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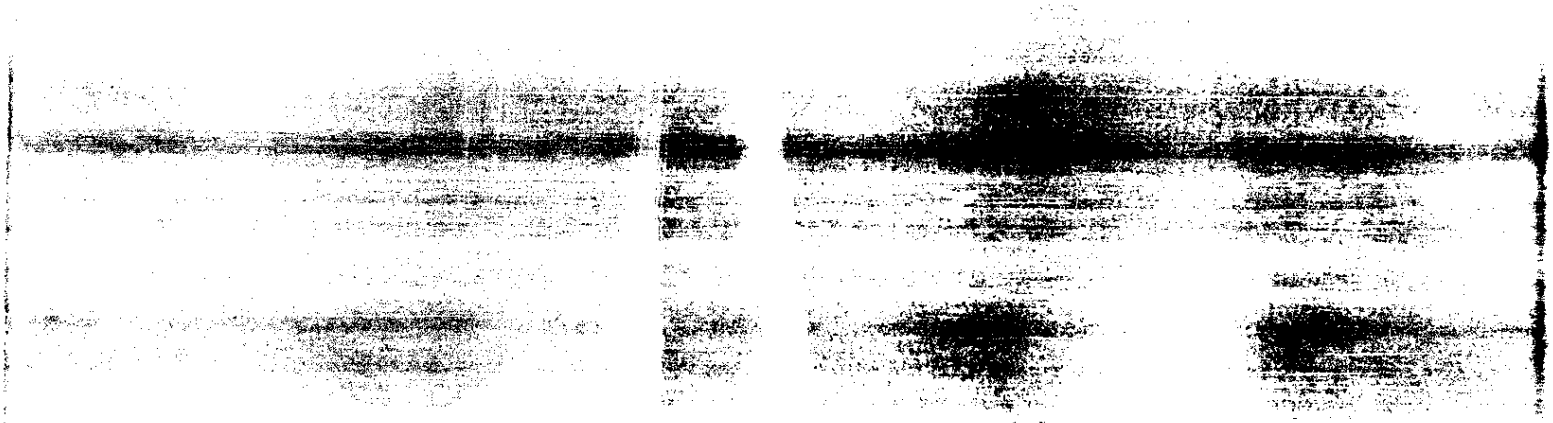

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ABSTRACT

The concentrations of plutonium, americium, and curium in a small stream that drains the chemical separations areas on the Savannah River Plant were determined. Typical concentrations for ^{238}Pu, $^{239,240}Pu$, ^{241}Am, and ^{244}Cm were 39, 9.5, 5.0, and 7.5 fCi/L, respectively. These concentrations are much lower than the maximum permissible concentrations in water of 30 nCi/L (U.S. Code of Federal Regulations, 10 CFR-20). Less than half of all transuranic activity was associated with particles that were one micron or greater in diameter. The distribution coefficients for plutonium, americium, and curium were 2.9×10^5, 6.1×10^5, and 8.3×10^5, respectively. The plutonium distribution coefficient is similar to that measured in Lake Michigan.

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TRANSURANICS IN A STREAM NEAR A NUCLEAR FUEL CHEMICAL SEPARATIONS PLANT

INTRODUCTION

The Savannah River Plant is the principal plutonium and tritium production facility for the U.S. Department of Energy. It began operation in 1953 and now has three heavy-water-moderated production reactors operating, two plants for the chemical separation of plutonium from fuel elements, a fuel fabrication facility, and a heavy water plant. Transuranics produced in the reactors include ^{238}Pu, $^{239,240}Pu$, ^{244}Cm, and ^{252}Cf.

The two chemical separations facilities (Figure 1) have discharged small amounts of radioactivity to Four Mile Creek and to the atmosphere via stacks.[1] These discharges have been monitored extensively,[1] and in most cases have been analyzed for specific isotopes except transuranics. Until 1971, the transuranics released were estimated by using gross alpha measurements at point of discharge and the conservative assumption that all alpha activity was due to $^{239,240}Pu$. Plutonium has been measured since 1971, and it was estimated that ~0.3 Ci of $^{239,240}Pu$ has been discharged to the aquatic environment since plant startup.[2]

The average water flow in Four Mile Creek upstream of any plant discharge is less than 15 L/sec and is increased by natural drainage and F- and H-Area effluents to 560 L/sec at Site 4 (Figure 1).[3] The flow in the creek can vary considerably (between 200 and 1400 L/sec) at Site 4.

Over the years, the Four Mile Creek floodplain and stream bed have accumulated radionuclides sorbed to the sediments. Measurements of transuranics in the water at any time would reflect this inventory as well as current discharges and watershed runoff.

The objective of this study was to investigate the behavior and transport of the transuranics in Four Mile Creek. It was expected that these results should be typical of transuranics discharged from nuclear installations into the natural freshwater stream environment of the southeastern U.S. coastal plain.

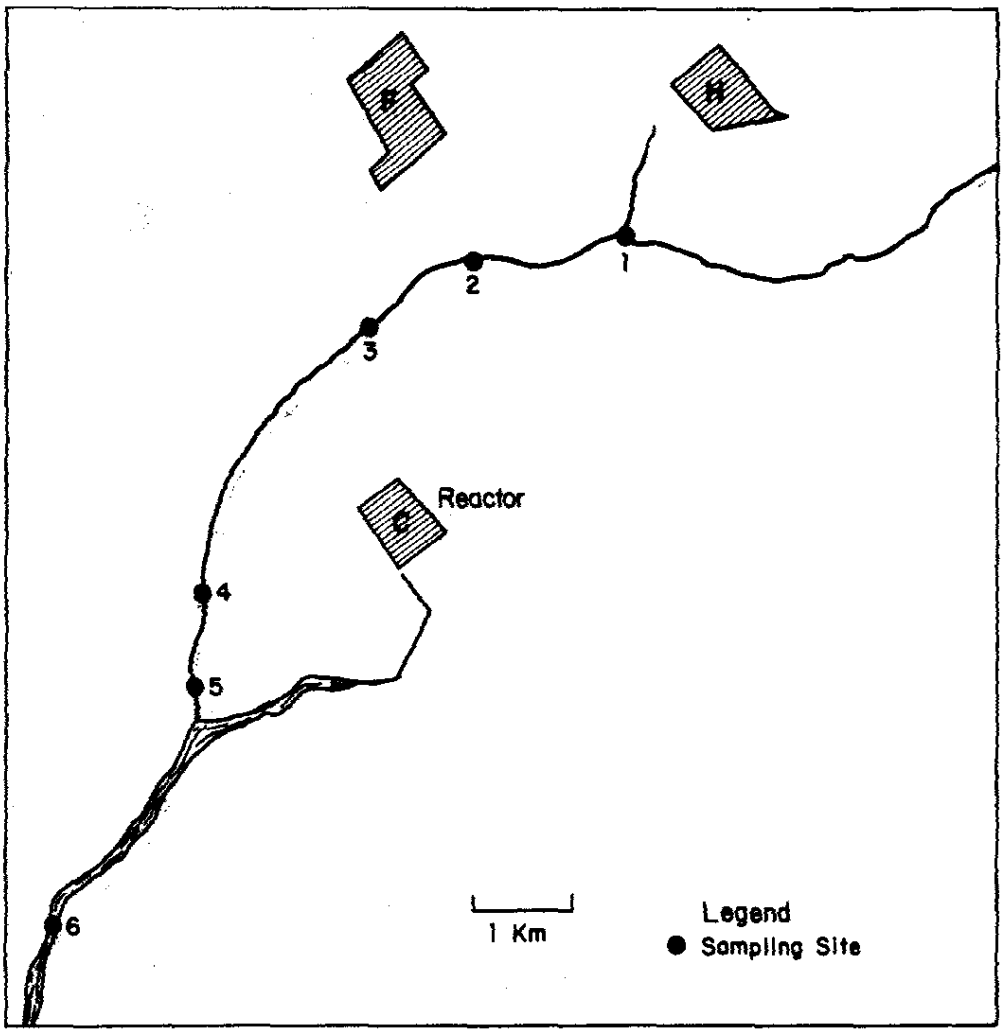

FIGURE 1. Location of Sampling Sites in Four Mile Creek. The areas marked F and H are the chemical separations areas.

SAMPLING AND ANALYSES

Water samples were taken from six locations in Four Mile Creek during January, 1975. Samples were collected each weekday and composited into a single sample for each site at the end of the month. Each composite sample contained a total of 19 L of water. The approximate depth of the creek at the sampling site was 30 cm, and the samples were taken from the water surface to minimize incorporation of sediments. The plutonium was removed from the water by an ion exchange column, and analyzed by a slightly modified procedure (Butler[4] and Sanders[5]). Concentrations of ^{137}Cs were determined by gamma ray spectroscopy without chemical separation.

A second set of water samples (50 L) was taken from Site 4 in 1979 for the analysis of plutonium, americium, and curium in the soluble and particulate phases. The transuranics were concentrated by an MnO_2 procedure,[6,7] and a slightly modified procedure of Bojanowski[8] was used for the radiochemical analysis of the transuranics.

Total transuranic concentrations were obtained by analyzing unfiltered stream water. Particulate fractions were separated from bulk water samples by filtering stream water through a cotton cartridge filter manufactured by the Carborundum Company.* The filter is rated to remove at least 95% of >1μ particles.

RESULTS AND DISCUSSION

The ^{238}Pu concentrations in Four Mile Creek decrease with downstream distance (Figure 2) from 223 fCi/L at Site 1 to 16.8 fCi/L at Site 6. This suggests that the original burden of ^{238}Pu present at Site 1 was diluted as the stream volume increased downstream. Stream flow rates, available during the sampling period at three of the sampling sites, increased substantially with downstream distance. The flow rates at sampling Sites 1, 2, and 4 were 212, 348, and 572 L/sec, respectively. The ^{238}Pu in transport at Site 2 is actually about 23% greater than is calculated from the concentration and flow rate data. This indicates an additional ^{238}Pu source between Sites 1 and 2, which is probably drainage from F Area or from the region between F and H separations areas. Stream bed remobilization of ^{238}Pu could also produce this result.

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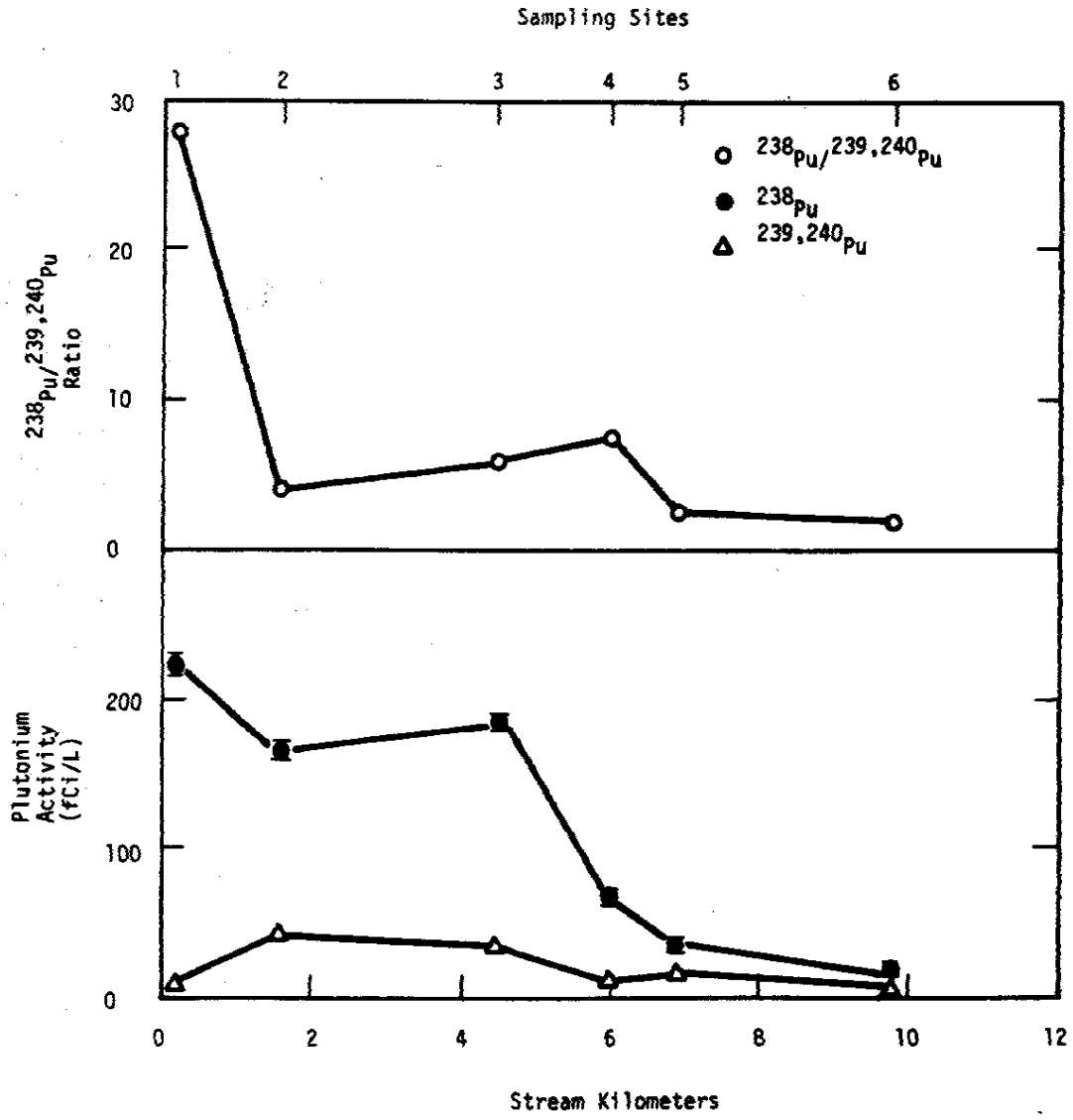

FIGURE 2. The Concentrations of ^{238}Pu and $^{239,240}Pu$ at the Six Sampling Sites in Four Mile Creek. Stream kilometers are measured going downstream, starting at Site 1.

The ^{238}Pu in transport decreased from Site 2 (58 ± 2 pCi/sec) to Site 4 (39 ± 3 pCi/sec), which may be due to sedimentation of plutonium-bearing particulates or stream bed sorption. In general, however, it appears that dilution is the major reason for the decrease in ^{238}Pu concentration with downstream distance.

The highest concentration of ^{238}Pu was detected at Site 1, but the highest concentration of $^{239,240}Pu$ was observed at Site 2. The $^{239,240}Pu$ flux increased by 860% at Site 2 compared to Site 1. This suggests that drainage or low-level discharges from F Area are a major source of $^{239,240}Pu$ to the stream. The loss of Pu to the streambed appears to be much greater for $^{239,240}Pu$ than for ^{238}Pu based on the flux calculations for Sites 2 and 4. The greater uncertainty of the $^{239,240}Pu$ values compared to ^{238}Pu may be responsible for much of this apparent difference, so we accept the ^{238}Pu flux calculations as being a more accurate indicator of loss to the streambed (Table 1).

TABLE 1

Plutonium, Cesium, and Gross Alpha Concentrations in Four Mile Creek

	Flow, L/sec	^{238}Pu, fCi/L	^{238}Pu Flux, pCi/sec	$^{239,240}Pu$, fCi/L	$^{239,240}Pu$ Flux, pCi/sec	^{137}Cs, pCi/L	Gross Alpha, fCi/L
Site 1	212	223 ± 6	47.3	8 ± 2	1.7	33 ± 5	720
Site 2	348	168 ± 5	58.5	42 ± 3	14.6	25 ± 5	1340
Site 3		186 ± 6		32 ± 2			
Site 4	572	68 ± 4	38.9	9 ± 2	5.0	17 ± 4	508
Site 5		34 ± 3		14 ± 2			
Site 6		17 ± 2		8 ± 2			

Researchers[9] have shown that a strong correlation often exists between ^{137}Cs and plutonium in soils and sediments, perhaps because both elements are tightly sorbed to soil particles. Our studies determined that a similar correlation exists for plutonium and cesium in stream water. A correlation coefficient of 0.986 ($p = 0.1$) exists between these two elements in stream water, which is similar to the correlation ($r = 0.965$) found between them in soils and sediments of midwestern watersheds (Figure 3).[9] Our results are based on total water samples which include both soluble and particulate forms.

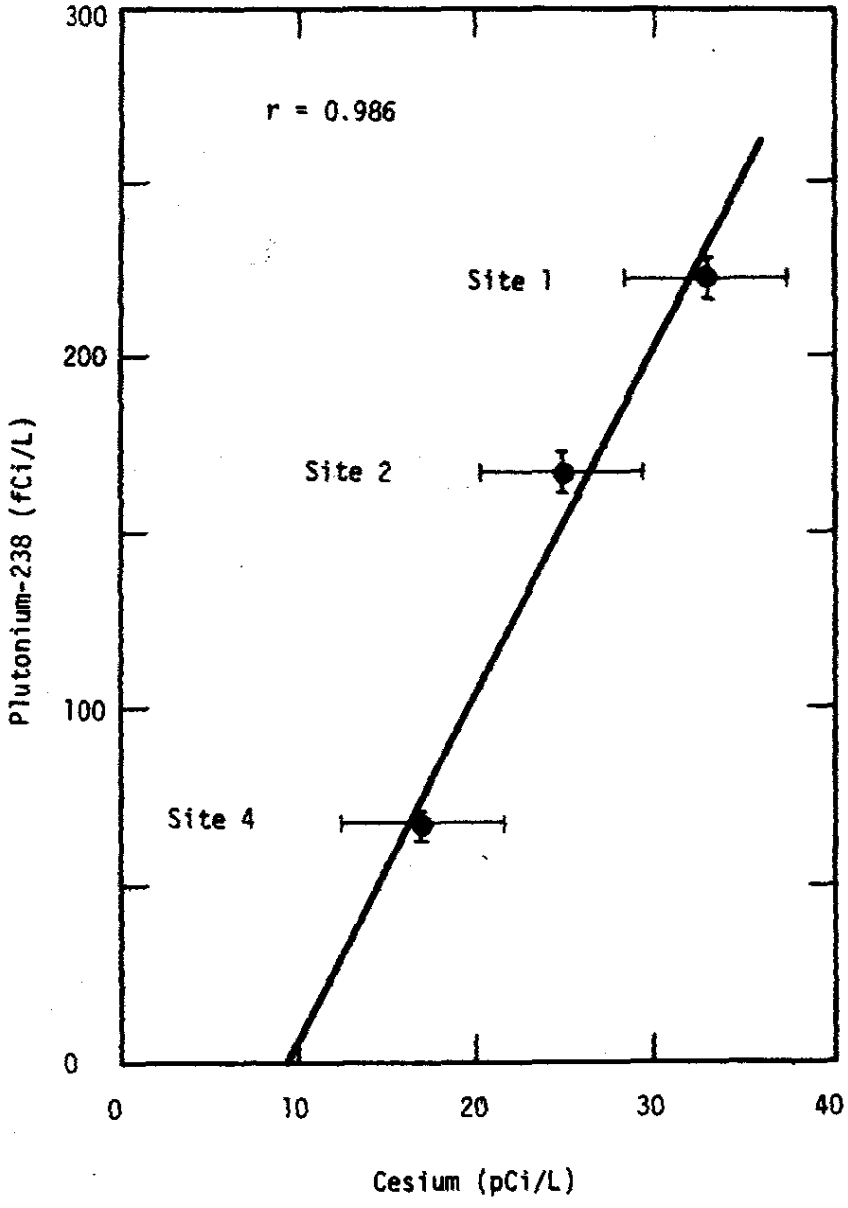

FIGURE 3. Correlation of ^{238}Pu and ^{137}Cs Concentrations in Water from Three Locations in Four Mile Creek. The line is the best fit calculated by the least square method.

Most of the ^{238}Pu was transported downstream during the sampling period with some net loss to the streambed. Recently, Kiser[10] noted similar transport behavior for stable cesium in Four Mile Creek. He released 130 g of cesium chloride in the upper portion of the creek and compared its transport with that of a dye released at the same location. He found that 88% of the stable cesium was transported downstream over a distance of 2.5 km. The cesium peak concentration lagged behind the peak dye concentration, indicating sorption-desorption behavior of the cesium. Our results seem to be consistent with Kiser's conclusions, considering the similarity in the transport behavior of ^{238}Pu and ^{137}Cs.

Table 2 shows additional data obtained in 1979 for transuranic elements in Four Mile Creek. The ^{239}Pu concentration for Site 4 is the same as the concentration measured four years earlier at the same location, but the ^{238}Pu concentration is somewhat lower than the earlier data. We have no discharge information that would explain this difference.

TABLE 2

Plutonium, Americium, and Curium Concentrations in Four Mile Creek at Site 4

Nuclide	Total Amount of Nuclide, 10^{-15} Ci/L	Nuclide in Particulate Phase, 10^{-15} Ci/L	Percent in Particulate Phase
^{238}Pu	39 ± 4	9 ± 0.5	23 ± 3
$^{239,240}Pu$	9.5 ± 2.0	2.8 ± 0.3	29 ± 6
^{241}Am	5.0 ± 1.0	1.9 ± 0.2	38 ± 8
^{244}Cm	7.5 ± 1.0	3.4 ± 0.2	46 ± 6

An interesting feature of the more recent data is that more than half of the transuranic activity was found in the soluble phase. Other researchers[11] have reported similar results, particularly when the suspended solid load is at low levels. The results shown in Table 2 were obtained from water samples when the suspended solids were 1 mg/L. This compares with an annual mean suspended solid concentration of 15 mg/L at that location. The flow rate of 220 L/sec was less than the mean flow rate for 1979 of 570 L/sec. The stream characteristics at the time of sampling are more typical of the relatively dry conditions of

summer rather than the average annual conditions. Assuming equilibrium exists between the soluble and particulate phase, it is possible to calculate distribution coefficients (K_d) for the data in Table 2. The distribution coefficient is defined as the activity of a given mass of particulate divided by the activity of an equal mass of the soluble phase. The distribution coefficients are given in Table 3.

TABLE 3

Distribution Coefficients for Transuranics in Four Mile Creek

Nuclide	K_d
^{241}Am	$(6.1 \pm 1.2) \times 10^5$
^{244}Cm	$(8.3 \pm 1.1) \times 10^5$
^{238}Pu	$(2.9 \pm 0.3) \times 10^5$
$^{239,249}Pu$	$(4.2 \pm 1.3) \times 10^5$

The distribution coefficients for americium and curium are only slightly different from the plutonium distribution coefficients. The usual range for plutonium distribution coefficients in natural aqueous environments is between 5×10^4 and 3×10^5. Wahlgren[12] proposed that the plutonium concentration of Lake Michigan water is controlled by an exchange reaction of the sedimentary plutonium with the overlying water following resuspension of the sediment particles. His data yield a distribution coefficient of ~3×10^5. Hetherington[13] and Pillai[14] also determined plutonium distribution coefficients, which were 5×10^4 and 9×10^4, respectively. Alberts[15] calculated a quantity similar to the distribution coefficient for Lake Michigan sediments and the overlying water; this quantity was called the concentration ratio. This is mathematically identical to the distribution coefficient, but equilibrium between the soluble and particulate phases is not assumed. Their concentration ratio was 1.7×10^5. Considering the variations in experimental methods, the similarity of these distribution coefficients reported by different authors is remarkable.

The gross alpha concentration is a conservative estimation for transuranic activity in Four Mile Creek. The gross alpha concentrations are at least a factor of two greater than the total transuranics (Table 1).

Although transuranic activity from the Savannah River Plant is detectable in Four Mile Creek, it is important to note that the observed levels are many orders of magnitude less than the maximum permissible concentrations for these transuranics (30 nCi/L) discharged in liquid effluent to the public domain.[16] The plutonium concentrations in the Savannah River downstream from Four Mile Creek are no higher - and in some cases lower - than plutonium concentrations in rivers where there are no nuclear installations.[2]

CONCLUSIONS

The results presented here indicate that a simple dilution model based on water volume is useful in describing the transport of plutonium in a small stream. However, actual downstream concentrations may be somewhat lower than that predicted by a dilution model due to stream bed sorption and sedimentation. Since transport mechanisms for plutonium and cesium are similar, cesium can in some cases be used as a tracer for plutonium. Distribution coefficients for americium and curium are similar to those of plutonium in this work, and the plutonium distribution coefficients are in good agreement with those of previous studies. The transport of transuranics in the soluble phase is significant in Four Mile Creek, at least during those times when the water contains relatively small amounts of suspended solids.

REFERENCES

1. C. A. Ashley and C. C. Ziegler. Releases of Radioactivity at the Savannah River Plant 1959 through 1974. USERDA Report DPSPU-77-25-1, E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC (1975).

2. D. W. Hayes and F. A. Cross. "Plutonium in Atlantic Coastal Estuaries in the Southeastern United States of America." Symposium on Transuranium Nuclides in the Environment, Vienna, 1976. Report No. IAEA-SM-199/84 (1976).

3. T. M. Langley and W. L. Marter. The Savannah River Plant Site. USAEC Report DP-1323, E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC (1973).

4. F. E. Butler. "Determination of Uranium and Americium-Curium in Urine by Liquid Ion Exchange." Analyt. Chem. 37, 340 (1965).

5. S. M. Sanders and S. C. Leidt. "A New Procedure for Plutonium Urinalysis." Health Phys. 6, 189 (1960).

6. K. M. Wong, G. S. Brown, and V. E. Noshkin. "A Rapid Procedure for Plutonium Separation in Large Volumes of Fresh and Saline Water by Manganese Dioxide Coprecipitation." J. Radioanal. Chem. 42, 7 (1978).

7. D. W. Hayes and J. H. Horton. "Plutonium and Americium Behavior in the Savannah River Marine Environment." Transuranic Elements in the Environment. USDOE Publication DOE/TIC-22800, U.S. Department of Energy, Washington, DC (1980).

8. R. Bojanowski, H. D. Livingston, D. L. Schneider, and D. R. Mann. "A Procedure for Analysis of Americium in Marine Environmental Samples." Reference Methods for Marine Radioactivity Studies II. IAEA Technical Reports Series No. 169, International Atomic Energy Agency, 77-86 (1975).

9. R. N. Muller, D. G. Sprugel, and B. Kohn. "Erosional Transport and Deposition of Plutonium and Cesium in Two Small Midwestern Watersheds." J. Environ. Qual. 7 (2), 171-174 (1978).

10. D. L. Kiser. Cesium Transport in Four Mile Creek of the Savannah River Plant. USDOE Report DP-1528, E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC (1979).

11. H. Singh and J. S. Marshall. "A Preliminary Assessment of ^{239,240}Pu Concentrations in a Stream Near Argonne National Laboratory." Health Phys. 32, 195 (1977).

12. M. A. Wahlgren, J. J. Alberts, D. M. Nelson, and K. A. Orlandini. "Study of the Behavior of Transuranics and Possible Chemical Homologues in Lake Michigan Water and Biota." Symposium on Transuranium Nuclides in the Environment, Vienna, 1976. Report No. IAEA-SM-199/27, 9-24 (1976).

13. J. A. Hetherington, D. F. Jeffries, N. T. Mitchell, R. J. Pentreath, and D. S. Woodlead. "Environmental and Public Health Consequences of the Controlled Dispersion of Transuranic Elements to the Marine Environment." Symposium on Transuranium Nuclides in the Environment, Vienna, 1976. Report No. IAEA-SM-199/11, 139-155 (1976).

14. K. C. Pillai and F. Mathew. "Plutonium in the Aquatic Environment." Symposium on Transuranium Elements in the Environment, Vienna, 1976. Report No. IAEA-SM-199/27, 25-45 (1976).

15. J. J. Alberts and R. N. Muller. "The Distribution of ^{239,240}Pu and ^{137}Cs in Various Particle Size Classes of Lake Michigan Sediments." J. Environ. Qual. 8, 20-22 (1979).

16. United States Code of Federal Regulations, Title 10, Part 20 (1979).

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