

Twentieth Century Atmospheric Metal Fluxes into Central Park Lake, New York City

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It is generally assumed that declining atmospheric lead concentrations in urban centers during the 1970s and 1980s were due almost entirely to the progressive introduction of unleaded gasoline. However, most environmental data are from monitoring programs that began only two to three decades ago, which limits their usefulness. Here, trace metal and radionuclide data from sediment cores in Central Park Lake provide a record of atmospheric pollutant deposition in New York City through the 20th century, which suggests that leaded gasoline combustion was not the dominant source of atmospheric lead for NYC. Lead deposition rates, normalized to known Pb-210 atmospheric influxes, were extremely high, reaching maximum values ($>70 \mu\text{g cm}^{-2} \text{ yr}^{-1}$) from the late 1930s to early 1960s, decades before maximum emissions from combustion of leaded gasoline. Temporal trends of lead, zinc, and tin deposition derived from the lake sediments closely resemble the history of solid waste incineration in New York City. Furthermore, widespread use of solid waste incinerators in the United States and Europe over the last century suggests that solid waste incineration may have provided the dominant source of atmospheric lead and several other metals to many urban centers.

Introduction

Great attention has been given to the strong correlations between decreases in the consumption of leaded gasoline during the 1970s and 1980s and coincident decreases in urban atmospheric lead or human blood lead levels, with most investigators suggesting a causal relationship (1–3). However, many sources of Pb, such as lead paints, solders, and various stack emissions were reduced during this same time period.

There has been relatively little work in urban areas based on analysis of environmental samples that provide data over longer time scales (4), which might discriminate between different major sources of atmospheric lead. Sediment cores in urban lakes, from areas of semi-continuous particle accumulation, can provide archives of past deposition rates of atmospheric contaminants. To derive atmospheric contaminant fluxes from lake sediment cores, three tasks need to be accomplished: the time period of accumulation of individual depth sections must be constrained, contaminant fluxes must be normalized to account for postdepositional particle focusing, and contaminant fluxes must be shown to have been primarily derived from atmospheric inputs rather than from other pathways such as stormwater runoff. Examination of temporal trends derived from such archives can illuminate sources, fate, and transport of environmental contaminants and assist in assessment of current and historical human exposures to toxic materials via atmospheric pathways.

In remote regions, the temporal rise and fall of leaded gasoline use has been well documented. Similarities between tetra-ethyl lead production records and temporal trends of Pb deposition derived from dated environmental samples (5, 6) indicate that leaded gasoline combustion has dominated the global atmospheric lead budget during most of the 20th century. Here sediment cores from Central Park (CP) Lake, a small freshwater recreational lake in Manhattan, are used to elucidate decadal-scale trends in atmospheric fluxes of heavy metal contamination for the New York metropolitan region during the 20th century.

Methods

Four sediment cores were collected on January 6, 1996, by pushing plastic tubes through holes chopped in the ice at two sites located near the center of the two main basins of CP Lake, known locally as the "rowboat lake" and situated southwest of the larger CP Reservoir. The four cores were of similar length ($50 \pm 4 \text{ cm}$) due to a hard layer past which penetration was not possible. CP Lake has a surface area of $7.1 \times 10^4 \text{ m}^2$ with a total drainage area, all contained within the park, of $7.0 \times 10^5 \text{ m}^2$. Park roads cover $5.2 \times 10^4 \text{ m}^2$ of the watershed, and road runoff drains into the lake via a series of six drainage pipes that include sediment traps designed to limit particulate inputs into the lake.

The sediment cores were extruded, sectioned at 2-cm intervals, and dried in an oven at 35°C under a flow of air filtered through a column of Fluorasil. Dried sediments were ground and homogenized to a fine powder with a mortar and pestle. Radionuclide activities were measured by γ spectrometry using either an intrinsic Ge detector or a lithium-drifted Ge detector. Activities were decay-corrected to the sample collection date. Trace metal analyses were made by AAS or ICP-MS on aliquots of total acid digests (7).

Depth distributions of two particle-reactive radionuclides, Cs-137 and Pb-210, were used to assign approximate dates of particle accumulation to individual depth sections of sediment cores. Global fallout from atmospheric testing of nuclear weapons first delivered measurable activities of Cs-137 ($t_{1/2} = 30 \text{ yr}$) to the land surface in the Northern Hemisphere in ca. 1954 and reached a maximum in 1963 (8). Knowledge of this input history of Cs-137 allows two depth horizons within a sediment core to be assigned probable dates of deposition. The sediment layer with maximum Cs-137 activity is likely to be a more reliable indicator of

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deposition time since diffusion of dissolved Cs-137 through pore waters should not appreciably affect the depth of the Cs-137 peak (9, 10).

Pb-210 is a natural radionuclide ($t_{1/2} = 22$ yr), ultimately derived from the radioactive decay of U-238, that has been used extensively for dating sediment cores (11). One of the precursors to Pb-210 is the short-lived gas Rn-222 ($t_{1/2} = 3.8$ day). A fraction of the Rn-222 that is generated by radioactive decay of Ra-226 escapes from upper layers of soils and bedrock to the atmosphere where it decays through several short-lived daughters to Pb-210. This Pb-210 becomes attached to atmospheric particles and is subsequently stripped from the atmosphere, primarily by precipitation, to the earth's surface. Atmospherically derived Pb-210 is called *excess* Pb-210 (Pb-210_{xs}) to differentiate it from *supported* Pb-210, which is ultimately derived from decay of in situ Ra-226 atoms residing within the sediments or soils. Total Pb-210, which is measured, is transformed to Pb-210_{xs} by assuming that supported Pb-210 is in secular equilibrium with Bi-214 and Pb-214, which are also measured by γ counting of each depth section. Supported Pb-210 is thus calculated as the mean activity and associated propagated error derived from measurements on two Pb-214 γ peaks and one Bi-214 γ peak. In an idealized sediment depth profile of Pb-210_{xs}, the activity is highest at the core surface and decreases exponentially with depth.

In addition to providing the approximate accumulation "ages" of sediment layers, Pb-210_{xs} and Cs-137 have a known history of cumulative atmospheric deposition per unit area and thus can be compared to total radionuclide sediment core inventories (depth-integrated activity/unit area of sediment). Dividing observed sediment radionuclide inventories by known rates of delivery from the atmosphere can provide a first-order correction for other atmospheric contaminant inputs for the magnitude of focusing via erosion and runoff from the watershed to sediments of a particular coring location (4, 12).

Results and Discussion

Sediment Dating. Depth profiles of Cs-137 and Pb-210_{xs} (Figure 1) in all four cores are consistent with semi-continuous sediment accumulation over ca. 90–130 yr. In the core chosen for trace metal analysis, dry densities increased with depth from 0.15 g/cm³ at the core top to 0.81 g/cm³ for the penultimate depth section, with an interval of constant dry density (0.45 ± 0.01 g/cm³) between 18 and 36 cm. We do not discuss data from the deepest section of the sediment core since its dry density of 1.1 g/cm³ was substantially greater than the rest of the core and more similar to the dry density of modern Central Park soils.

Approximate dates for accumulation of individual sediment depth sections are well constrained in the upper 20 cm of the core by both the well-pronounced Cs-137 peak and small counting uncertainties for Pb-210_{xs}. The Cs-137 depth profile also indicates that physical mixing that decreases approximately exponentially with depth was not a significant process at this site, allowing the effects of such mixing on the Pb-210 age model to be largely ignored; if exponential mixing were an important process here, the difference in Pb-210 derived ages of the depth section containing the maximum Cs-137 activity and the depth section containing the first presence of Cs-137 (activity $> 2\sigma$ counting uncertainty) would have been much greater.

Best-fit linear regressions of log Pb-210_{xs} versus mass depth provide clear evidence for two mean particle accumulation rates: (i) 1.5×10^{-1} g cm⁻² yr⁻¹ for the older portions of the core (prior to ca. 1970); (ii) 5.9×10^{-2} g cm⁻² yr⁻¹ from ca. 1970 through 1995. These mean particle accumulation rates result in the penultimate depth section having accumulated

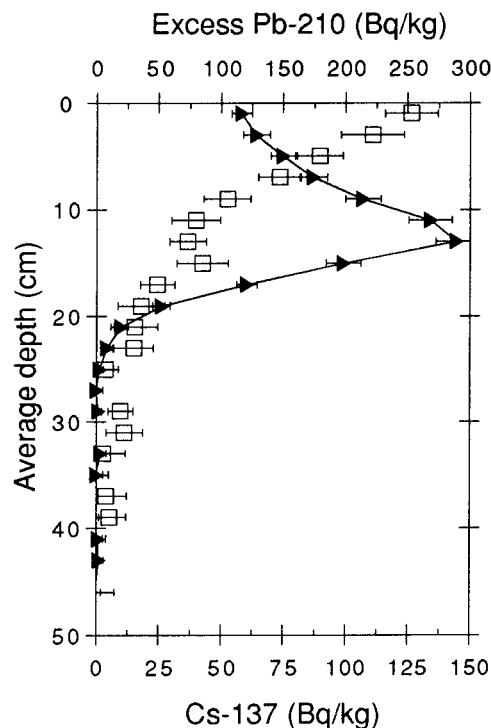


FIGURE 1. Depth profiles of Cs-137 (solid triangles) and Pb-210_{xs} (open squares) in the CP Lake sediment core selected for trace metal analyses. Three other cores collected on the same date from two locations on CP Lake had almost identical radionuclide depth profiles. Error bars indicate $\pm 1\sigma$ counting statistics. 1 Bq is 1 disintegration/s.

in the mid-1860s, consistent with the original excavation of the lake.

Independent support for this Cs-137/Pb-210 dating model for the past three decades comes from vanadium (V). V is enriched in certain fuel oils, especially the relatively inexpensive, sulfur-rich petroleums imported from Venezuela (13). In 1966, ca. 35% of the residual fuel oil used in the New York City (NYC) area was imported from Venezuela (13). Restrictions on the sulfur content of fuel oil combusted in NYC led to the phasing out of Venezuelan fuel oil imports to this area starting in October 1966 (13). This conversion to low sulfur fuel oil after 1966 is well documented in the lake sediments by V_{xs} concentrations (Figure 2).

The relatively large uncertainties in Pb-210_{xs} activities in the deeper layers of the core permit an alternative age model that indicates deposition of the penultimate depth section in the early-1900s. If this model were accurate, the sediment core would consist of particles accumulated since 1903, a year of significant dredging in CP Lake related to concerns over outbreaks of malaria (14). We have chosen to use the best fit age model that assigns the penultimate depth segment to the mid-1860s. It should be emphasized, however, that use of the alternative Pb-210 dating model would not significantly affect our major conclusions because age assignments of the most important depth sections (the top 20 cm of the core) are well constrained, with most of the age differences between the two dating models occurring in the bottom 16 cm of the core.

Trace Metals. Maximum Pb, Zn, and Sn concentrations in the CP Lake core are 57, 13, and 13 times their respective background concentrations for uncontaminated fine-grained sediments. Excess metal concentrations (total – background concentrations) display very similar trends through most of the core (Figure 3). From a depth of 12 cm to the core top, Pb_{xs} concentrations diverge slightly above the Zn_{xs} and Sn_{xs}

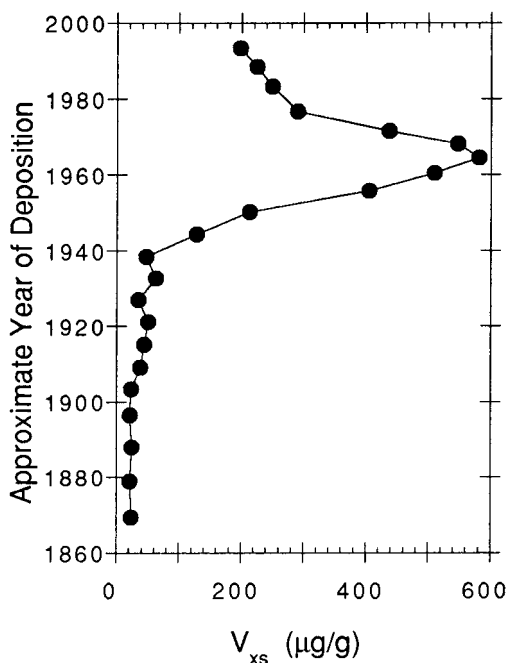


FIGURE 2. Temporal trends of excess V concentrations in the CP Lake core, assuming a background concentration of $83 \mu\text{g/g}$ in fine-grained sediments (29). V_{xs} is a tracer of high sulfur Venezuelan fuel oil use, which declined in NYC after 1966 due to restrictions on the sulfur content of fuel oil (see text).

trends. The timing of the start of this second-order deviation is about 1970, consistent with the period of maximum use of leaded gasoline in the late 1960s to early-1970s providing additional inputs of Pb to the NYC environment. It is clear, however, that these additional inputs of Pb contributed a relatively small fraction of the total Pb inputs to the site.

Anthropogenic Pb, Zn, and Sn deposition rates, normalized to the decay-corrected Pb-210_{xs} delivery and averaged over 3–6 yr (the approximate time interval represented by each core section), reached maximum values between the 1930s and early-1960s (Figure 4). The Pb and Zn deposition rates derived for the 1950s to early-1960s were $65\text{--}80 \mu\text{g cm}^{-2} \text{yr}^{-1}$. A single depth section deposited in the 1930s, which had unusually high concentrations of Pb and Sn (but not Zn), suggested a Pb deposition rate of $100 \mu\text{g cm}^{-2} \text{yr}^{-1}$.

These maximum Pb and Zn deposition rates derived from the CP Lake sediments are quite elevated as compared to published values for atmospheric fluxes in urban centers. Precipitation and aerosol samples collected in several U.S. cities between 1965 and 1975 yielded a mean Pb flux of $14.5 \pm 11.1 \mu\text{g cm}^{-2} \text{yr}^{-1}$ (4). Bulk precipitation samples collected in 1969–1970 in NYC resulted in estimated Pb and Zn fluxes of 35 and $32 \mu\text{g cm}^{-2} \text{yr}^{-1}$, respectively (15). Peak Pb deposition rates derived from sediment layers deposited during the early-to mid-1970s in two lakes in Dallas, TX, and Atlanta, GA, normalized to whole core radionuclide delivery ratios were $27\text{--}32 \mu\text{g cm}^{-2} \text{yr}^{-1}$ (4).

Based on sediment core data from more than 30 lakes and peat bogs (12, 16, 17), the Pb-210_{xs} normalized (4, 12) regional deposition of atmospheric Pb to the northeastern United States over the last 100–150 yr averages $1.0\text{--}1.3 \text{ g/m}^2$. For CP Lake, similarly calculated total deposition of Pb_{xs} is $70\text{--}100 \text{ g/m}^2$.

Although CP Lake was potentially susceptible to metal runoff inputs from park roads, several lines of evidence indicate that atmospheric inputs have dominated. Measured core inventories of both radionuclides agreed closely with independent estimates of their integrated atmospheric inputs. The Cs-137 core inventory of $4.62 \pm 0.11 \text{ kBq/m}^2$ is 1.6 times

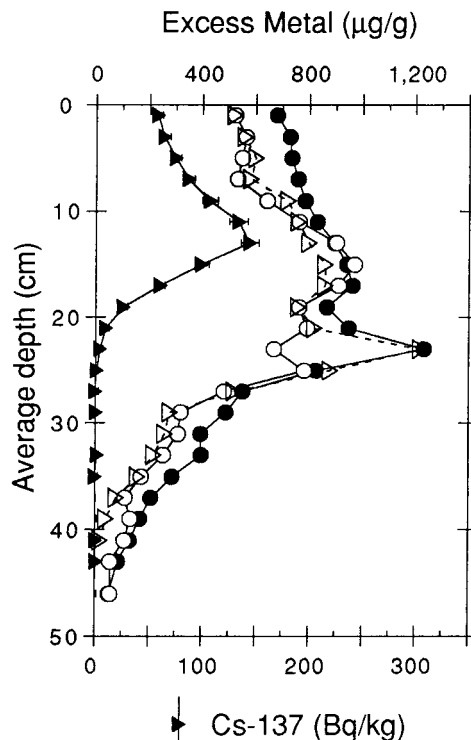


FIGURE 3. Depth profiles of average concentrations of excess metals and Cs-137 activities in the CP Lake core: Pb_{xs} (solid circles), Zn_{xs} (open circles), (20-Sn_{xs}) (open triangles), and Cs-137 (solid triangles). Natural background concentrations in fine-grained sediments were estimated as follows: Pb $\approx 26 \mu\text{g/g}$ (30); Zn $\approx 80 \mu\text{g/g}$ (31). For Sn, we assumed that background concentrations are similar to average shale value of $6 \mu\text{g/g}$ (29). Duplicate and/or triplicate digests of separate aliquots of seven of the 24 depth sections displayed relative standard deviations of $<10\%$ for Pb and $<5\%$ for Zn and Sn. The 22–24-cm sample, digested in triplicate, displayed considerably more variation, having relative standard deviations of ca. 30% for Pb, Zn, and Sn.

the decay-corrected integrated fallout measured in NYC of 2.93 kBq/m^2 (18). The Pb-210_{xs} core inventory of $5.95 \pm 0.66 \text{ kBq/m}^2$ is 1.0–1.5 times the two end-member estimates for the regional, integrated atmospheric input of Pb-210 (see Figure 4 caption).

Four soil cores from CP grasslands collected in October 1996 were analyzed for radionuclides and trace metals. Cs-137 inventories of these cores were similar to integrated atmospheric fallout, and the average Cs-137 normalized, whole-core Pb_{xs} inventory of $57 \pm 14 \text{ g/m}^2$ derived from these soil cores was quite similar to the CP Lake Cs-137 normalized inventory of 64 g/m^2 (Table 1). This similarity supports our conclusion that CP Lake sediments primarily reflect atmospheric inputs of Pb.

The occurrence of a distinct decrease in particle accumulation rate in the CP Lake core at a time of decreasing dustfall in NYC also suggests that CP Lake sediments record changes in atmospheric inputs. For the deepest sections of the core, the best-fit Pb-210 age model results in particle accumulation rates averaging $0.15 \text{ g cm}^{-2} \text{yr}^{-1}$. However, around 1970, the particle accumulation rate decreased to a value of ca. $0.06 \text{ g cm}^{-2} \text{yr}^{-1}$. Dustfall collections in NYC (19) between 1936 and 1960 yield atmospheric particle delivery rates between 0.03 and $0.05 \text{ g cm}^{-2} \text{yr}^{-1}$. The next available data, collected between 1969 and 1972, indicate dustfall rates of ca. $0.01 \text{ g cm}^{-2} \text{yr}^{-1}$.

Other Sources of Atmospheric Metals. Since inputs related to gasoline combustion appear to account for only a small portion of the Pb_{xs} in the CP Lake core, additional

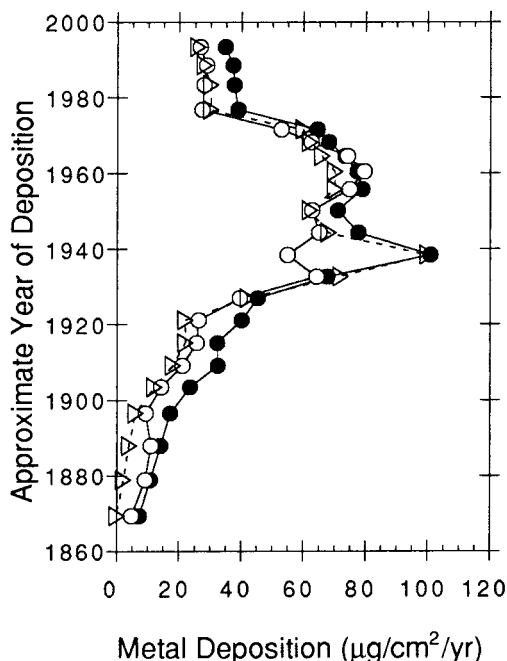


FIGURE 4. Pb (solid circles), Zn (open circles), and (20-Sn) (open triangles) deposition rates, normalized to Pb-210_{xs} delivery, as a function of time in the CP Lake core. To calculate normalized trace metal fluxes for individual core sections, we have used the equation below and the average of the two independent end-member estimates for the regional, annual flux of Pb-210. Estimates of Pb-210_{xs} inventories in the northeastern United States range from 4.09 kBq/m² (32) to 5.70 kBq/m² (12). Atmospheric fluxes derived from these inventories assuming steady state are 0.13–0.18 kBq m⁻² yr⁻¹, respectively. $M_{flux} = M_{xs}G(A/Y)D$, where M_{xs} is the excess metal concentration, G is the dry mass of the depth section, A is the area of core = 25 cm² ± 2%, Y is the number of years of deposition represented in the depth section, and D is the delivery ratio calculated as the average annual atmospheric flux of Pb-210 divided by the decay-corrected, best fit Pb-210_{xs} deposition rate for the depth section.

TABLE 1. Contaminant Pb Inventories in Central Park Lake Core CPH and Central Park Soil Cores

	Cs-137 (kBq/m ²)	Pb (g/m ²) ^a	normalized Pb (g/m ²) ^b
Central Park Lake core CPH	4.62	102	64 ^d
Central Park soil cores			
CPS-3	2.90	24.6	25
CPS-5	2.71	74.7	81
CPS-6	2.52	71.0	83
CPS-7	4.49	62.9	41
av			57
± SD/sqrt (n)			±14
atmospheric fallout	2.93 ^c		

^a Assuming 26 μg/g background. ^b Normalized to whole core Cs-137 inventory. ^c Ref 18. ^d Pb normalized to whole core Pb-210_{xs} inventory is 70–100 g/m². Pb normalized to depth-dependent Pb-210_{xs} delivery ratio (see Figure 4 caption) is 57 g/m².

sources of Pb and other metals to the atmosphere must be considered. Potentially important sources include coal combustion, metal smelting, and municipal solid waste (MSW) incineration.

Historical records indicate that coal use for space heating was predominant in NYC until about 1930 (20) and is expected

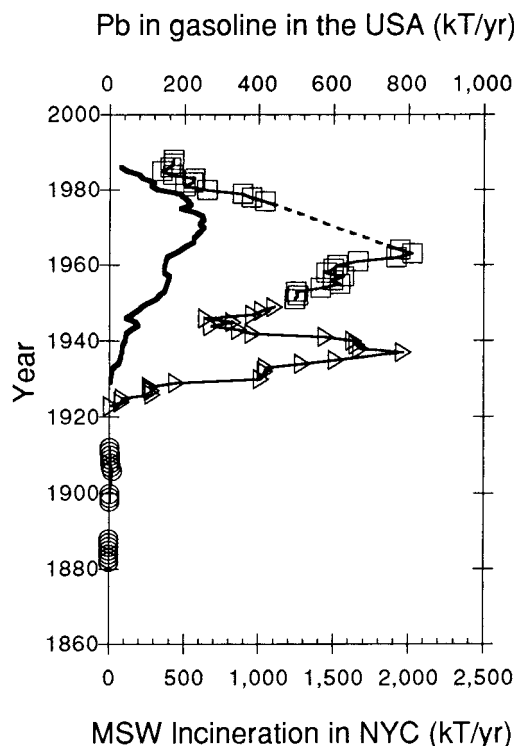


FIGURE 5. History of incineration of MSW in NYC at municipal facilities (symbols) and total production of Pb for leaded gasoline in the United States (18, 33) (heavy line). The 1930s peak in MSW incineration accounted for almost half of the total MSW disposed by NYC at that time. During the mid-1960s, MSW incineration comprised 30–40% of the total. The progressive decline in incineration rate assumed for 1964–1975 is consistent with the history of plant closings (24, 25) (see text). The primary data on MSW was reported in different units over the period of interest (20, 24): 1882–1915 (circles) cartloads; 1917–1949 (triangles) yd³; 1951–1988 (squares) short tons. Conversion factors were derived assuming the total amount of MSW disposed in NYC (data not shown) was the same in 1915 and 1917, as well as in 1949 and 1951: 2.48 yd³/cartload, 0.177 short ton/yd³, and 9.07×10^{-4} kT/short ton.

to have declined fairly soon thereafter with the transition to oil as the preferred fuel source. It has been reported (21) that coal combustion has never been a significant source of Pb aerosols in the global atmosphere. Our data support this conclusion. The history of coal use in NYC is not contemporaneous with the extended period of high Pb deposition (1930s to early-1960s) observed in the CP Lake sediments.

Primary smelting and refining of metals was also probably not a dominant source of the high Pb and Zn deposition in NYC from the 1930s to early-1960s. In the New York/New Jersey metropolitan area, major metal refining activities were concentrated in New Jersey counties adjacent to the Arthur Kill Waterway, west of Staten Island. Trace metal measurements on sediment cores from the Arthur Kill (18, 22) together with economic records of the primary refineries (23) document that maximum Pb and Zn emissions occurred from the late-1800s to the early-1930s.

The source for atmospheric metals most consistent with the data reported here for the CP Lake core is incineration of MSW. Rates of MSW incineration at municipal centers in NYC as a function of time are plotted in Figure 5. A bimodal pattern of incineration rates is observed with peak rates in the late-1930s and the mid-1960s, corresponding to two major incinerator construction programs (24). New incinerator construction in NYC was halted in 1964 (24), and subsequent Federal air pollution laws led to progressive closure of existing facilities, leaving seven operating plants by 1970 (19). An

TABLE 2. Radionuclide and Metal Data on Central Park Lake Core CPH (Control No. R1080)

depth (cm)	dry mass (g)	Cs-137 (Bq/kg)	$\pm 1\sigma$	excess Pb-210 (Bq/kg)	$\pm 1\sigma$	supported Pb-210 (Bq/kg)	$\pm 1\sigma$	av V ($\mu\text{g/g}$)	av Pb ($\mu\text{g/g}$)	av Zn ($\mu\text{g/g}$)	av Sn ($\mu\text{g/g}$)	av Cd ($\mu\text{g/g}$)	av Ag ($\mu\text{g/g}$)	av Sb ($\mu\text{g/g}$)	Cr ($\mu\text{g/g}$)	av Co ($\mu\text{g/g}$)
0–2	7.41	58	4	253	21	45	3	280	710	600	32	3.2	1.9	6.2	104	17
2–4	7.31	64	5	222	26	55	5	310	750	640	34	3.3	1.9	7.0	110	17
4–6	8.18	75	5	180	19	51	4	330	760	630	36	3.5	2.3	7.0	109	16
6–8	11.00	87	6	147	17	45	4	370	790	610	35	3.3	2.2	7.1	101	15
8–10	12.01	107	7	105	19	54	5	520	810	720	42	4.0	3.2	8.5	118	17
10–12	13.27	134	9	80	20	52	5	630	860	840	44	4.7	2.7	8.9	111	17
12–14	15.34	144	8	73	15	54	4	665	920	985	46	5.3	2.7	8.5	121	15
14–16	16.65	99	7	85	21	47	5	590	970	1050	49	5.2	2.5	8.8	128	18
16–18	19.95	60	4	49	14	59	4	490	990	990	49	4.7	2.2	8.6	134	19
18–20	22.45	26	3	36	19	53	5	300	890	845	44	3.5	1.8	6.8	110	16
20–22	23.03	10	3	30	19	58	5	210	980	870	47	3.1	1.8	7.4	108	19
22–24	21.81	4.0	2.3	30	16	49	4	130	1260	750	67	1.86	1.6	6.7	69	18
24–26	22.23	1.4	1.4	7	10	57	3	146	850	860	50	1.92	2.4	8.0	101	18
26–28	22.17	0.1	2.6	–17	17	48	5	118	580	560	32	1.27	3.6	12	85	21
28–30	23.28	0.8	1.4	19	10	48	3	134	520	400	19.8	0.88	0.98	4.2	77	20
30–32	23.60	–2.1	2.0	23	15	51	4	127	420	390	19.2	0.83	0.83	4.0	na ^a	22
32–34	22.54	1.5	2.4	5	18	55	5	121	420	340	17.0	0.73	0.67	2.9	84	20
34–36	22.54	0.1	2.6	–9	18	50	5	107	315	250	13.7	0.52	0.58	1.7	na	21
36–38	30.72	–1.4	2.1	8	16	64	5	105	240	193	9.6	0.32	0.36	1.1	75	20
38–40	34.89	–0.9	1.8	10	14	46	4	108	196	210	8.5	0.31	0.46	1.0	76	22
40–42	34.62	0.7	1.8	–5	13	48	4	105	157	192	7.3	0.28	0.37	0.8	74	21
42–44	40.43	1.0	1.9	–20	14	57	4	107	114	137	5.4	0.24	0.22	0.6	85	19
44–47	83.68	–0.2	2.2	0	15	43	4	87	79	139	4.0	0.25	0.25	0.5	76	17

^a na, not analyzed.

additional component of local solid waste combustion, residential, and commercial-building incinerators is not included in Figure 5. In NYC, between the late-1800s and early-1900s most combustible solid waste was burned together with coal for space heating (20). A NYC law requiring waste incinerators in new residential buildings resulted in construction of approximately 17 000 residential incinerators between 1951 and 1966 (19, 24). Following NYC air pollution legislation in 1966, residential incinerators were progressively closed (24, 25).

The similarities between the history of MSW incineration in NYC and accumulation of trace metals in CP Lake sediments (Figure 4) are consistent with incineration being a major source of several metals to the NYC atmosphere. As a source of metals, MSW analyzed as fly ash and/or atmospheric particulate samples from municipal incinerators is relatively enriched in Pb, Zn, and Sn (26, 27) with over 95% of the Pb leaving the incinerator stacks being less than 2 μm in diameter (26). Other metals highly enriched in refuse incineration include Sb, Ag, and Cd (26, 27), all of which were found in elevated levels (between ca. 10 and 40 times above typical background levels in fine-grained sediments) in the CP Lake core (Table 2). Refuse incineration is not a significant source of Cr or Co (26, 27), and levels of Cr and Co in the core are less than 2.1 times typical background values.

Analysis of sediments from urban lakes is one of the few methods available for assessing multi-decadal trends in atmospheric contaminant levels and thus human exposure prior to systematic environmental monitoring programs that began in the 1960s and 1970s. Observations presented here indicate that MSW combustion was probably the dominant source of Pb and several other metals to the NYC atmosphere during much of the 20th century. On the basis of widespread use of MSW incinerators over the last century (28), we propose that solid waste incineration has been generally underestimated as a source of atmospheric metals, particularly to urban centers in the United States, the U.K., and Germany. For the specific case of Pb, it appears that combustion of leaded gasoline provided a relatively small fraction of the total atmospheric burden in NYC during the 20th century.

Acknowledgments

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