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## Science of the Total Environment

journal homepage: <www.elsevier.com/locate/scitotenv>

## Sediment accumulation and mixing in the Penobscot River and estuary, Maine



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#### HIGHLIGHTS

#### GRAPHICAL ABSTRACT

- Sediment variability increased from Mendall Marsh/Orland River < Penobscot estuary < Penobscot River.
- Generally, sediment mixing was limited (maximum Db ranged from 3-6 cm<sup>2</sup>  $y^{-1}$ ; mixed layers of ~1-3 cm).
- A wide range in sediment accumulation rates was observed  $(0.10-1.85 \text{ cm y}^{-1})$ , and was expected.
- Extensive lateral transport of sediment (and total Hg) was limited to parts of the Penobscot River.
- Sediment (and Hg) accumulation in the Penobscot River was most random, focused inside meanders, and coves.

#### article info abstract

Article history: Received 29 June 2017 Received in revised form 26 March 2018 Accepted 3 April 2018 Available online xxxx

Keywords: Geochronology Sedimentation rates River-estuarine system Sediment transport Penobscot River

Composite sediment accumulation rates determined by radionuclides and total Hg for each part of the Penobscot River an/d estuary system (uncertainties reported at 1  $\sigma$ ).



Mercury (Hg) was discharged in the late 1960s into the Penobscot River by the Holtra-Chem chlor-alkali production facility, which was in operation from 1967 to 2000. To assess the transport and distribution of total Hg, and recovery of the river and estuary system from Hg pollution, physical and radiochemical data were assembled from sediment cores collected from 58 of 72 coring stations sampled in 2009. These stations were located throughout the lower Penobscot River, and included four principal study regions, the Penobscot River (PBR), Mendall Marsh (MM), the Orland River (OR), and the Penobscot estuary (ES). To provide the geochronology required to evaluate sedimentary total Hg profiles, 58 of 72 sediment cores were dated using the atmospheric ra-<br>dionuclide tracers <sup>137</sup>Cs, <sup>210</sup>Pb, and <sup>239,240</sup>Pu. Sediment cores were assessed for depths of mixing, and fo determination of sediment accumulation rates using both geochemical (total Hg) and radiochemical data. At most stations, evidence for significant vertical mixing, derived from profiles of <sup>7</sup>Be (where possible) and porosity, was restricted to the upper ~1-3 cm. Thus, historic profiles of both total Hg and radionuclides were only minimally distorted, allowing a reconstruction of their depositional history. The pulse input tracers  $137Cs$  and <sup>239,240</sup>Pu used to assess sediment accumulation rates agreed well, while the steady state tracer <sup>210</sup>Pb exhibited weaker agreement, likely due to irregular lateral sediment inputs.

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### 1. Introduction

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The Penobscot River is approximately 425 km in length (including the West and South branches), drains a basin of  $\sim$ 22,300 km<sup>2</sup>, and is the second largest river system in New England, after the Connecticut

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River. The Penobscot River estuary extends approximately from the city of Bangor, Maine (ME), south to the city of Searsport, ME, where it meets Penobscot Bay. With a surface area of ~90 km<sup>2</sup>, this estuary is the largest in Maine, and part of one of the largest embayments on the eastern coast of the U.S. The estuary has an appreciable tidal range (over 3 m), with measurable tidal influence extending 35 km up river to the city of Bangor.

Bedrock of the Penobscot River watershed is comprised of lower Paleozoic age metasedimentary and igneous rocks [\(Osberg et al., 1985](#page--1-0)). These rocks were shaped and overlain by deposits associated with the Laurentide Ice sheets. Deglaciation here generally proceeded from SE to NW between 16,000–12,000 radiocarbon years BP ([Borns et al.,](#page--1-0) [2004](#page--1-0)). Late Pleistocene age glacial tills cover parts of the upper Penobscot River watershed, with glacio-fluvial sediments concentrated in valleys [\(Thompson and Borns, 1985\)](#page--1-0). The history of sea level change here is complex, and is reflected by thick deposits of glacio-marine silts and clays in the central and lower elevation portions of the watershed. As ice retreat proceeded, marine transgression followed, inundating the depressed land surface. This transgression began 16,000 years BP, producing a relative sea level (RSL) high stand of 70 m above present at the coast ([Thompson and Borns, 1985](#page--1-0)). Isostatic uplift commenced as deglaciation proceeded, leading to rapid RSL fall ending at  $\sim$ 12,000 years BP, with a low stand at 60 m below present ([Kelley](#page--1-0) [et al., 2010\)](#page--1-0). By 11,500 years BP, RSL along the Maine coast rose to  $\sim$ 20 m below the present level. From 11,500 to 7000 years BP, RSL rose by 5 m, and then increased, first rapidly, and then more slowly to its present elevation. The central and lower parts of the Penobscot River channel is floored by rock, coarse gravels, and muds ([Kelley and](#page--1-0) [Kelley, 2006\)](#page--1-0).

The overall focus of this and two other, related articles [\(Yeager et al.,](#page--1-0) [2018;](#page--1-0) [Santschi et al., 2017\)](#page--1-0) is the total mercury (Hg) associated with sediments in portions of the lower Penobscot River, Mendall Marsh, the Orland River and the upper Penobscot River estuary. A primary, known point source of Hg pollution within the lower river basin is the Holtra-Chem chlor-alkali production facility, which was in operation from 1967 to 2000. Total Hg concentrations measured in sediment collected from the Penobscot River upstream of the limit of tidal influence are of the order of 100 ng  $g^{-1}$  dry weight, which is comparable to those of other New England rivers ([Morgan, 1998](#page--1-0); [Kamman et al., 2005\)](#page--1-0). In the Penobscot River estuary, total Hg concentrations in surficial, bottom sediments have been reported to range between 125 and 2750 ng  $g^{-1}$ [\(Merritt and Amirbahman, 2007\)](#page--1-0). The highest total Hg concentration reported in the literature is 230,000 ng  $g^{-1}$ , which corresponds to sediment collected within Holtra-Chem's discharge zone ([Morgan, 1998](#page--1-0)). 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 parts of the system (particularly Mendall Marsh), the physical and geochemical setting has been conducive to the sustained production of highly-elevated methyl Hg ( $CH<sub>3</sub>Hg<sup>+</sup>$ ) concentrations ([Gilmour et al., 2018\)](#page--1-0), with consequent and demonstrable impacts on ecosystem health (e.g., [Sullivan and Kopec, 2018](#page--1-0)).

The specific objectives of these three articles include: (1) Quantifying sediment mixing and accumulation rates throughout the system focusing on the upper ~1 m of sediment (corresponding to the relevant historical period (50–100 years)); (2) Quantifying inventories of total Hg in and fluxes to bottom sediments throughout [\(Yeager et al., 2018](#page--1-0)); and (3) Quantifying time scales of natural recovery from Hg contamination throughout the system [\(Santschi et al., 2017\)](#page--1-0). The research described here is focused on the first of these objectives.

#### 2. Materials and methods

#### 2.1. Field sampling and initial processing

During the field season of 2009, 72 sediment-coring stations were established and sampled throughout the lower Penobscot River, Mendall Marsh, the Orland River and the upper Penobscot Estuary. Due to the scale of fieldwork, cooperating research personnel from the University of New Orleans (UNO), and from Normandeau Associates Ltd. assisted in field operations. At each site, sediment cores were collected in triplicate (216 total) utilizing vibracoring, a common sediment coring method (e.g., [Ward et al., 2008;](#page--1-0) [Pre et al., 2011;](#page--1-0) [Yeager et al.,](#page--1-0) [2012\)](#page--1-0). These 72 stations were distributed from just south of Veazie Dam, near Eddington, ME, south through the lower Penobscot River and estuary to Islesboro Island. The locations of these stations were chosen based on preliminary data gathered by the Penobscot River Mercury Study (PRMS), field reconnaissance, and the desire to collect an adequate depth of relatively undisturbed sediment sufficient to represent the relevant historical period (50–100 years). Four key areas are represented, including (1) the Penobscot River (PBR) from well north to well south of the location of the Holtra-Chem facility near Orrington, ME [\(Fig. 1](#page--1-0)), (2) Mendall Marsh (MM), (3) the Orland River (OR), and (4) the upper Penobscot River estuary (ES).

All cores were returned to the laboratory for inspection and processing. All three cores from each station were split longitudinally and visually inspected. One core was then selected with the primary objective being to select that core that appeared least disturbed based on observation of the sedimentary section sampled. The other two cores from that station were discarded. The core sectioning protocol produced, with minimal exceptions, 40 sediment samples per core (1–20 cm sectioned at 1 cm, 20–40 cm at 2 cm, and 40–90 cm at 5 cm). A pre-screening process was then enacted, where aliquots from all intervals, from the selected core from all stations, were analyzed for total Hg. Based primarily on considerations that included the maximum concentrations and shape of total Hg profiles in each cored section, cores were either fully analytically characterized, or rejected. This process resulted in the rejection of cores collected from 16 stations, with cores from 56 stations being fully characterized (producing 58 cores, with two cores processed from two stations [PBR\_21, ES\_08]). These 58 cores were distributed throughout the four study areas as follows, (1) PBR – 24, (2) MM – 11, (3) OR – 5, and (4) ES – 18.

#### 2.2. Physical and chemical variables

Sediments from each core interval were weighed, dried, and reweighed to determine physical variables including dry bulk density (Bd), fraction of water (fw), sediment porosity ( $\varphi$ ), sediment mass depths, and cumulative mass depths (supplementary materials). Sediment textures (grain size) were assessed by quantifying the mineral grain size distribution, and sand, silt, and clay fractions as defined by the Wentworth Scale ([Wentworth, 1922\)](#page--1-0). Texture samples were dried at ~75 °C for 24–48 h, gently disaggregated, wet-sieved through 2 and 0.5 mm sieves, and treated with dilute  $H_2O_2$  to destroy organic binding agents [\(Hillier, 2001;](#page--1-0) [Yeager et al., 2005\)](#page--1-0). Samples were then analyzed using a Malvern Mastersizer S-2000, a laser-optical particle size characterization instrument capable of accurately resolving particles over a size range of 0.02 to 2000 μm. Particulate organic carbon (POC) concentrations were determined via elemental analysis according to methods given in [Santschi et al. \(2001\)](#page--1-0). Flett Research Ltd. ([http://www.](http://www) flettresearch.ca/) determined concentrations of total Hg. Total Hg in sediments was measured using a DMA-80 analyzer according to EPA Method 7473.

#### 2.3. Radiochemistry

High-resolution gamma spectrometry was employed to resolve <sup>7</sup>Be (where possible) ( $t_{1/2}$  = 53 d, E $\gamma$  = 477 keV), <sup>137</sup>Cs ( $t_{1/2}$  = 30 y, E $\gamma$  = 661 keV) and <sup>226</sup>Ra (t<sub>1/2</sub> = 1601 y, E $\gamma$  = 352 keV) using Canberra HPGe well detectors and multi-channel analyzer, model DSA-1000. Samples were contained in plastic test tubes (i.d. 1.3 cm, ht. 9.4 cm), and standards (<sup>137</sup>Cs and <sup>226</sup>Ra: NIST, SRM #4357; <sup>7</sup>Be: Isotope Products Laboratories CN #6007) were prepared and run on each detector in geometries Download English Version:

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