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EVALUATION OF ISOTOPE MIGRATION - LAND BURIAL Water Chemistry at Commercially Operated Low-Level Radioactive Waste Disposal Sites

QUARTERLY PROGRESS REPORT
JANUARY - MARCH 1977

P. COLOMBO, A.J. WEISS, AND A.J. FRANCIS

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PROCESS TECHNOLOGY DIVISION
DEPARTMENT OF NUCLEAR ENERGY
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UPTON, NEW YORK 11973

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SUMMARY

This is the fourth quarterly progress report of water chemistry at commercially operated low-level radioactive waste disposal sites. Water samples were collected from ten trenches at the Maxey Flats (Morehead), Kentucky, disposal site in September 1976. Precautions were taken to collect and process the samples under anoxic conditions in accordance with procedures developed for this purpose at Brookhaven National Laboratory. The methods used for the preparation of samples and the analytical techniques employed for inorganic and radiochemical analyses are described. Analytical results of the dissolved radiochemical constituents indicate that all trench water samples contain tritium, strontium-90, and plutonium isotopes. Cobalt-60 and cesium-137 were also detected in most trench water samples. The concentrations of the dissolved inorganic and radiochemical constituents are tabulated for each trench.

I. BACKGROUND

The disposal of low-level radioactive wastes at commercially operated disposal sites began in 1962 at Beatty, Nevada. Since that time, the industry has expanded to include three private companies operating six sites. The sites are located in Maxey Flats (Morehead), Kentucky; Beatty, Nevada; Sheffield, Illinois; Barnwell, South Carolina; West Valley, New York; and Richland, Washington. The three companies operating these sites are the Nuclear Engineering Company (Illinois, Kentucky, Nevada, and Washington), Chem Nuclear Systems (South Carolina), and Nuclear Fuel Services (New York). Although these facilities are managed by private industry, they are located on Federal or State owned land. In general, the disposal sites are regulated by the state in which they are located, according to provisions of the agreements between the individual states and the United States Nuclear Regulatory Commission (NRC).⁽¹⁾ The one exception is the site located in Sheffield, Illinois, which is regulated by both the State of Illinois and the NRC.

The disposal sites are located in remote areas of sparse population selected for their hydrogeological environment and were considered suitable at the time of selection for the purpose of shallow land burial of low-level radioactive wastes. In addition, these locations are in proximity to centers of nuclear industry to minimize long distance transportation of wastes.

Projections have been made of the available capacity of the commercial sites with the prediction that the current burial capacity of all six existing sites may be exceeded between 1988⁽²⁾ and 1998⁽³⁾ when existing sites may have to be closed or enlarged or new sites will have to be established. At the present time, the West Valley, New York, site has discontinued disposal operations and the Maxey Flats, Kentucky, site has reduced its disposal volume.

Recently, questions have been raised regarding the adequacy of these disposal sites.^(4,5) With the increasing growth of the nuclear energy industry, important considerations must be given to the selection of any new sites for disposal of radioactive wastes.

The United States Geological Survey (USGS) funded by the United States Congress and with concurrence of the individual states is conducting hydrogeological and geochemical investigations at all commercially operated low-level radioactive waste disposal sites except Richland, Washington.

The United States Nuclear Regulatory Commission (NRC) has the responsibility for establishing criteria for selection and operation of low-level radioactive waste disposal sites.

In support of USGS investigations, NRC funds Brookhaven National Laboratory (BNL) to perform analyses for USGS of natural waters and of waste leachate that have accumulated in waste trenches at the disposal sites.

II. SCOPE

The low-level radioactive waste disposal sites in use today throughout the United States are located in different hydrogeological environments. Until the various physical and geochemical processes governing transport are understood, the continued monitoring of these sites to detect movement, if any, of radionuclides from the disposal trenches is needed.

One of the goals of the USGS program is to collect hydrogeological and geochemical data at each site and to correlate these data with theoretical and laboratory results to develop a transport model for each disposal site. The ultimate goal is to establish criteria for selection, operation, and monitoring of low-level radioactive waste disposal sites.

In support of the USGS program, samples of materials from the disposal sites will be taken at selected intervals and the analysis of the chemical and radioactive species will be performed at Brookhaven National Laboratory. Methods for collecting and analyzing samples from each of the burial sites will be developed. The study will include the measurement of inorganic, organic, and radioactive constituents in water samples.

Soil column tests will be performed in the laboratory to simulate the hydrogeological conditions at each of the disposal sites and to measure the values of a number of factors involved in the migration rates of the various wastes.

III. INTRODUCTION

The first trench water samples for this study were obtained from the Maxey Flats, Kentucky, low-level radioactive waste disposal site in April 1976. The purpose was to evaluate tentative procedures for the collection, handling, and analysis of trench water, and to determine gross levels of the constituents in the trench water. The samples were subsequently filtered and analyzed for dissolved inorganic, organic, and radiochemical constituents.⁽⁶⁾ It was observed that upon standing some of the water samples became cloudy with the formation of a brown ferric hydroxide precipitate. This precipitate, which was present prior to and during the filtration, is probably due to a change in oxidation state of dissolved iron in the trench water. A reducing environment probably exists in the trenches which favors the ferrous state of iron. After trench water is removed from the ground and exposed to air, the dissolved iron is probably oxidized to the ferric state. As such, it is much less soluble than iron in the ferrous state and produces the observed precipitate.

Since ferric hydroxide is known to act as a scavenger of radionuclides, it was postulated that some dissolved radionuclides could have been lost from the aqueous part of the trench water samples by adsorption onto the precipitate. Therefore, anoxic procedures have been developed that prevent the precipitation from occurring during the collection and filtration of the trench water samples. The details of these procedures are described in a previous report.⁽⁷⁾

IV. EXPERIMENTAL

A. Sample Collection

On September 16, 1976, a second set of trenches were sampled at the Maxey Flats disposal site to measure the dissolved components in the trench water. Previous samples⁽⁶⁾ were collected aerobically which resulted in ferric hydroxide precipitation. Therefore, precautions were taken to collect the samples under anoxic conditions in accordance with procedures developed for this purpose.⁽⁷⁾

Water samples were collected from trenches 2, 7, 18, 19S, 26, 27, 32, 33L, and 37 as shown in Figure 1. Only trenches 7, 18, 19S, and 33L, of the trenches previously sampled, contained sufficient water. Therefore, samples were collected from trenches 2, 26, 27, 32, and 37 to obtain additional data. Water samples were collected from two riser pipes in trench 33L (i.e. 33L4 and 33L9). The "L" series trenches are a special group of side by side slit trenches having one or more riser pipes for water removal which are identified by the riser number following the trench number. Samples from trenches 18 and 26 were obtained from riser pipes designated with the letter "A".

Approximately eight liters of water were collected from each of the trenches indicated. The samples were maintained in an anoxic environment in glass collection bottles⁽⁷⁾ at ~4°C prior to analysis. In addition to the above samples, 100 ml water samples were taken from each trench for field measurements of color, pH, temperature, and specific conductance. These parameters are subject to change and were measured as soon as possible after collecting the sample.

NOTE: SCALE APPLIES ONLY TO FENCELINE. POSITION AND DIMENSIONS OF TRENCHES ARE APPROXIMATE.

Figure 1. Sketch of Maxey Flats Radioactive Waste Disposal Site (updated from sketch by EMCON Associates). (8)

B. Sample Processing

The water samples which were collected and stored at $\sim 4^{\circ}\text{C}$ in an anoxic environment were divided into several fractions as described below. A schematic flow chart of these procedures is shown in Figure 2.

- a. A fraction for the determination of alkalinity was obtained by applying gentle nitrogen pressure to the inlet valve on the collection bottle and discharging some water sample through the outlet valve on the bottle.
- b. An "organic" fraction was filtered through a 0.45 micron silver membrane under anoxic conditions.⁽⁷⁾ This was achieved by purging the air from the filtration system with nitrogen, discharging trench water from the sample bottle by applying gentle nitrogen pressure, as above, and filtering under nitrogen pressure. The silver membrane acts as a bactericide to prevent bacterial action from altering the organic nature of the sample. The filtrate was stored in glass containers at $\sim 4^{\circ}\text{C}$ until the various organic analyses were conducted.
- c. A portion of the water sample was filtered through a 0.45 micron cellulose membrane under anoxic conditions, as described above, for the analysis of tritium, anions, and dissolved residue. This portion was stored in a polyethylene container at $\sim 4^{\circ}\text{C}$.
- d. A fourth fraction for the analysis of metals and radio-nuclides was also filtered through a 0.45 micron cellulose membrane into a polyethylene bottle. This fraction was immediately preserved by acidifying with 15 ml of nitric acid per liter of filtrate to prevent losses to the container walls. An all plastic filtration system was used to minimize the possibility of

introducing metallic impurities into the filtrate. This acidified sample was stored in a polyethylene container at room temperature.

Figure 2. Schematic treatment of trench water sample for analysis of various constituents.

V. SAMPLE ANALYSIS

The processed samples were analyzed for dissolved inorganic, organic, and radiochemical constituents. The results of the inorganic and radiochemical analyses are presented in this report.

A. Inorganic Analysis

Field measurements of pH, temperature, specific conductance, and color were performed within 15 minutes after collection. These results are given in Table 1.

Dissolved metals, viz. calcium, iron, lithium, magnesium, manganese, potassium, and sodium were analyzed on the filtered, acidified samples by atomic absorption spectroscopy techniques. The concentration of dissolved metals found in the trench water samples is shown in Table 2.

Non-metals such as chloride, nitrate, nitrite, silica, and sulfate were determined on the filtered, non-acidified samples with a Technicon Auto Analyzer II.* A precipitate formed in the analyzer manifold during the analysis of some trench water samples which interfered with the analysis. Dilution of the samples 1:10 or greater eliminated this problem. Results of the analyses of previous samples from Maxey Flats⁽⁶⁾ by direct reading and standard addition methods indicated that matrix effects were not present in diluted trench water samples.

Total alkalinity was determined immediately after removal from the sample collection bottle by potentiometric titration of the unfiltered water sample using standard hydrochloric acid. Carbonate and bicarbonate were calculated from the titration curve where the carbonate end point is pH 8.3 and the bicarbonate end point is pH 4.5.

* Technicon Instrument Corporation, Tarrytown, New York

TABLE 1

Field Measurements of Water Samples Taken from
Maxey Flats September 1976

<u>Sample</u>	<u>Color</u>	<u>pH</u>	<u>Temperature*</u> <u>(°C)</u>	<u>Specific Conductance</u> <u>(micromhos/cm at 25°C)</u>
Trench 2	gray	6.7	20.0	3.40×10^3
Trench 7	no color	6.9	22.5	2.53×10^3
Trench 18A	light brown	7.0	21.5	3.45×10^3
Trench 19S	light green	6.6	21.0	2.34×10^3
Trench 26A	black	6.8	21.0	2.91×10^3
Trench 27	yellow-green	6.0	20.0	12.0×10^3
Trench 32	green-black	7.3	20.0	5.75×10^3
Trench 33L4	yellow	12.3	20.0	7.60×10^3
Trench 33L9	light yellow	5.6	22.0	6.15×10^3
Trench 37	light green	5.1	20.0	6.90×10^3

* Water temperatures were measured after removal from the ground and could have been affected by air temperature. The values should not be taken as the actual water temperature in the ground.

TABLE 2

Concentration of Dissolved Metals in Water Samples
 Taken from Maxey Flats September 1976
 (mg/liter)

<u>Sample</u>	<u>Calcium</u>	<u>Iron</u>	<u>Lithium</u>	<u>Magnesium</u>	<u>Manganese</u>	<u>Potassium</u>	<u>Sodium</u>
Trench 2	29	40	0.99	79	0.75	66	700
Trench 7	130	61	0.56	73	0.50	140	240
Trench 18	14	33	18	160	0.03	50	540
Trench 19S	58	150	0.23	130	0.76	25	100
Trench 26	31	65	0.34	130	0.74	39	240
Trench 27	600	1200	1.9	430	70	120	670
Trench 32	75	16	0.15	230	1.2	210	700
Trench 33L4	1400	<0.005	0.23	0.03	<0.005	160	160
Trench 33L9	2000	34	0.08	190	20	13	110
Trench 37	250	1100	0.30	730	42	20	680

Hardness is calculated from the calcium and magnesium concentrations. The excess of carbonate hardness (calcium+magnesium hardness) over total alkalinity is reported as non-carbonate hardness.

Dissolved residue was determined from a filtered, non-acidified aliquot of trench water. The sample was evaporated to dryness and then heated for two hours at 180°C.

The concentration of dissolved non-metals found in these trench water samples is shown in Table 3. Nitrogen as $\text{NO}_3^{-1} + \text{NO}_2^{-1}$ is derived from only NO_3^{-1} since NO_2^{-1} was not detected in any of the samples. A preliminary analysis was made for nitrogen as ammonia in three randomly selected samples, viz. 7, 19, and 33L4, since most of the trenches showed little or no nitrate+nitrite. Each of these trenches contains more than 10 mg/l of nitrogen as ammonia.

B. Radiochemical Analysis

All of the radiochemical measurements, except for tritium, were made on a filtered, acidified aliquot of trench water. Tritium was measured on a non-acidified, filtered aliquot because of its potential volatility from an acidic solution. The radiochemical procedures used for analyzing trench water samples have been described.⁽⁶⁾ The following is a summary of these procedures:

- a. Gross alpha and gross beta activities were measured by gas flow proportional counting. An aliquot of the filtered, acidified sample was evaporated in a two inch diameter planchet and counted in a two channel alpha/beta counter.
- b. Tritium was distilled under vacuum, condensed onto a cold finger, and subsequently counted in a liquid scintillation counter. Only volatile forms of

TABLE 3

Concentration of Dissolved Non-Metals in Water Samples

Taken from Maxey Flats September 1976
(mg/liter)

Sample	Total Alkalinity (CaCO ₃)	HCO ₃ ⁻¹	CO ₃ ⁻²	Cl ⁻¹	Hardness Ca+Mg (CaCO ₃)	Hardness Non-Carbonate (CaCO ₃)	N (NO ₃ ⁻¹ +NO ₂ ⁻¹)	Residue (180°C)	SiO ₂	SO ₄ ⁻²
Trench 2	1560	1910	0	310	400	0	<0.05	2160	14	11
Trench 7	1090	1330	0	220	630	0	<0.05	1410	12	<10
Trench 18	2050	2500	0	310	670	0	<0.05	2240	13	18
Trench 19S	980	1200	0	150	680	0	<0.05	1670	9.0	<10
Trench 26	1320	1600	0	290	590	0	<0.05	1580	9.0	<10
Trench 27	432	528	0	4200	3300	2800	1.56	7620	14	69
Trench 32	2720	3320	0	370	1200	0	<0.05	3810	14	11
Trench 33L4	1810	0	974	320	3600	1800	21	4870	1.7	<10
Trench 33L9	358	436	0	90	5700	5300	900	9560	9.0	390
Trench 37	125	152	0	180	3600	3500	13	11200	35	8000

tritium were measured by this procedure. Tritium in the trench water that became part of a non-volatile organic compound by exchange is not detected by the distillation method.

- c. Strontium-90 was determined by separating and counting the equilibrium yttrium-90 in the sample with a low-level anti-coincidence counter. The yttrium-90 was first purified by solvent extraction with diethylhexyl-phosphoric acid (HDEHP), followed by hydroxide and oxalate precipitations. Yttrium carrier was added to correct for chemical yield.
- d. Gamma emitters and americium-241 were measured with a lithium drifted-germanium detector, Ge(Li), system.
- e. Plutonium-238, 239, 240 isotopes were separated from other alpha emitting radionuclides by anion exchange chromatography. Plutonium-242 tracer was added to the sample to correct for chemical yield. After purification, the plutonium was electroplated on a stainless steel disc. The plutonium isotopes were measured by counting the disc in an alpha spectrometer system incorporating a surface barrier silicon detector.

The concentration of dissolved radionuclides found in the trench water samples is shown in Table 4. Plutonium isotopes were found in all trench water samples. The concentrations range from $5.0-1.7 \times 10^5$ picocuries/liter for ^{238}Pu and from $<1.0-2.1 \times 10^4$ picocuries/liter for $^{239,240}\text{Pu}$. Quantitative values for $^{238,239,240}\text{Pu}$ in trench 33L9 are not available at this time. There are indications that ^{242}Pu is present in this sample which interferes with the plutonium analysis since ^{242}Pu was added as a tracer for chemical yield determination. This measurement is being repeated using ^{236}Pu tracer in place of ^{242}Pu tracer.

TABLE 4

Concentration of Dissolved Radionuclides in Water Samples
Taken from Maxey Flats September 1976
(Picocuries/liter)

Sample	Gross Alpha	Gross Beta	Tritium (a)	^{134}Cs	^{137}Cs	^{60}Co	^{90}Sr (a)	^{241}Am	^{238}Pu	$^{239,240}\text{Pu}$
Trench 2	$1.15 \times 10^4 \pm 4.3\%$	$2.81 \times 10^4 \pm 3.6\%$	2.50×10^7	$< 1 \times 10^2$ (b)	$< 1 \times 10^2$	$1.35 \times 10^4 \pm 1.9\%$	6.76×10^3	$4.31 \times 10^3 \pm 3.9\%$	$3.76 \times 10^3 \pm 2.7\%$	$4.09 \times 10^2 \pm 8.6\%$
Trench 7	$2.66 \times 10^3 \pm 5.3\%$	$5.25 \times 10^6 \pm 0.5\%$	4.43×10^8	$< 1 \times 10^2$	$4.63 \times 10^3 \pm 2.9\%$	$2.45 \times 10^3 \pm 5.1\%$	1.99×10^6	$< 2 \times 10^1$	$5 \pm 20\%$	< 1.0
Trench 18	$1.46 \times 10^4 \pm 4.8\%$	$1.48 \times 10^5 \pm 1.4\%$	4.47×10^8	$< 1 \times 10^2$	$4.93 \times 10^3 \pm 3.0\%$	$2.18 \times 10^4 \pm 1.5\%$	4.70×10^4	$< 2 \times 10^1$	$5.92 \times 10^2 \pm 5.9\%$	$5.1 \times 10^1 \pm 20\%$
Trench 19S (c)	$2.27 \times 10^5 \pm 0.9\%$	$7.54 \times 10^5 \pm 0.7\%$	6.90×10^7	$< 1 \times 10^2$	$3.24 \times 10^3 \pm 9.8\%$	$1.32 \times 10^3 \pm 23\%$	2.56×10^5	$7.67 \times 10^2 \pm 7.0\%$	$1.72 \times 10^5 \pm 7.6\%$	$2.1 \times 10^4 \pm 24\%$
Trench 26	$4.12 \times 10^4 \pm 2.7\%$	$1.28 \times 10^5 \pm 1.6\%$	1.97×10^8	$3.07 \times 10^2 \pm 18\%$	$7.51 \times 10^3 \pm 2.0\%$	$1.26 \times 10^3 \pm 6.8\%$	3.46×10^4	$1.01 \times 10^3 \pm 5.7\%$	$3.09 \times 10^4 \pm 6.8\%$	$2.72 \times 10^3 \pm 2.2\%$
Trench 27	$4.83 \times 10^4 \pm 2.3\%$	$7.15 \times 10^5 \pm 0.6\%$	3.06×10^8	$< 1 \times 10^2$	$2.28 \times 10^4 \pm 2.4\%$	$2.02 \times 10^4 \pm 3.2\%$	2.02×10^5	$1.48 \times 10^4 \pm 3.8\%$	$1.28 \times 10^4 \pm 3.1\%$	$1.74 \times 10^3 \pm 13\%$
Trench 32	$8.58 \times 10^4 \pm 1.4\%$	$1.15 \times 10^6 \pm 0.6\%$	2.08×10^8	$4.24 \times 10^2 \pm 24\%$	$6.00 \times 10^3 \pm 4.6\%$	$6.03 \times 10^3 \pm 5.6\%$	3.84×10^5	$< 4 \times 10^1$	$3.58 \times 10^3 \pm 4.2\%$	$1.07 \times 10^2 \pm 25\%$
Trench 33L4	$7.60 \times 10^3 \pm 5.2\%$	$7.02 \times 10^4 \pm 1.9\%$	6.20×10^7	$< 1 \times 10^2$	$< 1 \times 10^2$	$< 3 \times 10^2$	2.42×10^4	$< 2 \times 10^1$	$9.88 \times 10^3 \pm 4.4\%$	$8.44 \times 10^2 \pm 40\%$
Trench 33L9	$2.15 \times 10^4 \pm 3.2\%$	$4.22 \times 10^4 \pm 2.6\%$	6.20×10^7	$< 1 \times 10^2$	$4.81 \times 10^3 \pm 5.3\%$	$3.23 \times 10^3 \pm 9.0\%$	1.13×10^4	$< 4 \times 10^1$	---	---
Trench 37 (d)	$3.17 \times 10^4 \pm 2.8\%$	$1.07 \times 10^5 \pm 1.9\%$	1.10×10^7	$1.65 \times 10^3 \pm 4.3\%$	$9.82 \times 10^3 \pm 2.1\%$	$4.99 \times 10^4 \pm 1.0\%$	1.93×10^3	$2.79 \times 10^4 \pm 1.0\%$	$1.79 \times 10^4 \pm 1\%$	$3.11 \times 10^2 \pm 2.6\%$

(a) The counting error for tritium and ^{90}Sr is less than 1%.

(b) < signifies not detected and the number following indicates the detection limit.

(c) ^{22}Na , $2.32 \times 10^1 \pm 11\%$ was found only in Trench 19S.

(d) ^{54}Mn , $3.66 \times 10^3 \pm 6.5\%$ and ^{65}Zn , $2.74 \times 10^3 \pm 16\%$ were found only in Trench 37.

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