Penobscot River Mercury Study

Phase II Study Plan

Submitted to:
Judge Gene Carter
U.S. District Court (District of Maine)
Portland, Maine

by

J.W.M. Rudd
President
R&K Research Limited
British Columbia, Canada
Chair, Penobscot River Mercury Study Panel

N.S. Fisher
Distinguished Professor
School of Marine and Atmospheric Sciences
Stony Brook University
Stony Brook, NY
Member, Penobscot River Mercury Study Panel

C.G. Whipple
Principal
Environ International Corporation
Emeryville, CA
Member, Penobscot River Mercury Study Panel

R.A. Bodaly
Project Leader
Penobscot River Mercury Study

May, 2008

Introduction

The primary objective of Phase I of the Penobscot River 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 manufacturing site at Orrington, ME.

Phase I of the study defined four criteria for determining whether mercury in the Penobscot system is likely to harm human health or wildlife. These criteria were:

- Do concentrations of mercury in the lower Penobscot River and Estuary exceed agency guidelines (NOAA, MDEP, EPA)?

- Are biota being harmed at current concentrations, based on analyses by internationally recognized toxicologists, and by comparison to published studies in the scientific literature?

- Do geographical patterns in water, sediments, and biota suggest that elevated mercury concentrations are associated with the HoltraChem site?

- How do the mercury concentrations in the Penobscot compare with known uncontaminated and contaminated freshwater and estuarine sites?

As described below, Phase I has demonstrated that mercury concentrations in sediments were high in comparison to NOAA toxicity guidelines, that mercury concentrations in certain biota are likely at toxic levels according to recognized toxicologists, that there is a strong geographical pattern suggesting that HoltraChem is the source of the mercury, and that mercury concentrations in certain areas of the ecosystem are as high or higher than other mercury contaminated ecosystems. While not all of the Phase I measurements indicated mercury contamination, for example, water and snails did not indicate concentrations or patterns suggestive of contamination from Holtrachem, the overall pattern of mercury does indicate that, based on the above four criteria, that harmful levels of mercury are present in the Penobscot system downstream of the plant.

Phase I Summary

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 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 mercury in their past operations and could be sources of mercury to the river. The reaches were also chosen in relation to the extent of tidal surges in the river that could have moved mercury 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 near-shore sites within each river reach and at 15 sites in the estuary of varying distances from the HoltraChem site.

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 mercury 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 total mercury in these bottom sediments were compared to those in a reference estuary (St. George River) which has no known point source of industrial mercury contamination.

Clear evidence for mercury contamination of the lower Penobscot River and upper estuary was found in suspended particles and in sediments of the Penobscot system. Mercury dissolved in water was not found to be elevated in the lower river and estuary as compared to reaches above the Veazie Dam. Total mercury attached to particles suspended in the water was found to be about two times 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 mercury relative to the upper reaches of the river. Total mercury (on a dry weight basis) 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 mercury, total mercury 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)[1] and in the upper estuary. Total mercury concentrations in the sediments of the lower Penobscot River and upper estuary were also found to be about twenty times that in sediments from the neighboring St. George estuary, which has no known history of point source mercury contamination. These results indicate that whereas the paper mills in the Penobscot have elevated mercury in the upper river to some degree, there has been a much larger mercury source or sources downstream of the Veazie Dam, consistent with a large source from the HoltraChem site.

The high concentrations of mercury in the sediments of the lower Penobscot River and upper estuary are similar to or higher than other contaminated sites in N. America and Europe. Perhaps most significantly, these concentrations are higher than NOAA guidelines for toxic effects on aquatic life.

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[1] The HoltraChem site is located below both the Veazie Dam and the Brewer mill. However, HoltraChem mercury impacts this upstream reach because of upstream tidal movement.

The concentration of mercury in inshore sediments of the Penobscot estuary decreased with increasing distance from the mouth of the river. Mercury in the offshore sediments of the Penobscot estuary was higher than in the inshore sediments. The offshore sediments were also 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.

Total mercury 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 mercury is known, much of the work that will be proposed for Phase II of the study will be confined to these areas of high mercury contamination.

We also assessed the potential use of measurements of the ratio of stable isotopes of mercury to determine the amount and extent of contamination of mercury from the HoltraChem site. We found that the isotope ratios of mercury sampled from the HoltraChem site were significantly different from mercury found outside of the aquatic influence of HoltraChem, indicating that the stable isotope fingerprinting techniques have potential for assessing unambiguously the contribution of mercury from HoltraChem.

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. Mercury in mussels was found to be high compared to other sites in Maine and the United States. Mercury in mussels and periwinkles showed a geographic pattern, being higher closer to the mouth of the Penobscot River. Mercury 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 mercury concentrations with increasing distance between the sampling site and the HoltraChem site.

Mercury 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. Mercury 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. Mercury in cormorant eggs[2] in the upper estuary approached levels thought to impair reproduction (Phase I report). Concentrations of mercury in wetland

[2] Mercury concentrations in cormorant eggs in the Penobscot were high compared to other Maine sites and other sites in N America (Phase I report). The highest in the upper Penobscot was 0.88 ug/g ww, compared to an overall mean of 0.28 at 8 Maine sites (range 0.11-0.45) and 0.28 and 0.26-0.27 at other N American sites. The san Francisco Bay-Delta, another contaminated site was similar to the upper Penobscot (0.17-1.17).

songbirds approached or exceeded levels expected to be toxic to songbirds (Phase I report).

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 mercury 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 mercury in sediments consistently exceed NOAA guidelines for toxicity to benthic fauna. Furthermore, some lobster in the upper estuary exceeded MDEP and USEPA criteria for protection of human health for consumption of MeHg in biota (0.2 and 0.3 µg MeHg per g of tissue, respectively). Twenty five percent of the individual lobsters sampled in the upper estuary exceeded the Maine criterion and 6% exceeded the EPA criterion. In the upper estuary, some mussels approach the Maine DEP guide line for mercury concentration.

With respect to Criterion 2: Mercury levels in some species of songbirds inhabiting wetlands in the lower Penobscot were found to be high enough to be of concern for the health of those species. For example, mercury in the Nelson's sharp-tailed sparrow was much higher than concentrations thought to be toxic by avian toxicologists in a related species. We were also advised by recognized toxicologists that mercury 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 mercury 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 mercury, 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 mercury to the lower Penobscot River below the Veazie dam.

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

Phase II - Penobscot River Mercury Study

1. There are two primary objectives for Phase II of the study.

- **To determine if the process of natural attenuation can reduce concentrations of mercury in the contaminated area of the Penobscot system to acceptable levels within a reasonable timeframe.**

- **To determine if active remediation measures could feasibly accelerate recovery.**

One of the main difficulties in remediation of large contaminated ecosystems such as the Penobscot River and Estuary is simply the large area involved. Within this larger area of contamination, there may also be sites where the mercury is concentrated into discrete locations, and/or where mercury methylation is especially important. These areas could be especially amenable to active mitigation strategies. However, there may, be at least one possibility for actively mitigating whole ecosystem (see section IV, 1). Many of the tasks presented below are aimed at gathering sufficient data to evaluate the practicality of possible active remediation measures, and to identify locations where environmental harm is occurring and where remediation may be feasible. Environmental harm includes both toxicity to biota and the loss of resource use due to mercury concentrations that restrict the use of seafood for human consumption. Particular attention will be placed on locations where environmental harm intersects high MeHg concentrations, e.g., wetland hotspots. We think that such locations are likely the best candidates for remediation. If we identify such locations during Phase II, we may recommend that specific mitigation measures be tested following the completion of Phase II.

For larger geographical areas that are contaminated, active remediation may not be feasible. In this case, it may be that the best resolution is for the system to be left alone to clean itself up by a process of natural attenuation, provided there are no significant ongoing point source inputs of mercury[3]. It is also possible that some combination of all the above approaches would be identified in our final recommendation.

Because MeHg is the most toxic form of mercury to higher animals, and because it bioaccumulates very efficiently to finfish and shellfish, one of the most important goals of Phase II will be to determine which environmental factors are most important in controlling rates of MeHg production, where and when MeHg is produced in the system, and how MeHg is transported and bioaccumulated in the lower river and upper estuary. This will enable us to see if there are any practical ways that this production or bioaccumulation could be reduced by active mitigation measures. Other important goals

[3] Natural attention occurs when cleaner surface sediments are naturally deposited at the sediment-water interface reducing surface sediment mercury concentrations, where mercury methylation occurs – in the Phase I study, surface sediment mercury concentration was shown to be an important factor controlling methylation rates.

will emphasize determining rates of ongoing input of mercury from the HoltraChem site and from other industrial and municipal sources, determining the rate of ongoing natural attenuation, and monitoring mercury concentrations in certain key species.

Specifically we propose:

- To quantify the spatial and temporal patterns of MeHg production in the Penobscot ecosystem, and to determine which environmental parameters control rates of MeHg production in sediments and wetlands.
- To continue an ongoing program of monitoring of mercury concentrations in certain key species, so that improvements can be demonstrated in the future if they occur.
- To estimate the time required for appreciable natural attenuation of mercury in the system.
- To determine ongoing inputs of mercury from the Orrington site and from other industrial and municipal sites on the lower Penobscot River.
- To do initial evaluations of the efficacy of certain active mitigation measures.
- To better characterize the mercury/MeHg levels in estuarine animals that are consumed by humans (lobsters, mussels, and fish).

2. Considerations with respect to remediation:

For areas of the ecosystem where mercury is widely dispersed, it may be that the best solution is for the system to be left alone (except for monitoring) to clean itself up by a process of natural attenuation. This will happen if sediments high in mercury are gradually covered by cleaner sediments. In this case the highest concentrations of mercury in the estuary and wetland would be left permanently buried below the depth of active mercury methylation in sediments or wetland soils. To evaluate the natural attenuation option we need to collect data that will enable us to estimate rates of natural attenuation of mercury in the ecosystem. This will also help us to evaluate the practicality and wisdom of possible site specific mitigation measures.

The U.S. Environmental Protection Agency defines monitored natural attenuation as the

"reliance on natural attenuation processes to achieve site-specific remediation objectives within a time frame that is reasonable compared to that offered by other more active methods. The 'natural attenuation processes' that are at work in such a remediation approach include a variety of physical, chemical, or biological processes that, under favorable conditions, act without human intervention to reduce the mass, toxicity, mobility, volume, or concentration of contaminants in soil or groundwater." In the context of the Penobscot River and estuary, the concentration of mercury and methylmercury in surface sediments, surface wetland soils, and biota are the measures of interest.

In considering the relative merit of active remediation versus natural attenuation, we need to know if natural attenuation is occurring and, if so, how long it will take to reduce the concentrations of mercury to levels typically found in Maine rivers and estuaries. If

we determine that all significant point sources of mercury have been stopped, and if sediment coring and dating indicates that recovery is ongoing, which will result in surface sediment mercury concentrations returning to regional backgrounds of about 100 to 150 ng/g DW within a reasonable time period, then we think that natural attenuation would be a preferred option to more widespread active remediation.[4] This is because active remediation such as dredging has the potential to release mercury that would otherwise be inactive. Other types of active remediation such as altering the flow of water into and out of the wetlands would be less likely to remobilize inactive mercury, but could introduce changes to the ecosystem that may be difficult to anticipate. However, if natural attenuation is unlikely to restore the system except over very long time scales, active remediation, if feasible, may well be a preferable option.

3. Geographical extent of Phase II studies:

During the summer of 2007 we completed a spatial survey of total mercury concentrations in 27 riparian wetlands located in the upper estuary and along the lower river (below the Veazie Dam). We also did a survey of mercury concentrations in upper estuary sediments along 5 transects located between Fort Point Cove and mid Vinalhaven Island (Fig. 1, and see Phase I report).

With one exception, the total mercury concentrations in wetland soils and in the intertidal sediments immediately in front of the wetlands located in the lower river and upper estuary were much higher (approximately 1000 ng/g organic carbon) than in wetlands in the lower estuary and Bagaduce River. The higher contamination in wetlands extended from wetlands below the Veazie dam down to the southerly tip of Verona Island, and also included wetlands in the Orland River area. At the time of writing of this proposal we do not yet have results back from the analytical laboratories on MeHg concentrations in the wetlands, but if they show the same pattern as the total mercury concentrations, Phase II studies of the wetlands will be mostly concentrated on the wetlands upstream of the southern tip of Verona Island.

The survey of estuarine sediments showed that the southerly extent of offshore estuarine sediment contamination is greater than for the wetlands and near-shore intertidal sediments. There is evidence of elevated total mercury concentrations in offshore estuarine sediments as far south as mid Islesboro Island (see Phase I report).

However, for Phase II of the study, the court has ordered that:

> "the conduct of Phase II of the study be restricted to the conditions of the Penobscot River and its environs north of Fort Point Light"

Fort Point light is located at the southerly end of Fort Point Cove, which is south of Verona Island, but north of Islesboro Island

[4] One of the objectives of the work presented in Section II, Task 1 is to determine the ongoing time trend of natural attenuation. Some early data collected during Phase I of the study indicates that this is already underway.

Accordingly, Phase II efforts to understand where and when MeHg is produced and factors that control its production as well as testing of possible mitigation measures will be confined to the area of the ecosystem downstream of the Veazie Dam and as far south as Fort Point Light. If, as the study proceeds, limited specific data needs arise that necessitate us taking a few control samples south of the Fort Point Light where sediment mercury concentrations are lower, to complete an objective[5], then permission will be requested from the court to carry out this limited sampling.

4. Proposed Duration of Phase II

We anticipate beginning the Phase II field work during May of 2008, and anticipate continuing sampling for at least one additional year, with earliest end date being November of 2009. After the field work has been completed, we will require a period of time for completion of sample analyses, for data analyses, and for the writing of the final report by the Project Leader. Our best estimate for the earliest date of completion of the final report is July of 2010.

However we want it to be clearly understood that these are our best time estimates at this time. It is not possible to predict fully the outcome of our studies, and additional work may be proposed, if warranted.

If Phase II of the study can not begin until the summer of 2008, we propose that the field studies would continue until the summer of 2010, so that data can be collected over at least two complete annual cycles. The final report would then be December 2010.

[5] For example, to complete the sediment coring and dating study, which is integral to the study of natural attenuation, we may need to take a few cores in the offshore sediments of the bay south of Fort Point light to complete our understanding of how mercury is being dispersed and buried in the bay sediments.

Proposed Phase II Studies

The tasks outlined below describe the work that the Study Panel and Project Leader foresee, at this time, as being necessary for the completion of Phase II of the study. Inevitably during studies of this type, and especially during this phase of the study, when factors underlying the production and bioaccumulation of MeHg in the Penboscot system are being investigated, additional topics of study are likely to emerge. Thus the work outlined below will be updated in response to data, and should not be considered to be all inclusive. Also, as detailed planning of the work outlined below proceeds with the contractors who will carry out this work, some of the details described in the tasks below will change.

The duration of the tasks presented below are given in Table 1.

I. Studies of ongoing inputs of mercury to the Penobscot River from the HoltraChem site, and other possible sites in the lower Penobscot River – Quantity and Quality

A. Estimates of the quantity of ongoing inputs of mercury to Penobscot River.

Objective:
To determine the ongoing mass flux (quantity) of total mercury and methyl mercury from the HoltraChem site and other point sources into the lower Penobscot River/Estuary.

Rationale:
Even though the HoltraChem plant is now shut down, there is still a possibility of some ongoing discharge of mercury into groundwater and stream flows that have passed through the contaminated HoltraChem site before entering the river. Below we describe why it is important to assess these comparatively small ongoing discharges, which could be supporting present day production of MeHg.

In the Penobscot River system, mercury methylation occurs in recently deposited sediments within a few cm of the sediment-water interface. Any ongoing discharge of mercury from the HoltraChem site would be deposited at the sediment-water interface or on the surface of wetland soils when wetlands are inundated. Because this newly deposited inorganic mercury would be deposited in the same physical location as the zone of mercury methylation, a relatively small amount of ongoing inorganic mercury input to the river may be sustaining ongoing mercury methylation in the ecosystem. This was shown to be the case in Lavaca Bay, TX, where relatively small ongoing mercury discharges (compared to very large, but mostly buried, historic deposits) were sufficient to sustain the present day production of MeHg and the contamination of the fishery. It was found that it was important to stop the ongoing discharges rather than remove the

more contaminated deeper sediments, which were buried deeply enough that they no longer were participating in the contamination problem. There were two reasons that the ongoing discharges were very important: 1) the physical location of newly deposited mercury onto surface sediments at the site of mercury methylation, and 2) the ongoing mercury export from the Lavaca Bay site was in a chemical form ($HgCl_2$)[6] that was very available for uptake and methylation by methylating bacteria in the river (i.e., it was very "bioavailable" to the methylating bacteria), as compared to inorganic mercury that had been in the river for many years, and was not nearly as bioavailable to the methylating bacteria. We think the same processes may be operating in the Penobscot system. Thus we need to quantify how much mercury is entering the river from the HoltraChem site (Part A described here), and we need to determine if the mercury in seeps from the HoltraChem site is in a highly bioavailable form (Part B described below). If this is the case, a small steady input of inorganic mercury from the HoltraChem site, because of its high bioavailability to methylating bacteria, could result in elevated rates of production of MeHg.

In addition to the HoltraChem site, there are several other possible ongoing industrial and municipal point sources of mercury to the lower Penobscot River/Estuary (e.g. the Brewer and Bucksport paper mills, and the Bangor sewage treatment outfall). It is also important to quantify these sources and to compare them directly to the quantity and quality of ongoing mercury discharges from the HoltraChem site. The quantity and quality of mercury in all of these ongoing inputs will be compared to the quantity of mercury accumulating in the surface sediments of the river and bay (see Section II). It is possible that these other sources are now bigger inputs of mercury to the lower river than is the HoltraChem site. If this was the case, stopping remaining ongoing inputs from the HoltraChem site might have a relatively small positive impact.

Tasks:

1. We propose to monitor mass flux of mercury from the HoltraChem site in stream flow during base flow periods and, most importantly, during periods of storm water flow, when most mercury transport is likely to occur. Estimates of the mass export of mercury to the river in stream flow will be obtained by multiplying the volume of stream water flow (m^3/sec) times the concentrations of total and methyl mercury in the stream water (ng/m^3). The volume of stream flow is presently being estimated by weirs installed on site. We propose to take water samples at this weir site, which will be analyzed for concentrations of THg, MeHg, chloride, TSS loads, and the isotopic signature of the THg (see I - 4. below). At other sites large exports of THg have been found to occur during brief intense storms, when personnel are often not on site to take mercury concentration samples. Thus, to ensure that we not miss possible large exports of mercury from the site during high stream flow events, we propose to monitor mercury concentrations continuously in the stream water on a 24 hour basis using

[6] Brine is used in the chlor-alkali process, so chloride concentrations are usually high in the ground water beneath chlor alkali plants. This promotes the formation of mercuric chloride ($MercuryCl_2$) which mobilizes mercury in ground water. Mercuric chloride is also one of the forms of mercury that is most bioavailable to mercury methylating bacteria.

Isco automated samplers installed to sample the stream water as it enters the river.

2. We also propose to study mass export of mercury to the river from ground water seeps. Our previous low-tide samplings of seep water at the cliff face below the HoltraChem site has demonstrated that concentrations of mercury in the seeps are much higher than in the river water, and therefore the mass flux of mercury to the river must be presently occurring. We need to estimate the mass of this flux to determine if this is an important ongoing input of total or MeHg to the river. We propose that the mass of mercury being exported to the river from the site in seeps be estimated using methods developed by RT Geosciences, and/or using methods developed at the Woods Hole Oceanographic Institution

   It was never our intention to produce a ground water model of mercury transport through and from the HoltraChem site. Nor do we propose to estimate mass flux of mercury to the river using a ground water model. Instead, at this time, we propose to monitor flux to the river taking the more direct approaches described above.

3. Using the same approaches as discussed above, we will also investigate the possibility that outfalls or seeps from other sites (e.g. the Brewer and Bucksport mills, Bangor sewage outfall), which may also be contributing importantly to the ongoing inputs of mercury to the river below Veazie Dam.

B. Examination of the quality (bioavailability) of mercury exported from the HoltraChem site to mercury methylating bacteria.

Objective:
To examine the bioavailability (quality) of exported inorganic mercury to mercury methylating bacteria active in the Penobscot River.

For inorganic mercury to be methylated it must first cross the cell membrane of mercury methylating bacteria and enter the cellular cytoplasm. In the natural environment, inorganic mercury is present in several chemical forms, some of these are bioavailable to methylating bacteria and some not. $HgCl_2$, which is the form of mercury found in ground water containing brine solutions, is one of the most bioavailable forms of mercury to methylating bacteria. Because $HgCl_2$ may be one of the important forms of mercury entering the river from the HoltraChem site in either ground water seeps or stream flow, we propose to measure the bioavailability of mercury in seep and stream water to methylating bacteria. We will also determine how this bioavailability changes with time after the mercury mixes with the complex variety of ligands present in the Penobscot River.

Tasks:
Samples of mercury-containing seep water and stream water will be taken from the HoltraChem site and from other significant industrial and municipal sources. Stream water from these same locations will be sampled during base flow and under storm flow conditions. In the laboratory, seep and stream water will be added to microcosms, which will consist of soft river sediments or wetland soils taken from either contaminated or non-contaminated sites[7]. Equal amounts of "old" mercury will be added to control microcosms. This mercury will be obtained form surface sediments and from surface wetland soils. All of the microcosms will be fed organic material in the inflow to maintain microbial activity. Further, to normalize differences in methylating activity in the sediments of each type of microcosm, the inflowing water of each microcosm will also contain isotopically labeled $HgCl_2$, which will be equally bioavailable to all of the microcosms.[8] Increases in methyl mercury concentrations in the sediments or soils over a period of months will be followed, which will be a quantitative estimate of net rates of mercury methylation. These rates of production will be compared to MeHg production rates from mercury downstream of the plant, which has been in the ecosystem for some time. This comparison will give us an understanding of the relative bioavailability to methylating bacteria of inorganic mercury now entering the river from HoltraChem and other sources in the lower river, as compared to mercury which has been in the river for many years, but has been recently remobilized and deposited at sites of methylation, such as wetlands.

During Phase I of the study we found that there was a unique stable isotopic signature for the mercury on the HoltraChem site, which enables us to distinguish HoltraChem mercury from mercury already in the river sediments or wetland soils. During Phase II we propose to use this isotopic signature to help us determine the bioavailability of HoltraChem inorganic mercury to methylators, as compared to other mercury that has been in surface sediments or wetland soils for many years. When the plant-derived inorganic mercury is added to microcosms containing either surface river sediments or wetland soils, the MeHg produced form the HoltraChem mercury will also have a unique isotopic signature. This will make it easier to assay MeHg produced from the added HoltraChem seep or stream water, and determine its relative bioavailability

[7] Microcosms containing uncontaminated sediments or wetland soils are to act as controls that will enable us to differentiate between MeHg production from inorganic mercury present in seep or stream water as compared to production of MeHg from inorganic mercury in that has been present in the contaminated sediments for many years.

[8] Sediments or wetlands soils taken for different locations differ in their potentials for production of MeHg. This is because of differences in the activity of mercury methylating bacteria and / or because of differences in the bioavailability of inorganic mercury to the methylating bacteria. Additions of equal amounts of isotopically labeled $HgCl_2$ to the microcosms, which is 100% bioavailable when added, will act as an internal standard that will enable us to normalize for the differences in potential of methylating activity amongst the microcosms. This normalization will enable us to determine the bioavailability of inorganic mercury added in the seep or stream water.

II. Natural Attenuation of Mercury in the lower Penobscot River and upper estuary.

Objective:
To determine the rate of natural attenuation of mercury contamination in the lower Penobscot River and upper estuary.

Rationale:
If no active mitigation measures are feasible, or if they are not cost effective, then natural attenuation of the mercury contamination in the lower Penobscot River and Estuary may be the only practical approach.

The Phase I study showed that in the Penobscot River/Estuary the rate of production of MeHg by bacterial methylators active in surface sediments and MeHg concentrations in surface sediments are primarily controlled by the concentration of inorganic mercury in surface sediments. Thus it is clear that reducing mercury concentrations in surficial aquatic sediments would improve the MeHg aquatic contamination problem (Fig. 1).

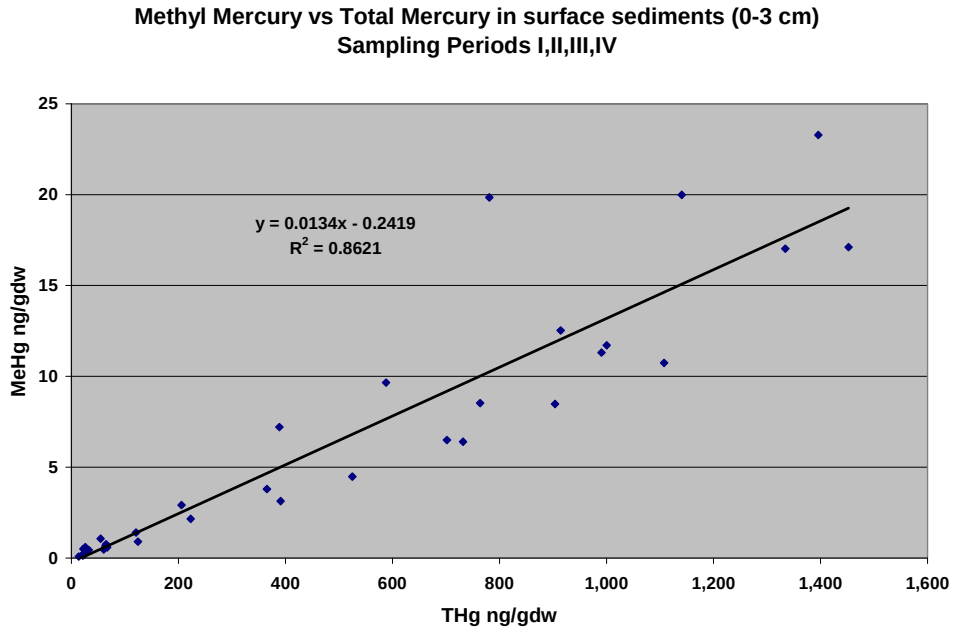

Figure 1. Concentrations of MeHg vs. total mercury in near shore surface sediments of the Penobscot River and Estuary.

The Phase I report also demonstrated that concentrations of mercury in surface sediments were highly correlated with concentrations of mercury in the aquatic food web. Initial deep profiles of THg concentration in river sediments showed that while the surface sediments were still contaminated their concentrations were much lower than in the older deeper sediments, which had been laid down during the time of early plant operation. Thus there has already been some natural attenuation and we need to know whether or

not more attenuation will occur and if so at what rate. To determine how quickly the mercury contamination of the Penboscot River will naturally attenuate it is important to understand how quickly mercury concentrations in surface sediments can be expected to decrease in the future. This rate of decrease of mercury concentrations in surface sediments is the natural mercury attenuation rate.

There are two sources of mercury to the surface sediments: 1) ongoing inputs of mercury to the river that are transported down river and deposited to the surface sediments, and 2) the mixing of historically deposited mercury upward into the surface sediments from deeper more contaminated sediments, in part mediated by bioturbation of benthic organisms. The mercury concentration of surface sediments is determined by the concentration of mercury in sedimenting particles, the mass flux of these particles to the sediment surface, and the mixing depth of the surface sediments. These same factors also determine the long term burial rate of mercury in deeper sediments. When mercury is buried in sediments at depths below the mixed sediments layer (usually greater than 4-5 cm below the sediment-water interface) it is no longer in the zone of mercury methylation, and as long as these sediments remain undisturbed the mercury in the sediments will not significantly contribute to the present day MeHg concentrations in biota.

New mercury containing particles are continuously being deposited at the sediment-water interface. If there are no ongoing point sources of mercury to the river the mercury concentration in the newly deposited particles will be lower than in the older particles already in the surface sediments, and natural attenuation will proceed. If however there are ongoing sources of mercury that maintain the concentrations of mercury in sedimenting particles at elevated levels, sedimentation of these particles will not lower surface sediments mercury concentrations and natural attention will not proceed. This is why it is so important that along with measurement of sedimentation rates (as described in this section of the Study Plan) we also estimate the mass of ongoing inputs of mercury to the lower river from all ongoing sources - including from HoltraChem other possible municipal and industrial sources in the lower river (as described in Section I).

Long term particle deposition and long term burial of mercury occurs at certain sites in the river (e.g., coves, Frankfort Flats) and in the estuary. These long term burial sites will be located as part of Task I (below). The burial rate at these sites (i.e., the sedimentation rate) is determined by the rate of particle transport down the river, which will be estimated as described in Task 2 (below). Particle transport varies inter-annually depending on river flow, and therefore there is inter-annual variability of the sediment accumulation rate. However, average sedimentation rates over many years can be obtained from the dating of cores (Task 1 below), and this average rate can be independently verified by estimates of average long term particle transport rates as described in Task 2 (below). The long term Hg burial rates and particle transport rates are assumed to be constant over time because they are determined by the rate of natural weathering of and transport of particles down river. Thus the historical sedimentation rate can be used to predict natural attenuation rates in the future.

Certain present day depositional sites are temporary, and the mercury that has accumulated at these sites will be transported further down river and to the bay during periods of very high river flow. When the resuspended mercury is deposited downstream onto surface sediments it stimulates mercury methylation Thus the temporary burial of mercury followed by later resuspension slows natural attenuation because it takes more time for the mercury to leave the system entirely (by deep burial in sediments and loss to the open ocean). Some of the coring work described below will determine the location and frequency of these temporary sites. This information will be needed to accurately estimate the natural attenuation of the Penobscot system.

Tasks:

1. Cores will be taken from areas of soft sediments and from wetlands in the lower river and upper estuary. The purpose of the cores is to determine: 1) which sites are long term burial sites and what the rates of mercury burial are at those sites, and 2) to determine which sites are short term burial sites, which may contribute to the ongoing problem if these sediments are remobilized and transported further downstream during periods of high river flow.

   At several depositional sites in the lower river (wetlands and soft river sediments) in coves, at the Frankfort Flats area, and in the estuary we propose to take long (60-70 cm) sediment cores, which will be sliced at 1 cm intervals. Using established methods, the following analyses will be done on each core slice – sediment porosity, x-radiography, and concentrations of organic carbon, total mercury, organic carbon, and radionuclides (^{210}Pb, ^{239,240}Pu ^{137}Cs, ^{7}Be, and if needed ^{239,240}Pu). These radionuclides decay at known rates after they have been deposited in the sediments on particulate material, and so they function as "clocks" that tell how long a particle has been in the sediments. The ^{7}Be isotope decays quickly and is used to determine the mixing depth of the surface sediments. Mercury buried below this mixing depth is buried below the zone of mercury methylation, and so this deeply buried mercury will not contribute to the present day contamination of biota by MeHg. All of these data will be used to estimate natural attenuation rates as described briefly in the above rationale and in more technical detail in Santschi et al. (Environ. Sci. Technol. 1999, 33: 378-391).

   During 2007 we took a few cores to determine if these procedures were likely to be successful. Our initial results were promising, so we now propose to take more cores at various depositional sites in the lower river and upper bay to determine the overall variability of sedimentation rates. Because of this natural variability the number of cores to be taken (maximum 60) will be fairly large. The actual number will be determined in the field during consultations with the contractor carrying out the work.

2. We also propose to determine rates of particle transport down the lower river and to the upper bay. The purpose of estimating particle transport rate is to constrain

the estimate of the pace of long term sedimentation rates in the Penobscot system, as determined in Task I. The rate of particle transport down the river determines the rate of downstream sediment accumulation in long term depositional areas. These two independent estimates (Tasks I & 2) will increase our confidence in estimated time for natural attenuation to occur.

To estimate particle transport we propose to do a mass flux study of suspended particles in the lower river using methods developed for the US Environmental Protection Agency (EPA/600/6-85/002b) by Steve Gherini and co-workers at Tetra Tech.

To apply this approach the following categories of data will be collected on several occasions at different river stages: 1) upstream mass inputs to the lower river; 2) mass transport down the lower river; and 3) delivery of suspended particles and mercury to the bay. These data will enable us to characterize the river under different flow conditions. Then we will be able to predict past (using archived USGS flow data from the Eddington station) and future (using average annual flow rates) particle transport by the river. The hind casting of mass particle flux will be an independent estimate of mass particle transport to the lower river and estuary, which will be compared to the particle deposition rates estimated from the core studies described in Task 1 above. The forecasting information will be useful to us for estimating future natural attenuation rates.

Data collections will include:
1) Upstream mass inputs: On a predetermined basis, and especially during periods of high flow, we will monitor mass inputs of particulates to the lower river by measuring flow volumes at the Veazie dam and in tributaries entering the river below the Veazie Dam and particle concentrations.

2) Transport down the river, and 3) discharge to the upper estuary: At two locations downstream of the Veazie dam (just downstream of HoltraChem and near Winterport), and at one location near the mouth of the river (just downstream of Mendall Marsh), we will measure TSS loads, temperature, salinity, current velocity and direction, as well as dissolved and particulate mercury concentrations. This will enable use to estimate net flux of water and particulates at these points in the tidal portion of the lower river using the EPA method described above.

III. Studies of methyl mercury production and transport in the Penobscot River and Estuary

MeHg is the most toxic chemical form of mercury to higher animals. It is produced from inorganic mercury by bacteria active in wetland soils and aquatic sediments. In the lower river and upper estuary of the Penboscot, MeHg concentrations are high in sediments and in wildlife (see Phase I summary and report). During Phase I of the study we

concentrated on studying MeHg concentration in riverine and estuarine sediments, and we found that a major factor controlling rates of MeHg production and MeHg concentrations in these aquatic sediments was inorganic mercury concentration in the surface of aquatic sediments (Phase I report, Fig. 1).

To date we do not have an analogous data set for MeHg concentrations in wetlands, and therefore we do not yet have the same level of understanding of MeHg production in the wetlands adjoining the shorelines of the lower river and upper estuary as we do for the aquatic sediments. MeHg production by bacteria active in mercury contaminated wetland soils is important to the entire ecosystem for two reasons: 1) Wetland soils are often especially active producers of MeHg, per unit of mercury, which leads to contamination of the food web in wetlands – for example, the very high concentrations of MeHg seen in song birds in Mendall marsh. 2) Some of the MeHg produced in wetlands may be exported to the adjoining river and estuary and result in MeHg contamination of the aquatic food web in the river (e.g. tomcod, cormorants, and osprey). The studies outlined below will further our understanding of MeHg production in both river sediments and wetlands, for the purpose of designing and testing mitigation measures that limit MeHg production.

A. Studies of MeHg production

Objective:
To understand when and where MeHg is being produced in the contaminated areas of the Penobscot ecosystem.

Rationale:
It would be inappropriate to recommend costly active mitigation measures intended to limit mercury methylation without a reasonable understanding of the microbial process that produces MeHg. The tasks proposed below will tell us where and when MeHg production is most active in the ecosystem, for example wetlands soils vs. offshore bay sediments vs. river sediments.

These studies will also give us a better understanding of which environmental parameters, in addition to inorganic mercury concentration, stimulate or inhibit the production of MeHg. This information will enable us to predict the likely success of active mitigation measures that may be proposed in the future. Also, the data produced by these tasks may suggest additional mitigation measures other than the three approaches that are outlined later in Section IV.

Tasks:

We will examine the timing and intensity of MeHg production in different types of wetlands, and factors controlling its production in these wetlands.

1. During our 2007 survey of wetlands we observed at least three basic types of wetlands, based on vegetation type. As early as possible in 2008, to confirm our observations, we propose to employ a wetland ecologist who is familiar with the wetlands of the lower Penboscot. The ecologist will classify the types of wetlands in the lower Penboscot and their distribution and surface area. At this time we anticipate that the primary criteria for characterizing the wetlands will be based on vegetation type. One of the primary factors controlling vegetation community structure in wetlands is hydrology, which in turn controls redox conditions in wetland soils and other biogeochemical factors affecting plant growth. These same factors are also known to stimulate or inhibit rates of MeHg production. Other related parameters used for wetland characterization will be total mercury concentrations, and % MeHg (from the 2007 survey), slope/elevation, frequency of inundation, and potential for the wetland to export MeHg to the river/estuary.

2. After the wetlands have been characterized, 2 or 3 wetlands of each type will be selected for further intensive study. They will be sampled biweekly at least three elevations during the summer field season over a two year period. The top 4 cm of wetlands soils will be taken analyzed for concentrations of THg, MeHg, organic carbon and porosity. For each sampling site the per cent of total mercury that is MeHg (% MeHg) is a good indicator of the recent intensity of mercury methylation at a given site, and the changes in % MeHg with time are a good indicator of seasonal changes in the net rate of MeHg production. This work will give us an understanding of the seasonality of MeHg production the year-to year variability of mercury methylation, and which types of wetlands have the highest rates of MeHg production. For comparison we will also sample some soft off-shore sediments in front of the wetlands.

3. At the same set of selected wetlands described above, on three occasions during a field season, wetland soils will be studied intensively for mercury methylation rates, as well as a detailed analysis of the biogeochemical factors that control these rates of microbial MeHg production. For this task, intact sediment cores will be taken from each site within a wetland. Isotopically labeled inorganic mercury will be injected into the undisturbed cores to assay methylating activity. The cores will then be sliced, and analyses will be done for other chemical parameters (e.g. pH, anions, sulfide, organic carbon, dissolved organic carbon quantity and quality) that are known to stimulate or inhibit rates of microbial mercury methylation.

4. Another factor that could stimulate MeHg production, particularly in Mendall Marsh, where MeHg concentrations are so high in birds, is the atmospheric deposition of reactive gaseous mercury (RGM). RGM deposition tends to be highest at the boundary between marine and continental air masses, which is where Mendall Marsh is located. RGM is an inorganic form of mercury that is very available for microbial mercury methylators. If RGM deposition is important at the Mendall Marsh, this could be one explanation for the very high

MeHg concentrations we found in birds sampled at Mendall Marsh, and we need to understand the possibility of this mercury source before recommending any active mitigation for Mendall marsh. We propose to continuously determine RGM concentrations in air over the Mendall Marsh, and estimate its deposition rate to the marsh over one entire field season using a Tekran mercury analyzer equipped with denuder tubes.

5. We will also study mercury methylation in offshore bay sediments located in Fort Point Cove using the methods described in Task 2. Samples from Fort Point Cove will be taken along the same transect and at the same locations as our 2007 sampling. In addition, sandy sediments have been found to be sites of mercury methylation in Chesapeake Bay, and so sandy sediments in the lower Penboscot and in Fort Point Cove will also be assayed for methylating activity. This work will be done on the same time schedule as for the wetlands. On three occasions the intensive methylation work described in Task 3 will also be done on these Fort Point Cove sediment samples.

B. Mercury transport and mass balance estimates MeHg production in the Penobscot ecosystem

Objective: To determine the relative importance of river sediments and wetlands as sources of MeHg to the river and upper bay.

Rationale:
In addition to understanding MeHg production, it is also important to know how MeHg moves from one place to another in the system. If, for example, the transport of MeHg from wetlands to the river is an important source of MeHg to the food web in the river, then active mitigation of wetland areas would improve the situation in both the wetland areas and in the river. On the other hand, if this transport is not as important a source of MeHg to the river as the river and bay sediments themselves, then mitigation measures that could enhance the rate of natural attenuation of mercury concentrations in the river would be the preferred choice. The tasks described below will help us explore these options.

Tasks:

1. The purposes of this work are to estimate the flux of MeHg from Mendall Marsh itself, and also to use these data to estimate fluxes to the river from other wetlands located in the lower river areas. To do this work, an acoustic doppler radar array will be installed at the water-sediment interface at the outflow of Mendall Marsh. This device will continuously measure net water flow. At the same location, an automatic ISCO sampler will be installed to take high frequency samples in the water column of the outflow of Mendall marsh to determine concentrations of

total and MeHg. The product of the net volume of water flux into and out of Mandel Marsh and the concentration of total or MeHg per volume of water will give us an estimate of mass flux of mercury out of Mendall Marsh and into the river.

Mendall Marsh is by far the largest wetland in the lower Penobscot. A per m^2 rate of MeHg flux from this marsh will be calculated and this areal flux rate will be prorated to other wetland areas along the main stem of the river to give us an estimate of the rate of flux from the total area of wetlands located in the lower river.

There are also extensive wetland areas and mud flat areas in the Orland River. Our 2007 wetland survey showed that these wetland areas are just as mercury contaminated as wetlands in the main stem of the river. We are presently exploring the technical possibility of determining net flux of total and MeHg out of the Orland River and into the main stem of the Penobscot River near its mouth. This would also be done using a second acoustic doppler array.

2. We will also estimate the mass flux of MeHg out of the river sediments to the river water. The mass flux of total and MeHg down the main stem of the lower river and to the bay will be estimated using a mass balance approach by adding measurements of total and MeHg concentrations to the measurements already being made to estimate particle and water volume flow at 3 locations in the main stem of the river (see Section II, task 2). The piggy-backing of the mercury concentrations onto particle flux work offers us a very cost effective opportunity to estimate MeHg and THg flux down the main stem of the river between the Veazie Dam and the mouth of the river just below the confluence of Penobscot and Marsh Rivers.

   The difference between the net flux of mercury at the mouth of the river (at the most southerly measurement point just below the Marsh River (described in Section II, Task 2), and the net flux of MeHg from wetlands (Task 1, above) is an estimate of the flux of MeHg from the sediments of the main stem of the river to the river water.

   In addition to the MeHg that diffuses from the surface sediments to the overlying water, there is also MeHg that is produced and stays in the sediments and is an important source of MeHg for the benthic food chain (e.g., insect emergence, consumption of worms or crustaceans by bottom feeding fishes). We will measure the timing of MeHg production in the surface sediments and estimate the change in total mass with time as described in Task A, 5 of this section.

   The integration of all of these data will tell us when and where MeHg is produced in the ecosystem and the relative importance to the entire ecosystem of MeHg produced in wetlands as compared to river and upper bay sediments. All of this information will be very useful in evaluating the type of active mitigation

strategies that we might recommend in the future. This mass transport work was proposed and approved for Phase I of the study, but the Study Panel and Project Leader decided not to do this work until Phase II.

IV. Preliminary testing of active mitigation measures

Objective:
To carry out preliminary testing of active mitigation measures that may reduce MeHg production in wetlands and/or river and bay sediments.

Rationale:
As we continue to digest the data collected in Phase I, and as we are developing Phase II of the study, our view of active remediation strategies is evolving. This evolution will continue as Phase II data begin to arrive. Our present plan is to use our understanding of methylation gained from the Phase I and Phase II data to test in a preliminary way, at the scale of laboratory experiments, the three possible mitigation strategies that we are now considering. More possible amelioration strategies may become evident during the course of the Phase II studies on factors controlling the production of MeHg in river sediments and wetland soils. If any of these preliminary laboratory tests are positive, we may recommend future field pilot projects to determine whether one of these approaches could be useful at a broader level.

For all of these active mitigation strategies we will investigate possible deleterious effects as well as the possible upside of reducing MeHg production and mercury concentrations in biota.

Tasks:
Three types of controlled experiments will be conducted with intact wetland soils and soft river sediments in lab based microcosms as preliminary tests of active mitigation measures.

1. *Actively increasing the rate of natural attenuation:* We propose to do a preliminary evaluation of an active mitigation method, which if successful would increase the rate of natural attenuation in the entire ecosystem (river and bay sediments as well as the wetlands)[9]. The method is based on actively suspending clean particulate material in the upstream river water and allowing the river flow to transport this clean material to downstream depositional areas in the lower river, bay, and wetlands. The increased rate of sedimentation of the clean material would decrease the mercury concentration in surface sediments and thus

[9] This proposed approach is quite different from traditional capping of sediments, where clean capping materials are applied to cover highly contaminated surface sediments at discrete locations. This approach, if successful, has the potential to treat the entire ecosystem, including wetlands, by actively increasing the pace of natural attenuation by upstream suspension of clean material followed by downstream dispersal and deposition by river currents.

reduce the rate of mercury methylation. (Phase I data demonstrated inorganic mercury concentration in surface sediments is an important driver of microbial mercury methylation (Fig. 1 above)). This whole-ecosystem active mitigation approach was studied and has been recommended for the English-Wabigoon in northwestern Ontario, which was also contaminated by mercury effluents from a chlor-alkali plant (Rudd et al. 1983. Can. J. Fish. Aquat. Sci. 40: 2206-2217). The English-Wabigoon River study also showed that there was more than one effect of suspension and downstream deposition of clean particulates. Not only did the deposition of this clean particulate material reduce the overall mercury concentration in surface sediments, but it also tightly bound the inorganic mercury, making it much less available for uptake and methylation by the methylating bacteria. A practical advantage of this approach is that there is the possibility of enabling us to treat the whole ecosystem (including the wetlands) and not just specific wetland sites. We propose to do some initial exploration of this mitigation approach by determining if clean clay material is present upstream in the Penboscot, which could be resuspended in the river by dredge spoiling, and/or if clean material could be transported from another location to the river at a reasonable cost. We also propose to do some preliminary laboratory investigations of the active mitigation approach using lab microcosms containing river sediments and wetland soils to determine if this approach limits mercury methylation in the Penobscot setting.

2. Lab microcosm experiments will be used to do preliminary testing of the possibility of Sedimite additions at certain Penobscot sites as a means of reducing mercury methylation rates. Sedimite is a manufactured proprietary material. It is primarily activated charcoal to which various ligands have been attached to bind metals – in this case mercury. In practice, Sedimite is applied to a surface of aquatic sediments or wetland soils, and it is mixed into the surface methylating layers by bioturbation. There it binds inorganic mercury making it unavailable to mercury methylating bacteria. Our preliminary testing of Sedimite will be done in laboratory microcosms where Sedimite will be mixed into surficial sediment and wetland soils and MeHg production will be followed with time. It would not be feasible to apply this active mitigation approach to the entire ecosystem, but it may be feasible to apply it to certain high priority locations such as Mendall Marsh. We would also explore the possible detrimental influences of Sedimite on benthic organisms such as restricting availability of other metals that are required for growth, e.g., iron and manganese.

3. *Control of periodic flooding:* Recent research in mercury contaminated wetlands in the San Francisco Bay Delta has shown that bacteria in wetland soils that are permanently inundated produce much less MeHg than bacteria in wetland soils that are periodically flooded and drained. (This is likely because of sulfide formation in the permanently flooded soils that binds the inorganic mercury as mercuric sulfide, making it much less bioavailable to the methylating bacteria.) We propose to do a preliminary test of this idea using Penobscot wetland soils, which are now periodically flooded naturally during high river events. These

initial tests would be conducted in lab based microcosms to determine if methylating bacteria in Penobscot wetland soils that are permanently inundated would produce less MeHg than wetland soils that are periodically drained, aerated and then reflooded. Permanently flooding a wetland such as Mendall Marsh would change the character of the marsh. As with the other possible active mitigation approaches, this possible downside would need to be investigated at a later date. However, a change in the character of the marsh might be a better overall solution than leaving it in its present state, which is putting wetland bird species and possibly other biota at risk of mercury toxicity.

V. Sampling during dredging operations

Objective:
To determine if there is significant mercury or MeHg released to the ecosystem during dredging operations that may be carried out by other parties during the course of Phase II of the study.

Rationale:
During Phase II of the study dredging may occur in Southerly Cove to remove high concentrations of mercury found there, and there may also be some dredging in the lower river to improve navigation and for site-specific environmental clean-up. We will have no control over this dredging, but if it occurs during Phase II of the study the resuspension of mercury during dredging could impact mercury concentrations of our bioindicator organisms (e.g., mussels, tom cod). We now have two years of bioindicator data and we are proposing to collect and additional two years of these data during Phase II of the study. In the future these data will become a valuable baseline of mercury concentrations, which will be used comparatively to detect changes in mercury concentration with time after implementation of active mitigation measures or as natural attenuation of mercury in the system progresses. We therefore propose to monitor mercury concentrations in water during dredging operations. The purpose of this monitoring is to improve our understanding of possible short-term fluctuations in baseline mercury concentrations of our bioindicator organisms that could occur following dredging disturbances, which may release buried mercury into the biologically active water column and surface sediments. These short-term fluctuations, if not understood, could confound our understanding of trends in the baseline bioindicator data and consequently limit our ability to demonstrate the success of active mitigation to alternatively to asses the pace of natural attenuation

We are not proposing at this time the use of dredging as an active mitigation method, and our limited monitoring of mercury resuspension during dredging is not designed to assess dredging as a possible mitigation approach.

Tasks:
If dredging operations occur, we will take water samples at several sites upstream and downstream of the dredging operation. Samples will be analyzed for dissolved and

particulate concentrations of total and MeHg. The sampling will be repeated several times during the dredging operation. These results will be used to evaluate any corresponding changes that might occur in the bioindicator data.

VI. Ongoing monitoring of mercury concentration in resident organisms

It is self-evident that it is necessary to establish tissue concentrations of mercury in resident organisms living in the mercury contaminated areas of the ecosystem. There are two overall goals for this bioaccumulation work:

1. To establish concentrations of mercury in certain indicator organisms that can be followed on the long term to determine if active mitigation measures or natural attention are successfully reducing the tissue concentrations of MeHg.
2. To assess mercury concentrations of certain species for which we do not presently have sufficient data.

Measurements from Phase I of this study have indicated that some organisms clearly display mercury concentrations in excess of those from reference sites. In some cases, for example, song birds and cormorants, these levels appear to be at or near toxic concentrations. Also, mercury concentrations in sediments where benthic organisms live were found to exceed NOAA guidelines for toxicity to these organisms. Furthermore, given the importance of some other organisms, notably mussels and lobsters, for commercial harvesting for human consumption, continued monitoring of mercury levels is warranted, especially since indications are that their levels in the northern part of the estuary are high and in some cases exceed health advisories. Additionally, mercury concentrations in fish samples from Phase I are still being processed. Other organisms need to be sampled more extensively or for the first time, and these would include bats, and possibly seals. Thus, we still need to evaluate mercury levels in some fish species that are eaten as seafood (e.g., eels) and those that may serve as conduits to higher-level predators (e.g., killifish). In this section, we describe tasks that would address the need for continued monitoring of bioindicator organisms, of important commercial species and of wildlife that may be at risk.

Objective:
The objective of this component of the work is to establish more comprehensive and longer term data bases for mercury concentrations in critically important bioindicator species and species for human consumption.

Rationale:
The rationales for conducting this task are: 1) we need to monitor continuously the commercially important species, such as mussels, eels and lobster, as possible conduits of mercury to people since our earlier measurements indicate that methylmercury concentrations approach or exceed health advisory levels (e.g., lobster); 2) in some cases certain species (e.g., mussels, tomcod, songbirds, cormorants) are especially appropriate to use as bioindicator organisms because their tissue concentrations of mercury reflect

ambient mercury concentrations in the ecosystem. Such data will very useful in establishing the efficacy of active mitigation measures or for monitoring long-term natural attenuation that may occur if active mitigation efforts are not implemented. These measurements will establish a baseline describing the "before" situation, which is necessary to compare with measurements from the "after" situation.

Tasks:

1. F*or species consumed by humans:* Continue to measure mercury and methylmercury in lobsters, primarily focusing on claw meat and tail meat, and tomalley in 20% of total samples. Samples from the same individual lobsters should be compared to determine the relative contamination of tail and claw meat by mercury and methylmercury. Lobsters should be sampled sufficiently frequently in sites nearest marshes to account for seasonal production of methylmercury. Eel fillets and mussels taken from the Penobscot are two other species eaten by humans, and their mercury concentrations will be monitored in the same manner as was done for Phase I of the study.

2. *For bioindicator organisms:* Continue to monitor inorganic and methylmercury in the common blue mussel, *Mytilus edulis* in various locations within the estuary. Based on results from Phase I, samples should focus more intensely on regions closest to where the Penobscot River enters the bay, such as around Verona Island. At least two samples per year from each location should be taken, one taken pre-spawning, the other post-spawning, since mussels are known to lose some metals (e.g., silver) very appreciably through spawning. Tomcod and killifish (where possible) should continue to be measured seasonally for mercury and methylmercury. As with lobsters, mercury concentrations should be related to size of the fish. These fish do not travel extensively throughout their lifetimes and so they are suitable as indicators of methylmercury that is available for accumulation in fish for a given region. Because the loss rates of methylmercury from fish are so slow, fish sampling will occur once per season, but the sampling will always be doen at the same time of year to account for any fluctuation in mercury concentration duet growth dilution.

   Certain bird species have proven to be very good bioindicators of exposure of the wetland food web to MeHg. We propose to continue the sampling of wetland bird species started in Phase I to establish baseline mercury concentrations against which any future improvement of mercury concentration can be gauged. Species anticipated to be followed at this time are: Nelson's sharp tailed sparrow, swamp sparrow, song sparrow, rails, and redwing blackbirds. When possible concentrations of mercury in blood and feathers of these bird species will be followed in the same wetlands that are also being intensively studied for MeHg production activity (see section III, A). Spiders are the main source of mercury for some wetland birds, including those most contaminated in the Penobscot system, so we will also gather spiders while sampling the wetland birds. This will enable us to compare MeHg in wetland food chains when birds are not available in a particular wetland.

We also propose to continue our study of mercury concentrations in cormorants. This bird is a good bioindicator species, because it is a fish consumer, has a wide distribution in the lower Penboscot, and is present in high enough numbers to give us statistically meaningful data.

3. Certain organisms were not sampled or were not well sampled during Phase I. During Phase II we propose to ensure that these species are not overlooked. These species include bats, striped bass, bluefish, and possibly seals and amphibians.

4. To better understand the bioaccumulation of MeHg in food webs we need sufficient information on the composition and MeHg content of the diet for those species that we propose to monitor, since MeHg is accumulated almost exclusively through diet. To help us interpret our MeHg concentrations in biota, we will have a literature survey produced of the known dietary relationships in Penobscot wetlands and for the dominant species that we will monitor. Where data are not available for the Penobscot system itself, we propose to apply known dietary relationships for these same species studied in other cool temperate rivers and estuaries.

Table 1. The following is our best estimate of the timing of the tasks outlined for Phase II of the Penobscot River mercury study. These durations could be lengthened or shortened as the data for Phase II are being analyzed. Month 1 of year 1 will be the month that field sampling begins on the Penobscot River.

	Year 1												Year 2												Year 3					
	1	2	3	4	5	6	7	8	9	10	11	12	1	2	3	4	5	6	7	8	9	10	11	12	1	2	3	4	5	6
I, Ongoing inputs																														
A, Mass inputs																														
1, 2 HoltraChem surface water & seeps																														
3, other discharges																														
B, Mercury quality																														
II, Natural Attenuation																														
1, mercury burial rates																														
2, Particle transport																														
III. Methylation																														
A. MeHg production																														
1, wetland characterization.																														
2, 3, wetland methylation																														
4, RGM																														
5, sediment methylation																														
IV. Hg transport & production																														
1, Mendall Marsh																														
2, river transport																														

Table 1 continued

	Year 1												Year 2												Year 3					
	1	2	3	4	5	6	7	8	9	10	11	12	1	2	3	4	5	6	7	8	9	10	11	12	1	2	3	4	5	6
IV. Mitigation testing																														
1, 2,3																														
V. Monitoring of dredging																														
VI. Hg bioaccumulation																														
1, bioindicators																														
2, additional species																														
Data analyses																														
Final report																														