SEEPAGE BASIN RADIONUCLIDE TRANSPORT IN SEDIMENTS AND VEGETATION (U)

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SEEPAGE BASINS RADIONUCLIDE TRANSPORT IN SEDIMENTS AND VEGETATION

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ABSTRACT

Radionuclide concentrations were measured in soil and vegetation growing adjacent to and in the Savannah River Laboratory Seepage Basins as part of the plan for closure of the basin system. The results of the measurements provide some information about the mobility of the radionuclides introduced into the basins. ^{90}Sr is the most mobile of the radionuclides in soil. Its high mobility and high relative uptake by vegetation cause ^{90}Sr to be distributed throughout the basin system. ^{137}Cs is not as mobile in the basin soil, limiting its uptake by vegetation growing on the edge of the seepage basins; however, it is readily taken up by the vegetation growing in the basins. Soil mobility and vegetation uptake is relatively low for all of the transuranic radionuclides. For the most part these radionuclides remain near the surface of the basin soils where they were absorbed from the waste-water.

The relative role of soil mobility and vegetation uptake on the distribution of radionuclide at the basins was further evaluated by comparing the vegetation concentration ratio and the half-depth of penetration of the radionuclides in the basin soil. The results suggest that vegetation processes dominate in determining the concentration of ^{60}Co and ^{137}Cs in the vegetation. The influences of soil and vegetation are more balanced for ^{90}Sr . The other radionuclides exhibit both low soil mobility and low vegetation uptake. The lack of soil mobility is seen in the lower concentrations found in vegetation growing on the edge of the basin compared to those growing in the basin.

INTRODUCTION

Seepage basins are designed to contain or pond aqueous waste so that the waste-water will flow into the soil at the bottom of the basin. Soils can be very effective filters for particulate suspended in water and generally are highly absorptive of cations in solution. Most soils have relatively low absorptive capacity for anionic solutes (Bohn et al. 1979). Even though seepage basins have been shown to be very effective in removing the majority of radionuclides, such as ^{137}Cs and the transuranic nuclides, from waste-water, the use of seepage basins for treatment has been discontinued at SRS. This is, in part, because of the inability to remove some radionuclides, such as ^3H . The measurements of radionuclide and metal contamination of vegetation growing adjacent to and in the Savannah River Laboratory (SRL) Seepage Basins was done as part of the plan for closure of this basin system.

Description

The SRL Basins and the sediments in the basins are described in Bransford et al. (1988). The following description of the SRL Basins is largely taken from that document.

The four Seepage Basins are located south of Road A-1 and west of Road D-1 in the northwestern section of the Savannah River Site. The basins are rectangular in shape and were constructed by removing the earth from within the basins to form the surrounding dikes. The construction of Basins 1, 2, and 3 primarily involved excavation of natural soils and the construction of limited perimeter dikes. The construction of Basin 4 required substantial filling at the north end (adjacent to Tims Branch) to achieve both the basin bottom and the dike crest elevation. The present soils are Udorthents, consisting of well drained soils of heterogenous spoils from the excavation. These soil were derived from the the original undisturbed soils of the Fuquay and Valcluse series (Rogers 1990).

The SRL Seepage Basins received waste-water from the Savannah River Technology Center (SRTC, formerly the Savannah River Laboratory). The dimensions and approximate capacity of the basins are:

Basin	Dimensions (LxWxD, m)	Volume (m ³)
1	40 x 19 x 2.0	1,520
2	40 x 40 x 2.0	3,200
3	53 x 38 x 2.7	5,440
4	94 x 46 x 3.4	14,700

Waste-water entered the first basin by a pipeline from the SRTC. The water overflowing Basin 1 entered Basin 2 and consecutively into Basins 3 and 4 when the water flow was greater than the infiltration capacity of the proceeding basins. Waste-water seldom entered Basin 4 because the infiltration capacity of the first three basins equaled or exceeded the input volume. Water entering the sediment at the base of the water column appears to have infiltrated downward, toward the groundwater.

Basins 1 and 2 have no vegetation growing on the basin bottom. The bottom of these basins has generally remained covered with water, even after discharges to the basins stopped. The slopes of the basin dikes are densely covered with young trees. Most of the trees were less than ten years in age at the time of this sampling. The majority of the trees are deciduous angiosperms (hardwoods), with a few scattered conifers (pines).

The bottom of Basin 3 also lacks any vegetation. At the time of this study the basin bottom was intermittently covered with water. The slopes of the dikes surrounding Basin 3 are more sparsely covered with woody vegetation, with a majority of the surface vegetated by grasses and herbaceous vegetation. The woody vegetation is largely hardwood trees with some scattered pines.

Basin 4 is the only basin which has trees growing on the bottom of the basin. Most of the basin is covered by a dense stand of young pines. A few hardwoods are also growing in the basin. The age of the trees was estimated to be less than ten years at the time of this study. The edge of the basin was characterized by a less dense stand of the same species. There is a small stand of older pine trees, estimated age 25 to 35 years, growing on the high dike on north side of Basin 4.

History of Operation

The first two basins were placed into operation in 1954. Basins 3 and 4 were added in 1958 and 1960, respectively. SRTC used the seepage basins to dispose of low-level radioactive liquid wastes. Pipes which tied into the low-level drains in the laboratories transferred the waste into one of four underground tanks. When one of the tanks accumulated approximately 20 m³ of waste, the contents were sampled. If analysis of the waste-water indicated concentrations not exceeding 100 d/m/mL alpha and/or 50 d/m/mL beta-gamma, the waste-water was discharge to the Seepage Basin. If concentrations exceeded these limits the waste-water was sent to another facility.

Releases to the SRL Seepage Basins stopped following 1982. During the 28-year loading history, approximately 130,000 m³ of water were discharged to the basins. Table 1 lists the radioactive releases between 1958 and 1980. An estimate of the total discharge of other contaminants was made on the basis of the concentrations in the low-level waste stream sampled during October of 1982.

Table 1. Radioactive Releases to the SRL Seepage Basins from 1958 to 1980 (Bransford et al. (1988))

<u>Radionuclide</u>	<u>Released Activity (Ci)</u>
³ H	105
^{89,90} Sr	0.4
¹³⁷ Cs	4.7
Natural U	0.022
²³⁸ Pu	0.009
²³⁹ Pu	0.003
²⁴¹ Am	0.001
^{242,244} Cm	0.001
^{103,106} Ru	1.4
⁶⁰ Co	0.1
^{141,144} Ce	2.7
Alpha (unidentified)	4.2
Beta-gama	10.6

METHODS

Soils

Five 6.6-m. cores samples were removed from the bottom of each basin. A total of twenty cores. The cores were bored with Shelby tube samplers. The top 0.66-m. of each Shelby sampler was cut into 7.6-cm. segments. The 7.6-cm soil samples were removed from the tube segments in the field. Below 0.66-m., composite samples over 0.98-m. intervals were used for analysis. A total of 280 sediment samples were collected.

Following collection the samples were shipped to a contract laboratory (Envirodyne) for analysis. A description of the analytical techniques can be found in Bransford et al. (1988).

Vegetation

Branches were collected from the trees selected for sampling. One kilogram of leaves and one kilogram of branch wood was collected for analysis. Each sample consisted of material from a single tree. The samples were collected in double polyethylene bags. Each bag was sealed before moving to the next sampling location.

Vegetation samples were located at the edge of the basins at each of the four corners. These locations were selected because the corners of the basins could be easily located if more sampling was needed later and the trees closest to the edge of the basins were likely to show the highest concentrations. In addition to the 16 locations at the edge of each basin, 12 trees growing on the bottom of Basin 4 were sampled. The four trees at each of three soil sampling locations were selected. Two additional trees were sampled near the top of the dike on the north side of Basin 3. A total of 60 samples were collected, 30 leaf samples and 30 branch wood samples.

The samples were sealed in double polyethylene bags in a DOT type 7A drum. The drum was shipped to TMA Thermo Analytical, Inc., Richmond, CA where the vegetation was analyzed for selected radionuclides. A description of the analytical techniques can be found in Murphy (1992).

RESULTS

Soils

Differences between Basins

Table 2 illustrates the integrated concentration of the seven radionuclides measured over the 6.56 meters of the soil. Because of the high variability of cores taken from the same basins, it is difficult to see any pattern in the deposition of the radionuclides in the basins soils. The only observable trend is the lower concentrations of many of the radionuclides in Basin 4, particularly when compared to Basins 1 and 2. As indicated previously Basin 4 was observed to receive much less water than the other basins.

Table 2. Integrated Radionuclide Content of Soils in SRL Seepage Basins (units, pCi/g-m², one standard deviation of mean in parenthesis)

	<u>Basin 1</u>	<u>Basin 2</u>	<u>Basin 3</u>	<u>Basin 4</u>
⁹⁰ Sr	79.6(33.1)	67.1(27.2)	139 (250)	20.2(10.3)
¹³⁷ Cs	226(69.7)	280 (101)	107 (30.9)	24.2(11.1)
^{243,244} Cm	62.9(45.9)	120(67.5)	12.2 (4.44)	19.1 (7.5)
²⁴¹ Am	4.75(3.55)	8.76(4.57)	1.81(0.434)	2.23(0.63)
²³⁸ Pu	5.55(2.49)	6.32(8.82)	0.835(0.437)	0.567(0.148)
^{239,240} Pu	49.4(34.2)	22.8(11.4)	0.450(0.293)	0.351(0.151)
⁶⁰ Co	0.485(0.291)	8.78(10.8)	1.69(0.955)	0.103(0.091)

Variation with Depth

With a few exceptions, the concentration of all the radionuclides measured in the basin soils show a marked decrease in concentration with distance from the surface. This is to be expected if the radionuclides are strongly absorbed by the soils. In many cases, a straight line relationship is found between concentration and depth when the concentration of a radionuclide collected at a particular depth is plotted against depth in logarithmic coordinates. Of the 28 combinations of 7 radionuclides and 4 basins 18 have negative regression slopes with regression coefficient of greater than 0.90, 22 have regression slopes of greater than 0.80. Only in the case of ²⁴¹Am are all regression coefficients less than 0.60. Examples are the average concentration of ¹³⁷Cs in the cores from Basins 2 and 4 (Figure 1) and the average concentration of ⁹⁰Sr in Basin 2 (Figure 2). The decrease in concentration with depth seen in both cores for ¹³⁷Cs and in the core from Basin 2 for ⁹⁰Sr is the pattern found for all of the other radionuclides in all of the cores (even the ²⁴¹Am cores, though with greater scatter). Only ⁹⁰Sr in Basin 4 shows the pattern of an almost constant concentration over the first meter.

Figure 1. ^{137}Cs concentration versus Depth from the Soil Surface.

Figure 2. ^{90}Sr concentration versus Depth from the Soil Surface.

This relationship can be interpreted in terms of the mobility of a particular radionuclide in the basin soil. Figure 3 shows the half-depth of penetration of each radionuclide. The half-depth was calculated by determining the logarithmic coordinate slope of the radionuclide versus depth relationship and extrapolating this depth to the point that half the radionuclide content of cores was above and half below that depth. The average half-depth of Basins 1 and 2 is plotted separately from the half-depth of Basin 4. The half-depths calculated for basin 3 are very similar to those for Basin 1 and 2.

Clearly all of the radionuclides, with the exception of ^{137}Cs have penetrated more deeply in the soil of Basin 4 than in Basins 1 and 2. This is in spite of the fact that Basins 1 and 2 received a much greater volume of waste-water. This indicates that the radionuclides were less mobile in the flooded soils of Basins 1 and 2 than in the more aerated, vegetated soil of Basin 4. This is particularly true for ^{90}Sr which has penetrated much further in Basin 4 than any other radionuclide. This is consistent with the behavior of ^{90}Sr in other environmental systems (Wicker and Schultz 1982).

Figure 3. Half-Depth of Penetration for Radionuclides in SRL Basin Soils.

Vegetation

Uptake of radionuclides from soil by plants is limited by the mobility of the radionuclide in the soil and the ability of the vegetation roots to extract the radionuclide from the soil particles and mobilize the radionuclide through the roots to the rest of the plant. The ability of the vegetation to remove radionuclides from the soil is shown in the ratio of the concentration in the vegetation divided by the concentration in the soil, called the concentration factor. The concentration factor can vary with the type of plant and with the part of the plant analyzed.

A linear plot of leaf concentration versus stem concentration indicated a straight line relationship for all radionuclides. The ratio of leaf concentration to stem concentration was calculated for each radionuclide from the slope of the straight line. Only in the case of ^{60}Co and ^{137}Cs were the slopes significantly greater than 1.0. In both cases the concentration in the leaves was about three times (3.21 for ^{60}Co , regression coefficient 0.91 and 2.72 for ^{137}Cs , regression coefficient 0.87) that for the stems.

Preliminary inspection of the contaminant concentrations suggested that the concentrations of some contaminants was higher in the hardwood trees than in the pines. Only in and around Basin 4 were there enough trees of each type to test the hypothesis that the mean concentrations were different. Ratios of the mean concentration in hardwoods to the mean concentration in pines were calculated for each contaminant. Then the hypothesis of different mean values was tested by applying a Student t-test. The t-test suggests that the mean is different only for ^{137}Cs ($p < 0.038$), and perhaps for ^{90}Sr ($p < 0.071$). The concentration of ^{137}Cs is lower in the hardwoods than the pines (ratio 0.4 hardwood/pine) while the concentration of ^{90}Sr is higher in the hardwoods than the pines (ratio 2.8 hardwood/pine).

There are two possible sources of variation that could lead to the differences between hardwoods and pines in Basin 4. As shown in Figure 4, the concentration of all radionuclides is higher for trees growing in Basin 4 than in trees growing on the edge of the basin or further away from the basin on the top of the dike. Most of the pines sampled are growing on the bottom of the basin while the most of the hardwoods are on the edge of the basin. This would lead one to expect the pines growing in the more highly contaminated basin soils would have higher concentrations than the hardwoods growing on the edge of the basin. Therefore, the higher ^{137}Cs concentration in the pines may not actually be a species effect but an effect of location. On the other hand, in spite of the effect of location there is higher concentration of ^{90}Sr in the hardwoods than in the pine. This suggests that the higher ^{90}Sr concentration might be a species effect.

Figure 4. Relative Concentration of Radionuclide as a Function of Distance from the Bottom of Basin 4.

Vegetation/Soil Concentration Ratios

Concentration ratios are used to estimate the concentration of radionuclide in one component of the environment given the concentration in another component that is the primary source of the radionuclide to that component. Vegetation-soil concentration ratios are used when the soil is the primary source of the radionuclide of interest. Vegetation-soil uptake ratios vary greatly because of the complex relationship between environmental factors which effect the uptake of materials from soils. One factor of importance under field conditions is the depth of rooting of the vegetation. The ratios presented here were calculated on the basis of an average concentration over the top 0.66-m. of soil. The same calculations for an average over twice that depth will result in ratios that are approximately half as large for contaminants retained in the upper few centimeters of the soil, e.g. ^{137}Cs , but have little difference for contaminants that are more uniformly distributed with depth, e.g. ^{90}Sr in Basin 4. For this reason, concentration ratios developed under similar conditions of soil type, water regime, soil contaminant distribution, and vegetation are used whenever possible.

Table 5 and Figure 5 show the concentration ratios calculated for the radionuclide concentrations measured in trees and soil. The concentration factors are for leaves. In the case of the two radionuclides which showed significant differences between leaf and wood concentrations, ^{60}Co and ^{137}Cs , the concentration ratios for wood are a factor of three less for leaves. The variability in concentration ratios, as illustrated by the one standard deviation (logarithmic transform) error bars in Figure 6, is, as expected, large. However, it is possible for the results to be used to provide an upper bound to the expected concentrations under similar conditions.

Table 5. Vegetation/Soil Concentration Ratios for Trees Growing on the Edge of the Normally Flooded, Seepage Basins 1 & 2, and Trees Growing in the Normally Dry Seepage Basin 4.

	Average of Basins 1&2	Basin 4	Literature	
			Mean	Range
^{90}Sr	0.41	1.69	2.93(2)	0.1-20(2)
^{137}Cs	0.0022	2.37	0.26(2)	0.1-10(3)
$^{234,244}\text{Cm}$	0.0009	0.0211	0.00085(1)	10^{-6} -1.7(2)
^{241}Am	0.0001	0.0001	0.0055(1)	0.0001-0.1(2)
^{238}Pu	6E-05	0.0047	0.00045(1)	0.006-0.1(2)
$^{239,240}\text{Pu}$	2E-05	0.0073	0.00045(1)	0.006-0.1(2)
^{60}Co	0.0087	0.61	0.06(1)	0.01-1(3)

Sources (1) Baes et al., 1984, (2) Coughtrey et al. 1985, and (3) Till and Meyer 1983.

Figure 5. The Geometric Mean Vegetation/Soil Concentration Ratio with a One Standard Deviation Error Bar for Trees growing on the Edge of Basins 1 & 2.

The concentration ratios for all radionuclide but ^{90}Sr are less than one. In agreement with most past studies, all of the transuranic radionuclide are less than 0.001 (Baes et al., 1984, Coughtrey et al. 1985, and Till and Meyer 1983). The relatively high concentration ratio of ^{90}Sr is in agreement with other studies. Uptake of Sr in soil is influenced by its role as an analog of Ca. The sandy, acid soils in the SRS are generally low in Ca, promoting Sr uptake.

The same thing would be expected for Cs which is an analog of K. The SRS soils are generally low in K which should promote Cs uptake (Coughtrey et al. 1985). The reason for the difference in the behavior of Cs compared to Sr may be its relative lack of mobility in the soil. ^{90}Sr is found at relatively high concentrations in the vegetation at the edge of the basins compared to the vegetation growing in the basin while ^{137}Cs is much lower in the trees at the edge of the basin than in trees in the basin (see Figure 4).

Table 5 and Figure 6 show the concentration ratios calculated for the pine trees growing on the bottom of Basin 4. The variation in the results, as expressed in the one standard deviation error bars, is less than that for the trees growing on the edge of Basin 1 and 2. With the exception of ^{137}Cs the ratios are similar to those found for trees growing at the edge of the basins. The ^{137}Cs concentration ratio is more like those reported for similar, low-K, soils (Coughtrey et al. 1985).

Figure 6. The Geometric Mean Vegetation/Soil Concentration Ratio with a One Standard Deviation Error Bar for Trees growing in Basin 4.

DISCUSSION

The results of the measurements of soil and vegetation radionuclide concentrations in and surrounding the SRL Seepage Basins provide some information about the mobility of the radionuclides introduced into the basin dissolved in the waste-water. ^{90}Sr is the most mobile of the radionuclides in soil. Its high mobility and high relative uptake by vegetation cause ^{90}Sr to be distributed throughout the basins, even beyond the edge of the basins.

^{137}Cs is not as mobile in the basin soil but is readily taken up by the vegetation. The combination of low soil mobility and relatively high uptake are the reason why the concentration ratio is high for trees growing in basin 4 but low for trees growing on the edges of all four of the basins.

Soil mobility and concentration ratios are low for all of the transuranic radionuclides. For the most part these radionuclides remain near the surface of the basin soils where they were initially absorbed from the waste-water.

The relative role of soil mobility and vegetation uptake on the concentration of the radionuclide measured in the SRL Basins are illustrated in Figure 7. Figure 7 shows the ratio of the vegetation concentration ratio and the half-depth of penetration of the radionuclides in the basin soil. If the concentration ratio is large compared to the half-depth, the ratio will be large and a strong influence of vegetation uptake is indicated. Conversely, if the half-depth is large compared to the concentration ratio, the influence of the soil is greatest. The results suggest that vegetation processes dominate in determining the concentration in vegetation for ^{60}Co and ^{137}Cs . The influences are more balanced for ^{90}Sr while the balance shifts towards greater soil influence for the other radionuclides. Figure 7 also shows the higher influence of soil mobility in vegetation growing on the edge of the basins than vegetation growing in the basins.

Figure 7. The Comparative Influence of Vegetation versus Soil on Vegetation Uptake as Measured by the Ratio of the Vegetation Uptake Concentration Ratio and the Half-Depth of Penetration into the Soil.

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