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CESIUM TRANSPORT IN FOUR MILE CREEK OF THE SAVANNAH RIVER PLANT

D. L. KISER

SAVANNAH RIVER LABORATORY
AIKEN, SOUTH CAROLINA 29801

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by

D. L. Kiser

Savannah River Laboratory
E. I. du Pont de Nemours and Company
Aiken, South Carolina 19801

Approved by

T. V. Crawford, Research Manager
Environmental Transport Division

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ABSTRACT

The behavior of a large radioactive cesium release to a Savannah River Plant (SRP) stream was examined using a stable cesium release to Four Mile Creek. Measurements following the release show that most of the cesium released was transported downstream; however, sorption and desorption decreased the maximum concentration and increased the travel time and duration, relative to a dye tracer, at sampling stations downstream. The study was made possible by the development of an analytical technique using ammonium molybdophosphate and neutron activation that permitted the measurement of stable cesium concentrations as low as 0.2 μg/L.

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CESIUM TRANSPORT IN FOUR MILE CREEK OF THE SAVANNAH RIVER PLANT

INTRODUCTION

Traces of ^{137}Cs produced by nuclear fission are found in the environment of the Savannah River Plant. The behavior of this radioisotope in an SRP creek must be known to predict the consequences of a large release. Because cesium interacts with the stream environment by sorption, stream models of conservative (no significant interaction with the stream or its environment) transport would be expected to overpredict concentrations downstream of a release. The magnitude of the sorption was determined in a field study with stable cesium.

LITERATURE SURVEY OF THE ELEMENT CESIUM

Chemical and Physical Characteristics

Cesium reacts as a univalent cation in all of its compounds (chemically it behaves similarly to potassium and rubidium). Twenty-one isotopes of cesium have been identified, ranging from Mass 123 to Mass 144.[2] All are radioactive except for its naturally occurring stable isotope of Mass 133. The radioactive isotopes of cesium decay with half-lives from less than one minute to 2.0×10^6 years (^{135}Cs). ^{137}Cs is important because of its relatively long half-life (30 years) and biological hazard (0.66 MeV gamma rays).

Cesium in Natural Streams

Stable cesium concentrations determined in United States streams in the 1950's were reported to be about 1 μg/L (1 ppb). With development of a technique using large volumes of water by Feldman and Rains,[3] the concentration range reported for U.S. fresh waters is 0.004 - 0.065 μg/L with an average of 0.018 μg/L.[4,5]

$^{134,137}Cs$ concentration in Four Mile Creek (stream of this study) is usually less than 100 pCi/L.[6] The drinking water standard as specified by DOE for ^{137}Cs is 2×10^4 pCi/L (230 pg/L).[7]

Sorption Behavior

In streams, cesium is accumulated in the biota, sediments, and suspended materials. The principle mechanisms are cation exchange with the clay minerals, chelation with naturally occurring organic acids, and surface exchange or metabolic processes with the biota. Kaolinite, the dominant clay mineral at SRP, has the lowest cation exchange capacity (3-15 meq/100 g) of the common clays.[8]

The interaction of ^{137}Cs with an aquatic environment was studied by Shih and Gloyna[9] in a model river system at the University of Texas. The model river system consisted of a flume with water recirculated from a supply tank. The flume contained Lake Austin sediments, rooted plants characteristic of aquatic systems, and several microorganisms. The Lake Austin sediment contained 30% clay (illite and vermiculite) and had a cation exchange capacity of 29 meq/100 g of sediment. Of the ^{137}Cs ion released, an average of 16% was sorbed by bed sediments, an average of 2% was sorbed by the aquatic plants, and an average of 14% was associated with the suspended materials following 120 days of continuous recirculation.

A study on the introduction of stable cesium into an aquatic ecosystem was done by Hakonson[10] while at Colorado State University. One kilogram of cesium as cesium chloride was released to East Twin Lake in the Rocky Mountains of northern Colorado. East Twin Lake is a public lake, with a volume of 146,000 to 160,000 m^3 and a surface area of about 50,000 m^2. Mean concentration of cesium in the water at the dosage point one and one-half hours after the release was 9.3 μg Cs/L. The pathway of the cesium was from the water to the invertebrates (zooplankton and amphipod), seston,* vegetation (lily and duckweed), and sediments. Cesium accumulation in trout muscle was observed to be from the invertebrates. Other routes were insignificant. Laboratory studies with ^{134}Cs also supported these results. Bottom sediments were identified as the major site of deposition of the stable cesium. The sediment 393 days after dosing accounted for 82% of the stable cesium in the lake compared to 3% for water and 14% for seston.

The uptake of radionuclides by a solid from a liquid is expressed as the distribution coefficient, K_d. The distribution coefficient is defined as the ratio of the concentration of ionic species in the solid to ionic species in the liquid.

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* Seston: minute material moving in water, including both living organisms (plankton and nekton) and nonliving matter (plant debris or suspended soil particles).

$$K_d = \frac{f_s/\text{mass of solid}}{f_e/\text{volume of solution}} = \frac{f_s}{f_e} \times \frac{\text{volume}}{\text{mass}}$$

where:

f_s is the fraction of radioactivity sorbed by the solid

f_e is the fraction of activity remaining in the solution

Prout[11] examined the equilibrium distribution coefficient of ^{137}Cs on SRP soil. He found that the K_d for ^{137}Cs was very dependent on the pH of the solution and on the ^{137}Cs concentration. The maximum K_d occurred near pH 8, and and the K_d increased with decreasing concentration of ^{137}Cs.

LABORATORY DETERMINATION OF THE ^{137}Cs DISTRIBUTION COEFFICIENT FOR SRP STREAMS

The distribution coefficients for three streambed sediments were evaluated: (1) a composite of samples taken near Road E, Road 4, and Road C on Four Mile Creek, referred to as Four Mile Creek-E, 4, C composite, (2) a composite of fifty samples collected from H Area to the swamp on Four Mile Creek and from P Area to the swamp on Steel Creek, referred to as Steel and Four Mile Creek Upland composite, and (3) a composite of twenty-four samples collected near the delta areas of Steel and Four Mile Creek in the Savannah River swamp, referred to as Steel and Four Mile Creek Swamp composite.

Table 1 lists the soil properties examined in addition to the distribution coefficients. All sediments were air dried, then oven dried, sieved through a U.S. Standard #10, and placed in a closed container prior to analysis. The particle size distribution (sand, silt and clay) was determined by the Bouyoucos hydrometer method.[12] The cation exchange capacity and total organic carbon were determined by methods from Agriculture Handbook No. 60.[13] The sodium of the cation exchange capacity method was measured with atomic absorption. The oxidizable organic was determined by weight loss after treating the soil with a 30% hydrogen peroxide solution. Sediments treated with the hydrogen peroxide solution did not show any organic carbon.

For the K_d determination, a minute volume of ^{137}Cs solution was added to 1 liter of Four Mile Creek water with a pH of approximately 7. The solution was placed in a mechanical shaker for five minutes prior to removing an aliquot for counting. To the remainder, 1 gram of dry sediment was added. The solution was then shaken in a mechanical shaker for two hours and allowed to stand for about twenty-four hours. The sample was centrifuged and an aliquot of the supernatent removed for counting. The solids were dried in a pre-weighed test tube. After determining the combined weight of the test tube and sediments, the sample was submitted for counting. Results were discarded where the radioactivity of the liquid and solid did not equal the radioactivity of the initial solution. Figure 1 shows the results, which agree with Prout's,[11] for pH 7. Because K_d appeared to correlate with the organic content rather than the clay content, K_d was measured for the hydrogen peroxide treated sediments. The results show that there are about 1150 ^{137}Cs K_d units per each percent organic matter present.

The K_d's of Rhodamine WT dye are also included in Table 1. For the same initial concentration (100 μg/L), the ^{137}Cs K_d from Figure 1 is 980. Rhodamine WT dye is a conservative tracer and is used in the development of stream models for SRP creeks. Rhodamine WT exhibits weak anionic behavior in water (very small sorption capability) and has a very low photochemical decay rate.[14]

TABLE 1. Characterization of Streambed Sediments from Four Mile and Steel Creeks

Analysis	Four Mile Creek E, 4, C Composite	Steel and Four Mile Creeks Upland Composite	Steel and Four Mile Creeks Swamp Composite
Sand, %	84	80	72
Silt, %	4	10	10
Clay, %	12	10	18
Cation Exchange Capacity, meq/100 g	5.51	5.04	6.60
Total Organic Carbon, %	0.82	0.37	1.02
Organic Matter, % (T.O.C. x 1.72)	1.41	0.64	1.75
Oxidizable Organic, %	1.0	0.5	1.0
^{137}Cs Kd, mL/g[a]	3420	2040	3960
^{137}Cs Kd for Peroxide Treated Sediments, mL/g[a]	1830	1230	2100
Rhodamine WT Kd, mL/g[b]	Not Analyzed	53	81

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[a] 100 to 200 pCi/mL (1.15 to 2.30 pg/mL) initial concentration and near pH 7.

[b] Near 100 μg/L initial concentration.

Of several concentration techniques evaluated, the best was found to be extraction with ammonium 12-molybdophosphate (AMP) followed by centrifugation. Solvent extraction with potassium hexacyanocobalt II ferrate II (KCFC-commonly used to extract ^{137}Cs from environmental water samples) resin was unacceptable, because the cobalt and iron produce long half-life activation products. AMP, a microcrystalline powder, was used by Feldman and Rains[3] in an elaborate batching technique with a recovery of 98.4%. ^{96}Mo activates to short half-life (66.6 hr) ^{99}Mo, which decays to insignificance in a few weeks. However, the elaborate batching technique would be very time consuming with very many 50-liter samples.

Zirconium phosphate (ZrP) resin[15] was evaluated, because activation products are not significant and a continuous flow column could be used to process the water. However, high Na^+ and K^+ concentrations interfere with the recovery[15] because the resin lacks selectivity. The Zr P resin was tested with a standard solution of stable cesium under conditions designed to simulate the range of concentrations expected in a postulated release. Fifty liters of water from Four Mile Creek near the proposed release point was pumped at 5 liters/hour through a resin-containing vial, which would be expected to give a recovery approaching 100%. Significant amounts of sodium and iron showed up as activation products in the resin; therefore, this method was abandoned.

Ammonium Molybdophosphate Methods

AMP was purchased from Bio-Rad Laboratories of Richmond, California. A series of standards and blanks was analyzed to determine the suitability of AMP. For processing 50 liters of creek water, water from a 55-liter polyethylene drum was transferred to two ~30-liter polyethylene carboys with the top portion removed. All containers used in this study were pickled with 1×10^{-4} M HCl prior to use. This was to remove any sorbants that might be on the container walls. To each carboy, 2.5 grams of AMP was added. The solution was mixed for about one hour on a magnetic stirrer-hot plate while heating to 40°C. The carboy was removed briefly from the magnetic stirrer-hot plate to add 10 mL of $Al(NO_3)_3$ solution (5 mg Al^{3+} per mL) and then returned for additional stirring. The aluminum solution aids in flocculation of the AMP. The stirred solution was then pumped through a Millipore filter (Cat. No. YY4014200) containing 0.8-μ filter paper (Millipore AAWP14200). Originally, 0.45-μ filter paper was used, but it plugged shortly after the start of pumping of the solution. The filter paper was removed from the holder, dried in an oven, and then ashed. The residue was placed in a 5-mL vial for neutron activation.

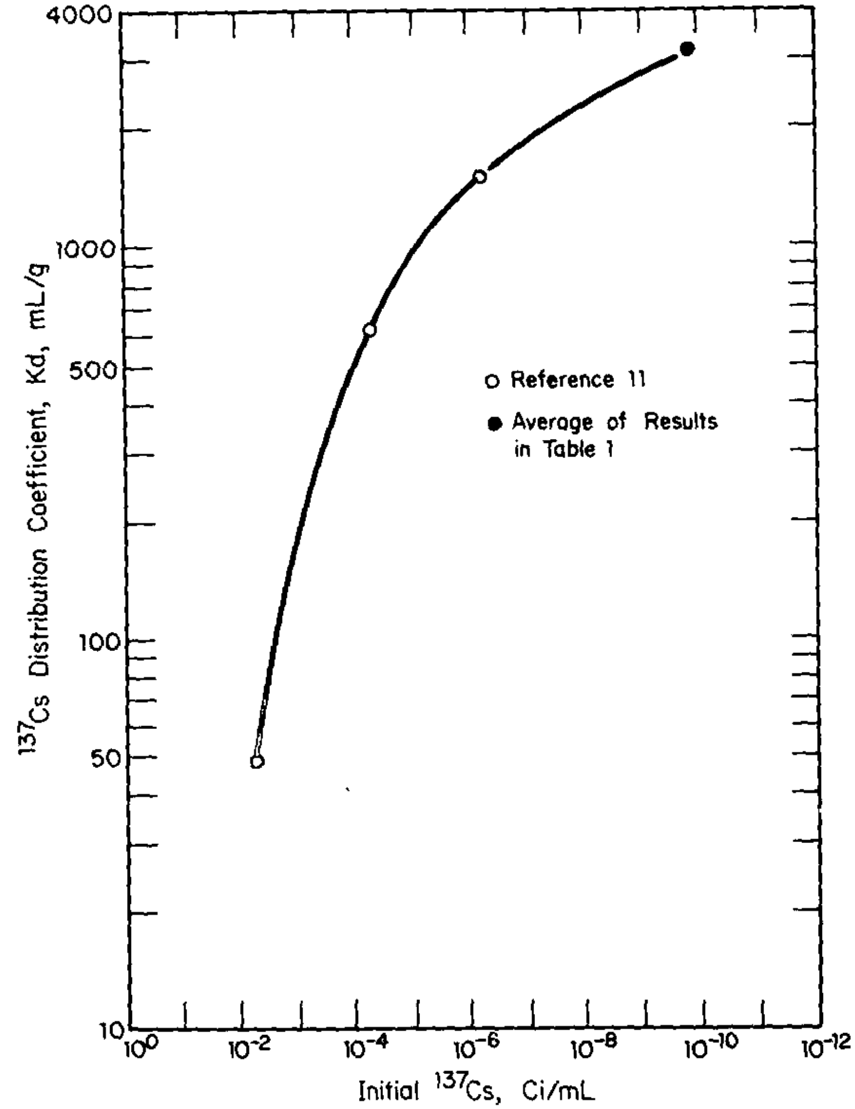

FIGURE 1. Effect of Cesium Concentration on Distribution Coefficient in Water at pH 7

AN ANALYTICAL TECHNIQUE TO DETERMINE VERY LOW CONCENTRATIONS OF STABLE CESIUM IN CREEK WATER

A technique was developed to determine very low stable cesium concentrations in creek water. The cesium from a large volume of water was concentrated and measured by neutron activation. Exposure of ^{133}Cs to thermal neutrons transforms ^{133}Cs to radioactive ^{134}Cs, which has a half-life of 2.06 yr. Samples were analyzed in the ^{252}Cf facility for Neutron Activation.

Five grams of AMP was analyzed in the neutron activation facility to determine its cesium content. To 50 liters of creek water, 500 µg of ^{133}Cs was added. This sample was referred to as AMP with intermediate level cesium. Table 2 lists the results of the neutron activation. The samples were allowed to decay for two weeks before counting. The sample "AMP with background" H_2O indicates the creek water has a background concentration of 0.24 µg/liter (unusually high, see page). Adjusting the sample "AMP with low level cesium" for background shows the result is above that expected.

TABLE 2. Cesium Determination with Ammonium Molybdophosphate Extractant (Filtered)

Sample	Total ^{133}Cs Expected, µg	^{133}Cs Found by Neutron Activation Analyses, µg
Stable Cesium Chloride Std. Soln.	5010	5380 ±50
AMP	None	None Detected
AMP with Bkg. H_2O	Unknown	12 ±4
AMP with Low Level ^{133}Cs	50 + Background	88 ±7
AMP with Intermediate Level ^{133}Cs	500 + Background	414 ±14

To further improve the study, a pair of Sorvall model SS-3 continuous flow centrifuges were substituted for the filtration step. These centrifuges were equipped with SS34 rotor assembly and a Szent, Gyorgyi, and Blume continuous flow system Type KSB-3. The operating speed was 15,000 rpm (27,000 RCF), and the flow was maintained at 10 liters/hour. About 400 mL of concentrate was collected from each sample. The concentrate was transferred to a one-liter beaker and evaporated to dryness on a hot plate. The dried solids were scraped from the beaker, weighed, and placed in a sample vial for neutron activation. Table 3 lists the results of the neutron activation. The samples were allowed to decay for four weeks before counting. Because only one sample of eight showed a background concentration (attributed to contamination in preparation) in this set, background concentration was taken as the limit of detection (about 10 µg). For the spiked samples as a group, the average recovery was 95.2 ±6.5%. Three samples, AMP with low level ^{133}Cs #1, AMP with intermediate level ^{133}Cs #1, and AMP with high level ^{133}Cs, had Rhodamine WT dye added to the creek water at concentrations of 5 μg/L, 50 μg/L, and μg/L, respectively. The purpose was to determine whether the dye had any significant effect on the cesium analysis. The results show no effect on cesium recovery from the presence of Rhodamine WT.

TABLE 3. Cesium Determination with Ammonium Molybdophosphate Extractant (Centrifuged)

Sample	Total ^{133}Cs Expected, μg	^{133}Cs Found by Neutron Activation Analyses, μg
AMP with Background H_2O #1	Unknown	None Detected
AMP with Background H_2O #2	Unknown	None Detected
Amp with Background H_2O #3	Unknown	None Detected
AMP with Background H_2O #4	Unknown	None Detected
AMP with Background H_2O #5	Unknown	None Detected
AMP with Background H_2O #6	Unknown	None Detected
AMP with Background H_2O #7	Unknown	None Detected
AMP with Background H_2O #8	Unknown	29 ±6
AMP with Low Level ^{133}Cs #1	50 + Background	60 ±7
AMP with Low Level ^{133}Cs #2	50 + Background	53 ±6
AMP with Low Level ^{133}Cs #3	100 + Background	96 ±7
AMP with Low Level ^{133}Cs #4	100 + Background	100 ±7
AMP with Intermediate Level ^{133}Cs #1	500 + Background	435 ±17
AMP with Intermediate Level ^{133}Cs #2	500 + Background	470 ±14
AMP with Intermediate Level ^{133}Cs #3	500 + Background	487 ±15
AMP with High Level ^{133}Cs	1002 + Background	946 ±28

FIELD EXPERIMENT

Area of Study

The creek selected to study stable cesium transport was Four Mile Creek from Road E near H Area to Road C-4 below F Area (Figure 2). The streambed material is primarily sand between Road E and Road 4. The sediments between Road C and Road C-4 contain more clay and organic matter content than elsewhere on this creek. The section between Road 4 and Road C is intermediate. At the time of release, flow measured by U.S.G.S. gauges was 31.2 L/sec at Road E, 144.4 L/sec at Road 4, and 237.9 L/sec at Road C.

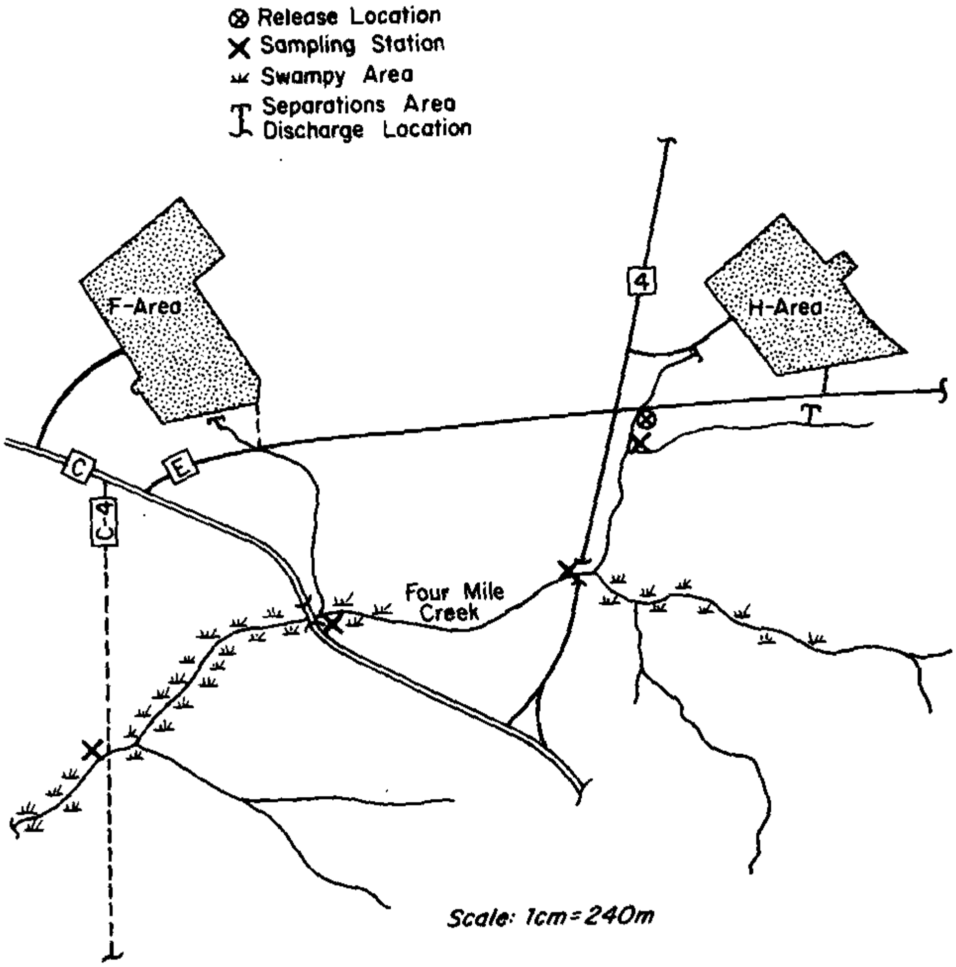

FIGURE 2. Four Mile Creek Region Below the Separation Areas at SRP

Chemical Toxicity

The literature of the chemical toxicity of cesium chloride was surveyed prior to tracer study to assure no significant impact on aquatic biota from the experiment. In a study of chemical toxicity of cesium chloride to rats, Cochran, et. al,[16] showed that the LD_{50}* was 1500 mg/kg (1,118 mg Cs/kg) of live weight. The LD_{50} for mice was determined to be 1420 mg/kg.[17] Sax[18] rates the toxicity hazard of cesium as slight (causes readily reversible changes which disappear after end of exposure).

Acute or chronic toxicity of stable cesium to humans is unknown. For aquatic life, Hakonson[10] reported the acute chemical toxicity of cesium chloride to invertebrates based on laboratory studies. There was 100% survival at a concentration of 6.9 mg ^{133}Cs/L for 5 days of exposure. The calculated LD_{50} was about 9 mg ^{133}Cs/L for zooplankton (**Cyclops sp.**) and amphipod (**Gammarus lacustris**).

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* LD_{50}: lethal dose to 50% of the population.

Release

One liter of 20% Rhodamine WT dye, a conservative tracer, was released to Four Mile Creek at Road E near H Area at about 9 AM on April 11, 1978. The creek was sampled for dye by dipping 250-mL bottles at the first sampling station near Road E (Figure 2). At the sampling stations at Road 4, Road C, and Road C-4, ISCO* Model 1680 automatic water samplers sampled the stream. The dye concentrations were determined using a Turner Model 111 fluorometer.

One hour after the dye release, 130 grams of cesium chloride (about 100 g ^{133}Cs) were released to Four Mile Creek at the same location as the dye. The dye-cesium releases were staggered to avoid any initial interaction between the dye (weak anionic) and the cesium (strong cationic). Sample collection frequency and sample size were adjusted at each of the sampling locations to ensure adequate cesium for analysis. At the release station near Road E, a 250-mL bottle was dipped at one minute intervals and composited to one liter. At Road 4, a one-liter bottle was dipped at one minute intervals and composited to 10 liters. At Road C, a two-liter bottle was dipped at one minute intervals and composited to 50 liters. At Road C-4, a two-liter bottle was dipped at two and half minute intervals and composited to 50 liters. Figures 3 through 6 show the concentration time profiles for cesium and dye at each of the sampling locations.

Discussion of Results

A background concentration of 0.4 μg/L stable cesium was subtracted from the concentrations plotted. This value is based on the average of four samples (one from each sampling station) and is near the lower limit of measurement by the analytical technique (0.2 μg/L). The reason for the elevated background concentration is unknown.

The cesium sorption decreased the ratio of the peak concentration at the last station to the peak concentration at the first station significantly in comparison with the dye. The dye ratio (Road C-4: Near Road E) is 3.57×10^{-3} but the cesium ratio (Road C-4: Near Road E) is 7.84×10^{-4}. However, the cesium desorption, dramatically illustrated by the large tailing effect at Road C (Figure 5) and Road C-4 (Figure 6), maintained most of the cesium in transport as shown by a comparison of the dye and the cesium time-integrated concentration ratio for the first and second sections. For the first section of stream the time-integrated concentration ratio (Road 4: Near Road E) for cesium is 88% of that for the dye. For the intermediate section, the time-integrated

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* Instrument Specialties Company, Lincoln, Nebraska.

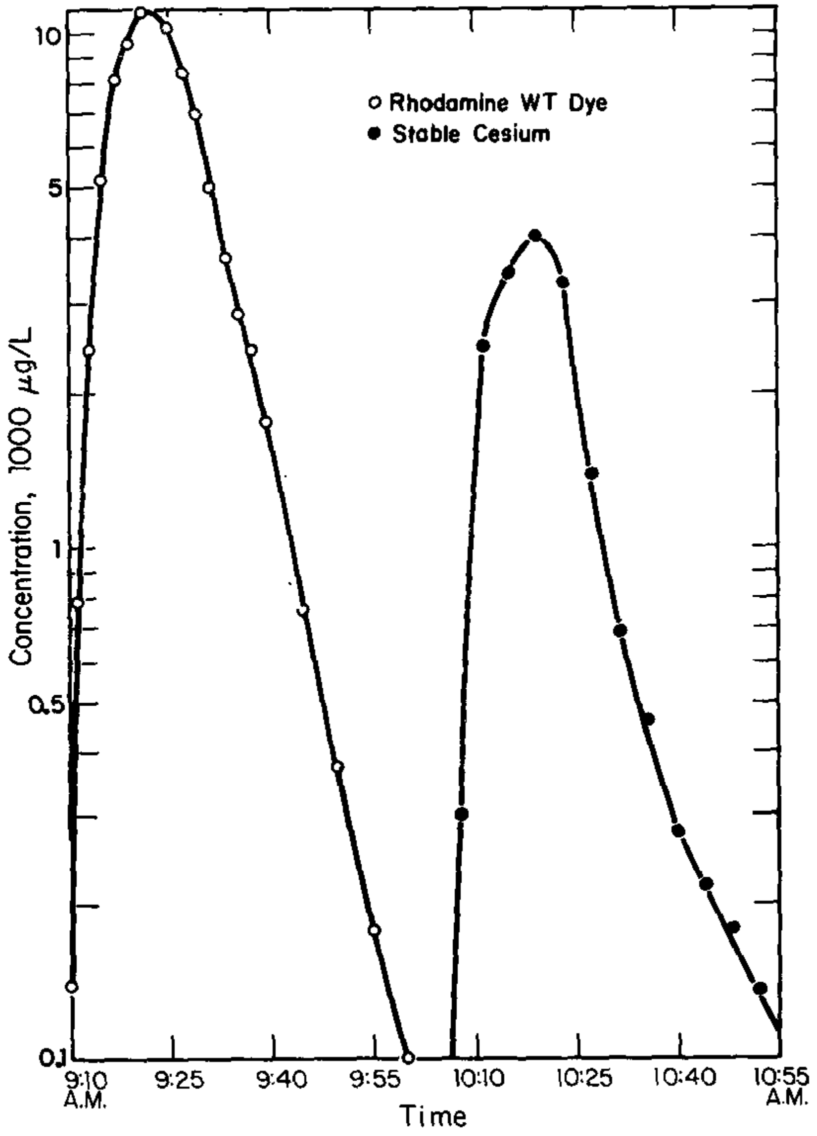

FIGURE 3. Passage of Dye and Cesium on Four Mile Creek Near Road E

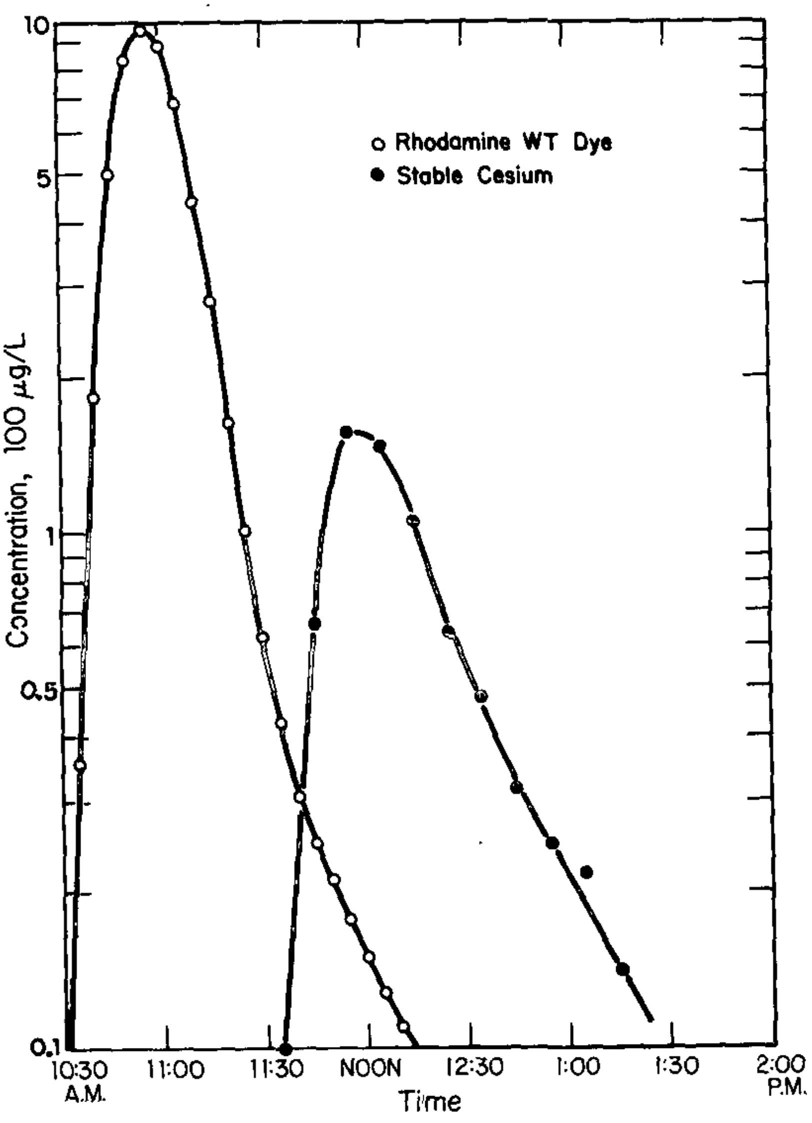

FIGURE 4. Passage of Dye and Cesium on Four Mile Creek at Road 4

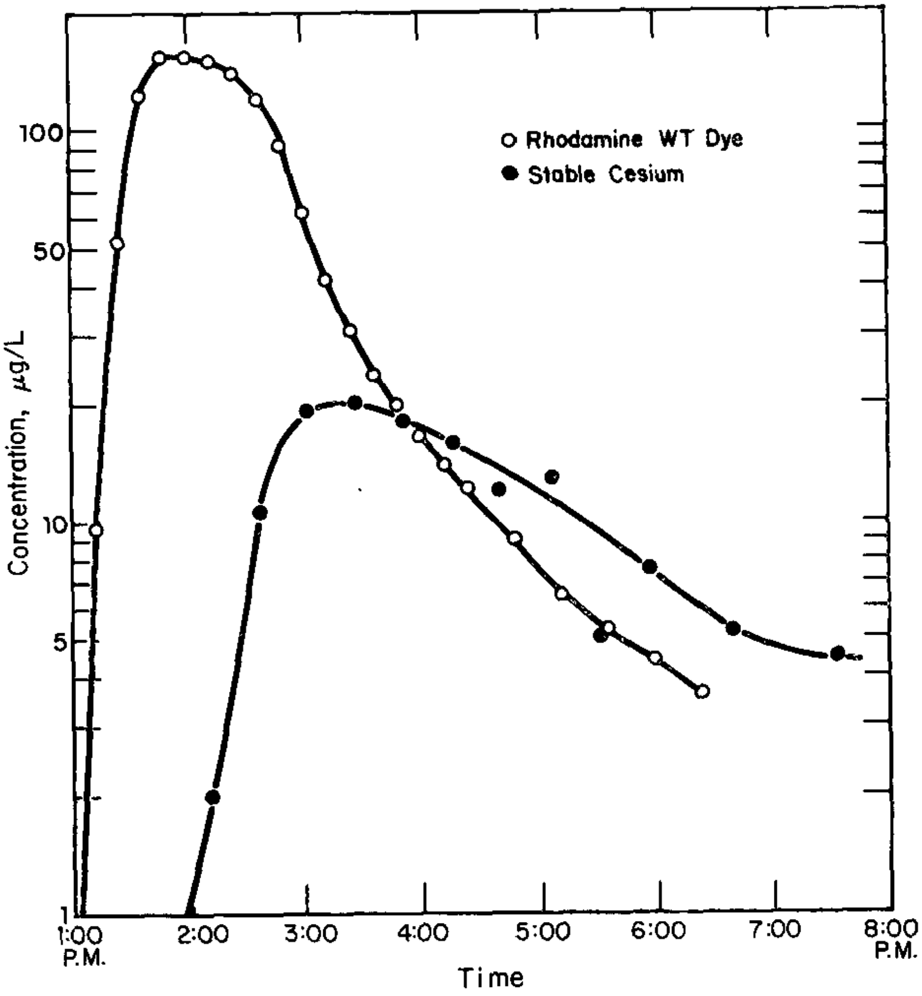

FIGURE 5. Passage of Dye and Cesium on Four Mile Creek at Road C

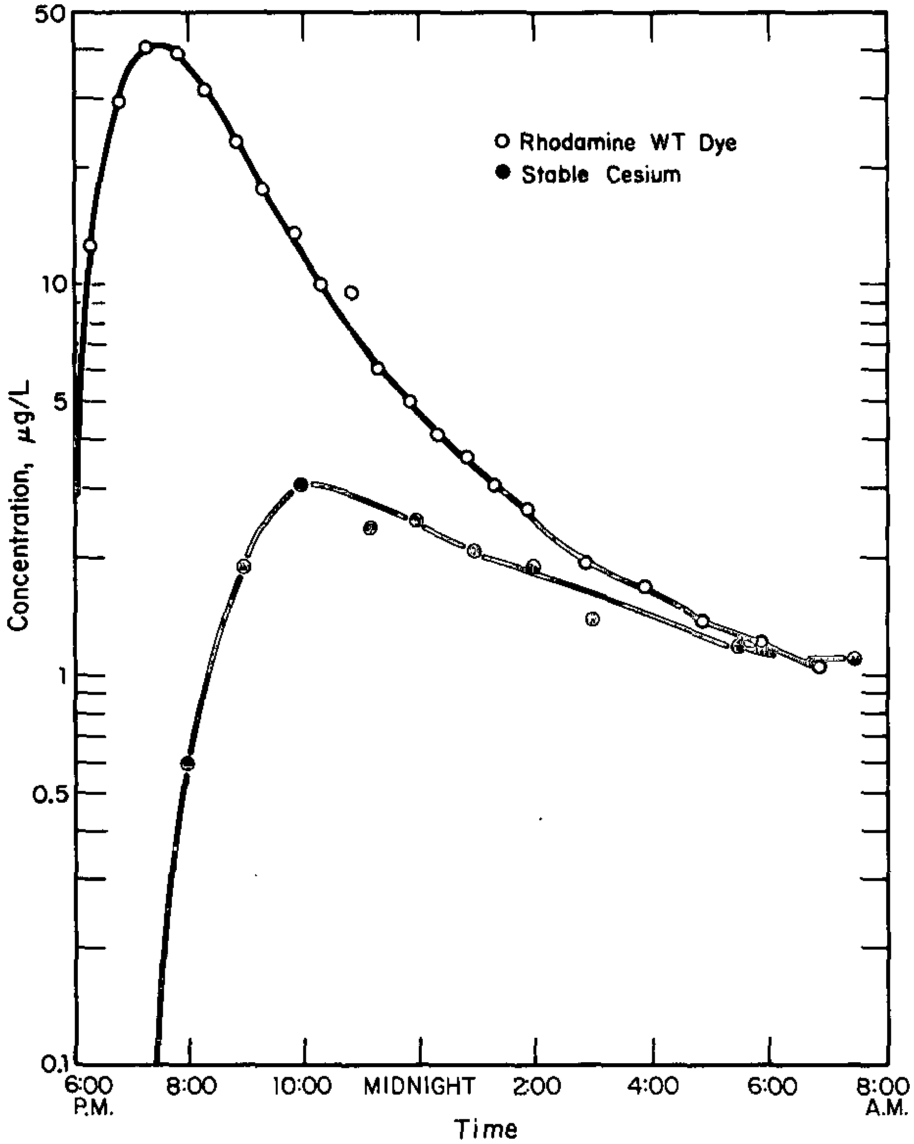

FIGURE 6. Passage of Dye and Cesium on Four Mile Creek at Road C-4

concentration ratio (Road C: Road 4) for the cesium is 89% of that for the dye. Cesium was not sampled long enough at Road C-4 to determine this ratio effect for the third section. The sorption-desorption increased the peak-to-peak travel time between the dye and the cesium from one hour at the stations Road 4 and near Road E to one and one-half hours at Road C station and two and one-half hours at Road C-4 station.

CONCLUSIONS

Most of the stable cesium released to Four Mile Creek was transported downstream. The sorption-desorption behavior of cesium decreased the maximum concentration and increased the travel time and duration in comparison to the Rhodamine WT dye.

An analytical technique using ammonium molybdophosphate extractant and neutron activation analysis was developed that successfully measures very low concentrations of stable cesium in creek water.

REFERENCES

1. Kirk-Othmer. **Encyclopedia of Chemical Technology**, 2nd ed., Vol. 4, pp. 855-868, Interscience, New York, NY (1964).

2. **Radiological Health Handbook.** January 1970 ed., pp. 301-303, U. S. Department of HEW. Public Health Service, Washington, DC (1970).

3. C. Feldman and J. C. Rains. "The Collection and Flame Photometric Determination of Cesium." **Anal Chem.** 36, 405 (1964).

4. C. Sreekumaran, K. C. Pillai, and J. R. Folsom. "The Concentrations of Lithrum, Potassium, Rubidium, and Cesium in Some Western American Rivers and Marine Sediments." **Geochimica et Cosmochimica Acta** 32, 1229 (1968).

5. D. P. Kharkar, K. K. Turekian, and K. K. Bertine. "Stream Supply of Dissolved Silver, Molydenum, Antimony, Selenium, Chromium, Cobalt, Rubidium, and Cesium to the Oceans." **Geochim. Cosmochimica Acta**, 32, 285 (1968).

6. C. Ashley, and C. C. Zeigler. **Environmental Monitoring at the Savannah River Plant - Annual Reports 1974, 1975, and 1976.** DPSPU 75-302, DPSPU 76-302, DPSPU 77-302.

7. **E.R.D.A. Manual.** Chapter 0524 "Standards for Radiation Protection." Approved March 30, 1977.

8. R. E. Grim. **Clay Mineralogy.** p. 27, McGraw-Hill Book Company, New York, (1953).

9. C. Shih and E. F. Gloyna. **Radioactivity Transport in Water-Transport of ^{85}Sr and ^{137}Cs in a Aquatic Environment.** Technical Report 10, EHE-01-6602, Civil Engineering Department, University of Texas, Austin, TX (1966).

10. T. E. Hakonson. **Cesium Kinetics in a Montane Lake Ecosystem.** Ph.D. dissertation, Radiology & Radiation Biology Dept., Colorado State University, Fort Collins, (1972).

11. W. E. Prout. **Adsorption of Fission Products by Savannah River Plant Soil.** USAEC Report DP-394, Savannah River Laboratory (1959).

12. G. J. Bouyoucos. "A Recalibration of the Hydrometer Method for Making Mechanical Analysis of Soils." **Agron. J. 43,** 434 (1951).

13. L. A. Richards, Ed. "Diagnosis and Improvement of Saline and Alkali Soils." **Agriculture Handbook No. 60,** U.S. Department of Agriculture. (1954).

14. P. L. Smart and I. M. S. Laidlaw. "An Evaluation of Some Fluorescent Dyes for Water Tracing." **Water Resources 13,** 15 (1977).

15. G. Seelye. **A Measurement of Low Level Cesium Isotope Concentration in a Fresh Water Lake.** COO-1795-7, Michigan State University, East Lansing, Department of Fisheries and Wildlife (1970).

16. K. W. Cochran, J. Doull, M. Marcella, and K. P. Dubois. "Acute Toxicity of Zirconium, Columbium, Strontium, Lanthanum Cesium, Tantalum, and Yttrium." **Arch. Ind. Hyg. Occupational Med.** 1, 644 (1950).

17. **The N.I.O.S.H. Toxic Substance List.** 1974 ed., U.S. Department of HEW, Publication No. 74-134 (1974).

18. N. I. Sax. **Dangerous Properties of Industrial Materials.** 4th ed., p. 530, Van Nostrand Reinhold Company, New York (1975).