

PLUTONIUM AND CESIUM RADIONUCLIDES IN THE HUDSON RIVER ESTUARY

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ABSTRACT

We have obtained a large set of cores from the Hudson Estuary covering nearly all of the ambient salinity range. A number of core sections have been analyzed for ^{137}Cs , ^{134}Cs , ^{60}Co and ^{40}K by direct gamma counting and for $^{239,240}\text{Pu}$ and ^{238}Pu by alpha-spectrometry. Rapid accumulation, up to 20 cm/year, of sediments containing $^{239,240}\text{Pu}$, ^{137}Cs , ^{134}Cs and ^{60}Co occurs in New York Harbor. Marginal coves upstream from the harbor also serve as depositional environments. The ratio of sediment $^{239,240}\text{Pu}$ to ^{137}Cs is higher than the fallout ratio in the seaward end of New York Harbor, despite the presence of a significant component of reactor ^{137}Cs in the sediments, but lower than the range of ratios observed by others for nearshore environments with low sediment deposition rates. A substantial portion of gamma emitting fission product and activation nuclides released from the Indian Point nuclear facility have accumulated in New York Harbor, more than 60 km downstream from the release area. We have not yet established whether local transuranic releases to the Hudson have occurred.

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PLUTONIUM AND CESIUM RADIONUCLIDES IN THE HUDSON RIVER ESTUARY

H.J. Simpson and S.C. Williams

INTRODUCTION

This report summarizes our activities during the second contract year of AT(11-1)2529, and provides a comprehensive data listing of radionuclide measurements made during both the initial and second year of the contract.

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 of 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 of 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.

Using a similar approach to that for the Hudson, we have collected and analyzed a limited number of samples from the Delaware Estuary during this 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 radio cesium in the Delaware will provide useful insights to the processes controlling the distribution in a large estuary of radionuclides derived only from fallout.

Some of the conclusions of our research on the Hudson are included in two preprints attached to this report, one of which has been accepted for publication at the present time.

SAMPLE COLLECTION

We have continued our program of sediment coring in the Hudson Estuary through the second 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 year 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.

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 during this year, 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. Up to now, we have used borrowed equipment from other research programs to obtain these samples, and will have to begin to acquire our own equipment for future large volume water and suspended particle sample collection.

As mentioned in the INTRODUCTION, we have begun collecting sediment samples in the Delaware Estuary. 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 will involve the gamm 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 from the immediate vicinity of a nuclear power plant at Millstone Point, Connecticut.

RESULTS - HUDSON ESTUARY

The results of our radionuclide measurements by gamma spectrometry for the first two 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 59.7. 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 Cs¹³⁷ 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 core from the harbor which we have analyzed (P-1.5E) has substantial activities of Cs¹³⁷ 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 Cs¹³⁷ activities. These sites, located upstream of New York harbor, are apparently in zones of little net accumulation

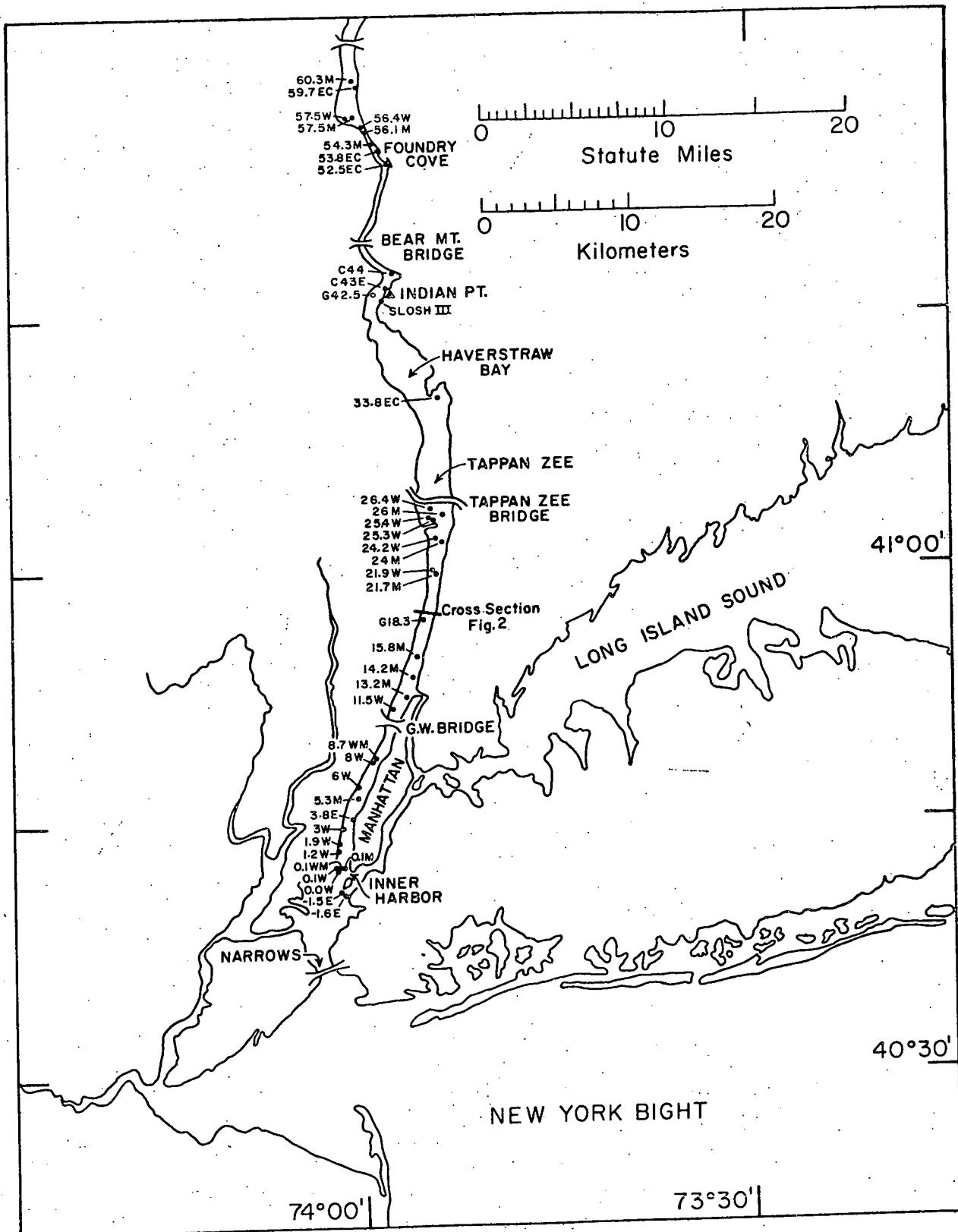


Table I
Gamma-emitting Radionuclides in Hudson Estuary Sediments

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

ACTIVITIES (a)
(pCi/kg)

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	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	⁴⁰ K $\times 10^3$
1.9 W	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	1084 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

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

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	ACTIVITIES (a) (pCi/kg)				K^{40} $\times 10^3$
				Cs ¹³⁷	Cs ¹³⁴	Co ⁶⁰		
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}	
	1025 A	0-7	112	240 ₋₁₅	25 ₋₈	65 ₋₁₁	13.2 _{-0.5}	
21.7 M	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
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}
	1032 A	0-7	128	610 ₂₁	79 ₁₉	120 ₂₁	16.1 _{0.5}
C 44	B	7-14	114	-1 ₁₁	-9 ₇	-14 ₁₅	17.4 _{0.6}
	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}
52.5 ECC	E	20-25	114	14 ₁₁	16 ₁₆	21 ₂₁	17.2 _{0.6}
	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}

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	ACTIVITIES (a) (pCi/kg)			
				Cs ¹³⁷	Cs ¹³⁴	Co ⁶⁰	K ⁴⁰ x 10 ³
	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
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

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	ACTIVITIES ^(a) (pCi/kg)			
				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
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

Location (Mile Point)	Sample #	Depth (cm)	Dry Weight (gm)	ACTIVITIES (a) (pCi/kg)			
				Cs ¹³⁷	Cs ¹³⁴	Co ⁶⁰	K ⁴⁰ x 10 ³
18	1088	Suspended Matter		1260+35	210+25	145+18	19.0+0.7
24	1114 A	Suspended Matter		1140+35	100+15	100+12	19.2+0.7

(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

of recent fine-grained sediment. Suspended particles (mp 18 and mp 24) from the same areas with low activities sediment have Cs¹³⁷ activities similar to the recently-deposited sediments of New York harbor. Thus the source of shoaling material in the harbor is clearly from upstream, and the particles are already labelled with Cs¹³⁷ (and Cs¹³⁴ and Co⁶⁰) long before they reach the harbor zone.

During the past year 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 the first two years of the contract are summarized in Table 2. The first year's data were collected using Pu²³⁶ as a yield tracer, but all of the second year data are based on a Pu²⁴² yield tracer. We have analyzed two different Pu²³⁶ spikes and one Pu²⁴² spike to determine their relative specific activities. The spikes cover a range of ~ 20%, when normalized to their labelled activities. All of the data in Table 2 have been reported relative to our Pu²⁴² spike, which lies between the two Pu²³⁶ spikes in normalized activity. Thus, although we currently have an absolute uncertainty on the order of 10%, due to uncertainty in spike calibrations, all of the data are based on a common yield tracer. We are continuing to work on a better absolute calibration of the Pu²⁴² spike.

We have collected several large composite surface sediment samples from the Hudson (SLOSS I, II, III, IV, V). These samples, especially SLOSS III which came from the immediate vicinity of the nuclear plant at Indian Point, are used as a laboratory internal calibration standard. We had considerable difficulty in homogenizing large sediment samples (> 10 kg) but hope to exchange them for intercalibration purposes with other laboratories. SLOSS III contains substantial amounts of Cs¹³⁴, a common constituent of power plant releases which is not present in the NBS Environmental Radioactivity standard (#4350). Tables 3 and 4 list data for samples of SLOSS III analyzed by gamma spectrometry and alpha

spectrometry at various times in our laboratory.

The general distribution of $\text{Pu}^{239,240}$ in Hudson sediments is reasonably similar to Cs^{137} . Long cores in the harbor which have layers of low $\text{Pu}^{239,240}$ between higher activity layers also have low Cs^{137} . The surface samples upstream of the harbor which are low in $\text{Pu}^{239,240}$ 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 $\text{Pu}^{239,240}$ in Hudson sediments has proven to be very successful in this estuary.

DISCUSSION - HUDSON ESTUARY

The activity of $\text{Pu}^{239,240}$ per gram of sediment varies greatly from place to place in the Hudson. The relative amounts of $\text{Pu}^{239,240}$ and Cs^{137} have much smaller variation than the absolute amount of either. Table 5 summarizes the ratios of $\text{Pu}^{239,240}$ to Cs^{137} , as well as the ratios of Cs^{134} and Co^{60} to Cs^{137} . The total reported range of $\text{Pu}^{239,240}$ to Cs^{137} ratios was 1-12% compared with the fallout ratio of 1-2%. There does appear to be a systematic downstream increase in this ratio, with the highest value (4-12%) in the two reported cores from upper New York Bay (-1.5E and -1.6E). Just upstream from the upper Bay the values range from 2-7%. Thus essentially all of the sediment samples in the harbor have higher $\text{Pu}^{239,240}$ 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 $\text{Pu}^{239,240}$ 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 $\text{Pu}^{239,240}$ from the dissolved phase (or both) must occur to some extent.

Table 2
Plutonium Isotopes in Hudson Estuary Sediments

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

Location (Mile Point)	Sample No.	Depth (cm)	Dry Wt. (gm)	% Yield	pCi/kg		
					239,240 _{Pu}	238 _{Pu}	238/239,240 _{Pu}
1 W	1066 A	0-5	37.2	48.1	24.5 _± 1.1	-	-
		B	72.3	32.7	31.4 _± 1.3	1.4 _± 0.2	0.04 _± 0.006
		C	69.9	35.0	28.1 _± 1.5	1.2 _± 0.2	0.04 _± 0.007
		D	63.5	56.5	33.6 _± 1.3	1.7 _± 0.2	0.05 _± 0.006
		E	77.6	-	-	-	-
		F	66.3	26.0	33.7 _± 1.5	1.4 _± 0.2	0.04 _± 0.006
		G	66.0	77.9	44.9 _± 1.0	2.3 _± 0.2	0.05 _± 0.004
		H	60.0	29.9	34.6 _± 1.2	-	-
		I	71.5	42.2	35.7 _± 1.2	2.2 _± 0.2	0.06 _± 0.004
		J	95.8	50.2	7.2 _± 0.4	-	-
		K	79.1	48.0	0.5 _± 0.1	N.D.	-
		L	97.8	5.0	1.6 _± 0.5	-	-
1 WM	1002 A	0-5	73.8	50.5	32.7 _± 2.4	-	-
		B	74.9	28.5	31.7 _± 3.2	-	-
		C	71.3	56.0	43.3 _± 3.0	2.2 _± 0.3	0.05 _± 0.008
		D	63.4	55.0	45.8 _± 3.2	2.1 _± 0.2	0.05 _± 0.006
		E	79.3	56.5	26.4 _± 1.2	1.2 _± 0.2	0.05 _± 0.009
		F	87.3	28.0	38.5 _± 1.3	1.5 _± 0.2	0.03 _± 0.004
		G	57.5	45.6	48.5 _± 1.6	-	-
1.9 W	1082 A	0-5	44.0	9.3	36.5 _± 2.6	-	-
		B	57.9	51.3	27.8 _± 1.0	1.2 _± 0.1	0.04 _± 0.006
		C	78.9	37.3	36.9 _± 1.5	-	-
		D	79.7	38.6	1.4 _± 0.2	-	-
		E	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
.6-5	1092 A	0-1	101.9	33.5	12.9+0.9	-	-
		B	108.1	43.6	12.4+0.8	-	-
		C	113.0	22.7	11.1+0.8	-	-
25.3 W	1086 A	0-6	66.3	21.8	39.4+1.7	-	-
		B	67.0	25.7	43.2+1.7	-	-
		C	72.7	32.3	12.2+0.7	0.3+0.1	0.02+0.007
		D	68.2	48.5	1.1+0.1	-	-
53.8 EC		0-5	53.1	26.2	69.2+6.4	3.0+0.6	0.04+0.009
		5-10	82.3	30.0	52.1+4.3	2.0+0.4	0.04+0.009
		10-15	87.8	38.3	5.7+0.4	-	-

Suspended Sediments

.1	1088	Surface	82.6	29.3	23.4+1.0	0.7+0.1	0.03+0.004
24.0	1114	Surface	102.8	27.6	18.9+0.9	1.0+0.2	0.05+0.01

Filtered Hudson River Water

24.0	1115	Surface	Volume liters	pCi/100 kg		
			1500	30.0	0.03+0.003	-

New York Bight Unfiltered Sea Water

Station No.

100	1102	Surface	660	12.5	0.06+0.01	-	-
104	1111	Surface	660	33.8	0.09+0.01	-	-
125	1104	Surface	660	13.5	0.11+0.01	-	-
136	1112	Surface	660	32.1	0.06+0.01	-	-
141	1107	Surface	660	17.5	0.07+0.01	-	-
148	1113	Surface	660	12.9	0.06+0.01	-	-
166	1109	Surface	660	10.4	0.09+0.01	-	-

Table 3
Laboratory Internal Standard

Sample #	Dry Weight	Gamma-Emitting Radionuclides in SLOSH III ^(a)			$K^{40} \times 10^3$
		Cs^{137}	Cs^{134} pCi/kg ^(b)	Co^{60}	
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 Q	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

- (a) SLOSS = Standard Lamont Observatory Sediment from Hudson is the surface oxidized layer of sediment removed from a series of grab samples taken in the Hudson adjacent to the Indian Point reactor at mp 43.
- (b) Sample specific activities are expressed in terms of dry weight and have been corrected for decay between time of coring and time of assay.
- (c) 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.
- (d) N.D. = Not Determined

Table 4
Laboratory Internal Standard

Nuclide Ratios in SLOSH III

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

Location (Site Point)	Sample No.	Depth (cm)	Dry Wt. (gm)	% Yield	239,240 Pu	238 Pu	pCi/kg 238/239,240 Pu
SLOSH III							
	1000 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 Q		400.2	28.2	26.5+0.5	1.1+0.1	0.04+0.004
	Pu 44		82.0	51.8	28.4+0.7	-	-
	1000 Q-2		88.5	25.3	26.0+2.0	1.5+0.2	0.06+0.009

N.D. = Not detectable

Table 5
Nuclide Ratios in Hudson Estuary Sediments

Location Site 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
-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}		134 _{Cs} x 10 ⁻²	60 _{Co} x 10 ⁻²
					137 _{Cs} x 10 ⁻²	137 _{Cs} x 10 ⁻²		
1.5	1048 A	0-10		260+21			-	-
	B	10-20		455+30			8.8+4.7	5.5+3.8
	C	20-30		345+23			-	-
	D	30-40		420+34			-	-
	E	40-50		350+26			17.1+5.3	32.5+5.7
	F	50-60		620+32			10.3+2.9	14.3+2.8
	G	60-70		1090+44			9.6+2.2	11.0+1.8
	H	70-80		950+36			14.5+2.4	10.0+1.6
	I	80-90		1080+42			14.0+2.2	9.7+1.4
	J	90-100		815+42			13.9+3.0	7.0+2.5
	K	100-110		635+33			8.5+2.9	9.1+2.7
	L	110-120		495+30			12.9+3.9	-
	M	120-130		290+18			12.4+4.2	-
	N	130-140		340+25			12.9+4.5	5.9+4.2
	O	140-150		350+18			-	6.9+2.9
	P	150-160		365+28			-	5.5+4.7
	Q	160-170		350+30			-	6.0+4.9
	R	170-180		395+21			-	6.0+3.0
	S	180-190		350+20			-	4.3+3.2
	T	190-200		200+22			-	9.0+7.0
	U	200-210		470+25			-	3.8+2.5
	V	210-220		425+27			-	6.1+3.3
	W	220-230		390+22			-	-
	X	230-240		335+18			-	8.3+3.0
	Y	240-250		500+27			-	5.6+2.8
	Z	250-260		46+11			-	-

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

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
	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		0-5		49+13		40.0+35.6	69.4+35.7
		5-10		< bkg		-	-
13.2 M		0-5		22+10		-	-
		5-10		< bkg		-	-
		10-15		6+11		-	-

Location (Mile Point)	Sample No.	Depth (cm)	239,240 Pu pCi/kg	137 Cs pCi/kg	239,240 Pu $\frac{137\text{Cs}}{\text{Cs}} \times 10^{-2}$	134 Cs $\frac{137\text{Cs}}{\text{Cs}} \times 10^{-2}$	60 Co $\frac{137\text{Cs}}{\text{Cs}} \times 10^{-2}$
14.2 M		0-5		155+33		61.0+30.8	29.9+21.8
18.6-3 M		0-5		150+13		24.0+8.3	33.0+11.6
		5-10		120+21		-	-
		10-15		8+20		-	-
18.6-5		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		0-7		240+15		10.4+3.4	27.1+4.9
21.9 W		0-5		450+50		-	27.3+9.4
		5-10		100+13		-	24.0+12.4
		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		0-5		285+19		24.9+7.9	30.5+6.3
		5-10		30+21		-	-

Location (Mile Point)	Sample No.	Depth (cm)	$^{239,240}\text{Pu}$ pCi/kg	^{137}Cs pCi/kg	$\frac{^{239,240}\text{Pu}}{^{137}\text{Cs}} \times 10^{-2}$	^{134}Cs $\frac{^{137}\text{Cs}}{^{137}\text{Cs}} \times 10^{-2}$	^{60}Co $\frac{^{137}\text{Cs}}{^{137}\text{Cs}} \times 10^{-2}$
	SLOSS III	Oxidized layer	~ 26	2700+72	~ 1	12.8+1.6	14.8+1.1
44	SLOSS IV	0-5		520+35		21.7+17.2	9.6+3.7
C 44		0-7		610+21		13.0+3.2	19.7+3.5
		7-14		-1+11		-	-
52.5 EC		0-5		2700+60		13.9+1.7	8.7+0.8
		5-10		2050+50		2.1+1.1	8.5+1.2
		10-15		735+30		-	4.6+3.5
		15-20		17+12		-	-
		20-25		14+11		-	-
53.8 EC		0-5	69.2+6.4	2475+63	2.8+0.3	4.0+1.1	2.8+1.1
		5-10	52.1+4.3	1825+68	2.9+0.3	-	-
		10-15	5.7+0.4	210+17	2.7+0.3	-	-
		15-20		26+17		-	-
		20-25		35+23		-	-
		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

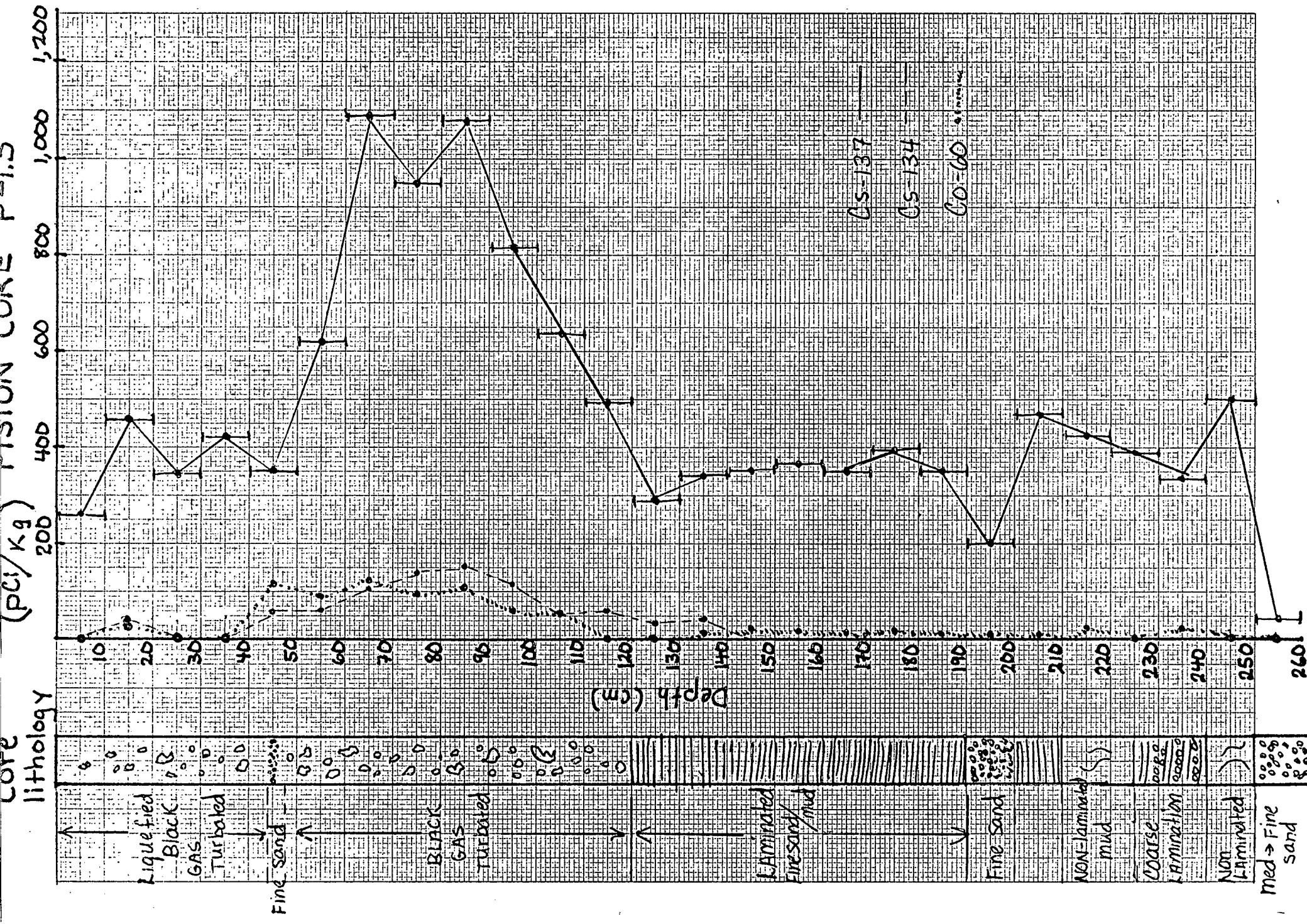
Location (Mile Point)	Sample No.	Depth (cm)	$^{239,240}\text{Pu}$ pCi/kg	^{137}Cs pCi/kg	$\frac{^{239,240}\text{Pu}}{^{137}\text{Cs}}$ $\times 10^{-2}$	^{134}Cs $\frac{^{137}\text{Cs}}{^{137}\text{Cs}}$ $\times 10^{-2}$	^{60}Co $\frac{^{137}\text{Cs}}{^{137}\text{Cs}}$ $\times 10^{-2}$
	FC 2			71+44			
	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+83		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		0-5		930+36			6.2+2.2
		5-10		15+15			
57.5 M		0-5		0+7			
59.7 EC		0-5		1870+52		2.2+1.4	0.6+0.6
		5-10		160+22			
		10-15		16+10			

Location (file Point)	Sample No.	Depth (cm)	$^{239,240}\text{Pu}$ pCi/kg	^{137}Cs pCi/kg	$\frac{^{239,240}\text{Pu}}{^{137}\text{Cs}}$ $\times 10^{-2}$	^{134}Cs $\frac{^{137}\text{Cs}}{^{137}\text{Cs}}$ $\times 10^{-2}$	^{60}Co $\frac{^{137}\text{Cs}}{^{137}\text{Cs}}$ $\times 10^{-2}$
18	1088	Suspended	23.4 ± 1.0	1260 ± 35	1.9 ± 0.1	16.7 ± 2.0	11.5 ± 1.9
24	1114	Suspended	18.9 ± 0.9	1140 ± 35	1.7 ± 0.1	8.8 ± 1.3	8.8 ± 1.1

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

Suspended particle samples collected during two recent periods from well upstream of the harbor have $\text{Pu}^{239,240}$ to Cs^{137} ratios approximately those of fallout, with absolute activities similar to sediments now accumulating opposite Manhattan (mp 0 to mp 6).

We have recently completed gamma counting of a piston core from the upper Bay (P-1.5E) which appears to penetrate through a zone of sediments containing a substantial fraction of reactor Cs^{137} into layers which contain only fallout Cs^{137} . We should be able to unravel the time history of $\text{Pu}^{239,240}$ deposition in Hudson sediments when analysis of that core is completed (Figure 2).



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°/‰. The upstream end of the series was ~ mp 78, near Wilmington, Delaware. This area of the estuary has salinities which range from about 5°/‰ 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 above. Results for about 30 samples are given in Table 6. Data for Co⁶⁰ are reported, although there is little likelihood of the presence of any measurable amounts of this nuclide in the sediments, to provide an indication of the uncertainty of the Co⁶⁰ data reported in Table 1 for the Hudson. Activities of Cs¹³⁷ 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 Cs¹³⁷ 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 Cs¹³⁷ 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 7). In general, samples with low Cs¹³⁷ also had low Pu^{239,240}, as was observed for the Hudson. The highest activity sample measured was ~ 16 pCi/kg of Pu^{239,249}, less than one quarter of the highest activity samples we reported for the Hudson.

Table 6

Gamma-Emitting Radionuclides in Delaware Estuary Sediments

Location	Sample #	Depth	Dry Weight	Cs ¹³⁷	Co ⁶⁰ (pCi/kg) ^(a)	K ⁴⁰ x 10 ³
D14-1 (mp ~ 10)	1154 A*	0-5	129	40 ⁺⁸ ^(b)	1 ⁺⁷	12.0 ^{+0.4}
	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 (mp ~ 36)	1151 A	0-5	78	165 ⁺¹⁵	12 ⁺¹³	14.8 ^{+0.5}
	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 (mp ~ 47)	1150 A	0-5	68	37 ⁺¹⁴	15 ⁺¹³	20.2 ^{+0.7}
	B	5-10	72	-7 ⁺¹²	-4 ⁺¹³	17.8 ^{+0.6}
D2-3 (mp ~ 50)	1149 A	0-5	62	335 ⁺²¹	23 ⁺¹⁵	17.0 ^{+0.6}
	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 (mp ~ 59)	1144 A	0-5	68	200 ⁺¹⁷	1 ⁺¹³	16.0 ^{+0.5}
	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 (mp ~ 60)	1142 A	0-5	100	32 ⁺¹⁰	25 ⁺¹¹	13.6 ^{+0.5}
	B	5-10	98	18 ⁺¹⁰	8 ⁺¹⁰	13.9 ^{+0.5}

Location	Sample #	Depth	Dry Weight	Cs ¹³⁷	Co ⁶⁰	K ⁴⁰ x 10 ³	pCi/kg (a)
J-2 (mp ~ 63)	1136 A	0-5	85	63+20	19+18	14.3+0.6	
	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 (mp ~ 65)	1134 A	0-5	123	110+10	11+8	12.5+0.4	
	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 (mp ~ 75)	1124 A	0-5	84	20+15	17+14	14.1+0.6	
	B	5-10	93	12+12	11+17	14.2+0.6	
B9-1 (mp ~ 78)	1120 A	0-5	93	10+18	19+18	12.6+0.6	
	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 <100 were rounded to the nearest pCi.

Table 7
Plutonium Isotopes in Delaware Estuary Sediments

Location (Mile Point)	Sample No.	Depth (cm)	Dry Wt. (gm)	% Yield	239,240 _{Pu}	238 _{Pu}	pCi/kg 238/239,240 _{Pu}
D 14-1 (~10)	1154 A	0-5	109.8	57.5	12.1 <u>±</u> 0.4	-	-
AS VII (~36)	1151 A	0-5	67.6	45.2	15.2 <u>±</u> 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 <u>±</u> 0.8	-	-
	B	5-10	70.4	58.1	16.3 <u>±</u> 0.4	-	-
	C	10-15	60.9	31.6	2.1 <u>±</u> 0.3	-	-
	D	15-20	58.5	43.7	1.4 <u>±</u> 0.2	-	-
CD 40 (~59)	1144 A	0-5	58.3	47.3	11.3 <u>±</u> 0.5	0.5 <u>±</u> 0.1	0.04 <u>±</u> 0.01
	B	5-10	52.9	11.4	9.9 <u>±</u> 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 <u>±</u> 0.3	-	-
	B	5-10	75.3	77.5	2.9 <u>±</u> 0.2	-	-
	C	10-15	62.6	42.0	0.4 <u>±</u> 0.1	-	-

N.D. = Not detectable

Table 8

Nuclide Ratios in Delaware Estuary Sediments

Location	Sample #	Depth	$^{239,240}\text{Pu}$ pCi/kg	^{137}Cs pCi/kg	$^{239,240}\text{Pu}/^{137}\text{Cs}$ $\times 10^{-2}$
D14-1 (mp ~ 10)	1154 A*	0-5	12.1 ± 0.4	40 ± 8	30.3 ± 6.1
	B*	5-10	N.A.	17 ± 7	-
	C*	10-15	N.A.	2 ± 8	-
AS VII (mp ~ 36)	1151 A	0-5	15.2 ± 0.6	165 ± 15	9.2 ± 0.9
	B	5-10	N.A.	110 ± 15	-
	C	10-15	N.A.	190 ± 20	-
	D*	15-20	N.A.	2 ± 8	-
J3-1 (mp ~ 47)	1150 A	0-5	N.D.	37 ± 14	-
	B	5-10	N.A.	-7 ± 12	-
D2-3 (mp ~ 50)	1149 A	0-5	16.5 ± 0.8	335 ± 21	4.9 ± 0.4
	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 (mp ~ 59)	1144 A	0-5	11.3 ± 0.5	200 ± 17	5.7 ± 0.5
	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	-

Discussion - Delaware Estuary

Most of the samples which had sufficient activity for confidence in reporting ratios of $\text{Pu}^{239,240}$ to Cs^{137} had values of about 5% (Table 8 - six samples). These samples were from areas with maximum seasonal salinities of $\sim 15\text{‰}$ or less and mean salinities of 10‰ or less. Two samples had appreciable higher ratios of $\text{Pu}^{239,240}$ to Cs^{137} , with the highest value of $\sim 30\%$ near the mouth of Delaware Bay. The next highest ratio was $\sim 9\%$, for a sample located between the highest ratio sample and those upstream with $\text{Pu}^{239,240}$ to Cs^{137} ratios of $\sim 5\%$.

Thus evidence for a downstream increase in $\text{Pu}^{239,240}$ 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 $\text{Pu}^{239,240}$ per unit of sediment surface area in the Delaware is equal to the fallout delivery rate of $\sim 2 \text{ mCi/km}^2$ 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 $\text{Pu}^{239,240}$ 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 $\text{Pu}^{239,240}$ 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

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

* Sandy Sediment

N.A. = Not Analyzed

N.D. = Not Detectable

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 distances 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 ~ 50). This particular site, near Artificial Island on the eastern shore of the Delaware, had the highest activity of $Pu^{239,240}$ of any of the 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 (C00-3563-47; C00-2379-10) recently completed an extensive survey of $Pu^{239,240}$ and Cs^{137} data from coastal sediments of the northwest 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 $\text{Pu}^{239,240}$ 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 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 $\text{Pu}^{239,240}$ to Cs^{137} ratios down the core. This is most likely the result of 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 long piston cores from the harbor before the depth trends of the $\text{Pu}^{239,240}$ to Cs^{137} ratio can be examined in detail in the Hudson.

Mobility of Plutonium in Aqueous Systems

One of the objectives of our research program is to help illuminate the pathways and magnitude of plutonium transport in the environment. The results discussed above relate primarily to the present distribution of fallout plutonium in Hudson and Delaware sediments. In some situations the distribution in sediments can provide valuable clues to the post-depositional migration of plutonium, as has been established by Livingston and Bowen. In nearly all shallow water aquatic systems the primary reservoir of fallout plutonium is the sediments, as in continental areas the primary reservoir is the soil zone.

One approach to understanding the mobility of plutonium which complements measurement of sediment concentrations is examination of water concentrations of plutonium in various environments. In nearly all aqueous systems except the deep ocean most of the fallout plutonium is predominantly in sediment or soil phases, but water concentrations can provide some insights into the transport rates between various compartments.

The Health and Safety Laboratory of ERDA has collected a long series of fallout nuclide measurements in New York area precipitation and New York City tap water. Since the water supply is derived from tributary streams within the Hudson drainage basin (and to a lesser extent the Delaware), the fallout nuclide data in drinking water can provide some clues about the mobility of those nuclides in the fresh waters draining into the Hudson Estuary. The fallout nuclides clearly may be affected in the transition from tributary runoff to tap water but this time series information is the best available for the Hudson area.

Data for Sr^{90} in rain and tap water have been collected since 1954, while Cs^{137} in tap water has been measured since 1966 and $\text{Pu}^{239,240}$ in tap water since 1973.

Averaged over the last two decades, the Sr^{90} content of tap water has been a small fraction of the rain water activity ($\sim 10\%$). However, as the rain water concentrations have declined from the mid-1960's peak levels to values of a few percent of the 1963 maximum, the ratio of tap water to rain water Sr^{90} contents has increased to approximately one. Thus the soils of the Hudson drainage basin appear to have reached a quasi-steady state with respect to supply and renewal of Sr^{90} . Since the annual rate of Sr^{90} fallout is now between 0.5 and 1% of the soil fallout Sr^{90} burden, a crude estimate of the residence time of soil Sr^{90} in the Hudson basin is 100-200 years.

The ratio of Cs^{137} to Sr^{90} in tap water is ~ 0.1 , more than an order of magnitude lower than the fallout ratio of 1.5. Thus the rate of release of Cs^{137} in solution from Hudson soils is still a small fraction of the current annual fallout increment (< 10%). This indicates that a first order minimum estimate of the residence time of Cs^{137} in Hudson soils is several thousand years.

The data base for $\text{Pu}^{239,240}$ in tap water is only for the period of 1973-1975, with concentrations averaging a few percent of the rain activities. Thus more than 90% of the annual increment of fallout plutonium stays in the soils, indicating that in the absence of soil particle loss from the drainage basin, the soil burden of $\text{Pu}^{239,240}$ would slowly increase as a rate of 0.5-1% per year as long as the ratio of the annual fallout increment to total soil burden remained at that level. The residence time of soil $\text{Pu}^{239,240}$ with respect to soluble phase loss would then have a minimum estimate of $\sim 10^4$ years.

The concentrations of $\text{Pu}^{239,240}$ in New York tap water during the period 1973-1975 ranged between 0.08 and 0.60 fCi/l , with a mean value of $\sim 0.3 \text{ fCi/l}$ (about 2% of the average rain activities during the same period).

During the past year we have analyzed a number of water samples in the New York Bight for plutonium under another contract (E[11-1] 2185). The range of those samples, collected during winter months, was 0.6 to 1.2 fCi/l , with the lower value closer to the average for water on the continental shelf. The tap water values of 0.3 fCi/l thus average about one half of the shelf waters. If Hudson River water were similar to tap water, then one would observe an increase of approximately a factor of 2 as fresh water passed down the Hudson and mixed into the shelf water. We have analyzed one large sample of filtered low salinity (< 5‰) Hudson water (~ 1500 liters) which had a $\text{Pu}^{239,240}$ activity of $0.3 \pm .03 \text{ fCi/l}$. This suggests that the tap water values may be reasonably good indicators of Hudson River $\text{Pu}^{239,240}$ activities during the period for which tap water data are available.

It is somewhat surprising that tap water $\text{Pu}^{239,240}$ activities are as similar to shelf ocean water values as they are (x 2). Observed concentrations of $\text{Pu}^{239,240}$ in the surface waters of the open Atlantic Ocean are approximately 1.2 fCi/l, representing only $\sim 20\%$ of the activity if all of the fallout delivered plutonium had remained in solution in the open ocean to the same extent as for Cs^{137} and Sr^{90} .

Farmer, Bowen, Noshkin and Gavini (COO-3568-6) have observed $\text{Pu}^{239,240}$ activities in Lake Ontario that are in the same range as those discussed here for the Hudson River and the New York Bight. During the period 1971 to 1973 the average $\text{Pu}^{239,240}$ activity for the entire lake declined from ~ 0.8 fCi/l to ~ 0.3 fCi/l. Thus if the values for Lake Ontario in 1973 are compared with tap water for New York City (1973-1975), Hudson River water for 1976 and New York Bight water in 1976, all of the activities are within a range of a factor of two. Considering the differences in these water types, the similarity in $\text{Pu}^{239,240}$ activities is surprising.

Most of the particulate phases in the upper levels of the sediments in these systems, or in the soil zone have average $\text{Pu}^{230,240}$ activities on the order of 20 pCi/kg (within a factor of two). If the water concentrations were determined by equilibration with the much larger reservoir of plutonium present on particulate phases, it is not unreasonable that they should have comparable activities, provided the partition coefficient between water and particles was not strongly dependent upon salinity or other dissolved phase properties. Using a partition coefficient (water/particles) of $\sim 1-3 \times 10^{-5}$, and a particle $\text{Pu}^{239,240}$ activity of ~ 20 pCi/kg, water concentrations of 0.2-0.6 fCi/l would be expected. It is difficult to understand why such a small range of partition coefficients would be compatible with waters having the range of other properties exhibited by tap water, turbid, low salinity Hudson Estuary water and New York Bight sea water.

Reports

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-cm³ 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 de-

fined 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 ⁴⁰K and daughters of ²³⁸U and ²³²Th, as well as from man-made nuclides such as ¹³⁷Cs (a fission product from bomb testing and nuclear reactor releases), ¹³¹Cs (an activation product almost exclusively from nuclear reactor operations), and ⁶⁰Co (an activation product predominantly from nuclear reactor operations).

The distribution of ¹³⁷Cs, ¹³¹Cs, and ⁶⁰Co 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 ¹³⁷Cs 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 ¹³⁷Cs, 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 ¹³⁷Cs than that in the lowest-activity example shown in Fig. 1 (labeled "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 ¹³⁷Cs 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 ¹³⁷Cs 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 km²), shallow (1 to 2 m) embayment on the east side of the Hudson River near West Point.

The third type of ¹³⁷Cs profile is illustrated by a core collected in New York harbor at MP 0.1. It shows high ¹³⁷Cs 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 ¹³⁷Cs activities from the surface to depths below 30 cm are clear indicators of rapid sediment accumulation.

There are two significant potential sources of ¹³⁷Cs 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 ¹³⁷Cs, 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, ⁹⁰Sr (4) and ¹³⁷Cs (5) are rapidly attached to particulates in the upper 10 to 20 cm of the soil. In the open ocean, fallout ⁹⁰Sr and ¹³⁷Cs 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 ⁹⁰Sr remains in solution, while a major fraction (> 90 percent) of fallout ¹³⁷Cs is now found attached to particulate phases in the sediments (7). In estuarine systems such as Chesapeake Bay, dissolved ¹³⁷Cs appears to be roughly proportional to sa-

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linity, 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 ^{131}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 ^{131}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.

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 ^{131}Cs and ^{60}Co , in proportions roughly similar to those in surface sediment samples collected near the reactor site (see Table 2: $^{131}\text{Cs}/^{137}\text{Cs}$ and $^{60}\text{Co}/^{137}\text{Cs} \sim 0.1$). The cove site approximately 15 km upstream from the release area has relatively low $^{131}\text{Cs}/^{137}\text{Cs}$ and $^{60}\text{Co}/^{137}\text{Cs}$ (~ 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/km² (11, 12). Using an area of water surface for the Hudson between MP 60 and MP 0 of 200 km², 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 ($\sim 4 \times 10^3$ curies for the total drainage basin of $\sim 3.5 \times 10^4$ km²). 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 $\sim 4 \times 10^6$ tons (dry weight) (15). Assuming that 50 percent of this mount is taken 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 $^{239,240}\text{Pu}$ (16). Concentration profiles of $^{239,240}\text{Pu}$ in the three sediment cores discussed earlier are shown in Fig. 3. Variations in the activity of $^{239,240}\text{Pu}$ 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 $^{239,240}\text{Pu}$ and ^{137}Cs . Covariance of fallout ^{137}Cs and $^{239,240}\text{Pu}$ in sediments of freshwater lakes has also been observed in the Great Lakes (7) and in soil profiles (5). In the ocean, $^{239,240}\text{Pu}$ is much more rapidly and completely associated with particulate phases than is ^{137}Cs (17).

We cannot establish budget terms for $^{239,240}\text{Pu}$ in the Hudson as well as for ^{137}Cs at the present time. The activities of $^{239,240}\text{Pu}$ present in global fallout are 1.5 to 2 percent of that of ^{137}Cs . The ratio of $^{239,240}\text{Pu}$ 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 $^{239,240}\text{Pu}$ 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 $^{239,240}\text{Pu}$ 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 $^{239,240}\text{Pu}$ 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 10⁴ 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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- Although we have observed sharp peaks of ¹³⁷Cs 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.
- Annual fallout ¹³⁷Cs delivery to the Hudson was computed from ⁸⁷Sr data for New York City (12), assuming a ¹³⁷Cs/⁸⁷Sr ratio of 1.5.
- USERDA Rep. HASL-294* (1975), Appendix.
- We estimated the drainage basin supply of ¹³⁷Cs as two components: particulate and dissolved. Assuming that all of the suspended solids in the Hudson River are soil particles with an average ¹³⁷Cs activity of 1 pCi/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 ¹³⁷Cs in 10 years is ~ 10 curies for an average river flow of 500 m³/sec and a suspended solids load of 60 parts per million. Three lines of evidence indicate that the supply of dissolved ¹³⁷Cs 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 ⁸⁷Sr and ¹³⁷Cs from both rainwater and New York City tap water (12) indicate that approximately 1 percent of the total fallout ¹³⁷Cs 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 ¹³⁷Cs occur in reservoir storage and water treatment. A second line of evidence is provided by published data on the annual average ¹³⁷Cs activity in the Hudson near the reactor site at MP 43. Activities declined from 1 to 2 pCi/liter to 0.01 to 0.1 pCi/liter between 1964 and 1968 (2), approximately following the trend in ¹³⁷Cs 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 ¹³⁷Cs in Chesapeake Bay during 1970 to 1972 (8), where freshwater and low-salinity (≤ 2 per mil) samples had activities (≤ 0.02 pCi/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 ¹³⁷Cs to the Hudson Estuary to be ~ 50 curies.
- 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.
- M. G. Gross, *Geol. Soc. Am. Bull.* 83, 3163 (1972).
- Samples were prepared by chemical procedure similar to those of K. M. Wong [*Anal. Chin. Acta* 56, 355 (1971)], using ²³⁹Pu and ²⁴⁰Pu as yield tracers and counted with an alpha-particle spectrometer. The peaks in the alpha-particle energy spectrum for ²³⁹Pu and ²⁴⁰Pu are not resolved, so data obtained by alpha spectrometry are reported as the sum for these two nuclides, ^{239,240}Pu.
- V. T. Bowen, K. M. Wong, V. E. Noshkin, *J. Mar. Res.* 29, 1 (1971); V. E. Noshkin, *Health Phys.* 22, 537 (1972).
- 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 ¹³⁷Cs 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 ¹³⁷Cs has left the soil in solution (11), then ~ 0.3 curie of ^{239,240}Pu would have reached the Hudson in solution from the drainage basin soils.
- 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)].
- 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).
- We thank J. Kelly of Consolidated Edison of New York, Inc. for providing copies of operating reports for Indian Point and the Health and Safety Laboratory of the Energy Research and Development Administration for supplying ²³²Pu and ²⁴²Pu spikes. Financial support was provided by ERDA contract E(11-1) 2529, Contribution No. 2190 from Lamont-Doherty Geological Observatory of Columbia University.

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

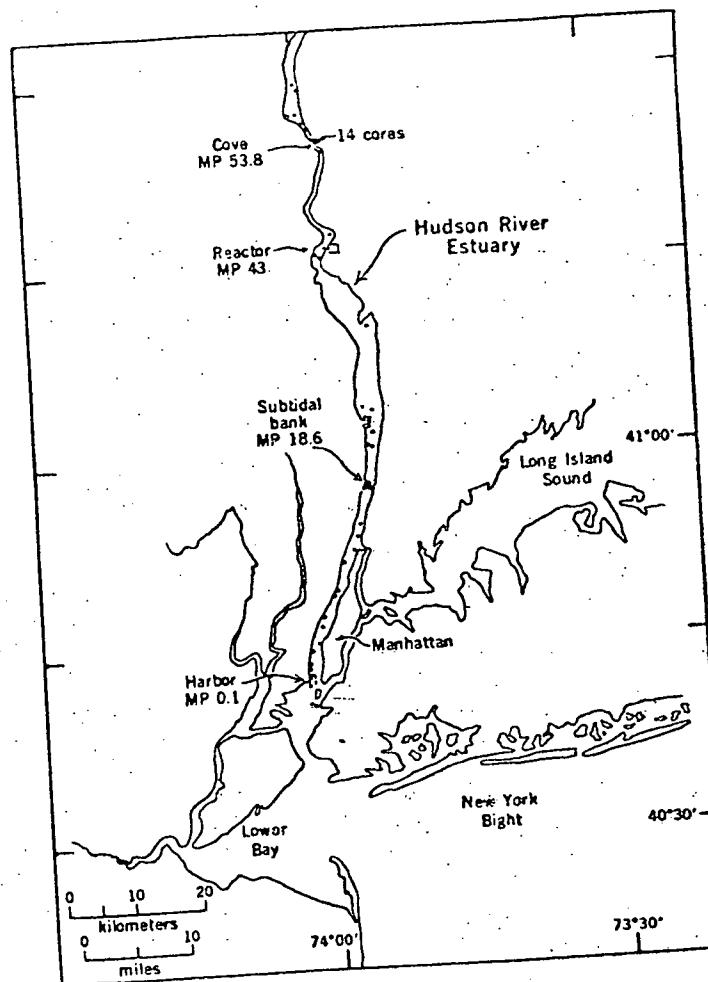
Table 3. Summary of budget estimates for ^{137}Cs and $^{239,240}\text{Pu}$ in the Hudson estuary. Values in parentheses have the largest uncertainties.

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

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 \oplus . The harbor salinity south of Manhattan ranges between 18 and 26 per mil. Saline water (≥ 0.1 per mil) intrudes upstream to \sim MP 15 during springtime high discharge of fresh water and to \sim MP 60 during average summertime conditions (20).

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 ($1 \text{ pc} = 10^{-12} \text{ curie}$). 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 comparable activity to 70 cm, while others reach background levels between 50 and 60 cm.

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



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FIG. 1

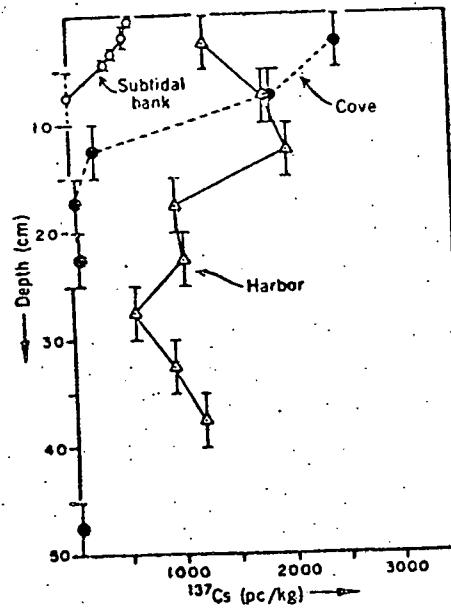
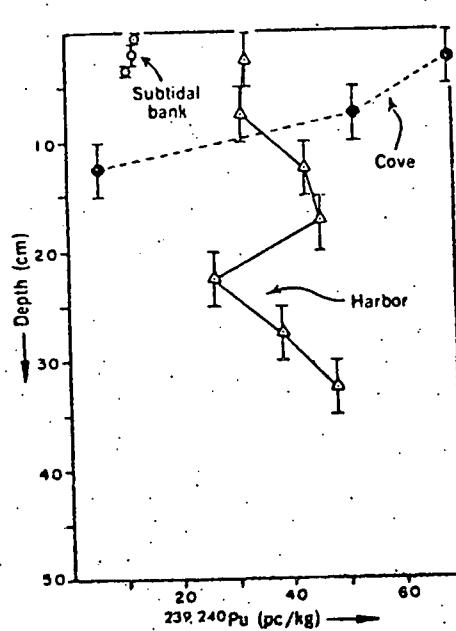
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A GEOCHEMICAL AND SEDIMENTOLOGICAL ANALYSIS OF THE SEDIMENTS
AND SEDIMENTARY STRUCTURES IN THE HUDSON ESTUARY^{1,2}

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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 150-400 mg; potassium 18-24 mg; zinc 50-350 μ g; copper 15-300 μ g; and lead 20-800 μ g. 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 sediment; but in the inner harbor area of New York they have been observed to sediment depths on the order of 60-80 cm. In some areas of the inner harbor the vertical distribution of these 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. The inner harbor sediments contain relatively high concentrations of trace metals, anthropogenic

radionuclides, and organic matter. ^{14}C analyses of this organic matter 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. Below the Tappan Zee coarse sand and 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 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. ^{14}C dating of the organic matter in the surface sediments indicates that the carbon is predominantly old, recycled carbon and not newly-fixed carbon from sewage disposal or phytoplankton activity.

One possible interpretation of the sedimentary structures that is consistent with the geochemical data is: (1) the laminated, fine-grained sediments are deposited when the Hudson transports relatively high concentrations of sediment, such as during severe storms; (2) the coarse sand-shell layers result from increased sand transport during high-energy (storm) conditions or from tidal scour under normal flow conditions; (3) the turbate zones represent periods of slower or no deposition

during normal flow conditions; and (4) the everyday tidal and estuarine processes are causing the rapid accumulation of recent "polluted" sediment in specific areas, such as in the inner harbor and in protected areas along the sides of the estuary, upstream of New York City.

INTRODUCTION

Numerous physical, chemical, and biological factors influence estuarine sedimentary processes and patterns. These include alternating tidal currents, flocculation effects, estuary morphology and its associated local hydrologic characteristics, bio-agglomeration and bioturbation, human activities, multiple sediment sources, and a non-tidal, two-layer estuarine circulation pattern. Non-tidal estuarine circulation is characterized by an upper fresh-water layer with a net seaward flow and a denser, more saline bottom layer having a net landward flow (Pritchard, 1955). Suspended matter is carried downstream in the upper layer and gradually settles into the more saline bottom layer. This settling may result from: (1) flocculation and aggregation of suspended particles due to an increase in salinity (Edzwald and O'Melia, 1975) or agglomeration by organisms (Meade, 1972), (2) variations in the current velocity associated with alternating tides, and (3) reductions in the current speed resulting from an increase in cross sectional area, isolated morphological features, such as coves, or artificial obstructions, such as piers or docks. Sedimentary

matter in the bottom layer is transported upstream, either under the influence of tidal processes (Van Straaten and Kuenen, 1958; Postma, 1967), or non-tidal estuarine flow (Meade, 1969). Consequently, estuaries act as effective sediment traps, and are sites where sediment accumulates rapidly.

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). Wide-spread deposition may also occur during slack water; however, during later tidal cycles this material may be resuspended, transported, and redeposited (Panuzio, 1965; 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 (Ball et al., 1967; Hayes, 1967; High, 1969; Reineck and Singh, 1972; Swift et al., 1972; Schubel, 1974a; Sanders and Kumar, 1975; Kumar and Sanders, in press), and several investigators have attributed deposits in the stratigraphic record to storm activity (Hobday and Reading, 1972; Ager, 1974; Kelling and Mullin, 1975).

In order to elucidate the sedimentary processes in the Hudson Estuary and to determine the character and distribution of the sediments, geochemical and sedimentological studies have been

conducted on a series of cores (Fig. 1) taken from the Narrows (mp -6) to as far upstream as Newburgh (mp 60.5). All mile point (mp) designations indicate the distance in statute miles along the channel upstream from the southern tip of Manhattan (mp 0). Particular attention has been given to a set of cores and grabs taken across the estuary at mp 18.6 (Figs. 2, 3, and 4). This area was chosen on the basis that: (1) it generally lies within the zone of two-layer estuarine circulation and undergoes large salinity variations during a tidal cycle, (2) there has been no dredging in the area as recorded by the Army Corps of Engineers, and (3) a piston core 6 m long (V32-02) had been taken in this area at the conclusion of R/V Vema Cruise 32, leg 1. An added advantage to this site is its proximity to L-DGO, allowing for rapid extrusion, examination, and analysis of the cores.

In this paper we first describe the kinds of sediments and the observed sedimentary structures for several cores taken in different areas of the estuary. Secondly, we present chronologies for several cores, including V32-02, on the basis of ^{14}C dates and the vertical distribution of ^{137}Cs , ^{134}Cs , and ^{60}Co in the sediment. Thirdly, we present trace metal data on both "polluted" and "non-polluted" sediment. Finally, we have attempted to correlate the geochemical data with the observed sediment kinds and sedimentary structures and thus to gain some understanding of sedimentary processes in the Hudson Estuary. Because estuarine organic silt has an affinity for radionuclides, hydrocarbons, and metals commonly associated

with domestic and industrial wastes, such an understanding is critical in the prediction of the dispersal and accumulation of many pollutants.

Hydrology and Geomorphology

The Hudson is one of several major estuarine systems on the northeastern Atlantic coastline of the United States and drains an area of approximately $35,000 \text{ km}^2$. For the purpose of this paper the Hudson Estuary is defined as extending from the Narrows (mp -6) to the upstream limit of the salinity intrusion ($>100 \text{ ppm Cl}$). The average fresh-water discharge is $550 \text{ m}^3/\text{sec}$, with mean monthly maxima and minima of $1200 \text{ m}^3/\text{sec}$ and $60 \text{ m}^3/\text{sec}$ (Simpson *et al.*, 1974). Three major storms in the last 75 years have caused flows of $6320 \text{ m}^3/\text{sec}$ in 1913; $6090 \text{ m}^3/\text{sec}$ in 1936; $5130 \text{ m}^3/\text{sec}$ in 1948 (U.S. Geol. Survey Water Resources Data for New York, 1973; Abood, 1974). Tidal flows average between 8000 and $11,000 \text{ m}^3/\text{sec}$, and thus completely mask the net fresh-water flow from any direct measurement. The tidal current reaches a maximum velocity of about 100 cm/sec , and tidal-stage fluctuations between 1 and 2 meters are felt as far upstream as the dam at Troy (mp 154).

The most fundamental physical parameter of any estuarine system is the salinity distribution. In addition to controlling net water motion, the salinity distribution is a critical parameter to the biological community and greatly affects the dynamics of the suspended load. The salinity distribution along the axis of the Hudson Estuary varies daily with tidal amplitude and seasonally with river flow (Pritchard *et al.*,

1962; Quirk et al., 1970; Simpson et al., 1974). During low fresh-water flow the salt intrusion varies from mp 60-80 (Abood, 1974) and during conditions of high fresh-water flow from mp 10-20.

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), a site 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.

Downstream of mp 0, the morphology is extremely complex, with numerous tidal straits, and dredged channels. Between mp 0 and mp 22, the Hudson is remarkably linear and can be classified as a partially mixed estuary, in which tidal currents go through a complete reversal at all depths, but vertical mixing is not complete (Panuzio, 1965). At the surface, the ebb velocity and the duration of the ebb cycle exceed the flood velocity and duration of the flood cycle, whereas at the bottom, the velocity and duration of the flood cycle exceed those of the ebb (Abood, 1974).

The regional geology and major morphological features in the vicinity of the Hudson Estuary have been summarized by

Berkey (1933), Schuberth (1968), and Sanders (1974). The Pleistocene stratigraphy, paleontology, and paleoecology of the Hudson Estuary have been discussed by McCrone and Schaffer (1966), Newman et al. (1969), Sanders (1974), and Weiss (1974). Saline estuarine conditions were established approximately 11,500 years ago (Weiss, 1974) based upon the appearance of foraminifers in the sedimentary record. Coring and geophysical profiling at Nyack (mp 24) indicate that the estuarine layer of organic silt ranges between 10 and 60 m thick (Worzel and Drake, 1959). If this layer were deposited during the last 11,500 years, the resulting sedimentation rate would be on the order of 1-5 mm/yr.

PROCEDURES

A 22-foot pontoon boat equipped with a small hydrographic winch has been used extensively for shallow-water work in the Hudson Estuary. Surface grab samples were taken with a shipek 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 Alpine piston corer, having a diameter of 3 cm, 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. The liners were kept in a vertical position, and the overlying water was usually drawn off with a syringe to prevent the surface sediment from sloshing around inside the liner. Upon returning to the lab, we immediately

extruded, split and air dried the cores. After drying for 3 to 7 days, lamination in the organic silts became apparent, resulting from the preferential drying of the fine sandy silt layers relative to the clay-rich layers.

Samples of estuary sediments were examined under a scanning electron microscope (SEM), and the elemental composition of individual particles was determined with an Energy-Dispersive X-ray Fluorescence analyzer (EDXRF). Approximately 300 particles were randomly examined in both the sand-size and silt-size range at mp 18.6 to determine the representative mineral composition. Percent carbonate was determined by adding dilute HCl to the sediment and measuring the volume of CO_2 emitted.

Subsurface shell layers were removed from several of the channel cores and analyzed for ^{14}C . To obtain enough carbonate (5-20 g) to date the shell layers at 21-23 cm and 50 cm in gravity core 18.6-1M (Fig. 3), several cores were taken at the same anchored station, and the shell layers were combined. Extreme care was taken to combine only the layers from cores which displayed identical stratigraphic relationships. ^{14}C analyses were also completed on the organic matter in the surface sediment at three different locations.

The activity of ^{137}Cs , ^{60}Co , ^{134}Cs , 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 with a Ge(Li) detector, and the spectral information was obtained with an 8000-channel analyzer. The sediment samples were air dried, broken up with a porcelain mortar and pestle,

and large debris such as rock fragments, shells, slag, and coal were removed. The samples were sealed in 100-cc aluminum cans or 6.5-cc plastic petri dishes to maintain a constant counting geometry. The samples were generally counted for 8-16 hours, and calibration was based on an environmental radioactivity standard (#4350) supplied by the National Bureau of Standards. Statistical errors have been reported in terms of 1 sigma.

Quantitative analyses for copper, zinc, and lead have also been determined as a function of depth in several cores. The metal concentrations were measured by atomic absorption, and the metal-extraction procedures have been described by Williams et al. (in preparation).

SEDIMENTS

The surface sediments in the Hudson Estuary frequently contain oil coatings, coal, tar, bricks, fly ash, and metaliferous slags. This anthropogenic detritus is generally confined to the top 10 cm, but may be as deep as 80 cm depending on the rate of deposition and the extent of surface sediment reworking.

The fine-grained sediments between mp 0 and mp 60 generally contain less than 3% carbonate and between 3 and 10% organic matter as determined from the weight loss-on-ignition at 550°C. This agrees reasonably well with the 5-6% oxidizable organic matter reported by McCrone (1967) in the channel sediments between mp 76 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. Loss-on-ignition values may exceed the amount of organic matter due to the partial decomposition of some minerals when heated to 550°C (Gross *et al.*, 1971); but the range of 3-10% organic matter in the fine-grained sediments of the Hudson is probably valid.

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 mineral composition of the sand- and silt-size fractions based on our data is given in Table 1. The percent SiO_2 has been determined on the basis of relative number using the SEM and EDXRF system (Table 1), and on a dry weight percent basis using differential thermal analysis (Table 4). The aluminosilicates were classified on the basis of the cations present, Al/Si ratios, and morphology. The heavy minerals consisted of amphiboles, pyroxenes, epidote, garnet, sphene, and tourmaline; the lack of magnetite is notable. It is also evident from the data presented in Table 1 that there is a greater percentage of feldspar relative to quartz in the silt-size range, probably a result of the greater potential of feldspar for physical and chemical breakdown.

Grain-size analyses were completed on surface samples taken from cores in both the channel (18.6-1M and 18.6-3M) and 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 the channel.

In the channel from mp 13 to mp 24 coarse sand and shell layers occur at the surface and at depth. These layers may consist almost entirely of coarse sand or of shell debris, but frequently they are a mixture of both, ranging from 30 to 70% carbonate material.

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 coarse material which characterized the surface channel sediments between mp 13 and mp 24 was not observed at the surface of the natural channel in the inner harbor, but layers of medium- to fine-grained sand were observed in several cores at depth.

Frequently the bottom sediments have a highly liquefied, olive-brown (oxidized) layer which lies at the sediment/water interface. The layer ranges from 0.1 to 3 cm thick and is composed of soupy, unconsolidated, fine-grained matter which may be easily eroded, transported, and redeposited during alternations of the tidal current. Similar oxidation layers have been observed in the James River Estuary (Nichols, 1972), in shallow marine bays (Oppenheimer, 1960), and in the Dutch Wadden Sea (Van Straaten, 1954). 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 monosulfides (hydrotroilite) and then to disulfides (pyrite) in the anoxic sediments below (Emery and Rittenberg, 1952; 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 hydrotroilite ($FeS \cdot nH_2O$), to which the black color has been attributed (Biggs, 1967). Several of the inner harbor cores penetrated gray sediment below 60 to 70 cm, but these cores were not long enough to establish the thickness of this gray layer or to determine if the gray color characterizes the sediments at lower depths.

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. In a few cores alternating bands of olive-gray and gray sediments were present (Fig. 7B).

Shells

The shells of the subsurface layers (channel, mp 13 to mp 24) consist almost entirely of a small clam, Mulinia lateralis, which lives in the muddy sediments of marine and brackish waters (Jacobson and Emerson, 1971). Although layer concentrates of Mulinia have been found in channel cores taken as far upstream as Piermont (mp 24), living Mulinia appear to be restricted to

the 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). The present scarcity of living Mulinia between mp 18 and mp 24 and their virtual restriction to the more saline waters is contradictory to the large number of shells concentrated in these subsurface layers. The old ¹⁴C ages of these layers (see Table 2 and section on ¹⁴C) indicate, however, a former more extensive distribution of living Mulinia in the estuarine muds north of the Battery. Their present rarity may be a result of 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, and attributed it to the rapid accumulation of silts, which may act to reduce the landward penetration of the salinity intrusion.

Several marine snail shells, Urosalpinx and Nassarius, have also been found in the layers of Mulinia. Other invertebrate shells that are frequently found in the estuarine sediments include oysters (Crassostrea virginica), clams (Macoma and Mya arenaria) and mussels (Mytilus and Mytilopsis).

SEDIMENTARY STRUCTURES

Alternating layers of fine sandy silts and clayey silts, on the order of a millimeter to a centimeter thick, characterize the fine-grained sediments in many of the cores examined (Fig. 5A). In cores taken in the channel between mp 13 and mp 24, these laminated sediments are interlayered with coarse sand and shell layers (Figs. 3 and 4), which are 1-20 cm thick and

occur at intervals of 10 to 50 cm. Occasionally these layers are graded (Fig. 5B), starting with whole shells and grading upward through coarse sand and shell fragments, and finally into laminated fine-grained sediment. The graded character of these layers suggest that they have been deposited as a unit, perhaps during high-energy events, such as storms. Frequently, however, these coarse sand-shell layers are not graded. These nongraded layers may be similar to the mixed sand-shell layer now found at the surface of the channel, where strong tidal currents winnow away much of the finer material.

Numerous animal escape traces, originating from the coarse sand-shell layer and migrating through the laminated silts, have been observed in the channel cores (Fig. 3). These escape traces suggest that the overlying non-turbated and finely laminated sediment was deposited very rapidly (Reineck and Singh, 1973).

In cores taken from the subtidal bank alternating layers of fine sandy silt and clayey silts dominate the sedimentary record. Coarse sand and shell layers are missing, but non-laminated zones, 1-10 cm thick, separate the zones of lamination (Fig. 6). These non-laminated zones occur at intervals of 10-50 cm and are probably a result of turbation. The top few centimeters of the sediment are also generally turbated.

The 6-meter core (V32-02) taken in the channel at mp 18.6 records a change in lithology at 300 cm (Fig. 4). Whereas the top 300 cm contains many coarse sand and shell layers,

the sediments below 300 cm consist almost entirely of laminated fine sand, silt, and clay and thus closely resemble the 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.

The top 10 to 15 cm of cores taken in the inner harbor consisted of highly liquefied black sediment, below which the sediment was more cohesive but extensively turbated. The turbation was a result not only of mechanical mixing by organisms but also of the release or entrapment of biochemically formed gases. Several of the meter-long cores were turbated over their entire length, showing only hints of lamination. Distinct zones of lamination, however, were occasionally observed. The laminated zones occurred more frequently in cores above mp 5 and in the channel. Subsurface layers of medium to fine-grained sand, from 1-15 cm thick, were also occasionally observed in several inner harbor cores below mp 0.

Secondary Gas Structures

Gas fissures, attributed to the production and release of methane (CH_4), were frequently observed in the Hudson estuarine sediments. Figure 7A shows a large gas bubble forming on the outside of a core which has just been extruded, and Figure 7B illustrates the gas fissures observed when the core was split. Similar gas fissures have been noted by Schubel (1974b) in the sediments of Chesapeake Bay.

Gases such as CO_2 , N_2 , H_2S , and CH_4 are released into the interstitial waters of the 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, oxidation continues by means of anaerobic bacterial processes - denitrification, sulfate reduction, and methane production. In recent years, extensive studies have been conducted on the physical, chemical, and ecological constraints which govern these anaerobic processes (Kaplan, 1974). It appears that methane is not produced at an appreciable rate until almost all of the dissolved sulfate in the interstitial waters is removed by sulfate-reducing bacteria (Martens and Berner, 1974). Reeburgh (1969) did not detect any CH_4 in the upper 10 to 20 cm in cores taken in Chesapeake Bay, but found that CH_4 could form bubbles at greater depths and concluded that embullition controls the maximum concentration of methane in the sediments.

In the interstitial waters of the Hudson Estuary, CH_4 abundance ranges from 1 $\mu\text{mole/l}$ to its in situ saturation value of 10^4 $\mu\text{mole/l}$ (Hammond, 1975). Bubbles may form at any depth when the concentration of CH_4 approaches its in situ saturation, and the release and partial solution of bubbles is an important factor in methane transport across the sediment/water interface (Hammond, 1975). Bubbles, however, may also become entrapped in the sediment, forming secondary gas structures and resulting in gaseous sedimentary zones that are acoustically turbid to

seismic profiling (Worzel and Drake, 1959; Schubel, 1974b).

Gas fissures often completely obliterated the primary sedimentary structures of several meter-long cores taken in the inner harbor. In other 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 structures. The marked increase in the occurrence of gas fissures in the sediments of the inner harbor may reflect the additional input of organic sewage to this area.

CARBON-14

To gain an understanding of the radiocarbon chemistry in the estuary and in an attempt to obtain an estimate of the net sedimentation rate, ^{14}C analyses were conducted on surface shells, subsurface shell layers, and the organic matter in the surface sediments. The type of material dated and the radiocarbon ages are listed in Table 2. The radiocarbon ages for Mulinia lateralis are layer averages based on 50 to 150 shells. Cores 18.6-1M (Fig. 3), V32-02 (Fig. 4), and P21.7M were taken in the channel between mp 18 and mp 22. Grab 0.1W was taken at the same location as core 0.1W (Fig. 1) and grabs G42.5W and G18.3W are located in Figure 1.

Interpretation

The shells at the sediment surface contain excess ^{14}C produced by nuclear weapons tests 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 dates indicate that the organic matter in the surface sediments at mp 18.3, as well as at mp 42.5, is predominantly recycled old carbon, perhaps from soils of the drainage basin or reworked from sediments in other areas of the Hudson River, and is not newly fixed carbon from sewage disposal or phytoplankton activity (Simpson et al., in preparation). The bomb-contaminated age of the organic matter at mp 0.1 reflects the additional input of recent organic sewage to this area.

The coarse surface layer of the natural channel between mp 18 and mp 22 also contains bomb-contaminated shell material, but the shell layers immediately below are very old. One possible interpretation of these data is that the subsurface shell layers are composed of older shells that have been exposed and reworked by channel erosion and migration. The coarse nature of the surface sediment indicates that the channel is presently undergoing tidal scour, and the change in lithology below 300 cm in core V32-02 may be a result of channel migration. The fact that the shells are generally concentrated into layers indicates that some reworking of the sediment has taken place. In addition, the older age of the Mulinia layer at 199 to 201 cm relative to the layer at 266 to 270 cm indicates that some of the Mulinia layers are composed of older reworked shells.

Another possible reason for the old age of the near surface shell layers is that prebomb $^{14}\text{C}/^{12}\text{C}$ ratios in rivers may be as much at 10-25% lower than atmospheric ratios due to the introduction of ^{14}C -free bicarbonate from the chemical weathering of rocks (Broecker and Walton, 1959). Consequently, carbonate shells which formed in the estuary before the onset of nuclear weapon testing may be depleted in ^{14}C with respect to the atmosphere and thus be expected to have an "inherited" initial age. Simpson et al. (in preparation) have suggested that the initial age can be estimated on the basis of the $\delta^{13}\text{C}$ of the carbonate sample. Without the $\delta^{13}\text{C}$ information, corrections on the order of 400 ± 400 years should be applied to radiocarbon dates for carbonate materials in the Hudson Estuary (Simpson et al., in preparation).

A minimum limit of ~ 1 mm/yr on the net rate of sedimentation can be obtained from the age of the oyster shell at 127-130 cm. A net sedimentation rate also of ~ 1 mm/yr has been obtained from one core (P21.7M) containing Mulinia layers for which the $\delta^{13}\text{C}$ corrected ages plot along a straight line with depth in the sediment (Fig. 8). Although sedimentation rates based on Mulinia layers are questionable due to their possible reworked nature, such a net sedimentation rate is consistent with the thickness of the organic silt layer relative to the beginning of estuarine conditions and also with net sedimentation rates observed in other estuaries (Rusnak, 1967; Oviatt and Nixon, 1975; Olsen, in press).

GAMMA-EMITTING RADIONUCLIDES

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. ^{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, cobalt-60 and other gamma-emitting nuclides in the Hudson Estuary have been studied by Lentsch et al. (1970), Hairr and Wrenn (1972), and Lentsch (1974). ^{137}Cs has been introduced into the Hudson Estuary via global fallout from nuclear weapons tests 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.

Figure 9 shows the annual fallout record for ^{137}Cs to the New York City area during the past 21 years. The major influx of fallout ^{137}Cs to the Hudson occurred in 1962-64. Figure 10 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 suspended-sediment transport and good indicators of sediment that has been recently deposited. In areas of rapid deposition, the 1971 reactor peak and 1962-64 fallout peak may be preserved in the sediment, thus providing definite time-stratigraphic units.

The gamma-spectrometric data for suspended matter and several cores in the inner harbor, river estuary, and marginal zones have been presented 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 by pumping water into 200 gallon tanks at mp 18 and allowing the suspended matter to settle out. The oxidized layer was removed from grab samples at mp 18.6 (SLOSS II) and at mp 42.8 (SLOSS III), approximately 100 meters south of the Indian Point outflow system.

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 cobalt-60 decreases rapidly with depth in the sediment. The activity was generally confined to the surface turbate zone (subtidal bank) or surface coarse layer (channel) and consequently was usually undetectable below 10 cm. In broad shallow areas (25.3W) and in protected areas along the sides of the estuary (53.8EC, 52.5EC), radiocesium was frequently detected to depths on the order of 15-25 cm, indicating that these areas are sites of more rapid deposition. These results are consistent with those presented by Hairr and Wrenn (1972) for several cores taken in the Hudson around the Indian Point nuclear power station (mp 43).

In the inner harbor, the vertical distribution of radiocesium and cobalt-60 in the sediment is much more extensive than in either the river estuary or marginal zones. In the

inner harbor, ^{134}Cs has been detected to depths greater than 60 cm. 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 , ^{60}Co , and ^{134}Cs are frequently just as high, if not higher, at 40-50 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. 11).

^{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 reflects the sediment composition.

Interpretation

Although the quartz-rich channel sands at the surface of cores 18.6-3M, C44, and 57.5M have lower ^{40}K activities, the ^{40}K activity of the suspended matter and fine-grained sediments shows little variation, indicating no major changes in the fine-grained sediment composition, in different areas of the estuary or with depth in the sediment. Consequently, we can assume that the peak concentration of ^{137}Cs , ^{134}Cs , and ^{60}Co at 30-35 cm in core 0.1W (Fig. 12) and at 50-55 cm in core -1.5E (Table 3) corresponds to the maximum reactor release in 1971 and not to a major change in sediment composition, and thereby obtain sedimentation rates on the order of 5-20 cm/yr. These rates are consistent with the amount of sediment that is

annually dredged in this area of the estuary to maintain present channel and harbor depths (Army Corps of Engineers, unpublished data). It thus appears that most of the suspended matter and much of the material that is resuspended, transported, and redeposited by tidal and estuarine processes is presently accumulating in the inner harbor.

Cores taken in recently dredged areas, such as core 8.7WM, taken in the annually dredged Weehawken-Edgewater Channel (mp 5-10), contained relatively low concentrations of radio-cesium 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 >50 cm. It thus appears that the distribution of radiocesium in the sediment may be helpful in distinguishing areas that have recently been dredged, and thereby exposing older "unpolluted" sediment near the surface, from areas undisturbed by dredging operations. This distinction is of primary importance when studying the effects of 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.

Cores taken in both Foundry Cove (53.8EC) and Indian Brook Cove (52.5EC) contained relatively high concentrations of ^{137}Cs , and detectable concentrations of ^{134}Cs and ^{60}Co (Table 3). Both coves are located approximately 10 miles (16 km) upstream

from the reactor at Indian Point (mp 43), indicating a transport of reactor-produced radionuclides upstream, and accumulation in protected areas.

For several cores, the vertical distribution of radiocesium was correlated with the observed sedimentary structures (Figs. 11 and 12). In core -1.6E (Fig. 11), the layer of medium- to fine-grained sand at 50-65 cm contained no 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 sands at mp 18.6 contained detectable concentrations of ^{137}Cs and ^{134}Cs 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 of these data 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. It is proposed that high energy events, such as severe storms, are necessary to transport and deposit sand in an area where mud deposition is normally taking place. The fact that the mud below 65 cm contains radiocesium indicates that extensive erosion did not occur before the deposition of the sand layer.

There also appears to be a correlation between the observed lamination in the fine-grained sediments and the radiocesium

activity. In cores taken in the river estuary, anthropogenic radionuclides were generally confined to the surface turbate zone and not observed in the laminated sediments below. In inner harbor 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. 12). This sharp decrease in radiocesium activity 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. In addition, the radiocesium activity rapidly decreases below 15 cm in core 1.9W where a zone of lamination (15-40 cm) was observed. Below this zone of lamination (>40 cm) there was no detectable radiocesium activity, although anthropogenic detritus (furnace slag and coal) were observed at 50 to 55 cm.

TRACE METALS

Quantitative analyses for copper, lead and zinc have been determined as a function of depth in core V32-02 from the channel at mp 18.6; core 53.8EC from Foundry Cove; cores 0.1W and 1.9W, taken in the inner harbor; and a surface grab sample (G18.6-5), taken on the subtidal bank adjacent to core V32-02. 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, as determined by loss-on-ignition, the total potassium concentration, as determined from the ^{40}K activity, and the weight percent quartz as determined by monitoring the α - β transition in a Differential Scanning Calorimeter at 1 atm pressure and 573°C, are also presented in Table 4. The trace metal analyses were conducted on cores for which the radiocesium and cobalt-60 activities have also been determined (Table 3).

Interpretation

Metal concentrations for core V32-02 show very little variation below 25 cm. Owens *et al.* (1974) have reported similar Zn concentrations in the organic silt between 20-50 meters in a drill core taken near the Newburgh Bridge (mp 61). It thus appears that the metals in the laminated fine-grained sediments below 25 cm in core V32-02 are at pre-industrial levels (Williams *et al.*, in preparation). This conclusion is consistent with the lack of ^{137}Cs below 25 cm, and with the old radiocarbon ages determined for the subsurface shell layers.

The coarse nature of the surface sample in core V32-02, as illustrated by its low organic and potassium values and its high quartz content, made it difficult to compare its metal concentrations with those obtained for the fine-grained sediments below. Consequently, surface organic silts from the adjacent subtidal bank were analyzed for the same metals. The ^{137}Cs and K data indicate that these organic silts have been recently deposited and are similar in K-mineral composition to the organic silts of V32-02. The higher metal concentrations

in G18.6-5 relative to those below 25 cm in V32-02 may be indicative of recent pollution.

The higher concentrations of Cu, Zn, and Pb in the surface sediments of Foundry Cove (53.8EC) relative to the sediments at 50 cm may also reflect the recent increase in human and industrial pollution in this area. The metal levels observed at 50 cm in core 53.8EC are similar to those below 25 cm in core V32-02.

In the inner harbor sediments, high metal concentrations were observed to extended depths. This finding is consistent with the vertical distribution of radiocesium and indicates that much of the recent "metal-polluted" sediment is rapidly accumulating in the inner harbor. Metal levels and percent organic matter in core 0.1W and core 1.9W have also been correlated with the sedimentary structures. As with radiocesium, the metal concentrations and percent organic matter appear to be lower in the laminated zones relative to the turbate zones. The sharp increase in the concentrations of Zn, Cu, and Pb above 53 cm in core 0.1W provides a good documentation of the increase in trace metal levels associated with urban pollution.

DISCUSSION

The sedimentary material deposited in the Hudson Estuary may be derived from several different sources. Waste discharge into the estuary from sewers, sewage-treatment plants and industries may amount to nearly 0.3 million tons per year (Gross, 1974) and may account for approximately 10% of the

shoaling material in the Lower Estuary (Panuzio, 1965). Long-shore drift of sand along the Long Island and New Jersey beaches funnel sediments into the estuary mouth south of the Narrows, and the extent to which this sandy sediment is transported up the estuary is still uncertain. Perhaps the sand layer at 50-65 cm in core -1.6E has such an origin. Sand, silt, and clay may also be derived from marginal sources such as Pleistocene till, landfill, and construction sites. Roberts and Pierce (1974) have shown that the unconsolidated, unvegetated and highly erodible soils of construction sites may provide large quantities of sediment to urban estuaries.

Till, construction sites, and agricultural areas in the drainage basin of the Hudson River also provide riverborne sediment to the estuary. The suspended-sediment load of the Hudson River has been estimated at 0.83 million tons per year (Panuzio, 1965). Concentrations of suspended sediment exhibit both diurnal and seasonal variations but typically increase with depth and decrease in a seaward direction. Some of this suspended material is deposited in wide shallow areas and in protected areas along the sides of the estuary, but a large quantity is transported to the inner harbor where it is deposited in submerged shoaling areas, principally along the west subtidal bank between mp 0 and mp 11 (Panuzio, 1965). Rapid shoaling in this area has been attributed to the narrowing of the estuary near the George Washington Bridge (mp 11), and the resultant constriction of the upstream flow (Panuzio, 1965; Gross, 1974). The increase in cross sectional area

below mp 0, flocculation during conditions of high river flow (when the Hudson transports most of its suspended load), and the additional sediment influx from waste discharge, possible marine sources and from the East, Passaic, and Hackensack rivers may also be responsible for the rapid shoaling in the inner harbor.

Alternating coarser and finer-grained layers, on the order of a mm to cm thick, are characteristic of the fine-grained sediment in the Hudson Estuary and have also been reported in other subtidal and intertidal environments (Van Straaten, 1954, 1959; Moore and Scruton, 1957; McKee, 1957; Coleman, 1964; Reineck, 1967; Reineck and Singh, 1973; Oertel, 1973). The genesis of this lamination is not yet clearly understood but 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 gasturbation, and on the rate of sedimentation.

Net sedimentation rates based on the thickness of the organic silt layer relative to the beginning of estuarine conditions and from radiocarbon ages, indicate that there may be a correlation between annual deposition (2 ± 1 mm/yr) and the observed lamination in the river estuarine environment. In such a dynamic environment, however, where there is con-

siderable tidal stirring and bioturbation of the surface sediment, lamination resulting from slow accumulation and seasonal variations would probably not be preserved.

Another 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, 1959; Reineck and Wunderlich, 1969; 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, there is some question whether enough mud can be deposited during the short time period of slack water to form the observed mud laminae (McCave, 1970). The formation of mud aggregates and fecal pellets (Klein, 1971; Haven and Morales-Alams, 1968) may increase the rate at which the mud settles from suspension, and the bedload deposition of mud aggregates (Oertel, 1973) may also be important in the formation of mud layers. In any case, unless there is an adequate sediment supply this tidal lamination will probably be destroyed by bioturbation or reworked during later tidal cycles.

When there is sufficient concentration of suspended matter, however, perhaps due to severe storms, deposition of fine-grained sediment will be relatively continuous (McCave, 1970, 1971; 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 lamination during periods when the Hudson transports large quantities of sediment, such as during severe storms.

The rapid deposition of the laminated unit during severe storms, 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 turbate zones may represent periods of slower deposition under normal flow conditions, and the sand-shell layers, periods of tidal scour or increased sand transport during high-energy conditions. In areas where deposition is relatively slow under normal flow conditions, such as along the subtidal bank of the river estuary, the turbate zones may represent periods when burrowing organisms obliterate the lamination to varying degrees. In the inner harbor, the turbate zones may represent periods when waste discharge forms a relatively greater proportion of the material deposited and the turbation is primarily a result of biochemically formed gases. Such an interpretation is consistent with the higher concentrations of trace metals, radionuclides, and organic matter observed in the turbate zones relative to the laminated zones. In addition, the lower values in the laminated zones would be expected if the laminated unit were composed predominantly of material eroded from the drainage basin or sediments reworked from upstream areas during severe

storms.

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 (1974a) 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 (Schubel and Zabawa, 1974). This laminated unit was separated from older bioturbated sediments by a sharp boundary (Schubel and Zabawa, 1974).

Kumar and Sanders (in press) have found that storm deposits may characterize the nearshore sediments off Long Island and identified the storm sequence as consisting of (1) a basal lag of coarse material and shell debris, (2) laminated sand and, (3) a burrow-mottled, fair-weather cap. Although the sediments in the river estuary are much finer in texture, these same sedimentary structures exist.

CONCLUSIONS

Geochemical and sedimentological evidence indicates that much of the suspended matter and sedimentary material that is deposited, resuspended, transported, and redeposited by tidal and estuarine processes is presently accumulating in the inner harbor and in protected areas along the sides of the estuary.

The vertical distribution of bomb- and reactor-produced radionuclides indicates sedimentation rates on the order of 5-20 cm/yr in some areas of the inner harbor and may be helpful in distinguishing areas that have been dredged from areas undisturbed by dredging operations. 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 New York Bight Apex may provide radioactive tracers for sediment transport in the Bight and across the shelf. The inner harbor sediments also contain relatively high concentrations of trace metals and organic matter reflecting the recent increase in human and industrial pollution.

In the natural channel and subtidal bank environments of the river estuary, upstream from the inner harbor, bomb- and reactor-produced radionuclides, anthropogenic detritus, and high trace-metal levels appear to be confined to the surface turbate zone (subtidal bank) or the coarse surface channel lag. Radiocarbon dating of subsurface shells and shell layers indicates a minimum net sedimentation rate of 1-3 mm/yr in the natural channel between mp 18 and mp 24. In contrast to the inner harbor, radiocarbon dating of the organic matter in the surface sediment along the subtidal bank of the river estuary indicates that the carbon is old, recycled carbon, perhaps eroded from the drainage basin or reworked from sediments in upstream areas of the Hudson River.

One possible interpretation of the geochemical and sedimentological data is that (1) the laminated fine-grained

sediments are deposited when the Hudson transports relatively high concentrations of sediment, such as during storms; (2) the coarse sand and shell layers result from increased sand transport during high-energy (storm) conditions or from tidal scour in the natural channel under normal flow conditions; and (3) the turbate zones may 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 "polluted" sediment in specific areas (e.g., inner harbor and marginal zones) but that storm deposits characterize the sedimentary record in subtidal bank and channel environments of the river estuary.

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Table 1. - Mineral composition of sediment at mp 18.6
in the Hudson Estuary

	SAND (0.65 mm diameter)	SILT + CLAY (<0.063 mm)
Quartz	67%	47%
Plagioclase	10%	15%
K-feldspar	12%	16%
Muscovite	2%	3%
Other K-Al-silicates ⁽¹⁾	-	8%
Biotite	1%	2%
Heavy minerals ⁽²⁾	7%	7%
Others ⁽³⁾	<1%	<1%

(1) Probably illite clay aggregates or extensively weathered K-feldspar.

(2) See text.

(3) Chlorite, apatite, clay aggregates.

Table 2. - Radiocarbon ages in the Hudson Estuary

Sample	Depth	Material	Age
Core 18.6-1 (Fig. 3)	0-5	<u>Crassostrea</u> , <u>Mya</u> , and <u>Macoma</u>	-400
	21-23	<u>Mulinia</u>	1240±50
	27-31	<u>Mulinia</u>	1260±60
	48-50	<u>Mulinia</u>	1320±60
Core V32-02 (Fig. 4)	0-10	<u>Crassostrea</u> , <u>Mya</u> , and <u>Macoma</u>	-350
	109-111	<u>Mulinia</u>	1120±300
	127-130	<u>Crassostrea</u>	1500±130
	199-201	<u>Mulinia</u>	2213±370
	266-270	<u>Mulinia</u>	2080±270
Core P21.7M	35-40	<u>Mulinia</u>	2160±70
	130-140	<u>Mulinia</u>	2710±80
	200-210	<u>Mulinia</u>	3180±90
GRAB G18.3	0-5	<u>Mya</u> and <u>Macoma</u>	-350
		Humic fraction	1990±230
		Residual organics	4370±430
GRAB G42.5	0-5	Humic fraction	1470±170
		Residual organics	4140±90
GRAB GO.1W	0-5	Residual organics	-2600±270

Table 3. - Gamma-emitting radionuclides in Hudson Estuary sediments

Depositional Environment	Core	Depth (cm)	ACTIVITIES ^(a)			
			Cs ¹³⁷	Cs ¹³⁴	Co ⁶⁰	K ⁴⁰
<u>INNER HARBOR</u>						
-1.6E	-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±500
-1.5E	-1.5E	60-65	27±14	- 6±15*	10±20	11,500±380
		65-73	455±24	11±10	24±18	16,200±630
		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	175±26	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

Table 3 (cont'd.)

	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
	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

Table 3 (cont'd.)

RIVER ESTUARY

Channel	18.6-3M	0-5	150±13	36±12	50±17	11,900±490
		5-10	120±21	-12±15*	32±30	16,700±800
		10-15	8±20	N.D. (c)	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
Cove	52.5EC	0-5	2700±60	375±45	235±22	18,400±650
		5-10	2050±50	44±22	175±25	18,800±650
		10-15	735±30	9±26	34±26	19,800±750
		15-20	17±12	32±18	-19±26*	19,100±630
		20-25	14±11	16±16	21±21	17,200±570

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)
0.1W	0-5	258	179	140	21	N.D. ^(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	0-5	343	227	828	21	N.D.	92	yes	Gas-turbated
	20-25	237	144	164	21	N.D.	65	?	Laminated
	50-55	247	178	245	19	N.D.	79	no	Gas-turbated
V32-02 ^(e)	0-10	48	35	55	17	565	26	yes	Sandy channel lag
	22-29	81	16	39	24	258	71	no	Laminated
	83-85	83	20	24	20	240	66	no	Laminated
	150-153	82	20	28	24	267	60	no	Laminated
	240-241	68	14	28	22	348	47	no	Laminated
	325-328	77	20	24	24	272	47	no	Laminated
	430-438	77	21	27	23	291	49	no	Laminated
	545-548	81	15	21	22	370	46	no	Laminated
G18.6-5 ^(e)	0-5	157	72	100	21	294	60	yes	Turbated

Table 3 (cont'd.)

53.8EC	0-5	2475±63	98±26	69±27	19,000±770
	5-10	1825±68	17±32	19±29	21,500±960
	10-15	210±17	- 8±10*	15±17	20,000±700
	15-20	26±17	-15±11*	30±25	18,800±780
	20-25	35±23	0±12*	7±27	20,700±870
	50-55	9±14	7±9	13±20	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		665±23	65±12	93±13	17,400±600
SLOSH III (mp 42.8) oxidized layer		2730±64	420±46	325±25	18,100±670

(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 (cont'd.)

53.8EC ^(e)	0-5	333	91	158	22	154	96	yes	Turbated
	50-55	84	18	26	23	203	75	no	N.D.

(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) Modified from Williams and others, in preparation.

Figure Captions

Fig. 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 preceding the mile number. SLOSH (Standard Lamont Observatory Sediment from the Hudson) is a composite of several surface Grab samples taken in the same anchored location.

Fig. 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 and core V32-02 is presented in Figure 4. Vertical exag. = 31.

Fig. 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

¹⁴C 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.

Fig. 4. - Core V32-02 is a 6 m core taken at mp 18.6. Several of the shell layers have been ¹⁴C dated (Table 2) and quantitative analyses for Zn, Cu, and Pb have been determined as a function of depth (Table 4).

Fig. 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).

Fig. 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.

Fig. 7. - (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 the core P18.6-5 was split. Note also the light color band at 7-9 cm.

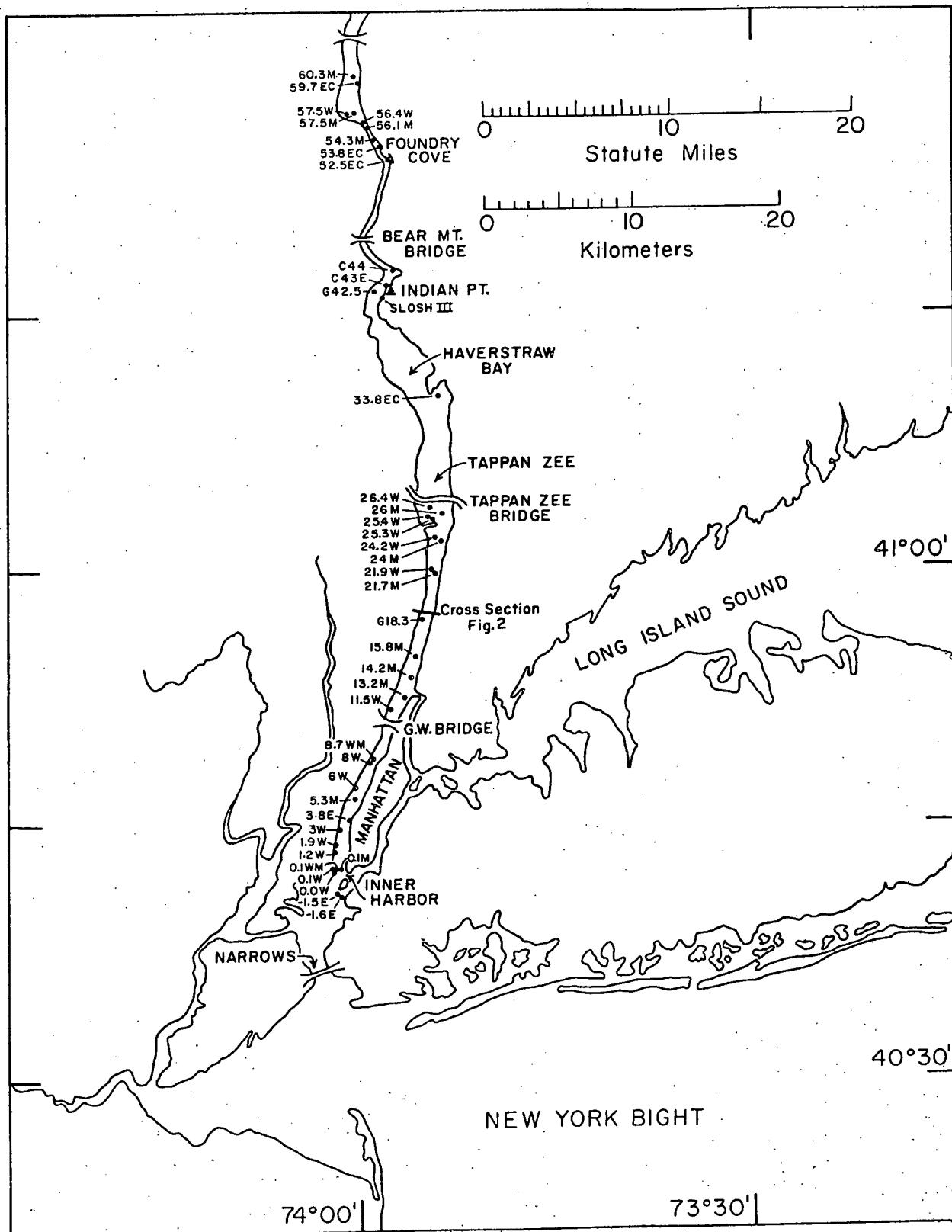
Fig. 8. - A plot of age versus depth for several of the shell layers in core P21.7M, indicating a net sedimentation rate of 1 to 2 mm/yr. The radiocarbon ages have been $\delta^{13}\text{C}$ corrected (Simpson et al., in preparation).

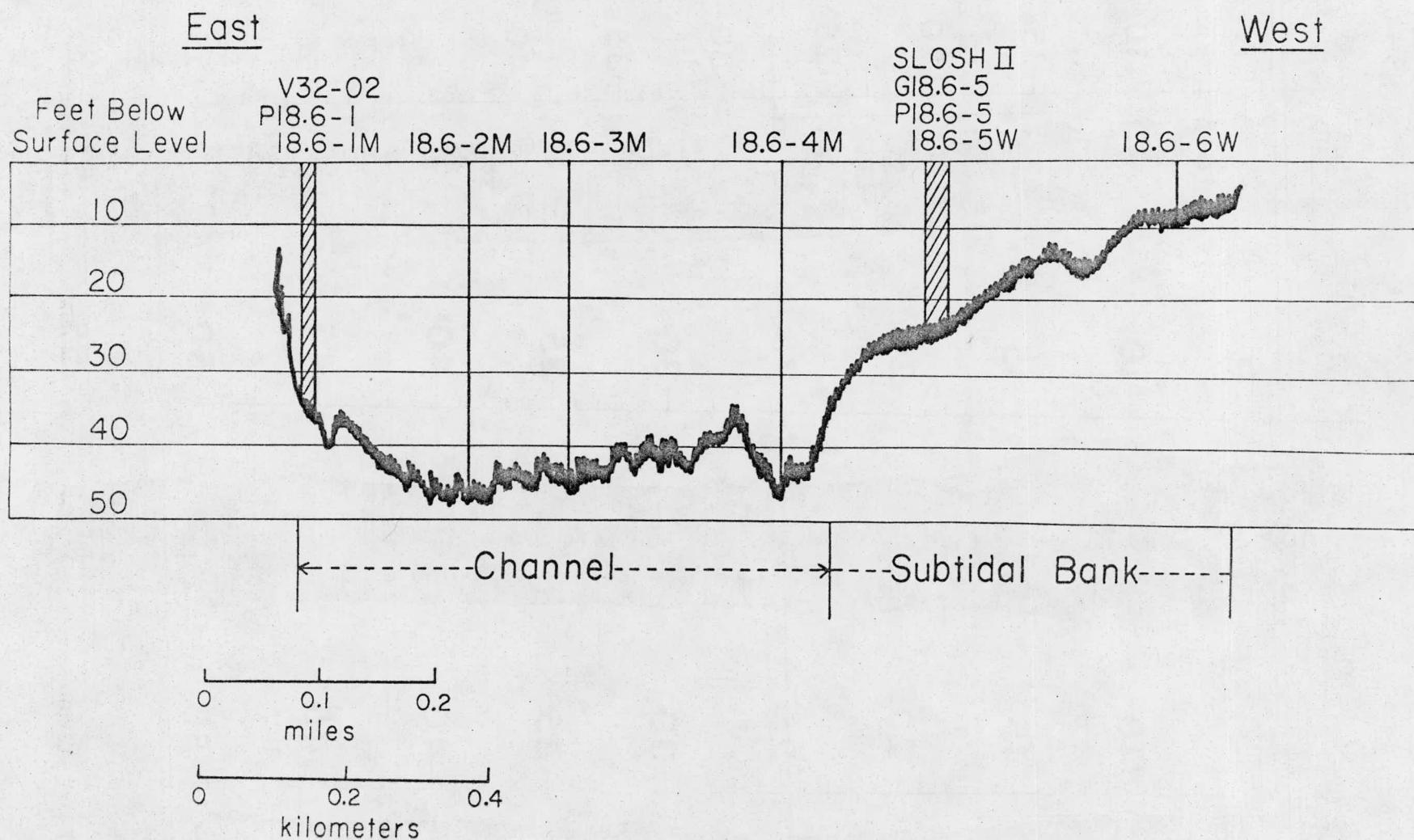
Fig. 9. - Annual deposition of ^{137}Cs from fallout at New York City. The ^{137}Cs data has been determined from ^{90}Sr precipitation data collected in lower Manhattan by the Health and Safety Laboratory of the Energy Research and Development Administration. The $^{137}\text{Cs}/^{90}\text{Sr}$ ratio is assumed to be 1.5.

Fig. 10. - Annual gross beta-gamma releases from the nuclear power plant at Indian Point (Booth, 1975). Note the peak in 1971.

Fig. 11. - 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 concentrations.

Fig. 12.- 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 Fig. 10). The sharp drop in ^{137}Cs activity below 53 cm may reflect a dredged boundary.

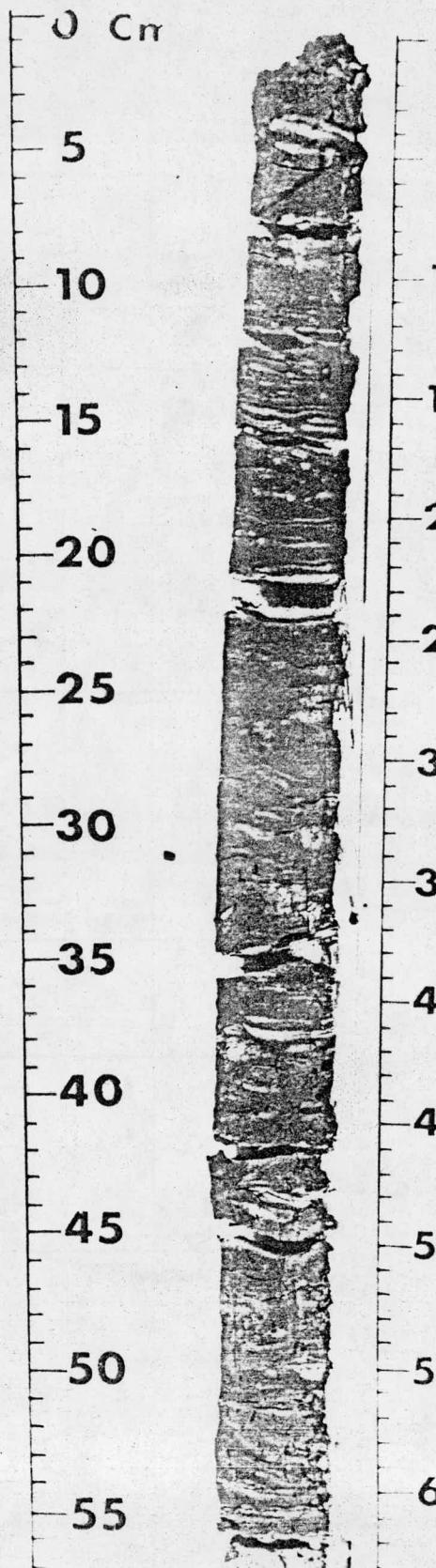
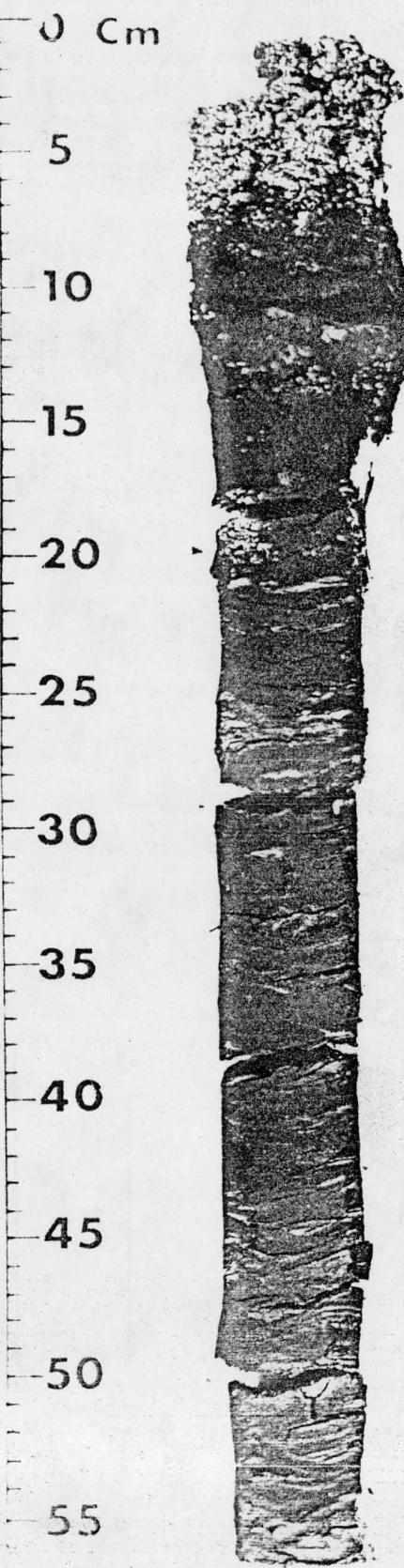
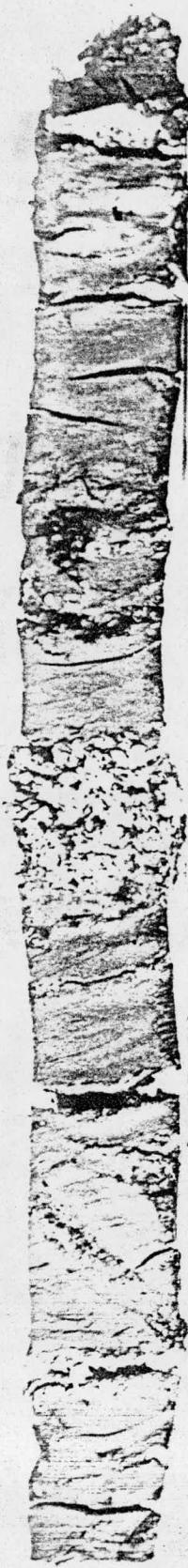


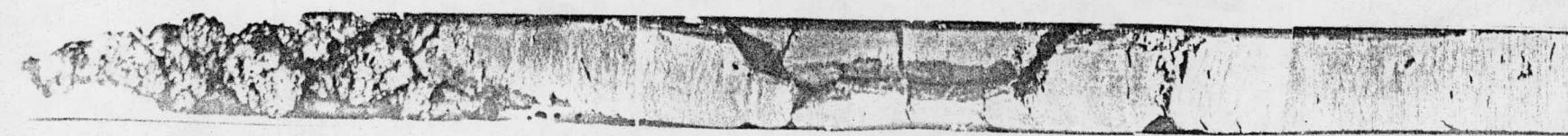
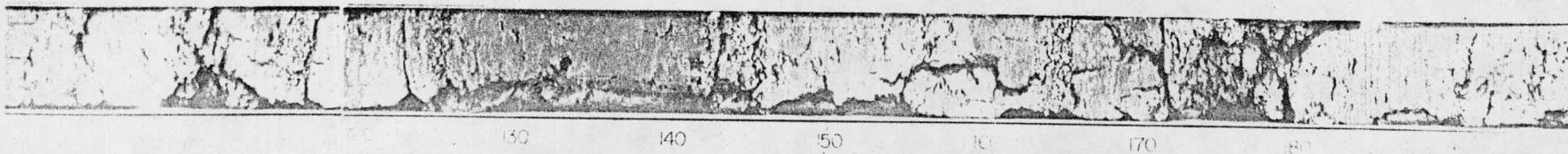
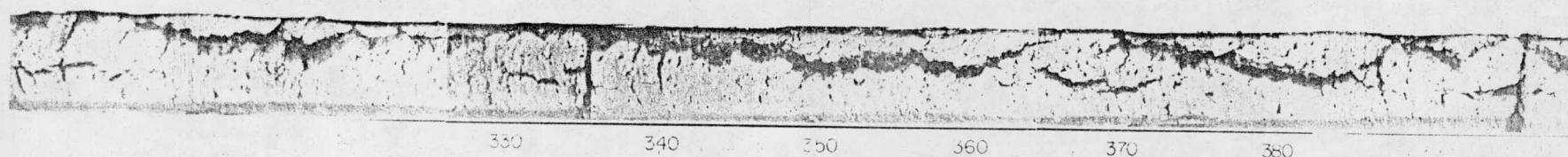


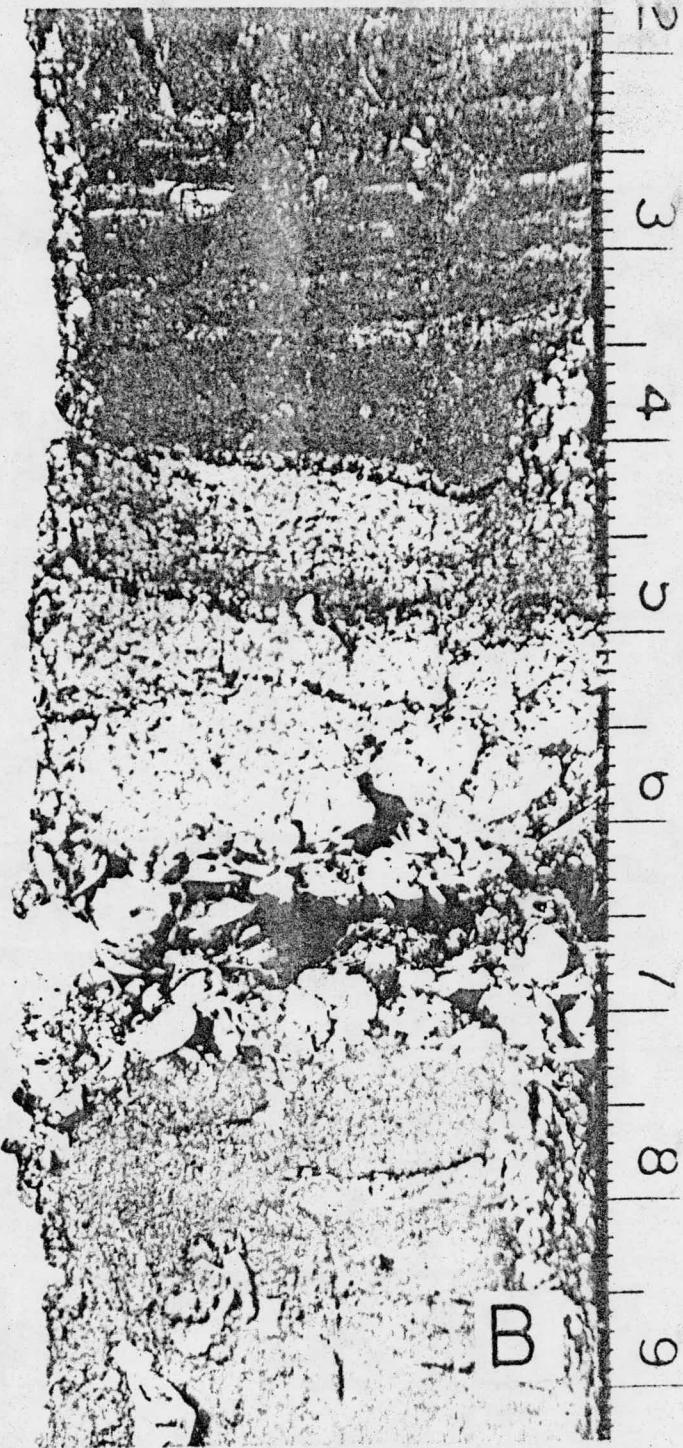
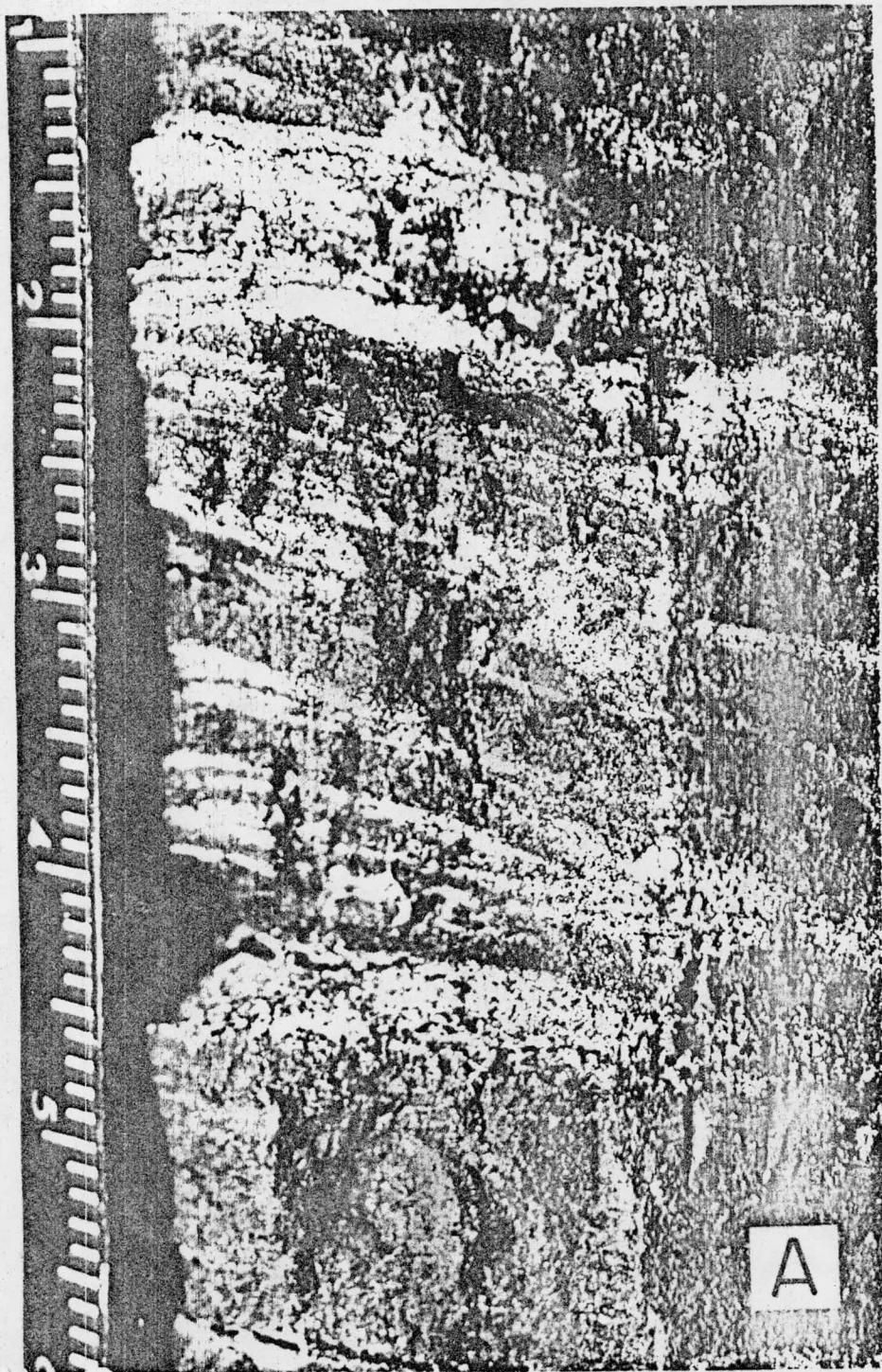
18.6-1M

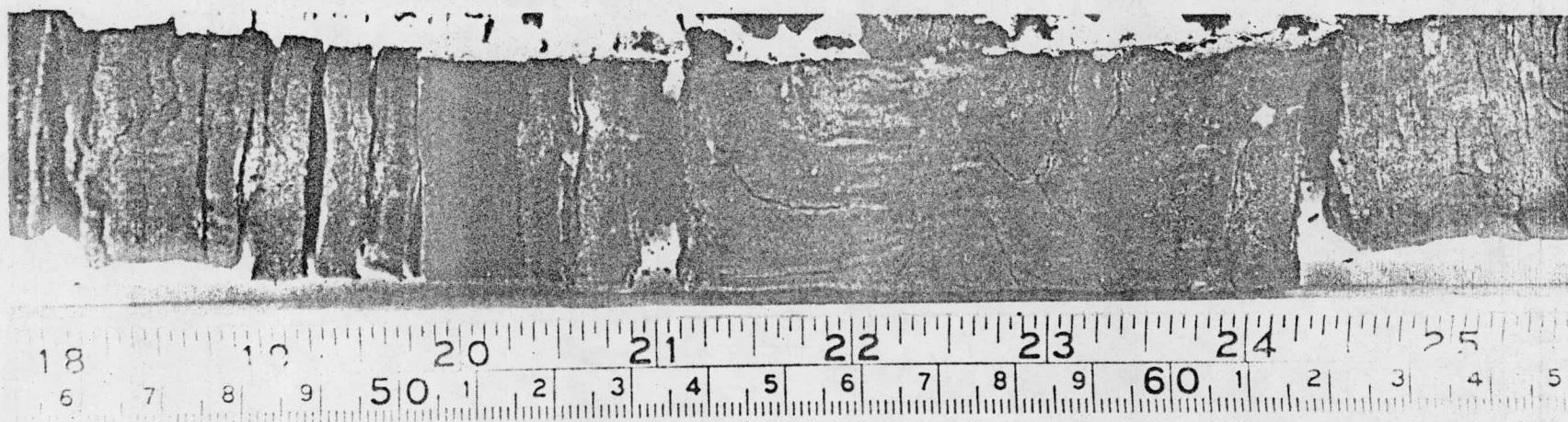
18.6-3M

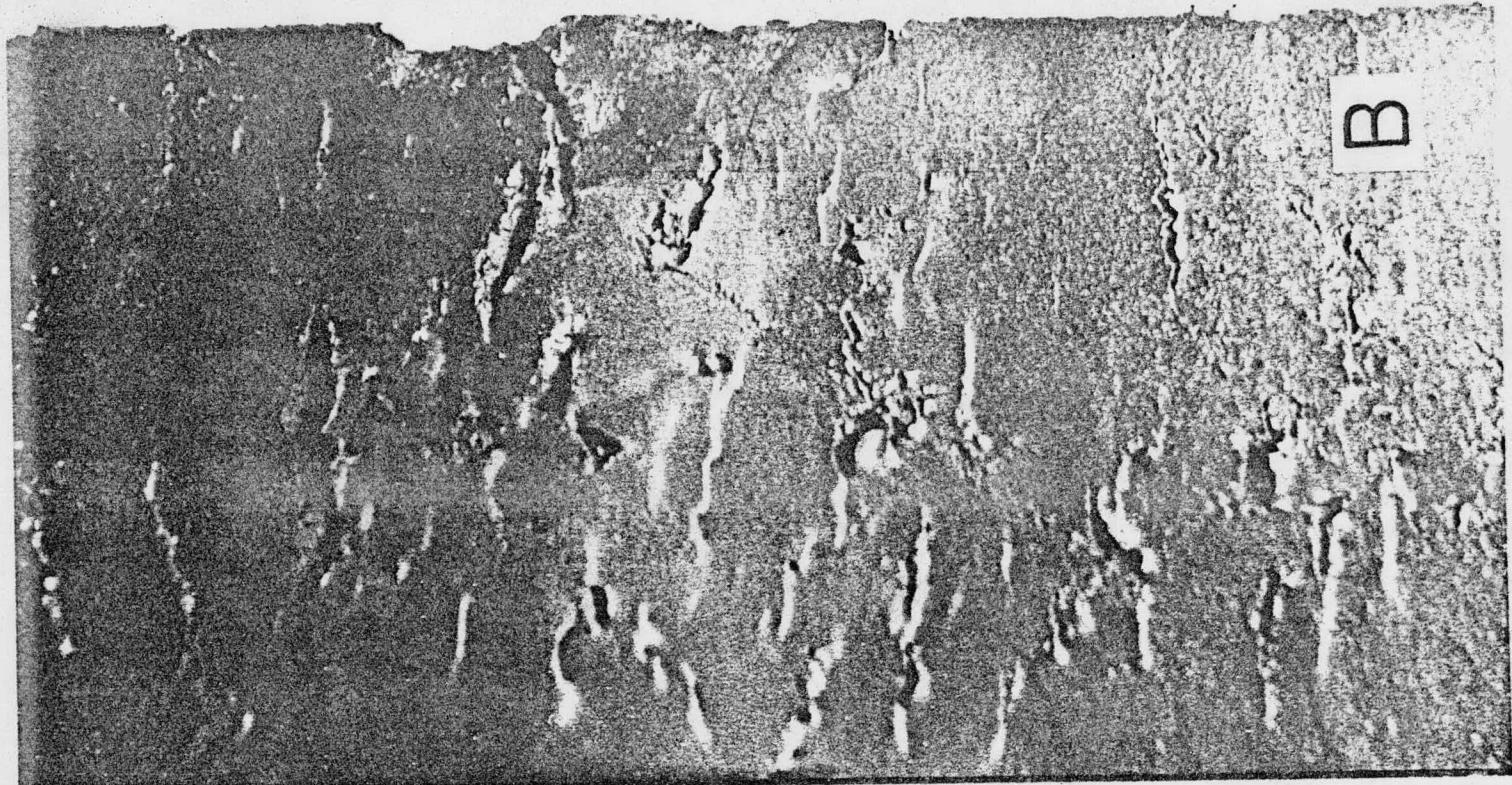
18.6-5W



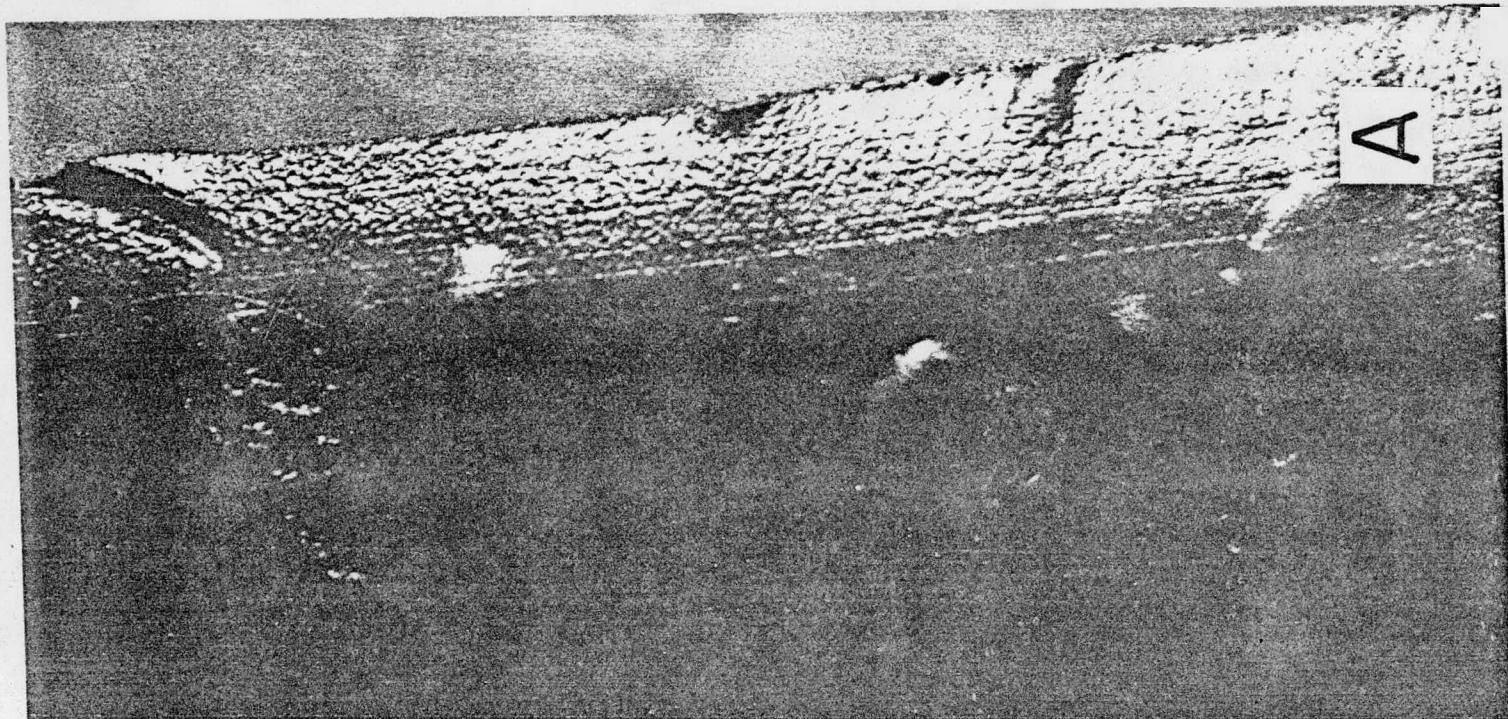
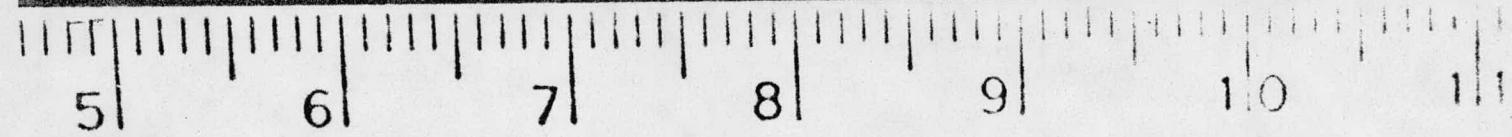








B

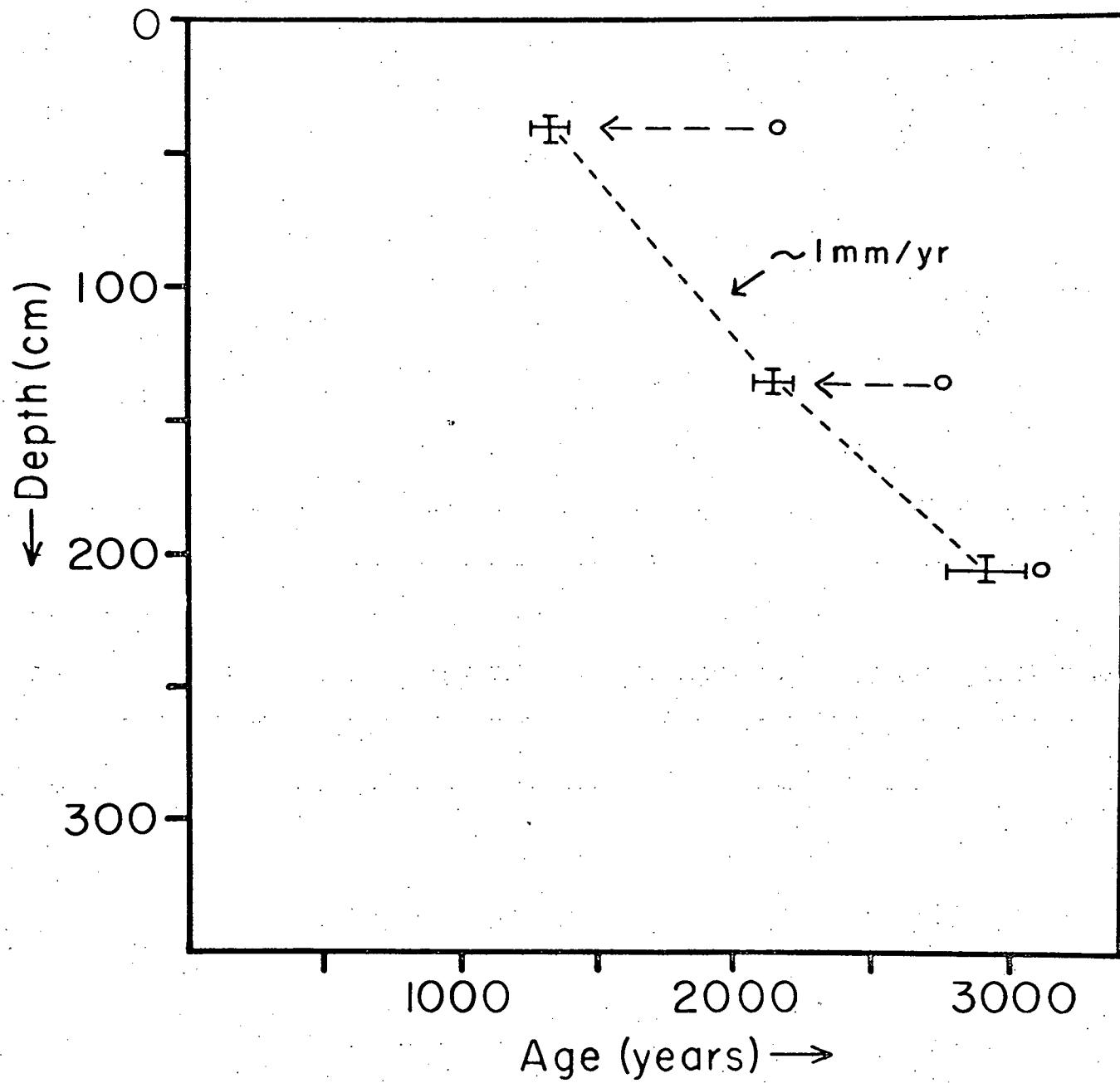


A

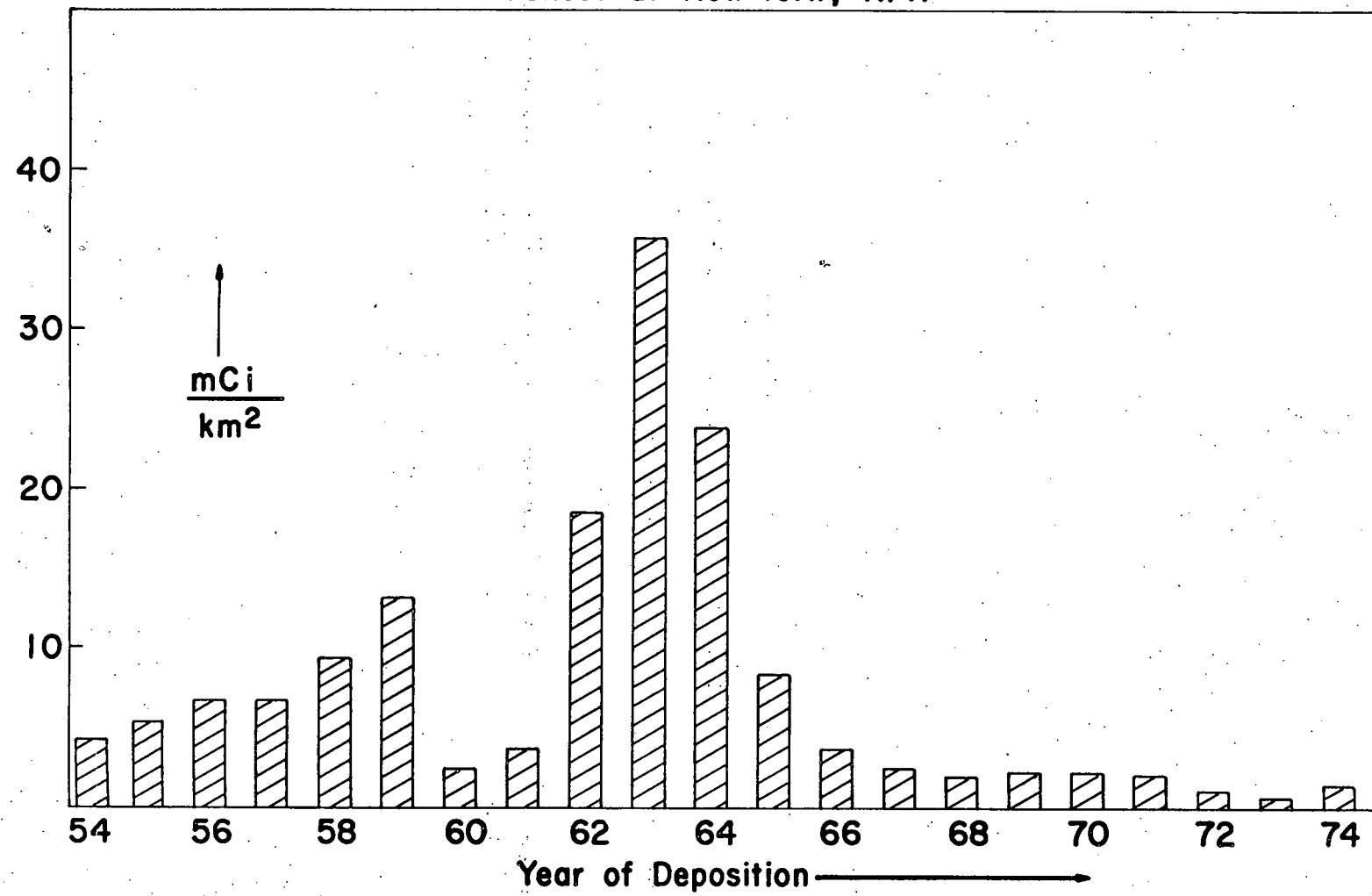


Subsurface Shell Dates

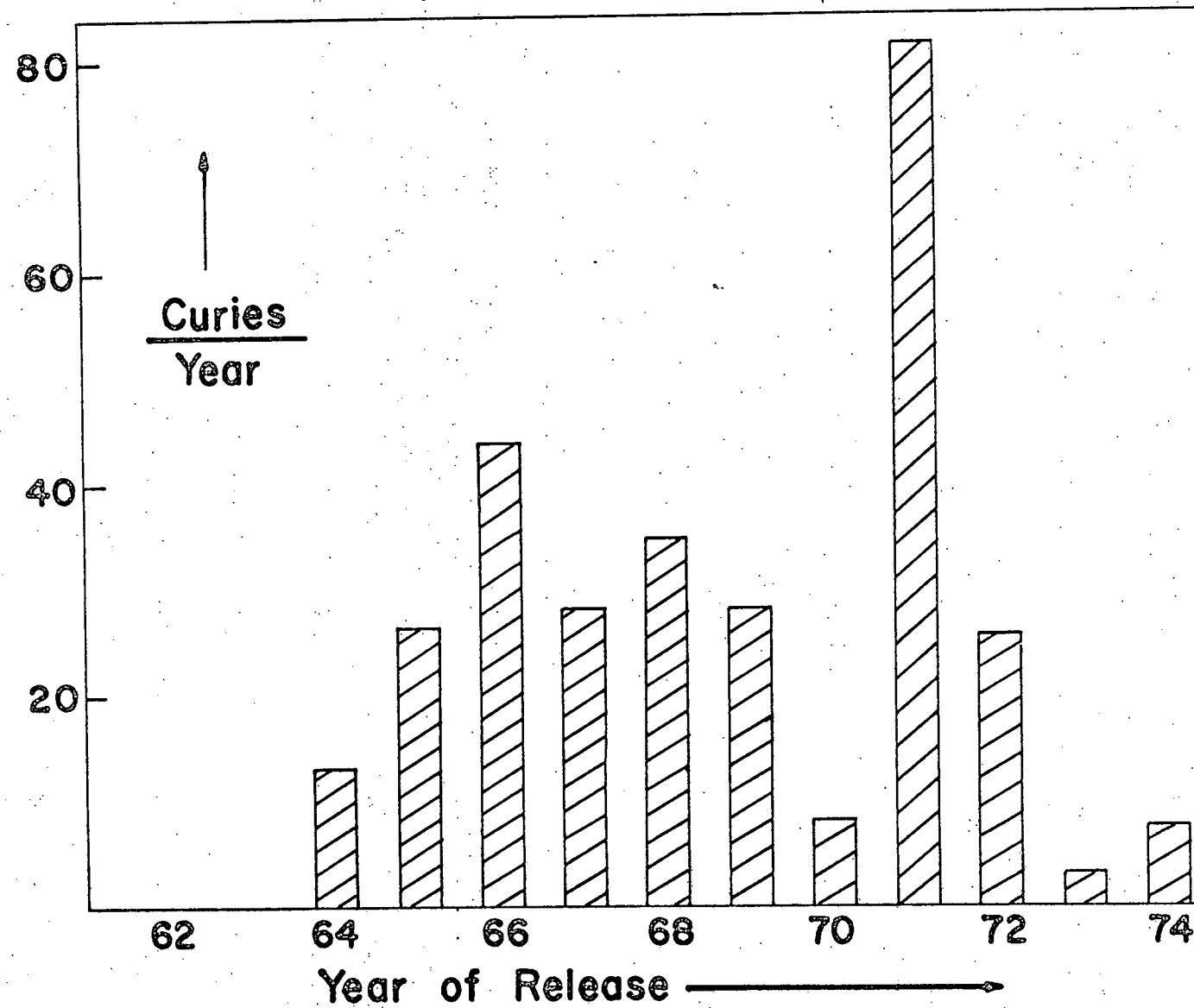
mp 21.7



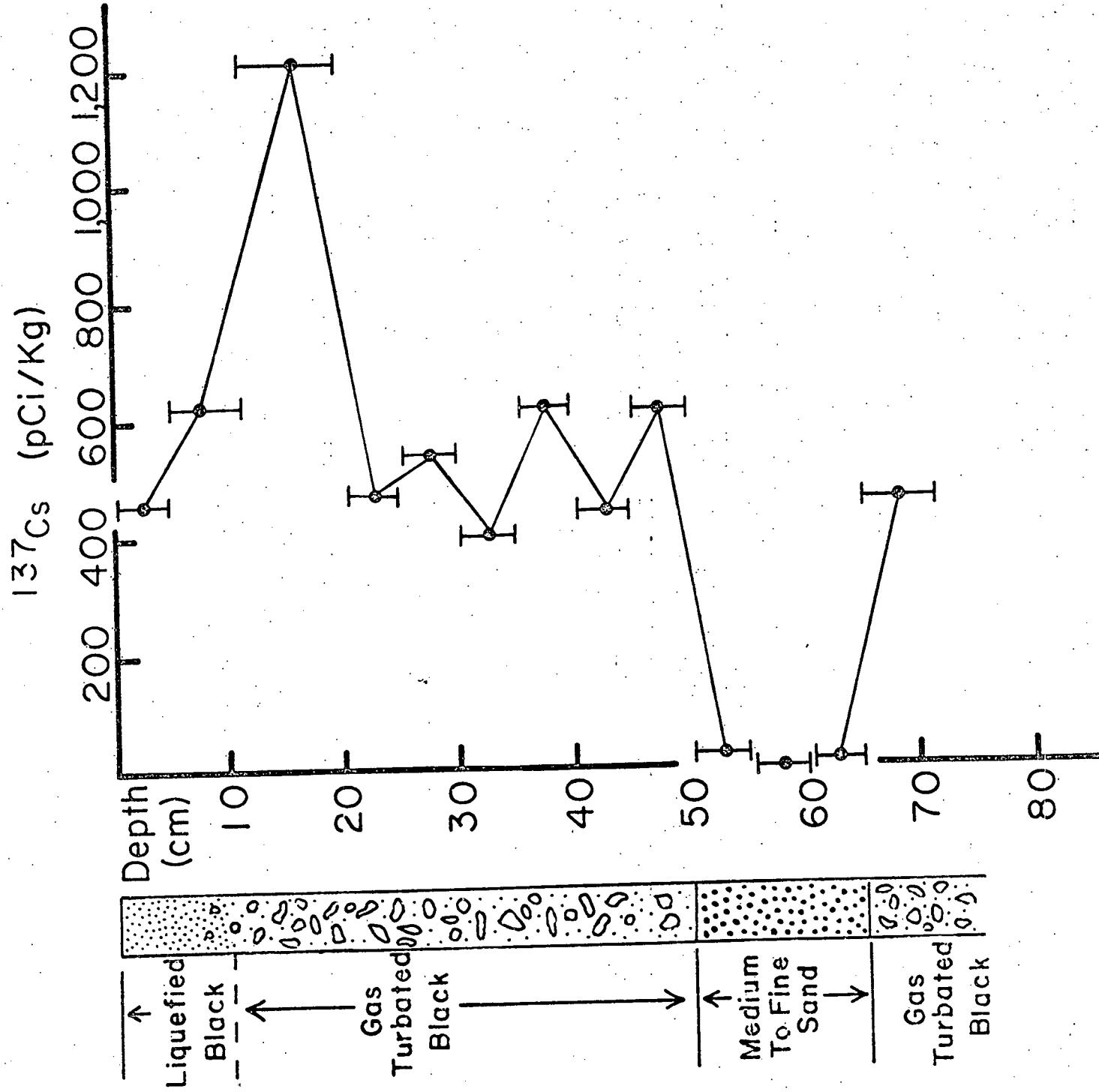
^{137}Cs Fallout at New York, N.Y.



**Gross beta and gamma releases from Indian Point
(Liquid Effluent)**



^{137}Cs Profile Core - 1.6 E



^{137}Cs Profile Core O.I.W

