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PLUTONIUM AND CESIUM RADIONUCLIDES IN THE HUDSON RIVER ESTUARY

Annual Technical Progress Report

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DISCLAIMER

SUMMARY

Plutonium and Cesium Radionuclides in the Hudson River Estuary and Other Environments

CONTRACT EY-76-S-02-2529

The major purpose of this project is to describe the behavior and distribution of plutonium in the Hudson River estuary and to establish the important transport pathways for transuranics in estuarine environments. To accomplish this goal a substantial number of cores, grab samples, suspended particle and water samples have been collected throughout the salinity range of the Hudson, and in a variety of depositional environments. The distribution of Cs^{137} has been shown in this project to be useful in defining the general outlines of the plutonium distribution pattern and this similarity will continue to be exploited.

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 $Pu^{239,240}$ within the system. The feasibility of using $Pu^{238}/Pu^{239,240}$ ratios as an indicator of transuranic reactor releases has been established and will be explored in more detail. A limited number of measurements in other aqueous environments, such as the Delaware estuary and near two other reactors, have been made and will be made to help formulate the most important environmental transport pathways of transuranics in such systems.

ABSTRACT

We have obtained a large set of sediment cores from the Hudson estuary through much of the ambient salinity range. A number of core sections and samples of suspended particles have been analyzed for Cs^{137}, Cs^{134} and Co^{60} by direct gamma counting, and for $Pu^{239,240}$ and Pu^{238} by alpha spectrometry. The distribution of both Cs^{137} and $Pu^{239,240}$ indicates rapid accumulation in marginal cove areas, and especially in the harbor region adjacent to New York City. The distributions of both Cs^{137} and $Pu^{239,240}$ are similar in surface sediments and with depth in cores, but there are deviations from the fallout ratio due to (1) addition of reactor Cs^{137} and (2) loss of Cs^{137} from the particle phases at higher salinities. Measureable amounts of reactor-derived Cs^{134} and Co^{60} are found in nearly all sediment samples containing appreciable Cs^{137}, between 15 km upstream of Indian Point and the downstream extent of our sampling, 70 km south of the reactor. Accumulations of $Pu^{239,240}$ in New York harbor sediments are more than an order of magnitude greater than the fallout delivery rate. The most likely explanation is accumulation of fine particles in the harbor which have been transported from upstream areas of the Hudson. Our evidence so far indicates that Indian Point is probably not a significant source of $Pu^{239,240}$ or Pu^{238} compared with the fallout burden of these nuclides already in the sediments.

PLUTONIUM AND CESIUM RADIONUCLIDES IN THE HUDSON RIVER ESTUARY

ERDA-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 the Indian Point nuclear reactor site, we have established that an appreciable amount of fine-grained sediments bearing reactor nuclides accumulate preferentially in New York harbor, more than 60 km downstream of the release area. Thus any monitoring program for release from Indian Point should take into account this aspect of the transport and accumulation of fine particles in the Hudson. Although there is considerable excess of $Pu^{239,240}$ and Pu^{238} in New York harbor sediments over the fallout delivery rate, our present evidence indicates that if any of this plutonium is reactor-derived, the amounts are not detectable in the presence of the fallout plutonium burden. Currently our explanation of the rapid accumulation of $Pu^{239,240}$ in New York harbor is that the fine-grained sediments from a considerable area upstream are moved toward the harbor and deposited after many episodes of sedimentation and resuspension.

From our research in the Delaware, there does not appear to be a zone of major accumulation of fine-grained sediment analogous to that which exists in the Hudson. The inventory of fallout $Pu^{239,240}$ in Delaware sediments is generally a small fraction of the fallout delivery rate. Thus a reactor sited on the shore of Delaware Bay or estuary might be expected to have a zone nearby with elevated levels of reactor release in the sediment, as we would predict for Artificial Island from our sampling there, but probably would have most of its releases transported and dispersed on fine particles over a large area. The Delaware estuary system does not appear to retain fine particles in Delaware Bay nearly so efficiently as occurs in New York harbor.

Sediment near the Oyster Creek and Millstone Point reactors both have relatively high activities of Co^{60}, and releases of Pu^{238} have occurred at Millstone Point which are readily measureable in the sediment. Thus the ratio of Pu^{238} to $Pu^{239,249}$ does appear to be useful as an indicator of reactor-released transuranics in sediment in some situations.

PLUTONIUM AND CESIUM RADIONUCLIDES IN THE HUDSON RIVER ESTUARY

H.J. Simpson and R.M. Trier

INTRODUCTION

This report summarizes our activities during the third contract year of EY-76-S-02-2529, provides a comprehensive data listing of radionuclide measurements made during all three years of the contract, and relates our findings up to now to several research objectives. One of our primary research goals 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 direct input to the Hudson by precipitation occurring during the early to mid 1960's. The problem of establishing the distribution of fallout plutonium 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.

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. 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 (Cs^{137}) and reactor releases (Cs^{137}, Cs^{134} and Co^{60}), 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 our 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.

In addition to analysis of sediment cores, we have collected and analyzed samples of suspended particles and large volume filtered water samples from the Hudson. The purpose of these measurements has been to help define some of the processes by which plutonium is transported in the Hudson and other river-estuarine systems.

Using a similar approach to our sediment analysis program in the Hudson, we collected and analyzed a limited number of samples from the Delaware estuary during the second contract year. The Delaware does not currently have a significant source of radionuclides from nuclear generating stations, although it will in the near future. Thus the distribution of plutonium and radiocesium in the Delaware will provide useful insights to the processes controlling the distribution in a large estuary of radionuclides derived only from fallout.

We have also sampled and analyzed sediments in the immediate proximity of two other reactors, Oyster Creek, New Jersey, and Millstone Point, Connecticut, to help characterize the isotopic composition of transuranic releases (if any) from other operating power reactors.

Intercomparison results from an exchange of samples with Vaughan Bowen's laboratory at Woods Hole and the Health and Safety Laboratory of ERDA are presented, as well as a brief summary discussion of river transport of plutonium.

Some of the conclusions of our research on the Hudson are included in appendices to this report, one of which has been published recently in SCIENCE, one presented at a USSR-USA symposium in May 1977, and two others which have been submitted to journals for publication.

SAMPLE COLLECTION

We have continued our program of sediment coring in the Hudson estuary through the third contract year. During the first year, we discovered that the sediments of New York harbor contained activity levels of reactor nuclides as high as sediment samples from the immediate vicinity of the reactor, about sixty kilometers upstream. As the result of this discovery, during the second and third years we collected a number of additional cores in the harbor. Most of these were gravity cores up to ∿ 70 cm in length, but a few were piston cores up to 3 meters in length.

During the last two contract years we concentrated our sediment sampling in the Hudson on the harbor region primarily because that area appeared to be the zone of greatest sediment and radionuclide deposition. This initial impression has been documented thoroughly and there is now little doubt that the primary zone of accumulation of long-lived releases from Indian Point is not the vicinity of the power plant, but is found sixty kilometers downstream in New York harbor.

During the past year we began collecting large volume water samples in the Hudson for measurement of both dissolved and suspended phase plutonium. We used a continuous flow centrifuge, followed by a high volume filtering system to separate suspended particles from water samples up to 1500 liters in total volume. We are now comparing results from this approach to those for large suspended particle samples obtained by gravity settling of water samples. Previously, we have used borrowed equipment from other research programs to obtain these samples, but have now begun to acquire our own equipment for future large volume water and suspended particle sample collection.

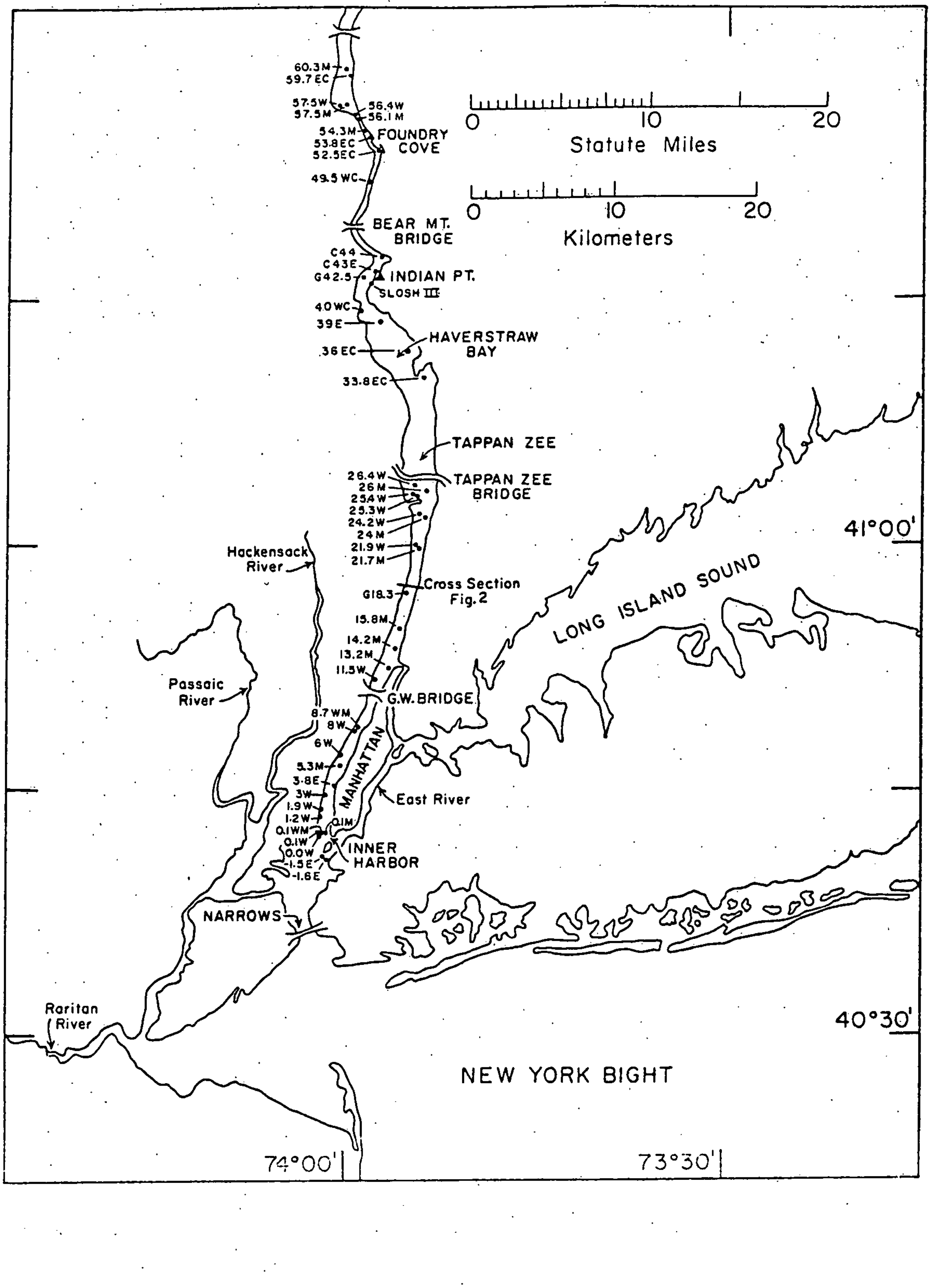
As mentioned in the INTRODUCTION, we collected sediment samples in the Delaware estuary, in addition to those in the Hudson. During the spring of 1976 we took gravity cores at 10 sites covering a significant portion of the salinity gradient of the Delaware. This field effort was made in cooperation with the University of Delaware's Department of Geology using a small research vessel operated by the College of Marine Studies.

We also participated in a substantial field sampling effort in Barnegat Bay during August of 1976. The primary support of the field program at Barnegat Bay was through a separate contract (E(11-1)2185), but much of the analytical work on those samples has involved the gamma spectrometry equipment at Lamont which was purchased under this contract and an EPA contract (R803113-01). The field program at Barnegat Bay was designed to evaluate the transport processes of radionuclides away from the nuclear power plant at Oyster Creek, New Jersey. We have also now collected a few samples of sediment from the immediate vicinity of a nuclear power plant at Millstone Point, Connecticut.

RESULTS - HUDSON ESTUARY SEDIMENTS

The results of our radionuclide measurements by gamma spectrometry for all three years of this contract are summarized in Table 1. The data are listed in geographical order along the axis of the Hudson, beginning with New York harbor (mile point -1.6) and extending upstream to mile point 91.3. The locations of most of the core samples are shown in Figure 1. Many of the harbor gravity core samples (between mp -1.6 and mp 6.0) contain Cs^{137} throughout the entire length of the core (40-70 cm) below the surface, while others drop to background levels within the core (25-60 cm). The only long piston

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
BEAR MT. BRIDGE
C44
C43E
G42.5
INDIAN PT.
SLOSH III
40WC
39E
36EC
HAVERSTRAW BAY
33.8EC
TAPPAN ZEE
TAPPAN ZEE BRIDGE
26.4W
26M
25.4W
25.3W
24.2W
24M
21.9W
21.7M
G18.3
Cross Section Fig. 2
15.8M
14.2M
13.2M
11.5W
G.W. BRIDGE
Hackensack River
Passaic River
8.7WM
8W
6W
5.3M
3.8E
3W
1.9W
1.2W
0.1WM
0.1M
0.1W
0.0W
-1.5E
-1.6E
MANHATTAN
East River
INNER HARBOR
NARROWS
Raritan River
LONG ISLAND SOUND
NEW YORK BIGHT
0
10
20
Statute Miles
0
10
20
Kilometers
41°00'
40°30'
74°00'
73°30'

Table

Gamma-emitting Radionuclides in Hudson Estuary Sediments

Location (Mile Point)	Sample #		Depth (cm)	Dry Weight (gm)	ACTIVITIES (a) (pCi/kg) Cs^{137}	Cs^{134}	Co^{60}	K^{40} x 10^3
-1.6 E	1051	A	0-5	80	450±26 (b)	12±11	51±23	15.7±0.7
		B	5-12	83	620±26	56±13	50±18	16.6±0.6
		C	12-20	82	1210±42	115±19	110±23	16.6±0.7
		D	20-25	99	470±25	37±12	43±19	16.6±0.7
		E	25-30	73	540±24	32±11	-13±15*	15.7±0.6
		F	30-35	63	390±18	20±8	44±15	15.8±0.6
		G	35-40	73	615±36	38±16	69±23	18.0±0.8
		H	40-45	51	535±95	-2±15*	54±35	17.9±0.9
		I	45-50	68	610±31	3±12	47±21	17.4±0.7
		J	50-55	112	29±11	-8±7*	-2±13*	15.5±0.6
		K	55-60	118	-19±9*	-9±7*	11±14	14.6±0.5
		L	60-65	138	27±14	-6±15*	10±20	11.5±0.4
		M	65-73	98	455±24	11±10	24±18	16.2±0.6
-1.5 E	1046	A	0-5	60	440±21	32±10	30±16	15.6±0.6
		B	5-12	58	430±26	18±12	30±22	15.1±0.7
		C	12-30	82	620±26	19±11	68±17	17.5±0.7
		D	20-25	86	440±23	13±10	56±15	15.7±0.6

				ACTIVITIES[(a)] (pCi/kg)			
Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	Cs^{137}	Cs^{134}	Co^{60}	K^{40} x 10^3
	E	25-30	54	390±28	175±26	28±25	18.4±0.8
	F	30-35	73	570±24	27±11	31±14	17.9±0.7
	G	35-40	57	605±24	43±11	37±17	15.7±0.6
	H	40-45	77	685±28	52±12	47±16	16.0±0.6
	I	45-50	67	635±30	36±13	43±19	16.0±0.7
	J	50-55	94	1140±50	135±25	105±25	16.9±0.8
	K	55-60	77	865±33	110±19	73±18	15.7±0.6
P-1.5 E	1048 A	0-10	61	260±21	14±15	12±13	12.7±0.6
	B	10-20	49	455±30	40±21	25±17	15.3±0.7
	C	20-30	53	345±23	3±14	10±14	14.7±0.7
	D	30-40	56	420±34	12±19	13±20	15.5±0.8
	E	40-50	56	350±26	60±18	115±18	13.0±0.6
	F	50-60	47	620±32	64±18	89±17	14.9±0.7
	G	60-70	48	1090±44	105±23	120±19	15.5±0.7
	H	70-80	51	950±36	140±22	95±15	14.4±0.6
	I	80-90	44	1080±42	150±23	105±15	14.0±0.7
	J	90-100	57	815±42	115±24	57±20	14.6±0.7

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	ACTIVITIES(a) (pCi/kg)			
				Cs^{137}	Cs^{134}	Co^{60}	K^{40} $x\ 10^3$
	K	100-110	48	635±33	54±18	58±17	14.9±0.7
	L	110-120	52	495±30	64±19	9±14	14.3±0.7
	M	120-130	74	290±18	36±12	8±11	11.9±0.5
	N	130-140	62	340±25	44±15	20±14	13.3±0.6
	O	140-150	66	350±18	-1±9	24±10	13.8±0.5
	P	150-160	53	365±28	-4±12	20±17	14.2±0.7
	Q	160-170	81	350±30	-16±12	21±17	15.1±0.7
	R	170-180	64	395±21	10±11	24±12	12.7±0.5
	S	180-190	66	350±20	-6±8	15±11	13.0±0.5
	T	190-200	79	200±22	-7±11	18±14	14.7±0.6
	U	200-210	75	470±25	12±15	18±12	14.1±0.6
	V	210-220	62	425±27	-10±11	26±14	13.6±0.6
	W	220-230	60	390±22	12±15	9±12	12.8±0.6
	X	230-240	91	335±18	13±12	28±10	12.6±0.5
	Y	240-250	52	500±27	-1±12	28±14	13.6±0.6
	Z	250-260	104	46±11	-5±8	6±9	12.3±0.5
0.1 W	1066 A	0-5	54	645±36	64±24	40±29	17.6±0.9
	B	5-10	80	890±28	63±12	60±16	19.7±0.7

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	ACTIVITIES[(a)] (pCi/kg)			
				Cs^{137}	Cs^{134}	Co^{60}	K^{40} x 10^3
	C	10-18	75	1040±50	87±22	110±33	18.7±0.9
	D	18-25	80	1480±40	125±18	170±21	19.4±0.7
	E	25-30	83	1260±40	130±19	175±22	17.1±0.6
	F	30-35	82	2310±70	300±38	205±32	18.4±0.8
	G	35-40	76	1210±60	130±25	90±31	19.0±0.9
	H	40-45	70	1140±50	110±21	84±26	17.8±0.8
	I	45-53	93	685±27	27±10	71±18	16.6±0.6
	J	53-60	106	105±14	7±8	38±15	17.7±0.6
	K	60-65	108	3±11	13±8	12±13	17.3±0.6
	L	65-70	109	2±12	6±8	15±14	18.4±0.6
0.1 WM	1002 A	0-5	77	1260±38	230±33	190±18	18.2±0.7
	B	5-10	68	1800±60	395±56	300±27	18.3±0.8
	C	10-15	74	2000±56	480±58	245±18	17.5±0.7
	D	15-20	66	960±44	135±33	65±20	17.0±0.8
	E	20-25	84	1030±40	175±33	125±19	16.3±0.7
	F	25-30	92	570±28	-1±10*	59±16	15.3±0.6

				ACTIVITIES(a) (pCi/kg)			
Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	Cs^{137}	Cs^{134}	Co^{60}	K^{40} x 10^3
	G	30-35	63	925±49	22±33	100±28	16.9±0.8
	H	35-40	63	1190±40	75±24	152±19	19.4±0.7
	SLOSH V	0-5		555±13	53±9	43±6	14.8±0.3
1.9 W	1082 A	0-5	58	1180±35	100±16	105±23	17.9±0.7
	B	5-10	63	375±19	12±9	64±19	15.2±0.6
	C	10-15	84	550±24	1±9	20±17	15.7±0.6
	D	15-20	85	57±13	-13±9*	8±15	17.5±0.6
	E	20-25	112	16±12	4±8	-6±16*	17.6±0.6
	F	25-30	99	16±13	0±9	14±18	16.5±0.6
	G	50-55	97	-1±11*	6±9	8±15	16.5±0.6
6.0 W	1085 A	0-5	72	720±47	90±30	65±22	18.1±0.9
	B	5-10	72	1000±39	63±21	68±20	17.4±0.7
	C	10-15	92	1010±38	140±22	115±16	15.6±0.6
	D	15-20	97	1370±45	150±25	135±17	17.0±0.7
	E	20-25	62	1040±32	84±15	105±13	15.8±0.6
	F	25-30	79	1050±43	5±18	120±19	18.0±0.7
	G	30-35	76	1280±44	15±14	135±17	18.7±0.7

				ACTIVITIES(a) (pCi/kg)			
Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	Cs^{137}	Cs^{134}	Co^{60}	K^{40} x 10^3
	H	35-40	77	895±35	-3±12	195±17	17.3±0.7
	I	40-45	80	870±35	10±13	250±19	17.5±0.7
	J	45-50	91	875±35	-6±12	105±16	17.1±0.7
	K	50-55	83	700±24	8±9	56±10	17.3±0.6
	L	55-60	82	415±31	-2±14	22±16	N.D.(c)
	M	60-65	94	36±13	4±12	34±11	16.8±0.6
	N	65-70	92	30±16	16±16	19±14	18.3±0.7
	O	70-75	96	24±10	24±11	5±9	17.0±0.6
11.5 W	1010 A	0-5	90	49±13	20±17	34±15	19.8±0.7
	B	5-10	90	< bkg	< bkg	41±40	16.5±1.0
13.2 M	1058 A	0-5	107	22±10	< bkg	15±13	18.2±0.6
	B	5-10	103	< bkg	< bkg	< bkg	18.4±1.0
	C	10-15	91	6±11	< bkg	17±15	19.5±0.7
14.2 M	1005 A	0-5	99	154±33	94±43	46±32	15.7±1.0
18.6-3 M	1089 A	0-5	68	150±13	36±12	50±17	11.9±0.5
	B	5-10	36	120±21	-12±15	32±30	16.7±0.8
	C	10-15	52	8±20	N.D.	10±29	16.0±0.8

				ACTIVITIES$^{(a)}$ (pCi/kg)			
Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	Cs^{137}	Cs^{134}	Co^{60}	K^{40} $\times 10^3$
18.6-5 W	1092 A	0-1	101	580±25	19±10	67±10	15.0±0.6
	B	1-3	108	525±45	22±21	64±21	16.6±1.0
	C	3-4	113	410±20	5±8	44±9	15.8±0.6
	D	4-5	111	335±25	35±13	51±16	14.8±0.7
	1090 A	5-10	100	-1±15	-11±10	-8±13	17.8±0.7
	SLOSH I	0-5		290±16	30±8	58±13	16.7±0.6
	SLOSH II	oxidized layer		660±23	60±11	89±13	17.4±0.6
21.7 M	1025 A	0-7	112	240±15	25±8	65±11	13.2±0.5
21.9 W	1021 A	0-5	80	450±50	15±28	125±40	18.9±1.2
	B	5-10	86	100±13	9±10	24±12	19.9±0.7
	C	10-15	79	< bkg	< bkg	< bkg	19.9±1.1
25.3 W	1086 A	0-5	75	1170±36	2±10	100±20	18.6±0.7
	B	5-12	76	780±34	-8±15	-11±22	18.9±0.7
	C	12-16	82	265±17	-12±8	-3±15	17.0±0.6
	D	16-21	77	48±18	3±11	3±21	18.4±0.7

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	ACTIVITIES[(a)] (pCi/kg) Cs^{137}	Cs^{134}	Co^{60}	K^{40} x 10^3
39.0	1100 A	0-7	107	88±12	4±8	16±9	17.6±0.6
	B	7-12	89	-12±15	N.D.	N.D.	18.1±0.7
	C	12-16	78	-7 ±21	N.D.	N.D.	20.4±0.8
40.0 WC	1095 A	0-5	77	2069±42	111±13	217±12	20.1±0.6
	B	5-10	83	619±28	8±13	-7±11	19.6±0.7
	C	10-15	64	109±118	1±13	10±13	19.8±0.7
	D	15-20	82	86±12	-8±9	18±14	19.7±0.6
	E	20-25	67	58±15	23±12	-18±11	19.0±0.6
	F	25-30	80	57±15	17±15	29±14	20.2±0.7
	G	30-35	83	57±14	10±11	-1±10	17.5±0.6
	H	35-40	103	20±9	N.D.	0±11	16.5±0.5
C 43 E	1043 A	0-5	95	285±19	71±22	87±17	21.9±0.8
	B	5-10	80	30±21	-13±13	-6±26	24.1±0.9
	SLOSH III	oxidized layer		2700±72	345±42	400±27	18.1±0.7
	SLOSH IV	0-5		520±35	24±19	50±19	20.6±0.8
C 44	1032 A	0-7	128	610±21	79±19	120±21	16.1±0.5
	B	7-14	114	-1±11	-9±7	-14±15	17.4±0.6

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	ACTIVITIES[(a)] (pCi/kg) Cs^{137}	Cs^{134}	Co^{60}	K^{40} x 10^3
49.5	1093 A	0-5	83	1500±37	95±16	160±12	18.8±0.6
	B	5-10	85	23±13	8±11	16±10	19.1±0.6
	C	10-15	72	21±15	16±15	-3±13	19.7±0.7
	D	15-20	96	-5±9	0±9	-2±7	16.2±0.5
	E	20-25	102	-1±9	6±9	11±7	15.3±0.5
52.5 EC	1039 A	0-5	79	2700±60	375±45	235±22	18.4±0.7
	B	5-10	104	2050±50	44±22	175±25	18.8±0.7
	C	10-15	106	735±30	9±26	34±26	19.8±0.8
	D	15-20	108	17±12	32±18	-19±26	19.1±0.6
	E	20-25	114	14±11	16±16	21±21	17.2±0.6
53.8 EC	1052 A	0-5	54	2475±63	98±27	69±19	19.0±0.8
	B	5-10	84	1825±68	17±32	19±20	21.5±1.0
	C	10-15	88	210±17	-8±10*	15±11	20.0±0.7
	D	15-20	92	26±17	-15±11	30±15	18.8±0.8
	E	20-25	97	35±23	0±12*	7±18	20.7±0.9
	F	50-55	82	9±14	7±9	13±15	18.5±0.7

				ACTIVITIES(a) (pCi/kg)			
Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	Cs^{137}	Cs^{134}	Co^{60}	K^{40} $\times 10^3$
	FC CIIC A	0-10	44	1380±65	-3±16	57±21	14.9±0.9
	B	10-15	58	1510±44	31±14	76±12	14.7±0.6
	C	15-20	56	1380±39	29±12	78±11	14.0±0.6
	D	20-25	63	795±26	-2±9	20±10	15.6±0.6
	E	25-30	69	58±15	10±10	30±9	16.3±0.6
	F	30-35	88	-18±16	4±14	6±14	19.2±0.7
	G	35-40	97	10±15	11±11	0±12	19.2±0.7
	FC 1	0-5	50	2140±96	8±24	103±30	18.6±1.2
	FC 2	0-5	75	71±44	1±30	8±35	16.4±1.3
	FC 3	0-5	58	1840±115	17±32	69±45	19.8±1.6
	FC 5	0-5	53	2220±135	2±32	32±43	17.7±1.6
	FC 6	0-5	41	2790±135	120±43	255±59	21.3±1.6
	FC 7	0-5	51	670±83	83±39	66±50	21.6±1.8

				ACTIVITIES(a) (pCi/kg)			
Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	Cs^{137}	Cs^{134}	Co^{60}	K^{40} x 10^{3}
	FC 8	0-5	62	1610±105	37±32	11±39	20.5±1.6
	FC 9	0-5	52	1730±66	10±17	34±27	21.9±1.1
	FC 10	0-5	82	2250±81	14±10	42±12	20.3±1.0
	FC 11	0-5	63	1490±54	34±14	66±21	20.6±0.9
	FC 12	0-5	51	2140±135	43±36	150±51	18.8±1.7
	FC 13	0-5	102	42±35	15±23	72±29	17.7±1.3
	FC 14	0-5	37	1840±69	21±21	96±30	18.1±0.9
	FCB1 A	0-7.5	75	1950±52	N.D.	42±14	15.6±0.6
	B	7.5-12.5	72	1380±34	N.D.	21±10	14.6±0.5
	C	12.5-17.5	67	720±21	N.D.	14±8	14.4±0.4
	D	17.5-22.5	79	56±18	N.D.	7±15	13.3±0.6
	E	22.5-27.5	81	7±7	N.D.	-1±7	14.2±0.4

				ACTIVITIES$^{(a)}$ (pCi/kg)			
Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	Cs^{137}	Cs^{134}	Co^{60}	K^{40} x 10^3
56.4 W	1069 A	0-5	87	930±36	10±22	58±20	18.5±0.7
	B	5-10	85	15±15	10±18	16±21	20.3±0.7
57.5 M	1035 A	0-5	155	0±7	10±22	8±11	12.9±0.5
59.7 EC	1091 A	0-5	64	1870±52	41±26	12±11	15.6±0.6
	B	5-10	79	160±22	3±28	-14±14	18.0±0.7
	C	10-15	105	16±10	-6±7	-19±7	15.1±0.6
75.5	1259 A	0-5	57	1890±39	-8±7	11±6	16.4±0.5
	B	5-10	73	2180±44	-9±10	-5±12	16.8±0.5
	C	10-15	64	330±18	4±11	14±10	16.3±0.5
	D	15-20	67	24±16	-12±14	-7±12	15.5±0.6
	E	20-25	80	22±13	3±10	5±10	15.4±0.5
76.1	1260 A	0-5	44	864±34	-14±12	13±11	16.2±0.7
	B	5-10	100	32±15	-19±11	3±11	16.4±0.6
91.3	1261 A	0-5	100	24±13	15±11	2±10	16.3±0.6
	B	5-10	125	-13±9	-13±7	4±7	14.7±0.5

(a) Sample specific activities are expressed in terms of dry weight and have been corrected for decay between time of coring and time of assay. Samples marked by * were uncorrected.

(b) Specific activities >10,000 were rounded to the nearest 100 pCi. Specific activities <10,000 but >1000 were rounded to the nearest 10 pCi. Specific activities <1000 but >100 were rounded to the nearest 5 pCi and specific activities <100 were rounded to the nearest pCi. Statistical errors are reported at 1 sigma. Errors for specific activities >10,000 were rounded to the nearest 10 pCi. Errors for specific activities <10,000 but >1000 were rounded to the nearest 5 pCi and errors for specific activities <1000 were rounded to the nearest pCi.

(c) N.D. = not determined

core from the harbor which we have analyzed (P-1.5E) has substantial activities of Cs^{137} to 250 cm. Assuming this core represents ∿ 10 years of time, the mean sedimentation rate in that area has been ∿ 25 cm/year. Several locations show very low Cs^{137} activities. These sites, located upstream of New York Harbor, are apparently in zones of little net accumulation of recent fine-grained sediment.

We have analyzed a substantial number of the samples which have been gamma counted (Table 1) for plutonium by radiochemical separation and alpha spectrometry. The results for all three years of the contract are summarized in Table 2. The general distribution of $Pu^{239,240}$ in Hudson sediments is reasonably similar to Cs^{137}. Long cores in the harbor which have layers of low $Pu^{239,240}$ between higher activity layers also have low Cs^{137}. The surface samples upstream of the harbor which are low in $Pu^{239,240}$ are also low in Cs^{137}. Thus our original proposal of using gamma counting for Cs^{137} as an aid to mapping the distribution of $Pu^{239,240}$ in Hudson sediments has proven to be very successful in this estuary.

RESULTS - HUDSON ESTUARY SUSPENDED SEDIMENTS

We have collected large samples of suspended particles in the Hudson throughout the salinity range, and during differing fresh water flow conditions. In general the amounts of suspended particles were greater during high fresh water flow conditions. Data on the radionuclide concentrations in the suspended matter are summarized in Table 3. Suspended particles throughout the Hudson have Cs^{137} activities similar to the recently-deposited sediments of New York harbor (Table 1), even in areas which showed little net deposition of recent sediments (mp 24 and mp 18). The activities of

Table 2

Plutonium Isotopes in Hudson Estuary Sediments

ocation (Mile Point)	Sample No.	Depth (cm)	Dry Wt. (gm)	% Yield	pCi/kg ^{239,240}Pu	 ^{238}Pu	 $^{238/239,240}$Pu
-1.6 E	1051 A	0-5	50.5	70.0	32.6±1.0	1.4±0.2	0.04±0.006
	B	5-12	73.1	56.2	29.4±0.7	1.4±0.1	0.05±0.004
	C	12-20	72.0	55.9	47.1±1.3	2.1±0.2	0.04±0.004
	D	20-25	78.9	30.1	45.4±1.8	1.8±0.2	0.04±0.005
	E	25-30	67.5	54.8	40.2±1.4	2.2±0.2	0.05±0.005
	F	30-35	58.4	61.1	41.3±1.2	2.0±0.2	0.05±0.005
	G	35-40	68.3	35.3	68.9±2.7	-	-
	H	40-45	37.6	60.8	61.5±1.8	-	-
	I	45-50	58.1	41.9	60.1±2.2	3.8±0.3	0.06±0.005
	J	50-55	92.8	36.9	0.8±0.1	N.D.	-
	K	55-60	97.7	33.5	0.2±0.1	N.D.	-
	L	60-65	107.4	32.1	0.5±0.1	N.D.	-
	M	65-73	87.9	40.0	45.7±1.4	-	-
-1.5 E	1046 A	0-5	55.9	36.0	28.2±1.3	1.3±0.2	0.05±0.008
	B	5-12	52.9	23.9	31.4±1.6	-	-
	C	12-20	76.7	32.2	38.1±1.2	1.8±0.2	0.05±0.006
	D	20-25	76.7	-	-	-	-
	E	25-30	48.8	18.2	34.6±1.7	1.4±0.3	0.04±0.009
	F	30-35	63.0	48.2	34.0±1.2	-	-
	G	35-40	52.0	17.4	34.0±1.7	1.6±0.3	0.05±0.01
	H	40-45	66.1	40.5	37.0±1.5	1.9±0.2	0.05±0.006
	I	45-50	59.5	25.8	42.3±3.5	2.5±0.3	0.06±0.009
	J	50-55	63.8	43.9	41.6±1.9	-	-
	K	55-60	68.9	41.7	54.5±2.7	1.9±0.2	0.04±0.005

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

Location (Mile Point)	Sample No.		Depth (cm)	Dry Wt. (gm)	% Yield	pCi/kg ^{239,240}Pu	pCi/kg ^{238}Pu	pCi/kg ^{238}/^{239,240}Pu
W	1066	A	0-5	37.2	48.1	24.5±1.1	-	-
		B	5-10	72.3	32.7	31.4±1.3	1.4±0.2	0.04±0.006
		C	10-18	69.9	35.0	28.1±1.5	1.2±0.2	0.04±0.007
		D	18-25	63.5	56.5	33.6±1.3	1.7±0.2	0.05±0.006
		E	25-30	77.6	-	-	-	-
		F	30-35	66.3	26.0	33.7±1.5	1.4±0.2	0.04±0.006
		G	35-40	66.0	77.9	44.9±1.0	2.3±0.2	0.05±0.004
		H	40-45	60.0	29.9	34.6±1.2	-	-
		I	45-53	71.5	42.2	35.7±1.2	2.2±0.2	0.06±0.004
		J	53-60	95.8	50.2	7.2±0.4	-	-
		K	60-65	79.1	48.0	0.5±0.1	N.D.	-
		L	65-70	97.8	5.0	1.6±0.5	-	-
[illegible].1 WM	1002	A	0-5	73.8	50.5	32.7±2.4	-	-
		B	5-10	74.9	28.5	31.7±3.2	-	-
		C	10-15	71.3	56.0	43.3±3.0	2.2±0.3	0.05±0.008
		D	15-20	63.4	55.0	45.8±3.2	2.1±0.2	0.05±0.006
		E	20-25	79.3	56.5	26.4±1.2	1.2±0.2	0.05±0.009
		F	25-30	87.3	28.0	38.5±1.3	1.5±0.2	0.03±0.004
		G	30-35	57.5	45.6	48.5±1.6	-	-
1.9 W	1082	A	0-5	44.0	9.3	36.5±2.6	-	-
		B	5-10	57.9	51.3	27.8±1.0	1.2±0.1	0.04±0.006
		C	10-15	78.9	37.3	36.9±1.5	-	-
		D	15-20	79.7	38.6	1.4±0.2	-	-
		E	20-25	94.1	37.6	0.5±0.1	-	-

Location (Mile Point)	Sample No.	Depth (cm)	Dry Wt. (gm)	% Yield	pCi/kg		
					^{239,240}Pu	^{238}Pu	$^{238/239,240}$Pu
-5	1092 A	0-1	101.9	33.5	12.9±0.9	–	–
	B	1-3	108.1	43.6	12.4±0.8	–	–
	C	3-4	113.0	22.7	11.1±0.8	–	–
25.3 W	1086 A	0-6	66.3	21.8	39.4±1.7	–	–
	B	6-12	67.0	25.7	43.2±1.7	–	–
	C	12-16	72.7	32.3	12.2±0.7	0.3±0.1	0.02±0.007
	D	16-21	68.2	48.5	1.1±0.1	–	–
53.8 EC	1052 A	0-5	53.1	26.2	69.2±6.4	3.0±0.6	0.04±0.009
	B	5-10	82.3	30.0	52.1±4.3	2.0±0.4	0.04±0.009
	C	10-15	87.8	38.3	5.7+0.4	–	–
59.7 EC	1091 A	0-5	54.8	35.9	40.7±1.5	1.2±0.2	0.03±0.005
	B	5-10	68.4	25.1	4.5+0.4	N.D.	

TABLE 3

Radionuclides on Suspended Particles of the Hudson Estuary and New York Bight

Location (Mile Point)	Sample #	Date Collected	Salinity (^{o}/oo)	^{137}Cs	^{134}Cs	^{60}Co
Hudson Shelf Channel	Sta. 133	1-15-76	30	123±63	N.D.	91±62
Long Island Shelf	Sta. 76	1- 4-76	30	210±133	N.D.	-63±97
N.Y. Bight Apex	Ambrose	1-20-76	30	257±62	N.D.	110±59
Mp-6	1178A	8- 2-76	21	505±39	57±31	3±25
Mp-6 (Storm)	1223B	10-27-76	15-21	1395±61	N.D.	150±18
Mp 0 (Storm)	He #5	10- 1-75	10	1435±170	N.D.	354±120
Mp 18.6 (Storm	1088	9-30-75	1.7	1260±35	210±25	145±18
Mp 24	1114A	5-14-76	N.D.	1140±35	101±15	100±12
Mp 47	1222B	10-27-76	0	1565±130	N.D.	190±70
Mp 61	1177A	7-31-76	0	1105±90	N.D.	-5±43

Location (Mile Point)	Sample #	Dry Weight (gms)	^{239,240}Pu (pCi/Kg)	^{238}Pu (pCi/Kg)
Mp 24	1114	102.8	18.9±0.9	1.0±0.2
Mp 18	1088	82.6	23.4±1.0	0.7±0.1

Co^{60} and Cs^{134} in the suspended particles throughout the Hudson downstream of the reactor site are comparable to the sediments near the release area and to recent sediments in the harbor. Thus the source of fine-grained shoaling material in the harbor is clearly from upstream, and the particles are already labeled with Cs^{137} (and Cs^{134} and Co^{60}) long before they reach the harbor zone.

DISCUSSION - HUDSON ESTUARY

The activity of $Pu^{239,240}$ per gram of sediment varies greatly from place to place in the Hudson. The relative amounts of $Pu^{239,240}$ and Cs^{137} have much smaller variation than the absolute amount of either. Table 4 summarizes the ratios of $Pu^{239,240}$ to Cs^{137}, as well as the ratios of Cs^{134} and Co^{60} to Cs^{137}. The total reported range of $Pu^{239,240}$ to Cs^{137} ratios was 1-13.8% compared with the fallout ratio of 1-2%. There does appear to be a systematic downstream increase in this ratio, with the highest values in the cores from upper New York Bay (-1.5E, -1.6E and P-1.5). Just upstream from the Upper Bay the values range from 2-7%. Thus essentially all of the sediment samples in the harbor have higher $Pu^{239,240}$ to Cs^{137} ratios than fallout.

The presence of Cs^{134} and Co^{60} in all of the gravity core samples from the harbor, except the deepest sample in core -1.6E indicates that reactor Cs^{137} must also be present in essentially all of the upper half meter of harbor sediments. Since an extra source of Cs^{137} would lower the $Pu^{239,240}$ to Cs^{137} ratio, there must be at least one additional process which affects the observed sediment ratios in the harbor. Loss of Cs^{137} to solution or gain of $Pu^{239,240}$ from the dissolved phase (or both) must occur to some extent.

Suspended particle samples collected during two recent periods from

Table 4

Nuclide Ratios in Hudson Estuary Sediments

Location le 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}
-1.6 E	1051 A	0-5	30.6±1.0	450±26	7.2±0.5	2.7±2.5	11.3±7.6
	B	5-12	29.4±0.7	620±26	4.7±0.2	9.0±2.1	8.1±2.9
	C	12-20	47.1±1.3	1210±42	3.9±0.2	9.5±1.6	9.1±1.9
	D	20-25	45.4±1.8	470±25	9.7±0.6	7.9±2.6	9.1±4.0
	E	25-30	40.2±1.4	540±24	7.4±0.4	5.9±2.0	-
	F	30-35	41.3±1.2	390±18	10.6±0.6	5.1±2.1	11.3±3.9
	G	35-40	68.9±2.7	615±36	11.2±0.8	6.2±2.6	11.2±3.8
	H	40-45	61.5±1.8	535±95	11.5±2.1	-	10.1±8.3
	I	45-50	60.1±2.2	610±31	9.9±0.6	-	7.7±3.5
	J	50-55	0.8±0.1	29±11	2.8±1.1	-	-
	K	55-60	0.2±0.1	-19±9	-	-	-
	L	60-65	0.5±0.1	27±14	1.9±1.1	-	-
	M	65-73	45.7±1.4	455±24	10.0±1.8	2.4±2.2	0.1±0.1
-1.5 E	1046 A	0-5	28.2±1.3	440±21	6.4±0.4	7.3±2.3	6.8±3.6
	B	5-12	31.4±1.6	430±26	7.3±0.6	2.8±1.9	7.0±5.2
	C	12-20	38.1±1.2	620±26	6.1±0.3	3.1±1.8	11.0±2.8
	D	20-25	-	440±23	-	3.0±2.3	12.7±6.6
	E	25-30	34.6±1.7	390±28	8.9±0.8	44.9±7.4	7.2±6.5
	F	30-35	34.0±1.2	570±24	6.0±0.3	4.7±1.9	5.4±2.4
	G	35-40	34.0±1.7	605±24	5.6±0.4	7.1±1.8	6.1±3.2
	H	40-45	37.0±1.5	685±28	5.4±0.3	7.6±1.8	6.9±2.4
	I	45-50	42.3±3.5	635±30	6.7±0.6	5.7±2.1	6.8±3.0
	J	50-55	41.6±1.9	1140±50	3.6±0.2	11.8±2.2	9.2±2.2
	K	55-60	54.4±2.7	865±33	6.3±0.4	12.7±2.2	8.4±2.1

Location (Mile Point)	Sample No.	Depth (cm)	^{239,240}Pu pCi/kg	^{137}Cs pCi/kg	^{239,240}Pu / ^{137}Cs x 10^{-2}	^{134}Cs / ^{137}Cs x 10^{-2}	^{60}Co / ^{137}Cs x 10^{-2}
.5	1048 A	0-10	19.2±0.9	260±21	7.4±0.7	-	-
	B	10-20	33.0±1.5	455±30	7.3±0.6	8.8±4.7	5.5±3.8
	C	20-30	25.0±1.8	345±23	7.2±0.7	-	-
	D	30-40	29.3±1.6	420±34	7.0±0.7	-	-
	E	40-50	24.4±2.0	350±26	7.0±0.8	17.1±5.3	32.5±5.7
	F	50-60	33.3±2.4	620±32	5.4±0.5	10.3±2.9	14.3±2.8
	G	60-70	40.9±1.4	1090±44	3.8±0.2	9.6±2.2	11.0±1.8
	H	70-80	38.5±2.0	950±36	4.1±0.3	14.5±2.4	10.0±1.6
	I	80-90	28.9±1.9	1080±42	2.7±0.2	14.0±2.2	9.7±1.4
	J	90-100	43.9±3.6	815±42	5.4±0.5	13.9±3.0	7.0±2.5
	K	100-110	51.3± 2.3	635±33	8.1±0.6	8.5±2.9	9.1±2.7
	L	110-120	42.8±3.0	495±30	8.6±0.8	12.9±3.9	-
	M	120-130	26.2±2.2	290±18	9.0±0.9	12.4±4.2	-
	N	130-140	32.0±1.3	340±25	9.4±0.8	12.9±4.5	5.9±4.2
	O	140-150	35.7±1.5	350±18	10.2±0.7	-	6.9±2.9
	P	150-160	32.5±1.8	365±28	8.9±0.8	-	5.5±4.7
	Q	160-170	29.9±1.9	350±30	8.5±0.9	-	6.0±4.9
	R	170-180	50.3±2.3	395±21	12.7±0.9	-	6.0±3.0
	S	180-190	42.3±2.5	350±20	12.1±1.0	-	4.3±3.2
	T	190-200	18.0±1.0	200±22	9.0±1.1	-	9.0±7.0
	U	200-210	37.4±2.0	470±25	8.0±0.6	-	3.8±2.5
	V	210-220	48.2±1.4	425±27	11.3±0.8	-	6.1±3.3
	W	220-230	53.9±2.5	390±22	13.8±1.0	-	-
	X	230-240	38.5±1.6	335±18	11.5±0.8	-	8.3±3.0
	Y	240-250	59.2±1.8	500±27	11.8±0.7	-	5.6±2.8
	Z	250-260	4.2±0.4	46±11	9.1±2.3	-	-

Location (Mile Point)	Sample No.	Depth (cm)	^{239,240}Pu pCi/kg	^{137}Cs pCi/kg	^{239,240}Pu/^{137}Cs x 10^{-2}	^{134}Cs/^{137}Cs x 10^{-2}	^{60}Co/^{137}Cs x 10^{-2}
W	1066 A	0-5	24.5±1.1	645±36	3.8±0.3	9.9±3.8	6.2±4.5
	B	5-10	31.4±1.3	890±28	3.5±0.2	7.1±1.4	6.7±1.8
	C	10-18	28.1±1.5	1040±50	2.7±0.2	8.4±2.2	10.6±3.2
	D	18-25	33.6±1.3	1480±40	2.3±0.1	8.4±1.2	11.5±1.5
	E	25-30	-	1260±40	-	10.3±1.5	13.9±1.8
	F	30-35	33.7±1.5	2310±70	1.5±0.1	12.9±1.7	8.9±1.4
	G	35-40	44.9±1.0	1210±60	3.7±0.2	10.7±2.1	7.4±2.6
	H	40-45	34.6±1.2	1140±50	3.0±0.2	9.6±1.9	7.4±2.3
	I	45-53	35.7±1.2	685±27	5.2±0.3	3.9±1.5	10.4±2.7
	J	53-60	7.2±0.4	105±14	6.9±1.0	-	36.1±15.0
	K	60-65	0.5±0.1	3±11	-	-	-
	L	65-70	1.6±0.5	2±12	-	-	-
W	SLOSH V	0-5		555±13		9.6±1.6	7.8±1.1
0.1 WM*	1002 A	0-5	32.7±2.4	1260±38	2.6±0.1	18.2±2.7	15.1±1.5
	B	5-10	31.7±3.2	1795±60	1.8±0.2	22.0±3.2	16.7±1.6
	C	10-15	43.3±3.0	1995±56	2.2±0.2	24.0±3.0	12.3±1.0
	D	15-20	45.8±3.2	960±44	4.8±0.4	14.0±3.5	6.8±2.1
	E	20-25	26.4±1.2	1030±40	2.6±0.2	17.0±3.3	12.1±1.9
	F	25-30	38.5±1.3	570±28	6.8±0.4	-	10.4±2.9
	G	30-35	48.5±1.6	925±49	5.2±0.3	-	10.8±3.1
	H	35-40				6.3±2.0	12.8±1.7
1.9 W	1082 A	0-5	36.5±2.6	1180±35	3.1±0.2	8.5±1.4	8.9±2.0
	B	5-10	27.8±1.0	375±19	7.4±0.5	3.2±2.4	17.1±5.1
	C	10-15	36.9±1.5	550±24	6.7±0.4	-	3.6±3.1

Location (Mile Point)	Sample No.	Depth (cm)	^{239,240}Pu pCi/kg	^{137}Cs pCi/kg	$\frac{^{239,240}\text{Pu}}{^{137}\text{Cs}}$ x 10^{2}	$\frac{^{134}\text{Cs}}{^{137}\text{Cs}}$ x 10^{2}	$\frac{^{60}\text{Co}}{^{137}\text{Cs}}$ x 10^{-2}
	D	15-20	1.4±0.2	57±13	2.5±0.7	-	-
	E	20-25	0.5±0.1	16±13	3.1±2.6	-	-
	F	25-30				-	-
	G	50-55				-	-
6.0 W	1084 A	0-5		720±47		12.5±4.2	9.0±3.1
	B	5-10		1000±39		6.3±2.1	6.8±2.0
	C	10-15		1010±38		14.0±2.2	11.5±1.6
	D	15-20		1370±45		11.0±1.9	10.0±1.3
	E	20-25		1050±32		8.0±1.5	10.2±1.3
	F	25-30		1050±43		-	11.5±1.9
	G	30-35		1280±44		-	10.4±1.4
	H	35-40		900±35		-	21.8±2.1
	I	40-45		870±35		-	28.7±2.5
	J	45-50		875±35		-	12.2±1.9
	K	50-55		700±24		-	8.0±1.5
	L	55-60		415±31		-	5.3±3.9
	M	60-65		36±13		-	-
	N	65-70		30±16		-	-
	O	70-75		24±10		-	-
11.5 W	1010 A	0-5		49±13		40.0±35.6	69.4±35.7
	B	5-10		< bkg		-	-
13.2 M	1058 A	0-5		22±10		-	-
	B	5-10		< bkg		-	-
	C	10-15		6±11		-	-

Location (Mile Point)	Sample No.	Depth (cm)	^{239,240}Pu pCi/kg	^{137}Cs pCi/kg	^{239,240}Pu/^{137}Cs x 10^{-2}	^{134}Cs/^{137}Cs x 10^{-2}	^{60}Co/^{137}Cs x 10^{-2}
2 M	1005 A	0-5		155±33		61.0±30.8	29.9±21.8
18.6-3 M	1089 A	0-5		150±13		24.0±8.3	33.0±11.6
	B	5-10		120±21		–	–
	C	10-15		8±20		–	–
18.6-5	1092	0-1	12.9±0.9	580±25	2.2±0.2	3.3±1.7	11.6±1.8
		1-3	12.4±0.8	525±45	2.4±0.3	–	12.2±4.1
		3-4	11.1±0.8	410±20	2.7±0.2	–	10.7±2.3
		4-5				10.4±3.9	15.2±4.9
		5-10				–	–
18.6	SLOSH I	0-5		290±16		10.3±2.8	20.0±4.6
6	SLOSH II	Oxidized layer		660±23		9.1±1.7	13.5±2.0
21.7 M	1025 A	0-7		240±15		10.4±3.4	27.1±4.9
21.9 W	1021 A	0-5		450±50		–	27.3±9.4
	B	5-10		100±13		–	24.0±12.4
	C	10-15		< bkg		–	–
25.3 W	1086 A	0-6	39.4±1.7	1170±36	3.4±0.2	–	8.5±1.7
	B	6-12	43.2±1.7	780±34	5.5±0.3	–	–
	C	12-16	12.2±0.7	265±17	4.6±0.4	–	–
	D	16-21	1.1±0.1	48±18	2.3±0.9	–	–
C 43 E	1043 A	0-5		285±19		24.9±7.9	30.5±6.3
	B	5-10		30±21		–	–

Location (Mile Point)	Sample No.	Depth (cm)	^{239,240}Pu pCi/kg	^{137}Cs pCi/kg	$\frac{^{239,240}\text{Pu}}{^{137}\text{Cs}}$ x 10^{-2}	$\frac{^{134}\text{Cs}}{^{137}\text{Cs}}$ x 10^{-2}	$\frac{^{60}\text{Co}}{^{137}\text{Cs}}$ x 10^{-2}
	SLOSH III	Oxidized layer	∿ 26	2700±72	∿ 1	12.8±1.6	14.8±1.1
44	SLOSH IV	0-5		520±35		21.7±17.2	9.6±3.7
C 44	1032 A	0-7		610±21		13.0±3.2	19.7±3.5
	B	7-14		-1±11		-	-
52.5 EC	1039 A	0-5		2700±60		13.9±1.7	8.7±0.8
	B	5-10		2050±50		2.1±1.1	8.5±1.2
	C	10-15		735±30		-	4.6±3.5
	D	15-20		17±12		-	-
	E	20-25		14±11		-	-
53.8 EC	1052 A	0-5	69.2±6.4	2475±63	2.8±0.3	4.0±1.1	2.8±1.1
	B	5-10	52.1±4.3	1825±68	2.9±0.3	-	-
	C	10-15	5.7±0.4	210±17	2.7±0.3	-	-
	D	15-20		26±17		-	-
	E	20-25		35±23		-	-
	F	50-55		9±14		-	-
	FCC II C	0-10		1380±65		-	4.1±1.5
		10-15		1520±44		21.±1.0	5.0±0.8
		15-20		1380±39		-	5.7±0.8
		20-25		795±26		2.1±0.9	2.5±1.3
		25-30		58±15		-	51.7±20.5
		30-35		-18±16		-	-
		35-40		10±15		-	-
	FC I			2140±96		-	4.8±1.4

Location (Mile Point)	Sample No.	Depth (cm)	$^{239,240}Pu$ pCi/kg	^{137}Cs pCi/kg	$^{239,240}Pu/^{137}Cs$ x 10^{-2}	$^{134}Cs/^{137}Cs$ x 10^{-2}	$^{60}Co/^{137}Cs$ x 10^{-2}
	FC 2			71±44		–	–
	FC 3			1840±115		–	3.8±2.5
	FC 5			2220±135		–	–
	FC 6			2790±135		4.2±5	9.2±2.2
	FC 7	0-5		670±33		12.4±6.0	9.9±7.6
	FC 8	0-5		1610±105		–	–
	FC 9	0-5		1730±66		–	–
	FC 10	0-5		2250±81		0.6±0.4	1.9±0.5
	FC 11	0-5		1490±54		2.3±1.0	4.4±1.4
	FC 12	0-5		2140±135		2.0±1.7	6.9±2.4
	FC 13	0-5		42±35		–	–
	FC 14	0-5		1840±69		–	5.2±1.6
56.4 W	1069 A	0-5		930±36		–	6.2±2.2
	B	5-10		15±15		–	–
57.5 M	1035 A	0-5		0±7		–	–
59.7 EC	1091 A	0-5		1870±52		2.2±1.4	0.6±0.6
	B	5-10		160±22		–	–
	C	10-15		16±10		–	–

*Core 0.1 WM was taken approximately 2 years before core 0.1 W.

well upstream of the harbor have $Pu^{239,240}$ to Cs^{137} ratios (Table 5) approximating those of fallout, with absolute activities similar to sediments now accumulating opposite Manhattan (mp 0 to mp 6).

One of the piston cores we collected from the upper Bay (P-1.5E) penetrates through a very thick sequence of sediments containing Cs^{137} (Figure 2). Reactor-derived Cs^{134} and Co^{60} are apparently confined to the upper half of the core (above 100-140 cm), where a broad peak in Cs^{137} occurs. We believe this Cs^{137} peak reflects the period of maximum reactor releases, while the sediments below contain Cs^{137} derived predominantly from fallout. The trend of $Pu^{239,240}$ with depth (Figure 3) in the same core is quite different than for Cs^{137}. There is no evidence of a peak in $Pu^{239,240}$ correlating with the peak in Cs^{137}. We interpret this as indicating that if releases of $Pu^{239,240}$ have occurred from Indian Point during the same period as the maximum release of Cs^{137}, they were too small to be observable in the sediments of New York harbor in the presence of the burden of fallout $Pu^{239,240}$ accumulating there. The ratios of Pu^{238} to $Pu^{239,240}$ in this core also show no anomalously high ratios in the depth range of maximum Cs^{137}. As will be shown later, changes in this ratio are observed in the immediate vicinity of some nuclear reactors, giving positive indication of the presence of transuranic releases.

We believe that identification of transuranic releases from reactors as the basis of anomalies in accumulation rates of $Pu^{239,240}$ per unit area of sediment will be very difficult, except in cases where the rates of releases are extrememly large, such as near Windscale. The rates of accumulation of fine-grained sediments in estuaries and other areas of coastal environment are so variable in space, and probably in time, that

TABLE 5

Nuclide Ratios in Hudson Estuary and New York Bight Suspended Particles

Location	Sample #	^{239,240}Pu pCi/Kg	^{137}Cs pCi/Kg	$\frac{^{239,240}\text{Pu}}{^{137}\text{Cs}}$ X10^{-2}	$\frac{^{134}\text{Cs}}{^{137}\text{Cs}}$ X10^{-2}	$\frac{^{60}\text{Cs}}{^{137}\text{Cs}}$ X10^{-2}
mp 61		--	1105±90	---	---	---
mp 47		--	1565±130	---	---	12.1±4.6
mp 24	1114	18.9±0.9	1140±35	1.7±0.1	8.8±1.3	8.8±1.1
mp 18	1088	23.4±1.0	1260±35	1.9±0.1	16.7±2.0	11.5±1.9
mp 0		--	1435±170	---	---	24.7±8.9
mp -6		--	505±39	---	11.3±6.2	---
mp -6		--	1395±61	--	---	10.8±1.4
Ambrose		--	257±62	--	---	---
Long Island Shelf		--	210±133	--	---	---
Hudson Channel (Shelf)		--	123±63	--	---	---

FIGURE 2. Cs^{137}, Cs^{134}, and Co^{60} 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 Cs^{137} in the lower half of the core is believed to be derived predominantly from global fallout.

Piston Core P-1.5
(pCi/kg)

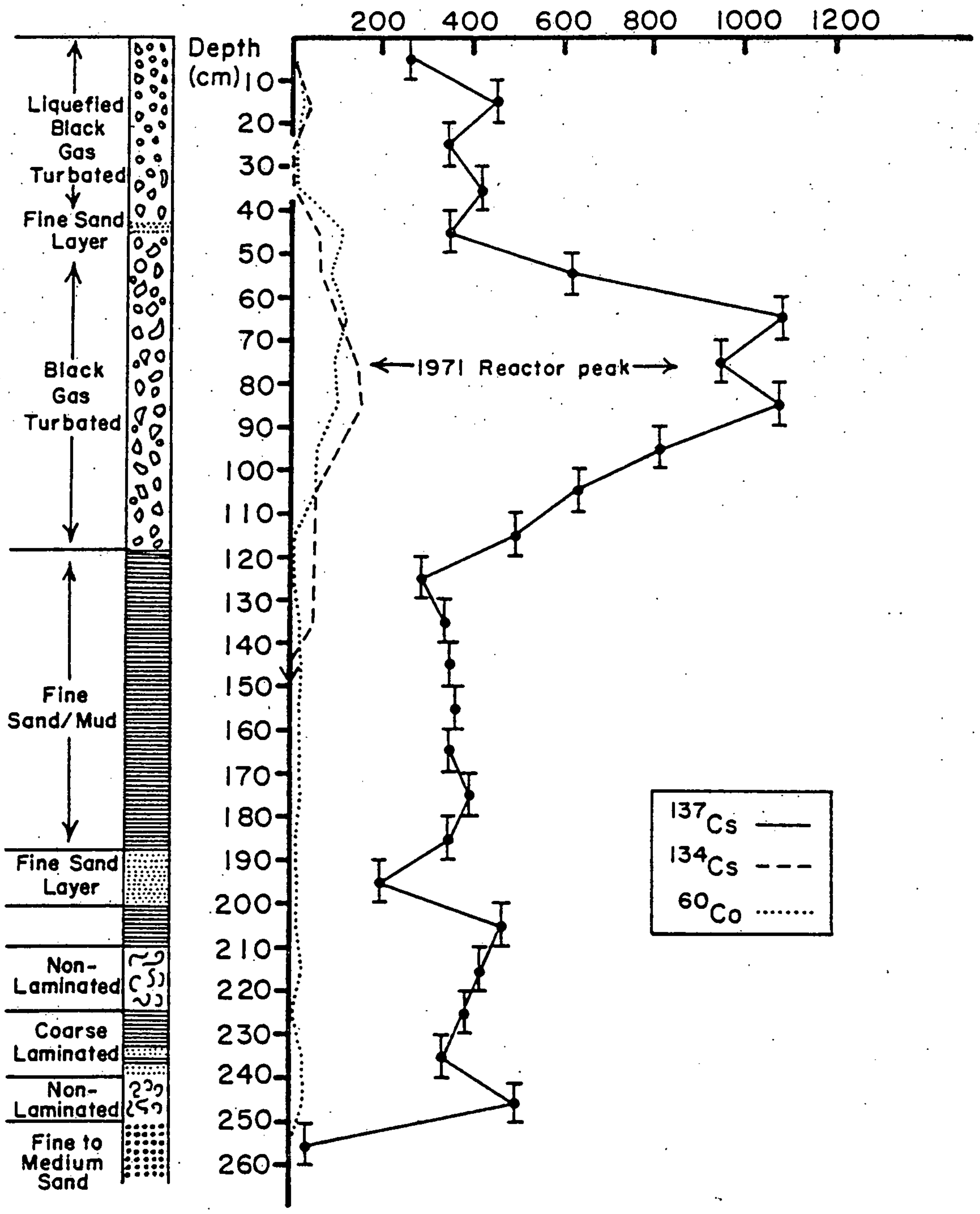
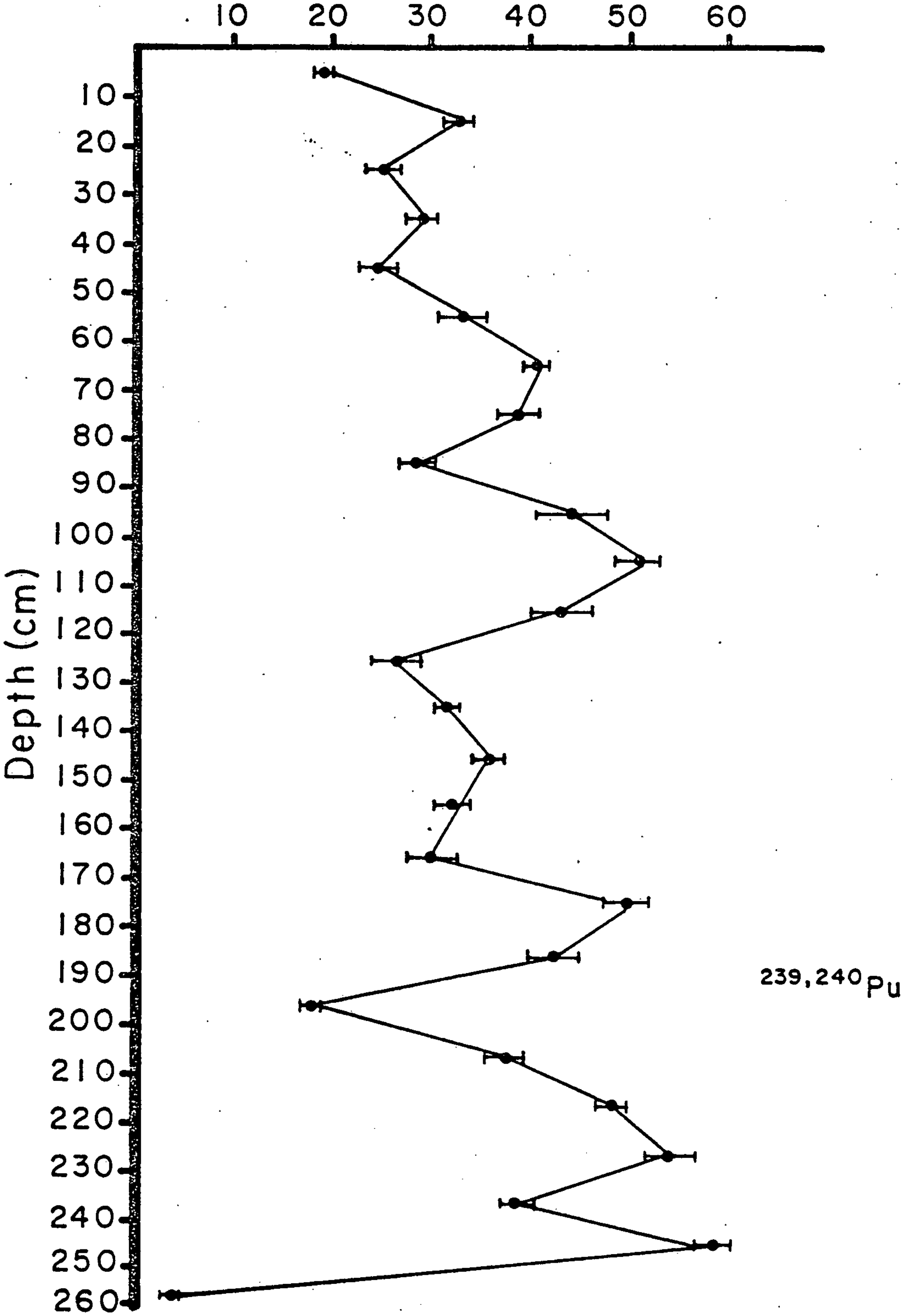


FIGURE 3. $Pu^{239,240}$ as a function of depth in the same piston core shown in Figure 2. There is no evidence of an increase in $Pu^{239,240}$ in the depth range (50 - 120 cm) which we believe correlates with the period (1971) of maximum reactor releases of gamma emitting nuclides.

Piston Core P-1.5



the presence of "extra" $Pu^{239,240}$ in addition to that derived from fallout would be very difficult to establish. In Tables 6 and 7 the accumulation rates of Cs^{137} and $Pu^{239,240}$ in Hudson sediments are given in terms of mCi per square kilometer. In the case of both nuclides, the observed rates of accumulation are in some locations much greater than the fallout delivery rate and in other locations much less. Since there is clearly a second important source of Cs^{137} in addition to fallout, the interpretation of Cs^{137} accumulation rates in sediments in terms of sources and processes which remove or add Cs^{137} to sediments is difficult. We believe appreciable Cs^{137} has been lost from particles to solution in the more saline parts of the Hudson, on the basis of the large deviation from the fallout ratio of $Pu^{239,240}$ to Cs^{137} in the lower part of the harbor piston core (Table 3). This is supported by the observation that the downstream increase in the $Pu^{239,240}$ to Cs^{137} ratio in the surface sediments is more clearly defined below mp 0 (Table 3). The mean specific activity of Cs^{137} in suspended particles from the New York Bight is also considerably lower than for Hudson suspended particles, even those from the most saline part of the estuary.

It is considerably more difficult to establish whether additional deposition of $Pu^{239,240}$ has occurred as opposed to loss of Cs^{137}. In Figures 4 and 5 the accumulation rates of Cs^{137} and $Pu^{239,240}$ per unit area are plotted as a function of mile point. The mCi/km^2 axis is logarithmic to allow presentation of the extremely large range of observed nuclide accumulation rates (more than two orders of magnitude). It is obvious that New York harbor has a very large accumulation rate of $Pu^{239,240}$ (more than an order of magnitude greater than the fallout

Table 6

Sediment Deposition of ^{137}Cs in the Hudson Estuary

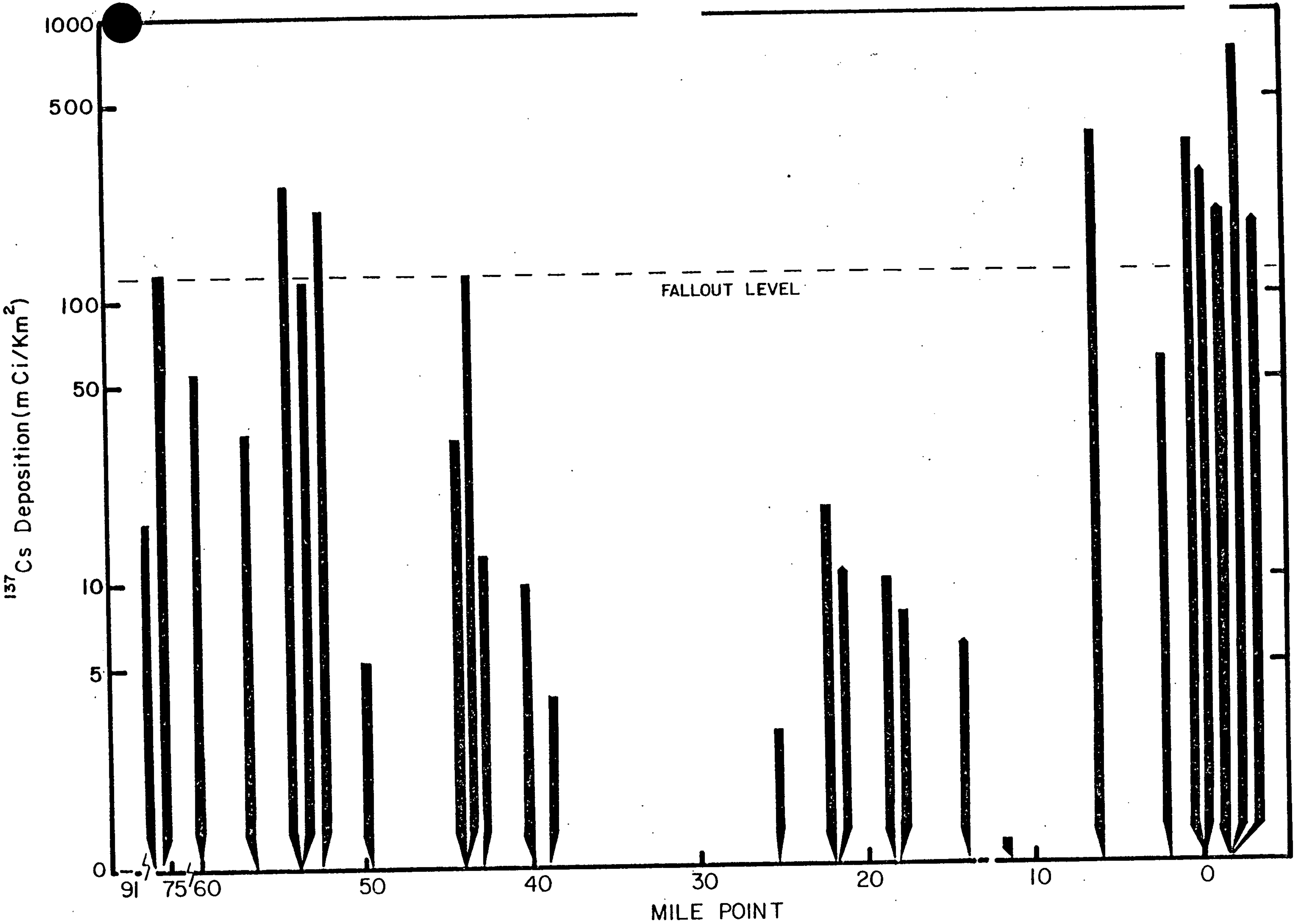
Location (mile point)	Sample No.	Core Depth (Cm)	^{137}Cs (mCi/Km^2)
-1.6 E	1051	73	184.9
-1.5 E	1046	60	202.9
P -1.5	1048	260	734.6
0.1 W	1066	70	345.1
0.1 WM	1002	40	285.1
1.9 W	1082	25	59.8
6.0 W	1085	75	371.0
11.5 W	1010	10	1.8
13.2 M	1058	10	0.9
14.2	1005	5	6.0
18.6-3 M	1089	15	7.6
18.6-5	1092	5	10.0
21.7	1025	7	10.9
21.9	1021	15	18.1
25.3 W	1086	21	70.4
39.0	1100	16	3.9
40 WC	1095	40	97.8
C 43 E	1043	10	12.2
43.8	FCBI	27.5	121.3
43.8	FCIICA	40	113.7
C 44	1032	14	31.7
49.5	1093	25	51.9
52.5	1039	25	206.2
53.8 EC	1052	20	125.3
56.4 W	1069	10	33.5
59.8 EC	1091	15	54.6
75.5	1259	25	118.1
76.1	1260	10	16.2
91.3	1261	10	0.8

Table 7

Sediment Deposition of $^{239,240}Pu$ in the Hudson Estuary

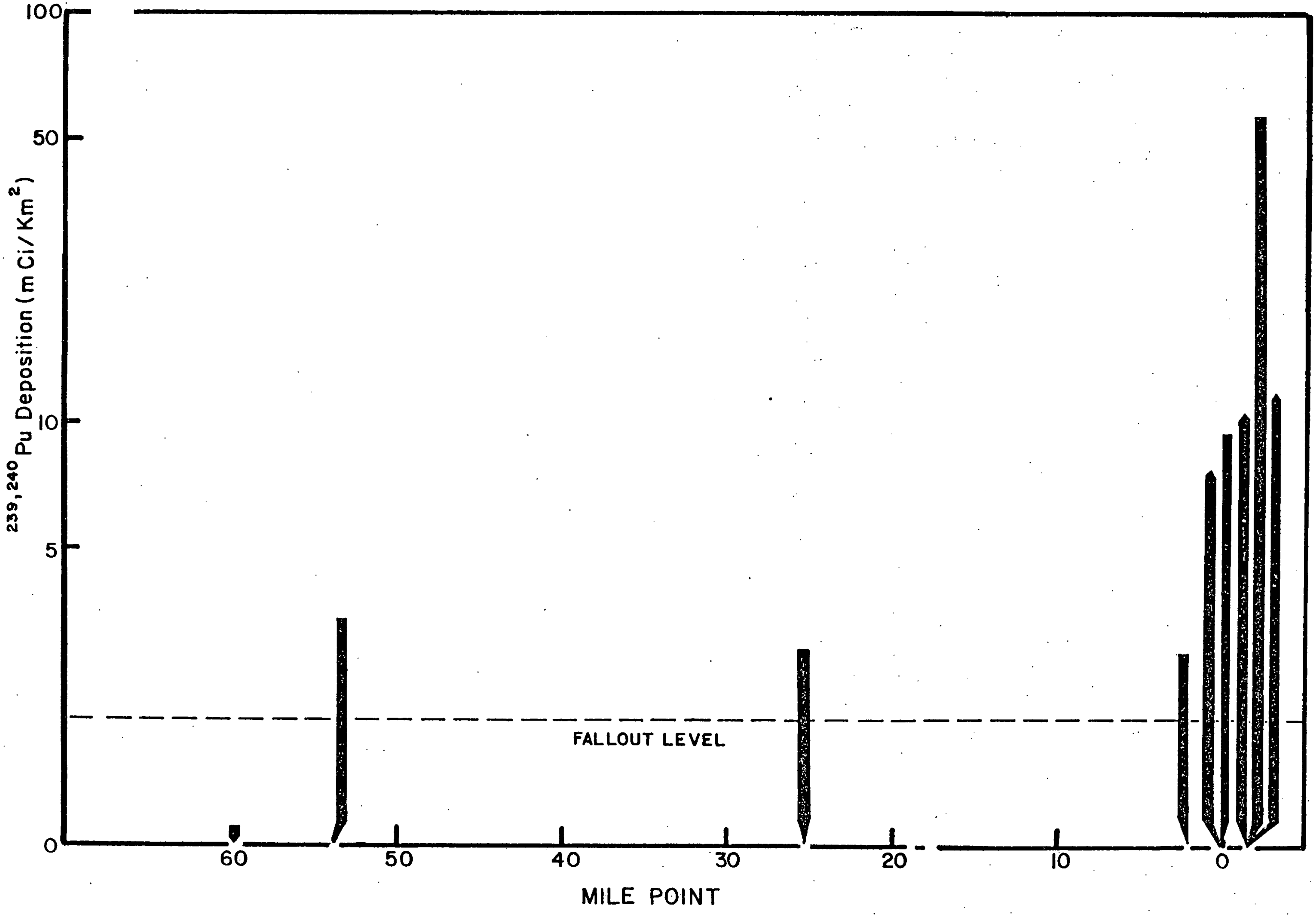
Location (mile point)	Sample No.	Core Depth (Cm)	$^{239,240}Pu$ (mCi/Km^2)
-1.6 E	1051	73	14.6
-1.5 E	1046	60	11.8
P-1.5	1048	260	57.2
0.1 W	1066	70	9.8
0.1 WM	1002	40	8.0
1.9 W	1082	25	2.9
18.6-5	1092	5	0.2
25.3 W	1086	21	2.9
53.8 EC	1052	20	3.5
59.8 EC	1091	15	1.2

FIGURE 4. Cs^{137} accumulation rate vs. mile point: the fallout delivery rate (decay corrected to 1975) is given as a reference line.



1000
500
100
50
10
5
0
FALLOUT LEVEL
^{137}Cs Deposition (m Ci/Km^2)
91
75
60
50
40
30
20
10
0
MILE POINT

FIGURE 5. $Pu^{239,240}$ accumulation rate vs. mile point: the fallout delivery rate is given as a reference line.



100
50
10
5
0
^{239,240}Pu Deposition (mCi/Km^2)
FALLOUT LEVEL
60
50
40
30
20
10
0
MILE POINT

delivery rate). The anomalously high accumulation of $Pu^{239,240}$ could be entirely due to the concentration of fine particles from a large area of the Hudson upstream of the harbor, all of which derived their $Pu^{239,240}$ from fallout delivered to the fresh water or low salinity area of the Hudson surface primarily more than a decade ago. A second possibility is that additional dissolved fallout $Pu^{239,240}$ from the coastal waters became associated with the fine particles brought downstream by the Hudson and deposited in the harbor. A third possibility, which we think is not very likely for reasons discussed earlier, is that the reactor has supplied additional $Pu^{239,240}$. At present we cannot evaluate the likelihood of the second possibility, but we definitely plan to pursue trying to do so.

RESULTS - DELAWARE ESTUARY

We collected gravity cores at ten sites in Delaware Bay and estuary, covering a total reach of the estuary of about 110 kilometers. The seaward end of the sample series was ∿ 15 km upstream of the mouth of Delaware Bay (mile point ∿ 10) in a region that has relatively constant salinity between 25 and $30^{o}/oo$. The upstream end of the series was ∿ mp 78, near Wilmington, Delaware. This area of the estuary has salinities which range from $5^{o}/oo$ down to fresh water, depending upon the discharge of the Delaware River.

The core samples were sectioned into 5 cm intervals and gamma counted in the same way as for the Hudson estuary samples reported earlier. Results for about 30 samples are given in Table 8. Data for Co^{60} are reported, although there is little likelihood of the presence of any measureable amounts of this nuclide in the sediments, to provide an indication of the uncertainty of the Co^{60} data reported in Table 1 for the Hudson.

Gamma-Emitting Radionuclides in Delaware Estuary Sediments

				pCi/kg(a)		
ation	Sample #	Depth	Dry Weight	Cs^{137}	Co^{60}	$K^{40} \times 10^3$
D14-1	1154 A*	0-5	129	40±8(b)	1±7	12.0±0.4
(mp ∿ 10)	B*	5-10	134	17±7	6±7	12.1±0.4
	C*	10-15	130	2±8	10±7	12.0±0.4
AS VII	1151 A	0-5	78	165±15	12±13	14.8±0.5
(mp ∿ 36)	B	5-10	93	110±15	17±12	11.3±0.4
	C	10-15	96	190±20	7±15	14.6±0.6
	D*	15-20	137	2±8	-2±7	7.5±0.3
J3-1	1150 A	0-5	68	37±14	15±13	20.2±0.7
(mp ∿ 47)	B	5-10	72	-7±12	-4±13	17.8±0.6
D2-3	1149 A	0-5	62	335±21	23±15	17.0±0.6
(mp ∿ 50)	B	5-10	91	250±21	-1±16	15.1±0.6
	C	10-15	72	19±13	3±13	16.7±0.6
	D	15-20	69	10±12	20±15	15.4±0.6
Cd #40	1144 A	0-5	68	200±17	1±13	16.0±0.5
(mp ∿ 59)	B	5-10	63	200±18	-6±13	17.9±0.6
	C	10-15	72	235±20	12±14	13.4±0.5
	D*	15-20	73	24±12	24±14	11.3±0.5
	E	20-25	73	215±16	27±14	13.6±0.5
	F	25-30	78	275±20	3±13	17.6±0.6
	G	30-35	73	400±20	31±12	16.7±0.6
C8-1	1142 A	0-5	100	32±10	25±11	13.6±0.5
∿ 60)	B	5-10	98	18±10	8±10	13.9±0.5

				pCi/kg(a)		
Location	Sample #	Depth	Dry Weight	Cs^{137}	Co^{60}	K^{40} x 10^3
[illegible] -	1136 A	0-5	85	63±20	19±18	14.3±0.6
(mp ∿ 63)	B	5-10	120	-6±11	18±13	12.4±0.5
	C	10-15	118	12±9	9±9	12.7±0.4
J-1	1134 A	0-5	123	110±10	11±8	12.5±0.4
(mp ∿ 65)	B	5-10	86	74±14	-2±13	14.0±0.5
	C	10-15	73	9±13	-3±13	16.2±0.6
C3-2	1124 A	0-5	84	20±15	17±14	14.1±0.6
(mp ∿ 75)	B	5-10	93	12±12	11±17	14.2±0.6
B9-1	1120 A	0-5	93	10±18	19±18	12.6±0.6
(mp ∿ 78)	B	5-10	92	19±16	-21±17	13.6±0.6

*Sandy Sediment

(a) Sample specific activities are expressed in terms of dry weight and have been corrected for decay between time of coring and time of assay.

(b) Specific activities >10,000 were rounded to the nearest 100 pCi. Specific activities <10,000 but >1000 were rounded to the nearest 10 pCi. Specific activities <1000 but >100 were rounded to the nearest 5 pCi and specific activities <100 were rounded to the nearest pCi. Statistical errors are reported at 1 sigma. Errors for specific activities >10,000 were rounded to the nearest 10 pCi. Errors for specific activities <10,000 but >1000 were rounded to the nearest 5 pCi and errors for specific activities <1000 were rounded to the nearest pCi.

Activities of Cs^{137} are considerably lower than for Hudson sediment samples (about an order of magnitude lower), and the only evidence of rapid accumulation of recent sediments was from a core in the Chesapeake and Delaware Canal (Cd #40, mp ∿ 59), where Cs^{137} was measured down to 40 cm.

Several of the sampling sites were in areas considered to be shoaling areas (mp ∿ 63, mp ∿ 65, mp ∿ 75), but none of these cores had Cs^{137} activities, or depth trends similar to shoaling areas in New York harbor.

We have analyzed a number of the Delaware sediment samples for plutonium (Table 9). In general, samples with low Cs^{137} also had low $Pu^{239,240}$, as was observed for the Hudson. The highest activity sample measured was ∿ 16 pCi/kg of $Pu^{239,240}$, less than one quarter of the highest activity samples we reported for the Hudson.

DISCUSSION - DELAWARE ESTUARY

Most of the samples which had sufficient activity for confidence in reporting ratios of $Pu^{239,240}$ to Cs^{137} had values of about 5% (Table 10 - six samples). These samples were from areas with maximum seasonal salinities of ∿ 15^{o}/oo or less **and** mean salinities of 10^{o}/oo or less. Two samples had appreciably higher ratios of $Pu^{239,240}$ to Cs^{137}, with the highest value of ∿ 30% near the mouth of Delaware Bay. The next highest ratio was ∿ 9%, for a sample located between the highest ratio sample and those upstream with $Pu^{239,240}$ to Cs^{137} ratios of ∿ 5%.

Thus evidence for a downstream increase in $Pu^{239,240}$ to Cs^{137} ratios which was suggested by the data in the Hudson appears to be even more dramatic in the Delaware.

The total accumulation of $Pu^{239,240}$ per unit of sediment surface area in the Delaware is equal to the fallout delivery rate of ∿ 2 mCi/km^2

Table 9.

Plutonium Isotopes in Delaware Estuary Sediments

Location (Mile Point)	Sample No.	Depth (cm)	Dry Wt. (gm)	% Yield	pCi/kg $^{239,240}Pu$	^{238}Pu	$^{238/239,240}Pu$
D 14-1 (~10)	1154 A	0-5	109.8	57.5	12.1±0.4	-	-
AS VII (~36)	1151 A	0-5	67.6	45.2	15.2±0.6	-	-
J 3-1 (~47)	1150 A	0-5	58.0	20.7	N.D.	N.D.	-
D 2-3 (~50)	1149 A	0-5	52.3	31.4	16.5±0.8	-	-
	B	5-10	70.4	58.1	16.3±0.4	-	-
	C	10-15	60.9	31.6	2.1±0.3	-	-
	D	15-20	58.5	43.7	1.4±0.2	-	-
CD 40 (~59)	1144 A	0-5	58.3	47.3	11.3±0.5	0.5±0.1	0.04±0.01
	B	5-10	52.9	11.4	9.9±1.1	-	-
	C	10-15					
	D	15-20	62.9	8.4	N.D.	-	-
J 1 (~65)	1134 A	0-5	103.3	44.8	5.7±0.3	-	-
	B	5-10	75.3	77.5	2.9±0.2	-	-
	C	10-15	62.6	42.0	0.4±0.1	-	-

N.D. = Not detectable

Table 10

Nuclide Ratios in Delaware Estuary Sediments

Location	Sample #	Depth	$^{239,240}Pu$ pCi/kg	^{137}Cs pCi/kg	$^{239,240}Pu/^{137}Cs$ $\times 10^{-2}$
D14-1	1154 A*	0-5	12.1±0.4	40±8	30.3±6.1
(mp ∿ 10)	B*	5-10	N.A.	17±7	-
	C*	10-15	N.A.	2±8	-
AS VII	1151 A	0-5	15.2±0.6	165±15	9.2±0.9
(mp ∿ 36)	B	5-10	N.A.	110±15	-
	C	10-15	N.A.	190±20	-
	D*	15-20	N.A.	2±8	-
J3-1	1150 A	0-5	N.D.	37±14	-
(mp ∿ 47)	B	5-10	N.A.	-7±12	-
D2-3	1149 A	0-5	16.5±0.8	335±21	4.9±0.4
(mp ∿ 50)	B	5-10	16.3±0.4	250±21	6.5±0.6
	C	10-15	2.1±0.3	19±13	11.1±7.8
	D	15-20	1.4±0.2	10±12	14.0±16.9
CD-40	1144 A	0-5	11.3±0.5	200±17	5.7±0.5
(mp ∿ 59)	B	5-10	9.9±1.1	200±18	5.0±0.7
	C	10-15	N.A.	235±20	-
	D*	15-20	N.D.	24±12	-
	E	20-25	N.A.	215±16	-
	F	25-30	N.A.	275±20	-
	G	30-35	N.A.	400±20	-

Location	Sample #	Depth	^{239,240}Pu pCi/kg	^{137}Cs pCi/kg	^{239,240}Pu/^{137}Cs x 10^{-2}
C8-1		0-5	N.A.	32±10	-
(mp ∿ 60)		5-10	N.A.	18±10	-
J-2		0-5	N.A.	63±20	-
(mp ∿ 63)		5-10	N.A.	-6±11	-
		10-15	N.A.	12±9	-
J-1	1134 A	0-5	5.7±0.3	110±10	5.2±0.5
(mp ∿ 65)	B	5-10	2.9±0.2	74±14	3.9±0.3
	C	10-15	0.4±0.1	9±13	4.4±6.5
C3-2		0-5	N.A.	20±15	-
(mp ∿ 75)		5-10	N.A.	12±17	-
B9-1		0-5	N.A.	10±18	-
(mp ∿ 78)		5-10	N.A.	19±16	-

* Sandy Sediment
N.A. = Not Analyzed
N.D. = Not Detectable

in only a few of the cores we collected. Most of the cores have inventories of only a small fraction of the delivery rate. This contrasts strongly with the situation in the Hudson, where in the zone of major sediment deposition in the harbor the accumulation rates of $Pu^{239,240}$ are an order of magnitude or more greater than the fallout delivery rate. Thus the areas we sampled in the Delaware do not have much accumulation of fallout plutonium, even though we specifically chose our sampling locations to maximize the probability of coring high sedimentation areas of fine-grained sediments. Much of the total area of the Hudson does have relatively low accumulation rates of $Pu^{239,240}$ in the sediments comparable to those we observed here for the Delaware. The major exceptions to this pattern are the high deposition areas of New York harbor and the shallow coves upstream of the harbor. It is possible that analogous areas exist in the Delaware, which could account for the large missing component of the fallout plutonium delivered to the Delaware. However, it now appears more likely that the Delaware estuary doesn't provide nearly as favorable an environment for deposition of recent, fine-grained sediments which contain most of the fallout plutonium as does the Hudson. If this is true, then any future local releases of transuranics to the Delaware might be expected to be transported on fine particles long distance away from the release area, rather than accumulating in a major deposition zone as appears to happen in the Hudson. This conclusion must be considered as tentative at the present time, until the possible existence of major recent sediment accumulation zones in the Delaware is more fully explored.

We did collect one core very close to the site of a large nuclear reactor complex now under construction (mp $\sim$ 50). This particular site, near Artificial Island on the eastern shore of the Delaware, had the highest activity of $Pu^{239,240}$ of any sediment samples we analyzed in the Delaware. Thus that site appears to be favorable for the accumulation of transuranics which might be released locally. This site offers a good potential for future monitoring of transuranic releases once the nuclear generating station becomes operational.

Livingston and Bowen (COO-3563-47; COO-2379-10) recently completed an extensive survey of $Pu^{239,240}$ and Cs^{137} data from coastal sediments of the north-west Atlantic Ocean. The range of values reported for the ratio of $Pu^{239,240}$ to Cs^{137} in several coastal environments was from 0.15 to 0.34. Shallow nearshore samples from a number of cores had total accumulations of $Pu^{239,240}$ comparable to the fallout delivery rate. The trend of the $Pu^{239,240}$ to Cs^{137} ratio in both the Delaware and the Hudson was toward higher downstream values approaching those observed at Woods Hole for coastal sediments. However, the deposition rate of $Pu^{239,240}$ in the Delaware was substantially lower than reported for nearshore cores, many of which were from Buzzards Bay or nearby.

One of the most intriguing observations made by Livingston and Bowen was systematically higher $Pu^{239,240}$ to Cs^{137} ratios in the upper part of the cores, with values near the surface up to five times higher than those twenty or more centimeters down the core. They suggested this indicated upward migration of plutonium relative to Cs^{137}, which could potentially release plutonium again to the water column.

We have not observed the same depth trends in the Hudson but the deposition rates of sediment in the Hudson are much higher than those studied by Livingston and Bowen, who suggested that their conclusions could not be extended directly to high deposition environments. In fact, the trend in one of the cores we have analyzed in Upper New York Bay is increasing $Pu^{239,240}$ to Cs^{137} ratios <u>down</u> the core. As we discussed, this probably results from a downward transition from sediment influenced by recent influxes of reactor Cs^{137}, with peak releases in 1971, into sediments dominated by fallout with the delivery peak in the mid 1960's. We will probably have to wait until analyses are completed on several more long cores from the harbor and other areas before the depth trends of the $Pu^{239,240}$ to Cs^{137} ratio can be examined in detail in the Hudson.

RESULTS AND DISCUSSION - RADIONUCLIDES IN SEDIMENTS NEAR OYSTER CREEK and MILLSTONE POINT REACTORS

As mentioned earlier, we participated in an extensive field sampling program at Barnegat Bay during 1976. The results of this study are discussed in the annual report for a separate contract (E11-17-2185; COO-2185-10). We have analyzed sediments from Oyster Creek, a small tributary of Barnegat Bay, which receives the cooling water from the Oyster Creek nuclear power plant, by gamma spectrometry, and for plutonium by alpha spectrometry. Our primary purpose for the plutonium measurements was to try to establish how sensitive the use of Pu^{238} to $Pu^{239,240}$ ratios might be for characterizing in sediments transuranic release from an operating reactor. In the Hudson the movement of fine particles past the reactor site and downstream is apparently quite vigorous, which tends to dilute the specific activity of reactor releases on sediment particles. In Oyster Creek, the movement of fine particles away from the immediate vicinity of the reactor site appears to be considerably less rapid than for the Hudson, since the observed concentrations of Co^{60} are approximately an order of magnitude higher than for the Hudson (Table 11). The specific activities of $Pu^{239,240}$ and Pu^{238} and the isotopic ratio of $Pu^{238}/Pu^{239,240}$ for the Oyster Creek sediments were not unusually high (Table 11), indicating that if transuranic releases have occurred from Oyster Creek, the increments of $Pu^{239,240}$ and Pu^{238} are not distinguishable from the burden of fallout plutonium in the sediments.

Samples of sediments from near the Millstone Point reactor, in an even more restricted sedimentary environment than for Oyster Creek, had very high Co^{60} activities (more than two orders of magnitude higher than found in the Hudson near Indian Point). These samples did have anomalously high Pu^{238} to $Pu^{239,240}$ ratios, although specific activity of $Pu^{239,240}$ is certainly within the range we have observed for fine-grained estuary and coastal sediments. Thus the ratio of Pu^{238} to $Pu^{239,240}$ does appear to be indicative of reactor releases in sediments in some situations.

LAMONT-DOHERTY QUALITY CONTROL AND LABORATORY INTERCOMPARISON PROGRAM

The most common type of sample analyzed for transuranics in our laboratory over the last three years has been fine-grained estuarine sediments. Usually these samples have been sections of sediment cores which were air-dried, ground to a fine powder with mortar and pestle, and analyzed by gamma spectrometry [Ge (Li) detector] using counts of 8 - 24 hours. The primary standard which we have used for reporting our gamma spectrometry results for ^{137}Cs and ^{60}Co is NBS River Sediment SRM 4350. ^{134}Cs activities have been related indirectly to the same NBS standard through a detector efficiency versus energy curve derived from a sample of Hudson sediments tagged with ^{226}Ra, assuming secular equilibrium with the

TABLE 11

Radionuclides Sediments near Oyster Creek and Millstone Point Reactors

Sample	^{137}Cs	^{134}Cs	^{54}Mn	^{60}Co	$^{239,240}Pu$	^{238}Pu	$^{238}Pu/^{239,240}Pu$
Oyster Creek (1200A)	907±32	58±15	298±43	4909±71	45.9±2.1	2.7±0.3	0.059
Oyster Creek (1200B)	889±33	80±17	290±44	4645±69	43.9±1.5	2.2±0.2	0.050
Oyster Creek (1200C)	-	-	-	-	50.8±1.8	2.3±0.3	0.045
Millstone Point	14,450±410	-	-	81,820±1550	38.2±1.3	5.6±0.4	0.15
Millstone Point	12,820±290	-	-	99,300±1700	39.1±1.2	7.4±0.4	0.19
Millstone Point	-	-	-	-	35.7±1.1	5.4±0.3	0.15
Indian Point (Slosh III)	2700±72	345±42	-	400±27	25.0±1.6	1.0±0.1	0.04

short-lived radioactive daughters produced by decay of ^{226}Ra. The yield tracer and basis of reporting of ^{239,240}Pu and ^{238}Pu activities has been a ^{242}Pu spike solution supplied to us by HASL.

One of the approaches we have taken in trying to assess the reproducibility of our laboratory results, other than repetitive analysis of primary standards, has been to mix a large quantity of Hudson sediment which we attempted to split into subsamples with uniform activities. This provided a large quantity of reasonably homogeneous sediment which we have used for testing analytical procedures and have analyzed repeatedly over a period of several years to obtain some indication of the overall reproducibility of our data. The sediment for this internal "control" substandard was obtained near the Indian Point nuclear power facility and contains readily measurable activities of several gamma-emitting nuclides (including ^{134}Cs which has not been reported for NBS River Sediment SRM 4350). Since our control substandard is very large, it provides a convenient sample for laboratory intercomparisons and the data are of more general interest, because of the obvious presence of reactor-derived fission and activation products.

To obtain the sediment for an internal substandard, we took a number of surface grab samples in early June, 1975, at several locations in the Hudson near Indian Point. These samples were counted immediately, while still wet, to establish the location with the highest specific activity of reactor nuclides. The location chosen (mile point 42.8) was approximately 30 meters from the east bank in 8 meters of water, and ∿ 100 meters downstream of the Indian Point cooling water discharge. Numerous surface grabs were then taken with a Shipek sampler at this location and combined into one large sample, SLOSH III (Standard Lamont Observatory Sediment from the

Hudson).

This composite sample, consisting of highly liquified light brown (oxidized) mud from the upper 10 cm of Hudson sediment, was wet sieved through a 250 micron sieve to remove any pebbles, shell fragments, coal, or large organic detritus that was present. The sediment was then air dried, ground with a mortar and pestle, and placed in a 60 liter plastic barrel (Deldrum^{Tm}) that was attached to a mechanism which caused it to tumble off center, producing fairly vigorous mixing as the barrel rotated. The sample size at this time was ∿ 25 kg.

After mixing for ∿ 4 hours, aliquots of ∿ 100 grams were withdrawn at intervals of 20 minutes as the barrel continued to rotate, and added at random to smaller containers. The subsampling operations required approximately two days to produce twenty composite samples of ∿ 1250 grams each, designated with index numbers 1000A - 1000T. Each of the smaller containers was then mixed, and was remixed prior to any withdrawal of smaller samples (50 - 100 grams). Aliquots from the kilogram-sized subsamples were taken and analyzed by gamma spectrometry (Table 12).

Data for ^{137}Cs in 22 samples of SLOSH III indicate that we were reasonably successful in producing a large number of subsamples of comparable activity (Table 12). Data reported for one of the samples (Composite) were averaged from 5 separate aliquots counted separately. One sample (1000 F-1) was quite different from the rest of the aliquots analyzed, including another (1000 F-2) from the same kilogram-size container. This sample was ∿ 10 sigma from the mean value of the rest of the data, and was excluded from calculation of the mean.

The ^{137}Cs activities of 6 of the 21 samples (excluding 1000 F-1) were further than 1 sigma from the mean of the samples, and 2 were between 2

Table 12

Laboratory Internal Standard

Gamma-Emitting Radionuclides in SLOSH III

		pCi/kg			
Sample #	Dry Weight	Cs^{137}	Cs^{134}	Co^{60}	K^{40} x 10^3
1000 A-2	95	2470±54(c)	370±43	365±17	16.6±0.6
1000 D-1	88	2600±52	435±48	320±15	17.5±0.6
1000 E-1	88	2510±52	375±43	345±16	16.9±0.6
1000 F-1	87	3960±94	665±77	570±30	18.0±0.8
1000 F-2	88	2720±65	445±52	280±19	18.0±0.7
1000 G-1	88	2680±54	445±49	295±15	17.8±0.6
1000 H-1	87	2610±77	445±58	665±35	18.1±0.4
1000 I-1	88	2610±52	375±43	365±15	17.5±0.6
1000 J-1	88	3010±97	480±67	340±34	17.5±0.9
1000 K-1	87	2580±52	375±43	340±16	18.3±0.6
1000 L-1	89	2450±51	325±37	290±15	17.2±0.6
1000 M-1	88	2490±50	355±40	380±16	17.7±0.6
1000 N-1	88	2500±54	370±43	340±17	18.2±0.6
1000 O-1	92	2470±57	310±38	295±18	17.0±0.6
1000 Q-1	88	2630±54	340±40	320±16	17.0±0.6
1000 Q-2	88	2920±65	500±54	385±29	19.1±0.7
1000 Q-3	90	2780±76	430±52	505±43	19.2±0.8
1000 Q-4	89	2700±72	345±42	400±37	19.2±0.8
Composite	400	2550±27	390±21	390±9	17.3±0.3
1000 R-1	88	2640±67	375±47	330±23	17.0±0.7
1000 S-1	87	2550±53	375±43	305±15	N.D.(d)
1000 T-1	88	2680±63	385±47	310±20	17.4±0.6

and 3 sigma from the mean. For ^{134}Cs, 8 of the 21 samples were between 1 and 2 sigma beyond the mean value (excluding 1000 F-1). Although these deviations from the mean are roughly consistent with a normal statistical distribution expected for a homogeneous sample, the data strongly indicate a distribution skewed toward higher specific activities. All three of the samples which were beyond 2 sigma from the mean ^{137}Cs activity [F-1 (excluded from the mean), J-1, Q-2] were greater than the mean. The mean deviation of the ^{137}Cs activities was approximately twice the mean of the statistical uncertainty (1 sigma) of the counting data of the individual samples. The standard deviation from the mean of the ^{137}Cs activities of the 21 samples was about three times the mean of the mean statistical uncertainty of the counting data of the individual samples. Thus there is clear evidence that we were not successful in producing a set of subsamples of a large (∿ 25 kg) sample of Hudson sediment which have identical ^{137}Cs specific activities within the counting uncertainty of individual aliquots. The primary deviation from homogeneity appears to be a skew toward higher ^{137}Cs activities in 10 - 15% of the samples. However, most of the samples (19 of 22) had specific ^{137}Cs activities within three times the statistical uncertainty (1 sigma) of the counting data of the individual samples, thus providing us with a large number of samples of comparable specific activity of ^{137}Cs for our own laboratory analytical procedure-testing work and for intercomparison purposes with other laboratories.

Our primary purpose in preparing SLOSH III was to obtain a large sample of fine-grained estuarine sediment which would be reasonably homogeneous in transuranic activity. We used ^{137}Cs as a convenient indicator for comparing activities of radionuclides between subsamples because it was the most abundant long half life gamma emitter released from Indian Point, and also because we could obtain reasonably good counting statistics (1 sigma ∿ 2%) with 8 - 16 hours of counting time for 50 - 100 gram samples. From the data we have obtained up to now the activities of both $^{239,240}Pu$ and ^{238}Pu appear to be reasonably constant in subsamples of SLOSH III (Table 13). The isotopic ratio of ^{238}Pu to $^{239,240}Pu$ in the samples is consistent with a fallout source for these nuclides in Hudson sediments, even in close proximity to Indian Point.

The activities of ^{134}Cs and ^{60}Co in SLOSH III are about an order of magnitude lower than for ^{137}Cs, so the uncertainty in the activities of these nuclides in the subsamples is somewhat greater (Table 12), but the variations of activity do appear to be related to variation in ^{137}Cs activity. The sample (1000 F-1) highest in ^{137}Cs was also highest in ^{134}Cs and ^{60}Co. Table 14 summarizes several nuclide ratios for subsamples of SLOSH III.

During the last year we have begun a program of sample exchange to provide some indication of the consistency of our data with that of other laboratories. Vaughan Bowen's group at Woods Hole and Herb Volchok and others at the Health and Safety Laboratory of ERDA have analyzed samples of SLOSH III for several nuclides (Table 15). Aliquots of subsample 1000 Q of SLOSH III were analyzed for ^{137}Cs, ^{134}Cs and ^{60}Co by gamma spectrometry at Lamont-Doherty with no chemical separations employed, and were then analyzed at WHOI and HASL by the procedures usually followed in those laboratories. The data listed for 1000 Q-6 in Table 15 were derived from Ge (Li) counting of dry sediment samples at HASL using basically the same technique as followed in our laboratory at Lamont-Doherty. The data for 1000 Q-1 which were determined at WHOI were obtained by Ge (Li) gamma

Table 13

Laboratory Internal Standard

Plutonium Isotopes in SLOSH III(a) Sediments

			pCi/Kg		
Sample #	Dry Weight	% Yield	$^{339,240}Pu$	^{238}Pu	$^{238/239,240}Pu$
Pu 44	82.0	51.8	28.4±0.7	-	-
1000 K-1	76.4	18.8	28.0±1.6	0.8±0.2	0.03±0.008
1000 Q-2	88.5	25.3	26.0±2.0	1.5±0.2	0.06±0.009
Q-3	89.7	15.2	26.3±2.3	0.9±0.2	0.03±0.008
Q-4	78.8	58.4	25.0±1.6	1.0±0.1	0.04±0.005
Composite	400.2	28.2	26.5±0.5	1.1±0.1	0.04±0.004

Table 14

Laboratory Internal Standard

Nuclide Ratios in SLOSH III

Sample #	$Pu^{239,240}$ pCi/kg	Cs^{137} pCi/kg	$\frac{Pu^{239,240}}{Cs^{137}}$ x 10^{-2}	$\frac{Cs^{134}}{Cs^{137}}$ x 10^{-2}	$\frac{Co^{60}}{Cs^{137}}$ x 10^{-2}
1000 A-2		2470±54		15.0±1.8	14.8±0.8
1000 D-1		2600±52		16.7±1.9	12.3±0.6
1000 E-1		2510±52		14.9±1.7	13.7±0.7
1000 F-1		3960±94		16.8±2.0	14.4±0.8
1000 F-2		2720±65		16.4±2.0	10.3±0.7
1000 G-1		2680±54		16.6±1.9	11.0±0.6
1000 H-1		2610±77		17.1±2.3	25.5±1.5
1000 I-1		2610±52		14.4±1.7	14.0±0.6
1000 J-1		3010±97		15.9±2.3	11.3±1.2
1000 K-1	28.0±1.6	2580±52	1.1±0.07	14.5±1.7	13.2±0.7
1000 L-1		2450±51		13.3±1.5	11.8±0.8
1000 M-1		2490±50		14.3±1.6	15.3±0.7
1000 N-1		2500±54		14.8±1.7	13.6±0.7
1000 O-1		2470±57		12.6±1.6	11.9±0.8
1000 Q-1		2630±54		12.9±1.5	12.2±0.7
1000 Q-2	26.0±2.0	2920±65	0.9±0.07	17.1±1.9	13.2±1.0
1000 Q-3	26.3±2.3	2780±76	0.9±0.08	15.5±1.9	18.2±1.6
1000 Q-4	25.0±1.6	2700±72	0.9±0.06	12.8±1.6	14.8±1.4
Composite	26.5±0.5	2550±27	1.0±0.02	15.3±0.8	15.3±0.4
1000 R-1		2640±67		14.2±1.8	12.5±0.9
1000 S-1		2550±53		14.7±1.7	11.9±0.6
1000 T-1		2680±63		14.4±1.8	11.6±0.8

Table 15

SLOSH III

INTERCOMPARISON EXPERIMENT

Sample #	^{239,240}Pu	^{238}Pu	pCi/kg ^{137}Cs	^{134}Cs	^{60}Co	$\frac{^{239,240}Pu}{^{137}Cs}$ X10^{-2}	$\frac{^{238}Pu}{^{239,240}Pu}$ X10^{-2}	$\frac{^{134}Cs}{^{137}Cs}$ X10^{-2}	$\frac{^{60}Co}{^{137}Cs}$ X10^{-2}
L.D.G.O.									
1000 Q-1	--	--	2630±54	340±40	320±16	--	--	12.9±1.5	12.2±0.7
-2	26.0±2.0	1.5±0.2	2920±65	500±54	385±29	0.9±0.07	5.8±0.9	17.1±1.9	13.2±1.0
-3	26.3±2.3	0.9±0.2	2780±76	430±52	505±43	0.9±0.08	3.4±0.8	15.5±1.9	18.2±1.6
-4	25.0±1.6	1.0±0.1	2700±72	345±42	400±37	0.9±0.06	4.0±0.5	12.8±1.6	14.8±1.6
-6	--	--	2610±50	410±50	300±20	--	--	15.7±1.9	11.5±1.6
W.H.O.I.									
1000 Q-1	25.3±1.4	1.08±0.23	2720±30	483±38	340±20	0.9±0.05	4.3±0.9	17.8±1.4	12.5±0.7
"	24.5±1.0	0.79±0.14	2720±30	445±62	--	0.9±0.04	3.2±0.6	16.4±2.3	--
H.A.S.L.									
1000 Q-6	--	--	2680±30	440±50	320±20	--	--	16.4±1.9	11.9±1.5

Lamont Doherty Geological Obs. (L.D.G.O.)
Woods Hole Oceanographic Inst. (W.H.O.I.)
Health and Safety Laboratory of ERDA (H.A.S.L.)

spectrometry for ^{137}Cs and ^{134}Cs after chemical separation, and by non-destructive Ge (Li) gamma spectrometry for ^{60}Co (photo peak at 1.33 Mev). ^{239,240}Pu and ^{238}Pu were analyzed at WHOI by the method which has been published previously by that laboratory. We have used the same analytical procedure for our plutonium data.

We are continuing our exchange of samples with the Woods Hole group, and report preliminary results of gamma spectrometry analysis of six samples provided to us by Woods Hole. These data were obtained during a period in which our Ge (Li) detector was degrading rapidly, and should be considered only as preliminary results. All but one of Woods Hole intercomparison samples (#4) appear to have considerably lower ^{137}Cs activities than most of the Hudson sediment samples which we have analyzed, and two of the samples (#5, #6) are not distinguishable from blank values by our analytical techniques. After our detector has been replaced by Princeton Gamma Tech (at no cost), we will reanalyze these samples by gamma spectrometry and then start them through our alpha spectrometry procedures. We also plan to analyze other intercalibration samples during the coming year.

Table 16

WOODS HOLE INTERCOMPARISON SAMPLES

(Preliminary Results)

Sample #		^{137}Cs dpm/g	^{137}Cs pCi/kg
1	A *	0.144±0.028	65±13
	B	0.186±0.015	84± 7
2	A	0.336±0.038	151±17
	B	0.373± 0.018	168± 8
3	A	0.229±0.034	103±16
	B	0.226±0.017	102± 8
4	A	1.265±0.059	570±27
	B	1.274±0.050	574±22
5	A	0.034±0.021	15± 9 **
6	A	0.018± 0.020	8± 9 **

* A and B represent duplicate runs of the same sample.

** We cannot distinguish these activities from blank values by our analytical technique.

TRANSPORT OF PLUTONIUM BY RIVERS

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 partitioning 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.

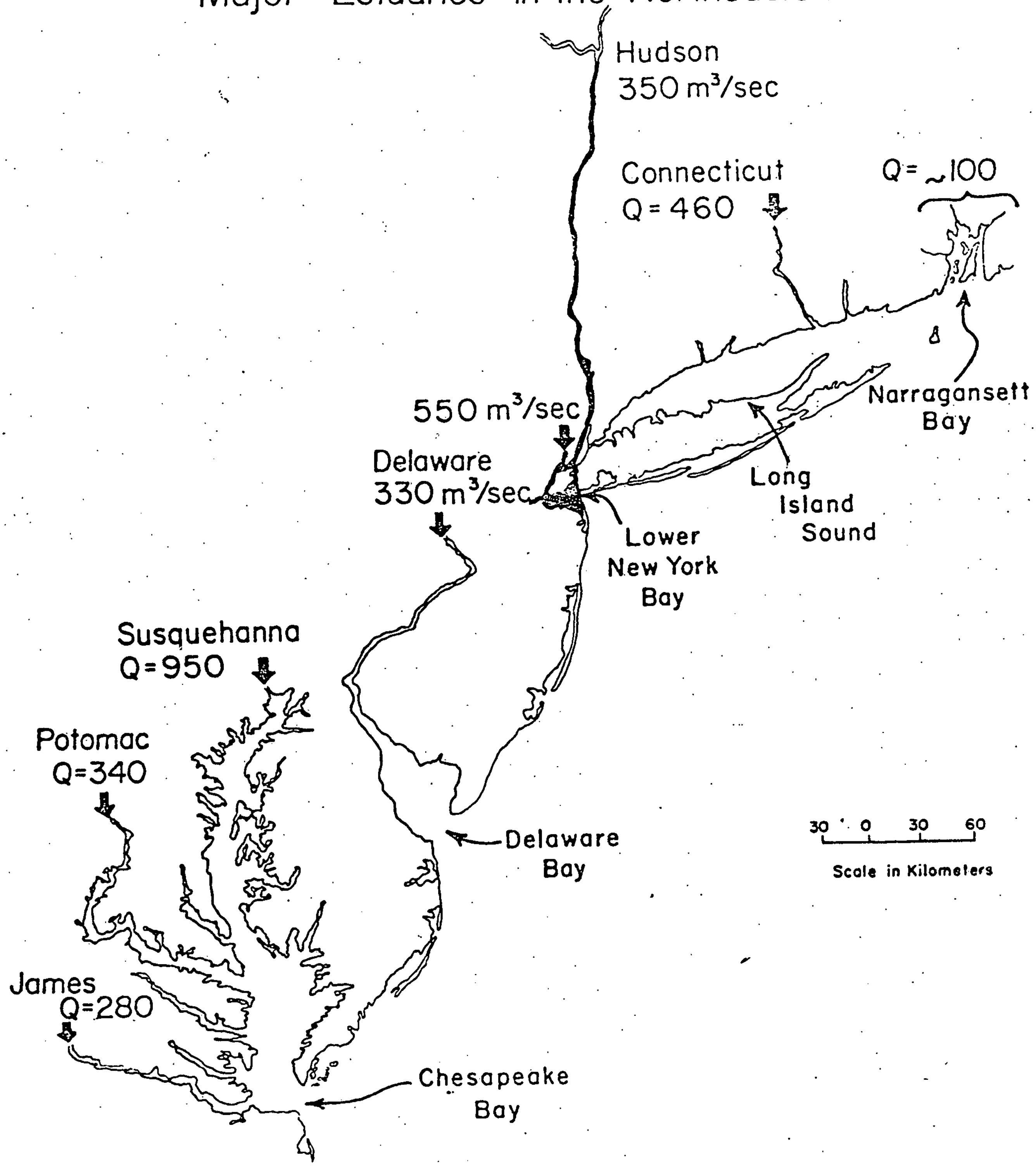
There have been relatively few field studies of point source releases of plutonium to river systems. Three areas in the eastern United States which have received such attention are the Savannah River and its tributary downstream of the reprocessing facility in South Carolina, the Miami River (a tributary of the Ohio River) downstream of Mound Labs in Ohio, and streams near Oak Ridge, Tennessee. 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 6). 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 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. 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

Major Estuaries in the Northeastern U.S.



is more than 60 km downstream of the reactor site, and the time scale of transport of fine particles labeled with reactor nuclides 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 plutonium which are resolvable in the presence of the burden of fallout plutonium. Thus the current distribution of plutonium 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 17 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/l, with the median value about 0.3 fCi/l. The current annual 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/l 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 a 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 ± 0.9 and 23.4 ± 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

TABLE 17

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).

transport of ^{239,240}Pu by suspended particles equals that in the dissolved phase when the concentration of suspended particles is ~15 mg/l, 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.

From the quantities of material dredged annually from New York harbor (~4 x 10^6 T) the downstream transport of particles by the Hudson must be about an order of magnitude higher than indicated by multiplying typical 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 downstream transport of ^{239,240}Pu is approximately an order of magnitude 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 ~10^3 years. 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 17). 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 are available for the period 1973-1975 (Table 17). 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/l, with a mean value of ∿ 0.3 fCi/l (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, Bowen, Noshkin and Gavini (COO-3568-6) have observed $^{239,240}Pu$ activities in Lake Ontario (Table 17). 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 ∿ 0.8 fCi/l to ∿ 0.3 fCi/l. The average $^{239,240}Pu$ activity in all five Great Lakes during 1972-1973 was ∿ 0.5 fCi/l (Table 17, Wahlgren and Marshall).

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 (∿ 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/l. Since the global average of suspended load in rivers is about 600 mg/l, 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 1-5 x 10^2.

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 transuranics 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.

APPENDIXES

A: Man-Made Radionuclides and Sedimentation in the Hudson River Estuary.

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

Sciences, 1976, V 194 pp. 179-183.

B: 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. Submitted to: Journal of Sedimentary Petrology, March 1976.

C: 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 presented at joint USSR-USA Symposium, May 1977.

D: Sources of Heavy Metals in Sediments of the Hudson River Estuary.

S.C. Williams, H.J. Simpson, C.R. Olsen and R.F. Bopp.

E: Abstracts from talks given during the past year.

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Man-Made Radionuclides and Sedimentation in the Hudson River Estuary

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

Man-Made Radionuclides and Sedimentation in the Hudson River Estuary

Abstract. *Recently deposited fine-grained sediments in the Hudson River estuary contain radionuclides from global fallout produced by atmospheric bomb tests as well as from low-level releases of a local nuclear reactor. Accumulation rates of these nuclides are dependent on rates of sediment deposition and vary with location in the estuary by more than two orders of magnitude. Within the Hudson estuary, New York harbor is currently the zone of most rapid deposition of sediments containing radionuclides, some of which were released from a nuclear reactor about 60 kilometers upstream of the harbor.*

Many estuaries are sites of rapid sediment accumulation. Shoaling of estuarine harbors often requires extensive dredging operations to maintain adequate navigation depths. There is some general understanding of the processes which cause sediment deposition in estuaries, but patterns of accumulation are usually sufficiently complicated to defy simple prediction (*1*). Many chemically reactive wastes, such as metals from the electroplating industry and some types of radionuclides, are rapidly bound to particles when discharged to turbid estuarine waters. Patterns of accumulation of estuarine sediments are thus of importance both for management of reactive waste discharges and for maintenance of adequate navigation depths. We have exploited the presence of man-made radionuclides in sediments to provide information concerning patterns of recent sediment accumulation in the Hudson River estuary.

The Hudson River forms one of several major estuarine systems which dominate the coastal environment in the densely populated northeastern United States. The New York metropolitan region is dependent on the Hudson estuary for water transportation as well as for disposal of wastes, including treated and untreated domestic sewage and a wide range of industrial chemicals. We have collected sediment cores, most of which were between 0.5 and 1 m in length, at more than 50 localities in the Hudson estuary (Fig. 1). Gravity cores (~ 6 cm in diameter) from sites throughout the range of salinity within the estuary were sectioned into depth intervals (usually 5 cm), dried, and sealed into 100-cm^3 aluminum cans. Gamma-ray emissions were measured by using a lithium-drifted germanium [Ge(Li)] detector and a multichannel analyzer. Because of the good energy resolution of a Ge(Li) detector (≤ 2 kev), a large number of clearly defined photopeaks whose energies are characteristic of individual radioactive nuclides can be observed in the gamma spectrum between 100 and 3000 kev. These peaks include those from naturally occurring radionuclides such as ^{40}K and daughters of ^{238}U and ^{232}Th, as well as from man-made nuclides such as ^{137}Cs (a fission product from bomb testing and nuclear reactor releases), ^{134}Cs (an activation product almost exclusively from nuclear reactor operations), and ^{60}Co (an activation product predominantly from nuclear reactor operations).

The distribution of ^{137}Cs, ^{134}Cs, and ^{60}Co in Hudson estuary sediment cores (Table 1) generally falls within one of three distinct types. The locations of cores representative of each type are shown in Fig. 1, and the distribution of ^{137}Cs with depth in the sediment for each of these three cores is shown in Fig. 2.

Cores from most areas of the estuary have relatively low activities of ^{137}Cs, generally confined to the upper 5 to 10 cm of sediment. Many cores from the sandy main navigation channel (water depth, 10 to 20 m) have an order of magnitude less ^{137}Cs than that in the lowest-activity example shown in Fig. 1 (labeled

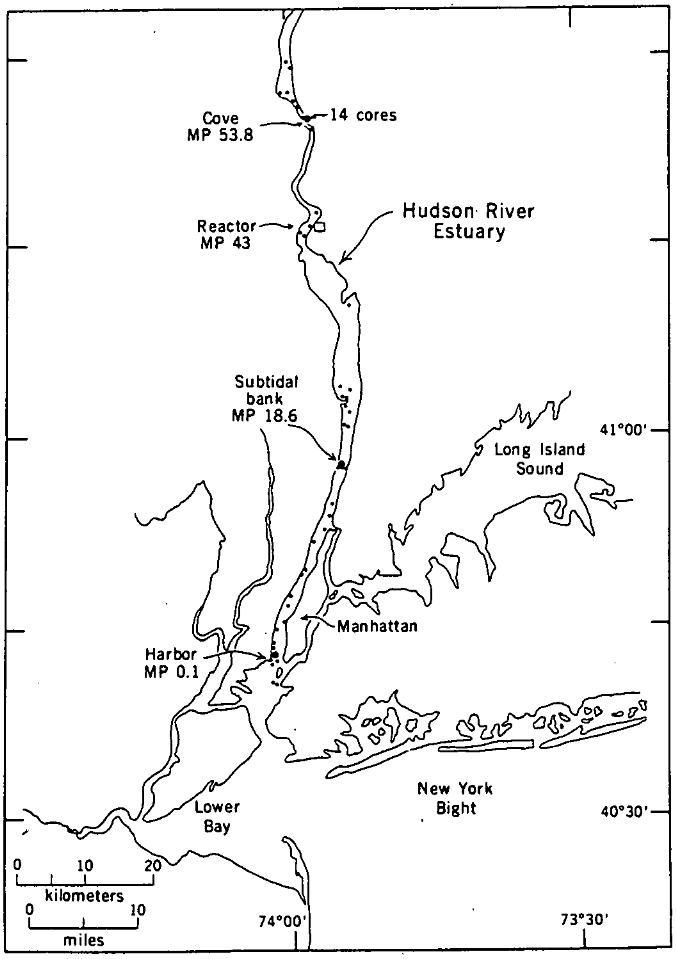

Fig. 1. Location map of analyzed cores from the Hudson estuary. Samples are designated in terms of the number of miles (MP = mile point) upstream from the southern tip of Manhattan. One representative core from each of three locations is indicated by ⊗. The harbor salinity south of Manhattan ranges between 18 and 26 per mil. Saline water (≥ 0.1 per mil) intrudes upstream to ~ MP 15 during springtime high discharge of freshwater and to ~ MP 60 during average summertime conditions (*20*).

"subtidal bank"). The core at mile point (MP) 18.6 was taken on a shallow bank (≤ 8 m) which is more conducive to the deposition of fine-grained particles than the adjacent sandy navigation channel.

Marginal coves show a second type of profile having much more ^{137}Cs in the top 5 cm of sediment per gram (dry weight) and measurable activities usually to 10 to 15 cm although sometimes to 30 to 40 cm. This type of ^{137}Cs depth profile has been reported for the Hudson estuary by other authors and interpreted as indicative of high sedimentation rates in shallow protected environments (*2*). The core reported here (Fig. 1; MP 53.8) is one of more than a dozen cores we have collected from Foundry Cove, a small (0.5 km^2), shallow (1 to 2 m) embayment on the east side of the Hudson River near West Point.

Table 1. Anthropogenic radionuclides in Hudson estuary sediments: discrete samples. Sample locations are given as mile point (MP) upstream of the southern tip of Manhattan (Fig. 1). Uncertainties reported for sediment activities are based only on 1 standard deviation counting statistics for both the photopeak and background adjacent to the photopeak. The ^{60}Co data are averages from two photopeaks. Activities are reported in terms of the date of sample collection. Samples with negative net activities are reported for the counting date and no attempt was made to extrapolate to the date of collection.

Collection date	Location of sample	Depth interval in sediments (cm) or sample type	Activity (pc/kg)			
			^{137}Cs	^{134}Cs	^{60}Co	^{239,240}Pu
25 October 1973	MP 0.1	0–5	1260 ± 38	230 ± 33	190 ± 18	32.7 ± 2.4
		5–10	1795 ± 60	395 ± 56	300 ± 27	31.7 ± 3.2
		10–15	1995 ± 56	480 ± 58	245 ± 18	43.3 ± 3.0
		15–20	960 ± 44	135 ± 33	65 ± 20	45.8 ± 3.2
		20–25	1030 ± 40	175 ± 33	125 ± 19	26.4 ± 1.2
		25–30	570 ± 28	−1 ± 10	59 ± 16	38.5 ± 1.3
		30–35	925 ± 49	22 ± 33	100 ± 28	48.5 ± 1.6
		35–40	1190 ± 40	75 ± 24	152 ± 19	
20 June 1975	MP 18.6	0–1	580 ± 25	19 ± 10	67 ± 10	12.9 ± 0.9
		1–3	525 ± 45	22 ± 21	64 ± 21	12.4 ± 0.8
		3–4	410 ± 20	5 ± 8	44 ± 9	11.1 ± 0.8
		4–5	335 ± 25	35 ± 13	51 ± 16	
		5–10	−1 ± 15	−11 ± 10	−8 ± 13	
11 June 1975	MP 43	0–10	2700 ± 72	345 ± 42	400 ± 27	25.0 ± 1.6
29 August 1973	MP 53.8	0–5	2475 ± 63	98 ± 26	69 ± 19	69.2 ± 6.4
		5–10	1825 ± 68	17 ± 32	19 ± 20	52.1 ± 4.3
		10–15	210 ± 17	−8 ± 10	15 ± 11	5.7 ± 0.4
		15–20	26 ± 17	−15 ± 11	30 ± 15	
		20–25	35 ± 23	0 ± 12	7 ± 18	
		50–55	9 ± 14	7 ± 9	13 ± 15	
30 September 1975	MP 18	Suspended solids*	1260 ± 35	210 ± 25	145 ± 18	23.4 ± 1.0
20 April 1976	MP 24	Suspended solids*	1135 ± 35	101 ± 15	100 ± 12	18.9 ± 0.9

*Samples of suspended sediments were collected with a continuous-flow centrifuge and large settling tanks during two high-runoff episodes, one following a period of abnormally heavy rainfall and the other during the normal spring discharge peak.

Table 2. Average radionuclide ratios in Hudson sediments and reactor releases. Ratios are reported in terms of the activities during the year indicated, even if collected at an earlier date. Ratios for 1975 are reported to 1 November, using half-lives of 30.0 years (^{137}Cs), 2.1 years (^{134}Cs), and 5.2 years (^{60}Co).

Year of reporting	Location and number of samples	Depth interval in sediments (cm)	Source	^{134}Cs/^{137}Cs	^{60}Co/^{137}Cs
1975	MP 43 (1)	0–10	Reactor site; large composite surface sediment	0.13	0.15
1975	MP −1.6 to MP 6 (6)	0–10	New York harbor cores	0.07–0.11	0.07–0.15
1975	MP 53.5 to MP 54 (13)	0–5	Foundry Cove (upstream of reactor site) cores	0.02	0.02
1971	MP 35 to MP 49 (5)	0–10	Reactor region; surface sediment (*2*)	0.6–0.7	0.12–0.36
1971			Reported reactor releases for year of maximum discharge (*8*)	0.73	0.21
1975			Reactor releases (decay corrected 1971 to 1975)	0.22	0.14

The third type of ^{137}Cs profile is illustrated by a core collected in New York harbor at MP 0.1. It shows high ^{137}Cs activities from the surface to the bottom of the core at 40 cm (Fig. 2). We have measured comparable activities to 250 cm in another core from the harbor (*3*). Sediment cores which have high ^{137}Cs activities from the surface to depths below 30 cm are clear indicators of rapid sediment accumulation.

There are two significant potential sources of ^{137}Cs in Hudson sediments: (i) fallout from atmospheric testing of nuclear weapons and (ii) periodic low-level releases from a commercial nuclear power generating station at MP 43 (Fig. 1). The major peak of fallout from weapons testing was delivered to the Hudson in the years 1962 to 1965. Strontium-90 and ^{137}Cs, both of which have radioactive half-lives of about 30 years, are the fission products from the peak fallout years which are found in greatest abundance in soils and the surface ocean throughout the globe. Most of the fallout arrived at the earth's surface in rain and snow. As rainwater percolates into soils of continental areas, ^{90}Sr (*4*) and ^{137}Cs (*5*) are rapidly attached to particulates in the upper 10 to 20 cm of the soil. In the open ocean, fallout ^{90}Sr and ^{137}Cs are predominantly in solution (*6*) and are frequently used as tracers of water circulation because of the minor role of particulate phases in the transport of these nuclides in seawater.

In freshwater lakes such as Lake Michigan and Lake Ontario, most of the ^{90}Sr remains in solution, while a major fraction (> 90 percent) of fallout ^{137}Cs is now found attached to particulate phases in the sediments (*7*). In estuarine systems such as Chesapeake Bay, dissolved ^{137}Cs appears to be roughly proportional to salinity, with very low values typical of salinities less than a few parts per thousand (*8*). Higher values in more saline estuarine waters are usually attributed to dissolved potassium decreasing the removal onto particles of ^{137}Cs supplied by rain. Thus, there appears to be substantial variation in the behavior of fallout ^{137}Cs in natural water systems, dependent on the salinity of the water and the degree of contact with solid phases such as soil particles.

The time history of ^{137}Cs release from the nuclear reactor at MP 43 cannot be defined quite as well as for fallout. The reactor began operating in 1962, but reported releases through 1964 were very low. Activities of specific nuclides in the releases have not been reported for the entire operating period, but estimates for earlier years can be made from the gross activity of all the nuclides in the releases, assuming a composition similar to the most recent data. The highest annual release of ^{137}Cs was in 1971, accounting for almost half of the total discharges of ^{137}Cs since 1962. During the period 1973 and 1974, very little ^{137}Cs activity was released (*9*).

In addition to ^{137}Cs, reactor releases of gamma-emitting nuclides to the Hudson have included ^{134}Cs (half-life = 2.1 years) and ^{60}Co (half-life = 5.2 years). Since ^{60}Co also becomes bound to particles in the Hudson, the proportions of ^{134}Cs and ^{60}Co relative to ^{137}Cs in Hudson sediments can be used in conjunction with the time history of release data to estimate the relative importance of fallout and reactor releases as a source of ^{137}Cs in the sediments.

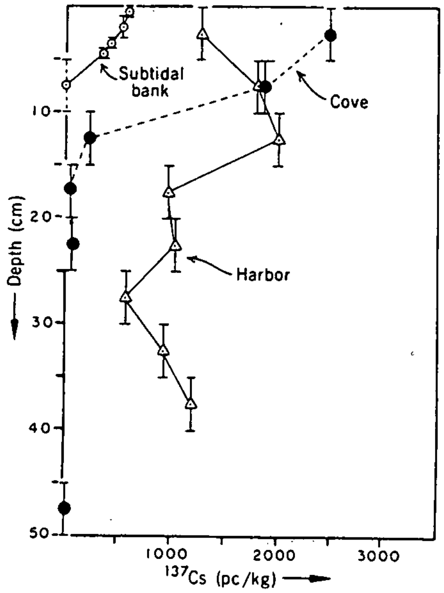

Fig. 2. Cesium-137 depth profiles in Hudson estuary sediments. All data were normalized to National Bureau of Standards environmental radioactivity standard 4350 and are expressed as activity per dry weight (1 pc = 10^{-12} curie). Activities at the core tops vary over a large range, presumably because of variations in sediment grain size, organic content, and percentage of fine-grained particulates which have been exposed to water with a relatively high ^{137}Cs activity over the past decade or so. The profile shown for the subtidal bank site is a composite of four points (0 to 5 cm) from a large grab sample sectioned at 1- to 2-cm intervals and one point (5 to 10 cm) from a core taken at the same site. The core from the harbor shown here has significant ^{137}Cs activity to the bottom of the sample (40 cm). Other harbor cores within a kilometer of this site have similar activities to 250 cm, while others reach background levels between 50 and 60 cm.

Table 3. Summary of budget estimates for ^{137}Cs and ^{239,240}Pu in the Hudson estuary. Values in parentheses have the largest uncertainties.

Source	Activity (curies)	
	^{137}Cs	^{239,240}Pu
Sediment burden		
High sedimentation areas (~ 10 percent of estuary)	~ 5	~ 0.1
Low sedimentation areas (~ 90 percent of estuary)	~ 5	~ 0.1
Direct inputs to estuary		
Fallout on water surface (rain and snow)	~ 25	~ 0.5
Reactor discharge to water	~ 35	?
Other inputs to estuary		
Soil erosion from drainage basin	(~ 10)	(~ 0.2)
Dissolved runoff from drainage basin	(~ 50)	(~ 0.3)
Removal from estuary		
Dredging	~ 20	~ 0.4
Outflow (by difference)	(~ 90)	(~ 0.4)

Nearly all of the sediment samples from the Hudson near the reactor site or downstream to New York harbor in which we have measured ^{137}Cs activity also contain ^{134}Cs and ^{60}Co, in proportions roughly similar to those in surface sediment samples collected near the reactor site (see Table 2; ^{134}Cs/^{137}Cs and ^{60}Co/^{137}Cs ~ 0.1). The cove site approximately 15 km upstream from the release area has relatively low ^{134}Cs/^{137}Cs and ^{60}Co/^{137}Cs (~ 0.02), indicating that a smaller proportion of the ^{137}Cs in this sediment is derived from the reactor than in the sediments of New York harbor.

On the basis of the nuclide ratios reported in Table 2, which include data from many more of our cores than the few itemized in the table plus data for sediments at several localities collected within about 12 km of the reactor site during 1971 by other authors (*2*), we conclude that a substantial fraction of the ^{137}Cs now found in New York harbor sediments was derived from reactor releases, primarily from the period of peak discharge about 5 years ago (*10*).

We can compare the ^{137}Cs activities in Hudson estuary sediments with the magnitudes of several source terms, some of which have reasonably small uncertainties, while others can only be roughly approximated from indirect evidence (Table 3). We estimate the present sediment ^{137}Cs burden between MP 60 and New York harbor to be ~ 10 curies (1 curie = 2.2 × 10^{12} disintegrations per minute), with approximately half of the total in zones of rapid sedimentation which cover ~ 10 percent of the total surface area.

The delivery of fallout ^{137}Cs to the water surface of the Hudson can be estimated more accurately than any of the other budget terms. The total deposition rate corrected for radioactive decay to 1975 has been about 120 mc/km^2 (*11, 12*). Using an area of water surface for the Hudson between MP 60 and MP 0 of 200 km^2, the total direct fallout deposition of ^{137}Cs to the estuary surface is ~ 25 curies.

The total supply of ^{137}Cs from the reactor since 1967 has been ~ 35 curies with ~ 20 curies released in 1971. Thus, direct fallout and reactor releases have supplied comparable amounts of ^{137}Cs to the Hudson estuary.

There is a very large reservoir of fallout ^{137}Cs in the soils of the Hudson drainage basin (~ 4 × 10^3 curies for the total drainage basin of ~ 3.5 × 10^4 km^2). Some of the ^{137}Cs from this soil reservoir has undoubtedly reached the Hudson estuary, but the magnitude of this source is not simple to establish. We have estimated the input from the drainage basin to be ~ 60 curies, but this number has a large uncertainty (*13*).

The rate of removal of ^{137}Cs from the Hudson estuary is also difficult to establish. Dredging of New York harbor is nearly continuous at a few extremely rapid shoaling areas, and occurs at least every few years at many other sites (*14*). During the past 15 years, the average annual removal of sediments has been ~ 4 × 10^6 tons (dry weight) (*15*). Assuming that 50 percent of this amount is taken

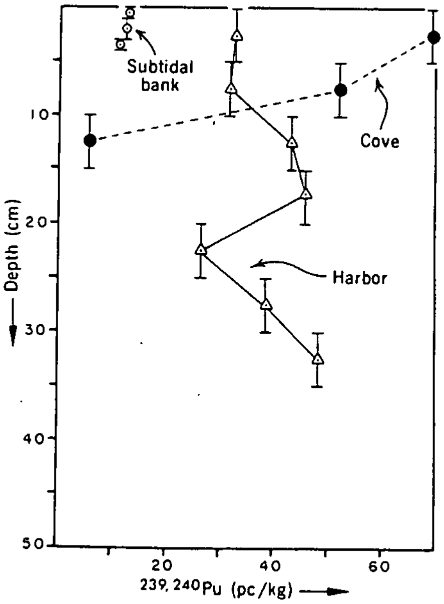

Fig. 3. Depth profiles of ^{239,240}Pu in Hudson estuary sediments, expressed as activity per dry weight. The distribution of ^{239,240}Pu in these cores is similar to that of ^{137}Cs (Fig. 2).

from shoaling areas with fine-grained sediments having a mean ^{137}Cs activity of 1 pc/g, then the total removed over 10 years has been ~ 20 curies. We have no direct means for estimating loss of ^{137}Cs in solution and on suspended particulates. Assuming our other budget estimates are correct, the total outflow loss has been ~ 90 curies.

We have also analyzed a number of Hudson sediment samples for ^{239,240}Pu (*16*). Concentration profiles of ^{239,240}Pu in the three sediment cores discussed earlier are shown in Fig. 3. Variations in the activity of ^{239,240}Pu with depth are not identical to those for ^{137}Cs in the harbor core, but the general similarity in the distribution of both radioactive elements within each of the three cores is obvious. This would be expected if transport and accumulation on particulates were important for both ^{239,240}Pu and ^{137}Cs. Covariance of fallout ^{137}Cs and ^{239,240}Pu in sediments of freshwater lakes has also been observed in the Great Lakes (*7*) and in soil profiles (*5*). In the ocean, ^{239,240}Pu is much more rapidly and completely associated with particulate phases than is ^{137}Cs (*17*).

We cannot establish budget terms for ^{239,240}Pu in the Hudson as well as for ^{137}Cs at the present time. The activities of ^{239,240}Pu present in global fallout are 1 to 2 percent of that of ^{137}Cs. The ratio of ^{239,240}Pu to ^{137}Cs in Hudson sediments reported here ranges between 1 and 7 percent, with most samples in the range 2 to 3 percent. By analogy to the ^{137}Cs budget for the Hudson, the sediment burden is ~ 0.2 curie (Table 3), while the direct input from fallout is ~ 0.5 curie and the loss by dredging is ~ 0.4 curie. We estimate the contribution of ^{239,240}Pu from the soils of the drainage basin to be ~ 0.5 curie (*18*), a smaller proportion of the input than was true for ^{137}Cs. Again, this input is not well determined. We do not have any direct estimate of ^{239,240}Pu which might be derived from the reactor at MP 43. If such releases have occurred, they do not appear to have been large compared with the fallout source.

Our primary conclusions follow.

1) Cesium-137 can be a valuable tracer for recent (last 10 to 15 years) accumulation of fine-grained sediment in estuaries (*19*).

2) Particulate phases play a significant role in the transport processes for both ^{137}Cs and ^{239,240}Pu in turbid estuarine waters.

3) Sedimentation in the Hudson estuary occurs primarily in the harbor adjacent to New York City and in freshwater or low-salinity shallow marginal coves. Accumulation rates of sediment in the harbor approach 5 to 10 cm/year over large areas, which is more than an order of magnitude greater than the long-term (last 10^4 years) average for the Hudson. Recent sediments within the harbor contain readily measurable activities of gamma-emitting nuclides derived from a nuclear reactor and released more than 60 km upstream of New York harbor. The activities of reactor nuclides in Hudson sediments are comparable to those of fallout nuclides found in surface soils on a global basis and do not currently appear to present a biological hazard to humans. The present distribution of nuclides derived from low-level releases of a reactor which has been in operation for more than a decade can provide valuable information as to the likely transport pathways of radionuclides in the event of a major release to the Hudson estuary.

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References and Notes

1. H. B. Simmons and F. A. Hermann, Jr., *Geol. Soc. Am. Mem. 133* (1972), p. 555; R. H. Meade, *ibid.*, p. 91.
2. J. W. Lentsch, thesis, New York University (1974); L. M. Hairr and M. E. Wrenn, paper presented at the annual meeting of the American Nuclear Society, Las Vegas, Nevada (1972); M. E. Wrenn, J. W. Lentsch, M. Eisenbud, G. J. Lauer, G. P. Howells, in *Proceedings of the Third National Symposium on Radioecology* (Oak Ridge, Tenn., 1971); M. E. Wrenn, S. M. Jinks, L. M. Hairr, A. S. Paschoa, J. W. Lentsch, in *Proceedings of the Second International Symposium on the Natural Radiation Environment*, J. A. S. Adams, W. M. Lowder, T. F. Gesell, Eds. (CONF-720805-P2, National Technical Information Service, Springfield, Va., 1972), p. 897; J. W. Lentsch, M. E. Wrenn, T. J. Kneip, M. Eisenbud, in *Proceedings of the Fifth Annual Health Physics Society Mid-Year Topical Symposium*, P. Voilleyue, Ed. (Idaho Falls, Idaho, 1970).
3. A few harbor cores collected in deep channels show low or undetectable ^{137}Cs activities. Boundaries between zones of high activity and zones of background activity within a core are often quite sharp. In one core in the harbor, a 15-cm-thick sandy layer with background activities of ^{137}Cs was observed between layers of fine-grained sediment with high activities.
4. H. L. Volchok, *J. Geophys. Res.* 71, 1515 (1966); G. H. Hamada, E. P. Hardy, J. T. Alexander, *USAEC Rep. HASL-33* (1958).
5. E. P. Hardy, *USAEC Rep. HASL-286* (1974).
6. T. R. Folsom, C. Sreekumaran, N. Hansen, J. M. Moore, R. Grismore, *USAEC Rep. HASL-217* (1970); W. S. Broecker, E. R. Bonebakker, G. R. Rocco, *J. Geophys. Res.* **71**, 1999 (1966); V. T. Bowen and W. Roether, *ibid.* **78**, 6277 (1973).
7. M. A. Wahlgren and D. M. Nelson, *Verh. Int. Ver. Limnol.* **19**, 317 (1975); M. A. Wahlgren and J. S. Marshal, in *International Symposium on Transuranium Nuclides in the Environment* (IAEA-SM-198/39, International Atomic Energy Agency, Vienna, 1975); V. T. Bowen and V. E. Noshkin, *USAEC Rep. C00-3568-3* (1973).
8. G. K. Riel, in *Proceedings of the Second International Symposium on the Natural Radiation Environment*, J. A. S. Adams, W. M. Lowder, T. F. Gesell, Eds. (CONF-720805-P2, National Technical Information Service, Springfield, Va., 1972), p. 883.
9. R. S. Booth, *USERDA Rep. ORNL-TM-3801* (1975); Consolidated Edison of New York, Inc., Indian Point semiannual operations reports 21, 22, 23, and 24 (1973–1974).
10. Although we have observed sharp peaks of ^{137}Cs activity corresponding to the year of maximum reactor release in some harbor cores not reported here, such a peak does not always occur. Several processes may act to obliterate this curve, including mixing of the sediments by organisms and gas bubbles formed within the sediments, and local resuspension and deposition of sediments during dredging operations. Also, it appears likely that a substantial fraction of the releases become bound to particles within a few kilometers of the point of discharge and are then transported downstream in the suspended load. Suspended particles from near the water surface of the Hudson collected about halfway between the reactor and New York harbor during 1975 and 1976 have man-made nuclide activities similar to those in surface sediments near the reactor and in sediments which have accumulated in New York harbor over the past several years (Table 1). Those suspended particles do not have nuclide ratios typical of fresh reactor releases (< 1 year since discharge) and probably include some fraction of particulates which have been deposited and resuspended many times as they pass down the estuary. Such a process would produce a complicated depth profile of radionuclides in sediments of a downstream zone of accumulation which may not be simply related to the time history of releases.
11. Annual fallout ^{137}Cs delivery to the Hudson was computed from ^{90}Sr data for New York City (*12*), assuming a ^{137}Cs/^{90}Sr ratio of 1.5.
12. *USERDA Rep. HASL-294* (1975), appendix.
13. We estimated the drainage basin supply of ^{137}Cs as two components: particulate and dissolved. Assuming that all of the suspended solids in the Hudson River are soil particles with an average ^{137}Cs activity of 1 pc/g, which is approximately equal to that found in the upper 5 cm of soil from a well-studied site on Long Island (*5*), the supply of ^{137}Cs in 10 years is ~ 10 curies for an average river flow of 500 m^3/sec and a suspended solids load of 60 parts per million. Three lines of evidence indicate that the supply of dissolved ^{137}Cs from the drainage basin is a very small fraction (~ 1 percent) of the total fallout delivered to the soil. New York City drinking water is derived primarily from tributary streams to the Hudson and Delaware rivers. Data on ^{90}Sr and ^{137}Cs from both rainwater and New York City tap water (*12*) indicate that approximately 1 percent of the total fallout ^{137}Cs has been removed in solution from the soil zone. This estimate is based on the assumptions that tap water is representative of Hudson tributaries and that negligible losses of ^{137}Cs occur in reservoir storage and water treatment. A second line of evidence is provided by published data on the annual average ^{137}Cs activity in the Hudson near the reactor site at MP 43. Activities declined from 1 to 2 pc/liter to 0.01 to 0.1 pc/liter between 1964 and 1968 (*2*), approximately following the trend in ^{137}Cs levels in precipitation (*11*, *12*) with absolute concentrations averaging a few percent of the precipitation values. A third line of evidence is provided by published data for dissolved ^{137}Cs in Chesapeake Bay during 1970 to 1972 (*8*), where freshwater and low-salinity ($\leq$ 2 per mil) samples had activities ($\leq$ 0.02 pc/liter) less than one-third of those of New York City tap water for the same years. Using reported data for the Hudson for 1964 to 1968 (*2*) and extrapolations from years before 1964 and after 1968 we estimated the total drainage basin input of soluble ^{137}Cs to the Hudson estuary to be ~ 50 curies.
14. F. L. Panuzio, *U.S. Dep. Agric. Agric. Res. Serv. Misc. Publ. 970* (1965); U.S. Army Corps of Engineers, New York, unpublished dredging records.
15. M. G. Gross, *Geol. Soc. Am. Bull.* **83**, 3163 (1972).
16. Samples were prepared by chemical procedures similar to those of K. M. Wong [*Anal. Chim. Acta* **56**, 355 (1971)], using ^{236}Pu and ^{242}Pu as yield tracers and counted with an alpha-particle spectrometer. The peaks in the alpha-particle energy spectrum for ^{239}Pu and ^{240}Pu are not resolved, so data obtained by alpha spectrometry are reported as the sum for these two nuclides, ^{239,240}Pu.
17. V. T. Bowen, K. M. Wong, V. E. Noshkin, *J. Mar. Res.* **29**, 1 (1971); V. E. Noshkin, *Health Phys.* **22**, 537 (1972).
18. At present our only line of evidence to estimate soluble ^{239,240}Pu released from soils is from analysis of New York City tap water for 1973 to 1975. The ratio of ^{239,240}Pu to ^{137}Cs in tap water averaged ~ 0.3 times the fallout ratio [B. G. Bennett, *USAEC Rep. HASL-306* (1976)]. Assuming

that only ~ 1 percent of the fallout ^{137}Cs has left the soil in solution (*13*), then ~ 0.3 curie of $^{239,240}Pu$ would have reached the Hudson in solution from the drainage basin soils.

19. A similar conclusion was reached in a recent study of low-level releases of radioactive wastes into Bombay harbor [B. Patel, C. D. Mulay, A. K. Ganguly, *Estuarine Coastal Mar. Sci.* 3, 13 (1975)].
20. H. J. Simpson, R. Bopp, D. Thurber, in *Hudson River Ecology, Third Symposium on Hudson River Ecology* (Bear Mountain, N.Y., March 1973), paper 9; K. A. Abood, *Ann. N.Y. Acad. Sci.* **250**, 39 (1974).
21. We thank J. Kelly of Consolidated Edison of New York, Inc., for providing copies of operating reports for Indian Point, N. Chu of the Health and Safety Laboratory of the Energy Research and Development Administration for advice on plutonium chemistry and for supplying ^{236}Pu and ^{242}Pu spikes, and G. Mathieu and P. Breland for help in the coring operations. Financial support was provided by ERDA contract E (11-1) 2529. Contribution No. 2390 from Lamont-Doherty Geological Observatory of Columbia University.

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A GEOCHEMICAL ANALYSIS OF THE SEDIMENTS AND SEDIMENTATION IN THE HUDSON ESTUARY[1,2]

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ABSTRACT

Geochemical and sedimentological studies have been conducted on one 6 m core and on a series of 1-3 m cores taken over ∿ 100 km of the Hudson Estuary. Each gram (dry-weight basis) of estuarine silt contains: organic matter 30-100 mg; carbonate generally < 30 mg; quartz 250-400 mg; potassium 18-24 mg; zinc 50-550 μg; copper 15-400 μg; and lead 20-800 μg. In most localities, the higher trace metal levels, as well as anthropogenic detritus (e.g., metalliferous slags, fly ash and coal), and reactor- and bomb-produced radionuclides (e.g., ^{137}Cs, ^{134}Cs and ^{60}Co) are confined to the upper 10 cm of the sediment; but in the inner harbor area of New York they have been observed to sediment depths of 250 cm. In some areas of the inner harbor the vertical distribution of anthropogenic radionuclides indicates sedimentation rates of 5-20 cm/yr.

The top 10 cm of the inner harbor sediment is highly liquefied and the top meter shows extensive turbation, although distinct sand layers and laminated zones are present. The turbation may have resulted from the release or entrapment of biochemically formed gases, as well as from mechanical mixing of sediment by organisms. ^{14}C analyses of organic matter in inner harbor surface sediments indicate that the major source of carbon is recent sewage, nearly all of which is discharged in this area of the estuary.

The fine-grained sediments of the natural channel and subtidal bank, upstream of the inner harbor, are characterized by alternating layers of fine sandy silts and clay-rich silts on a mm to cm scale. Downstream from the Tappan Zee, sand-shell layers, 1-20 cm thick, occur at the channel surface and are interlayered

with zones of laminated, fine-grained sediment at depth. Radiocarbon dating of shells and shell layers indicates a minimum net sedimentation rate of 1-3 mm/yr in this area of the estuary during the past 3000 years. In cores taken on the subtidal bank, coarse sand-shell layers are absent, but turbate zones, 1-10 cm thick, occur at the surface and are interlayered with the laminated zones at depth. One possible interpretation of the sedimentary structures is: (1) the laminated, fine-grained sediments are deposited when the Hudson transports relatively high concentrations of sediment, such as during and after severe storms or other short term events of large scale resuspension; (2) the sand-shell layers result from increased sand transport during high-energy (storm) conditions or from tidal scour under normal flow conditions; and (3) the turbate zones represent periods of slower or no deposition during normal flow conditions.

Consequently, it appears that everyday tidal and estuarine processes are causing the rapid accumulation of recent "polluted" sediment in specific areas, such as the inner harbor, whereas storm deposits characterize the sedimentary record in the channel and subtidal bank environments of the river estuary, upstream of New York City. This large variation in both patterns and rates of sediment deposition in the Hudson Estuary has not been previously identified on the basis of grain-size and mineralogical analyses.

INTRODUCTION

Numerous physical, chemical and biological factors affect estuarine sedimentary processes and patterns. These include alternating tidal currents, two-layer (non-tidal) estuarine circulation, estuary morphology, flocculation and bio-agglomeration of fine particles in the water column, mixing of sediments by organisms, human activities and multiple sediment sources. Non-tidal estuarine circulation can be characterized as a lower salinity surface layer with a net seaward flow and a denser, more saline bottom layer having a net landward flow (Pritchard, 1955). Suspended matter carried downstream in the upper layer gradually settles into the more saline bottom layer. Sedimentary matter in the bottom layer can then be transported upstream, either under the influence of tidal processes (Van Straaten and Kuenen, 1958; Postma, 1967) or non-tidal estuarine flow (Meade, 1969). Deposition is most pronounced in areas where the tidal or estuarine flow is hindered or dissipated, such as in coves, around piers, docks and natural points, and near the end of the salinity intrusion (Ippen, 1966; Schubel, 1968). Widespread deposition may also occur during slack water; however, during later tidal cycles such material may be resuspended, transported and redeposited (Schubel, 1968; Nichols, 1972; Oviatt and Nixon, 1975).

This picture of estuarine sedimentation involves only processes that occur on a regular basis during conditions of average river flow. Recently, however, more attention has been given to the erosional and depositional aspects of storms in coastal areas (Reineck and Singh, 1972; Swift *et al*., 1972; Schubel, 1974; Kumar and Sanders, 1976) and several investigators have attributed deposits in the stratigraphic record to storm activity (Ager, 1974; Kelling and Mullin, 1975).

Sedimentological studies in estuaries and near-shore marine environments have traditionally utilized grain-size, mineralogical and suspended solid measurements. Previous work of this type on the Hudson Estuary includes that of Panuzio (1965), McCrone (1967) and Coch (1976). Panuzio (1965) discussed circulation and siltation processes in the estuary, while McCrone (1967) studied the general lithology and cation exchange capacities of the sediment and found no significant differences in the mineralogical and chemical properties of the sediment with depth or in a downstream direction. Coch (1976) conducted grain-size analyses on sediment and concluded that net deposition is occurring along the entire length of the salt-intruded reach of the estuary. No previous study has described the recent sedimentary structures nor has used data related to the time scale of deposition to determine whether the grain-size or mineralogical studies were conducted on recently deposited sediments or on older sediments exposed by erosion, dredging, or no net deposition.

On the basis of anthropogenic radionuclide tracers, ^{14}C ages, trace metal concentrations and sedimentary structures, we have found a large variation in the patterns and rates of sediment deposition in the Hudson. Consequently, previous conclusions based on grain-size and mineralogical studies have been misleading. We show that: (1) most of the suspended matter and sediment transported by everyday estuarine and tidal processes is rapidly accumulating in specific areas (i.e. inner harbor) at rates more than two orders of magnitude greater than net sedimentation rates in upstream areas of the river estuary; (2) the fine-grained material responsible for the harbor shoaling is river-borne, and not material transported landward from marine sources by estuarine (Meade, 1969) or tidal (Van Straaten and Kuenen, 1958) processes; and (3) the sedimentary record of upstream areas in the river estuary reflects storm processes rather than everyday estuarine or tidal processes.

Hydrology and Geomorphology

The Hudson River forms one of several major estuarine systems on the north-eastern Atlantic coastline of the United States and drains an area of approximately 35,000 km^2. For the purpose of this paper the Hudson Estuary is defined as extending to the upstream limit of the salinity intrusion during low fresh water flow. The salinity distribution along the axis of the Hudson Estuary varies daily with tidal amplitude and seasonally with river flow (Simpson et al., 1974). Location designations in the Hudson are usually given in terms of mile point (mp), which indicates the distance in statute miles along the channel upstream from the southern tip of Manhattan (mp 0). The mean annual fresh water discharge past mp 0 is 550 m^3/sec. During low fresh water flow (∿ 60 m^3/sec) the salt intrusion varies from mp 60-80 (Abood, 1974) and during conditions of seasonal high fresh water flow (∿ 1200 m^3/sec) from mp 10-20 (Fig. 1). Tidal flows average between 8,000 and 11,000 m^3/sec. Tidal-stage fluctuations between 1 and 2 meters occur as far upstream as the dam at Troy (mp 154), and tidal currents reach a maximum velocity of about 100 cm/sec.

On the basis of the sedimentary record, depth profiles and morphology, the estuary can be subdivided into three regions: (1) the inner harbor, which extends from the Narrows (mp -6) to the George Washington Bridge (mp 11), with extensive areas of rapid shoaling; (2) the river estuary, extending from the George Washington Bridge to the upstream limit of the salinity intrusion, generally characterized by channel and subtidal bank depositional environments, as illustrated in Figure 2; and (3) marginal zones, which include broad, shallow reaches, such as Haverstraw Bay, and protected areas along the sides of the estuary, such as Foundry Cove.

SAMPLING AND ANALYTICAL PROCEDURES

The locations and site designation numbers for sediment samples analyzed are indicated in Figure 1. Surface grab samples were collected with a Shipeck grab sampler and cores 50 to 70 cm long, having a diameter of 5.5 cm, were taken with a 150-pound Mills gravity corer. A small diameter (3 cm) Alpine piston corer has also been used to take 2-3 meter cores. Both the piston and gravity core barrels were equipped with removable plastic liners which eliminated onboard extruding procedures and provided maximum protection during transport. Upon returning to the lab, we immediately extruded, split and air dried the cores.

Detailed descriptions, grain-size analyses and mineralogical studies have been conducted on a set of cores and grabs taken across the estuary at mp 18.6 (Figs. 2 and 3). A 6 m long piston core (V32-02) has also been taken in this area at the conclusion of R/V VEMA cruise 32, leg 1. The representative mineral composition was determined at mp 18.6 by examination of individual particles with a scanning electron microscope and an associated Energy Dispersive X-Ray Fluorescence analyzer (EDXRF).

The dry weight percent quartz was determined by monitoring the α-β transition in a Differential Scanning Calorimeter at 1 atm pressure and 573°C. Weight percent carbonate was measured by adding dilute HCl to the sediment and measuring the volume of CO_2 emitted. The total potassium concentration was determined from the ^{40}K activity and the weight percent organic matter was measured by loss-on-ignition. It should be noted that loss-on-ignition values may exceed the actual amount of organic matter due to the partial decomposition of some minerals when heated to 550°C (Gross et al., 1971). Quantitative analyses for total copper, zinc and lead have also been made as a function of depth in several cores by atomic absorption spectrometry.

^{14}C analyses were completed on the organic matter in the surface sediment at several locations in the estuary and on subsurface shell layers from several channel cores. The activity of ^{137}Cs, ^{134}Cs, ^{60}Co and ^{40}K as a function of depth in the sediment has been determined for almost all the cores located in Figure 1. The activities were measured by gamma spectrometry using a Ge(Li) detector and a multi-channel analyzer. Each sample was usually counted for 8-16 hours, and calibration was based on an NBS environmental radioactivity standard (#4350).

SEDIMENT DESCRIPTIONS AND STRUCTURES

Surface sediments in the Hudson Estuary frequently contain oil coatings, coal, tar, brick fragments, fly ash, and metalliferous slags. This anthropogenic detritus is generally confined to the top 10 cm, but may be as deep as 200 cm depending on the rate of deposition. Sand generally comprises < 10% (by weight) of the surface sediment except for a few areas near the mouth of the estuary (mp -6 to -3), in the channel (mp 13 to 24) and along the shore.

The fine-grained sediments between mp 0 and mp 60 generally contain less than 3% carbonate and between 3 and 10% organic matter. This agrees reasonably well with the 5-6% oxidizable organic matter reported by McCrone

(1967) in the channel sediments between mp 75 and mp 41, and is slightly lower than the loss-on-ignition values obtained by Gross et al. (1971) for areas south of mp 0.

The dominant clay minerals in the sediments have been reported to be illite and chlorite with some montmorillonite, kaolinite, and mixed-layer clays also present (McCrone, 1967; Owens et al., 1974). Representative particle composition (based on relative numbers) for suspended matter and sediment in both the sand- and silt-size fractions is given in Table 1. The heavy minerals consisted of amphiboles, pyroxenes, epidote, garnet, sphene, and tourmaline; the lack of magnetite is notable. It is evident from the data presented in Table 1 that there is a greater percentage K-Al-silicates (feldspar, muscovite, illite) relative to quartz in the smaller size ranges, probably reflecting the resistance of quartz to physical and chemical breakdown.

Frequently the bottom sediments have a highly liquefied, olive-brown (oxidized) layer at the sediment/water interface. The layer ranges from 0.1 to 3 cm thick and is composed of fine-grained flocculent matter which may be easily eroded, transported, and redeposited during alterations of the tidal current. It has been proposed that the brownish color of the layer is due to the presence of iron-hydroxides (Van Straaten, 1954), which in time are reduced to iron-monosulfides and then to disulfides (pyrite) in the anoxic sediments below (Biggs, 1967). The sediments below the oxidation layer were either black, gray or olive-gray in color. Black sediments are characteristic of cores taken in the inner harbor. Within a few hours after extrusion, the black sediments faded to olive-gray, presumably reflecting the instability of iron-monosulfides to which the black color has been attributed (Berner, 1973). Cores from the river estuary and from marginal shallow zones were olive-gray in color, frequently over their entire length, but occasionally a black layer a few centimeters thick was found near the surface or at depth.

In the inner harbor the surface sediments consist of fine sandy silts with reported median grain diameters between 10 and 60 microns (Gross, 1974). The top 10 cm of the harbor sediment was highly liquefied and the top meter generally showed extensive turbation, although distinct layers of medium- to fine-grained sand (5 to 15 cm thick) and zones of lamination (alternating layers of fine sandy silts and clayey silts, on a mm to cm scale) were observed in several cores at depth. The turbation was a result not only of mechanical mixing by organisms but also of the release or entrapment of methane (CH_4) gas bubbles which are released into the interstitial waters of sediment by the biochemical degradation of organic matter. In the Hudson estuary, oxidation of organic matter by free oxygen is confined to the olive-brown layer near the sediment/water interface. Below this zone, degradation of organic matter continues by means of anaerobic bacterial processes, such as denitrification, sulfate reduction and methane production (Kaplan, 1974). Methane bubbles form at any depth in the sediment when the concentration of CH_4 approaches its *in situ* saturation value (Fig. 4). In the interstitial waters of the Hudson sediments, CH_4 often attains its *in situ* saturation concentration of 10^4 μmole/ℓ (Hammond, 1975). In most areas of the estuary, gas fissures generally occur below 30 cm in the sediment (Fig. 3) and are not so numerous as to destroy the primary sedimentary structures. The marked increase in occurrence of gas fissures in the sediments of the inner harbor may reflect the very large input of organic sewage particles to this area.

The fine-grained sediments in the channel and subtidal bank environments of the river estuary, upstream of the inner harbor, are characterized by alternating layers of fine sandy silts and clayey silts on the order of a mm to a cm thick (Fig. 5A). Sand and shell layers (1 to 20 cm thick) were observed at the surface and were interlayered with the laminated fine-grained sediments (at depth intervals of 10 to 50 cm) in cores taken along the channel between mp 13 and mp 24.

Occasionally these layers are graded (Fig. 5B), starting with whole shells and grading upward through sand and shell fragments, and finally into laminated fine-grained sediment. Along the adjacent subtidal bank, sand and shell layers were missing, but non-laminated zones (1 to 10 cm thick) separate the zones of lamination (Fig. 6).

The 6 meter core (V32-02) taken in the channel at mp 18.6 records a change in lithology at 300 cm. Whereas the top 300 cm contains many sand and shell layers, the sediments below 300 cm consist almost entirely of laminated fine sand, silt and clay and thus closely resemble 3 m cores taken on the adjacent subtidal bank. This change in lithology may record a shift from a subtidal bank to a channel environment.

Grain-size analyses were completed on surface samples taken from cores (Figs. 2 and 3) in both the channel (18.6-1M and 18.6-3M) and on the subtidal bank (18.6-5W) at mp 18.6. The median diameter for the surface sediment in both channel cores was 0.65 mm and the fines (< 0.063 mm) composed only 9% (18.6-1M) and 3% (18.6-3M) of the total sediment weight. On the subtidal bank (18.6-5W), however, fines composed 98% of the surface sediment, indicating that the subtidal bank is presently more conducive to the deposition of suspended sediment or is less affected by the strong tidal currents which may winnow away much of the fine material in this area of the channel.

Interpretation of the Sedimentary Structures

Laminated fine-grained sediments, characteristic of the Hudson Estuary, have also been reported in other subtidal and intertidal environments (Van Straaten, 1954; McKee, 1957; Reineck and Singh, 1973; Oertel, 1973). The genesis of this lamination may be a result of tidal, wave, seasonal or storm processes. The relative importance of each of these processes will vary in different depositional environments, depending upon the water depth, supply and type of sediments, and the climatic and hydrologic conditions. In addition, the extent to which the lamination is preserved in the sedimentary record will depend on the amount of bio- and gas-turbation, and on the rate of sedimentation.

One mechanism which may produce the observed lamination is the deposition of fine sands and sandy silts during periods of current or wave activity and of clay-rich silts (mud) during periods of slack water (Johnson, 1922; Van Straaten, 1954; Reineck and Singh, 1973). Variations in the suspended sediment concentration during tidal cycles indicates that some deposition occurs during slack water (Panuzio, 1965; Meade, 1969); however, unless there is an adequate sediment supply this tidal lamination will probably be destroyed by bioturbation or reworked during the later tidal cycles. When there is sufficient concentration of suspended matter, perhaps due to severe storms, deposition of fine-grained sediment will be relatively continuous (McCave, 1970; Oertel, 1973). Sorting during the deposition of suspension clouds (Reineck and Singh, 1973) and/or during the deposition of the bedload (Kuenen, 1966), perhaps in response to tidal cycles or other variations in current or wave activity, may give rise to the observed sedimentary lamination. Consequently, it is proposed that the laminated fine-grained sediments are deposited very rapidly when the Hudson transports large quantities of sedimentary material, such as during and after severe storms or other short term events of large scale resuspension.

The rapid deposition of the laminated unit followed by periods of slower deposition, non-sedimentation or even erosion, is consistent with the observed laminated zones alternating with turbate zones (inner harbor, subtidal bank) and sand-shell layers (channel, inner harbor). The graded character of some of the sand-shell layers (Fig. 5B) suggest that they have been deposited as a unit, perhaps as a result of high-energy events such as severe storms. Several animal escape traces, originating from the sand-shell layer and migrating through the laminated silts, have been observed (Fig. 3), also suggesting that the overlying non-turbated and finely laminated sediment was deposited very rapidly (Reineck and Singh, 1973). The turbate zones may represent periods of slower deposition under normal flow conditions when burrowing organisms obliterate the lamination to varying degrees or when organic waste discharge forms a relatively greater proportion of the material deposited resulting in increased turbation by biochemically formed gases.

It is well-documented that high-energy events, such as severe storms, can rework, erode, transport and deposit enormous quantities of sediment and may thus have profound (and preferentially preserved) effects on the sedimentary record (Kelling and Mullin, 1975). Schubel (1974) found that during Hurricane Agnes, 22-28 June 1972, the Susquehanna River discharged more sediment than it had previously discharged during the past 10 years. The effects of Agnes on the sedimentary record of Chesapeake Bay were depositional rather than erosional, resulting in 4 to 20 cm of laminated silts and clays (Zabawa and Schubel, 1974). This laminated unit was separated from older bioturbated sediments by a sharp boundary (Zabawa and Schubel, 1974).

GEOCHEMICAL ANALYSIS OF THE SEDIMENTS

^{14}C analyses were conducted on surface shells, subsurface shell layers, and organic matter in surface sediments. The location and type of material dated and the radiocarbon ages are listed in Table 2. The radiocarbon measurements for *Mulinia lateralis* are layer averages based on 50 to 150 shells.

The activity of ^{137}Cs, ^{134}Cs, ^{60}Co and ^{40}K has been determined in the suspended matter and as a function of depth in the sediment for almost all the cores located in Figure 1. ^{40}K is a naturally-occurring radionuclide with a half-life of 1.27×10^{9} years. Approximately 0.012% of the potassium in K-bearing minerals such as muscovite, biotite, illite and K-feldspar is ^{40}K. Consequently, the activity of ^{40}K in the sample simply reflects the sediment composition.

^{137}Cs, ^{134}Cs and ^{60}Co are anthropogenic radionuclides with half-lives of approximately 30, 2 and 5 years respectively. The ecological pathways of radiocesium, ^{60}Co and other gamma-emitting nuclides in the Hudson Estuary have been studied by Lentsch et al. (1970), and Lentsch (1974). ^{137}Cs has been introduced into the Hudson Estuary via global fallout from nuclear weapons testing and through low level releases from the nuclear power plant at Indian Point (mp 43). Reactor releases are the only source for the observed ^{134}Cs and ^{60}Co activity in the Hudson sediments and they can be very useful in defining the time scale and pattern of sediment transport along the axis of flow of the Hudson (Simpson et al., 1976).

Figure 7A shows the annual fallout record for ^{137}Cs to the New York City area during the past 23 years. The major influx of fallout ^{137}Cs to the Hudson occurred in 1962-1964. Figure 7B shows the annual release of gross beta and gamma activity in the liquid effluent from the Indian Point nuclear power plant. The major influx of reactor-produced ^{137}Cs, ^{134}Cs and ^{60}Co to the Hudson occurred in 1971. These radionuclides are adsorbed to the surface of the suspended matter and consequently provide excellent tracers for river-borne sediment transport and good indicators of sediment that has been recently deposited. In areas of rapid deposition, the 1971 reactor peak and 1962-1964 fallout peak may be preserved in the sediment, thus providing definite time-stratigraphic reference levels.

Gamma-spectrometric data for suspended matter and several cores in each of the three estuarine regions (i.e. inner harbor, river estuary and marginal zones) are given in Table 3. The highest anthropogenic radionuclide concentrations were observed in the suspended matter and in the liquefied olive-brown (oxidized) layer at the sediment-water interface. The suspended matter was collected at mp 18 and the oxidized layer was removed from grab samples at mp 18.6 (SLOSH II) and at mp 42.8 (SLOSH III), approximately 100 m downstream of the Indian Point cooling water discharge system.

Quantitative analyses for total copper, lead and zinc have been determined as a function of depth in a 6 m core (V32-02) from the channel at mp 18.6; core

53.8EC from Foundry Cove; and cores -1.5E, 0.1W and 1.9W taken in the inner harbor. The metal concentrations are listed in Table 4. Because organic-rich, fine-grained sediments have a greater affinity for trace metals, as well as for radionuclides, variations in sediment composition should be considered when examining the vertical distribution of trace metals or radionuclides in sediments. Therefore, the percent organic matter, the total potassium concentration and the weight percent quartz are also presented in Table 4. The trace metal analyses were conducted on cores for which the radiocesium and ^{60}Co activities have also been determined (Table 3).

Results and Interpretation

Carbon-14 - The shells at the sediment surface contain excess ^{14}C produced by nuclear weapons testing and thus give future ages when presented in radiocarbon years (Table 2). These bomb-contaminated ages indicate that the surface shell material formed during recent times, i.e., < 20 years ago, and that this recent material was not lost during coring operations. Although the surface sediment at mp 18.3 contained carbonate shells with measurable bomb ^{14}C, the humic acid fraction of the organic matter yielded an age of ∿ 2000 years, while the residual organic matter gave an age of ∿ 4000 years. These old apparent ages for the organic matter in the surface sediments on the subtidal bank at mp 18.3, as well as at mp 42.5, may result from the recycling of old carbon from soils of the drainage basin or from sediments in other areas of the Hudson River. The interpretation of these old organic matter ^{14}C ages, however, is complicated in that it may reflect recent contamination with petroleum hydrocarbons. The bomb-contaminated age of the organic matter at mp 0.1, is in marked contrast with the surface organic matter ages in upstream areas of the river estuary and reflects the additional input of recent organic sewage to the inner harbor.

The sand-shell surface layer of the natural channel between mp 18 and mp 22 also contains bomb ^{14}C in shell materials, but the shell layers immediately below are free of bomb ^{14}C and have old ^{14}C ages. One possible interpretation of these data is that the subsurface shell layers are composed of older shells that have been exposed by channel erosion and migration. The coarse nature of the surface sediment indicates that the channel at mp 18 has undergone tidal scour, and the change in lithology below 300 cm in core V32-02 may be a result of channel migration.

The subsurface shell layers consist almost entirely of a small clam, *Mulinia lateralis*. Living *Mulinia* appear to be restricted to areas south of Yonkers (mp 17) and are found most often in the muddy sediments of more saline waters near the Battery (mp 0) (Shaw, 1975). Their present rarity in the surface sediments of upstream areas where they are abundant in subsurface layers may reflect a decrease in estuarine salinity. Weiss (1974) noted that such a decrease has occurred during the past 2300 ± 700 years, on the basis of foraminiferal evidence. A net sedimentation rate of a few mm/yr has been obtained from one core (P21.7M) containing *Mulinia* layers for which the radiocarbon ages plot along a straight line with depth in the sediment. Such a net sedimentation rate is consistent with the total thickness of the estuarine organic silt layer (10 to 60 meters; Worzel and Drake, 1959) relative to the beginning of estuarine conditions (11,500 yrs Bp; Weiss, 1974) and also with net sedimentation rates observed in other estuaries (Oviatt and Nixon, 1975).

Anthropogenic Radionuclides - The distribution of ^{137}Cs, ^{134}Cs and ^{60}Co in the Hudson estuary sediment cores generally falls within one of three distinct types reflecting the different sedimentary processes which occur in the three estuarine regions (Simpson et al., 1976). In the channel (18.6-3M, C44, 57.4M) and along the subtidal banks (18.6-5W, C43E, 56.4W) of the river estuary, the activity of radiocesium and ^{60}Co decreases rapidly with depth in the sediment. The activity was usually undetectable below 10 cm and was generally confined to the surface turbate zone (subtidal bank) or surface coarse layer (channel) and not observed in the laminated sediments below. In broad shallow areas (25.3W) and in protected areas along the sides of the estuary (53.8EC, 40WC), radiocesium was frequently detected to depths on the order of 15-35 cm, indicating that these areas are sites of more rapid deposition.

In the inner harbor, the vertical distribution of radiocesium and ^{60}Co in the sediment is much more extensive than in either the river estuary or marginal zones. ^{137}Cs has been detected to a depth > 250 cm in one harbor core. Although this sediment is extensively turbated, mechanical mixing of the surface sediment cannot account for the observed activity at lower depths because (1) the activities of ^{137}Cs, ^{134}Cs and ^{60}Co are frequently just as high, if not higher, at 50-70 cm as in the near-surface sediment, and (2) relatively high concentrations of radiocesium were occasionally observed in the fine-grained sediments below distinct sand layers (Fig. 8) and laminated zones. Correlation of the radionuclide distribution in the harbor sediments with that of the suspended matter and with the reactor release data (Fig. 7B) indicate that the fine-grained sediment is river-borne and that it is accumulating at rates of 5 to 20 cm/yr. This rate is more than two orders of magnitude greater than net deposition rates in the channel and more than one order of magnitude greater than on the subtidal banks of upstream areas in the river estuary.

Cores taken in recently dredged areas, such as core 8.7WM, taken in an area which is dredged annually (mp 5-10), contained relatively low concentrations of radiocesium in the top 5 cm and undetectable levels in the sediments below. Cores taken on the banks of the dredged channel, such as core 6.0W, contained radiocesium to depths > 60 cm. In addition, in core 0.1W the ^{137}Cs activity sharply decreases from 685 pCi/kg in the turbated black sediment (45-53 cm) to background levels in the laminated gray sediment below 53 cm (Fig. 9). This sharp decrease in radiocesium activity correlates with a sharp decrease in trace metal levels (Table 4) and may represent a dredged boundary between the older, laminated gray sediment, exposed by past dredging activities, and the recent, turbated black sediment deposited after the dredging episode. It thus appears that the distribution of radiocesium in the sediment may be helpful in distinguishing areas that have recently been dredged from areas undisturbed by dredging operations. This distinction is of primary importance when studying the effects of sedimentation and pollution in urban estuaries. In addition, the dredging of this ^{137}Cs-, ^{134}Cs- and ^{60}Co-laden inner harbor sediment and the consequent dumping of this dredged material in the apex of the New York Bight may provide radioactive tracers for sediment transport in the Bight and across the shelf.

In harbor core -1.6E (Fig. 8), a layer of medium- to fine-grained sand at 50-65 cm contained no measurable radiocesium, contrasting with the high concentrations of radiocesium observed in the turbated, fine-grained sediment both above and below this sandy layer. Although a reduction in the activity of radiocesium is expected with increasing grain size, the fact that the coarse channel sediment at mp 18.6 contained detectable concentrations of ^{137}Cs and ^{134}Cs (Table 3) indicates that the increase in grain size alone cannot account for the lack of radiocesium in this layer. Piston cores 2 to 3 meters long taken in the same area also penetrate several similar medium- to fine-grained sand layers. One possible interpretation is that these sand layers consist of material transported up the estuary from marine sources or derived from construction sites along the sides of the estuary.

<u>Trace Metals</u> - Metal concentrations for the 6 m core (V32-02) taken in the channel at mp 18.6 show very little variation below the sandy surface layer. Anthropogenic radionuclides are only present in the surface layer and subsurface shell layers have radiocarbon ages > 1000 years. Consequently, it appears that the metals in the laminated fine-grained sediments below 300 cm in core V32-02 are indicative of pre-industrial levels. Owens <u>et</u> <u>al</u>. (1974) have reported similar Zn

concentrations in the organic silt between 20 and 50 meters in a drill core taken near the Newburgh Bridge (mp 61).

In the inner harbor sediments, Cu, Zn and Pb concentrations are 2 to 6 times higher than pre-industrial levels and were observed to extended depths. This finding is consistent with the vertical distribution of radiocesium and indicates that much of the trace metal pollutants introduced into the Hudson are accumulating with the fine-grained sediment in the inner harbor. In addition, the sharp increase in Cu, Zn and Pb above 53 cm in core 0.1W provides a good documentation of the increase in trace metal levels associated with urban pollution.

In Foundry Cove (53.8EC), concentrations of Cu, Zn and Pb decrease with depth in the sediment to 40 cm below which they are similar to the pre-industrial levels in V32-02. The higher metal concentrations in the surface sediment relative to the sediments below 40 cm reflects the recent increase in human and industrial pollution in this area. Trace metal pollution appears to have occurred over a somewhat longer time period (> two decades) than for fallout ^{137}Cs which is confined to the upper 20 cm of sediment (Table 3).

CONCLUSIONS

Geochemical and sedimentological evidence indicates that:

(1) There is a large variation in the patterns and rates of sediment deposition in the Hudson Estuary, and this dynamic character has not been previously identified on the basis of grain-size and mineralogical analyses;

(2) Much of the suspended matter and sedimentary material that is resuspended, transported and redeposited by everyday estuarine and tidal processes is presently rapidly accumulating in the inner harbor and in protected areas along the sides of the estuary;

(3) The vertical distribution of bomb- and reactor-produced radionuclides indicate sedimentation rates of 5 to 20 cm/yr in the inner harbor which is

more than two orders of magnitude greater than net sedimentation rates in the channel and more than one order of magnitude greater than on the subtidal banks of the river estuary;

(4) The fine-grained material responsible for shoaling in the inner harbor is primarily river-borne (labelled with reactor nuclides released ∿ 60 km upstream of the harbor) and not material transported landward from marine sources by estuarine or tidal processes;

(5) Distinct coarser-grained layers (containing no man-made radionuclides) in New York Harbor may come from marine or local marginal sources;

(6) The distribution of anthropogenic radionuclides in the sediment is helpful in distinguishing areas that have been dredged from areas undisturbed by dreding operations, and the consequent dumping of dredged sediment (tagged with ^{137}Cs, ^{134}Cs and ^{60}Co) in the New York Bight may provide radioactive tracers for sediment transport in the Bight and across the shelf;

(7) The sedimentary record in the channel and on the subtidal bank of the river estuary, upstream of the inner harbor, is characterized by storm deposits or other short term events of large scale resuspension rather than reflecting everyday estuarine and tidal processes.

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REFERENCES

ABOOD, K.A., 1974, Circulation in the Hudson Estuary: In: Roels, O.A. (Ed.), Hudson River Colloquium, Annals of the New York Academy of Sciences, v. 250, p. 39-111.

AGER, D.V., 1974, Storm deposits in the Jurassic of the Moroccan High Atlas: Palaeogeogr. Palaeoclimatol. Palaeoecol., v. 15, p. 83-93.

BERNER, R.A., 1973, Pyrite formation in the oceans: In: Ingerson, E. (Ed.), Proceedings of Symposium on Hydrogeochemistry and Biogeochemistry, v. 1, Clark Company, Tokyo, p. 402-417.

BIGGS, R.B., 1967, The sediments of Chesapeake Bay: In: Lauff, G.H. (Ed.), Estuaries, Amer. Assoc. Adv. Sci. Publ. No. 83, Washington, D.C., p. 239-260.

BOOTH, R.S., 1975, A compendium of radionuclides found in liquid effluents of nuclear power stations, ORNL-TM-3801, Oak Ridge National Laboratory, 22 pp.

COCH, N.K., 1976, Temporal and areal variations in some Hudson River Estuary sediments (Abs.), Geol. Soc. Amer. N.E. Sect. Mtg., v. 8, No. 2, p. 153.

GROSS, M.G., BLACK, J.A., KALIN, R.J. SCHRAMEL, J.R. and SMITH, R.N., 1971, Survey of Marine waste deposits, New York Metropolitan Region. Technical Report No. 8, Marine Sciences Research Center, SUNY, Stony Brook.

__________, 1974, Sediment and waste deposition in New York Harbor: In: Roels, O.A. (Ed.), Hudson River Colloquium, Annals of the New York Academy of Sciences, v. 250, p. 112-128.

HAMMOND, D.E., 1975, Dissolved gases and kinetic processes in the Hudson River Estuary, Ph.D. Thesis, Columbia University, 161 pp.

IPPEN, A.T., 1966, Sedimentation in estuaries: In: Ippen, A.T. (Ed.), Estuary and Coastline Hydrodynamics, McGraw Hill, New York, p. 648-672.

JOHNSTON, W.A., 1922, The character of the stratification of the sediments in the recent delta of Fraser River, British Columbia, <u>Canadian Jour. Geology</u>, v. 30, p. 115-129.

KAPLAN, I.R. (Ed.), 1974, Natural Gases in Marine Sediments, Marine Science, v. 3, Plenum Press, New York, 324 pp.

KELLING, G. and MULLIN, P.R., 1975, Graded limestones and limestone-quartzite couplets: Possible storm-deposits from the Moroccan carboniferous, <u>Sedimentary Geology</u>, v. 13, p. 161-190.

KUENEN, P.H., 1966, Experimental turbidite lamination in a circular flume, <u>Jour. Geology</u>, v. 74, p. 523-545.

KUMAR, N., and SANDERS, J.E., 1976, Characteristics of shoreface storm deposits: Modern and ancient examples, <u>Jour. Sed. Petrology</u>, v. 46, p. 145-162.

LENTSCH, J.W., WRENN, M.E., KNEIP, T.J. and EISENBUD, M., 1970, Man-made radionuclides in the Hudson River Estuary, Proceedings of the Fifth Annual Health Physics Society Mid-Year Topical Symposium, Idaho Falls, Idaho.

__________, 1974, The fate of gamma-emitting radionuclides released into the Hudson River Estuary and an evaluation of their environmental significance, Ph.D. Thesis, New York University, 608 pp.

McCAVE, I.N., 1970, Deposition of fine-grained suspended sediment from tidal currents, <u>Jour. Geophys. Res.</u>, v. 75, p. 4151-4159.

McCRONE, A.W., 1967, The Hudson River Estuary: Sedimentary and geochemical properties between Kingston and Haverstraw, New York, <u>Jour. Sed. Petrology</u>, v. 37, p. 475-486.

McKEE, E.D., 1957, Primary structures in some recend sediments, <u>Amer. Assoc. Petroleum Geologists Bull.</u>, v. 41, p. 1704-1747.

MEADE, R.H., 1969, Landward transport of bottom sediments in estuaries of the Atlantic coastal plain, <u>Jour. Sed. Petrology</u>, v. 39, p. 222-234.

NICHOLS, M.M., 1972, Sediments of the James River Estuary, Virginia: In: Nelson, B.W. (Ed.), Environmental Framework of Coastal Plain Estuaries, Geol. Soc. Amer. Mem., 133, p. 169-212.

OERTEL, G.F., 1973, Examination of textures and structures of mud in layered sediments at the entrance of a Georgial tidal inlet, Jour. Sed. Petrology, v. 43, p. 33-41.

OVIATT, C.A. and NIXON, S.W., 1975, Sediment resuspension and deposition in Narragansett Bay, Estuarine and Coastal Marine Science, v. 3, p. 201-217.

OWENS, J.P., STEFANSSON, K. and SIRKIN, L.A., 1974, Chemical mineralogic and palynologic character of the Upper Wisconsin-Lower Holocene fill in parts of the Hudson, Delaware and Chesapeake Estuaries, Jour. Sed. Petrology, v. 44, p. 390-408.

PANUZIO, F.L., 1965, Lower Hudson River siltation: In: Proceedings of the Federal Inter-Agency Sedimentation Conference, 1963, Miscellaneous Publication No. 970, Agricultural Research Service, p. 512-550.

POSTMA, H., 1967, Sediment transport and sedimentation in the estuarine environment: In: Lauff, G.H. (Ed.), Estuaries, Amer. Assoc. Adv. Sci. Publ. No. 83, Washington, D.C., p. 158-179.

PRITCHARD, D.W., 1955, Estuarine circulation patterns, Proc. Amer. Soc. Civil Engrs., v. 81, p. 717/1-717/11.

REINECK, H.E. and SINGH, I.B., 1972, Genesis of laminated sand and graded rhythmites in storm-sand layers of shelf mud, Sedimentology, v. 18, p. 123-128.

__________, __________, 1973, Depositional Sedimentary Environments, Springer-Verlag, New York, 439 pp.

SCHUBEL, J.R., 1968, Turbidity maximum of the northern Chesapeake Bay, Science, v. 161, p. 1013-1015.

__________, 1974, Effects of tropical storm Agnes on the suspended solids of the northern Chesapeake Bay: In: Gibbs, R.J. (Ed.), Suspended Solids in Water, Marine Science, v. 4, Plenum Press, New York, p. 113-132.

SHAW, F.C., 1975, Distribution of shelled macroinvertebrate benthos in the Hudson Estuary (Abs.): In: Programs and Abstracts of the 7th Annual Long Island Sound Conference, January 11, 1975.

SIMPSON, H.J., BOPP, R. and THURBER, D, 1974, Salt movement patterns in the lower Hudson, Third Symposium on Hudson River Ecology, 1973, Ch. 9, 34 pp., Hudson River Environmental Society, New York.

SIMPSON, H.J., OLSEN, C.R., TRIER, R.M. and WILLIAMS, S.C., 1976, Man-made radionuclides and sedimentation in the Hudson River Estuary, Science, v. 194, p. 179-183.

SWIFT, D.J.P., DUANE, D.B. and PILKEY, O.H. (Eds.), 1972, Shelf Sediment Transport: Process and Pattern, Dowden, Hutchinson and Ross, Stroudsburgh, Pennsylvania, 656 pp.

VAN STRAATEN, L.M.J.U., 1954, Composition and structure of recent marine sediments in the Netherlands, Leidse Geol. Mededel., v. 19, p. 1-110.

__________, and KUENEN, P.H., 1958, Tidal action as a cause of clay accumulation, Jour. Sed. Petrology, v. 28, p. 406-413.

WEISS, D., 1974, Late Pleistocene stratigraphy and paleoecology of the lower Hudson River Estuary, Geol. Soc. Amer. Bull., v. 85, p. 1561-1570.

WORZEL, J.L. and DRAKE, C.L., 1959, Structure section across the Hudson River at Nyack, New York from seismic observation, Annals of the New York Academy of Science, v. 80, p. 1092-1105.

ZABAWA, C.F. and SCHUBEL, J.R., 1974, Geologic effects of tropical storm Agnes on Upper Chesapeake Bay, Maritime Sediments, v. 10, p. 79-84.

Table 1

Particle Composition of Sediments and Suspended Matter at mp 18.6 in the Hudson Estuary

Elemental[1] Composition	Sand (0.65 mm diameter)	Silt	Suspended Matter	Mineralogy[2]
SiO_2 (Al,Fe)	67%	47%	27%	Quartz, diatoms
Na(Ca)Al-Si	10%	15%	3%	Plagioclase feldspar
K-Al-Si	12%	16%	(22%)	K-feldspar
K-Al-Si-Fe	2%	3%		Muscovite
K-Al-Si-P(Fe)	-	8%	(30%)	Illite-organic aggregates
Mg-K-Fe-Al-Si	1%	2%		Biotite
(Na)(Mg)(K)(Ca)(Fe)-Al-Si	7%	7%	7%	Heavy minerals[3]
Others	1%	2%	11%	Clay-organic aggregates, oxides, carbonates, fly ash, coal

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(1) Elements in parentheses are occasionally present. Statistical variation was based on the examination of ∿ 300 particles in both the sand and silt size range and ∿ 1000 particles of suspended matter.

(2) Mineralogical classification of the aluminosilicates was based on the cations present, Al/Si ratios and cleavage. Al/Si ratios and cleavage data was not obtained for the suspended matter. Consequently muscovite and biotite could not be specifically identified and the percentage of K-feldspar in the suspended matter has a greater uncertainty than in the sediment.

(3) See text.

Table 2

Radiocarbon Data in the Hudson Estuary

Sample	Depth	Material	Apparent ^{14}C Age
Core 18.6-1 (Fig. 3)	0-5	*Crassostrea*, *Mya*, and *Macoma*	-3010 ± 80
	21-23	*Mulinia*	1290 ± 100
	27-31	*Mulinia*	1290 ± 130
	48-50	*Mulinia*	1320 ± 130
Core V32-02 (Fig. 4)	0-10	*Crassostrea*, *Mya*, and *Macoma*	-450 ± 150
	109-111	*Mulinia*	1120 ± 300
	127-130	*Crassostrea*	1500 ± 130
	199-201	*Mulinia*	2210 ± 370
	266-270	*Mulinia*	2080 ± 270
Core P21.7M	35-40	*Mulinia*	2240 ± 230
	130-140	*Mulinia*	2740 ± 190
	200-210	*Mulinia*	2980 ± 130
Grab G18.3	0-5	*Mya* and *Macoma*	-2450 ± 100
		Humic Fraction	1890 ± 520
		Residual Organics	4600 ± 500
Grab G42.5	0-5	Humic Fraction	1470 ± 340
		Residual Organics	4140 ± 160
Grab G0.1W	0-5	Residual Organics	-2590 ± 270

Table 3

Gamma-emitting Radionuclides in Hudson Estuary Sediments

			ACTIVITIES[a] (pCi/kg)			
Depositional Environment	Core	Depth (cm)	Cs^{137}	Cs^{134}	Co^{60}	K^{40}
INNER HARBOR	-1.6E	0-5	450±26[b]	12±11	51±23	15,700±690
		5-12	620±26	56±13	50±18	16,600±630
		12-20	1210±42	115±19	110±23	16,600±690
		20-25	470±25	37±12	43±19	16,600±650
		25-30	540±24	32±11	-13±15*	15,700±590
		30-35	390±18	20±8	44±15	15,800±590
		35-40	615±36	38±16	69±23	18,000±800
		40-45	535±95	- 2±15*	54±35	17,900±910
		45-50	610±31	3±12	47±21	17,400±720
		50-55	29±11	- 8±7*	- 2±13*	15,500±550
		55-60	-19±9*	- 9±7*	11±14	14,600±550
		60-65	27±14	- 6±15*	10±20	11,500±380
		65-73	455±24	11±10	24±18	16,200±630
	-1.5E	0-5	440±21	32±10	30±16	15,600±610
		5-12	430±26	18±12	30±22	15,100±680
		12-20	620±26	19±11	68±17	17,500±650
		20-25	440±23	13±10	56±15	15,700±610
		25-30	390±28	2±13	28±25	18,400±830
		30-35	570±24	27±11	31±14	17,900±650
		35-40	605±24	43±11	37±17	15,700±620
		40-45	685±28	52±12	47±16	16,000±620
		45-50	635±30	36±13	43±19	16,000±680
		50-55	1140±50	135±25	105±25	16,900±810
		55-60	865±33	110±19	73±18	15,700±630
	0.1W	0-5	645±36	64±24	40±29	17,600±850
		5-10	890±28	63±12	60±16	19,700±660
		10-18	1040±50	87±22	110±33	18,700±900
		18-25	1480±40	125±18	170±21	19,400±700
		25-30	1260±40	130±19	175±22	17,100±630

Table 3 cont'd

		30-35	2310±70	300±38	205±32	18,400±800
		35-40	1210±60	130±25	90±31	19,000±920
		40-45	1140±50	110±21	84±26	17,800±780
		45-53	685±27	27±10	71±18	16,600±610
		53-60	105±14	7±8	38±15	17,700±620
		60-65	3±11	13±8	12±13	17,300±600
		65-70	2±12	6±8	15±14	18,400±630
	1.9W	0-5	1180±35	100±16	105±23	17,900±700
		5-10	375±19	12±9	64±19	15,200±600
		10-15	550±24	1±9	20±17	15,700±590
		15-20	57±13	-13±9*	8±15	17,500±630
		20-25	16±12	4±8	- 6±16*	17,600±630
		25-30	16±13	0±9	14±18	16,500±590
		50-55	- 1±11*	6±9	8±15	16,500±590
RIVER ESTUARY						
Channel	18.6-3M	0-5	150±13	36±12	50±17	11,900±490
		5-10	120±21	-12±15*(c)	32±30	16,700±800
		10-15	8±20	N.D.	10±29	16,000±830
	C44	0-7	610±21	79±19	120±21	16,100±530
		7-14	- 1±11*	- 9±7*	-14±15*	17,400±590
	57.5M	0-5	0±7*	10±22	8+11	12,900±450
Subtidal Bank	18.6-5W	0-5	140±20	10±17	45±31	20,500±830
		5-10	24±13	13±14	35±25	19,400±750
		10-15	-27±20*	14±23	-41±31*	20,100±970
	C43E	0-5	285±19	71±22	87±17	21,900±750
		5-10	30±21	-13±13*	- 6±26	24,100±940
	56.4W	0-5	930±36	10±22	58±20	18,500±720
		5-10	15±15	10±18	16±21	20,300±720
MARGINAL ZONES						
Wide, shallow	25.3W	0-6	1170±36	2±10	100±20	18,600±670
		6-12	780±34	- 8±15*	-11±22*	18,900±730
		12-16	265±17	-12±8*	- 3±15*	17,000±600
		16-21	48±18	3±11	3±21	18,400±730

Table 3 cont'd

Cove	40WC	0-5	2070±42	110±13	215±12	20,100±590
		5-10	620±28	8±13	- 7±11*	19,600±680
		10-15	110±18	1±13	10±13	19,800±700
		15-20	85±12	- 8±9*	18±14	19,700±600
		20-25	58±15	23±12	-18±11*	19,000±650
		25-30	57±15	17±15	29±14	20,200±740
		30-35	57±14	10±11	- 1±10*	17,500±590
		35-40	20±9	N.D.	0±11	16,500±500
	53.8EC	0-5	2475±63	98±26	69±19	19,000±770
		5-10	1825±68	17±32	19±20	21,500±960
		10-15	210±17	- 8±10*	15±11	20,000±700
		15-20	26±17	-15±11*	30±15	18,800±780
		20-25	35±23	0±12*	7±18	20,700±870
		50-55	9±14	7±9	13±15	18,500±670
<u>SUSPENDED MATTER</u>						
HE No. 4	(mp 18)		1260±35	210±25	145±18.	17,700±630
<u>SLOSH</u>						
SLOSH II	(mp 18.6)	oxidized layer	660±23	60±11	89±13	17,400±600
SLOSH III	(mp 42.8)	oxidized layer	2700±72	345±42	400±27	19,200±780

(a) Sample specific activities are expressed in terms of dry weight and have been corrected for decay between time of coring and time of assay. Samples marked by * were uncorrected.

(b) Specific activities >10,000 were rounded to the nearest 100 pCi. Specific activities <10,000 but >1000 were rounded to the nearest 10 pCi. Specific activities <1000 but >100 were rounded to the nearest 5 pCi and specific activities <100 were rounded to the nearest pCi. Statistical errors are reported at 1 sigma. Errors for specific activities >10,000 were rounded to the nearest 10 pCi. Errors for specific activities <10,000 but >1000 were rounded to the nearest 5 pCi and errors for specific activities <1000 were rounded to the nearest pCi.

(c) N.D. = Not Determined

Table 4

Trace Metals in the Hudson Estuary Sediments
(per gram dry weight)

Sample	Depth (cm)	Zn µg	Cu µg	Pb µg	K mg	Quartz mg	LIG(a) mg	Radio-cesium(b)	Sediment(c)
-1.5E (Harbor)	0-5	337	248	202	18	N.D.(d)	75	Yes	Gas-turbated
	35-40	399	294	247	19	N.D.	94	Yes	Gas-turbated
	45-50	472	338	286	19	N.D.	100	Yes	Gas-turbated
	55-60	557	416	345	19	N.D.	110	Yes	Gas-turbated
0.1W (Harbor)	0-5	258	179	140	21	N.D.	85	Yes	Gas-turbated
	18-25	215	198	197	22	N.D.	98	Yes	Gas-turbated
	45-53	224	285	177	20	N.D.	88	Yes	Gas-turbated
	60-65	53	12	<28	20	N.D.	49	No	Laminated
1.9W (Harbor)	0-5	343	227	828	21	N.D.	92	Yes	Gas-turbated
	20-25	237	144	164	21	N.D.	65	No?	Laminated
	50-55	247	178	245	19	N.D.	79	No	Gas-turbated
V32-02 (Channel mp 18.6)	0-10	48	35	55	17	570	26	Yes	Sandy channel lag
	22-29	81	16	39	24	260	71	No	Laminated
	83-85	83	20	24	20	240	66	No	Laminated
	150-153	82	20	28	24	270	60	No	Laminated
	240-241	68	14	28	22	350	47	No	Laminated
	325-328	77	20	24	24	270	47	No	Laminated
	430-438	77	21	27	23	290	49	No	Laminated
	545-548	81	15	21	22	370	46	No	Laminated
53.8EC (Cove)	0-5	316	88	147	22	250	96	Yes	Turbated
	10-15	305	80	121	23	250	96	Yes	Turbated
	20-25	124	31	49	24	290	71	No?	Turbated
	30-35	90	18	25	N.D.	310	68	N.D.	Turbated
	40-45	84	18	20	N.D.	290	69	N.D.	Turbated
	50-55	82	17	23	22	320	75	No	Turbated
Error (e)		±6	±4	±12	±12%	±10%	±9%		

(a) Organic matter as determined by loss-on-ignition at 550°C. See text for interpretation.

(b) See Table 3.

(c) Fine-grained estuarine silts, unless otherwise noted.

(d) N.D. = Not determined.

(e) Potassium error is one sigma based on counting statistics. Error reported for trace metals and loss-on-ignition is based on several duplicate analyses. Error for quartz is based on duplicate analyses and comparison with values determined by x-ray diffraction.

FIGURE CAPTIONS

Figure 1 - A location map of the cores and surface grab samples collected in the Hudson Estuary. The sample locations are specified by the number of statute miles along the channel upstream from the southern tip of Manhattan (mp 0) and by a letter indicating the depositional environment; M = channel, W = west subtidal bank, E = east subtidal bank, and C = cove. Grab samples have the letter G preceeding the mile number. SLOSH (Standard Lamont Observatory Sediment from the Hudson) is a composite of several surface grab samples taken at the same anchored location.

Figure 2 - A bottom profile across the Hudson at mp 18.6, illustrating the channel and subtidal bank environments. The locations of gravity cores, 18.6-1M, 18.6-2M, 18.6-3M, 18.6-4M, 18.6-5W and 18.6-6W; piston cores, P18.6-1 and P18.6-5; the 6 meter VEMA core, V32-02, and surface grab samples, G18.6-5 and SLOSH II are also located with respect to the bottom profile. Cores 18.6-1M, 18.6-3M and 18.6-5W are presented in Figure 3. Vertical exag. = 31.

Figure 3 - Gravity cores 18.6-1M, 18.6-3M and 18.6-5W taken at mp 18.6 and located in Figure 2. The photos were taken ∿ 7 days after the cores were extruded and split, in order for the lamination to become visible. The parting, due to shrinkage upon drying, generally occurs along fine sand layers. Core 18.6-1M contains shell layers that have been ^{14}C dated (Table 2). Note the diagonal animal escape trace through the laminated fine-grained sediments at 45-50 cm. Core 18.6-3M clearly illustrates the coarse channel lag observed in this area of the Hudson. Core 18.6-5W, taken on the subtidal bank, lacks the coarse sand-shell layers observed in the channel cores. Note the formation of methane (CH_4) gas fissures below 45 cm.

Figure 4 - (A) Methane (CH_4) gas bubbles forming on the outside of core P18.6-5 immediately after extrusion (scale in cm).

(B) Gas fissures observed when core P18.6-5 was split.

Figure 5 - (A) An enlarged photo taken between 530 and 540 cm in core V32-02 illustrating the mm to cm scale lamination (scale in cm).

(B) An enlarged photo taken between 50 and 60 cm in core P18.6-1 illustrating the graded character of some of the sand and shell layers (scale in cm).

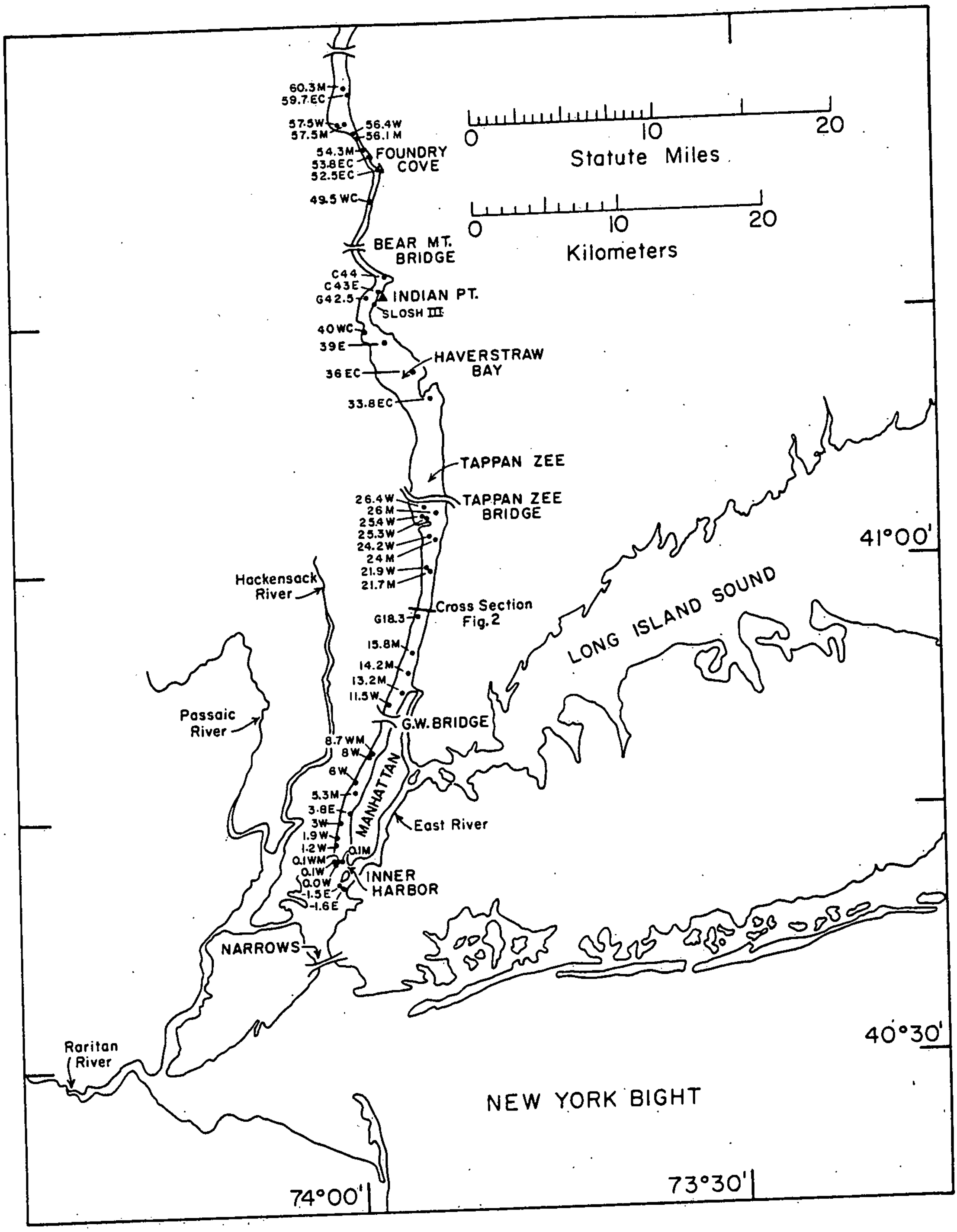
Figure 6 - An enlarged photo taken at 45-65 cm in core P18.6-5 illustrating a turbate zone (54 to 61 cm) between two laminated zones. The photo was taken ∿ 7 days after the core was extruded in order for the lamination to become visible. The parting generally occurs along fine sand layers.

Figure 7 - (A) Annual deposition of ^{137}Cs from fallout at New York City. The ^{137}Cs data has been determined from ^{90}Sr precipitation data collected in lower Manhattan by the Health and Safety Laboratory of ERDA. The $^{137}Cs/^{90}Sr$ ratio is assumed to be 1.5. Data for 1976 is for only half of the year (January to June).

(B) Annual gross beta-gamma releases (excluding tritium) in the liquid effluent from the nuclear power plant at Indian Point (Booth, 1975). Note peak in 1971.

Figure 8 - Schematic drawing illustrating the vertical distribution of ^{137}Cs in core -1.6E. Note that the medium- to fine-grained sand layer at 50-65 cm contains undetectable levels of ^{137}Cs, whereas the turbated sediment both above and below this sandy layer has relatively high ^{137}Cs activities.

Figure 9 - Schematic drawing illustrating the vertical distribution of ^{137}Cs in core 0.1W. Note the maximum at 30-35 cm and the sharp drop from 685 pCi/kg to background levels at the boundary between the laminated and turbated sediment. The maximum concentration of ^{137}Cs as well as other reactor-produced radionuclides (see Table 3) at 30-35 cm is correlated to the 1971 maximum reactor release (see Figure 7B). The sharp drop in ^{137}Cs activity below 53 cm may reflect a dredged boundary.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57.5W
57.5M
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53.8EC
52.5EC
FOUNDRY COVE
49.5WC
BEAR MT. BRIDGE
C44
C43E
G42.5
INDIAN PT.
SLOSH III
40WC
39E
HAVERSTRAW BAY
36EC
33.8EC
TAPPAN ZEE
26.4W
26M
25.4W
25.3W
24.2W
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21.9W
21.7M
TAPPAN ZEE BRIDGE
Cross Section Fig. 2
G18.3
15.8M
14.2M
13.2M
11.5W
G.W. BRIDGE
8.7WM
8W
6W
5.3M
3.8E
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NARROWS
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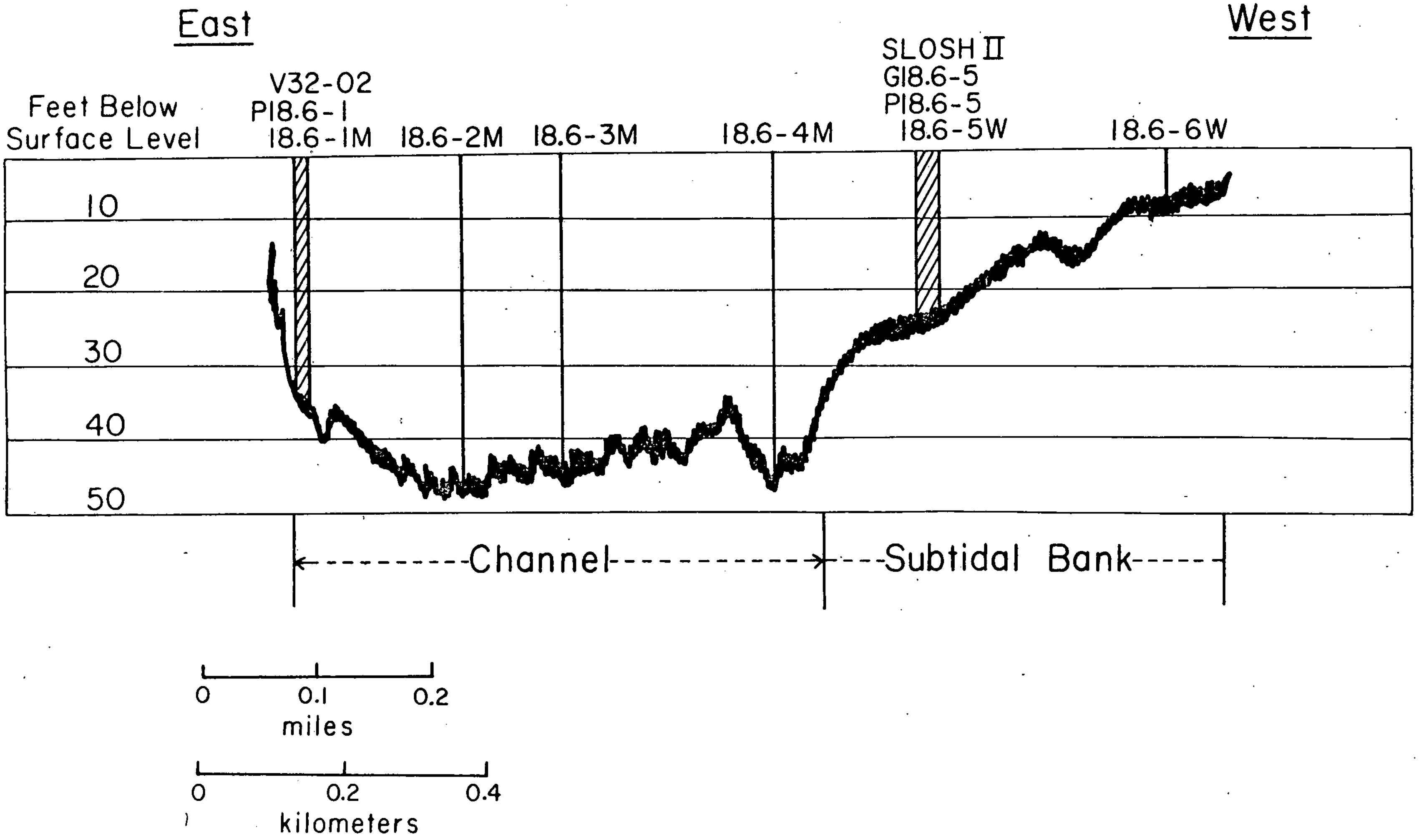
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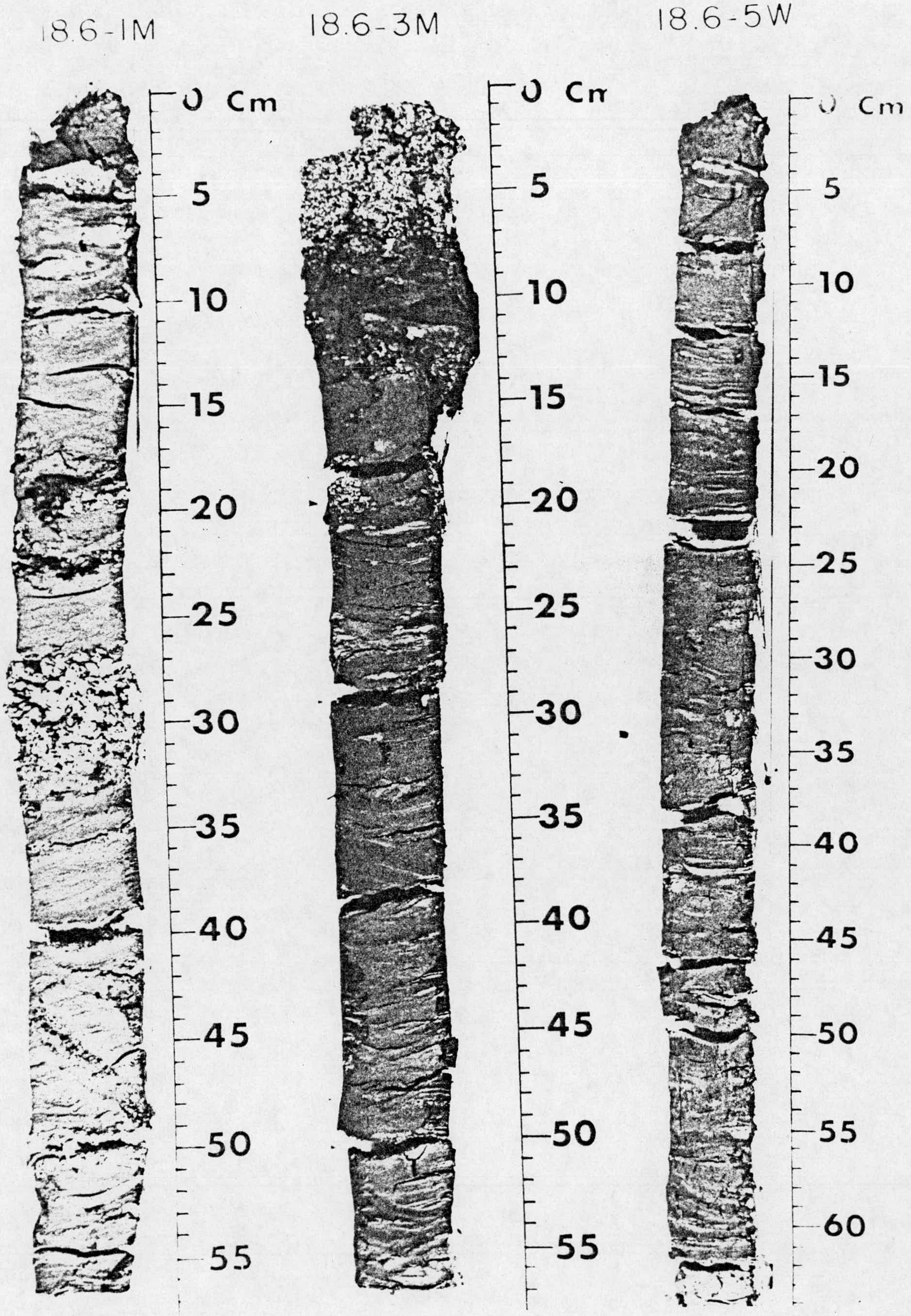
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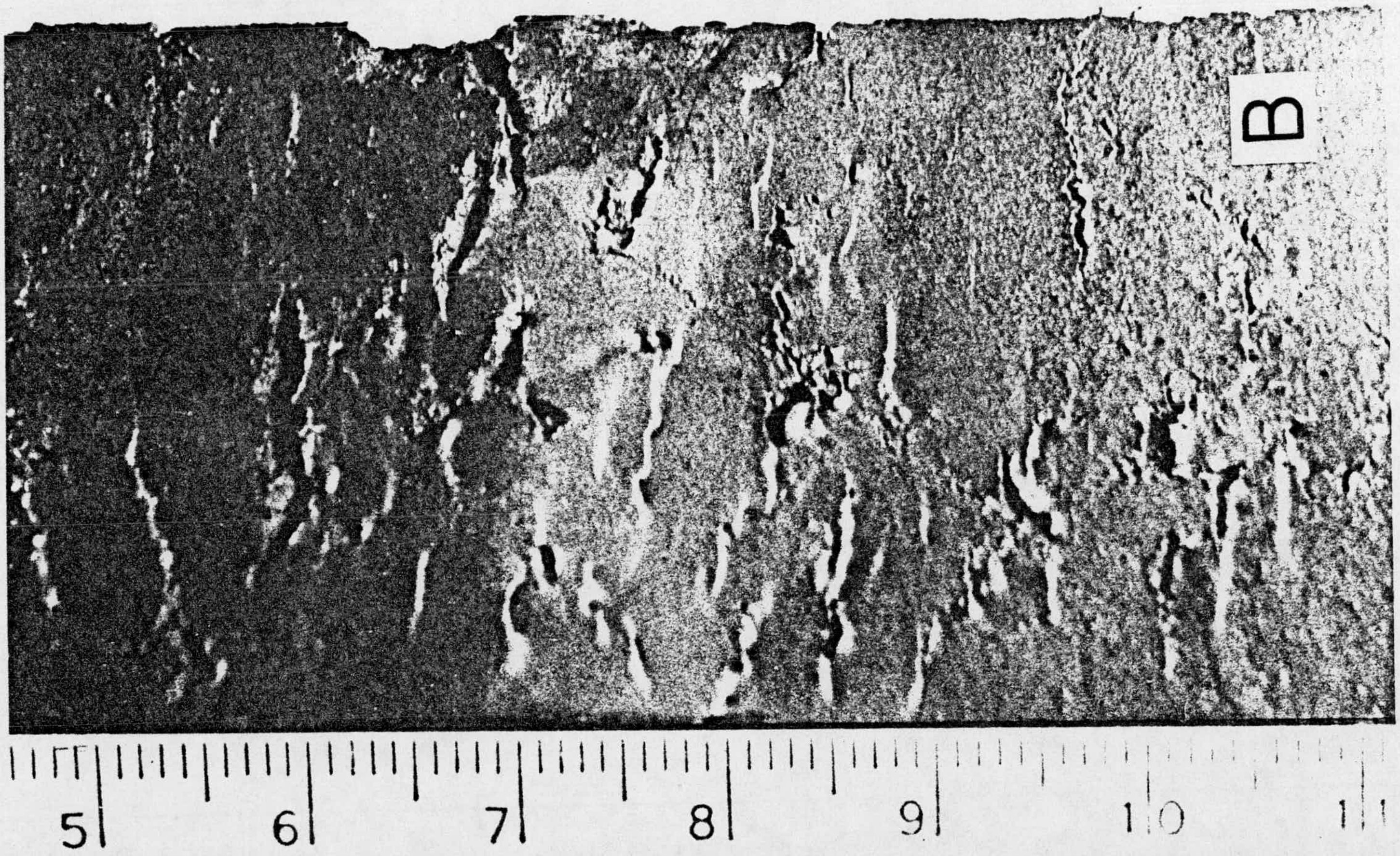
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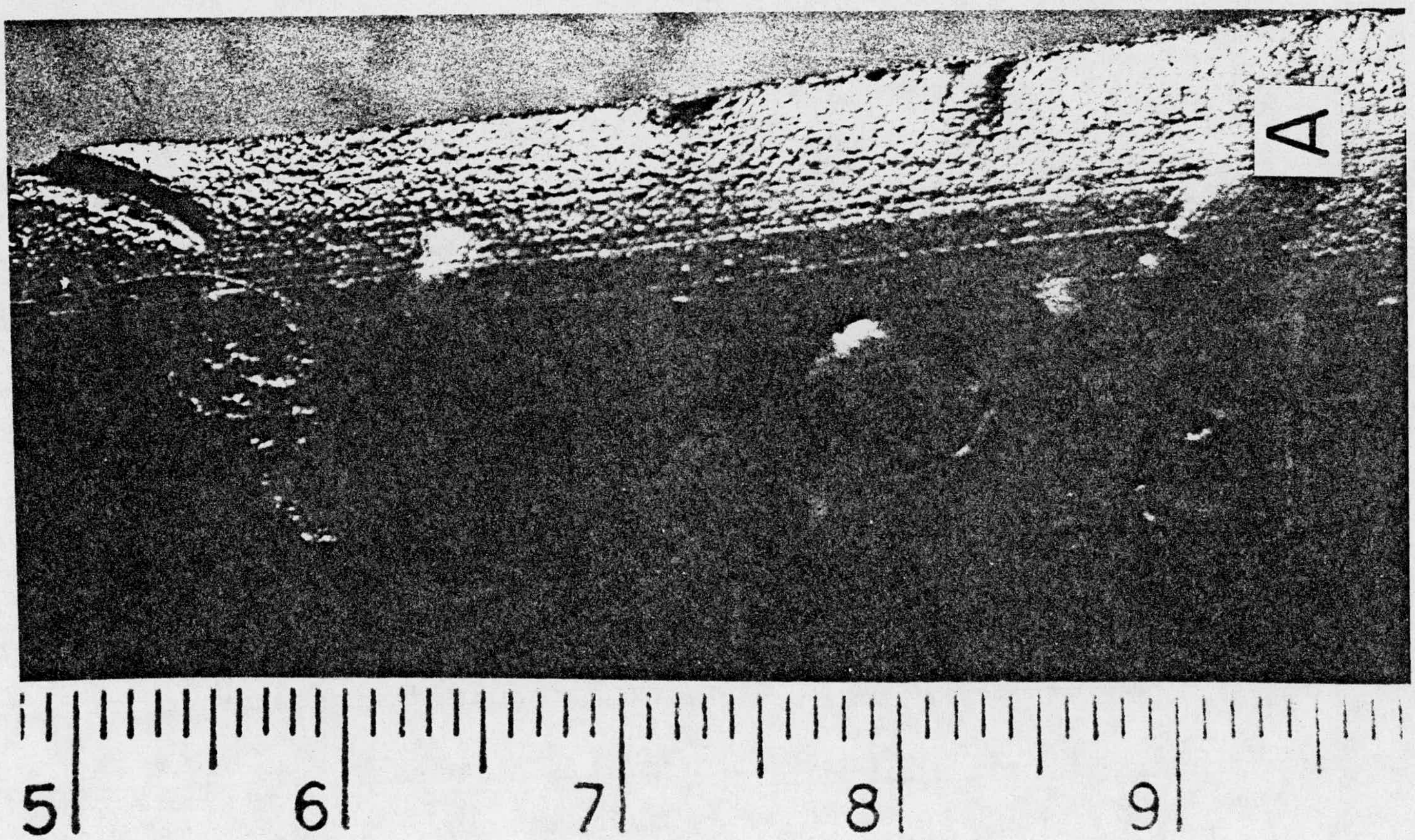
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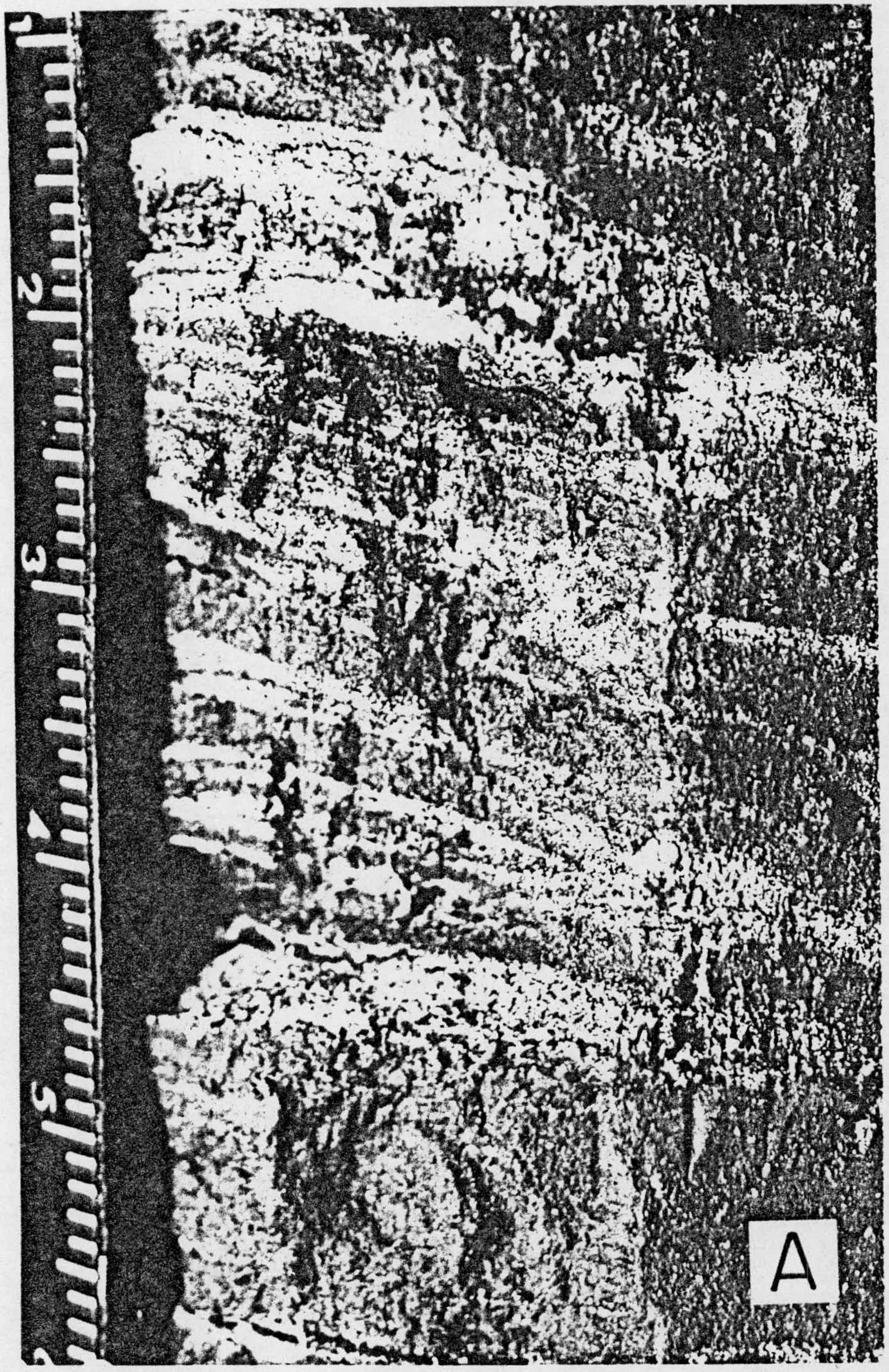
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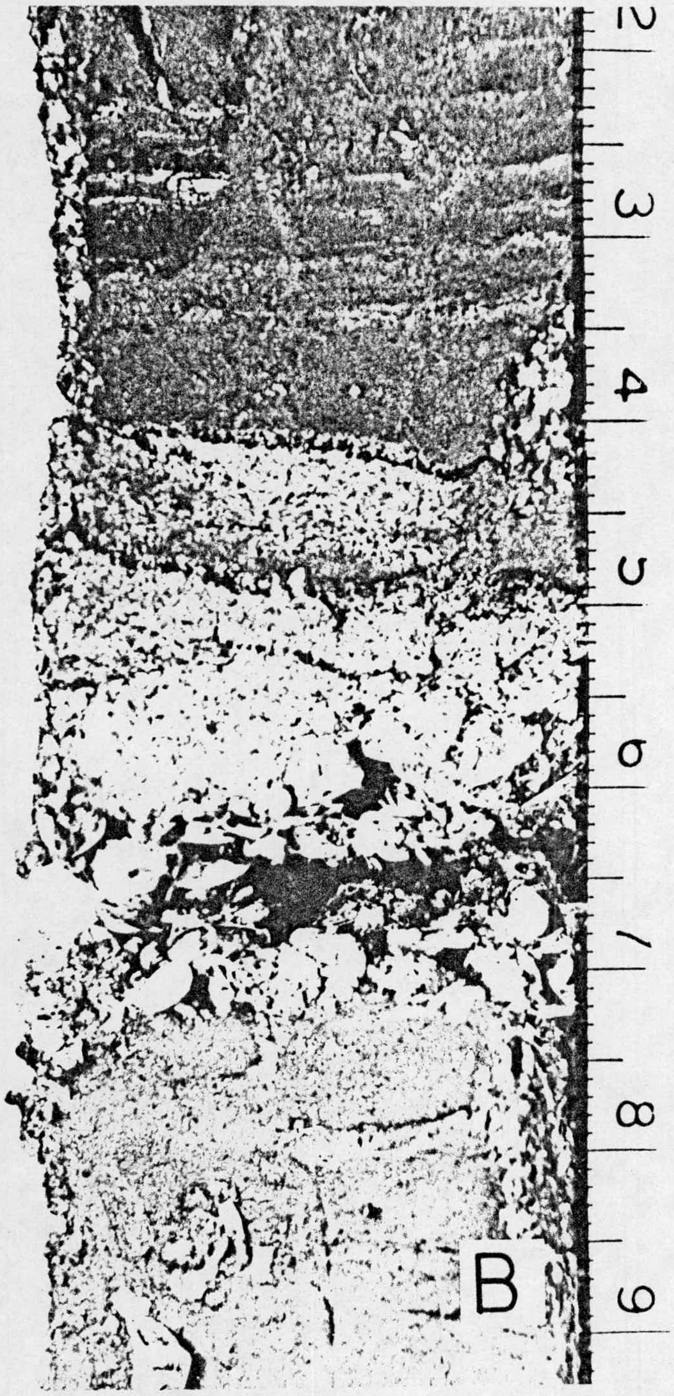
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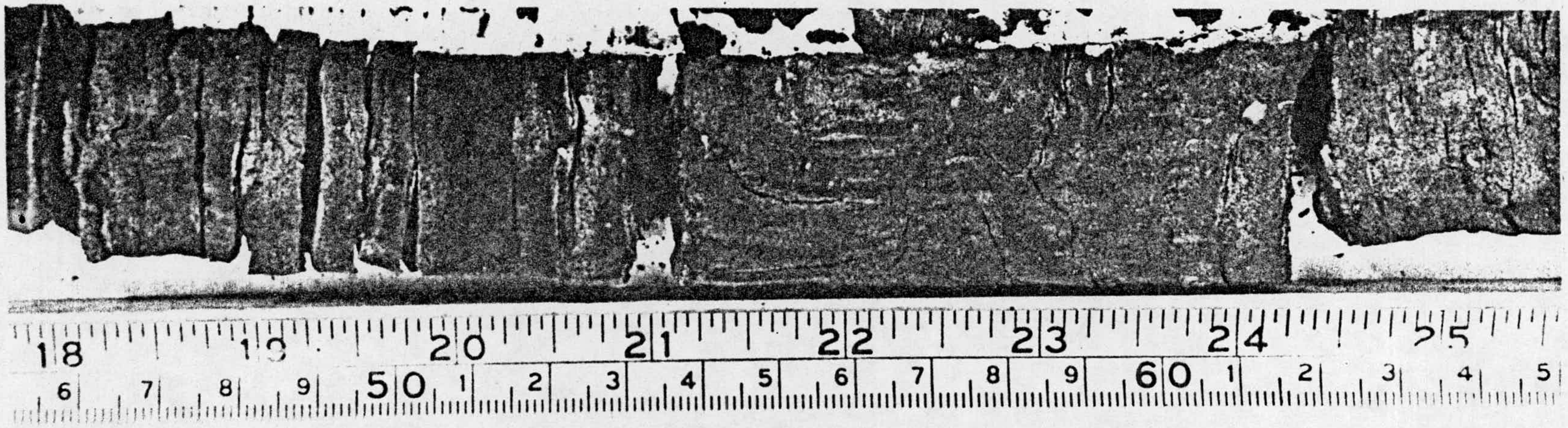
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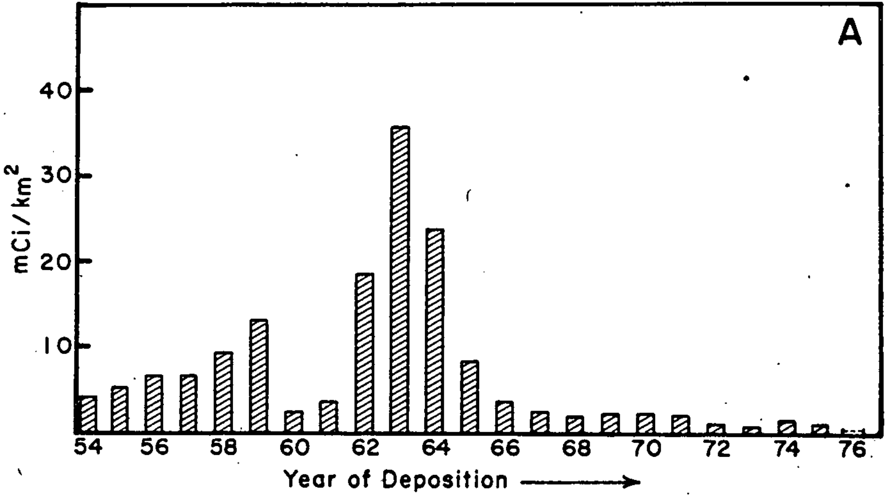
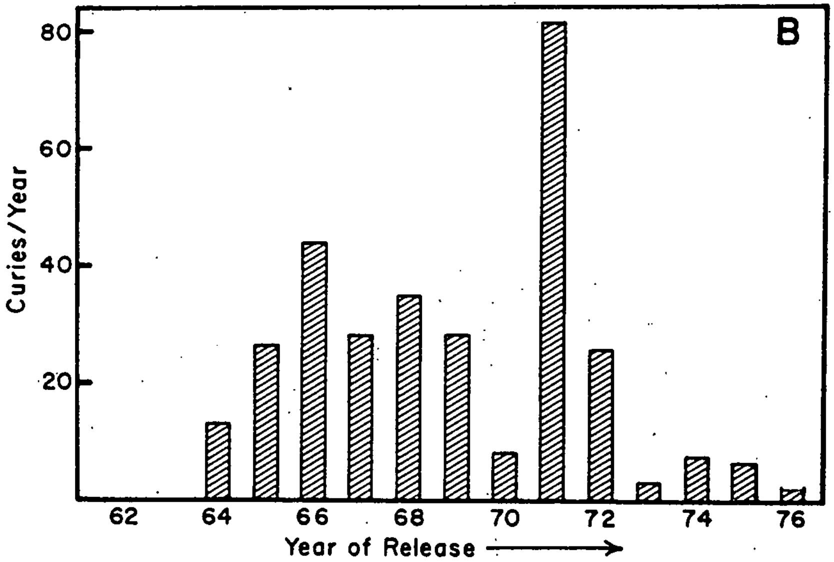
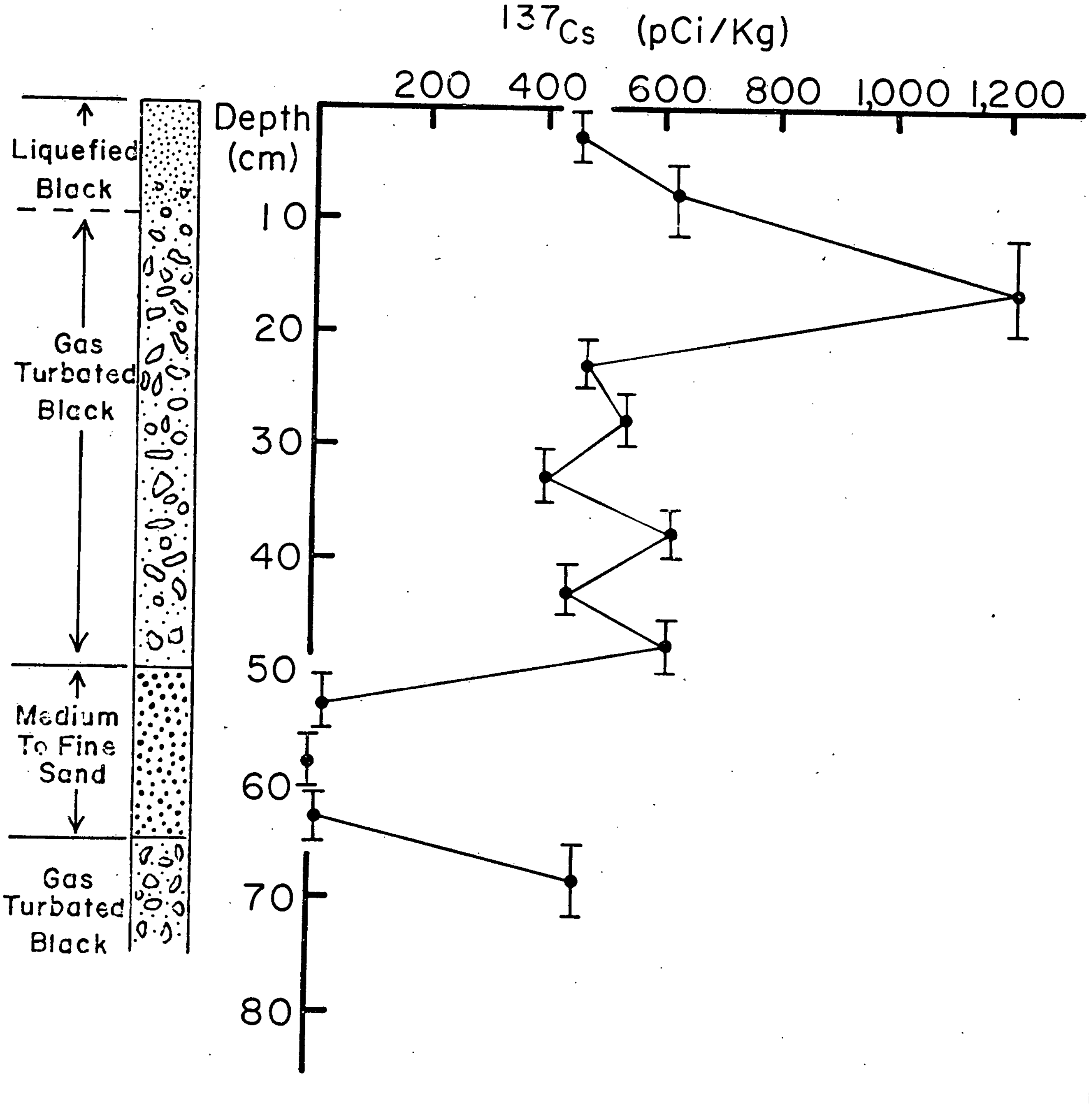
^{137}Cs Fallout at New York, N.Y.



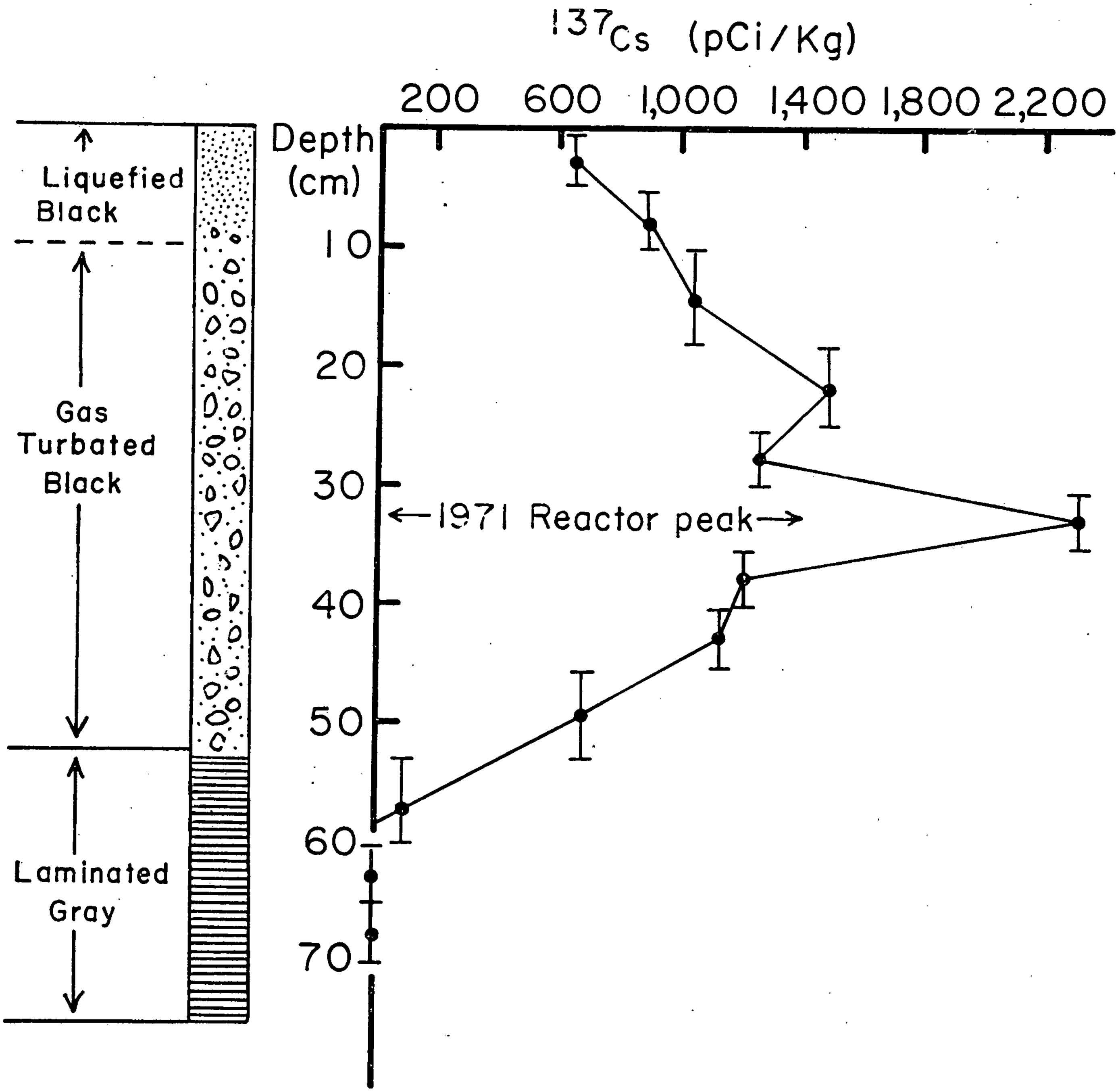
Gross Beta and Gamma Releases from Indian Point (Liquid Effluent, Excluding Tritium)



^{137}Cs Profile Core - 1.6 E



^{137}Cs Profile Core 0.1W



Cesium-137 as a Tracer for Reactive Pollutants in Estuarine Sediments

H.J. Simpson, R. Bopp, C.R. Olsen, R.M. Trier and S.C. Williams

Abstract

Many reactive pollutants discharged to natural waters become associated with fine-grained particulates. Accumulation and transport patterns of fine particles in estuaries and other natural water systems can be quite complex and difficult to predict. Cesium-137, a fission product with a 30 year half life, has been added in readily measureable quantities to natural waters throughout the globe as a result of fallout from atmospheric nuclear weapons testing. Measurement of Cs-137 in estuarine sediments can be used to rapidly establish the distribution of recent (last two decades) fine-grained sediments. In the sediments of the Hudson River Estuary (U.S.A.) the amount of Cs-137 has been found to correlate with the distribution of a wide range of reactive pollutants in terms of both depth profiles in the sediments, as well as surface sediment concentrations. The pollutants for which we have found such a covariance with Cs-137 include Pu-239,240, PCB's, Zn, Cu, Pb, Cd and Ni.

Cesium-137 as a Tracer for Reactive Pollutants in Estuarine Sediments

Introduction

A substantial number of the pollutants discharged to natural waters can be classified as "reactive" in terms of their propensity to be associated with particles, either in the original effluent or after becoming dispersed in the receiving water. For example, metals from the electroplating industry or some types of artificial radionuclides released from power plants are transported and accumulated on particles in natural waters, as well as in solution. The particles which are most important in reactive pollutant transport are usually relatively small and often contain both organic and inorganic components. We will not discuss the composition or pollutant binding characteristics of these fine particles, but instead will describe some of their characteristics as vectors of pollutant dispersal and accumulation.

In estuaries, fine particles (< 63 microns) are quite mobile and often undergo many episodes of deposition and resuspension by the variable currents of tidal waters. In theory, it should be possible to describe and predict the pathways of fine particle transport in estuaries, based on the physics of the particle motions and numerical models of sufficient complexity or from properly-scaled physical models. Actually, it is more practical to make direct field measurements of particle transport or to use tracers to infer the net motion of particles over extended periods of time. The approach described here uses the presence of a "natural" tracer (Cesium-137) associated with fine particles in estuaries as a guide to the distribution and transport of several types of pollutants. The pattern of accumulation of fine particles in estuarine sediments is quite complex and essentially unique to each estuary. As a first approximation, estuarine sediments can be grouped into three end members: (1) large mineral particles, such as quartz sands, which are relatively unimportant in the transport of reactive pollutants; (2) fine particles (roughly those particles smaller than 63 microns) which have not acquired significant quantities of pollutants, probably primarily because they have had relatively little contact with soluble phase pollutants (these will be referred to here as "old" fines); and (3) fine particles with readily measureable quantities of pollutants, which will be denoted here as "recent" fines. Obviously the degree of contamination of recent fines can be extremely variable, but as will be shown in the case of the Hudson River Estuary (U.S.A.), there is often a relatively uniform dispersal of reactive pollutants in recent fine particles over large areas and a surprisingly coherent distribution of several types of pollutants.

Cesium-137 as an Indicator of Recent Sediments

Atmospheric testing of large nuclear weapons during the 1950's and 1960's, predominantly by the U.S.A. and U.S.S.R., dispersed a great variety of radioactive materials over the entire earth. A number of these products have sufficiently long radioactive half-lives to have been valuable as tracers of global scale processes. The pattern and time scale of deposition of Strontium-90 ($t_{1/2} \sim 29$ years), especially in the northern hemisphere, have been followed closely (Volchok, 1966; Volchok and Kleinman, 1971) because of its long half-life, potentially serious biological impact, and the existence of relatively direct pathways by which this nuclide can reach man. The deposition history of Cs-137 ($t_{1/2} \sim 30$ years) has not been documented to the extent as has Sr-90 because it does not appear to be of nearly as much biological concern to man as Sr-90. The available data indicate that the pattern of delivery of fallout Cs-137 to the earth's surface can be assumed to be identical to Sr-90, with an activity ratio of Cs-137 to Sr-90 of ∿ 1.5 (Hardy, 1974). The peak delivery of fallout Cs-137 to the earth's surface by rain and snow occurred during the years 1962-1964, and the quantities deposited since then have been relatively small.

In the open ocean, both Sr-90 and Cs-137 appear to have remained predominantly in solution (Broecker _et al_., 1966; Folsom _et al_., 1970; Bowen and Roether, 1973), although there is some indication of more removal of Cs-137 than Sr-90 into the sediments (Noshkin and Bowen, 1973). The fraction of the total fallout Cs-137 delivered to the ocean which is now in the sediments is quite small. In most fresh water lakes, Sr-90 stays in solution to the first approximation, but Cs-137 is nearly completely removed onto particles (Wahlgren and Marshall, 1975; Farmer _et al_., 1977). In rivers and estuaries, the amount of fallout Cs-137 associated with particles as compared with that which passed through these systems in solution is not well-defined (Riel, 1972), but sufficient amounts are found in the sediments of estuaries which we have studied to be readily measureable.

We usually measure Cs-137 in estuarine sediments by gamma counting of 50-100 gram samples of dried sediment which have undergone no chemical steps to enrich the specific activity of the samples. Our counting equipment consists of a high resolution lithium-drifted germanium detector and a multichannel analyzer, which allows us to simultaneously measure the activity of many other radionuclides (both natural and artificial) as well as the Cs-137 gamma emission peak at 662 Kev. Because of our ability to measure Cs-137 at "normal" environmental levels in sediments with non-destructive gamma counting, we are able to process a large number of samples with relatively little laboratory preparation effort, compared with the analytical techniques required for most pollutant measurements.

Cesium-137 and Other Anthropogenic Components in Hudson Estuary Sediments

The total delivery of fallout Cs-137 to the Hudson Estuary, decay corrected to 1975, has been about 120 mCi/km^2 (USERDA, 1975). There is an additional supply of Cs-137 from a nuclear electrical-generating facility located near the upstream end of the salinity intrusion in the Hudson. The total release of Cs-137 from this facility over more than a decade of operation has been comparable to the amount supplied by rain to the surface of the Hudson Estuary from global fallout. Thus the direct supply of Cs-137 to the Hudson Estuary is roughly a factor of two greater than might be expected if fallout were the only source.

The specific activity of Cs-137 in surface sediments in the Hudson ranges over more than two orders of magnitude, with the lowest values in sandy sediments typical of areas scoured of fine particles by strong currents. The fine-grained surface sediments usually range between 0.2 and 2 pCi/g of Cs-137, which is comparable to fallout Cs-137 activity in surface soils throughout the northern hemisphere (Hardy, 1974; Ritchie _et_ _al_., 1975). There is also a large variation in the depth to which Cs-137 is found in the sediments. In some areas the activity is confined to the upper 5 cm, whereas in others it extends to nearly 3 meters. In general, the geographical areas of lowest surface activity also have smaller depth accumulations of measureable Cs-137 activity. Thus the integrated amount of Cs-137 per unit area is extremely non-uniform, and ranges over approximately three orders of magnitude. As a result relatively limited geographical areas account for large portions of the total sediment burden of Cs-137. In the Hudson the dominant areas of Cs-137 accumulation are the harbor and shallow coves upstream of the harbor. These areas are not in close proximity to the site of localized discharge of Cs-137 to the Hudson, and primarily reflect the zones in which fine particles are accumulating rapidly (Simpson _et_ _al_., 1976).

We have found the distribution of other man-made reactive contaminants in Hudson sediments to be very similar to that of Cs-137, despite significant differences in chemistry, and mode of input to the system. The locations of sediment sampling sites for data reported here are shown in Figure 1. These sites extend from approximately the upstream limit of salinity intrusion during summer months, to the harbor area which typically has salinities of approximately two-thirds of sea water.

In Figure 2A, activities of Pu-239,240, determined by alpha spectrometry (Wong, 1971), are plotted against Cs-137 in the same samples. The covariance over two orders of magnitude of these two parameters in Hudson sediments is clear. Thus if the present distribution of Pu-239,240 in Hudson sediments (mostly derived from fallout) were to be measured the most efficient procedure would be to use the distribution of Cs-137, which is relatively easy to measure by gamma spectrometry, to guide the selection of samples for analysis by alpha spectrometry for Pu-239,240. (The alpha particle energies of these two plutonium isotopes are nearly identical and the sum of their activities is usually reported.)

In Figure 2B, the concentration of polychlorinated biphenyls (PCB's) in Hudson sediments is plotted against Cs-137. Although our data is limited at this time, the covariance of these constituents is also obvious. The levels of PCB's are high in sediments over large areas of the Hudson because of industrial releases for a period of approximately two decades at two sites more than 200 km upstream of the locations of our sampling area. Considering the great differences in chemistry between Cs-137 and PCB's, it is perhaps surprising to find their sediment distributions to be as similar as they are, but their covariance is a good indicator of the ability of fine particles to transport and accumulate quite a variety of reactive pollutants.

In Figure 3A is shown the covariance of several trace metals with Cs-137. Zinc, copper and lead concentrations in Hudson sediments are several times the levels which existed in pre-industrial sediments. All of the samples shown in Figure 3A are upstream of the harbor area, and thus reflect diffuse sources of these metals to the Hudson over a number of decades. Samples in the harbor are somewhat higher in all three metals, because of discharges from the electroplating industry. Vertical distributions of all three metals in harbor sediments also are similar to that of Cs-137.

In Figure 3B the concentrations of cadmium and nickel in a small cove are plotted against Cs-137. High level contamination of the sediments of this small (∿ 0.5 km^2) shallow (mean depth ∿ 1-2 meters) area by effluent from a battery factory has resulted in Cd concentrations ranging from a few percent to ∿ 100 ppm. Some surface sediments in the cove which are apparently in areas of active current scouring contain relatively low amounts of Cd and Ni, and also of Cs-137. Thus Cs-137 can be useful in mapping the pattern of trace metal accumulation in sediments in relatively small, highly contaminated areas, as well as, as for diffuse sources over large areas.

Cesium-137 as a Pollutant Tracer in other Aqueous Systems

Fallout Cs-137 has been used as an indicator of recent sediments in the Delaware Estuary. Sites with appreciable activity of Cs-137 in surface sediments also have hydrocarbon constituents typical of recent pollution, whereas surface sediments free of Cs-137 have hydrocarbons typical of unpolluted marsh sources (Wehmiller, personal communication).

In lake sediments, the distribution of Cs-137 has been shown to be nonuniform in large lakes and to covary with fallout Pu-239,240 (Edgington *et al*., 1976). We believe it is likely that similar covariance in fine-grained coastal sediments is likely, based on the data available.

Thus the concentration of a number of reactive pollutants in estuarine sediments, although complicated in both surface and depth distributions, has been shown to have considerable coherence from one pollutant to another and to have a strong correlation with Cs-137. This coherence greatly simplifies the task of establishing the sediment distribution of reactive pollutants in complicated sedimentary regimes through the use of a "natural" tracer, Cs-137.

Figure Captions

Figure 1 Locations of cores for which data are reported in this paper are indicated by ⊘.

The most-northerly sampling sites are near the upstream limit of salt water intrusion of the Hudson Estuary during summer months, and the harbor sites adjacent to New York City usually have salinities of one half to two thirds of that of sea water. The Hudson is tidal for approximately 250 km upstream of New York City. The locations of discharge of polychlorinated biphenyls (PCB's), cadmium and nickel (Cd, Ni), and radioactive cesium (Cs-137) are also indicated.

Figure 2-A Activities of Cs-137 and Pu-239,240 in Hudson Estuary sediment samples are given for sites indicated in Figure 1. Data are for samples well below the sediment-water interface as well as surface sediment samples. Two suspended particulate samples (△) collected near the middle of the sampling range are also included. All data is expressed as activity per dry weight of sediment.

Figure 2-B Activities of Cs-137 and concentrations of polychlorinated biphenyl's (PCB's) in samples of surface sediment are given for Hudson Estuary sites included in Figure 1. All data is expressed in terms of dry weight.

Figure 3-A Activities of Cs-137 and concentrations of zinc (X), copper (Δ) and lead (O) in Hudson Estuary sediment samples are plotted for sites indicated in Figure 1. Data are for samples well below the sediment-water interface as well as surface sediment samples. The data are presented on a linear plot to indicate the observed range of pre-industrial concentrations in Hudson sediments on the Y axis. All data is expressed in terms of dry weight.

Figure 3-B Activities of Cs-137 and concentrations of cadmium(●) and nickel (⊗) are given for sediment samples from a small cove in the Hudson receiving effluent from a battery factory (see insert in Figure 1 where locations of samples are indicated by x's). Some of the lowest concentrations are from surface samples of fine-grained sediment in areas which are apparently scoured by tidal currents and are thus kept free of recently deposited sediments. Samples from these scoured sites have a similar physical appearance to those from the highly contaminated sites.

References

Bowen, V.T. and Roether, W. (1973). Vertical distributions of strontium-90 cesium-137 and tritium near 45° North in the Atlantic, J. Geophys. Res., 78, pp. 6277-6285.

Broecker, W.S., Bonebakker, E.R. and Rocco, G.G. (1966). The vertical distribution of cesium-137 and strontium-90 in the oceans, 2, J. Geophys. Res., 71, pp. 1999-2003.

Edgington, D.N., Alberts, J.J., Wahlgren, M.A., Karttuenen, J.O. and Reeve, C.A. (1976). Plutonium and americium in Lake Michigan sediments, Transuranium Nuclides in the Environment, IAEA-SM-199/47, IAEA, Vienna, pp. 493-516.

Farmer, J.G., Bowen, V.T. and Noshkin, V.E. (1977). Long-lived artificial radionuclides in Lake Ontario, I. Supply from fallout, and concentrations in lake water of plutonium, americium, strontium-90 and cesium-137, submitted to Limnology and Oceanography.

Folsom, T.R., Sreekumaran, Hansen, N., Moore, J.M. and Grismore, R. (1970). Some concentrations of Cs-137 at moderate depths in the Pacific 1965-1968, USAEC Rep. HASL-217, pp. 1-9.

Hardy, E.P. (1974). Depth distributions of global fallout Sr-90, Cs-137 and Pu-239,240 in sandy loam soil, USAEC Rep. HASL-286, pp. 2-10.

Noshkin, V.E., and Bowen, V.T. (1973). Concentrations and distributions of long-lived fallout radionuclides in open ocean sediments, Radioactive Contamination of the Marine Environment, IAEA, Vienna, pp. 671-686.

Riel, G.K, (1972). The distribution of fallout cesium-137 in the Chesapeake Bay, Proceedings of the Second International Symposium on the Natural Radiation Environment, Adams, J.A.S., Lowder, W.M., Gesell, T.F., Eds., pp. 883-896.

Ritchie, J.C., Hawks, P.H. and McHenry, J.R. (1975). Deposition rates in valleys determined using fallout cesium-137, Geol. Soc. Amer. Bull., 86, pp. 1128-1130.

Simpson, H.J., Olsen, C.R., Trier, R.M. and Williams, S.C. (1976). Man-made radionuclides and sedimentation in the Hudson River Estuary, Science, 194, pp. 179-183.

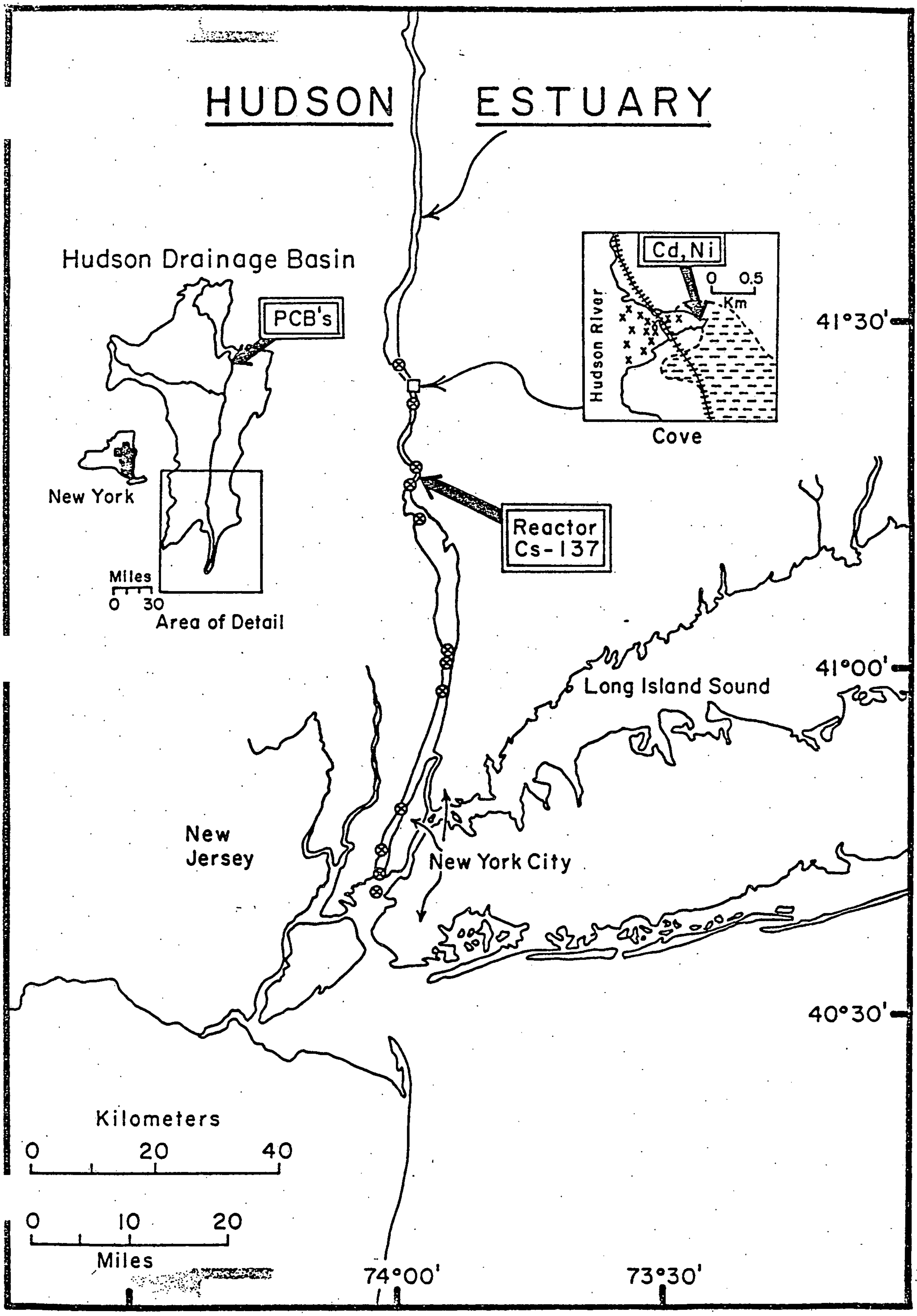
USERDA Rep. HASL-294 (1975), appendix, pp. 68-70.

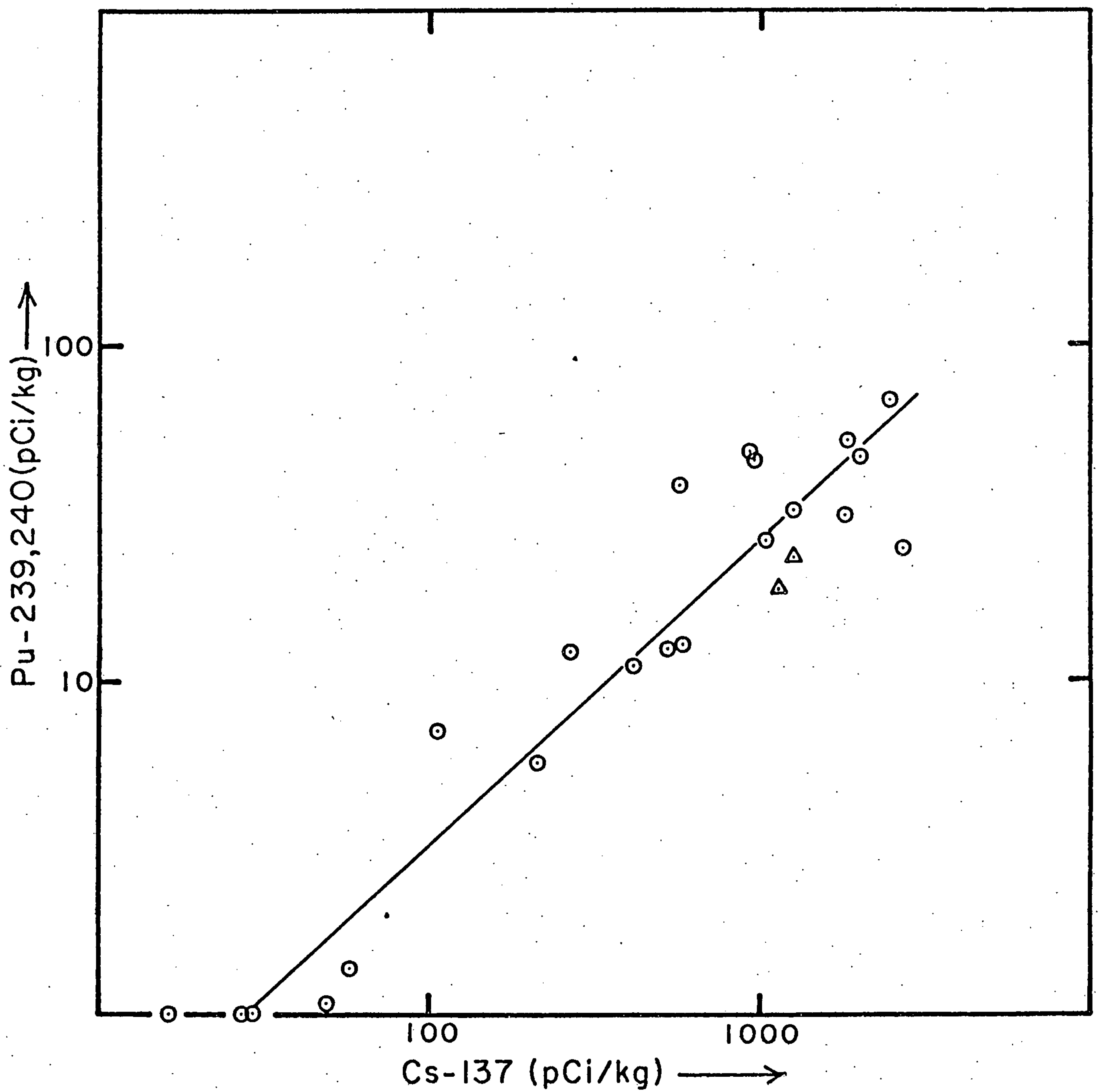
Volchok, H.L. (1966). The global strontium-90 budget, J. Geophys. Res., 71, pp. 1515-1518.

Volchok, H.L. and Kleinman, M.T. (1971). Global Sr-90 fallout and precipitation: Summary of the data by 10 degree bands of latitude, USAEC Rep. HASL-245, pp. 2-83.

Wahlgren, M.A. and Marshall, J.S. (1975). The behavior of plutonium and other long-lived radionuclides in Lake Michigan: I. Biological transport, seasonal cycling and residence times in the water column, International Symposium on Transuranium Nuclides in the Environment, IAEA-SM-198/39, IAEA, Vienna, pp. 227.

Wong, K.M. (1971). Radiochemical determination of plutonium in sea-water, sediments and organisms, Anal. Chim. Acta, 56, pp. 355-364.


HUDSON ESTUARY
Hudson Drainage Basin
PCB's
New York
Miles
0 30
Area of Detail
Cd, Ni
0 0.5
Km
Hudson River
Cove
41°30'
Reactor
Cs-137
41°00'
Long Island Sound
40°30'
New Jersey
New York City
Kilometers
0 20 40
0 10 20
Miles
74°00'
73°30'


Pu-239,240 (pCi/kg) →
100
10
100
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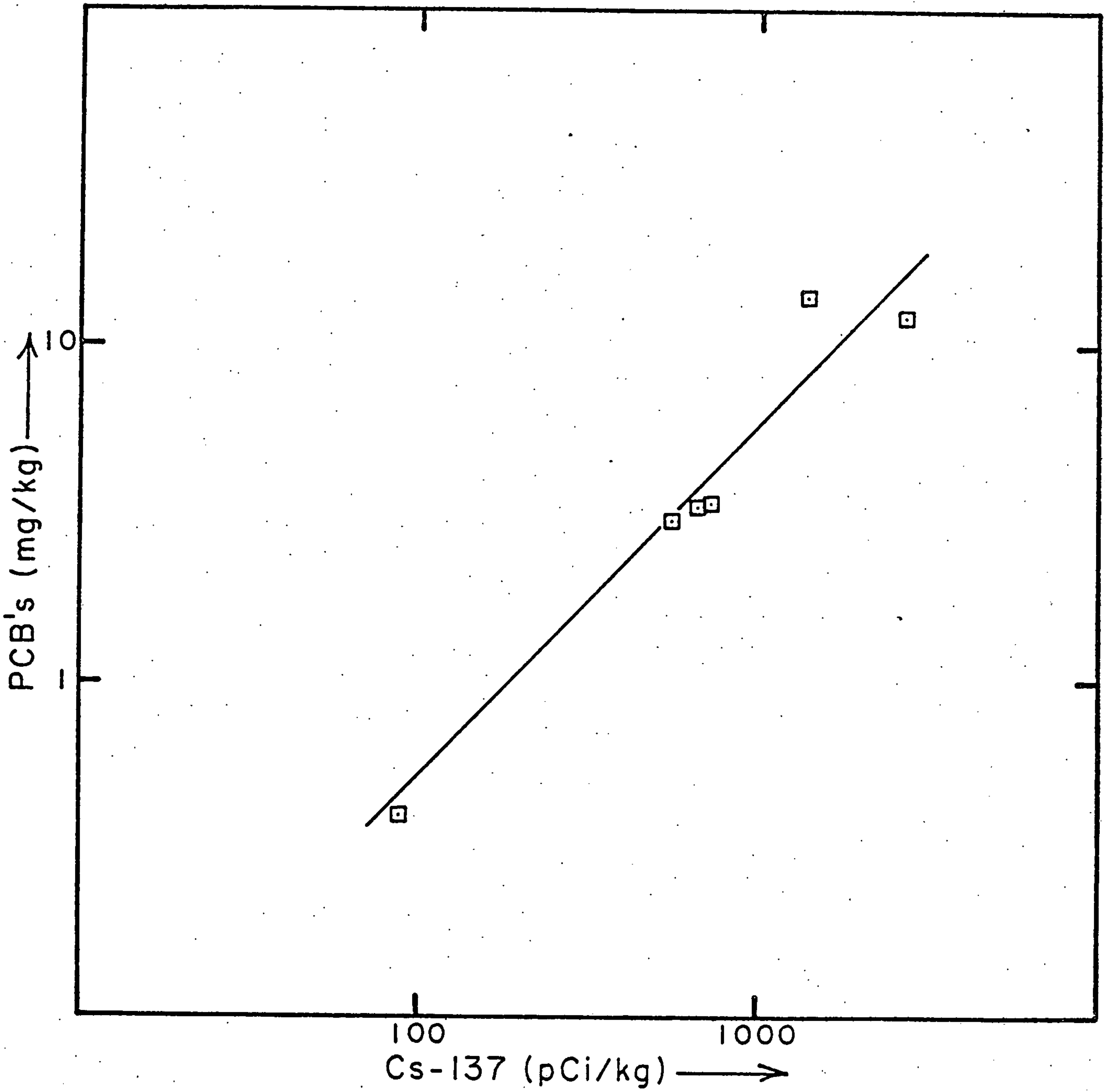
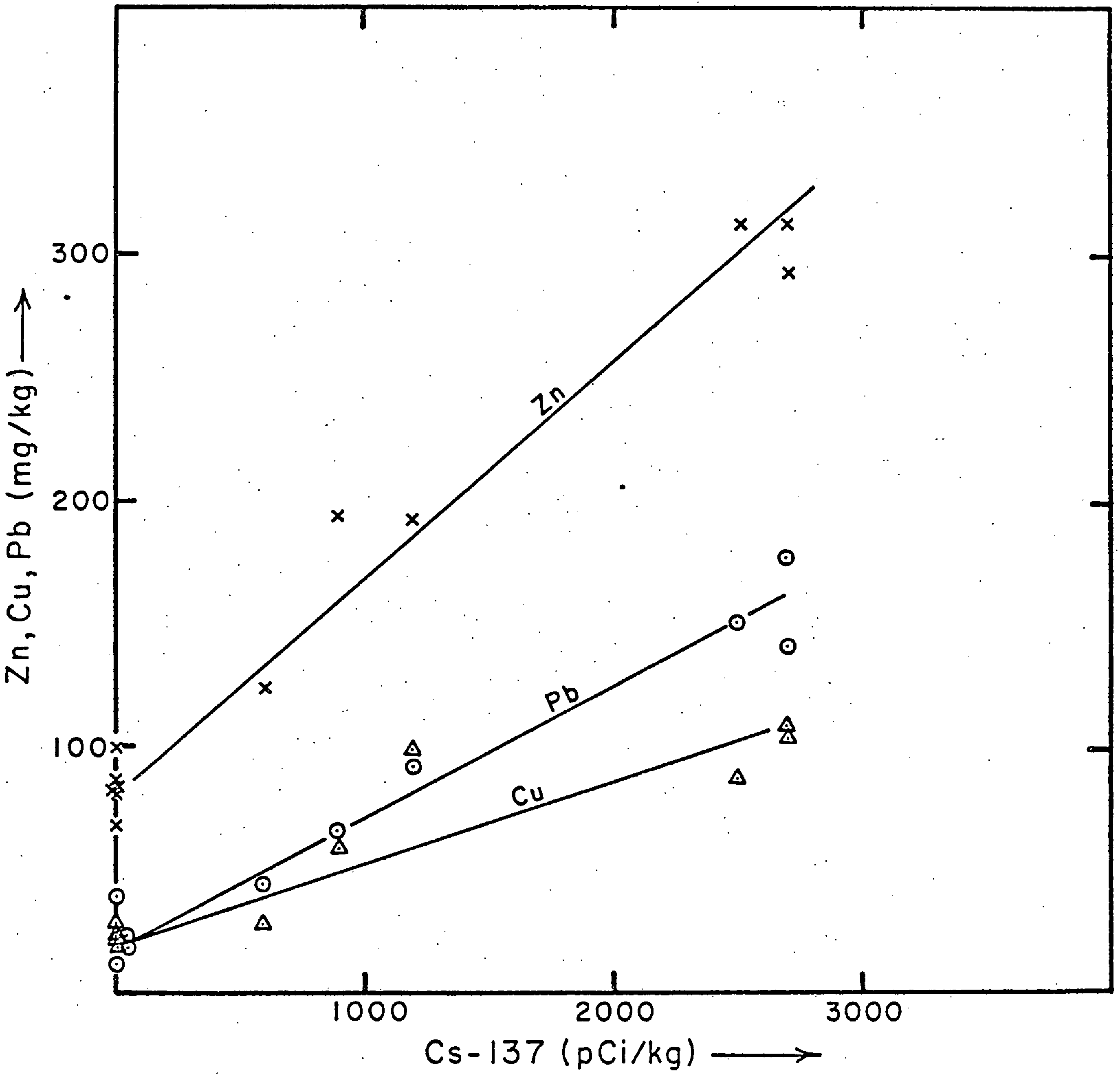
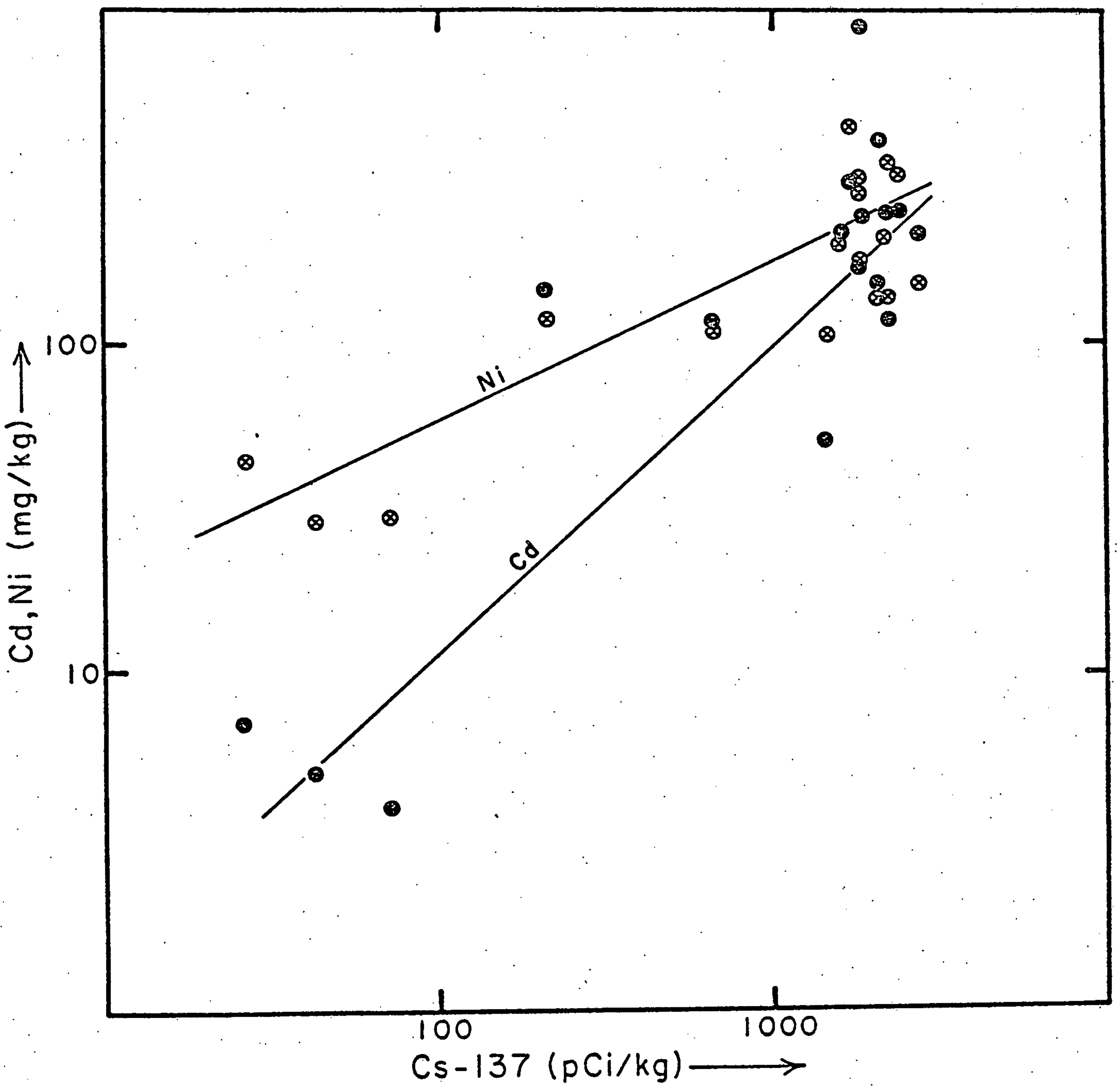
Cs-137 (pCi/kg) →


10
1
PCB's (mg/kg)
100
1000
Cs-137 (pCi/kg)


Zn
Pb
Cu
300
200
100
1000
2000
3000
Zn, Cu, Pb (mg/kg) ⟶
Cs-137 (pCi/kg) ⟶



100
10
100
1000
Ni
Cd
Cd, Ni (mg/kg) ⟶
Cs-137 (pCi/kg) ⟶

SOURCES OF HEAVY METALS IN SEDIMENTS OF THE HUDSON RIVER ESTUARY

by

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Sources of Heavy Metals in Sediments of the Hudson River Estuary

ABSTRACT

Sediments in the Hudson Estuary contain zinc, copper and lead from metal pollutants discharged to the harbor in the New York City area, from dispersed sources of contamination introduced upstream, and from natural weathering processes. The magnitude of the contribution from each of these three sources to particular sites can be estimated on the basis of total metal abundances, relative proportions of several metals, and other sediment properties. The pattern of recent heavy metal contamination in Hudson sediments closely follows the distribution in sediments of cesium-137 which was derived over the past two decades from global fallout and local releases from a commercial nuclear reactor. Several simple empirical corrections related to grain size and mineralogy variations are suggested for comparing heavy metal contamination levels of sandy continental shelf sediments with fine-grained estuarine and coastal sediments. Iron has little variation in Hudson sediments while manganese is greater in surface sediment of some low salinity and fresh water areas than deeper in the sediments, and generally less in the high salinity area of rapid sediment deposition in New York harbor. Much of the pollutant Cu added to the harbor appears to be rapidly deposited in the sediments.

Sources of Heavy Metals in Sediments of the Hudson River Estuary

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INTRODUCTION

Heavy metals discharged into estuaries and coastal waters with domestic and industrial wastes are often present as particulates, or have strong affinities for solid phases in the receiving waters (Morel *et al*., 1975). Present distributions of metals in sediments can serve as an indication of the time history and extent of pollutant discharge in specific areas (Chow *et al*., 1973; Bruland *et al*., 1974; Erlenkeuser *et al*., 1974; de Groot *et al*., 1976). To interpret the present metal content of sediments, several factors must be considered including the "natural" preindustrial metal content of sediments in the area in question. Components such as clay minerals, organics and iron and manganese oxides are important phases in both the natural and pollutant heavy metal content of sediments. Other phases such as quartz and calcium carbonate, which tend to occur in relatively large particles, are usually less important in binding heavy metals, and thus serve primarily to "dilute" the heavy metal bearing phases which dominate the fine particle size fraction. In environments where particle size distributions and mineralogy are heterogeneous, substantial variability in heavy metal concentrations would thus be expected, even in the absence of significant local sources of metal contamination (de Groot *et al*., 1976). The pattern of sediment accumulation in environments such as estuaries or coastal waters is generally sufficiently complicated to make it difficult to establish the time history of accumulation at any particular sampling site.

We have analyzed sediments from the Hudson estuary and adjacent coastal area for zinc (Zn), copper (Cu), lead (Pb), manganese (Mn) and iron (Fe), as well as man-made radionuclides such as cesium-137, to establish the present distribution of heavy metals in a system subject to substantial pollutant loading. We have related the present sediment metal concentrations to those typical of the Hudson prior to industrial pollutant discharge, and to the patterns of recent sediment accumulation.

SAMPLING AND ANALYTICAL METHODS

Cores were taken from near the upstream limit of saline water intrusion in the Hudson to the middle of New York harbor (Figure 1) where salinities are usually about two thirds of deep ocean water salinities. Large grab samples of the upper 10 centimeters of sediment in the coastal waters adjacent to the Hudson were collected out to beyond the edge of the continental shelf (Figure 2). Most of the cores were collected in plastic liners with a six centimeter diameter gravity corer, and were usually about 0.5 meters in length. One of the cores from nineteen miles upstream of the southern tip of Manhattan Island (mile point 19) was a 6 meter piston core taken from one of Columbia University's oceanographic vessels (R/V VEMA). Another of the cores (mp 24) which had a total length of about 12 meters was taken in sections by hand drilling in a marsh which currently is under water during only very high tides.

Sediment samples were air dried, ground in a mortar and pestle and passed through a 250 μ nylon sieve. Samples for heavy metal determinations were dried at 105°C to constant weight and stored in a dessicator, while those for measurement by gamma spectrometry were sealed in teflon-lined aluminum cans and counted with a large volume lithium-drifted germanium detector and multichannel analyzer.

Metals were leached from sediments by fuming samples with concentrated HCl/HNO_3 (1:3) for several hours, followed by treatment with 70% $HClO_4$ until the solid phase was essentially pure white. Final solutions of acid were made up to approximately 5% in HCl after driving off most of the $HClO_4$. Sediment suspensions were filtered and washed through a Gooch crucible with preweighed glass fiber filters. Filtrates were diluted and analyzed by flame atomic absorption spectrometry and the white sediment residue was reweighed after drying. The difference in sediment weight resulting from treatment with strong acids is reported as "loss on leaching" (LOL). The difference in weight for fresh sediment samples dried at 105°C to constant weight and then heated to 500°C for a number of hours is reported as "loss on ignition" (LIG). Quartz was determined by monitoring the α-β transition in a differential scanning calorimeter, potassium from the gamma ray emission peak for ^{40}K at 1.46 Mev and ^{137}Cs from the gamma ray emission peak at 0.662 Mev.

RESULTS AND DISCUSSION

Heavy metal data and related measurements are reported in Tables 1, 2 and 3 for 56 samples of sediment and one of sewage sludge from a large New York City sewage treatment plant. The data are listed beginning at the upstream end of the sampling locations in the Hudson, with the data in Table 2 from New York harbor and in Table 3 from the sediments of the continental shelf. Samples from the Hudson estuary were generally fine-grained silts relatively uniform in physical properties, while several of those from the shelf were predominantly sand despite our attempts to sample the narrow zone of fine grained sediment between the mouth of the Hudson Estuary and the Hudson Submarine Canyon.

We chose the analytical scheme outlined above for Hudson sediments after trying a number of procedures. The reported data for Zn, Cu, Pb and Mn are probably good estimates of the total metal concentration in our samples, but the reported values for Fe may be systematically low (15-20%). We have analyzed a number of samples after total dissolution with HF and found the procedure to be somewhat less convenient and reproducible than the strong acid leaching method employed here. Data reported in Table 4 provide some comparison of metal concentrations in a Hudson sample which has been analyzed by three separate procedures. The sample (mp 43) used for the intercomparison of analytical techniques is typical of recent Hudson Estuary sediments upstream of New York harbor. We have also analysed seven replicates of a sample typical of preindustrial Hudson sediments (mp 19, 525 cm) which has substantially lower concentrations of Zn, Cu and Pb than the mp 43 sample. Statistical variation (2σ) in that data set was: Zn 8%, Cu 13%, Pb 39%, Mn 3% and Fe 6%. The uncertainties of Pb and Cu were higher than for the sample at mp 43 because we were approaching the detection limits by our procedures for those two metals (Pb: average 23 ppm, detection limit ∿ 10 ppm; Cu: average 19 ppm, detection limit ∿ 4 ppm).

During the past several years we have developed some understanding of the general accumulation patterns of recent sediments in the Hudson through the use of ^{137}Cs (Simpson et al., 1976; Olsen et al., 1977). This tracer has been added to the Hudson estuary from two sources: (1) global fallout from atmospheric nuclear weapons testing over the last two decades with peak deliveries from rainfall during the years 1963-1965; (2) low level releases from a nuclear power reactor at Indian Point (mp 43) over the last decade, with peak discharges in the years 1971-1972. Enough ^{137}Cs is associated with particles in the Hudson to provide a readily measureable tracer of sediment accumulation rates in areas of high deposition, and of transport along the axis of the Hudson downstream of the reactor site, when used in conjunction with two other radionuclides, ^{134}Cs and ^{60}Co which have been supplied to the Hudson almost exclusively by reactor releases. Cesium-137, which has a half life of about 30 years, is an unequivocal indicator of sediment which has been in contact with the water column within the last two decades, and primarily serves here as a "label" for recent (last two decades) sediments. Using this tracer we have found that accumulation rates of fine grained sediments in the Hudson range over approximately two orders of magnitude, with the dominant deposition zone being New York harbor. We have found cores with ^{137}Cs to nearly 3 meters below the sediment surface, and accumulation rates of 10-20 cm per year typical of large areas of the harbor. Marginal coves in the lower salinity reaches of the estuary (mp 45 - mp 56) have sediment accumulation rates of a few cm per year (Wrenn et al., 1971; Simpson et al., 1976) while most of the total area of the estuary has little net accumulation of recent fine-grained sediments labeled with ^{137}Cs (less than a cm per year).

From Tables 1 and 2 it is clear that sediments containing ^{137}Cs are significantly higher in the metals Zn, Cu and Pb. Two of the reported cores (mp 54 and mp 19) penetrate well into sediments with relatively low and constant concentrations of Zn, Cu and Pb which appear to be typical of pre-industrial sediments in the Hudson. One of the cores (mp 19) contains many subsurface layers of estuarine carbonate shells, four of which we have analyzed for ^{14}C and found apparent ages of 1000-2000 radiocarbon years. Interpretation of radiocarbon data from estuarine carbonates is not unambiguous, but we are confident that the lower half of this core (3-6 meters), which lies below the four radiocarbon dated carbonate layers in the upper 3 meters, is representative of preindustrial fine-grained sediments in the Hudson.

When data from Tables 1 and 2 are plotted in terms of the concentrations of Zn and Cu, (Figure 2) the samples fall into three relatively distinct groups. Approximately one third of the Hudson samples, from seven of the eleven coring sites, contain similar Zn and Cu concentrations below some level in each of the cores. This can be seen most clearly in three cores (mp 56, mp 24 and mp 19) two of which were the longest collected. Similar "baseline" values were found throughout the salinity range in the estuary, and none of the estuary samples in the group with Zn concentrations of ∿ 80 ppm and Cu concentrations of ∿ 20 ppm contain ^{137}Cs. We consider these samples to be typical of preindustrial fine-grained sediments throughout the Hudson.

The remainder of the samples (all of which contain ^{137}Cs and thus are recent sediments) can be represented as members of two groups. One group has a number of samples with Zn concentrations of ∿ 300 ppm and Cu concentrations of ∿ 100 ppm, as well as others which fall along a mixing line between these values and preindustrial sediment Zn and Cu values. All of these samples were collected upstream of New York harbor (Table 1) and appear to be representative of the level of heavy metals in recent Hudson sediments upstream of New York City resulting from many diffuse sources of Zn and Cu. One of the upstream sites for which we are reporting data (mp 54) is a small cove which is grossly contaminated with Cd and Ni effluent from a battery factory (Kneip et al., 1974). Bower (1976) has found Zn, Cu and Pb concentrations in this cove to be unrelated to the local metal contamination of Cd and Ni (up to ∿ 0.1% by weight of both metals) and to be typical of other recent estuarine sediments in the Hudson in terms of these three metals. The third group of samples all from New York harbor (Table 2), fall off of the trend line for recent sediments upstream of the harbor (Figure 2) and extend up to considerably higher concentrations of both zinc (∿ 550 ppm) and copper (∿ 400 ppm). Metal contamination added directly to New York harbor sediments appears to be richer in Cu relative to Zn than the diffuse recent contamination of sediments transported down the Hudson toward the harbor. The sewage sludge sample we analyzed had very high Cu (∿ 1400 ppm) relative to Zn (∿ 700 ppm), supporting the suggestion of relatively high Cu proportions in the metal discharge to the harbor (Table 3).

A similar distribution of data from Tables 1 and 2 is produced if Zn is plotted against Pb (Figure 3) rather than Cu, although the relative proportions of contaminant Pb and Zn appear more similar in both recent harbor sediments and those upstream of the harbor than was suggested for Cu and Zn. Approximate concentrations for Zn, Cu and Pb in the three "end member" types of fine grained Hudson sediments indicated on Figures 2 and 3 are listed in Table 5. The recent harbor sediments do not have metal contents which tend to cluster around a small concentration range and thus and end member composition for recent harbor sediments is more artificial than for preindustrial or for recent sediments upstream of the harbor, but a representative high concentration sample is included for comparison purposes.

Heavy metal data have been used to delineate the extent of spreading of wastes such as sewage sludge and dredge spoils dumped in the coastal area about 15 km outside of the mouth of the Hudson Estuary (Gross et al., 1971; Gross, 1972; Carmody et al., 1973). The sensitivity of such tracers is relatively high because the dumped wastes are rich in heavy metals, and because most of the coastal sediments off New York City are sandy and contain very low heavy metal concentrations. Three of the coastal sediment samples reported here (Table 3) from the vicinity of the shelf break (mp -136, mp -147 and mp -157) have Zn and Cu concentrations similar to preindustrial Hudson sediments (Figure 2). These samples are very fine grained and are from an area far enough off shore to be reasonably free of much recent metal contamination. The metal data for the other three shelf samples in Table 3 (mp -38, mp -67 and mp -117) are more difficult to interpret. All three samples are relatively low in Zn, Cu and Pb but all three have relatively high proportions of sand (> 63 μ) which would tend to dilute the metal concentrations. Conceptually, it is logical to "correct" metal concentration data in samples with a high percentage of sand to provide a more representative comparison with fine-grained sediments and thus to perhaps obtain an indication of the source of the metals in a particular sample of sandy sediment. The most satisfactory procedure for developing such a correction is not clear, however. A number of possibilities are reasonable to suggest: (1) assume the metals in sediments are primarily bound in organic phases, and multiply the observed concentration of metals in sandy sediments by the ratio of organic content in fine grained sediments to that of organic matter in the sandy sediments; (2) assume that particles below a certain size (such as 63 μ) are the only ones important in binding heavy metals (de Groot et al., 1976), and multiply metal data from sandy sediments by the ratio of the proportion of sample weight less than 63 μ particles in typical fine grained sediments to that in the sandy sediments; (3) assume quartz or other metal poor phase is not significant in binding metals and calculate metal concentrations on a "quartz free" basis (Thomas, 1972; Thomas, 1973). There are many other possible approaches, such as normalizing to a constant surface area, but no single procedure appears conceptually superior.

We have tried several normalization procedures for metal data on the three sandy shelf samples mentioned above (Table 6). All of the procedures increase the observed metal concentrations but the amount of increase varied from one approach to another. The "corrected" values based on weight loss on ignition (LIG), weight loss on acid leaching (LOL) and the Fe content of sediments were all reasonably similar, and the "corrected" values are plotted in Figure 5, which is an expanded version of the preindustrial sediment portion of Figure 3. Two of the sandy shelf samples (mp -67 and mp -117) appear to be relatively free of recent metal contamination, whereas the shelf sample (mp -38) closest to the estuary mouth and to the area of dredge spoil and sewage sludge discharge (∿ mp -25) appears to have a significant proportion of recent contamination. The "corrections" suggested are obviously crude and simple minded, but are relatively convenient to apply from observational data on sediment properties, and suggest reasonable conclusions on the basis of the few shelf samples discussed here.

A previous study of the distribution of cation exchange capacity, and organic material in Hudson sediments (McCrone, 1967) indicated that the estuarine silts of the Hudson had relatively uniform properties (when compared with variations found on the adjacent continental shelf) over an extended reach upstream of ∿ mp 22. We have not tried to apply any normalization scheme to metal data from the estuary sediments reported in Tables 1 and 2. Some variations in grain size proportions and organic content do occur within the estuary samples, as indicated by the reported values for LIG and LOL. These variations probably contribute to some of the spread in Zn, Cu and Pb concentrations observed in Figures 2 and 3.

Concentrations of Fe are relatively constant in Hudson sediments, but Mn has considerable variation. Except for the surface sample at mp 24, which is really more of a marsh soil than an estuarine sediment sample, the range of Mn to Fe ratios is 0.008 to 0.067. Two generalizations about the Mn data are suggested: (1) at locations with significant variation down the core, the highest values of Mn/Fe are usually near the surface; (2) lower values of Mn/Fe especially in the upper sediment layers are more common in the harbor and shelf sediments, compared with sites in the Hudson where fresh or low salinity waters are found (mp 40-60). Manganese is quite reactive in the transition zone between river water and sea water, and a number of studies of Mn in estuaries have been made (Lowman _et_ _al_., 1966; Windom _et_ _al_., 1971; Evans and Cutshall, 1973; Graham _et_ _al_., 1976; Evans _et_ _al_., 1977). A recent study in Narragansett Bay (Graham _et_ _al_., 1976) which summarized the behavior of Mn in estuarine environments, suggested that desorption of Mn from particles at low salinities was followed by removal onto particles at higher salinities of dissolved manganese, probably as Mn (II) was slowly oxidized to Mn (IV). The time constant suggested for Mn oxidation at salinities of ∿ two-thirds of sea water was of the order of 2 days for Narragansett Bay, whereas the desorption at low salinity was very rapid. The sediment data in the Hudson suggest that desorption of Mn from suspended particles, and accumulation of these particles in New York harbor is one of the major processes affecting the behavior of Mn in this estuary. A similar conclusion has been suggested by Klinkhammer (1977) on the basis of an extensive survey including both dissolved and suspended Mn phases in the Hudson.

Precipitation of Mn within the Hudson Estuary does not appear to be nearly as important as in Narragansett Bay. If Mn precipitation is primarily the result of slow oxidation of Mn (II), the short residence time of dissolved components in the Hudson during high river discharge (∿ 5 days), and heavy sewage loading from the New York City area (∿ 100 m^3/sec) may explain the lack of evidence for this process within the Hudson Estuary. The mobility of Mn (II) in reducing sediments is well known (Lynn and Bonatti, 1965; Li et al., 1969; Bender, 1971, Elderfield, 1976) and can lead to high Mn concentrations in the upper layers of sediment. We have no direct evidence that upward diffusion process in the sediments of reduced Mn followed by precipitation of oxidized Mn near the sediment-water interface is responsible for the tendency for some cores in the fresh water and low salinity reach of the Hudson to have higher Mn near the sediment surface, but such a process does provide a plausible explanation for the observed data.

SUMMARY AND CONCLUSIONS

Concentrations of Zn, Cu and Pb in Hudson sediments indicate those metals can be considered in terms of three general sources of comparable magnitude: (1) preindustrial natural sources of weathering; (2) diffuse recent contamination throughout the estuary; and (3) sewage, industrial effluent and urban runoff, reaching the harbor from the New York City area.

The preindustrial levels of Zn, Cu and Pb in Hudson sediments are comparable to those reported for average shale (Turekian and Wedepohl, 1961) and average continental crust (Taylor, 1964) except for relatively low Cu in the Hudson (Table 7). Our data for Hudson sediments is very similar to background values suggested for Ottawa River sediments (Table 7, Oliver, 1973). A previous study of sediments from an engineering boring at about mp 60, an area which is now usually fresh water (Owens et al., 1974), reported heavy metal data on seven "estuarine" silt samples which averaged as follows: Zn ∿ 80 μg/g, Mn ∿ 1.6 mg/g, Fe ∿ 4.2%. Except for Fe, where our values are ∿ 20% lower, these data are quite consistent with our values from a large number of cores throughout the range of salinity.

The distribution of ^{137}Cs in Hudson sediments corresponds well with that of sediments with recent metal contamination, and provides a good indication of the pattern of pollutant metal accumulation. The depth to which pollutant metals are found in Hudson sediments is usually very similar to the depth distribution of ^{137}Cs.

The metal concentration of shelf sediments consisting of appreciable fractions of sand-sized particles can be compared with fine-grained estuarine sediments by normalizing the sandy sediment metal data on the basis of empirical measurements of sediment properties. Similar conclusions about the amount of recent metal contamination in sandy shelf sediments were reached using several normalization parameters including the fraction of weight loss upon heating from 105°C to 500°C, the fraction of weight loss due to leaching with strong oxidizong acids, and the Fe concentration in the sediment.

The amount of Fe in fine-grained Hudson estuary sediments is reasonably constant, but the concentration of Mn varies over approximately an order of magnitude, with greater values near the sediment surface in low salinity and fresh water areas and lesser values deep in cores and in the higher salinity zones such as New York harbor.

The harbor sediments are relatively enriched in Cu, and a first order budget for Cu released to the harbor can be estimated. Assuming a mean concentration of 250 ppm Cu in harbor sediments, and 100 ppm Cu for recent fine grained sediments delivered to the harbor from upstream, the net increase is 150 ppm Cu. Approximately 4 x 10^6 tons per year (dry weight) of dredge spoils are discharged in the coastal water off New York City (Gross, 1972). We do not have any direct information on the average metal composition of the dredge spoils which would be significantly influenced by the proportion of the total which was sandy sediment removed from areas such as lower New York Bay. Most of the dredge spoils do appear to be derived from New York harbor in areas of fine particle deposition (Panuzio, 1965). If we assume half (this estimate is probably a reasonable minimum value) of the total mass of dredge spoils consists of recent fine-grained harbor sediment, then ∿ 300 tons of Cu per year added to the harbor sediments are being removed from the harbor by dredging. Klein _et_ _al_. (1975) estimate a total delivery of ∿ 2 tons/day of Cu to New York harbor. Thus based on an assumption that at least half of the dredge spoils consist of recent harbor sediments, the current removal rate of Cu by dredging appears to be at least half of the loading rate of Cu. For Zn, since ∿ 5 tons/day are added (Klein, _et_ _al_., 1975) the fraction (∿ 15-20%) going into the harbor sediments appears considerably smaller than for Cu. A similar suggestion about the relative behavior of Zn and Cu added to New York harbor has been made by Klinkhammer (1977) on the basis of dissolved phase distributions of Zn and Cu in the Hudson.

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REFERENCES

Bender, M.L., 1971. Does upward diffusion supply the excess manganese in pelagic sediments? J. Geophys. Res., 76:4212-4215.

Bower, P.M., 1976. Cadmium and nickel in the sediments of Foundry Cove. Master's Thesis, Queens College of the City University of New York, Flushing, New York, 162 pp. with appendicies.

Bruland, K.W., Bertine, K., Koide, M., and Goldberg, E.D., 1974. History of metal pollution in Southern California coastal zone, Environmental Science and Technology, 8:425-432.

Carmody, D.J., Pearce, J.B., and Yasso, W.E., 1973. Trace metals in sediments of New York Bight. Marine Pollution Bulletin, 9:132-135.

Chow, T.J., Bruland, K., Bertine, K., Soutar, A., Koide, M., and Goldberg, E.D., 1973. Lead pollution: records in Southern California coastal sediments. Science, 181:551-552.

de Groot, A.J., Salomons, W. and Allersma, E., 1976. Processes affecting heavy metals in estuarine sediments. In: J.D. Burton and P.S. Liss (Editors), Estuarine Chemistry. Academic Press, London, pp. 131-157.

Elderfield, H., 1976. Manganese fluxes to the oceans. Marine Chemistry, 4:103-132.

Erlenkeuser, H., Suess, E., and Willkomm, H., 1974. Industrialization affects heavy metal and carbon isotope concentrations in recent Baltic Sea sediments. Geocheimica et Cosmochimica Acta, 38:823-842.

Evans, D.W., and Cutshall, N.H., 1973. Effects of ocean water on the soluble-suspended distribution of Columbia River radionuclides. In: Radioactive Contamination of the Marine Environment. International Atomic Energy Agency, Vienna, pp. 125-138.

Evans, D.W., Cutshall, N.H., Cross, F.A., and Wolfe, D.A., 1977. Manganese cycling in the Newport River estuary, North Carolina. Estuarine and Coastal Marine Science, 5:71-80.

Graham, W.F., Bender, M.L., and Klinkhammer, G.P., 1976. Manganese in Narragansett Bay. Limnology and Oceanography, 21:665-673.

Gross, M.G., 1972. Geologic aspects of waste solids and marine waste deposits, New York metropolitan region. Geological Society of America Bulletin, 83:3163-3176.

Gross, M.G., Black, J.A., Kalin, R.J., Schramel, J.R., and Smith, R.N., 1971. Survey of marine waste deposits, New York metropolitan region. Marine Science Research Center Technical Report 8, State University of New York, Stony Brook, 72 pp.

Klein, L.A., Lang, M., Nash, N., and Kirschner, S., 1975. Sources of metals in New York City waste waters, Department of Water Resources Report, City of New York.

Klinkhammer, G.P., 1977. The distribution and partitioning of some trace metals in the Hudson River Estuary. Master's Thesis, University of Rhode Island, Kingston, Rhode Island, 136 pp.

Kneip, T.J., Hernandez, T., and Re, G., 1974. Cadmium in an aquatic ecosystem: transport and distribution. In: Trace contaminants in the environment. Proceedings of the 2nd annual NSF-RANN trace contaminants conference, Asilomar, Pacific Grove, California.

Li, Y.H., Bishoff, J., and Mathieu, G., 1969. The migration of manganese in the Arctic Basin sediment. Earth and Planetary Science Letters, 7:265-270.

Lowman, F.G., Phelps, D.K., McClin, R., DeVega, V.R., DePodovani, I.O., and Garcia, R.J., 1966. Interactions of the environmental and biological factors on the distribution of trace elements in the marine environment. In: Disposal of radioactive wastes into seas, oceans, and surface waters. International Atomic Energy Agency, Vienna, pp. 249-265.

Lynn, D.C. and Bonatti, E., 1965. Mobility of manganese in diagenesis of deep sea sediments. Marine Geology, 3:457-474.

McCrone, W.S., 1967. The Hudson River Estuary: sedimentary and geochemical properties between Kingston and Haverstraw, New York. Journal of Sedimentary Petrology, 37:475-486.

Morel, F.M.M., Westall, J.C., O'Melia, C.R., and Morgan, J.J., 1975. Fate of trace metals in Los Angeles County waste water discharge. Environmental Science and Technology, 9:756-761.

Oliver, B.G., 1973. Heavy metal levels at Ottawa River sediments. Environmental Science and Technology, 7:135-137.

Olsen, C.R., Simpson, H.J., Bopp, R.F., Williams, S.C., Peng, T.H. and Deck, B.L., 1977. A geochemical analysis of the sediments and sedimentation in the Hudson estuary. Submitted to Journal of Sedimentary Petrology.

Owens, J.P., Stefansson, K., and Sirkin, L.A., 1974. Chemical, mineralogic and palynologic character of the Upper Wisconsin-Lower Holocene fill in parts of Hudson, Delaware and Chesapeake estuaries. Journal of Sedimentary Petrology, 44:390-408.

Panuzio, F.L., 1965. Lower Hudson River siltation. In: Proceedings of the Federal Interagency Sedimentation Conference. Miscellaneous Publication No. 920, Agricultural Research Service, pp. 512-520.

Simpson, H.J., Olsen, C.R., Trier, R.M., and Williams, S.C., 1976. Man-made radionuclides and sedimentation in the Hudson River Estuary. Science, 194:179-183.

Taylor, S.R., 1964. Abundance of chemical elements in the continental crust: a new table. Geochimica et Cosmochimica Acta, 28:2173-2185.

Thomas, R.L., 1972. The distribution of mercury in the sediments of Lake Ontario. Canadian Journal of Earth Sciences, 9:636-651.

Thomas, R.L., 1973. The distribution of mercury in the surficial sediments of Lake Huron. Canadian Journal of Earth Sciences, 10:194-204.

Turekian, K.K., and Wedepohl, K.H., 1961. Distribution of elements in the earth's crust. Geological Society of America Bulletin, 72:174-192.

Windom, H.L., Beck, K.C., and Smith, R., 1971. Transport of trace metals to the Atlantic Ocean by three southeastern rivers. Southeast Geology, 12:169-181.

Wrenn, M.E., Lentsch, J.W., Eisenbud, M., Laver, G.J., and Howells, G.P., 1971. Radiocesium distribution in water, sediment and biota in the Hudson River estuary from 1964 through 1970, Proceedings of the Third National Symposium on Radioecology, Oak Ridge, Tennessee.

Table 1

Hudson Estuary Sediment Composition

Location (Mile Point)	Depth (cm)	Cs-137[a] (pCi/g)	Zn (μg/g)	Cu (μg/g)	Pb (μg/g)	Mn (mg/g)	Fe (%)	LIG[b] (%)	LOL[c] (%)	K[d] (%)	Quartz[e] (%)	Fine Fraction (% < 63u)
56	0-5	0.9	190	58	66	1.70	3.3	6.6	19	2.2		
	5-10	0.0	100	26	22	2.40	3.6	7.0	20	2.4		
54	0-5	2.5	315	88	150	0.75	3.7	9.6	25	2.2	25	
	5-10	1.8	320	86	130	0.75	3.8	9.4	25	2.5	24	
	10-15	0.2	305	80	120	0.73	3.7	9.6	24	2.4	25	
	15-20	0.0	225	65	110	0.66	3.5	8.8	23	2.2	24	97
	20-25	0.0	125	31	49	0.57	3.4	7.1	24	2.4	29	98
	25-30	-	97	24	33	0.57	3.5	7.2	21		30	98
	30-35	-	90	18	25	0.55	3.5	6.8	21		31	96
	35-40	-	80	17	20	0.51	2.8	6.9	20		31	96
	40-45	-	84	18	20	0.51	3.4	6.9	21		29	96
	45-50	-	80	17	23	0.52	3.3	7.3	20		30	96
	50-55	0.0	82	17	23	0.52	3.5	7.5	21	2.2	32	97
53	0-5	2.7	290	110	175	1.15	2.4	8.4	20	2.2		
	5-10	2.1	295	97	130	1.05	2.9	7.7	18	2.2		
	10-15	0.7	245	83	105	.92	3.0	7.4	18	2.3		
	15-20	0.0	170	55	74	.77	2.7	6.3	16	2.3		
	20-25	0.0	87	18	13	.61	2.2	6.0	14	2.0		
44	0-7	0.6	125	29	45	0.53	2.3	3.5	14	1.9		
	7-14	0.0	96	27	45	0.65	2.3	4.6	15	2.1		
	14-21	-	65	26	37	0.44	1.8	3.2	10			
43	0-10	2.7	315	107	140	1.39	3.3			2.1		95
25	0-6	1.2	190	99	92	0.45	2.6	4.6	14	2.2		
	6-12	0.8	205	115	92	0.51	2.5	4.9	16	2.3		
	12-16	0.3	220	105	98	0.52	2.8	5.6	16	2.0		
	16-21	0.1	170	89	72	0.48	2.6	4.5	14	2.2		
24	0-3	( )	230	115	175	2.45	3.1	38.5	50			
	633-635	-	82	23	22	0.52	3.2	6.8	20			
	1175-1177	-	105	26	24	0.48	4.0	7.5	24			
19	0-10	( )	48	35	55	0.32	0.7	2.6	9	1.7	57	
	22-29	-	81	16	39	1.15	3.2	7.1	23	2.4	26	
	83-85	-	83	20	24	0.84	3.6	6.6	24	2.0	24	
	150-153	-	82	20	28	1.00	3.8	6.0	24	2.4	27	
	240-241	-	68	14	28	0.96	2.9	4.7	18	2.2	35	
	325-328	-	77	20	24	0.84	3.5	4.7	23	2.4	27	
	430-438	-	77	21	27	0.90	3.4	4.9	23	2.3	29	
	545-548	-	81	15	21	0.83	3.1	4.6	19	2.2	37	90

a) Cs-137 was measured by gamma spectrometry (photo peak at 0.662 MEV). Samples reported as 0.0 were analyzed and found to be free of Cs-137; those indicated with dashed lines (-) were assumed to be free of Cs-137.

(b) Loss on Ignition (LIG) indicates weight loss upon heating from 105°C to 500°C.

(c) Loss on Leaching (LOL) indicates weight loss of a sample dried at 105°C after treatment with strong acids (HCl, HNO_3, $HClO_4$).

(d) Potassium (K) was measured by gamma counting of the K^{40} peak at 1.46 MEV.

(e) Quartz was measured by monitoring the α-β transition with a differential scanning colorimeter.

Table 2

New York Harbor Sediment Composition

Location (Mile Point)	Depth (cm)	Cs-137[a] (pCi/g)	Zn (μg/g)	Cu (μg/g)	Pb (μg/g)	Mn (mg/g)	Fe (%)	LIG[b] (%)	LOL[c] (%)	K[d] (%)	Quartz[e] (%)	Fine Fraction (% < 63μ)
2	0-5	1.2	345	225	830	0.31	3.7	9.2	26	2.1		
	20-25	0.0	235	145	165	0.29	3.3	6.5	22	2.1		
	50-55	0.0	245	180	245	0.26	3.2	7.8	23	1.9		
0	0-5	0.7	260	180	140	0.26	3.3	8.5	24	2.1		$\sim$95
	18-25	1.5	215	200	200	0.30	4.0	9.8	28	2.3		$\sim$95
	45-53	0.7	225	285	175	0.33	3.3	8.8	24	2.0		$\sim$95
	60-65	0.0	53	12	< 28	0.56	3.6	4.9	23	2.0		$\sim$95
-2	0-5	0.4	337	248	202	0.55	3.3	7.5	22	1.8		
	12-20	0.6	434	344	271	0.50	3.3	9.4	25	2.1		
	25-30	0.4	459	348	253	0.60	4.2		27	2.2		
	35-40	0.6	399	294	247	0.47	3.6	9.4	23	1.9		
	45-50	0.6	472	338	286	0.53	3.5	10.0		1.9		
	55-60	0.9	557	416	345	0.46	3.7	11.0		1.9		

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a) Cs-137 was measured by gamma spectrometry (photo peak at 0.662 MEV). Samples reported as 0.0 were analyzed and found to be free of Cs-137; those indicated with dashed lines (-) were assumed to be free of Cs-137.

(b) Loss on Ignition (LIG) indicates weight loss upon heating from 105°C to 500°C.

(c) Loss on Leaching (LOL) indicates weight loss of a sample dried at 105°C after treatment with strong acids (HCl, HNO_3, $HClO_4$).

(d) Potassium (K) was measured by gamma counting of the K^{40} peak at 1.46 MEV.

(e) Quartz was measured by monitoring the α-β transition with a differential scanning calorimeter

Table 3

New York Bight Sediment Composition

Location (Mile Point)	Depth (cm)	Cs-137[a] (pCi/g)	Zn (μg/g)	Cu (μg/g)	Pb (μg/g)	Mn (mg/g)	Fe (%)	LIG[b] (%)	LOL[c] (%)	K[d] (%)	Quartz[e] (%)	Fine Fraction (% < 63μ)
Sewage Sludge		0.1	692	1440	375	0.20	1.1	72.4	84	0.7	1	
-38	0-10	0.2	110	27	67	0.18	2.1	5.0	14	1.5		33
-67	0-10	0.1	38	3	< 20	0.10	1.0	2.5	7	1.5		17
-117	0-10	0.0	17	2	< 20	0.28	0.7	1.3	6	1.3		5
-136	0-10	0.0	79	19	< 20	0.28	2.6	9.3	28	2.3		87
-147	0-10	0.0	90	25	< 20	0.30	2.6	10.2	30	2.3		98
-157	0-10	0.1	69	20	30	0.32	2.6	10.8	33	2.1		93

a) Cs-137 was measured by gamma spectrometry (photo peak at 0.662 MEV). Samples reported as 0.0 were analyzed and found to be free of Cs-137; those indicated with dashed lines (-) were assumed to be free of Cs-137.

(b) Loss on Ignition (LIG) indicates weight loss upon heating from 105°C to 500°C.

(c) Loss on Leaching (LOL) indicates weight loss of a sample dried at 105°C after treatment with strong acids (HCl, HNO_3, $HClO_4$).

(d) Potassium (K) was measured by gamma counting of the K^{40} peak at 1.46 MEV.

(e) Quartz was measured by monitoring the α-β transition with a differential scanning calorimeter.

Table 4

Comparison of Metal Concentration by Several Analytical Techniques

	Zn (μg/g)	Cu (μg/g)	Pb (μg/g)	Mn (mg/g)	Fe (%)
mp 43[a]	336	111	141	1.42	3.5
mp 43[a]	327	103	145	1.30	3.3
mp 43[a]	302	105	144	1.25	2.8
mp 43[a]	294	107	131	1.57	3.5
Average mp 43	315	107	140	1.39	3.3
2σ	(12%)	(9%)	(14%)	(18%)	(25%)
mp 43[b]	(570)	79	144	1.27	4.0
mp 43[b]	255	108	114	1.09	3.5
mp 43[c]	305	102	-	1.39	4.3

[a] Each of these samples was analyzed by the techniques used for all of the data reported in Tables 1, 2 and 3. They were analyzed on different days with groups of other samples.

[b] These samples were analyzed by a total dissolution technique using HF. Erratic Zn values were common in a number of other samples we analyzed by total dissolution.

[c] Reported analytical data for a blind determination of sample composition by a commercial instrument company using emission spectroscopy.

Table 5

Trace Metal End Members in Hudson Sediments

	Zinc (ppm)	Copper (ppm)	Lead (ppm)
1. Old Hudson Sediments	80	20	25
2. Recent Hudson Sediments	300	100	135
3. Recent Harbor Sediments	550	400	350
4. (Recent - Old) Hudson Sediments [#2 - #1]	220	80	110
5. Recent (Harbor - Hudson) Sediments [#3 - #2]	250	300	215

Table 6

Normalization Procedures for Comparing Heavy Metals in Sandy Sediments with Fine Grained Sediment

Sediment Parameter	Fine Grained Hudson and/or Shelf Sediment	Shelf Sediments		
		mp -38	mp -67	mp -117
Weight fraction $< 63\mu$	∿ 100%	33%	17%	5%
Normalization factor	1	3.0	5.5	20
Weight loss on ignition (LIG)	8%	5%	2.5%	1.3%
Normalization factor	1	1.6	3.2	6.2
Weight loss on acid leaching (LOL)	25%	14%	7%	6%
Normalization factor	1	1.8	3.6	4.2
Weight fraction iron	3.5%	2.1%	1.0%	0.7%
Normalization factor	1	1.7	3.5	5.0

Table 7

Natural Abundances of Heavy Metals

	Zn	Cu	Pb
Hudson Sediments (this study) (preindustrial)	80	20	25
Average Shale (Turekian and Wedephol, 1961)	95	45	20
Average Continental Crust (Taylor, 1964)	70	55	12
Ottawa River Sediments - Background (Oliver, 1973)	84	28	26

FIGURE CAPTIONS

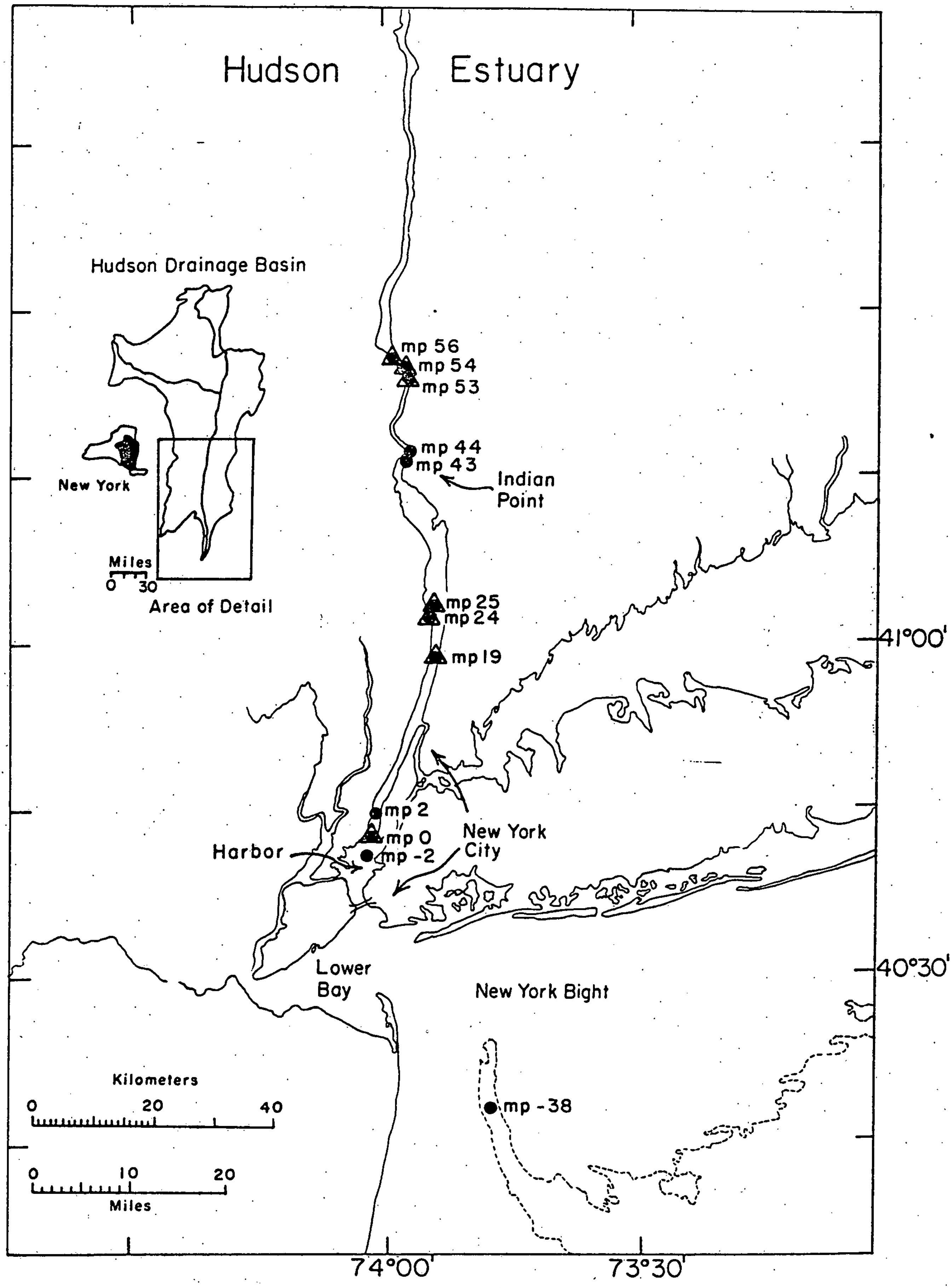
Figure 1 Location map for cores collected in the Hudson River Estuary. Sites where background levels of heavy metals were observed at some depth in the core are indicated with triangles. New York harbor is the site of greatest sediment accumulation in the Hudson. Locations in the Hudson are generally described in terms of the number of miles (mile point = mp) upstream of the southern tip of Manhattan Island. Saline water intrudes to ∿ mp 20 during high river discharge and to ∿ mp 60 during normal seasonal low discharge.

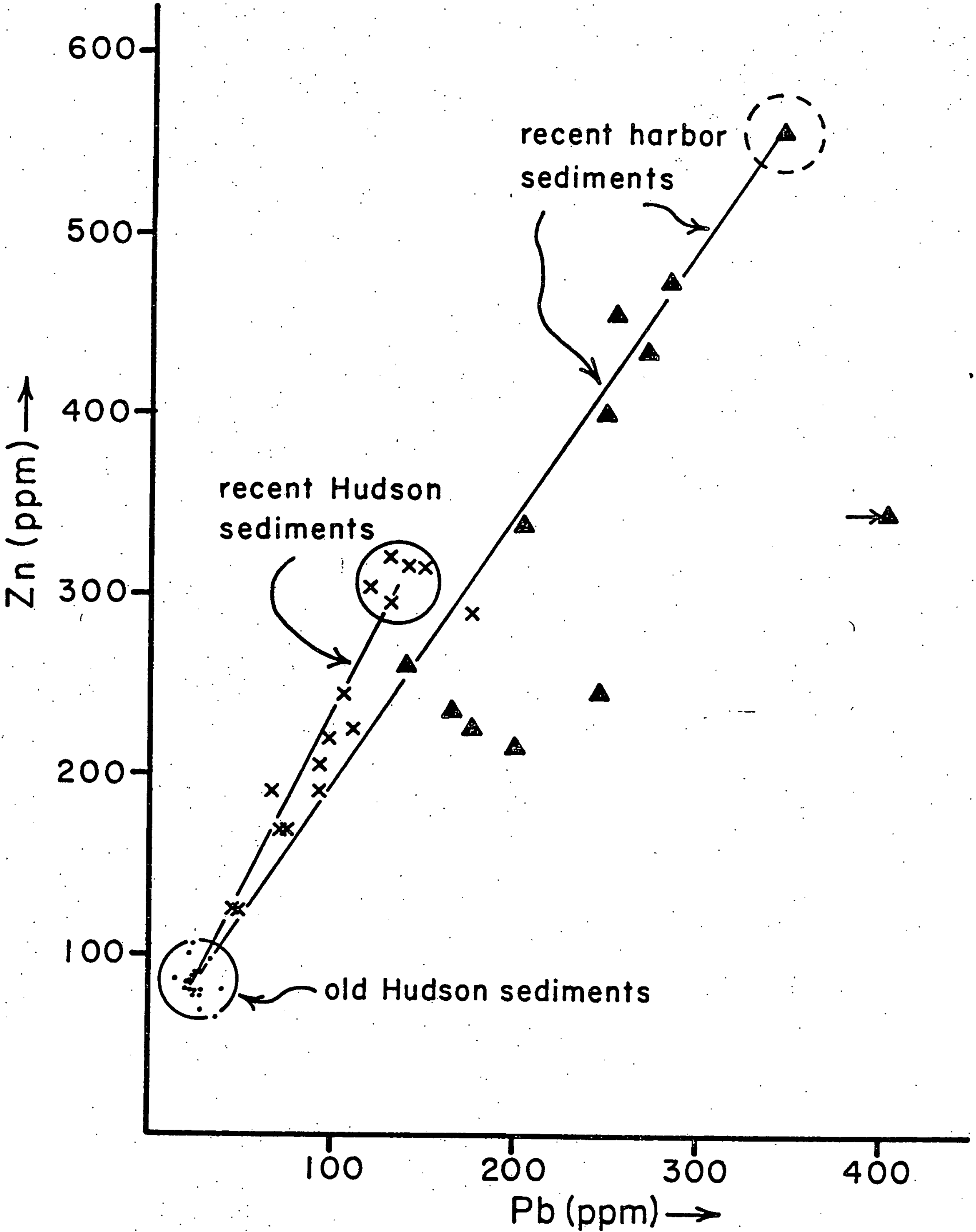
Figure 2 Location map for cores collected on the continental shelf. Sites are indicated with negative numbers, indicating the number of miles downstream from the origin of the mile point reference system. The sampling sites generally follow the narrow band of fine grained sediments in the Hudson Shelf Valley and Hudson Submarine Canyon.

Figure 3 Plot of zinc and copper concentrations in Hudson sediments, with individual samples indicated by one of four symbols. Dots indicate old sediments, free of any cesium-137 or elevated metal levels. Triangles indicate recent harbor sediments (mp 2 to mp -2) containing ^{137}Cs, and x's indicate recent estuary sediments upstream of the harbor containing ^{137}Cs. Shelf samples are indicated with S's. The estuary data can be described in terms of three groups, with the circles indicating typical end member compositions for the three sediment types. Preindustrial sediment and recent sediments upstream of the harbor cluster fairly well, but the concept of an end member composition for the harbor sediments is somewhat more artificial.

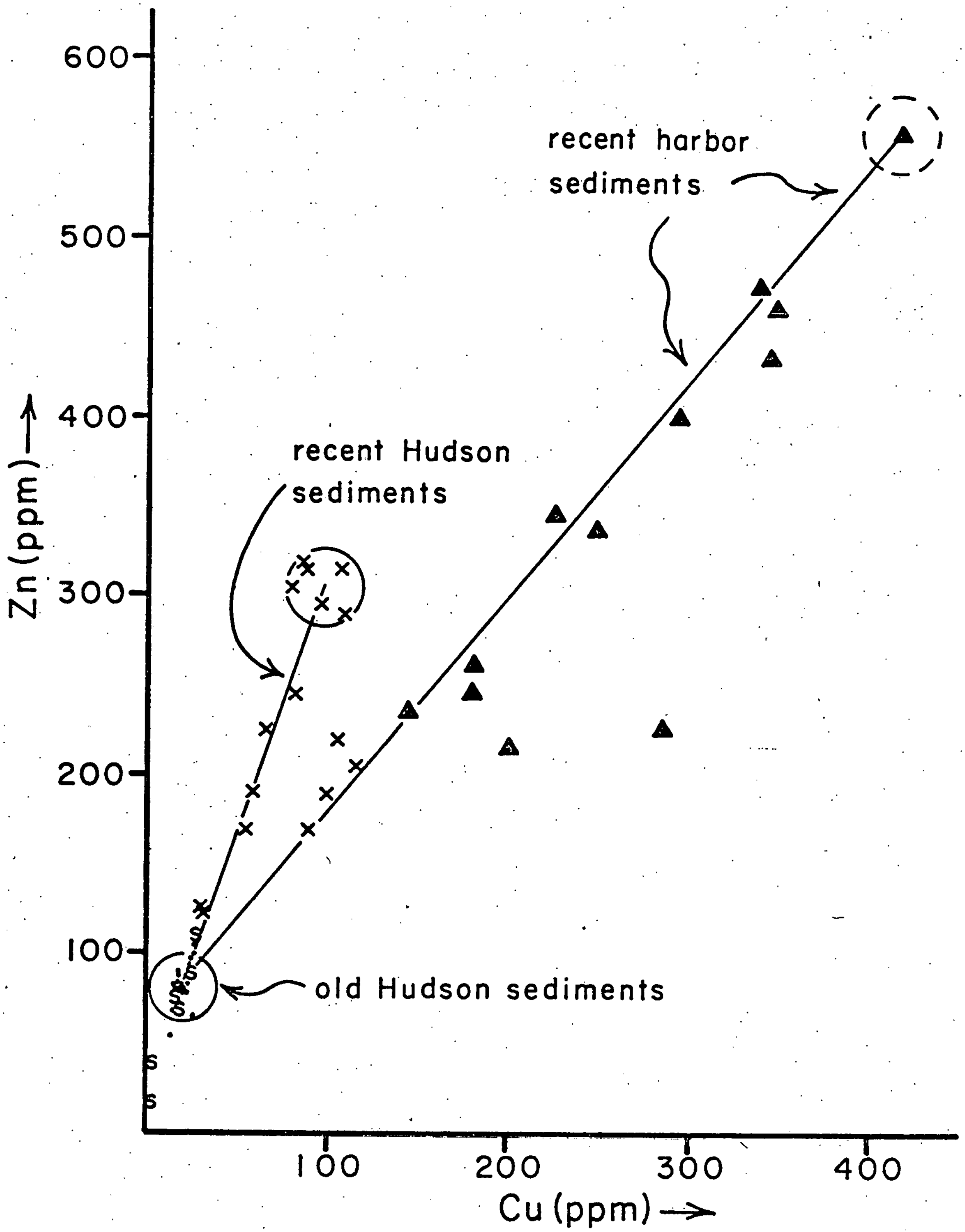
Figure 4 Plot of zinc and lead concentrations in Hudson sediments. The format and conventions are the same as for Figure 3. Lead does not seem quite as useful in distinguishing metal sources as is copper, but the sediment abundance patterns and degree of contamination are similar.

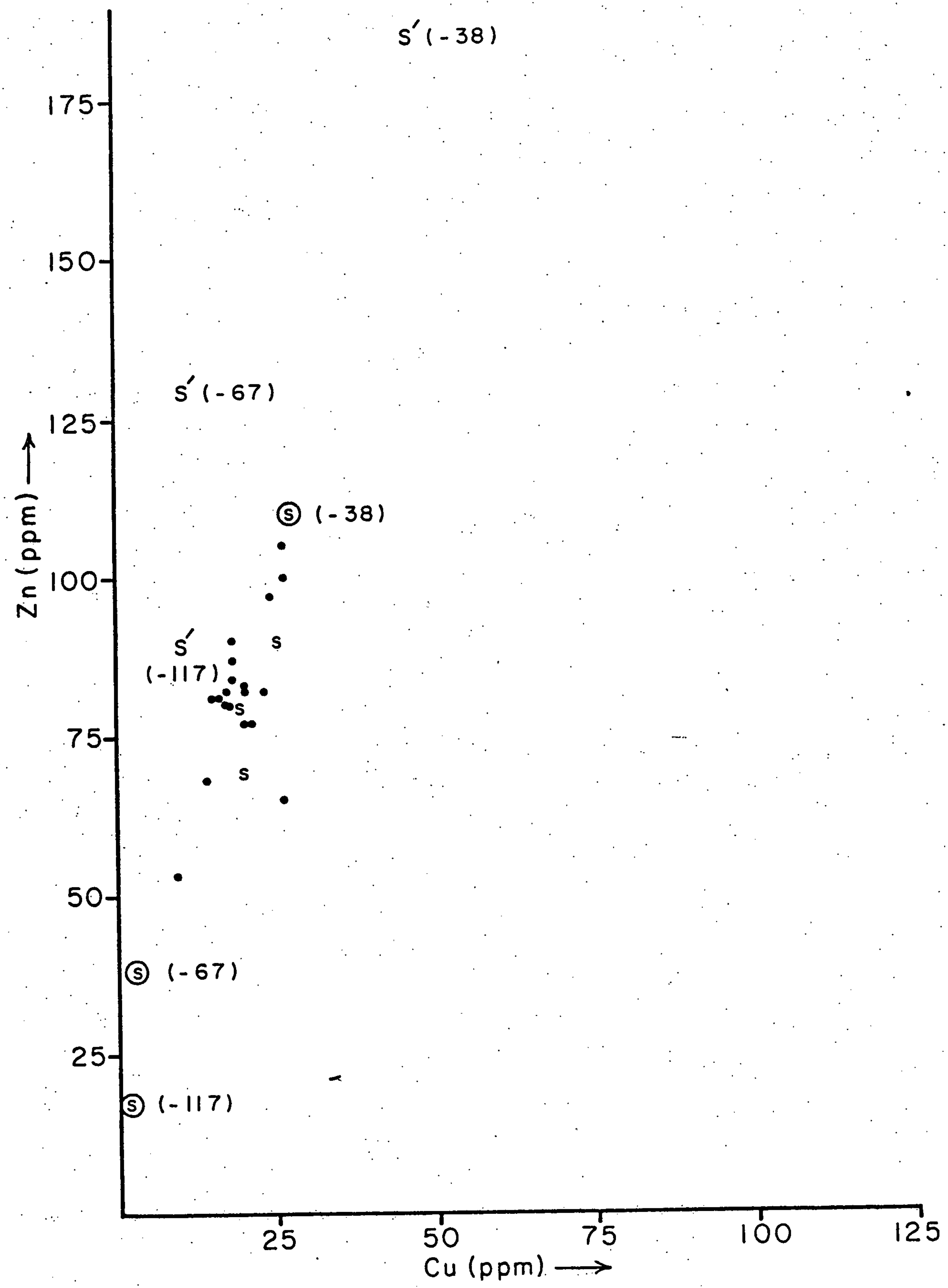
Figure 5 Expanded plot of a portion of Figure 3 to illustrate the effect of transforming observed metal concentrations in sandy shelf (mp -38, mp -67 and mp -117) by the use of normalizing factors (Table 3) for comparison with fine grained sediments. Normalized shelf sediment metal concentrations ar indicated as S' in the figure. Only one of the shelf samples (mp -38) appears to be contaminated appreciable above preindustrial heavy metal concentrations.


Hudson Estuary
Hudson Drainage Basin
New York
Miles
0 30
Area of Detail
mp 56
mp 54
mp 53
mp 44
mp 43
Indian Point
mp 25
mp 24
mp 19
41°00'
mp 2
mp 0
mp -2
Harbor
New York City
Lower Bay
New York Bight
40°30'
Kilometers
0 20 40
0 10 20
Miles
mp -38
74°00'
73°30'

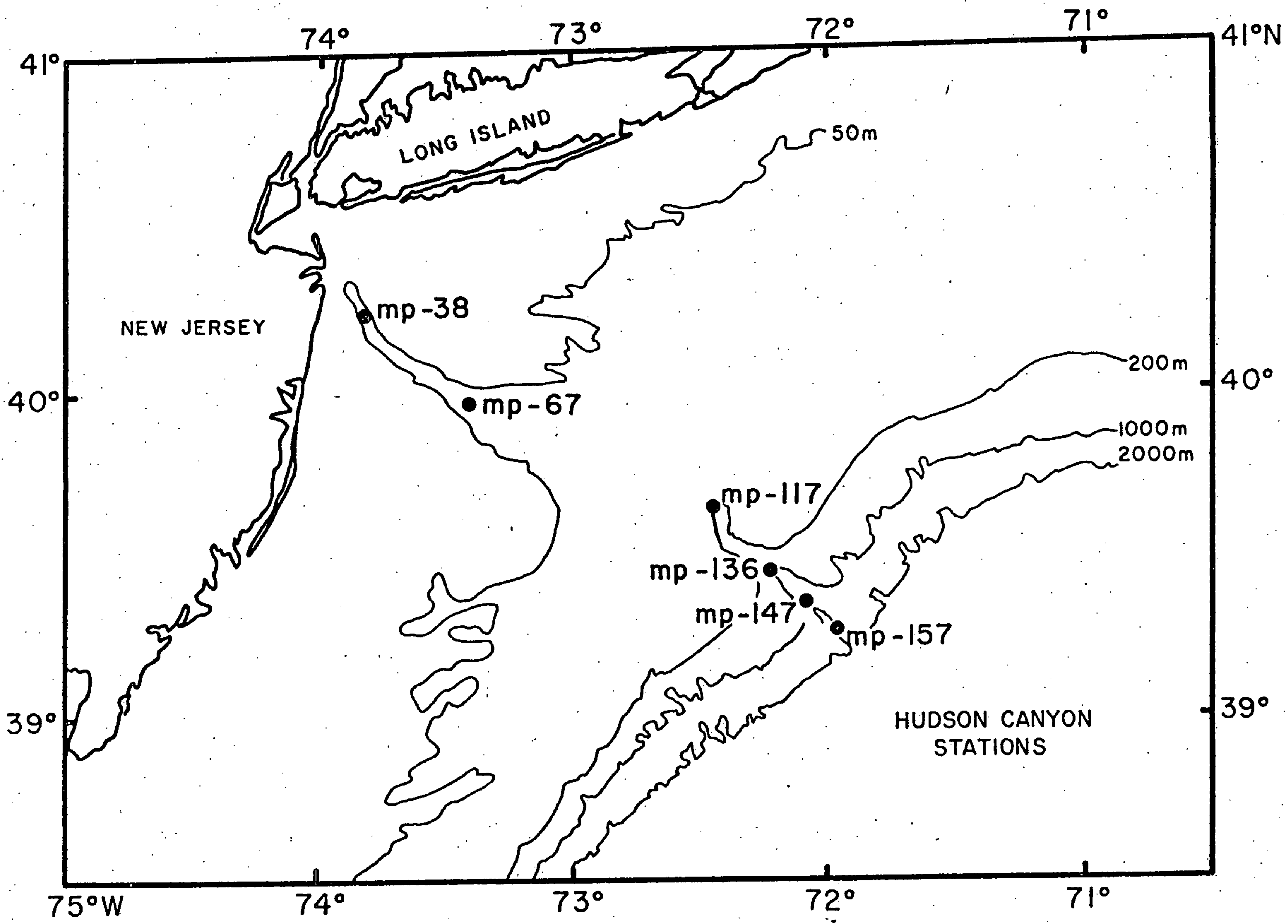


600
500
400
300
200
100
Zn (ppm) →
100
200
300
400
Pb (ppm) →
recent harbor sediments
recent Hudson sediments
old Hudson sediments


recent harbor sediments
recent Hudson sediments
old Hudson sediments
Zn (ppm) →
Cu (ppm) →
600
500
400
300
200
100
100
200
300
400



S′ (-38)
175
150
S′ (-67)
125
Zn (ppm)
Ⓢ (-38)
100
S′
(-117)
s
s
75
s
50
Ⓢ (-67)
25
Ⓢ (-117)
25
50
75
100
125
Cu (ppm)


41°
74°
73°
72°
71°
41°N
LONG ISLAND
50m
NEW JERSEY
mp-38
mp-67
40°
200m
40°
1000m
2000m
mp-117
mp-136
mp-147
mp-157
39°
39°
HUDSON CANYON
STATIONS
75°W
74°
73°
72°
71°

The Distribution of Anthropogenic Radionuclides
in Hudson Estuary Sediments

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THE DISTRIBUTION OF ANTHROPOGENIC RADIONUCLIDES IN HUDSON ESTUARY SEDIMENTS.

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Radiochemical studies have been conducted on a series of 0.5 to 3 meter cores taken over ∿ 100 km of the Hudson Estuary. Reactor and bomb-produced radionuclides (e.g., Cs-137, Cs-134, Co-60 and Pu-239,240) were found to be useful in distinguishing areas of recent sediment accumulation, from areas undergoing erosion or effected by recent dredging activities. In most areas of the estuary anthropogenic radionuclides were confined to the upper 5 to 10 cm of sediment indicating sedimentation rates < 1 cm/yr. In marginal coves measurable Cs-137 activities were frequently observed to depths of 20 cm and sometimes to 30 to 40 cm, indicating higher rates of sedimentation in shallow protected environments. In New York harbor, the vertical distribution of anthropogenic radionuclides in the sediment is much more extensive. One harbor core had measurable Cs-137 activities to 230 cm and the correlation of peak concentrations with reactor release data indicate sedimentation rates on the order of 10-20 cm/yr.

The anthropogenic radionuclide data have also been correlated with trace metal concentrations and with sediment types and structures. Recent sediment containing anthropogenic radionuclides also contains relatively high trace metal concentrations.

Cesium-137 as a Tracer for Reactive Pollutants in Estuarine Sediments

H.J. Simpson, R. Bopp, C.R. Olsen, R.M. Trier and S.C. Williams

Abstract

Many reactive pollutants discharged to natural waters become associated with fine-grained particulates. Accumulation and transport patterns of fine particles in estuaries and other natural water systems can be quite complex and difficult to predict. Cesium-137, a fission product with a 30 year half life, has been added in readily measureable quantities to natural waters throughout the globe as a result of fallout from atmospheric nuclear weapons testing. Measurement of Cs-137 in estuarine sediments can be used to rapidly establish the distribution of recent (last two decades) fine-grained sediments. In the sediments of the Hudson River Estuary (U.S.A.) the amount of Cs-137 has been found to correlate with the distribution of a wide range of reactive pollutants in terms of both depth profiles in the sediments, as well as surface sediment concentrations. The pollutants for which we have found such a covariance with Cs-137 include Pu-239,240, PCB's, Zn, Cu, Pb, Cd and Ni.

USSR - USA Symposium May 1977

E O S Trans. Amer. Geophys. Union, V. 58,p. 406

ANTHROPOGENIC RADIONUCLIDES AS TRACERS FOR RECENT SEDIMENT DEPOSITION IN THE HUDSON ESTUARY

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Cs-137, Cs-134 and Co-60 are anthropogenic radionuclides with half-lives of approximately 30, 2 and 5 years respectively. They have been introduced into the Hudson Estuary via global fallout from nuclear weapons testing and through low-level releases from a nuclear reactor, situated ∿ 70 km upstream from the southern tip of Manhattan. These radionuclides are adsorbed to fine-grained particles and consequently provide excellent tracers for sediment transport and are useful in distinguishing areas of rapid deposition from areas undergoing erosion or affected by dredging activities.

Radiochemical studies have been conducted on a series of 0.5 to 3 meter cores taken over 100 km of the Hudson Estuary. In most areas, anthropogenic radionuclides were confined to the upper 5 to 10 cm of sediment indicating sedimentation rates <1 cm/yr. In marginal coves, however, measurable Cs-137 activities were frequently observed to depths of 20 to 30 cm, indicating higher rates of sedimentation in these areas. In New York Harbor, measurable Cs-137 activities have been observed to 250 cm and the correlation of peak concentrations with reactor release data indicate deposition rates of 10 to 20 cm/yr. Consequently, it appears that there is a large variation in the patterns and rates of sediment deposition in the Hudson Estuary and that the fine-grained material responsible for the rapid shoaling in the harbor is primarily river-borne.

E O S Trans. Amer. Geophys. Union, V. 58, p.407

PCB'S AND CS-137 IN SEDIMENTS OF THE HUDSON ESTUARY

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During the past two decades, significant quantities of both PCB's and Cs-137 have been introduced into the Hudson River. The two major sources of Cs-137 are global fallout from nuclear weapons testing and low level releases from a nuclear reactor situated ∿70 km upstream from the southern tip of Manhattan. The single largest source of PCB's in the Hudson River is the discharge from two capacitor plants located ∿60 km upstream from Albany, New York.

Cs-137 is adsorbed to fine-grained particles and has been used to trace recent sedimentation in the Hudson Estuary. PCB's are also associated with fine-grained sediments and we have found excellent correlation between the amounts of PCB's and Cs-137 in sediment samples. The correlation suggests that the relatively easy Cs-137 measurement can be used to screen large numbers of samples to determine which ones would be appropriate for the much more difficult PCB analysis.

The preliminary results of our measurements indicate that recent sediments of the Hudson Estuary are contaminated with PCB's to levels of several ppm on a dry weight basis. This represents a severe contamination problem which has already resulted in the closing of the Hudson River to commercial fishing. Areas most affected include New York Harbor and marginal areas where much of the recent sedimentation takes place. The continuous dredging of the river channel and shoaling areas in the harbor has resulted in a significant transport of PCB's to the continental shelf where the dredge spoils are dumped.