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THE SAVANNAH RIVER PLANT BURIAL GROUND

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ABSTRACT

The low-level radioactive waste burial ground at the Savannah River Plant is a typical shallow-land-burial disposal site in a humid region. Studies of waste migration at this site provide generic data for designing other disposal facilities. A program of field, laboratory, and modeling studies for the SRP burial ground has been conducted for several years. Recent results of lysimeter tests, soil-water chemistry studies, and transport modeling are reported. The lysimeter experiments include ongoing tests with 40 lysimeters containing a variety of defense wastes, and recently concluded lysimeter tests with tritium and plutonium waste forms. The tritium lysimeter operated 12 years. In chemistry studies, measurements of soil-water distribution coefficients (K_d) were concluded. Current emphasis is on identification of trace organic compounds in groundwater from the burial site. Development of the dose-to-man model was completed, and the computer code is available for routine use.

INTRODUCTION

Low-level waste migration studies are conducted at the Savannah River Plant (SRP) to obtain a fundamental understanding of the soil/waste/water system of the SRP burial ground.¹ The ongoing program also provides generic information on migration of radionuclides from an operating shallow land burial site in a humid region. Three subtasks for the DOE Low-Level Waste Management Program² are: lysimeter tests, soil-water chemistry, and transport modeling. Much of the earlier work on these activities was reported previously.³⁻⁷ This paper summarizes the most recent results.

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LYSIMETER TESTS

In the lysimeter subtask, radionuclide migration is measured from known sources emplaced in lysimeters. The data provide a link between laboratory and field conditions and thus aid in predicting migration from a humid shallow land burial site. Leaching and migration from typical unencapsulated defense wastes have been studied by analysis of percolate water, vegetation, and soil cores from the lysimeters.

Several types of long-term lysimeter experiments are located at the SRP burial ground. Numerous lysimeters contain typical SRP solid wastes.⁸ In addition, several lysimeters were installed to investigate specific radionuclides. In this paper, results are summarized for 40 defense waste lysimeters now operated 5 to 7 years, for a tritium waste lysimeter operated 12 years, and for a plutonium waste lysimeter operated 2 years.

Defense Waste Lysimeters

The defense waste lysimeter program at SRL is designed to investigate (in a controlled manner) the leaching and migration characteristics of eight typical SRP solid wastes under actual burial conditions. The data generated by this program are useful for predicting the future behavior of the burial ground and provide many of the input parameters for the transport model. A schematic drawing of a lysimeter is shown in Figure 1. The lysimeters are designed as models of burial trenches in the SRP radioactive waste burial ground.

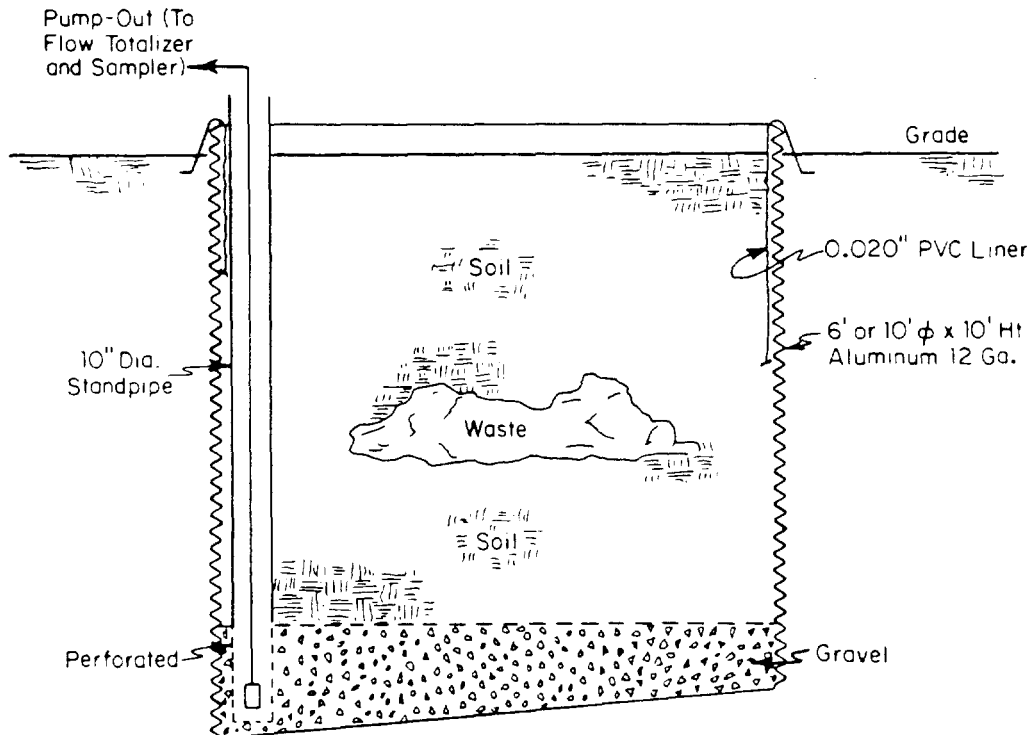


Figure 1. Defense waste lysimeter cross section.

Four six-foot and one ten-foot diameter lysimeters each contain one of the eight types of waste, for a total of forty lysimeters. Two six-foot-diameter lysimeters which contain no waste are maintained as controls. Pine trees were planted on the ten-foot lysimeters to measure the vegetative uptake from the waste. Percolate water samples are collected from each of the lysimeter sumps every six to seven weeks to determine which radionuclides are migrating through the soil. Nearly all other aspects of the operation of the lysimeters, such as maintaining grass as a ground cover, fertilization rates, rainfall, and physical environment are identical to those for the rest of the burial ground. Principal differences are much less depth to the water table and no runoff permitted for the lysimeters. Placement of the lysimeters in a portion of the burial ground ensures that the climatic conditions at the lysimeter site are the same as those at the trenches.

In the seven years since the first defense lysimeters were placed in operation, strontium, alpha-emitting radionuclides, and seven gamma-emitting radionuclides have been detected in the lysimeter effluents. In addition, strontium and cesium have been detected in at least one of the pine trees.

Radionuclide concentrations in the lysimeter effluents during the past year are similar to those reported in previous years,^{3,4} except for Cs-137 which has increased by an order of magnitude in one lysimeter. In general, each radionuclide observed has been found in only a few of the lysimeters, and source terms are known or can be estimated for each. Analyses for Sr-90 and the alpha emitters Pu-238, Pu-239, and Am-241 are performed for those lysimeters with known source terms. All lysimeters are analyzed for gamma emitters, with Mn-54, Co-60, Ru-106, Sb-125, Cs-137, Th-234, and U-235 observed to date. Source terms are not available for Sb-125. All of these gamma emitters appear to be from the waste except Th-234 (a daughter of U-238) and U-235. These two radionuclides are found in the effluents several months after the lysimeters are fertilized and are believed to originate from phosphate salts in the fertilizer.

Some other observations follow: The lysimeters containing laboratory-type wastes release the greatest fraction and variety of radionuclides to the effluent water. There are no apparent differences between the saturated and unsaturated lysimeters in terms of radionuclide releases. Ru-106 and Sb-125 are only observed in anionic forms in the lysimeter effluents. Sr-90 has the highest fractional release rate in these lysimeters and is also observed in the highest concentration.

Fractional release rates of the radionuclides from the lysimeters are summarized in Table 1. These release rates can be directly related to the fraction of each radionuclide which will decay in place (in the lysimeter) if the assumption is made that the release is a first order process. This assumption reduces the problem to the kinetics of competing first-order processes, with the calculations summarized graphically in Figure 2. The fraction of the material that decays in place is plotted against the half-life of the radionuclide for a number of different release rates.

TABLE 1. RADIONUCLIDE RELEASE RATES
FROM DEFENSE WASTE LYSIMETERS

Radionuclide	Fractional Release Rate (fraction/yr)
Mn-54	10^{-9}
Co-60 ^a	10^{-8} to 10^{-7}
Sr-90	$<10^{-4}$
Ru-106	10^{-6} to 10^{-5}
Cs-137	10^{-7}
Pu-238,239	10^{-10} to 10^{-8}
Am-241	10^{-8} to 10^{-6}

a. Except shielded cell waste, 10^{-5} /yr.

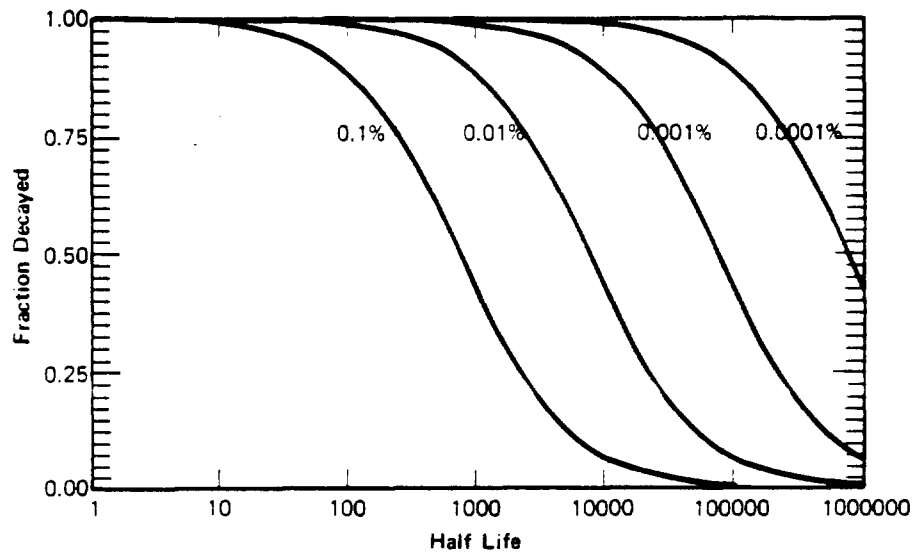


Figure 2. Fraction of source material lost by decay in the lysimeter as a function of half-life. Curves are shown for typical release rates.

The maximum release rate was 10^{-4} per year for Sr-90. With a 30-year half-life, less than 0.5% of the Sr-90 originally placed in the lysimeter will ever be released if there is no change in rates or mechanism. Similarly for cobalt, which had the next highest release rate, less than 0.01% of the original activity will be released. As these results indicate, the lysimeters containing all eight waste types retain the radionuclides in the wastes very well, with minimal releases of radioactivity to the effluent water. As the lysimeters are models of the SRP burial ground trenches, the results show that the burial ground should continue to perform quite well. Releases from the burial ground to the groundwater will be significantly lower than those observed from the lysimeters because of the longer soil column through which the water must pass and the longer time for decay.

Several of the ten-foot-diameter lysimeters have pine trees of sufficient size that the roots probably have extended four feet deep and into the waste. Samples from two of these trees were collected by cutting branches of the tree between July 1983 and February 1984. Total mass of the tree was estimated using established relationship between trunk diameter and tree mass.⁹ Measurement of the mass was not possible because the trees are still growing in a continuation of the experiment. Needles and woody portions of the trees were both analyzed for cesium and strontium. The results are shown in Table 2. The wastes involved were shielded cell waste and canyon pipe jumpers.

The tree growing over the canyon pipe jumpers is not as large as some others, and the roots may not be in the waste zone. However, the source terms for both Sr-90 and Cs-137 were known for this lysimeter. At the time of collection of the samples, the source terms were 2.6 microcuries of Sr-90 and 1.7 microcuries of Cs-137. Concentrations in the pine needles and wood were below 2 pCi/g (dry weight basis) for both isotopes. The fraction of each radionuclide taken up into the tree is approximately 10^{-4} . However, because the observed concentrations are so low and there is not a suitable control for pine trees grown in the burial ground, it is not certain that the observed activity is from the waste.

TABLE 2. UPTAKE OF RADIONUCLIDES BY PINE TREES GROWING ON DEFENSE WASTE LYSIMETERS

Radionuclide	Sample	Concentration (pCi/g)	
		Canyon Pipe Jumpers	Shielded Cell Waste
Sr-90	Needles	2	400
	Wood	1	370
Cs-137	Needles	0.8	34
	Wood	0.6	37

The pine needles and wood from the tree growing over the shielded cell waste contain 400 pCi/g Sr-90 and 35 pCi/g Cs-137. The source term for Cs-137 in this lysimeter was 38 mCi at the time of sample collection. The Sr-90 source term is not known, but is estimated to be the same as that for Cs-137. Using an estimate of the tree mass, the uptake fractions have been calculated as 10^{-5} for Sr-90 and 10^{-6} for Cs-137. These fractions are at least an order of magnitude greater than the fraction of material released through the lysimeter effluent over the four years this lysimeter had been in operation.

Thus, uptake by pine trees can be a major pathway for migration of Sr-90 and Cs-137 and can result in greater releases than the effluent pathway, if roots are able to contact the waste.

Tritium Lysimeter

Several wasteforms typical of SRP tritium waste were emplaced in a ten-foot-diameter lysimeter in 1973, and the lysimeter operated continuously for 12 years until the experiment was concluded in 1985. The wasteforms are stainless-steel crucibles containing spent lithium-aluminum melts. The tritium lysimeter and a control lysimeter that contains no buried waste are similar in design and construction to the defense waste lysimeters.¹⁰ The volume of percolate water and its tritium concentration were determined periodically.

In this experiment, the tritium source term was not measured directly, but is estimated as 450 Ci from other data. After 12 years, 400 Ci were accounted for by decay or release to percolate water. The remaining 50 Ci, now decayed to 25 Ci, were still being slowly released at the conclusion of the test.

Over the 12-year period the data show numerous seasonal fluctuations caused by periods of high or low infiltration through the lysimeter. Such fluctuations are not related to the underlying release mechanisms but can be suppressed by examining the effluent tritium concentration as a function of cumulative volume of effluent (rather than as a function of time). The data treated in this manner show three distinct regions of behavior and suggest that at least three mechanisms are responsible for release of tritium from the wasteform.

About 17% of the original tritium was released in the first year by a mechanism clearly identified as washoff. The amount rapidly washed off agrees well with results of a separate immersion experiment with a similar wasteform. An additional 33% of the tritium was released at a relatively constant rate over the first three years by a second mechanism. Thereafter, about 25% was released at a slowly declining rate over the next 9 years, by what appears to be yet another mechanism. Decay in the wasteform accounts for 19% of the original tritium, leaving an estimated 6% unreleased after 12 years. Although the nature of the postulated second and third mechanisms cannot be determined from the lysimeter data, any plausible mechanism is likely to involve diffusion of tritium from the wasteform or dissolution of the wasteform.

These results may be applied to the filled portion of the SRP burial ground, which has had no tritium burials since 1972. Tritium migration at this site should be dominated by groundwater hydrology of the area; contribution of tritium from continued leaching of sources should now be negligible.

Plutonium Lysimeter

Soil cores were taken from a small lysimeter⁴ that contained one of the more mobile forms of plutonium, Pu(VI) as $\text{PuO}_2(\text{NO}_3)_2$ on a specially prepared wasteform. Analyses of the cores revealed the distribution of plutonium in the lysimeter soil to be mainly adjacent to and below the wasteform. This lysimeter had been operated for 24 months in the actual SRP burial ground environment. During this period, rainfall percolate water collected from the lysimeter did not contain detectable plutonium. Soil coring thus provided information on radionuclide migration that was not otherwise obtainable.

Approximately 90% of the plutonium found in the soil had migrated less than 7.5 cm (3 inches) below the source, and most of this was within 2.5 cm (1 inch) of the source. About half of the plutonium remained on the wasteform after two years of leaching in the unsaturated soil. Soil distribution coefficients (K_d) for plutonium calculated from the core data range from 9 to 35 mL/g. These values are in good agreement with laboratory K_d values for Pu(VI) in SRP soil as determined by batch tests.¹¹

The source for this experiment consisted of 480 microcuries of Pu-239 in the form of plutonyl nitrate solution dried on a paper filter disc. During the two years of operation, a total of 79 L of percolate water passed through the lysimeter. The detection limit for plutonium analyses of percolate water was 2.5 pCi/L. Thus, an upper bound for the amount of plutonium that could have migrated from the lysimeter prior to soil coring is 200 pCi, for a fractional release of $<4 \times 10^{-7}$.

The lysimeter was 33 cm in diameter and 51 cm deep, with the source emplaced at a depth of 21 cm. Vertical soil cores were collected from seven locations in the lysimeter. Each core was divided into one-inch segments for analysis. A total of 82 selected segments were analyzed for Pu-239,240.

The results show that radial migration was small compared to downward migration. The data also demonstrate that Pu(VI) is mobile under conditions at the SRP burial ground, but the migration rate is small, about 15 cm/yr based on the maximum depth at which plutonium was detected in the lysimeter. This is an upper bound for plutonium migration rates in the SRP burial ground because most of the buried plutonium is expected to be in other, less soluble and less mobile valence states.

SOIL-WATER CHEMISTRY

In the soil-water chemistry subtask, laboratory and field data required to understand the fundamental processes of the soil/waste/water system of the SRP burial ground are developed. In the laboratory, K_d values are determined for radionuclides of interest. In the field, groundwater samples from numerous locations at the SRP burial ground have been collected for measurements of chemical properties that may be correlated with radionuclide transport mechanisms.

In the past year, several additional K_d studies of narrow scope were performed to supplement extensive previous work.^{3,11,12} Because K_d values were found to be very sensitive to pH, a study of factors that affect pH in the SRP burial ground was completed. Studies of chemical properties have focused on methods for identifying organics in groundwater from the SRP burial ground.

Radionuclide Distribution Coefficients

Several new laboratory studies have been performed. First, the effects of organic carbon on radionuclide sorption were examined. Second, interactive effects of multiple radionuclides on individual radionuclide sorption were evaluated. Third, the effect of pH on Cs-137 sorption was investigated. Fourth, iodine sorption onto columns of SRP soil was evaluated and compared to recent batch sorption studies.

Burial ground well waters containing elevated levels of total organic carbon (TOC) decreased Co-60, Sr-85, Sb-125, and Cs-137 sorption onto SRP soil. However, no firm correlation was observed between TOC and the radionuclide distribution coefficient (K_d). Apparently, a knowledge of the specific organics present in these well waters is necessary to establish a relationship between radionuclide sorption and organic content of well waters.

Interactive effects of multiple radionuclides on sorption of individual radionuclides were found to be small. Behavior of K_d with pH for Co-60, Sr-85, Sb-125, or Cs-137 with the other three radionuclides present was similar to the behavior of each alone, especially within the pH range of 3.4 to 7.2 expected in the burial ground. Because the interactions are small, valid K_d measurements may be obtained with single radionuclides or with mixtures.

The effect of pH on the sorption of cesium-137 onto SRP soil was evaluated. Over the pH range of the burial ground, cesium K_d varies from 90 mL/g (at pH 3.4) to a maximum of 2400 mL/g (at pH 6.9). Cesium sorption is less pH-dependent than strontium or cobalt sorption.

Studies of iodine sorption onto columns of SRP soil were conducted to supplement batch studies. Column K_d values for iodide were found to be 0.6 mL/g, which is at the low end of the batch K_d range of 0.5 to 6.6 mL/g. Column flow rates in these tests were several times groundwater velocity in the burial ground, so the column K_d values are probably lower than field K_d values. The field K_d for iodide is expected to be intermediate between the batch and column K_d values.

Prior batch studies at SRL indicate that the equilibrium pH of the groundwater/soil system is a major variable that determines the extent of radionuclide sorption.³ Even changes in sorption which result from increases or decreases in cation concentrations or clay content of the soil are due apparently to the minor changes in pH that result. As such, it is important to understand the major factors that control and regulate the groundwater pH.

Titration curves for several burial groundwaters and soils show that the ability of a water to resist changes in pH varies widely within the burial ground. The ability of a soil to resist pH changes is a function of the clay content. An estimate of the final pH of various groundwater/soil combinations may be obtained from the titration curves. The ratio of groundwater to soil determines whether the groundwater, the soil, or some combination of each is the pH controlling factor. In batch studies, where the ratio of water to soil was 15 mL/g, either the water or the soil may determine the pH; in most instances both are important. In the field, where a representative ratio of water to soil is 0.267 mL/g, the soil determines the pH.

Groundwater Analyses for Organics

A search of burial records was conducted to determine the history of organic waste disposal in the SRP burial ground. The search covered three areas: organics which were buried; organics which are known degradation products of TBP extractant or spent solvent; organics which have been detected in burial ground waters. The types of materials known to have been buried or stored include spent solvent, liquid scintillation solutions, various oils, and various decontamination chemicals. A limited number of groundwater analyses for specific organic compounds were performed between 1971 and 1976. Compounds found in certain wells included various long-chain hydrocarbons, butyric acid, hexanoic acid, and TBP.

A new scoping study to characterize the organics present in groundwaters from the SRP burial ground showed that 25 to 30 well waters contained greater than 1 ppm total organic carbon (TOC). Analyses of groundwaters have now been performed for EDTA, TBP, and oxalate ion. Only four groundwaters out of 26 contained measurable (sub-ppm) amounts of EDTA. Only one of 9 groundwaters analyzed contained measurable amounts of TBP (0.15 ppm). No oxalate ion (<1 ppm) was found in 11 groundwaters analyzed. The work to date has: identified the wells in the burial ground which contain elevated levels of TOC; identified the types of organics that might be in these groundwaters; shown that no simple field correlations exist among TOC, radionuclide activity, phosphorous concentration, and areas known to be previously contaminated with organics.

More complete information on the types and amounts of organics in the groundwater is being obtained through gas chromatography coupled with mass spectrometric analyses (GC/MS). A reproducible extraction procedure has been developed to separate groundwater organics into hydrophilic and hydrophobic fractions prior to GC/MS analysis. Among compounds identified so far are low molecular weight mono- and di-carboxylic acids, possibly degradation products from spent solvent. Benzene, toluene, and naphthalene, which are characteristic of liquid scintillation wastes, also have been detected. Many organics found in the groundwater have not yet been identified.

Work is continuing on characterization of groundwater from the SRP burial ground. The relationship between organic compounds found and radionuclide mobility will be investigated.

TRANSPORT MODELING

In this subtask, the Savannah River DOSTOMAN code¹³⁻¹⁶ has been developed. The DOSTOMAN code (1) solves a set of differential equations that simulate radionuclide transport; (2) performs calculations for a number of pathways, with scenarios such as hydrologic transport or future land occupation; (3) estimates environmental impacts as dose commitments; (4) evaluates site performance, radionuclide disposal limits, improved concepts for land disposal of waste, and decommissioning alternatives. The code uses site-specific input data.

During the past year, biotic transport calculations⁴ were completed, new transuranic (TRU) waste disposal limits³ were studied for implementation at SRP, and a Technical Manual was prepared for the DOSTOMAN code. Currently the code is being used for human health risk assessments for hazardous chemical and mixed waste sites at SRP.

The DOSTOMAN Technical Manual, which gives complete details of the code and its applications, will be distributed to DOE sites. Publication of the DOSTOMAN Technical Manual marks the completion of the transport modeling subtask.

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