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PLUTONIUM AND AMERICIUM BEHAVIOR IN THE SAVANNAH RIVER MARINE ENVIRONMENT

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D. W. Hayes and J. H. Horton

Savannah River Laboratory
E. I. du Pont de Nemours and Company
Aiken, South Carolina 29801

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Transuranic Elements in the Environment

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INTRODUCTION

Thirteen power reactors, two fuel fabrication plants and a Department of Energy nuclear production complex, the Savannah River Plant (SRP), are operating on rivers or in coastal regions of the Southeastern United States. Rivers and estuaries are a major geographic feature of this region and can represent both transport paths and sinks for transuranics. Studies are in progress to establish the distribution and transport properties of transuranic elements in the rivers and estuaries of this region. Of particular interest is the Savannah River and its estuary because located on the watershed are the Savannah River Plant and three commercial power reactors. These facilities make the Savannah River watershed one of the most intensively developed

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nuclear watersheds in the U.S. The Savannah River Plant consists of three production reactors, two fuel separation plants, a fuel fabrication facility, and a heavy water plant. The SRP has been in operation since 1952, whereas the three power reactors located at the headwaters of the Savannah River have operated for less than ten years. Included in this report are estimates of watershed loss rates for plutonium, measurements of plutonium and americium concentrations in the water and sediments of the Savannah River and estuary, estimates of plutonium concentrations in seafood, and dose rate estimates for seafood ingestion.

SAVANNAH RIVER BASIN AND ESTUARY

The Savannah River Basin has a surface area of 27,400 km^2. It may be divided into three physiographic provinces that transect the basin (Figure 1). The Blue Ridge Mountains include portions of North Carolina, South Carolina, and Georgia and have elevations that range from 5500 feet at the headwaters to about 1000 feet at the piedmont plateau. The hilly plateau descends from 1000 feet to about 200 feet near Augusta, Georgia. The gently rolling (upper) to nearly level (lower) Atlantic coastal plains extend from Augusta, Georgia to the ocean.

Of the 16 rivers in the Southeastern U.S. with flows greater than 30 m^3/sec (1000 cfs), the Savannah River is fourth in volume flow with an average of about 340 m^3/sec (12,000 cfs). The flow is regulated by two large impoundments, the Clark Hill and Hartwell Reservoirs, located in the piedmont region (Figure 1).

Each reservoir has a capacity exceeding 3 km^3 and can contain the equivalent of one half to one year's flow for the region of the river where it is located.

The Savannah River Estuary is relatively narrow (about 0.5 km) and maintained at a minimum depth of 11 m for shipping throughout its length of 35 km. To maintain the depth of 11 m in the harbor requires practically continual dredging and the dredge materials are dumped on adjacent areas to the north of the harbor. The harbor region has a tidal range of 2.1 to 2.4 m. The estuary classification is that of a moderately stratified one. The estuary has been heavily polluted with raw sewage and industrial waste, but these pollutants have been reduced considerably in the last few years.

SAMPLING

Water, sediment, and seafood samples were collected and analyzed to permit transport, inventory, and dose-to-man calculations for the Savannah River and its estuary. Location of the sampling station for plutonium transport in the Savannah River watershed is shown in Figure 1. This location was chosen because the sampler could be easily located to take water samples near mid-stream. Monthly composite samples were collected. To avoid sample bias, four samples per day, about 300 cc each, were taken from a depth of 1 m by an automatic compositing sampler.

Estuary water and sediment sample locations are shown in Figure 2. Sediment was collected in marshes where vegetation would lend some stability to the sediments. The sediments were collected by inserting a 3.6 cm diameter core barrel into the sediments. The cores were then extruded, sectioned, and bottled. Estuary water samples were 50 L grab-samples from a depth of 1 m.

Oysters and crab meat were obtained from a local wholesaler who obtained the oysters from Wassaw Island and the crabs from crab pots located in Wassaw Sound (Figure 2). Clams were collected from Port Royal Sound which is about 32 km north of the mouth of the Savannah River Estuary. Shad were netted in the Savannah River, and mullet and speckled trout were obtained from a local wholesaler whose boats work in the Savannah River estuary and nearby waters.

ANALYTICAL METHODS

The procedure developed by Wong (1) for concentrating plutonium from large volumes of water was adapted for use on these water samples for both plutonium and americium analysis. In the modified method the 50 L of water in the drum was adjusted to pH 2 using hydrochloric acid. ^{236}Pu and ^{243}Am spikes were added and the sample equilibrated for 7 to 10 days. At the end of the equilibrium period, 40 cc of saturated potassium permanganate was added and the pH adjusted to 8 with sodium hydroxide. The potassium permanganate was reduced using a slight excess of sodium bisulfite. The hydrated manganese oxide was collected on a 1 micron cotton filter by continually recirculating the sample at 12 L/min through the filter for 25 min. Recirculation had the advantage of keeping the water vigorously stirred as it was continually passed through the manganese oxide bed being collected on the filter. The samples were ashed while wet to avoid rapid combustion. The plutonium analyses were performed according to a procedure developed by Butler (2) and Sanders (3) or by LEF Environmental Analysis Laboratories, Richmond, California. All americium analyses were done by the LFE Laboratories.

SOURCES OF TRANSURANICS

The Savannah River receives transuranics by direct deposition of fallout from nuclear weapons tests, watershed runoff, and discharges from nuclear facilities. The estuary also receives transuranics from the ocean via the movement of salt water some 35 km up the estuary.

Estimates of the total amount of transuranics deposited on the watershed from nuclear weapons tests are based on analyses of soil cores in the Southeastern U.S. These estimates range from 1.5 to about 2.2 mCi/km^2 with $^{238}Pu/^{239,240}Pu$ ratios of about 0.04 to 0.18 (4). If fallout deposition is uniform over the Savannah River watershed, then the inventory is approximately 55 Ci — of which about 1.5 Ci was deposited on the water impoundments. No americium data are available for estimating its inventory in the Southeastern U.S.

Data on plutonium releases from the Savannah River Plant, located 256 km from the mouth of the Savannah River, are available (5). Atmospheric releases have totaled about 3.7 Ci since fuel reprocessing operations began in 1954. Of the 3.7 Ci, about 2 Ci was released in 1955 prior to installation of high efficiency filters on the air exhaust system and about 0.8 Ci in 1969 when a sand filter failed. Currently, atmospheric releases average about 10 mCi per year. Most of the plutonium from SRP operations is probably on site because analyses of soil cores from the plant perimeter and offsite soils have about the same concentration, 1.96 $\pm$0.7 mCi/km^2 at the plant perimeter, 1.81 $\pm$0.58 mCi/km^2 at 160 km, and other values at the same latitude are about 2 mCi/km^2.

Savannah River Plant plutonium releases to surface water were estimated to be about 0.3 Ci for the 20-year period from 1954 to 1974 and were fairly consistent over this interval (6). Until 1971, plutonium was estimated by measuring only gross alpha and by assuming all alpha activity was from plutonium. Since then, plutonium has been analyzed for specifically. The water after release is subjected to cleanup by onsite streams (about 16 km in length), an onsite swamp, and by the Savannah River before it reaches the estuary.

The fate of ^{137}Cs released to surface water by SRP has been extensively studied and can be used to estimate the fate of Pu released to surface waters. Plutonium and cesium have similar transport properties in most environmental systems due to their strong association with the very fine suspended solids in stream water and with stream bed sediments (7). Some 500 Ci of ^{137}Cs has been discharged to effluent streams and only 90 Ci (about 18%) has been measured at Highway 301, Figure 1. It is estimated that about 58% of the 500 Ci of ^{137}Cs that has been discharged has been deposited in SRP streams prior to reaching the onsite swamp, and the swamp is estimated to contain about 120 Ci or about 24% of cesium that has been discharged (8). If this ^{137}Cs data is extrapolated to plutonium, about 0.054 Ci plutonium is estimated to have left the SRP site since startup.

Using the above data, the total amount of plutonium on the watershed is estimated to be about 59 Ci.

RESULTS AND DISCUSSION

Savannah River

Plutonium concentrations in the Savannah River water are lower than would be predicted by considering the concentration in other fresh waters. Concentrations measured for three months in the Savannah River at Highway 301 (Figure 1) varied from 0.13 to 0.32 fCi/L. In comparison, Lake Michigan contains 0.6 fCi/L (9), the Great Miami River in Ohio about 1 fCi/L (10), and the Neuse and Newport Rivers in North Carolina contain about 1.2 and 1.7 fCi/L (6), respectively. Concentrations in the Savannah River appear to be greatly influenced by reservoirs. Sedimentation in the two large reservoirs on the Savannah River should remove all except some of the very small Pu-bearing particles. About 73% of the Savannah flow originates above Clark Hill Dam, Figure 1. Erosion is greater in the hilly piedmont region above the reservoirs than in the coastal plains so that removal of Pu-bearing particles from the watershed below the reservoirs would be less rapid than above the reservoirs. In contrast, the Great Miami River of Ohio and the Neuse and Newport Rivers of North Carolina do not have large water impoundments on them. Also, the percentage of the watershed which is under cultivation is only one-third as large for the Savannah as for the Great Miami.

The calculated rate of plutonium removal from the Savannah River watershed is about 0.1 as large as the rate calculated for the Great Miami River. Based on the three month average plutonium concentration and measured flow rates (see Table 1), the estimated plutonium transport in the Savannah River at Highway 301 is 0.22 mCi/month or 2.6 mCi/year. The area of the Savannah River watershed above Highway 301 is 81% of the total watershed. So the amount of plutonium in the watershed above the sampling point is 0.81 x 55 Ci from nuclear weapons fallout plus 4 Ci released by SRP or a total of 48.6 Ci. Thus, the annual removal rate is approximately 0.005%. The value reported for the 1400 km^2 watershed of the Great Miami River is 0.05% (10).

Information concerning the transport and fate of americium in rivers and estuaries is limited. The concentration of ^{241}Am in Savannah River water has not been accurately determined, but from a few samples collected at Highway 301 - it is about 0.05 fCi/L as compared with $^{239,240}Pu$ concentration of 0.25 fCi/L. The

^{241}Am concentration is the same as in the Mediterranean (11) and Lake Michigan (12) water where ^{241}Am concentration are 3 to 5% of the $^{239,240}Pu$ concentration. If the same percentage existed in Savannah River water, the concentration of ^{241}Am would be only about 0.01 fCi/L.

Savannah River Estuary

The Savannah River estuary does not have plutonium concentrations much different than other estuaries in the Southeastern U.S., in fact the concentrations are lower than in some. $^{239,240}Pu$ water concentrations were determined in the Neuse and Newport River estuaries of North Carolina for comparison with concentrations in the Savannah River estuary. The results (Figure 3) show that the concentrations in the Newport estuary are about three times greater than in the Neuse or Savannah estuaries which are about equal.

The three estuaries and the rivers supplying them are quite different. The Neuse and Savannah Rivers flow through both the piedmont plateau and Atlantic coastal plains. Only the Savannah has its flow regulated by reservoirs. The volume of flow in the Savannah River is about twice that of the Neuse River and ten times that of the Newport River. The Newport estuary is extremely small and shallow with depths of less than 1 m at mean low water as compared with at least 4 m in the other two. Suspended solid (5 micron fraction) concentrations in the Newport estuary are about one and one-half times greater than in the Savannah or Neuse estuary (6) and may be due to shallow water in the Newport which could resuspend bottom sediments throughout its depth. These sediments are likely to be very fine since the Newport River flows entirely in the coastal plains where the slope is small. The higher plutonium concentration in the Newport estuary could be due to the larger quantity of suspended solids.

Within the Savannah River estuary the plutonium concentrations in the sediment from the tidal fresh water region and near the mouth of the estuary were comparable (Table 2). The values are not greatly different from other locations that only received transuranic input from nuclear weapons fallout. Plutonium cencentrations have been reported for Great Lake sediments [up to 200 fCi/gm] (9), Atlantic coastal waters like Buzzard's Bay [about 60 to 70 fCi/gm] (13), and previous values in the Savannah River system [about 10 to 30 fCi/gm] (6). Fallout ^{238}Pu to $^{239,240}Pu$ ratios are generally less than 0.1. If ratios are greater than this, it is usually indicative of other sources of plutonium in the system. The ratios for the Savannah River estuary cores are reported in Table 2 and only in the fresh water core in the upper 0 to 15 cm were ratios found to be different from fallout. These ratios were about a factor of two greater than fallout ratios and presumably resulted from SRP releases to the river system.

If americium dynamics are different in estuaries than in fresh or sea water systems, this difference would be evidenced from the ^{241}Am to $^{239,240}Pu$ ratios. The average value for such ratios in shallow near-shore sediments (13) and in Lake Michigan sediments (9) varies from 0.14 to 0.34, with an average of 0.22; and no fractionation between americium and plutonium has been found in these sediments, even when the radionuclides are being lost from the sediment following upward migration (13). With the exception of one value of 0.66, the ^{241}Am to $^{239,240}Pu$ ratios for two sediment cores reported in Table 1 are not significantly different from those quoted in the literature. The indication is that the chemistries of americium and plutonium are similar in this estuarine system and that the ^{241}Am has grown in from ^{241}Pu.

The transuranic alpha activity in these cores represents less than 1% of the gross alpha activity from the natural radionuclides that are present. Indeed, modern civilization's impact on the alpha activity of these cores is small compared to the natural background.

Seafood

At present plutonium levels, seafoods make a very minor contribution to the overall radiation dose commitment to the populations in the Southeast. Seafood samples that represent all trophic levels consumed by people in the Southeastern United States were collected in and near the Savannah River estuary and analyzed for plutonium. The plutonium concentrations decreased as expected from the molluscs to the fish, with the oyster having the highest concentration, 0.12 fCi/kg, compared to 0.001 fCi/kg for shad

(Table 3). The 50-year bone dose commitment from consuming 5.9 kg per year of oysters is less than 0.0004% of the annual background radiation dose of about 120 mrem that is received by man in the Southeastern U.S.

CONCLUSIONS

Nuclear facilities operating on the Savannah River watershed have contributed less than 10% as much plutonium to the watershed as has nuclear weapons fallout. Transport of plutonium from the watershed to the estuary is very slow and appears to be influenced by two large reservoirs which serve as sinks for suspended plutonium-bearing particles. Consequently, plutonium concentrations in Savannah River water and estuary sediments are no higher — and in some cases lower — than plutonium concentrations in other rivers and estuaries on which there are no nuclear facilities.

At present plutonium levels, seafoods make a very minor contribution to the overall dose commitment to population in the Southeastern U.S.

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TABLE 1

Plutonium Transport in the Savannah River

Sampling Period	River Flow, L/Period	$^{239,240}Pu$, fCi/L	$^{239,240}Pu$ Transport, mCi/Sampling Period
6/8 to 7/7, 1976	1.25×10^{12}	0.13 ±0.09	0.16
7/7 to 8/4, 1976	1.20×10^{12}	0.27 ±0.08	0.32
8/5 to 9/7, 1976	7.31×10^{11}	0.26 ±0.11	0.19
Average	1.06×10^{12}	0.22	0.22

TABLE 2

Plutonium, Americium, and Gross Alpha Activities in the Savannah River Estuary (all are dry weights)

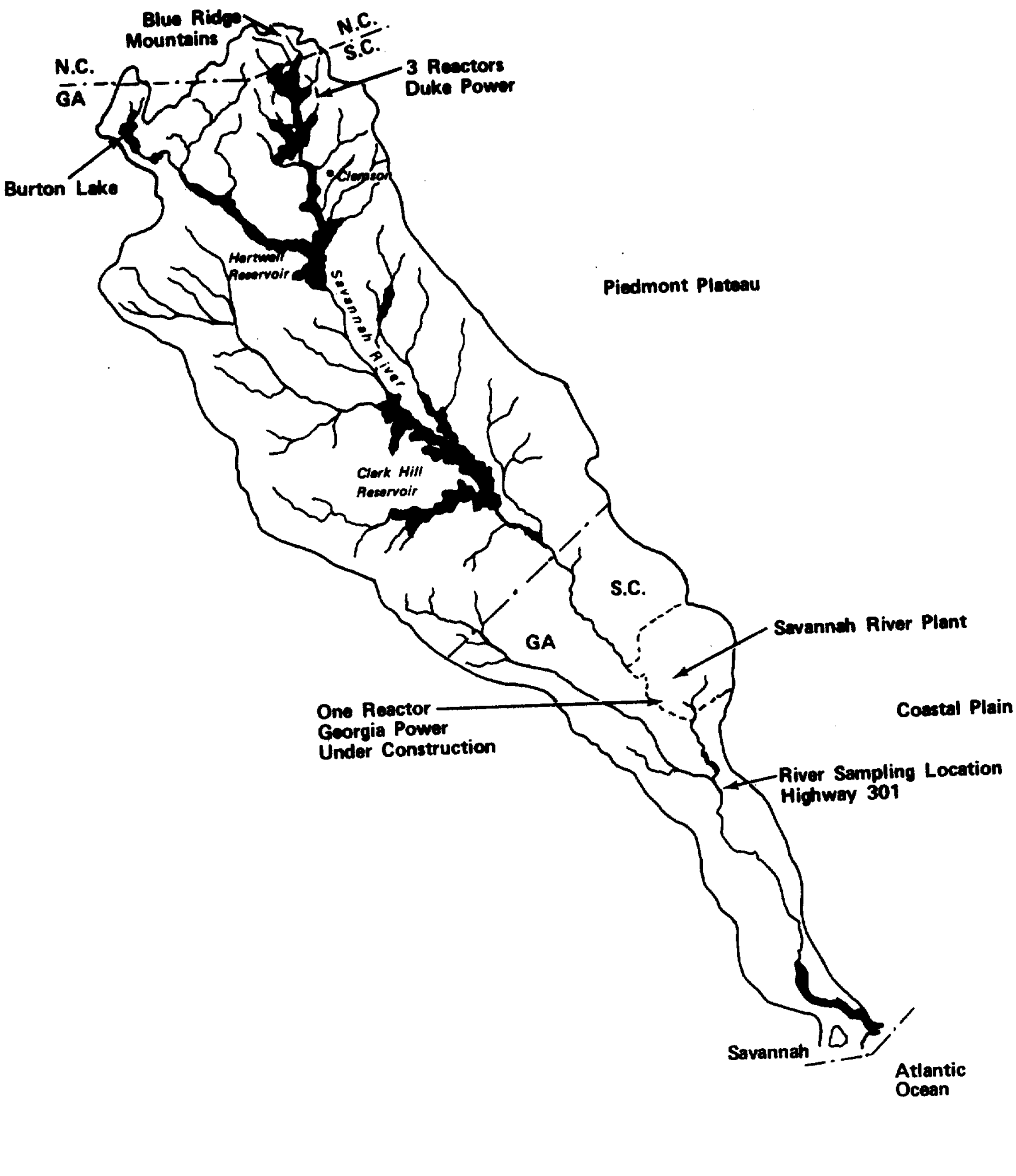
Location	Core Depth Interval, cm	^{238}Pu, fCi/gm	$^{239,240}Pu$, fCi/gm	^{238}Pu / $^{239,240}Pu$ Ratio	^{241}Am, fCi/gm	^{241}Am / $^{239,240}Pu$ Ratio	Gross Alpha, fCi/gm
Tidal Fresh Water	0 to 5	4.3 ±1.3	27.2 ±3.3	0.16 ±0.05	11.5 ±4.1	0.42 ±0.16	24,000
	5 to 15	6.1 ±1.2	35.5 ±2.8	0.17 ±0.04	4.0 ±1.9	0.11 ±0.05	20,000
	15 to 30	2.8 ±1.3	30.9 ±2.8	0.09 ±0.04	5.4 ±1.9	0.17 ±0.06	20,000
	30 to 50	0.4 ±0.4	10.6 ±1.0	0.04 ±0.04	2.0 ±0.8	0.19 ±0.08	18,000
	50 to 70	0.05 ±0.10	0.05 ±0.05	- -	0.23 ±0.23	- -	18,000
Mouth of Estuary	0 to 5	3.2 ±1.1	50.6 ±4.1	0.06 ±0.02	11.1 ±1.7	0.22 ±0.04	10,000
	5 to 15	1.7 ±1.0	21.6 ±2.2	0.08 ±0.05	3.0 ±2.3	0.14 ±0.11	13,000
	15 to 25	0.2 ±0.2	2.9 ±0.5	0.07 ±0.07	1.8 ±0.6	0.62 ±0.23	12,000
	45 to 65	-	0.5 ±0.5	-	-	-	13,000

TABLE 3

Southeastern Seafood Plutonium Dose Commitments

	pCi/kg Wet Weight[a]	50-Year[a] Bone Dose Commitment, mrem
Oysters	0.12	5.5×10^{-4}
Clams	0.05	2.3×10^{-4}
Crabs	0.007	2.4×10^{-5}
Mullet	0.005	5×10^{-5}
Speckled Trout	0.004	3.9×10^{-5}
Shad	0.001	1.3×10^{-5}

a. Consumption assumed for the dose calculation, 5.9 Kg/year molluscs, 11.8 Kg/year fish.


Blue Ridge
Mountains
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Duke Power
Burton Lake
Clemson
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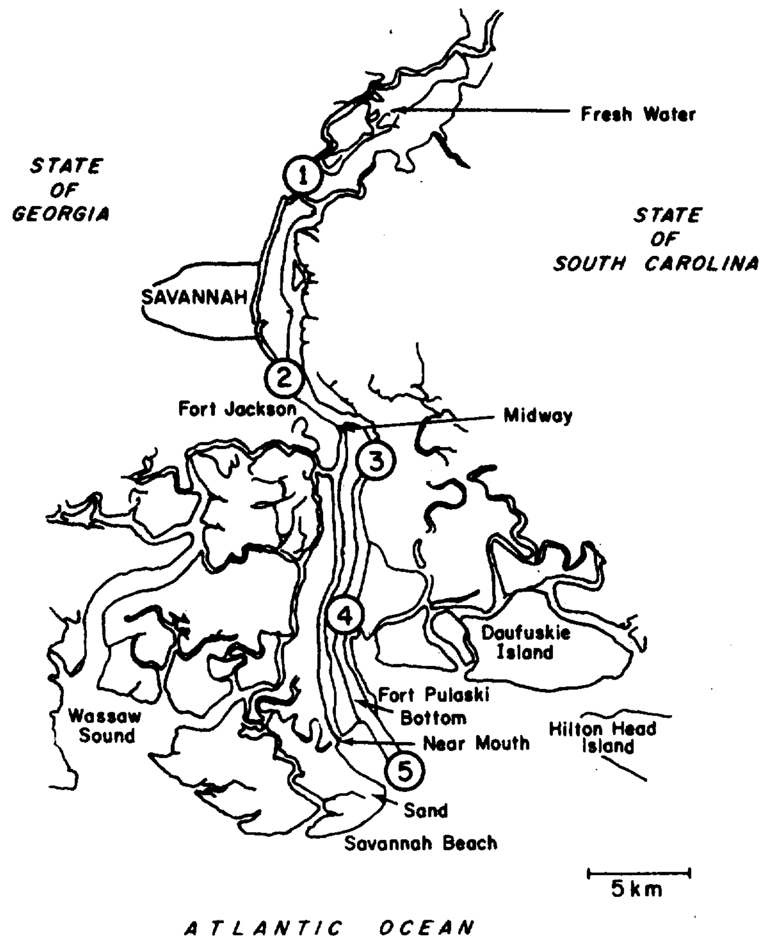
Savannah River
Piedmont Plateau
Clark Hill
Reservoir
S.C.
GA
Savannah River Plant
One Reactor
Georgia Power
Under Construction
Coastal Plain
River Sampling Location
Highway 301
Savannah
Atlantic
Ocean


Fresh Water
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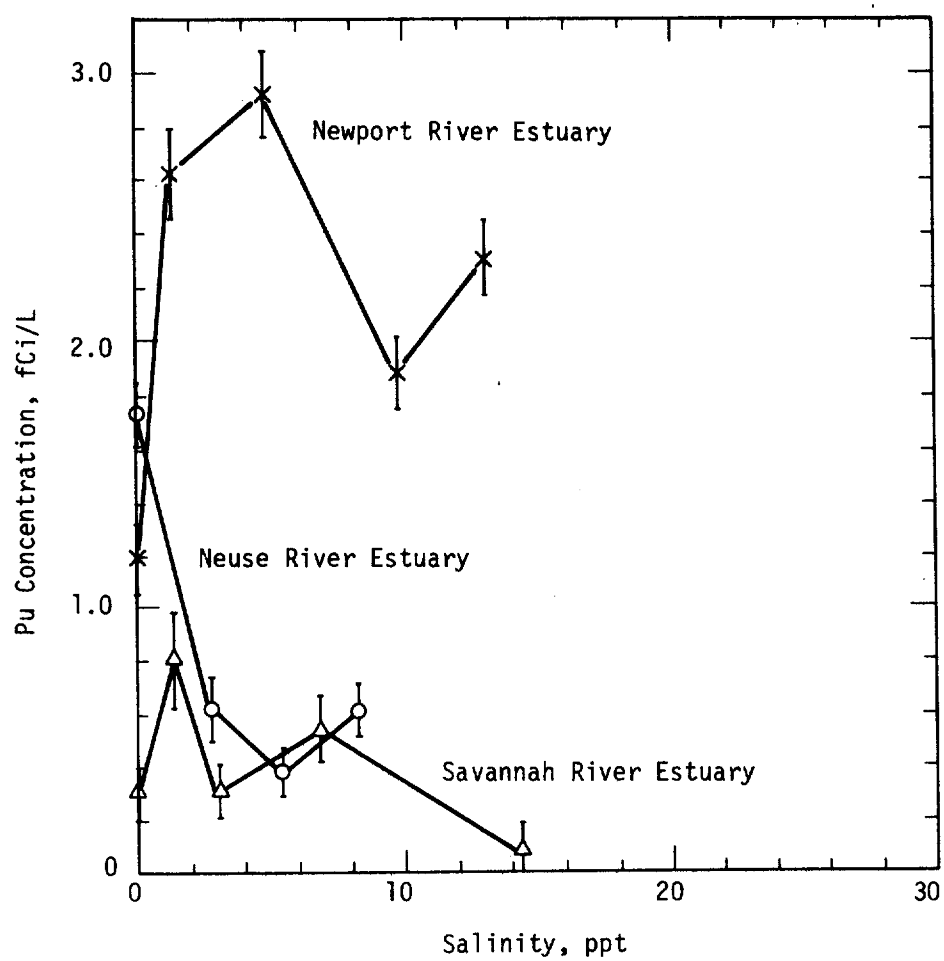

FIGURE 3. Total Plutonium Concentrations in the Waters of 3 Southeastern U.S. Estuaries

CC: D. M. Peek, DOE-SR
R. E. Naylor, Wilm.
C. H. Ice, SRL
J. A. Porter - T. V. Crawford
D. W. Hayes
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TO DISTRIBUTION

Attached is a copy of the following:

DP-MS-78-56, "Plutonium and Americium Behavior in the Savannah River Marine Environment," by D. W. Hayes and J. H. Horton.

This paper is proposed for publication in *Transuranic Elements in the Environment*, a DOE publication.

If there are comments about its release notify the TIS office (Ext. 3598).

For any technical clarification, we suggest you call:

T. V. Crawford, Research Manager
Environmental Transport Division
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