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Heavy Metals in the Sediments of Foundry Cove, Cold Spring, New York

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■ The distribution of Cd and Ni in the sediments of Foundry Cove, a shallow embayment of the Hudson River, is determined as a function of distance from an industrial point-source of Cd–Ni release. Concentrations of Cd and Ni decrease smoothly by 4 and 3 orders of magnitude, respectively, to the cove margin. High values in surface sediments decrease to preindustrial background levels of 2 ppm Cd and 30 ppm Ni at sediment depths of ~20 cm. Budget calculations indicate that 20–50 tons of Cd and 10–30 tons of Ni are now present in the sediments of the cove. Cd is less mobile than Ni in the immediate vicinity of the industrial outfall due to the formation of a solid, mixed, calcium–cadmium carbonate phase from the alkaline waste discharge. Cesium-137 activity is useful in defining areas of sediment scour or recent dredging activities. Concentrations of Zn, Cu, and Pb are related to diffuse metal sources common to all recent Hudson sediments, and not to the local source of Cd and Ni. Preindustrial values of Pb, Cu, and Zn are found at approximately twice the depth for which background values of Cd and Ni are found.

Foundry Cove, a small embayment of the Hudson River, located about 60 km north of New York City (Figure 1) is a site where very high concentrations of cadmium (Cd) and nickel (Ni) in sediments are found as a result of waste discharge from a battery factory (1952–1971). Foundry Cove is shallow (1–3 m at low tide) and divided by a railroad causeway into two sections connected by a channel of about 25 m width. The Hudson River is tidal for more than 200 km upstream of New York City, with semidiurnal stage changes of 1–2 m, producing relatively vigorous tidal current flushing of both the eastern and western sections of the cove. Foundry Cove and the adjacent main channel of the Hudson contain fresh water during much of the year. Saline water (up to 3–6 ‰) is present nearly every year during low freshwater flow months, and especially during years of lower than average river discharge. A very small stream enters the eastern end of the cove, but exchange of water in the cove is completely dominated by tidal processes.

Most of the previous studies of the distribution of Cd and Ni in Foundry Cove (1–4) were related to a court-ordered dredging operation (5) whose purpose was to remove sediments from the most heavily contaminated zone in the eastern end of the eastern section of the cove. Surveys of sediment Cd and Ni in the eastern cove (2, 5) indicate significant differences in the distribution pattern before and after dredging, with the primary change being a shift in the location and a broadening of the zone of highest contamination. Research (6) characterizing the nature of the Cd and Ni solid phase in sediments near the factory outfall was also useful to this present study.

Purpose

This study was undertaken primarily to provide information on the abundance and distribution of Cd and Ni in sediments contaminated by industrial releases. Studies of the horizontal and vertical distributions of Cd and Ni in the sediments were made to put constraints on postdepositional

migration and to provide baseline data for further studies of Cd and Ni geochemistry and for assessment of a potential Cd public health hazard. Other parameters such as ¹³⁷Cs, Zn, Co, Pb, Mn, and Fe were measured to determine their relation or response to Cd and Ni contamination and movement.

Sample Collection and Analytical Procedures. During October 1974 we collected cores at 14 sites in Foundry Cove, 12 of which were in the western section of the cove (Figure 1). The core from station 15 was collected in May 1973. The cores, which were about 6 cm in diameter and up to 50 cm in length, were sectioned into 5-cm-depth intervals at the field site, transported to the lab, and frozen. Following thawing, the samples were dried overnight at 100 °C and pulverized with a mortar and pestle. Aliquots of about 2 g of the total sample weight of 80 g were leached with 20 mL of conc. HNO₃ and 5 mL of conc. HCl at room temperature for 30 min. The samples were heated slowly until most of the organic matter was destroyed, and then reduced in volume on a hot plate to ~10 mL. After cooling, 20 mL of 70% HClO₄ was added, and the sample slowly evaporated to near dryness. After cooling, 10 mL of conc. HCl and ~40 mL of distilled water were added to the white residue of sediment. Then the sediment was filtered, the residue rinsed, and the final filtrate volume brought to 100 mL in a volumetric flask.

The resultant acid solutions were analyzed by flame atomic absorption spectrometry (Perkin-Elmer 303), using mixed-metal standards for Cd, Ni, Zn, Cu, Pb, Co, Mn, and Fe. The residual sediment, which consisted of a fine white powder, was dried and weighed, and the fraction of weight loss of the original sediment sample is reported as “loss on leaching”

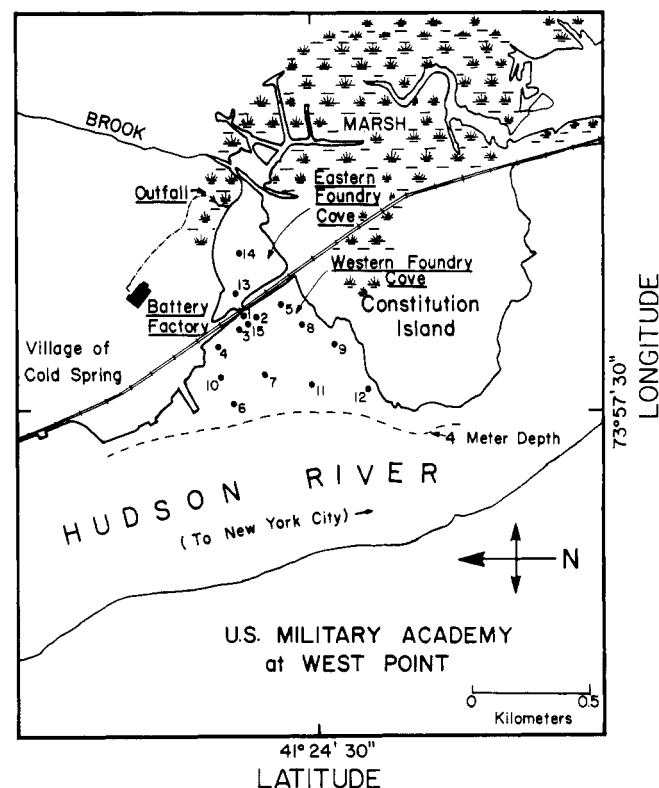


Figure 1. Location of Foundry Cove and sampling stations

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Table I. Foundry Cove Core No. 15

Depth (cm)	Cd (ppm)	Ni (ppm)	Zn (ppm)	Cu (ppm)	Pb (ppm)	Co (ppm)	Mn (ppm)	Fe (%)	Quartz (%)	LIG (%)	LOX (%)	¹³⁷ Cs (pCi/g)
0-5	258	325	316	88	147	23	752	3.7	25	9.6	25	2.47 ± 0.06
5-10	246	284	321	86	132	24	751	3.8	24	9.4	25	1.82 ± 0.07
10-15	147	120	305	80	121	20	729	3.7	2.5	9.6	24	0.21 ± 0.02
15-20	7	44	224	65	109	21	656	3.5	24	8.8	23	0.03 ± 0.02
20-25	14	49	124	31	49	20	571	3.4	29	7.1	24	0.03 ± 0.02
25-30	4	36	97	24	33	20	567	3.5	30	7.2	21	nd
30-35	2	31	90	18	25	16	546	3.5	31	6.8	21	nd
35-40	2	30	80	17	20	16	513	2.8	31	6.9	20	nd
40-45	2	33	84	18	20	16	510	3.4	29	6.9	21	nd
45-50	1	31	80	17	23	15	518	3.3	30	7.3	20	nd
50-55	2	32	82	17	23	15	524	3.5	32	7.5	21	nd
W ^a	±1.3%	±1.5%	±2.6%	±2.3%	±4.0%	±9.5%	±2.8%	±1.4%	±2.1%	±2.9%	±3.4%	...

^a W, the coefficient of variation, is the same for similar analyses in Tables II and III. W is calculated from the std. dev. σ and the mean \bar{x} as follows: $W = \sigma/\bar{x} \cdot 100\%$.

(LOL) by strong acids. Separate dry sediment samples were heated overnight to 500 °C, and the weight loss is reported as "loss on ignition" (LIG).

Small aliquots (~40 mg) of dry sediment were analyzed for quartz content by monitoring the α - β transition of quartz in a differential scanning calorimeter at 1 atm and 573 °C. After the frozen sediment samples were thawed, dried, and ground to a fine powder, and before subsampling and treatment with strong acids for metal analysis, they were placed in 100-cc aluminum cans and radioassayed for ¹³⁷Cs using a lithium-drifted germanium [Ge(Li)] gamma ray detector.

Results

Table I reports values for trace metals, ¹³⁷Cs, weight loss on leaching by strong acids (LOL), weight loss on ignition at 500 °C (LIG), and quartz content in Foundry Cove core No. 15 (all on a dry weight basis). The slight decrease in LOL values with depth in this core reflects increasing quartz content and decreasing organic content. Water bound by clays and hydrous oxides was measured with a moisture analyzer and was found to be no more than 4% of the total dry weight

of any sample. Since quartz content varies only slightly down the core, quartz normalization (7) makes no significant difference to any of the conclusions or calculations that follow.

Table II includes depth distributions of metals, ¹³⁷Cs, LOL, and LIG for the two additional cores (No.'s 6 and 10), while Table III indicates surface sediments from all stations.

Global fallout is the primary source of ¹³⁷Cs to Foundry Cove sediments, with the peak delivery by rainfall occurring between 1962 and 1965. A local nuclear power reactor located about 15 km downstream of Foundry Cove has also supplied ¹³⁷Cs to the sediments with the peak delivery from monitored, low-level releases during the early 1970's. However, reactor releases are a small fraction of the total sediment radionuclide burden in Foundry Cove (8).

The distribution of Cd and Ni in surface sediments clearly indicates that both have been transported well beyond the outfall area, through the opening in the railroad causeway, and out into the western part of the cove. Information from previous studies (1-6) combined with the data of Table III indicates that surface sediment concentrations of Cd and Ni form a "halo" decreasing smoothly by 4 and 3 orders of magnitude,

Table II. Foundry Cove Cores No. 6 and No. 10

Depth (cm)	Cd (ppm)	Ni (ppm)	Zn (ppm)	Cu (ppm)	Pb (ppm)	Co (ppm)	Mn (ppm)	Fe (%)	LIG (%)	LOX (%)
Core no. 6										
0-5	214	152	358	104	179	25	920	3.9	9.0	28
5-10	261	184	342	100	174	28	1090	4.2	9.1	28
10-15	79	115	314	87	124	22	790	4.1	8.8	27
15-20	4	44	235	67	109	19	590	3.6	8.7	25
20-25	4	42	219	59	97	19	630	3.5	7.9	25
25-30	2	31	119	28	60	15	440	2.9	5.4	19
30-35	2	30	79	15	26	14	420	3.2	4.5	18
Core no. 10										
0-5	249	356	309	85	129	23	740	4.0	8.3	24
5-10	69	119	307	85	133	20	710	3.8	8.3	25
10-15	7	59	274	82	125	20	720	3.7	8.2	24
15-20	4	56	238	70	111	19	610	3.5	8.1	24
20-25	2	42	168	45	87	18	530	3.5	(6.8	23
25-30
30-35	2	40	106	26	43	17	550	3.5	5.6	26
35-40	2	40	107	26	31	17	560	3.4	...	23

Table III. Foundry Cove Surface Sediments

Core no.	Cd (ppm)	Ni (ppm)	Zn (ppm)	Cu (ppm)	Pb (ppm)	Co (ppm)	Mn (ppm)	Fe (%)	LIG (%)	LOX (%)	¹³⁷ Cs (pCi/g)
1	417	211	320	97	169	29	710	...	9.3	30	2.13 ± 0.09
2	4	31	76	16	24	14	440	...	5.0	19	0.07 ± 0.04
3	170	179	323	86	134	22	750	...	11.5	29	1.84 ± 0.11
4	89	122	369	99	165	21	770	...	8.6	25	2.22 ± 0.13
5	118	139	388	84	119	22	710	...	8.5	28	2.78 ± 0.13
6	214	152	358	104	179	25	920	3.8	...	28	0.67 ± 0.08
7	115	108	247	95	132	22	780	...	8.5	28	1.61 ± 0.10
8	214	200	323	88	125	20	940	...	7.9	26	1.72 ± 0.07
9	301	429	324	83	130	23	830	...	7.1	25	2.25 ± 0.08
10	249	356	309	85	129	23	740	4.0	...	24	2.32 ± 0.05
11	51	105	328	83	118	22	920	...	14.9	26	1.48 ± 0.05
12	153	137	379	100	173	25	1040	...	8.9	27	2.13 ± 0.13
13	5	28	82	14	25	13	320	...	5.0	16	0.04 ± 0.03
14	908	325	362	94	158	33	690	...	7.8	33	1.83 ± 0.07
15	258	325	316	88	147	23	750	3.7	9.6	25	2.47 ± 0.06

respectively, from the outfall to the westernmost part of the cove. Surface sediment concentrations of Cd and Ni obtained in this study are consistent with previous data for the limited area of overlap in sample coverage. Surface samples from two cores (No.'s 2 and 13) show very low values of Cd and Ni, but they also have very low values of ¹³⁷Cs indicating a lack of recent sediment in these samples, probably due to either dredging or tidal scour. Both of these samples have the same fine texture as the rest of the Foundry Cove samples. The fine fraction (<62 μm) of all sediment samples ranges from 85 to 99% of dry weight with an average of 96 ± 2%.

Core No. 15 shows a dramatic drop from high surface values to constant, low, background values below 20 cm. Core No.'s 6 and 10 show similar drops, although the concentration profiles in the upper 10 cm are not identical for the three cores. All three cores have similar background concentrations of Cd and Ni (2 and 30 ppm, respectively) in the deeper parts of the sediments. The fact that Cd in preindustrial sediment from cores taken in Wisconsin lakes has been found to range from 0.2 to 2.7 ppm with an average value of 1.7 ppm (9) and that shales from the Great Plains region with organic contents >1% contain an average of 2 ppm Cd (10) indicates that the background values for Cd in Foundry Cove sediments (2 ppm) are of more than local significance. The depth distribution of ¹³⁷Cs is similar to that of Cd and Ni. This is not surprising since both inputs occurred during the same approximate time span, and both become associated with the fine fraction of the sediment. Thus, the present distribution of ¹³⁷Cs in sediments offers a convenient indicator of other contaminants (11).

Concentration depth profiles for Zn, Pb, Cu, and ¹³⁷Cs in Core No. 15 all show a similar, relatively uniform concentration in the upper 10–15 cm, followed by a broad and steady decline to background levels near 35 cm. Surface samples from Core No.'s 2 and 13 (Table III) show background levels of Zn, Pb, and Cu, strengthening the argument that these surface sediments are not of recent origin.

A convenient statistical approach to establishing interrelationships among the various parameters measured in Foundry Cove sediments is Factor Analysis (12). The data were grouped into two sets, one including 24 samples from three cores (Table I and II), and another including 12 surface samples (Table III). Through a factor analysis computer program (FACTO, in IBM System/360 Scientific Subroutine Package), a rotated factor matrix (varimax) was obtained for each set of data. The cutoff point for the eigenvalue was set as 0.1. In the varimax obtained for the data from the three

cores, the first factor is characterized by heavy loading of Zn, Cu, and Pb, likely representing diffused industrial sources of those metals. The second factor is dominated by Cd and Ni from the discharge of the battery factory. Co is moderately related to the above two factors, which is not unreasonable, since Co is supplied to the cove by diffuse industrial sources and was also used intermittently as a substitute for Ni in the battery fabrication process. The remainder of the factors are dominated by individual components, LIG, LOL, and Fe and are not relevant.

In the varimax obtained from the surface sediment data, one factor is dominated by Cd and Ni as would be expected, and a second by Cu and Pb. Surprisingly, a third factor is strongly dominated by Zn and ¹³⁷Cs. Zn and ¹³⁷Cs may follow similar pathways in an aquatic environment (both are mostly in ionic forms in solution and are absorbed on the surface of fine particles by ionic exchange), though in this case they have completely different origins. The remainder of factors are again dominated by individual components, LIG, LOL, and Mn.

Discussion

From the data reported here, we can estimate the total burden of metals released from the battery factory to the eastern part of the cove now present in the western part of Foundry Cove. As a reasonable lower limit, we estimate the amount of Cd in the sediments of the western part of Foundry Cove to be ~2 metric tons, based on the following assumptions:

- Surface area of the western part of Foundry Cove = 3×10^5 cm²
- Average depth of Cd contamination in the sediment = 20 cm
- Average surface concentration for Cd = 100 ppm
- The average concentration of Cd over the 20-cm sediment depth equals one-half of the surface concentration value
- Density of wet sediment = 1.3 g/cm³; density of dry sediment = 0.8 g/cm³.

Making the same assumptions for Ni, the total amount of Ni in the western part of Foundry Cove is also ~2 metric tons.

Using previously published data (1–3) for surface sediment concentrations of Cd from the eastern, more highly contaminated area of Foundry Cove, and assuming the same type of depth profile as found for our cores in the western part of the

cove, the total Cd content of the eastern section of the cove, including the highly contaminated outfall area, is 17 metric tons. The total Cd present in all the sediments of Foundry Cove would then be 20 metric tons. Both of these calculations should be considered as a minimum estimate of the present sediment metal burden because surface sediment values were chosen very conservatively. For example, all surface sediments between the 10^3 and 10^4 ppm contours were assigned the lowest value of 10^3 ppm. Furthermore, there is substantial uncertainty about the depth at which Cd reaches background values in the eastern part of the cove. Some data from court records (5) indicate that Cd penetration may exceed 50 cm in many locations. Our budget calculation assumed a mean depth of contamination of 20 cm as we observed from cores taken in the western part of the cove. Using values for the surface sediment concentrations interpolated from our data and other published data and estimating the penetration depth to be 30 cm for sediments in the eastern part of the cove, the total Cd burden in Foundry Cove would be 50 metric tons. Thus, the range of estimates of total Cd in the cove suggested here is 20–50 metric tons, with 4–10% of the total now residing in the western part of Foundry Cove.

Comparison of Cd/Ni values provides information on the relative mobilities of Cd and Ni. Ratios of Cd to Ni were calculated from all available surface sediment data (1–6) as well as the new data reported here (after subtracting 2 ppm Cd and 30 ppm Ni as a correction for background). The ratios were then grouped into three regions: the outfall area, the eastern cove, and the western cove. The range and average of Cd/Ni for each region are given below.

Region	No. of samples	Av Cd-2/Ni-30	Range of Cd-2/Ni-30
Outfall	7	3.20	1.67–6.90
East Cove	30	1.58	0.92–3.14
West Cove	13	1.20	0.65–2.29

The results show a trend of decreasing Cd/Ni with distance from the source in the outfall area, with a particularly dramatic decrease in Cd/Ni from the outfall area to the main body of the cove.

The original Cd/Ni ratio in the industrial wastewater that emptied into the outfall area was 1.75 (3.87 Kg Cd per day/2.22 Kg Ni per day) (5). The outfall area, therefore, shows a concentration or retention of Cd relative to Ni with respect to the original dumping ratio of 1.75. The main body of the cove shows a depletion of Cd relative to Ni.

While the differences in Cd/Ni between the eastern and western sections of the cove may be significant and produced by differential solubility of Cd and Ni species on the basis of water column measurements during the dredging operation (1–3), the trend is only suggested by the sediment data. However, physical transport of sediment particles by tidal currents almost completely masks any such differential chemical mobility of Cd and Ni in the main body of the cove.

Of more significance is the decrease in Cd/Ni by approximately a factor of two from the outfall area on the adjacent, eastern part of the cove. Figure 2 shows this difference in Cd/Ni for sediments with a similar range of Cd (>2000 ppm). Higher Cd/Ni for the outfall area can be explained by comparing the chemistry of the industrial effluent waters to the water chemistry of the main body of Foundry Cove and by utilizing data from previous studies of the chemical forms of Cd and Ni in sediments of the cove and Eh-pH diagrams for Cd and Ni.

The pH of the industrial waste waters is known (5) to be ~ 12 (a 0.05 N solution of NaOH with colloidal Cd and Ni hydroxides and dissolved Cd^{+2} and Ni^{+2}). The chemistry of the

Foundry Cove waters is characterized by a $[\Sigma\text{CO}_2] = 10^{-3}$ mol/L, a pH slightly above 7, and dissolved O_2 at approximately saturation levels (2). The water temperature varies between 0 and 25 °C, but this range does not affect significantly the conclusions from the thermodynamic calculations which follow.

The chemical form of Cd in sediments from the outfall area has been found to be a mixed calcium–cadmium carbonate, while Ni in the same sediments was associated with the organic fraction (6). However, in sediments 100 m from the outfall area, both Cd and Ni were found to be associated with the organic fraction.

Significantly, Eh-pH diagrams for Cd (13) and Ni (14) support these findings by showing that NiCO_3 is not a stable species at the levels of ΣCO_2 found in Foundry Cove waters, but that CdCO_3 has a broad field of stability. The industrial effluent with a pH of 12 and laden with colloidal $\text{Cd}(\text{OH})_2$ and $\text{Ni}(\text{OH})_2$, when mixed with Foundry Cove waters (pH = 7), would undergo a rapid, large decrease in pH. Precipitation of CdCO_3 occurs in the pH range of 11–8. Since an insoluble carbonate phase of Ni is not found (6), the formation of a mixed $(\text{Cd,Ca})\text{CO}_3$ could explain the relative retention of Cd in the outfall area. Regardless of particle size, Cd and Ca are associated with particles of density $>2.7 \text{ g/cm}^3$, which is to be expected, since the densities of Cd and Ca carbonate are 4.2 and 2.7 g/cm^3 , respectively (6). Physical transport can remove the Ni, which is associated closely with the less dense organic fraction (6), more readily than it can the denser $(\text{Cd,Ca})\text{CO}_3$ phase.

The distribution of Ni in Foundry Cove sediments can be estimated from the Cd budget for the minimum burden of Cd in the system and from the average Cd/Ni values given previously.

For western Foundry Cove, if Cd/Ni = 1.2 and Cd ~ 2 tons, then Ni is also ~ 2 tons.

For eastern Foundry Cove, if Cd/Ni = 1.6 and Cd ~ 8 tons, then Ni is ~ 5 tons.

For the outfall area, if Cd/Ni = 3.2 and Cd ~ 8 tons, then Ni ~ 3 tons.

For the whole cove, therefore, $\Sigma\text{Ni} \sim 10$ tons. Since $\Sigma\text{Cd} = \sim 18$ tons (for the three regions listed above), then $\Delta\text{Cd}/\Sigma\text{Ni} = 1.80$, which is consistent with the original effluent ratio of 1.75.

Conclusions

The results of our sediment analyses, when combined with data from earlier studies, indicate that Cd and Ni have similar distributions (over several orders of magnitude in concentration) in the sediments of a shallow, estuarine fresh water cove, except in the immediate vicinity of the industrial waste outfall. This indicates that the predominant transport process for

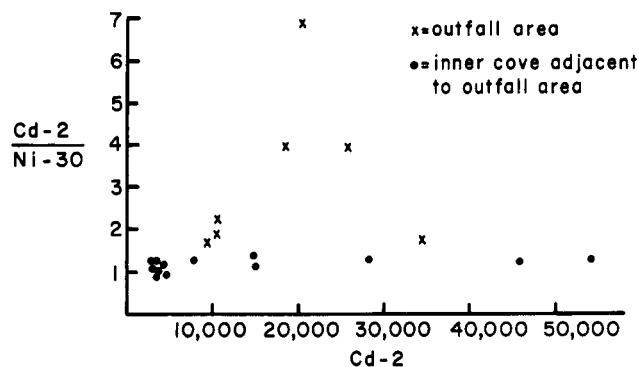


Figure 2. Cd/Ni vs. Cd for Cd > 2000 ppm in outfall and adjacent area

these metals is association with fine particles which can be expected to be deposited and resuspended many times by tidal currents. In the area of direct, alkaline, waste discharge Cd is less mobile than Ni, apparently because of the formation of an insoluble CdCO_3 phase.

Baseline data are provided on the distribution of Cd and Ni in the sediments of a cove, which, because of localized, industrial pollution, can serve as a useful environment for the study of Cd and Ni biogeochemistry.

The toxicity and potential health hazards to humans have been the subject of much recent study (15, 16) and are most dramatically expressed in the outbreak of "itai-itai" syndrome in Japan (17, 18). While the results of this study do not prove the existence of a public health hazard due to Cd contamination, they do show that the potential for such a hazard exists, should be studied, and that, due to the mobility of Cd, concern for such a hazard must include the western as well as the eastern part of the cove.

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Seasonal Trends in Denver Atmospheric Lead Concentrations

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■ During the 42-month period from January 1972 to June 1975, monthly average atmospheric lead concentrations at five metropolitan Denver sampling sites displayed maxima during winter months that coincided with minima in mixing heights. Monthly atmospheric lead inputs estimated on the basis of city-wide consumption of leaded gasoline showed a general downward trend with maxima during summer months. Atmospheric lead concentrations correlated well with the dispersion factor, the product of the mixing height and the mean wind. Although the data suggested a long-term declining trend at three sites, monthly atmospheric lead concentrations did not correlate with estimated monthly atmospheric lead inputs.

The major source of lead in urban atmospheres is combustion of leaded gasoline (1). Although there is a divergence of scientific opinion regarding some of the possible environmental and health effects of automotive lead (2, 3), there is widespread agreement that a general trend toward rising urban atmospheric lead exposures would be undesirable. The National Academy of Sciences Panel on Airborne Lead (1)

reported in 1972 that urban atmospheric lead concentrations in the United States had increased very little over several decades in spite of substantial increases in the consumption of lead antiknock additives. Shorter-term increasing trends have been reported for certain sites, however (4, 5). The general implication is that meteorological factors, urban geometry, and traffic routing may enter strongly in determining urban atmospheric lead concentrations. However, very little quantitative information has been compiled concerning the relative sensitivity of atmospheric lead concentrations in a specific urban center to changes in local lead additive consumption. The need for quantitative information has assumed major importance since regulations have been adopted to reduce ambient atmospheric lead exposures in the United States by phasing down the lead content of the total gasoline pool (6). These regulations are based upon a linear rollback formula, i.e., changes in atmospheric lead concentrations are directly proportional to changes in lead additive consumption.

While no totally satisfactory mathematical model has been developed for predicting pollutant dispersion within an urban complex, the atmospheric concentration of a pollutant emitted from an area source is often proportional to the emission rate and inversely proportional to the product of the mean wind