



Mercury inputs and redistribution in the Penobscot River and estuary, Maine

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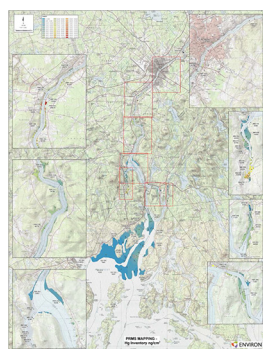
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HIGHLIGHTS

- Total Hg was rapidly distributed and deposited throughout this system.
- The calculated cumulative total sedimentary Hg (9.28 metric tons) throughout the system falls within the range of total Hg (6–12 metric tons) believed to have been released from HoltraChem.
- Differences between distributions of total Hg inventories, near-surface (upper 3 cm) total Hg concentrations, and contemporary total Hg fluxes show that total Hg is being redistributed throughout the system.
- Mean, near-surface (upper 3 cm) total Hg concentrations are greatest in the Orland River (1,120 ng g⁻¹) > Penobscot River (815 ng g⁻¹) > Mendall Marsh (673 ng g⁻¹) > Penobscot Estuary (526 ng g⁻¹).
- Hg(o) values at different sites were similar, though individual total Hg profiles were heterogeneous.

GRAPHICAL ABSTRACT



Simple contour map (kriging) showing the distributions of total sedimentary Hg inventories (ng cm⁻²) throughout the Penobscot system.

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ABSTRACT

We examined total mercury (Hg) distributions in sediments from the Penobscot River and estuary, Maine, a site of extensive Hg releases from HoltraChem (1967–2000). Our objectives were to quantify: (1) bottom sediment Hg inventories (upper ~1 m; 50–100 y); (2) sediment accumulation rates; and (3) contemporary Hg fluxes to bottom sediments; by sampling the Penobscot River (PBR), Mendall Marsh (MM), the Orland River (OR) and the Penobscot estuary (ES). Hg was rapidly distributed here, and the cumulative total (9.28 metric tons) associated with sediments system-wide was within the range released (6–12 metric tons). Evidence of sediment/Hg remobilization was observed in cores primarily from the PBR, and to a lesser extent the ES, whereas cores from MM, most of the OR, the ES, and half from the PBR exhibited sharp peaks in Hg concentrations at depth, followed by gradual decreases towards the surface. Based on background PBR sediment Hg concentrations (100 ng g⁻¹), “elevated” (300 ng g⁻¹), or “highly elevated” (600 ng g⁻¹) Hg concentrations in sediments, and resulting inventories, we assessed impact levels (“elevated” ≥ 270, or “highly elevated” ≥ 540 mg m⁻²). 71% of PBR stations had “elevated”, and 29% had “highly elevated” Hg inventories; 45% of MM stations had “elevated”, and 27% had

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“highly elevated” inventories; 80% of OR stations had “elevated” inventories only; and 17% of ES stations had “elevated” inventories only. Most “highly elevated” stations were located within 8 km of HoltraChem, in MM, in the PBR, and in the OR. Near-surface sediments in the OR, PBR and MM were all “highly elevated”, while those in the ES were “elevated”, on average. Mean Hg fluxes to bottom sediments were greatest in the OR (554), followed by the PBR (469), then MM (452), and finally the ES (204 ng cm⁻² y⁻¹).

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1. Introduction

Prior to the late 1700's, the only sources of pollution to the Penobscot River included sewage and periodically high sediment loads driven by land clearing. Large-scale logging began here in the 1770's, and it has been estimated that ~4% of all logs that floated down-river sank to the bottom, which equates to 400 million board feet of wood (Davies, 1972). Beginning at this same time, saw mills were constructed along the river and its tributaries, and contributed sawdust, edgings and bark to the waterways, a process which continued through the 1950's (Cutting, 1959). Throughout much of the 1800's, industrial development in the lower Penobscot River basin continued, and included the establishment of flour mills, shipbuilding, as well as cotton and tanning. The rise of pulp and paper mills in the lower river basin began in 1889 (Goode, 1934), and as this industry rapidly grew, it resulted in the discharge of large quantities of organic matter and industrial solids to the river and its tributaries. These discharges drove rapid depletion of dissolved oxygen in receiving waters. By 1960, numerous pulp and paper mills, leather and textile plants were located in the lower river basin. Residents began to voice concerns about the effects of pollution on fisheries and drinking water supplies (Judd and Beach, 2003). In 1972, the University of Maine conducted the Penobscot River Study, which concluded that the river was periodically overloaded by oxygen-demanding wastes, and as a result was unable to support most fish species, or be used as a municipal water supply (Penobscot River Study Team, 1972). Passage of the Clean Water Act (1972) resulted in the beginning of some pollution reduction and mitigation in the Penobscot River and estuary. Industrial and municipal outfalls began to be monitored, and improvements were mandated where needed. Monitored pollutant loads to the river decreased by 85%, and all of Maine's Department of Environmental Protection sampling sites improved in water quality and the health of macroinvertebrate communities (Davies, 1999).

The HoltraChem Manufacturing Company in Orrington, Maine is located on a 235-acre property on the banks of the Penobscot River. Approximately 50 acres are developed, including the manufacturing facility, five landfills, a surface impoundment and a scrap metal area. The immediate plant area covers approximately 12 acres. The facility opened in 1967 and manufactured chlorine, caustic soda (sodium hydroxide), chlorine bleach (sodium hypochlorite), hydrochloric acid and the pesticide chloropicrin. The plant closed in September 2000. As of 2011, some limited demolition (including the mercury (Hg) cell building, but without removal of its concrete floor) and waste removal from the site was completed, but much of the infrastructure and all of the landfills remained. Surface and groundwater on the plant site, and surface water in the adjacent Penobscot River, was subjected to detailed characterization prior to the plant closing in 2000 (Camp et al., 1998). The results indicated that groundwater within the plant was contaminated with Hg (Hg concentrations > 1000 µg l⁻¹ in some wells), and that surface runoff was also elevated in Hg. The presence of the plant could be detected by sampling Hg in Penobscot River surface water near the plant outfalls (Hg concentrations up to 70 ng l⁻¹ compared with upstream background of <5 ng l⁻¹). Calculations suggested that the net effect of all plant discharges and the estimated groundwater flux would raise average Hg concentration in the river by ~2 ng l⁻¹ at a river discharge of 4000 ft³ s⁻¹ (113 m³ s⁻¹). Total Hg loading to the

Penobscot River from the plant site was estimated at ~20 g d⁻¹. This loading was compared to that carried by the river from upstream under various flow conditions. At average river discharge (16,400 ft³ sec⁻¹, 464 m³ sec⁻¹) and assuming ambient total Hg concentration in river water (4.3 ng l⁻¹), Camp et al. (1998) estimated river loading was 172 g d⁻¹. Direct discharges of wastewater from the onsite water treatment plant to the Penobscot River were significantly reduced following plant closure in 2000, although groundwater discharges and storm water runoff continued. Total Hg loading to the Penobscot River from the HoltraChem site has not been rigorously evaluated since prior to cessation of production, but some data collection (surface water and groundwater sampling) has continued.

Elevated levels of methyl Hg (CH₃Hg⁺) in sediments and biota led to legal action by the Maine People's Alliance in the 1990's. This group joined with the Natural Resources Defense Council (NRDC) to bring a lawsuit against HoltraChem, which came to trial in 2002. As a result, in 2003, the U.S. District Court (District of Maine) ordered an independent scientific study, and implemented the Penobscot River Mercury Study (PRMS). The PRMS had the following three charges, to determine: 1. The extent of existing harm resulting from Hg contamination to the Penobscot River system south of the HoltraChem site; 2. The need for, and feasibility of a remediation plan to effectively address the present effect of such existing harm, if any; and 3. The elements and timetable for the execution of the appropriate remediation plan to address the harm existing as a result of Hg contamination. Two phases of study have been completed. Phase I addressed objective one, concluding that there was extensive contamination of the lower river and estuary due to Hg released from HoltraChem, which was sufficient to threaten the health of biota. This warranted the initiation of Phase II of the study (of which the research described herein was integral). Phase II of the study addressed objective two, concluding that there was a need for a remediation program and that the remediation approaches, which have been recommended, were scientifically feasible. Data collected during Phase II have provided the scientific basis for recommendations to the federal court to proceed on to the third objective of the implementing order, to order a remediation program to speed the recovery of the Penobscot River from its present state of Hg contamination.

The focus of this study is the total Hg associated with sediments in portions of the lower Penobscot River, Mendall Marsh, the Orland River and the Penobscot estuary. As discussed, a primary known point source of Hg pollution within the lower river basin is HoltraChem, which was in operation from 1967 to 2000 (with most direct releases of Hg occurring from ~1967–1970, Penobscot River Mercury Study Panel [PRMS], 2013). Total Hg concentrations measured in sediment collected from the Penobscot River upstream of the limit of tidal influence are ~100 ng g⁻¹ dry weight, which is comparable to those of other New England rivers (Morgan, 1998; Kamman et al., 2005). In the Penobscot estuary, total Hg concentrations in surficial, bottom sediments have been reported to range between 125 and 2750 ng g⁻¹ (Merritt and Amirbahman, 2007). The highest total Hg concentration reported in the literature is 230,000 ng g⁻¹, which corresponds to sediment collected within HoltraChem's discharge zone (Morgan, 1998). This Hg pollution has resulted in the widespread distribution of elevated concentrations of total Hg in sediments throughout the lower Penobscot River and estuary. In some parts of the system, the physical and

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