APPENDIX 1-2

Penobscot River Mercury Study

Phase I of the Study: 2006-2007

Report to:
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U.S. District Court (District of Maine)
Portland, Maine

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EXECUTIVE SUMMARY

This report provides an interim summary of the results of Phase I of the Penobscot River Mercury Study. The Penobscot River Mercury Study was ordered in 2002 by the United States District Court, District of Maine under the Resource Conservation and Recovery Act. The primary objective of Phase I of the study was to determine whether mercury levels in fish, shellfish, and wildlife found in the lower Penobscot River (Maine) and in Penobscot Bay are of concern with regard to possible human consumption or to the species themselves, particularly in relation to the location of the HoltraChem chemical factory site at Orrington, ME. The purpose of this report is to provide results from water, sediment and biota sampling from Phase I of this study. Other results concerning additional species of biota will be provided in a later updated report.

Sampling of water, sediments, benthic invertebrates, fish, shellfish, birds and mammals was carried out in the Penobscot River and estuary in 2006 and 2007 to examine mercury (Hg) and methyl mercury (MeHg) levels and spatial patterns in the river and estuary. The design of sampling for aquatic components of the river and estuary divided the river and estuary into five study "reaches". These reaches were chosen with reference to the location of the HoltraChem site and to the location of paper mills on the river. These mills may have used Hg in their past operations and could be sources of Hg to the river. The reaches were also chosen in relation to the extent of tidal surges in the river that could have moved Hg upstream from the HoltraChem site as far north as the Veazie Dam. Temporal changes in mercury concentrations were studied by sampling each reach six times between July 2006 and July 2007. Water, sediments, and benthic invertebrates were sampled at each of five discrete nearshore sites within each river reach and at 15 sites in the estuary.

Sampling of fish, birds, and mammals was, by necessity, more opportunistic, and was determined by the spatial and temporal distribution of the various species of interest.

During the summer of 2007, to determine the geographic extent of the Hg pollution, we conducted a spatial survey of wetlands which are hypothesized to be sites of potentially high rates of production of MeHg. We also conducted a spatial survey of the bottom sediments of Penobscot Bay. Concentrations of Hg in these bottom sediments were compared to those in a reference estuary (St. George River) which has no known point source of industrial Hg contamination.

Various quality assurance/quality control (QA/QC) measures were put into place to ensure the quality of the data produced. These measures included ultra-clean sampling

techniques for water, analyses of standard materials by the analytical laboratories, field blanks, field and analytical duplicates, and inter-laboratory comparisons of various sample types among three internationally recognized laboratories. Detailed methods and results for QA/QC are provided in the main body of the report.

Clear evidence for Hg contamination of the lower Penobscot River and upper estuary was found in suspended particles and in sediments of the Penobscot system. Hg dissolved in water was not found to be elevated in the lower river and estuary as compared to reaches above the Veazie Dam. Hg attached to particles suspended in the water was found to be about 2X higher downstream of the Orrington site. It appears that river flows cause the suspension of significant amounts of small particles in the lower river that are contaminated with Hg relative to the upper reaches of the river. Hg in sediments was found to be significantly elevated in the lower Penobscot River and estuary. Compared to the reference area in the East Branch of the Penobscot River, which has no known point source input of industrial Hg, Hg in sediments was approximately three times as concentrated downstream of three paper mills in the upper river, but was twenty times more concentrated in the lower river (downstream of the Veazie dam and Brewer) and in the upper estuary. Hg concentrations in the sediments of the lower Penobscot River and upper estuary were also found to be much higher than in sediments from the neighbouring St. George estuary, which has no known history of point source Hg contamination. These results indicate that whereas the paper mills in the Penobscot have elevated Hg in the river to some degree, there has been a much larger Hg source or sources downstream of the Veazie Dam, and are consistent with a large source from the HoltraChem site.

The concentration of Hg in inshore sediments of the Penobscot estuary decreased with increasing distance from the mouth of the river. The high concentrations of Hg in the sediments of the lower Penobscot River and upper estuary are similar to other contaminated sites in N. America and Europe. Perhaps most significantly, these concentrations are higher than NOAA levels of concern for toxic effects on aquatic life.

Mercury in the offshore sediments of the Penobscot estuary were highest in the upper estuary and decreased in a regular pattern to Vinalhaven Island, where they were similar to those in the uncontaminated reference estuary. Hg concentrations in riparian wetlands located in the lower river and upper estuary were also high, but showed an abrupt decrease south of Verona Island. Taken together, these results indicate that the most severe contamination of the Penobscot system is between Brewer on the lower river and about Fort Point or Sears Island in the upper estuary. Now that this spatial distribution of Hg is known, most of the work that will be proposed for Phase II of the study will be confined to these areas of high Hg contamination.

Hg in periwinkles in the Penobscot estuary were higher closer to the mouth of the river but were not high compared to concentrations at other polluted sites. Hg in freshwater snails were higher in the river reaches within tidal influence as compared to the reach immediately upstream of the Veazie dam, however it was highest in the East Branch reference area. Hg in lobster in the greater Penobscot estuary were similar to other sites in Maine but were generally higher closer to the HoltraChem site. Some individual lobsters were found to have levels of MeHg in claw and tail muscle that exceeded the Maine DEP and USEPA criteria for protection of human health for consumption of MeHg in biota. Hg in mussels was found to be high compared to other sites in Maine and the United States. Hg in mussels and periwinkles showed a geographic pattern, being higher closer to the mouth of the Penobscot River. Hg in tomcod was higher in the lower Penobscot River than at stations sampled in the estuary. Thus, in fish, shellfish and sediments there was a general pattern of lower Hg concentrations with increasing distance between the sampling site and the HoltraChem site.

Hg concentrations in the blood of three species of songbirds inhabiting wetlands adjacent to the lower Penobscot River in the Frankfort Flats area were found to be very high compared to songbirds in reference areas in other parts of Maine, and high compared to levels of concern for possible toxic effects on the birds themselves. Hg levels in cormorant eggs were relatively high compared to other locations in Maine, and were higher closer to HoltraChem, consistent with results for sediments, shellfish and fish. Hg in cormorant eggs in the upper estuary approached or exceeded levels thought to impair reproduction.

We attempted to assess Hg concentrations in mammals (otter and mink) in areas adjacent to contaminated and uncontaminated reaches of the Penobscot River. However, the numbers of individuals that we were able to obtain were too small to give us statistically defensible results. Samples obtained did not show consistent patterns related to possible exposure mercury from the HoltraChem site. Concentrations were similar to other regions of North America and the world. Levels in otters were generally not high enough to cause concern related to toxic effects on that species but Hg concentrations in the fur of mink were often above the level at which toxic effects have been demonstrated for that species, particularly at sites potentially contaminated by Hg from the Holtrachem site.

We also assessed the potential use of measurements of the ratio of stable isotopes of Hg to determine the amount and extent of contamination of Hg from the HoltraChem site. We found that the isotope ratios of Hg sampled from the HoltraChem site were significantly different from Hg found outside of the aquatic influence of HoltraChem Hg,

indicating that the stable isotope fingerprinting techniques have potential for assessing the impact of HoltraChem.

Four criteria were used to decide whether the environment and biota of the Penobscot River and estuary have high enough levels of mercury to be of concern to an extent that justifies us proceeding to Phase II of the project and whether the source of that mercury appears to the HoltraChem plant site. These four criteria were:

1. Comparison of Hg data from the Penobscot with agency guidelines (National Oceanic and Atmospheric Administration, Maine Department of Environmental Protection, United States Environmental Protection Agency) for toxic effects on biota and for human consumption of fish and shellfish.
2. Evaluations of data by internationally recognized toxicologists and comparison to the scientific literature on toxic effects.
3. Geographical patterns of Hg in water, sediments, and wildlife within the Penobscot system, especially in relation to the HoltraChem plant site.
4. Comparisons of Hg concentrations data to other known uncontaminated and contaminated freshwater and estuarine sites.

Based on the above criteria, we conclude that there is sufficient weight of scientific evidence to conclude that the Penobscot River and estuary are contaminated with Hg to an extent that poses endangerment to some wildlife species and possibly some limited risk for human consumers of fish and shellfish. We further conclude that these data justify our recommendation for the study to proceed to its second phase.

The specific data that lead us to this conclusion are:

With respect to Criterion 1: Downstream of Brewer and in the upper estuary, concentrations of total Hg in sediments exceed NOAA guidelines for toxicity to benthic fauna. Some lobster in the upper estuary exceeded MDEP and USEPA criteria for protection of human health for consumption of MeHg in biota (25% of the lobster sampled in the upper estuary exceeded the Maine criterion and 6% exceeded the EPA criterion).

With respect to Criterion 2: Hg levels in some species of songbirds inhabiting wetlands in the lower Penobscot were found to be of concern for the health of those species. For example, Hg in the Nelson's sharp-tailed sparrow was much higher than concentrations thought to be toxic by avian toxicologists in a related species. Hg in the eggs of cormorants was also probably high enough to impair reproduction.

With respect to Criterion 3: We conclude that there has been a large point source of Hg to the ecosystem from a location downstream of Veazie Dam. The pattern of contamination of the sediments of the Penobscot River and estuary was not consistent with contamination from paper mills on the river or from regional atmospheric deposition of Hg, but was consistent with a large source from the HoltraChem site at Orrington. The spatial pattern of contamination of various species of biota, such as periwinkles, mussels, lobsters, tomcod (fish) and cormorants (birds) was also consistent with elevated inputs of Hg to the lower Penobscot River below the Veazie dam.

With respect to Criterion 4: Hg concentrations in sediments, songbirds, cormorant eggs, and mussels were high compared to uncontaminated sites and as high as many other sites known to be contaminated with point sources of Hg.

We therefore recommend that the Study proceeds to Phase II. Phase II of the study will concentrate on understanding where and when MeHg is produced in the system, and how it is transported and bioaccumulated in the lower river and upper estuary. Emphasis will also be placed on determining rates of ongoing input from the HoltraChem site and from other industrial and municipal sources. These data are needed to estimate rates of natural attenuation of Hg in the ecosystem, which will be an important topic of study in Phase II. All of these data will be used to evaluate the practicality of possible mitigation measures.

TABLE OF CONTENTS

INTRODUCTION

The purpose of this report is to summarize the results of Phase I of the Penobscot River Mercury Study. This study resulted from a ruling from the U.S. District Court, District of Maine (Civil No. 00-69-B-C; Plaintiffs Maine People's Alliance and Natural Resources Defence Council, Inc. Vs. Defendants Holtrachem Manufacturing Company LLC and Mallinckrodt Inc.) in 2002 under the Resource Conservation and Recovery Act that a study of mercury in the Penobscot River and estuary would be carried out. The Penobscot River Mercury Study is managed independently of the Defendants and the Plaintiffs. A Study Panel consisting of three scientists was appointed by the Court to oversee the study and to set the overall study plan. One member of the Panel was appointed by the Defendants, one member was appointed by the Plaintiffs, and a third (Chair) was recommended by the two existing Panel members. All members of the Panel were independent of either party once appointed. The Panel recommended the hiring of a Project Leader to coordinate and oversee the operational aspects of the Study, to analyze data resulting from the study and to be the primary author of study reports. The Study Panel also authored the Study Plan (Penobscot River Study Panel 2005), which was subsequently approved by the Court.

The Study Plan was implemented by a series of specific research proposals that were taken from the broad objectives of the Study Plan. These proposals were drafted by the Project Leader after discussion with the Study Panel, and re-drafted with comments from the Study Panel. Each proposal was then submitted to the Court and, if approved by the Court, was implemented by the Project Leader, usually using outside contractors, with continuing advice from the Study Panel.

Mercury (Hg) cycling in aquatic environments is complex. It is relatively straightforward to measure Hg concentrations in various media (e.g. sediments, biota). However, it is much more difficult to understand the movements and chemical transformations of Hg or the impact that Hg is having on animals and plants. Hg can not only be transported physically in the environment, it can also be transformed chemically into different compounds and bioconcentrated in food chains. In general, most of the Hg in the environment is in various inorganic forms, such as ionic Hg and elemental Hg (Munthe et al. 2007). However, it is the organic compound methyl mercury (MeHg), which is produced from inorganic Hg, that poses the greatest risk to wildlife (Scheuhammer et al. 2007) and to people eating fish and shellfish (Mergler et al. 2007). Inorganic forms of Hg do not biomagnify progressively as one moves up food chains, but MeHg does (Wiener et al. 2003). For example, concentrations of MeHg in fish muscle are greater than in the food they eat and typically one million to ten million times those in the water in which they live (Wiener et al. 2003). The conversion of ionic Hg to MeHg is done by

microorganisms (primarily sulphate-reducing bacteria) that live in sediments and water (Wiener et al. 2003). The rate of conversion of inorganic Hg to MeHg is controlled by many factors, including temperature, microbial activity, the availability of inorganic Hg, salinity, pH, and the presence of various compounds of organic carbon, sulphur and selenium which can complex with Hg (Munthe et al. 2007). MeHg can be broken down by microbial activity and by light. MeHg probably enters the base of the food chain mainly by uptake directly from water but transfers and biomagnifies with each step in the food chain (trophic level) so that it reaches higher concentrations at higher trophic levels (Wiener et al. 2003). Human activities can contribute to higher concentrations of MeHg in aquatic food chains, including activities that increase the amount of Hg in the atmosphere such as the burning of fossil fuels (Hammerschmidt and Fitzgerald 2006), the direct discharge of Hg to water (Southworth et al. 2000; Herut et al. 1996), and the flooding of land for reservoir creation (Bodaly et al. 2007).

The purpose of the Penobscot River Mercury Study is to provide an independent assessment of whether there are elevated levels of Hg in the lower Penobscot River and estuary as a result of contamination from the HoltraChem chlor alkali plant site at Orrington, Maine. The Study Plan set out the overall objectives of the Study and the main components of the Study. The Study Plan (Penobscot River Study Panel 2005) outlined a phased approach to the study of Hg in the river and estuary. Specifically, in the first phase of the study "...the whole ecosystem will be monitored in the river and estuary to determine if concentrations of mercury in fish and wildlife are high enough to be of concern." Additional studies of mercury in potential methylation hotspots and the extent of contamination in the Penobscot estuary were carried out in response to comments and recommendations from the parties. The Study Plan states that if it is concluded that concentrations of Hg are high enough to be of concern, the study would move to the second phase. This second phase would concentrate on understanding factors controlling the production, transport and bioaccumulation of MeHg and its toxicity, so that mitigative measures, if practical, could be recommended.

It is the purpose of this report to provide a summary of data collected in the first phase of the Study. Although not all data collected could be included here, the weight of evidence available to us enables us to recommend moving on to Phase II now, so that field work can continue in the summer of 2008. Additional information, in the form of updates to this report, will be provided later. These updates will include data on Hg in nearshore sediments from the fifth and sixth sampling periods, mercury in a variety of biota including annelid worms, scallops, crabs and clams, eels, estuarine fish, and data on the MeHg content of wetlands and offshore sediments. Bird data including those from eagles, osprey and kingfishers will also be available, as will additional data on Hg in cormorants and songbirds.

METHODS

Field and analytical contractors

Aquatic sampling, including water, sediment and aquatic biota was conducted by Normandeau Asociates, Inc. (Yarmouth ME and Bedford NH), under the direction of Marcia Bowen. Field sampling of birds and mammals was conducted by Biodiversity Research Institute (Gorham ME) under the direction of David Evers. Water, sediments and biotic tissues were analyzed for mercury by Battelle Marine Sciences Laboratory, Sequim WA (Gary Gill and Brenda Lasorsa), Studio Geochimica, Seattle WA (Nicolas Bloom) and Flett Research Ltd., Winnipeg, Canada (Robert Flett). Samples for stable isotope analysis were analyzed by Trent University, Department of Chemistry, Peterborough, Canada (Holger Hintelmann). All of the above laboratories, except for Studio Geochimica, were involved in interlab comparisons.

Sampling design

Phase I of the study had two primary focuses. The first was to determine if sediments and biota in the area of the HoltraChem site are contaminated with Hg. The second focus was to determine if the source of the Hg was from regional atmospheric deposition of Hg, from Hg lost from paper mills in the area, or if most of the contamination originated from below the Veazie dam where HoltraChem is located. To accomplish these goals, five reaches of the river and estuary were sampled during six separate periods. The river and estuary was divided into five reaches, four in the river, plus the estuary (Figure 1). These reaches were chosen with reference to the location of the Orrington plant site and the five currently operating or previously operating paper mills on the river, which may have used Hg in the past. The division of the river and estuary into five sampling areas was done to allow direct comparisons of the effects of different influences of Hg on the river, including the Orrington plant. The five reaches were:

1. East Branch (EB) (Figure 2): the downstream section of the East Branch of the Penobscot River. This sampling reach was intended to serve as a reference section of the river. It is upstream of all paper mills, the tidal influence of the river, and of significant human population. This background reach does, however, receive Hg from natural weathering of crustal material and from atmospheric deposition of Hg.
2. Old Town – Veazie (OV) (Figure 3): the reach of the main stem of the Penobscot River between the city of Old Town and the Veazie Dam. This sampling reach is downstream of three paper mills (Millinocket, Lincoln and Old Town) but is upstream of any tidal influence. This reach receives Hg from weathering, atmospheric deposition,

and possibly from the three upstream paper mills, that probably used Hg in their past operations. Any Hg that originated from Orrington would therefore not be able to move upstream into this sampling reach by a water route.

3. Brewer – Orrington (BO) (Figure 4): the reach of the main stem of the Penobscot River between the paper mill at Brewer and the HoltraChem chlor-alkali plant site. This reach is downstream of an additional paper mill and is upstream of Orrington but within the influence of Orrington due to tidal movements.

4. Orrington – Bucksport (OB) (Figure 5): the reach of the main stem of the Penobscot River between the Orrington chemical plant site and the paper mill at Bucksport. This reach is downstream of the Orrington site.

5. Estuary (ES) (Figure 6): the upper part of the Penobscot River estuary, from Bucksport to middle of Islesboro Island. The estuary receives Hg from all of the above named sources.

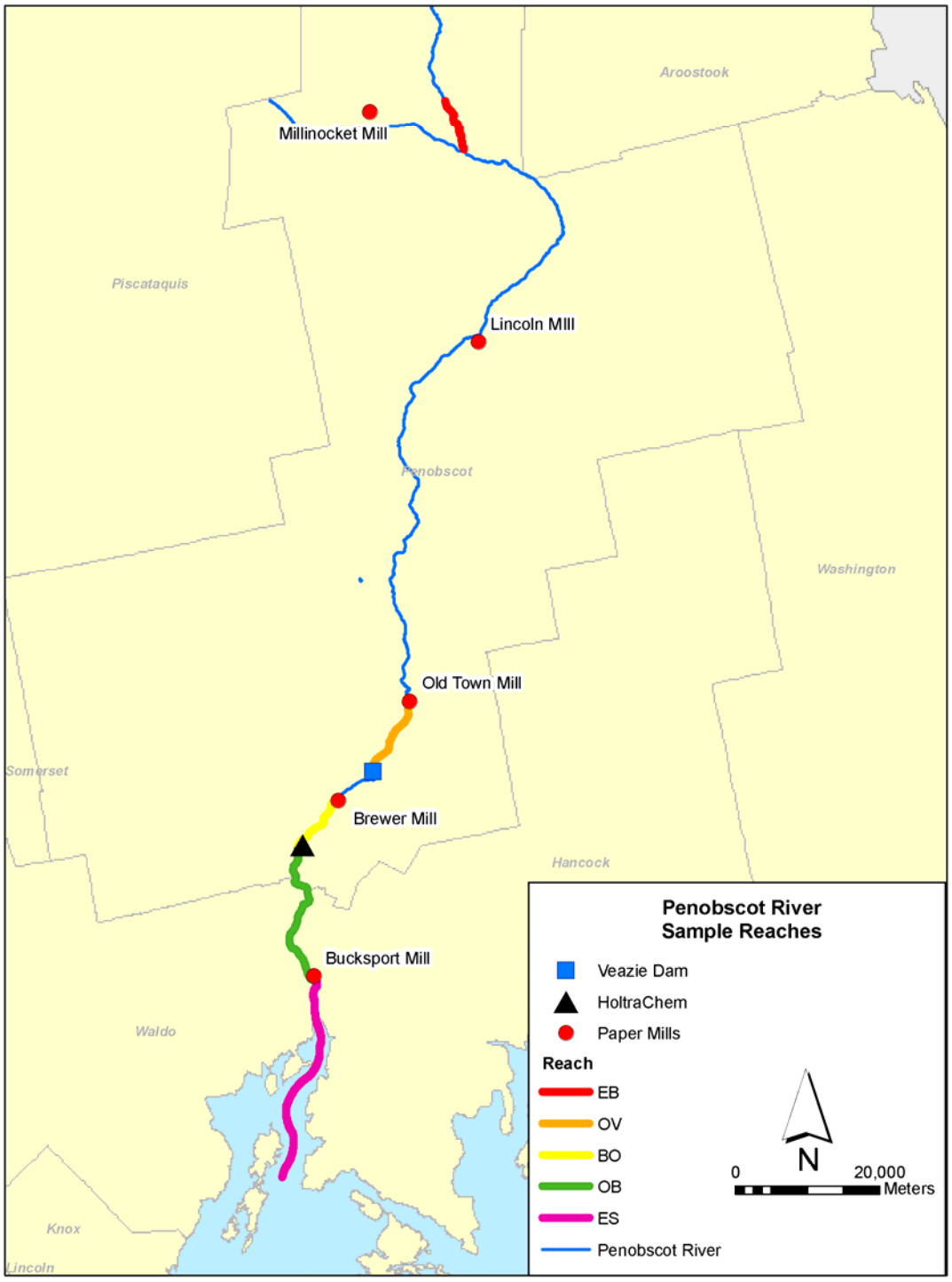

Figure 1. Map of the Penobscot River and estuary showing the locations of sampling reaches, active and inactive paper mills, the Veazie dam, and the HoltraChem site at Orrington. The locations of sampling sites within each reach are shown in Figures 2, 3, 4, 5, and 6.

Five sites were sampled within each of the four river reaches, and 15 sites were sampled in the estuary. The locations of each site are shown on maps in Figures 2, 3, 4, 5, and 6. A site was defined as a shallow (intertidal in the lower river and estuary) section of shore 50 m long with fine grained sediments (sediments fine enough that they could be cored by a 3 inch diameter piston corer). Of the five sites in the river reaches, three were randomly chosen and two were purpose-chosen to represent areas that may have higher rates of mercury methylation due to the presence of higher densities of aquatic vegetation, and more organic-rich fine grained sediments. In the estuary, 7 of the fifteen sites were randomly chosen, three were chosen to duplicate sampling sites used in 2004 by Dr. Celia Chen of Dartmouth College, and five were chosen to be "hotspots".

Aquatic sampling was conducted during six periods to determine temporal differences in concentrations of total Hg and MeHg in the Penobscot River and estuary. Sampling was confined to warmer, ice-free months and was intended to complete an annual cycle, as follows:
Sampling I: late July to early August 2006 (July 31 - August 8)
Sampling II: early September 2006 (September 6 - September 11)
Sampling III: late September to early October 2006 (September 26 - October 1)
Sampling IV: late October to early November 2006 (October 22 - November 6)
Sampling V: late May to early June 2007 (May 29 - June 1)
Sampling VI: early July 2007 (July 9 - July 11)

Aquatic sampling included water, sediments, some invertebrates (e.g. snails and periwinkles) and some fish samples (e.g. cyprinids). Other biota samples were taken, by necessity, where the animals could be found. This sampling included birds, mammals, and some fish (e.g., eels, tomcod) and shellfish (e.g. lobster, mussels). Sampling methods are detailed below.

Sampling methods

All samples were accompanied by chain-of-custody forms. Forms were completed in the field when samples were taken. Forms were signed and dated by a member of the sampling crew when they were relinquished to another person (generally the head of the field laboratory or the person shipping the samples), who signed the form as having received the samples. The same procedure was used when samples were received by the analytical laboratory.

Water - Water sampling in the field was conducted using strict trace metal clean protocols. All samples were collected and stored in acid-cleaned, double-bagged Teflon bottles or new, double-bagged glass I-Chem bottles. Samplers wore new, powder free latex gloves. One person ("clean hands") touched only the outside of the sampling bottle. The other person ("dirty hands") touched only the outside of coolers, bags holding sampling bottles, and the pump and associated tubing. All water samples were placed on ice packs in coolers in the field and were shipped to the analytical laboratory on the same day they were collected so they could be preserved in the lab within 48 hours of collection.

All water sampling in the tidal reaches of the river and the upper estuary was conducted on falling tides.

For the first sampling round, water was taken whole in the field, shipped overnight to the analytical laboratory (Studio Geochimica) and filtered, as required, in the lab. Four bottles of whole water were taken at each site. Two of these were used for unfiltered determinations and two were filtered in the lab for dissolved determinations. For other sampling rounds, two samples of unfiltered water and two of filtered water were taken at each site. Water was filtered in the field using a peristaltic pump, acid-cleaned Teflon tubing and 0.45 micron in-line cartridge filters (Whatman Polycap Groundwater Capsule filter). All samples were shipped overnight to Battelle Marine Sciences Laboratory or Studio Geochimica for preservation within 48 hours of collection. Studio Geochimica was replaced by Battelle Marine Sciences Laboratory after the first sampling period and unanalyzed samples were sent from Studio Geochimica to Battelle for analysis.

Suspended solids (total suspended sediments, or TSS) samples were taken at each station using Nalgene bottles, rinsed with ambient water before filling. TSS in water was determined using standard method 2540D (Greenberg et al. 1992). TSS is the material retained on a standard glass fiber filter. All TSS samples were analyzed in duplicate in the laboratory.

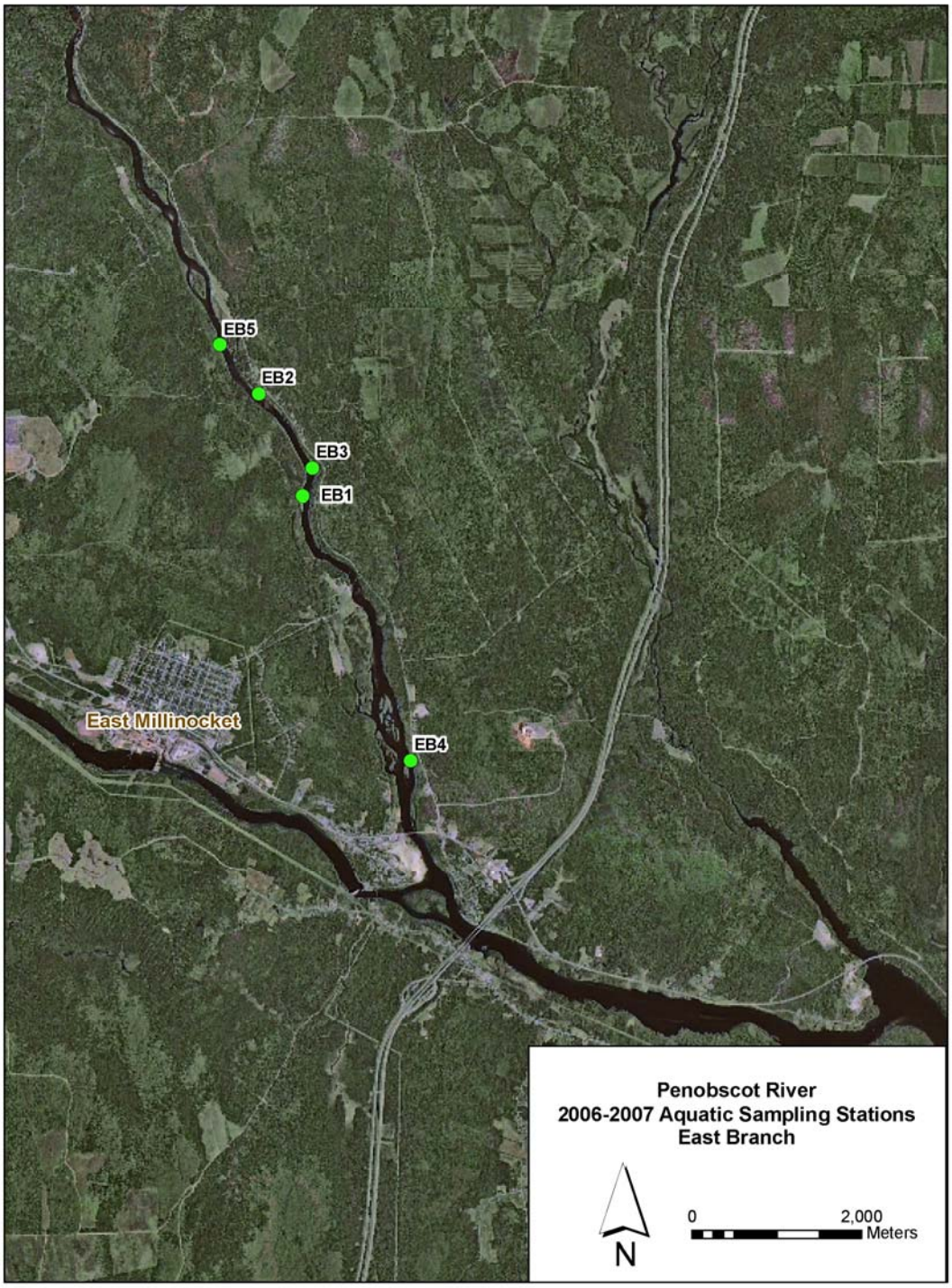

Figure 2. Map of East Branch sampling sites, 2006-2007.

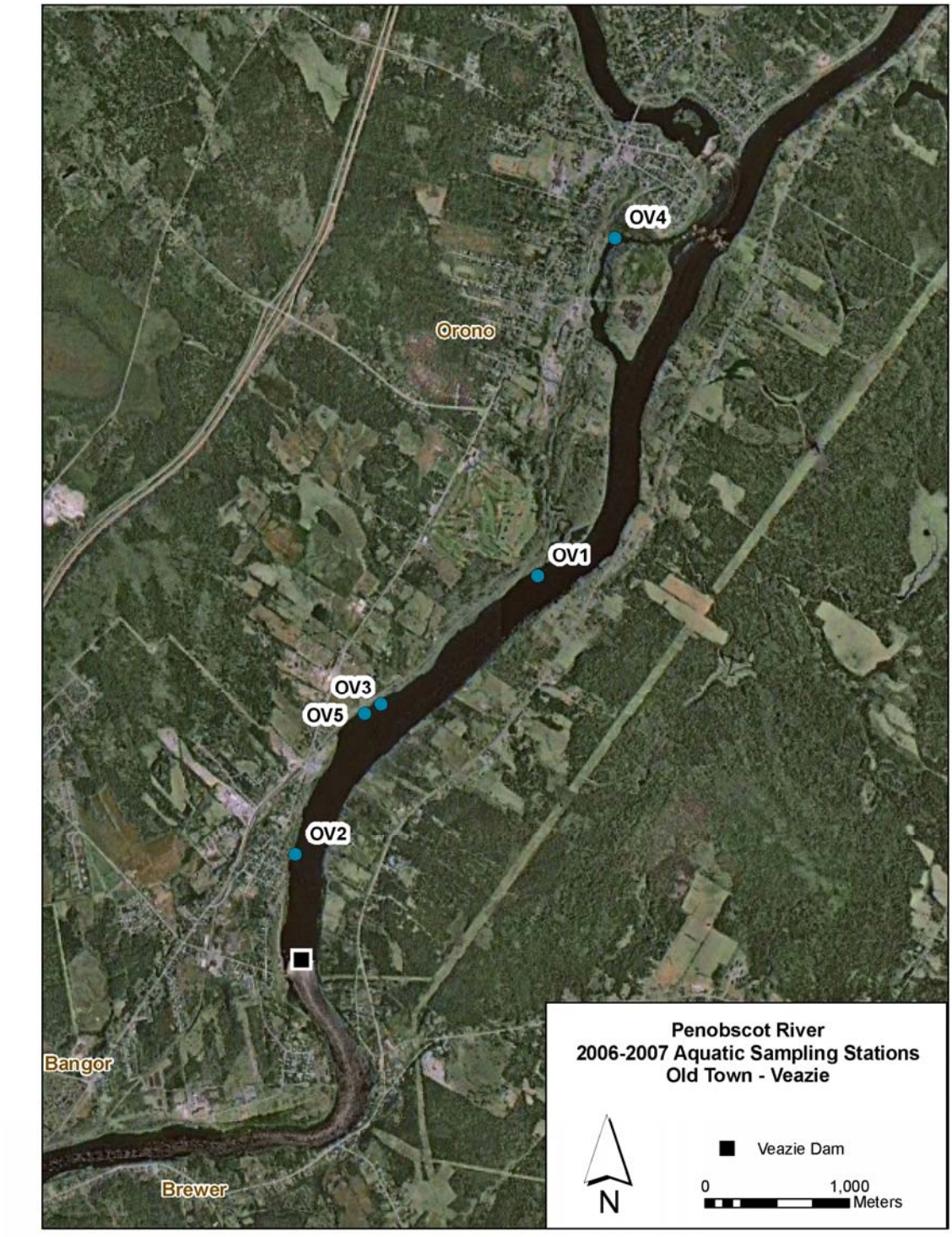

Figure 3. Map of Old Town – Veazie sampling sites, 2006-2007.

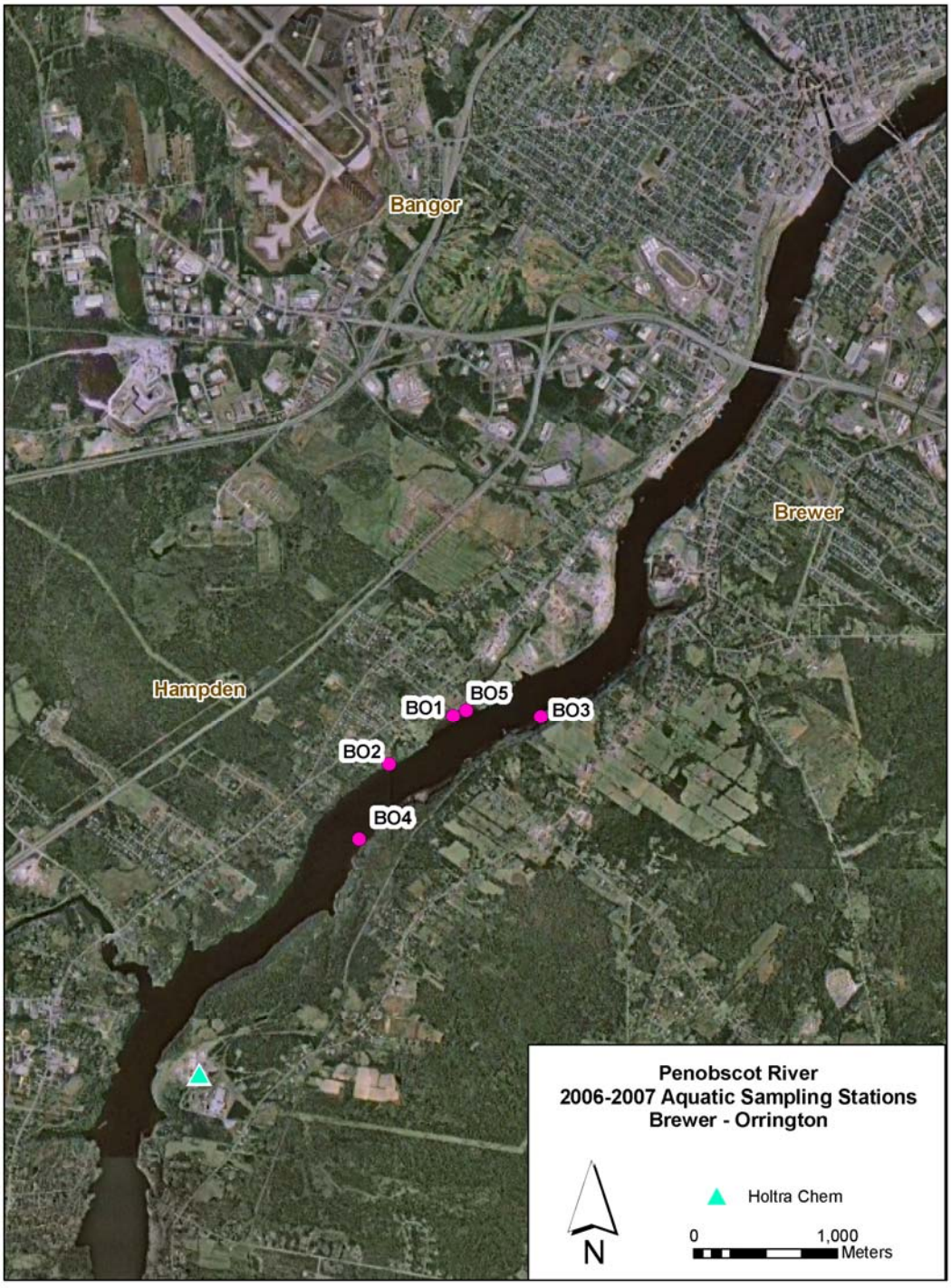

Figure 4. Map of Brewer – Orrington sampling sites, 2006-2007.

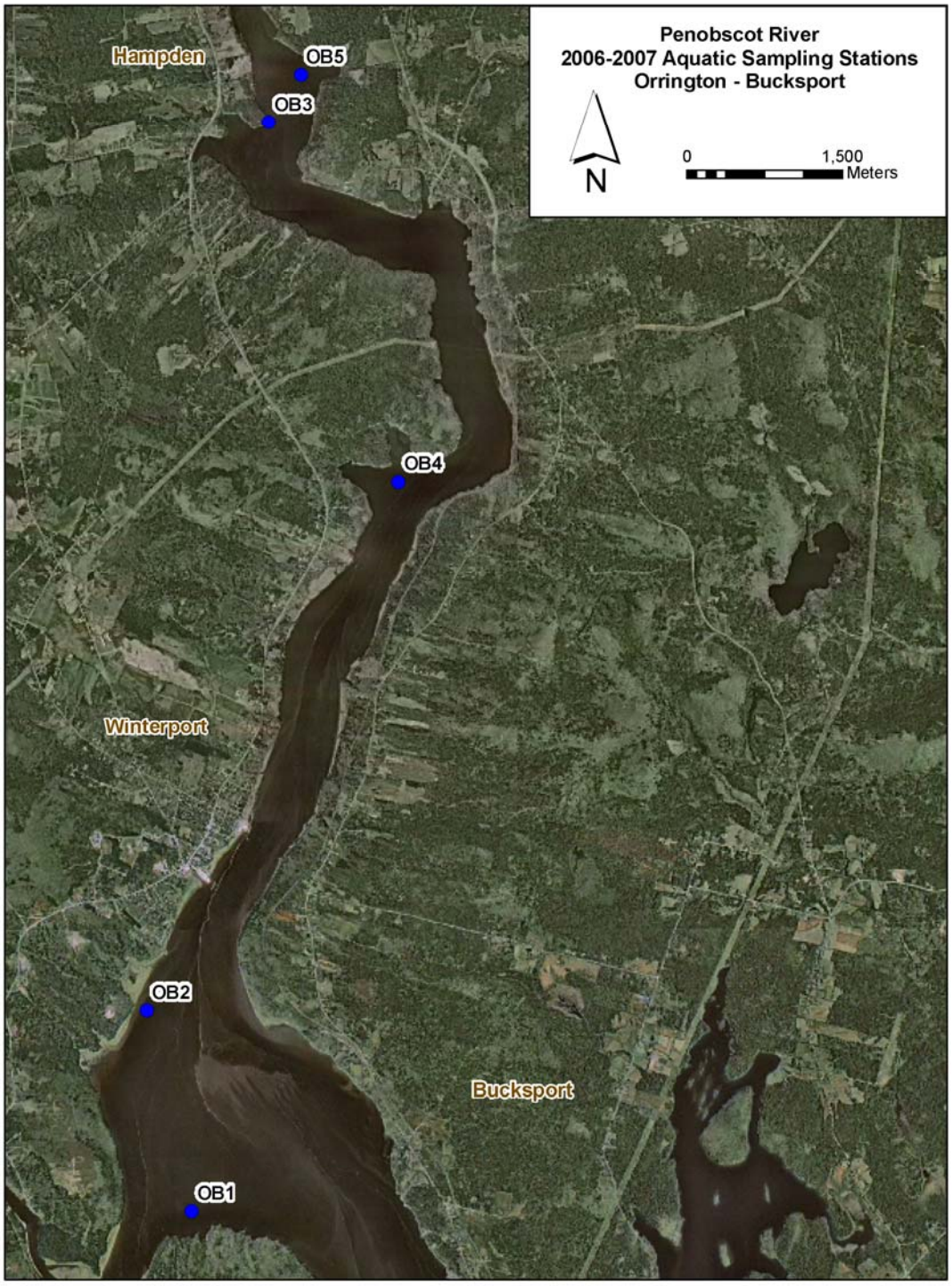

Figure 5. Map of Orrington – Bucksport sampling sites, 2006-2007.

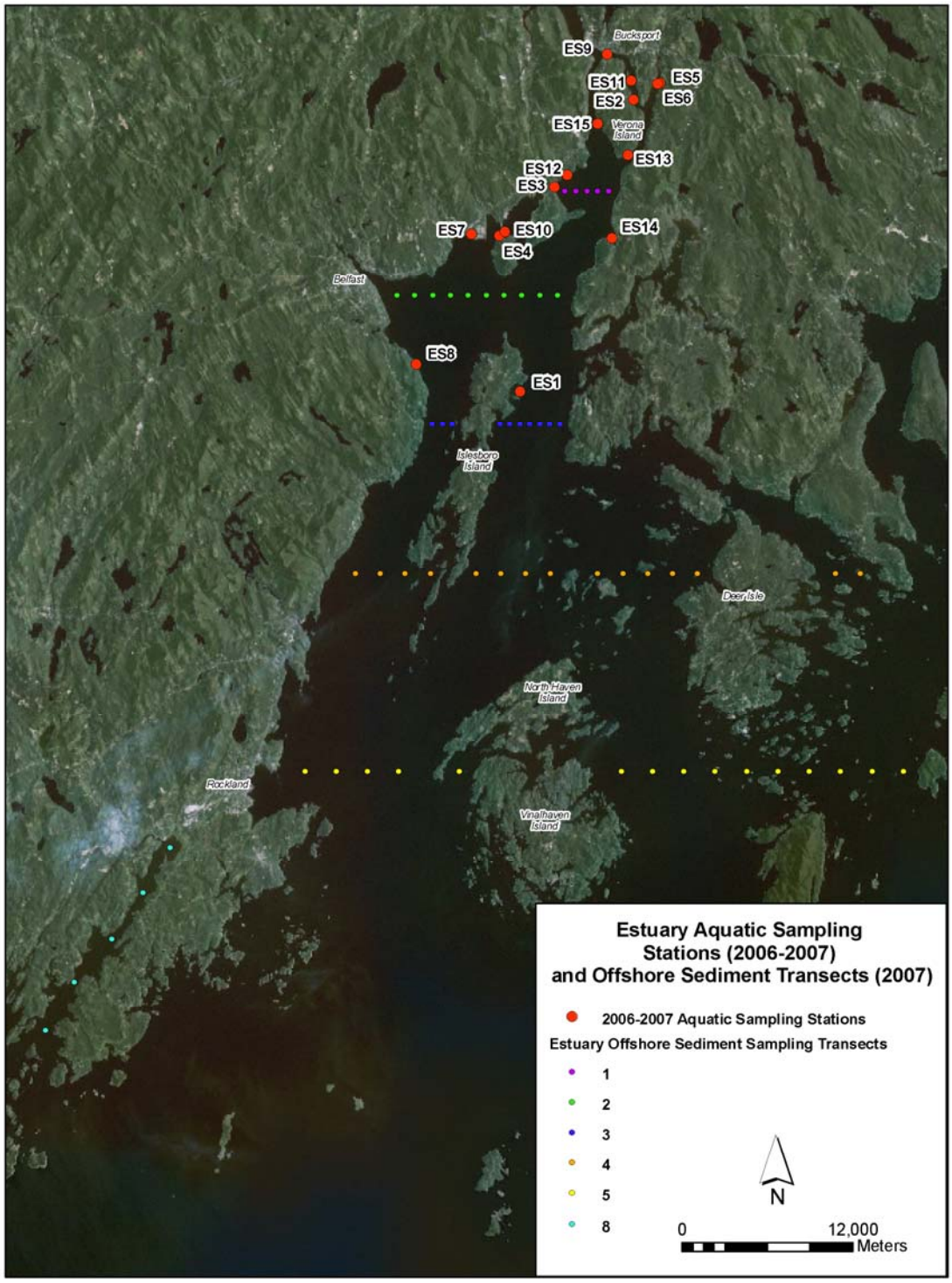

Figure 6. Map of Estuary (ES) aquatic sampling sites, 2006-2007. Also shown are the locations of sampling transects and sites in the estuary sampled for offshore surficial sediments in August 2007. Transects 1-5 are in the Penobscot estuary; Transect 8 is in the St. George estuary.

Sediments – Sediments were sampled using three different approaches:

1. For the six rounds of aquatic sampling in 2006 and 2007, a 3-inch diameter piston corer was employed at each of the five sites in the four river reaches and the 15 sites in the estuary. Cores were extruded and sliced in the field. Mercury is reported as concentrations over the top 3 cm of sediment. Ten cm cores were sliced every 1 cm and concentrations were reported for the top 3 cm as the arithmetic average of the top three slices. The top 3 cm of sediment was sampled as a single slice for 3 cm cores.

2. In August 2007, samples were taken from the offshore sediments of the Penobscot River estuary. Five to 15 stations on a series of transects that ranged from Fort Point Cove in the North to Vinalhaven Island in the South were sampled. Transect and station locations are shown in Figure 6. Five stations in the estuary of the St. George River were also sampled to serve as a reference for the Penobscot system (Figure 6). Stations in the Bagaduce river estuary were planned but could not be sampled because of the lack of soft substrate.

Sediments were sampled using a stainless steel Van Veen dredge (area 0.04 m^2) operated by a hydraulic winch. The Van Veen dredge was thoroughly washed between each station using ambient water. The top 3 cm of sediment was removed with a stainless steel spoon and placed into a washed, stainless steel bowl. Samples were then well mixed, and split into subsamples for the determination of total and methyl mercury, organic carbon, and grain size composition. Sediments for mercury determination were frozen within one minute of exposure to air by placing them on dry ice. Also, at every fifth station, a subsample was taken for mercury stable isotope analyses. Samples were not obtained at every site on Transects 2, 4, and 5 because of occasional rocky substrates.

3. Also in August 2007, sediments and soils from 27 wetlands adjacent to the river and upper estuary were sampled to determine the degree and extent of contamination by mercury and methyl mercury. There were 11 wetlands sampled in the lower river, 6 in the Bagaduce River vicinity (including near the mouth of the Bagaduce estuary), and 10 in the upper estuary (Figure 7). Wetlands were initially identified from an aerial survey of the lower river and a boat survey to confirm wetland characteristics. Forty wetlands were identified but wetlands that appeared to be similar to other nearby wetlands were not sampled to reduce costs. A map showing the locations of wetlands is shown in Figure 7 and the geographic coordinates of the wetlands surveyed and sampled are shown in Appendix 3. At each wetland, samples of sediment/soil were taken from four elevations: intertidal below the vegetated zone, intertidal within the vegetated zone but

below the frequent high tide mark, intertidal within the vegetated zone but above the frequent high tide mark and a very high elevation sample that was above the debris line indicating infrequent high tides. Not all elevations could be sampled at every site because of the occasional presence of hard substrate. Sediment was removed to 3 cm depth over an area of approximately 75 cm^2 at three sites within the wetland (at each elevation) with stainless steel knives, placed into washed, glass bowls and mixed thoroughly with stainless steel scissors and knives. The mixed, combined sample was then subsampled into portions for total and methyl mercury determination, grain size analysis and organic carbon content determination. Sediments for mercury determination were frozen within one minute of exposure to air by placing them on dry ice. Subsamples for mercury stable isotope analysis were also taken at five wetlands - two in the lower river, two in the Bagaduce River, and one in the upper estuary (Appendix 3).

For the first three aquatic sample rounds, separate subsamples were taken for total mercury and methyl mercury. Total mercury samples were kept cool whereas methyl mercury samples were frozen on dry ice within one minute of being exposed to air. For the last three aquatic rounds of samples, estuary sampling and wetland sampling, a combined sample for total mercury and methyl mercury was taken and frozen within one minute of being exposed to air.

Biota – Freshwater snails (*Lymnaea megasoma*) were collected at all stations on the Penobscot River, during all four of the aquatic sampling periods in 2006. Snails were frozen soon after sampling and shipped to the analysis laboratory frozen, where the soft tissues were freeze-dried and weighed. Statistical tests showed that Hg in snails during Sampling II (August 2006) were higher than the other sampling periods (which were not significantly different), so snails from Sampling II were treated separately. Snails larger than 0.1 g dry weight were eliminated from the analysis in order to facilitate comparisons reaches by eliminating statistically significant differences in snail weights among reaches. All mercury data were log transformed to create normal distributions required for statistical tests. Averages by reach were presented as geometric means because of the disproportionately large amount of variation seen in samples from the Old Town-Veazie sampling reach.

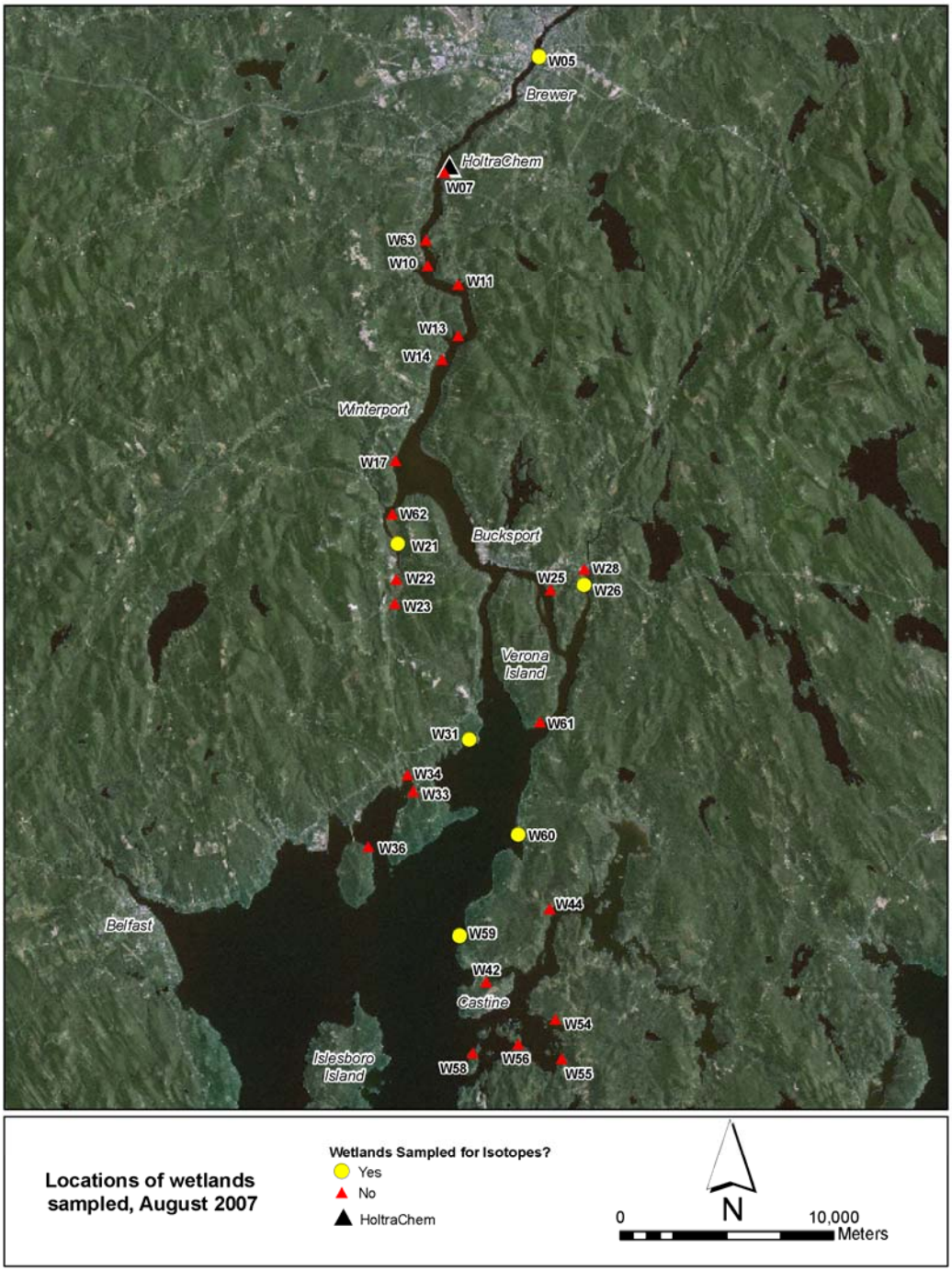

Figure 7. Locations of wetlands sampled for mercury in August, 2007. Symbols indicate whether each wetland was sampled for mercury only (red triangle) or mercury and mercury stable isotope ratios (yellow circle). See Appendix 3 for the geographic coordinates of wetlands surveyed and sampled.

Common periwinkles (*Littorina littorea*) were collected at twelve of the estuary sampling sites during all four of the aquatic sampling periods in 2006. Periwinkles were frozen soon after sampling and shipped to the analysis laboratory frozen, where the soft tissues were freeze-dried and weighed. Hg in periwinkles was not statistically different by sampling time, so data from all sampling periods were considered together. Hg was not adjusted for animal size because the proportion of the variation in Hg or MeHg that was explained by animals size was small (R^2's were 0.01 – 0.1). Statistical comparisons were performed on log transformed Hg data to provide normal distributions.

Samples of blue mussels (*Mytilus edulis*) were taken at ten of the estuary sampling stations, during early September, 2006 and during late September/early October, 2006. Mussels were sampled wherever they occurred naturally. They were not found at stations in the upper estuary/lower river. Mussels were frozen soon after sampling and shipped to the analytical laboratory frozen, where the soft tissues were freeze-dried and weighed.

American lobsters (*Homarus americanus*) were sampled in the Penobscot estuary on three dates in 2006: September 6, 12, and 29. A biological sampler accompanied a commercial lobsterman. Sites were specific to lobster and were therefore not the same as regular aquatic sampling sites. The 18 sites ranged from the upper estuary, adjacent to Fort Point Cove, to the lower estuary, between Cape Rozier and Islesboro Island, approximately mid-island. The length of each lobster was determined in the field (carapace length), sex was determined, and a claw was removed for mercury analysis. A subsample of animals was collected whole for comparison of mercury in tomalley, tail muscle, and claw muscle. Mercury data were not standardized for lobster size because there was only a weak, statistically non-significant relationship between mercury and carapace length (Appendix 12). All mercury data were log transformed to create normal distributions required for statistical tests. There was little relationship between %MeHg and total Hg in lobster claw muscle (Appendix 11).

Atlantic tomcod (*Microgadus tomcod*) were sampled in the lower parts of the Penobscot River and in the upper estuary using trawl nets during the period September 9, 2006 to October 9, 2006. Fish length (total length) and fresh weight was determined for each fish. Total mercury was determined on samples of muscle on all fish, and methyl mercury concentrations were determined on muscle samples from every tenth fish. Because total mercury concentrations were usually significantly related to fish length,

mean mercury concentrations were adjusted for fish length by linear interpolation to a standardized total length of 140 mm.

The eggs of double-crested cormorants (*Phalacrocorax auritus*) were sampled in July and August 2006. Five sites were sampled, ranging from Fort Point in the upper Penobscot estuary to Robinson Rock in the lower estuary. The locations and sampling dates are shown in Appendix 14. Sample sizes ranged from one to 10 eggs. Eggs were analyzed for total mercury by Battelle Marine Sciences Laboratory.

Nelson's sharp-tailed sparrow (*Ammospiza caudacuta*), song sparrows (*Melospiza melodia*) and swamp sparrows (*Melospiza georgiana*) were sampled at two sites on the lower Penobscot River, Mandell Marsh and Winterport, in July and August 2006. Mist nets were used to capture birds live and samples of blood were taken from veins located in each bird's wing. The location of sampling sites and sampling dates are given in Appendix 15.

Tissue samples from mink (*Mustela vison*) and river otter (*Lontra canadensis*) were taken in 2006. Sampling was conducted from carcasses of animals captured by trappers and from living animals that were live-captured, sampled, and released back to their environment. Only blood and fur samples were taken from live-captured animals whereas brain, fur, liver and muscle samples were taken from carcasses obtained from trappers. Sites where animals were sampled were classified, based on their location, as being either potentially contaminated with mercury from the Orrington HoltraChem site (on the lower Penobscot River or estuary, downstream of the Veazie Dam, or on water directly connected to the lower Penobscot) or as reference sites (not on the lower Penobscot or estuary or on water directly connected to the lower Penosbscot). For mink, reference sites included the East Branch of the Penobscot River as well as other sites in Penobscot River watershed, distant from the river. The one potentially contaminated site for mink was the S. Branch of the Marsh R., near the town of Prospect. For otter, reference sites also included the East Branch of the Penobscot River as well as other sites in Penobscot River watershed, distant from the river. Two potentially contaminated areas were sampled for otter, one on Reeds Brook (Hampden) and on the Bagaduce R. (near the town of Castine). Potentially contaminated sites were compared to reference sites by unpaired t-tests performed assuming equal or unequal variances after an F-test to determine whether sample variances were statistically significantly different.

Invertebrate tissues were analyzed for both total Hg and MeHg. Fish muscle or whole fish were analyzed for total Hg, with a 10% subset analyzed for MeHg. Bird tissues were analyzed for total Hg only, except for brain which was analyzed for MeHg and total

Hg. In mammals, all tissues were analyzed for both total Hg and MeHg except for blood, which was analyzed for total Hg only.

Analytical methods

At Battelle, total mercury in water was analyzed by US EPA Method 1631e, using atomic fluorescence, following distillation, by the method of Horvat et al. (1993), and then digested with bromine monochloride oxidation for a minimum of 24 hours. Then, mercury in the sample was reduced to Hg^{o} with $SnCl_2$, and purged onto gold traps. Mercury vapour was thermally desorbed to a second analytical gold trap and carried into a fluorescence cell by an inert gas. The MDL was 0.188 ng/L. At Flett Research, total mercury in water was determined by EPA Method 1631e, by oxidation, purge and trap and CVAFS (cold vapour atomic fluorescence spectrophotometry). The method detection limit (MDL) was estimated to be 0.04 ng/L and the estimated uncertainty was 14.7% at 0.2 – 50 ng/L. Reference material run with each day's samples consisted of large batches of water made with THg concentrations within the range of usual samples and Baker Quality Control Solution with a certified concentration of 1000 ng/L.

Methyl mercury in water was analyzed by EPA Method 1630 (atomic fluorescence) after distillation at both Battelle and Flett Research. Methylmercury in the sample was ethylated, purged onto carbon traps, and then thermally desorbed into a fluorescence cell. Methyl mercury in water was analyzed at Flett Research by EPA Method 1630, using distillation, ethylation, purge and trap, and CVAFS. The MDL was 0.048 ng/L and estimated uncertainly was 22% at 0.05 ng/L. Reference material was MeOPR (1000 ng/L).

At Battelle Marine Sciences Laboratory, sediment samples were analyzed for total mercury by EPA Method 7473 (thermal decomposition, amalgamation and atomic spectrophotometry) using a Direct Mercury Analyzer (DMA). The standard reference material used was IAEA-405 and the criterion for recovery from the standard material was 80-120%. Sediment samples were analyzed for methyl mercury at Battelle by EPA method 1630 with extraction. Sediments were extracted by the method of Bloom et al. (1997), followed by ethylation and then concentration onto carbon traps. The ethylated methyl mercury was thermally desorbed into a fluorescence cell. The standard reference material used was IAEA-405. The criterion for recovery was 65-135%.

At Flett Research, sediment samples were analyzed for total mercury by EPA Method 1631e after digestion (CVAFS). The MDL was 2.4 ng/g. The certified reference material used was Mess-2 from the National Research Council of Canada. Sediment

samples were analyzed for methyl mercury at Flett Research by EPA Method 1630 with distillation (CVAFS). Ethylation was followed by purge and trap and the sample was then thermally desorbed into a fluorescence cell. The MDL was 0.02 ng/g. The certified reference material used was IAEA 405.

Matrix effects spikes were carried out on each day that samples were run at both Flett Research and Battelle, by adding a known quantity of total mercury or methyl mercury to sediments to determine the recovery efficiency of the method.

Sediments were analyzed for organic carbon content by Northeast Laboratories using method SW-846 9060. Sediments were analyzed for grain size distribution by Sevee and Maher Engineers or Normandeau using ASTM C92-95 (2005) (Standard Test Methods for Sieve Analysis and Water Content of Refractory Materials, Dry Sieve Analysis #8212).

Biota - Total mercury in tissues from animals were analyzed by Flett Research using direct combustion, amalgamation, and cold vapor atomic spectrophotometry in a Direct Mercury Analyzer (DMA) (US EPA method 7473) and by Battelle by atomic fluorescence (US EPA method 1631). All analytical runs were accompanied by tissues with known amounts of mercury added (sample spikes) to determine instrument recovery. Methyl mercury in tissues was analyzed by KOH digestion, ethylation, purge and trap, and CVAFS by US EPA method 1630. Method blanks were analyzed with all runs, as were matrix sample spikes to determine recovery values. The MDL for methyl mercury was 0.2 ng/g.

Preliminary sampling of mercury for stable isotope signatures

On October 4 and 5, 2006, samples of sediments were taken at six sites for analysis for stable isotope ratios of Hg. The locations chosen for these samples were intended to provide a contrast between sediments likely contaminated with Hg at the HoltraChem site and sediments outside the direct (water) influence of the HoltraChem site, to determine whether differences could be detected. Three of the samples were soils/sediment taken from the Orrington site. Orrington Sample 1 was from the Northern Ditch (003), in a cattail stand just upstream of the V-notch sampling weir. Orrington Sample 2 was sediment on a paved area near the cell building. Orrington Sample 3 was from the Southerly Stream, downstream of the V-notch sampling weir. The East Branch sample was taken from the eastern shore of the Penobscot River, East Branch. The St. George River sample was taken near the outlet of Sennebec Lake. The Eddington sample was taken near the boat launch at Eddington on the Penobscot

River, upstream of the Veazie dam. All samples were kept cool and shipped to Trent University in coolers with blue ice for analysis. Appendix 19 gives the geographic coordinates of the sampling locations. Analysis was by a multi-collector Inductively Coupled Plasma Mass Spectrometer (ICP-MS).

Data from samples taken in 2007 for Hg stable isotope analysis (wetlands and offshore sediments in the estuary) will be presented in an updated report.

Quality assurance/quality control methods

The laboratory air at the field laboratory (Winterport ME) was tested on two occasions for levels of dissolved gaseous mercury, using passive gold traps. These gold traps were analyzed by Studio Geochimica.

To test for possible contamination of water samples caused by Hg in the sampling apparatus or by handling of sample bottles, field blanks for water were performed at each sampling period. Deionized (DI) or Milli-Q water was sent to the field in large Teflon bottles. This water was then poured into sample bottles, using clean techniques for unfiltered field blanks or was filtered through the Teflon tubing and in-line filters for filtered field blanks. In addition, in 2007, “trip blanks” consisting of sealed Milli-Q water were sent to the field and returned unopened to test for possible contamination due to shipment. Because all water samples were duplicated in the field during sampling periods II to VI, the variation between these duplicates was analyzed to provide a measure of the total variation from sampling, handling and analytical methods. Water taken at the same time and place was also sent to three analytical laboratories (Battelle, Flett and Trent) to provide interlaboratory comparisons.

In order to examine core-to-core variation in cores taken at one site, field replicates of sediment samples were taken, typically 10 or more duplicate samples for total mercury and methyl mercury analysis during each sampling period. Analytical variation was examined by doing analytical duplicates of single sediment samples. In addition, samples of sediments were taken, split and sent to three laboratories (Battelle, Flett and Trent) for interlab comparisons.

Samples of biological tissues were also the subject of interlaboratory comparisons for both total Hg and MeHg. Three laboratories were involved (Battelle, Flett, and Trent). Samples of various tissues (except fish) from the Penobscot system were freeze-dried at Flett Research, split, and shipped to other labs for analysis. For fish, frozen samples

of macerated, mixed fish muscle from a mixture of species was used. This sample was a proficiency sample from Fisheries and Oceans Canada

Quality assurance/quality control results

Laboratory air - The air in the Winterport field laboratory, where all samples were taken for processing and shipment to analytical laboratories, was tested on two occasions for gaseous mercury concentrations. The first test was made in July 2006 when construction of the inside of the lab was taking place. Construction activities and the installation of new materials such as carpets are known to raise the concentration of mercury in building air. The concentration was found to be 65.9 ng/m^3. This concentration is considered to be above that desirable for a laboratory handling low level mercury samples. The lab air was therefore re-tested, in September 2006, after construction was completed and the lab was in use. Gaseous mercury in the lab air had decreased to 17.6 ng/m^3, a concentration considered to be acceptable for laboratories handling low-level mercury samples (Gill and Fitzgerald 1987).

Water - Table 1 shows the results for the analysis of samples of blank water handled in the field in 2006. In Sampling period I, blanks were analyzed only for MeHg in unfiltered samples; all samples were below the detection limit of 0.006 ng/gL. Field blank values were above the detection limits for both THg (means ranged from 0.143 to 0.479 ng/L) and MeHg (means ranged from 0.017 to 0.040 ng/L). For Sampling periods II and III, THg in unfiltered blanks averaged 0.479 and 0.439 ng/L, respectively. It is considered desirable that field blanks are below 0.3 ng/L total mercury and the unfiltered field blanks taken in Sampling periods II and III had values higher than this. Unfortunately, Studio Geochimica, who provided the blank water to field crews, did not analyze the blank water used in Sampling periods I, II and III for total mercury or methyl mercury, so it is not known whether these values indicate contamination of samples by field procedures, or whether the blank water was above the detection limit in total mercury and methyl mercury. Most unfiltered field blanks in these two sampling periods were 0.4 to 0.5 ng/L, showing relative consistency.

Differences among field replicates of regular water samples were also examined to check for the possibility of contamination. During the first three sampling periods in 2006, variation among field replicates was quite low (see below). However, filtered field blanks showed less consistency, ranging from 0.2 to 0.8 ng/L

For Sampling period IV, blank water was supplied and analyzed by Battelle Marine Science Laboratories. It was found that both total mercury and methyl mercury in the

blank water supplied for this sampling period were below the level of detection (0.121 ng/L and 0.0192 ng/L, respectively). After handling in the field, unfiltered blanks ranged from below detection to 0.16 ng/L total mercury and from below detection to 0.017 ng/L methyl mercury, indicating that very little mercury was added by the sampling procedure.

In most cases, concentrations in filtered blanks were not significantly higher than concentrations in unfiltered blanks, indicating that the filtration apparatus did not result in contamination of the water being filtered. There were three occasions, however, when filtered THg values were more than 50% higher than unfiltered values, indicating a possible problem in rinsing the filtration line after the previous sample for those samples.

Table 2 shows the results of the analysis of blank water samples collected during 2007 (Sampling V and Sampling VI). For both sampling rounds, filtered and unfiltered blanks were satisfactorily and consistently low (near the detection limit), indicating that the sampling techniques being used in the field were adding only very small amounts of mercury and methyl mercury to the water samples. Filtered blanks (water that had been pumped through the filtered apparatus including the filter cartridge) were very similar to unfiltered blanks (water not pumped through the filter apparatus), showing that the filtering apparatus and the filter cartridges being used were not adding significant amount of mercury or methyl mercury to water samples that were filtered. Trip blanks were very similar to filtered or unfiltered blanks. Overall, these results indicate a lack of significant contamination of samples by field sampling and handling procedures.

Table 1. Results for field blanks carried out during each sampling period in 2006. Analyses were done at Battelle Marine Sciences Laboratory. n.d. = non detectable. s.d. = standard deviation. n/a=not applicable.

Sampling Period		THg unfiltered ng/L	THg filtered ng/L	MeHg unfiltered ng/L	MeHg filtered ng/L
I	range	n/a	n/a	n.d. to n.d.	n/a
I	**mean**	**n/a**	**n/a**	**0.0015**	**n/a**
I	s.d.	n/a	n/a	0.000	n/a
I	n	n/a	n/a	6	n/a
II	range	0.39-.55	0.40-0.83	n.d. to 0.19	n.d. to 0.14
II	**mean**	**0.479**	**0.528**	**0.040**	**0.033**
II	s.d.	0.054	0.144	0.073	0.050
	N	7	7	6	6
III	range	0.21-0.56	0.22-0.72	n.d. to 0.04	n.d. to 0.032
III	**mean**	**0.439**	**0.452**	**0.022**	**0.024**
III	s.d.	0.123	0.150	0.014	0.008
	N	7	7	7	7
IV	range	n.d. to 0.16	n.d. to 0.44	n.d. to 0.024	n.d. to 0.023
IV	**mean**	**0.143**	**0.235**	**0.017**	**0.015**
IV	s.d.	0.018	0.105	0.006	0.006
	N	6	8	6	8

Table 2. Concentrations of methyl mercury and total mercury in field blanks, filter blanks and trip blanks analyzed for Sampling V and Sampling VI, 2007. All analyses by Battelle Marine Sciences Laboratory.

Description	Number of samples	Mean Methyl Mercury (ng/L)	Range – Methyl mercury (ng/L)	Mean Total mercury (ng/L)	Range – Total mercury (ng/L)
Blank water	3	Under detection limit	n/a	Under detection limit	n/a
Unfiltered field blank (Sampling V)	5	0.019	Below detection-0.0196	0.213	Below detection-0.258
Filtered field blank (Sampling V)	5	0.018	Below detection-0.0304	0.192	Below detection-0.283
Unfiltered field blank (Sampling VI)	7	0.024	Below detection-0.0451	0.241	0.199-0.300
Filtered field blank (Sampling VI)	7	0.026	Below detection-0.0409	0.224	Below detection-0.285
Trip blank (Sampling VI)	3	0.0282	0.0259-0.0318	0.2219	0.199-0.254
Detection limit	n/a	0.0188		0.188	

The reproducibility of pairs of analytical duplicates for total and methyl mercury in water was excellent for Battelle Marine Sciences Laboratory and Flett Research. These data are shown in Table 3.

Table 3. Relative percent differences between pairs of analytical duplicate water samples (duplicate subsamples taken from the same water sample). All samples were filtered. Data are composited from both Flett Research Ltd. and Battelle Marine Sciences Laboratory.

	Total Mercury	Methyl Mercury
Average RPD	4.13%	7.37%
Stand. Dev.	7.55%	9.55%
n	17	10

Field replicates are samples taken at the same time and place in the field, but in separate bottles, and analyzed separately in the laboratory. Variation among field replicates are expected to be higher than for laboratory duplicates because of possible variation in the water actually sampled. Table 4 provides the relative percent differences (RPD = difference between the two replicates divided by the average of the two replicates) between pairs of field replicates taken in 2006 and 2007 and analyzed for total Hg. RPD's for field replicates of total mercury in both filtered and unfiltered samples were generally low, ranging from 4.1 – 11.4%, as compared to the average variation among laboratory replicates of 4.1%, indicating excellent reproducibility of sampling and lack of contamination. RPD's for field replicates of filtered MeHg samples were 15.6 – 23.0 %, as compared to the average RPD for analytical duplicates of 7.4% (Table 5). The highest values for average RPD in field replicates were for unfiltered MeHg samples (12.2 – 33.4%). This was likely due to the low concentrations in these samples (most less than 0.15 ng/L), which means that even a few particles containing MeHg could provide a significant difference in two different field samples.

Table 4. Relative percent Difference (RPD) for replicate water samples analyzed for filtered total mercury and replicate samples analyzed for unfiltered total mercury.

	THg filtered ng/L			**THg unfiltered ng/L**		
	Average RPD	Std Dev	n	Average RPD	Std Dev	n
Period II	11.4%	11.1%	33	6.4%	7.5%	32
Period III	10.5%	13.0%	37	5.0%	5.5%	37
Period IV	9.6%	8.1%	35	10.3%	9.0%	34
Period V	6.7%	8.6%	35	5.9%	8.1%	35
Period VI	4.1%	3.0%	35	11.4%	14.9%	35

Table 5. Relative percent Difference (RPD) for replicate water samples analyzed for filtered methyl mercury and replicate samples analyzed for unfiltered methyl mercury.

	MeHg filtered ng/L			**MeHg unfiltered ng/L**		
	Average RPD	Std Dev	n	Average RPD	Std Dev	n
Period II	19.2%	30.5%	38	28.1%	23.8%	34
Period III	23.0%	22.5%	36	25.8%	24.8%	37
Period IV	21.8%	15.2%	28	33.4%	23.8%	29
Period V	15.6%	15.8%	35	12.2%	8.2%	35
Period VI	18.5%	21.5%	35	15.9%	15.0%	35

Interlaboratory comparisons of total Hg and MeHg in water showed good comparability (Figures 8 and 9). Better agreement was evident at higher than at lower concentrations, as would be expected.

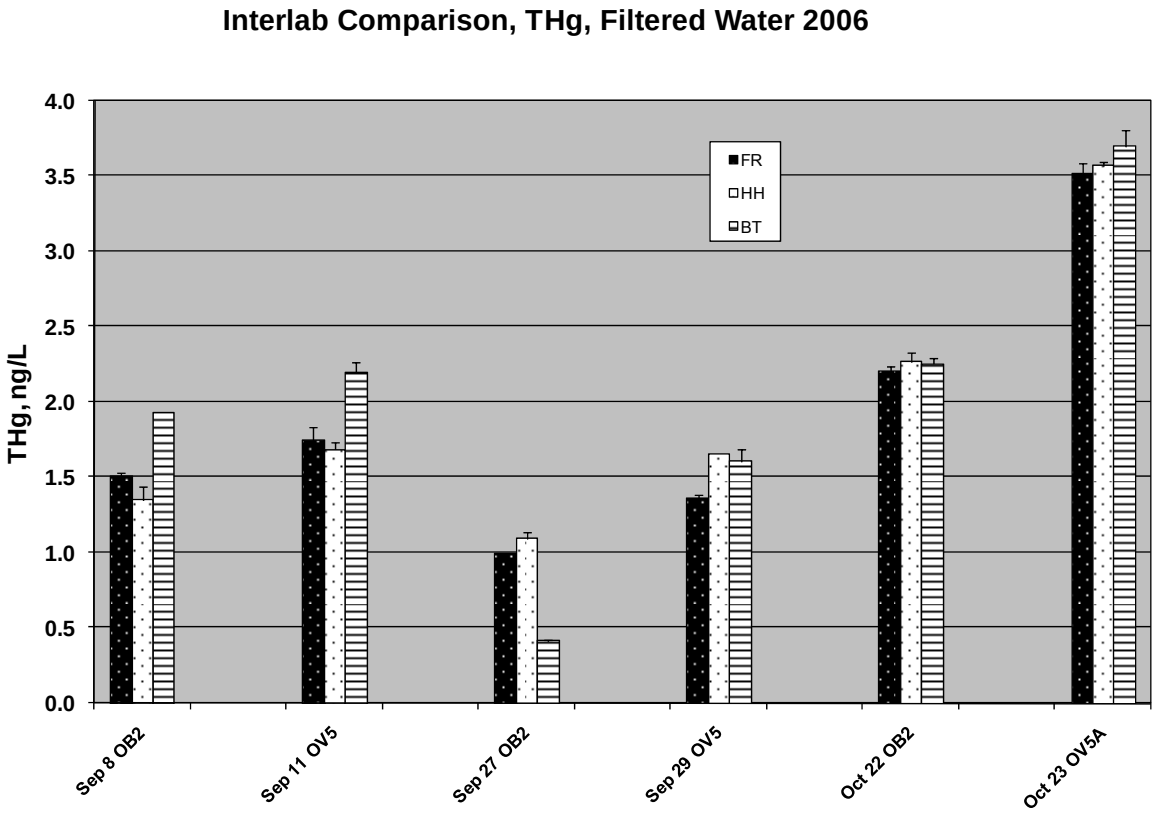

Figure 8. Interlaboratory comparisons of total mercury in water among three laboratories. FR=Flett Research; HH=Trent University; BT=Battelle Marine Sciences Laboratory. Error bars are one standard deviation.

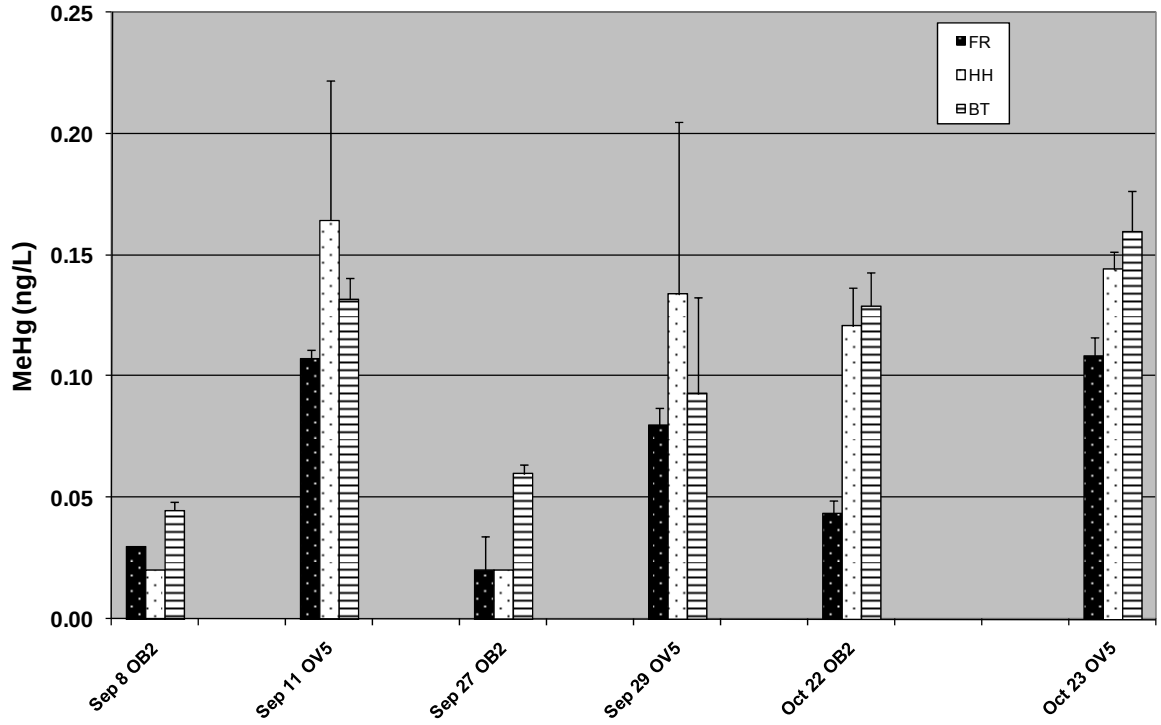

Figure 9. Interlaboratory comparisons of the determination of methyl mercury in water. FR=Flett Research; HH=Trent University; BT=Battelle Marine Sciences Laboratory. Error bars are one standard deviation.

Because all total suspended solid (TSS) determinations were made in duplicate, the variation between laboratory duplicates could be made (Table 6). In sampling periods IV, V and VI, the relative percent difference was quite low, averaging 10.6 to 16.7% in the different sampling periods. This is considered to be acceptable variability for this method. (Note that TSS was not done in sampling periods I, II, and III, as discussed in the later section on acceptable hold times for TSS samples.)

Table 6. Relative Percent Difference (RPD) for laboratory duplicate determination of water samples analyzed for total suspended solids. Data from Battelle Marine Sciences Laboratory.

	Total Suspended Solids		
	Average RPD	Std Dev	n
Period IV	16.7%	14.1%	34
Period V	10.6%	8.0%	35
Period VI	12.9%	11.6%	35

Sediments - The variation among pairs of duplicate sub-samples of sediment core sections analyzed for total mercury and methyl mercury was examined at Flett Research and Battelle Marine Sciences Laboratory. Variation was acceptably low for both total mercury analyses and methyl mercury analyses at both laboratories (Tables 7 and 8).

Table 7. Relative percent differences between pairs of duplicate sub-samples of sediment core sections. Data are from Battelle Marine Sciences Laboratory.

	Total Mercury	Methyl Mercury
Average RPD	6.52%	8.66%
Stand. Dev.	5.63%	7.43%
n	13	9

Table 8. Relative percent differences between pairs of duplicate sub-samples of sediment core sections. Data are from Flett Research Ltd.

	Total Mercury	Methyl Mercury
Average RPD	9.83%	9.26%
Stand. Dev.	5.51%	7.73%
n	4	10

Interlaboratory comparisons were carried out for total mercury and methyl mercury in sediments. Total mercury results are shown in Figure 10, which demonstrates good comparability among labs. Discrepancies between two methods of determining methyl mercury concentrations in sediments (distillation vs. extraction) have been uncovered as part of the interlaboratory comparisons performed as part of the Penobscot study. These differences have not been noted previously in the scientific literature and they appear to be peculiar to the sediments of the Penobscot River. They are under investigation at the time of writing this report.

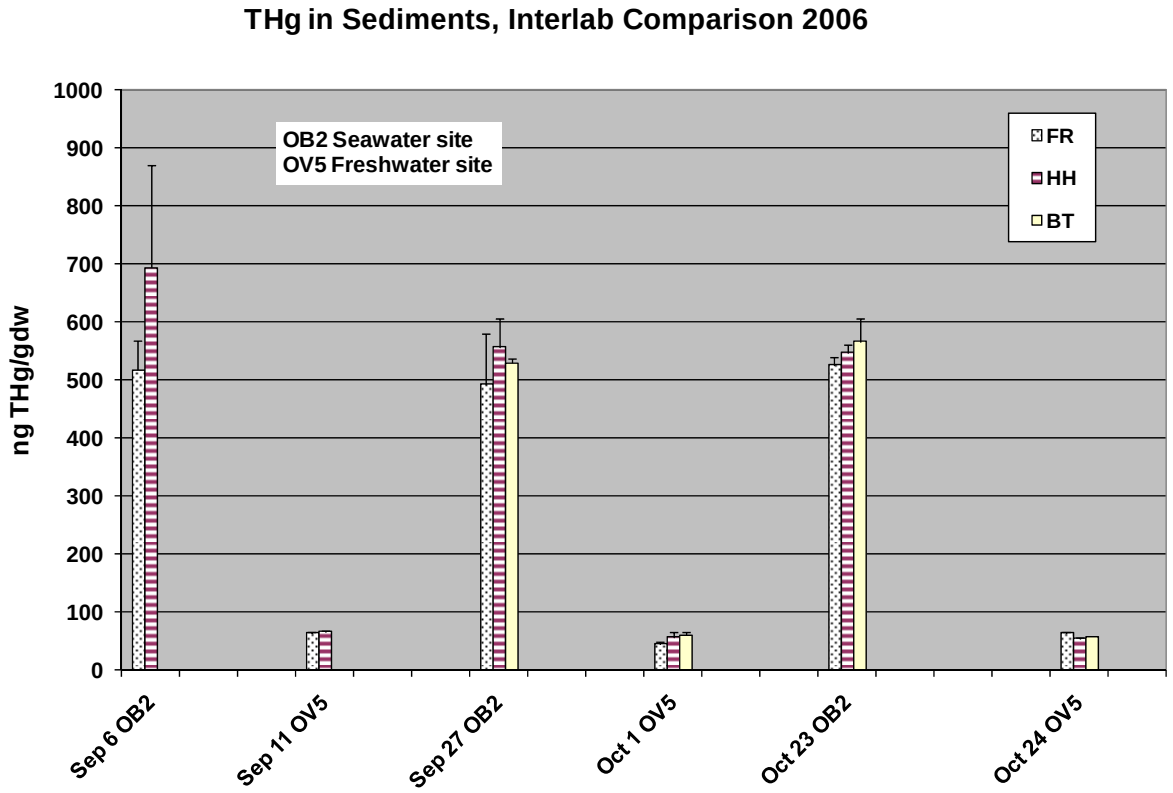

Figure 10. Results of interlaboratory comparisons of total mercury in sediments from two sites, one estuarine and one freshwater, from the Penobscot system. FR=Flett Research; HH=Trent University; BT=Battelle Marine Sciences Laboratory.

Biota - Results of the interlaboratory comparisons for biological tissues showed excellent agreement among the three different laboratories. For total mercury in tissues, nine tissues were analyzed in two different labs (Figure 11). Mean RPD

(relative percent difference) was 2.5%, with a range of 0.6-7.2. For methyl mercury in tissues, nine tissues were analyzed (Figure 12). Mean RPD was 5.3% with a range of 1.1 – 7.25%.

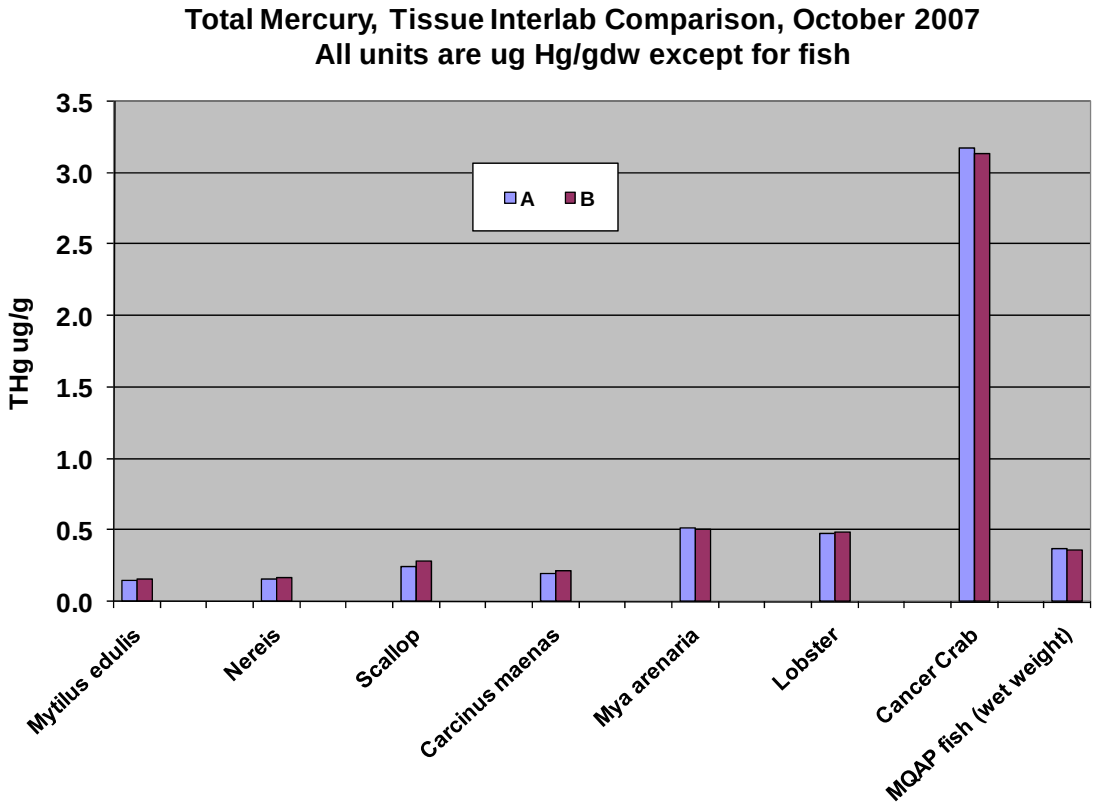

Figure 11. Results of interlab comparisons for the analysis of total mercury in biological tissues between Flett Research (Lab A) and Battelle Marine Sciences Laboratory (Lab B). Each bar represents one determination. All tissues from the Penobscot estuary except for fish which was a proficiency sample from Fisheries and Oceans Canada.

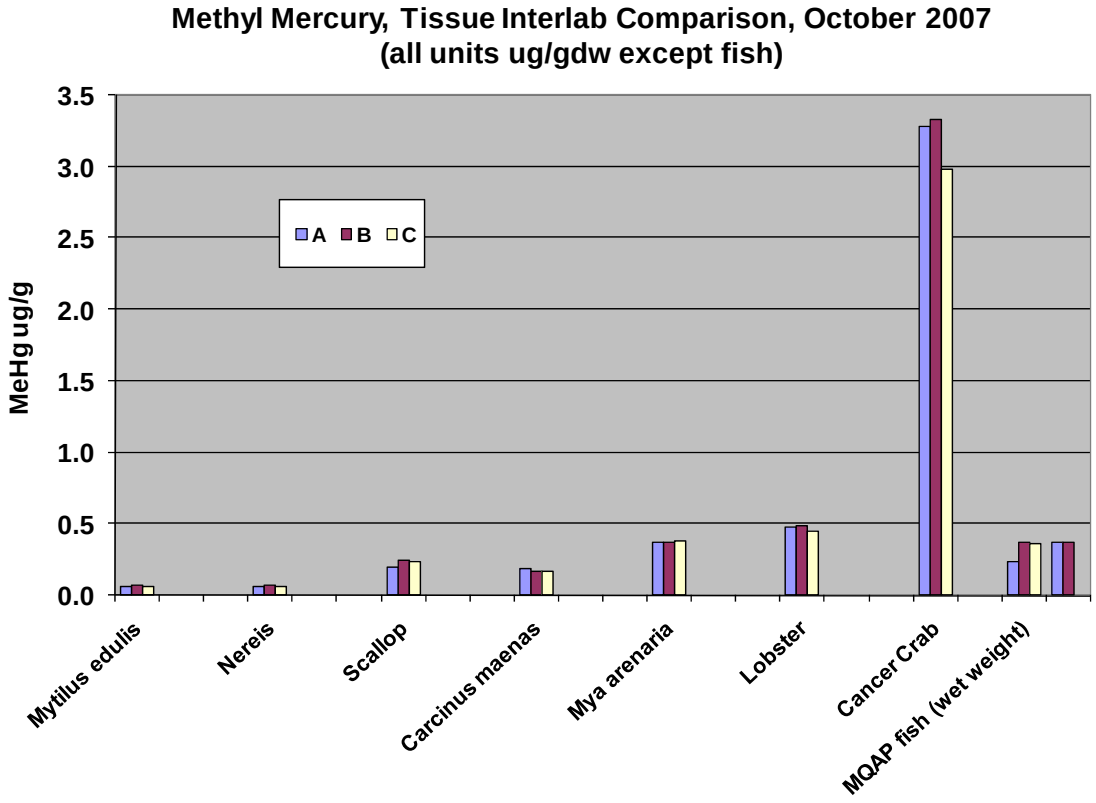

Figure 12. Results of interlab comparisons for the analysis of methyl mercury in biological tissues among Flett Research (Lab A), Battelle Marine Sciences Laboratory (Lab B), and Trent University (Lab C). Each bar represents one determination. All tissues from the Penobscot estuary except for fish which was a proficiency sample from Fisheries and Oceans Canada.

Hold times - Because of concerns related to the change in analytical laboratories from Studio Geochimica to Battelle Marine Sciences Laboratory for the analysis of water and sediment samples, a detailed analysis of the duration that water samples were held in storage before they were analysed (hold times) was conducted. The United States Environmental Protection Agency (USEPA) prescribes maximum hold times for water samples to be analyzed for total mercury and methyl mercury. EPA method 1631 for total mercury in water prescribes maximum hold times of 90 days, if samples are preserved within 48 hours of sampling, and EPA method 1630 (draft 2001) for methyl mercury in water notes that samples are stable for at least 180 days, if samples are preserved within 48 hours of sampling. Hold times for the first four aquatic sampling events are shown in Table 9. All samples were preserved within 48 hours of collection. All samples for THg were analyzed within the USEPA recommended time of 90 days,

with the exception of four samples that were held for up to 93 days before being analyzed. The concentrations seen in those four samples were similar to others at the same or adjacent sites, so were accepted as being accurate. All samples for MeHg were analyzed within the USEPA recommended time of 180 days.

Table 9. Mean (days) and range (days) that water samples were held before analysis for the early August (Sampling I), early September (Sampling II), late September/early October (Sampling III), and late October (Sampling IV), 2006.

	Early August	Early Sept.	Sept./ October	Late October
Total mercury	15.5 d (11-19)	87.6 d (80-90)	72.3 d (46-93)*	63.7 (22-78)
Methyl mercury	95.6 d (*25*-177)	115.1 d (94-136)	68.4 d (48-106)	33.6 d (6-77)

* Number of samples with hold times >90 days = 4

For Sampling events V and VI (late May/early June and July, 2006), all samples were analyzed for THg and MeHg within the USEPA recommended hold times. During Sampling V, the maximum hold time for THg samples was 21 days and for MeHg samples was 49 days. During Sampling VI, the maximum hold time for THg samples was 36 days and for MeHg samples was 33 days. All samples were preserved within 48 hours of collection.

The maximum hold time for water samples to be analyzed for total suspended solids (TSS) is 7 days (Greenberg et al. 1992). Due to unacceptably long hold times, TSS data from sampling events I, II, and III were not used. The maximum hold time for samples taken during Sampling event IV was 6 days, the maximum hold time for samples taken during Sampling V was 6 days, and the maximum hold time for samples taken during Sampling VI was also 6 days.

All sediment samples were analyzed for total mercury and methyl mercury within the USEPA suggested hold time of 6 months.

The maximum hold time recommended by the USEPA for biological tissues to be held frozen before analysis for mercury is 1 year. All tissue samples for the Penobscot study have been analyzed within this hold time.

RESULTS AND DISCUSSION

Mercury in water and suspended particles

Total Hg dissolved in water is shown for the five sampling reaches, averaged over all six sampling periods, in Figure 13. Average concentrations ranged from about 0.75 ng/L to about 2.6 ng/L. These concentrations are typical of unimpacted sites receiving no point sources of Hg and with low rates of atmospheric deposition. For example, St. Louis et al. (1994) found concentrations of 1.4 – 13.4 ng/L total Hg in streams in a remote area of northwestern Ontario where there is no industry and atmospheric deposition rates of mercury are at background levels. Dissolved total Hg was higher in the river and lower in the estuary, especially at outer estuary sites. In the river, concentrations of dissolved total Hg were slightly higher on average in the Old Town to Veazie reach of the river (Figure 12). This pattern was quite consistent over the six sampling times. Dissolved total Hg was lower in the estuary than in the river during all times of the year and was highest in the East Branch, Old Town to Veazie or Brewer to Orrington, with decreasing concentrations further downstream. Dissolved total Hg in the river showed no pattern in relation to the location of the HoltraChem site.

MeHg dissolved in water averaged about 0.02 to 0.31 ng/L and was also higher in the river, as compared to the estuary (Figure 14). Concentrations were generally higher in the Old Town to Veazie reach than other river reaches, as for dissolved total Hg. Also as for total Hg, concentrations seen are typical of unimpacted sites (e.g. St. Louis et al. 1994). This pattern was very consistent at different times of the year. Thus, dissolved MeHg showed no relationship to the HoltraChem site.

The observation of lower concentrations of dissolved Total Hg and MeHg in the lower river and upper estuary as compared to further upstream is consistent with other studies, which demonstrate that mercury tends to absorb more to particles as the water becomes more brackish and salinity increases (Turner et al. 2001; 2002). The lower concentrations at the outer estuarine sites may also have been caused by dilution of river water by sea water.

The lower reaches of the Penobscot River carried much higher loads of suspended particles than the upper river reaches or the estuary (Figure 15). Concentrations of suspended particles are usually related to water turbulence. In the East Branch, total suspended solids (TSS) generally averaged about 2 mg/L, increasing to about 5 mg/L in the Old Town-Veazie and Brewer-Orrington reaches, and to 10 mg/L or higher in the river downstream of Orrington. In the estuary, average TSS values ranged from 2 to 25 mg/L. River flows apparently cause the suspension of significant amounts of fine particles in the river, especially downstream of Orrington. TSS was highest in the Orrington to Bucksport reach in late October and July but was highest in the estuary in late May/early June.

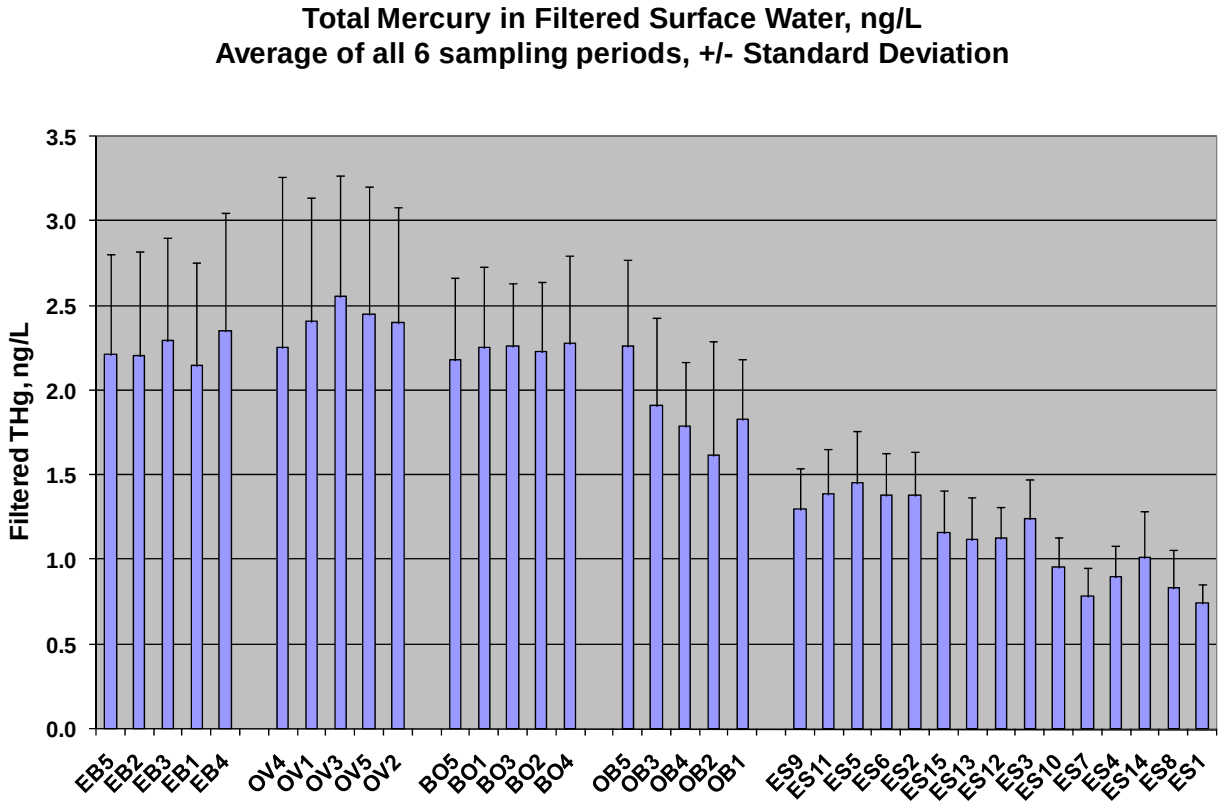

Figure 13. Mean concentrations (+/- 1 standard deviation) of total mercury (ng/L) in filtered water in the Penobscot River and estuary during the six sampling periods in 2006 and 2007. With the exception of some stations during Sampling period I, each site was sampled in duplicate during each sampling period, so most means are from 12 determinations at each site. EB=East Branch, OV=Old Town-Veazie, BO=Brewer-Orrington, OB=Orrington-Bucksport, ES=Estuary. Stations are plotted in geographic order from upstream to downstream (north to south); sites are mapped in Figures 1-6.

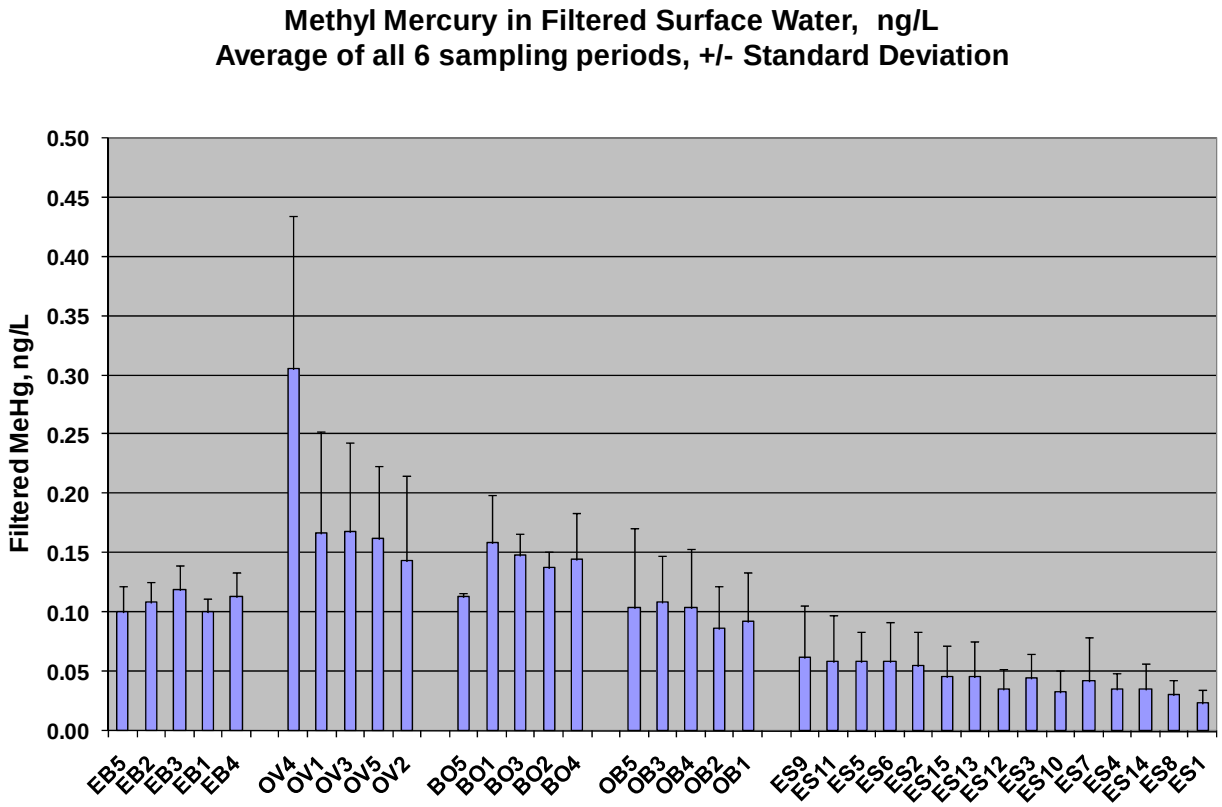

Figure 14. Mean concentrations (+/- 1 standard deviation) of methyl mercury (ng/L) in filtered water in the Penobscot River and estuary during the six sampling periods in 2006 and 2007. With the exception of some stations during Sampling period I, each site was sampled in duplicate during each sampling period, so most means are from 12 determinations at each site. EB=East Branch, OV=Old Town-Veazie, BO=Brewer-Orrington, OB=Orrington-Bucksport, ES=Estuary. Stations are plotted in geographic order from upstream to downstream (north to south); sites are mapped in Figures 1-6.

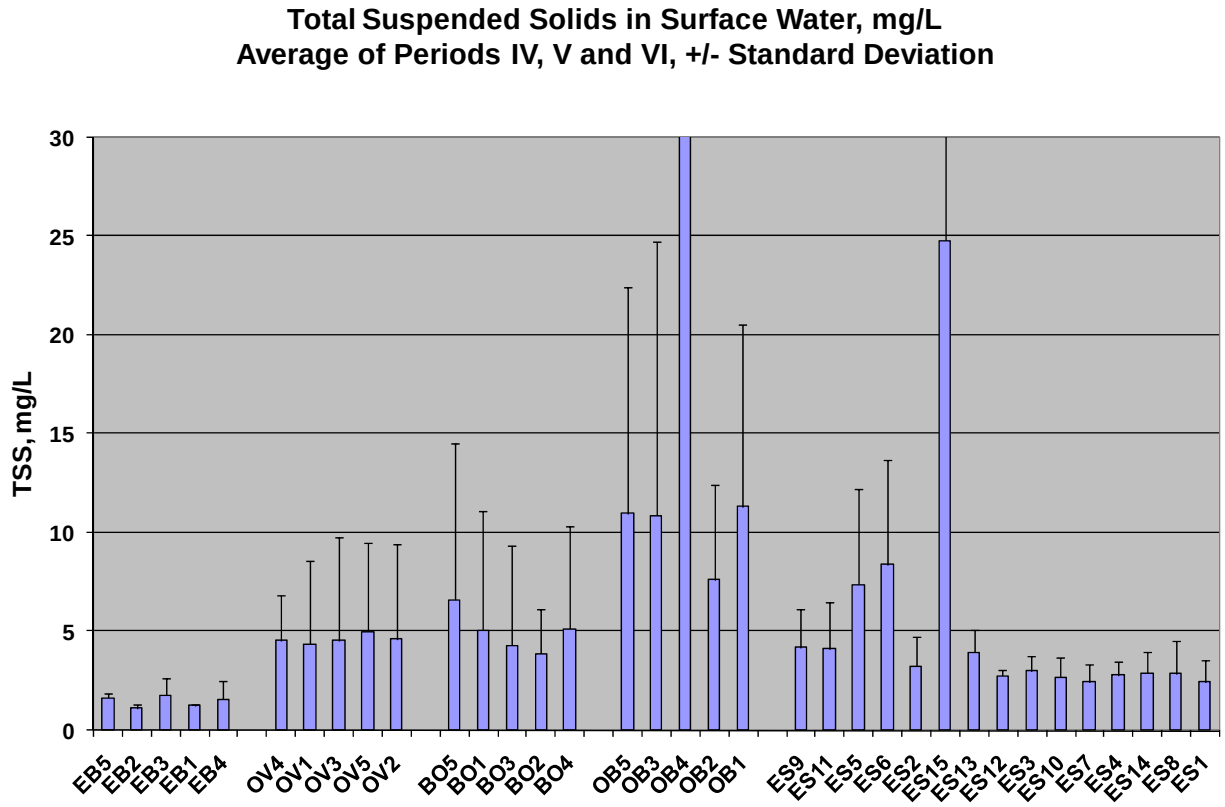

Figure 15. Mean concentrations (+/- 1 standard deviation) of suspended sediments (mg/L) in surface water in the Penobscot River and estuary during Sampling periods IV, V, and VI, 2006 and 2007. All water samples were analyzed in duplicate in the laboratory and therefore each mean is from duplicate analyses taken at three different sampling times. EB=East Branch, OV=Old Town-Veazie, BO=Brewer-Orrington, OB=Orrington-Bucksport, ES=Estuary. Actual mean for OB4 was 107.0 and s.d. was 180.6. Standard deviation for ES15 was 26.1. Stations are plotted in geographic order from upstream to downstream (north to south); sites are mapped in Figures 1-6.

The concentration of total Hg on particles suspended in river and estuary water was relatively constant in the upper river (about 0.3 µg THg/g), but increased noticeably downstream of Orrington to about 0.7 µg/g, and decreased with distance out into the estuary (Figure 16). This pattern was quite consistent among the three sampling periods (Late October, late May/early June and July). Concentrations were much less than were observed in the mercury contaminated Elbe River (Germany) (Wilken and Hintelmann 1991).

Because the total amount of suspended particles also increased in the lower river, the load of Hg carried by particles increased more than based only on concentration. TSS was typically 2 times as high in the reach downstream of HoltraChem (OB) as compared to upstream of HoltraChem (BO) and total Hg concentrations on particles were also typically twice as high. Therefore, the total load of total Hg on suspended averaged about 4 times as high downstream of HoltraChem.

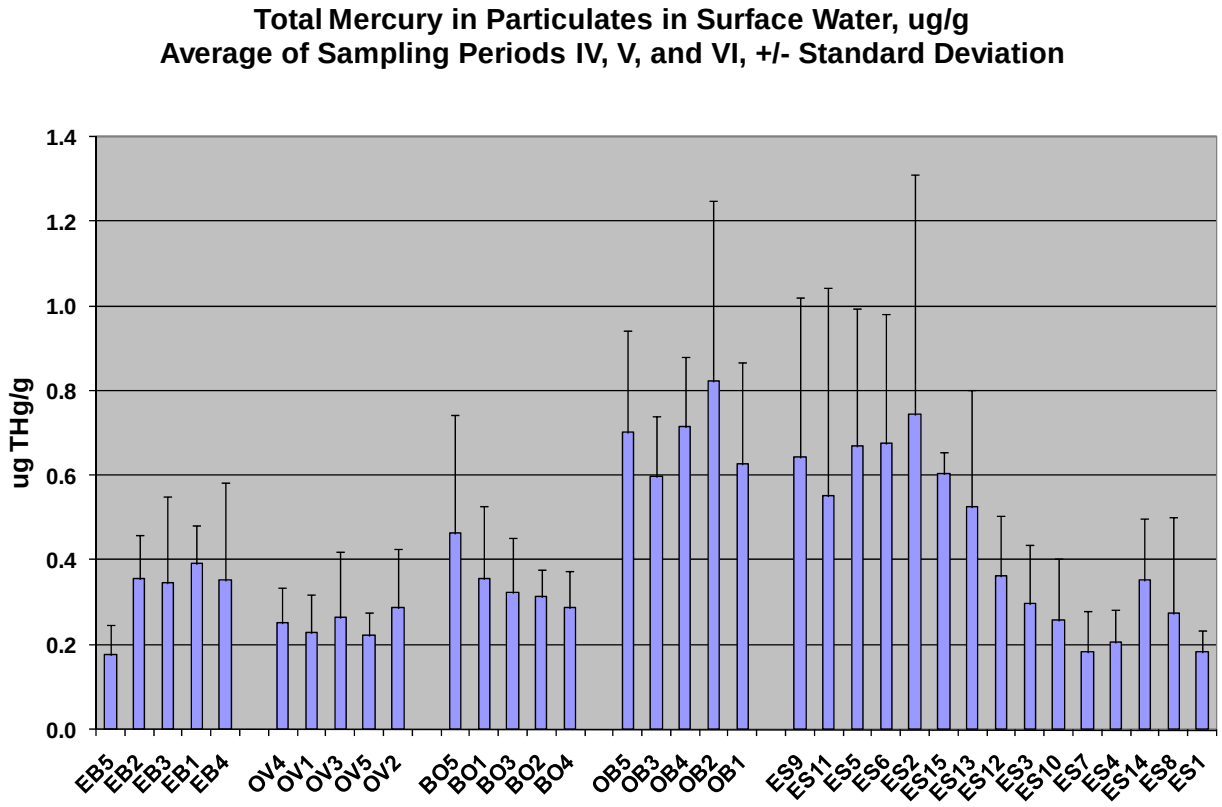

Figure 16. Mean concentrations (+/- 1 standard deviation) of total mercury (µg/g) on suspended particles in the Penobscot River and estuary during sampling periods IV, V, and VI, 2006 and 2007. Means for each of the three sampling periods were calculated from field duplicate samples for total mercury in unfiltered filtered water and from laboratory duplicate determinations of suspended solids. EB=East Branch, OV=Old Town-Veazie, BO=Brewer-Orrington, OB=Orrington-Bucksport, ES=Estuary. Stations are plotted in geographic order from upstream to downstream (north to south); sites are mapped in Figures 1-6.

MeHg on particles in the Penobscot River and estuary averaged from 0.005 to 0.04 µg/g and did not show noticeable or consistent differences over the study area (Figure 16). MeHg on particles did not show patterns related to the location of the HoltraChem site, although on average, it tended to be lower in the East Branch and the lower estuary (Figure 17).

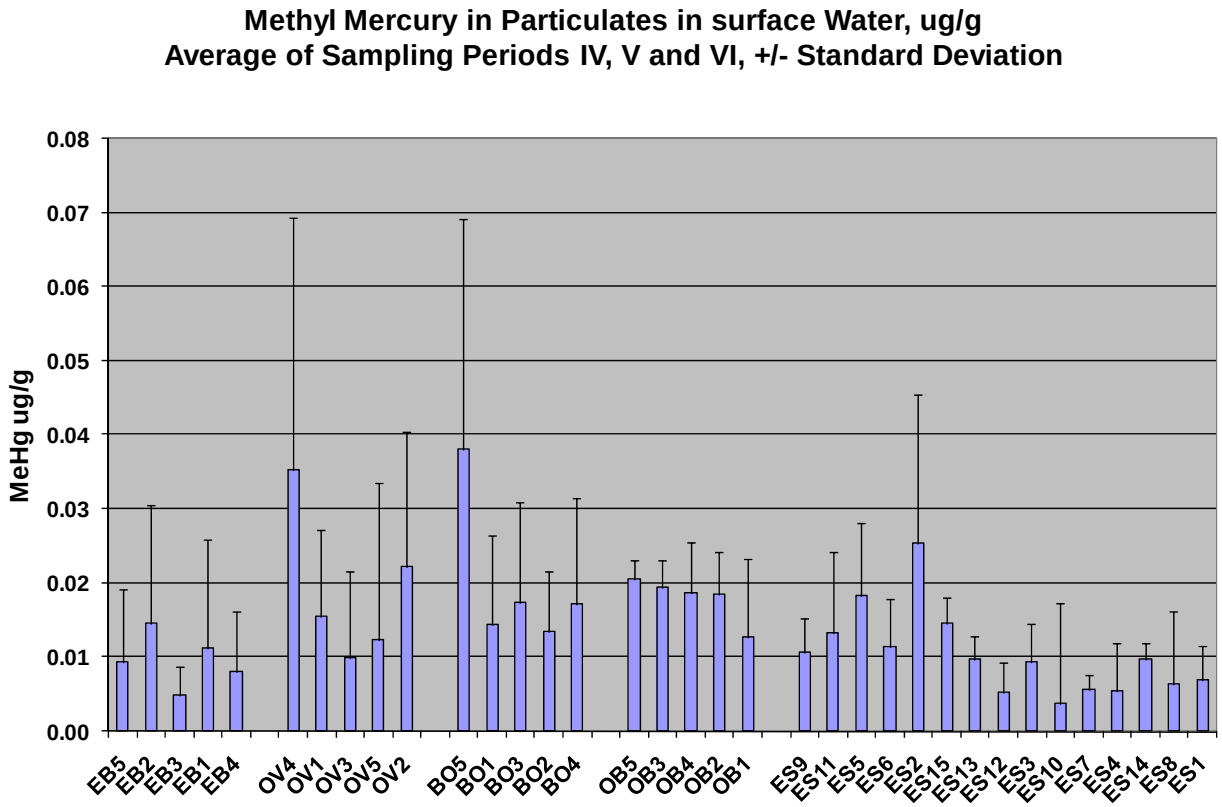

Figure 17. Mean concentrations (+/- 1 standard deviation) of methyl mercury (µg/g) on suspended particles in the Penobscot River and estuary during sampling periods IV, V, and VI, 2006 and 2007. Means for each of the three sampling periods were calculated from field duplicate samples for total mercury in unfiltered filtered water and from laboratory duplicate determinations of suspended solids. EB=East Branch, OV=Old Town-Veazie, BO=Brewer-Orrington, OB=Orrington-Bucksport, ES=Estuary. Stations are plotted in geographic order from upstream to downstream (north to south); sites are mapped in Figures 1-6.

We have assessed the degree of mercury enrichment on the suspended particles throughout the Penobscot system, relative to their surrounding water. This was done for total Hg and for MeHg and these data are summarized in Figures 18 and 19,

respectively. They show that particles south of Orrington are enriched 2-5 x 10^5 times in total Hg relative to ambient water, regardless of whether water was fresh or more saline (Figure 18). These partition coefficients, or Kd values, are not atypical of THg for particulate matter in other waters. Note that the Kd values were consistently lower, about 1-2 x 10^5, for waters above Orrington. This suggests that most of the particulate matter heavily enriched in HoltraChem total Hg is being transported downstream. The Kd values for MeHg are also high (Fig. 19), but less than those of total Hg, again consistent with some earlier reports. As with total Hg, the Kd values on particulate matter north of the Hotrachem site are lower than those south of the site. The higher Kd's in the OB reach and in the estuary are consistent with other studies (Turner et al. 2001; 2002) that have found that higher chloride concentrations in brackish waters result in mercury partitioning from the dissolved fraction to particulate material.

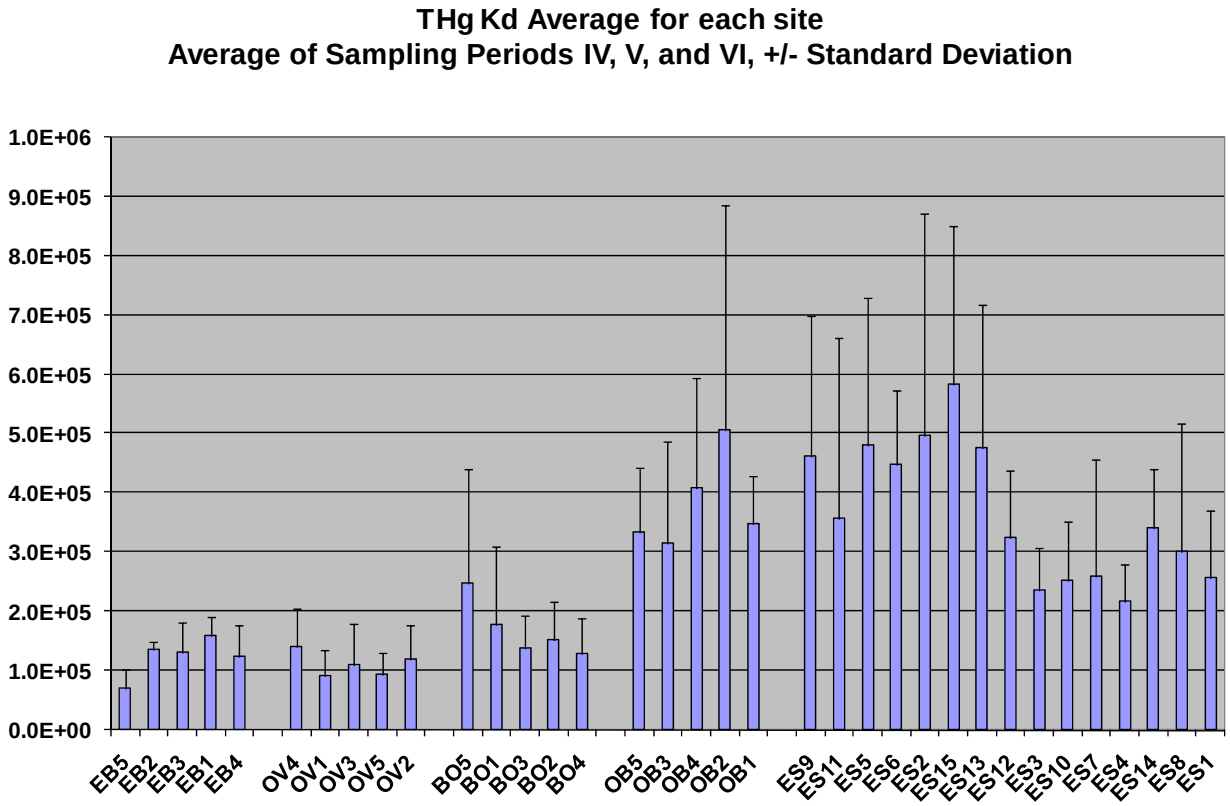

Fig. 18. Mean partition coefficients (Kd's) of THg on suspended particles at discrete stations from 5 different reaches within the Penobscot system. Kd values indicate degrees of THg enrichment in the particles relative to surrounding water at the same site. Note that most values are $\geq$ 1 x 10^5. Stations are plotted in geographic order from upstream to downstream (north to south); sites are mapped in Figures 1-6.

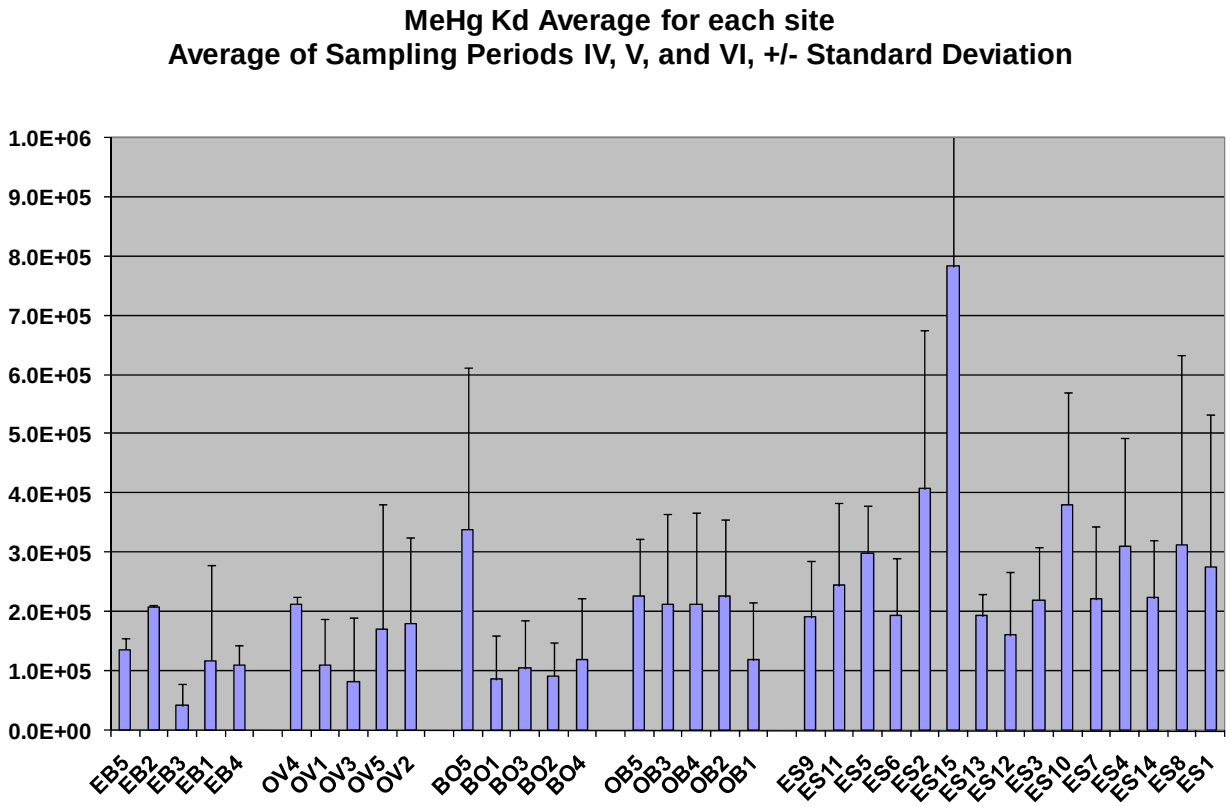

Fig. 19. Mean partition coefficients (Kd's) of MeHg on suspended particles at discrete stations from 5 different reaches within the Penobscot system. Kd values indicate degrees of MeHg enrichment in the particles relative to surrounding water at the same site. Note that most values are $\geq 5 \times 10^4$. Stations are plotted in geographic order from upstream to downstream (north to south); sites are mapped in Figures 1-6.

Mercury in sediments

Data on mercury in near-shore fine-grained sediments indicate a large source of mercury in the lower Penobscot River, downstream of the Veazie dam (within tidal influence). Total Hg in nearshore surficial sediments (0-3 cm in depth) of the reference area in the East Branch were quite low (Figure 20). Mean dry weight concentrations, in the East Branch during the first four sampling periods ranged from 29 – 44 ng/g d.w. THg. These are typical concentrations for uncontaminated areas in the region (e.g. Sowles 1999). Concentrations increased noticeably (2.3 to 3.6 times) in the Old Town to Veazie reach, averaging 67-106 ng/g dw, indicating local sources of Hg from human activities, but at a relatively low level. The most likely sources of this Hg are from paper

mills at Millinocket, Lincoln and Old Town, which have probably used Hg in slimicides in their operations in the past.

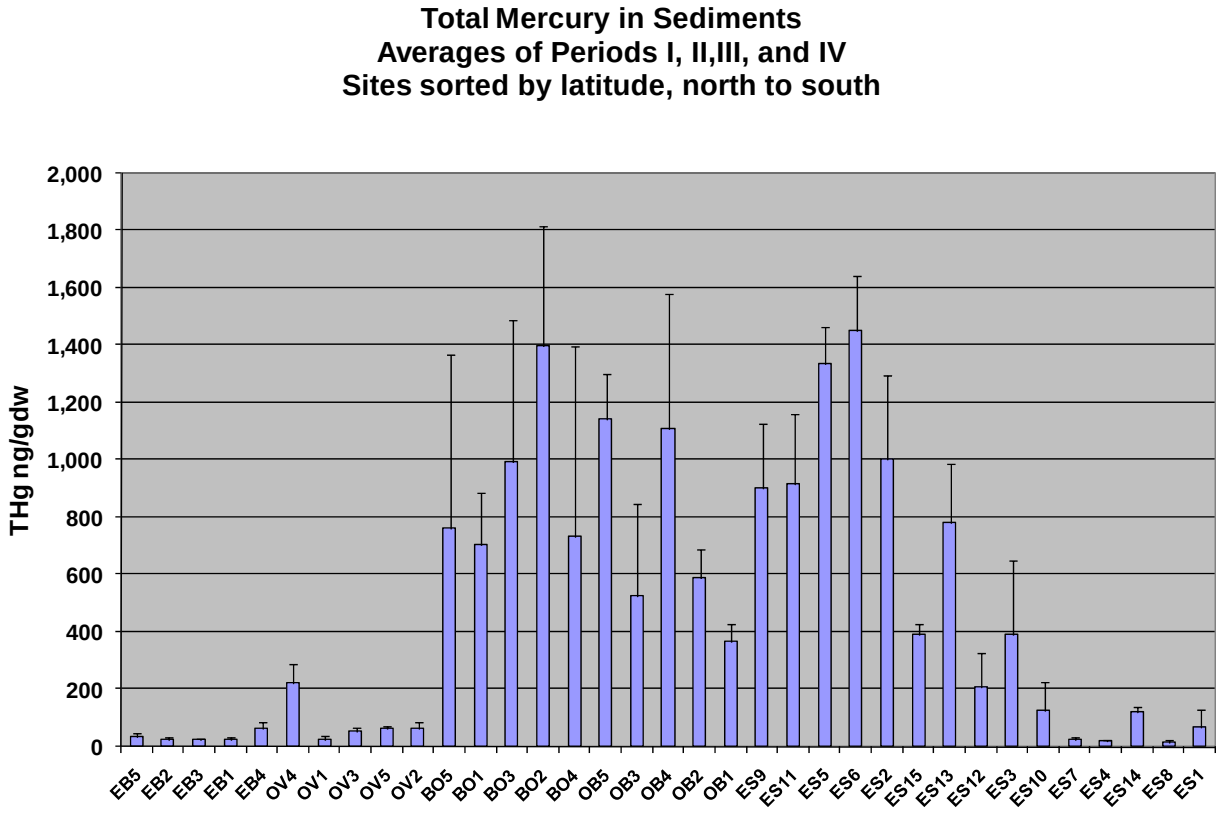

Figure 20. Total mercury (ng/g dry weight) in near-shore surficial sediments (0-3 cm) of the Penobscot River and estuary. Each bar is the mean from Sampling periods I, II, III, and IV (+/- s.d.). Stations are plotted in geographic order from upstream to downstream (north to south); sites are mapped in Figures 1-6.

Hg in sediments was much higher in the lower Penobscot River, downstream of the Veazie dam and Brewer (within tidal influence) than above the dam. Downstream of the Veazie dam, in the reaches of the river influenced by the tide (Veazie to Old Town and Brewer to Orrington sampling reaches), total Hg concentrations were often ten times higher than in the reaches of the river upstream of the Veazie Dam. In the Brewer to Orrington reach, average concentrations were 741 to 1016 and in the Orrington to Bucksport reach were 600 to 822 ng/g dw. In the upper estuary in the vicinity of Verona Island, the Orland River and Fort Point Cove (sites ES, 11, 5, 6, 2, 15, 13, 12, Figure 20) concentrations were as high as in the river lower river. Further south in the estuary

(as far mid Islesboro Island), concentrations declined by more than an order of magnitude (e.g. sites ES8, ES1)

Because the sediments in the Old Town – Veazie sampling reach, which is downstream of three paper mills, were only slightly elevated compared to the East Branch, it would appear that paper mills are not large sources of Hg to the Penobscot River. Rather, the data are consistent with a large source of Hg downstream of the Veazie dam. This source has apparently contaminated the downstream reaches of the river within tidal influence and the upper estuary.

Total Hg concentrations in sediments in the lower Penobscot River and upper estuary are similar to other areas known to be contaminated from chlor-alkali facilities and other Hg sources. The total range seen in the Penobscot downstream of Veazie was 0.007 – 1.94 µg/g d.w. when sampling periods were taken separately, but average values for the lower river and upper estuary ranged between 0.4 - 1.4 µg/g dw. In San Francisco Bay, total Hg in sediments ranged from 0.1 – 0.35 µg/g dw; at Lavaca Bay (TX) near the point of discharge sediments ranged from 0.3 – 0.7 µg/g; in Venice Lagoon (Italy) sediments average 1.35 µg/g; and in the Saguenay Fjord (Canada), sediments averaged about 3 µg/g (Heim et al. 2007; Bloom et al. 2004; Suchanek et al. 1998; Smith and Loring 1981). In the Sudbury River (MA), levels in surficial sediments ranged from about 0.3 – 20 ug/g d.w., higher than the Penobscot (although the highest concentrations in the Sudbury River were seen in mainstem reservoirs) (Frazier et al. 2000). In sediments of the Hudson River (NY), total Hg averaged about 1 µg/g in the upper 25 cm (Heyes et al. 2004). Total Hg in the sediments of Baltimore Harbor (MD) can be as high as 1 µg/g (Mason et al. 1999). For comparison, concentrations in areas without point sources of Hg are generally not higher than about 0.2 µg/g dw (St.Louis et al. 2004; Wiener et al. 1990). NOAA considers that levels in sediments of 0.004-0.051 µg/g d.w. are indicative of background conditions (NOAA 2004).

NOAA (National Oceanic and Atmospheric Administration) considers that freshwater sediments with more than 0.174, 0.486 and 0.560 µg/g d.w. total Hg exceed the Threshold Effects Level, Probable Effects Level and Upper Effects Threshold, respectively for likely toxicity to organisms living in the sediments (NOAA 2004)[1]. In the lower Penobscot River, sediments usually exceeded the Threshold Effects Level (8 to 10 of the 10 sites sampled, depending on the sampling period), the Probable Effects

[1] NOAA defines Threshold Effects Level as the concentration below which adverse effects are expected to occur only rarely. Effects Range – Low represents the value at which toxicity may begin to observed in sensitive species. Effects Range – Medium is the median concentration of just toxic samples. Probable Effects Level is defined as the level above which toxic effects are frequently expected. Upper Effects Threshold is defined as the concentration above which adverse biological impacts would always be expected. (NOAA 2004).

Level (6 to 8 of the 10 sites sampled) and the Upper Effects Threshold (5 to 8 of the 10 sites sampled). For marine sediment, NOAA considers that sediments with more than 0.130, 0.150, 0.696, and 0.710 µg/g d.w. exceed the Threshold Effects Level, the Effects Range-Low, Probable Effects Level and Effects Range Medium thresholds, respectively (NOAA 2004; Long and MacDonald 1982). In the upper Penobscot estuary (sites ES-2, 3, 5, 6, 9, 11, 12, 13, 14, and 15), sediments usually exceeded the Threshold Effects Level (9 to 10 of 10 sites sampled, depending on the sampling period), and the Effects Range-Low threshold (8 to 9 of 10 sites sampled), and often exceeded the Probable Effects Level (4 to 6 of 10 sites sampled) and the Effects Range Medium threshold (3 to 6 of 10 sites sampled).

It should be noted that although in the scientific literature concentrations of Hg in sediments are commonly presented on a dry weight basis, site-to-site comparisons using this metric are problematic. This is because the organic carbon content of sediments is highly variable and mercury binds tightly to this organic material. So, the inorganic material in sediments with a low % organic carbon (such as the Penboscot downstream of the Veazie Dam) essentially dilutes the Hg in the dried sediment sample. Thus sediments with a high percentage of organic carbon on content on a dry weight basis appear to have relatively high Hg concentrations when compared to sediments with low per cent organic carbon. Overall, this artefact may minimize differences between contaminated an uncontaminated sites or it may maximize differences among contaminated sites (e.g. Sudbury River vs. the lower Penboscot).

To minimize the impact of this artefact, we normalized mercury concentrations to the organic carbon content of the sediments. When this was done, Hg in the Old Town to Veazie reach showed smaller increases from the East Branch stations than were seen with dry weight concentrations (Figure 21). Total Hg normalized to organic carbon content was similar or up to twice as concentrated in the Old Town to Veazie reach as compared to the East Branch but 5 to 10 times as concentrated in the sediments in the lower river downstream of Veazie, and 3.8 to 5.6 times as concentrated in the estuary than upstream of Veazie (Figure 21). Whereas dry weight total Hg concentrations in sediments were generally higher immediately upstream of Orrington, concentrations standardized for organic carbon were, on average, more concentrated downstream of Orrington as compared to upstream sampling stations. For the estuarine sampling sites, the patterns of Hg expressed on a dry weight basis suggested that contamination in the estuary is confined mostly to the most northerly sites - as far south as Fort Point Cove (sites ES-11, 5, 6, 2, 15, 13, 12, Figures 6 and 20). The more southerly sites (ES 7, 4, 14, 8, and 1) appeared to be much less contaminated on a dry weight basis. However, Hg normalized to organic carbon content longitudinal gradient was much less pronounced, suggesting that the level of contamination is similar at least as far south as

mid Islesboro Island (Figure 21). There was a very similar picture when we sampled the offshore sediments of the bay (see below). These offshore data also show that the mercury concentrations normalized to organic carbon are still elevated at mid Islesboro Island, but decrease to regional background levels at mid Vinalhaven Island near the mouth of the estuary.

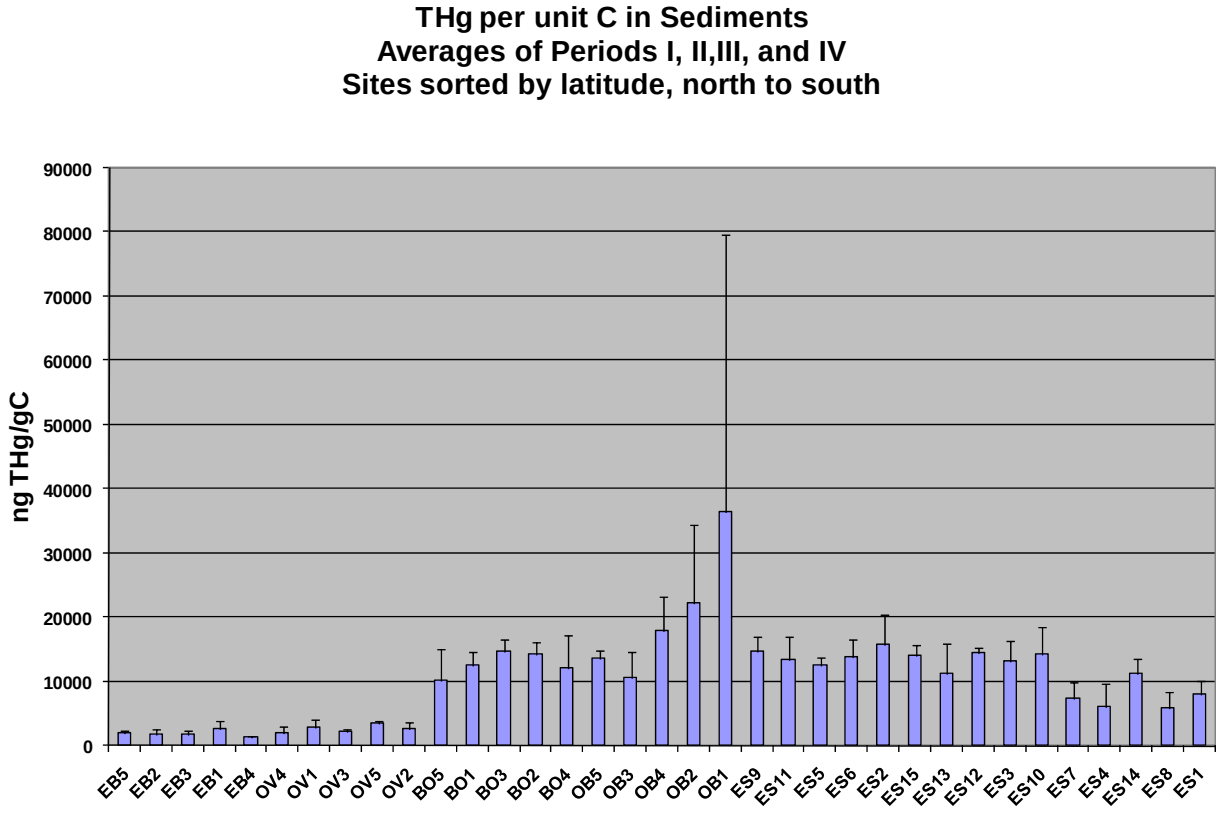

Figure 21. Total mercury in near-shore surficial sediments (0-3 cm) of the Penobscot River and estuary, standardized to organic carbon content of the sediments. Data are averaged from Sampling periods I, II, III and IV, 2006. Stations are plotted in geographic order from upstream to downstream (north to south); sites are mapped in Figures 1-6.

Average MeHg concentrations in surficial sediments in the lower river and upper estuary ranged from 1.4 – 23 ng/g d.w. (Figure 22). The methyl mercury concentrations in the Penobscot are similar to other contaminated sites. In San Francisco Bay (CA), Venice Lagoon (Italy), Lavaca Bay (TX), and Clear Lake (CA), methyl mercury in sediments ranged up to about 16 ng/g d.w. (Heim et al. 2007; Bloom et al. 2004; Suchanek et al. 1998). In the Hudson River (NY), MeHg concentrations in surficial sediments average about 1.3 ng/g (Heyes et al. 2004). Methyl mercury concentrations in the sediments of polluted Baltimore Harbor (MD) range as high as 10 ng/g (Mason et al. 1999).

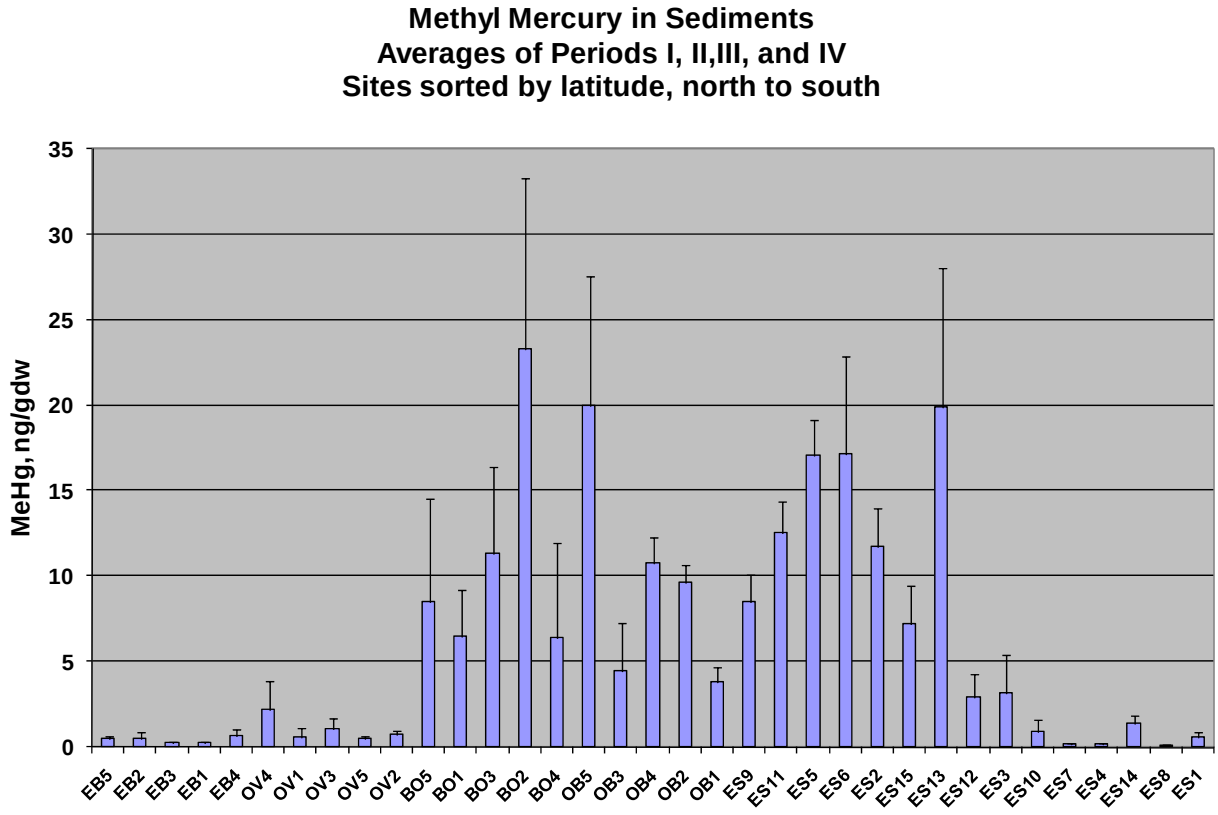

Figure 22. Methyl mercury in near-shore surficial sediments (0-3 cm) of the Penobscot River and estuary, expressed per dry weight of sediment. Data are averaged from Sampling periods I, II, III and IV, 2006. Stations are plotted in geographic order from upstream to downstream (north to south); sites are mapped in Figures 1-6.

The geographic pattern of MeHg in sediments normalized to organic carbon content was very similar to that of concentrations expressed on a dry weight basis (Figure 23).

MeHg concentrations were closely related to total mercury concentrations (Figure 24). This strong relationship between total Hg and MeHg (R^2 = 0.75) suggests that in the sediments of the Penobscot River and estuary the concentration of inorganic mercury (Total Hg is > 95% inorganic mercury) is an important factor limiting rates of MeHg production. The proportion of the total Hg present in sediments that was MeHg averaged 0.81 to 2.7 % at all sites (extreme range 0.2 – 4.5%), and did not show noticeable geographic patterns over the areas sampled (Figure 25). This constant percentage of MeHg in sediments over a wide range of total Hg concentrations further supports our conclusion that Hg concentration, and not some other environmental factor such as pH, is the primary factor controlling rates of net MeHg production in the Penobscot system.

If inorganic Hg concentrations in surficial sediments (0-3 cm), where most mercury methylation occurs, could be lowered by either direct intervention or by natural attenuation, MeHg production rates in sediments would decrease. It should be noted, however, that this dataset does not include MeHg concentrations in wetlands, which may prove to be different from nearshore sediments in their ability to methylate Hg.

Percent MeHg appeared to be lower later in the season (especially during late October) which would be expected because the metabolic activity of the methylating bacteria would decrease at lower temperatures. MeHg production has also been found to be seasonal in Lavaca Bay (TX), an estuary that has also been contaminated by Hg from a chlor alkali plant (Bloom et al. 1999).

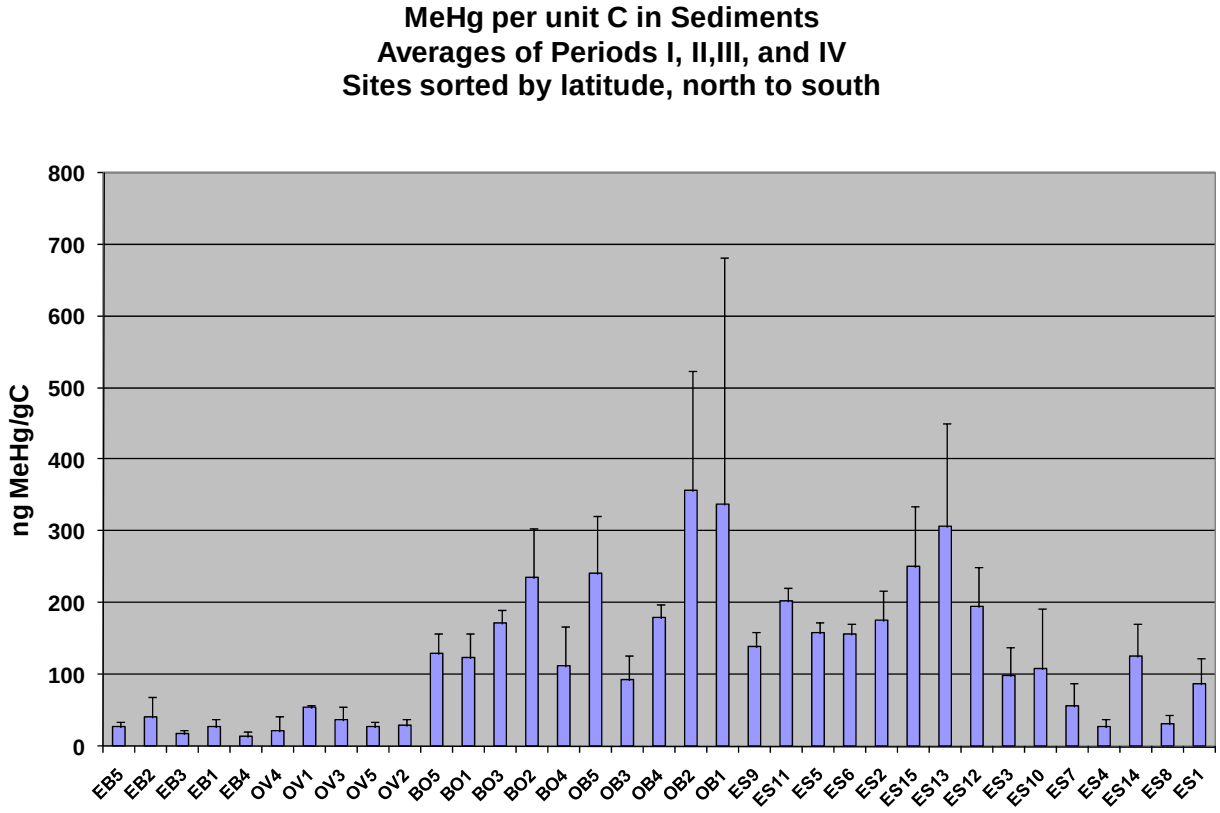

Figure 23. Methyl mercury in near-shore surficial sediments (0-3 cm) of the Penobscot River and estuary, standardized to organic carbon content of the sediments. Data are averaged from Sampling periods I, II, III and IV, 2006. Stations are plotted in geographic order from upstream to downstream (north to south); sites are mapped in Figures 1-6.

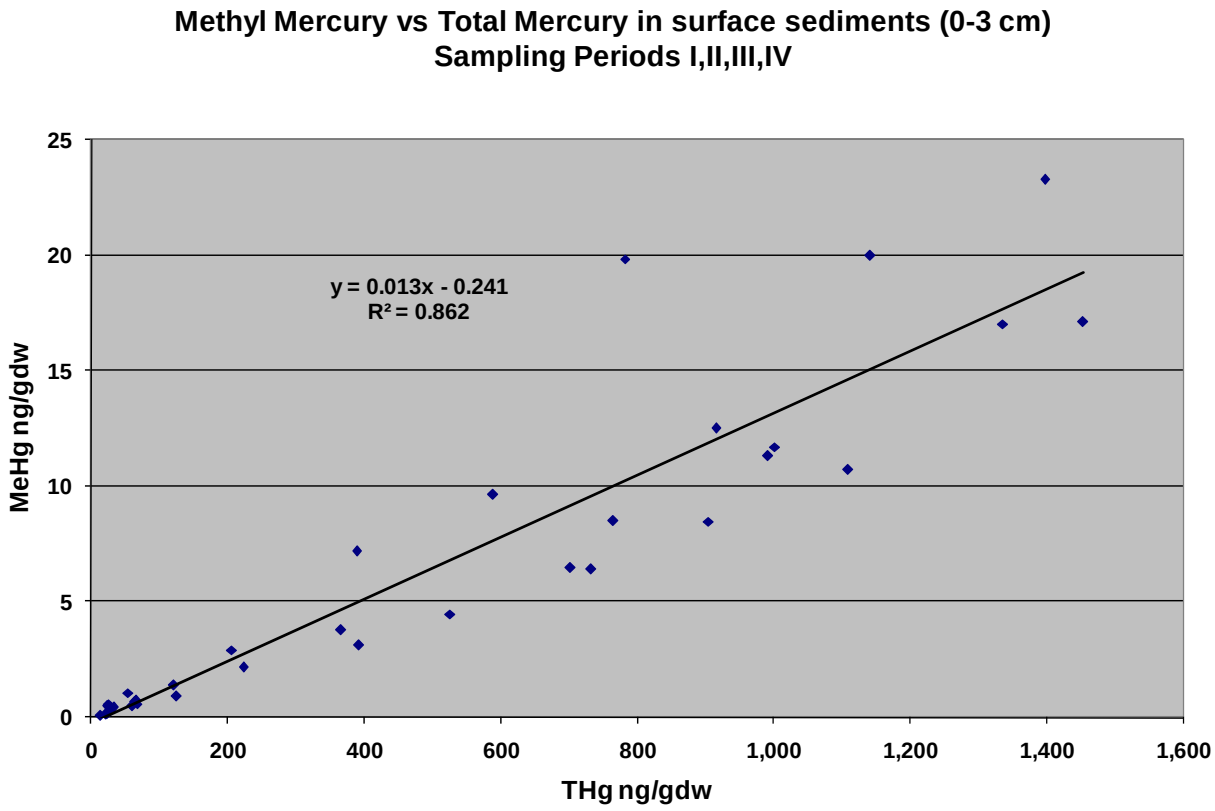

Figure 24. Relationship between methyl mercury concentrations and total mercury concentrations in near-shore surficial sediments of the Penobscot River and estuary. Mean values for each sampling station from sampling periods I, II, III, and IV are plotted.

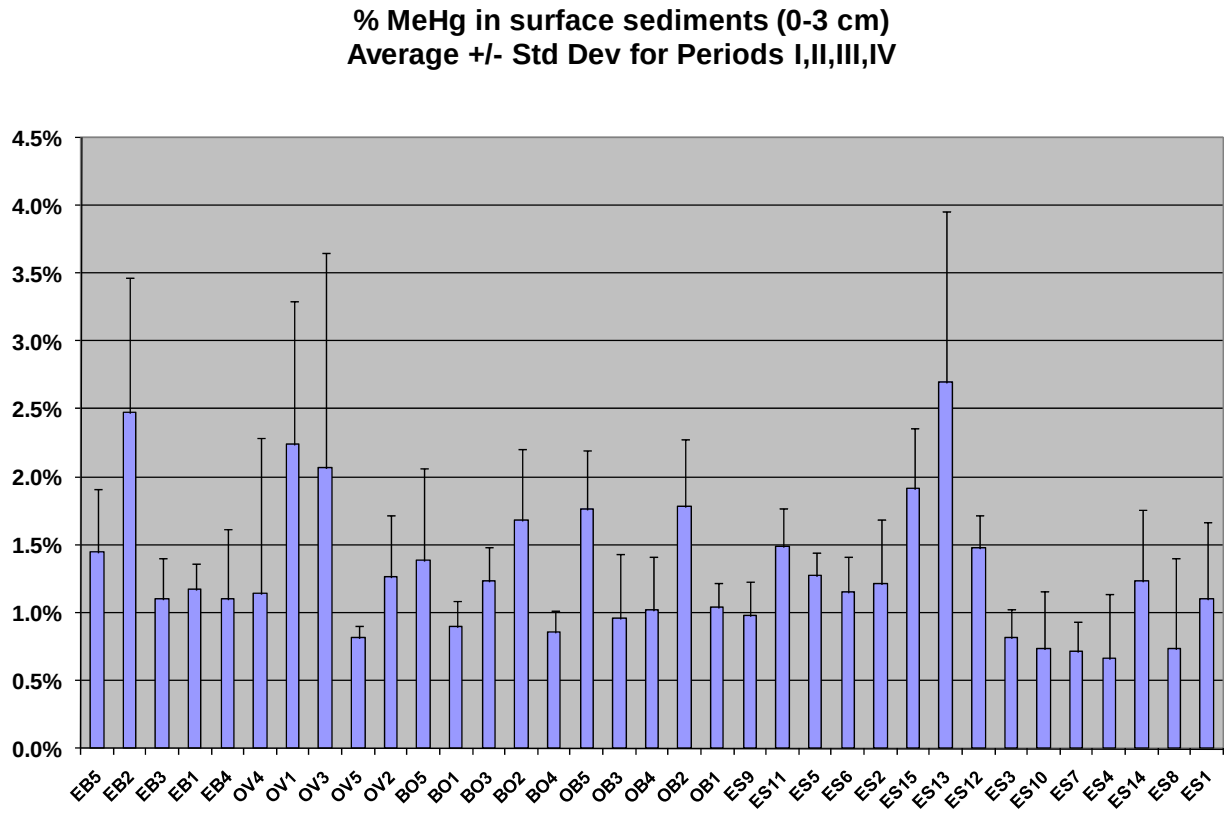

Figure 25. Percentage of the total mercury present in near-shore surficial sediments (0-3 cm depth) that was methyl mercury. Each bar is the average from Sampling periods I, II, III, and IV (+/- s.d.). Stations are plotted in geographic order from upstream to downstream (north to south); sites are mapped in Figures 1-6.

Total Hg in offshore surficial (0-3 cm) sediments of the estuary of the Penobscot River sampled in August 2007 showed a remarkably regular pattern of decreasing concentrations from near the south end of Verona Island (Fort Point Cove) to Vinalhaven Island (Figures 26 and 27). On a dry weight basis, Hg was highest at the furthest north transect and decreased in a regular manner to the most southerly transect (Figure 26). Background concentrations, similar to those in the St. George estuary, were reached only at the stations near Vinalhaven Island. Total Hg normalized to organic carbon was highest at both of the transects in the northern part of the estuary (near Fort Point Cove and between Sears Island and Islesboro Island) and decreased in a regular manner to the most southerly transect (Figure 27). As was seen for

concentrations expressed on a dry weight basis, concentrations normalized to organic carbon reached background levels, similar to those in the St. George estuary, at the most southerly stations near Vinalhaven Island. This pattern was very similar to that seen for nearshore sediments (Figure 21).

Concentrations of total Hg in surficial (0-3 cm) sediments taken from offshore sites between Fort Point Cove and mid Isleboro Island (Figure 26) are about a factor of two greater than concentrations of total Hg in samples taken at near-shore sites in the same northern part of the estuary (Figure 21). The relationship between total Hg concentrations and MeHg concentrations (Figure 26) suggests that methylation rates in the offshore sediments may be greater than in the near shore sediments, at least on a mass balance basis. This question will be addressed as part of Phase II of the study.

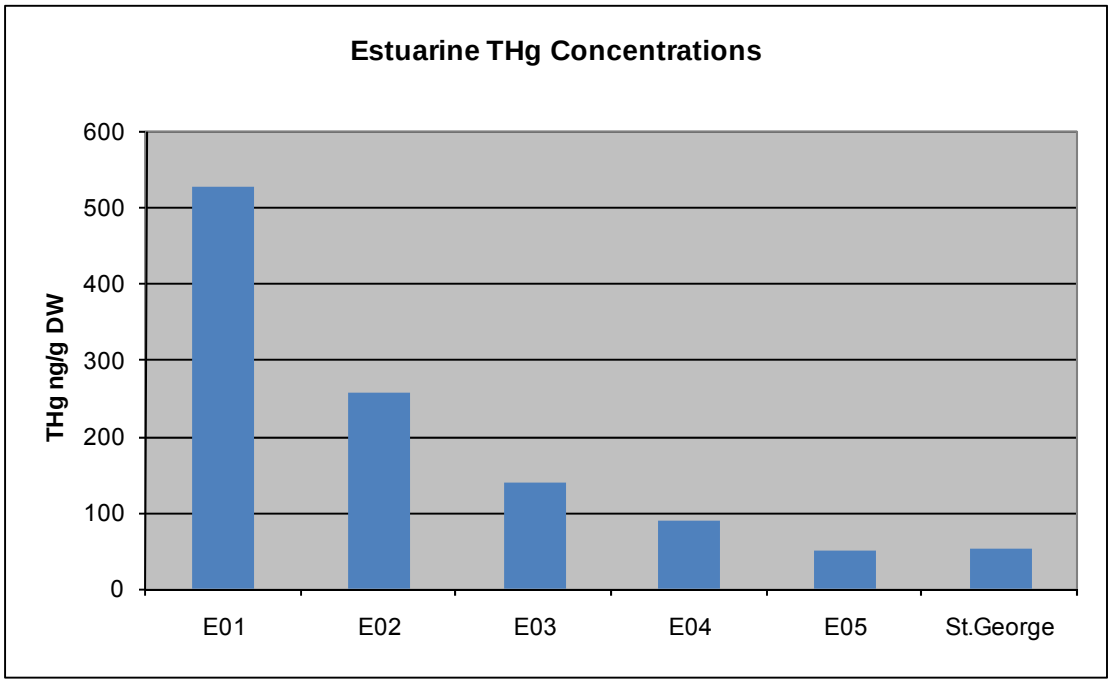

Figure 26. Mean concentrations of total mercury (ng/g d.w.) in offshore surficial (0-3 cm) sediments of the Penobscot and St. George river estuaries. The locations of transects are shown in Figure 6. All transects in the Penobscot River were oriented in an east-west direction. E01 was near Fort Point Cove, E02 was between Sears Island and Islesboro Island, E03 intersected with the north part of Islesboro Island, E04 intersected with the south end of Islesboro Island and E05 intersected with Vinalhaven Island. Samples were taken in August 2007.

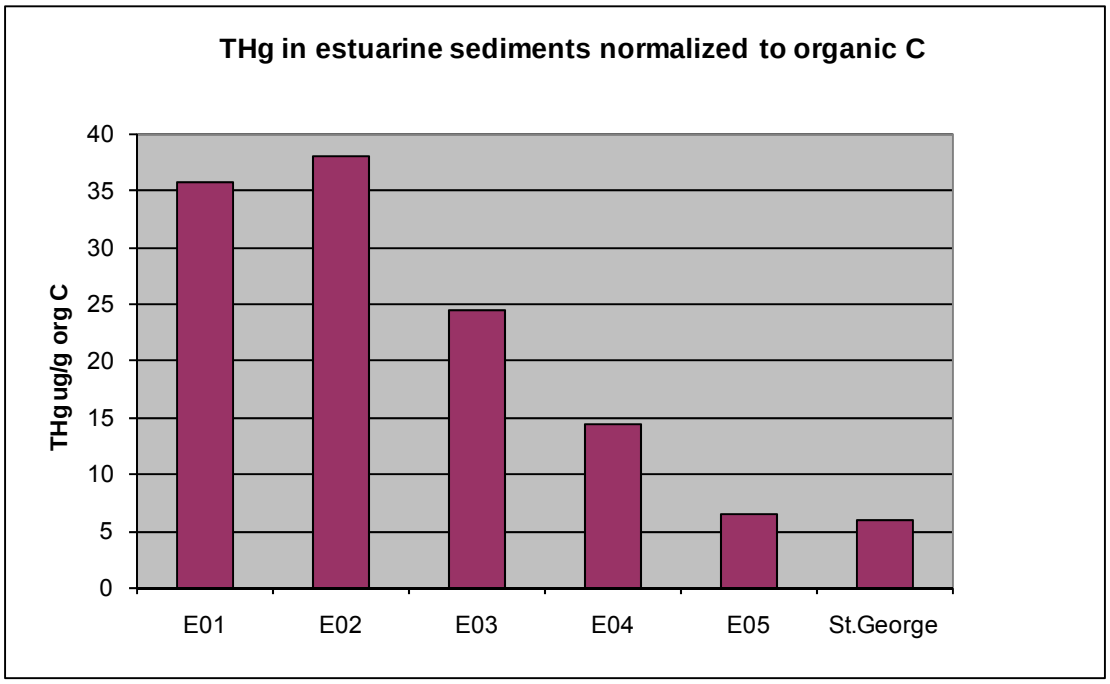

Figure 27. Mean concentrations of total mercury, normalized to organic carbon content (ng/g organic carbon), in offshore surficial (0-3 cm) sediments of the Penobscot and St. George river estuaries. The locations of transects are shown in Figure 6. All transects in the Penobscot River were oriented in an east-west direction. E01 was near Fort Point Cove, E02 was between Sears Island and Islesboro Island, E03 intersected with the north part of Islesboro Island, E04 intersected with the south end of Islesboro Island and E05 intersected with Vinalhaven Island. Samples were taken in August 2007.

Twenty seven wetlands located adjacent to the Penboscot River and estuary were sampled for total mercury concentrations in August 2007 (Figure 7). Each wetland was sampled at four elevations. One site was intertidal sediments at the front of the wetland. The other three sites were of wetland soils at increasing elevation towards the back of each wetland. The data for each of the wetlands (Figures 28 and 29) are presented as averages of these four sampling elevations. Mercury in these wetlands showed a spatial pattern of total Hg contamination that was quite consistent with samples taken at near-shore stations (Figures 20 and 21) and at offshore stations in the estuary (Figures 26 and 27). The spatial pattern was similar whether total Hg was expressed on a dry weight basis or normalized for organic carbon content of the wetland soils and sediments. All wetlands downstream of HoltraChem to the south end

of Verona Island (W61), including those in the Frankfort/Mendall Marsh area and the Orland River had high and remarkably similar concentrations of total Hg.

The most northerly wetland sampled (W05) had quite low concentrations of Hg. This site is upstream of the most upstream aquatic sampling site in the Brewer-Orrington sampling reach (See Figures 4 and 7). These results suggest that significant amounts of mercury from HoltraChem did not contaminate the river upstream of this site. This hypothesis will be confirmed as part of Phase II of the Study.

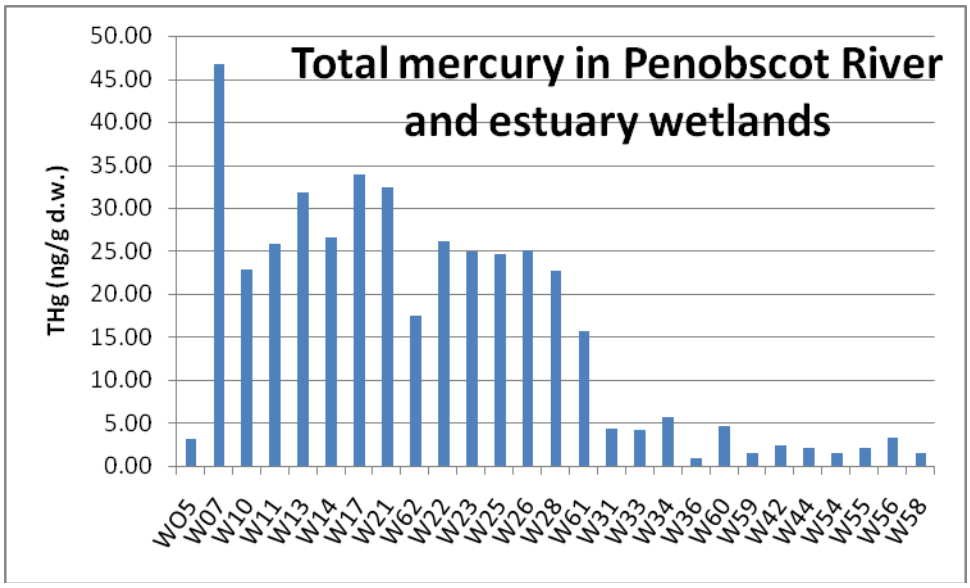

Figure 28. Total mercury concentrations (ng/g d.w.) in soils and sediments in wetlands adjacent to the Penobscot River and estuary. Each bar represents the mean of samples taken at four different elevations in each wetland. Wetlands are in order of distance from the HoltraChem site. Wetlands 05, 07, 10, 11, 13, 14, and 17 are adjacent to river in the Brewer-Orrington and Orrington-Bucksport reaches. Wetlands 21, 62, 22, and 23 are in the Frankfort/Mendall Marsh. Wetlands 25, 61, 31, 33, 34, 36, 60, and 59 are adjacent to the upper estuary. Wetlands 26 and 28 are adjacent to the Orland River and wetlands 42, 44, 54, 55, 56, and 58 are in the estuary of the Bagaduce River. A map of sites is shown as Figure 7. All Samples were taken in August, 2007.

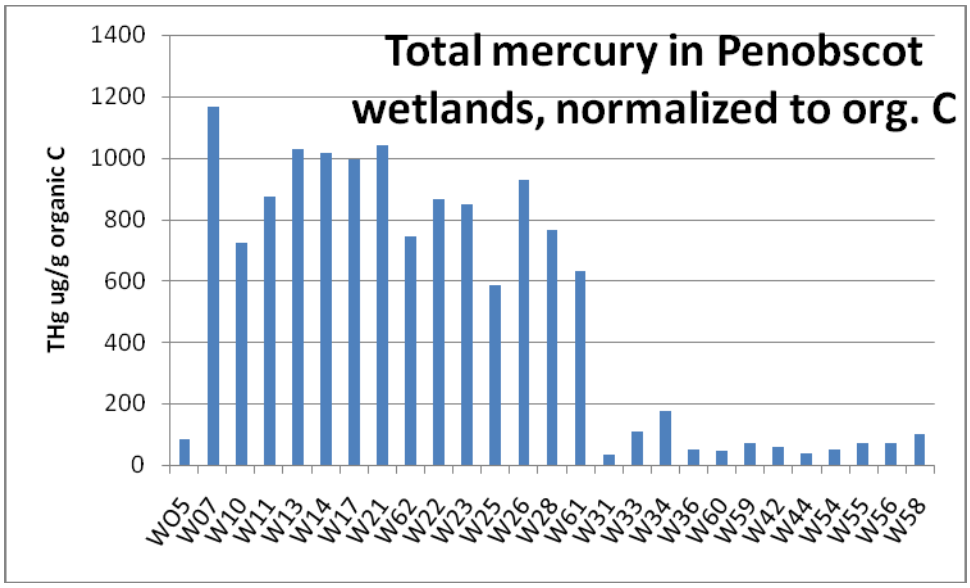

Figure 29. Total mercury concentrations normalized to organic carbon content (µg/g organic carbon) in soils and sediments in wetlands adjacent to the Penobscot River and estuary. Each bar represents the mean of samples taken at four different elevations in each wetland. Wetlands are in order of distance from the HoltraChem site. Wetlands 05, 07, 10, 11, 13, 14, and 17 are adjacent to river in the Brewer-Orrington and Orrington-Bucksport reaches. Wetlands 21, 62, 22, and 23 are in the Frankfort/Mendall Marsh. Wetlands 25, 61, 31, 33, 34, 36, 60, and 59 are adjacent to the upper estuary. Wetlands 26 and 28 are adjacent to the Orland River and wetlands 42, 44, 54, 55, 56, and 58 are in the estuary of the Bagaduce River. A map of the sites is shows as Figure 7. All Samples were taken in August 2007.

Hg was noticeably lower in wetlands further south in the estuary (Fort Point Cove and further south), including those in the Bagaduce River estuary. These results are somewhat in contrast with those from intertidal sediments and from offshore sediments discussed previously, which suggest a gradual decline in Hg in the sediments of the estuary that extends much further south down the estuary. However, the results from wetland sampling should be treated with some caution because grain size analysis results are not yet available and it is important to normalize Hg concentrations to the proportion of fine sediments in the samples to confirm geographic patterns. Also, methyl mercury results for wetland sediments are not yet available, but these should provide some indication of the relative conditions for mercury methylation in the wetlands in different parts of the river and estuary.

Mercury in invertebrates, shellfish and fish

Freshwater snails – Lymnaed (freshwater) snails (*Lymnaea megasoma*) were found in all four river reaches, but not in the estuary. Total mercury in the soft tissues of snails varied among river reaches, but did not present a simple pattern, or a strong pattern related to the location of HoltraChem. Supporting data are shown in Appendices 4 and 5. Because Hg in freshwater snails was found to be significantly higher during sampling period II, data for this period were analyzed and presented separately from the other three sampling periods in 2006. Patterns were consistent, however, between Sampling II and the other times (Figures 30 and 31). Hg was statistically significantly higher at the reference sites (EB) relative to the other three river reaches. These higher concentrations are probably related to site-specific environmental influences in the East Branch that are unrelated to Hg in the environment, such as pH, temperature or river productivity (that could influence snail growth rates and ages). Snails from the BO and OB reaches adjacent to the HoltraChem site had significantly higher total Hg levels than snails sampled in the OV reach, immediately upstream of any tidal influence. This is consistent with observations of higher Hg in suspended particles and sediments in the lower Penobscot River.

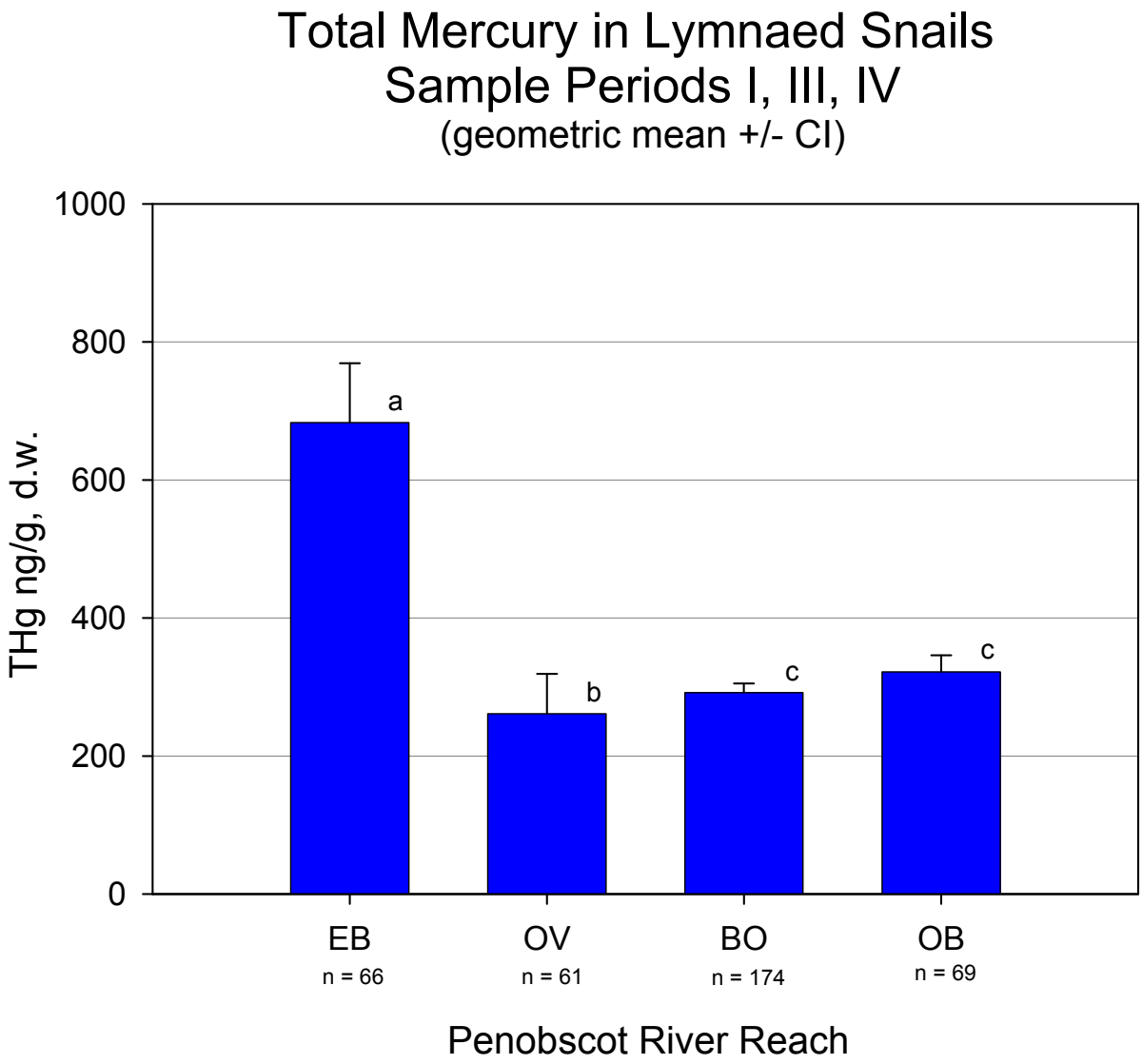

Figure 30. Geometric means (+/- 95% confidence intervals) of total mercury (ng/g d.w.) in freshwater snails in the Penobscot River, Sampling Periods I (late July/early August, 2006), III (late September/early October, 2006) and IV (late October/early November, 2006). Sample sizes are shown under each bar. Lower case letters above each bar indicate statistical differences or similarities (Tukey's Multiple Comparison Test); the same letter indicates that means are not statistically significantly different whereas different letters indicates significant differences.

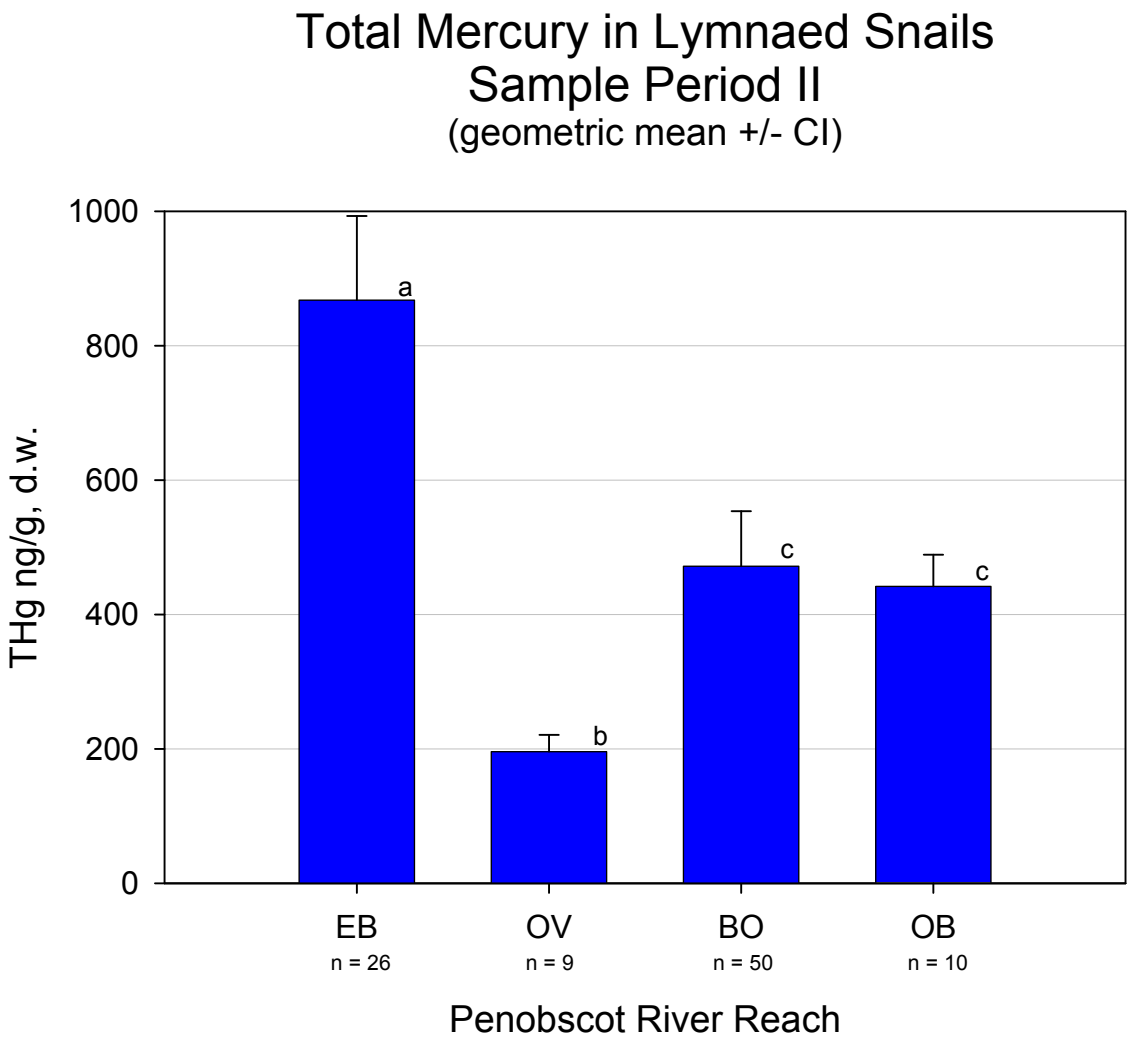

Figure 31. Geometric means (+/- 95% confidence intervals) of total mercury (ng/g d.w.) in freshwater snails in the Penobscot River, Sampling Period II (September, 2006). Sample sizes are shown under each bar. Lower case letters above each bar indicate statistical differences or similarities (Tukey's Multiple Comparison Test); the same letter indicates that means are not statistically significantly different whereas different letters indicates significant differences.

Periwinkles – Periwinkles (*Littorina*) were found at almost all estuary sites, but not in the Penobscot River. Periwinkles were sampled at the regular aquatic sampling sites, in 2006. Total mercury in the soft tissues of periwinkles varied significantly among sites in the Penobscot estuary. Average concentrations ranged from 155 ng/g d.w. to 539 ng/g (Figure 32). Hg decreased with increasing distance from HoltraChem (Figure 33, Appendix 6). Table 10 shows the raw data by site. An analysis of variance showed that variation among sampling times was not statistically significant; data from different sampling times were therefore combined and considered together (Appendix 6). Most of the variation in Hg in periwinkles was explained by distance from the HoltraChem site, with lesser amounts being explained by snail weight and % moisture (Appendix 6). Methyl mercury comprised, on average, 28% of the total mercury in periwinkles.

Concentrations of mercury in periwinkles in the Penobscot estuary were not high compared to other polluted sites but were higher than pristine sites. In a salt marsh polluted by a chlor-alkali facility in Georgia, Windom et al. (1976) found total mercury concentrations of 1,600 – 9,400 ng/g d.w., of which 3 – 10% was methyl mercury. In the polluted Limfjord, Denmark, Kiorbe et al. (1983) found about 10,000 ng/g d.w. THg in periwinkles. In Southampton Water (UK), Leatherland and Burton (1974) found 750 ng/g d.w. in perwinkles. Hg in periwinkles in the Penobscot was similar to those found in the Elbe estuary, Germany (about 400-800 ng/g d.w.) (Zauke 1977). The Elbe River is considered to have elevated concentrations in biota compared to pristine sites (Zauke 1977). Hg in perwinkles in other areas were generally lower than levels seen in the Penobscot (Severn estuary, UK: 300 ng/g, Tay Region, Scotland: 200 ng/g, Fjord of Kiel, Baltic Sea, Germany: 50-250 ng/g, Helgoland, North Sea, Germany: 250 ng/g (see Zauke 1977)).

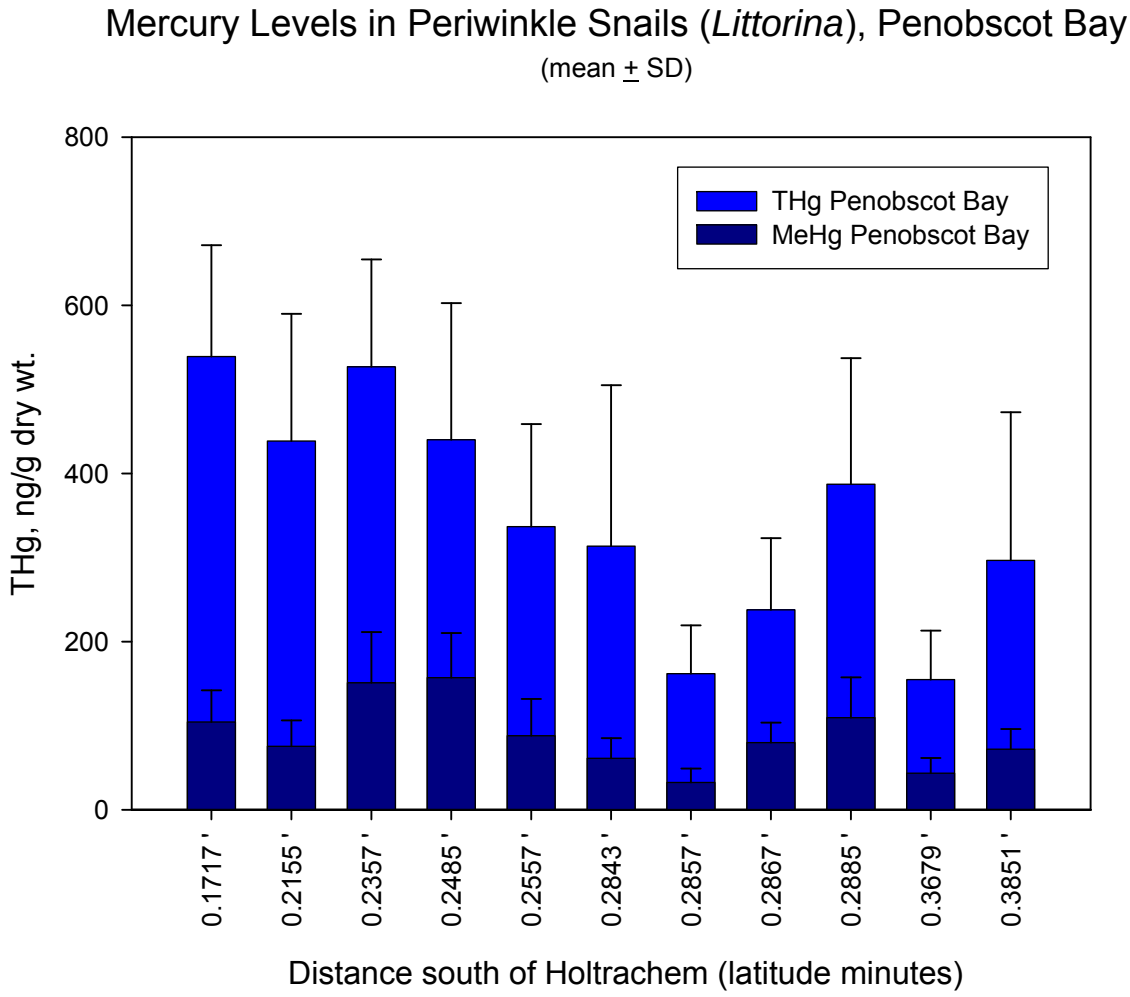

Figure 32. Mean total mercury concentrations (+/- 1 s.d.) in periwinkles sampled from the Penobscot estuary in 2006. Site means are plotted in order of distance from the HoltraChem site. Also shown are mean methyl mercury concentrations (+/- 1 s.d.) for each site.

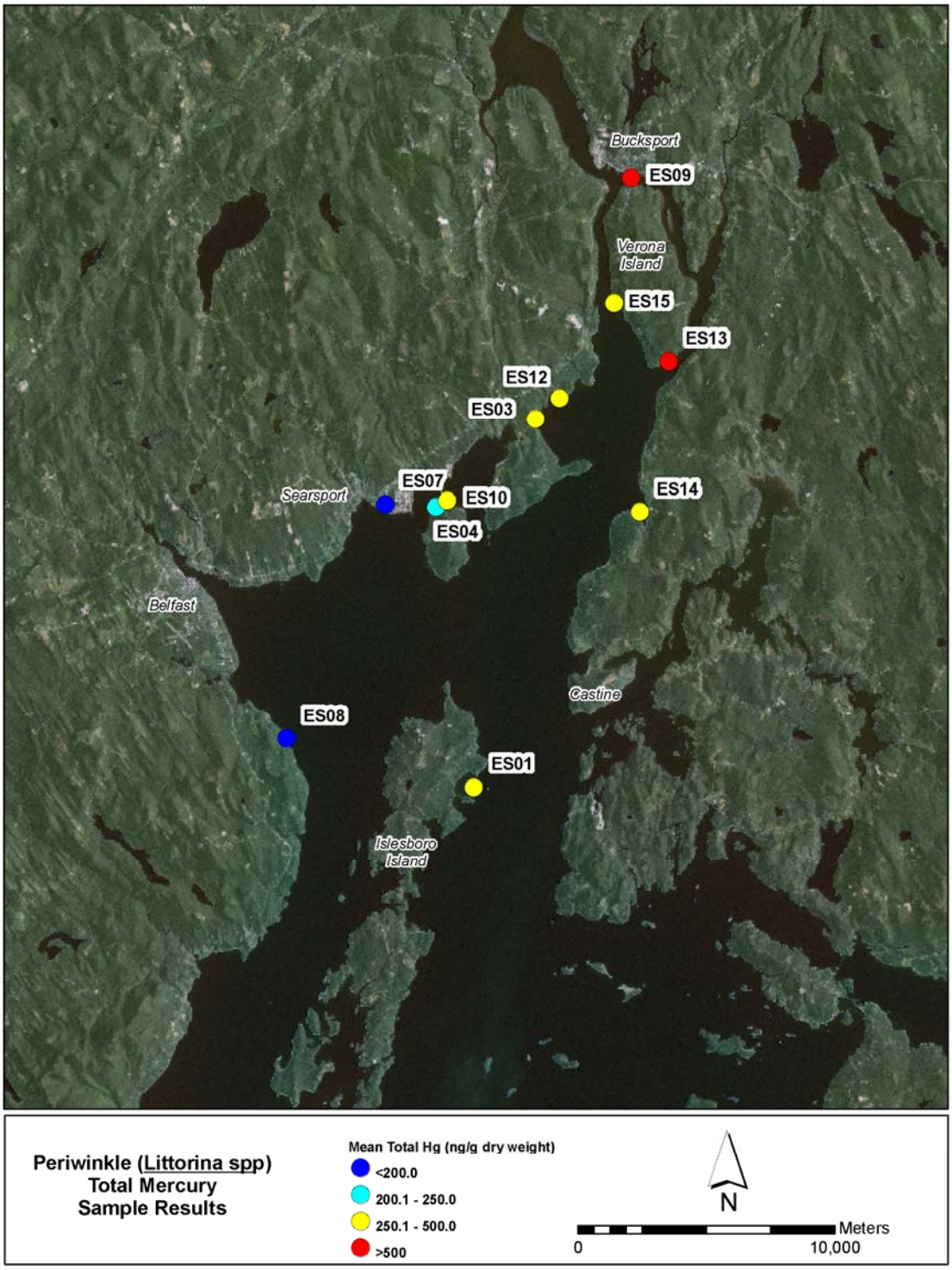

Figure 33. Map figure of average total mercury concentrations in periwinkles in the Penobscot estuary, sampled during four sampling periods in 2006.

Table 10. Mean total mercury concentrations in periwinkles sampled at 11 sites in the Penobscot estuary, 2006. Data from four sampling periods were combined. Also shown are the standard deviations, sample sizes (n), and range of total mercury concentrations.

Site	Mean (THg ng/g d.w.)	Standard Deviation	n	Range (THg ng/g d.w.)
ES-01	296.6	176.2	40	79-1150
ES-03	336.7	121.9	39	155-596
ES-04	237.9	85.2	49	116-519
ES-07	162.0	57.3	48	46-342
ES-08	154.8	58.3	40	61-376
ES-09	539.1	132.3	40	307-853
ES-10	310.1	191.2	50	70-902
ES-12	440.0	162.6	40	66-797
ES-13	526.7	127.9	40	289-860
ES-14	387.0	150.0	30	176-892
ES-15	438.5	151.3	50	135-790

Mussels – Mussels (*Mytilus*) were found at all sites in the estuary except those at the north and east site of Verona Island and those in the Orland River. Total mercury in the soft tissues of mussels showed a large amount of variation among sites, ranging from 146 to 1262 ng/g d.w. in early September and from 101 to 1279 ng/g d.w. in late September/early October (Table 11). Mercury concentrations in mussels were higher in the upper estuary (southern end of Verona Island and Fort Point Cove) and were lower at sites further south in the lower estuary (Searsport, Islesboro) (Figure 34). The rankings of sites were similar at both sampling times. Differences among sites were found to be statistically significant by analysis of variance and analysis of covariance on total mercury data (w.w.), whether animal size was used as a covariate or not (Appendices 7 and 8).

From early September to late September/early October, total mercury concentrations generally decreased (Table 11). However, the average concentration of MeHg in mussels stayed about the same over the same time period. Therefore, the proportion of the total mercury that was MeHg increased. The proportion of total Hg that was MeHg averaged 32% in the first sampling as compared to 43% in the second sampling. It would be expected that MeHg concentrations would be less changeable that total Hg concentrations based on the physiology of MeHg vs. inorganic mercury; MeHg is known

to have longer turnover times in biotic tissues than inorganic Hg. The % MeHg did not show any geographic patterns in either sampling period, so the geographic patterns seen for total Hg were also present for MeHg.

Table 11. Mean concentrations of total mercury (ng/g d.w.) in mussels sampled in the Penobscot River estuary, 2006. N=10 for all means. Late September sampling period was September 7 – 11, 2006. Sept/Oct sampling period was September 27 – October 2, 2006. Sites listed in geographic order from North to South.

Site	Mean (Late Sept)	Standard Deviation (Late Sept)	Mean (Sept/Oct)	Standard Deviation (Sept/Oct)
ES15	857.6	217.8	513.3	145.1
ES13	1262.0	272.3	1278.9	309.2
ES12	884.0	202.5	850.2	172.1
ES3	985.1	209.4	431.0	87.9
ES10	174.7	96.1	134.2	22.1
ES7	146.0	35.4	101.3	15.5
ES4	231.9	75.1	181.9	51.9
ES14	834.4	325.2	803.4	247.6
ES8	172.1	43.0	170.9	58.3
ES1	304.4	126.2	269.0	68.4

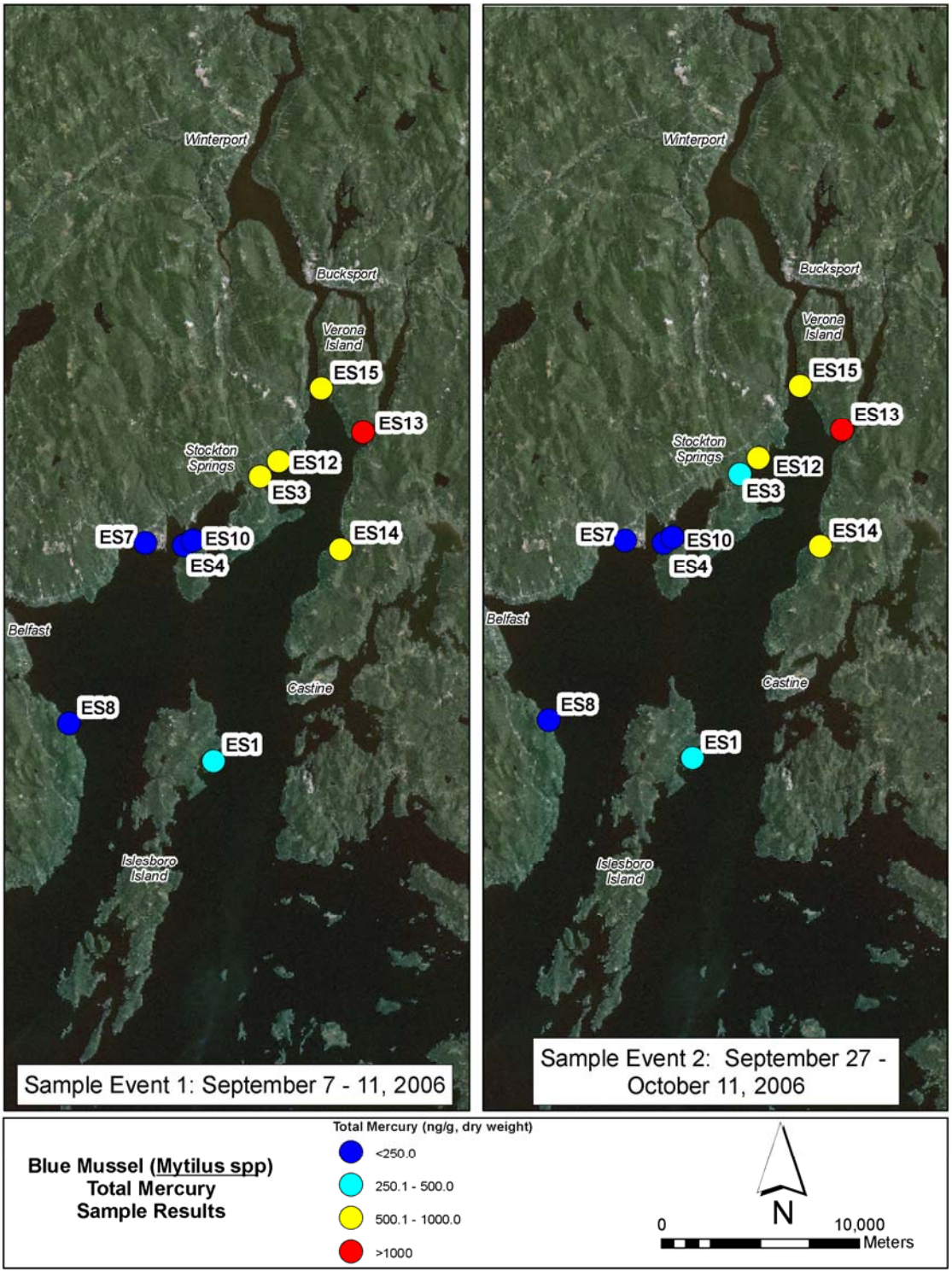

Figure 34. Average concentrations of total mercury (ng/g d.w.) in mussels in the Penobscot estuary, sampled in early September, 2006 (left map) and late September/early October, 2006 (right map).

Average total Hg concentrations in mussels at all sites in the Penobscot estuary in 2006 (both sampling times combined) ranged from about 150 to 1270 ng/g d.w., but were 685 – 1270 ng/g in the upper estuary. The concentrations determined in 2006 were usually similar to those reported in other recent studies for the Penobscot, for example by Livingston (2000), Mussel Watch (www8.nos.noaa.gov), Gulf Watch (www.gulfofmaine.org), and Maine DEP (www.maine.gov/dep). It does appear, however, that present-day levels are significantly lower than concentrations in the 1990's. At the Sears Island Mussel Watch site, mean concentrations were always higher than 300 ng/g d.w. from 1990 – 1997, compared to the present concentration of 120 ng/g. At the Pickering Island Mussel Watch site, concentrations were also noticeably higher in the early 1990's than at present (www8.nos.noaa.gov). These data suggest that there may already have been some natural attenuation of Hg pollution in the Penboscot River and Estuary. The topic of rates of natural attenuation of Hg contamination of the Penboscot ecosystem will be addressed by several of the tasks that are being planned for Phase II of the study.

The concentrations of total Hg in mussels in the upper Penobscot estuary are high relative to other sites in the region. In 2006, mean concentrations in the upper estuary (both sampling times combined) ranged from 685 to 1270 ng/g d.w. In 2005, the median concentration for Maine from Mussel Watch data was 166 ng/g and the 85th percentile for Maine was 304 ng/g. For the Gulf Watch data, 36 of 38 sites had medians less than 790 ng/g. Maine DEP found means ranging from 76 to 518 ng/g d.w. at eight sites in Maine outside of the Penobscot estuary in 2001. None of the mussels sampled in the Penobscot estuary exceeded the most protective criteria for protection of human health for consumption of MeHg in biota (set by Maine DEP at 0.2 µg/g w.w or approximately 1,000 ng/g d.w. and by the USEPA at 0.3 µg/g w.w. or approximately 1,500 ng/g d.w.).

Lobster – Lobsters were sampled in the estuary from Fort Point to Islesboro Island. The average concentrations of total mercury in lobster claw muscle at various sites in the Penobscot estuary ranged from 46 to 211 ng/g w.w. (for samples greater than one individual) (Appendix 10). Average methyl mercury concentrations ranged from 39 to 176 ng/g w.w. and MeHg comprised, on average, about 76% of the total Hg in lobster claw muscle. A subsample of lobsters was analyzed for Hg and MeHg in tail muscle and tomalley (hepatopancreas) and it was found that 75% of the Hg in tail muscle was MeHg, similar to claw muscle. Concentrations of Hg in tail muscle were on average 53% higher than in claw muscle (n=8). Therefore, some of the lobsters sampled from the upper estuary exceeded the Maine DEP and USEPA concentrations of 200 and 300 ng/g w.w. that serve as criteria for the protection of human health due to consumption of

MeHg in biota (Figure 35). At the eight upper estuary sites (see map Figure 36), of 67 lobster sampled, 25% exceeded the MDEP criterion of 200 ng/g w.w. MeHg and 6% exceeded the USEPA criterion of 300 ng/g. This was calculated from the mean of total Hg in claws and tails (from individual total Hg concentration in claws assuming tail muscle was 53% higher in total Hg) and that 75% of the total Hg in both tissues was MeHg.

There was an apparent relationship between Hg in lobster claw muscle and distance from the HoltraChem site, with mercury decreasing with distance from Orrington, although this relationship was not statistically significant (Figures 35 and 36).

Maine DEP found total mercury in lobster near Verona Island to average 120 ng/g w.w. in 1995, which is slightly lower than was seen in 2006 near the southern end of Verona Island. The levels of Hg seen in lobsters in the Penobscot estuary overlap with those from other Maine estuaries. Sowles (1997) summarized data for Hg in lobsters in Maine in 1995 and noted that means in lobster muscle (claw vs. tail muscle not specified) ranged from 82 to 208 ng/g w.w. at seven sites outside the Penobscot system.

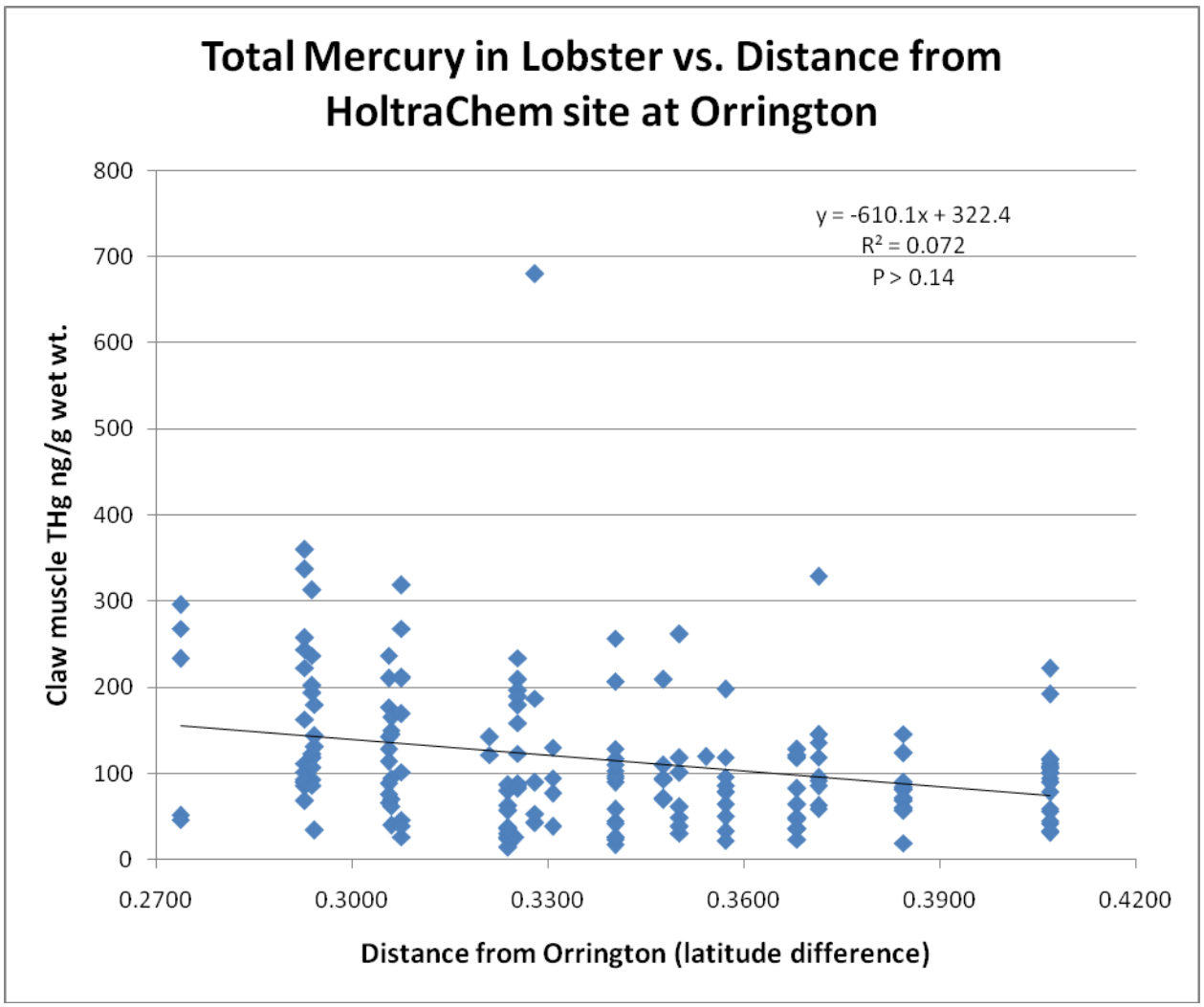

Figure 35. Total mercury concentrations (ng/g w.w.) in individual lobsters vs. distance from the HoltraChem site at Orrington, ME. Relationship is not statistically significant.

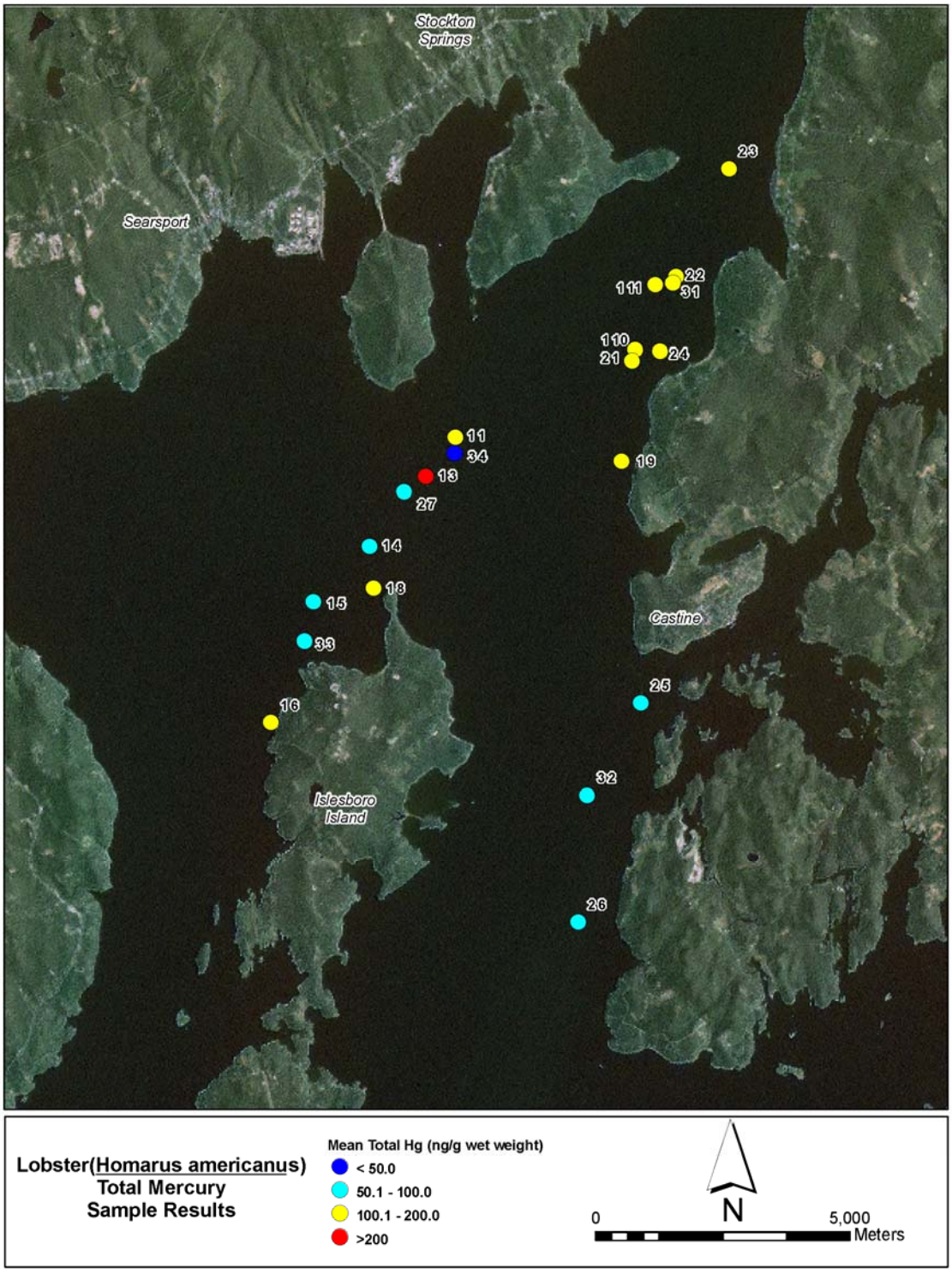

Figure 36. Total Hg concentrations in the claw muscle of lobster, Penobscot estuary, 2006.

Tomcod – Tomcod were found in the lower Penobscot River (Brewer – Orrington and Orrington – Bucksport reaches) and in the upper estuary, as far south as the south end of Verona Island. Average mercury concentrations in tomcod ranged from 104 to 238 ng/g w.w. (adjusted to a standard fork length) in two reaches of the lower river and seven sites in the upper estuary (Figure 37; Appendix 13). Mercury in tomcod was much higher in the lower Penobscot River (Brewer-Orrington and Orrington-Bucksport reaches) than in the estuary (Figures 38 and 39). Adjusted concentrations decreased significantly with distance from the HoltraChem site (Figure 38).

The proportion of the total mercury in tomcod muscle that was methyl mercury averaged 105% for all samples (14 determinations). Although it is not possible that methyl mercury can constitute more than 100% of the total amount of mercury present, this average is within the combined analytical errors of the determinations of total Hg and MeHg.

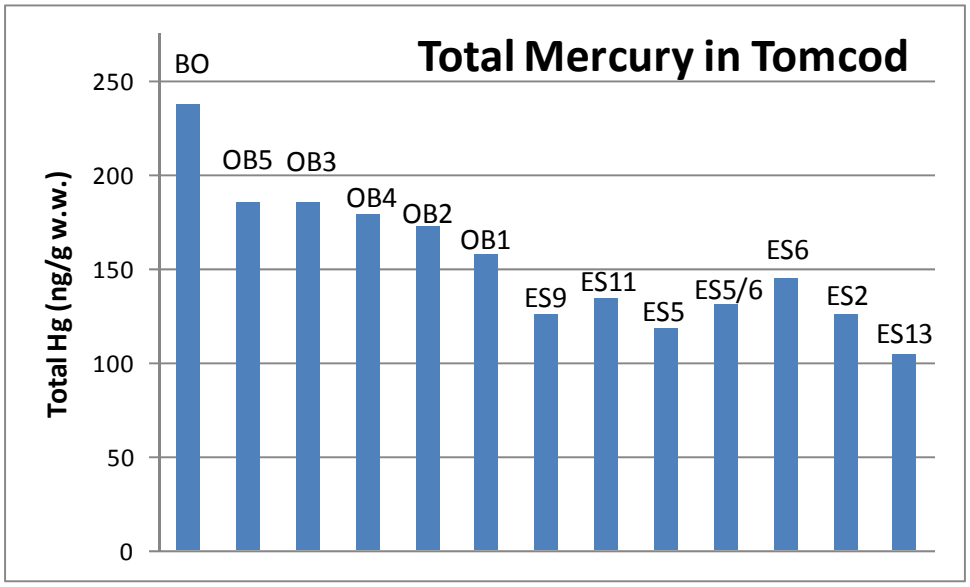

Figure 37. Mean concentrations of total mercury (ng/g w.w.) in tomcod muscle in the lower Penobscot River and upper estuary. All samples were caught September 9 to October 9, 2006. Mean mercury concentrations were adjusted by linear interpolation to 140 mm total length. BO = Brewer-Orrington; OB = Orrington-Bucksport, ES = stations in the Penobscot estuary. Sites ordered from north to south. Because of small sample sizes, all fish caught in the BO reach were considered to be one sample. Means were calculated separately for each site in the OB reach and the estuary.

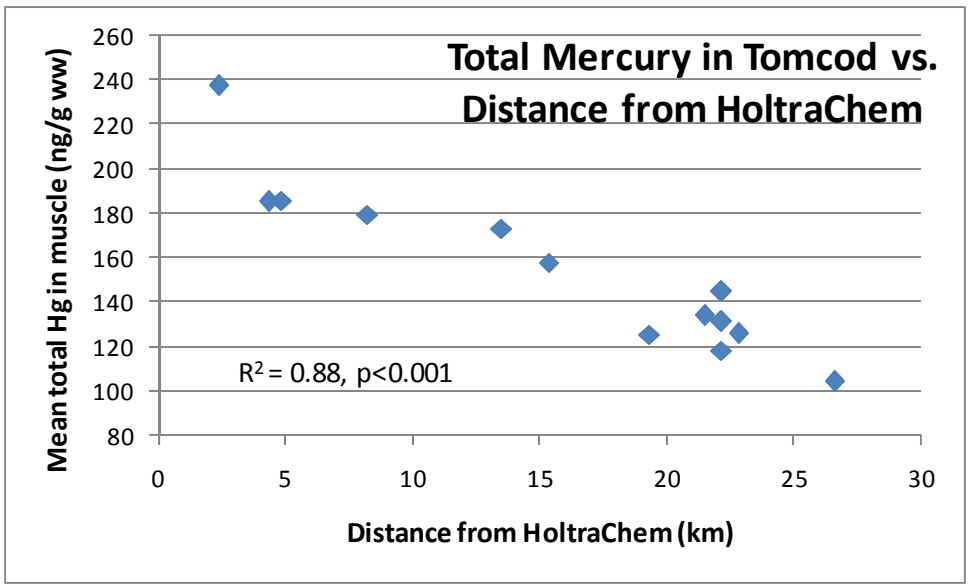

Figure 38. Total mercury in the muscle of tomcod (ng/g w.w.) at sites in the Penobscot River and estuary plotted vs. distance from HoltraChem. Mean mercury values were adjusted to 140 mm total fish length by linear interpolation of plots of mercury vs. length. Because of small sample sizes, all fish caught in the BO reach were considered to be one sample. Means were calculated separately for each site in the OB reach and the estuary.

We were not able to obtain a good sample of large, predatory fish in the lower Penobscot ecosystem, although eels caught on the river will be aged and Hg in eel muscle will be determined. The tomcod is a small fish that feed lower in the food chain than large predatory species that are often used for human consumption and that would be expected to have lower Hg concentrations than those large, predatory species. However, concentrations of MeHg in tomcod at the four sites closest to HoltraChem were near or above the Maine DEP level that serves as a criteria for the protection of human health due to consumption of MeHg in biota.

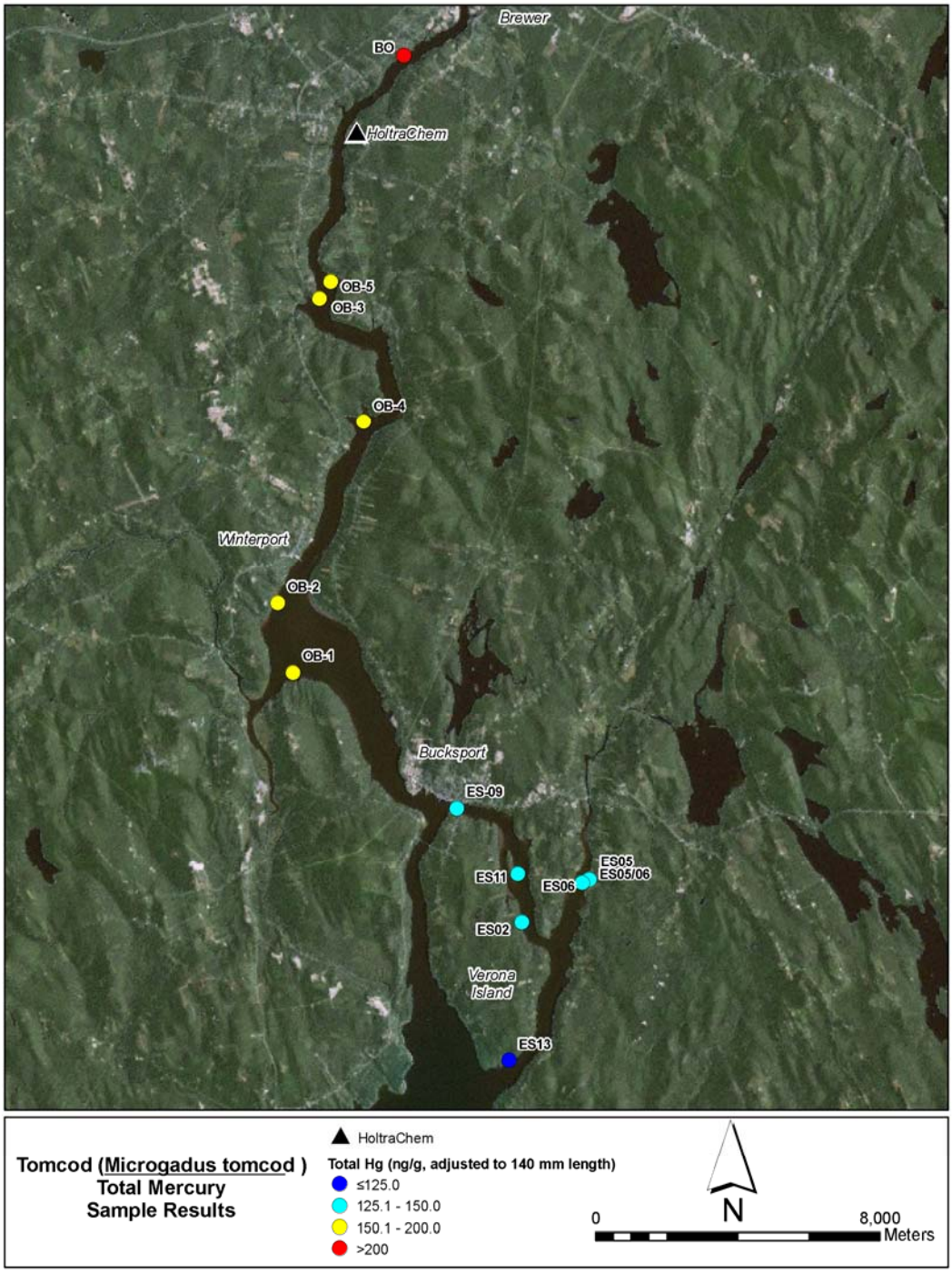

Figure 39. Map of average total mercury concentrations in tomcod at sites in the lower Penobscot River and upper estuary, 2006. Mean mercury values were adjusted to 140 mm total fish length by linear interpolation of plots of mercury vs. length. Because of small sample sizes, all fish caught in the BO reach were considered to be one sample. Means were calculated separately for each site in the OB reach and the estuary.

Mercury in birds and mammals

Cormorants - In 2006, mercury was measured in double-crested cormorant eggs sampled at six sites in the Penobscot estuary from Fort Point in the north to the south end of Islesboro Island. Mean values for each site ranged from 0.192 µg/g w.w. to 0.880 µg/g w.w., a difference of over four times (Appendix 14). Mean values showed a noticeable geographic gradient, with the highest mean value occurring at Fort Point, the most northerly site sampled and the lowest mean value occurring at Robinson Rock, the most southerly site sampled (Figure 40).

The levels of mercury seen in cormorant eggs at most of the Penobscot estuary sites are relatively high compared to other sites in Maine. BioDiversity Research Institute (Goodale et al., BRI, unpublished data) reports an overall mean concentration of 0.28 µg/g w.w. at eight sites in Maine (number of eggs sampled was 46, range 0.11 – 0.45 µg/g). Levels in the Penobscot are also relatively high compared to many other sites in N. America. Burgess and Braune (2001) found a mean mercury concentration of 0.28 µg/g in double-crested cormorant eggs from the Bay of Fundy, Canada and Henny et al. (1989) found 0.26 – 0.27 µg/g Hg in double-crested cormorant eggs from northwestern Washington. The range of concentrations in the Penobscot estuary was similar to the range seen in the mercury-impacted San Francisco Bay-Delta, which ranged from 0.17 – 1.17 µg/g in cormorant eggs (Davis et al. 2005).

Studies specific to cormorants to establish the concentrations of mercury that cause reproductive impairment or other effects have not been done. However, studies have been done on the effect of mercury in common loons (also a relatively large, fish-eating bird). Concentrations in loon eggs that are higher than 1.3 µg/g w.w. are known to cause reproductive and other effects (Evers et al. 2003; Evers et al., in press). Sandheinrich (2007) suggested that a level of 0.8 µg/g w.w. in bird eggs will be associated with reproductive impairment and Scheuhammer et al. (2007) stated that concentrations in bird eggs greater than 1 µg/g w.w. are associated with impaired hatchability. Thus, the levels seen in cormorants at the most upstream site on the Penobscot exceed levels thought to be toxic by one expert and approach toxic levels as defined by two other sources. However, studies have not been conducted on cormorants themselves.

Sampling has been carried out in 2007 at 10 sites, including all of the sites sampled in 2006 (Figure 40). 105 eggs were sampled in total. This sampling was conducted to confirm concentrations measured in 2006 and to extend the geographic range of samples from those taken in 2006 to both the north and the south. Data from 2007 sampling was not available in time to be included in this report.

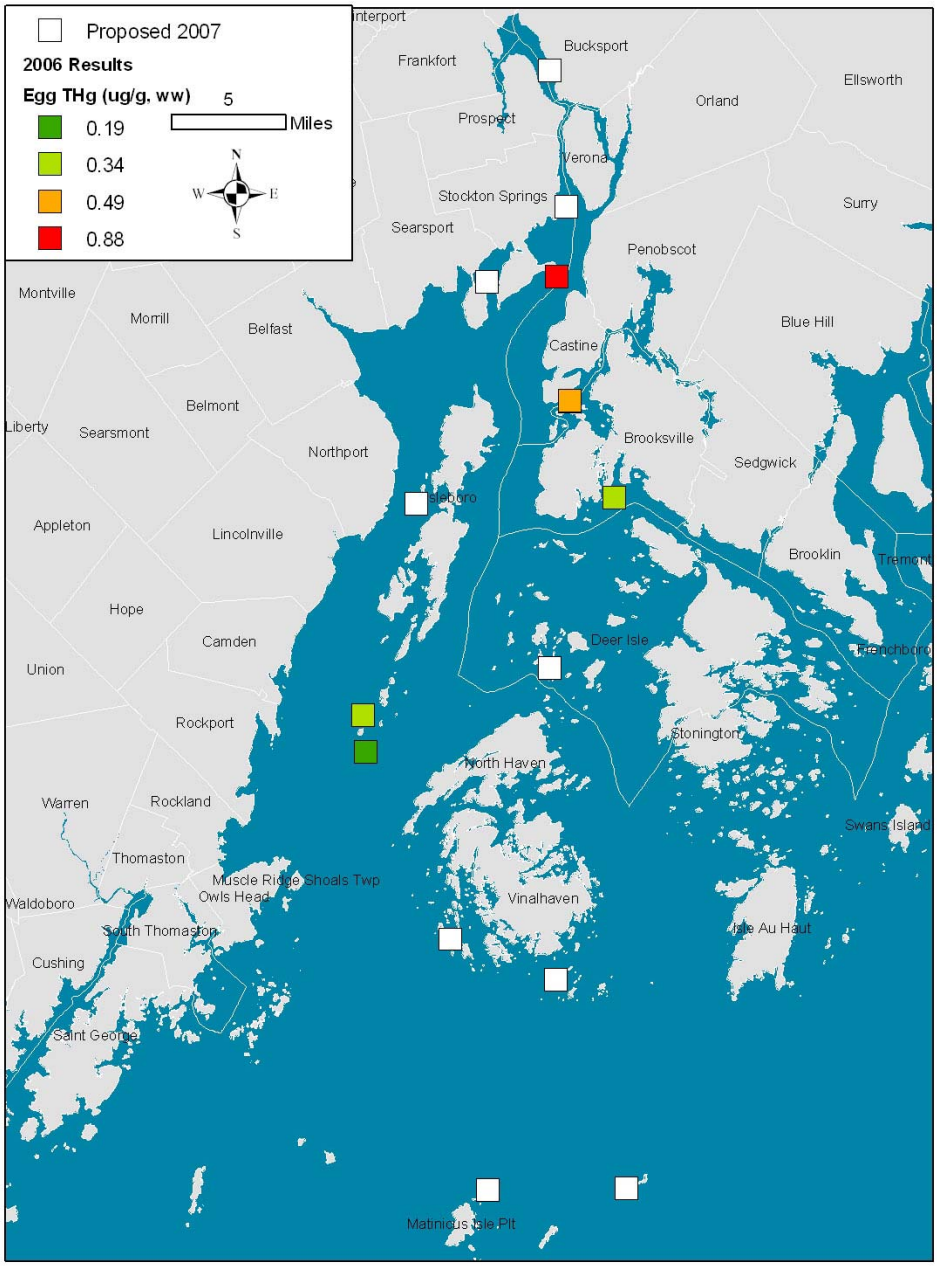

Figure 40. Mean mercury concentrations in the eggs of cormorants sampled in July and August 2006 in the Penobscot River estuary, overlain on a map of the estuary. Sampling sites are, from north to south: Fort Point, Castine, Thrumcap Island, E. Goose Rock, and Robinson Rock. Also shown (white squares) are sites that were sampled in 2007.

Wetland songbirds - Mercury in songbirds was determined in 2006 by sampling three sites adjacent to the lower Penobscot River, downstream of the Orrington site. Nelson's sharp-tailed sparrows, swamp sparrows and song sparrows were sampled at Mendall Marsh, Prospect Marsh, and at a Marsh near the town of Winterport. Mercury in the blood of sharp-tailed sparrows was much higher than in other species; means ranged from over 4 to almost 6 µg/g w.w. (Figure 41). Mean concentrations in swamp sparrows were 0.7 and 1.6 µg/g at two sites while mercury in song sparrows averaged from 0.4 – 1.9 µg/g at three sites.

Concentrations of mercury in sharp-tailed sparrows in the Penobscot were much higher than at other sites in Maine (Figure 42).

Based on recent studies of tree swallows, a concentration of 1.18 µg/g w.w. in blood is considered to be the level of concern for reproductive effects in song birds (Heinz, Evers et al. Unpublished data). Sandheinrich (2007) suggested that reproductive effects will be present when blood levels exceed 2 µg/g w.w. All three songbird species sampled in the Mendall Marsh adjacent to the Penobscot River had average concentrations of mercury in their blood that were higher than 1.18 µg/g and sharp-tailed sparrows exceeded both suggested levels of concern for reproductive effects. Therefore, biological effects due to mercury are probably occurring in these populations.

Additional sampling of wetland songbirds was carried out in 2007 to extend the geographic coverage of samples and to confirm the concentrations seen in 2006. Over 200 adult birds were sampled. Sampling included reference areas in the outer Penobscot estuary and in southern Maine. Data were not available to include in this report.

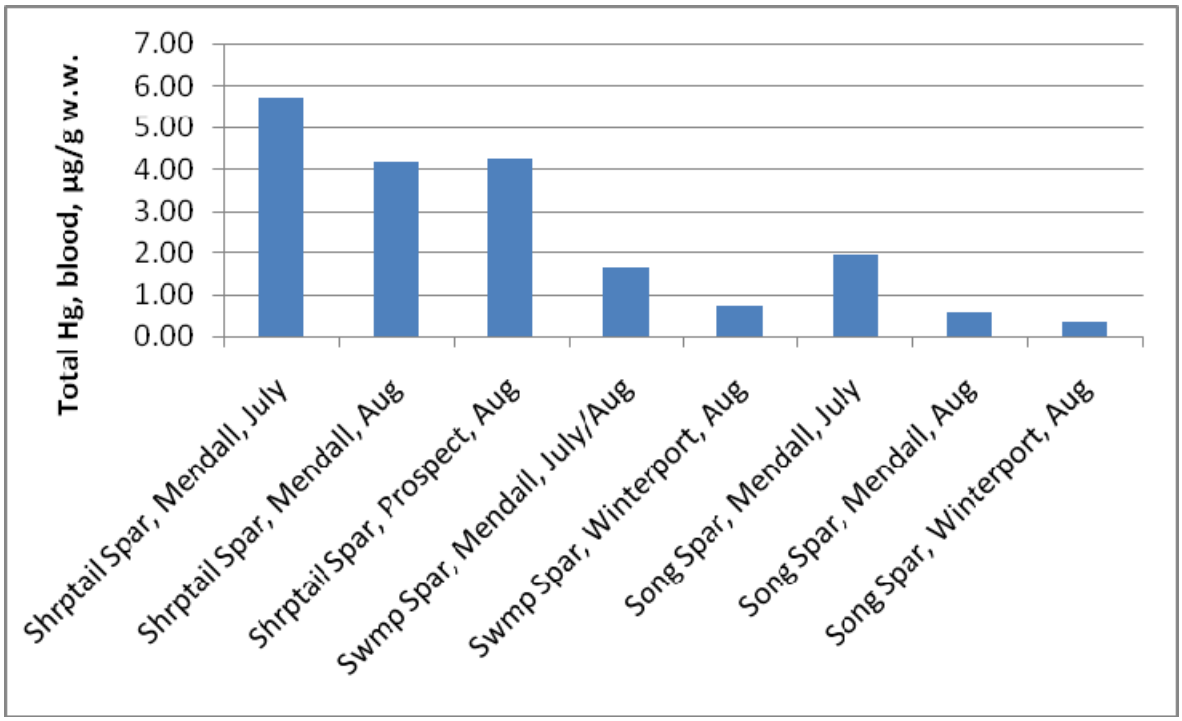

Figure 41. Mean concentrations of total mercury (µg/g w.w.) in blood of wetland songbirds sampled adjacent to the Penobscot River, 2006. Shrptail Spar = Nelson's sharp-tailed sparrow. Swmp Spar = Swamp sparrow. Song Spar = Song sparrow. Mendall = Mendall Marsh. Prospect = Prospect Marsh.

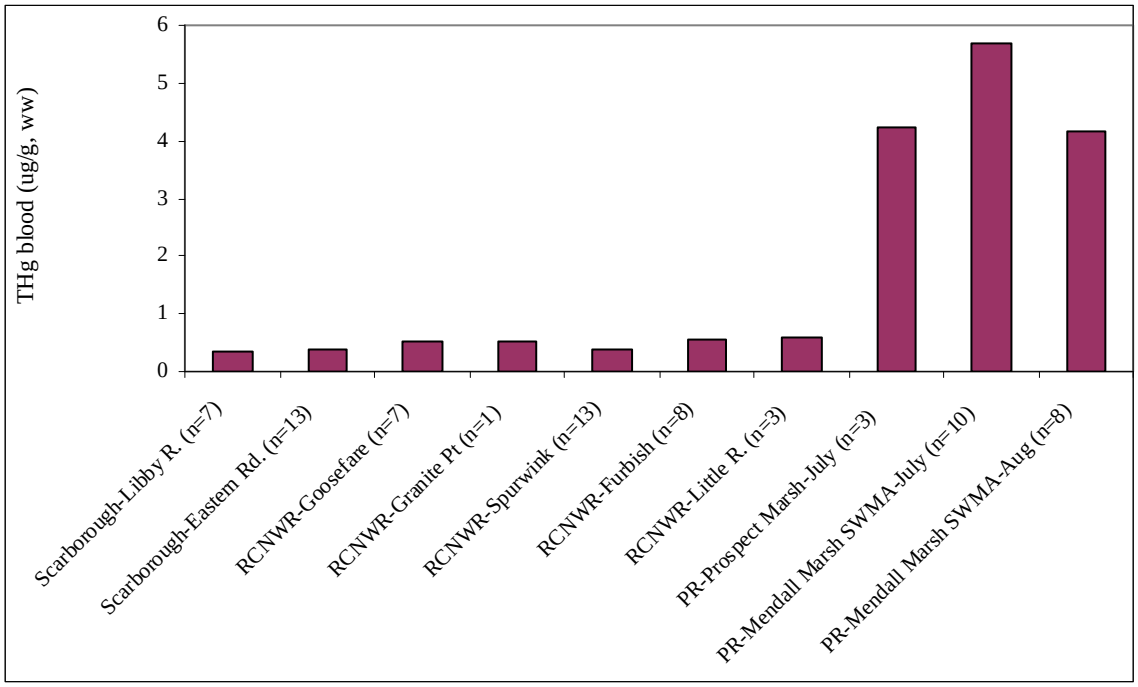

Figure 42. Mean blood THg (µg/g w.w.) in Nelson's Sharp-tailed Sparrows, Maine 2004-2006. RCNWR=Rachel Carson National Wildlife Refuge; PR=Penobscot River; SWMA=State Wildlife Management Area.

Mammals – Attempts were made to assess Hg concentrations in the tissues of both mink and river otter. We attempted to compare concentrations in animals from potentially contaminated to non-contaminated sites. Unfortunately, few animals were collected from contaminated sites, so no firm conclusions can be drawn from these data. Nevertheless, a description of the results follows.

Mink were sampled at one site potentially contaminated by Hg from HoltraChem (S. Branch, Marsh River) and four reference sites (East Branch Penobscot River, Alger Pond, Carley Brook and Pushaw Lake). Otters were sampled at two sites potentially contaminated by Hg from HoltraChem (Bagaduce River and Reeds Brook) and at six reference sites (East Branch Penobscot River, Carley Brook, Fields Pond, Jordan Brook, Pushaw Lake and Souadabscook Stream). Recent results from sampling of mercury in wetlands in the Bagaduce River estuary indicate, however, that this area

may not be contaminated with Hg from HoltraChem, thus limiting the value of the comparisons involving those sites.

Mercury in the tissues of mink sampled in the vicinity of the Penobscot was usually higher at sites that were potentially contaminated by mercury from the Orrington HoltraChem site than at reference sites (Table 12). Average mercury concentrations in three of the four tissues analyzed (brain, fur, and muscle) were higher at potentially contaminated sites than at reference sites, although none of the comparisons of the data were statistically significant (Table 12). Sample sizes were small for potentially contaminated sites, limiting the power of statistical comparisons.

Table 12. Mean concentrations of total mercury (µg/g w.w.) found in the tissues of mink sampled in the vicinity of the Penobscot River in 2006. Sampling sites were classified as being potentially contaminated with mercury from the Orrington HoltraChem site or as reference sites by proximity to the lower Penobscot River and estuary. Also shown are sample sizes, ranges, standard deviations and the p value for one-tailed t-tests (assuming equal or unequal variances as appropriate after an F-test to compare sample variances) for statistical comparisons between potentially contaminated and reference sites.

Tissue	Site Classification	Average mercury (THg µg/g w.w.)	Sample size	Range (THg µg/g w.w.)	Sample Variance	P value compared to α=0.05
Brain	Reference	0.46	17	0.16-1.15	0.07	P=0.23
	Contaminated	0.79	3	0.27-1.44	0.35	
Fur	Reference	20.38	17	11.4-33.3	52.3	P=0.28
	Contaminated	29.93	3	14.5-56.9	551.2	
Liver	Reference	2.93	15	0.58-18.4	19.21	P=0.39
	Contaminated	2.15	3	0.76-3.69	2.17	
Muscle	Reference	1.04	17	0.32-2.06	0.27	P=0.20
	Contaminated	1.66	3	0.76-2.77	1.05	

Concentrations of mercury in the tissues of mink at both reference and potentially contaminated sites in the Penobscot area were generally similar to other sites in North America. Comparisons to concentrations reported in the literature are shown in Appendix 17.

In mink, it is known that concentrations of total mercury in the brain higher than 4.1 µg/g cause negative alterations to the brain's cholinergic system (Basu et al. 2006) and Sandheinrich (2007) suggested that brain concentrations exceeding 5 µg/g will be associated with reduced reproductive success in mammals, but none of the individual otter sampled in this study approached these concentrations in their brains. Sublethal and lethal effects are known in mink at concentrations of 20-30 µg/g in liver (Halbrook et al. 1994; Mierle et al. 2000), but again, no animals sampled had concentrations this high. However, levels of greater than 20 µg/g total mercury in fur have been associated with reduced survivorship (Halbrook et al. 1994; Mierle et al. 2000) and some animals had mercury concentrations in fur higher than 20 µg/g. The mean concentration in fur at potentially contaminated sites was about 30 µg/g and the

highest individual had about 57 µg/g total mercury in fur. At reference sites, the mean concentration in fur was about 20 µg/g, the highest concentration observed was 33 µg/g and a number of individuals had concentrations above 20 µg/g. Given the small number of animals sampled at sites that were potentially contaminated with mercury from the Orrington HoltraChem site, it may be worthwhile to attempt to perform more complete sampling in the future.

Average concentrations of mercury in the tissues of otter sampled in the vicinity of the Penobscot River were always lower at sites that were potentially contaminated by mercury from the Orrington HoltraChem site than at reference sites, although only one of the three statistical comparisons that were possible were significant (Table 13). Sample sizes were small for three of the four tissues at potentially contaminated sites.

Table 13. Mean concentrations of total mercury (µg/g w.w.) found in the tissues of river otter sampled in the vicinity of the Penobscot River in 2006. Sampling sites were classified as being potentially contaminated with mercury from the Orrington HoltraChem site or as reference sites by proximity to the lower Penobscot River and estuary. Also shown are sample sizes, ranges, standard deviations and the p value for one-tailed t-tests (assuming equal or unequal variances as appropriate after an F-test to compare sample variances) for statistical comparisons between potentially contaminated and reference sites.

Tissue	Site Classification	Average mercury (THg µg/g w.w.)	Sample size	Range (THg µg/g w.w.)	Sample Variance	P value compared to α=0.05
Brain	Reference	0.50	11	0.38-0.64	0.006	P=0.18
	Contaminated	0.28	2	0.14-0.42	0.037	
Fur	Reference	23.18	10	17.0-31.9	35.7	P=0.003
	Contaminated	15.29	9	8.13-21.7	21.8	
Liver	Reference	1.90	11	1.10-3.00	0.34	P=0.52
	Contaminated	1.08	2	0.56-1.61	0.55	
Muscle	Reference	0.94	12	0.35-1.49	1.01	n/a
	Contaminated	0.91	1	n/a	n/a	

Concentrations of mercury in otter tissues from the Penobscot were generally similar to other sites in North America and Europe and were within the range seen at other sites in Maine. Comparisons to concentrations reported in the literature are shown in Appendix 18.

In otter, it is known that concentrations of total mercury in fur that exceed 20 µg/g are associated with reduced survivorship (Halbrook et al. 1994; Mierle et al. 2000). The mean concentration of mercury seen in otter fur at reference sites was about 23 µg/g, with a number of animals having concentrations higher than 20 µg/g. At contaminated sites, the mean concentration in otter fur was about 15 µg/g and two individual animals had concentrations slightly above 20 µg/g. Sandheinrich (2007) suggests that brain concentrations exceeding 5 µg/g will be associated with reduced reproductive success in mammals; none of the individual otter sampled in this study approached this concentration in their brains.

Preliminary sampling of mercury for stable isotope signatures

Because Hg exists in nature as seven different stable isotopes, six of which are quite abundant (greater than about 6% of the total), stable isotope fingerprinting has the potential to be able to trace movements and transformations of Hg in the environment. New, ultra-sensitive instrumentation can be used to detect small differences in the ratios of various Hg stable isotopes in environmental samples. To determine the potential for using measurements of the stable isotope ratios of Hg found in the Penobscot system to assess the extent and degree of contamination from HoltraChem, we examined Hg found in aquatic sediments at six locations in and near the Penoboscot system in 2006. Three of the locations were at the HoltraChem site and three of the sites were outside of the direct aquatic influence of HoltraChem. Sampling locations and raw data are in Appendix 19.

The ratio of two of the stable isotopes of Hg sampled from the Orrington chemical plant site were significantly different from samples taken from other Maine sites out of the direct aquatic influence of the Orrington site (Figure 43). Therefore, Hg found on the Orrington site is isotopically different from Hg found at other sites subject mainly atmospheric deposition of Hg. The sample taken from the Southerly Stream (Orrington 3) was somewhat different from the other two Orrington samples, apparently due to the mixing of Hg from the site and background Hg in the watershed of the Southerly Stream. The sample from Eddington was in between those from HoltraChem and sites

further away. This would occur if the Eddington area received mercury that was released into the air from the HoltraChem plant and then deposited onto the ground and water. Overall, these results indicate a reasonable possibility that stable isotope fingerprinting will be a useful technique to examine the contribution of HoltraChem Hg to the Penobscot River and estuary. A number of additional samples were taken in 2007 to test further the utility of mercury stable isotope ratios to trace Hg from the HoltraChem site, including from offshore sediments in the estuary, from wetlands adjacent to the lower river and upper estuary, and in sediment cores taken from sites in the lower river, but those results were not available for this report.

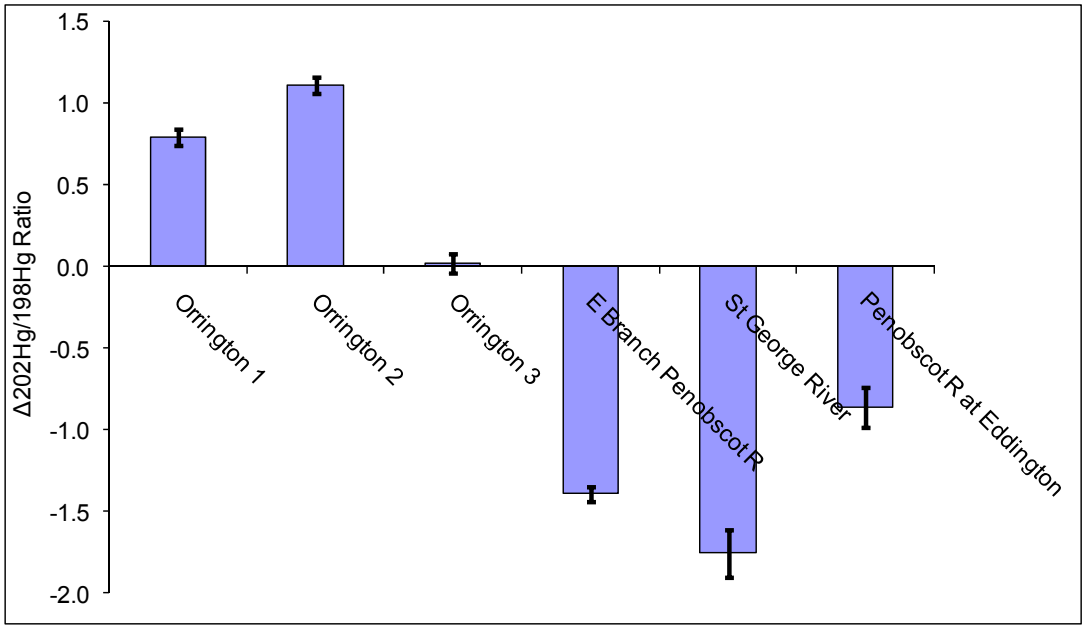

Figure 43. Deviations from the ratio of two stable isotopes of mercury (202/198) from three samples taken from the Orrington site and three samples taken outside of the aquatic influence of Orrington. Error bars are +/- 2 s.d.

DISTRIBUTION, TRANSPORT, AND BIOACCUMULATION OF MERCURY IN THE PENOBSCOT RIVER AND ESTUARY

Distribution of mercury

The data presented in this report clearly demonstrate that there has been a large input of mercury to the Penobscot River below the Veazie Dam. Our data show that mercury contamination of the ecosystem extends from river sampling sites upstream of the HoltraChem plant to sites as far south as southern Islesboro Island in the estuary. The highest levels of contamination extended from reaches of the river below the dam into the upper estuary to a point just south of Sears Island, with the peak of contamination occurring at Frankfort Flats. There is an area of lesser contamination in the estuary that extends to southern end of Islesboro Island. The details of this distribution are described below.

Overall, the distribution of the elevated mercury in the ecosystem generally follows a consistent pattern in river sediments, and it is quite coherent. In inshore depositional sediments, total mercury concentrations in the river downstream of the Veazie Dam are much higher than upstream of the dam - about 10 fold higher if the data are plotted on a dry weight basis (Figure 19). Dry weight total mercury concentrations of inshore sediments appear to decrease quite quickly in the upper estuary (about Site ES10, Figure 19 – see map Figure 6).

Patterns of total mercury concentrations in sediments (on a dry weight basis) can be misleading because Hg preferentially binds to organic carbon in sediments, and different sediment sites contain different concentrations of organic carbon. When the dry weight mercury data are normalized to organic carbon content a more accurate site-to-site comparison of mercury contamination can be made. For inshore sediments, the contamination appears to extend as far south as we sampled, which was site ES-0I at mid Islesboro Island (Figure 20 – see map Figure 6).

The southern geographic limit of the contamination was clearly shown by the transects of offshore bay sediments, which were sampled further south than the inshore sediments. In these offshore sediments, total mercury concentrations were elevated as far south as southern Islesboro Island (Transect EO4, Figure 2, and Figure 22), but were at regional background levels (i.e. the same as the St. George River sediments) by the next transect further south, which was at mid Vinalhaven Island (Transect EO5, Figures 6 and 26).

A difference between the inshore intertidal and offshore subtidal sediments in the bay is that the subtidal sediments are twice to three times as high in mercury concentration as the intertidal sediments (Figures 20 and 26). This is important because we have seen a definite relationship between total mercury concentrations and methyl mercury concentrations (Figure 19), so production of methyl mercury in offshore subtidal sediments may be higher than in inshore intertidal sediments. This possibility will be examined as part of phase II of the study.

Total mercury concentrations in the wetlands sampled had a similar geographic pattern to the river and estuarine sediments. On a dry weight basis or normalized to organic carbon concentrations were high and similar in the river upstream and downstream of HoltraChem, in the Frankfort Flats/Mendall Marsh area, in the Orland River, and in the upper estuary as far south as the southern tip of Verona Island (Figures 27 and 28). Total mercury concentrations were much lower in estuarine wetlands south of Verona Island and in the Bagaduce River, but we do not know if they are at regional background levels because we did not sample wetlands south of Islesboro Island as was done for the offshore bay sediments.

Based on these data for Hg in intertidal sediments, subtidal sediments and wetland soils, we conclude that the Penobscot ecosystem has been contaminated by mercury from a point source or sources located downstream of the Veazie Dam. This contamination extends from the river reaches we sampled south of the dam to approximately the southern end of Islesboro Island. As a result of this finding, Phase II of the study will be concentrated primarily within these geographic limits.

Transport of mercury

Understanding the processes affecting the transport and deposition of mercury in the river and estuary is important because this explains the distribution of mercury described above. With respect to mercury transport, the Penobscot River and estuary are operating in manner similar to other estuaries (e.g. Cossa et al. 1988). In the East Branch of the river, where suspended particulate concentrations are much lower than at downstream sites (Figure 14), total mercury is primarily (90%) transported in a dissolved form attached to dissolved organic carbon. Below the HoltraChem site, where the water is more brackish and turbulent, the concentration of particles (suspended sediment) is much higher (Figure 14). In the lower river, 60 - 90% of the mercury is being transported down the river on particles. The binding and transport of mercury on particles is further enhanced in the lower river by the higher Kd's in these brackish parts of the river (Figures 17 and 18). Higher Kd's mean that relatively more mercury is

bound to particles than is dissolved. This is thought to be caused by the salting out of dissolved organic carbon at the marine/freshwater interface. The organic carbon takes the Hg out of solution with it, because Hg is tightly bound to organic carbon (Turner et al. 2001; 2002). This salting out of dissolved Hg has been observed elsewhere in brackish waters (Turner et al. 2001; 2002). In the Penobscot system, the location of greatest deposition of Hg appears to be in the Frankfort Flats area. There are two reasons for this. One is that Frankfort Flats is the first large downstream depositional area for suspended particulates. The second is that the concentration of Hg in sediments (normalized to carbon) is the highest at Frankfort Flats (sites OB1 and OB2, Figure 20, see map Figure 5), likely because of the salting out process.

Downstream of the Frankfort Flats area, particulate Hg concentrations in water decline as the Hg laden particulate material sediments to the sediment/water interface in the bay (Fig. 20).

Wetlands are known to be areas with high rates of mercury methylation. Unfortunately for the Penobscot system, the location of the greatest accumulation of mercury rich sediments, at Frankfort Flats, is also the confluence of the Marsh River, and the location of the largest single wetland in the ecosystem, Mendall Marsh. The combination of a wetland environment and the higher inorganic mercury concentrations, which stimulate mercury methylation (Figure 19), may explain the very high mercury concentrations seen in songbirds at this location (Figures 38 and 39). This will be further explored as part of Phase II of the study.

Mercury bioaccumulation

In other Hg contaminated estuaries, such as the San Francisco Bay-Delta, MeHg concentrations in the food web have been found to be primarily controlled by MeHg concentrations in the local environment, rather than by factors within the food web itself (Marvin-DiPasquale et al. 2007). The Penobscot estuary appears to be operating in the same manner. In the southern-most sampled sites of the estuary, where total Hg concentrations were lowest, MeHg concentrations in biota were also at a minimum. In general, MeHg concentrations in biota increased in a northerly direction as the biota sampling sites approached the mouth of the river, where MeHg concentrations in sediments were higher. This was true for periwinkles (Figure 32), mussels (Figure 33), lobster (Figure 35), tomcod (Figure 38) and for cormorant eggs (Figure 29). Thus, mercury concentrations in the biota were most determined by how close they were to the areas of highest Hg contamination in the Penobscot system.

CONCLUSIONS AND RECOMMENDATIONS

The primary objective of Phase I of the Study was to determine whether concentrations of Hg in shellfish, fish and wildlife are high enough to be of concern. If so, a recommendation would be made to proceed to Phase II of the Study which will concentrate on understanding factors controlling the production, transport and bioaccumulation of MeHg, so that possible remediation measures can be tested in the future.

Four criteria were used to decide whether the environment and biota of the Penobscot River and estuary have high enough levels of Hg to be of concern and whether the source of that mercury appears to be the HoltraChem plant site. These four criteria were: 1. Comparison of concentrations of Hg seen in the Penobscot system to available agency guidelines (NOAA, MDEP, and USEPA) for toxic effects on benthic organisms and for human consumption, 2. Evaluations of Hg concentrations found in the Penobscot system by toxicologists and comparison to the scientific literature on toxic effects, 3. Geographical patterns of the distribution of Hg within the Penobscot system, especially in relation to the HoltraChem plant site, and 4. Comparisons of Hg concentrations in the Penobscot to other known uncontaminated and contaminated sites.

We have found that there is conclusive evidence of contamination of the Penobscot River and estuary with Hg. The distribution of Hg in the lower Penobscot River and upper estuary is consistent with releases of Hg from the HoltraChem site, but other sources in the lower river may have also contributed. This will be investigated as part of Phase II of the Study. Although not all data from sampling in 2006 and 2007 were able to be included in this report, the weight of evidence is sufficient to make the conclusion that the Penobscot system is Hg contaminated and we therefore recommend that the Study proceed to Phase II. The overall conclusion and recommendation will not change with the addition of further data. The data that satisfy our four stated criteria are as follows:

1. Agency guidelines: Concentrations of Hg in sediments in the lower Penobscot River and upper estuary were found to be higher than accepted levels of concern for toxic effects to animals living in the sediments using NOAA guidelines. Many lobsters sampled in the upper Penobscot estuary had Hg concentrations that were higher than the USEPA's fish and shellfish tissue criterion for MeHg for the protection of human

health and the comparable criterion derived by Maine. Average Hg concentrations in tomcod in the lower river approached or exceeded the Maine criterion.

2. Toxic concentrations in biota: Hg levels in songbirds sampled in wetlands adjacent to the lower river (in the area of contamination of mercury) were higher than concentrations known to cause reproductive effects in related species. Hg in cormorant eggs in the upper estuary approached or exceeded levels thought to impair reproduction in other birds species.

3. Geographical patterns: The pattern of contamination of the sediments and wetland soils of the Penobscot River and estuary was not consistent with contamination from paper mills on the river but was consistent with large inputs of Hg from the Holtrachem site directly into the Penobscot River. Sediments and wetland soils were much higher in Hg downstream of the Veazie dam, within the tidal influence of the HoltraChem site. Hg in sediments was higher in the upper estuary, closer to HoltraChem, and lower in the estuary, further away from HoltraChem. The patterns of contamination of various species of biota, such as mussels, lobsters, periwinkles, tomcod (fish) and cormorants (birds) were also consistent with large inputs of mercury from the HoltraChem site. Hg concentrations in these species were to be higher in the river or the upper estuary (closer to the mouth of the river) than lower down in the estuary.

4. Comparison of Hg in the Penobscot with other systems: Concentrations of Hg in sediments in the lower river and upper estuary were found to be as high as other sites known to be contaminated with Hg from chlor-alkali plants and other industrial facilities. Hg in mussels and cormorants were high relative to other sites in Maine. Hg in songbirds in wetlands adjacent to the Lower Penobscot River was much higher than at other sites in Maine.

We therefore recommend that the Study needs to proceed to Phase II to examine the dynamics and toxicity of mercury in the Penobscot River and estuary with a focus on the possible mitigation of mercury in the river and estuary, including an attempt to estimate the rate of natural attenuation of this problem.

The results of the first two years of study will be critical to help guide the studies in Phase II, and indeed, many of the studies conducted were designed to lay the groundwork to determining possible mitigation options for the Penobscot system. For example, in 2007, the estuary of the Penobscot was sampled for surface sediments to determine the extent of Hg contamination in the estuary. The results from sediment sampling at nearshore stations in 2006 suggest that the most severe contamination does not extend further south than Fort Point Cove or Sears Island, and sampling of

offshore sediments and wetlands adjacent to the river and estuary in 2007 supports this conclusion. Thus, the geographical extent of the contamination and the areas that need to be included in further work are now known.

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APPENDICES

Appendix 1. Data for total mercury in offshore sediments in the Penobscot River estuary. Concentrations are given as ng/g d.w. All samples were taken in August, 2006. Sampling locations shown on Figure 6. Transects 1-5 are in the Penobscot estuary, ranging from near Fort Point Cove (Transect 1) to Vinalhaven Island (Transect 5). Transect 8 is in the St. George River estuary.

	n	mean	range	Stand. Dev.
Transect 1	5	526.8	278-672	163.2
Transect 2	8	256.1	192-321	46.4
Transect 3	10	137.8	80.5-213	41.3
Transect 4	13	87.9	43.6-115	18.7
Transect 5	12	50.0	14.2-89.5	20.5
Transect 8	5	51.5	45.9-55.3	3.8

Appendix 2. Data for total mercury in bottom sediments in the Penobscot River estuary, normalized for organic carbon content of the sediments. Concentrations are given as ng/g org. C. All samples were taken in August, 2006. Sampling locations shown on Figure 6. Transects 1-5 are in the Penobscot estuary, ranging from near Fort Point Cove (Transect 1) to Vinalhaven Island (Transect 5). Transect 8 is in the St. George River estuary.

	N	mean	range	Stand. Dev.
Transect 1	5	35.7	18.5-45.1	10.5
Transect 2	8	38.0	29.1-45.9	6.0
Transect 3	10	24.5	11.7-44.4	9.2
Transect 4	13	14.5	7.0-21.0	3.7
Transect 5	12	6.6	0.4-11.9	3.4
Transect 8	5	6.1	3.8-7.7	1.5

Appendix 3. Geographic coordinates of wetlands surveyed and sampled, August 2007.

Wetland #	North latitude (deg, min, sec or digital degrees)	West longitude (deg, min, sec or digital degrees)	Sampled?	Isotope samples taken?
W05	44 47 08.4	68 46 18.3	Yes	Yes
W07	44 44 13.8	68 49 40.4	Yes	No
W10	44 41 52.6	68 50 16.7	Yes	No
W11	44.68991	68.82006	Yes	No
W12	44 40 33	68 48 59.7	No	No
W13	44.66850	68.81994	Yes	No
W14	44.65855	68.82944	Yes	No
W15	44 37 46 5	68 50 52.8	No	No
W16	44 37 21	68 50 30	No	No
W17	44 36 56.3	68 51 25.6	Yes	No
W19	44 35 46	68 51 30	No	No
W21	44 34 51.7	68 51 20.2	Yes	Yes
W22	44 33 57	68 51 23	Yes	No
W23	44 33 21.0	68 51 27.6	Yes	No
W25	44 33 40.3	68 45 58	Yes	No
W26	44 33 47.4	68 44 46.8	Yes	Yes
W27	44 33 34	68 44 45	No	No
W28	44 34 12.0	68 44 46.8	Yes	No
W30	44 30 39.4	68 48 33.8	No	No
W31	44 29 53.9	68 48 49.7	Yes	Yes
W32	44 29 43.2	68 49 35.8	No	No
W33	44 28 37.0	68 50 48.4	Yes	No
W34	44 29 01.3	68 51 00.8	Yes	No
W36	44 27 12.7	68 52 23.7	Yes	No
W42	44 23 48.2	68 48 14.5	Yes	No
W43	44 24 32.0	68 46 20.1	No	No
W44	44 25 37.9	68 46 01.4	Yes	No
W46	44 26 17.1	68 44 37.3	No	No
W47	44 27 38.0	68 43 40.9	No	No
W52	44 27 09.8	68 42 05.9	No	No
W54	44 22 51.4	68 45 49.8	Yes	No
W55	44 21 52.0	68 45 35.9	Yes	No
W56	44 22 14.2	68 47 08.3	Yes	No
W58	44 22 00.6	68 48 44.8	Yes	No
W59	44.41602	68.81985	Yes	Yes
W60	44 27 30.6	68 47 06.4	Yes	Yes
W61	44 30 20.3	68 46 20.9	Yes	No
W62	44 35 37	68 51 32	Yes	No
W63	44 42 31.6	68 50 19.7	Yes	No
W64	44 41 32	68 49 57	No	No

Appendix 4. Data for total mercury and methyl mercury in freshwater snails, Penobscot River, Sampling Periods I (late July/early August, 2006), III (late September/early October 2006) and IV (late October/early November, 2006). EB=East Branch; OV=Old Town-Veazie; BO=Brewer-Orrington; OB=Orrington-Bucksport. Where two numbers are given for n, the first is for THg and the second is for MeHg.

Sampling reach	n	Mean THg ng/g d.w.	s.d.	Range	Mean MeHg ng/g d.w.	s.d.	Range
EB	66	762.8	379.3	148-2190	305.5	144.5	80-712
OV	61, 60	376.0	391.3	101-1770	133.1	126.0	0-526
BO	174	304.0	85.4	81-565	123.2	41.8	0-312
OB	69, 59	336.0	98.1	140-617	165.6	75.9	33-437

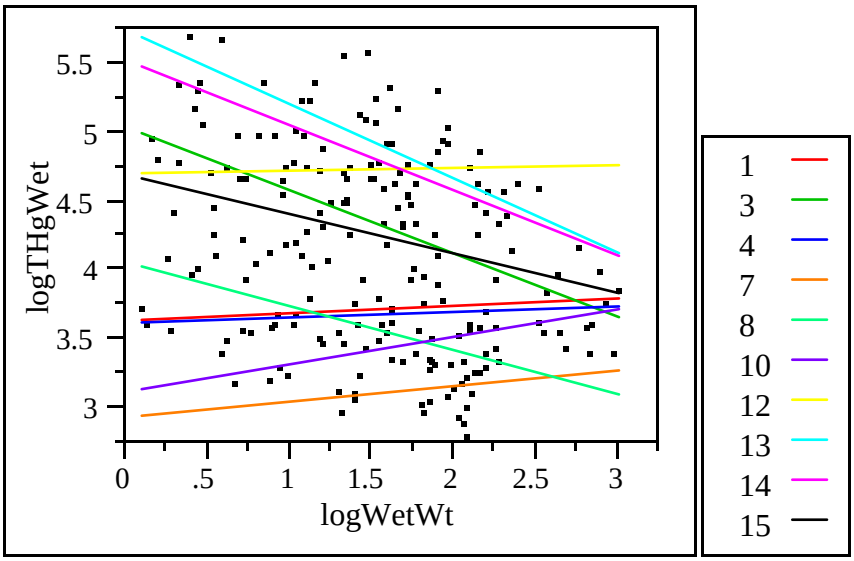
Appendix 5. Data for total mercury and methyl mercury in freshwater snails, Penobscot River, Sampling Period II (early September, 2006). EB=East Branch; OV=Old Town-Veazie; BO=Brewer-Orrington; OB=Orrington-Bucksport.

Sampling reach	n	Mean THg ng/g d.w.	s.d.	Range	Mean MeHg ng/g d.w.	s.d.	Range
EB	26	922.2	328.0	463-1550	375.8	153.0	117-699
OV	9	198.1	28.3	135-238	102.8	27.4	67-139
BO	50	632.2	952.5	275-6520	193.4	163.7	79-1060
OB	10	445.9	62.9	359-546	148.4	46.0	79-211

Appendix 6. Analysis of variance table for total mercury data in periwinkles, Penobscot estuary, 2006.

Source	Sum of Squares	Degrees of freedom	Mean Square	F-Ratio	P
Distance from HoltraChem	76.257	10	7.626	59.435	0.000
Snail weight	9.826	1	9.826	76.584	0.000
Sampling period	0.109	1	0.109	0.849	0.357
% water	3.716	1	3.716	28.960	0.000
Error	58.378	455	0.128		

Appendix 7. Plot of log total mercury vs individual animal weight for mussels from the Penobscot estuary, 2006. Data was analyzed by analysis of covariance with an interaction term to partition variation related to sampling site and mussel size. Sampling sites are ES-1, ES-3, ES-4, ES-7, ES-8, ES-10, ES-12, ES-13, ES-14 and ES-15. Data from both sampling periods were combined for this analysis.



Appendix 8. Analysis of covariance table for total mercury concentrations in mussels sampled in the Penobscot estuary in 2006. Source of variation, number of parameters, degrees of freedom, sums of square, F-ratios and probability of a greater F ratio are shown.

Effect Tests

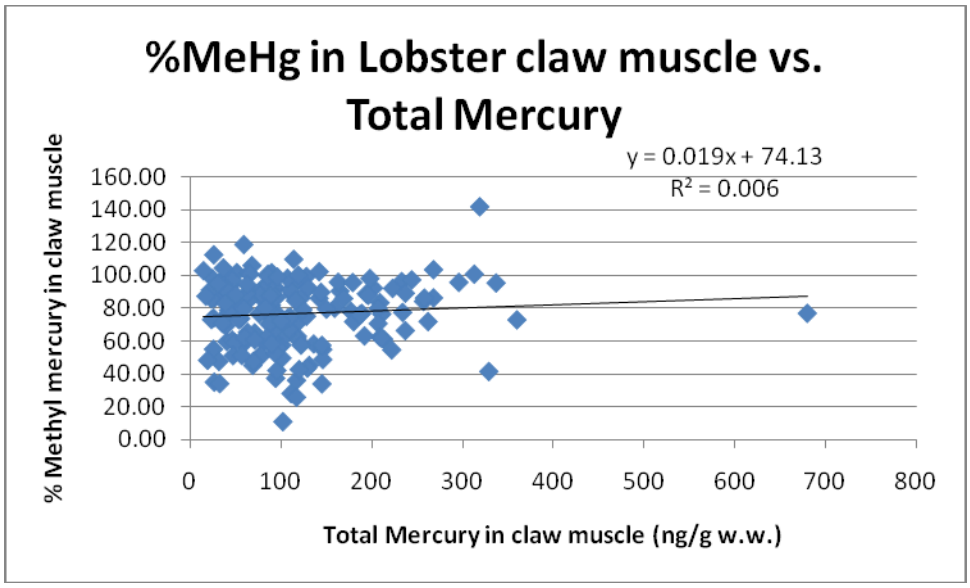
Source	Nparm	DF	Sum of Squares	F Ratio	Prob > F
logWetWt	1	1	0.919818	9.9310	0.0019
site	9	9	52.491125	62.9703	<.0001
site*logWetWt	9	9	3.525334	4.2291	<.0001

Appendix 9. Sampling dates, sites, number of animals sampled, and site locations for lobsters sampled in the Penobscot estuary, 2006.

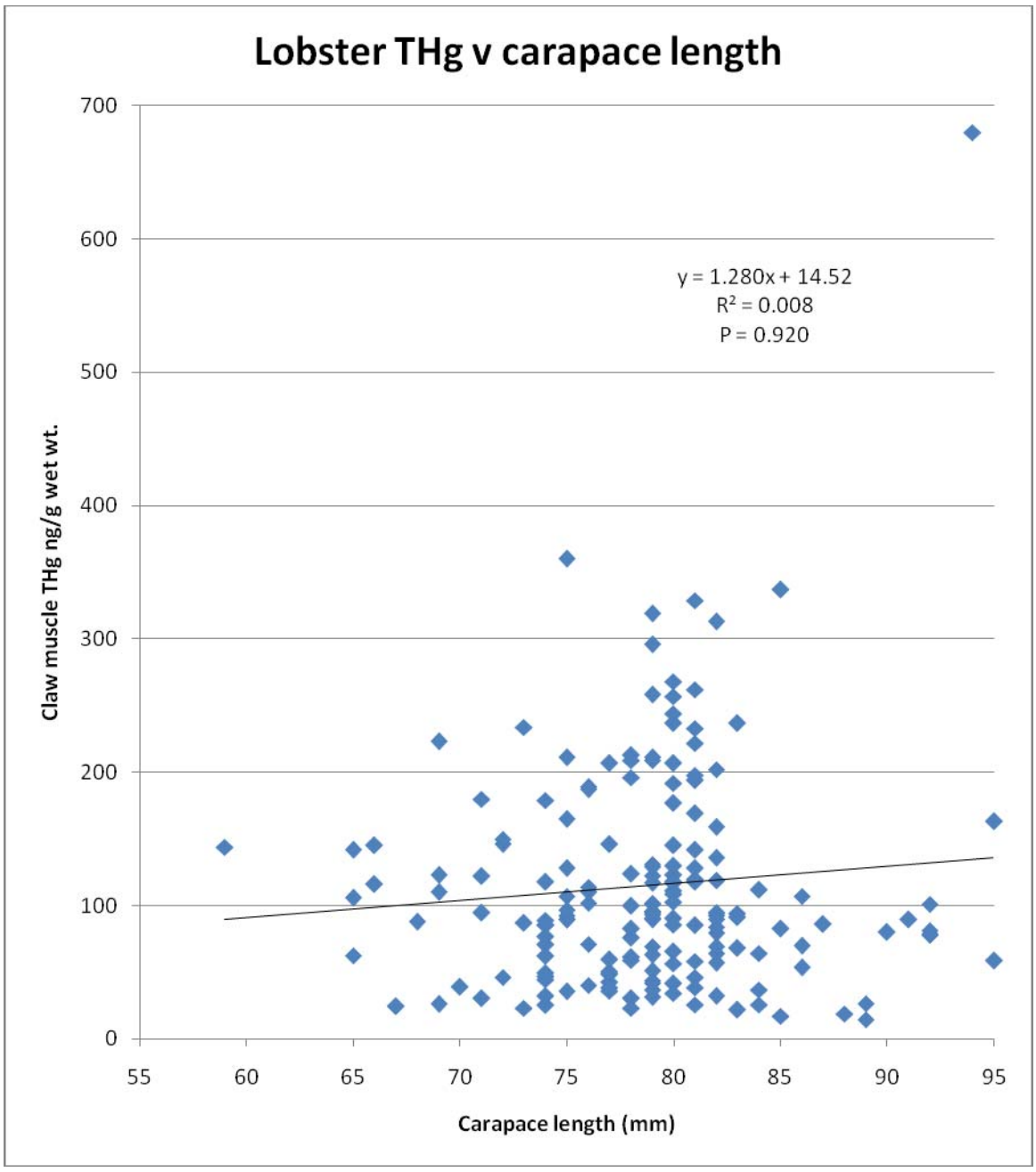
Site	Date	# animals analyzed for mercury	Latitude of site	Longitude of site
1 1	Sept. 6, 2006	2	44.41955	68.86478
1 2	Sept. 6, 2006	1	44.41567	68.86798
1 3	Sept. 6, 2006	5	44.41258	68.87208
1 4	Sept. 6, 2006	17	44.40027	68.88598
1 5	Sept. 6, 2006	7	44.39043	68.89978
1 6	Sept. 6, 2006	10	44.36912	68.91033
1 7	Sept. 6, 2006	1	44.38640	68.89823
1 8	Sept. 6, 2006	6	44.39285	68.88505
1 9	Sept. 6, 2006	10	44.41517	68.82377
1 10	Sept. 6, 2006	9	44.43493	68.82038
1 11	Sept. 6, 2006	4	44.44635	68.81530
2 1	Sept. 12, 2006	9	44.43297	68.82115
2 2	Sept. 12, 2006	13	44.44785	68.81020
2 3	Sept. 12, 2006	5	44.46685	68.79708
2 4	Sept. 12, 2006	7	44.43460	68.81421
2 5	Sept. 12, 2006	10	44.37243	68.81908
2 6	Sept. 12, 2006	20	44.33372	68.83472
2 7	Sept. 12, 2006	3	44.40982	68.87735
3 1	Sept. 29, 2006	10	44.44668	68.81098
3 2	Sept. 29, 2006	10	44.35613	68.83238
3 3	Sept. 29, 2006	10	44.38342	68.90200
3 4	Sept. 29, 2006	10	44.41665	68.86485

Appendix 10. Means, ranges, and standard deviations of total mercury and methyl mercury in lobsters sampled in the Penobscot estuary, 2006. See Appendix 9 for sample sizes and site locations.

Site	Mean Total Mercury ng/g w.w.	Range Total Mercury ng/g w.w.	Standard deviation Total Mercury ng/g w.w.	Mean Methyl Mercury ng/g w.w.	Range Methyl Mercury ng/g w.w.	Standard Deviation Methyl Mercury ng/g w.w.
1 1	132.00	122-142	14.14	113.50	105-122	12.02
1 2	25.80	n/a	n/a	29.00	n/a	n/a
1 3	210.66	43-680	268.45	161.00	33.0-522	206.41
1 4	96.90	17.2-257	71.05	67.94	14.0-216	56.84
1 5	94.43	30.6-262	80.61	64.00	29.0-188	57.25
1 6	126.90	59.1-329	77.72	69.80	40.0-136	29.36
1 7	120.00	n/a	n/a	51.00	n/a	n/a
1 8	107.75	69.4-209	51.93	65.50	33-130	39.97
1 9	154.64	82.6-234	55.77	127.50	62-181	47.54
1 10	137.62	65.6-237	60.14	117.11	58-161	39.70
1 11	122.35	34.4-180	62.19	85.50	25-129	52.11
2 1	154.87	26.4-319	106.63	144.25	11-452	145.24
2 2	171.23	68.0-360	101.75	148.54	62-321	90.10
2 3	178.92	46.4-296	120.87	176.20	45-283	118.84
2 4	103.81	40.3-165	49.32	75.86	24-149	43.31
2 5	70.55	23.3-128	39.53	50.10	17-96	25.44
2 6	94.91	31.6-222	48.00	60.80	11-121	32.09
2 7	84.75	38.1-130	38.23	70.25	35-120	40.72
3 1	159.16	86.0-313	74.46	151.20	81-315	71.66
3 2	80.02	19.1-146	35.40	47.02	9.2-98	24.39
3 3	82.46	22.0-198	49.95	79.60	20-194	49.96
3 4	45.57	14.6-86.8	24.65	38.90	15-68	18.41



Appendix 11. Percent methyl mercury in lobster claw muscle vs total mercury in claw muscle. Relationship is not statistically significant. MeHg values above 100% are due to the combined analytical errors for total Hg and MeHg.



Appendix 12. Relationship between total mercury in claw muscle and lobster size (carapace length) for all lobsters sampled at all sites in the Penobscot estuary in 2006.

Appendix 13. Mercury in tomcod sampled in the lower Penobscot River and upper estuary, September 7 to October 7, 2006. Adjusted mean mercury adjusted by linear interpolation of plots of mercury vs. fish total length to 140 mm. Data from all BO sites combined due to small sample sizes.

	Mean Total mercury ng/g w.w.	Number of samples	Range of total mercury	Standard deviation	Adjusted mean total mercury
BO	273.8	12	158-341	58.03	237.6
OB-1	160.6	10	103-334	64.38	157.6
OB-2	181.2	10	95-275	56.98	172.9
OB-3	217.1	10	130-416	90.72	185.7
OB4	172.5	8	108-281	39.67	179.3
OB-5	196.6	10	81-372	83.68	185.2
ES-02	147.6	17	46.8-288	70.79	125.8
ES-05	114.3	5	92.2-141	17.92	118.3
ES-05/06	128.6	30	68.4-235	40.75	131.2
ES-06	133.2	5	78.1-256	70.38	144.8
ES-09	121.3	20	65.5-226	38.25	125.4
ES-11	137.0	18	56.2-296	61.12	133.9
ES-13	113.3	7	85.4-139.5	16.89	104.3

Appendix 14. Sample locations, sampling dates, and mercury data for double-crested cormorant eggs, 2006.

Sampling site	Sample date	Coordinates of site	Number of eggs sampled	Mean Mercury (ug/g w.w.)	Standard deviation	range
Robinson Rock	July 13, 2006	44.160446 68.978167	10	0.192	0.0896	0.108-0.365
Castine	August 16, 2006	44.381362 68.797649	1	0.487	n/a	n/a
Thrumcap Island	July 13, 2006	44.320980 68.758180	10	0.362	0.1139	0.139-0.510
E. Goose Rock	July 13, 2006	44.183447 68.979321	9	0.342	0.0803	0.179-0.442
E. Goose Rock	July 23, 2006	44.183447 68.979321	8	0.341	0.0353	0.297-0.388
Fort Point	July 13, 2006	44.461089 68.810042	2	0.884	0.0096	0.869-0.891

Appendix 15. Sampling information for wetland songbirds, Penobscot, 2006. Species sampled, location, dates, geographic coordinates of sites, and number of birds sampled are given.

Species	Location	Dates	Geographic coordinates	Number of birds sampled
Nelson's sharp-tailed sparrow	Mendall Marsh	July 20-21	44.58289 N 68.86156 W	10
Nelson's sharp-tailed sparrow	Mendall Marsh	August 9	44.58289 N 68.86156 W	8
Nelson's sharp-tailed sparrow	Prospect Marsh	July 20	44.55651 N 68.85770 W	3
Swamp sparrow	Mendall Marsh	July 21 and August 9	44.58289 N 68.86156 W	3
Swamp sparrow	Winterport	August 8	44.63204 N 68.84745 W	4
Song sparrow	Mendall Marsh	July 20-24	44.58289 N 68.86156 W	4
Song sparrow	Mendall Marsh	August 9	44.58289 N 68.86156 W	10
Song sparrow	Winterport	August 8	44.63204 N 68.84745 W	3

Appendix 16. Mercury data for wetland songbirds, Penobscot, 2006.
Species/location/date (see Appendix 15), number of samples, mean blood total mercury (µg/g w.w.), range of concentrations, and s.d. are given for each sample.

Species	Location	Dates	Mean Mercury concentrations (µg/g w.w.)	Range	s.d.
Nelson's sharp-tailed sparrow	Mendall Marsh	July 20-21	5.72	3.63-8.11	1.382
Nelson's sharp-tailed sparrow	Mendall Marsh	August 9	4.17	2.09-5.72	1.281
Nelson's sharp-tailed sparrow	Prospect Marsh	July 20	4.24	3.63-4.62	0.532
Swamp sparrow	Mendall Marsh	July 21 and August 9	1.62	1.06-2.04	0.503
Swamp sparrow	Winterport	August 8	0.73	0.36-1.38	0.474
Song sparrow	Mendall Marsh	July 20-24	1.93	0.67-2.67	0.955
Song sparrow	Mendall Marsh	August 9	0.55	0.04-1.88	0.575
Song sparrow	Winterport	August 8	0.35	0.03-0.59	0.289

Appendix 17. Comparisons of concentrations of mercury observed in the tissues of mink in the Penobscot River Mercury Study with other areas in North America. All concentrations given as µg/g w.w.

Location	Sample size	Mean in muscle	Range in muscle	Mean in brain	Range in brain	Mean in liver	Range in liver	Mean in fur	Range in fur	Reference
Connecticut	8						1.1 - 8.5			Major and Carr 1991
Massachusetts	4						0.01 - 1.9			Major and Carr 1991
New York	60						0.25 - 7.66			Foley et al. 1988
Ohio	n/a					0.1				Lynch 1973
Ontario	94		0.01 - 4.1		0.3 - 0.7		0.01 - 7.5			Wren et al. 1987
Ontario	39-316			0.96		3.71		30.1		Fortin et al. 2001
Quebec	n/a	1.9		0.8		9.2				Desai-Greenway and Price 1976
Quebec	n/a	2.4	0.41 - 6.2			8.3	2.2 - 20.0			Langis et al. 1999
Saskatchewan	1					58.2				Wobeser and Swift 1976
Tennessee	1							104		Stevens et al. 1997
Wisconsin	39	1.3		0.5		2.1		7.6		Sheffy and St.Amant 1982
Maine	92			0.55	0.1 - 2.6	1.64	0.1 - 8.0	20.92	1.8 - 68.5	Yates et al. 2004
Penobscot Reference	15 - 17	1.0	0.3 - 2.1	0.46	0.2 - 1.2	2.9	0.6 - 18.4	20.4	11.4 - 33.3	Penobscot River Mercury Study
Penobscot Contaminated	3	1.7	0.8 - 2.8	0.79	0.3 - 1.4	2.2	0.8 - 3.7	29.9	14.5 - 56.9	Penobscot River Mercury Study

Appendix 18. Comparisons of concentrations of mercury observed in the tissues of river otter in the Penobscot River Mercury Study with other areas in North America and Europe. All concentrations given as µg/g w.w.

Area	Sample sizes	Mean in muscle	Range in muscle	Mean in brain	Range in brain	Mean in liver	Range in liver	Mean in fur	Range in fur	Reference
Britain	7						0.2 - 4.3			Mason 1988
Denmark	69						0.03 - 12.4			Mason and Madsen 1992
Georgia	n/a	4.4c, 1.5i				7.5c		24.3c, 15.2i		Halbrook et al. 1994
Ireland	32						0.15 - 17.03			Mason and Sullivan 1993
Manitoba	38				0.04 - 9.5		1.3 - 21.7			Kucera 1983
Mass.	96					1.9	0.5 - 4.8			Organ 1989
New York	34						0.01 - 6.95			Foley et al. 1988
Nova Scotia	23				0.07 - 1.8c, 0.5 - 10.2i					Burgess et al. 2002
Ontario[1]	1	36		30		96		47		Wren 1985
Ontario	n/a	0.9				2.9				Wren et al. 1980
Ontario	84		0.1 - 4.3		0.2 - 7.2		0.2 - 17.4			Wren et al. 1986
Ontario	n/a						1.0 - 3.5			Wren and Stokes 1988
Ontario	130			2		6.7		13.8		Mierle et al. 2000
Vermont	21							13.58	4.91 - 46.5	BRI unpubl. Data
Wisconsin	49	1.4		0.7		3.3		6.5		Sheffy and St.Amant 1982
Maine	69			0.55	0.06 - 3.25	1.76	0.24 - 8.66	25.9	1.1 - 234	BRI unpubl. data
Penobscot (Reference)	10-12	0.9	n/a	0.5	0.38 - 0.64	1.9	1.10 - 3.00	23.2	17.0 - 31.9	Penobscot River Mercury Study
Penobscot (Contaminated)	1-9	0.9	0.35 - 1.49	0.28	0.14 - 0.42	1.1	0.56 - 1.61	15.3	8.1 - 21.7	Penobscot River Mercury Study

c= coastal, i=inland, 1 = single individual found near former chlor-alkali plant, apparently dead due to mercury exposure

Appendix 19. Sample site locations, number of analyses per sample, deviations from 202/198 isotope ratios and standard deviations for samples taken for mercury stable isotope ratios. Samples taken October 4 and 5, 2006.

Sample	Geographic location	Number of replicate analyses	$\Delta^{202}Hg/^{198}Hg$ ratio, corrected	Standard deviation
Orrington site 1	44.74022 68.82731	4	0.790	0.051
Orrington site 2	44.74083 68.82675	4	1.112	0.052
Orrington site 3	44.73865 68.82712	4	0.020	0.062
East Branch, Penobscot River	45.64168 68.54623	3	-1.395	0.045
St. George River	44.23760 69.27882	3	-1.757	0.143
Penobscot River @ Eddington	44.84372 68.69552	4	-0.864	0.123