COO-2529-10

PLUTONIUM AND CESIUM RADIONUCLIDES IN THE HUDSON RIVER ESTUARY AND OTHER ENVIRONMENTS

Annual Technical Progress Report

H.J. Simpson and R.M. Trier

Principal Investigators

Lamont-Doherty Geological Observatory

of

Columbia University

Palisades, New York

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DISCLAIMER

TABLE OF CONTENTS

SUMMARY

PLUTONIUM AND CESIUM RADIONUCLIDES IN THE HUDSON RIVER ESTUARY AND OTHER ENVIRONMENTS

The major purpose of this project is to describe the behavior and distribution of plutonium and other radionuclides in the Hudson River estuary and to establish the important transport pathways for transuranics and other radionuclides of interest in the nuclear fuel cycle in estuarine environments. To accomplish this goal a substantial number of cores, grab samples, suspended particles and water samples have been collected throughout the salinity range of the Hudson, and in a variety of depositional environments.

The possibility of transuranic release from the reactor site at Indian Point has been and will continue to be explored, and the distribution of reactor-released gamma-emitting nuclides has been and will be employed to obtain a better understanding of the estuarine sediment transport and accumulation patterns which control the distribution of $^{239,240}Pu$ within the system. The feasibility of using ^{238}Pu to $^{239,240}Pu$ ratios as an indicator of transuranic reactor releases has been established and will be explored in more detail. Measurements in other aqueous environments, such as the Delaware estuary, near two other reactors in coastal New Jersey and Connecticut, and in lakes of unusual chemical environments valuable in radionuclide chemical speciation studies, have been made and will be made to help describe the most important environmental transport pathways of transuranics and other radionuclides in natural waters.

PLUTONIUM, CESIUM AND URANIUM SERIES RADIONUCLIDES IN THE HUDSON RIVER ESTUARY AND OTHER ENVIRONMENTS

ABSTRACT

We have measured radionuclide activites in a large number of sediment cores and suspended particle samples throughout the salinity range of the Hudson River estuary. Activities of ^{137}Cs, ^{134}Cs and ^{60}Co determined by gamma spectrometry and $^{239,240}Pu$ and ^{238}Pu determined by alpha spectrometry indicate reasonably rapid accumulation rates in the sediments of marginal cove areas, and very rapid deposition in the harbor region adjacent to New York City. General distributions of ^{137}Cs and $^{239,240}Pu$ are similar in surface sediments and with depth in cores, but there are deviations from the fallout ratio due to (1) addition of ractor ^{137}Cs and (2) loss of ^{137}Cs from the particle phases at higher salinities. Measurable amounts of reactor-derived ^{134}Cs and ^{60}Co are found in nearly all sediment samples containing appreciable ^{137}Cs between 15 Km upstream of Indian Point and the downstream extent of our sampling about 70 Km south of the reactor. Accumulations of $^{239,240}Pu$ in New York harbor sediments are more than an order of magnitude greater than the fallout delivery rate, probably primarily due to the accumulation of fine particles containing fallout plutonium in the harbor which have been transported from upstream areas of the Hudson. Depth profiles of radionuclides and variations of activities with particle size at low salinities in the Hudson indicate the importance of organic phases, including large flocculent particles greater than 180μ, in binding plutonium, and no evidence of significant chemical migration within the sediments. Measurements of water column fallout $^{239,240}Pu$ in a saline lake

with a high carbonate ion concentration yielded activities about two orders of magnitude greater than has been found for fallout plutonium in other continental waters, indicating extensive mobility in some natural water environments.

PLUTONIUM AND CESIUM RADIONUCLIDES IN THE HUDSON RIVER ESTUARY AND OTHER ENVIRONMENTS

DOE-DIRECTED ACCOMPLISHMENTS

The primary area of our field research, the Hudson River estuary, is a system which is heavily used for energy generating activities at present and promises to be utilized to an even greater extent in the future. From our studies of the present distribution of radionuclides released over a number of years from Indian Point, we have established that an appreciable amount of fine-grained sediments bearing reactor nuclides accumulate in New York harbor more than 60 Km downstream of the release area. Thus any monitoring program for Indian Point releases should be designed with those findings incorporated. The large inventory of $^{239,240}Pu$ in the harbor sediments, although present in depth integrated amounts about an order of magnitude greater than total fallout delivery, was probably derived from fine-grained sediments containing fallout plutonium from a considerable area upstream, which was moved downstream toward the harbor and deposited after many episodes of sedimentation and resuspension.

Depth profiles of radionuclides and variations of activities with particle size at low salinities in the Hudson indicate the importance of organic phases, including large flocculent particles greater than 180 microns, in binding plutonium, and no evidence of significant chemical migration within the sediments.

Several zones of rapid sediment accumulation in the tidal fresh water reach of the Hudson have been identified which could be expected to accumulate

radionuclides if significant sources in addition to fallout were to be present in the future. Thus, the pattern of large variations in radionuclide accumulation rates which we have demonstrated for the saline intruded reach of the Hudson is also typical of the fresh water tidal reach, and should be incorporated explicitly in any radionuclide monitoring program for that portion of the Hudson in the future.

Measurements of water column fallout $^{239,240}Pu$ in a saline lake with a high carbonate ion concentration yielded activities about two orders of magnitude greater than has been found for fallout plutonium in other continental waters, indicating extensive mobility of this element in some natural water environments.

Findings of our research related to the Delaware estuary and the sediments near the Oyster Creek and Millstone Point reactors are discussed in last years summary of ERDA-Directed Accomplishments (COO-2529-3).

PLUTONIUM AND CESIUM RADIONUCLIDES IN THE HUDSON RIVER ESTUARY AND OTHER ENVIRONMENTS

H.J. Simpson and R.M. Trier

I. INTRODUCTION

This report summarizes our activities during the period December 1, 1977 to November 30, 1978 under contract EY-76-S-02-2529. A comprehensive data listing of radionuclide measurements made during the first three years of this contract was given in our last annual report (COO-2529-3). Here we will provide a compilation of data collected during the last year and a discussion of our most significant findings during that period.

One of our primary research goals up to now has been to develop a better understanding of the behavior of transuranics, especially plutonium, in natural aqueous environments. We began by attempting to establish the present distribution of plutonium in the sediments of the Hudson River estuary. The plutonium was assumed to be predominantly derived from fallout from nuclear weapons testing, with the primary input to the Hudson by precipitation occurring during the early to mid 1960's. The problem of establishing the distribution of fallout radionuclides in Hudson sediments is made more complex by the presence of a nuclear generating station at Indian Point, approximately sixty kilometers upstream of New York Harbor.

Most of the data we have collected up to now has been for samples of sediment from the Hudson estuary. We are attempting to use this data, plus that for sediments and water samples from other natural water environments to help understand the transport and accumulation processes which govern the behavior of radionuclides, especially transuranics in natural water systems.

II. SUMMARY OF REPORTS DISCUSSING OUR RESULTS TO DATE

Some of our conclusions have been presented in three previous annual technical progress reports:

COO-2529-1 Plutonium and Cesium Radionuclides in the Hudson River Estuary (Dec. 1, 1974-Nov. 30, 1975).

COO-2529-2 Plutonium and Cesium Radionuclides in the Hudson River Estuary (Dec. 1, 1975-Nov. 30, 1976).

COO-2529-3 Plutonium and Cesium Radionuclides in the Hudson River Estuary (Dec. 1, 1976-Nov. 30, 1977).

In addition to these annual technical progress reports, several appendices to those reports and related publications have summarized findings by our laboratory resulting from research carried out under this contract. A listing of these reports and publications are provided below, with report numbers assigned to conform with the procedure used for our earlier technical progress reports. Most of the reports listed below have been provided previously, and are not included as part of this document. This list will be updated in the future, with publication citations provided as information is reported in the literature.

COO-2529-4 Man-made Radionuclides and Sedimentation in the Hudson River Estuary, H.J. Simpson, C.R. Olsen, R.M. Trier and S.C. Williams, Science 194, 179-183, 1976.

COO-2529-5 A Geochemical Analysis of the Sediments and Sedimentation in the Hudson Estuary. C.R. Olsen, H.J. Simpson, R.F. Bopp, S.C. Williams, T.H. Peng and B.L. Deck, Journal of Sedimentary Petrology 48, 401-418, 1978.

COO-2529-6 Sources of Heavy Metals in Sediments of the Hudson River Estuary, S.C. Williams, H.J. Simpson, C.R. Olsen and R.F. Bopp, Marine Chemistry 6, 195-213, 1978.

COO-2529-7 Cesium-137 as a Tracer for Reactive Pollutants in Estuarine Sediments, H.J. Simpson, R.F. Bopp, C.R. Olsen, R.M. Trier and S.C. Williams, USSR-USA Symposium and Report on Marine Pollution, (in press), 1978.

COO-2529-8 (and COO-2185) Reactor-Released Radionuclides and Fine-Grained Sediment Transport and Accumulation Patterns in Barnegat Bay, New Jersey and Adjacent Shelf Waters, C.R. Olsen, P.E. Biscaye, H.J. Simpson, R.M. Trier, N. Kostyk, R.F. Bopp, Y.-H. Li and H.W. Feely.

COO-2529-9 Transport of Plutonium by Rivers, H.J. Simpson, R.M. Trier and C.R. Olsen, to be included as part of the DOE/DBER publication Transuranic Elements in the Environment, TID-22800 (ed. W.C. Hanson).

III. TRANSPORT AND ACCUMULATION OF RADIONUCLIDES IN HUDSON ESTUARY SEDIMENTS

Deposition of sediments in the Hudson occurs in a complicated pattern, which is both time dependent and highly variable in rate. As a result, mapping the distribution of plutonium in the sediments requires measurement of a large number of samples (Figure 1). We have chosen to attack this problem by measurement of other man-made radionuclides in conjunction with plutonium. There are several nuclides in Hudson sediments derived from fallout (^{137}Cs) and reactor releases (^{137}Cs, ^{134}Cs and ^{60}Co) which we have analyzed by gamma counting dried sediment samples with no chemical separations employed. We have found these nuclides to be very effective as indicators of the levels of plutonium in Hudson sediments. Using direct gamma counting as out initial mapping approach, we have been able to make rapid progress in describing the present distribution of plutonium in the sediments of a large, complicated estuarine environment.

The activity of $^{239,240}Pu$ per gram of sediment varies greatly from place to place in the Hudson. The relative amounts of $^{239,240}Pu$ and ^{137}Cs have much smaller variation than the absolute amounts of either. As discussed in our last annual report (COO-2529-3) the total reported range of $^{239,240}Pu$ to ^{137}Cs ratios in Hudson sediment is 1 - 14% with most samples falling in the range of 3-10%.

A. Radionuclide accumulation trends in the lower Hudson estuary

One of the piston cores we collected from the upper Bay (P-1.5E) penetrates through a very thick sequence of sediments containing ^{137}Cs (Figure 2). Reactor-derived ^{134}Cs and ^{60}Co are apparently confined to the upper half of the core (above 100-140 cm), where a broad peak in ^{137}Cs occurs.

FIGURE 1. Locations of Hudson estuary cores: numbers indicate miles upstream (or downstream for negative numbers) of the southern tip of Manhattan; letters indicate direction from center of channel (east (E), west (W), middle (M), eastern cove (EC)).

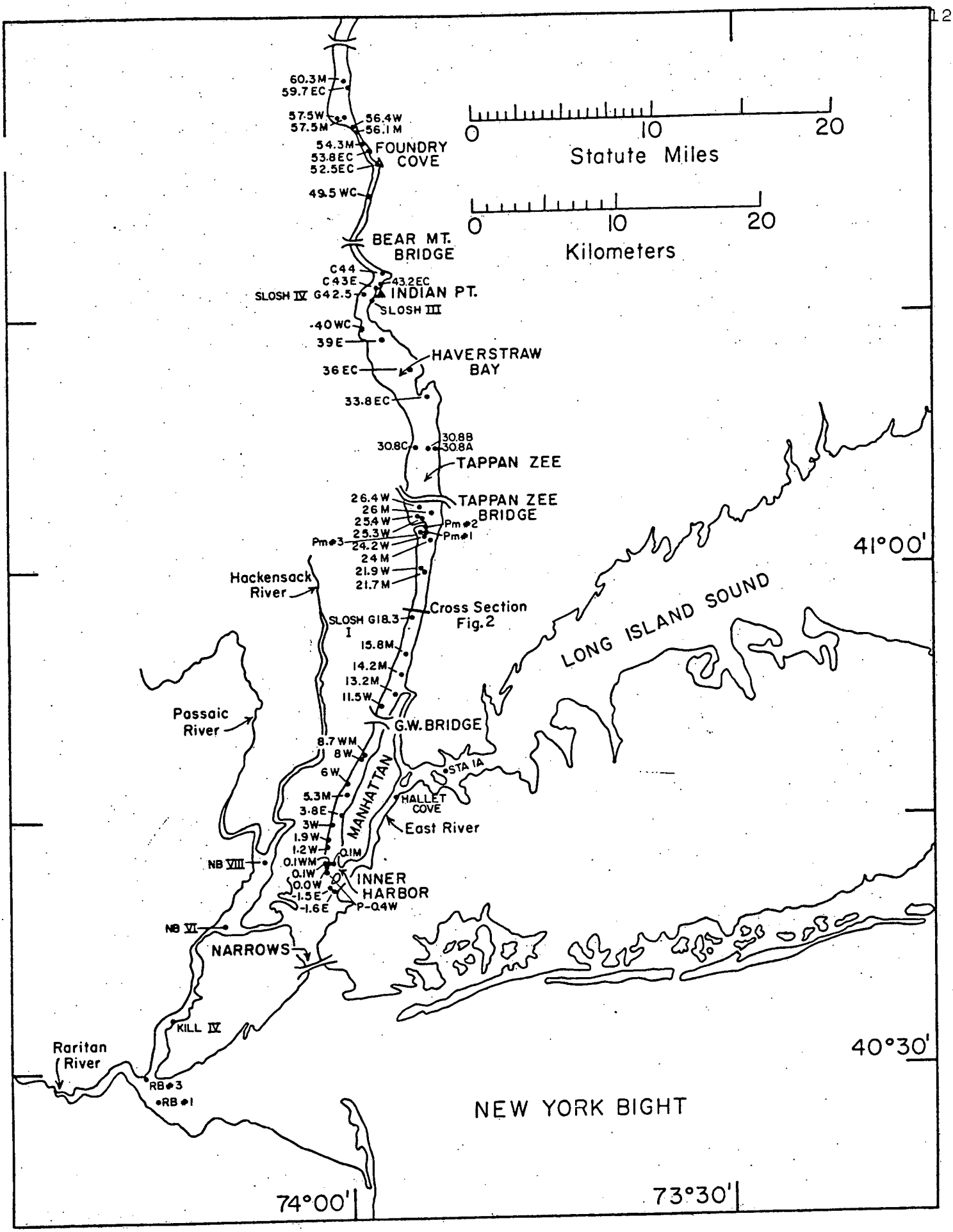
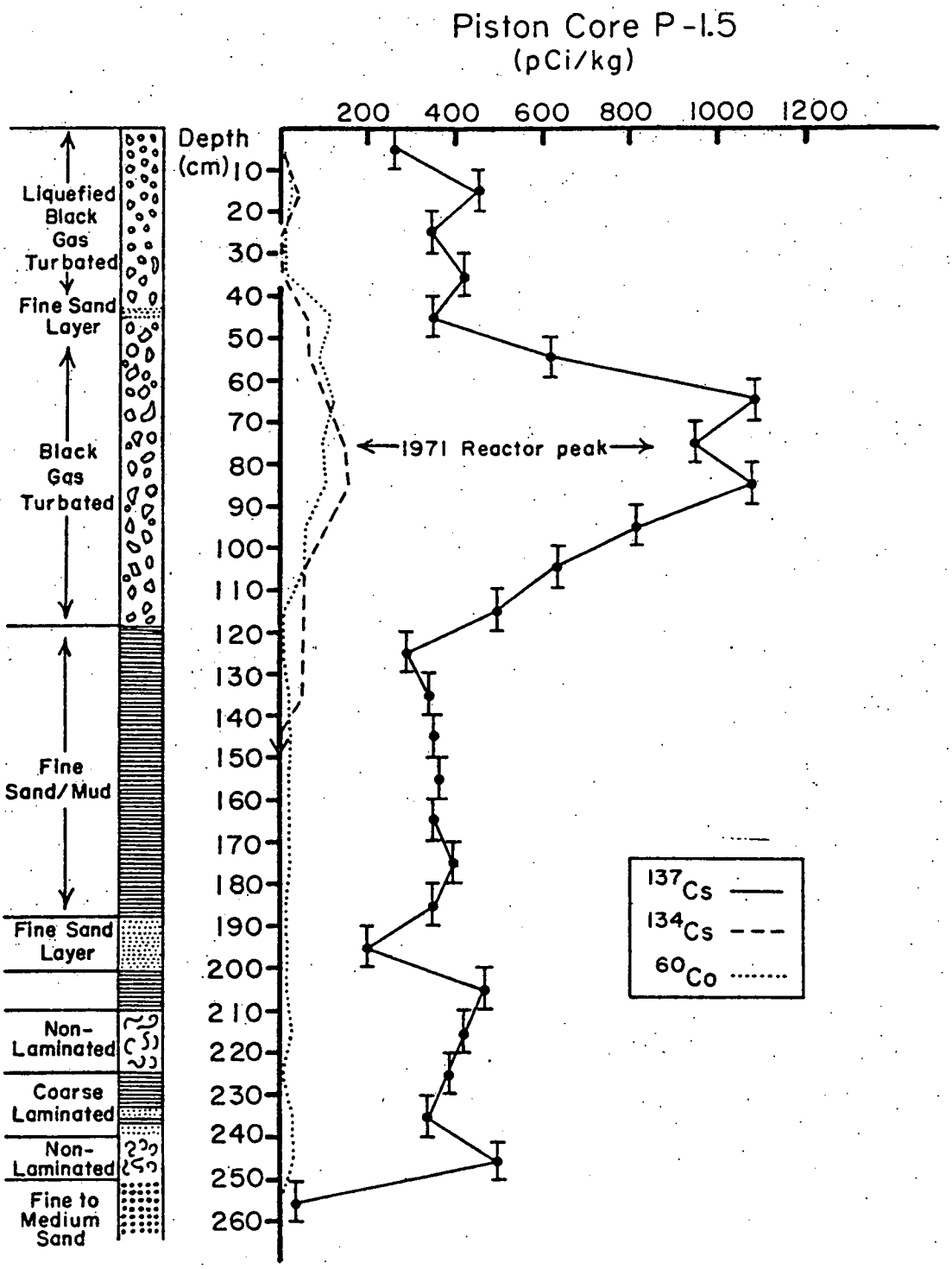

60.3M
59.7EC
57.5W
57.5M
56.4W
56.1M
54.3M
53.8EC
52.5EC
FOUNDRY COVE
49.5WC
0
10
20
Statute Miles
0
10
20
Kilometers
BEAR MT. BRIDGE
C44
C43E
43.2EC
SLOSH IV
G42.5
INDIAN PT.
SLOSH III
40WC
39E
HAVERSTRAW BAY
36EC
33.8EC
30.8C
30.8B
30.8A
TAPPAN ZEE
26.4W
26M
25.4W
25.3W
Pm#2
Pm#1
Pm#3
24.2W
24M
21.9W
21.7M
TAPPAN ZEE BRIDGE
Hackensack River
SLOSH I
G18.3
Cross Section Fig.2
15.8M
14.2M
13.2M
11.5W
G.W. BRIDGE
LONG ISLAND SOUND
41°00'
Passaic River
8.7WM
8W
6W
5.3M
3.8E
3W
1.9W
1.2W
0.1M
0.1WM
0.1W
0.0W
-1.5E
-1.6E
P-0.4W
NB VIII
NB VI
NARROWS
INNER HARBOR
MANHATTAN
STA 1A
HALLET COVE
East River
KILL IV
Raritan River
RB#3
RB#1
NEW YORK BIGHT
40°30'
74°00'
73°30'

FIGURE 2. ^{137}Cs, ^{134}Cs, and ^{60}Co as a function of depth in a piston core collected from New York harbor (upper New York Bay) at mile point - 1.5: the broad peak in all three nuclides is believed to correlate with the period (1971) of maximum releases of reactor nuclides from Indian Point. The ^{137}Cs in the lower half of the core is believed to be derived predominantly from global fallout.


Piston Core P-1.5
(pCi/kg)
200 400 600 800 1000 1200
Depth
(cm)
10 20 30 40 50 60 70 80 90 100 110 120 130 140 150 160 170 180 190 200 210 220 230 240 250 260
Liquefied Black Gas Turbated
Fine Sand Layer
Black Gas Turbated
Fine Sand/Mud
Fine Sand Layer
Non-Laminated
Coarse Laminated
Non-Laminated
Fine to Medium Sand
←1971 Reactor peak→
137Cs
134Cs
60Co

We believe this ^{137}Cs peak reflects the period of maximum reactor releases, while the sediments below contain ^{137}Cs derived predominantly from fallout. The trend of $^{239,240}Pu$ with depth (Figure 3) in the same core is quite different than for ^{137}Cs. There is no evidence of a peak in $^{239,240}Pu$ for correlating with the peak in ^{137}Cs. We interpret this as indicating that if releases of $^{239,240}Pu$ have occurred from Indian Point during the same period as the maximum releases of ^{137}Cs, they were too small to be observable in the sediments of New York Harbor in the presence of the burden of fallout $^{239,240}Pu$ accumulating there. The ratio of ^{238}Pu to $^{239,240}Pu$ (Table 1) in the same harbor core samples are shown in Figure 4. The samples near the bottom of the core have lower amounts of ^{238}Pu relative to $^{239,240}Pu$ than those nearer the upper half of the core. At present, we do not believe that definite conclusions can be drawn about the sources of ^{238}Pu in New York harbor sediments.

After our initial survey of plutonium distributions in Hudson sediments in which the zone of major accumulation was discovered to be New York harbor, we concentrated much of our effort on harbor sediment analysis to obtain better estimates of the total inventory of radionuclides in that area of the estuary. From data obtained for several other harbor cores (-1.6E, -1.5E, 0.1W, 0.1WM, 1.9W-COO-2529-3, Table 2; and P-0.4W listed in Table 12 of this report) we have clearly demonstrated that the extremely high rate of accumulation of recent sediment containing fallout $^{239,240}Pu$ shown in Figure 3 is indicative of conditions for a substantial area of the harbor. There is now little doubt that this area is one of the major zones of recent sediment accumulation in the Hudson estuary, and represents a significant portion of the total inventory of fallout $^{239,240}Pu$ present in the sediments

FIGURE 3. $^{239,240}Pu$ as a function of depth in the same piston core shown in Figure 2. There is no evidence of an increase in $^{239,240}Pu$ in the depth range (50-120 cm) which we believe correlates with the period (1971) of maximum reactor releases of gamma-emitting nuclides.

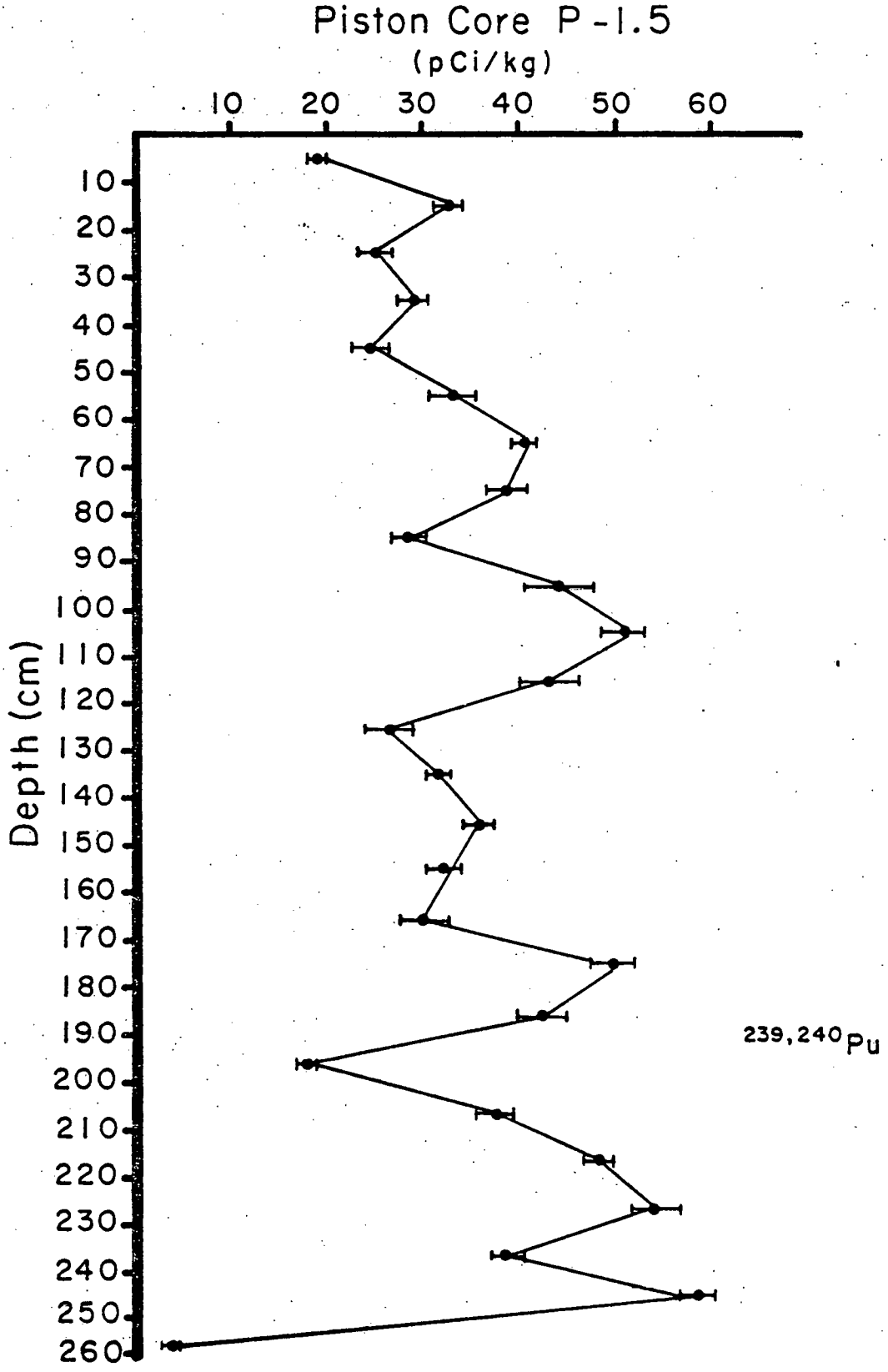
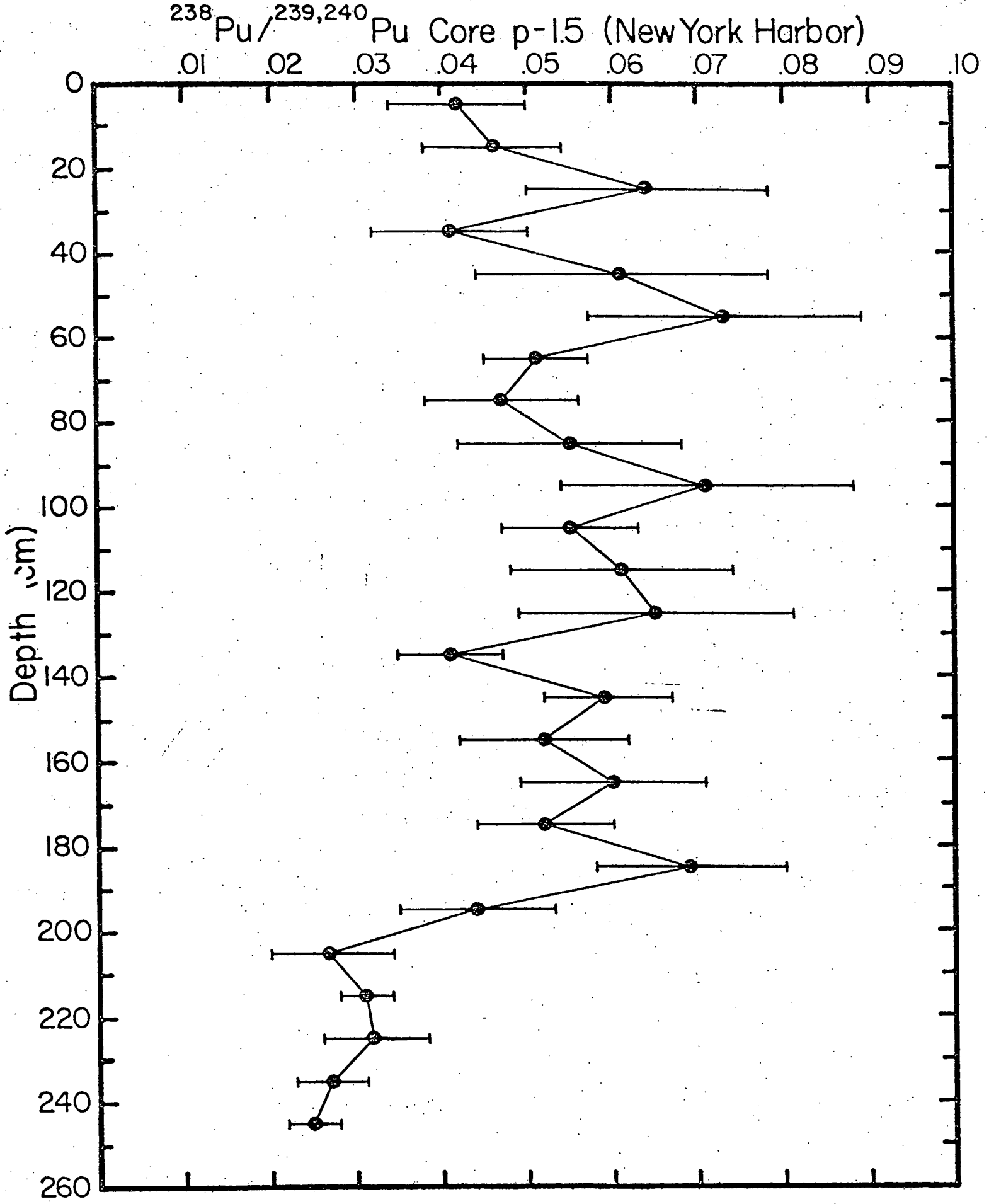

Piston Core P-1.5
(pCi/kg)
10
20
30
40
50
60
Depth (cm)
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
190
200
210
220
230
240
250
260
239,240Pu

TABLE 1. Plutonium Isotopes in Piston Core - 1.5

Location (Mile Point)	Sample No.	Depth (cm)	Dry Wt. (gm)	% Yield	$^{239,240}Pu$ (pCi/Kg)	^{238}Pu (pCi/Kg)	$^{238/239,240}Pu$ (pCi/Kg)
P - 1.5	1048A	0-10	50.7	44.2	19.2±0.9	0.8±0.2	0.042±.008
	B	10-20	38.5	25.3	33.0±1.5	1.5±0.3	0.046±.008
	C	20-30	43.4	20.0	25.0±1.8	1.6±0.3	0.064±.014
	D	30-40	45.5	24.9	29.3±1.6	1.2±0.3	0.041±.009
	E	40-50	41.7	15.2	24.4±2.0	1.5±0.4	0.061±.017
	F	50-60	37.2	19.0	33.3±2.4	2.4±0.5	0.073±.016
	G	60-70	38.3	62.8	40.9±1.4	2.1±0.2	0.051±.006
	H	70-80	40.7	14.6	38.5±2.0	1.8±0.3	0.047±.009
	I	80-90	33.8	25.0	28.9±1.9	1.6±0.4	0.055±.013
	J	90-100	42.1	12.3	43.9±3.6	3.1±0.7	0.071±.017
	K	100-110	38.2	32.4	51.3±2.3	2.8±0.4	0.055±.008
	L	110-120	42.2	15.7	42.8±3.0	2.6±0.5	0.061±.013
	M	120-130	57.5	12.4	26.2±2.2	1.7±0.4	0.065±.016
	N	130-140	52.9	41.0	32.0±1.3	1.3±0.2	0.041±.006
	O	140-150	52.1	20.7	35.7±1.5	2.1±0.3	0.059±.008
	P	150-160	42.7	32.0	32.5±1.8	1.7±0.3	0.052±.010
	Q	160-170	65.9	22.0	29.9±1.9	1.8±0.3	0.060±.011
	R	170-180	53.7	25.0	50.3±2.3	2.6±0.4	0.052±.008
	S	180-190	51.0	17.6	42.3±2.5	2.9±0.4	0.069±.011
	T	190-200	63.7	17.8	18.0±1.0	0.8±0.2	0.044±.009
	U	200-210	59.6	16.7	37.4±2.0	1.0±0.2	0.027±.007
	V	210-220	46.7	45.9	48.2±1.4	1.5±0.2	0.031±.003
	W	220-230	50.5	10.2	53.9±2.5	1.7±0.3	0.032±.006
	X	230-240	76.0	15.2	38.5±1.6	1.0±0.2	0.027±.004
	Y	240-250	39.3	65.6	59.2±1.8	1.5±0.2	0.025±.003
	Z	250-260	84.0	16.2	4.2±0.4	--	--

FIGURE 4. Ratios of ^{238}Pu to $^{239,240}Pu$ in New York harbor core P - 1.5.


$^{238}Pu/^{239,240}Pu$ Core p-1.5 (New York Harbor)
.01
.02
.03
.04
.05
.06
.07
.08
.09
.10
0
20
40
60
80
100
120
140
160
180
200
220
240
260
Depth (cm)

of the Hudson.

B. Radionuclide accumulation near the upstream end of the salinity intrusion in the Hudson

Although the harbor is of major interest for budget calculations, and as an indicator of the rate of transport of recent sediments along the axis of the estuary, the combination of rapid sediment accumulation rates and dredging activities make it difficult to interpret vertical distributions of plutonium in terms of possible mobility in the sediments. To examine the importance of this type of process in the Hudson, we have turned to areas in which recent sediments are accumulating more slowly than in the harbor, and where the possibility of disturbance by dredging activities is considerably less. One area for which we have considerable information is Foundry Cove, a small embayment in the Hudson at mile point 54, about 18 kilometers upstream of the reactor site at mile point 43. From detailed studies of the trace metal and radionuclide distribution in a number of cores from that cove (COO-2529-3, Table 1) we have shown that recent (last two decades) sediments are confined to approximately the upper 20-30 cm. We collected three cores from within a few centimeters of each other in Foundry Cove and sectioned each of them into 1 cm intervals. These samples were then gamma counted to determine if the ^{137}Cs activity in the depth sections were comparable in all three cores. When the activities had been shown to be within the counting error of each other for all three cores, the sections were combined for each depth interval, and then counted for longer periods to obtain the data reported in Table 2. After the gamma-emitting nuclides were measured, each of the depth segments were analyzed for plutonium isotopes (Table 3). Nuclide ratios for one centimeter intervals down to 26 cm are given in Table 4. Data from Tables 2 and 3

TABLE 2. Gamma Emitting Radionuclides in Core 54 EC (Foundry Cove)

Locationn (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	Cs^{137} (pCi/Kg)	Cs^{134} (pCi/kg)	Co^{60} (pCi/Kg)	K^{40} (pCi/g)
mp 54 EC	1240A	0-1	61	1530±38	28±13	73±10	18.7±0.6
(March 1977)	B	1-2	66	1610±44	47±16	55±13	17.2±0.7
	C	2-3	75	2000±48	75±16	98±13	20.0±0.7
	D	3-4	62	2400±56	75±16	145±13	18.2±0.6
	E	4-5	57	2240±55	30±16	110±14	19.4±0.7
	F	5-6	62	2060±56	-7±13	89±15	18.2±0.7
	G	6-7	67	2010±48	5±10	67±12	17.8±0.6
	H	7-8	62	2180±54	12±11	83±13	18.2±0.7
	I	8-9	70	2130±54	-7±11	95±13	17.6±0.7
	J	9-10	73	1970±49	-7±10	100±12	17.9±0.6
	K	10-11	70	2120±50	1±10	70±11	17.7±0.6
	L	11-12	72	2130±59	-14±13	41±15	17.8±0.7
	M	12-13	64	2170±57	-13±12	67±14	18.4±0.7
	N	13-14	65	2140±68	-10±16	47±19	18.0±0.8
	O	14-15	69	1990±48	-18±10	36±11	18.4±0.7
	P	15-16	68	1680±55	4±16	43±18	18.3±0.8
	Q	16-17	71	1440±38	2±9	17±10	18.9±0.7
	R	17-18	72	1160±42	-10±14	16±15	17.9±0.7
	S	18-19	69	770±29	-8±11	-4±14	18.0±0.7
	T	19-20	68	580±29	-11±9	-8±10	18.8±0.7
	U	20-21	78	380±21	-5±11	24±13	19.6±0.7
	V	21-22	80	200±18	1±11	13±13	19.7±0.7
	W	22-23	85	105±15	8±10	26±11	19.2±0.7
	X	23-24	83	22±12	8±9	-14±11	18.7±0.6
	Y	24-25	83	19±11	-6±9	8±9	19.5±0.6
	Z	25-26	85	-9±13	-5±10	-6±10	18.7±0.7

TABLE 3. Plutonium Isotopes in Core 54EC (Foundry Cove)

Location (Mile Point)	Sample No.	Depth (cm)	Dry Wt. (gm)	% Yield	$^{239,240}Pu$ (pCi/Kg)	^{238}Pu (pCi/Kg)	$^{238/239,240}Pu$ (pCi/Kg)
mp 54EC	1240A	0-1	51	25.9	27.9±1.4	1.1±0.2	.039±.008
	B	1-2	51	21.9	28.6±1.7	1.4±0.3	.049±.010
	C	2-3	66	15.9	30.9±1.4	1.5±0.2	.049±.008
	D	3-4	52	13.9	34.7±1.9	1.7±0.3	.049±.010
	E	4-5	48	9.2	35.4±1.9	2.2±0.3	.063±.010
	F	5-6	52	8.5	41.4±2.7	2.3±0.5	.056±.012
	G	6-7					
	H	7-8	52	16.5	47.5±2.2	1.6±0.3	.034±.006
	I	8-9	60	15.6	53.4±1.9	1.7±0.2	.032±.004
	J	9-10	63	26.3	49.6±2.3	1.9±0.3	.038±.006
	K	10-11	55	39.5	54.9±2.3	1.6±0.2	.029±.004
	L	11-12	63	11.9	68.3±2.6	1.9±0.3	.028±.004
	M	12-13	54	39.0	73.8±2.0	1.8±0.2	.025±.003
	N	13-14	51	5.5	76.6±5.3	2.3±0.5	.030±.007
	O	14-15	54	17.2	55.6±1.9	1.7±0.2	.031±.004
	P	15-16	59	24.0	53.1±2.1	1.7±0.2	.032±.005
	Q	16-17	56	60.7	48.6±1.9	1.2±0.2	.025±.004
	R	17-18	62	16.4	39.8±2.1	1.0±0.2	.025±.005
	S	18-19	59	37.0	25.9±0.8	0.6±0.1	.023±.004
	T	19-20	58	26.2	20.4±0.9	0.4±0.1	.022±.006
	U	20-21	68	29.8	12.5±0.6	0.4±0.1	.030±.007
	V	21-22	71	29.9	6.3±0.4	0.25±0.07	.040±.012
	W	22-23	75	10.1	3.2±0.3	N.D.	—
	X	23-24	67	13.3	0.81±0.01	N.D.	—
	Y	24-25	73	52.2	0.47±0.1	N.D.	—
	Z	25-26	75	50.8	0.22±0.1	N.D.	—

N.D. Not Detectable

TABLE 4

Nuclide Ratios in Core 54EC (Foundry Cove)

Location (Mile Point)	Sample No.	Depth (cm)	$^{239,240}Pu$ (pCi/Kg)	^{137}Cs (pCi/Kg)	$\frac{^{239,240}Pu}{^{137}Cs}$ x 10^{-2}	$\frac{^{134}Cs}{^{137}Cs}$ x 10^{-2}	$\frac{^{60}Co}{^{137}Cs}$ x 10^{-2}
mp 54EC	1240A	0-1	27.9±1.4	1530±38	1.82±.10	1.83±.85	4.77±.66
	B	1-2	28.6±1.7	1610±44	1.78±.12	2.92±1.0	3.42±.81
	C	2-3	30.9±1.4	2000±48	1.55±.08	3.75±.81	4.90±.66
	D	3-4	34.7±1.9	2400±56	1.45±.09	3.13±.67	6.04±.56
	E	4-5	35.4±1.9	2240±55	1.58±.09	1.34±.72	4.91±.64
	F	5-6	41.4±2.7	2060±56	2.01±.14	-	4.32±.74
	G	6-7	-	2010±48	-	-	3.33±.60
	H	7-8	47.5±2.2	2180±54	2.18±.12	-	3.81±.60
	I	8-9	53.4±1.9	2130±54	2.51±.11	-	4.46±.62
	J	9-10	49.6±2.3	1970±49	2.52±.13	-	5.08±.62
	K	10-11	54.9±2.3	2120±50	2.59±.12	-	3.30±.52
	L	11-12	68.3±2.6	2130±59	3.21±.15	-	1.92±.70
	M	12-13	73.8±2.0	2170±57	3.40±.13	-	3.09±.65
	N	13-14	76.6±5.3	2140±68	3.58±.27	-	2.20±.89
	O	14-15	55.6±1.9	1990±48	2.79±.12	-	1.81±.55
	P	15-16	53.1±2.1	1680±55	3.16±.16	-	2.56±1.1
	Q	16-17	48.6±1.9	1440±38	3.38±.16	-	1.18±.69
	R	17-18	39.8±2.1	1160±42	3.43±.22	-	-
	S	18-19	25.9±0.8	770±29	3.36±.16	-	-
	T	19-20	20.4±0.9	580±29	3.52±.23	-	-
	U	20-21	12.5±0.6	380±21	3.29±.24	-	-
	V	21-22	6.3±0.4	200±18	3.15±.35	-	-
	W	22-23	3.2±0.3	105±15	3.05±.52	-	-
	X	23-24	0.81±0.01	22±12	3.68±2.0	-	-
	Y	24-25	0.47±0.1	19±11	2.47±1.5	-	-
	Z	25-26	0.22±0.1	-9±13	-	-	-

are plotted as a function of depth in Figures 5 and 6, respectively.

The depth distributions of gamma-emitting nuclides and plutonium isotopes are consistent with the known history of both fallout delivery of radionuclides and reactor releases to the Hudson. Assuming uniform sedimentation rates, and relatively little vertical mixing of the sediments by organisms or other processes, the average accumulation rate of fine sediments in this core for the last two decades has been about 1 cm per year. This sedimentation rate is about an order of magnitude less than we have observed for New York harbor.

One of the most interesting features of the data in Figures 5 and 6 is the lack of indication of vertical mixing processes. The measured activites of ^{134}Cs and ^{60}Co (Figure 5) could have been derived only as the result of upstream transport by tidal currents from the reactor site 18 km downstream. If significant vertical mixing in the sediments had occurred, the activities of these nuclides would not show such large variations with depth. The variations with depth of ^{137}Cs (Figure 5) and plutonium isotopes (Figure 6) also indicate that the specific activities of the suspended particles at the time of deposition have been reasonably well preserved. The peak in ^{137}Cs at 3-4 cm was probably derived from reactor releases (1971), based on the coincidence of peaks in ^{134}Cs and ^{60}Co at that depth, while the initial rise in ^{137}Cs peaking at 12-13 cm was probably produced by the maximum in fallout delivery (1963-65).

The depth profile of $^{239,240}Pu$ can be reasonably interpreted in terms of maximum fallout deposition for the interval 12-14 cm, and gradually decreasing $^{239,240}Pu$ specific activities in the suspended load transported down the Hudson since the peak fallout years, with present activities

FIGURE 5. Gamma-emitting nuclide activites as a function of depth in a Hudson estuary core 18 Km upstream of Indian Point. The peak of ^{60}Co, ^{134}Cs activities and the uppermost peak in ^{137}Cs are believed to correlate with the period of maximum release of nuclides from the reactor (1971). The initial rise in ^{137}Cs is believed to have resulted from the peak fallout years (1963-65).

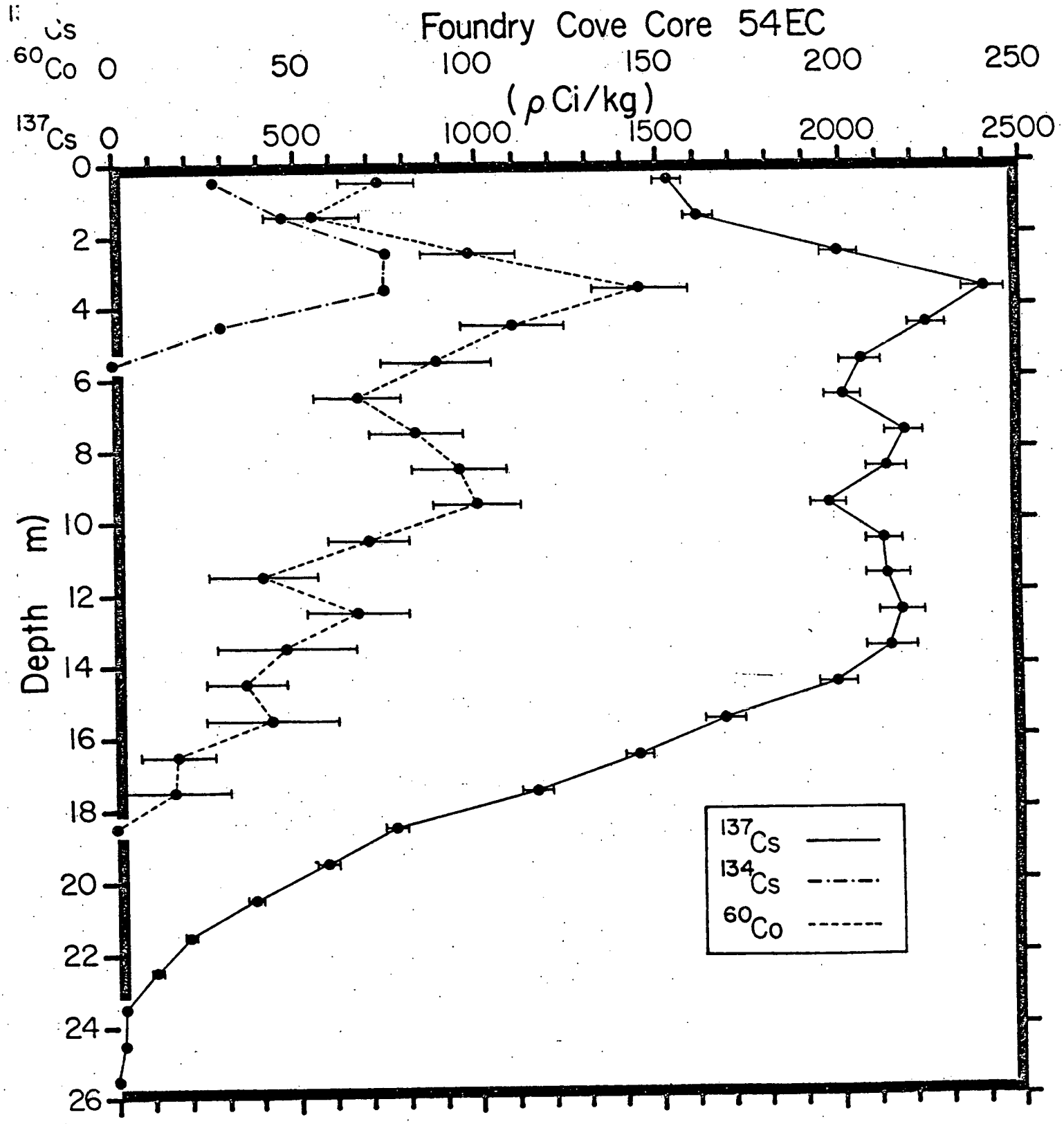
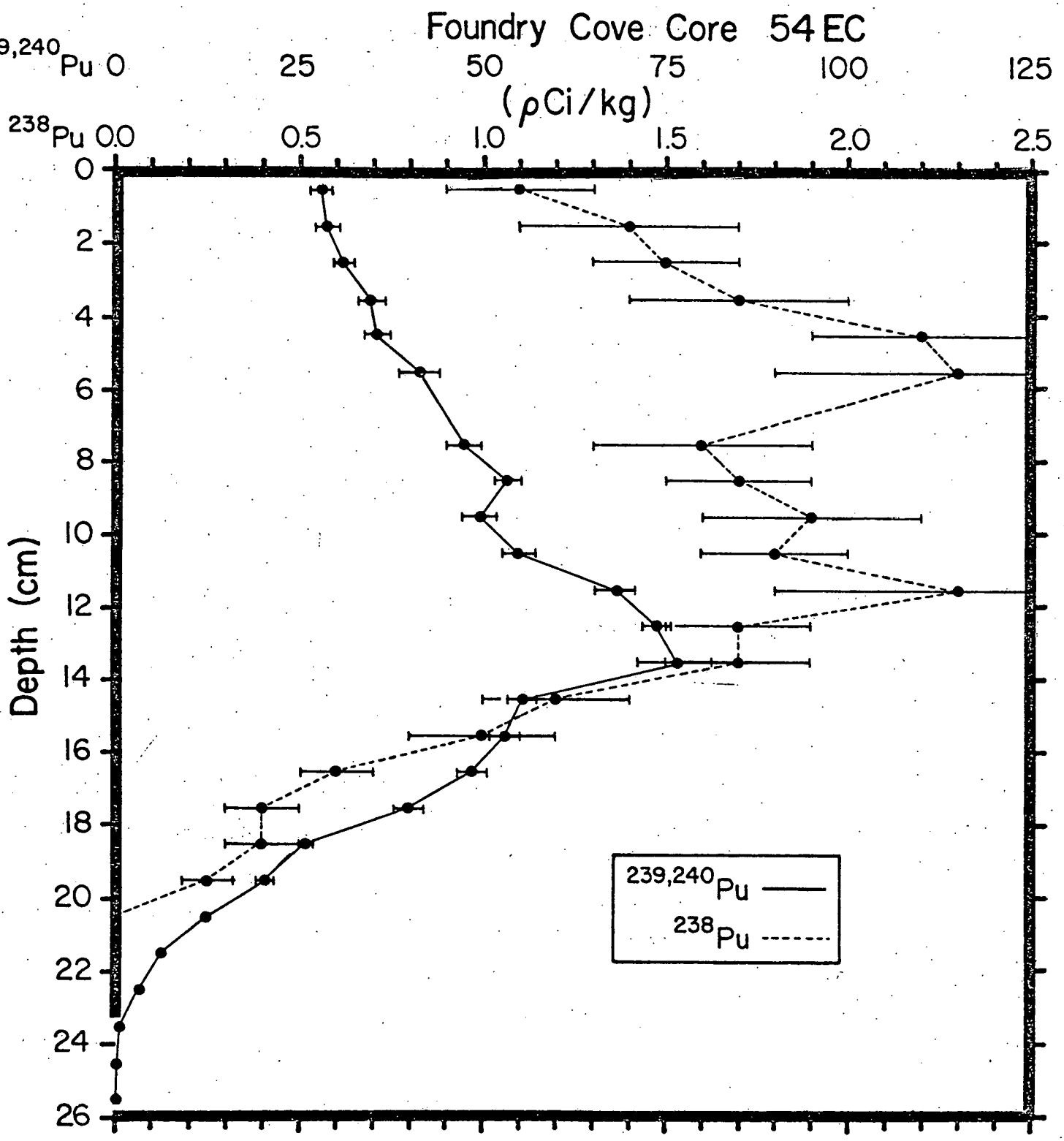

Foundry Cove Core 54EC
137Cs
60Co 0 50 100 150 200 250
(ρCi/kg)
137Cs 0 500 1000 1500 2000 2500
Depth m)
0 2 4 6 8 10 12 14 16 18 20 22 24 26
137Cs
134Cs
60Co

FIGURE 6. Plutonium isotope activites as a function of depth in a Hudson estuary core 18 Km upstream of Indian Point. The peak in $^{239,240}Pu$ activity is believed to have resulted from the peak fallout years (1963-65).


Foundry Cove Core 54 EC
239,240Pu 0
25
50
75
100
125
(ρCi/kg)
238Pu 0.0
0.5
1.0
1.5
2.0
2.5
Depth (cm)
0
2
4
6
8
10
12
14
16
18
20
22
24
26
239,240Pu
238Pu

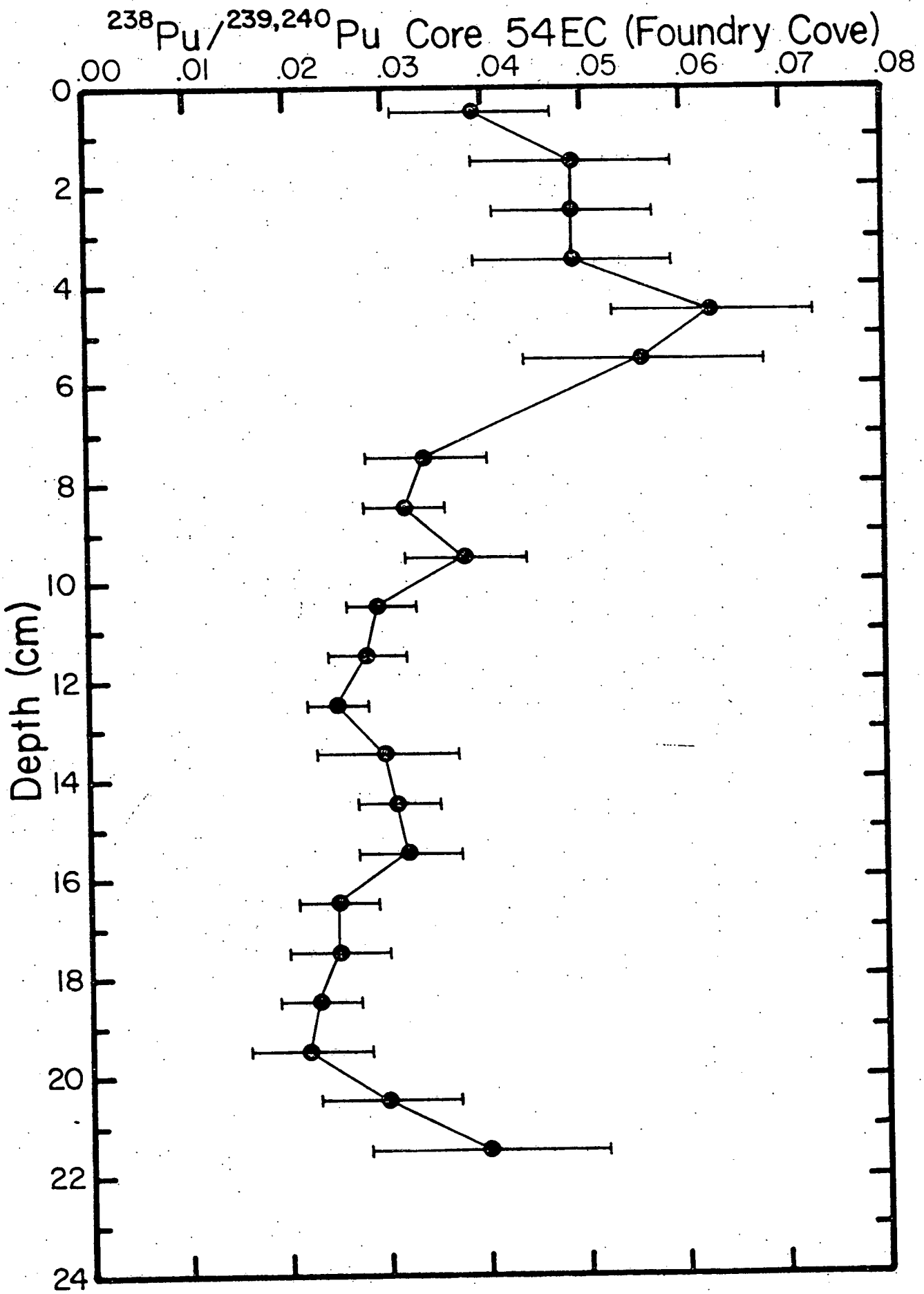
approximately one third of those in the years 1963-65. There does not seem to be any reasonable likelihood of a significant source of $^{239,240}Pu$ for the Hudson sediments other than fallout and the gradual movement of drainage basin soils into tidal water by erosion. The time trends in $^{239,240}Pu$ suspended load activity indicated from the data for the low salinity end of the estuary (Figure 6) are very similar to that recorded in the harbor sediments at salinities of about 2/3 of sea water (Figure 3).

The depth profile of ^{238}Pu in Foundry Cove (Figure 6) is not so straightforward to interpret. The trend for ^{238}Pu does not appear to be the monotonic decline in $^{239,240}Pu$ activity following the peak fallout years. Considering the much larger counting error on our ^{238}Pu data than for $^{239,240}Pu$ (Figure 6), it is difficult to draw firm conclusions from the depth trend of ^{238}Pu. However, the variations in ^{238}Pu to $^{239,240}Pu$ ratios with depth in the Foundry Cove core (Figure 7) are similar enough to the trends observed for New York harbor (Figure 4) to suggest a consistent picture for these two areas of the estuary separated by about 80 km. At present, it is not possible to establish whether or not a second significant source of ^{238}Pu has contributed to the distribution of plutonium in Hudson sediments. We plan to continue our examination of this line of research.

C. Radionuclide analysis of size fractions in Hudson estuary sediments

The transport and accumulation of plutonium and other radionuclides associated with particulate phases is influenced by the size and type of particles involved. In very general terms, much of the particle transport of plutonium is associated with fine particles rather than coarser material, and sandy sediments have lower specific activities of a whole spectrum of fallout and reactor-released nuclides than muddy sediments in contact

FIGURE 7. Ratios of ^{238}Pu to $^{239,240}Pu$ in Hudson estuary core 54 EC (Foundry Cove), located 18 Km upstream of Indian Point, in the low salinity region of the estuary.


$^{238}Pu/^{239,240}Pu$ Core 54EC (Foundry Cove)
.00
.01
.02
.03
.04
.05
.06
.07
.08
0
2
4
6
8
10
12
14
16
18
20
22
24
Depth (cm)

with the same ambient water. Since estuarine sediments often show a large range in mean particle size from one depositional environment to another, one can expect to find major differences in specific activities of radionuclides, including plutonium and other transuranics, from one area to another. The heteorogenity of particle size distributions and depositional environment in the Hudson was one of the primary reasons we chose to use direct-gamma counting of sediments as a means of preliminary screening of sample prior to radiochemical analysis. In general we have found the distribution of ^{137}Cs and other gamma-emitting nuclides in Hudson sediments to be very useful in guiding us in selection of sediment samples for alpha spectrometry. Much of our analytical work has been directed toward fine-grained sediments in the Hudson because of their higher specific activities, and greater potential for reconstructing depositional histories. Many of the areas in the Hudson with coarse sediments are in the main navigation channel, and are predominantly lag deposits left by active current scour.

The areas of the Hudson which have significant net deposition of recent fine-grained particles (New York harbor, subtidal banks, and marginal cores) all have specific activities of ^{137}Cs of the order of 0.5-2 pCi/g and of $^{239,240}Pu$ of the order of 20-60 pCi/Kg. The suspended load activities of these nuclides throughout the estuary are similar to those of the fine-grained sediments, although there is clear evidence for both desorption of ^{137}Cs in the higher salinity areas and supply of ^{137}Cs from reactor releases in addition to that delivered from fallout. Thus, the predominant process regulating the accumulation pattern of radionuclides in Hudson sediments is the transport, deposition and resuspension of the suspended load.

As part of our effort to examine the role of fine-particles in the transport of radionuclides in the Hudson, we have analyzed several size fractions of a large sample of sediments from a marginal cove in the low salinity region of the estuary. A composite sample was prepared by collecting a number of large grab samples from an area which was predominantly fine particles (Foundry Cove, near the site of Core EC) to which was added a few large grab samples from a sandy area at the bottom of a nearby tidal current channel (also in Foundry Cove). This large composite sample was homogenized as well as possible immediately after collection, and then size fractioned by wet sieving through a set of decreasing mesh size screens, followed by settling and centrifugation. The size fractions obtained and the specific activities of several gamma emitting nuclides are listed in Table 5. Of the total sample (several kilograms dry weight - all size fractioning was done while still wet), however, most of the mass was found in the 20-63μ fraction and in the greater than 250μ fraction, with much of the latter consisting of coal and ash fragments typical of lag deposits in the Hudson (Figure 8). Plutonium data and nuclide ratios for the same size fractions are reported in Tables 6 and 7, respectively.

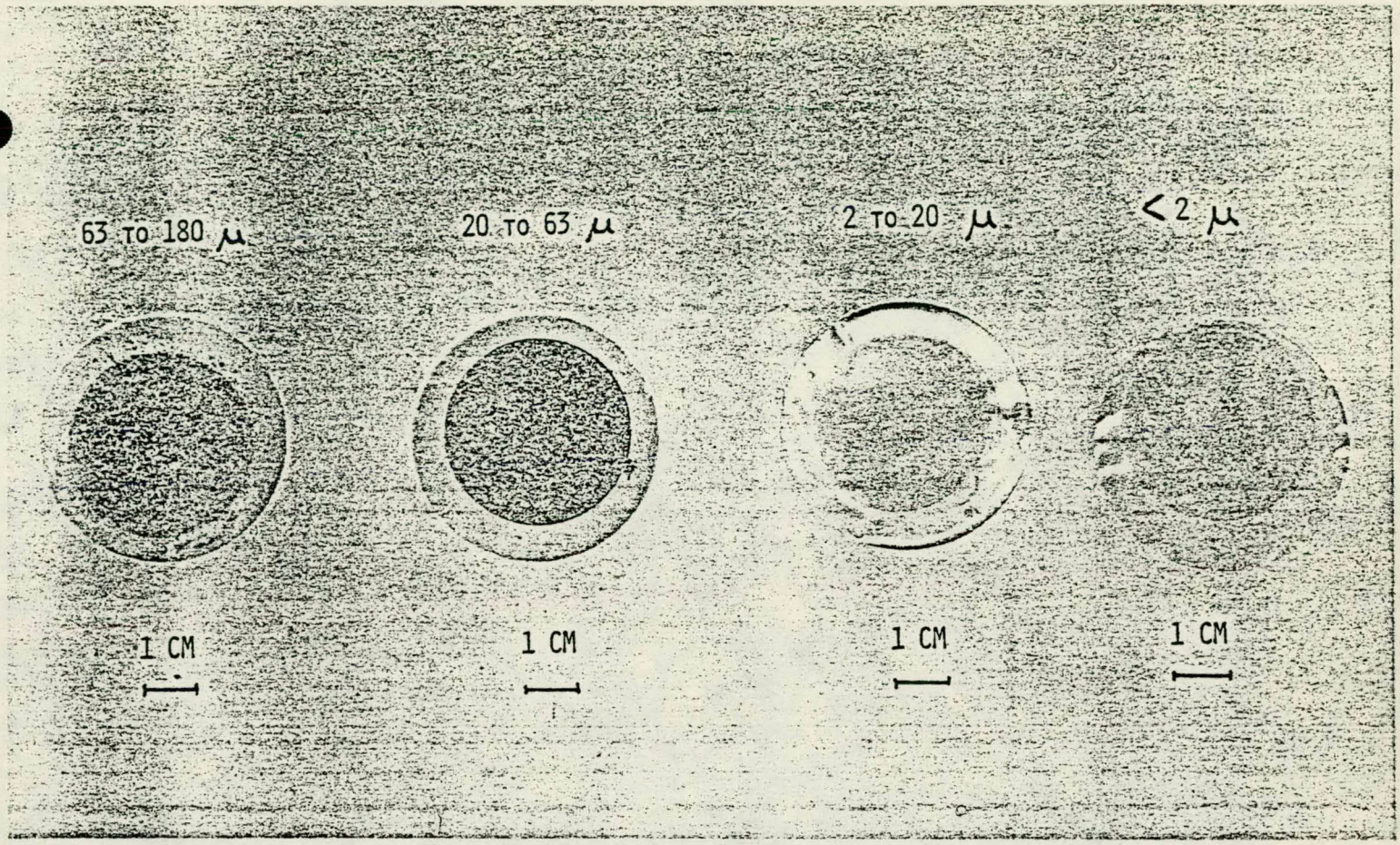
The specific activities of both the gamma-emitting nuclides and the plutonium isotopes were significantly higher in the silt (2-63μ) and clay (<2μ) size fractions than in coarse sand (180-250μ) as would be expected. The activities in the size range 63-180μ were comparable to the finer particles indicating that much of this material was probably aggregations of the silt particles. We did not attempt to disaggregate the sediments with ultrasonic agitation, or any other mechanical processes which are sometimes employed in size fraction preparations of clay minerals because

TABLE 5

γ-Emitting Radionuclides in Foundry Cove Composite Sample Size Fractions

Sediment Size (μ)	Sample No.	^{137}Cs (pCi/Kg)	^{134}Cs (pCi/Kg)	^{60}Co (1Ci/Kg)	^{40}K (pCi/Kg)
0-2	1352 G	810±30	96±21	60±15	10.1±0.5
2-20	F	1575±38	26±13	82±13	20.1±0.7
20-63	E	1045±45	53±22	81±22	16.3±0.8
63-180	D	1100±33	39±14	67±14	15.4±0.6
180-250 (Floc)	C	1470±165	650±142	515±142	11.3±1.6
180-250 (Sand)	B	375±27	46±20	57±19	10.7±0.6
>250	A	61±15	31±14	25±14	3.3±0.2

FIGURE 8. Photograph of the composite sediment sample from Foundry Cove illustrating the different size fractions for which the gamma-emitting radionuclide and plutonium activites are presented in Tables 5 and 6, respectively.


63 TO 180 μ
20 TO 63 μ
2 TO 20 μ
< 2 μ
1 CM
1 CM
1 CM
1 CM

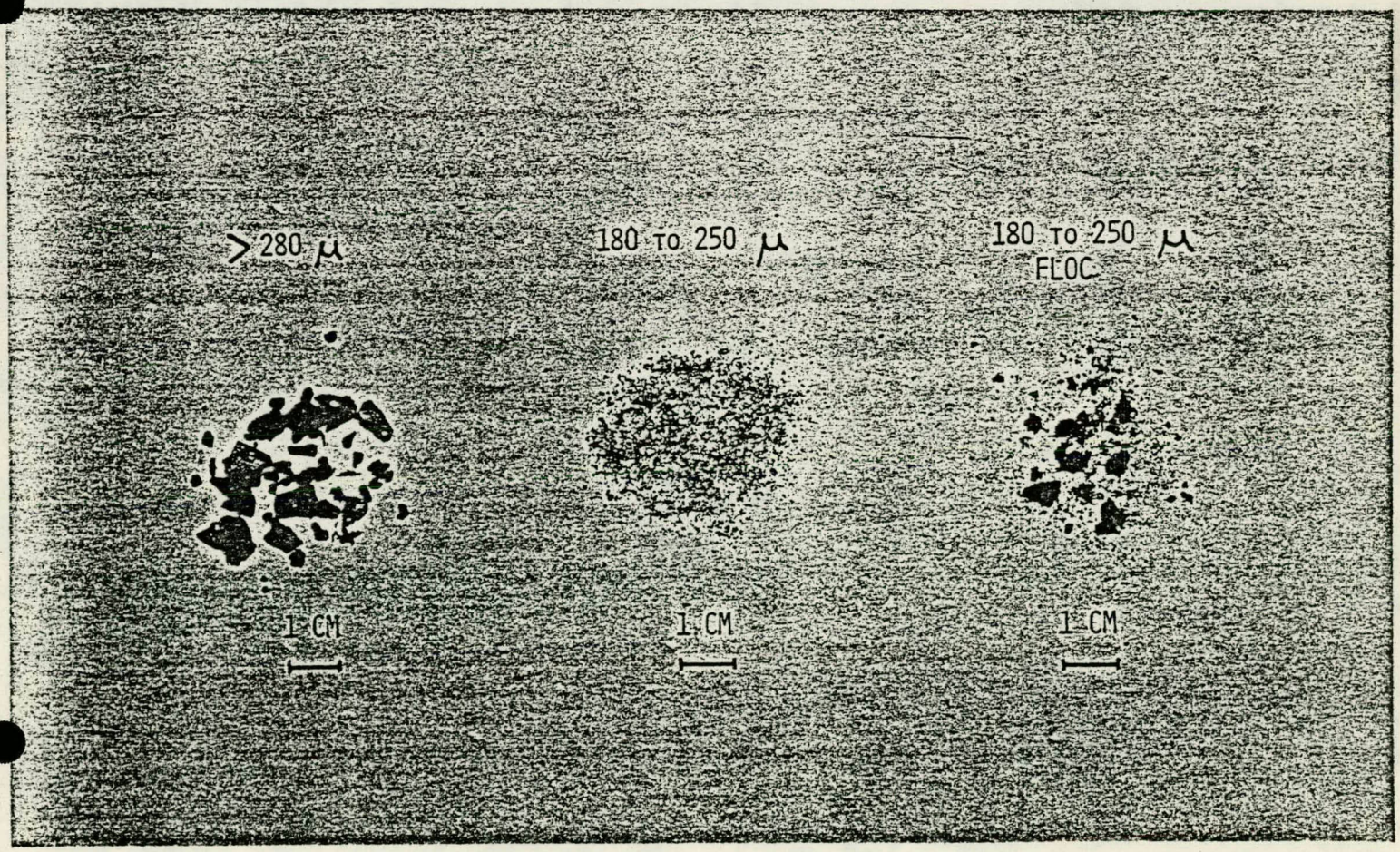

> 280 μ
180 TO 250 μ
180 TO 250 μ
FLOC
1 CM
1 CM
1 CM

TABLE 6

Plutonium Isotopes in Foundry Cove Composite Sample Size Fractions

Sediment Size (μ)	Sample No.	$^{239,240}Pu$ (pCi/Kg)	^{238}Pu (pCi/Kg)	$^{238}Pu/^{239,240}Pu$ (pCi/Kg)
0-2	1352G	12.4±1.0	0.71±.12	.057±.011
2-20	F	20.5±0.8	0.86±.13	.042±.007
20-63	E	19.0±1.2	0.70±.22	.037±.012
63-180	D	26.4±1.0	1.21±.15	.046±.006
63-180	D-1	26.4±2.3	N.D.	
180-250 (Floc)	C	43.6±3.7	N.D.	
180-250 (Sand)	B	20.7±1.8	0.96±.32	.046±.016
>250	A	3.7±0.3	0.14±.05	.038±.014

TABLE 7

Nuclide Ratios in Foundry Cove Composite Sample Size Fractions

Sediment Size (μ)	Sample No.	$^{239,240}Pu$ (pCi/Kg)	^{137}Cs (pCi/Kg)	$\frac{^{239,240}Pu}{^{137}Cs} \times 10^{-2}$	$\frac{^{134}Cs}{^{137}Cs} \times 10^{-2}$	$\frac{^{60}Co}{^{137}Cs} \times 10^{-2}$
0-2	1352G	12.4±1.0	810±30	1.53±.14	11.9±2.6	7.4±1.9
2-20	F	20.5±0.8	1575±38	1.30±.06	1.7±.9	5.2±0.8
20-63	E	19.0±1.2	1045±45	1.82±.14	5.1±2.1	7.8±2.1
63-180	D	26.4±1.0	1100±33	2.40±.12	3.6±1.3	6.1±1.3
180-250 (Floc)	C	43.6±3.7	1475±165	2.96±.42	44.2±10.9	35.0±8.0
180-250 (Sand)	B	20.7±1.8	375±27	5.52±.63	12.3±5.4	15.2±5.2
>250	A	3.7±0.3	61±15	6.07±1.57	50.8±26.1	41.0±25.1

we wished to preserve as much as possible the size distribution of the sediments as they relate to transport potential in the estuary. The dominant size fraction collected by our procedures was 20-63μ, accounting for more than 90% of the total sample less than 250μ.

One aspect of the data reported in Tables 5 and 6 which was somewhat surprising was the apparent decrease in specific activity of most of the nuclides in the smallest size fraction (<2μ). This was most noticeable for plutonium, but was also indicated for ^{137}Cs to a somewhat lesser extent. Since the dominant material <2μ should be clay minerals, one possible interpretation of the lower Pu activity is that organic material may be more important in contributing binding sites than clay minerals in Hudson sediment. Further support of this interpretation is indicated by the higher specific activity in the 180-250μ fraction designated as "floc". Sediments which passed through the 250μ sieve, but were trapped by the 180μ sieve fell into two quite distinct categories. The sandy mineral grains settled rapidly while a large quantity of organic floc remained in suspension or floated. This latter material accounted for about 10% of the dry weight mass of the 180-250μ size fraction, and was higher in specific activity for all the radionuclides measured than any of the other fractions. Much of the radionuclide activity in the sandy portion of the 180-250μ fraction was probably also associated with this type of material since we did not attempt to remove all of the flocculant particles from the phase which settled out fairly rapidly. Considering the high specific activity of $^{239,240}Pu$ measured for the large (180-250μ) organic particles, it seems reasonable to assume that organic phases play a significant role in the

accumulation of plutonium (and other radionuclides) in Hudson sediments. Because of the binding between organics and mineral particles, especially clay minerals, in estuary sediments, it is generally not possible to separate these two types of material mechanically (except in relatively unusual situations such as that for the the 180-250μ particles described here) and any chemical separation procedure is likely to significantly alter the distribution of radionuclides which exists in the sediments in a natural water system. Thus, although organic phases appear to be significant in the sediment binding and suspended particle transport of radionuclides, we cannot clearly establish from our work up to now the relative importance of organic material and a variety of mineral phases in the sediments and suspended load of the Hudson.

D. Radionuclide analysis of sediments in tributaries of the Hudson estuary

To help characterize the supply of fallout nuclides from the soils of the Hudson drainage basin to the sediments of the estuary, we have collected and analyzed samples of fine-grained sediments from several of the tributaries of the tidal Hudson. Data on gamma-emitting nuclides for two size fractions are reported in Table 8. In general, the sediments <63μ have significantly higher specific activities than those >63μ, as would be expected. The same size fraction trend is observed for plutonium isotopes in the tributary sediments (Table 9). The activities of ^{137}Cs and $^{239,240}Pu$ in the <63μ fraction of all of the tidal water tributaries (Mohawk River and Catskill, Esopus, Roundout, Wappinger and Fishkill Creeks) are somewhat lower than fine-grained Hudson estuary sediments from the areas currently accumulating sediments (harbor, subtidal banks, and marginal coves), but the primary explanation may be a systematic difference in mean grain size. The

TABLE 8

γ-Emitting Radionuclide in Hudson Tributaries and Adjacent Bays

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	Cs^{137} (pCi/Kg)	Cs^{134} (pCi/kg)	Co^{60} (pCi/Kg)	K^{40} (pCi/ g)
Mohawk River	1333C	<63μ	112	680±22			17.0±0.6
(mp156)	1333A	>63μ	125	270±14			17.1±0.5
Catskill Creek	1334B	<63μ	87	565±23			15.3±0.5
(mp113)	1334A	>63μ	140	190±12			14.9±0.5
Esopus Creek	1335B	<63μ	92	870±32			12.8±0.5
(mp102)	1335A	>63μ	112	280±16			8.6±0.3
Roundout Creek	1336B	<63μ	102	585±24			11.7±0.5
(mp91)	1336A	>63μ	134	100±8			6.7±0.2
Wappinger Creek	1338B	<63μ	89	675±24			15.9±0.5
(mp67)	1338A	>63μ	148	170±10			12.5±0.4
Fishkill Creek	1337B	<63μ	86	315±17			16.9±0.6
(mp60)	1337A	>63μ	144	145±11			12.0±0.4
Newark Bay	1350A	0-5	75	220±16	6±9	6±10	14.4±0.5
	1350B	5-10	76	30±25	25±12	24±12	15.8±0.6
	1350C	10-16	89	-3±21	13±8	15±8	15.8±0.5
Arthur Kill	1346A	0-2	8	345±54	-30±40	90±109	13.3±0.9
	B	2-4	9	440±55	-42±38	35±60	13.1±0.9
	C	4-6	11	495±47	2±29	99±61	14.1±0.9
	D	6-8	9	470±49	-59±33	62±39	13.6±0.9
	E	8-11	26	535±32			16.6±0.8
	F	11-13	20	490±40	-7±23	29±26	15.3±1.0
	G	13-16	28	510±36			16.0±0.8
	H	16-19	33	485±30			16.4±0.8
	I	19-22	46	140±21			14.1±0.6
	J	22-25	60	245±16			15.0±0.6
	K	25-30	87	60±14			10.5±0.5
	L	30-35	120	2±5			8.5±0.3
	M	35-40	88	-11±10			17.1±0.6
Raritan Bay	1343A	0-7	67	70±13	-12±9	16±17	14.3±0.6
	B	7-11	94	-6±13	-7±8	4±4	17.5±0.6
East River	1340B	<63μ	30	825±40	84±17	61±15	17.4±0.8
(Hallet Cove)	A	>63μ	66	38±11	19±8	17±8	8.6±0.4
East River	1339B	<63μ	31	380±33	31±18	-8±19	14.4±0.8
(Hells Gate)	A	>63μ	134	35±11	15±7	11±8	10.9±0.4

TABLE 9

Plutonium Isotopes in Hudson Tributaries and Adjacent Bays

Location (mile point)	Sample No.	Depth (cm)	Dry Wt. (gms)	% Yield	$^{239,240}Pu$ pCi/Kg	^{238}Pu pCi/Kg	$^{238}/^{239,240}Pu$
Mohawk River	1333C	<63μ	92	30.5	8.9±0.5	0.3±0.1	0.030±.008
(mp 156)	1333A	>63μ	115	3.1	2.4±0.4	N.D.	--
Catskill Creek	1334B	<63μ	77	24.8	10.2±0.6	0.4±0.1	0.039±.009
(mp 113)	1334A	>63μ	131	5.9	2.2±0.3	N.D.	--
Esopus Creek (mp102)	1335B	<63μ	81	28.0	11.0±0.6	0.7±0.1	0.059±.010
Roundout Creek (mp 91)	1336B	<63μ	82	14.7	8.7±0.6	N.D.	--
Wappinger Cr.	1338B	<63μ	79	10.5	10.4±0.8	0.5±0.1	0.047±.014
(mp 67)	1338A	>63μ	138	32.6	1.6±0.1	N.D.	--
Fishkill Creek (mp 60)	1337B	<63μ	76	23.4	3.6±0.3	N.D.	--
East River							
(Hallet Cove)	1340B	<63μ	25	7.7	31.0±2.6	1.7±0.5	.055±.016
(Hells Gate)	1339B	<63μ	26	20.4	26.3±1.9	1.2±0.3	.046±.013
Long Island Sound		Grab	92	16.0	20.4±0.8	0.9±0.1	0.043±.006

tributary samples were obtained in non-tidal waters where the current regime is quite different than in tidal waters, being dominated by spring maximum flows followed by moderate to low flows through most of the remainder of the year. In general, the kind of depositional environment in tidal waters, characterized by reversing current directions separated by periods of little or no current velocity, shouldn't be reproduced in any of the tributaries. The Mohawk River is ponded behind the Troy dam on the main stem of the Hudson somewhat below the area from which our sample was collected, so the depositional environment there should be more like that of a reservoir than in the other tributaries discussed here. The specific activity of ^{137}Cs and $^{239,240}Pu$ in a bulk sample produced by combining the two size fractions for each of the tributaries would be significantly lower than for the <63μ fraction alone, and for fine sediments from the estuary. Since the proportion of sample >63μ is so much greater in the tributary samples than in fine sediments from the Hudson (see Tables 5, 6, 7 for Foundry Cove), it is likely that the <63μ fraction from the tributaries may also have larger mean particle size than for the same size fraction in the estuary, or be systemically different in some other property, such as the type or amount of organic matter.

The ratio of $^{239,240}Pu$ to ^{137}Cs in the tributary sediments (Table 10) is comparable to the fallout delivery ratio, indicating little or no systematic difference in the mobility of these fallout nuclides in the drainage basin soils. One simple explanation for the data is that both ^{137}Cs and $^{239,240}Pu$ are relatively tightly bound to the soil particles, and the activities found in the tributary sediments reflect particle erosion of drainage basin soils and transport of the nuclides by the particles into

TABLE 10

Nuclide Ratios in Hudson Tributaries and Adjacent Bays

Location (mile point)	Sample No.	Depth (cm)	$^{239,240}Pu$ pCi/Kg	^{137}Cs pCi/Kg	$\frac{^{239,240}Pu}{^{137}Cs} \times 10^{-2}$	$\frac{^{134}Cs}{^{137}Cs} \times 10^{-2}$	$\frac{^{60}Co}{^{137}Cs} \times 10^{-2}$
Mohawk River (mp 156)	1333C	<63μ	8.9±0.5	680±22	1.31±.08	--	--
	1333A	>63μ	2.4±0.4	270±14	.89±.16	--	--
Catskill Cr. (mp 113)	1334B	>63μ	10.2±0.6	565±23	1.81±.13	--	--
	1334A	<63μ	2.2±0.3	190±12	1.16±.17	--	--
Esopus Cr. (mp 102)	1335B	<63μ	11.0±0.6	870±32	1.26±.08	--	--
	1335A	>63μ	--	280±16	--	--	--
Roundout Cr. (mp 91)	1336B	<63μ	8.7±0.6	585±24	1.49±.12	--	--
	1336A	>63μ	--	100±8	--	--	--
Wappinger (mp 67)	1338B	<63μ	10.4±0.8	675±24	1.54±.13	--	--
	1338A	>63μ	1.6±0.1	170±10	.94±.08	--	--
Fishkill Cr	1337B	<63μ	3.6±0.3	315±17	1.14±.11	--	--
	1337A	>63μ	--	145±11	--	--	--
Newark Bay	1350A	0-5	--	220±16	--	--	--
	B	5-10	--	30±25	--	--	--
	C	10-16	--	-3±21	--	--	--
East River (Hallet Cove)	1340B	<63μ	31.0±2.6	825±40	3.76±.36	10.1±2.1	7.4±1.9
	1340A	>63μ	--	38±11	--	50.0±25.5	44.7±24.7
(Hells Gate)	1339B	<63μ	26.3±1.9	380±33	6.92±.78	8.2±4.8	--
	1339A	>63μ	--	35±11	--	42.9±24.1	--

estuary with no significant loss of either ^{137}Cs or $^{239,240}Pu$ from soil particles in the tidal fresh water reach of the Hudson.

During the past several years we have been collecting a substantial amount of data on the suspended load characteristics of the Hudson estuary, since this is one of the primary factors controlling the transport and accumulation of radionuclides in the Hudson. We are in the process of preparing a report on that study at the present time, and should be able to provide a complete summary in our next annual report. One of our primary observations is that the suspended load mass concentration in the Hudson estuary are extremely variable in both space and time. Although it is possible to draw some general conclusions about zones of fine particle sediment accumulation and ranges of observed suspended load mass concentrations, we believe it is not going to be feasible to make much progess in using such data to predict very precisly where radionuclides are likely to accumulate, and what the net transport patterns are over many tidal cycles. We believe this applies to both qualitative analysis of the suspended particle mass data and to numerical model simulations of the transport based on the same data. Our general impression is that the major input of data for tuning numerical model simulations of radionuclide transport by the particle load should be from what are, in effect, long term tracer experiments in the natural water system of interest. Thus, the measured distribution of tracers, such as the gamma-emitting nuclides from a reactor release area, or fallout nuclides in an area where such tracers are not already present, offers a much better long-term average of particle transport properties in a system of interest than any extrapolation of suspended particle mass measurements can hope to provide.

We have begun to try to use the distribution of natural radionuclides in conjunction with reactor and fallout nuclides to help understand the origin and net transport of fine-grained particles in the Hudson. Activities of several natural radionuclides in tributary sediments are reported in Table 11, based on gamma counting of ^{40}K and of radioactive daughters of ^{226}Ra and ^{228}Ra. From these data, plus similar data for natural radionuclides in the sediments of the Hudson estuary, we believe that the tributaries of the tidal Hudson (Catskill, Esopus, Roundout, Wappinger and Fishkill Creeks) make relatively small contribution to the total supply of particles to the tidal Hudson, despite the fact that the drainage basin downstream of the dam at Troy accounts for more than a third of the total water budget. The major source of particles, based on the natural radioclide data collected so far, appears to be the Mohawk River. We hope to explore further the potential of this type of fine-particle province indicator as we obtain samples from other parts of the drainage basin.

Data for sediments from marginal areas of the lower Hudson estuary are also reported in Tables 8-11. The East River, a portion of the harbor tidal complex, has definite activities of reactor-derived nuclides (^{134}Cs and ^{60}Co) which must have been supplied to the system originally at mp43 from Indian Point. Thus, from the viewpoint of radionuclide accumulation patterns, the parts of the East River which have appreciable fine particle sedimentation should be considered as part of the harbor zone of deposition for nuclides supplied to the Hudson well upstream of the harbor complex. Other marginal areas of lower estuary for which we have samples (Newark Bay, Raritan Bay and the upper East River leading to Long Island Sound) do not show evidence of reactor gamma-emitting nuclides, but the specific

TABLE 11

Natural Radionuclides in Hudson Tr :aries & Adjacent Bays

Location (mp of Mouth)	Sample #	Depth (cm)	Dry Wt. (g)	Org.Matter (%)	^{40}K (pCi/g)	^{226}Ra (pCi/Kg)	^{228}Ra (pCi/ .g)
Mohawk River	1333C	<63μ	112	3.0	17.0±0.6	865±142	910±97
(mp 156)	1333A	>63μ	125		17.1±0.5	380± 75	375±50
Catskill Creek	1334B	<63μ	87	6.2	15.3±0.5	660±117	1050±122
(mp113)	1334A	>63μ	140		14.9±0.5	445± 84	755±82
Esopus Creek	1335B	<63μ	92	5.6	12.8±0.5	825±150	1095±125
(mp 102)	1335A	>63μ	112		8.6±0.3	440± 89	610±71
Roundout Creek	1336B	<63μ	102		11.7±0.5	820±145	1020±112
(mp 91)	1336A	>63μ	134		6.7±0.2	345±62	365±43
Wappinger Creek	1338B	<63μ	89		15.9±0.5	690±121	1260±133
(mp 67)	1338A	>63μ	148		12.5±0.4	220± 54	665±71
Fishkill Creek	1337B	<63μ	86	6.9	16.9±0.6	905±145	1250±142
(mp 60)	1337A	>63μ	144		12.0±0.4	295±64	505±61
Newark Bay	1350A	0-5	75		14.4±0.5	655±116	980±118
	B	5-10	76		15.8±0.6	740±135	1145±140
	C	10-15	89		15.8±0.5	740±122	1050±120
Raritan Bay		0-7	67	6.4	14.3±0.6	565±98	910±127
		7-11	94	5.7	17.5±0.6	560±107	1035±123
East River (Hallet Cove)		<63μ	30		17.4±0.8	810±135	1100±175
East River (Hells Gate)		<63μ	31		14.4±0.8	975±166	775±137

activities of ^{137}Cs in our samples and the rates of recent sediment accumulation are sufficiently low that it is difficult to establish the lack of reactor contribution with much certainty. Another part of the harbor complex, the Arthur Kill, is definitely a zone of fairly rapid recent sediment accumulation, and there is some hint of reactor activity (^{60}Co), but the data obtained so far for that area are not sufficient to draw any firm conclusions about the source of particles accumulating there.

E. Radionuclide data collected in the tidal Hudson during the last contract year

A comprehensive summary of our radionuclide analytical data for Hudson estuary sediments are given in Tables 1 and 2 of our last annual report (COO-2529-3). Data collected during this contract year are given in tables discussed earlier in this report (Tables 2, 3, 4, 5, 6, 7, 8, 9, 10, 11). Additional data collected during this contract year, and not reported in Tables 2-11 are included in Tables 12, 13, and 14. The data in these last three tables extends our sediment sample coverage over almost the entire reach of the tidal Hudson. Several zones of rapid sediment accumulation in the tidal fresh water reach of the Hudson were identified as a result of this initial survey (see cores 87.0C, 109.5WCI, P143.4 in Table 12). These cores, plus a few additional ones such as 91.8 indicate zones in which accumulation of radionuclides could be expected in the tidal fresh water reach of the Hudson if significant sources in addition to fallout were to be present in the future. Thus, the pattern of large variations in radionuclide accumulation rates which we have demonstrated for the saline intruded reach of the Hudson is also typical of the fresh water tidal reach of the Hudson, and should be incorporated explicitly in any

TABLE 12

γ-Emitting Radionuclides in the Hudson River Estuary[1]

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	Cs^{137} (pCi/Kg)	Cs^{134} (pCi/kg)	Co^{60} (pCi/Kg)	K^{40} (pCi/ g)
Inner Harbor							
P-0.4W	1375A	0-10	51	585±25	98±27	37±25	16.0±0.6
(18 Oct. 1975)	B	10-20	46	1030±35	185±31	150±18	17.1±0.6
	C	20-30	49	1610±60	255±51	190±29	17.2±0.8
	D	30-40	36	1030±45	130±42	56±23	17.1±0.8
	E	40-50	40	760±29	57±26	34±15	17.0±0.6
	F	50-60	44	985±34	170±29	85±17	15.8±0.6
	G	60-70	55	545±24	57±24	33±15	14.3±0.5
	H	70-80	39	710±50	97±53	2±32	16.4±0.9
	I	80-90	37	555±32	-2±16	2±23	16.8±0.7
	J	90-100	60	250±19	12±11	18±13	14.4±0.6
	K	100-110	83	275±15	13±8	22±8	15.1±0.5
	L	110-120	81	245±16	14±9	-2±15	14.5±0.5
	M	120-130	81	245±15	5±8	13±13	14.8±0.5
	N	130-140	85	235±17	14±9	4±10	14.7±0.5
	*O	140-150	73	520±22	57±22	92±15	15.4±0.5
	P	150-159	51	320±21	17±12	45±28	14.4±0.6
	Q	159-170	104	47±12	-3±9	-4±13	17.0±0.6
	R	170-180	98	13±10	8±8	5±11	16.2±0.5
	S	180-190	95	21±10	2±7	-2±11	17.1±0.5
	T	190-200	97	33±7	9±5	12±5	16.8±0.5
	U	200-210	82	8±12	2±9	-6±15	18.0±0.6
Piermont Marsh							
PM #1-4	Surf	0-1	68	700±17	52±7	175±8	20.2±0.6
(May 20, 1977)	Composite	1-2	71	775±18	19±6	61±6	21.7±0.6
PM #1	1254A	2-5	58	845±42	-6±18	3±4	17.4±0.8
(May 20, 1977)	B	5-10	101	425±27	-13±14	24±14	16.8±0.7
	C	10-15	84	110±18	10±11	8±9	16.8±0.7
	D	15-20	95	21±13	7±11	22±12	17.1±0.6
	E	20-25	105	16±15	7±13	11±16	17.1±0.7

γ-Emitting Radionuclides in the Hudson River Estuary[1]

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	Cs^{137} (pCi/Kg)	Cs^{134} (pCi/kg)	Co^{60} (pCi/Kg)	K^{40} (pCi/g)
PM #2	1255A	2-5	59	30±18	-10±16	22±30	18.9±0.8
(May 20, 1977)	B	5-10	76	2±18	33±17	7±9	19.0±0.8
PM #3	1256A	2-5	51	330±29	-3±18	94±22	19.1±0.6
(May 20, 1977	B	5-10	75	305±16	13±9	41±10	18.7±0.6
	C	10-15	75	425±31	37±17	42±18	19.0±0.8
	D	15-20	98	58±13	-11±20	21±16	18.8±0.6
	E	20-25	87	-3±20	-5±14	11±18	19.7±0.8
Tappan Zee							
30.8A	1356A	0-2	34	1535±60	45±29	350±29	20.0±0.9
(August 19, 1977)	B	2-4	39	1440±51	-31±18	240±25	19.6±0.8
	C	4-8	64	1035±32	-3±10	2±3	18.2±0.7
	D	8-10	61	300±20	-1±12	6±16	19.5±0.7
	E	10-14	86	23±22	-6±11	21±12	18.3±0.8
	F	14-18	99	-7±21	-8±12	5±11	19.0±0.7
30.8B	1357A	0-4	43	575±33	40±23	70±20	18.3±0.8
(Aug. 19, 1977)	B	4-8	91	87±15	26±12	12±15	19.0±0.7
30.8C	1358A	0-4	51	265±23	37±16	57±25	17.8±0.7
(Aug. 19, 1977)	B	4-8	81	26±14	-9±11	2±15	19.2±0.7
	D	12-16	90	-10±12	2±9	0±13	18.3±0.7
36 E	1096A	0-5	75	425±34	0±17	40±40	21.4±1.0
(27 Oct. 1976)	B	5-10	75	18±16	-3±12	-12±14	21.8±0.8
	C	10-15	83	12±7	-3±6	3±7	19.4±0.6
39 E	1100A	0-7	107	88±12	4±8	16±9	17.6±0.6
(14 Aug. 1975)	B	7-12	89	-12±15	--	--	18.1±0.7
	C	12-16	78	+7±21	--	--	20.4±0.8
SLOSH III[e]	1000Q-4	Oxidized layer	89	2700±72	345±42	400±27	19.2±0.8
(June 11, 1975)	Organics	>250μ	20	2360±110	555±70	700±37	10.9±0.5
SLOSH IV (June 11,1975)	1000D-1	0-10	67	520±35	24±19	50±19	20.6±0.8
Lents Cove							
43.2 ECI	1264A	0-2	32	1090±51	82±25	96±21	15.6±0.8
(July 11, 1977)	B	2-4	27	1130±54	54±25	95±21	13.8±0.7
	C	4-8	58	1730±66	120±28	195±27	13.3±0.7
	D	8-12	57	4140±93	395±29	420±20	14.5±0.6
	E	12-16	71	3220±70	255±22	280±17	12.1±0.5
	F	16-20	62	2200±46	160±12	175±9	13.2±0.4
	G	20-24	78	1590±43	78±14	115±14	12.8±0.5
	H	24-28	61	1280±38	52±15	190±16	14.0±0.6
	I	28-32	75	970±32	32±13	99±14	14.3±0.6
	J	32-36	71	860±41	15±16	100±19	15.4±0.7

γ-Emitting Radionuclides in the Hudson River Estuary[1]

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	Cs^{137} (pCi/Kg)	Cs^{134} (pCi/kg)	Co^{60} (pCi/Kg)	K^{40} (pCi/ g)
Lents Cove cont'd.							
	1264K	36-40	55	1020±35	7±13	145±16	14.5±0.6
	L	40-44	82	845±35	19±9	130±12	14.4±0.5
	M	44-48	75	520±35	4±18	47±19	14.8±0.8
	N	48-52	85	410±19	8±9	47±11	16.4±0.6
	O	52-56	76	255±21	11±13	17±16	25.3±0.9
	P	56-60	100	11±10	-9±9	3±9	14.9±0.5
	Q	60-64	98	-5±10	14±8	-5±9	15.2±0.5
Hudson River							
72.6[2]	1281A	0-2	30	1020±160			
(Luly 11, 1977)	1281B	2-4	34	550±96			
	C	4-8	78	462±65			
	D	8-12	88	575±55			
	E	12-16	93	545±49			
	F	16-20	105	-44±33			
	G	20-24	109	52±32			
83.2 EC[2]	1284A	0-2	35	1158±165			
(July 11, 1977)	B	2-4	28	1328±81			
	C	4-8	60	1258±123			
	D	8-12	63	995±127			
	E	12-16	80	1248±100			
	F	16-20	77	537±73			
	G	20-24	100	31±33			
	H	24-28					
	I	28-32	101	-5±3			
87.0A[2]	1287A	0-2	25	1389±180			
	B	2-4	34	930±128			
	C	4-8	91	136±46			
	D	8-12	120	-9±32			
	E	12-14	54	-41±44			
87.0B[2]	1288A	0-2	47	38±65			
(July 11, 1977)	B	2-4	39	33±72			
	C	4-8	91	-24±39			
	D	8-12	97	-17±40			

γ-Emitting Radionuclides in the Hudson River Estuary[1]

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	Cs^{137} (pCi/Kg)	Cs^{134} (pCi/kg)	Co^{60} (pCi/Kg)	K^{40} (pCi/ g)
87.0B[2] cont'd	1288E	12-16	98	26±38			
	F	16-20	117	-30±33			
87.0C[2]	1289A	0-2	28	1382±141			
(July 11, 1977)	C	4-8	50	833±105			
	D	8-12	66	942±84			
	F	16-20	43	2809±150			
	H	24-28	62	5154±200			
	J	32-36	56	2270±96			
	L	40-44	56	2124±134			
	N	48-52	64	1965±111			
	Ø	52-56	62	1941±98			
	P	56-60	72	1949±138			
	Q	60-64	60	2604±159			
91.8[2]	1329A	0-2	25	1049±76			
(July 12, 1977)	B	2-4	28	1115±75			
	C	4-8	66	1658±118			
	D	8-12	62	1776±140			
	E	12-16	66	1630±137			
	F	16-20	67	1486±129			
	G	20-24	68	1991±137			
	H	24-28	67	2335±70			
	I	28-32	70	1084±78			
	J	32-36	67	36±20			
	K	36-40	71	20±45			
108A[2]	1321A	0-2	69	28±25			
(July 14, 1977)	B	2-4	77	5±8			
	C	4-8	125	8±15			
	D	8-12	119	19±10			
108B[2]	1322A	0-2	56	152±54			
(July 14, 1977)	B	2-4	38	90±18			
	C	4-8	110	53±12			

γ-Emitting Radionucl s in the Hudson River Estuary[1]

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	Cs^{137} (pCi/Kg)	Cs^{134} (pCi/kg)	Co^{60} (pCi/Kg)	K^{40} (pCi/ .g)
109.5WCI	1319A	0-2	31	995±52		17±18	18.2±0.9
(July 15, 1977)	B	2-4	27	950±42		17±14	19.5±0.9
	C	4-8	65	895±26		8±8	17.8±0.6
	D	8-12	62	950±32		4±2	18.2±0.7
	E	12-16	63	875±30			18.1±0.7
	F	16-20	65	780±38			16.7±0.8
	G	20-24	68	1140±40			16.1±0.7
	H	24-28	78	1030±31			17.7±0.6
	I	28-32	75	1420±35			18.2±0.6
	J	32-36	79	1790±38			19.6±0.6
	K	36-40	87	1660±59			18.4±0.8
	L	40-44	90	2870±55			18.7±0.6
	M	44-48	88	1600±44			19.6±0.7
	N	48-52	82	1100±29			20.3±0.7
	O	52-56	78	1740±58			21.3±0.9
	P	56-60	79	4560±100			19.6±0.8
P143.4	1298A	0-10	57	1452±50			
(July 14, 1977)	B	10-20	72	1685±47			
	C	20-30	60	2120±61			
	D	30-40	51	2060±62			
	E	40-50	63	3165±76			
	F	50-60	45	2769±79			
	G	60-70	50	159±27			
	H	70-80	46	89±16			
	I	80-90	31	84±25			
	J	90-100	39	67±20			
	K	100-110	59	65±16			
	L	110-120	43	54±16			
	M	120-130	42	7±16			
	N	130-140	76	45±12			
	Ø	140-150	67	37±12			
	P	150-160	48	14±13			
	Q	160-170	55	8±11			
	R	170-180					
	S	180-190	65	6±10			
	T	190-200					
	U	200-210	75	-9±10			

TABLE 13

Plutonium Isotopes in the Hudson River Estuary

Location (mile point)	Sample No.	Depth (cm)	Dry Wt. (gm)	% Yield	$^{239,240}Pu$	^{238}Pu	$^{238/239,240}Pu$
36 EC	1096A	0-5	74	26.3	7.9±0.5	0.3±0.8	0.039±.011
	B	5-10	65	30.3	0.3±0.1	N.D.	--
39	1100A	0-7	71	13.7	1.0±0.2	N.D.	--
40 WL	1095A	0-5	67	42.5	33.1±1.3	1.4±0.2	0.043±.006
	B	5-10	--	--	--		--
	C	10-15	55	20.4	2.5±0.2	--	--
109.5WCI	1319P	56-60	69	25.1	40.1±1.6	0.8±0.1	0.020±.003
143.4A	1298A	0-10	47	9.9	15.8±0.9	0.8±0.2	0.051±.011
	B	10-20	56	30.9	18.3±0.8	0.8±0.1	0.043±.007
	C	20-30	45	15.5	20.4±1.4	1.6±0.3	0.081±.016
	D	30-40	-	--	--	--	--
	E	40-50	48	23.3	34.9±1.8	1.7±0.3	0.048±.008
	F	50-60	33	18.3	33.0±2.3	0.8±0.3	0.026±.008
	G	60-70	-	--	--	--	--
	H	70-80	35	37.1	0.8±0.1	N.D.	--
	I	80-90	20	11.6	1.8±0.5	N.D.	--

TABLE 14

Nuclide Ratios in the Hudson River Estuary Sediments

Location (mile point)	Sample No.	Depth (cm)	$^{239,240}Pu$ pCi/Kg	^{137}Cs pCi/kg	$\frac{^{239,240}Pu}{^{137}Cs} \times 10^{-2}$	$\frac{^{134}Cs}{^{137}Cs} \times 10^{-2}$	$\frac{^{60}Co}{^{137}Cs} \times 10^{-2}$
36 EC	1096A	0-5	7.9±0.5	425±34	1.86±.19	--	--
	B	5-10	0.3±0.1	18±16	--	--	--
39 E	1100A	0-7	1.0±0.2	88±12	1.14±.28	--	--
40WC	1095A	0-5	33.1±1.3	--	--	--	--
	B	5-10	--	--	--	--	--
	C	10-15	.25±0.2	--	--	--	--
109.5WCI	1319P	56-60	40.1±1.6	4560±100	.88±.04	--	--
143.4A	1298A	0-10	15.8±0.9	1452±50	1.09±.07	--	--
	B	10-20	18.3±0.8	1685±47	1.09±.06	--	--
	C	20-30	20.4±1.4	2120±61	0.96±.07	--	--
	D	30-40	--	2060±62	--	--	--
	E	40-50	34.9±1.8	3165±76	1.10±.06	--	--
	F	50-60	33.0±2.3	2769±79	1.19±.09	--	--
	G	60-70	--	159±27	--	--	--
	H	70-80	0.8±0.1	89±16	.90±20	--	--
	I	80-90	1.8±0.5	84±25	2.14±.87	--	--

radionuclide monitoring program for that portion of the Hudson in the future. The sampling sites in the tidal fresh water Hudson reported here should not be considered as representative of the proportion of various depositional environments, since we were generally attempting to identify zones of most rapid sediment accumulation and chose our first coring sites with that in mind.

One of the cores reported in Table 12 is from New York harbor (P-0.4W). The depth profiles of ^{137}Cs, ^{134}Cs and ^{60}Co show a large peak at 20-30 cm which probably resulted from the peak reactor release year (1971), much as we observed in core P-1.5. Thus, the pattern of rapid recent accumulation of sediments in the harbor which we have observed in previous cores is found in this section as well. This core should be useful in helping confirm the accumulation history of plutonium in the zone of highest deposition in the Hudson.

Several cores from a large marsh area situated at intermediate salinities in the Hudson are also reported in Table 12 (Piermont Marsh). This area is known to have been shoaling fairly rapidly over the past century and might reasonally be expected to be an important accumulation zone for radionuclides. From the core samples we have analyzed up to now, the rate of radionuclide accumulation near Piermont marsh appears to be faster than much of the total area of the estuary, but is considerably lower than the harbor area and somewhat lower than for marginal coves further upstream.

One of the coring sites (43.2ECI) in a small marginal cove near the reactor appears to offer the most detailed and continuous record of the release of gamma-emitting nuclides from Indian Point of any site we have located up to now. The peak in ^{137}Cs, ^{134}Cs and ^{60}Co activities occurs at a depth of 8-12 cm in the core we collected, with a second

smaller maximum in ^{137}Cs and ^{60}Co at 36-40 cm. The general shape of the depth profile of nuclides is very similar to the time release history of these nuclides which has been reconstructed from discharge information.

As documented in an earlier report (COO-2529-3 - Table 12) we have prepared a large composite sample of surface sediment from near the reactor site to be used primarily as a laboratory secondary standard for our own quality control and for exchange with other laboratories. During the initial preparation of this sample we removed coarse organic debris by screening through a 250μ sieve. Analysis of this fraction by gamma spectrometry indicates specific activities of ^{137}Cs, ^{134}Cs, and ^{60}Co as high or higher than for the finer particles which passed through the sieve (Tables 12, Slosh III organics). Considering that this fraction contains a substantial portion of macroscopic plant debris such as root fragments, the activity of gamma-emitting nuclides in the organic floc of this coarse fraction is probably as high as any material we have analyzed in the Hudson. This observation is similar to our findings for the coarse fraction organic floc in Foundry Cove (Tables 2, 3, 4) and helps to confirm the importance of coarse organic detritus in transporting radionuclides by particulate matter in estuaries.

Data on plutonium isotopes for samples not discussed earlier are included in Table 13, and a summary of isotope ratios is included in Table 14. The general coherence in the pattern of ^{137}Cs and $^{239,240}Pu$ activities found up to now for Hudson sediment is also observed in these data. Sites of low but measurable activity of ^{137}Cs also had $^{239,240}Pu$ activities of a few percent of that for areas of rapid deposition of recent fine-grained particles (Table 13: 36EC, 5-10cm; 39; 0-7cm; 40WC, 10-15cm; 143.4A, 70-80 cm, 80-90cm). Ratios of $^{239,240}Pu$ to ^{137}Cs in the tidal fresh water reach of the Hudson (Table 14) were very similar to the fallout ratio of these nuclides, as indicated for the tributary sediments (Table 10).

F. Integrated radionuclide activities in the Hudson cores

One of the goals of our research in the Hudson is to establish the present distribution of radionuclides in the sediments. This information is needed to estimate the total burden of radionuclides in the sediment, and to help define the transport and accumulation processes in estuaries which are important on the time scale of years to decades. Vertical profiles of radionuclide activities in sediment cores have been discussed in previous sections of this report. The total integrated activity for each of the cores is also of interest for budget calculations and for describing the overall pattern of accumulation of radionuclides. The integrated activities of ^{137}Cs, ^{134}Cs, ^{60}Co and $^{239,240}Pu$ for a number of Hudson cores are listed in Table 15. The accumulation rates of $^{239,240}Pu$, expressed as mCi/Km^2, range from 10% of fallout delivery ($\sim$2 mCi/Km^2) to more than 30 times fallout delivery. We believe the primary factor which is responsible for this large range in accumulation of radionuclides per unit area is the heterogenity in depositional environments of fine particles. Some areas serve as very efficient settling basins, probably because of decreased current velocities due to shore line configuration or large scale estuarine circulation features which serve to trap or focus particle deposition in restricted zones. Other areas are scoured of fine particles by tidal currents, even though temporary accumulation may occur for parts of each tidal cycle or for much longer periods before removal takes place. The dynamics of estuarine circulation are sufficiently complicated in both space and time to defy detailed description of most of the processes which are important in establishing the net fine particle depositional pattern over the time scale of years to decades. The most reasonable approach appears to be to use the observed distribution of nuclides to deduce the long-term transport features.

TABLE 15

Integrated Radionuclide Activities [1] in the Sediments of the Hudson Estuary, Adjacent Bays and Fallout

Location (mile point)	Sample #	^{137}Cs (mCi/Km^2)	^{134}Cs (mCi/Km^2)	^{60}Co (mCi/Km^2)	$^{239,240}Pu$ (mCi/Km^2)
Fallout [2]		116	-	-	1.77
Raritan Bay	1343	2.0	N.D. [5]	N.D.	-
Arthur Kill	1346	42	N.D.	N.D.	-
Newark Bay	1350	7.1	N.D.	N.D.	0.4
-1.6E [3]	1051	185	10	14	15
P-1.5E [3]	1048	822	53	42	64
P-0.4W	1375	502	52	34	
0.1W [4]	1066	344	32	32	10
0.1WM [4]	1002	278	23	28	8.0
1.9W	1082	60	2.6	4.1	2.9
6.0W	1085	370	18	44	-
11.5W	1010	1.7	N.D.	0.9	-
13.2M	1058	0.9	N.D.	N.D.	-
14.2M	1005	5.7	N.D.	N.D.	-
18.6-1M	1229	8.8	N.D.	N.D.	-
18.6-3M	1089	12	1.3	2.3	-
18.6-5W	1232	4.7	N.D.	N.D.	-
18.6W	1090	22	2.7	2.2	-
G18.6-5W [6]	1092	16	0.3	2.3	0.3
21.7M	1025	10	0.5	2.2	-
21.9W	1021	17	N.D.	3.1	-
PM#1	1254	55	N.D.	3.3	-
PM#2	1255	N.D.	N.D.	N.D.	-
PM#3	1256	44	2.6	7.7	-
25.3W	1086	71	N.D.	3.1	3.0
30.8A	1356	83	1.2	11	-
30.8B	1357	14	1.4	1.6	-
30.8C	1358	6.7	1.9	1.5	-
33.8EC	1915	39	4.8	6.2	-
36E	1096	13	N.D.	N.D.	0.3
39E	1100	3.9	N.D.	N.D.	0.04
40WC	1095	116	4.9	7.9	2.3
C43	1043	11	1.3	2.5	

Integrated Radionuclide Activities (1) in the Sediments of the Hudson Estuary, Adjacent Bays and Fallout

Location (mile point)	Sample #	$^{137}Cs_2$ (mCi/Km^2)	$^{134}Cs_2$ (mCi/Km^2)	$^{60}Co_2$ (mCi/Km^2)	$^{239,240}Pu$ (mCi/Km^2)
43.2EC	1264	568	54	71	-
C44	1032	30	1.9	4.6	-
49.5W	1093	53	4.6	6.3	-
52.5EC	1039	198	7.1	11.4	-
53.8EC	1052	121	1.1	1.2	3.6
54EC	1240	341	3.6	13	8.6
Foundry Cove	FC CIIC	117	N.D.	6.0	-
	FCBI	125	N.D.	2.1	-
56.4W	1069	32	N.D.	1.6	-
57.5M	1035	N.D.	N.D.	N.D.	-
59.7EC	1091	52	N.D.	N.D.	1.4
72.6	1281	80	-	-	-
75.5	1259	113	N.D.	N.D.	-
76.1	1260	15	N.D.	N.D.	-
83.2EC	1284	152	-	-	-
87.0A	1287	34	-	-	-
87.0B	1288	N.D.	-	-	-
87.0C(3)	1289	585	-	-	-
91.8	1329	363	-	-	-
91.3	1261	N.D.	-	-	-
108A	1321	N.D.	-	-	-
108B	1322	7.6	-	-	-
109.5WC(3)	1319	773	N.D.	N.D.	-
P143.4	1298	961	-	-	9.8

(1) Activities are integrated and corrected to October 15, 1975, small differences between the integrated values in this table relative to integrated values reported previously, reflect corrections to a specific date rather than to the date of core collection and also incorporation of a more exact measurement of the core diameter.

(2) As determined from monthly ^{90}Sr precipitation data for New York City assuming $^{137}Cs/^{90}Sr = 1.5$ and $^{239,240}Pu/^{90}Sr = 0.017$ at deposition.

(3) Sediment Core did not penetrate to sediments containing no ^{137}Cs activity.

(4) Sediment record disturbed by dredging (see Olsen et al., 1978).

(5) N.D. ≈ No detectable activity, dashes indicate that the sample was not analyzed.

(6) Grab sample assumed sediment density of 0.7 g/cm^3.

(7) Local "hotspot" for ^{60}Co, may reflect a local source for ^{60}Co other than Indian Point.

The integrated activities of ^{137}Cs, ^{134}Cs and ^{60}Co are also listed in Table 15. The large range in accumulation rates observed for $^{239,240}Pu$ appears to be typical of the gamma-emitting nuclides as well. Interpretation of the overall distributions of these nuclides will be somwhat more difficult than for $^{239,240}Pu$. In the case of ^{137}Cs there are two significant sources, fallout and reactor-releases, and also clear evidence of some desorption of ^{137}Cs from particles in the higher salinity region of the estuary. Thus, the heterogeneous field of accumulation of fine particles bearing ^{137}Cs must also be considered in terms of these other complication. The distribution of ^{134}Cs and ^{60}Co are also somewhat more difficult to explain than $^{239,240}Pu$, although the only source is reactor releases, because of the combination of a complicated time history of releases and relatively short half-lives (2 yrs and 5 yrs, respectively). We should be able to place constraints on a number of important transport processes for all of these radionuclides, however, and will be summarizing some of these efforts in our next annual report.

The major features of the variations in accumulation of radionuclides along the axis of the Hudson can be seen in Figure 9 and 10, which show a log plot of the integrated activities of ^{137}Cs and $^{239,240}Pu$ as a function of mile point. The zone of fine particle accumulation in the harbor is evident as high integrated activities for both ^{137}Cs and $^{239,240}Pu$, even though some ^{137}Cs has been lost by desorption from the particles accumulating in the harbor. Areas of high accumulation rate are also indicted for a few localities in the tidal river portion of the Hudson (Figure 9).

The present sediment integrated activities of reactor-derived nuclides, ^{134}Cs and ^{60}Co, are shown in Figures 11 and 12. It is clear that the primary

FIGURE 9. An updated version of the total ^{137}Cs deposition (mCi/Km^2) along the axis of the Hudson Estuary. Note that the vertical axis is in log scale and that the black columns along the mile point baseline indicate no detectable activity. The greatest accumulation of both fallout and reactor-released ^{137}Cs occurs in the inner harbor area of New York and in coves (indicated by the letter C next to the column) upstream along the estuary margin.

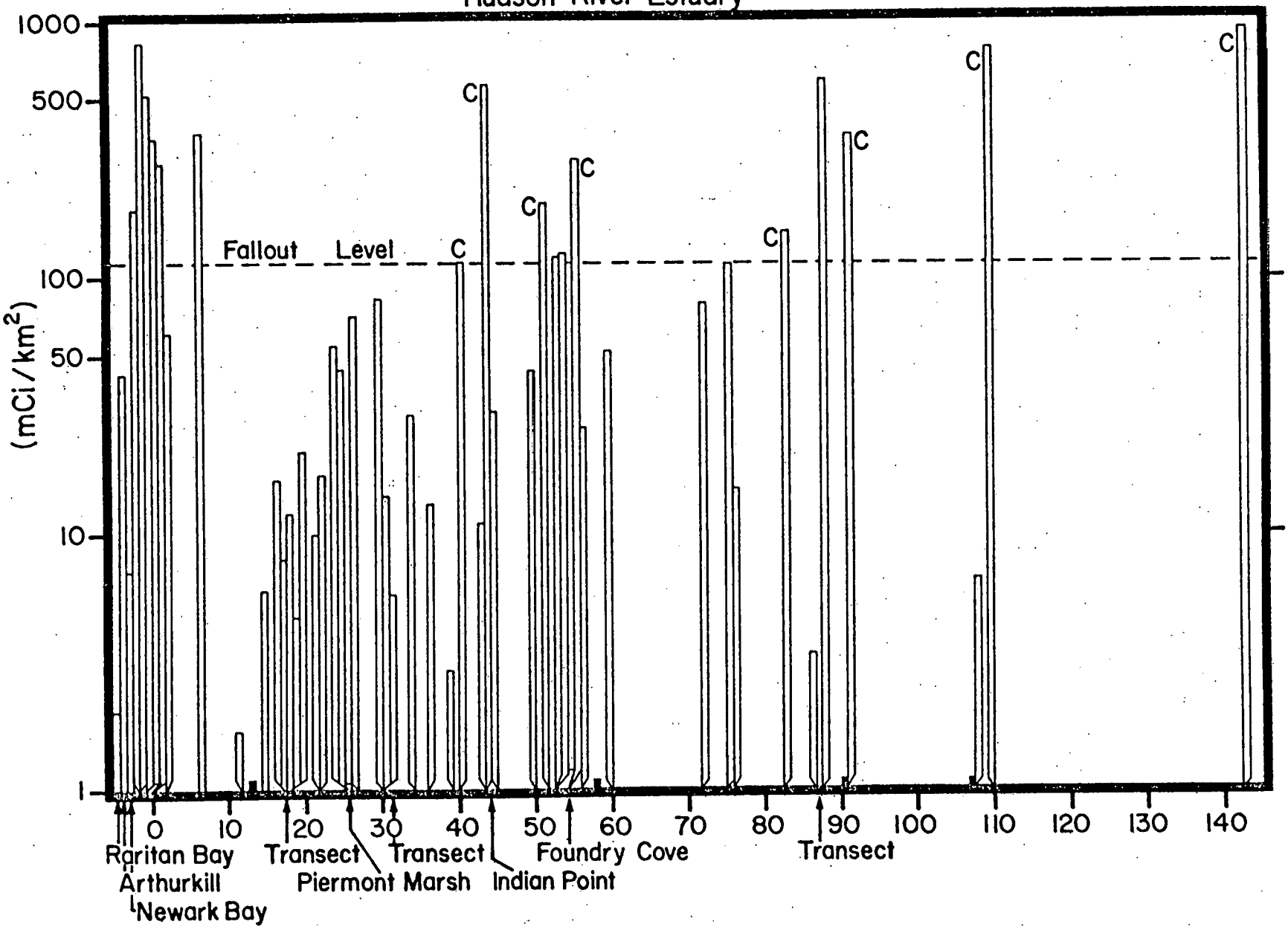

^{137}Cs Accumulation (mCi/km^2) vs. Mile Point
Hudson River Estuary
1000
500
100
50
10
1
(mCi/km^2)
Fallout Level
C
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
Raritan Bay
Arthurkill
Newark Bay
Transect
Transect
Piermont Marsh
Indian Point
Foundry Cove
Transect

FIGURE 10. An updated verson of the total $^{239,240}Pu$ deposition (mCi/Km^2) along the axis of the Hudson Estuary. The greatest accumulation of $^{239,240}Pu$ (like that of ^{137}Cs) occurs in the inner harbor area of New York and in coves upstream along the estuary margin.

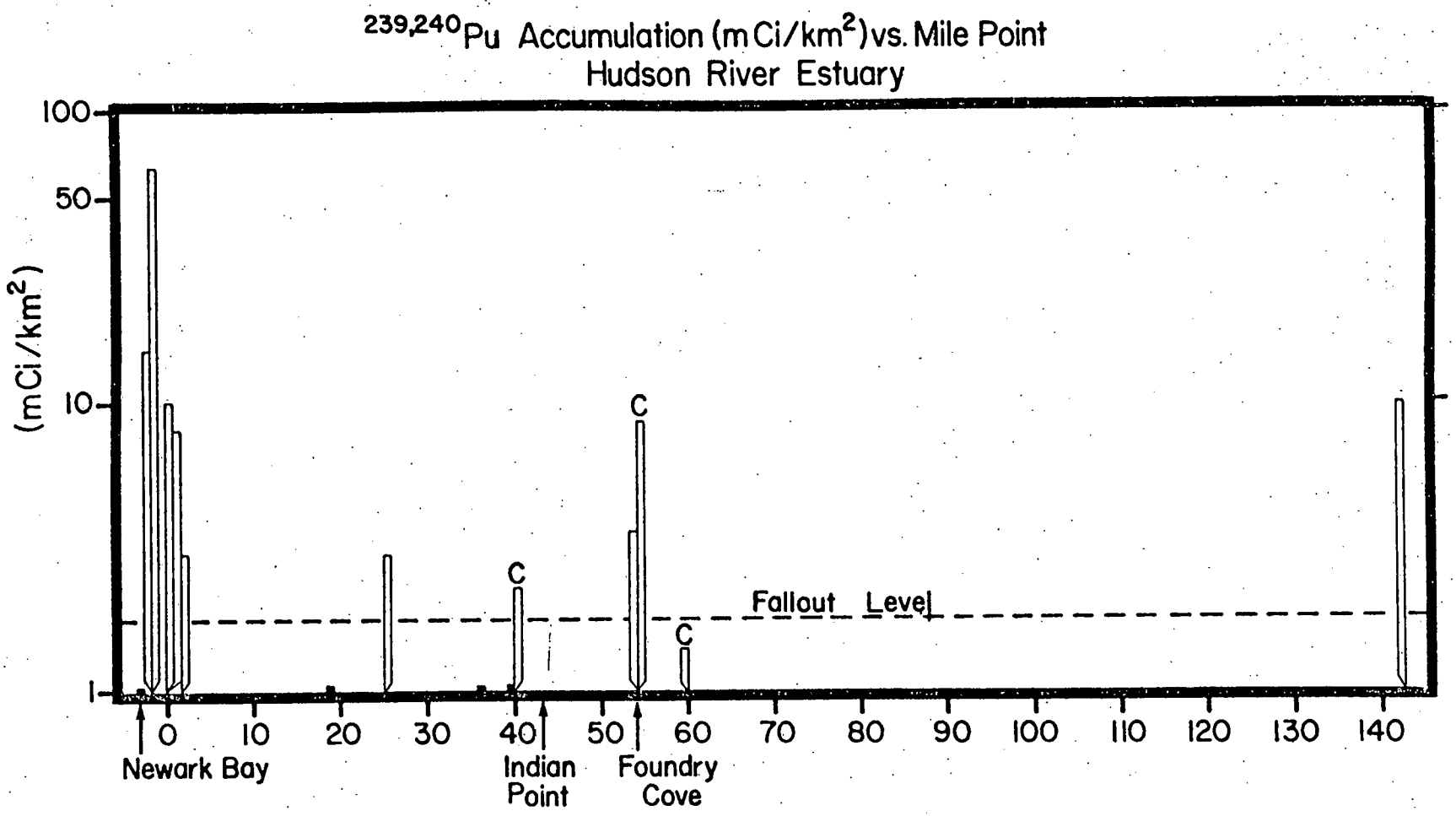

^{239,240}Pu Accumulation (m Ci/km^2) vs. Mile Point
Hudson River Estuary
100
50
10
1
(mCi/km^2)
C
C
C
Fallout Level
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
Newark Bay
Indian Point
Foundry Cove

FIGURE 11. Total ^{134}Cs deposition (mCi/Km^2) along the axis of the Hudson Estuary. The greatest accumulation of reactor-released ^{134}Cs occurs in the inner harbor area of New York and in coves (indicated by the letter C next to the column) upstream along the estuary margin. The blank columns indicate no detectable ^{134}Cs activities at these locations.

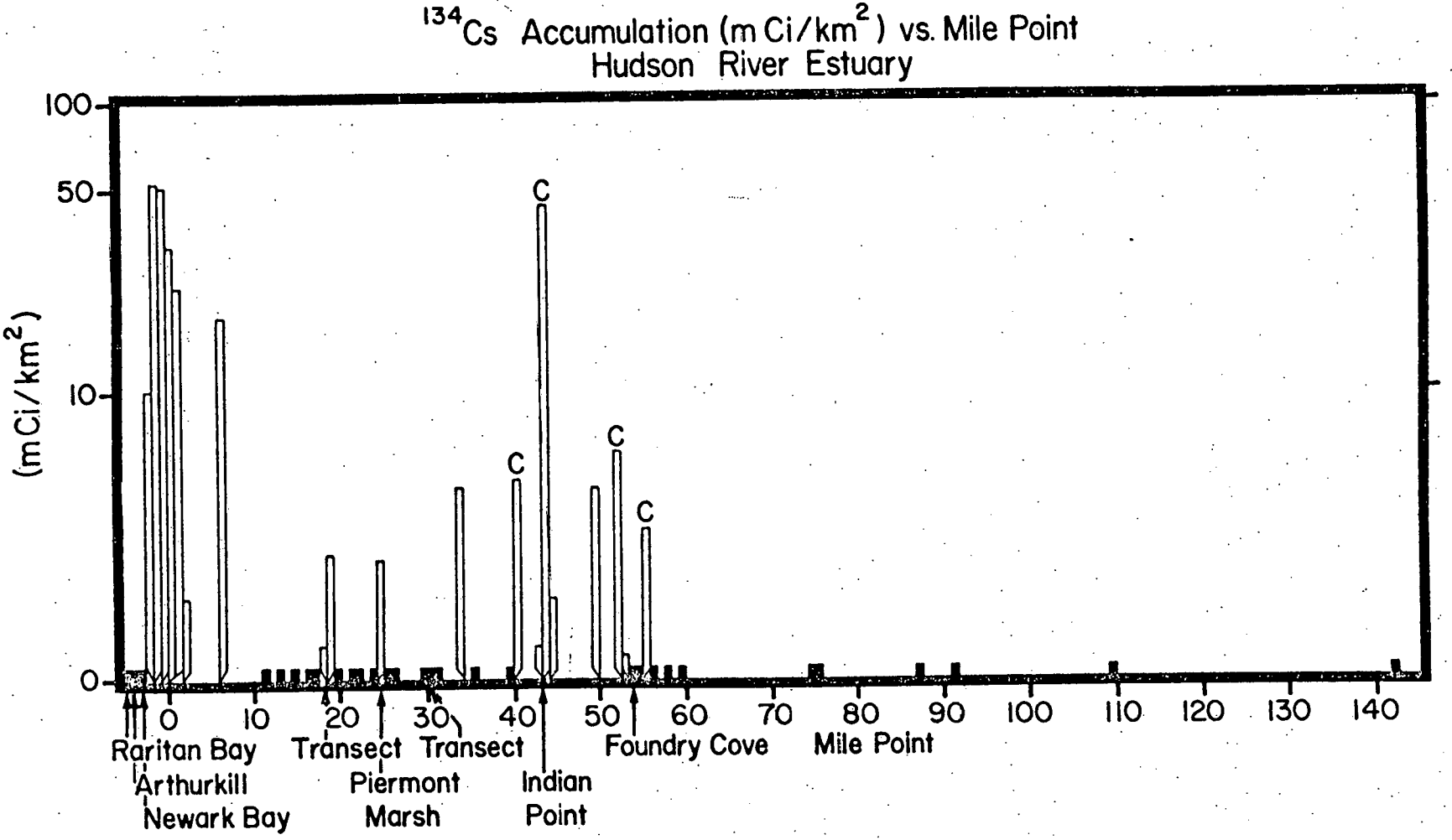
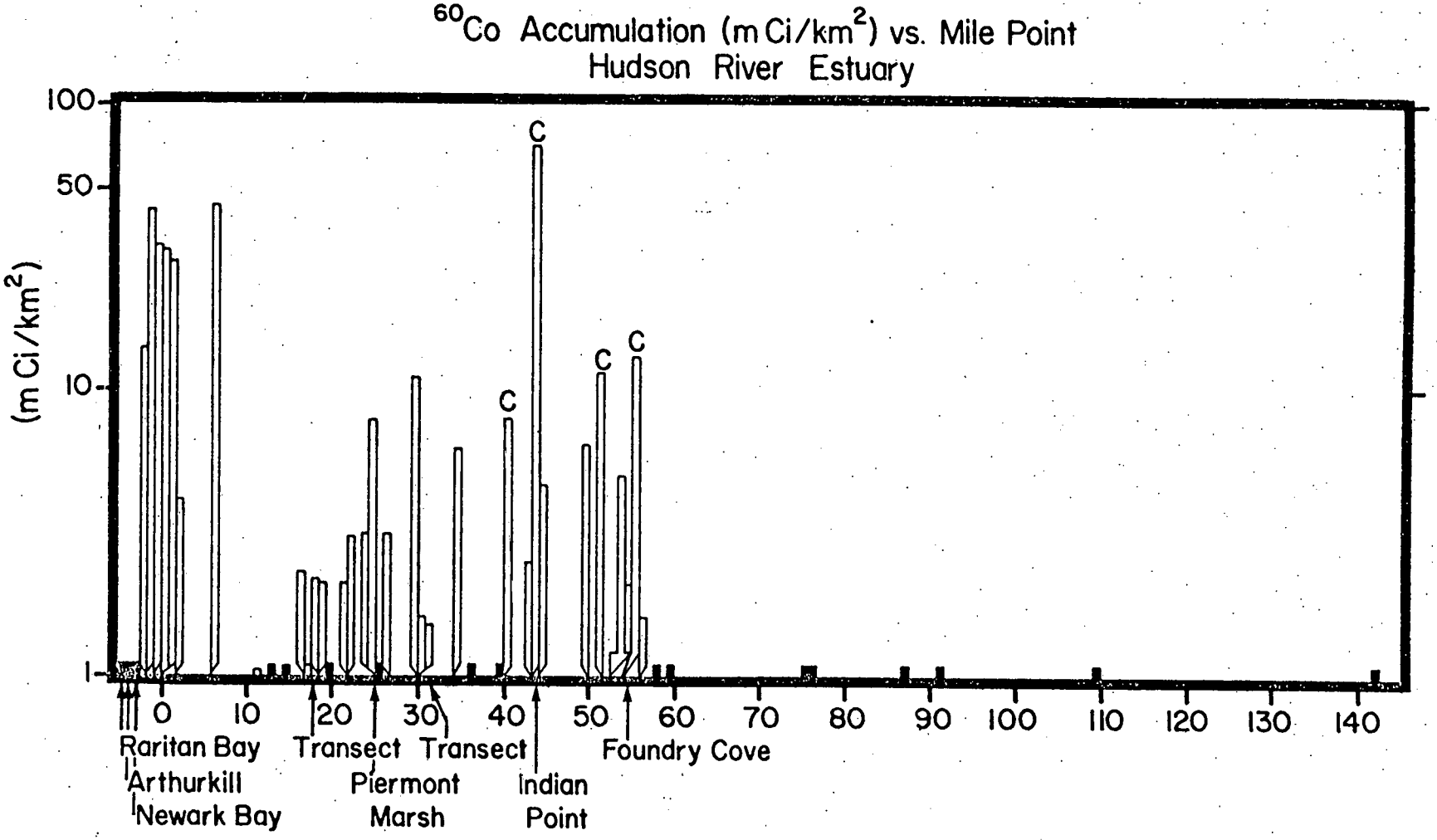

^{134}Cs Accumulation (m Ci/km^2) vs. Mile Point
Hudson River Estuary
(m Ci/km^2)
100
50
10
0
C
C
C
C
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
Raritan Bay
Arthurkill
Newark Bay
Transect
Piermont
Marsh
Transect
Indian
Point
Foundry Cove
Mile Point

FIGURE 12. Total ^{60}Co deposition (mCi/Km^2) along the axis of the Hudson Estuary. The greatest accumulation of ^{60}Co occurs in the inner harbor of New York and in coves along the estuary margin.


^{60}Co Accumulation (m Ci/km^2) vs. Mile Point
Hudson River Estuary
(m Ci/km^2)
100
50
10
1
C
C
C
C
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
Raritan Bay
Arthurkill
Newark Bay
Transect
Piermont
Marsh
Transect
Indian
Point
Foundry Cove

area of accumulation of these nuclides is well downstream from the original release point, and that the lack of significant integrated activities near the reactor site is not indicative of removal of these nuclides from the estuary. The most sensitive indicator of the present distribution of reactor gamma-emitting in the sediments is ^{60}Co, in part because of its half-life is longer than ^{134}Cs and also because there doesn't seem to be as much likelihood of desorption loss from higher salinity sediments as for ^{134}Cs.

IV. RADIONUCLIDE DISTRIBUTIONS IN MONO LAKE

A. Introduction

One of the observations which we have discussed in previous annual reports is the relatively constant concentration of dissolved fallout plutonium measured in a variety of natural water systems, including the Hudson estuary, Hudson River, New York Bight sea water, New York City tap water and the St. Lawrence Great Lakes. The range of suspended loads and ionic strengths of these waters is very large, yet the spread of dissolved ^{239,240}Pu concentrations is relatively small. It is important to establish what processes or chemical equilibria are responsible for maintaining the dissolved concentrations at the observed levels.

To help define what the critical processes are in determining the dissolved concentrations of plutonium in natural waters, we have begun a limited program of measurement of fallout plutonium and other radionuclide activities in a few unusual natural water systems which offer the potential of providing significant new insights. The data to be discussed here is for Mono Lake, a highly alkaline closed basin lake, which offers an unusual opportunity to examine the possibility that carbonate ions may play an important role in complexing plutonium in natural waters. As will be discussed below, we believe that we have made a significant advance in understanding the environmental behavior of plutonium as the result of this preliminary effort.

Mono Lake is located in a closed structural depression at the base of the Sierra Nevada escarpment in eastern California. The lake overlies a thick sequence of low-density lacustrine sediment and volcanic debris,

which fills the basin to a depth of ∿ 1000 meters. Mono Lake is the remnant of a much larger pluvial lake, and is fed by streams draining plutonic and metamorphic terrain in the Sierra Nevada to the west, as well as by numerous springs, including thermal springs. The most important feature of Mono Lake for the purpose of this research is its unusual chemical composition. Due to continual concentration by evaporation of the fresh waters supplying dissolved weathering products to the basin, the lake has developed a major element chemistry quite unlike sea water, or most other saline lakes. In Mono Lake, which has a total concentration of dissolved ions about twice sea water, the dominant anions balancing the cations in solution are bicarbonate and carbonate and not chloride as is true for sea water. The total carbonate ion concentration in Mono Lake is more than two orders of magnitude greater than in sea water. The pH of Mono Lake is ∿ 10, and the concentrations of Ca and Mg are very low as the result of precipitation of $CaCO_3$ at the lake margins and apparently the formation of authigenic magnesium silicate minerals.

From previous work under an AEC contract at Mono Lake about a decade ago, we have measured several radionuclides in the lake water and tributary streams. A summary of some of the characteristics of the lake, and an estimate of fallout ^{137}Cs delivery as of 1966 are given in Table 16. The measured activity of ^{137}Cs in Mono Lake in 1966 was very similar to that predicted from the regional fallout data and mean annual precipitation over the lake surface.

TABLE 16

Mono Lake ^{137}Cs Fallout Budget (1966)

Surface elevation (1965)	1948 m
Lake area (A) (1965)	198 Km^2
Lake volume (V) (1965)	3.7 Km^3
Average depth (V/A)	18.7 m
Drainage basin area	1,646 Km^2
Lake area/basin area	12 %
Precipitation over lake (annual average)	20 cm
Precipitation over basin (annual average)	43 cm
Deposition rate of ^{90}Sr over lake[a] (1966)	17 mCi/Km^2
Total deposition of ^{90}Sr (1966)	3370 mCi
Predicted lake ^{90}Sr activity (1966)	0.9 pCi/ℓ
Predicted lake ^{137}Cs activity [b]	1.35 pCi/ℓ
Measured ^{137}Cs Surface Water (1966)	0.93 pCi/ℓ
Measured ^{137}Cs Deep Water (1966)	1.59 pCi/ℓ

a) Deposition rate estimated from fallout data for sites in California, Oregon and Washington plotted as a function of mean annual precipitation.

b) Assuming $^{137}Cs/^{90}Sr = 1.5$.

B. Cesium-137 in Mono Lake Water (1978)

During May of this year we collected a few preliminary samples of water and sediment from Mono Lake to analyze for plutonium isotopes and other radionuclides. The field work was done in cooperation with Professor Doug Hammond and coworkers at the University of Southern California. Our major effort in the field was involved in collecting and processing water at the lake site to facilitate analysis of large samples.

Samples of water (∿ 240 liters) for ^{137}Cs analysis were transferred from the lake into portable processing tanks using a submersible pump and about 30 meters of hose. The lake water was then acidified with concentrated HCl to pH ∿ 2 and spiked with ^{134}Cs. After mixing with a large paddle and allowing up to several hours for equilibration. ammonium molybdophosphate (AMP), an ion exchange resin, was dispersed in the sample, stirred for about half an hour and allowed to settle overnight. The clear water was pumped away from the AMP in two steps, using a smaller container (80ℓ) as a second settling basin. The volume of AMP and water returned to the lab for analysis was several liters per sample.

Measurement of ^{137}Cs was made by gamma spectrometry using the AMP resin (after drying) in our standard sediment sample counting geometry. Yield estimates were made on the basis of measured ^{134}Cs activities.

Data for four large volume (∿ 240ℓ) samples are reported in Table 17. The measured activity of ^{137}Cs averaged ∿ 4.8 pCi/ℓ, with one of the samples consisting of a composite of portions of three other large samples which had been previously processed for removal of plutonium. Our primary goal was to obtain samples for plutonium analysis, and only two of the samples (C1 and C4) were processed for ^{137}Cs without having been previously stripped of plutonium by iron hydroxide precipitation.

TABLE 17

Mono Lake ^{137}Cs Activity in Water Samples

Field Notation	Sample Number	Volume (liters)	Amp activity (pCi/g)	Amp Amount (g)	Yield (%)	Activity (pCi/ℓ)
C1 (ML #5)	1368	240	13.21	44.3	51.5	4.74±0.27
C2 (ML#2)	1369	230	5.32	47.4	20.6	5.33±0.34
C3 (Composite ML#'s 1,3,4)	1370	240	5.45	42.0	24.0	3.97±0.24
C4 (ML #6)	1371	240	3.54	49.7	14.4	5.08±0.35

The measured activity of ^{137}Cs in 1978 of $\sim$ 4.8 pCi/ℓ is about three times that for 1966. The major fresh water stream tributaries of Mono Lake have been diverted from the basin since the 1940's to provide drinking water for Los Angeles, and as a result the lake level has decreased steadily over that period. We estimate the lake volume in 1978 to be less than half of that in 1966. Thus the primary explanation of the increase in ^{137}Cs activity in Mono Lake between 1966 and 1978 is probably evaporative concentration of the lake water. We plan to treat the budget of ^{137}Cs in this lake in more detail in a future report.

C. Plutonium isotopes in Mono Lake water

Our main purpose in collecting samples at Mono Lake during this preliminary survey effort was to analyze several large volume water samples for fallout plutonium isotopes to establish the approximate total concentration in the water column. We pumped water from about a meter below the surface into processing tanks on the shore and acidified the water to pH ∿3 with concentrated HCl. A yield tracer ^{242}Pu and iron carrier were added, the sample was stirred thoroughly with a paddle and allowed to equilibrate up to a few hours, and then iron hydroxide was precipitated by adding NH_4OH to raise the pH to ∿ 10. After the precipitate settled several hours, or in some cases over night, the supernatent was pumped away and the hydroxide precipitate transferred to a smaller settling tank, or directly into shipping containers. The large volume samples were generally processed first to remove plutonium, and then reacidified and AMP was added to concentrate ^{137}Cs. Samples of a range of total volumes (80ℓ to 240ℓ) were spiked with the same amount of ^{242}Pu to vary the spike to sample plutonium ratio. Data for plutonium isotopes in Mono Lake water are reported in Table 18.

The measured values of $^{239,240}Pu$ ranged from 19.0±0.7 to 22.9±7 pCi/1000ℓ, with an average value of ∿ 20 pCi/1000ℓ. The measured activity of ^{238}Pu ranged from 0.65±0.10 to 1.25±0.30 pCi/ℓ, with an average value of ∿ 0.9 pCi/1000ℓ. These activities of plutonium are dramatically different from those found for fallout plutonium in other lakes and estuaries, being almost two orders of magnitude greater than the activities we have measured in the Hudson. Clearly fallout plutonium is found in the water column of Mono Lake to a much greater extent than would be expected on the basis of fallout data from other continental water systems.

TABLE 18

Mono Lake Plutonium Activity in Water Samples

Field Notation	Sample Number	Volume (liters)	Yield (%)	$^{239,240}Pu$ (pCi/1000ℓ)	^{238}Pu (pCi/1000ℓ)	$^{238}Pu/^{239,240}Pu$
P 1 (ML #1)	1361	80	26.3	19.5±0.7	0.85±0.1	0.044±0.006
P 4 (ML #4)	1362	80	26.9	19.0±0.7	0.65±0.1	0.034±0.005
P 2 (ML #2)	1363	240	3.9	22.9±1.6	1.25±0.3	0.055±0.012
P 3 (ML #3)	1364	160	17.1	19.1±0.8	0.78±0.1	0.041±0.006

As discussed earlier, the activity of ^{137}Cs in Mono Lake is, to the first approximation, consistent with the regional fallout delivery data, assuming essentially all of the fallout arriving at the lake surface in precipitation is still in solution and no additional significant supply from the drainage basin streams. The ratio of $^{239,240}Pu$ to ^{137}Cs which we measured in Mono Lake water is about 0.004 (0.02 pCi/ℓ $^{239,240}Pu$/4.8 pCi/ℓ ^{137}Cs). This ratio is about 25% of the fallout delivery ratio, decay corrected to 1978, indicating that a major portion of the fallout plutonium is still in the water column in Mono Lake, and that probably both the sediments and the water column in the lake account for significant portions of the current inventory of fallout plutonium.

Our original purpose in measuring the present distribution of fallout plutonium in Mono Lake was to explore the possibility that carbonate ion complexing may be important in determining the amount of plutonium in the aqueous phase of natural water systems. We now have very dramatic evidence of increased mobility of fallout plutonium in a natural water system with high carbonate concentrations (∿100 times sea water) and a high pH (∿ 10). At present it is not possible to establish from our data what the primary factors are in maintaining plutonium in the water column of Mono Lake, but it should be possible to further constrain the major controlling processes with further research.

One interesting observation about the data reported in Table 18 is that the overall yield of sample P2 is considerably lower than for the other samples. For most samples we bubbled helium through the processing tanks after acidification to strip out carbon dioxide before adding NH_4OH to raise the pH and precipitate iron hydroxide. The amount of

total inorganic carbon in our large volume samples was very great (requiring ∿ 14 liters of concentrated HCl per 240 liter sample to neutralize the carbonate buffer capacity), and without removing CO_2 with a stripping gas, significant amounts probably remained in solution unless the sample was allowed to stand many hours before the pH was increased again. Sample P2 was not stripped of CO_2 with He, and the iron hydroxide precipitate formed was considerably less flocculent than for other samples. We can't establish a definite explanation for the unusually low yield of sample P2, but it may have been related to the presence of significant inorganic carbon (primarily as carbonate ion) during the latter stages of processing.

D. Cesium-137 in Mono Lake Sediments

We collected a few sediment samples during the period in which we were processing large volume water samples. At present, the only analytical data for the sediments which have been completed were determined by gamma counting. Data for ^{137}Cs for a core collected a few hundred meters off shore in Mono Lake are reported in Table 19. The specific activities of ^{137}Cs in the sediments are relative low, and are confined to the upper 3 cm of the core we analyzed. The integrated activity of ^{137}Cs per unit are in the core is a few percent of the fallout delivery rate.

Considering the concentrations of ^{137}Cs in the water and the relatively low activities in the sediments, it would appear that most of the fallout ^{137}Cs has been retained in the water column of Mono Lake. Such a finding is consistent with the general understanding of the factors which control the behavior of ^{137}Cs in natural waters, considering the relatively high dissolved solids concentration in Mono Lake. However, we cannot establish budget terms very precisely at present with our limited information for the sediments.

TABLE 19

Gamma-emitting Radionuclides in Mono Lake Sediments

Sample No.	Depth (cm)	Dry Weight (gm)	^{137}Cs (pCi/Kg)	^{40}K (pCi/g)
1373 A	0-1	4.93	132±50	15.8±1.3
1373 B	1-2	7.00	275±36	14.0±1.1
1373 C	2-3	5.29	260±51	12.7±1.1
1373 D	3-4	5.62	16±36	12.6±1.0
1373 E	4-5	5.00	60±42	12.8±1.1
	Grab[a] (∿ 0-6)	120	14±8	24.0±0.7

a) Sample retrieved from anchor in nearshore area:
Sediment much coarser and from a different locality than core 1373.

V. Intercomparison and Quality Control Program

The most common type of sample analyzed for transuranics in our laboratory over the last four years has been fine-grained estuarine sediments. We have discussed in our last annual report (COO-2529-3) the preparation and repeated analysis in our laboratory of a large sample of surface sediments in the Hudson estuary. Several other groups (WHOI, EML-DOE, OSU) have helped us in our intercalibration efforts by analyzing portions of our large Hudson estuary sample in their own laboratories. Some of that data was discussed in our last annual report (COO-2529-3, Table 15). We have continued to participate in intercomparison experiments during this past year, and have been in the process of analyzing several sediment samples provided us by WHOI, and the Environmental Measurements Laboratory of DOE. In addition, we have been analyzing other sediment standards.

We have almost completed our analysis of these samples, but will discuss them in our next annual report as part of a summary of all of our intercomparison sample data. At that time the results of the samples distributed by EML-DOE to a number of laboratories will be available.

APPENDIX A

Transport of Plutonium by Rivers*

H.J. SIMPSON, R.M. TRIER AND C.R. OLSEN

Transuranic Elements in the Environment

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COO-2529-9

*Lamont-Doherty Geological Observatory Contribution Number 0000.

TRANSPORT OF PLUTONIUM BY RIVERS*

H.J. Simpson, R.M. Trier and C.R. Olsen

INTRODUCTION

A number of nuclear power plants are now located on rivers or estuaries and many more probably will be in the future. The only major reprocessing facility currently operating in the United States is located on a small tributary of the Savannah River. Thus, knowledge of the transport pathways of transuranics in rivers is essential for proper monitoring of the routine operations of these facilities and for developing plans for dealing with any abnormally large releases of transuranics which might occur.

In principle, rivers can carry plutonium and other transuranics either in solution or as part of the suspended load. These two transport pathways are probably strongly coupled by some type of quasi-equilibrium partioning between the two phases, and thus cannot really be considered separately. As with many elements which are reactive in natural waters, the classifications of "dissolved" and "particulate" plutonium are based largely on operational procedures such as whether or not material will pass through a filter of a certain nominal pore size. The actual species distribution of plutonium in natural waters is probably some kind of continuum from small molecular weight complexes through silt or sand-sized particles. To further complicate matters, particles may be transported in suspension or as bed load in a stream, or accumulated in depositional environments, and either buried or resuspended at a later time.

There have been relatively few field studies of point source releases of plutonium to river systems. Three areas in the eastern United States which have received such attention are the Savannah River and its tributary downstream of the reprocessing facility in South Carolina (Hayes & Horton, 1979), the Miami River (a tributary of the Ohio River) downstream of Mound Labs in Ohio (Sprugeland & Bartelt, 1978) and streams near Oak Ridge, Tenn. These river systems

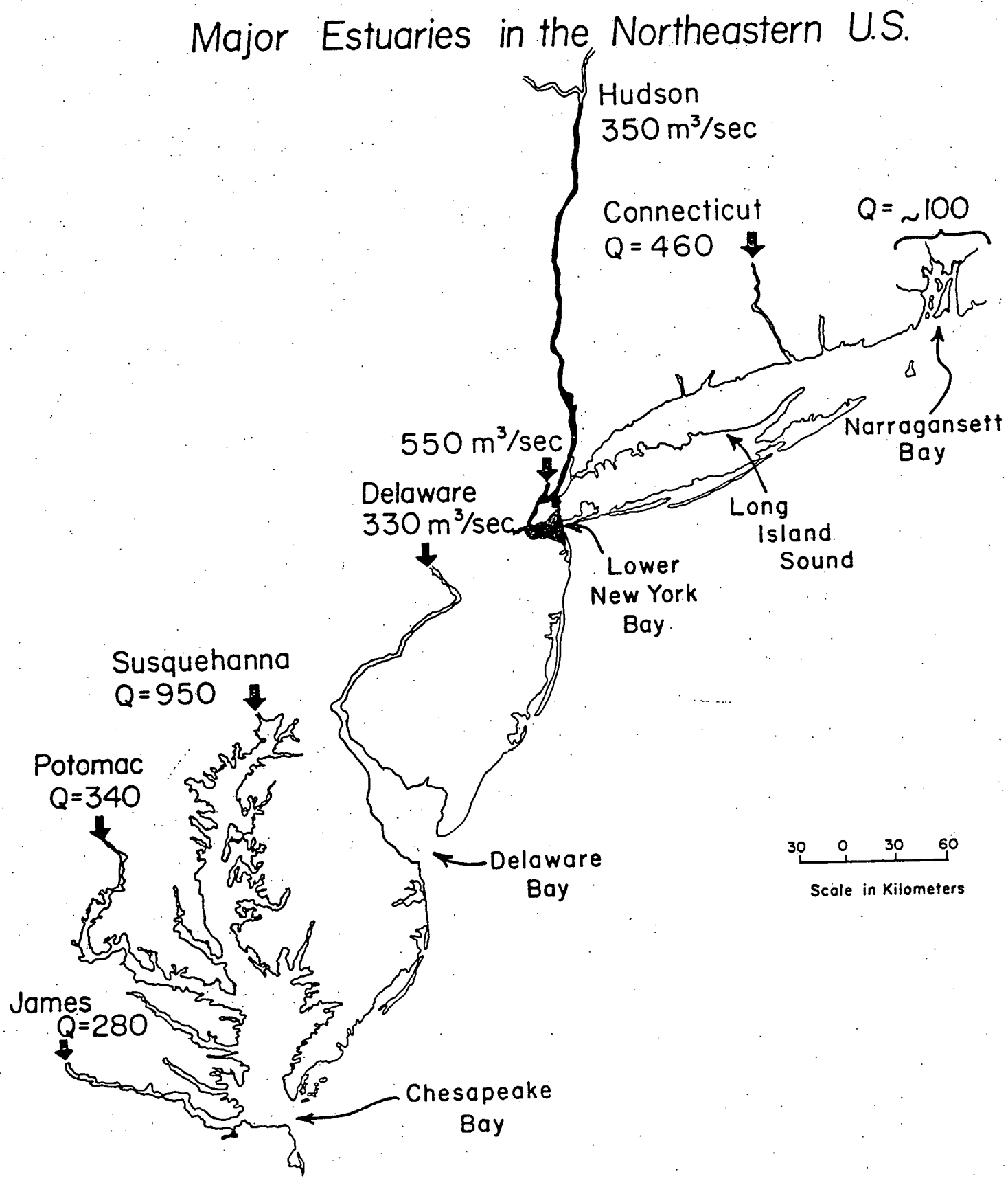
are the focus of ongoing research programs which should provide considerable information about the transport by rivers of plutonium derived from point sources. The approach taken here will be to discuss the distribution of fallout plutonium in a few natural systems, including the Hudson River and estuary, and attempt to derive some first-order principles by which the transport pathways of plutonium in other river systems could be predicted. The Hudson estuary is now the site of three nuclear reactors, and at least half a dozen other units which are planned for the next two decades.

Plutonium in the Hudson River Estuary

The Hudson River discharges into one of the large estuarine systems which dominate much of the coastal environment of the northeastern United States (Figure 1). The Hudson has an unusually long, narrow reach of tidal water (>250 km), most of which is usually fresh. Saline water intrudes only about 40 km from the coast line during seasonal high fresh water discharge, and reaches as far inland as 120 km during summer and early fall months of drought years. The near-surface suspended load of the Hudson is relatively low (10-20 mg/l), as it is for nearly all of the larger rivers in the northeastern United States, except during maximum spring runoff and following major storms.

From studies of the distribution of fallout nuclides and gamma-emitting nuclides released from Indian Point, the patterns of suspended particle transport and recent sediment accumulation in the Hudson have been described (Simpson et al., 1976; Olsen et al., 1978, Simpson et al., 1978). Much of the Hudson has relatively little net accumulation of fine particles, while a few areas, such as marginal coves and especially New York harbor, account for a major fraction of the total deposition of fine particles containing fallout and reactor nuclides. The zone of major sediment accumulation is more than 60 km downstream of the reactor site, and the time scale of transport of fine particles labeled with reactor nuclides

FIGURE 1. Location map of the major estuarine systems of the northeastern United States.


Major Estuaries in the Northeastern U.S.
Hudson
350 m³/sec
Connecticut
Q = 460
Q = ~100
Narragansett
Bay
550 m³/sec
Delaware
330 m³/sec
Long
Island
Sound
Lower
New York
Bay
Susquehanna
Q=950
Potomac
Q=340
Delaware
Bay
James
Q=280
Chesapeake
Bay
30 0 30 60
Scale in Kilometers

from the release area to burial in the harbor sediments varies from probably less than a month to years. At present there is no evidence in Hudson sediments, including New York harbor, of releases of reactor $^{239,240}Pu$ which are resolvable in the presence of the burden of fallout $^{239,240}Pu$. Thus the current distribution of $^{239,240}Pu$ in the Hudson appears to be governed primarily by the delivery of global fallout to the drainage basin mostly more than a decade ago and the transport processes which have occurred since delivery.

In Table 1 are given concentrations of $^{239,240}Pu$ in large volume Hudson water samples which have had the suspended load removed by settling for 24-48 hours or by passing through a continuous flow centrifuge followed by a 0.45 micron filter. The range of observed values for samples collected in 1975 and 1976 was 0.12 to 0.88 fCi/1, with the median value about 0.3 fCi/1. The current transport of $^{239,240}Pu$ in the "dissolved" phase in the Hudson can be estimated to be $\sim 5 \times 10^{-3}$ Ci, assuming a concentration of 0.3 fCi/1 and a mean annual river discharge of 550 m^3/sec. This represents somewhat less than 0.01% of the fallout burden of $^{239,240}Pu$ ($\sim$ 80 curies) in the soils of the Hudson drainage basin ($\sim 3.5 \times 10^4$ km^2). Soluble phase release of fallout $^{239,240}Pu$ from Hudson soils thus has a half time of the order of 10^4 years, and supplies an insignificant amount of dissolved $^{239,240}Pu$ to the coastal ocean, compared with that transported onto the shelf from surface waters of the deep ocean.

The suspended load activity of $^{239,240}Pu$ for two of the Hudson samples listed in Table 3 averaged $\sim$ 20 pCi/kg (18.9 $\pm$ 0.9 and 23.4 $\pm$ 1.0 pCi/kg). The distribution coefficient ((K_D) of $^{239,240}Pu$ between the dissolved phase and suspended particles for those two samples was $\sim 1.5 \times 10^{-5}$. Thus the transport of $^{239,240}Pu$ by suspended particles equals that in the dissolved phase when the concentration of suspended particles is $\sim$ 15 mg/1, a value which is reasonably typical of moderate and low fresh water flow periods in the Hudson. During periods of higher suspended load, the transport of $^{239,240}Pu$ in the Hudson is predominantly on particles.

TABLE 1

Dissolved ^{239,240}Pu in Continental Waters

Location [a]	fCi/l	Volume (l)	Sample #
Hudson River (mp 61) (S)	0.32±0.01	660	1177B
Hudson Estuary (mp 19) (S)	0.88±0.07	625	1175B
Hudson Estuary (mp 18) (F)	0.47±0.03	490	1176
Hudson River (mp 47) (S)	0.27±0.02	570	1222A
Hudson Estuary (mp 19) (S)	0.12±0.02	570	1221A
Hudson Estuary (mp -8)(S)	0.15±0.02	570	1223A
Hudson Estuary (mp 24) (F)	0.30±0.03	1500	1115
New York Bight (S)	0.25±0.03	380	1179B
New York Bight (U)	0.59±0.09	660	1102
New York Bight (U)	0.68±0.05	660	1112
New York Bight (U)	0.68±0.09	660	1107
New York Bight (U)	0.68±0.09	660	1113
New York Bight (U)	0.91±0.14	660	1111
New York Bight (U)	0.95±0.14	660	1109
New York Bight (U)	1.18±0.14	660	1104
New York City Tap water (1973-1975)[b]	∿ 0.3		
Lake Ontario (1973)[c]	∿ 0.3		
Great Lakes (1972-73)[d]	∿ 0.5		

a) The pretreatment procedure of the large volume samples is indicated by one of three letters:
   (U) - unfiltered
   (F) - filtered after passing through a continuous flow centrifuge.
   (S) - suspended particles allowed to settle, usually for 24-48 hours before the clarified water was transferred to another tank for processing.

b) Bennett, B.G., 1976, USAEC Rep. HASL-306.
c) Farmer, Bowen, Noshkin and Gavini (COO-3568-6).
d) Wahlgren, M.A. and Marshall, J.S., 1975, IAEA-SM-198/39, 240 (Table VI).

From the quantities of material dredged annually from New York harbor ($\sim 2 \times 10^6$ T) the downstream transport of particles by the Hudson must be about a factor of four higher than indicated by multiplying typical near-surface suspended load concentrations times mean annual flow. The "extra" transport of particles is probably accomplished by some combination of very high suspended loads coinciding with the highest fresh water flow rates and bed load transport, which in the Hudson appears to consist largely of resuspension and deposition of fine particles in the lowest meter of the water column on the time scale of a tidal cycle. Thus in the Hudson the total downstram transport of $^{239,240}Pu$ is approximately a factor of four greater than that in the soluble phase, indicating a half time for removal of fallout $^{239,240}Pu$ from the drainage basin, largely on particles, of $\sim 10^3$ years. Similar calculations for the Savannah River (Hayes and Horton, 1979) and the Greater Miami River (Sprugel and Bartelt, 1978) suggest drainage basin removal times of $\sim 2 \times 10^4$ years and 2×10^3 years, respectively. Again, this supply term to the coastal ocean is not significant relative to advection of deep ocean fallout $^{239,240}Pu$ onto the shelf. In the case of the Hudson, most of the delivery of $^{239,240}Pu$ on particles to the coastal ocean is accomplished by dumping of dredge spoils rather than estuarine discharge of suspended particles.

Plutonium in the New York Bight

The concentrations of dissolved $^{239,240}Pu$ in the coastal waters off the New York City area are 2-3 times that in the Hudson (Table 1). The suspended loads in the New York Bight are almost two orders of magnitude lower than in the Hudson and fine-grained sediments in the Bight have activities of $^{239,240}Pu$ comparable to those in the Hudson. Thus the transport of $^{239,240}Pu$ in the shelf environment appears to be largely in the dissolved phase, in contrast to the situation in the Hudson River and estuary.

Plutonium in Other Fresh Waters

Data for the concentration of $^{239,240}Pu$ in New York City tap water (Bennett, 1976) are available for the period 1973-1975 (Table 1). The water supply for New York City is derived from tributaries of the Hudson and Delaware Rivers. The activities ranged from 0.08 to 0.60 fCi/1, with a mean value of $\sim$ 0.3 fCo/1 (about 2% of the average rain activities during the same period). The range and mean value of the tap water $^{239,240}Pu$ concentrations are almost identical with the values observed for the Hudson River and estuary.

Farmer, et al., 1973 have reported $^{239,240}Pu$ activities in Lake Ontario (Table 1) that are in the same range as the data discussed here for the Hudson and New York Bight. During the period 1971 to 1973 the average $^{239,240}Pu$ activity for the entire lake declined from $\sim$ 0.8 fCi/1 to $\sim$ 0.3 fCi/1. The average $^{239,240}Pu$ activity in all five Great Lakes (Wahlgren and Marshall, 1975; Wahlgren et al., 1976) during 1972-1973 was $\sim$ 0.5 fCi/1 (Table 1).

Transport of Fallout Plutonium to the Oceans

The data available indicate that the range of variation of soluble phase $^{239,240}Pu$ in fresh waters is relatively small. The transport by rivers of fallout $^{239,240}Pu$ in "solution" can thus be estimated relatively easily, based only knowing the rate of fresh water discharge. The concentrations in fresh waters appear to be "buffered" to some extent by the large reservoir of fallout $^{239,240}Pu$ in soils, and the relative uniformity of the specific activity of $^{239,240}Pu$ on soil particles and river suspended particles ($\sim$ 20 pCi/kg). The distribution of fallout $^{239,240}Pu$ between soluble phases and particles in rivers can probably be approximated by a partition coefficient of $\sim 10^{-5}$. The total delivery of dissolved fallout $^{239,240}Pu$ to the oceans by rivers is probably about 10 Ci/year, assuming a discharge rate for all rivers of $\sim 10^6$ m^3/sec, and a concentration of $\sim$ 0.3 fCi/1. Since the global average of suspended load in rivers is about 600 mg/1, the transport of fallout

$^{239,240}Pu$ by rivers will clearly be dominated by particles. Assuming the specific activity of all river suspended matter is similar to surface soils, the total delivery of fallout $^{239,240}Pu$ to the ocean by rivers is $\sim 5 \times 10^2$ Ci/year, about 50 times the soluble phase delivery. The specific activity of particles in rivers with very high suspended loads is probably somewhat lower, due to the presence of more large silt and sand sized particles, so a more reasonable estimate for the total annual delivery of fallout $^{239,240}Pu$ to the ocean by rivers is probably $1\text{-}5 \times 10^2$ Ci.

Transport of Plutonium by Rivers Added at Point Sources

The distribution of fallout $^{239,240}Pu$ provides information about the partitioning of plutonium between soluble and suspended particle phases in rivers, and about the processes by which transuranic transport occurs in rivers. For point source addition of plutonium to a river, the most important transport pathway appears to be binding to the suspended load and the mobile portions of the fine-grained sediments, and downstream movement with the fine particles. Since the effective concentrations of suspended particles, including the upper few centimeters of fine-grained sediment, in a river will be far greater than 10-15 mg/l, the dominant transport of plutonium would be in association with particles. The kinetics and downstream transport pathways of a particular river system will depend on many factors, such as the frequency and duration of deposition and resuspension episodes for the suspended particles. In the tidal reach of the Hudson, the downstream movement of fine particles tagged with reactor nuclides is distributed such that some particles require several years to move 50 km while others probably require considerably less than a few months. In other rivers such as the Columbia above tidal influence, the downstream transport of some portions of the suspended load is probably similar to the rate of water

transport, while other portions of the suspended particles are trapped for long periods, perhaps indefinitely, behind dams.

The distribution of fallout nuclides can provide valuable information about which areas of the bottom in a river system are actively scoured, and which portions accumulate fine-grained sediments rapidly, but probably cannot provide a very detailed picture of the kinetics of downstream transport of fine particles. A tracer added relatively uniformly to the earth's surface, as was weapons testing fallout, is not very powerful for providing such information. Fortunately, the river systems for which the kinetics of fine particle movement are most important to understand for predicting transport of transurancs are also ones for which point source tracers are available. Many nuclear power plants and reprocessing facilities release fission or activation products in sufficient quantities during normal operations to be useful as indicators of fine particle transport pathways. The behavior of these radioactive tracers cannot be expected to be identical to transuranics in river systems, but some of them are associated with particles sufficiently to provide very valuable information about the patterns and kinetics of movement and accumulation of fine particles of most importance for evaluating the transport pathways of point source releases of transuranics.

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