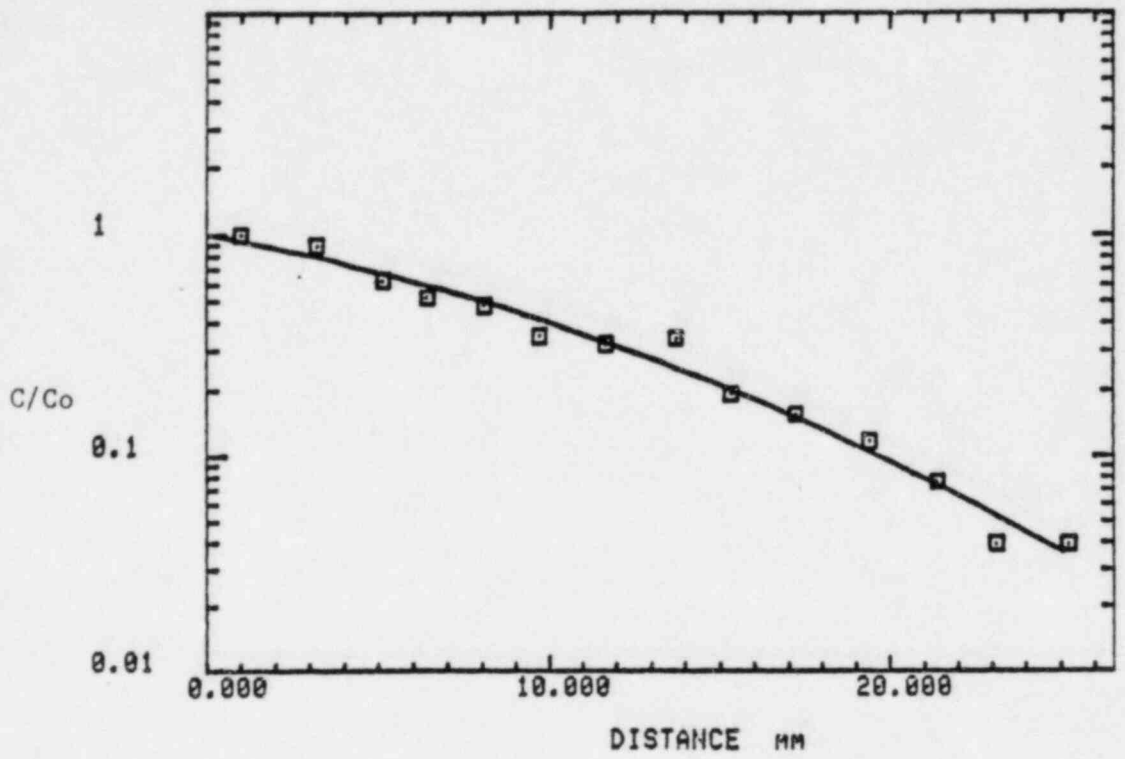

Figure 3.5 Diffusion profile of ^{3}H in a repacked sediment core.

Table 3.1

Radionuclide Diffusion Coefficients[a]

Radionuclide	Diffusion Coefficient cm^2/s
^{85}Sr	1.2×10^{-7}
^{134}Cs	6.3×10^{-8}
^{137}Cs	7.6×10^{-8}
^{60}Co	1.1×10^{-7}
^{3}H	1.2×10^{-5}

[a]Includes the effects of sorption and tortuosity.

As expected, the diffusion coefficients of both ^{134}Cs and ^{137}Cs show good agreement. Relative to the cesium isotopes, both ^{60}Co and ^{85}Sr have a higher diffusion rate because of their lower reactivity with soil particles. A diffusion rate of 1.2×10^{-5} cm^2/s for tritium essentially represents a non-sorptive, diffusion behavior including the effects of tortuosity. Relative to tritium, cesium and cobalt and strontium migrate at rates two to three orders of magnitude lower, showing the extent of retardation by sorption.

3.2 Retardation Factors

3.2.1 Introduction

Radionuclide retardation experiments are in progress using sediment from the Hawthorne formation at Barnwell and a simulated Barnwell trench 6D1 water. The general theory, experimented procedure and composition of the sediment and trench water was given in an earlier report (Pietrzak and Dayal, 1982a; 1982b). The experimental procedure was based on the recommendation of Relyea (1981).

Experiments are in progress to measure the rate of transport of ^{85}Sr, ^{137}Cs and ^{60}Co present in a simulated Barnwell trench 6D1 water through a small (1.5 x 6.0 in.) column of repacked soil from the Hawthorne formation at Barnwell. The cumulative fraction released curves will then be used to calculate radionuclide retardation factors. The column retardation factors (R_f) will be compared with the calculated values based on previously determined batch sorption coefficients (Pietrzak and Dayal, 1982c) for each radionuclide according to the following equation:

$$R_f = 1 + K_d \frac{\rho}{\varepsilon} \qquad (2)$$

where

ρ = bulk density of the repacked soil, and
ε = porosity of the repacked soil.

4. LEACHING OF WASTE FORMS IN SIMULATED TRENCH ENVIRONMENTS

4.1 Introduction

Experiments have been initiated to leach solidified waste under realistic burial conditions. Materials simulating the Barnwell disposal site were selected for this study. The general description of the experiments has been reported earlier (Pietrzak and Dayal, 1982a and 1982b).

4.2 Results

The cumulative fractional releases of ^{85}Sr and ^{137}Cs in the HDPE columns are given in Table 4.1. In the column containing Barnwell soil, no activity has been observed in the effluent after three months of leaching. We are planning to discontinue the soil column experiment, and by sectioning the sediment column beneath the waste form, we should be able to locate the position of the radionuclide front.

Additional experiments planned involve leaching at flow rates higher than that considered in the above experiment and leaching under complete saturation conditions for short periods, followed by drying periods. Specific details of these experiments will be given in the next quarterly report.

Table 4.1

Volume-to-Surface Area Normalized Cumulative Fractional Releases (V/S · CFR) of ^{85}Sr and ^{137}Cs From a 2x4-Size Resin Waste Composite Embedded in a HDPE Bead Column[a]

Time (Days)	V/S · CFR (cm) ^{85}Sr (10^{-3}) No. 1	^{85}Sr (10^{-3}) No. 2	^{137}Cs (10^{-2}) No. 1	^{137}Cs (10^{-2}) No. 2
21.69	0.139	—[b]	0.263	—[b]
22.73	0.241	—	0.456	—
23.70	0.313	—	0.605	—
24.70	0.387	—	0.761	—
25.72	0.450	—	0.896	—
26.71	0.501	—	1.02	—
27.67	0.541	—	1.12	—
28.68	0.583	—	1.23	—
29.67	0.623	—	1.32	—
30.68	0.667	—	1.41	—
31.71	0.712	—	1.55	—
32.69	0.763	—	1.68	—
33.68	0.799	—	1.78	—
34.67	1.05	0.173	2.16	0.271
35.69	1.17	0.295	2.41	0.504
36.90	1.28	0.383	2.64	0.506
37.73	1.37	0.431	2.82	0.619
38.70	1.52	0.482	3.04	0.743
39.64	1.63	0.562	3.24	0.890
40.69	1.71	0.629	3.42	1.04
41.68	1.76	0.679	3.56	1.14
42.68	1.81	0.734	3.71	1.25
43.75	1.86	0.809	3.87	1.38
44.71	1.93	0.860	4.03	1.47
45.72	1.97	0.907	4.19	1.58
46.70	2.03	0.946	4.32	1.66
47.69	2.07	0.988	4.45	1.76
49.00	2.13	1.06	4.68	1.88
50.00	2.19	1.10	4.85	1.95
51.69	2.26	1.15	5.01	2.04
51.70	2.34	1.20	5.17	2.13
52.69	2.39	1.24	5.29	2.20
53.68	2.44	1.28	5.41	2.27
54.69	2.49	1.32	5.54	2.34
55.69	2.53	1.36	5.66	2.41
56.68	2.57	1.40	5.77	2.48
58.69	2.61	1.44	5.87	2.64
59.70	2.63	1.46	5.95	2.77
60.69	2.66	1.49	6.02	2.83
61.69	2.68	1.51	6.10	2.90
62.69	2.71	1.55	6.18	3.05
65.69	2.78	1.66	6.40	3.30
68.86	2.84	1.80	6.65	3.58
72.69	2.93	1.97	6.94	3.87

[a]Data presented for duplicate columns No. 1 and No. 2.

[b]No ^{137}Cs and ^{85}Sr activities were observed in column 2 prior to the sampling at 34.7 days.

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