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PLUTONIUM AND CESIUM RADIONUCLIDES IN THE HUDSON RIVER ESTUARY

Annual Technical Progress Report

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DISCLAIMER

SUMMARY

Plutonium and Cesium Radionuclides in the Hudson River Estuary

CONTRACT AT(11-1)-2529

The major purpose of this project is to describe the behavior and distribution of fallout plutonium in the sediments of the Hudson River Estuary. To accomplish this goal a substantial number of cores and grab samples will be collected throughout the salinity range of the Hudson River Estuary, and in as many depositional environments as possible. The distribution of fallout Cs-137 has been shown in this project to be useful in defining the general outlines of the plutonium distribution pattern, and this similarity will continue to be exploited. The possibility of transuranic releases from the reactor site at Indian Point will be explored, and the distribution of reactor-released gamma-emitting nuclides will be employed to obtain a better understanding of the estuarine sediment transport and accumulation patterns which control the distribution of fallout Pu-239,240 within the system. The feasibility of using Pu-238/Pu-239,240 ratios as an indicator of transuranic reactor releases will be explored in some detail.

ABSTRACT

We have obtained a large set of gravity cores from the Hudson Estuary through much of the ambient salinity range. A number of core sections have been analyzed for Cs-137, Cs-134, Co-60 and K-40 by direct gamma counting, and for Pu-239,240 by alpha-spectrometry. The distribution of both Cs-137 and Pu-239,240 indicates rapid accumulation in marginal cove areas and in the harbor region adjacent to New York City. The distribution of both Cs-137 and Pu-239,240 in the sediments is quite similar in surface sediments, and the trends with depth in cores are also similar. The ratio of sediment Pu-239,240 to Cs-137 throughout the sampled salinity range (0-20 o/oo) approximates that in fallout, except near the nuclear reactor at Indian Point where releases of Cs-137 result in a ratio lower (0.004-0.008) than typical of fallout (0.015). Measureable amounts of reactor-derived Cs-134, Co-60 and Mn-54 are found in nearly all of the samples containing appreciable Cs-137. These samples were between 15 km upstream of Indian Point and the downstream extent of our sampling, 70 km south of the reactor.

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INTRODUCTION

This report summarizes our activities during the initial contract year of AT(11-1)-2529. The data presented here were collected through approximately the first two-thirds of the contract period.

One of the primary goals of our research is to develop a better understanding of the behavior of plutonium in the natural aqueous environment. We propose to measure the distribution of global fallout-derived plutonium in the sediments of the Hudson River Estuary. The measured plutonium distribution in the sediments will be examined in terms of a number of variables, such as salinity of the overlying water, type of depositional environment, composition of the sediment, and the distribution of other anthropogenic radionuclides.

Our primary accomplishments up to now will be described in more detail below, but they can be summarized as follows:

(1) Collection of a substantial number of gravity cores throughout most of the salinity gradient within the Hudson River Estuary.

(2) Purchase and installation of gamma spectrometry equipment for direct counting of sediment samples, and an initial survey of a substantial number of Hudson River Estuary sediment samples.

(3) Initiation of our analytical program for plutonium, and determination of the plutonium concentrations by alpha-spectrometry of a number of samples on which we had obtained direct gamma counting data.

RESULTS & DISCUSSION

Sample Collection

We have used at least three types of equipment during our initial survey of the sediments of the Hudson, but our most extensive sampling has been done with 6.3 cm diameter gravity cores up to about 1 meter in length. Figure 1 shows the locations of most of the gravity cores we have collected, spread over a range of about 100 km of the Hudson River Estuary, encompassing a mean salinity range from about 20 o/oo down to essentially fresh water. The cores are designated in terms of a reference system which is nearly universally employed in describing locations within the Hudson River, that of the numbers of statute miles upstream of the southern tip of Manhattan. Table 1 lists the core sites and the salinity and temperature of the water at the time of collection. At most of the approximately 30 locations, at least three cores were taken and two were frozen for later sampling. One of the cores from each site was split into 5-cm depth segments, dried, and sealed in 100 ml aluminum cans for direct gamma counting. These gamma-spectrometry data are given in the following section.

We have also collected a number of grab samples of surface sediment, primarily in two general areas, one near the site of the

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FIGURE 1. Location map of gravity cores collected in the Hudson River Estuary. Locations are specified by the number of statute miles upstream from the mouth of the Hudson River opposite the southern tip of Manhattan (approximately the location of core 0.1 WM). Salinities average about 20-25 o/oo at mile point 0, and average between 1 and 5 o/oo at mile point 50.

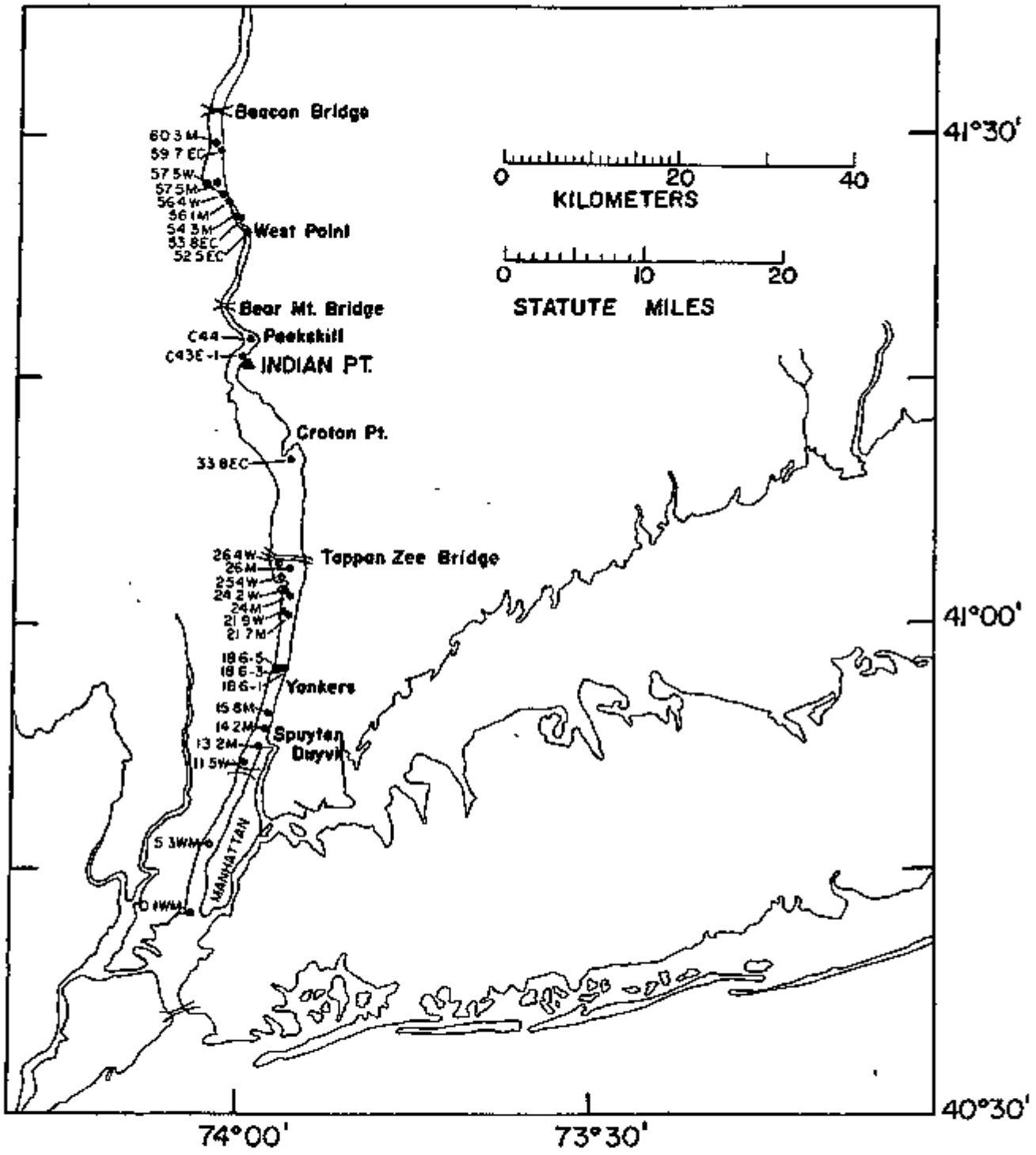
Beacon Bridge
West Point
Bear Mt. Bridge
Peekskill
INDIAN PT.
Croton Pt.
Tappan Zee Bridge
Yonkers
Spuyten Duyvil
MANHATTAN
KILOMETERS
STATUTE MILES
41°30'
41°00'
40°30'
74°00'
73°30'

TABLE 1

HUDSON RIVER ESTUARY CORE LOCATIONS,
WATER DEPTHS, BOTTOM SALINITIES AND TEMPERATURES

CORE* (mile point)	WATER DEPTH (m)	BOTTOM WATER SALINITY (o/oo)	BOTTOM WATER TEMPERATURES (^{o}C)
0.1 WM	14	27.4	14.5
5.3 WM	15	26.2	15.1
11.5 W	7	13.8	25.6
13.2 M	17	17.6	24.6
14.2 M	14	16.2	25.4
15.8 M	9	15.0	25.1
18.6-1	12	8.0	--
18.6-3	15	8.0	--
18.6-5	10	8.0	--
21.7 M	16	11.7	23.6
21.9 W	9	9.8	24.0
24.0 M	18	9.7	24.6
24.2 W	4	6.0	25.5
25.4 W	3	6.1	27.0
26.0 M	15	8.1	26.2
26.4 W	4	3.4	27.2
33.8 EC	2	4.0	27.0
43.0 E-1	5	2.8	11.0

- table continued -

*Refer to figure 1 for core location map. C = cove, E = east bank, M = midchannel, W = west bank, EC = east bank cove, WM = west of midchannel

TABLE 1 continued p. 2

CORE (mile point)	WATER DEPTH (m)	BOTTOM WATER SALINITY (o/oo)	BOTTOM WATER TEMPERATURES (°C)
44.0	22	2.8	11.0
52.5 EC	2	<1	27.3
53.8 EC	2	<1	27.0
54.3 M	19	<1	26.5
56.1 M	23	<1	27.1
56.4 W	17	<1	27.1
57.5 M	11	<1	26.1
57.5 W	2	<1	27.0
59.7 EC	3	<1	28.0
60.3 M	13	<1	27.0

nuclear power facility at Indian Point (m.p. 43, salinity 0-5 o/oo). and the other downstream about halfway between Indian Point and New York Harbor (Alpine, New Jersey, m.p. 18, salinity 5-12 o/oo). These samples provide larger samples of sediment down to about 10 cm (up to 100 g/cm of depth) than are obtained from our gravity cores (15-25 g/cm of depth). The locations of these samples are not shown explicitly in figure 1.

We have collected a few large samples (greater than 1 kg) which are composites of a number of surface grab samples. We collected these samples to provide a large supply of material for control of our laboratory procedures, and perhaps to serve as interlaboratory comparison samples. These composite samples are designated as SLOSH I, II, III in our data tables. One of these samples, (SLOSH III), collected near the reactor at Indian Point contains Cs-137 activities comparable to the NBS environmental radioactivity sample now available. We have not completed homogenizing this sample sufficiently for inter-laboratory comparisons, but we do have quite a large sample (more than 10 kg) and the activity of Cs-134 is readily measureable, as well as that of several other reactor-derived nuclides.

Cs-137 Measurements and Other Gamma-Spectrometry Data

One of the observations we made during a preliminary survey prior to submission of our original proposal was that the distribution of anthropogenic nuclides in Hudson River Estuary sediments is nonuniform and difficult to predict. This observation can also be made using the

more extensive data now available to us. Because of the complexity of sediment accumulation patterns in the Hudson, we chose direct gamma counting of sediment samples as a screening procedure before selecting samples for chemical separation and alpha-spectrometry for plutonium. This provides information which can be useful in the interpretation of measured plutonium concentrations. We were especially interested in obtaining measurements of Cs-137 and Cs-134 on samples which were analyzed for plutonium, since they would provide an indication of the amount of fallout plutonium which might be expected in a sample, as well as how significant reactor releases might be.

We have invested a substantial amount of money and time in setting up gamma-spectrometry equipment at L-D.G.O. during the past six months, and the data we have been able to obtain during the past two months indicate that the effort will more than be justified in terms of the goals of this research. The major pieces of equipment we purchased included a Princeton Gamma Tech Ge(Li) detector and a Tracor Northern multichannel analyzer, each of which costs about $13,000. ERDA funds under this contract were used to purchase the multichannel analyzer (8000 channels) and EPA funds (EPA-R803113-01) were used to purchase the detector (about 15% efficiency). The system we have assembled is substantially more sensitive for our purposes than we originally planned to acquire, but no additional cost to ERDA over that originally proposed was incurred because approximately half of the total costs were supplied by EPA. We have encountered substantial difficulties with slow delivery times and malfunctioning of both of

the major equipment items, but we feel that after most of the bugs are worked out the system will be almost ideal for the purposes of this contract.

Table 2 summarizes data for direct gamma counting obtained in our laboratory for Cs-137, Cs-134, Co-60 and K-40 in pCi/kg. The range of Cs-137 activity we observed in surface sediments was approximately two orders of magnitude, which confirmed our impression of nonuniform accumulation rate. In some cores the Cs-137 activity dropped off by more than a factor of ten within 10 cm of the surface, while in one core collected in New York Harbor, the activity at 25 cm is comparable to that at the surface. In general, we did not observe a decrease in Cs-137 activity with increasing salinity. The highest activities were observed in marginal coves and bays, and in New York Harbor.

The samples we selected for plutonium measurement were core samples with relatively high Cs-137 activity at the surface plus the large samples representing a composite of a number of grab samples at the same location.

Plutonium Measurements

Our analytical program for plutonium is now proceeding on a routine basis after several months of experimentation to determine a dependable procedure for our laboratory. We were fortunate to have the advice of Norton Chu at HASL during this period, as well as the experience of Robert Trier, a Research Scientist in the geochemistry laboratory of Lamont who has been working on the chemistry of radium

TABLE 2

HUDSON RIVER ESTUARY SEDIMENTS

GAMMA-SPECTROMETRY DATA

CORE	DEPTH INTERVAL (cm)	SAMPLE DRY WEIGHT (g)	ACTIVITIES[a] (pCi/kg) Cs-137	Cs-134	Co-60	K-40 $\times 10^{-3}$
0.1 WM	0- 5	76.5	1210[b] ±35[c] (3%)[d]	130 ±20[e] (14%)	145 ±20 (12%)	18.2 ±0.7 (4%)
	5-10	67.7	1720 ±60 (3%)	225 ±30 (14%)	255 ±30 (12%)	18.3 ±0.8 (4%)
	10-15	73.9	1920 ±50 (3%)	275 ±35 (12%)	220 ±20 (10%)	17.5 ±0.7 (4%)
	15-20	66.1	920 ±40 (5%)	76 ±19 (26%)	59 ±23 (40%)	17.0 ±0.8 (4%)
	20-25	84.4	985 ±40 (4%)	96 ±18 (18%)	105 ±20 (20%)	16.3 ±0.7 (4%)
	25-30	92.5	545 ±30 (5%)	< background[f]	53 ±17 (32%)	15.3 ±0.6 (4%)
	30-35	62.5	890 ±50 (5%)	12 ±18 (149%)	74 ±29 (39%)	16.9 ±0.8 (5%)
	35-40	62.6	1140 ±40 (3%)	41 ±13 (32%)	120 ±20 (17%)	19.4 ±0.7 (4%)
11.5 W	0- 5	89.5	47 ±12 (26%)	11 ±9 (88%)	27 ±12 (46%)	19.8 ±0.7 (3%)
	5-10	79.7	< background	< background	32 ±31 (98%)	16.5 ±1.0 (6%)
13.2 M	0- 5	107.4	21 ±10 (47%)	< background	12 ±10 (83%)	18.2 ±0.6 (3%)
	5-10	102.5	< background	< background	< background	18.4 ±1.0 (6%)
	10-15	90.7	6 ±11 (181%)	< background	13 ±12 (88%)	19.5 ±0.7 (3%)

- table continued -

TABLE 2 continued p. 2

CORE	DEPTH INTERVAL (cm)	SAMPLE DRY WEIGHT (g)	ACTIVITIES (pCi/kg) Cs-137	Cs-134	Co-60	K-40 $\times 10^{-3}$
14.2 M[g]	0- 5	99.2	148 ±32 (21%)	51 ±23 (45%)	36 ±25 (70%)	15.7 ±1.0 (6%)
18.6-3[g]	0- 5	68.4	210 ±20 (9%)	36 ±12 (33%)	64 ±17 (26%)	14.8 ±0.6 (4%)
	5-10	36.5	170 ±30 (18%)	< background	20 ±29 (148%)	16.6 ±1.0 (5%)
	10-15	52.1	11 ±28 (258%)	< background	28 ±31 (112%)	15.8 ±1.0 (5%)
SLOSH I[h]	--	118.5	285 ±15 (6%)	30 ±8 (28%)	58 ±13 (22%)	16.7 ±0.6 (4%)
SLOSH II[i]	--	113.8	660 ±25 (4%)	59 ±11 (18%)	89 ±13 (15%)	17.4 ±0.6 (4%)
18.6-5	0- 1	101.9	580 ±25 (4%)	19 ±10 (50%)	80 ±14 (18%)	15.0 ±0.5 (4%)
	1- 3	108.1	525 ±45 (9%)	22 ±21 (95%)	86 ±30 (35%)	16.6 ±1.0 (6%)
	3- 5	113.0	410 ±20 (5%)	5 ±8 (155%)	62 ±12 (20%)	15.8 ±0.6 (4%)
21.7 M[g]	0- 7	112.2	240 ±15 (6%)	25 ±8 (30%)	65 ±11 (17%)	13.2 ±0.5 (4%)
21.9 W	0- 5	79.8	450 ±50 (11%)	15 ±28 (189%)	123 ±40 (33%)	18.9 ±1.2 (6%)
	5-10	86.0	102 ±13 (13%)	9 ±10 (111%)	24 ±12 (50%)	19.9 ±0.7 (3%)
	10-15	79.3	< background	< background	< background	19.9 ±1.1 (6%)

- table continued -

TABLE 2 continued p. 3

CORE	DEPTH INTERVAL (cm)	SAMPLE DRY WEIGHT (g)	ACTIVITIES (pCi/kg) Cs-137	Cs-134	Co-60	K-40 x10^{-3}
33.8 EC[g]	0- 5	132.3	725 ±30 (4%)	87 ±15 (17%)	180 ±20 (11%)	14.9 ±0.6 (4%)
	5-10	153.6	9 ±8 (81%)	< background	5 ±9 (159%)	12.5 ±0.4 (4%)
C43E-1	0- 5	95.3	270 ±20 (7%)	33 ±10 (32%)	73 ±14 (19%)	21.9 ±0.8 (3%)
C44[g]	0- 7	128.9	560 ±65 (12%)	73 ±31 (43%)	37 ±45 (122%)	16.7 ±1.3 (8%)
SLOSH III[j]	--	88.9	2715 ±65 (2%)	395 ±45 (11%)	330 ±25 (7%)	18.1 ±0.7 (4%)
52.5 EC	0- 5	78.9	2730 ±60 (2%)	207 ±25 (12%)	191 ±18 (9%)	19.5 ±0.7 (4%)
53.8 EC	0- 5	54.3	3370 ±90 (3%)	30 ±17 (59%)	54 ±22 (41%)	23.8 ±0.9 (4%)
	5-10	84.3	1850 ±70 (4%)	10 ±18 (189%)	32 ±23 (71%)	22.7 ±1.0 (4%)
	10-15	88.0	215 ±15 (8%)	< background	13 ±13 (100%)	21.1 ±0.7 (4%)
	15-20	91.8	27 ±18 (67%)	< background	54 ±19 (36%)	19.9 ±0.8 (4%)
FC-1	0- 5	50.0	3040 ±135 (4%)	11 ±35 (308%)	108 ±48 (44%)	23.4 ±1.5 (6%)
FC-10	0- 5	82.2	2450 ±60 (2%)	15 ±11 (74%)	26 ±15 (58%)	21.6 ±0.7 (4%)

- table continued -

TABLE 2 continued p. 4

CORE	DEPTH INTERVAL (cm)	SAMPLE DRY WEIGHT (g)	ACTIVITIES (pCi/kg) Cs-137	Cs-134	Co-60	K-40 $\times 10^{-3}$
56.4 W	0- 5	87.5	940 +35 (4%)	5 +12 (218%)	48 +16 (34%)	19.6 +0.8 (4%)
57.4 M[g]	0- 5	154.9	< background	< background	17 +8 (49%)	12.9 +0.5 (4%)
Brookhaven (soil)	--	161.4	1140 +35 (3%)	12 +8 (66%)	< background	5.4 +0.3 (6%)
NBS #4350 River sediment	--	96.4	2700 +65 (2%)	31 +16 (51%)	4000 +95 (2%)	14.6 +0.6 (4%)

a Sample specific activities are expressed in terms of dry weight. Samples range over approximately a factor of 2 or 3 in dry weight per 5-cm depth interval and in general the denser sandy samples have a lower specific activity than the lighter organic silt samples.
b Specific activities > 1000 were rounded to the nearest 10 pCi. Specific activities < 1000 but > 100 were rounded to the nearest 5 pCi. Specific activities < 100 were rounded to the nearest pCi.
c Statistical errors reported at 1 sigma. Errors for specific activities > 100 were rounded to the nearest 5 pCi. Errors for specific activities < 100 were rounded to nearest pCi.
d Errors > 10% were rounded to the nearest per cent. Errors < 10% were rounded to the nearest 0.1%.
e Error in the efficiency for Cs-134 was estimated at 10% and was determined from a plot of the detection efficiency vs. energy for K-40, Co-60 and Cs-137.
f No detectable counts above background.
g Sandy sample.
h Grab sample at mile point 18.6.
i Liquefied olive brown (oxidized) layer approximately 1-2 cm thick at the sediment water interface at mile point 18.6.
j Liquefied olive brown (oxidized) layer near Indian Point.

and thorium separations for several years. The procedure which we have finally adopted is essentially that of Wong (1971). The method consists of a nitric acid and hydrogen peroxide leach, followed by separation of plutonium from the leachate on AG1-X4 resin (Bio-Rad). The anion-exchange separations are done first with nitric acid and then with hydrochloric acid. Plutonium eluted from the final resin column is electroplated on stainless steel discs and the activity of spike Pu-236, and fallout Pu-239,240 and Pu-238 are measured by alpha-spectrometry.

Initially we had problems with Po-210 being carried through the separation. Then, as this problem was resolved, Th-228 interference became a problem. The key step to reducing the thorium contamination in our laboratory appears to have been the introduction of a hydrochloric acid column for the second anion-exchange column, a procedure which we had not followed when we used a separation procedure developed for soils (Chu, 1971). It is still not clear why our initial separations were not satisfactory since we were following a procedure which had previously been successfully employed on Hudson River Estuary sediments for us by Norton Chu at HASL.

Table 3 summarizes information on the samples and the chemical yield of plutonium obtained for Hudson River Estuary sediments. The chemical separations for eight of the listed samples (indicated by footnotes) were done at HASL, and the remainder were done in our laboratory. The alpha-spectrometry data were collected at Lamont. The data are presented with the most recently completed measurements

TABLE 3

HUDSON RIVER ESTUARY SEDIMENT PLUTONIUM MEASUREMENTS:
SAMPLE IDENTIFICATION AND CHEMICAL YIELDS[a]

SAMPLE[b]	DESCRIPTION (m.p., core depth)	DRY WEIGHT (g)	COUNTING TIME (min)	Pu-236 ADDED (dpm)	Pu-236 RECOVERED (counts)	CHEMICAL YIELD (%)
Pu-59	53.8 EC/10-15 cm	88[c]	1370	2.64	690	76
Pu-58	53.8 EC/5-10 cm	82[c]	1290	2.58	312	38
Pu-57	53.8 EC/0-5 cm	53[c]	1103	2.61	236	33
Pu-56	0.1 WM/15-20 cm	63[c]	1565	2.44	656	69
Pu-55	0.1 WM/10-15 cm	71[c]	1565	2.27	621	70
Pu-54	0.1 WM/5-10 cm	75[c]	967	2.54	218	36
Pu-53	0.1 WM/0-5 cm	74[c]	1565	2.22	548	63
Pu-52	G 18.6-5/3-4 cm	113	967	2.52	215	35
Pu-51	G 18.6-5/1-3 cm	108	401	2.67	161	60
Pu-50	G 18.6-5/0-1 cm	102	507	2.74	137	40
Pu-45	SLOSH III	100	2915	3.03	58	5
Pu-44	SLOSH III	90	1586	2.48	1170	65
Pu-43	SLOSH II	100	1586	3.09	902	74
Pu-42	SLOSH II	100	2759	2.66	1413	77
Pu-41	SLOSH II	100	5590	2.47	1701	49

- table continued -

TABLE 3 continued p. 2

SAMPLE	DESCRIPTION (m.p., core depth)	DRY WEIGHT (g)	COUNTING TIME (min)	Pu-236 ADDED (dpm)	Pu-236 RECOVERED (counts)	CHEMICAL YIELD (%)
Pu-40	SLOSH II	100	3177	2.21	635	36
Pu-39	SLOSH II	100	1185	2.42	491	68
Pu-38	SLOSH II	100	2074	2.36	308	25
Pu-35-1	F43E-1	100	2880	2.32	24	<5
Pu-35-2	Pu-236 spike	---	2966	2.32	105	
Pu-34	Pu-236 spike	---	3360	2.53	2174	>100
Pu-33	Pu-236 spike	---	2883	2.54	2848	>100
Pu-32-1	SLOSH I	100	960	2.18	--	--
Pu-32-2	Pu-236 spike	---	2782	2.18	35	>5
Pu-31	SLOSH I	100	--	--	--	--
Pu-30[d]	SLOSH I	100	2658	2.92	1390	72
Pu-29[d]	SLOSH I	100	2682	3.03	548	27
Pu-28	Pu-236 spike	---	4412	3.17	4965	>100
Pu-27[e]	SLOSH I	100	3041	2.77	576	27
Pu-26[e]	SLOSH I	100	1536	2.92	712	63

- table continued -

TABLE 3 continued p. 3

SAMPLE	DESCRIPTION (m.p., core depth)	DRY WEIGHT (g)	COUNTING TIME (min)	Pu-236 ADDED (dpm)	Pu-236 RECOVERED (counts)	CHEMICAL YIELD (%)
HR-1[d]	42 E	209	1081	2.20	221	37
HR-3[d]	44 M	241	2843	2.08	948	64
HR-8[d]	18 M	269	2902	2.14	1159	75
HR-11[d]	43 E	208	2965	2.53	1741	93

a Chemical yield calculation assumes a counting efficiency of 25%. Yields of greater than 100% represent an overestimate of the counting efficiency for that sample.

b Samples Pu-38, and Pu-40 through 59 were analyzed by the method of Wong (1971). All other samples were analyzed by the method of Chu (1971).

c Dry weight after 2 hours at 110°C. All other dry weights air-equilibrated.

d Analysis performed by Dr. Norton Chu, HASL.

e Acid leaching steps performed at L-DGO, ion-exchange and electroplating done at HASL.

listed first. Samples Pu-1 to Pu-25 were either spike experiments to test various steps in the chemical procedures or were relatively unsuccessful attempts to obtain spectra free of uranium and thorium series contamination using SLOSH I samples.

Table 4 lists the activities of Pu-239,240 and Pu-238 for the same samples for which yield data are reported in table 3. In general the statistical uncertainty is lowest in the samples which have the highest specific activities, since roughly comparable sample sizes and counting times were used for all of the samples. The measured activities of Pu-239,240 ranged over somewhat more than an order of magnitude, from about 4 to about 56 pCi/kg. The data reported for Pu-238 should be considered very preliminary, since the total activities are small and the problems of detector background and tailing of the Pu-236 spike peak have not yet been resolved in our laboratory. The reported Pu-238/Pu-239,240 ratios should likewise be treated as preliminary.

In general there is sufficient Pu-239,240 activity in Hudson River Estuary sediments to use samples of approximately 100 grams, as we have done in our initial survey.

Comparisons of Pu-239,240 and Cs-137 Distribution

Table 5 summarizes data for both Pu-239,240 and Cs-137 in core samples and surface grab samples from the Hudson River Estuary. The samples are listed in order of decreasing salinity, with the core collected at mile point 0.1 having a near-bottom salinity of 20-25 o/oo,

TABLE 4

HUDSON RIVER ESTUARY SEDIMENT PLUTONIUM MEASUREMENTS:
Pu-238 AND Pu-239,240 ACTIVITIES

SAMPLE	TOTAL COUNTS		ACTIVITIES (pCi/kg)		Pu-238 / Pu-239,240
	Pu-238	Pu-239,240	Pu-238	Pu-239,240	
Pu-59	32	195	0.4 ±0.1[a]	3.9 ±0.4	0.11 ±0.03
Pu-58	47	920	1.6 ±0.3	42.0 ±3.8	0.04 ±0.01
Pu-57	37	591	2.5 ±0.6	55.9 ±5.3	0.04 ±0.01
Pu-56	126	1387	1.7 ±0.3	37.0 ±1.8	0.05 ±0.01
Pu-55	87	1503	1.8 ±0.4	35.0 ±1.7	0.05 ±0.01
Pu-54	36	362	0.2 ±0.1	25.5 ±2.2	0.01 ±0.00
Pu-53	142	1060	0.9 ±0.3	26.5 ±1.4	0.03 ±0.01
Pu-52	17	160	0.4 ±0.2	7.6 ±0.8	0.06 ±0.03
Pu-51	15	134	0.7 ±0.3	9.3 ±1.1	0.07 ±0.03
Pu-50	26	132	1.6 ±0.3	11.8 ±1.5	0.14 ±0.03
Pu-45	--	80	--	24.3	--
Pu-44	159	1949	1.0 ±0.2	22.8 ±0.7	0.19 ±0.03
Pu-43	62	560	0.8 ±0.1	8.7 ±0.5	0.09 ±0.02

- table continued -

TABLE 4 continued p. 2

SAMPLE	TOTAL COUNTS		ACTIVITIES (pCi/kg)		Pu-238 / Pu-239,240
	Pu-238	Pu-239,240	Pu-238	Pu-239,240	
Pu-42	107	899	0.9 ±0.1	7.7 ±0.3	0.04 ±0.01
Pu-41	169	1117	0.6 ±0.1	7.4 ±0.3	0.08 ±0.01
Pu-40	56	520	0.5 ±0.1	8.2 ±0.5	0.06 ±0.02
Pu-39	Po-210 not separated				
Pu-38	--	242	--	--	--
Pu-35-1	Po-210 not separated				
Pu-35-2	Pu-236 spike experiment				
Pu-34	Pu-236 spike experiment				
Pu-33	Pu-236 spike experiment				
Pu-32-1	Po-210 not separated				
Pu-32-2	Pu-236 spike experiment				
Pu-31	Po-210 not separated				
Pu-30	--	309	--	3.0	--
Pu-29	--	104	--	2.6	--
Pu-28	Pu-236 spike experiment				

- table continued -

TABLE 4 continued p. 3

SAMPLE	TOTAL COUNTS Pu-238	TOTAL COUNTS Pu-239,240	ACTIVITIES (pCi/kg) Pu-238	ACTIVITIES (pCi/kg) Pu-239,240	Pu-238 / Pu-239,240
Pu-27	--	141	--	3.1	--
Pu-26	--	166	--	3.1	--
HR-1	55	1208	1.2	24.8	0.05
HR-3	291	4690	1.2	23.4	0.05
HR-8	139	1560	0.4	5.4	0.07
HR-11	389	7028	1.2	23.0	0.05

a The uncertainties stated represent one sigma with respect to the statistics of counting only. No estimates of other contributions to the uncertainty of the reported values are given, such as systematic errors in coring or weighing samples; or in the absolute values of primary standards.

TABLE 5

HUDSON RIVER ESTUARY SEDIMENTS

SUMMARY OF Pu-239,240 AND Cs-137 DATA

SAMPLE	DEPTH INTERVAL (cm)	Pu-239,240 (pCi/kg)	Cs-137 (pCi/kg)	$\frac{\text{Pu-239,240}}{\text{Cs-137}}$ $\times 10^2$	$\frac{\text{Cs-134}}{\text{Cs-137}}$	$\frac{\text{Co-60}}{\text{Cs-137}}$
0.1 WM	0- 5	26.5 ±1.4	1210 ±35	2.2	0.11 ±0.02	0.12 ±0.02
	5-10	25.5 ±2.2	1720 ±60	1.5	0.13 ±0.02	0.15 ±0.02
	10-15	35.0 ±1.7	1920 ±50	1.8	0.14 ±0.02	0.11 ±0.01
	15-20	37.0 ±1.8	920 ±40	4.0	0.09 ±0.02	0.06 ±0.02
	20-25	--	985 ±40	---	0.10 ±0.02	0.11 ±0.02
	25-30	--	545 ±30	---	--	--
G 18.6-5	0- 1	11.8 ±1.5	580 ±25	2.0	0.03 ±0.02	0.14 ±0.02
	1- 3	9.3 ±1.1	525 ±45	1.8	0.04 ±0.04	0.16 ±0.06
	3- 4	7.6 ±0.8	410 ±20	1.9	0.01 ±0.02	0.15 ±0.03
SLOSH I	surface[a]	3.0	285 ±15	1.0	0.11 ±0.03	0.20 ±0.04
SLOSH II	surface[a]	7.7 ±0.3	660 ±25	1.2	0.09 ±0.02	0.14 ±0.02
SLOSH III	surface[a]	22.8 ±0.7	2715 ±65	0.8	0.15 ±0.02	0.12 ±0.01

- table continued -

TABLE 5 continued p. 2

SAMPLE	DEPTH INTERVAL (cm)	Pu-239,240 (pCi/kg)	Cs-137 (pCi/kg)	Pu-239,240 / Cs-137 x10^2	Cs-134 / Cs-137	Co-60 / Cs-137
53.8 EC	0- 5	55.9 ±5.3	3370 ±90	1.7	0.02 ±0.01	0.02 ±0.01
	5-10	42.0 ±2.8	1850 ±70	2.3	0.01 ±0.02	0.02 ±0.01
	10-15	3.9 ±0.4	215 ±15	1.8	--	0.08 ±0.08
HR-8 (18M)	surface	5.4	990	0.6	--	--
HR-1 (42E)	surface	24.8	5130	0.5	--	--
HR-3 (44M)	surface	23.4	5810	0.4	--	--
HR-11 (43E)	surface	23.0	5220	0.4	--	--
NBS-SRM[b]	--	37.8	270 ±65	1.4	--	--
Brookhaven[c] composite	surface	18.9	1140[f] ±35	1.7	--	--
Eastham[d] soil	0-30	5.9	370	1.6	--	--

a See Table 2 notes.
b National Bureau of Standards - Standard Reference Material 4350. Environmental radioactivity standard: river sediment.
c ERDA-HASL standard.
d Hardy 1974.
e Hardy et al. 1972.
f H. Volchok, personal communication.

while that at mile point 53.8 is in a location that has an average salinity of less than 2 o/oo and is free of any saline water for a significant portion of the year. These two cores represent the extreme range of salinity conditions for sediment samples discussed in this report and will be referred to below as the Battery core (m.p. 0.1) and the Foundry Cove core (m.p. 53.8).

Both cores have surface activities of Pu-239,240 and Cs-137 greater than a typical surface activity for soils in this area (see table 5 for data on a Brookhaven soil sample which is a HASL intercalibration standard consisting of a composite of a large number of surface [0-5 cm] soil samples).

The Foundry Cove core has surface activities approximately twice that of the Battery core, but nearly all of the activity is confined to the upper 10 cm, while the Battery core has significant activity extending below 30 cm.

The average ratio of Pu-239,240 to Cs-137 in both cores is a little less than 2%, which is typical for fallout, and for soil data, two examples of which are given at the bottom of table 5. The NBS Environmental Radioactivity Standard: River Sediment also has a similar Pu-239,240/Cs-137 ratio. In general samples with ratios which substantially differ from fallout are the large composite surface samples collected near Indian Point in June of 1975 (SLOSH III) and the four surface samples collected in April of 1973 (HR-1, 3, 8, 11), three of which were collected within a few kilometers of Indian Point. These samples have Pu-239,240/Cs-137 ratios substantially lower (0.4 to 0.8) than fallout (about 1.8). The most

reasonable explanation of these low ratios is the presence of reactor Cs-137 in addition to that from fallout. Reactor-derived Cs-134 and Co-60 are both present in the Battery core and in SLOSH III, with similar ratios to Cs-137; while these two nuclides are much less significant, relative to Cs-137, in the Foundry Cove core collected 15 km upstream of the reactor site.

It seems reasonable to assume that some Cs-137 should be removed from Hudson sediments in water with salinities of about 70% of sea water. Lentsch (1974) has suggested that Cs-137, Cs-134, Mn-54 and Co-60 are all released to some extent from Hudson sediments near Indian Point when saline water intrudes into that region. There is also ample evidence that significant quantities of Cs-137 are added to the system from Indian Point. The general similarity of the Pu-239,240 to Cs-137 ratio of the Battery core with that of fallout, soils, and the Foundry Cove core is difficult to explain at this time. It is conceivable that some fallout Cs-137 removal has occurred, and that this has been approximately balanced by additional reactor Cs-137. Whatever the detailed processes are, it seems reasonable to conclude that the first order behavior of Pu-239,240 and Cs-137 from fallout are similar in terms of their sediment binding and particulate phase transport properties in the Hudson River Estuary, up to salinities of about 70% of sea water. Thus on the average one could predict the Pu-239,240 content of a sample of Hudson River Estuary sediment to within a factor of 2 if the Cs-137 content of the sample were measured and the fallout ratio was assumed to be typical of the sediment content.

The major deviation from this simple picture appears to be for SLOSH III collected near the reactor site in June 1975, and surface samples collected near the reactor site and downstream about 35 km in April 1973. These samples show low Pu-239,240/Cs-137 ratios, apparently due to the presence of substantial amounts of reactor Cs-137 compared with their fallout burden.

Pu-239,240 and Cs-137 Budgets in the Hudson River Estuary

It is not possible to compute an accurate budget for Pu-239,240 and Cs-137 on the basis of the limited number of samples reported here, but a crude attempt can be made to indicate the order of magnitude of the terms.

Figure 2 gives the annual delivery of fallout Cs-137 to the New York City area over the last 21 years. This figure was constructed from the annual Sr-90 delivery data at the HASL fallout collection station in New York City (U.S. Energy Research and Development Administration 1975), using a Cs-137/Sr-90 ratio of 1.5. The best available estimate of the time history of fallout Pu-239,240 to the Hudson would probably be to use the Sr-90 delivery data multiplied by a constant Pu-239,240/Sr-90 ratio. Using figure 2, the Pu-239,240 delivery rate would be about 0.015 times that for Cs-137.

The major peak of fallout Cs-137 and Pu-239,240 delivery to the Hudson was 1962-64, and the total deposition rate (not corrected for decay of Cs-137) was about 150 mCi/km^2. Assuming a mean life of 10 years, the decay-corrected Cs-137 burden at the surface should be

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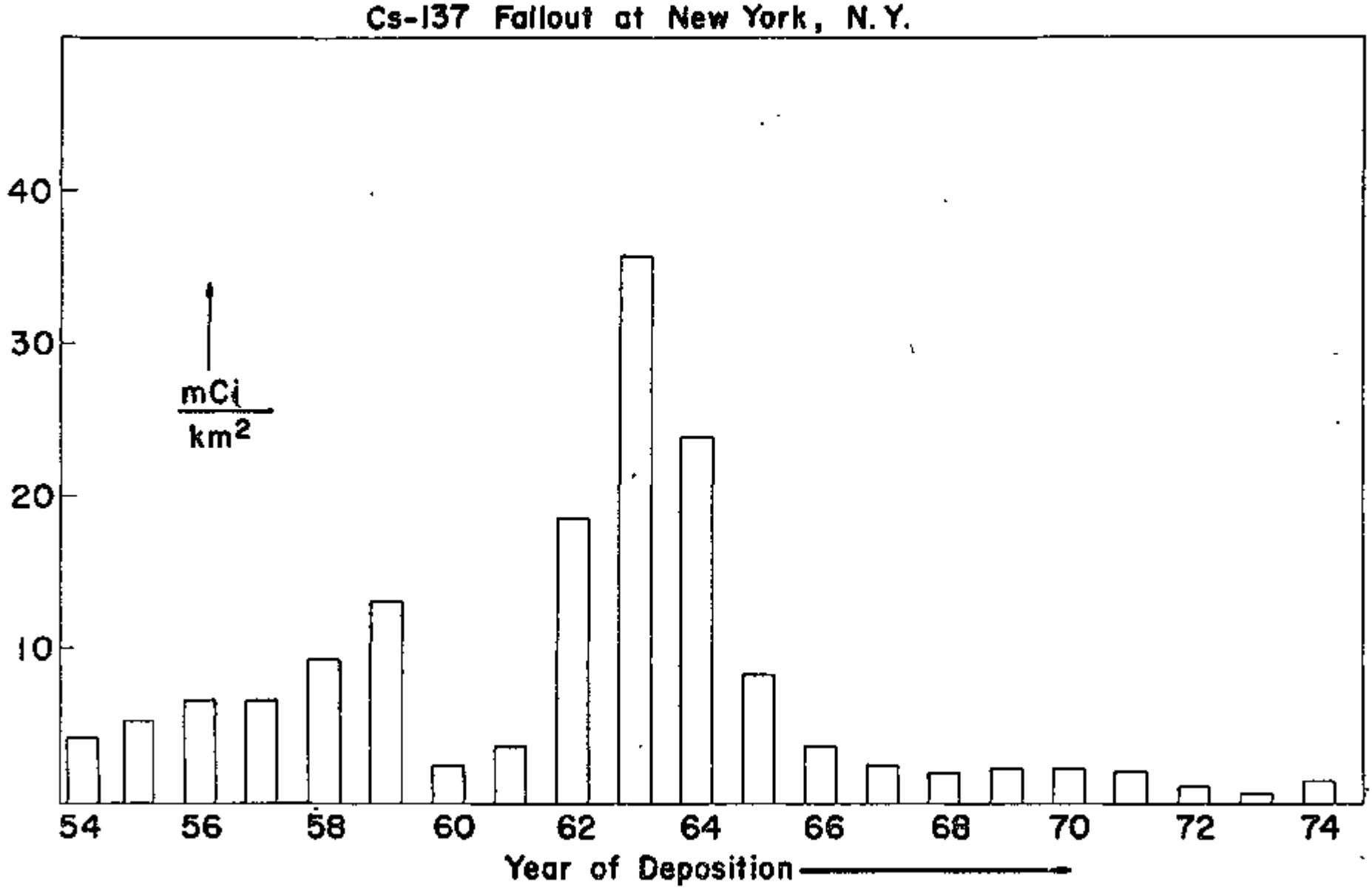
FIGURE 2. Annual deposition of Cs-137 from fallout at New York City. The basis for this plot is the Sr-90 in precipitation data collected at ERDA-HASL in lower Manhattan. The Cs-137/Sr-90 ratio is assumed to be 1.5.


Cs-137 Fallout at New York, N.Y.
40
30
20
10
mCi
km²
54
56
58
60
62
64
66
68
70
72
74
Year of Deposition

about 120 mCi/km^2. The mean decay-corrected Cs-137 burden for the latitude band of 40°-50° N is about 95 mCi/km^2 based on a Sr-90 burden of 63 mCi/km^2 (Volchok 1974). Thus the rate of fallout delivery to the New York area is 10-15% higher than the average for this latitude.

The surface area of the Hudson in the region covered by our cores is about 200 km^2 (m.p. 0-60). Thus the estimated fallout delivery to the water surface is about 20 Ci. There should be a comparable amount delivered to the surface of the Hudson between mile point 60 and the upstream end of tidal influence (m.p. 150), because the total surface area is similar to the area between mile points 0 and 60.

The contributions of the dissolved Cs-137 (and Pu-239,240) from runoff of small streams is difficult to estimate, as is the supply of radionuclides adsorbed onto soil from the drainage basin.

Assuming a suspended particulate Cs-137 activity of 1000 pCi/kg (approximately that found for the Brookhaven soil composite surface sample listed in table 5), a mean suspended load of 60 ppm and a flow rate of 500 m^3/sec for the Hudson - the suspended load could deliver about 1 Ci/yr of Cs-137. Over 10 years the supply of Cs-137 on particulates would be about 10 Ci, about half of the amount supplied directly to the water surface. No estimate of the bed load contribution from soil particulates is included in this calculation, so it should be considered a lower limit.

The drainage basin supply of dissolved Cs-137 (and Pu-239,240) is difficult to estimate. One possibility is to use the data available for Sr-90 and Cs-137 in New York City drinking water, a large fraction

of which is derived from within the Hudson drainage basin adjacent to the tidal reach of the Hudson. Table 6 summarizes the available data (U.S. Energy Research and Development Administration 1975). The tap water concentrations average 12-15% of rain water concentrations, and the Cs-137 concentration averages about 7% of the fallout ratio of 1.5. Thus if one assumes that tap water Cs-137 concentrations are at all typical of stream runoff Cs-137 concentrations, then only about 1% (0.07 x 0.15) of Cs-137 delivered to the drainage basin exclusive of the Hudson water surface would reach the Hudson in solution. This presumes that drinking water treatment procedures and reservoir impoundments do not drastically alter the dissolved Cs-137 and Pu-239,240 content. Such an assumption certainly must be more critically examined, but it is at least a starting point. The ratio of drainage basin area to water surface area in the Hudson having tidal influence is about 20-25. Thus the dissolved Cs-137 supply rate to the tidal Hudson, exclusive of direct infall, would be about 5 Ci (25/1 x 1% x 20 Ci/yr).

Thus a first order fallout budget for Cs-137 would suggest a delivery of about 40 $\pm$20 Ci to the Hudson River Estuary, about half of which could be supplied by direct precipitation on the water surface between mile points 0 and 60.

The second major source of Cs-137 to the Hudson River Estuary is from reactor releases at Indian Point. Figure 3 shows the time history of releases in terms of gross beta and gamma activity. This figure was prepared using data summarized by Lentsch (1974), Booth

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FIGURE 3. Annual gross beta-gamma releases from Indian Point (Booth 1975).

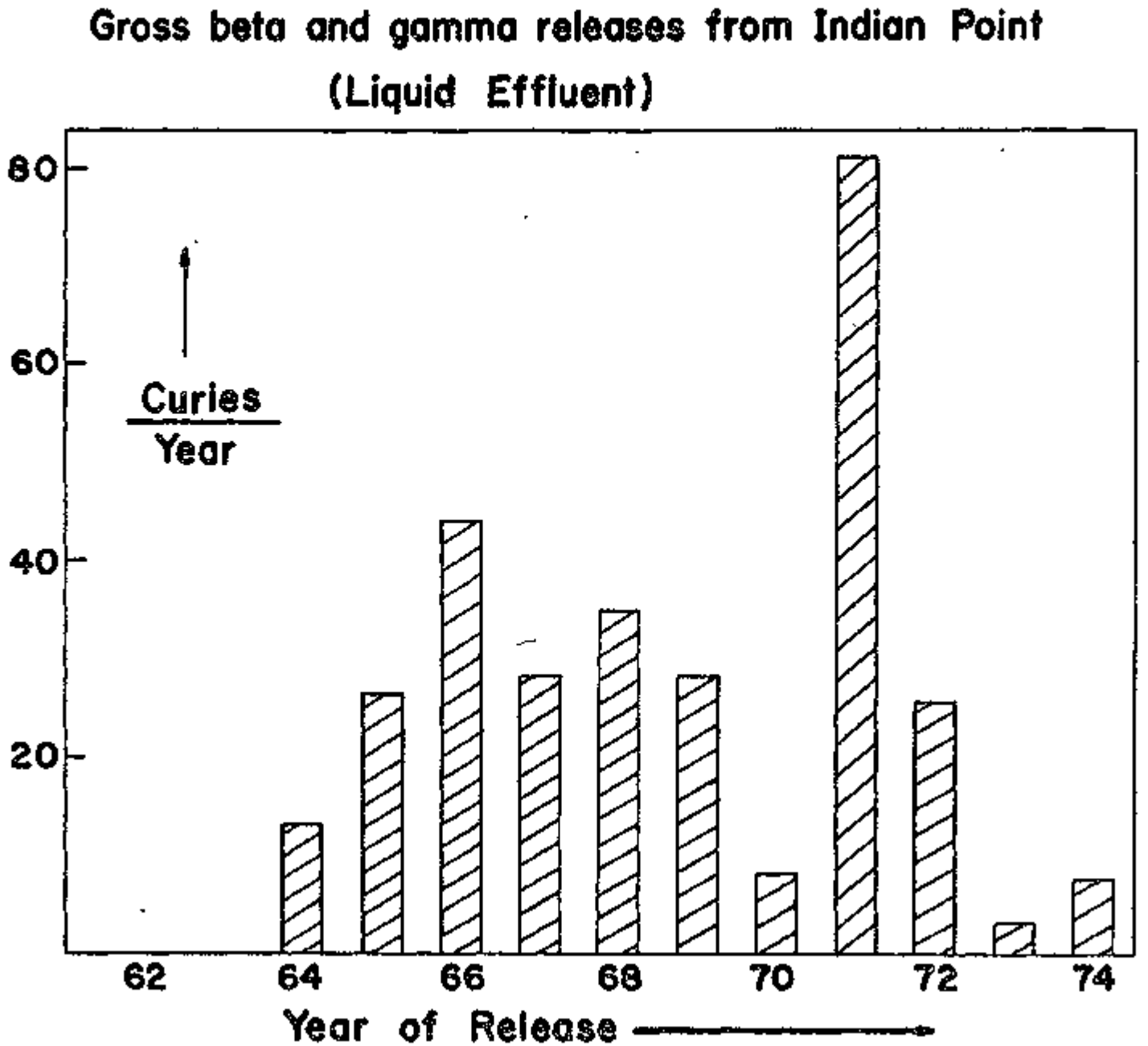
Gross beta and gamma releases from Indian Point
(Liquid Effluent)
80
60
40
20
Curies
Year
62
64
66
68
70
72
74
Year of Release

TABLE 6

Cs-137 AND Sr-90 RUNOFF IN THE HUDSON RIVER ESTUARY[a]

YEAR	Sr-90 DEPOSITION (mCi/km^2)	ANNUAL PRECIPITATION (cm)	Sr-90 RAIN (pCi/l)	Sr-90 TAPWATER (pCi/l)	Cs-137/Sr-90 TAPWATER
1954[b]	2.76	86.2	3.20	0.06	--[c]
1955	3.57	101.4	3.52	0.10	--
1956	4.43	92.1	4.81	0.15	--
1957	4.44	92.7	4.79	0.14	--
1958	6.16	104.0	5.92	0.14	--
1959	8.68	98.5	8.81	0.49	--
1960	1.58	117.8	1.34	0.47	--
1961	2.43	99.9	2.43	0.32	--
1962	12.33	94.4	13.06	0.72	--
1963	23.79	87.1	27.32	1.45	--
1964	15.85	83.8	18.92	1.79	--
1965	5.53	66.3	8.34	1.45	0.13
1966	2.43	101.3	2.40	1.11	0.16
1967	1.64	124.7	1.31	0.60	0.09
1968	1.32	110.7	1.19	0.58	0.10
1969	1.43	123.3	1.16	0.71	0.08
1970	1.48	89.6	1.65	0.67	0.11
1971	1.41	144.2	0.98	0.66	0.11
1972	0.75	170.3	0.44	0.56	0.10
1973	0.42	145.4	0.29	0.51	0.08
1974	0.93	121.1	0.77	0.50	0.10
Average 1954-1974		about 108	5.36	0.63	--
1962-1974			5.98	0.87	0.11

a Data from U.S. Energy Research and Development Administration 1975.
b January data not included.
c No Cs-137 data 1954-1964.

(1975), and the semiannual operating reports for Indian Point prepared by Consolidated Edison of New York, Inc. (1973a, 1973b, 1974a, 1974b). Figure 4 shows a similar presentation for Cs-137 releases, which were also obtained from the sources cited. The maximum releases were in 1971, about eight years after the fallout peak, and the total release of about 40 Ci is comparable to that estimated earlier for the fallout delivery.

We can reasonably estimate the fallout delivery of Pu-239,240 as about 0.6 $\pm$0.3 Ci, based on analogy with the Cs-137 budget, using an input ratio of 1.5%. There is no simple way to estimate reactor releases of Pu-239,240 since such data have not been previously collected. Two samples of primary coolant water from Indian Point were readily analyzed at HASL for a number of nuclides, including Cs-137 and Pu-239,240. The ratio of Pu-239,240 to Cs-137 from Indian Point I was about 1×10^{-3}, a factor of ten lower than fallout; while that from Indian Point II was lower by more than two or three orders of magnitude. Thus if the samples analyzed were at all representative of the material released in previous years, the amount of Pu-239,240 supplied to the Hudson from Indian Point should have been only a small fraction of that supplied from fallout (less than 10%).

The total burden of Pu-239,240 and Cs-137 in Hudson sediments can be only crudely estimated from the data listed in tables 2 and 5. The Battery core has a Cs-137 content of about 180 mCi/km^2 (1.2 pCi/g x 0.5 g/cm^3 x 30 cm) and the Foundry Cove core about 150 mCi/km^2

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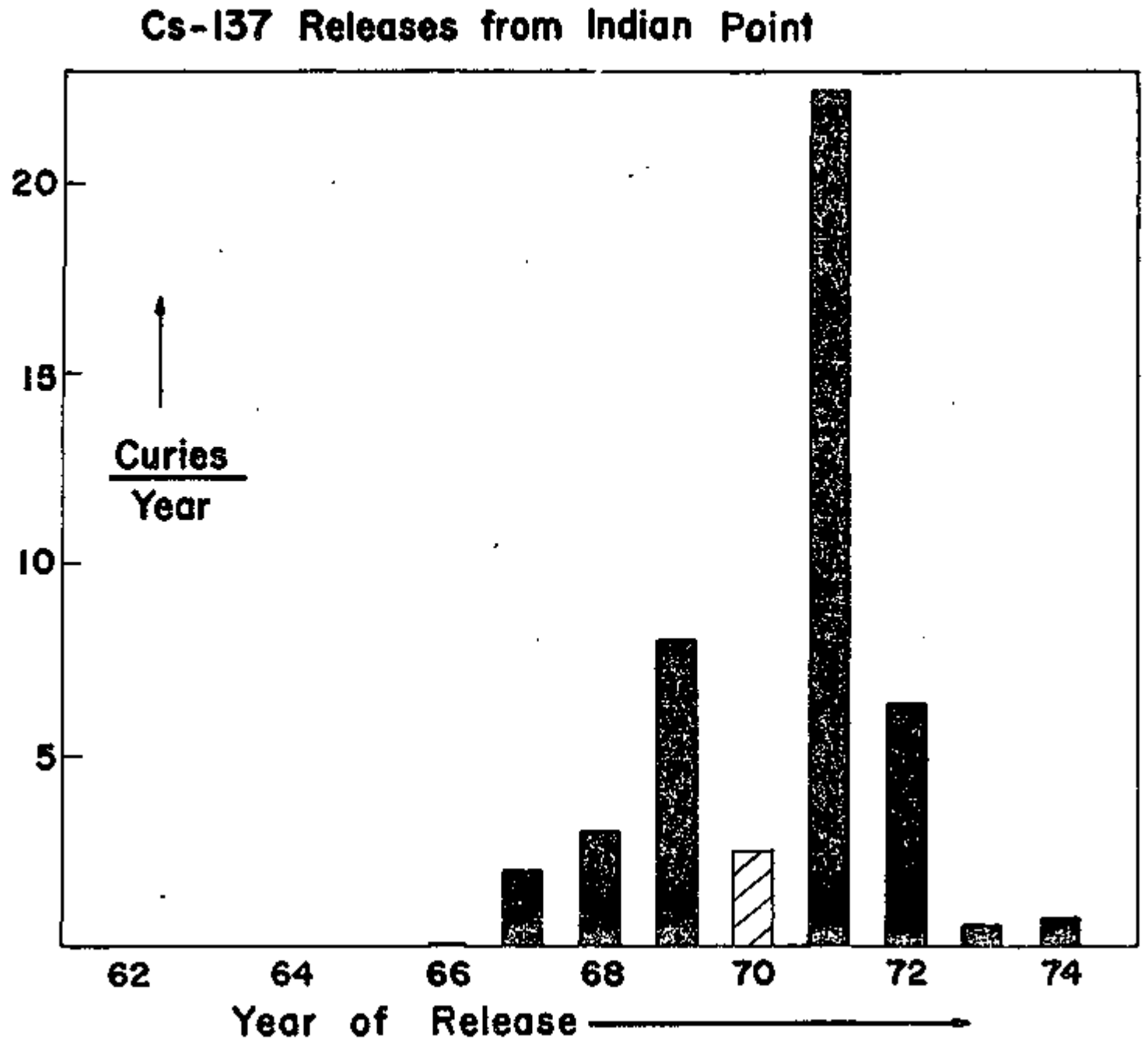
FIGURE 4. Indian Point Cs-137 releases based on the following sources: 1966-69 from Lentsch (1974), 1971-72 from Booth (1975), and 1973-74 from Consolidated Edison of New York, Inc. (1973a, b; 1974a, b). The 1970 release was estimated assuming about 25% of the gross beta-gamma release was Cs-137.


Cs-137 Releases from Indian Point
20
15
10
5
Curies
Year
62
64
66
68
70
72
74
Year of Release

(2.5 pCi/g x 0.6 g/cm x 10 cm), both of which are somewhat higher than the direct fallout deposition rate (about 120 mCi/km^2). Most of the remainder of the samples in tables 2 and 5 have surface concentrations on the order of 10-20% of the Foundry Cove core, with some sites having less than 5%.

If areas of high sediment deposition, such as that where the Battery core was collected, and marginal shallow areas such as Foundry Cove make up about 10% of the total estuary surface area and have about twice the Pu-239,240 and Cs-137 deposition rate as supplied by direct fallout, while the remainder of the estuary (90%) has a factor of 10 less deposition per unit area than the total burden of Pu-239,240 and Cs-137 now in Hudson River Estuary sediments is on the order of 40% of that supplied by direct fallout, or about 20% of that supplied by fallout from all sources.

A summary of the input budget terms for Pu-239,240 and Cs-137, as well as order of magnitude estimates of the sediment burden of these nuclides, are given in table 7. The numerical values of the listed terms should obviously only be considered as a starting point for further refinement. The terms with the best control are direct fallout (A), and reactor discharge (D). The inputs due to soil erosion (B) and dissolved runoff (C) are largely guesses, and the sediment burden terms (M & N) will require much more extensive sampling to reduce the uncertainties to within a factor of 2. The relative magnitudes of the less terms (W, X, Y and Z) cannot be even sensibly estimated at this time, and will be left without numerical values until we can constrain them in some way.

TABLE 7

HUDSON RIVER ESTUARY Pu-239,240 AND
Cs-137 BUDGET SUMMARY

	Cs-137 (Ci)	Pu-239,240 (Ci)
INPUTS		
A. Direct fallout	20	0.30
B. Soil erosion	10	0.15
C. Dissolved runoff	5	0.07
D. Reactor discharge	40	< 0.06
SEDIMENT BURDEN		
M. High sedimentation areas (10%)	4	0.06
N. Low sedimentation areas (90%)	4	0.06
LOSSES		
W. Direct dissolved outflow	?	?
X. Re-solution by sea water	?	?
Y. Suspended sediment outflow	?	?
Z. Dredging	?	?

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