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Estimates of recovery of the Penobscot River and estuarine system from mercury contamination in the 1960's

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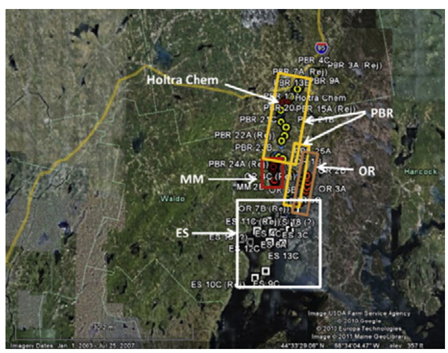
HIGHLIGHTS

- For the first time, recovery times have been estimated in a large, Hg contaminated river-estuarine system.
- Estimated recovery is a slow process lasting many decades.
- Redistribution of Hg from sites with higher to those with lower concentrations is taking place.

GRAPHICAL ABSTRACT



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ABSTRACT

Mercury (Hg) was discharged in the late 1960s into the Penobscot River by a chlor-alkali production facility, HoltraChem. Using total Hg concentration profiles from 56 stations (58 sediment cores) in the Penobscot River (PBR), Mendall Marsh (MM), Orland River (OR) and Penobscot Estuary (ES), and sediment accumulation rates derived using detailed profiles of total Hg concentrations and radionuclide activities (^{137}Cs, ^{239,240}Pu, ^{210}Pb), recovery from system-wide Hg pollution was assessed. Total Hg concentration profiles showed sharp maxima at depths attributed in time to a 1967 release date, and were divided into two sections: the first 21 years (1967–1988; rapid recovery), and the recent 21 years (1988–2009; slower recovery). The recent 21 years of Hg input were used to estimate 'apparent' recovery rates, yielding exponentially decreasing total Hg concentrations. Apparent recovery half-times ($T_{1/2} = \ln 2/\alpha$) were calculated from an exponential fit of $Hg(t) = Hg(t = 21) * \exp(-\alpha * t) + Hg(\infty)$ to total Hg concentration profiles over the past 21 years (assuming $Hg(\infty)$ of 0, 100, or 400 ng g^{-1}). Mean $T_{1/2}$ values were, at PBR 31 years (16 of 24 cores), at MM 22 years (9 of 11 cores), at ES 20 to 120 years (mean of 78 years; 12 of 18 cores), and at OR 69 years (3 of 5 cores). In 18 out of 57 cores, concentrations either increased towards the surface or remained the same, indicating slower or incomplete 'communication' with the larger system. The Penobscot River and Estuary system has recovered substantially since 1967, and top 1 cm sediment Hg concentrations (Hg(0)) from areas in rapid communication with the larger system are converging to 600–700 ng g^{-1} (1967 maxima of 70,000^{+} ng g^{-1}). However, to recover from Hg(0) of 700 ng g^{-1} to a $Hg(\infty)$ of <100 ng g^{-1} would require 3 or more half-times.

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1. Introduction

The Penobscot River in Maine drains a watershed of approximately 19,350 km^2 and is the second largest river system in New England. The lower Penobscot River is defined by a long, narrow estuary (mean width 0.75 km), and measurable tidal influence extends 35 km to the city of Bangor. Point sources of Hg pollution within the estuary include a closed (2000) chlor-alkali production facility (HoltraChem) that operated from 1967, as well as several upriver paper mills. Sediment total Hg concentrations upstream of the limit of tidal influence are of the order of 100 ng g^{-1} (d.w.), comparable to those of other New England rivers (Morgan, 1998). Surface-sediment total Hg concentrations in the Penobscot Estuary have been previously reported to range between 125 and 2750 ng g^{-1} (Merritt and Amirbahman, 2007). The highest sediment total Hg concentration reported in the published literature is 230,000 ng g^{-1}, which was collected from a site within the HoltraChem discharge zone (Morgan, 1998). Mercury is among the most hazardous of contaminants in aquatic environments, with human health risks as well as ecological and toxicological effects that depend on its chemical speciation (e.g., Jarup, 2003; Horvat et al., 2003; Boening, 2000). A major concern in the Penobscot River and Estuary system is that large quantities of inorganic Hg are, and will continue to be, available to be converted to methyl-Hg (CH_3Hg^+), a potent neurotoxin that is produced in wetlands (Gilmour et al., 2017) and readily accumulated by aquatic biota (e.g., Watras et al., 1998; Wolfe et al., 1998; Mason et al., 2000; Mergler et al., 2007; Selin, 2009).

A critically important question is: how long will it take for these elevated total Hg concentrations in sediment to decrease to an acceptable level? Or, alternatively, how long will it take for the Penobscot River and Estuary to 'recover' from Hg pollution? Extensive Hg pollution here has produced greatly elevated methyl-Hg concentrations in wetland sediments and wildlife (Gilmour et al., in this volume), threatening organism and ecosystem health (Kopec et al., in this volume). The purpose of this study was to assess time scales of Hg recovery, using an approach whereby total Hg concentrations and sediment accumulation rates (Yeager et al., 2017a, this volume) were determined using the 1963 bomb fallout event markers ^{137}Cs and ^{239,240}Pu, and the atmospherically delivered steady-state marker excess-^{210}Pb ($^{210}Pb_{xs}$ = Total ^{210}Pb − ^{226}Ra-supported ^{210}Pb) in 58 sediment cores taken from throughout the Penobscot River and Estuary.

2. Methods

2.1. Field and initial processing

A total of 58 sediment cores, collected from 56 stations throughout the Penobscot River and Estuary were analyzed after collections during the field season of 2009 (Fig. 1). Coring stations were located in the Penobscot River (PBR), Mendall Marsh (MM) (a wetland located ~15 km downstream of HoltraChem with high Hg methylation rates, Gilmour et al., in this volume), Penobscot Estuary (ES), and the Orland River (OR) (a tributary of the Penobscot River, with confluence ~25 km downstream from HoltraChem, situated in a region near the turbidity maximum of the Penobscot River, Geyer et al., in this volume).

Due to the scale of field work required, it involved cooperating research personnel from the University of New Orleans, and from Normandeau Associates, Inc. Similarly, given the scale of the analytical work, multiple laboratories were involved, including those at Texas

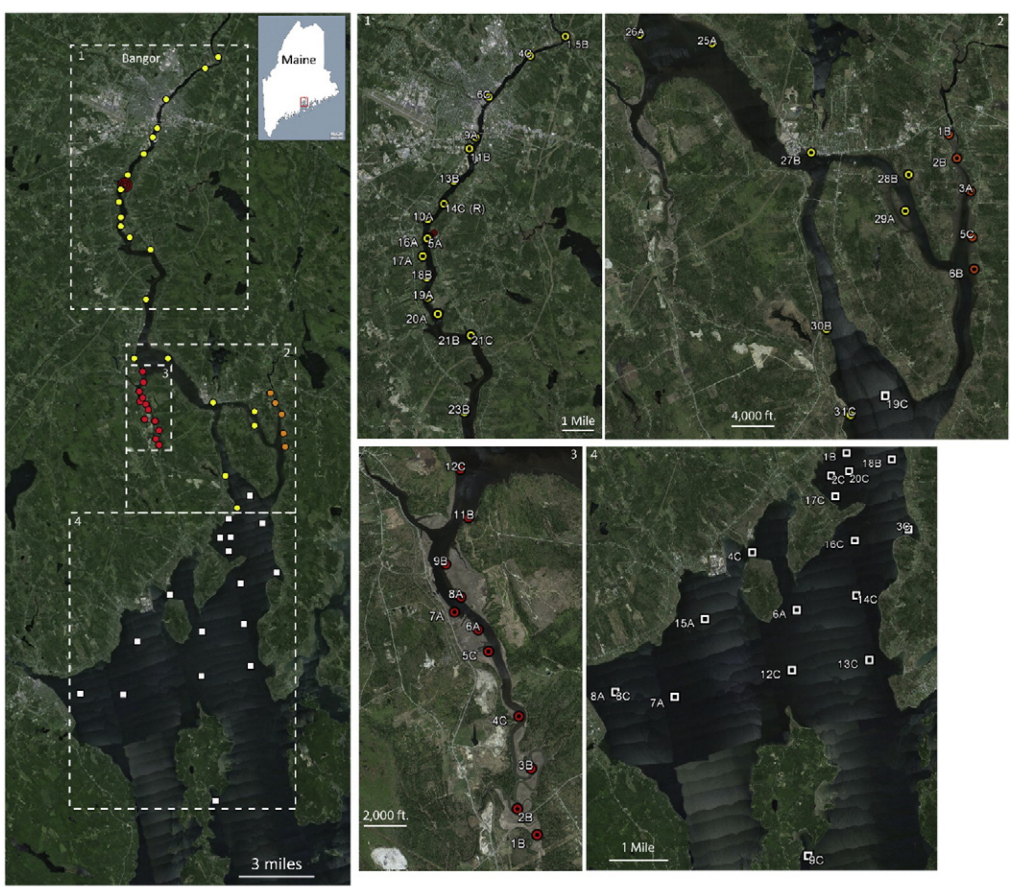

Fig. 1. All coring stations shown in far left panel, HoltraChem denoted by red bullseye. Inset panel one shows most of the Penobscot River (PBR) stations; inset panel two shows the remaining PBR stations, all Orland River (OR) stations, and one of the Penobscot Estuary (ES) stations; inset panel three shows all Mendall Marsh (MM) stations, and inset panel four shows the remaining ES stations. Only those stations which were fully characterized are shown.
Imagery courtesy of Google Earth (2015).