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Atmospheric Deposition and Fluxes of Mercury in Remote and Urban Areas of the Hudson River Basin

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Current environmental mercury concentrations are significantly elevated compared to pre-industrial levels due to anthropogenic inputs from fossil fuel combustion and incineration of municipal wastes. The contamination of remote lakes by the trace metal has been attributed to a rise in deposition of anthropogenic mercury from the atmosphere on a global scale.

Total mercury fluxes were reconstructed from analysis of archived sediment cores collected from natural waters of the Hudson River Basin. Sampling sites included: Black Rock Forest, a preserve near West Point, receiving primarily atmospheric inputs; Central Park Lake in Manhattan, reflecting urban atmospheric and runoff inputs; and the New York/New Jersey Harbor. Black Rock Forest fluxes were found to be on the order of atmospheric fluxes reported for remote areas. Fluxes to Central Park Lake sediments were over an order of magnitude higher than those to Sutherland Pond in Black Rock Forest. Total flux to NY/NJ Harbor sediments was an order of magnitude greater than to Central Park Lake, indicating that the dominant sources of mercury to the harbor were from direct industrial and wastewater inputs. While fluxes between the areas varied over orders of magnitude, total mercury concentrations in the sediments were similar (within a factor of 2). Sutherland Pond, with the smallest atmospheric flux of mercury, received a proportionally smaller particulate flux from its catchment area resulting in mercury concentrations similar to those in Central Park Lake and NY/NJ Harbor sediments. In response to such circumstances remote areas resembling the Black Rock Forest lake may exhibit problematic levels of mercury independent of anthropogenic inputs.

Abstract

Current environmental mercury concentrations are significantly elevated compared to pre-industrial levels due to anthropogenic inputs from fossil fuel combustion and incineration of municipal wastes. The contamination of remote lakes by the trace metal has been attributed to a rise in deposition of anthropogenic mercury from the atmosphere on a global scale.

Total mercury fluxes were reconstructed from analysis of archived sediment cores collected from natural waters of the Hudson River Basin. Sampling sites included: Black Rock Forest, a preserve near West Point, receiving primarily atmospheric inputs; Central Park Lake in Manhattan, reflecting urban atmospheric and runoff inputs; and the New York/New Jersey Harbor. Black Rock Forest fluxes were found to be on the order of atmospheric fluxes reported for remote areas. Fluxes to Central Park Lake sediments were over an order of magnitude higher than those to Sutherland Pond in Black Rock Forest. Total flux to NY/NJ Harbor sediments was an order of magnitude greater than to Central Park Lake, indicating that the

dominant sources of mercury to the harbor were from direct industrial and wastewater inputs. While fluxes between the areas varied over orders of magnitude, total mercury concentrations in the sediments were similar (within a factor of two). Sutherland Pond, with the smallest atmospheric flux of mercury, received a proportionally smaller particulate flux from its catchment area, resulting in mercury concentrations similar to those in Central Park Lake and NY/NJ Harbor sediments. In response to such circumstances remote areas resembling the Black Rock Forest lake may exhibit problematic levels of mercury independent of anthropogenic inputs.

Sediment Core Sampling Sites

- **Sutherland Pond, Black Rock Forest (SUP B)**

One of several freshwater ponds found in a forest preserve set aside for research in the early 20th century, Sutherland Pond is located in the Hudson Highlands 50 miles north of New York City. This pond provides us with an integrated sample of primarily atmospheric inputs over the past century.

- **Central Park Lake, Manhattan, NY (CPE)**

This small freshwater recreational lake was created in the 1860s-70s and dredged in 1903. It samples urban atmospheric input and runoff signals from the early 1900s to 1996.

- **NY/NJ Harbor (-1.7W)**

Surrounded by industrial activity, this area of the Harbor receives contributions from wastewaters and direct industrial inputs in addition to the urban atmospheric signal.

Introduction

- Sediments of the Hudson River Basin receive Hg from direct industrial inputs and atmospheric deposition.
- Historically, a limited amount of data has been collected on mercury levels in sediments of the basin.
- In this study dated sediment samples are used to describe temporal and spatial trends in mercury fluxes.

Methods

- Radionuclide analysis of ^{137}Cs and ^{210}Pb was conducted in our lab using Gamma Spectroscopy.
- Distribution of ^{137}Cs with depth in a vertical sediment core is the primary tool used for dating sediments, allowing us to identify sediments deposited at specific time horizons.
- Two distinct time horizons are easily identified in an ideal core:

1954 = first visibly significant inputs from global fallout due to large-scale atmospheric testing

1963 = year of peak ^{137}Cs fallout

Refer to posters H22D-11 and H22D-12, also in this session, for further explanation and figures on radionuclide analysis.

- Sedimentation rates and fluxes were determined using ^{137}Cs and ^{210}Pb measurements. For Sutherland Pond the net particle accumulation rate was based on pollen dating of other cores (Maenza-Gmelch, *Can J Botany*, 75, pp. 431-39, 1996).
- Samples were analyzed for Hg using a nitric acid digest followed by a gold amalgamation technique, and cold vapor AA analysis.

Results and Discussion

- Total Hg fluxes to Sutherland Pond were on the order of 0.02 to 0.05 $\mu\text{g}/\text{cm}^2\text{yr}$ (see table). This flux, for a site about 50 miles from NYC, is about an order of magnitude greater than values of atmospheric Hg fluxes reported for remote areas (Mason et al., *Geochim Cosmochim Acta*, 58, pp. 3191-98, 1994).
- Recent fluxes of Hg to Central Park Lake were on the order of 0.15 to 0.4 $\mu\text{g}/\text{cm}^2\text{yr}$ (see table), consistent with observations that a significant fraction of anthropogenic emissions of Hg to the atmosphere are deposited near the source (Mason, et al., 1994).
- Fluxes of Hg to NY/NJ Harbor sediments along the main stem of the Hudson ranged from about 3 $\mu\text{g}/\text{cm}^2\text{yr}$ in the early 1970s to about 1 $\mu\text{g}/\text{cm}^2\text{yr}$ in the 1990s (see table). These fluxes are much higher than those observed in Central Park Lake and indicate that urban atmospheric deposition accounts for only a small fraction of the Hg in NY/NJ Harbor sediments.

Other Hg Sources

- Total Hg analyses have been conducted on dated sediment samples from other areas of the harbor (see figure).
- The results indicate the existence of a number of major point sources of Hg in western NY/NJ Harbor including a Superfund site on the Hackensack River, a metal smelting facility on the Arthur Kill, and an unidentified source in the upper Passaic River Basin.
- These industrial sources appear to dominate the inputs of Hg to the western harbor and could be responsible for a significant fraction of the Hg observed in harbor sediments from along the main stem of the Hudson.

Conclusions

- Analysis of dated sediment samples is an effective tool for monitoring sources and trends of particle-associated contaminants such as Hg.
- Results from our three primary sampling sites in the Hudson Basin yield Hg fluxes that range over more than three orders of magnitude reflecting large spatial changes in atmospheric deposition and major direct inputs of Hg to the NY/NJ Harbor.
- Lakes similar to Sutherland Pond, with low sediment fluxes, may exhibit problematic levels of Hg even with relatively low atmospheric Hg fluxes.

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Hg Flux Data

| <i>Core</i> | <i>Depth (cm)</i> | <i>Year</i> | <i>Total Hg (ppm)</i> | <i>Total Hg flux ($\mu\text{g}/\text{cm}^2 \text{ yr}$)</i> |
|-------------|-------------------|-------------|-----------------------|--|
| SUP B | 0-20 cm | 1850-1996 | 3.96 | 0.0305 (0.0458)* |
| SUP B | 20-40 cm | 1700-1850 | 1.80 | 0.0156 (0.0234)* |
| SUP B | 44-49 cm | pre 1700 | 1.89 | 0.0126 (0.0189)* |

sed rate = 0.008g/cm² yr

**denotes ²¹⁰Pb normalized Hg flux, using a focusing factor of .6*

| | | | | |
|-------|----------|-----------|------|-------|
| CPE | 0-4 cm | 1990s | 2.02 | 0.154 |
| CPE | 6-8 cm | late 70s | 2.00 | 0.221 |
| CPE | 12-14 cm | early 60s | 2.07 | 0.321 |
| CPE | 14-16 cm | 50s-60s | 2.32 | 0.303 |
| CPE | 18-20 cm | 1950s | 1.66 | 0.324 |
| CPE | 22-24 cm | 40s-50s | 1.55 | 0.219 |
| CPE | 24-26 cm | 1940s | 1.31 | 0.292 |
| CPE | 30-32 cm | 1930s | 1.72 | 0.429 |
| CPE | 36-38 cm | 1920s | 1.47 | 0.417 |
| **CPE | 14-16 cm | 50s-60s | 2.09 | 0.273 |
| **CPE | 22-24 cm | 40s-50s | 1.70 | 0.241 |

sed rate = 0.1 - 0.29g/cm² yr

**** denotes wet sample**

| | | | |
|---------------|-----------|------|-------|
| -1.70W 0-2 cm | 1996 | 1.30 | 0.975 |
| -1.71W 0-2 cm | 1996 | 1.58 | 1.19 |
| -1.69W 0-2 cm | 1994 | 1.42 | 1.07 |
| -1.68W 0-2 cm | 1989 | 2.07 | 1.55 |
| -1.7W 8-10 cm | early 70s | 4.23 | 3.17 |

sed. rate = 0.75g/cm² yr
