

**Chlorinated Hydrocarbons and Water Quality Issues
in the New York City Municipal Supply***

**Richard F. Bopp*, Bruce L. Deck*,
H. James Simpson and Steven D. Warren**

**Lamont-Doherty Geological Observatory and
Department of Geological Sciences of Columbia University
Palisades, New York 10964**

***Also Department of Environmental Science,
Barnard College, New York, N.Y. 10027**

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Water quality of the municipal supply of New York City (NYC) is quite high by almost any standard of measurement. However, perturbations are under consideration which could significantly degrade that quality with respect to toxic chlorinated compounds. During a major drought in 1985, 7% of the total supply was derived from the Hudson River to supplement water derived primarily from the Catskill Mountains. Current plans for further expansion of the base supply for NYC envision Hudson River withdrawals at triple the rate during the 1985 drought. A second possible perturbation involves joint use of Catskill reservoirs for water supply and pumped-storage generation of electricity. Both of these proposed changes could lead to increases of toxic chlorinated compounds in finished water provided to the NYC delivery system.

New York City derives its drinking water from three groups of surface water impoundments (the Croton, Catskill and Delaware Systems), located up to 150-200 km northwest of the city (Figure 1). Drought emergency water has also been pumped from the Hudson River at Chelsea, N.Y. during two previous episodes (1965-66 and 1985). Normal treatment procedures for NYC involve only addition of fluoride and moderate chlorine doses due to generally ideal watershed characteristics including very low suspended particle concentrations. Hudson River water

receives alum and much larger chlorine doses when this source is used to supplement the base supply.

OBSERVATIONS DURING 1985 DROUGHT

Sample Collection and Analytical Program

During the northeastern USA drought of 1985, water was pumped from the Hudson River (Chelsea) into the NYC municipal supply system for a period of five months, beginning on July 10, in response to seriously depleted reservoir storage following very low runoff during the winter and spring of 1984-85. On October 29, we collected a large volume sample (40 liters) of untreated Hudson intake water for polychlorinated biphenyl (PCB) measurements and a similar sample of Hudson water after alum and chlorine addition. We also collected a 12 liter sample of untreated Hudson intake water for laboratory trihalomethane (THM) production experiments and samples of the treated Hudson water, in biological oxygen demand (BOD) bottles, for time-series THM measurements. The water temperature (16°C), withdrawal rate ($3.1 \times 10^5 \text{ m}^3/\text{day}$ with 4 pumps operating) and chlorine addition rate ($3.6 \times 10^3 \text{ kg/day}$) were typical of mid-fall operating conditions at the Chelsea station (mean Cl_2 dosage of 12 ppm). On the same day we collected 6 liters of untreated Delaware System water leaving Rondout Reservoir and entering the head of the Delaware Aqueduct, which should

be representative of the high quality supplies that dominate the NYC raw water sources during normal conditions.

A second similar set of samples (excluding PCB samples) was collected from the same three sources on November 26, a few weeks prior to termination of Hudson River withdrawals for NYC on December 11, 1985. Water temperatures were 7-9°C at the three sampling points. The Chelsea pumping rate was $3.1 \times 10^5 \text{ m}^3/\text{day}$ and chlorination rate was $3.4 \times 10^3 \text{ kg/day}$ (mean Cl_2 dosage of 11 ppm) averaged over 24 hours. At the time (9:30 AM) our treated Chelsea water samples were collected, one of the chlorine tanks was being changed and Cl_2 residual levels were 7.3 ppm, which was probably 15-20% below the average for the day.

Laboratory chlorination experiments were performed using raw water from both the Hudson River and Rondout Reservoir to provide direct comparison of the amounts of THM compounds produced from these two source types under the same experimental conditions. All of the THM measurements were made using solvent extraction procedures and detection by electron-capture gas chromatography.^{1,2}

We also measured total dissolved organic carbon (DOC) in the laboratory after filtration to remove particulate carbon, acidification and gas stripping to remove dissolved inorganic carbon, and elevated temperature

oxidation of the DOC in sealed glass vials using potassium persulfate.^{3,4} The carbon dioxide produced by this oxidation procedure was then quantified by gas chromatography using a thermal conductivity detector.

PCBs were measured on a large sample (40 liters) of untreated Hudson River water which was passed through pre-fired (400°C overnight) 0.7 micron glass fiber filters (Whatman GF/F) to provide separate determination of the dissolved and suspended particle components.⁵ A second large sample of Chelsea water that had been chlorinated and treated with alum was allowed to settle for several days. The clear water above the alum-flocculated suspended particles was then decanted and analyzed for PCBs to establish the amounts which could be expected to remain dissolved in the water following treatment.

Trihalomethane Data

Concentrations of four THM compounds (the most abundant of which was chloroform) as a function of time since collection of the treated Chelsea water samples are listed in Table I. Residual chlorine levels⁶ are also reported. Total trihalomethane (TTHM) concentrations ranged from 110 to 236 ppb in the treated samples collected on October 29, 1985 and from 98 to 189 ppb for treated samples collected on November 26, 1985. In both cases, raw Hudson River water contained less than 1 ppb

TTHM. Thus, these compounds were not present in appreciable amounts prior to chlorination, but within five hours after treatment had TTHM levels comparable to the current USEPA maximum permissible value for large municipal water supplies (100 ppb).⁷ After 200-250 hours, these samples had increased to approximately 200 ppb TTHMs. Data for the samples collected on November 26, 1985 were the most consistent (Figure 2), and illustrated continued buildup of chloroform and other THM compounds throughout a 10-day period following collection.

Two groups of laboratory incubation experiments were performed, the first, using samples collected October 29, included (1) unfiltered Chelsea raw water, (2) filtered Chelsea raw water, and (3) unfiltered Rondout raw water (Table II). Over the course of 100 hours, the TTHM levels in these three sample sets reached 191, 165, and 33 ppb, respectively. Thus, both of the Chelsea sample sets were 5-6 times higher in THMs than the Rondout samples. The filtered Chelsea samples were usually 5-15% lower than the unfiltered Chelsea set, and all three sets had monotonically increasing chloroform (CHCl_3) levels with time since chlorination began (Figure 3).

The second group of laboratory incubation experiments done with water samples collected on November 26, gave quite similar results. Unfiltered Chelsea raw water reached 197 ppb in TTHM after 100 hours, while unfiltered

Rondout raw water reached 33 ppb in TTHM. Filtered samples from Chelsea and Rondout achieved TTHM levels of 178 ppb and 35 ppb, respectively. Thus, the yield of THMs from Chelsea water was again 5-6 times greater than for Rondout water and filtered Chelsea samples were usually 10-15% lower in chloroform than unfiltered Chelsea samples.

Dissolved Organic Carbon Data

Dissolved organic carbon (DOC) concentrations in 13 samples from the Hudson River and 5 samples from the upper Delaware reservoirs and streams collected during October and November 1985 are listed in Table III. The average of 8 samples of Chelsea raw water from the NYC intake (5.57 mg/l) was approximately the same as 5 surface water samples (5.25 mg/l) collected from different sites along the Hudson River and its major tributary, the Mohawk. The average of all of these 13 samples (5.45 mg/l) was about 2.4 times the average of 5 samples from the Delaware System (2.31 mg/l). Thus, Hudson River water during late fall 1985 had 2-3 times the DOC levels of water in the Delaware watershed reservoirs and streams. This difference is consistent with the higher yield of THMs from chlorination of Hudson River water compared with Rondout Reservoirs waters, but the "efficiency" of production of THMs per unit of DOC appears to be about

twice as great for the Hudson River samples. DOC levels in the Hudson River, 2-3 times greater than those in the Delaware System water, produced 5-6 times the TTHMs during our chlorination experiments.

Polychlorinated Biphenyl Data

The largest inventory of PCBs in the Hudson River is currently found in the sediments both upstream of the Green Island Dam located about 150 km upstream of Chelsea and downstream of the dam in tidal waters. These sediments, containing several hundred tons of PCBs are distributed throughout the axis of the Hudson, beginning about 320 km upstream of New York harbor. They interact with both the water and biota of the system to sustain elevated levels of contamination, despite termination of direct releases from the major upstream industrial source more than a decade ago.⁸ Thus, water withdrawn from the Hudson at Chelsea contains PCBs originally discharged to the river far upstream of tidal waters over a period of several decades.

Total dissolved PCBs in the filtered raw water and decanted treated water from Chelsea were 17 ng/l and 14 ng/l, respectively (Table IV). To the first approximation, no appreciable removal of dissolved PCBs occurred as a result of chlorination, alum addition and settling. Total particulate phase PCBs in the raw water

from Chelsea was 26 ng/l, and the total raw water PCB concentration was 44 ng/l (Table IV). These PCBs values were derived by summing the contribution of 14 separate components distinguished by packed-column chromatography in each sample.^{8,9} Peak 11 chromatogram integrals were modified (reduced) to remove the contribution of pp'DDE, and peaks 2 and 3 in the treated sample were estimated assuming the same peak ratios relative to the average of peaks 4-15 as observed in the untreated dissolved PCB sample. In the sample receiving chlorine, there was substantial interference with the early eluting peaks due to newly synthesized chlorinated byproducts generated during treatment.

The ratio of particulate phase to dissolved total PCBs, expressed per unit weight of particles and solution was 1.14×10^5 , ranging from 0.40×10^5 for peak 2 PCB components to 4.4×10^5 for peak 13 components (Table IV). Thus, the distribution coefficients for different PCB components ranged over at least an order of magnitude. The composition of the dissolved PCBs prior to and after treatment was quite similar, indicating that sorption of PCBs to alum-flocculated particles was not significantly different, either in extent or in relative sorption efficiency for the various components, from the equilibrium already established between dissolved PCBs and suspended particle PCBs in the Hudson prior to treatment. Using the

PCB distribution coefficients in Table IV and measured surface sediment PCB concentrations in the Ashokan Reservoir of the Catskill system (20 ppb), we estimate the average dissolved PCB concentration in upstate NYC drinking water reservoirs to be about 0.18 ng/l. Thus, treated Hudson River water at Chelsea has about 80 times more dissolved PCBs than the base supply of NYC municipal water derived from the Catskill Mountains. During the 1985 drought, average dissolved PCBs in the NYC water supply were probably increased by a factor of about 6 as a result of the 7% of the total supply derived from the Hudson. Although even undiluted treated Hudson River water does not exceed federal or state standards for dissolved PCBs in drinking water, this calculation does illustrate how sensitive high quality municipal supplies can be to degradation from lower quality supplementary supplies.

OBSERVATIONS ON SOME WATER QUALITY IMPACTS OF PUMPED-STORAGE RESERVOIR OPERATIONS

The Power Authority of the State of New York (PASNY) has proposed construction of a new reservoir above and adjacent to the Schoharie Reservoir, one of NYC's drinking water reservoirs in the Catskill System. By cycling water between the two impoundments, up to 100 MW of electricity

could be generated during periods of high demand using water pumped uphill from Schoharie Reservoir with electricity produced at other localities during low demand periods (primarily at night). The closest analogue to conditions that might be expected if the above proposed project were constructed is an operating pump-storage electric-generating station, the Blenheim-Gilboa Project (BGP), located about 5 km north of Schoharie Reservoir. Water for the BGP reservoirs, which are managed exclusively for generation of electricity and are not linked to the NYC municipal water network, is primarily derived from overflow and releases from the Schoharie Reservoir just upstream. The bedrock, soil and vegetation regimes of the BGP drainage basin appear to be quite similar to those of the Schoharie and Ashokan Reservoirs of the NYC municipal supply.

Sampling and Analytical Program

We measured DOC in 15 individual samples of water collected from Schoharie and Ashokan Reservoirs (NYC Catskill System) during the first six months of 1981, using analytical procedures identical to those outlined earlier for Hudson River samples.^{3,4} The coefficient of variation of several DOC measurements for replicate samples taken in different bottles was $\pm 1.6\%$ and six replicate runs from the sample bottle was $\pm 1.5\%$. Five

surface waters from streams and ponds in the area (Dog Hill) to be submerged by the proposed new pumped-storage reservoir adjacent to Schoharie Reservoir were also collected.

For comparison to the Schoharie and Ashokan Reservoir samples, 15 samples from the BGP reservoirs were collected during the same period of months, including samples from both of the pumped storage reservoirs at various times during the pumping cycle.

Laboratory chlorination experiments were conducted on a representative group of samples from each of the above locations. Residual chlorine measurements⁶ were also made to establish that positive residuals were obtained at the levels common in municipal water supply systems in the USA. THM concentrations were measured two days after chlorination.

Results and Comparisons

Concentrations of DOC in the 17 NYC reservoir samples averaged 2.11 mg/l with a standard error of the mean of ± 0.15 mg/l. No statistically significant difference was observed between the means of the two drinking water reservoirs (Schoharie and Ashokan). Dissolved organic carbon concentrations in the BGP samples averaged 2.85 mg/l with a standard error of the mean of ± 0.11 mg/l, approximately 35% higher than for the drinking water reservoirs (Figure 4). The two populations of DOC values

are statistically distinct at the 99% confidence level, having a mean difference of 0.74 ± 0.54 mg/l (t-test).

Surface waters from the site of the proposed new reservoir had DOC concentrations which ranged from 2.7 mg/l to 9.0 mg/l, with the highest values for samples from swampy areas that are typical of a significant portion of the area.

Total trihalomethane concentrations developed after two days of exposure to chlorine in laboratory experiments ranged from approximately 30 to 350 ppb, and were strongly correlated with the DOC concentration of the sample prior to chlorination (Figure 5). While there is some indication of this correlation in the two sets of reservoir samples (NYC Catskill System and BGP reservoirs), the observed trend is predominately the result of the high DOC samples from swampy areas and is consistent with a number of other studies.¹⁰

We cannot explain with any assurance why the BGP samples had somewhat higher DOC concentrations than samples from the NYC drinking water reservoirs. The pumped-storage reservoirs experience water level changes each day of the order of 10 meters, and there are some plausible mechanisms such as enhanced sediment pore water exchange with the water column which could result in higher water column DOC concentrations. Although we do not have sufficient data to explain the cause of the

difference in DOC levels, there clearly does appear to be additional DOC in the pumped-storage reservoir waters.

IMPLICATIONS OF PROPOSED PROJECTS TO FUTURE NYC WATER QUALITY

At present there is not a consensus as to the human health significance of the concentrations of chlorinated organic compounds typically observed in treated drinking water supplies.^{11,12} The limited TTHM data available for NYC drinking water generally average <50% of the level currently permitted in the USA for large municipal supplies (100 ppb TTHMs)⁷, and it is unlikely that the water supply system as a whole would exceed the current standard even if the Hudson River water were used for 20-30% of base supply and one of the Catskill System reservoirs was operated as part of a pumped-storage project. However, each of these perturbations has clear potential to generated a considerable percentage increase in DOC of raw water supplies and of additional chlorination byproducts in treated water. For non-volatile chlorination products,¹³⁻¹⁵ it is likely that drawing 20-30% of base supply from the Hudson River would approximately double the resultant treated-water burden of such compounds in the total system. The effect of one pumped-storage project in the municipal water system would probably be considerably less, but still measurable.

Several epidemiological studies suggest higher incidences of gastrointestinal and urinary tract cancer deaths for populations in the USA drinking chlorinated vs. non-chlorinated water.^{16,17} Since THMs are only a small portion of the spectrum of chlorinated organics present in drinking waters, and the expense for removal of such compounds on the scale of a very large municipal system is so high, one of the most effective management practices would be to minimize precursor organic compounds. Thus, decisions concerning water supply perturbations should require careful consideration of possible effects on raw water DOC levels. The most prudent general policy would appear to be to work toward reduction of DOC in water supply systems¹⁸, even ones with such a large supply of high quality raw water as that of NYC. Both of the proposed perturbations of the NYC municipal water system would accomplish just the opposite effect.

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Table I. Treated Hudson River Water From Chelsea.

Time Since Cl ₂ Added (hrs)	Cl ₂ (mg/l)	CHCl ₃ (ppb) ³	CHBrCl ₂ (ppb) ²	CHBr ₂ Cl (ppb)	CHBr ₃ (ppb) ³	TTHM (ppb)
<u>Samples of October 29, 1985</u>						
0	10.4	0	0	0	0	0
5	-	98	11.0	<0.8	<0.4	110
6	-	106	12.0	<0.7	<0.4	119
144	4	213	21.9	<1.3	<0.4	236
200	-	210	13.2	<1.3	<0.4	224
<u>Samples of November 26, 1985</u>						
0	7.3	<1	<0.1	<0.1	<0.1	<1
5	6.8	94	4.0	0.3	<0.1	98
25	5.0	114	6.6	0.3	<0.1	121
50	4.9	133	8.4	0.5	<0.1	142
75	4.6	142	8.8	0.6	<0.1	152
100	4.3	153	9.9	0.6	<0.1	164
250	3.1	177	11.1	0.6	<0.1	189

Table II. Laboratory Chlorination Experiments^a
 (Samples collected October 29, 1985).

Time Since Cl ₂ Added (hrs)	Cl ₂ (mg/l)	CHCl ₃ (ppb) ³	CHBrCl ₂ (ppb) ²	CHBr ₂ Cl (ppb)	CHBr ₃ (ppb) ³	TTHM (ppb)
<u>Chelsea Raw Water (unfiltered Hudson River)</u>						
0	6.8	0	0	0	0	0
1	4.0	55	5.3	0.6	<0.4	61
5	3.1	78	7.7	0.8	<0.4	87
25	2.0	106	11.3	1.1	<0.4	118
100	0.4	162	28	1.4	<0.4	191
<u>Chelsea Filtered Water (filtered Hudson River)</u>						
0	6.8	0	0	0	0	0
1	4.2	58	5.6	0.7	<0.1	64
5	3.2	75	8.0	0.8	<0.4	84
25	2.1	103	11.3	1.1	<0.1	115
100	0.7	147	17.0	1.4	<0.1	165
<u>Rondout Raw Water (unfiltered Delaware System)</u>						
0	6.8	0	0	0	0	0
1	5.6	13	1.1	<0.2	<0.1	14
5	4.0	14	0.9	<0.1	<0.1	15
25	4.2	20	1.4	<0.2	<0.3	21
100	2.9	31	2.3	<0.2	<0.3	33

^aSamples stored at 4°C in the dark during the experiment.

Table III. Dissolved Organic Carbon Concentrations.

Sample site	Sample date (1985)	DOC (mg/l)	Sample site	Sample date (1985)	DOC (mg/l)
<u>Hudson River</u>			<u>Delaware System</u>		
Chelsea raw	10/29	6.63	Pepacton ^a	11/02	1.62
Chelsea raw	10/29	5.94	Rondout	10/29	3.00
Chelsea raw	10/29	5.56	Rondout	10/29	2.45
Chelsea raw	10/29	5.52	Rondout	10/29	1.83
Chelsea raw	10/29	5.06	Rondout	11/26	2.64
Chelsea raw	10/29	5.64	Average		2.31
Chelsea raw	10/29	4.93			
<u>Chelsea raw</u>	<u>11/26</u>	<u>5.31</u>			
Average		5.57			
Cornwall	11/20	5.42			
Highland	11/12	5.01			
Wonton Is.	11/12	4.93			
Mechanicville	11/20	5.53			
<u>Rexford^b</u>	<u>11/19</u>	<u>5.38</u>			
Average		5.25			
Average of all					
Hudson River values		5.45			

^a sample of outflow stream of the Pepacton Reservoir
^b Mohawk River sample.

Table IV. PCBs in Chelsea Water Withdrawn from the Hudson.^a

Peak # ^b	RRT ^c	Dissolved Chelsea Raw (ng/l)	Particulate Chelsea Raw (ug/g)	Dissolved Chelsea treated	Chelsea Raw Distribution Coeff. Particulate PCBs ÷ dissolved PCBs (x10 ⁵)
2	28	3.45	0.138	(2.79)	0.40
3	32	1.71	0.080	(1.38)	0.47
4	37	1.92	0.371	1.65	1.93
5	40	0.83	0.117	0.65	1.42
6	47	3.78	0.301	2.78	0.80
7	54	1.80	0.116	1.31	0.64
8	58	1.06	0.130	0.77	1.23
9	70	0.97	0.250	0.63	2.57
10	78, 84	0.59	0.130	0.60	2.20
11	98	(0.26)	(0.050)	(0.37)	(1.95)
12	104	0.43	0.084	0.63	1.95
13	125	0.20	0.088	0.19	4.43
14	146	0.20	0.072	0.16	3.58
15	174	0.15	0.043	0.08	2.98
2-15		17.4	1.97 (26.2 ng/l)	14.0	1.14

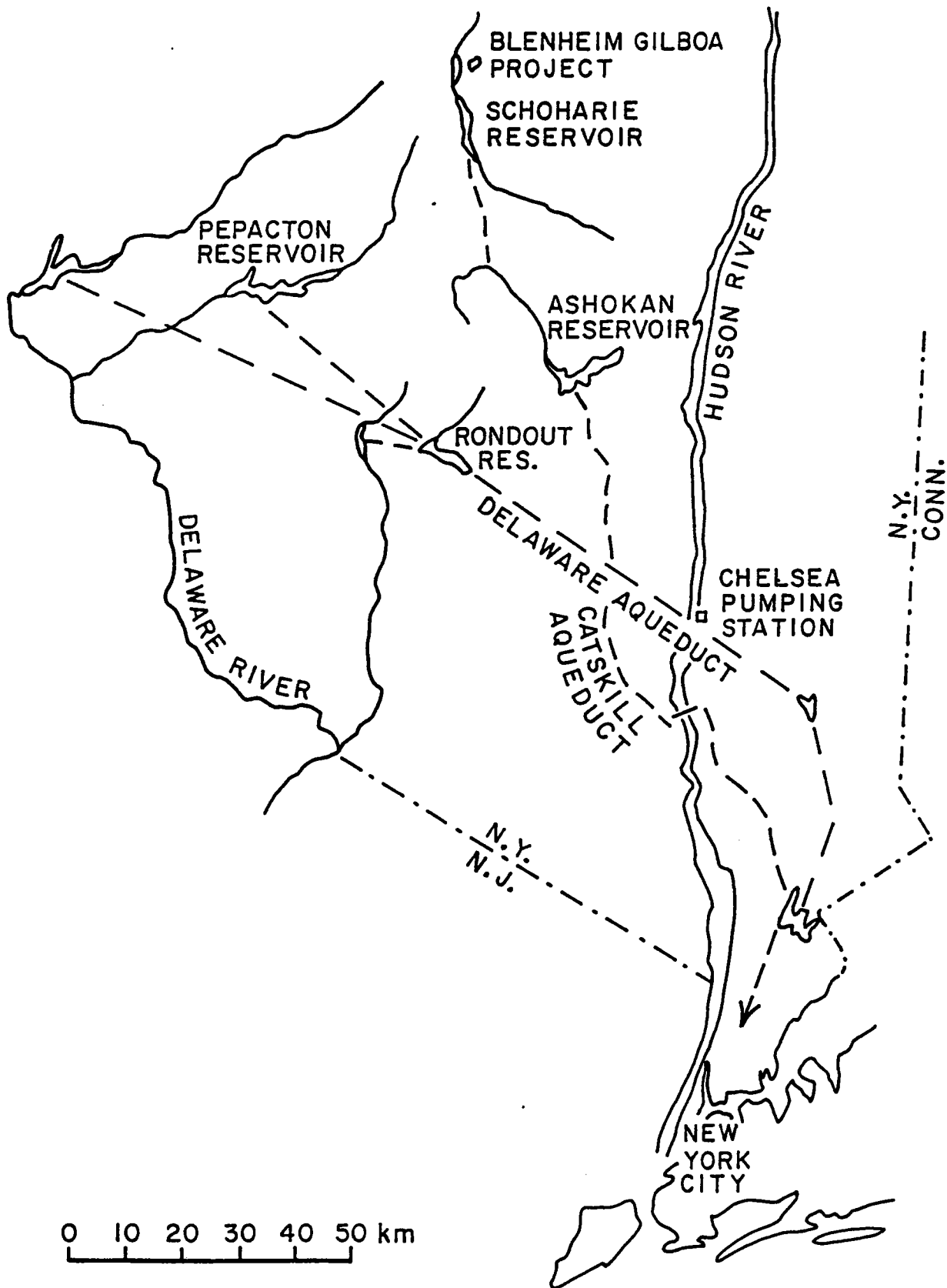
^a 13.3 mg/l suspended particles, sampled on 10/29/85.

^b Peak numbering system described elsewhere.

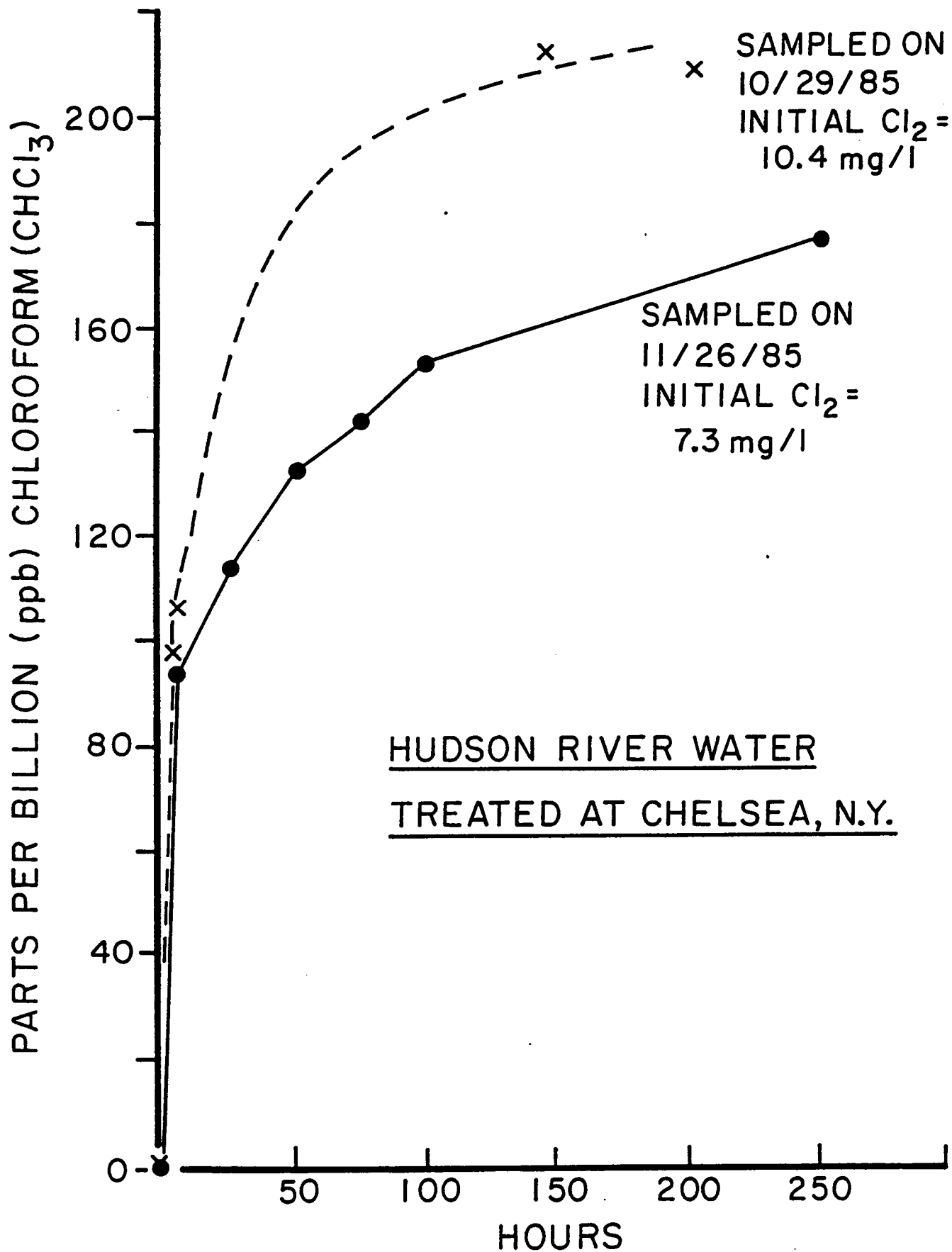
^c Retention time on 3% SE-30 column^g relative to pp'DDE = 100, measured from first appearance of solvent.

Figure Captions

- Figure 1. Municipal water supply distribution system of the City of New York, including drought emergency pumping station on the Hudson River at Chelsea. Reservoirs operated by the PAsNY for pumped storage electricity generation are located at Blenheim-Gilboa.
- Figure 2. Chloroform vs time in Hudson River water treated with alum and chlorine at Chelsea, N.Y.
- Figure 3. Chloroform vs time in laboratory chlorination experiments using Hudson River and Rondout Reservoir (Delaware System) water.
- Figure 4. Concentrations of dissolved organic carbon (DOC) in surface waters from the Catskills: open squares from the drinking water reservoirs, Schoharie and Ashokan; filled squares from the operating pumped-storage project, Blenheim-Gilboa; squares with x's from organic-rich streams and ponds at the site of the proposed pumped-storage reservoir.
- Figure 5. Concentrations of total trihalomethanes (TTHM) produced in laboratory experiments and dissolved organic carbon from Catskill surface waters: open circles from the drinking water reservoirs, filled circles from the operating pumped-storage project, x's from organic-rich streams and ponds at the site of the proposed pumped-storage reservoir.



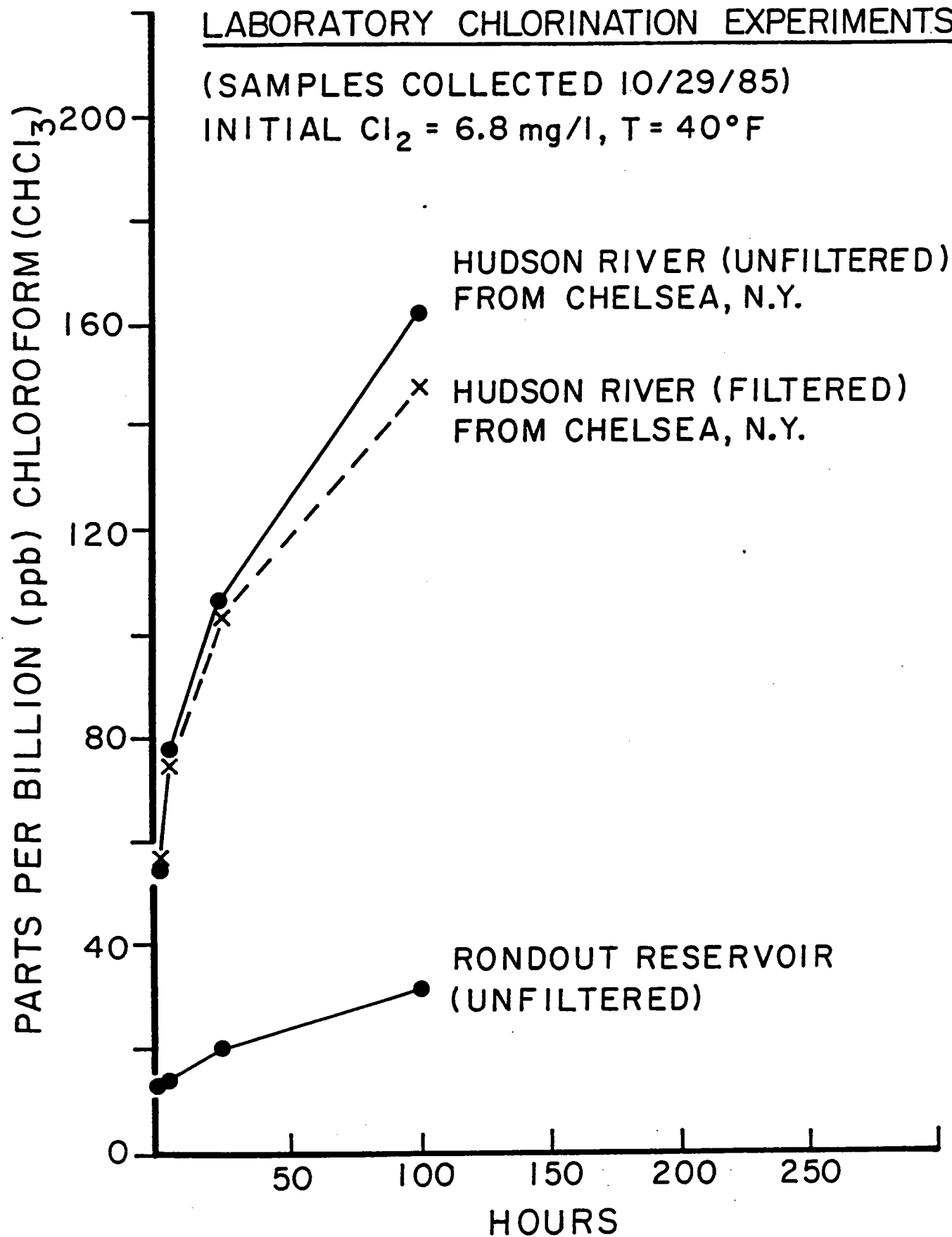
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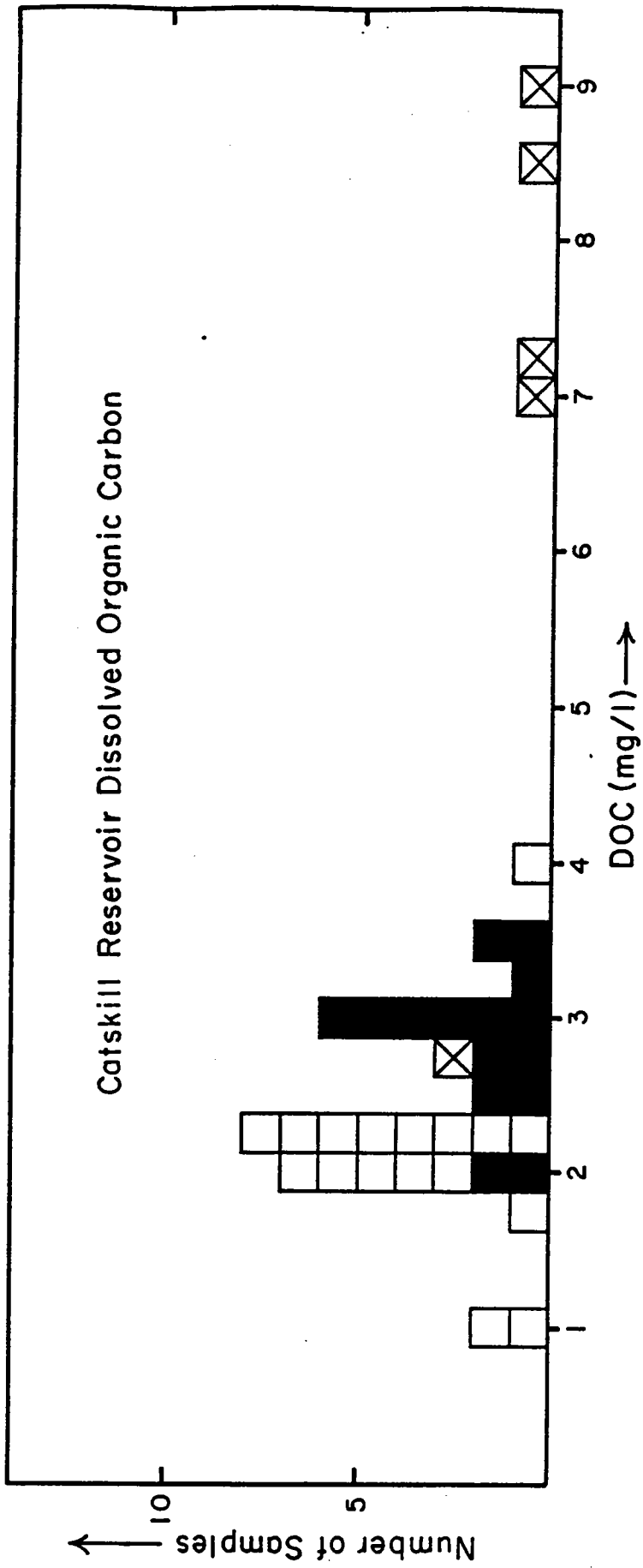
LABORATORY CHLORINATION EXPERIMENTS

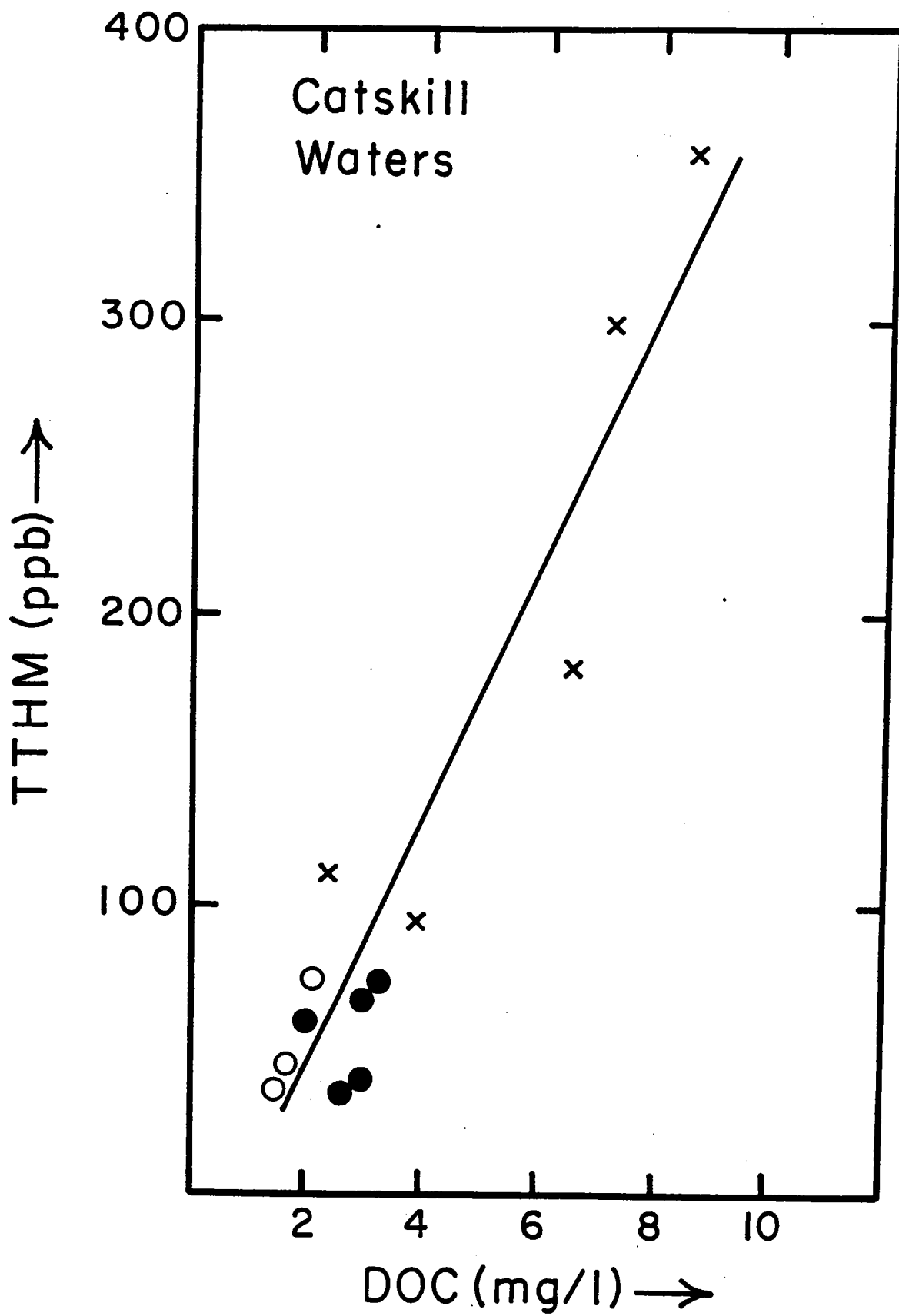
(SAMPLES COLLECTED 10/29/85)

INITIAL $\text{Cl}_2 = 6.8 \text{ mg/l}$, $T = 40^\circ\text{F}$



Catskill Reservoir Dissolved Organic Carbon







Sediment-Derived Chronologies of Persistent Contaminants in Jamaica Bay, New York¹

RICHARD F. BOPP²
H. JAMES SIMPSON
STEVEN N. CHILLRUD
DAVID W. ROBINSON
*Lamont-Doherty Geological Observatory of
Columbia University
Palisades, New York 10964*
and
*Columbia University
Department of Geological Sciences
New York, New York 10017*

ABSTRACT: Chronologies of particle-associated contaminant levels were developed for Jamaica Bay, an embayment of the Atlantic Ocean on southwestern Long Island. A major potential source of contaminants to the system is wastewater discharge, which comprises most of the freshwater input to the bay. Sediment core sections were analyzed for Cs-137, Pu-239, 240, and Be-7 activities which were then used to establish net particle accumulation rates. Samples deposited from the early 1950s through the late 1980s were analyzed for metals and chlorinated hydrocarbon contaminants. Trace metals, including copper, lead, chromium, zinc, and mercury, were elevated to levels several times those of pre-industrial concentrations. These metals decreased by about 50% between the mid 1960s and the late 1980s. Chlorinated hydrocarbon concentrations, including polychlorinated biphenyls, chlordane, and dichlorodiphenyltrichloroethane-derived compounds, decreased by a factor of five to ten between the late 1960s and the late 1980s. Local improvements in wastewater treatment and national efforts to regulate the uses and releases of specific chemicals are the most likely explanation of declines in contaminant levels in these sediments.

Introduction

Jamaica Bay (Fig. 1) is a coastal ecosystem that has been significantly impacted by development (Dewling et al. 1970; Jamaica Bay Environmental Study Group 1971). Its islands belong to the Gateway National Recreation Area system, but about three quarters of the original surrounding wetlands no longer exist, having been filled decades ago. In the eastern part of the bay, former marshes are now covered by the John F. Kennedy (JFK) International Airport. Most of the drainage basin is occupied by residential housing.

Tidal currents exchange approximately one third of the volume of Jamaica Bay each semidiurnal cycle. Freshwater inputs total approximately one half of one percent of the bay volume per day (O'Brien & Gere Engineers, Inc. 1990). Over two thirds of these inputs consist of discharge from secondary sewage treatment plants and approximately ten percent enter from combined sewer

overflow. This characteristic makes Jamaica Bay a good location for monitoring trends in contaminants derived from sewage effluent.

Jamaica Bay was chosen as one of approximately 200 coastal systems to be studied as part of the National Oceanic and Atmospheric Administration's National Status and Trends (NST) Program. Analyses of sediment grab samples indicated that Jamaica Bay was among the most contaminated systems in the study (National Oceanic and Atmospheric Administration 1988). Recently, the NST Program set out to obtain data on temporal trends in contaminant levels through the analysis of dated sediment core sections. This approach has been applied to numerous natural water systems (Alderton 1985) including several in the vicinity of Jamaica Bay (Bopp and Simpson 1989; Bopp et al. 1991). This paper presents the results of an initial NST study.

Methods

Gravity cores were collected on June 21, 1982 (JB 6) and October 1, 1988 (JB 13) from Jamaica Bay at a site adjacent to JFK International Airport (Fig. 1). The site is located in one of the deepest

¹ Lamont-Doherty Geological Observatory contribution number 5022.

² Present address: Rensselaer Polytechnic Institute, Department of Earth and Environmental Sciences, Troy, New York 12180.

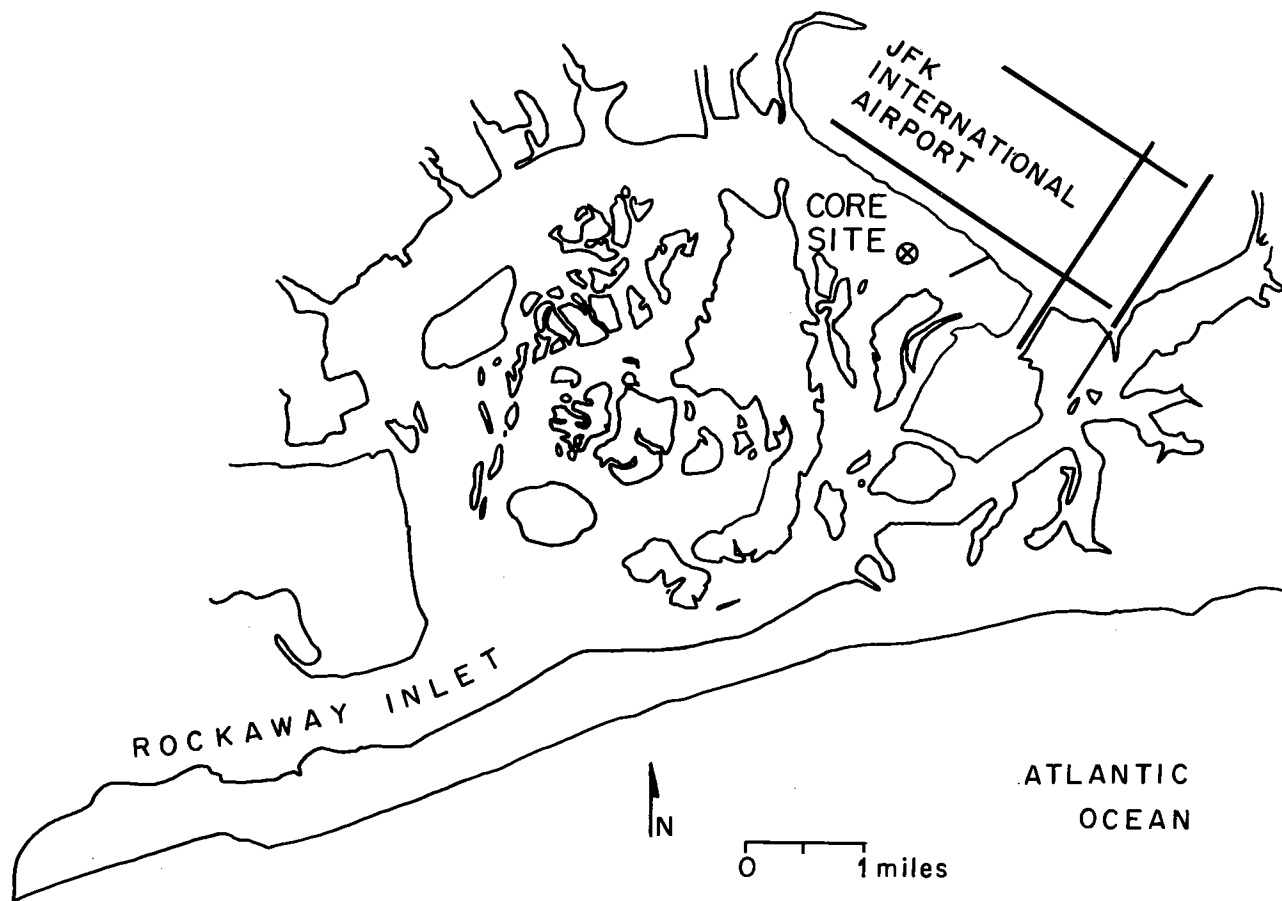


Fig. 1. Map of Jamaica Bay showing the location of our coring site near the John F. Kennedy International Airport.

parts of the bay, which was dredged at about the time of airport runway construction in 1938 (Jamaica Bay Environmental Study Group 1971). Since dredging formed an environment favorable to fine-particle deposition, it appeared likely that our cores would contain a record of the last several decades of sediment and associated contaminant accumulation. Both cores were comprised of sediments dominated by silt and clay throughout their lengths.

Cores were divided into section intervals of 2–4 cm and dried in an oven at 35°C using a flow of air passed through a column of magnesium silicate (florisil) to minimize potential atmospheric contamination of chlorinated hydrocarbons. Cs-137, K-40, and Be-7 analyses were carried out by gamma spectrometry using lithium-drifted germanium or intrinsic germanium detectors, and Pu isotopes were measured by alpha spectrometry. Reported radionuclide activities have been decay-corrected to the date of sediment core collection. Chlorinated hydrocarbon analyses were performed on subsamples (c. 1 g) of dried sediment core sections ground to a fine powder in a mortar and pestle as

described in Bopp et al. (1982). Trace metals were analyzed by atomic absorption spectroscopy following the procedures of Williams et al. (1978), modified to include digestion with HF. The only exception was Hg, which required digestion of a separate aliquot of sample in a nitric acid bomb. Hg analyses were performed by the gold amalgamation technique (Gill and Fitzgerald 1987) and were carried out in the laboratory of William Fitzgerald at the University of Connecticut. Weight loss on ignition (WLOI) was determined by heating subsamples of dried core sections overnight at 375°C. This provides an indication of the total organic matter content of the samples. Total carbon and nitrogen were determined on a commercial C-N analyzer using high temperature combustion and thermal conductivity analysis of evolved gases. The inorganic carbon content of samples was determined by acidification and measurement of the evolved CO₂ in a coulometer. Organic carbon content was calculated by subtracting the inorganic carbon from the total carbon content of the sample.

TABLE 1. Radionuclide data from sediment core JB 6 (CN 1739^a, collected June 21, 1982).

Depth (cm)	Dry Weight (g)	WLOI ^b (%)	Cs-137 ^c (pCi kg ⁻¹)	Cs-137 (mCi km ⁻²) ^d	Pu-239,240 (pCi kg ⁻¹)	Pu-239,240 (mCi km ⁻²)	Pu-239,240 Cs-137	Pu-238 (pCi kg ⁻¹)	Pu-238		K-40 (pCi g ⁻¹)	Be-7 (pCi g ⁻¹)
									Pu-239,240 (×100)			
0-2	5.12	17.3	313 ± 49	0.65	30.5 ± 2.9	0.063	0.097	1.5 ± 0.6	4.8 ± 2.1	10.4 ± 0.9	1.6 ± 0.7	
2-4	5.44	16.8	334 ± 80	0.74	40.8 ± 3.0	0.090	0.12	2.6 ± 0.7	6.3 ± 1.8	10.6 ± 1.4	0.3 ± 1.1	
4-8	12.65	16.2	252 ± 38	1.30	42.8 ± 2.3	0.220	0.17	1.6 ± 0.4	3.7 ± 0.9	12.5 ± 0.8	0.5 ± 0.6	
8-12	16.07	17.9	289 ± 34	1.89	34.6 ± 2.1	0.226	0.12			11.3 ± 0.7		
12-16	22.60	17.2	243 ± 53	2.23	43.4 ± 1.7	0.399	0.18	1.2 ± 0.2	2.8 ± 0.6	9.6 ± 1.3		
16-20	14.98	19.2	306 ± 75	1.86	71.1 ± 3.3	0.433	0.23	6.2 ± 0.8	8.1 ± 1.2	12.5 ± 1.8		
20-24	17.68	15.4	560 ± 32	4.03	97.7 ± 3.4	0.702	0.17	5.4 ± 0.6	5.5 ± 0.6	13.6 ± 0.7		
24-28	27.97	12.8	652 ± 41	7.41	142.1 ± 2.8	1.616	0.22	3.5 ± 0.2	2.4 ± 0.2	16.4 ± 0.9		
28-32	35.25	10.7	387 ± 28	5.55	102.4 ± 6.2	1.467	0.26	3.2 ± 0.7	3.1 ± 0.7	15.5 ± 0.6		
32-36	33.26	10.5	286 ± 27	3.87	87.6 ± 1.8	1.184	0.31	1.1 ± 0.1	1.2 ± 0.1	15.1 ± 0.6		
36-40	41.79	12.5	65 ± 38	1.10	24.0 ± 1.8	0.408	0.37	0.9 ± 0.3	3.7 ± 1.2	16.4 ± 1.0		
40-44	33.33	13.8	25 ± 28	0.34	5.9 ± 0.4	0.080	0.24			14.2 ± 0.7		
44-47	22.69	10.8	-1 ± 28	0.00	0.6 ± 0.1	0.006				14.1 ± 0.8		
$\overline{6.15 \text{ g cm}^{-1}}$				$\overline{31.0}$		$\overline{6.81}$	$\overline{0.22}$					

^a Control Numbers (CN) are used to identify sediment cores from the collection at the Lamont-Doherty Geological Observatory.

^b WLOI is weight loss on ignition at 375°C (16 h).

^c Uncertainties indicate ± one sigma counting statistics, including the effects of uncertainties in background regions.

^d Calculated inventories are based on the core diameter of 5.6 cm.

Results

SEDIMENT CORE DATING

Our interpretation of the history of sediment accumulation in cores JB 6 and JB 13 is based on the depth profiles of the activities of several radionuclides (Tables 1 and 2). Cs-137 and Pu-239,240, derived as fallout from atmospheric testing of nuclear weapons, first entered natural water systems on a global scale in significant amounts in the early 1950s. In cores with continuous and relatively rapid sediment accumulation (on the order of 1 cm

yr⁻¹ or greater), the maximum levels of fallout Cs-137 and Pu-239,240 can be associated with years of peak fallout delivery (mid 1960s) (Hardy 1977; Olsen et al. 1981; Environmental Measurements Laboratory 1982). An increased ratio of Pu-238 to Pu-239,240 in global fallout has been associated with the disintegration of the Pu-238 powered satellite SNAP-9A upon entering the atmosphere of the southern hemisphere in 1964. The pulse of Pu-238 reached the northern hemisphere in about 1966 and has been used as a geochronological marker for establishing that time horizon in sedi-

TABLE 2. Radionuclide data from sediment core JB 13 (CN 2057^a, collected October 1, 1988).

Depth (cm)	Dry Weight (g)	WLOI ^b (%)	Total C (%)	Inorganic C (%)	Total N (%)	Organic C/N (mol)	Cs-137 ^c (pCi kg ⁻¹)	Cs-137 (mCi km ⁻²) ^d	K-40 (pCi g ⁻¹)	Be-7 (pCi g ⁻¹)	
											0-2
2-4	8.58	15.3	7.22	0.07	0.71	11.7	96 ± 35	0.34	13.5 ± 0.9	0.4 ± 0.4	
4-6	12.05	14.0	7.05	0.12	0.72	11.2	135 ± 24	0.66	14.1 ± 0.8	0.0 ± 0.3	
6-8	10.55	12.9	6.59	0.19	0.65	11.6	168 ± 32	0.72	15.5 ± 1.0		
8-12	19.70	12.5	6.25	0.16	0.62	11.5	151 ± 22	1.21	14.4 ± 0.8		
12-16	20.33	12.6	6.32	0.17	0.61	11.8	182 ± 30	1.50	13.4 ± 0.8		
16-20	22.02	11.6	6.37	0.15	0.58	12.5	235 ± 25	2.10	13.7 ± 0.8		
20-24	18.79	14.3	7.56	0.16	0.69	12.6	271 ± 38	2.07	13.5 ± 0.9		
24-28	19.85	13.7	7.11	0.16	0.63	12.8	291 ± 25	2.35	14.5 ± 0.8		
28-32	18.49	15.0	8.48	0.19	0.73	13.3	330 ± 29	2.48	13.2 ± 0.8		
32-36	21.67	13.5	7.41	0.16	0.64	13.1	667 ± 41	5.88	14.4 ± 0.8		
36-40	27.63	11.8	5.79	0.11	0.49	13.6	408 ± 29	4.58	16.4 ± 0.9		
40-44	30.31	11.3	5.98	0.13	0.49	13.8	385 ± 25	4.74	15.2 ± 0.8		
44-48	31.83	10.7	5.31	0.28	0.45	13.0	275 ± 20	3.56	15.3 ± 0.8		
48-52	36.01	10.9	5.39	0.09	0.46	13.6	173 ± 15	2.53	15.4 ± 0.8		
52-54	26.89	9.7	4.48		0.38		83 ± 15	0.91	15.7 ± 0.9		
$\overline{6.16 \text{ g cm}^{-1}}$								$\overline{35.9}$			

^a Control Numbers (CN) are used to identify sediment cores from the collection at the Lamont-Doherty Geological Observatory.

^b WLOI is weight loss on ignition at 375°C (16 h).

^c Uncertainties indicate ± one sigma counting statistics, including the effects of uncertainties in background regions.

^d Calculated inventories are based on the core diameter of 5.6 cm.

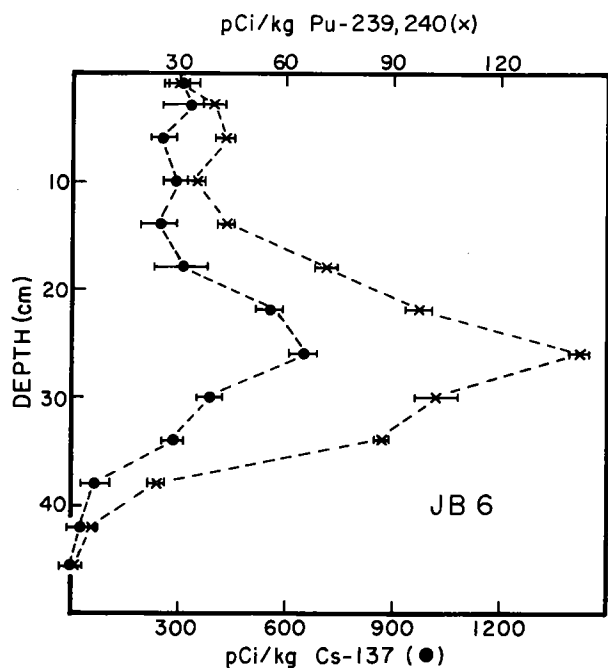


Fig. 2. Profiles of the activities of Cs-137 and Pu-239,240 as a function of depth in core JB 6. Error bars indicate one sigma counting uncertainties.

ment cores. In sediments of Lake Cayuga in western New York State, the 1966 horizon was marked by an increase in the ratio of Pu-238 to Pu-239,240 above 0.05 (Heit et al. 1986). Be-7 is a cosmic ray-produced radionuclide with a half life of 53 d. Detectable activity of Be-7 is characteristic of near-surface sediment samples containing a significant component of particles deposited within approximately 1 yr prior to sample collection.

The Cs-137 and Pu-239,240 profiles in core JB 6 are shown in Fig. 2. We associate the initial activity of these nuclides near the bottom of the core with the early 1950s and the maximum levels in the 24–28 cm section with the years of maximum fallout (1963–1964). The deepest core section with a ratio of Pu-238 to Pu-239,240 greater than 0.05 was at 20–24 cm (Table 1), consistent with deposition of this layer at approximately 1966. The location of these three horizons and the collection date of the core (1982), all indicate a net particle accumulation rate in this core of about 1.4 cm yr^{-1} . Due to possible compression of fine-grained sediments during gravity coring (Nevissi et al. 1989; Crusius and Anderson 1991), this rate could significantly underestimate the actual sediment accumulation at the site. Our estimation of the time of accumulation of core sections, however, would not be significantly affected by such an artifact since it is based on interpolations between time horizons rather than specific depths.

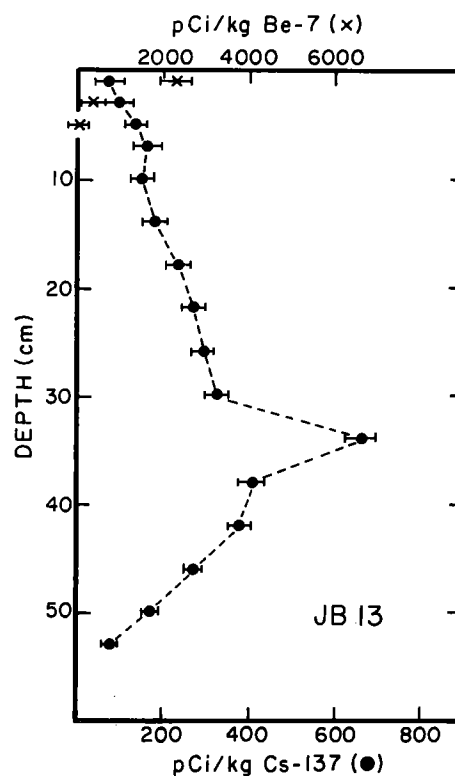


Fig. 3. Profiles of Cs-137 and Be-7 as a function of depth in core JB 13.

The Cs-137 and Be-7 profiles in core JB 13 are shown in Fig. 3. We associate the Cs-137 maximum in the 32–36 cm section with the fallout maximum of 1963–1964. We interpret the low Cs-137 activity in the bottom section of the core as an indicator of deposition in the mid 1950s. This information and the collection date of this core (1988) can be used to calculate a net sediment accumulation rate of about 1.4 cm yr^{-1} between the mid 1960s and late 1980s and a slightly higher rate, about 1.6 cm yr^{-1} , between the mid 1950s and late 1980s. These rates are consistent with the observation that activity of Be-7 more than two sigma above the counting uncertainty was found only in the 0–2 cm section. The confinement of Be-7 to the top layer also indicates that vertical mixing of the sediments on the time scale of a year does not significantly affect the distribution of particles or associated contaminants over a depth of more than a few centimeters. The two cores, JB 6 and JB 13, appear to represent a near ideal situation: relatively high rates of net particle accumulation and restricted sediment mixing yield a time resolution for particle-associated contaminant chronologies on the order of a few years.

The inventories of fallout Cs-137 and Pu-239,240 in these Jamaica Bay cores can also provide infor-

