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Spatial and temporal trends of mercury in the aquatic food web of the lower Penobscot River, Maine, USA, affected by a chlor-alkali plant

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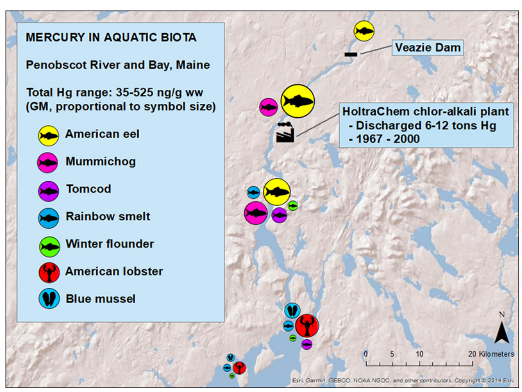
HIGHLIGHTS

- Elevated mercury residues persist in riverine biota 20 years after chlor-alkali plant closure
- Mercury in fish, lobster and blue mussels decrease with distance from plant site
- Evidence of declining trends in mercury concentrations in pelagic food web
- No change in mercury concentrations in benthic food web over time
- Elevated lobster mercury concentrations forced fishery closure in upper Penobscot Bay

GRAPHICAL ABSTRACT



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ABSTRACT

Mercury (Hg) concentrations in aquatic biota, including fish and shellfish, were measured over the period 2006–2012 in the lower Penobscot River and upper estuary (Maine, USA). The Penobscot is a system contaminated with Hg by a chlor-alkali plant that operated from 1967 to 2000, discharging 6–12 tons of mercury into the river. Mercury levels in aquatic biota were highest at sites downstream of the chlor-alkali plant and spatial trends were similar to those of sediments. Mean total Hg concentrations in fish muscle (adjusted for size or age) in the most affected areas were 521 (480, 566; 95% CI) ng/g ww in American eels, 321 (261,395) in mummichog, 121 (104, 140) in rainbow smelt, 155 (142,169) in tomcod, 55.2 (42.7,71.4) in winter flounder, and 328 (259,413) in American lobster tail and 522 (488,557) ng/g dw in blue mussel. Levels exceeded the 50 ng/g ww considered protective for piscivorous predators and were of concern for human health, with American eels and American lobster exceeding Maine's mercury action level of 200 ng/g ww. Calculations of trophic position (using nitrogen isotopes) suggested that the spatial patterns observed in total Hg concentrations were not due to changes in feeding habits of the species. Fish feeding in benthic food webs, as defined by stomach content and stable carbon isotope analyses, showed no change in Hg concentrations over time. In contrast, declining trends in Hg were found in two species dependent on pelagic food webs. The absence of declines in Hg

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American lobster (*Homarus americanus*)
Blue mussel (*Mytilus edulis*)
Stable isotopes

concentrations in the benthically-based food webs, despite the fact that most Hg was discharged into the system >40 years ago, is consistent with the long recovery predicted from dated sediment cores and from similar studies elsewhere.

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1. Introduction

Mercury (Hg) remains a global concern for aquatic ecosystems (Krabbenhoft and Sunderland, 2013) due to anthropogenic loadings via atmospheric emissions (Fitzgerald et al., 1998) and Hg point sources discharging directly into freshwater and marine environments. Localized discharges from mercury-cell chlor-alkali plants are among the greatest point sources of Hg to aquatic systems (Bloom et al., 1999; Fimreite et al., 1971). While this production method has been largely discontinued, past discharges continue to pose a threat to aquatic systems long after plant operations have ceased (Munthe et al., 2007; Neff et al., 2012).

Inorganic Hg is the primary form of Hg discharged into aquatic systems yet, in the upper layers of aquatic sediment and in water, bacteria transform inorganic Hg into methyl Hg (Mitchell and Gilmour, 2008). Methyl Hg is highly bioaccumulative in organisms, and biomagnifies in aquatic food webs (Chen et al., 2008). This form of Hg is a potent neurotoxin that harms fish (Depew et al., 2012) and fish consumption remains the primary exposure route for humans (Zahir et al., 2005) and wildlife (Scheuhammer and Sandheinrich, 2008; Wolfe et al., 1998).

Mercury's biomagnification through an aquatic food web can be traced through the prey consumed at each trophic level. Methyl Hg, like carbon and nitrogen, enters the base of the aquatic food web through either benthic or pelagic primary producers (Chen et al., 2014). The source of methyl Hg and its transfer through the food web can be identified using stable isotope ratios of carbon and nitrogen which define the primary energy sources to and relative trophic levels of fish and other upper trophic level organisms (Lavoie et al., 2010; Peterson, 1999; Peterson et al., 2017). Identifying the source of methyl Hg to the food web through benthic or pelagic carbon may allow for focused and more effective remediation of a contaminated system.

The Penobscot River drains the largest watershed in New England, 22,600 km^2, and flows into Penobscot Bay along the central Maine coast. Between 1967 and 2000 the HoltraChem mercury-cell chlor-alkali plant operated along the east bank of the lower tidal portion of the river. This plant (HoltraChem) discharged an estimated 6–12 tons of Hg to the river via direct discharge of contaminated brine and via surface and subsurface runoff of Hg from the site (Turner et al., 2018). Assessments of the Hg contamination of the Penobscot system proceeded as unique court-ordered studies, and Hg levels in sediments, water and biota were determined with the ultimate goal of remediating the problem (Rudd et al., 2018).

Our study goals were to define the geographic extent of Hg contamination in aquatic biota in the lower Penobscot, identify temporal trends in Hg concentrations, and to define the accumulation and transfer of Hg (as total and methyl Hg) in the food web of the Penobscot River and estuary. We hypothesized that Hg concentrations in biota would diminish with distance from the chlor-alkali plant, that concentrations would decline following closure of the plant, and that Hg concentrations in target species feeding in the benthic food web would reflect sediment Hg concentrations.

2. Methods

2.1. Study area and sample collection

The study area extended for 43 km along the lower Penobscot River, from the town of Orono south to the river's mouth at Bucksport (Fig. 1) and south for an additional 27 km into Penobscot Bay. HoltraChem was located along the river in the town of Orrington, 23 km north of the river mouth. Samples were collected over several years (2006–2012; see Table 1) from five reaches within the study area, two upstream and three downstream of the plant site. The upstream OV reference reach was outside of the aquatic influence of the HoltraChem discharges in an impounded reservoir behind the former Veazie Dam, the first head-tide dam upstream of the river's mouth. All other reaches were under tidal influence. The BO reach extended upstream of the HoltraChem discharge site, the OB reach extended downstream, south, from the HoltraChem site to the river's mouth, upper ES ran south from the river's mouth at Bucksport to Fort Point in Penobscot Bay, and lower ES extended south into Penobscot Bay to the island of Islesboro (Fig.1).

The salinity gradient within the study area, from freshwater in the OV reach above the dam to full seawater in Penobscot Bay, imposed habitat boundaries on the species sampled and prevented sampling the same species throughout the entire study area (Fig. 1, Table 1). American eels (*Anguilla rostrata*) were sampled in the OV reference reach and the river upstream and downstream of the plant site (BO and OB reaches). Mummichog (*Fundulus heteroclitus*) and Atlantic tomcod (*Microgadus tomcod*) were collected both upstream and downstream of the plant site (BO and OB reaches). Rainbow smelt (*Osmerus mordax*) and winter flounder (*Pleuronectes americanus*) were sampled in the river downstream of the plant, and in both reaches of the Bay (OB and ES reaches). Blue mussels (*Mytilus edulis*) and American lobster (*Homarus americanus*) were sampled only in Penobscot Bay (ES reaches), except for one subtidal mussel collection made opportunistically in the OB reach.

Muscle or whole-body (see below) samples from all fish were analyzed for total Hg and methyl Hg was analyzed in 10% or more of the fish sampled. Fresh weight and total length were recorded from each fish prior to necropsy and sampling.

American eels were collected in the BO and OB reach using eelpots baited with salted herring during July in most years, with collections in August and September in 2008 and 2009. In the freshwater OV reference reach eels were electrofished between June and August, except for collections in September in 2008. Yellow, immature eels were exclusively sampled to determine Hg exposure at the sampling site. Sexually mature silver eels migrate through the study area in the fall from upstream lakes and rivers on their way to Atlantic spawning grounds. In 2008 several of the eels electrofished from the OV reach showed characteristics indicative of migratory silver eels. We collected additional external measurements and gonad weights from all OV eels and a subset of the BO eels, to confirm whether these eels were non-migratory yellow eels or silver eels migrating down the Penobscot River from upstream sites. One female eel, OV5-1, was found to be 16 years old, had a total length of 750 mm, weighed 890 g, had a gonadosomatic index of 4.96 (GSI; gonad weight/total weight $*$ 100), and a Pankhurst eye index of 11.7 (PEI; $[((\text{horizontal} + \text{vertical eye diameters}/4)2 * 3.14159)/\text{body length}] * 100$; Cottrill et al., 2002; Pankhurst, 1982)). Given the evidence this eel was determined to be a silver eel migrating downstream in the Penobscot River and omitted from the dataset. All eels were aged by counting the annular rings on otoliths necropsied from each eel; eel length is highly variable with age and could not be used as a surrogate for age.

Mummichog were collected in the Penobscot River and bordering wetlands by beach seine, bottom trawl and in unbaited minnow traps

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