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RADIONUCLIDE RELEASE FROM LOW-LEVEL WASTE IN FIELD LYSIMETERS

by

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ABSTRACT

A field program has been in operation for 8 years at the Savannah River Plant (SRP) to determine the leaching/migration behavior of low-level radioactive waste using lysimeters. The lysimeters are soil-filled caissons containing well characterized wastes, with each lysimeter serving as a model of a shallow land burial trench. Sampling and analysis of percolate water and vegetation from the lysimeters provide a determination of the release rates of the radionuclides from the waste/soil system. Vegetative uptake appears to be a major pathway for migration. Fractional release rates from the waste/soil system are less than 0.01% per year. Waste-to-soil leach rates up to 10% per year have been determined by coring several of the lysimeters. The leaching of solidified wasteforms under unsaturated field conditions has agreed well with static, immersion leaching of the same type waste in the laboratory. However, releases from the waste/soil system in the lysimeter may be greater than predicted based on leaching alone, due to complexation of the radionuclides by other components leached from the wastes to form mobile, anionic species.

INTRODUCTION

In 1978 the Savannah River Laboratory (SRL) established a lysimeter program to evaluate the rate of leaching and migration of radionuclides from low-level, solid, radioactive wastes buried under actual field conditions. Lysimeters have been used for many years in agricultural studies to measure the loss of soluble constituents from the soil to the percolate water. By emplacing radioactive wastes with well characterized sources of radionuclides in the lysimeters, leaching and migration of the nuclides may be studied. Provision for collection and analysis of the

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percolate water closes the system and allows a mass balance to be calculated. The SRL lysimeters are located within the Savannah River Plant (SRP) Low-Level Radioactive Waste Burial Ground, which they are meant to model and are operated in the same manner as this burial ground. Used in this manner, the lysimeters are a very site-specific measurement technique.

Most of the SRL lysimeters contain radioactive wastes typical of those routinely disposed of at the SRP. Approximately 40 of the lysimeters contain unconsolidated low-level wastes from eight different sources. These wastes include glove box or shielded cell wastes, process piping, and wastes generated in various maintenance operations. Radioactivity in these wastes is predominantly from fission and activation products. A second set of 10 lysimeters contains wasteforms from two different commercial power reactors. These wasteforms contain evaporator concentrate wastes solidified in Portland cement or vinyl ester-styrene (VES) polymer. An additional set of 10 smaller (mini-) lysimeters contains known amounts of actinides in specific oxidation states to allow determination of the migration rates of these species in the SRP shallow-land burial environment. Control lysimeters containing no waste are operated for each lysimeter design.

These lysimeters are part of a long-term program to study the migration of radionuclides in order to determine the consequences of past burial practices and to improve future practices. The lysimeters have been in operation for as long as 8 years. For many of the lysimeters it has taken nearly this long for any detectable activity to appear in the percolate water. This paper summarizes the available information from the lysimeters and demonstrates their usefulness as a technique for measuring migration and leach rates of radionuclides from low-level radioactive wastes.

LYSIMETER DESIGNS

The lysimeters containing unconsolidated wastes are either 6 or 10 ft in diameter and 10 ft deep. The details of design and construction of these lysimeters and a complete description of the wastes and source terms are given elsewhere.[1] A line drawing of the lysimeter is given in Figure 1. In addition to the piping, which allows collection and volume measurement of the percolate water, the lysimeters are equipped to maintain the waste in either a saturated or unsaturated zone. Operation with the waste in a saturated zone is to model perched water in the burial trenches. Several of the 10-ft-diameter lysimeters have pine trees growing on them to determine if vegetative uptake is an important migration pathway for the system.

The solidified wasteforms are contained in 6-ft-diameter lysimeters, which are of essentially the same design as those lysimeters that contain the unconsolidated wastes. Design details, source terms, and solidification agents have been described previously.[2] These lysimeters are all operated in an unsaturated mode, as this is the expected manner of disposal. Two additional sampling features are included in these lysimeters. They

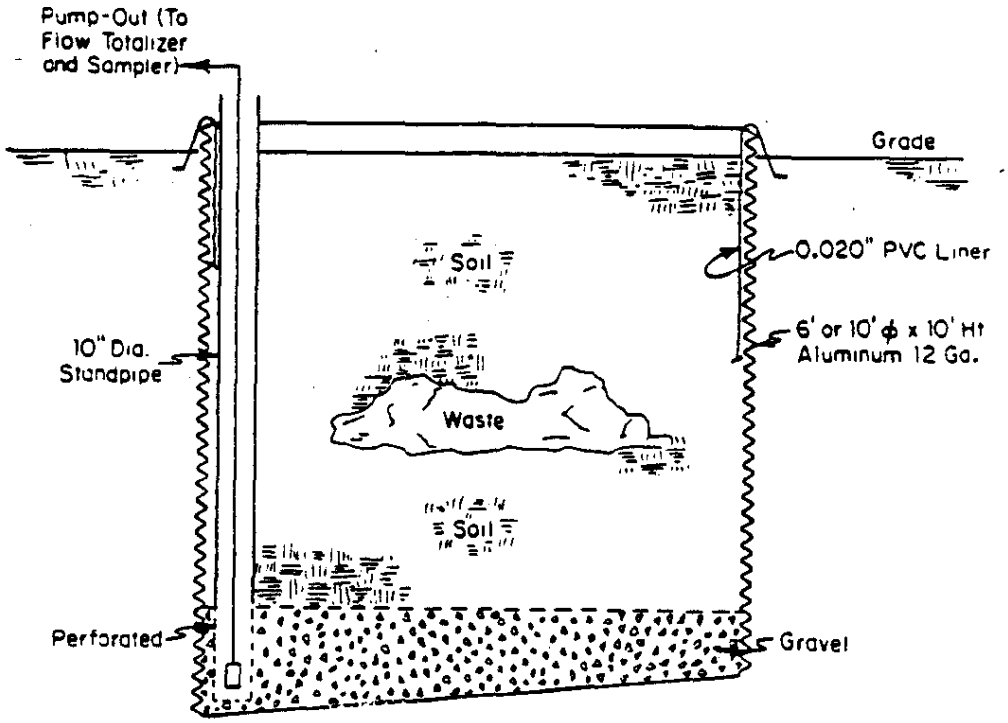

Figure 1. Large lysimeter cross section.

are access ports to allow horizontal soil cores to be collected at two depths beneath the wasteforms, and porous ceramic cups to allow collection of soil moisture from the unsaturated zone beneath the waste. These features are shown in Figure 2.

The mini-lysimeters are smaller and are operated only under unsaturated conditions. They consist of 13-gal polyethylene carboys with the bottoms removed, as shown in Figure 3. Each contains a known quantity of a single isotope in a known chemical form and oxidation state. None of these lysimeters have released any radioactivity via the percolate water (detection limit of 3 pCi/L). The only mini-lysimeter that is of further concern in this paper is one that was cored after 2 years of operation to obtain leaching and migration data. This lysimeter contained 480 μCi of $PuO_2(NO_3)_2$ on a paper filter disk.

For all three types of lysimeters, the wastes were intentionally made available for leaching. In the lysimeters containing unconsolidated wastes, this consisted of cutting open plastic bags and cartons and spreading the waste before burial. For the solidifed wasteforms, this meant removing the shipping drum and liner from the wasteform prior to burial.

SAMPLING AND ANALYSIS

Samples of the percolate water from the lysimeters are routinely collected and immediately concentrated by absorption onto mixed bed ion exchange resin. Typically, a 20-L sample is concentrated. At the time the sample is collected, the total amount of effluent water that has percolated through the lysimeter is recorded. The resin column is analyzed by pulse-

height-analysis (PHA) gamma spectroscopy to determine the concentrations of gamma-emitting nuclides in the effluent water. Strontium or alpha analyses are performed, if needed, by ashing and leaching the resin followed by standard radiochemical separation. The control lysimeters are sampled and analyzed in an identical manner to those containing waste.

Analyses of soil cores are performed either by drying the soil and obtaining a PHA gamma spectrum for a known weight of the soil, or by leaching followed by radiochemical separation. These analyses have been limited to Co-60, Cs-137, and Pu-239. Standards were prepared in about the same concentration by adding tracers to clean soil samples, drying them, and analyzing the standards by the same method as the core samples.

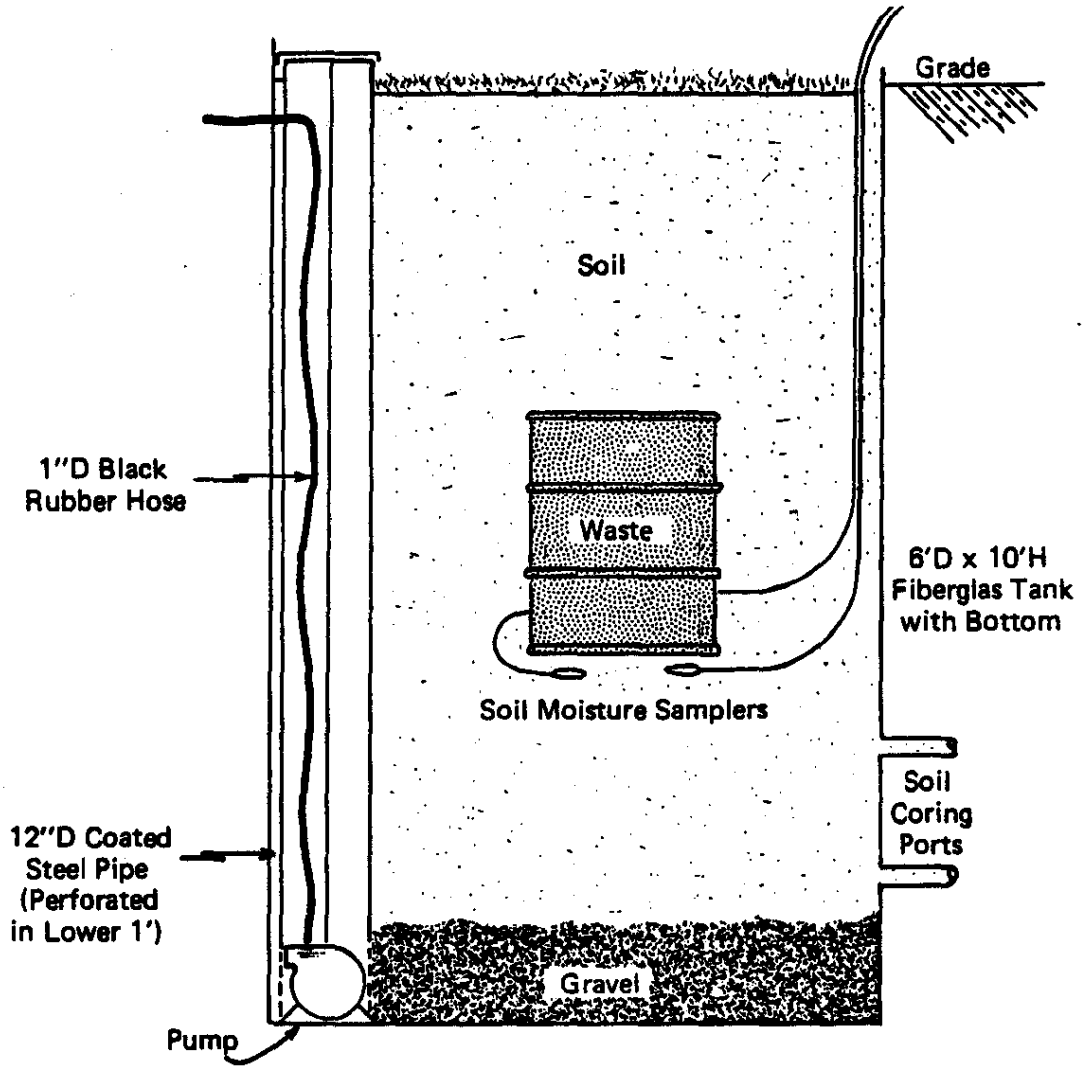

Figure 2. Solidified wasteform lysimeter cross section.

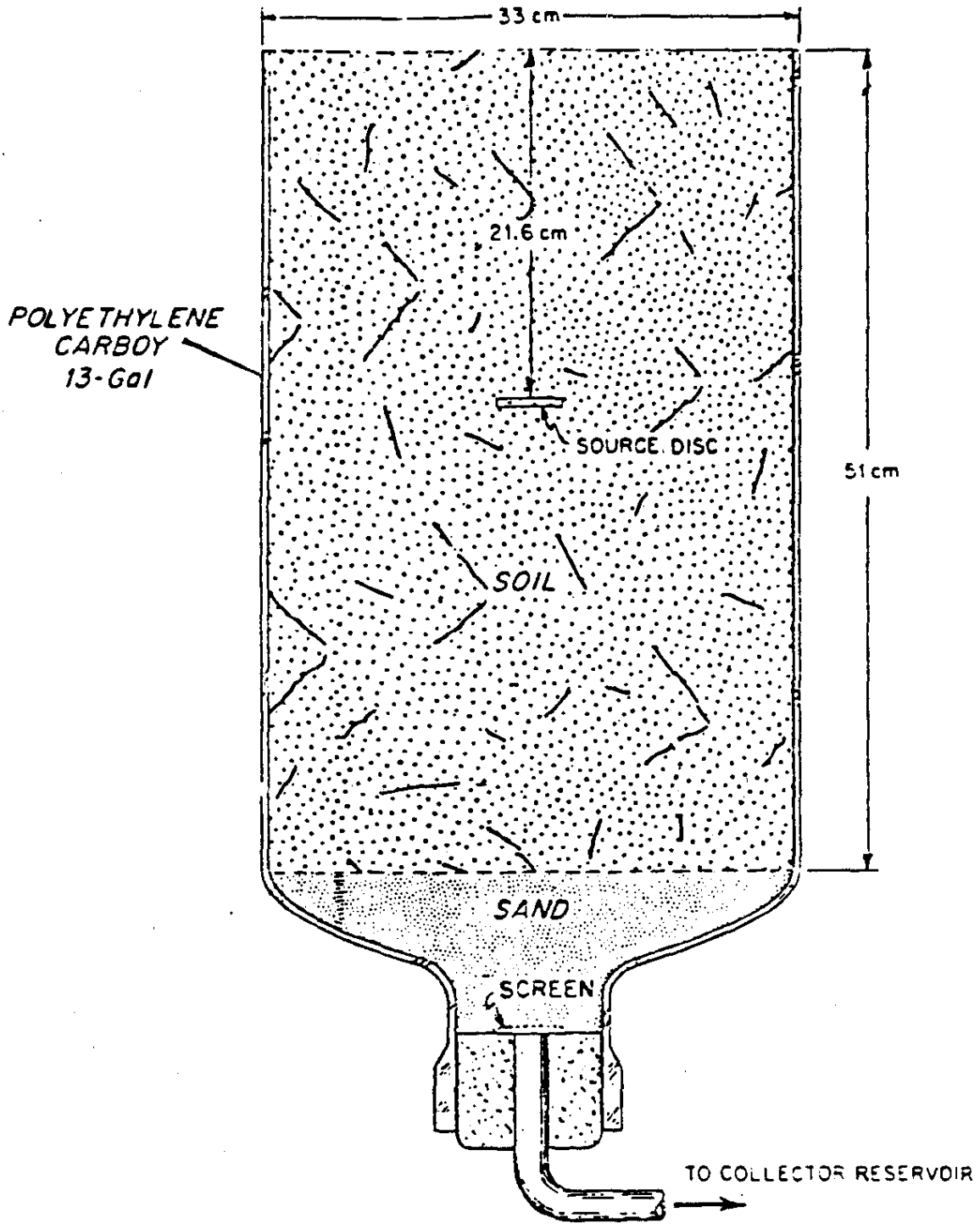

Figure 3. Mini-lysimeter cross section.

In addition to these water and soil samples, the pine trees growing over the unconsolidated wastes are occasionally sampled to estimate the vegetative uptake. For these samples several branches are removed from the tree and split into needle or woody portions. These portions are then digested and isotopes of interest determined by radiochemical separations.

RADIONUCLIDE RELEASE VIA PERCOLATE WATER

Seven isotopes have been detected in the percolate water from the lysimeters: Sr-90, Co-60, Ru-106, Mn-54, Pu-238/239, Am-241, and Cs-137. Concentrations have ranged from a fraction of a pCi/L to 2,000 pCi/L. All of the data have been summarized elsewhere.[2,3]

A more useful representation of the data is to express the results as fractional release rates. These values are obtained by counting the curies as they are released from the lysimeter over a given time period, and dividing by the original source term for the lysimeter. These annual fractional release rates are reported in Table 1 for the seven isotopes released from the lysimeters. It is important to point out that these values do not represent the leaching of radionuclides from the waste, but are a combination of leaching and retardation by the soil. Leach rates must be determined by other means, unless detailed knowledge is available for the retardation of the nuclide through the particular soil under the particular flow conditions.

TABLE 1. FRACTIONAL RELEASE RATES

Isotope	Fractional Release Rate (fraction per year)
Sr-90	10^{-4}
Co-60	
Shielded cell	10^{-5}
Scrap metal	10^{-8} to 10^{-7}
Portland cement	10^{-6} to 10^{-5}
VES polymer	10^{-8} to 10^{-7}
Ru-106	10^{-6} to 10^{-5}
Mn-54	10^{-9}
Pu-238, -239	10^{-10} to 10^{-8}
Am-241	10^{-8} to 10^{-6}
Cs-137	10^{-8}

While interpretation of leach rates would require a decay correction for the source term, the release rate is intentionally calculated without such a correction. This allows for an estimation of the fraction of the material that will decay within the lysimeter if release is assumed to be a first order process. This assumption reduces the problem to one of competing first order reactions (decay and release) with the results graphically summarized in Figure 4. The fraction of the material that decays within the lysimeter is plotted against the half-life of the radionuclide for various release rates.

Sr-90 showed the largest fractional release rate of 10^{-4} per year from the lysimeters. Even at this release rate, less than 0.5% of the original source term will ever be released from the lysimeter. For Co-60, with a release rate of 10^{-5} per year and a 5.3-year half-life, less than 0.01% of the source term will be released. As these results indicate, all of the wasteforms that have been tested are performing excellently. It is also clear that only those radionuclides that are very mobile or have long half-lives will be appreciably released.

RADIONUCLIDE RELEASE BY VEGETATIVE UPTAKE

Besides the release of radionuclides to the environment by migration through the groundwater, the lysimeters allow for measurement of the uptake by pine trees. Pine trees are the predominant ground cover for the SRP site. Samples were collected from two of the trees after approximately 5 years of growth. The total mass of the tree was estimated using established relationships between trunk diameter at breast height and tree mass.[4] The data are shown in Table 2 for both trees. Only Sr-90 and Cs-137 were detected in the trees.

TABLE 2. TREE UPTAKE DATA

	Shielded Cell (pCi/g)	Canyon Pipe Jumpers (pCi/g)
Cs-137		
Needles	34	0.8
Wood	37	0.6
Fractional uptake	10^{-6}	10^{-4}
Sr-90		
Needles	400	2
Wood	370	1
Fractional uptake	10^{-5}	10^{-4}

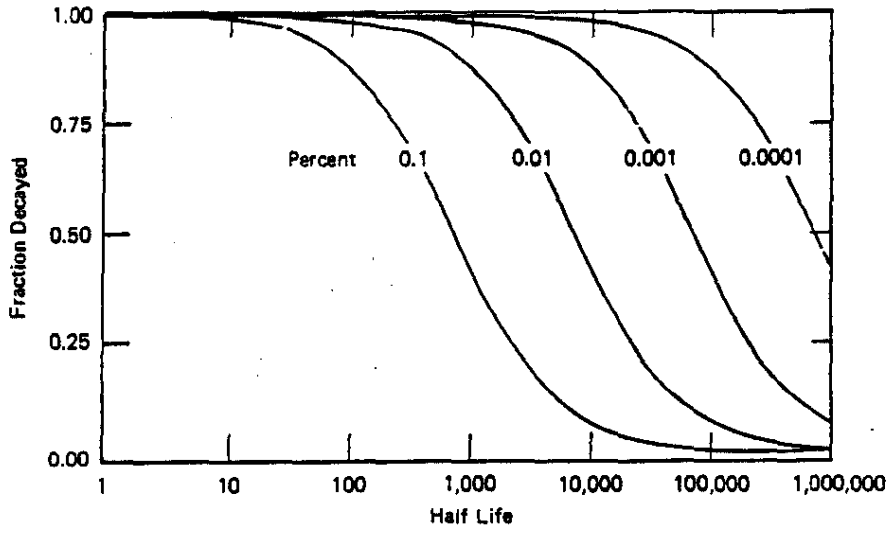

Figure 4. Fraction of material lost by decay within the lysimeter as a function of half life. (Curves are shown for various release rates.)

The tree growing over the shielded cell wastes had taken up a large fraction of activity when compared to that released to the groundwater. The fraction of Cs-137 taken up by the tree is 10^{-6} and that for Sr-90 (estimating the Sr-90 source term to be equal to that for Cs-137) is 10^{-5}. Both of these are at least in order of magnitude greater than the fraction released via the effluent water for the same lysimeter. For the tree growing over the canyon pipe jumpers, the fraction of the source that was taken up by the tree was 10^{-4} for both isotopes. These results indicate that plant uptake can be a major pathway for the migration of radionuclides from waste in shallow land burial, if the roots are allowed to contact the wastes.

WASTE-TO-SOIL LEACH RATES

Because the calculated release rate cannot be usually separated into leaching and retardation components, several coring studies have been carried out to determine the waste-to-soil leach rates. This allows the calculation of the radioactivity that has leached from the source in a given period of time, and gives a profile of the activity on the soil column below the waste.

Four cores were collected from the mini-lysimeter containing plutonyl nitrate after 2 years of operation. One core contained the source disk, with the other three at different distances from the center of the lysimeter. Analysis of the source disk indicated that 50% of the plutonium had been leached from the source in this time, but none of it had reached the lysimeter sump. The majority of it had migrated downward, but a small portion moved up the soil column. This is believed to be due to movement of the soil moisture upwards during dry periods when there is considerable evaporation. This upward migration is particularly evident since the waste is within a foot of the surface of the lysimeter.

The centerline concentration profile is plotted in Figure 5. C/C_o is the measured ratio of the concentration at depths below the waste to that immediately below the source. Also shown are curves predicted using various soil/water distribution coefficient (K_d) values, as calculated by a simple one-dimensional, unsteady-state model. The details of this model are given elsewhere.[1] This calculation, although somewhat simplistic, gives a good estimate of the range for the K_d for Pu(VI) in SRP soils. The values of 10 to 30 mL/g are in good agreement with values measured in the laboratory using the same soil.[5]

The bend in the experimental curve in Figure 5 cannot be calculated by the model. A possible explanation is that conversion of some of the Pu(VI) to a lower oxidation state, and less mobile form, may have taken place. A second explanation is that the evaporative cycling caused the material to move down, then up, and finally down again. The net result would be a reduced migration rate, or an increased observed sorption. This cycling should be less pronounced at greater depths and would result in a gradual lowering of the K_d with increasing depth. The simple model cannot take these factors into account.

Horizontal cores have been collected from lysimeters containing solidified wasteforms at two depths beneath the wasteforms. This method was chosen to maintain functioning lysimeters after coring. The results indicate that both Cs-137 and Co-60 had leached from the wasteforms in significant amounts. The migration was dominated by downward migration rather than radial diffusion. Leach rates for the two isotopes are estimated from these corings at 1% over a 2-year period for Co-60 and between 20 and 50% for the Cs-137 over the same time period. These values compare well with the leach rates as determined on duplicate wasteforms leached by the ANS 16.1 procedure. This procedure was extended to 320 days, at which time 0.1% of the Co-60 and 20% of the Cs-137 had been leached from a full size wasteform.[6]

For both isotopes the leach rates as measured from the coring studies are orders of magnitude greater than the release rates measured via the effluent water. This demonstrates that the soil in the lysimeter is sorbing most of the activity and allowing it to decay within the lysimeter. This also points out the need to look at the entire waste/soil/water system when evaluating or estimating the behavior of a shallow land burial system.

LYSIMETER RELEASES COMPARED TO IMMERSION LEACHING

The agreement between the leach rates as determined by immersion leaching and by coring the solidified wasteform containing lysimeters has already been noted. It would be expected that two wasteforms that showed the same leach rate for a given isotope should have the same release rate, provided they are emplaced in the same soil.

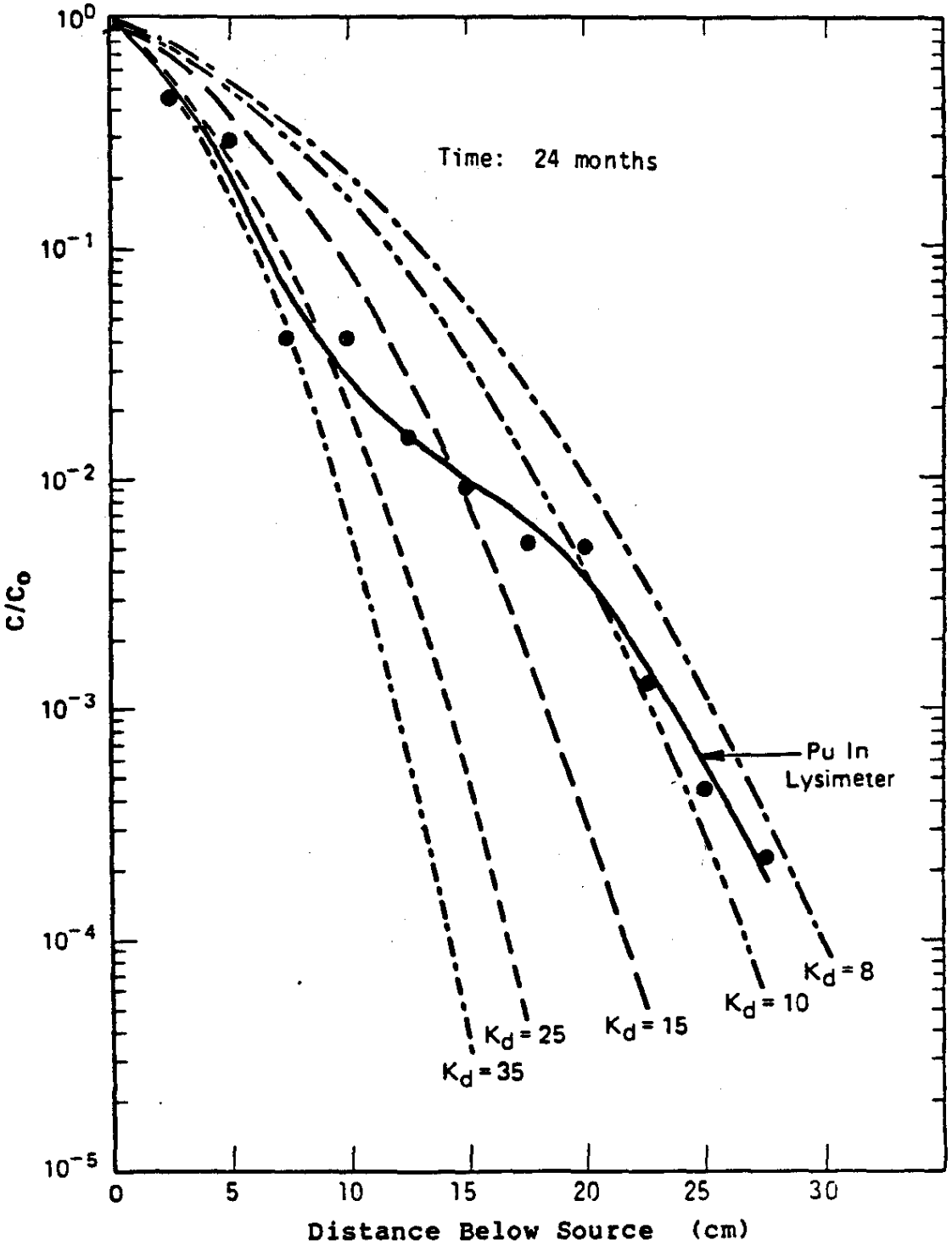

Figure 5. Plutonium migration from $PuO_2(NO_3)_2$ source in mini-lysimeter.

For Sr-90 this has been true. The VES polymer wasteforms leached strontium more rapidly than the concrete wasteforms during the immersion leaching, as was expected with the chemical binding of strontium by the concrete. The same trend was observed in the lysimeters, where Sr-90 is released from the polymer wasteform-containing lysimeters, but not from those containing cement wasteforms.

Leach rates for Co-60 from the immersion testing were within a factor of 2 for the concrete and polymer wasteforms. However, the lysimeter releases have shown a factor of 10 greater release from the lysimeters containing concrete wasteforms, and have shown this trend for over 3 years. Investigation of the apparent discrepancy determined that the Co-60 released is present as a labile, anionic complex.

The literature indicates that organic reagents used in decontamination agents can form anionic complexes with metals, and that these organics are released approximately 10 times faster from VES polymer wasteforms than from Portland cement.[7] This enhancement of the migration by complex formation can explain the difference in release rates while having the same leach rate. It also indicates that Co-60 is migrating as two different species with different rates of transport. Currently, the release rate for cobalt only measured the anionic species. At some time in the future, when cationic cobalt finally breaks through the soil column into the effluent water, different release rates will be measured.

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