# PENOBSCOT RIVER MERCURY STUDY

# Chapter 3

## Total mercury loading to the Penobscot River from the HoltraChem Plant Site, Orrington, Maine and from other point sources

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# **1 SUMMARY**

This work had two primary objectives: 1) to quantify total mercury (Hg) export from the HoltraChem site to the Penobscot River under current (2009-2010) site conditions and operations, and over a range of surface water and groundwater discharges, and 2) to identify and quantify other industrial and non-industrial point sources of Hg to the Penobscot River downstream of the Veazie Dam and above the town of Bucksport.

An important focus of the HoltraChem site work was to quantify Hg export from the site by continuously monitoring Hg transport in the two streams flowing over the site. Previous studies of Hg export in these streams have been done by discrete time interval sampling, irrespective of flow rate. Our goal was to continuously monitor Hg export, sampling more intensively at times of high flow when transport is likely to be high, e.g., during storm events. Discharge rate and total Hg content of surface water runoff from the site was monitored for slightly longer than one year. During the year we monitored these streams, we were fortunate to be characterizing Hg export during a very large (11.4 cm) precipitation event. Results from this event provided better understanding of the impact of very high stream flow event on Hg transport from the HoltraChem site to the river.

A second aspect of the HoltraChem site work was to assess groundwater loading of Hg to the river from the site. This was done using conventional seepage meters installed on the foreshore of the HoltraChem site, and by a radon tracer technique to determine groundwater flow, which along with beach seepage and porewater Hg concentrations, was used to estimate groundwater loss of Hg to the river. Estimates using both approaches were similar. Groundwater Hg loading to the river was assessed twice (spring and late summer) using the tracer technique, seepage meters installed in the foreshore and sampling of foreshore seepages and porewater. Subtidal water sampling in the vicinity of the suspected groundwater discharge zone on low ebbing tides in 2011 did not detect Hg concentrations in excess of upstream concentrations.

Over the period of study approximately 2.3 kg of Hg were discharged to the Penobscot River by the two surface streams that drain the site. For one stream (Southerly), 90% of the total loading (1.9 kg) occurred in a few hours following a large rainfall (11.4 cm). The same storm accounted for only 17% of the total load (0.39 kg) for the other stream (Northerly). Average daily loading for these streams were 1.12 and 5.66 g/day, respectively, for the Northerly and Southerly Streams. Northerly Stream exhibited a higher mean total Hg concentration (10,800 ng/L) and higher average Hg content of suspended matter (392  $\mu$ g/g dry wt.) compared with Southerly Stream (2,460 ng/L, 64  $\mu$ g/g dry wt.). The combined total loading from the two surface streams for the period of study (422 days) was 2.3 kg (5.4 g/day) with most (78%) of this combined loading associated with a single large storm. Thus, in the absence of this storm the combined loading loading estimate would have been 0.51 kg (1.16 g/day).

Groundwater seepage rates from the site, as estimated from both the tracer and seepage meter methods were in the 3 to 4 cm/day range and, when combined with a best estimate of the area of groundwater discharge (11,000 m<sup>2</sup>) and average seepage/porewater Hg concentration (242 ng/L, UCL95), yielded a loading of 0.22

g/day (80 g/year) for site groundwater. This estimate of recent Hg loading from groundwater is substantially less than that (17 g/day) reported in the late 1990s.

None of the municipal or industrial point sources of Hg to the river between Veazie and Bucksport exceeded 1 g/day individually nor was the aggregate loading of all such sources > 2 g/day (based on State of Maine data). The HoltraChem site Hg loading can also be compared to that by the three largest tributary streams downstream of Veazie Dam, Kenduskeag, Souadabscook and North Branch Marsh River. Hg loadings for these tributaries were estimated by other project scientists to contribute 3.8, 4.8 and 2.9 g/day, respectively, to the Penobscot River. The combined loading of all tributaries between Bangor and Bucksport was estimated at ~15 g/day (5.5 kg/year). Higher tributary stream loadings than these average values would be expected during freshet and other times of higher flow. Based on sampling (discharge-weighted mean total Hg =3.9 ng/L) and historical discharge data (406 m<sup>3</sup>/s) the Penobscot River at Veazie dam contributes ~140 g/day (50 kg/year) to the downstream reach depending on river discharge.

# 2 INTRODUCTION

The HoltraChem Manufacturing Company in Orrington, Maine is located on a 235-acre property on the banks of the Penobscot River. Approximately 50 acres are developed and include the manufacturing facility, five landfills, a surface impoundment and a scrap metal area. The immediate plant area covers approximately 12 acres. The facility opened in 1967 and manufactured chlorine, caustic soda (sodium hydroxide), chlorine bleach (sodium hypochlorite), hydrochloric acid and the pesticide chloropicrin. The plant closed in September, 2000. As of 2011 some limited demolition (including demolition of the mercury (Hg) cell building, but not removal of its concrete floor) and waste removal from the site has been completed, but much of the infrastructure and all the landfills remain.

Groundwater and surface water on the plant site as well as surface water in the adjacent Penobscot River was subjected to detailed characterization prior to the plant closing in 2000 (e.g., Camp, Dresser and McKee [CDM] 1998). These results indicated that groundwater within the plant was Hg-contaminated (reported concentrations > 1,000  $\mu$ g/L in some wells), and that surface runoff was also elevated in Hg. The presence of the plant could be detected by sampling Hg in Penobscot River surface water near the plant outfalls (reported concentrations up to 70 ng/L compared with upstream background of <5 ng/L). In fact, calculations suggested that the net effect of all plant discharges and the estimated groundwater flux would raise average Hg concentration in the river by about 2 ng/L at a river discharge of 4000 cfs (113 m<sup>3</sup>/s). The total Hg loading to the Penobscot River from the plant site was estimated at approximately 20 g/day. This loading was then compared to that carried by the river from upstream under various river flow conditions. At average river discharge (16,400 cfs, 464 m<sup>3</sup>/s) and assuming ambient total Hg concentration in river water (4.3 ng/L), CDM (1998) estimated river loading was 172 g/day.

Direct discharges of wastewater from onsite water treatment plant to the Penobscot River were significantly reduced following plant closure in 2000 although groundwater discharges and storm water runoff continued. Beginning in January 2005 a groundwater extraction and treatment system began operating to capture discharge of contaminated groundwater from the area of Landfill 1. Total Hg loading to the Penobscot River from the site has apparently not been summarized or rigorously evaluated since prior to cessation of production but some data collection (surface water and groundwater sampling) has continued.

## 2.1 Review of HoltraChem and Other Monitoring Data

Licensed discharges of Hg to the river - publicly available data for discharges of Hg to the Penobscot River - can be accessed via the Toxic Release Inventory (TRI) and Discharge Monitoring Reports (DMR). These data are from a sampling site at the upper end of pipe running to Outfall 001 and were reported by the facility to the federal (TRI) and state (DMR) agencies that maintain these databases<sup>1</sup>. Figure 3-1 summarizes Hg

<sup>&</sup>lt;sup>1</sup> TRI link: <u>http://oaspub.epa.gov/enviro/tris\_control\_v2.tris\_print?tris\_id=04474LCPCHROUTE#p2report</u>. DMR data were obtained directly from the Maine Department of Environmental Protection.

loading data at this outfall from 1987 to 2011 in lbs/yr (as recorded in original databases) with horizontal grid lines allowing conversion to equivalent grams/day, the units used everywhere else in this report. The data presented represent measured discharges through a permitted outfall (001) only, and do not include other losses to the river from discharges of the Northerly and Southerly Streams, or from non-point sources such as groundwater and non-sampled overland flow from the site. The large decrease in loading indicated for the 1999-2000 period corresponds with cessation of production at the plant. The smaller decrease in loading circa 2009 is related to reconfiguration of wastewater routing upstream of the monitored outfall which no longer captured the Hg-contaminated groundwater infiltration.



Figure 3-1. Hg loading data for Outfall 001 retrieved from the TRI and DMRs for the HoltraChem Manufacturing Facility.

An estimate of yearly and cumulative Hg loading (Figure 3-2) to the Penobscot River since the plant began operating in 1967 was also prepared to support modeling efforts by other Penobscot River Mercury Study (PRMS) scientists. The assumptions used and the results are provided below and in tabulated annual form in Appendix 3-6.

#### 2.1.1 Pre-1970 Losses

Flewelling (1971): An average 100 ton Cl/day plant had total Hg losses of 39 lbs/day (17,700 g/day) prior to mid-1970. Total losses were estimated as including 25% to 60% losses to sewer (rivers, lakes, bays). Thus, Holtrachem producing 180 ton Cl/day would have lost 4400 to 10,600 g Hg/day to Penobscot River prior to 1970. Two

measurements are available from 1970, both assumed to include only Outfall 001, and not groundwater or streams:

- July 14, 1970 = 2.65 lbs/day (1200 g/day)
- August 19, 1970 = 0.22 lbs/day (100 g/day)

This large change is probably due to redirecting brine sludges to an onsite pond after regulators intervened.

#### 2.1.2 Losses between 1970 and 1987

Aquatic losses for typical plants were regulated in the earliest part of this period to 45 g/day.

No info for ground water or surface discharges found for this period.

Assumed to be 75 g/day, i.e lower than earlier but higher than later.

#### 2.1.3 Losses after 1987

TRI and Maine DMRs are available (see spreadsheet in Appendix 3-6). Assume these reflect outfalls, streams but not groundwater.



Figure 3-2. Estimated or Measured Annual and Cumulative Hg Losses to the Penobscot River, 1967-2011. See Appendix 3-6 for tabulation.

Some outfall-specific data (time series discharge and concentration) were obtained directly from the facility to assist in designing the present study. Discharge and Hg concentration data are collected by plant staff at two streams (Outfall 003 and Southerly Stream) on a weekly basis (Figure 3-3). The Outfall 003 sampling site is located on the Northerly Stream upstream of the site that we established for the PRMS. The CDM's sampling site on Southerly Stream was also upstream of the PRMS sampling site on Southerly Streams were sampled and the discharge recorded by plant staff on a weekly basis. Detailed data for the period January 2005 to October 2007 were reviewed for this study and are summarized in Table 3-1.

Table 3-1: Summary discharge, Hg concentration and loading data for two surfacestreams on HoltraChem site, January 2005 to October 2007 (dataprovided by CDM)								
Parameter	Northerly stream (Outfall 003)	Southerly Stream						
Mean discharge (gpm)	17.0	52.9						
Max discharge (gpm)	123	784						
Min discharge (gpm)	0.015	2.19						
Mean [Hg] (ng/L)	862	299						
Max [Hg] (ng/L)	8000	1564						
Min [Hg] (ng/L)	179	66						
Mean Hg Loading (g/day)	0.10	0.098						
Max Hg Loading (g/day)	1.88	1.28						
Min Hg Loading (g/day)	0.00003	0.001						

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When summed, the average Hg loading from these two streams amounts to about 0.2 g/day with a maximum combined loading of just over 3 g/day. Examination of the time series loading data for these streams (Figure 3-3) indicates that highest loadings occur between November and June. The highest loading recorded for this time interval occurred in October 2005 when monthly total rainfall (33.8 cm) was almost 25 cm above normal.



Figure 3-3. Hg loading at Northerly (Outfall 003) and Southerly Stream locations monitored by plant staff (data provided by CDM).

Groundwater has been pumped since February 2005 from an extraction well (MW-601), located near the downgradient edge of Landfill 1, to a water treatment plant. The extracted groundwater averaged 140,000 ng/L during the first six months of 2009 according to court testimony (pg 14, Mallinckrodt 2010). The effluent from this water treatment plant is routed through Outfall 001 that discharges to the Penobscot River through a submerged pipe below the high tide level. The data for Hg concentration and discharge from the water treatment plant, which were reviewed for this report began in January 2005 and end in October 2007, although only loading data were reviewed after April 2006. The treatment works is a "batch system" and thus discharge is not continuous. Figures 3-4 and 3-5 illustrate the trends in these data sets and show an apparently stable loading of about 0.02 g/day at a total Hg concentration of ~ 300 ng/L.

Note that the submerged pipe that transports effluent from the treatment plant also carries groundwater that infiltrates into the pipe and other underground structures upstream of the point of discharge to the Penobscot River. Access to the far end of this pipe in the river is obviously limited and thus no data are known to exist that might help quantify the combined loading from both the treatment plant and groundwater that infiltrates the entire length of this pipeline. As noted earlier, infiltration from the upper

portion of the pipe was captured prior to 2011 and monitored at Outfall 001. Efforts were made during the summer of 2011 to locate and sample the outflow of this pipe (see Appendix 3-7). It could not be located and is thought to be buried by sand within an intertidal area observed to be unstable (fluidized) and have extensive seepage (see later section on seepage sampling in 2009-2010).



Figure 3-4. Average monthly Hg concentrations and discharges for the HoltraChem Wastewater Treatment Plant January 2005 to April 2006 (data provided by CDM)



Figure 3-5. Hg loading for HoltraChem Wastewater Treatment Plant, January 2005 to October 2007 (data provided by CDM).

#### 2.1.3.1 Penobscot River surface water samples

Surface water samples were also collected by the first author for the PRMS in October 2004 near-low tide at the edge of water (0.5 to 1.0 m below surface) at ten (10) locations spanning the length of the intertidal beach adjacent to plant and Southerly Cove (Figure 3-6). In addition, two seepage samples were collected at the northern end of the beach. The objective of this limited sampling program was to determine whether evidence of continued Hg loading from the site could be detected in the adjacent river or groundwater seepages as had been the case while the plant was still operating (e.g., CDM 1998). By design this sampling event was intended to detect and quantify foreshore Hg concentrations below Landfill 1 prior to implementation of groundwater capture and treatment. All samples were analyzed for dissolved Hg (0.45 micron filtered) and a few analyzed for total Hg (unfiltered). Results for this sampling are shown below. The dissolved Hg concentration in this set of foreshore samples did not suggest presence of a nearby source. However, the beach seepage sample results were elevated 10-fold or more over river water and coincidently very similar to results reported for the same seepages by Acheron in the 1990s (CDM 1998). Total Hg (unfiltered) results are too limited to draw any inferences. Although not shown in the figure, suspended sediment Hg concentrations at HC-1, HC-3 and HC-10 ranged from 0.8 to 1.3 µg/g dry wt. These values are somewhat higher than those for suspended sediment values reported for the river downstream of Orrington (0.3 to 0.8  $\mu$ g/g, e.g., see Fig 16 in Phase 1 Penobscot River Mercury Study: 2006-2007, PRMS 2008 - refer to appendix of Chapter 1 – appendix 1-2) and thus did not suggest a strong local source in the foreshore at the time of this sampling.



Pointer lat 44.737783° lon -68.829483° elev 7 ft Streaming [[[]]] 100% Figure 3-6. Locations of near-shore and seep sampling, October 9, 2004.



Figure 3-7. Hg concentrations in seepage and near-bottom water adjacent to HoltraChem site on October 9, 2004 (see Figure 3-5 for locations).

# 2.1.3.2 Mercury Fluxes Measured Downstream of HoltraChem Site in October 2004

The objective of this investigation conducted by the first author of this report was to quantify the net downstream flux of Hg in the vicinity of the HoltraChem site near Orrington, Maine. To accomplish this objective, numerous measurements of river discharge and Hg concentrations were taken over a 2-day period beginning October 7, 2004 at a location ~2 km downstream of the HoltraChem plant site. In addition, water samples were collected from the Penobscot River near Veazie Dam and from several large tributaries upstream of the plant site (e.g., Kenduskeag, Souadabscook and Sedgeunkedunk) to provide data for completing the mass balance. A full description of this investigation and results are provided in Appendix 3-1.

This study was unable to derive an unequivocal estimate of the net downstream flux of Hg in the vicinity of the HoltraChem site. Nonetheless, the results did allow some estimates of the maximum possible flux from the site that ranged from 7 to 28 g/day with an indication (based on analysis of salinity profiles) that the actual HoltraChem flux was probably less than 7 g/day during the 2-day period of investigation when river discharge at Veazie Dam was at a seasonal low (mean=209 m<sup>3</sup>/sec). Upstream sources (Penobscot River at Veazie Dam, major tributary streams and outfalls) during the same time were estimated at ~33 g/day. The study also documented that bidirectional flow was often present at the transect location and greatly complicated measurement and interpretation of Hg fluxes.

#### 2.1.3.3 Groundwater

Prior to installation/operation of a groundwater extraction and treatment system in January 2005 at the HoltraChem site, groundwater discharge to the Penobscot River from beneath Landfill 1 was estimated by (CDM 1998) to average 3.56 gpm (685 feet<sup>3</sup>/day). CDM used four somewhat independent methods to arrive at this average but the range of estimates was remarkably constrained. The daily discharge volume was multiplied by the average Hg concentration in site groundwater beneath Landfill 1 (880,000 ng/L) to yield an estimated loading of about 17 g/day for this source to the river.

Subsequently, CDM conducted a pump test at Landfill 1 in November 2003 to assess the effect of groundwater extraction on the Hg loading to the river. Calculations using the pump test results suggested that Hg flux from Landfill 1 to the river was significantly lower (3.3 g/day) than in 1998 due to lower plume concentrations (mean = 140,000 ng/L). The test results also indicated that approximately 1.7 g Hg/day could be recovered from groundwater at a 3 gpm pumping rate. In 2006, when the groundwater extraction and treatment system was operational, it was capturing about 12 gpm (2300 feet<sup>3</sup>/day) for treatment in the water treatment plant. Thus, the net reduction of Hg loading to the river may be significantly lowered by pumping this groundwater to the treatment plant for Hg removal before releasing it to the river through outfall 001.

#### 2.1.3.4 Reconnaissance Sampling October 2007

Surface water samples were collected at the HoltraChem site in late October 2007 in part to assess the differences, if any, between surface water samples collected at the regularly monitored upstream locations sampled previously by CDM (Outfall 003 and SS) and the locations where each drainage (Northerly Stream and Southerly Stream) enter the Penobscot River (Figure 3-8). As indicated in Table 2 below, concentrations of both total Hg and methyl Hg were substantially higher at the upstream monitored locations than at the locations near the river. In addition, total Hg in the three seeps was highly variable with two values substantially exceeding those measured earlier (see Figure 3-7 this report; CDM 1998).

Table 3-2: Results of reconnaissance sampling of surface streams, one spring andforeshore seepage on the HoltraChem site in October 2007.							
Sample ID	NS-UP	NS-DN	SS-UP	SS-DN	SS-SPR		
Date and time	10/26/07 14:45	10/26/07 14:00	10/26/07 16:24	10/26/07 15:20	10/26/07 16:00		
Specific Conductivity(µS/cm)	661	614	2186	2240	1364		
Total Hg (ng/L)	478	104	133	51.4	75.8		
Dissolved Hg (ng/L)	166	48.4	47.1	20.2	38.2		
Suspended Hg (µg/g)	442	90.6	139	20.7	0.51		

Table 3-2: Results of reconnaissance sampling of surface streams, one spring andforeshore seepage on the HoltraChem site in October 2007.							
Sample ID	NS-UP	NS-DN	SS-UP	SS-DN	SS-SPR		
Methyl Hg (ng/L)	2.59	0.13	0.23	0.16	0.13		
Dissolved Methyl Hg (ng/L)	1.55	0.08	0.20	0.14	0.08		
Suspended Methyl Hg (µg/g)	1.47	0.08	0.06	0.02	0.0006		

Sample ID	Seep 1	Seep 2	Seep 3
Date and time	10/26/07 17:05	10/26/07 17:30	10/26/07 17:45
Specific Conductivity (µS/cm)	8636	2402	3266
Dissolved total Hg (ng/L)	265	15.3	1882
Dissolved methyl Hg (ng/L)	0.077	0.023	0.137



Figure 3-8. Locations of reconnaissance sampling in October 2007

# **3 OBJECTIVE**

The overall objective of this work was to attempt to determine the size of the ongoing sources of Hg to the Penobscot estuary from the HoltraChem site and from other possible industrial and municipal sources below the Veazie Dam. The size of these ongoing sources of Hg from the HoltraChem site and the "other" sources to the estuary are two of the factors that are determining the present-day recovery rate of the estuary from Hg pollution.

#### 3.1 Approach

A multi-faceted approach was used to assess the present day loss of Hg to the river from the HoltraChem site. This included measurements of Hg fluxes to river from surface water streams and groundwater seeps (intertidal and subtidal) as well upstream-downstream measurements of Hg concentration in the river water during several tidal cycles. Figure 3-9 shows the general locations of all water sampling on and off of the HoltraChem site. The distal end of Outfall 001 that discharges subtidally could not be located for sampling and thus assessment of Hg loading data from this outfall relied on DMRs from Maine Department of Environmental Protection (MDEP). To achieve the objective of loading assessment from the HoltraChem site, this PRMS study improved the existing infrastructure for measurement of surface water discharges and employed a method (natural radioactive tracer) to assess groundwater discharges that had not been applied previously to the site. The existing infrastructure for estimating surface water discharges consists of two monitoring locations fitted with V-notch weirs. Flows had previously only been recorded at time of sampling (grab type) that occurred once per week. Neither location captures all surface runoff that enters the Penobscot River from the site. As shown by the sampling results from October 2007 (summarized above), concentrations can differ significantly between these monitoring locations and the points of discharge to the river.

Continuous flow monitoring and water sampling equipment (Teledyne Isco composite samplers and bubble meters) were installed downstream of the existing weirs and operated for slightly longer (422 days) than one year to obtain an improved estimate of Hg loading from these streams. Monthly grab sample data for total (unfiltered), dissolved (filtered) Hg and suspended matter were also collected to provide an estimate of the loading of Hg in a form (dissolved) that may be immediately bioavailable as well as to assess the Hg content of suspended matter leaving the site through these two streams.



Figure 3-9. Water sampling locations to assess Hg loading from HoltraChem site.

# 4 METHODS

#### 4.1 Surface Streams

The two main surface streams (Northerly, NS and Southerly, SS) that receive runoff from the plant site were instrumented with flow monitoring/logging equipment and automated flow-proportional samplers. Although v-notch weirs existed in the upper sections of each stream, new downstream measurement locations closer to the high tide level were established and fitted with a Parshall flume (Northerly Stream) or a 90degree weir (Southerly Stream), bubble tube and automatic sampler. Discharge was calculated from the depth of water in each control structure (see Appendix 3-2 photographs) as measured by the bubble meter. The automatic samplers were programmed to collect weekly discharge-proportional composite samples of unfiltered water. In addition the samplers were also set up occasionally to collect composite samples over shorter intervals, e.g., while storm flow was occurring or about to occur. The composite sampling rate had to be adjusted regularly to avoid overfilling the bottles while still insuring that at least one subsample (set to 100 mL) was collected per day. Ideally, the 2500 mL composite sample bottle would fill exactly in 7 days (about three 100 mL subsamples collected per day). Overfilling occurred on several occasions on both streams. Whenever possible the amount of overfilling was measured and recorded but on one occasion (June 2009) after an 11.4 cm overnight rainfall, both sampling systems were overwhelmed. No samples that had experienced overfilling were discarded but the analytical results were flagged in the master spreadsheet.

The automatic samplers were modified slightly to assure collection and preservation of "clean" samples. All tubing was changed to silicon (pump tube) and Teflon (intake line) and replaced regularly. Sample containers were glass and specially-cleaned by the analytical laboratory prior to each use. Equipment blanks, representing reagent or spring water with known Hg concentration that has been run through the sampling train were prepared periodically to evaluate contamination.

Collection of weekly composite samples was augmented with collection of monthly grab samples for analysis of Hg (total and dissolved) and total suspended solids (TSS). The resulting data allowed calculation of the range of suspended Hg concentrations ( $\mu$ g/g) in these streams [Hg<sub>susp</sub>=(Hg<sub>tot</sub> – Hg<sub>diss</sub>)/TSS].

Sample collection and flow monitoring began April 21, 2009 and continued until June 16, 2010. Freezing conditions precluded automatic sampling from mid-December 2009 to mid-March 2010. However, monthly grab sampling and flow monitoring continued through this period whenever conditions allowed. Northerly Stream never froze while Southerly Stream was periodically frozen and impossible to sample or to estimate discharge.

A non-recording rain gauge was installed near the Northerly Stream monitoring station to measure cumulative rainfall between times that the composite sample bottles were replaced. Results (see Table 3-4 in RESULTS section) from this gauge compared reasonably well to rainfall amounts reported for the Bangor Airport and were used to verify that increases in flow at the monitored streams were due to rainfall in the watersheds.

## 4.2 Intertidal Seepage and Porewater Sampling

Intertidal seepage water samples were collected on several occasions along the sandgravel foreshore below Landfill 1 and once along the rock bluff below Landfills 3, 4 and 5 (Figures 3-9 and 3-27). Seepage samples were collected by dipping using a clean wide-mouth transfer container and then filtered immediately using a Nalgene disposable filtration unit. Seepage samples from the rock bluff (see Appendix 3-2 photograph) were collected unfiltered directly into a sample bottle. All the seepage samples from the rock bluff were "clear".

Porewater samples from the foreshore below Landfill 1 were collected using a clean 1inch diameter PVC well screen inserted approximately 12 inches into the beach substrate (see Appendix 3-2 photograph). After porewater partially filled the screen, Teflon tubing connected to a peristaltic pump was inserted. The pump was set to operate at a low flow rate (~100 mL/min) and the flow routed through the flow cell of a YSI 556 multimeter to measure temperature, specific conductivity, salinity and dissolved oxygen. Readings were allowed to stabilize and then recorded. The tubing was removed from the flow cell and attached to an inline capsule filter (Pall Aqua-Prep or similar) to collect a dissolved sample for analysis.

#### 4.3 Groundwater

The main approach to quantifying groundwater input to the river entailed use of the natural-occurring radionuclide, radon (<sup>222</sup>Rn) (Burnett and Dulaiova 2003; Burnett et al. 2006; Mulligan and Charette 2006). Over the past decade, naturally occurring radionuclides such as radon-222 (<sup>222</sup>Rn) have gained popularity as tracers of groundwater flow due to their enrichment in groundwater relative to other sources (river water, rainfall) and the fact that they can provide integrated water flux estimates over a large area. The enrichment of these tracers is due to the fact that the water-sediment ratio in aquifers is usually quite small and that aquifer sediments/rock are enriched in uranium/thorium and their decay products; radon, which is a noble gas, can easily partition into the aqueous phase. Maine groundwater has among the highest radon levels in the world, making this an ideal candidate for the project: if appreciable subsurface flow into the Penobscot River is occurring, then the flux of this element via groundwater will lead to enrichment in the surface water body that is well above background levels. A simple mass balance/box model (Figure 3-10) was constructed for the system under study, where all sources other than groundwater are subtracted from the total inventory of the radon and losses are added back in (atmospheric exchange, decay). The residual inventory (I), or "excess", was then divided by the concentration of the radon in the discharging groundwater, which was measured from seeps, wells, and piezometers.





Although changing radon concentrations in river waters could be in response to a number of processes, advective transport of groundwater (pore water) through sediment of Rn–rich solutions is usually the dominant process. Thus, if one can measure or estimate the radon concentration in the advecting fluids (e.g., river water and intertidal sediment porewater and/or site groundwater), we can convert <sup>222</sup>Rn fluxes to water fluxes. The complete procedure for estimating groundwater fluxes from continuous radon measurements in a tidally influenced river may be summarized by the following steps:

We first performed continuous measurements of <sup>222</sup>Rn activities (Becquerel Per Cubic Meter [Bq/m<sup>3</sup>]) in the river water column, water depth, water and air temperatures, wind speed, and atmospheric <sup>222</sup>Rn concentrations. All of these measurements were performed using automated sensors with data logging capabilities deployed on a float.

We then calculated excess (unsupported by <sup>226</sup>Radium (Ra)) <sup>222</sup>Rn inventories for each measurement interval, i.e.,

I (Bq/m<sup>2</sup>) = Excess<sup>222</sup>Rn (Bq/m<sup>3</sup>) × water depth (m) Ex<sup>222</sup>Rn (Bq/m<sup>3</sup>) = total <sup>222</sup>Rn - <sup>226</sup>Ra (Bq/m<sup>3</sup>)

Excess<sup>222</sup>Rn activities in the water column were estimated from measured <sup>222</sup>Rn less an assumed river-water <sup>226</sup>Ra activity (80 dpm/m<sup>3</sup>). Though <sup>226</sup>Ra was not measured, the value we used is a conservative, upper limit <sup>226</sup>Ra estimate that is typically less than 1-2% of the typical <sup>222</sup>Rn activity.

The calculated inventories were next normalized to mean tidal height to remove the effect of changing inventory due simply to tidal height variations. This normalization was done for each measurement interval by multiplying the unit change in water depth (m)

over the measurement interval times the <sup>222</sup>Rn activity (Bq/m<sup>3</sup>) during the flood tide and by activities of <sup>222</sup>Rn for the ebb tide. The flood tide corrections are negative (since the inventory would be increasing due simply to an increase in water depth) and the ebb tide correction is positive.

We next corrected the tide normalized inventories for atmospheric evasion losses during each measurement interval. The total flux across the air–water interface depends on the molecular diffusion produced by the concentration gradient across this interface and turbulent transfer, which is dependent on physical processes, primarily governed by wind speed. Standardized equations from the literature that relate trace gas concentration, temperature and wind speed are used for this correction of the unsupported inventory. The final loss term is decay, which is accounted for as the product of the <sup>222</sup>Rn inventory (I) and the <sup>222</sup>Rn decay constant ( $\lambda = \ln(2)/t_{1/2}$ ).

Net <sup>222</sup>Rn fluxes ( $F_{net}$ ) was estimated by evaluating the change in the corrected inventories over each time interval ( $\Delta t$ )

$$F_{net}$$
 (Bq/m<sup>2</sup>/sec) =  $\Delta I$  (Bq/m<sup>2</sup>)/ $\Delta t$  (sec)

These fluxes represent the observed fluxes of <sup>222</sup>Rn into the river water column with all necessary corrections except loss via mixing with lower concentration waters from upstream, downstream or from opposite shore of the river. These net fluxes are likely minimum values as we base the estimate on what remains in the system (what can be measured) and higher mixing rates could be compensated by higher fluxes. The mixing loss from the box is the inventory at a given time point minus the minimum Rn activity during the time series (basically considered as the background radon value delivered to the site via tide driven mixing) times 1.9 tides per day. Together these represent export via mixing (Bq/m<sup>2</sup>/sec) corrected for river-air loss and decay. At steady state, the sum of these three "outputs" is balanced by the net Rn input to the box via groundwater.

Note that diffusion is not considered in these calculations because advection is the dominant process in fluvial surface water-groundwater interactions.

In order to convert radon flux estimates into water flux we measured  $^{222}Rn$  concentrations in site groundwater and intertidal porewater within several seepage zones. Water fluxes,  $\omega$  (m/sec), were calculated by dividing the estimated total  $^{222}Rn$  fluxes by the excess  $^{222}Rn$  in porewater ( $^{222}Rn_{pw}$ ), where excess is calculated as for river water,

$$\omega$$
 (m/sec) = F<sub>total</sub>/Ex<sup>222</sup>Rn<sub>pw</sub>

For convenience we express groundwater fluxes in cm/day in the balance of this report.

As an additional measure of groundwater discharge at this site, a number of manual seepage meters (Lee 1977) were deployed in the intertidal zone below Landfill 1. In its simplest form, the seepage meter is the top of a 55-gal drum with a small opening through which the flow is channeled into a plastic bag (see Appendix 3-2 photograph). The volume collected in the bag over a fixed period of time was combined with the cross-sectional area of the drum to provide an estimate of flow. Seepage meters must

be submerged in order to operate. Given the large intertidal area at this study site, they could miss a potentially large component of the groundwater flux. However, they worked in the subtidal zone as well as within the intertidal for portions of each tide cycle, and provided a valuable comparison to the fluxes obtained via the radon approach.

The total shoreline length bordering the HoltraChem property is approximately 1000 m, of which 300 m consisted of a gently sloping silty sand and gravel beach. The other 700 m consists of a relatively steep rock wall with one narrow beach located below Landfills 3, 4 and 5. We assumed that groundwater seepage occurs over a 25 m wide seepage face along the gently sloping beach below Landfill 1 and over a 5 meter wide seepage face over 700 m of rock wall. Thus, the effective seepage zone is 11,000 m<sup>2</sup>.

#### 4.3.1 Subtidal Water Sampling to Identify Groundwater Source

While the radon method of assessing groundwater inputs to the Penobscot River should have captured water fluxes from both intertidal and subtidal zones, no data on subtidal porewater Hg concentrations were obtained to combine with water fluxes to assess subtidal Hg flux. Any significant subtidal fluxes of Hg should be revealed by sampling near-bottom water in zones of suspected subtidal groundwater discharge. During summer of 2011 we conducted near-bottom water sampling in the vicinity (Figures 3-9 and 3-28) of the HoltraChem site on three occasions: July 7, September 9 and October 26. For all three sampling events we chose sampling times to correspond with late ebbing tides to maximize the hydraulic head and chemical gradients. The elevation difference between the river surface and upland water table increases as the tide elevation decreases producing a higher driving force for groundwater discharge to the river. Near low tide is also a period when river water adjacent to the HoltraChem site is likely to be least affected by upstream movement of seawater and river water that might have already acquired Hg from site groundwater discharges. Sampling always began upstream of likely groundwater discharge zones (e.g., river section below Landfill 1) and proceeded downstream. Field measurements of specific conductance (salinity) were used to verify that only river water was present at depth and as a possible indicator of "brine" inputs for the HoltraChem site. Distinguishing between a local source of brine and intrusion of seawater at depth was not expected to be straight forward but might be resolved if evidence of brine or seawater began exactly adjacent to the site. As discussed later, no evidence of either was detected by the near-bottom salinity surveys. Water samples for Hg analysis were collected along several transects from foreshore to mid-river (July 7 and September 9) or parallel to river flow direction (October 26) along bottom of main channel (Figures 3-9 and 3-28). These samples were collected by pumping (ShurFlo diaphragm pump) water through a weighted intake line (C-flex) protected at the inlet by a plastic cage and held ~10 cm above the sediment-water interface.

## 4.4 Other Mercury Sources

Other point sources of Hg to the Penobscot River, including especially those between Veazie and Bucksport, were sought by queries to the Maine DEP, specifically to obtain discharge monitoring and TRI data. A state-widesampling and analysis program (Maine DEP 2001) to assess Hg sources throughout the state of Maine also provided data for

Hg concentrations in outfalls that could be combined with design wastewater discharge data to calculate loading from point sources.

In addition to retrieving data from other agencies, PRMS scientists also conducted sampling of the Penobscot River at Eddington (below Veazie Dam) and the major tributary streams, including Kenduskeag, Souadabscook, Sedgeundedunk and North Marsh River. Discharge data for these streams was not available for the sampling times. However, mean annual discharges for these streams were estimated using watershed area and other properties (Dudley 2004). Tributary Hg loadings were calculated as the product of these discharges and measured Hg concentrations. Appendix 3-8 provides the data, graphics and calculations used to estimate loadings for the Penobscot River near Veazie Dam and all major tributaries between Bangor and Bucksport.

#### 4.5 Sample Handling and Analysis

All water samples for Hg analysis were shipped overnight to Battelle's analytical laboratory in Sequim, Washington. This laboratory has participated successfully in the PRMS's QA/QC program. The weekly composite samples were shipped directly to the analytical laboratory and oxidized (BrCl) in the same container used for collection (EPA Method 1631e, Section 8.5.1). After analysis the containers were cleaned by the laboratory and returned for reuse. Similarly, grab samples were iced, shipped overnight and then oxidized on arrival at the laboratory. Grab samples for "dissolved" Hg were always field-filtered using Nalgene 0.45 micron pore size disposable filtration units with nitrocellulose membranes. Samples for TSS were iced and also shipped immediately after collection to facilitate analysis within the 7-day holding time for this analyte.

Field quality control/assurance actions included preparation of field and equipment blanks. Equipment blanks for the Isco autosampler system were prepared by pumping locally purchased spring water (Poland Springs) through all the tubing used for sampling. Both new and used tubing were included. Field blanks were prepared by filling sample bottles with the same purchased spring water used to prepare the equipment blanks. Results for equipment and field blanks are summarized in Table 3-3. Although some equipment blanks showed concentrations higher than the water used to generate these blanks, no equipment blanks exceeded 1% of the lowest concentration observed for any sampling system.

Table 3-3: Quality control/assurance results for field and equipment blanks.								
Date	Location	Туре	Equipment	Result (ng/L)	Notes			
4/14/2009	SS	EQ-Unfilt	Pump/Intake Tubing	<0.1	New tubing			
4/14/2009	NS	EQ-Unfilt	Pump/Intake Tubing	<0.1	New tubing			
4/14/2009		Field	na	<0.1	Poland Spring			
9/17/2009		Field	na	0.15	Poland Spring			
9/18/2009	Foreshore	EQ-Filt	Well screen	0.1	Tubing/Used filter			
9/19/2009	Foreshore	EQ-Filt	Nalgene filter/C-flex	0.31	Tubing/New filter			
10/12/2009	NS	EQ-Unfilt	Pump/Intake Tubing	1.7	Used tubing			
10/12/2009	SS	EQ-Unfilt	Pump/Intake Tubing	1.4	Used tubing			
3/9/2010	SS	EQ-Unfilt	Pump/Intake Tubing	0.11	New tubing			
3/9/2010	NS	EQ-Unfilt	Pump/Intake Tubing	<0.1	New tubing			
3/9/2010	SS	EQ-Unfilt	Pump/Intake Tubing	0.29	New tubing			
3/9/2010	NS	EQ-Unfilt	Pump/Intake Tubing	18.8	Used tubing			
4/20/2010		Field	na	0.18	Poland Spring			
4/20/2010	NS	EQ-Unfilt	Pump/Intake Tubing	0.50	New tubing			

Generally no field replicate samples could be collected as part of the flow-proportional composite sampling of the Southerly and Northerly Streams due to the nature of the sampling system. However, on one occasion (May 2009) several replicates were collected over a few minutes using the pump and tubing associated with the Isco sampler. The relative standard deviation (%RSD) for these replicates was 3.9%. Field replicates (FDs) were collected for the monthly grab sampling program and for the foreshore seep and porewater program. Laboratory matrix duplicates (MDs) were also run typically at the rate of one per batch of 10 or fewer samples. Results of the field and laboratory duplicate analyses are summarized in the Appendix 3-4. The grand mean %RPD of these replicates is 1.1% with a range from -12% to 20%.

# 5 RESULTS

#### 5.1 Surface Stream Monitoring

Figures 3-11 and 3-12 present flow monitoring results and a summary of the number of subsamples collected as part of the Isco composite program on the Northerly and Southerly Streams.

Flow monitoring at the Northerly Stream proceeded mostly without problems. During the high rainfall event of June 19, 2009 the temporary dam holding the Parshall flume was

briefly (< 30 minutes) overtopped (by 3.1 cm). The dam was overtopped (by 3.8 cm) a second time (<30 minutes) on December 3, 2009 following an intense 2.5 cm inch rainfall on frozen ground. No physical damage was sustained from either of these overtoppings but the highest discharges associated with these storms were underestimated and the composite sample bottle was overfilled during the June 2009 storm. Of the nearly 40,000 15-minute interval discharge values recorded at this monitoring station only 16 values exceeded the maximum recommended discharge (835 gpm) for this size Parshall flume. The location of the flume on this stream near the elevation of maximum tide resulted in a few occurrences (12) of tidewater rising into the throat of the flume. These events were relatively rare and occurred on especially high equinoctial tides. These were easily detected in, and removed from, the discharge records and had only a minimal effect on sample compositing because of the short duration of the tidal interference.

Flow monitoring at the Southerly Stream also experienced some problems due to the large storm (11.9 cm) that occurred on June 19-20, 2009. The site had been set up to use the Manning Equation to calculate flow through the 35-inch (88.9 cm) plastic culvert pipe that runs under the forest road that crosses the stream. The bubble tube and sampling line intake were located on the downstream end of this pipe. During the June storm very high peak runoff from the Penobscot Energy Recovery Company (PERC) plant, which enters Southerly Stream just downstream of the culvert pipe, washed out part the road, partially blocked flow from the pipe and buried both the bubble tube and sample intake. The discharge records for most of this storm were thus rendered unusable and the composite sample compromised. The bubble tube and intake were unburied on June 22 and the composite bottle replaced. However, backwater effects from the PERC stream continued to frustrate acquisition of reliable flow data until the monitoring site was moved and a 90-degree weir plate installed on the upstream end of the pipe on July 9, 2009. Reconstructed discharge data for the period June 19 through July 9 were obtained by regression analysis of Northerly Stream 15-minute discharge data to predict discharges for Southerly Stream. For this period discharges on Southerly Stream (Q<sub>SS</sub>) were calculated as:

 $Log Q_{SS} = -0.742 + 1.574 \times Log Q_{NS}$ 

Details of the regression analysis, which used Northerly and Southerly Stream 15minute discharge data collected prior to the storm, are provided in Appendix 3-5. Although one composite sample from this period was compromised by severe overfilling the total Hg concentration (50,000 ng/L) was retained for loading calculation. A grab sample collected on June 22 was down to1200 ng/L unfiltered Hg while the discharge was 346 gpm. As discussed subsequently, including or excluding the loading value (1700 grams) for the weekly composite had a significant effect on the average Hg loading of the Southerly Stream.



Figure 3-11. Discharge at Northerly Stream and the number of composite subsamples collected per day.

Tables 3-4 and 3-5 provide complete data for cumulative discharge, Hg concentration and loading values derived from the composite sampling program on the Southerly and Northerly Streams. Composite sample concentrations ranged from 2,150 to 47,200 ng/L for Northerly Stream and from 213 to 50,000 ng/L for Southerly Stream (Figure 3-12, Table 3-6). Total Hg concentrations in composite samples from Northerly Stream greatly exceeded those in samples from Southerly Stream on most occasions (Figure 3-12). Loading (grams/day) is the product of cumulative discharge (gallons) for each composite interval and concentration (ng/L) in the composite sample divided by the number of days of compositing. Total loading (grams) for the period of monitoring is the sum of the individual loads as measured by each composite. Figures 3-14 and 3-15 illustrate the average daily loading for each composite period. A grand average daily loading (grams/day) was calculated by dividing the total load by the total number of days.

Average loading values are highly influenced by the very high estimated loading that occurred on both streams due to the June 2009 storm, accounting for 90% of the total load on Southerly Stream and 17% of that on Northerly Stream (Table 3-6). Observing Hg transport during storm events was the major objective of this study. Other studies

have shown that the majority of pollutant transport in streamflow occurs during these rare but important events, and this was the case for both the Northerly and Southerly Stream.

Analysis of the loading data using the cumulative frequency approach (Figure 3-16) also revealed that more than 80% of the time the loading from the Northerly Stream was greater than that for the Southerly Stream. The use of cumulative frequency curves is a better way of representing loading for these two streams than the use of averages.



Figure 3-12. Discharge at Southerly Stream and the number of composite subsamples collected per day. Discharge for period June 22, 2009 through July 9, 2008 was "reconstructed" by regression analysis using data from the Northerly Stream. "Questioned" flow rates are those where either a backwater condition or ice existed.



Figure 3-13. Total Hg concentrations in composite samples from streams.

Table 3-4: [	Table 3-4: Discharge and Total Hg Loading at Northerly Stream, April 2009 – June 2010								
Sample Collection Date	Rainfall (in)	Cumulative Flow (gallons)	Volume /Period (gallons)	Volume /Day (gallons)	Mean flow (gpm)	[Total Hg] ng/L	Total Hg Load/sample period (grams)	Days /sample period	Total Hg Loading (g/day)
04/21/09	0.42	277,220	277,220	69,305	48	2,515	2.639	4	0.660
04/24/09	1.75	476,990	199,770	66,590	46	4,710	3.561	3	1.187
04/29/09	0.01	716,290	239,300	47,860	33	3,510	3.179	5	0.636
05/06/09	0.52	970,950	254,660	36,380	25	5,215	5.027	7	0.718
05/13/09	1.10	1,209,280	238,330	34,047	24	9,210	8.308	7	1.187
05/20/09	0.62	1,407,770	198,490	28,356	20	8,935	6.713	7	0.959
05/26/09	0.05	1,548,550	140,780	23,463	16	12,300	6.554	6	1.092
06/01/09	1.25	1,694,990	146,440	29,288	20	11,500	6.374	5	1.275
06/08/09	0.01	1,816,770	121,780	17,397	12	13,600	6.269	7	0.896
06/12/09	1.60	1,924,460	107,690	26,923	19	44,300	18.057	4	4.514
06/18/09	0.42	2,044,600	120,140	20,023	14	14,000	6.366	6	1.061
06/22/09	4.75	2,822,080	777,480	194,370	135	22,950	67.536	4	16.884
06/25/09	0.44	3,069,740	247,660	82,553	57	5,400	5.062	3	1.687
07/01/09	0.84	3,373,470	303,730	50,622	35	6,170	7.093	6	1.182
07/07/09	1.60	3,707,970	334,500	55,750	39	11,900	15.066	6	2.511
07/15/09	1.10	4,074,220	366,250	45,781	32	5,170	7.167	8	0.896
07/24/09	1.25	4,394,040	319,820	35,536	25	6,440	7.796	9	0.866
07/30/09	0.70	4,574,270	180,230	25,747	18	5,100	3.479	7	0.497
08/06/09	0.70	4,856,070	281,800	40,257	28	8,870	9.461	7	1.352

Table 3-4: Discharge and Total Hg Loading at Northerly Stream, April 2009 – June 2010									
Sample Collection Date	Rainfall (in)	Cumulative Flow (gallons)	Volume /Period (gallons)	Volume /Day (gallons)	Mean flow (gpm)	[Total Hg] ng/L	Total Hg Load/sample period (grams)	Days /sample period	Total Hg Loading (g/day)
08/14/09	0.01	5,063,800	207,730	25,966	18	3,140	2.469	8	0.309
08/20/09	0.00	5,215,460	151,660	25,277	18	4,850	2.784	6	0.464
08/31/09	2.80	5,460,180	244,720	22,247	15	16,100	14.913	11	1.356
09/08/09	0.00	5,584,580	124,400	15,550	11	7,180	3.381	8	0.423
09/14/09	0.02	5,647,100	62,520	10,420	7	7,460	1.765	6	0.294
09/23/09	0.02	5,699,700	52,600	5,844	4	3,350	0.667	9	0.074
09/29/09	1.22	5,735,010	35,310	5,885	4	31,500	4.210	6	0.702
10/05/09	1.75	5,835,600	100,590	16,765	12	24,700	9.404	6	1.567
10/12/09	1.00	5,935,340	99,740	14,249	10	2,150	0.812	7	0.116
10/16/09	0.46	5,988,650	53,310	13,328	9	2,190	0.442	4	0.110
10/23/09	0.53	6,072,770	84,120	12,017	8	2,380	0.758	7	0.108
10/27/09	1.65	6,216,620	143,850	35,963	25	7,810	4.252	4	1.063
11/04/09	0.13	6,380,810	164,190	20,524	14	3,080	1.914	8	0.239
11/12/09	0.08	6,491,360	110,550	13,819	10	3,460	1.448	8	0.181
11/18/09	2.60	6,760,850	380,040	63,340	44	8,100	11.651	6	1.942
11/25/09	0.44	6,967,860	207,010	29,573	21	4,410	3.455	7	0.494
11/30/09	1.25	7,156,600	188,740	37,748	26	6,430	4.593	5	0.919
12/07/09	1.50	7,619,550	462,950	66,136	46	7,720	13.527	7	1.932
12/11/09	1.05	7,796,890	177,340	44,335	31	2,900	1.947	4	0.487

Table 3-4: Discharge and Total Hg Loading at Northerly Stream, April 2009 – June 2010									
Sample Collection Date	Rainfall (in)	Cumulative Flow (gallons)	Volume /Period (gallons)	Volume /Day (gallons)	Mean flow (gpm)	[Total Hg] ng/L	Total Hg Load/sample period (grams)	Days /sample period	Total Hg Loading (g/day)
12/17/09	0.70	8,045,150	248,260	41,377	29	4,880	4.586	6	0.764
03/12/10	0.00	11,798,400	109,550	36,517	25	3,800	1.576	3	0.525
03/19/10	0.00	11,998,560	200,160	28,594	20	2,780	2.106	7	0.301
03/26/10	2.15	12,279,880	281,320	40,189	28	6,650	7.081	7	1.012
04/02/10	2.80	12,932,080	652,200	93,171	65	9,845	24.303	7	3.472
04/08/10	0.46	13,289,010	356,930	59,488	41	4,390	5.931	6	0.988
04/20/10	0.82	13,822,820	533,810	44,484	31	4,025	8.132	12	0.678
04/27/10	0.30	14,019,740	196,920	28,131	20	5,220	3.891	7	0.556
05/06/10	0.38	14,239,180	219,440	24,382	17	10,200	8.472	9	0.941
05/11/10	0.62	14,337,050	97,870	19,574	14	11,400	4.223	5	0.845
05/18/10	0.10	14,434,890	97,840	13,977	10	15,600	5.777	7	0.825
05/25/10	0.62	14,527,400	92,510	13,216	9	23,500	8.229	7	1.176
06/01/10	0.26	14,593,390	65,990	9,427	7	30,200	7.543	7	1.078
06/05/10	0.84	14,664,620	71,230	17,808	12	38,400	10.353	4	2.588
06/07/10	0.80	14,705,775	41,155	20,578	14	47,200	7.352	2	3.676
06/17/10	0.56	14,746,880	41,105	4,111	3	13,850	2.155	10	0.215
	Total Loading 4/17/2009 thru 6/17/2010 = 392 grams							Mean = 1.	12 g/d



Figure 3-14. Time series plot of Hg loading at Northerly Stream

Table 3-5: Discharge and Total Hg Loading at Southerly Stream, April 2009 – June 2010								
Sample Collection Date	Cumulative Flow (gallons)	Volume /Period (gallons)	Volume/Day (gallons)	Mean flow (gpm)	[Total Hg] ng/L	Total Hg Load/sample period (grams)	Days /sample period	Total Hg Loading (g/day)
04/21/09	670,900	670,900	167,725	116	437	1.110	4	0.277
04/24/09	2,735,300	2,064,400	688,133	478	3,080	24.066	3	8.022
04/29/09	3,617,300	882,000	176,400	123	266	0.888	5	0.178
05/06/09	4,206,200	588,900	84,129	58	337	0.751	7	0.107
05/13/09	5,133,600	927,400	132,486	92	540	1.896	7	0.271
05/20/09	5,623,400	489,800	69,971	49	339	0.628	7	0.090
05/26/09	5,884,000	260,600	43,433	30	556	0.548	6	0.091
06/01/09	6,189,700	305,700	61,140	42	637	0.737	5	0.147
06/08/09	6,479,900	290,200	41,457	29	859	0.944	7	0.135
06/12/09	6,818,100	338,200	84,550	59	3,220	4.122	4	1.030
06/18/09	7,192,300	374,200	62,367	43	377	0.534	6	0.089
06/22/09		9,041,811	2,260,453	1,570	50,000	1711	4	427.
06/25/09	74,496,900	1,383,543	461,181	320	1,190	6.232	3	2.077
07/01/09	77,581,100	1,244,078	207,346	144	1,140	5.368	6	0.895
07/07/09	83,126,500	1,627,769	271,295	188	1,940	11.953	6	1.992
07/09/09	84,886,900	428,580	214,290	149	824	1.337	2	0.668
07/15/09	85,425,700	502,800	100,560	70	567	1.079	5	0.216
07/24/09	85,836,500	410,800	45,644	32	340	0.529	9	0.059

Table 3-5: Discharge and Total Hg Loading at Southerly Stream, April 2009 – June 2010								
Sample Collection Date	Cumulative Flow (gallons)	Volume /Period (gallons)	Volume/Day (gallons)	Mean flow (gpm)	[Total Hg] ng/L	Total Hg Load/sample period (grams)	Days /sample period	Total Hg Loading (g/day)
07/30/09	86,232,100	395,600	56,514	39	773	1.157	7	0.165
08/06/09	86,547,600	315,500	45,071	31	4,310	5.147	7	0.735
08/14/09	86,644,400	96,800	12,100	8	286	0.105	8	0.013
08/20/09	86,702,600	58,200	9,700	7	213	0.047	6	0.008
08/31/09	87,122,500	419,900	38,173	27	2,880	4.577	11	0.416
09/09/09	87,212,100	89,600	9,956	7	530	0.180	9	0.020
09/14/09	87,232,500	20,400	4,080	3	851	0.066	5	0.013
09/23/09	87,254,700	22,200	2,467	2	617	0.052	9	0.006
09/29/09	87,311,100	56,400	9,400	7	2,480	0.529	6	0.088
10/05/09	87,628,500	317,400	52,900	37	14,500	17.420	6	2.903
10/12/09	87,870,100	241,600	34,514	24	554	0.507	7	0.072
10/16/09	87,980,600	110,500	27,625	19	638	0.267	4	0.067
10/23/09	88,127,200	146,600	20,943	15	534	0.296	7	0.042
10/27/09	88,721,800	594,600	148,650	103	3,830	8.620	4	2.155
11/04/09	88,927,300	205,500	25,688	18	1,280	0.996	8	0.124
11/12/09	89,076,200	148,900	18,613	13	501	0.282	8	0.035
11/18/09	90,254,600	1,178,400	196,400	136	5,920	26.405	6	4.401
11/25/09	90,594,800	340,200	48,600	34	816	1.051	7	0.150
11/30/09	91,272,400	677,600	135,520	94	1,890	4.847	5	0.969

Table 3-5: D	Table 3-5: Discharge and Total Hg Loading at Southerly Stream, April 2009 – June 2010							
Sample Collection Date	Cumulative Flow (gallons)	Volume /Period (gallons)	Volume/Day (gallons)	Mean flow (gpm)	[Total Hg] ng/L	Total Hg Load/sample period (grams)	Days /sample period	Total Hg Loading (g/day)
12/07/09	92,788,400	1,516,000	216,571	150	1,800	10.329	7	1.476
12/11/09	94,158,415	927129	231,782	161	758	2.660	4	0.665
03/19/10	111,019,400	745,100	74,510	52	644	1.816	10	0.182
03/27/10	112,810,300	1,790,900	223,863	155	1,120	7.592	8	0.949
03/30/10	114,226,600	1,416,300	472,100	328	2,640	14.152	3	4.717
04/02/10	116,670,400	2,443,800	814,600	566	907	8.390	3	2.797
04/08/10	117,945,400	1,275,000	212,500	148	683	3.296	6	0.549
04/20/10	119,679,200	1,733,800	144,483	100	1,070	7.022	12	0.585
04/27/10	120,014,300	335,100	47,871	33	1,790	2.270	7	0.324
05/06/10	120,290,100	275,800	30,644	21	870	0.908	9	0.101
05/11/10	120,413,600	123,500	24,700	17	1,100	0.514	5	0.103
05/18/10	120,497,500	83,900	11,986	8	517	0.164	7	0.023
05/25/10	120,611,100	113,600	16,229	11	1,290	0.555	7	0.079
06/01/10	120,667,800	56,700	8,100	6	876	0.188	7	0.027
06/05/10	120,735,400	67,600	16,900	12	3,190	0.816	4	0.204
06/07/10	120,875,700	140,300	70,150	49	2,910	1.545	2	0.773
06/17/10	121,086,600	210,900	21,090	15	634	0.506	10	0.051
	Total Loading 4/17/2009 thru 6/17/2010 = 1909 grams Mean = 5.66 g/d							



Figure 3-15. Time series plot of Hg loading at Southerly Stream

Table 3-6: Summary discharge (15-minute average), composite concentration and loading data.							
	Northerly Stream	Southerly Stream					
Discharge Mean (gpm)	24	73					
Discharge Minimum (gpm)	1.3	0.5					
Discharge Maximum (gpm)	1301	26319					
Concentration Mean (ng/L)	10800	2460					
Concentration Minimum (ng/L)	2150	213					
Concentration Maximum (ng/L)	47200	50000					
Loading Total (g) <sup>a</sup>	392 (17%) <sup>b</sup>	1909 (90%) <sup>b</sup>					
Loading Mean (g/day)	1.12	5.66					
Loading Median (g/day)	0.90	0.18					
Loading Minimum (g/day)	0.074	0.006					
Loading Maximum (g/day)	17	428					
<sup>a</sup> Total for period April 21, 2009 throug <sup>b</sup> Percentage due to June 19-20, 2009	h June 17, 2010 storm flow event)						



Figure 3-16. Comparison of cumulative frequencies of Hg loading from two surface streams draining the HoltraChem site.

#### **Grab sampling** 5.2

Complete results of the monthly grab sampling program are given in Appendix 3-4 and summarized below in Table 3-7 and Figures 3-17 through 3-19. There was no

expectation that the grab sampling program would generate mean unfiltered Hg concentrations for NS-DN, SS-PIPE and SS-PIPEN that would be similar to the mean unfiltered concentrations derived from the flow-proportional composite sampling (Table 3-6). Nonetheless the means from each program were of similar order of magnitude and ratio to each other (grab vs. composite - NS-DN: 5807 vs 10800 ng/L; SS-PIPEN:1690 vs 2460 ng/L). The higher concentrations in the composite sample sets are consistent with higher mean discharges being included in the composite sets.

The purpose of the grab sampling program was to investigate partitioning of Hg between filter-passing (dissolved) and particulate phases and to assess the Hg content of the suspended phase. The fraction of Hg that was filter-passing was quite variable on both streams as indicated by the standard deviations, and ranged from <1% to 68%. In general the mean fraction that was filter-passing was slightly greater on Northerly Stream ( $29 \pm 18\%$ ) than on Southerly Stream ( $22 \pm 17\%$ ) but the difference was not significant. Higher fractions of filter-passing Hg tended to occur during late summer, fall and winter when TSS concentrations were lower. While there was no significant correlation between particulate Hg (ng/L) and stream flow rates on either stream, higher particulate concentrations generally occurred at the higher flow rates.

Hg on suspended solids (TSS-Hg) carried by the two surface streams varied from 42 to 1862  $\mu$ g/g dry wt. (Table 3-7, Figure 3-19) with TSS-Hg on Northerly Stream averaging much higher (557 ± 405  $\mu$ g/g dry wt.) than Southerly Stream (110 ± 64  $\mu$ g/g dry wt.). Higher flow rates were associated with somewhat lower and more stable TSS-Hg concentrations on both streams. Background soil and sediment concentrations are generally <0.5  $\mu$ g/g and thus these suspended solids values are quite elevated above expected background and also much higher than measured in suspended solids in the Penobscot River in the vicinity and downstream of the HoltraChem site (0.5 to 2  $\mu$ g/g dry wt., Figure 16 in PRMS 2008).

Additional grab samples were collected downstream of SS-PIPEN at SS-DN at the location where Southerly Stream enters Southerly Cove. A paired-samples t-test was conducted to compare unfiltered and filtered Hg concentrations in grab samples from SS-PIPEN and SS-DN. As noted the latter station is closer to the Penobscot River and included runoff from both SS-PIPEN and PERC. There was not a significant difference (N=15, P>0.05) in either unfiltered Hg (means=880, 930 ng/L) or filtered Hg (means=123, 113 ng/L) concentrations between these monitoring locations. This suggests that any inputs of Hg, e.g., from groundwater, below the continuously monitored location at SS-PIPEN, or via runoff from the PERC site, were insufficient to alter concentrations of Hg entering the Penobscot River at SS-DN but would have resulted in a higher loading due to higher discharge and non-zero Hg concentrations, e.g., from the PERC site. Runoff from the PERC site would have increased discharge at SS-DN but the relatively low concentrations of Hg (Unfiltered=57 + 61 ng/L, Filtered=5 + 2 ng/L) in this runoff would probably not have added significantly to the loading from the Southerly Stream at its point of discharge to the river. In the absence of discharge data for SS-DN it is not possible to estimate any contribution of groundwater inputs downstream of SS-PIPEN. This means only that Hg loading to the Penobscot River from Southerly Stream, as calculated from the SS-PIPEN discharge and concentration
data, is likely to be slightly underestimated because it does not include groundwater inputs to the stream below the monitored location but above the river.

Table 3-7: Summary of results of grab sampling of surfacestreams.									
Location	NS-DN	SS-PIPEN <sup>b</sup>	SS-DN	PERC					
Unfiltered Hg (ng/L)									
N	19	19	15	11					
Mean	5807	1690	770	56.6					
Standard Deviation	9805	2836	786	60.8					
Minimum	104	126	51.4	10.1					
Maximum	38000	11900	2480	215					
		Filtered Hg (ng/	L)						
N	18	18	13	11					
Mean	660	118	115	4.79					
Standard Deviation	334	71.9	205	2.39					
Minimum	48.4	46.5	17.8	2.51					
Maximum	1470	287	837	9.09					
	% F	ilter-passing (dis	solved)						
N	18	18	13	11					
Mean	29	22	18	14					
	18	17	14	8.4					
Minimum	1.5	0.9	2.0	4.0					
Maximum	68	57	43	26					
		TSS (mg/L)							
N	17	17	13	11					
Mean	17.3	22.0	15.8	24.2					
Standard Deviation	28.1	38.2	14.1	20.9					
Min	0.6	0.3	1.5	3.5					
Max	95.0	148	50.5	74.4					

Table 3-7: Summary of results of grab sampling of surface streams.					
Location	NS-DN	SS-PIPEN <sup>b</sup>	SS-DN	PERC	
		TSS-Hg (μg/g dw	/)	·	
Ν	17	17	12	11	
Mean	557	110	109	1.9	
Standard Deviation	405	64	212	0.61	
Min	52	42	12	0.83	
Max	1862	290	830	2.8	
	-	Discharge (gpm)	) <sup>a</sup>		
N	22	17			
Mean	45	251			
Standard Deviation	43	314			
Min	2.9	2.0			
Max	150	909			
<sup>a</sup> Discharge at ti	me of sampli	ng. <sup>b</sup> Includes data fro	m SS-PIPE	•	



Figure 3-17. Hg and TSS in Northerly Stream grab Samples



Figure 3-18. Hg and TSS in Southerly Stream grab samples



Figure 3-19. Hg on suspended solids (TSS) in grab samples.

# 6 GROUNDWATER DISCHARGE TO PENOBSCOT RIVER

The following sections present and discuss seepage meter and radon results obtained in March and September 2009.

Seepage Meters (March 2009) – Locations of seepage meters for this time period are shown in Figure 3-20. Flow determined via seepage meters in March was in the range of 1.0-56 cm day<sup>-1</sup> with a grand average (average of the mean flow at each meter) of 22 cm day<sup>-1</sup> (Table 3-8). Flow was highest in the middle of the along shore transect at sites SM2 and SM3 (20 and 30 cm day<sup>-1</sup>, respectively); these locations were river-ward of a large (10 m wide by 30 m long) seep observed emanating from the beach face during low tide. On both sides of this visible seepage zone, groundwater flow was significantly lower ranging between 4.1 and 9.6 cm day<sup>-1</sup> on average (sites SM1 and SM4). At the shore perpendicular transect, located in line with the meter at site SM2, only meter 5

yielded reliable results as meter 6 was exposed by the ebbing tide prematurely by the rapidly receding tide. At site 5, flow ranged from 32-56 cm day<sup>-1</sup>, for measurements that were made during the outgoing tide. Such high values are consistent with the high flows observed along this section of the river at sites SM2 and SM3 and the nearby visible seep. In all, these seepage rates are typical or slightly elevated in comparison with studies at other river systems where groundwater discharge is known to be important (Burnett et al. 2006).

Due to extreme tidal ranges and water temperatures, seepage meter bags were not exchanged on regular intervals other than at low tide. Therefore, the majority of the data is either from a 24-hour deployment or a 15-30 minute deployment collected at or near low tide. We were unable to determine from the seepage meter data that river water level was a controlling factor in groundwater flow rate (i.e., due to a change in hydraulic gradient between river level and the aquifer water table). However, at site SM2 where three time points are available, the 24-hr measurement was 6.1 cm day<sup>-1</sup>, a factor of 4-5 times lower than the two low tide measurements. The large change in water level is more than likely large enough to change the hydraulic gradient and explain this difference between daily averages and low tide measurements. The range in groundwater flow rates observed with the seepage meters is testament to the patchy nature of groundwater flow combined with the fact that the meters are measuring flow over a relatively small area (~0.25 m<sup>2</sup>) of river bottom; inhomogeneity in sediment permeability on this spatial scale are usually the cause of such variability.



Figure 3-20. Map of study area with seepage meter locations (round symbols), radon monitor time series station (square symbol), and groundwater radon sampling stations (diamond symbol) for the March 2009 study. For reference the Landfill Area 1 is shown in the background.

	Date and	Date and	Deployment	Seepage
ID	Time On	Time Off	Time (hr)	Rate (cm/d)
SM1	3/22/09 16:00	3/22/09 16:30	0.5	6.4
	3/22/09 16:35	3/23/09 17:10	25	1.0
	3/24/09 16:00	3/24/09 16:57	1.0	25
	3/24/09 17:00	3/24/09 17:53	0.9	7.1
	3/25/09 16:00	3/25/09 16:23	0.4	9.3
	3/25/09 16:25	3/25/09 16:44	0.3	8.5
	3/25/09 16:48			exposed by tide
SM2	3/22/09 15:45	3/22/09 16:15	0.5	31
	3/22/09 16:20	3/22/09 16:50	0.5	24
	3/22/09 16:53	3/23/09 16:45	24	6.1
	3/23/09 16:50			lost down river
SM3	3/22/09 16:05	3/22/09 16:36	0.5	22
	3/22/09 16:40	3/22/09 16:54	0.2	37
	3/23/09 16:55			lost down river
SM4	3/22/09 16:08	3/22/09 16:42	0.6	4.1
	3/22/09 16:43			lost down river
SM5	3/25/09 15:20	3/25/09 15:41	0.3	56
	3/25/09 15:44	3/25/09 15:55	0.2	32
	3/25/09 15:58			exposed by tide
SM6	3/25/09 15:05			exposed by tide

 Table 3-8: Seepage meter data for March 2009.

Seepage Meters (September 2009) - Locations of seepage meters for this time period are shown in Figure 3-21. The September seepage meter data for 22-25 hr monitoring was in the range of 0.15-2.8 cm day<sup>-1</sup> with a grand average (average of the mean flow at each meter) of 0.99 cm day<sup>-1</sup> (Table 3-9). Flow was highest at the southern end of the along shore transect at sites SM10 and SM11 (Figure 3-21) with averages of 1.3 and 2.6 cm day<sup>-1</sup>, respectively; unlike March, these locations were to the south of the large seepage zone emanating from the beach face during low tide. To the north of the high seepage meter readings, groundwater flow was significantly lower ranging between 0.30 and 0.40 cm day<sup>-1</sup> (average for sites SM12 and SM13, respectively). At the shore perpendicular transect, located in line with the meter at site SM12, seepage increased in a landward direction: 0.15, 0.33, 4.9, 3.6, and 4.7 cm day<sup>-1</sup> (SM13-SM23 Table 9). Because of their position in the upper intertidal, meters SM21-SM23 could be used to obtain only short-term seepage rates. Seepage continued after these positions were exposed by the ebbing tide. Inclusion of seepage rates for these positions in the grand average yields 2.7 cm day<sup>-1</sup>.

Based on visual evidence as well as the measurements from the seepage meters, we surmise that the majority of the groundwater discharge originates from the intertidal zone but we cannot unequivocally rule out subtidal groundwater discharge with this data set (see later discussion of subtidal sampling). Due to technical complications associated with ice floes, few daily-integrated seepage measurements were available for the March study. Hence, we are unable to determine with the seepage method if

there are statistically significant differences between spring and late summer 24 hour groundwater discharges at the HoltraChem site.



Figure 3-21. Seepage meter and radon time series locations for the September 2009 study.

	Date and	Date and	Deployment	Seepage
ID	Time On	Time Off	Time (hr)	Rate (cm/d)
SM10	9/16/09 15:25	9/17/09 15:52	24	1.4
	9/17/09 16:04	9/18/09 16:39	25	1.2
SM11	9/16/09 15:28	9/17/09 15:11	24	2.8
	9/17/09 15:33	9/18/09 16:44	25	2.4
SM12	9/16/09 15:36	9/17/09 15:24	24	0.44
	9/17/09 15:53			Bag Lost
	9/18/09 18:03	9/19/09 17:04	23	0.15
SM13	9/16/09 15:29	9/17/09 15:34	24	0.33
	9/17/09 15:42	9/18/09 16:53	25	0.47
SM14	9/16/09 15:32	9/17/09 15:43	24	0.42
	9/17/09 15:41	9/18/09 16:54	25	0.25
SM20	9/18/09 18:40	9/19/09 16:33	22	0.33
SM21	9/19/09 15:15	9/19/09 15:45	0.5	4.9
SM22	9/19/09 15:30	9/19/09 16:01	0.5	3.6
SM23	9/19/09 15:59	9/19/09 16:30	0.5	4.7

Table 3-9 <sup>.</sup> Seenage	meter	data for	Septembe	r 2009
Table J-J. Occhage	Inclui	uata ioi	Ocptenibe	2003.

*Radon (March 2009)* - For the two days in March when the radon monitor was deployed for a full 8-hour interval, radon-222 concentrations from the continuous radon monitor ranged from 2.5-8.9 dpm L<sup>-1</sup> and averaged 4.1 dpm L<sup>-1</sup>. On both days there was trend of increasing radon with decreasing water level (Figure 3-22), which could be due to increasing groundwater seepage as a result of an increase in the hydraulic gradient as the water level in the river recedes. In addition to the trend with water level, wind was likely an overarching factor in the average radon concentration difference between day 1 and 2; as a noble gas, radon is subject to river-air gas transfer, a process that is well known to be a function of wind speed. On day 1 (avg. Rn = 3.0 dpm/L), wind speed averaged 3.4 m/s compared with 2.2 m/s on day two (avg. Rn = 5.4 dpm/L).

Upland groundwater radon ranged from 66-2800 dpm L<sup>-1</sup> with an average of 1200 dpm L<sup>-1</sup> (Figure 3-23, Table 3-10, n=16). These upland data were not ultimately used in calculations of groundwater flux. Instead, intertidal groundwater samples taken from the beach seepages and directly beneath the beach face using push point piezometers (Figure 3-20, 3-Table 10 "piezo" values), were used to calculate how much groundwater is needed to explain the radon levels observed in the river. We assumed the following: (1) radon loss to the atmosphere is driven by wind speed, and (2) the radon excess was driven by the intertidal groundwater concentrations (average = 160 dpm L<sup>-1</sup>). Given these assumptions, the model requires an average groundwater seepage rate of 2.7 cm day<sup>-1</sup> over the two 8 hour deployments (range = 0.9-6.1 cm day<sup>-1</sup>; Figure 3-20). This result is in excellent agreement with the two March seepage meter samples that encompassed a full tidal cycle (Table 3-8: SM1, SM2, 1.0-6.1 cm day<sup>-1</sup>), which is encouraging given that these are two completely independent techniques.



Figure 3-22. River water level (A), river radon concentrations (B), and radon-derived groundwater seepage (C) during the course of the March 2009 experiment.

	Sampling Date	Radon
ID	and Time	(dpm/L)
B304-B1	3/23/09 12:00	514
B304-O1	3/23/09 12:50	392
B307-B2	3/23/09 16:00	1611
B321-B2	3/24/09 11:00	2786
B326-O3	3/23/09 12:30	777
MW402-01	3/23/09 14:30	1091
MW501-01	3/24/09 13:30	1482
MW505-B1	3/24/09 13:45	2675
MW505-B2	3/24/09 13:00	822
MW512-01	3/23/09 11:45	1250
MW513-01	3/23/09 13:45	1795
P13	3/24/09 11:55	66
P2A	3/23/09 17:10	2808
GW1 (piezo.)	3/23/09 16:00	151
GW2 (piezo.)	3/23/09 16:35	151
GW3 (seep)	3/23/09 17:30	190

 Table 3-10:
 Upland groundwater radon concentrations for March 2009.

*Radon (September 2009)* – During the three-day record in September, radon-222 concentrations from the continuous radon monitor (see Figure 3-20) ranged from 1.6-19 dpm L<sup>-1</sup> and averaged 6.0 dpm L<sup>-1</sup>. On all days there was a trend of increasing radon with decreasing water level (Figure 3-24), similar to the trend observed for March (Figure 3-22). In addition, the early morning negative tides produced radon maxima as much as 50% higher than for the late afternoon low. This further bolsters the seepage meter observations of the importance of the intertidal zone for groundwater discharge: negative tides result in an increased time interval for accumulation of discharges from this zone within the river.

Radon in intertidal groundwater ranged from 24-900 dpm L<sup>-1</sup> with an average of 250 dpm L<sup>-1</sup> (Figure 3-25, Table 3-11, n=29). In general, groundwater radon increased from south to north along the seepage zone. In order to determine whether or not there is a groundwater component to stream flow on the site, we collected radon samples at Northerly Stream and Southerly Stream (NS and SS in Table 3-11). If a radon value of 250 dpm L<sup>-1</sup> is considered pure groundwater, then Northerly Stream flow is fed by groundwater during this time period. A suspected spring discharge in the lower reaches of South Stream is nearly pure overland flow according to its radon content (SS Spring, Table 3-11).



Figure 3-23. Groundwater monitoring wells used to obtain samples for radon analysis, March 2009.

	Sampling Date	Radon
ID	and Time	(dpm/L)
HOLC 1	9/16/09 13:53	71
HOLC 2	9/16/09 14:33	125
HOLC 3	9/16/09 14:25	112
NS	9/17/09 11:06	5
SS	9/17/09 13:40	39
SS Spring	9/17/09 13:48	14
PW1	9/17/09 14:45	26
PW2	9/17/09 15:05	49
PW3	9/17/09 15:35	200
PW4	9/17/09 15:55	295
PW5	9/17/09 16:10	179
PW6	9/17/09 16:30	165
PW7	9/17/09 16:55	100
PW8	9/17/09 17:25	388
PW9	9/17/09 17:50	461
PW10	9/18/09 15:30	279
PW11	9/18/09 15:55	122
PW12	9/18/09 16:20	139
PW13	9/18/09 16:45	191
PW14	9/18/09 17:10	24
PW15	9/18/09 17:35	899
PW16	9/18/09 17:55	237
PW17	9/18/09 18:25	500
TS1	9/18/09 14:35	66
TS2	9/18/09 15:08	163
TS3	9/18/09 15:30	227
TS4	9/18/09 16:00	238
TS5	9/18/09 16:30	297
TS6	9/18/09 17:04	311
TS7	9/18/09 17:32	326
TS8	9/18/09 18:07	492
TS9	9/18/09 18:40	407

 Table 3-11:
 Groundwater radon concentrations for September 2009.



Figure 3-25. Locations of intertidal seepage and porewater sampling sites for radon and total Hg concentrations, September 2009. Images overlap and are not at same scale.

Given the large amount of flow observed emanating from the intertidal zone of the study area, we assumed some fraction must be recycled river water, i.e. water that fills the pore spaces of the river bank sediments during high tide and drains during low tide. If this is true, the recirculated river water will carry a lower radon concentration in comparison to a pure groundwater end member. To test this idea, we installed a well point into the center of the seepage zone and collected radon samples every 30 minutes during a falling tide (Figure 3-26). Consistent with our theory, radon was low in the initial water pulse after high tide (66 dpmL-1) and increased steadily to maximum of ~500 dpmL-1 four hours later (see TS1-8, Table 11). Hence for this location early discharge from the seepage zone may contain as much as 85% recycled river water. Analysis of the same sample series for Hg showed a low and near constant concentration (7 to 9 ng/L) over the period that radon was increasing, suggesting that while the sampling probe was intersecting site groundwater it was not capturing the Hg plume from Landfill 1.

Using the same box model and assumptions (except for the average groundwater radon) for the March radon time series, we calculate an average groundwater seepage rate of 4.1 cm day<sup>-1</sup> over the 72 hour deployment (range = 0.8-14 cm day<sup>-1</sup>). Groundwater discharge is highest at the negative low tides and lowest at all high tides, consistent with a river water level control on the land-sea hydraulic gradient. This value is in good agreement with the weighted seepage meter average of 2.7 cm day<sup>-1</sup>. The radon-derived rate for March was lower (2.7 cm day<sup>-1</sup>), however, this average does not include the negative tides as ice cover on the river precluded 24-hour deployments. Hence, we are unable to determine with certainty whether or not the seasonal difference is significant.



Figure 3-24. River water level (A), river radon concentrations (B), and radon-derived groundwater seepage (C) during the course of the September 2009 experiment.



Figure 3-26. Time series of radon (diamonds) and tidal height (squares) for groundwater collected from the seepage face bordering the HoltraChem site.

In conclusion, two independent techniques were used to quantify groundwater seepage through the bottom sediment of the Penobscot River bordering the Holtrachem site. The seepage meters indicated that mean daily flow was 3.6 cm day<sup>-1</sup> for March and 2.7 cm day<sup>-1</sup> for September. Meters deployed for shorter intervals around the time of low tide and within the intertidal zone yielded significantly higher seepage rates, a likely function of a higher hydraulic gradient between the aquifer and river level. The radon derived seepage rate was 2.7 cm day<sup>-1</sup> for March and 4.1 cm day<sup>-1</sup> for September. The September record covered three 24-hour time intervals, which revealed 50% higher flow during negative tides.

The total shoreline length bordering the HoltraChem property is approximately 1000 m, of which 300 m consisted of a gently sloping silty sand and gravel beach. The other 700 m consists of a relatively steep rock wall with one narrow beach located below Landfills 3, 4 and 5. If we assume that groundwater seepage occurs over a 25 m wide seepage face along the gently sloping beach below Landfill 1 and over a5 meter wide seepage face over 700 m, then the effective seepage zone is 11,000 m<sup>2</sup>. Combined with the two independently quantified seepage measurements, the volumetric groundwater flow is in the range of 299-396 m<sup>3</sup> day<sup>-1</sup> for March and 299-440 m<sup>3</sup> day<sup>-1</sup> for September. As discussed in the next section, average Hg concentrations for intertidal seepage and sediment porewater were used in conjunction with these groundwater discharge values, to estimate the groundwater-derived Hg flux from this site to the Penobscot River.

Groundwater seepage is driven by a number of forcing functions including water level in the aquifer and river. The differences in groundwater discharge between March and September 2009 when we expected that the summer dry season would lead to lower aquifer water inventory, and therefore reduced groundwater seepage to the river, were not evident. This may be due in part to a wetter-than-average summer and/or the reduced temporal coverage of our March samples due to ice conditions on the river (we were not able to capture the negative tides that resulted in 50% higher groundwater flux

during September). Because of these uncertainties related to seasonality and/or incomplete data it is conservative to assume groundwater discharge could be 100% higher on an annual basis than the values derived from the available data. Thus we used 900 m<sup>3</sup>/day as the average for a full year in calculating Hg flux to the river by groundwater.

## 6.1 Mercury Concentrations in Intertidal Seepage and Porewater

Sampling of seepages and porewater for total Hg concentrations was focused mainly on the intertidal zone immediately downhill from Landfill 1 (Figure 25). This zone is characterized as a sandy gravelly beach with several areas where the substrate is mildly fluidized and/or freely and profusely draining water as the tide recedes.

Earlier (2007) sampling of seepages in two areas (see Figure 3-8, just below Landfill 1 and near Outfall 001) had shown Hg concentrations up to 1880 ng/L (see Table 3-2). In September 2009 the more comprehensive sampling of porewater and seepage did not detect any Hg concentrations greater than 437 ng/L (Tables 3-13 and 3-14). This highest value was observed in the same area where the highest value October 2007 occurred (SEEP3). As shown in Figure 3-27, which summarizes all the seep and porewater data since 2007, there are two areas where higher concentrations occur, immediately below Landfill 1 (near SEEP3, Figure 3-8) and near the submerged outfall pipe (SEEP 1). The beach is locally fluidized near SEEP 1, possibly reflecting submarine discharge from the buried Outfall 001 pipe or a preferred flow path for groundwater due to coarser backfill associated with the two pipes (one from HoltraChem and one from PERC facility) that pass under this area of the beach.

The mean concentration of all the 2007 and 2009 seepage and porewater samples is 125 ng/L, with values ranging widely from <1 to 1880 ng/L. For the purpose of calculating the groundwater flux of Hg, the UCL of the mean (UCL95), 242 ng/L, was multiplied by the estimated daily groundwater discharge volume (900 m<sup>3</sup>) assuming 11,000 m<sup>2</sup> as the area of discharge. Thus, the combined daily loading from the intertidal zone downhill of Landfill 3, 4 and 5 and extending southward into Southerly Cove is estimated at 0.22 g/day, or 82 g/year.

To test the decision include all 1000 m of HoltraChem shoreline in the estimation of groundwater Hg flux, we collected five samples of water seeping out of rock fractures below Landfills 3, 4 and 5 (Figure 3-28). All of these seepages were at or below the high tide elevation and flowing at  $\leq$ 100mL/min. All were sampled at or shortly after low tide to minimize the inclusion of river water. The highest measured unfiltered Hg concentration (Table 3-14) was 5.7 ng/L and the mean was 2.6 ng/L. Thus it appears the annual loading value given above is likely an overestimate.

Table 3-12:         Results of intertidal porewater sampling, September 17-19, 2009							
Station	WPT	Datetime	Temp ©	ElecCond (µS/cm)	Salinity (ppth)	DO (mg/L)	Diss Total Hg (ng/L)
PW-1	494	9/17/2009 14:45	18.82	4866	2.62	3.81	3.27
PW-2	495	9/17/2009 15:05	19.45	5665	2.74	4.98	2.87
PW-3	496	9/17/2009 15:35	19.22	4512	2.42	3.78	8.20
PW-4	497	9/17/2009 15:55	19.2	4445	2.37	3.78	6.07
PW-5	498	9/17/2009 16:10	18.82	4685	2.51	5.81	5.22
PW-6	499	9/17/2009 16:30	19.78	4766	2.56	6.36	3.93
PW-7	500	9/17/2009 16:55	19.27	3893	2.06	5.86	5.59
PW-8	502	9/17/2009 17:25	19.11	2865	1.49	2.56	10.6
PW-9	504	9/17/2009 17:50	18.39	3172	1.66	3.56	9.09
PW-10	505	9/18/2009 15:30	18.24	3387	1.78	4.32	9.66
PW-11	506	9/18/2009 15:55	18.34	3921	2.08	5.92	14.7
PW-12	507	9/18/2009 16:20	18.36	3924	2.08	3.69	7.42
PW-13	508	9/18/2009 16:45	18.02	3849	2.04	1.55	18.9
PW-14	509	9/18/2009 17:10	18.97	3889	2.06	5.97	22.2
PW-15	510	9/18/2009 17:35	18.25	2005	1.03	0.66	290
PW-16	511	9/18/2009 17:55	17.31	3600	1.9	10.17	16.9
PW-17	512	9/18/2009 18:25	18.34	2322	1.2	3.94	11.8
PW-18	514	9/19/2009 16:15	16.72	2012	1.03	5.17	4.46
PW-19	515	9/19/2009 16:35	16.95	2052	1.06	0.20	0.394
PW-20	516	9/19/2009 17:05	16.64	992	0.49	0.28	0.363

Table 13. Results of intertidal seepage sampling, September 19, 2009.								
Station	WPT	Datetime	Temp ©	ElecCond (µS/cm)	Salinity (ppth)	DO (mg/L)	Diss Total Hg (ng/L)	
SEEP 1	517	9/19/2009 17:25	16.57	2613	1.36	7.9	14.7	
SEEP 1A	518	9/19/2009 17:35	15.82	2094	1.08	8.53	11.5	
SEEP 1B	519	9/19/2009 17:45	16.29	2695	1.4	7.72	7.80	
SEEP 2	520	9/19/2009 18:05	15.30	2475	1.28	8.64	14.9	
SEEP 2B	524	9/19/2009 18:45	15.97	2324	1.2	8.26	36.6	
SEEP 3	521	9/19/2009 18:15	15.07	2279	1.18	8.09	134	
SEEP 3A	523	9/19/2009 18:38	15.33	2483	1.29	8.33	36.0	
SEEP 3B	522	9/19/2009 18:25	15.66	1753	0.89	8.01	437	



Figure 3-27. Summary of filter-passing (dissolved) Hg concentrations in intertidal seepage and porewater collected below Landfill 1 between October 2007 and September 2009.



Figure 3-28. Locations of seepage samples collected below Landfills 3, 4 and 5 May 18, 2010

Table 3-14. Concentrations of Hg in unfiltered samples of seepages collected from fractures in rock during final stage of ebbing tide.							
Waypoint	Time	Latitude (NAD83)	Longitude (NAD83)	Total Hg (ng/L)			
637	8:02	44.74520	68.82439	0.80			
638	8:14	44.74439	68.82508	1.34			
639	8:29	44.74366	68.82554	4.41			
640	8:39	44.74323	68.82600	0.91			
641	8:50	44.74240	68.82707	5.67			

#### 6.2 Subtidal Water Sampling to Identify Groundwater Source

Limited surveys (Figure 3-29) of near-bottom specific conductance failed to detect evidence of either seawater intrusion or presence of brine. In general, the specific conductance of near- bottom water was very similar to specific conductance of river water at Veazie Dam (~50  $\mu$ S/cm). The exceptions were for river water collected in July near (within 20 ft) the confluence with Northerly Stream (205  $\mu$ S/cm), Northerly Stream (558  $\mu$ S/cm), and seepage water emanating from the intertidal near the former location of Outfall 001 (1200  $\mu$ S/cm).

During the July nearshore survey, Hg concentrations were uniformly low (2.0 to 2.6 ng/L) except for the same locations that had higher specific conductances, i.e., near Northerly Stream confluence (7.3 ng/L), Northerly Stream (52 ng/L) and seepage at 001 (230 ng/L) (Figure 3-29). The September near-bottom sampling was focused on three subtidal areas: immediately upstream of the beach below Landfill 1, immediately downstream of the former location of Outfall 001 and downstream of Southerly Cove. At each location, up to five samples were collected along transects from nearshore to midriver. As indicated in Figure 3-31, all values were low and no pattern related to location was evident. The October survey was limited to the deepest part of the river channel along the western edge and again revealed no anomalous Hg concentration in nearbottom water (Figure 3-32). Overall, these results suggest that there was no significant subtidal discharge of Hg. Obviously a small subtidal groundwater discharge could have been missed given the spacing between sample points and the high velocities of the river near the bottom at near low tide. The latter condition would tend to quickly disperse and dilute all but the most concentrated brine or Hg discharge. Choosing reasonable parameters<sup>2</sup> to estimate whether such a subtidal discharge could be detected suggested that Hg concentration in bottom water (10 cm interval above interface) would be increased by 1.7 ng/L and thus be detectable against the upstream background of ~4 ng/L.



Figure 3-29. Locations of subtidal sampling of near-bottom water and approximate bathymetry (inset).

<sup>&</sup>lt;sup>2</sup> Assuming area of subtidal discharge = 100 m<sup>2</sup>, groundwater discharge = 3 cm/day (3000 L/day), Hg in discharging groundwater = 0.5 mg/L, mixing depth = 10 cm, river bottom current velocity = 1 m/sec (10 m<sup>3</sup>/sec) yields the following: Hg loading rate = 1.5 g/day (17,360 ng/sec), Hg concentration in bottom water = 1736 ng/m<sup>3</sup> (1.7 ng/L).



Figure 3-30. Hg concentrations measured in near-shore near-bottom water, Northerly Stream and seepage near former Outfall 001, July 7, 2011. All samples collected on ebbing tide. Stations 2 through 8 run from upstream to downstream.



Figure 3-31. Hg concentrations measured in near-bottom water upstream of Landfill 1 beach (UP1 to UP3), downstream of Outfall 001 (MID-1 to MID-5) and downstream of Southerly Cove (DN-1 to DN-4).



Figure 3-32. Hg concentrations measured in near-bottom water along transect over deepest part of river channel from north to south running past HoltraChem site, October 26, 2011.

### 6.3 Other Sources of Mercury Between Veazie Dam and Bucksport

In 1998 the Maine DEP initiated a state-wide program of sampling permitted discharges for Hg using clean sampling techniques and low level detection (Maine DEP 2001). Results for the reach of the Penobscot River between Veazie Dam and Bucksport were obtained from Stirling Pearce (Maine DEP) and are summarized in Table 3-15 and Figures 3-33 and 3-34. The HoltraChem plant was not included in this exercise but data from the DMRs for Outfall 001 and from CDM for the two surface streams have been added to Figure 3-34. None of the point sources of Hg to the river between Veazie and Bucksport exceeded 1 g/day individually nor was the aggregate loading of all such sources > 2 g/day.

Kenduskeag Stream, which flows through urban Bangor, was estimated from sampling on October 12, 2009 (total Hg = 2.0 to 2.4 ng/L) and historical discharge (10.7  $m^3/s$ ) to contribute about 2 g/day to the river during a typical October. Additional sampling (2010-11) and analysis of Kenduskeag water and that of other major tributaries by PRMS scientists (see Appendix 3-8) has determined average Hg loadings for these streams using average discharges for each stream derived from watershed properties (Dudley 2004). Total Hg concentrations in these streams varied from ~0.7 to 7.2 ng/L depending on TSS concentrations that ranged from 0.5 to 121 mg/L. The combined average total Hg loading by these tributaries was estimated to be ~15 g/day (see Appendix 3-8, Table 3). Higher tributary stream loadings would be expected during freshet and other times of higher flow. Based on sampling (discharge-weighted mean total Hg =3.9 ng/L) and historical discharge data (406 m<sup>3</sup>/s) the Penobscot River at Veazie dam contributes ~140 g/day (50 kg/year) to the downstream reach depending on river discharge (see Appendix 3-8, Table 3). Detailed measurements and estimates by PRMS scientists for the period 2008 to 2011 showed annual total Hg loadings for the Penobscot River at Veazie ranging from 156 to 189 g/day (57 to 69 kg/year). Total average particle (TSS)

loading by the Penobscot River at Veazie during the same period ranged from 51,700 to 59,100 T/year (see Appendix 3-8, Table 2).

Table 3-15:Loading of Hg from point sources on the Penobscot River (data from Maine DEP, 1998-2009, Clean Sampling Program)						
Point Sources (POTWs/NON-POTWs)	[Hg] Average (ng/L)	Std Dev (ng/L)	Discharge (mgd)	Hg Loading (g/day)		
SEARSPORT, TOWN OF	19.1	9.5	0.2	0.0145		
BUCKSPORT, TOWN OF	19.0	7.6	0.46	0.0330		
WINTERPORT SEWERAGE DISTRICT	17.5	5.5	0.11	0.0073		
MATTAWAMKEAG, TOWN OF	9.6	14.5	0.09	0.0033		
OLD TOWN, CITY OF	8.0	5.0	3.5	0.1061		
RED SHIELD ACQUISITION LLC	7.5	6.0	24.4	0.6949		
BANGOR, CITY OF	6.9	2.9	18	0.4705		
PENOBSCOT ENERGY RECOVERY CORP	6.2	5.1	0.0225	0.0005		
ORONO, TOWN OF	5.1	2.6	1.84	0.0352		
LINCOLN SANITARY DISTRICT	4.8	2.7	1.07	0.0196		
VEAZIE SEWER DISTRICT	4.2	1.7	0.35	0.0055		
HOWLAND ,TOWN OF	3.5	3.2	0.3	0.0040		
VERSO BUCKSPORT LLC	1.7	1.7	18	0.1173		
BREWER, CITY OF	1.7	0.7	5.19	0.0334		
KATAHDIN PAPER COMPANY LLC	1.6	0.8	43	0.2552		
HOLTRACHEM			·	0.52		
Sum						
Sum of average loadings reported in DMRs (1999-2009) for Outfall 001 and by CDM (see also Table 3-1) for Outfall 003 (Northerly Stream) and Southerly Stream.						



Figure 3-33. Average and standard deviation of Hg concentrations in point sources of Hg to Penobscot River (data from Maine DEP Clean Sampling Program).



Figure 3-34. Average Hg loading by point sources to Penobscot River. Except for HoltraChem all data are from Maine DEP's Clean Sampling Program. Data for HoltraChem is the sum of company reported average loading for Outfall 001 (0.32 g/day, 1999-2009) and CDM average loading (0.20 g/day, 2005-2007) for Outfall 003 (Northerly Stream) and Southerly Stream (see also Table 3-1 and Figure 3-3).

# 7 SUMMARY AND CONCLUSIONS

The primary objective of this investigation was to quantify total Hg loading from the former HoltraChem site under current (2009-2010) site conditions and operations, and over a range of surface water and groundwater discharges. A secondary objective was to identify and quantify other point sources of Hg to the Penobscot River downstream of Veazie Dam and above Bucksport. Surface water runoff from the site was monitored for slightly longer than one year while groundwater loading was assessed twice using a tracer technique, conventional seepage meters installed in the foreshore and sampling of foreshore seepages and porewater. Over the period of study approximately 2.3 kg of Hg was discharged to the Penobscot River by the two surface streams that drain the site. For one stream (Southerly) 90% of the total loading (1.9 kg) occurred in a few hours following a large rainfall (28 cm). The same storm accounted for only 17% of the total load (0.39 kg) for the other stream (Northerly). Average daily loading for these streams were 1.12 and 5.66 g/day, respectively, for the Northerly and Southerly Streams. Northerly Stream exhibited higher mean total Hg concentration (10,800 ng/L) and higher average Hg content of suspended matter (392  $\mu$ g/g dry wt.) compared with Southerly Stream (2,460 ng/L, 64 µg/g dry wt.). The combined total loading from the two surface streams for the period of study (422 days) was 2.3 kg (5.4 g/day) with most (78%) of this combined loading associated with a single large storm. Thus, in the absence of this storm the combined loading estimate would have been 0.51 kg (1.16 g/day).

Groundwater seepage rates from the site, as estimated from both the tracer and seepage meter methods, were in the 3 to 4 cm/day range and, when combined with a best estimate of the area of groundwater discharge (11,000 m<sup>2</sup>) and average seepage/porewater Hg concentration (242 ng/L, UCL95), yielded a loading of 0.22 g/day (80 g/year) for site groundwater. This estimate of recent Hg loading from groundwater is substantially less than that (17 g/day) reported in the late 1990s.

None of the municipal or industrial point sources of Hg to the river between Veazie and Bucksport exceeded 1 g/day individually nor was the aggregate loading of all such sources > 2 g/day (based on State of Maine data). The HoltraChem site Hg loading can also be compared to that by the three largest tributary streams downstream of Veazie Dam, Kenduskeag, Souadabscook and North Branch Marsh River. Hg loadings for these tributaries were estimated by other project scientists to contribute 3.8, 4.8 and 2.9 g/day, respectively, to the Penobscot River. The combined loading of all tributaries between Bangor and Bucksport was estimated at ~15 g/day (5.5 kg/year). Higher tributary stream loadings than these average values would be expected during freshet and other times of higher flow. Based on sampling (discharge-weighted mean total Hg =3.9 ng/L) and historical discharge data (406 m<sup>3</sup>/s) the Penobscot River at Veazie dam contributes ~140 g/day (50 kg/year) to the downstream reach depending on river discharge.

Overall this study has shown that average current Hg loading (~5 g/day) from the HoltraChem site is relatively low compared to historical loading (>20 g/day) and similar to the loads carried by the larger tributaries including Kenduskeag (3.8 g/day),

Souadabscook (4.8 g/day) and North Marsh Streams (2.9 g/day) that are present on the Penobscot River between Veazie Dam and Bucksport (Figure 3-35). While this current loading is also low with respect to that carried by the river, it may still represent a significant source to the immediate receiving area (i.e., Southerly Cove) adjacent to the site. For example, the high concentrations of Hg in site runoff water and high content of Hg on suspended matter carried by the two surface streams, especially Northerly Stream, may continue to supply Hg-contaminated water and sediment to Southerly Cove and adjacent areas.



Figure 3-35. Summary of Hg loadings to Penobscot River between Veazie Dam and Bucksport, Maine. The Veazie values were calculated using a discharge-weighted average total Hg concentration of 3.9 ng/L (from PRMS data) and mean river discharge (406 m<sup>3</sup>/s). Note that loading axis is logarithmic.

## 8 **REFERENCES**

- Burnett, W.C. and H. Dulaiova. 2003 Estimating the dynamics of groundwater input into coastal zone via continuous radon-222 measurements. Journal of Environmental Radioactivity. 69:21-35.
- Burnett, W.C., P.K. Aggarwal, H. Bokuniewicz, J.E. Cable, M.A. Charette, E. Kontar, S. Krupa, K.M. Kulkarni, A. Loveless, W.S. Moore, J.A. Oberdorfer, J. Oliveira, N. Ozyurt, P. Povinec, A.M.G. Privitera, R. Rajar, R.T. Ramessur, J. Scholten, T. Stieglitz, M. Taniguchi, and J.V. Turner. 2006 Quantifying submarine groundwater discharge in the coastal zone via multiple methods. Science of the Total Environment. 367:498-543.
- CDM. 1998. Site Investigation Report, HoltraChem Manufacturing Site, Orrington, Maine. Camp Dresser & McKee Inc., December 22, 1998.
- Dudley, RW. 2004. Estimating monthly, annual, and low 7-day, 10-year stream flows for ungaged rivers in Maine. USGS Scientific Investigations Report 2004-5026. 22 p.
- Lee, D. R. 1977. A device for measuring seepage in lakes and estuaries. Limnology and Oceanography 22:140-147.
- Mallinckrodt. 2010. Mallinckrodt's Post Trial Brief. <u>http://www.maine.gov/dep/spills/holtrachem/documents/mallinckrodt\_spostrialbrief.p\_df.</u>
- MDEP. 2001. Status of Mercury Discharged from Wastewater Treatment Facilities in Maine. Maine Department of Environmental Protection. 29 p.
- Mulligan, A.E. and M.A Charette. 2006. Intercomparison of submarine groundwater discharges from a sandy unconfined aquifer. Journal of Hydrology. 327:411-425.
- PRMS. 2008. Penobscot River Mercury Study Phase 1 of the Study: 2006-2007, January 24, 2008.

# **APPENDIX 3-1**

2004 Flux Study

## Measurements of Mercury Fluxes in Penobscot River Downstream of HoltraChem Site, October 2004

Conducted by Ralph Turner RT Geosciences Inc and George Southworth Oak Ridge National Laboratory

for Penobscot River Mercury Study

### ABSTRACT

The objective of this investigation was to quantify the net downstream flux of mercury (Hg) in the vicinity of the HoltraChem site near Orrington, Maine. To accomplish this objective numerous measurements of river discharge and Hg concentrations were taken over a 2-day period beginning October 7, 2004 at a location ~2 km downstream of the HoltraChem plant site. In addition water samples were collected from the Penobscot River near Veazie Dam and from several large tributaries upstream of the plant site (e.g., Kenduskeag, Souadabscook and Sedeunkedunk) to provide data for completing the mass balance.

This study was unable to derive an unequivocal estimate of the net downstream flux of Hg in the vicinity of the Holtrachem site. Nonetheless, the results did allow some estimates of the maximum possible flux from the site that ranged from 7 to 28 g/day with an indication (based on analysis of salinity profiles) that the actual HoltraChem flux was probably less than 7 g/day during the 2-day period of investigation when river discharge at Veazie Dam was at a seasonal low (mean=209 m<sup>3</sup>/s). Upstream sources (Penobscot River at Veazie Dam, major tributary streams and outfalls) during the same time were estimated at ~33 g/day. The study also documented that bidirectional flow was often present at the transect location and greatly complicated measurement and interpretation of Hg fluxes.

#### **Background and Rationale**

The investigation described in this report was among the first to try to quantify the downstream transport of Hg from the HoltraChem plant site using measurements within the Penobscot River. At the time of the study (fall 2004) sources of mercury were believed to include: 1) resuspension and downstream movement of sediments presently in the river bottom (particularly from Southerly Cove), 2) ongoing outfalls and surface runoff from the plant site, and 3) movement of Hg contaminated groundwater from the plant site into the river. Neither the total mass contribution of all of these sources to Hg in the river nor their individual contributions were very well known at the time. Establishing the mass fluxes of Hg down the river was necessary to determine if there were reductions in downstream transport after the completion of proposed and planned Hg amelioration measures, e.g. dredging of Southerly Cove, capture and treatment of contaminated groundwater. The latter amelioration measure was to be undertaken in early 2005 and thus this field investigation was planned and completed in advance of this measure.

### **Objectives and Approach**

The objective of this investigation was to quantify the net downstream flux of Hg in the vicinity of the HoltraChem site. To accomplish this objective numerous measurements of river discharge and mercury concentrations were taken over a 2-day period beginning October 7, 2004 at a location ~2 km downstream of the HoltraChem plant site (**Figure 1**). In addition water samples were collected from the Penobscot River near Veazie

Dam and from several large tributaries upstream of the plant site (e.g., Kenduskeag, Souadabscook and Sedeunkedunk) to provide data for completing the mass balance.

### Methods

In advance of sampling and discharge measurements on the river, four buoys were set across the direction of flow of the river and a bathymetric survey run to define the dimensions of the transect (Figure 2). Current velocity measurements and water sampling began on October 7, 2004 at 5:41AM and were completed on October 8, 2004 at 5:01PM with an overnight gap between 6:21PM on October 7 and 8:36AM on October 8. Eight combined sampling and velocity profiling events and four velocity profiling only events were conducted over the two day period. Each combined event required approximately one hour to complete. Velocity measurements were conducted using a General Oceanics Model 2135 digital current meter (see Photograph 1) at 1 meter intervals to the bottom at each transect location. Salinity and temperature were measured at the same times and depths as velocity measurement using a YSI 556 Multimeter. Water samples were typically collected at 2 meter, but in some cases at 1 meter, intervals. Channel discharge (Q) for a given event was calculated by summing discharges for each cell within the cross section of flow where each cell was typically 1 m high and 0.25 times the length from bank to bank. The vertical profile in velocities and salinities often showed bidirectional flow indicating presence of a salt wedge at depth. Inward (upstream, flood) velocities were recorded as negative values while outward (downstream, ebb) velocities were recorded as positive values.

Water sampling employed a 12-volt marine diaphragm pump and C-flex tubing with a plastic-coated weight attached to the intake end (see **Photograph 2**). This sampling system was deployed from an inflatable boat tethered to each buoy. The pump and tubing were first flushed for at least one minute with water from the target sampling depth. Bottles (500-mL Teflon for Hg samples, 1-L HDPE for TSS) were then filled with unfiltered water from a short piece of C-flex tubing attached to the pump discharge. Where filter-passing Hg samples were collected the pump was then shut off briefly to allow installation of an inline filter (0.45  $\mu$ M pore size). The pump was restarted and the filter flushed with ~3x filter volumes (500 mL) before filling a 500-mL Teflon bottle labeled to contain a filtered (dissolved) sample.

All samples for Hg analysis (EPA Method 1631) were shipped on ice unpreserved overnight to Flett Research Laboratory in Winnipeg, Manitoba. Samples for total suspended solids (TSS) (modified Standard Methods, 2450D) were shipped to Frontier Geosciences in Seattle, Washington. Hg detection limit was 0.2 ng/L while that for TSS using 0.4 micron membrane filters was 0.5 mg/L. Sample abbreviations used hereafter in this report are as follows:

TSS = Total suspended solids

THg = Total (unfiltered) mercury

FTHg = Filter-passing total mercury

Instantaneous flux (ng/sec) in a cell was calculated as the product of cell discharge (m<sup>3</sup>/s) and the concentration of Hg in the sample collected in the sample cell. Fluxes were separated between inflow (flood, negative) and outflow (ebb, positive) and used to calculate discharge-weighted means for each group. Discharge-weighted means for each group were calculated as sum of instantaneous fluxes divided by the sum of instantaneous discharges. Total flux was then calculated as the product of the weighted mean Hg concentration and total discharge for each flow direction. This procedure yields flux units of ng/s; for easier reference and comparisons these units were also converted to g/day.

The discharge of the Penobscot River during the measurement period was stable (207 to 214  $m^3$ /s). Two water samples collected below Veazie dam averaged 1.6 ng/L in total Hg leading to a calculated loading of about 30 g/day



Figure 1. Location of transect used to measure fluxes of Hg in Penobscot River, October 2004.



Figure 2. Sample locations (looking downstream) relative to bathymetry at Penobscot River transect, October 2004. All Units are meters.

### Results

Complete results of discharge measurements and mercury loading calculations are provided in Attachment while Table 1 is a summary of these.

**Hydrology** - As a rough check on calculated discharges for the transect, discharge over Veazie Dam can be compared to river discharges at the transect location during two tidal periods, near high tide and low tide. At these times discharge in the river should be approximately equal to the sum of Veazie discharge and the major tributary discharges (Kenduskeag, Souadabscook and Sedgeunkedunk) between Veazie and the transect location. Discharge measurements corresponded with these tidal stages three times: high tide at 10/7 5:41-7:07, low tide at 10/7 11:55-13:02 and low tide at 10/8 12:58-14:05 (Figure 3). Ebb discharges were 200, 224 and 263 m<sup>3</sup>/s, respectively, at these times and thus correspond reasonably well with the mean daily Veazie discharges of 207 and 214 m<sup>3</sup>/s without adjustment for additional tributary inputs. Based on estimated mean annual discharges for the three tributaries (25, 21 and 2 m<sup>3</sup>/s) adjusted for seasonally lower discharges in October (assuming 50% of mean annual) suggests the river discharge at transect location was in the range of 230 to 237 m<sup>3</sup>/s during the flux measurement period. A secondary check on hydrologic measurements is possible using the mean value of flood discharge: it should be approximately equal to the river discharge, because net tidal discharges should be zero if minimum and maximum tidal elevations are similar over the period being studied. The mean flood discharge (196  $m^{3}$ /s) is slightly lower than the estimated river discharge but also reasonably comparable. Overall there were more discharge measurements on ebb tides (8) than flood tides (5) which may introduce some bias in estimating "mean" values. However, considering all discharge measurements, including those when there was bidirectional flow, there were more flood discharge measurements (10) than ebb discharge measurements (8). Given the limitations of the technology (mechanical current meter velocity readings at discrete depths and at only four locations across the river) used to estimate discharge, the agreement with independently derived river discharge values is reassuring.

**Mercury Concentrations and Loading** – As indicated in Table 1 both the arithmetic and Q- weighted mean Hg concentrations for flood flows were higher than those for ebb flows. The difference in arithmetic means (3.01 and 5.53 ng/L) was significant (F-test,  $P \le 0.002$ ) even when three outlier values (28, 35 and 74 ng/L) were removed from the flood flow data set (3.64 ng/L after outlier removal). The three values removed are suspected to have been possibly affected by sampling gear touching the bottom as all three values were from the deepest sample. However, these could have been valid (uncompromised) values and indicative of near-bottom water being higher in Hg due to local scouring of particles (two of the three samples were also higher in TSS compared with shallower samples). Considering all the data (Attachment) it does appear that samples from the deepest depths were often higher than samples from shallower depths. More detailed analysis of this issue is not possible because analysis of TSS was generally limited to one sample depth per location per sampling time. No outliers were removed for the calculation of Hg loading.



Figure 3. Time series plots of tide levels and discharge, October 7-8, 2004.

The mean ebb flow Hg loading (61 g/day) was substantially lower that the flood flow mercury loading (101 g/day). As noted previously three flood flow Hg concentrations could have been artifacts of sampling and the difference in mean loading between ebb and flood would be less if these values are excluded. Hg loading at Veazie Dam was 30 g/day and thus the mean ebb flow loading at the transect (61 g/day) has to include components of tributary loading (~2 g/day), municipal/industrial outfall loading (~1 g/day), Holtrachem site loading and loading carried over from the previous flood flow. The last component cannot be derived from the available data but if it is assumed to be zero all the other loadings sum to ~33 g/day and the Holtrachem loading should not be greater than 28 g/day (61 minus 33 g/day). Another estimate of the Hg loading from Holtrachem site using this data set can be made using the ebb flow loading for the two times when measurements corresponded with times of low, or near low, tide (10/7 11:55-13:02 and 10/8 12:58-14:05). Ebb flow loadings (Table 1) at these times were 458 and 590 µg/s, respectively, or 40 and 51 g/day. Subtraction of the Veazie, tributary and outfall loadings yield net loadings possibly attributable to the Holtrachem site of 7 and 18 g/day. This estimate is based on the assumption that late ebb tide flows do not include any water that moved upstream during the previous flood tide. Examination of the salinity data for these times (Attachment) shows ebbing water values (1 ppth or greater) indicative of incomplete flushing of seawater from the river reach upstream of the transect. Thus, the loadings for the Holtrachem site derived by this method also likely represent maximum values.

		Arithmetric			Ebb	Flood		
Start	End	Mean Hg	Ebb Q	Flood Q	Q-watC	Q-wqtC	Ebb J	Flood J
Datetime	Datetime	(ng/L)	(m <sup>3</sup> /s)	(m <sup>3</sup> /s)	(ng/L)	(ng/L)	(µg/s)	(µg/s)
10/07/04	10/07/04	4.22	200	30.6	2.29	10.15	458	311
5:41	7:07							
10/07/04	10/07/04	3.26	698	18.4	1.94	2.61	1354	48.0
9:00	10:18							
10/07/04	10/07/04		548	79.5				
11:10	11:30							
10/07/04	10/07/04	2.66	224	66	2.08	2.55	466	168
11:55	13:02							
10/07/04	10/07/04	3.5	34.9	250	4.89	4.03	171	1008
14:40	15:44							
10/07/04	10/07/04		0	727				
16:09	16:50							
10/07/04	10/07/04	10.4	0	472		15.3		7223
17:06	18:21							
10/08/04	10/08/04	3.2	407	0	2.09		851	
8:36	9:55							
10/08/04	10/08/04		879	0				
10:34	11:27							
10/08/04	10/08/04	2.46	263	37.4	2.24	2.06	590	77.0
12:58	14:05							
10/08/04	10/08/04		0	169				
15:02	15:49							
10/08/04	10/08/04	4.88	0	508		4.8		2439
16:02	17:11							
Grand		4.32	271	196	2.59	5.93	702	1165
Mean								
Values		0.04					00.0	404
	EDD	3.01				g/day	60.6	101
	Flood	5.53						

Table 1 . Summary of transect discharges (Q), Hg concentrations (C) and fluxes (J) for October 2004 study in Penobscot River downstream of HoltraChem site

## Conclusions

This study was unable to derive an unequivocal estimate of the net downstream flux of Hg in the vicinity of the Holtrachem site. Nonetheless, the results did allow some estimates of the maximum possible flux from the site that ranged from 7 to 28 g/day with an indication (based on analysis of salinity profiles) that the actual Holtrachem flux was probably less than 7 g/day during the 2-day period of investigation when river discharge at Veazie Dam was at a seasonal low (209 m<sup>3</sup>/s). Upstream sources (Penobscot River at Veazie Dam, major tributary streams and outfalls) during the same time were estimated at ~33 g/day. The study also documented that bidirectional flow was often present at the transect location and greatly complicated measurement and interpretation of Hg fluxes.
## ATTACHMENT

Penobsco	t River Transec	t Data-October	2004															
							Directional			Cell	Cell							
			Depth	Temp	SpecCond	Salinity	Velocity		THg	Discharge	Hg Load	DHg	TSS	TSS-Hg	Kds	Sample		
Station	Date	Time	(m)	(oC)	(uS/cm)	(ppth)	(m/sec)	Direction	(ng/L)	(m3/sec)	(ng/sec)	(ng/L)	(mg/L)	(ug/g)	(mL/g)	Time	Notes	
T4	10/07/04	5:41	1	13.9	3224	1.69	0.205	Out	2.02	20.5	41393					5:58		
T4	10/07/04	5:43	2	13.9	3535	1.87	0.152	Out		15.2	0							
T4	10/07/04	5:45	3	14.1	4660	2.51	0.071	Out	2.78	7.1	19729					6:00		
T4	10/07/04	5:47	4	14.2	9886	5.61	0.004	Out		0.4	0							
T4	10/07/04	5:49	5	14.2	16510	9.74	0.000	Out	4.83	0.0	0					6:02		
T4	10/07/04	5:51	6	13.6	27181	16.69	-0.090	In		-9.0	0							
T4	10/07/04	5:53	7	13.5	28964	17.92	-0.133	In	14.2	-13.3	-189333	0.95	14.9	0.89	4.8	6:04	Bottom=7.3	
T3	10/07/04	6:11	1	13.8	3047	1.6	0.285	Out	1.73	28.5	49348	1.01	1.03	0.70	6.0	6:28	High Tide at 6:00	0
T3	10/07/04	6:13	2	14.2	4496	2.41	0.177	Out		17.7	0							
Т3	10/07/04	6:15	3	14.2	4927	2.66	0.076	Out	2.53	7.6	19179							
Т3	10/07/04	6:17	4	14.3	7363	4.09	0.000	Out		0.0	0							
Т3	10/07/04	6:18	5	14.2	16875	9.96	-0.024	In	4.73	-2.4	-11262							
Т3	10/07/04	6:19	6	13.6	27349	16.8	-0.003	In		-0.3	0							
T3	10/07/04	6:21	7	13.6	28380	17.52	0.000	In	8.05	0.0	0					6:31		
Т3	10/07/04	6:23	7.8	13.5	28488	17.6	0.000	In		0.0	0						Bottom=7.8	
T2	10/07/04	6:35	1	13.9	3069	1.61	0.258	Out	2.22	25.8	57350					6:50		
T2	10/07/04	6:37	2	14.2	3930	2.11	0.161	Out		16.1	0							
T2	10/07/04	6:38	3	14.3	5780	3.15	0.110	Out	2.27	11.0	24897							
T2	10/07/04	6:41	4	14.3	6867	3.81	0.049	Out		4.9	0							
T2	10/07/04	6:42	5	14.2	15217	8.91	0.018	Out	3.42	1.8	6125						Taken @ 4.5	
T2	10/07/04	6:42	6	13.7	25490	15.54	-0.056	In	3.3	-5.6	-18333	1.2	12.3	0.17	4.9	6:53	Bottom=6.0	
T1	10/07/04	6:59	1	14	3720	2	0.210	Out	2.95	21.0	61855	1.04	1.24	1.54	5.9	7:09		
T1	10/07/04	7:01	2	14.2	4750	2.53	0.066	Out		6.6	0							
T1	10/07/04	7:03	3	14.2	5131	2.78	0.076	Out	2.33	7.6	17752							
T1	10/07/04	7:04	4	14.3	5850	3.22	0.064	Out		6.4	0							
T1	10/07/04	7:05	5	14.3	12956	7.5	0.014	Out	3.14	1.4	4348						Taken @4.5	
T1	10/07/04	7:07	6	13.7	25647	15.69	0.003	Out	7.03	0.3	2038					7:13	Bottom=6.0	
									Mean	SumAlIQ								
Sta Name	Latitude	Longitude						Ebb	2.95	199.8								
T1	N 44 43.364	W 068 49.809	)					Flood	7.57	-30.6								
T2	N 44 43.336	W 068 49.914								SumQ	SumJ	Q-wgtC	ng/sec	g/day				
Т3	N 44 43.345	W 068 49.976	5					Ebb		132.5	304014	2.29	458353	39.60				
T4	N 44 43.337	W 068 50.042						Flood		-21.6	-218929	10.15	-310430	-26.82				

# ATTACHMENT. Complete field and analytical data for Penobscot River transect sampling October 7-8, 2004

Penobscot	River Transec	t Data																
							Directional			Cell	Cell							
			Depth	Temp	SpecCond	Salinity	Velocity		THg	Discharge	Hg Load	DHg	TSS	TSS-Hg	Kds	Sample		
Station	Date	Time	(m)	(oC)	(uS/cm)	(ppth)	(m/sec)	Direction	(ng/L)	(m3/sec)	(ng/sec)	(ng/L)	(mg/L)	(ug/g)	(mL/g)	Time	Notes	
T4	10/07/04	9:00	1	13.7	2027	1.04	0.442	Out	1.68	44.2	74245	1	0.77	0.88	6.1	9:18		
T4	10/07/04	9:07	2	13.8	2381	1.23	0.326	Out		32.6								
T4	10/07/04	9:09	3	13.8	3317	1.71	0.194	Out	1.81	19.4	35032							
T4	10/07/04	9:11	4	14.2	7960	4.4	0.302	Out		30.2								
T4	10/07/04	9:13	5	14.3	9609	5.43	0.210	Out	2.78	21.0	58248							
T4	10/07/04	9:14	6	14.2	14530	8.46	0.134	Out		13.4								
T4	10/07/04	9:15	6.5	14.1	19050	11.34	0.170	Out	3.92	17.0						9:25	Bottom=6.5	
T3	10/07/04	9:30	1	13.8	1407	0.71	0.656	Out	1.65	65.6	108197							
T3	10/07/04	9:32	2	13.9	3108	1.63	0.610	Out		61.0								
T3	10/07/04	9:34	3	14	3646	1.93	0.580	Out	2.02	58.0	117226	1.07	1.25	0.76	5.9			
T3	10/07/04	9:35	4	14.1	5650	3.04	0.412	Out		41.2							Wire<30	
T3	10/07/04	9:36	5	14.2	9910	5.59	0.244	Out	2.78	24.4	67706						Wire<45	
T3	10/07/04	9:37	6	14.3	15400	9.03	0.211	Out		21.1							Wire<45	
T3	10/07/04	9:38	7	14	21300	12.88	0.036	Out	10.5	3.6							Bottom=7.0	
T2	10/07/04	9:54	1	13.9	1502	0.76	0.707	Out	1.78	70.7	125767	1.09	0.78	0.88	6.1			
T2	10/07/04		2	13.9	2267	1.17	0.611	Out	1.83	61.1	111900							
T2	10/07/04	9:57	3	13.9	2741	1.43	0.482	Out	2.1	48.2	101213							
T2	10/07/04		4	14	5113	2.75	0.425	Out		42.5							Wire<30	
T2	10/07/04		5	14.2	11840	6.78	0.226	Out	3.27	22.6								
T1	10/07/04	10:12	1	13.9	3207	1.69	-0.020	In	1.95	-2.0	-3900							
T1	10/07/04	10:14	2	13.9	3220	1.69	-0.074	In	2.12	-7.4	-15729	1.13	1.14	0.87	5.9			
T1	10/07/04		3	14	5055	2.69	-0.081	In	3.23	-8.1	-26244							
T1	10/07/04		4	14.3	10800	6.2	-0.009	In		-0.9								
T1	10/07/04	10:18	5	14.3	13790	8.04	0.000	In	8.73	0.0								
										SumAlIQ								
								Ebb		697.7								
								Flood		-18.4								
										SumQ	SumJ	Q-wgtC	ng/sec	g/day				
								Ebb		412.5	799535	1.94	1352356	116.84				
								Flood		-17.5	-45873	2.61	-48214	-4.17				

Penobsco	t River Transeo	t Data																
							Directional			Cell	Cell							
			Depth	Temp	SpecCond	Salinity	Velocity		THg	Discharge	Hg Load	DHg	TSS	TSS-Hg	Kds	Sample		
Station	Date	Time	(m)	(oC)	(uS/cm)	(ppth)	(m/sec)	Direction	(ng/L)	(m3/sec)	(ng/sec)	(ng/L)	(mg/L)	(ug/g)	(mL/g)	Time	Notes	
T4	10/07/04	11:10	1	13.9	1877	0.96	0.628	Out		62.8								
T4	10/07/04		2	13.9	2785	1.48	0.434	Out		43.4								
T4	10/07/04		3	14	2719	1.52	0.292	Out		29.2								
T4	10/07/04		4	14	5709	3.11	0.338	Out		33.8								
T4	10/07/04		5	14.1	11030	6.26	0.180	Out		18.0								
T4	10/07/04		5.5	14.1	13095	7.57	0.152	Out		7.6							Bottom=5.5	
T3	10/07/04	11:20	1	13.8	1597	0.81	0.633	Out		63.3								
T3	10/07/04		2	13.9	2850	1.51	0.554	Out		55.4								
T3	10/07/04		3	13.9	3975	2.1	0.548	Out		54.8								
T3	10/07/04		4	14.1	9776	5.53	0.295	Out		29.5								
T3	10/07/04		5	14.1	12533	7.21	0.147	Out		14.7								
T3	10/07/04		6	14.1	14612	8.53	0.102	Out		10.2								
T2	10/07/04	11:25	1	13.9	2326	1.22	0.498	Out		49.8								
T2	10/07/04		2	14	2900	1.53	0.377	Out		37.7								
T2	10/07/04		3	14	3605	1.9	0.236	Out		23.6								
T2	10/07/04		4	14	8490	4.79	0.139	Out		13.9								
T1	10/07/04	11:30	1	14.1	3051	1.6	-0.311	In		-31.1								
T1	10/07/04		2	14	3372	1.8	-0.267	In		-26.7								
T1	10/07/04		3	14.1	7707	4.35	-0.200	In		-20.0								
T1	10/07/04		4	14.2	14190	8.25	-0.015	In		-1.5								
T1	10/07/04		4.5	14.2	14332	8.34	-0.003	In		-0.1								
										SumAlIQ								
								Ebb		547.7								
								Flood		-79.5								

Penobsco	t River Transec	t Data																
							Directional			Cell	Cell							
			Depth	Temp	SpecCond	Salinity	Velocity		THg	Discharge	Hg Load	DHg	TSS	TSS-Hg	Kds	Sample		
Station	Date	Time	(m)	(oC)	(uS/cm)	(ppth)	(m/sec)	Direction	(ng/L)	(m3/sec)	(ng/sec)	(ng/L)	(mg/L)	(ug/g)	(mL/g)	Time	Notes	
T4	10/07/04	11:55	1	13.9	1845	0.94	0.497	Out	1.86	49.7	92390							
T4	10/07/04		2	14	3050	1.62	0.364	Out	2.17	36.4	78974	1.05	2.52	0.44	5.6			
T4	10/07/04		3	14	3905	2.08	0.263	Out	2.16	26.3	56880							
T4	10/07/04	12:00	4	14	7750	4.31	0.005	Out		0.5								
T4	10/07/04		5	14.1	14950	8.52	0.002	Out	4.55	0.2	711							
T4	10/07/04		5.5	14.1	15880	9.33	0.002	Out		0.1								
T3	10/07/04	12:15	1	13.9	2230	1.16	0.466	Out	1.82	46.6	84734							
T3	10/07/04		2	13.9	2686	1.4	0.413	Out	1.98	41.3	81840	1.03	0.97	0.98	6.0			
T3	10/07/04		3	14.0	5305	2.86	0.229	Out	3.02	22.9	69168							
T3	10/07/04		4	14.1	10985	6.26	0.000	Out		0.0								
T3	10/07/04	12:25	5	14.1	13520	7.84	-0.027	In	3.78	-2.7							Low tide at	12:26
T3	10/07/04		5.5	14.1	16008	9.35	0.000	In		0.0								
T2	10/07/04	12:40	1	14.2	3648	1.94	-0.009	In	2.16	-0.9	-1851							
T2	10/07/04		2	14.0	6589	3.64	-0.088	In	2.31	-8.8	-20342	1.27	1.43	0.73	5.8			
T2	10/07/04		3	14.1	10856	6.19	-0.002	In	2.87	-0.2	-456							
T2	10/07/04		3.7	14.1	12967	7.49	-0.020	In	3.43	-1.4	-4734							
T1	10/07/04	12:55	1	14.2	2748	1.43	-0.290	In	3.08	-29.0	-89320							
T1	10/07/04		2	14.0	6336	3.47	-0.203	In	2.18	-20.3	-44315							
T1	10/07/04		3	14.0	7838	4.36	-0.010	In	2.23	-1.0	-2262	1.09	1.3	0.88	5.9			
T1	10/07/04	13:02	4	14.0	8274	4.62	-0.008	In	3	-0.8	-2344							
T1	10/07/04		4.5	14.0	8024	4.47	-0.012	In		-0.6								
										SumAllQ								
								Ebb		224								
								Flood		-66								
										SumQ	SumJ	Q-wgtC	ng/sec	g/day				
								Ebb		223.3	464697	2.08	465803	40.25				
								Flood		-65.0	-165624	2.55	-167153	-14.44				

Penobscot	t River Transec	t Data																
							Directional			Cell	Cell							
			Depth	Temp	SpecCond	Salinity	Velocity		THg	Discharge	Hg Load	DHg	TSS	TSS-Hg	Kds	Sample		
Station	Date	Time	(m)	(oC)	(uS/cm)	(ppth)	(m/sec)	Direction	(ng/L)	(m3/sec)	(ng/sec)	(ng/L)	(mg/L)	(ug/g)	(mL/g)	Time	Notes	
T4	10/07/04	14:40	1	14.7	2570	1.34	-0.071	In	2.05	-7.1	-14558							
T4	10/07/04		2	14.4	2645	1.37	-0.053	In	1.83	-5.3	-9705							
T4	10/07/04		3	14.2	2733	1.43	-0.077	In	2.09	-7.7	-16077	1.02	0.82	1.30	6.1			
T4	10/07/04		4	14.2	3292	1.74	-0.078	In		-7.8								
T4	10/07/04	14:48	5	14.1	11630	6.66	-0.152	In	3.17	-15.2	-48282							
T4	10/07/04		5.5	14.1	13730	7.97	-0.035	In		-1.8								
T3	10/07/04	15:02	1	14.6	2392	1.24	-0.230	In	1.8	-23.0	-41311							
T3	10/07/04		2	14.6	2521	1.31	-0.180	In		-18.0								
T3	10/07/04		3	14.3	2977	1.55	-0.110	In	1.92	-11.0	-21029							
T3	10/07/04		4	14.1	3390	1.79	0.000	In		0.0								
T3	10/07/04		5	14.1	11340	6.49	0.130	Out	3.11	13.0	40277							
T3	10/07/04	15:12	6	14.2	15145	8.86	0.116	Out		11.6								
T3	10/07/04		6.5	13.9	23185	14.09	0.206	Out	7.13	10.3	73563	1.13	6.5	0.92	5.2			
T2	10/07/04	15:25	1	14.6	2373	1.23	-0.095	In	1.76	-9.5	-16788							
T2	10/07/04		2	14.2	3026	1.59	0.000	In		0.0								
T2	10/07/04		3	14.1	3432	1.81	-0.071	In	2.16	-7.1	-15429	1.01	1.27	0.91	5.9			
T2	10/07/04		4	14	7750	4.31	-0.241	In	2.74	-24.1	-66030							
T2	10/07/04		5	14	18712	11.15	-0.202	In	11.3	-20.2	-227738							
T1	10/07/04	15:44	1	14.4	2550	1.32	-0.024	In	1.78	-2.4	-4228							
T1	10/07/04		2	14.3	2951	1.55	0.000	In		0.0								
T1	10/07/04		3	14	5529	3.02	-0.173	In	2.88	-17.3	-49703							
T1	10/07/04		4	14.1	15699	9.24	-0.280	In	3.23	-28.0	-90546							
T1	10/07/04		5	13.9	20600	12.43	-0.313	In	7.1	-31.3	-221875	1.08	7.68	0.78	5.1			
T1	10/07/04		5.5	13.8	23350	14.17	-0.268	In		-13.4								
										SumAllQ								
								Ebb		34.9								
								Flood		-250.1								
										SumQ	SumJ	Q-wgtC	ng/sec	g/day				
								Ebb		23.3	113841	4.89	170680	14.75				
								Flood		-209.1	-843297	4.03	-1008668	-87.15				

Penobsco	t River Transec	t Data																
							Directional			Cell	Cell							
			Depth	Temp	SpecCond	Salinity	Velocity		THg	Discharge	Hg Load	DHg	TSS	TSS-Hg	Kds	Sample		
Station	Date	Time	(m)	(oC)	(uS/cm)	(ppth)	(m/sec)	Direction	(ng/L)	(m3/sec)	(ng/sec)	(ng/L)	(mg/L)	(ug/g)	(mL/g)	Time	Notes	
T4	10/07/04	16:09	1	14.1	2874	1.5	-0.147	In		-14.7								
T4	10/07/04		2	14.1	2896	1.51	-0.103	In		-10.3								
T4	10/07/04		3	14	3196	1.69	0.000	In		0.0								
T4	10/07/04		4	14.1	8160	4.5	-0.327	In		-32.7								
T4	10/07/04		5	13.4	29206	18.08	-0.344	In		-34.4								
T4	10/07/04	16:18	6	13.4	30299	18.82	-0.341	In		-34.1								
T4	10/07/04		6.5	13.4	30390	18.88	-0.307	In		-15.3								
T3	10/07/04	16:23	1	14.6	2438	1.26	-0.254	In		-25.4								
T3	10/07/04		2	14.4	2690	1.4	-0.195	In		-19.5								
T3	10/07/04		3	14.1	4307	2.3	-0.248	In		-24.8								
T3	10/07/04		4	14.1	10350	5.85	-0.465	In		-46.5								
T3	10/07/04		5	13.6	25450	15.58	-0.475	In		-47.5								
T3	10/07/04	16:31	6	13.4	28559	17.65	-0.384	In		-38.4								
T3	10/07/04		7	13.4	29225	18.09	-0.297	In		-29.7								
T2	10/07/04	16:37	1	14.6	2503	1.3	-0.270	In		-27.0								
T2	10/07/04		2	14.2	3334	1.76	-0.244	In		-24.4								
T2	10/07/04		3	14.1	6876	3.75	-0.308	In		-30.8								
T2	10/07/04		4	13.9	15821	9.27	-0.603	In		-60.3								
T2	10/07/04		5	13.6	25760	16.03	-0.500	In		-50.0								
T2	10/07/04	16:47	6	13.4	29022	17.98	-0.308	In		-30.8								
T1	10/07/04	16:50	1	14.6	2627	1.37	-0.171	In		-17.1								
T1	10/07/04		2	14.3	4142	2.2	0.000	In		0.0								
T1	10/07/04		3	14,1	12217	7.01	-0.308	In		-30.8								
T1	10/07/04		4	13.9	17276	10.28	-0.382	In		-38.2								
T1	10/07/04		5	13.5	27450	16.89	-0.274	In		-27.4								
T1	10/07/04		6	13.4	29824	18.5	-0.170	In		-17.0								
										SumAlIQ								
								Ebb		0								
								Flood		-727.2								

Penobscot	River Transec	t Data																
							Directional			Cell	Cell							
			Depth	Temp	SpecCond	Salinity	Velocity		THg	Discharge	Hg Load	DHg	TSS	TSS-Hg	Kds	Sample		
Station	Date	Time	(m)	(oC)	(uS/cm)	(ppth)	(m/sec)	Direction	(ng/L)	(m3/sec)	(ng/sec)	(ng/L)	(mg/L)	(ug/g)	(mL/g)	Time	Notes	
T4	10/07/04	17:06	1	14.4	3402	1.81	-0.127	In	2.43	-12.7	-30755							
T4	10/07/04		2	14.2	4336	2.32	-0.178	In		-17.8								
T4	10/07/04		3	14.2	5024	2.72	-0.246	In	2.75	-24.6	-67623							
T4	10/07/04		4	13.6	26670	15.5	-0.232	In		-23.2								
T4	10/07/04		5	13.3	31035	19.11	-0.259	In	35.1	-25.9	-908143							
T4	10/07/04		6	13.3	31030	19.34	-0.326	In		-32.6								
T4	10/07/04	17:17	7	13.2	31340	19.52	-0.285	In	74.1	-28.5	-2111850	0.91	48.2	1.52	4.3			
T3	10/07/04	17:28	1	14.5	3450	1.82	-0.316	In	1.86	-31.6	-58800							
T3	10/07/04		2	14.3	4250	2.32	-0.244	In		-24.4								
T3	10/07/04		3	14.2	5770	3.17	-0.238	In	2.54	-23.8	-60377							
T3	10/07/04		4	14.1	9068	4.96	-0.398	In		-39.8								
T3	10/07/04		5	13.4	29158	18.05	-0.227	In	7.75	-22.7	-176250	0.98	8.6	0.79	5.1			
T3	10/07/04		6	13.3	30318	18.81	-0.258	In		-25.8								
T3	10/07/04	17:38	7	13.2	31020	19.32	-0.244	In	10	-24.4	-244262							
T3	10/07/04		8.3	13.2	31075	19.35	-0.103	In		-13.4								
T2	10/07/04	17:50	1	14.7	3886	2.07	-0.128	In	2.09	-12.8	-26778							
T2	10/07/04		2	14.6	3920	2.08	0.000	In		0.0								
T2	10/07/04		3	14.5	4374	2.34	0.000	In	2.11	0.0	0							
T2	10/07/04		4	14	14125	8.02	-0.237	In	2.53	-23.7	-59901	0.98	1.85	0.84	5.7			
T2	10/07/04		5	13.1	22775	13.76	-0.230	In		-23.0								
T2	10/07/04	17:58	6	13.3	30076	18.67	-0.015	In	6.74	-1.5	-10060							
T1	10/07/04	18:11	1	14.5	2882	1.49	-0.048	In	2.05	-4.8	-9791	1.05	1.06	0.94	6.0			
T1	10/07/04		2	14.5	3807	1.68	-0.136	In		-13.6								
T1	10/07/04		3	14.5	3545	1.88	-0.073	In	2.1	-7.3	-15422							
T1	10/07/04		4	14,2	5728	3.12	-0.083	In		-8.3								
T1	10/07/04		5	13.7	22447	13.65	-0.002	In	4.34	-0.2	-668							
T1	10/07/04		6	13.4	28485	17.6	0.000	In		0.0								
T1	10/07/04	18:21	7	13.3	30145	18.72	-0.058	In	8.75	-5.8	-50845							
										SumAlIQ								
								Ebb		0.0								
								Flood		-472.1								
										SumQ	SumJ	Q-watC	na/sec	⟨⟨⟨av				
								Ebb		0.0			-					
								Flood		-250.2	-3831524	15.31	-7228262	-624.52				

Penobscot	t River Transec	t Data																
							Directional			Cell	Cell							
			Depth	Temp	SpecCond	Salinity	Velocity		THg	Discharge	Hg Load	DHg	TSS	TSS-Hg	Kds	Sample		
Station	Date	Time	(m)	(oC)	(uS/cm)	(ppth)	(m/sec)	Direction	(ng/L)	(m3/sec)	(ng/sec)	(ng/L)	(mg/L)	(ug/g)	(mL/g)	Time	Notes	
T4	10/08/04	8:36	1	13.3	3169	1.67	0.291	Out	1.73	29.1	50278	1.00	0.83	0.88	6.1			
T4	10/08/04		2	13.3	3717	1.97	0.184	Out		18.4								
T4	10/08/04		3	13.6	5325	2.91	0.251	Out	1.94	25.1	48659							
T4	10/08/04		4	13.7	7258	4.02	0.031	Out		3.1								
T4	10/08/04		5	13.7	12986	7.5	0.056	Out	3.07	5.6	17056							
T4	10/08/04		6	13.5	23404	14.2	0.000	Out		0.0								
T4	10/08/04	8:47	7	13.2	30230	18.78	0.000	Out	8.12	0.0	0							
T3	10/08/04	9:00	1	13.4	3332	1.76	0.462	Out	2.1	46.2	97082	0.96	0.68	1.68	6.2			
T3	10/08/04		2	13.5	3565	1.88	0.411	Out		41.1								
T3	10/08/04		3	13.5	4952	2.68	0.292	Out	1.6	29.2	46710							
T3	10/08/04		4	13.7	8495	4.76	0.145	Out		14.5								
T3	10/08/04		5	13.7	11274	6.49	0.044	Out	2.99	4.4	13021							
T3	10/08/04		6	13.5	22260	13.44	0.000	Out		0.0								
T3	10/08/04		7	13.2	29810	18.48	0.000	Out	8.36	0.0	0							
T3	10/08/04		7.5	13.2	31249	19.47	0.000	Out		0.0								
T2	10/08/04	9:25	1	13.5	3360	1.76	0.502	Out	1.96	50.2	98327	0.96	0.88	1.14	6.1			
T2	10/08/04		2	13.5	4224	2.26	0.374	Out		37.4								
T2	10/08/04		3	13.6	5903	3.21	0.372	Out	2.16	37.2	80380							
T2	10/08/04		4	13.7	9050	5.07	0.158	Out	2.49	15.8	39457							
T2	10/08/04		5	13.7	13130	7.59	0.065	Out	3.02	6.5	19514							
T2	10/08/04	9:35	5.5	13.6	18730	11.15	0.166	Out		8.3								
T1	10/08/04	9:46	1	13.5	3744	1.99	0.167	Out	1.88	16.7	31333	0.97	0.87	1.05	6.1	9:57		
T1	10/08/04		2	13.5	3940	2.1	0.004	Out		0.4					-			
T1	10/08/04		3	13.6	6610	3.67	0.101	Out	2.29	10.1	23242							
T1	10/08/04		4	13.6	8160	4.57	0.052	Out	2.6	5.2	13619							
T1	10/08/04	9.55	5	13.7	15900	9.23	0.030	Out	4 93	3.0	14920					10.03		
	10/00/01	0.00	Ū		10000	0.20	0.000	out		SumAllQ	11020					10100		
								Ebb		407.4								
			_					Flood		0.0								
										Sums	Sums	Q-watC	na/sec	g/dav				
								Ebb		284.2	593597	2.09	850789	73.51				1
								Flood		0.0								

Penobscot	River Transec	t Data															
							Directional			Cell	Cell						
			Depth	Temp	SpecCond	Salinity	Velocity		THg	Discharge	Hg Load	DHg	TSS	TSS-Hg	Kds	Sample	
Station	Date	Time	(m)	(oC)	(uS/cm)	(ppth)	(m/sec)	Direction	(ng/L)	(m3/sec)	(ng/sec)	(ng/L)	(mg/L)	(ug/g)	(mL/g)	Time	Notes
T4	10/08/04	10:34	1	13.5	2390	1.24	0.375	Out		37.5							
T4	10/08/04		2	13.5	2780	1.46	0.242	Out		24.2							
T4	10/08/04		3	13.5	4020	2.15	0.311	Out		31.1							
T4	10/08/04		4	13.6	6570	3.56	0.279	Out		27.9							
T4	10/08/04		5	13.7	11000	6.22	0.243	Out		24.3							
T4	10/08/04		6	13.6	16300	9.49	0.370	Out		37.0							
T4	10/08/04	10:43	6.3	13.6	21350	12.91	0.309	Out		30.9							
T3	10/08/04	10:52	1	13.5	2460	1.27	0.605	Out		60.5							
T3	10/08/04		2	13.5	2698	1.4	0.519	Out		51.9							
T3	10/08/04		3	13.6	5020	2.71	0.645	Out		64.5							
T3	10/08/04		4	13.6	6962	3.84	0.585	Out		58.5							
Т3	10/08/04		5	13.7	14700	8.65	0.430	Out		43.0							
T3	10/08/04		6	13.6	19500	11.6	0.331	Out		33.1							
T3	10/08/04	11:01	7	13.4	26270	16.17	0.234	Out		23.4							
T2	10/08/04	11:08	1	13.6	2280	1.2	0.838	Out		83.8							
T2	10/08/04		2	13.5	2780	1.46	0.600	Out		60.0							
T2	10/08/04		3	13.5	3460	1.83	0.654	Out		65.4							
T2	10/08/04		4	13.6	5860	3.16	0.600	Out		60.0							
T2	10/08/04	11:14	5	13.6	19138	11.43	0.292	Out		29.2							
T1	10/08/04	11:20	1	13.7	3274	1.72	0.013	Out		1.3							
T1	10/08/04		2	13.6	3293	1.74	0.000	Out		0.0							
T1	10/08/04		3	13.6	3390	1.79	0.044	Out		4.4							
T1	10/08/04		4	13.6	14070	8.28	0.039	Out		3.9							
T1	10/08/04	11:27	5	13.6	21200	14.4	0.232	Out		23.2							
			-							SumAllQ							
								Ebb		879.0							
								Flood		0.0							

Penobsco	t River Transeo	t Data																
							Directional			Cell	Cell							
			Depth	Temp	SpecCond	Salinity	Velocity		THg	Discharge	Hg Load	DHg	TSS	TSS-Hg	Kds	Sample		
Station	Date	Time	(m)	(oC)	(uS/cm)	(ppth)	(m/sec)	Direction	(ng/L)	(m3/sec)	(ng/sec)	(ng/L)	(mg/L)	(ug/g)	(mL/g)	Time	Notes	
T4	10/08/04	12:58	1	13.9	2668	1.37	0.443	Out	2.46	44.3	108885							
T4	10/08/04		2	13.9	3350	1.77	0.280	Out	2.98	28.0	83538	0.79	0.97	2.26	6.0			
T4	10/08/04		3	13.9	4630	2.48	0.168	Out		16.8								
T4	10/08/04		4	13.8	7250	4.13	0.234	Out	2.95	23.4	68992							
T4	10/08/04		5	13.6	14986	8.76	0.066	Out	4.30	6.6	28219							
T4	10/08/04	13:05	5.5	13.5	23380	14.19	0.007	Out		0.4								
T3	10/08/04	13:19	1	13.8	2434	1.24	0.474	Out	1.5	47.4	71066					13:27		
T3	10/08/04		2	13.7	3603	1.89	0.392	Out	1.87	39.2	73267							
T3	10/08/04		3	13.7	4402	2.36	0.223	Out		22.3								
T3	10/08/04		4	13.6	11096	6.35	0.040	Out	2.61	4.0	10513							
T3	10/08/04		5	13.6	16590	9.8	0.000	Out	4.81	0.0	0	1.39	4.48	0.76	5.3	13:33	TMHg/DMH	g also
T3	10/08/04	13:26	5.5	13.6	17600	10.41	0.000	Out		0.0							Low tide at	13:26
T2	10/08/04	13:40	1	14	2845	1.49	0.205	Out	1.67	20.5	34183					13:48		
T2	10/08/04		2	13.9	3514	1.86	0.049	Out	1.88	4.9	9131							
T2	10/08/04		3	13.9	4156	2.22	0.015	Out	2.07	1.5	3090							
T2	10/08/04	13:46	3.5	13.7	7470	4.13	0.083	Out	2.36	4.1	9740	0.95	1.73	0.82	5.8	13:52	TMHg/DMH	g also
T1	10/08/04	13:59	1	14.1	2382	1.24	-0.037	In	1.59	-3.7	-5939					14:08		
T1	10/08/04		2	14	2590	1.35	-0.146	In	1.61	-14.6	-23511							
T1	10/08/04		3	13.9	3294	1.74	-0.170	In	2.52	-17.0	-42919							
T1	10/08/04	14:05	4	13.8	4918	2.63	-0.020	In	2.23	-2.0	-4530	0.89	1.87	0.72	5.7			
										SumAllQ								
								Ebb		263.3								
								Flood		-37.4								
										SumQ	SumJ	Q-wgtC	ng/sec	g/day				
								Ebb		223.8	500622	2.24	588941	50.88				
								Flood		-37.4	-76898	2.06	-76898	-6.64				

Penobsco	t River Transec	t Data																
							Directional			Cell	Cell							
			Depth	Temp	SpecCond	Salinity	Velocity		THg	Discharge	Hg Load	DHg	TSS	TSS-Hg	Kds	Sample		
Station	Date	Time	(m)	(oC)	(uS/cm)	(ppth)	(m/sec)	Direction	(ng/L)	(m3/sec)	(ng/sec)	(ng/L)	(mg/L)	(ug/g)	(mL/g)	Time	Notes	
T4	10/08/04	15:02	1	14.3	2666	1.39	-0.032	In		-3.2								
T4	10/08/04		2	14.3	2752	1.43	0.000	In		0.0								
T4	10/08/04		3	14.3	2903	1.52	-0.022	In		-2.2								
T4	10/08/04		4	13.9	5091	2.75	-0.140	In		-14.0								
T4	10/08/04		5	13.6	11148	6.38	-0.111	In		-11.1								
T4	10/08/04	15:10	5.5	13.6	16150	9.45	-0.098	In		-4.9								
T3	10/08/04	15:15	1	14.4	2764	1.45	-0.013	In		-1.3								
T3	10/08/04		2	14.2	3315	1.76	-0.019	In		-1.9								
T3	10/08/04		3	14	4147	2.21	-0.161	In		-16.1								
T3	10/08/04		4	13.9	4058	2.17	-0.084	In		-8.4								
T3	10/08/04		5	13.7	10265	5.83	-0.138	In		-13.8								
T3	10/08/04		6	13.6	21041	12.6	-0.108	In		-10.8								
T3	10/08/04	15:24	6.3	13.4	25990	15.9	-0.102	In		-3.0								
T2	10/08/04	15:29	1	14.2	3932	2.1	-0.110	In		-11.0								
T2	10/08/04		2	14.1	4013	2.14	-0.191	In		-19.1								
T2	10/08/04		3	14.1	3792	2	-0.165	In		-16.5								
T2	10/08/04		4	13.8	4962	2.82	-0.116	In		-11.6								
T2	10/08/04	15:37	4.5	13.8	5807	3.17	-0.139	In		-7.0								
T1	10/08/04	15:41	1	14.4	2606	1.32	-0.103	In		-10.3								
T1	10/08/04		2	14.4	2640	1.37	-0.017	In		-1.7								
T1	10/08/04		3	14.1	3772	1.94	0.000	In		0.0								
T1	10/08/04		4	14.1	3454	1.83	-0.005	In		-0.5								
T1	10/08/04	15:49	4.8	13.7	6120	3.35	-0.004	In		-0.3								
										SumAllQ								
								Ebb		0.0								
								Flood		-168.9								

Penobsco	t River Transec	t Data																
							Directional			Cell	Cell							
			Depth	Temp	SpecCond	Salinity	Velocity		THg	Discharge	Hg Load	DHg	TSS	TSS-Hg	Kds	Sample		
Station	Date	Time	(m)	(oC)	(uS/cm)	(ppth)	(m/sec)	Direction	(ng/L)	(m3/sec)	(ng/sec)	(ng/L)	(mg/L)	(ug/g)	(mL/g)	Time	Notes	
T4	10/08/04	16:02	1	14.3	2832	1.48	-0.091	In	2.29	-9.1	-20753							
T4	10/08/04		2	14	3300	1.74	-0.003	In		-0.3								
T4	10/08/04		3	14.1	3472	1.83	-0.129	In	1.79	-12.9	-23053							
T4	10/08/04		4	14.1	3744	1.99	-0.011	In		-1.1								
T4	10/08/04		5	13.6	10620	6.04	-0.155	In	3.39	-15.5	-52439							
T4	10/08/04	16:10	5.9	13.6	19260	11.49	-0.192	In	4.99	-17.3	-86256	1.04	4.15	0.95	5.4	16:20	TMHg/DMHg	also
T3	10/08/04	16:24	1	14.3	3080	1.62	-0.272	In	1.90	-27.2	-51617							
T3	10/08/04		2	14.3	3210	1.69	-0.218	In		-21.8								
T3	10/08/04		3	14	4169	2.24	-0.207	In	2.15	-20.7	-44410							
T3	10/08/04		4	13.7	7378	4.11	-0.282	In		-28.2								
Т3	10/08/04		5	13.6	21408	12.89	-0.393	In	4.91	-39.3	-193071							
T3	10/08/04		6	13.4	25120	15.35	-0.384	In		-38.4								
T3	10/08/04	16:33	6.7	13.3	28259	17.4	-0.233	In	28.0	-16.3	-457333	1.07	22.2	1.21	4.7	16:40		
T2	10/08/04	16:43	1	14.3	3152	1.66	-0.169	In	1.89	-16.9	-32008							
T2	10/08/04		2	14.2	3272	1.72	-0.164	In		-16.4								
T2	10/08/04		3	13.7	6550	3.62	-0.268	In	2.89	-26.8	-77377							
T2	10/08/04		4	13.6	16640	9.9	-0.450	In	3.83	-45.0	-172350							
T2	10/08/04		5	13.4	24640	14.68	-0.334	In	4.64	-33.4	-155174	1.02	5.18	0.70	5.3	16:59	TMHq/DMHq	also
T2	10/08/04	16:50	5.3	13.4	25395	15.47	-0.343	In		-10.3								
T1	10/08/04	17:01	1	14.2	3318	1.75	-0.172	In	1.69	-17.2	-29090							
T1	10/08/04		2	14.2	3335	1.76	-0.130	In	1.81	-13.0	-23609							
T1	10/08/04		3	13.8	8050	4.53	-0.221	In		-22.1								
T1	10/08/04		4	13.6	16420	9.65	-0.171	In	4.38	-17.1	-74884							
T1	10/08/04		5	13.4	24881	15.18	-0.302	In	7.46	-30.2	-225023	1.11	6.18	1.03	5.2			
T1	10/08/04	17:11	5.5	13.4	26126	16.01	-0.239	In		-11.9								
										SumAlIQ								
								Ebb		0.0								
								Flood		-508.2								
										SumQ	SumJ	Q-wgtC	ng/sec	g/day				
								Ebb		0.0			_					
								Flood		-357.8	-1718447	4.80	-2440670	-210.87				

Photographs



3-inch Parshall flume installation at Northerly Stream



90-Degree weir installation on 35-inch pipe at Southerly Stream



Foreshore seepage below Landfill 1, HoltraChem Site



Rock bluff seepage below Landfills 3, 4 and 5, HoltraChem Site



Foreshore porewater collection using well screen



Lee Type seepage meter installed in foreshore prior to rising tide



Continuous radon monitoring float - March 2009

**Field And Matrix Replicates** 

Field and Matrix Replicates												
Date	Sample ID	Sample ID Type Analyte Rep 1 Rep 2 Rep 3 %F										
3/23/2010	NS-DN-T	MD	THg	37900	38100		-0.53					
3/23/2010	NS-DN-T	MD	TSS	94	95.6		-1.69					
2/25/2010	SS-PIPEN-D	MD	THg	82.1	77.9		5.25					
6/1/2010	NS-DN-COMP	MD	THg	30200	30800		-1.97					
6/17/2010	NS-DN-COMP	MD	THg	14500	13200		9.39					
4/27/2010	NS-DN-T	MD	THg	2440	2390		2.07					
6/5/2010	NS-DN-T	MD	THg	35500	37000		-4.14					
6/5/2010	NS-DN-T	MD	TSS	87.7	88.7		-1.13					
4/21/2009	NS-DN-COMP	MD	THg	2400	2630		-9.15					
4/24/2009	NS-DN-COMP	MD	THg	4780	4640		2.97					
5/6/2009	NS-DN-COMP	MD	THg	5310	5120		3.64					
5/20/2009	NS-DN-COMP	MD	THg	8820	9050		-2.57					
6/12/2009	NS-DN-COMP	MD	THg	43200	45400		-4.97					
6/22/2009	NS-DN-COMP	MD	THg	22800	23100		-1.31					
7/24/2009	NS-DN-COMP	MD	THg	6340	6540		-3.11					
8/20/2009	NS-DN-COMP	MD	THg	4860	4840		0.41					
9/14/2009	NS-DN-COMP	MD	THg	7450	7470		-0.27					
10/12/2009	NS-DN-COMP	MD	THg	2160	2140		0.93					
10/27/2009	NS-DN-COMP	MD	THg	7810	7810		0.00					
11/25/2009	NS-DN-COMP	MD	THg	4410	4440		-0.68					
12/11/2009	NS-DN-COMP	MD	THg	2910	2890		0.69					
4/2/2010	NS-DN-COMP	MD	THg	10100	9590		5.18					
4/20/2010	NS-DN-COMP	MD	THg	3970	4080		-2.73					
4/29/2010	NS-DN-COMP	MD	THg	5160	5280		-2.30					
5/18/2010	NS-DN-COMP	MD	THg	15900	15300		3.85					
3/25/2009	SS-SPR-UNF	MD	THg	87.3	91.1		-4.26					
3/26/2009	SS-UP-FILT	MD	THg	336	275		19.97					
4/21/2009	SS-PIPE-D	MD	THg	124	132		-6.25					
5/26/2009	SS-PIPE-T-AUTO	MD	THg	123	118		4.15					

Field and Matrix Replicates												
Date	Sample ID Type Analyte		Rep 1	Rep 2	Rep 3	%RPD						
5/26/2009	SS-PIPE-T-AUTO	FD	THg	111	126		-12.66					
5/26/2009	SS-PIPE-T-PIPE	FD	THg	136	133	126	3.90					
7/30/2009	NS-DN-T	MD	THg	201	194		3.54					
8/20/2009	SS-DN-T	MD	THg	531	476		10.92					
9/29/2009	NS-DN-D	MD	THg	354	352		0.57					
10/27/2009	NS-DN-T	MD	THg	3240	3160		2.50					
10/12/2009	K-NORTH-THG	MD	THg	2.42	2.58		-6.40					
1/26/2010	NS-DN-T	MD	THg	2160	2110		2.34					
1/26/2010	SS-PERC-T	MD	TSS	30.2	30.1		0.33					
12/29/2009	NS-DN-T	MD	THg	1616	1634		-1.11					
9/18/2009	PW-13	MD	THg	18.9	16.5		13.56					
9/18/2009	PW-13	FD	THg	18.9	16.1		16.00					
9/19/2009	SEEP1B	MD	THg	7.8	8.04		-3.03					
9/19/2009	SEEP3	FD	THg	134	126		6.15					
						Mean	1.12					
						StDev	6.21					

**Grab Sample Data** 

Results of Grab Sample Analysis at HoltraChem Surface Streams										
Location	Datetime	Discharge (gpm)	Unfiltered Total Hg (ng/L)	Filtered Total Hg (ng/L)	%Diss	TSS (mg/L)	Suspended Hg (ug/g)			
NS-DN	10/26/07 14:00		104	48.4	46.5	0.615	90.6			
NS-DN	10/25/08 0:00		580							
NS-DN	3/26/09 12:15	25.00	1,190	809	68.0					
NS-DN	4/21/09 12:55	129.63	11,700	783	6.7	43.3	252			
NS-DN	5/26/09 11:00	14.26	6,230	544	8.7	15.4	369			
NS-DN	6/22/09 11:00	57.68	4,420	1470	33.3	4.34	680			
NS-DN	7/30/09 11:55	27.51	201	74.8	37.2	2.44	52			
NS-DN	8/20/09 16:20	9.28	1,220	503	41.2	2.48	289			
NS-DN	9/29/09 12:50	2.87	1,060	354	33.4	1.08	654			
NS-DN	10/27/09 10:00	17.51	3,160	1130	35.8	1.09	1862			
NS-DN	11/30/09 11:00	30.03	2,050	890	43.4	1.17	991			
NS-DN	12/29/09 13:30	28.60	1,616	772	47.8	1.26	670			
NS-DN	1/7/10 13:05	21.56	1,280							
NS-DN	1/15/10 15:00	17.18	1,740							
NS-DN	1/25/10 12:00	18.60	1,630							
NS-DN	1/26/10 9:00	118.06	2,110	850	40.3	3.88	325			
NS-DN	2/12/10 0:00		2,140							
NS-DN	2/19/10 16:49	13.08	1,500							
NS-DN	2/25/10 8:45	62.69	16,900	487	2.9	38.1	431			
NS-DN	2/26/10 14:04	35.51	1,650	482	29.2	2.24	521			
NS-DN	3/23/10 14:30	149.85	38,000	581	1.5	95	394			
NS-DN	3/30/10 11:05	78.50	3,128	1030	32.9	4.15	506			
NS-DN	4/27/10 13:10	17.29	2,440	684	28.0	2.03	865			
NS-DN	5/25/10 16:20	5.98	3,635	464	12.8	4.65	682			
NS-DN	6/5/10 9:57	112.62	35,500	586	1.7	87.7	398			
Mean			5,807	660	29	17	557			
Standard	Deviation		9,825	334	18	29	397			
Minimum			104.00	48.40	1.53	0.62	51.72			

Maximum	ו		38,000 1,470			68	95	1,862			
Results of Grab Sample Analysis at HoltraChem Surface Streams											
Location	Datetime	Discharge (gpm)	Unfiltered Total Hg (ng/L)	Unfiltered Filtered Total Hg (ng/L) (ng/L)		TSS (mg/L	_) S	uspended Hg (ug/g)			
NS-UP	10/26/07 14:00		478	166	34.7	0.706	442				
NS-UP	3/26/09 12:35		2,570	1090	42.4						
PERC	6/22/09 14:15		35	4.72	13.3	20.6	1.5				
PERC	9/29/09 15:05		77	4.07	5.3	42.2	1.7				
PERC	10/27/09 12:00		11	3.00	26.3	3.47	2.4				
PERC	11/30/09 0:00		22	3.57	16.3	9.58	1.9				
PERC	12/29/09 0:00		32	2.51	7.8	20.1	1.5				
PERC	1/26/10 0:00		66	5.02	7.6	30.2	2.0				
PERC	2/25/10 9:45		215	8.56	4.0	74.4	2.8				
PERC	2/26/10 13:20		106	9.09	8.6	36.7	2.6				
PERC	3/30/10 11:40		37	6.92	18.8	13.1	2.3				
PERC	4/27/10 14:00		11	2.71	25.1	6.38	1.3				
PERC	5/25/10 16:55		10	2.52	25.0	9.18	0.8				
Mean			57	5	14	24	2				
Standard	Deviation		61	2	8	21	1				
Minimum			10	3	4	3	1				
Maximum			215	9	26	74	3				

	Results of Grab Sample Analysis at HoltraChem Surface Streams										
Location	Datetime	Discharge (gpm)	Unfiltered Total Hg (ng/L)	Filtered Total Hg (ng/L)	%Diss	TSS (mg/L)	Suspended Hg (ug/g)				
SS-DN	10/26/07 0:00		51	20.2	39.3	1.51	20.7				
SS-DN	10/25/08 0:00		57								
SS-DN	3/26/09 13:05		456	125	27.4						
SS-DN	7/30/09 13:32		2,480	837	33.8	1.98	830				
SS-DN	8/20/09 17:45		531	42	7.9	7.85	62				
SS-DN	9/29/09 15:20		108	26.2	24.3	6.93	12				
SS-DN	10/27/09 12:15		295	128	43.4	1.54	108				
SS-DN	11/30/09 0:00		285	80.4	28.2	4.28	48				
SS-DN	12/29/09 0:00		278			8.81					
SS-DN	1/26/10 0:00		1,450	47.8	3.3	32.8	43				
SS-DN	2/25/10 10:15		1,620	44.5	2.7	50.5	31				
SS-DN	2/26/10 13:35		2,410	483	20.0	31.9	60				
SS-DN	3/30/10 11:50		634	55.1	8.7	13.1	44				
SS-DN	4/27/10 14:20		479	25.7	5.4	24.4	19				
SS-DN	5/25/10 17:10		412	17.8	4.3	20.3	19				
Mean			770	149	19	16	108				
Standard	Deviation		790	231	14	15	219				
Minimum			51	18	3	2	12				
Maximum			2,480	837	43	51	830				

Results of Grab Sample Analysis at HoltraChem Surface Streams										
Location	Datetime	Discharge (gpm)	Unfiltered Total Hg (ng/L)	Filtered Total Hg (ng/L)	%Diss	TSS (mg/L)	Suspended Hg (ug/g)			
SS-PIPE	3/26/09 13:20		869	227	26.1					
SS-PIPE	4/21/09 14:30	279	1,010	124	12.3	13.5	66			
SS-PIPE	5/26/09 12:00	27.9	126	46.5	36.9	0.71	113			
SS-PIPE	6/22/09 14:15	346	1,200	258	21.5	7.14	132			
SS-PIPEN	7/30/09 13:09	30.7	301	143	47.5	1.03	153			
SS-PIPEN	8/20/09 17:25	2.0	344	61.7	17.9	3.68	77			
SS-PIPEN	9/29/09 14:50	8.5	136	77.9	57.3	0.33	175			
SS-PIPEN	10/27/09 11:45	31.9	876	287	32.8	2.03	290			
SS-PIPEN	11/30/09 12:30	96.5	663	153	23.1	2.68	190			
SS-PIPEN	12/29/09 0:00	129	430	113	26.3	4.49	71			
SS-PIPEN	1/7/10 13:54	46.9	187							
SS-PIPEN	1/26/10 10:00	909	1,640	57.7	3.5	26	61			
SS-PIPEN	2/25/10 9:30	NA	2,120	82.1	3.9	29.9	68			
SS-PIPEN	2/26/10 13:10	561	3,670	64.3	1.8	49.2	73			
SS-PIPEN	3/23/10 14:44	688	11,900	113	0.9	148	80			
SS-PIPEN	3/30/10 11:24	873	659	76.5	11.6	9.76	60			
SS-PIPEN	4/27/10 13:50	24.1	205	59.2	28.9	1.04	140			
SS-PIPEN	5/25/10 16:45	5.3	171	62.5	36.5	2.56	42			
SS-PIPEN	6/5/10 10:05	211	5,600	117	2.1	72.8	75			
Mean		·	1,690	118	22	22	110			
Standard D	eviation		2,836	72	17	38	64			
Minimum			126	47	1	0.3	42			
Maximum			11,900	287	57	148	290			

Results of Grab Sample Analysis at HoltraChem Surface Streams												
Location	Datetime	Discharge (gpm)	Unfiltered Total Hg (ng/L)	Filtered Total Hg (ng/L)	%Diss	TSS (mg/L)	Suspended Hg (ug/g)					
SS-SPR	10/26/07 0:00		76	38.2	50.4	75.3	0.51					
SS-SPR	3/25/09 17:25		87									
SS-UP	3/26/09 13:55	128.00	1,060	336	31.7							
SS-UP	10/26/07 0:00		133	47.1	35.4	0.618	139					
WTP-EFF	3/26/09 10:00		4,870									

Regression Analysis to Simulate Discharge on Southerly Stream, June 20-July 9, 2009.



Estimates of Annual Mercury Loading to Penobscot River, 1967-2011.

Year	Outfalls g/day	Streams g/day	GW g/day	Total g/day	mt/yr	cum tons	Notes
1967	7500	100	0	7600	2.77	0.42	Brine waste to river
1968	7500	100	0	7600	2.77	3.19	Brine waste to river
1969	7500	100	0	7600	2.77	5.97	Brine waste to river
1970	500	10	0	510	0.19	6.15	First measurements July-August- brine waste directed to on site pond
1971	100	10	20	130	0.05	6.20	
1972	45	10	20	75	0.03	6.23	
1973	45	10	20	75	0.03	6.26	
1974	45	10	20	75	0.03	6.28	
1975	45	10	20	75	0.03	6.31	
1976	45	10	20	75	0.03	6.34	
1977	45	10	20	75	0.03	6.37	
1978	45	10	20	75	0.03	6.39	
1979	45	10	20	75	0.03	6.42	
1980	45	10	20	75	0.03	6.45	
1981	45	10	20	75	0.03	6.48	
1982	45	10	20	75	0.03	6.50	
1983	45	10	20	75	0.03	6.53	
1984	45	10	20	75	0.03	6.56	
1985	45	10	20	75	0.03	6.58	
1986	45	10	20	75	0.03	6.61	
1987	12	10	20	42	0.02	6.63	TRI-DMR Data starts
1988	12	10	20	42	0.02	6.64	
1989	12	10	20	42	0.02	6.66	
1990	12	10	20	42	0.02	6.67	
1991	12	10	20	42	0.02	6.69	
1992	12	10	20	42	0.02	6.70	
1993	12	10	20	42	0.02	6.72	
1994	12	10	20	42	0.02	6.73	
1995	12	10	20	42	0.02	6.75	

Year	Outfalls g/day	Streams g/day	GW g/day	Total g/day	mt/yr	cum tons	Notes
1996	12	10	20	42	0.02	6.77	
1997	12	10	20	42	0.02	6.78	
1998	12	10	20	42	0.02	6.80	
1999	12	10	20	42	0.02	6.81	
2000	6	6	20	32	0.01	6.82	Plant closes
2001	6	6	20	32	0.01	6.83	
2002	6	6	20	32	0.01	6.85	
2003	6	6	20	32	0.01	6.86	
2004	6	6	20	32	0.01	6.87	GW P&T starts
2005	0.1	6	1	7.1	0.003	6.87	
2006	0.1	6	1	7.1	0.003	6.88	
2007	0.1	6	1	7.1	0.003	6.88	
2008	0.1	6	1	7.1	0.003	6.88	
2009	0.1	6	1	7.1	0.003	6.88	
2010	0.1	6	1	7.1	0.003	6.89	
2011	0.1	6	1	7.1	0.003	6.89	

Memo Describing Effort Using Divers to Locate Submerged Discharge Pipe at HoltraChem Site.

Study Panel

Penobscot River Mercury Study

# Report on Effort to Locate Submerged Outfall Pipe (Outfall 001) at HoltraChem Site

At 9:00 AM on September 8, 2011 I met two Normandeau divers (team leader Erik Feldotto) and boat operator at Hamlin Marina in Hampden, Maine. We proceeded down river to coordinates I had scaled from a custom bathymetric chart I had prepared earlier. The chart displayed a suspect feature slightly north of where the submerged outfall pipe was thought to exist based on its trace within the intertidal and upland mapping. A temporary buoy was set at these coordinates to provide an underwater position to begin the search. As the search began (9:30) the tide was ebbing with a relative strong current speed (2-3 ft/sec) and water clarity was poor (<1 foot). At or near this location the divers identified a debris pile consisting of several large logs (up to 1 ft in diameter) apparently partially buried in a local mound of sediment but no evidence of a pipe beneath or in the vicinity of the debris. The search was then shifted southward about 50 feet and extended from shallower water (intertidal zone) where the divers positively identified a concrete slab capping the pipe in the upper intertidal and a shallow pit with survey flag known to be a seepage zone probably associated with the pipe or its backfill. From the latter reference point the divers searched westward, towards deeper water, out to a maximum water depth of 29 feet without finding any evidence of the pipe. At this point more than two hours and four tanks of breathing air had been expended to locate the pipe. After a short phone consultation with Drew Bodaly the search was terminated and we returned to the boat launch, arriving at ~12:30PM.

My conclusion from this diver search and my own wading search during a very low tide (-1.7 ft) in July is that the original end of the pipe is now buried, or long ago swept away by ice, and its current discharge is via the seepage visible in the intertidal zone that we (and others) have sampled for mercury on several occasions.

Ralph Turner

#### Data and Calculations Supporting TSS and Mercury Loading For Penobscot River at Eddington, Maine and For Selected Downstream Tributaries

#### **Objectives**

The main objective of this exercise was to estimate particle (TSS) and mercury (Hg) loading (fluxes) from all major fluvial sources between Veazie Dam in Bangor and Bucksport, Maine. These sources included Penobscot River at Eddington, Kenduskeag Stream, Souadabscook Stream, Penjajawoc Stream, Reeds Brook, Cove Brook, North Marsh River, Colson Stream (South Marsh River), BO5 Stream, Eaton Brook, Fells Brook, Sedgeunkedunk Stream and Mill Creek. Particle fluxes were needed to support modelling of the natural recovery rate from historical mercury inputs at HoltraChem site in Orrington, Maine. Estimation of particulate Hg loading also required good particle flux data as Hg has a strong affinity for particles and its fluvial transport is often dominated by particle transport.

#### <u>Methods</u>

More than one method of TSS and Hg fluxes was employed whenever possible to improve confidence of the calculated fluxes. Simpler methods employed mean concentration (discharge-weighted when possible) values and mean annual discharge, whereas more sophisticated methods employed regression analyses of relationships between the following:

- Discharge and stage height
- Discharge and watershed properties (Dudley 2004)
- Penobscot River Discharges at Eddington and West Enfield Gauging Stations
- Turbidity and Total Suspended Solids (TSS)
- Discharge and Filter-passing Mercury (ng/L)
- Discharge and Particulate Mercury (µg/g dw)

For the Penobscot River these relationships (except discharge and watershed properties) were exploited to generate daily flux values that could be summed annually. For example, turbidity was recorded continuously by the USGS in the Penobscot River at Eddington whereas TSS was measured by sampling and laboratory analysis only periodically in order to develop a calibration between these measurements. Similarly, useful relationships were observed between discharge and total, filter-passing and particulate Hg which were exploited to generate daily total, filter-passing and particulate Hg flux values that could be summed annually. Generally the period used for flux calculation was defined by availability of daily turbidity values and spanned part of 2007, all of 2008 and 2009, and parts of 2010 and 2011. For tributary fluxes other than the main Penobscot River at Eddington we used the best available analytical data from
sampling and average annual discharge for each tributary and contributing area as derived from watershed properties (Dudley 2004).

## Penobscot River near Veazie Dam

Discharge (Q) for the Penobscot River at Eddington was required to estimate TSS and Hg loading upstream of the influence from Hg inputs from the HoltraChem site. Discharge has not recently been reported continuously at the USGS Eddington gage, only river stage height (H), and for a limited period, velocity. However, discharge is reported continuously upstream of Veazie Dam at the USGS West Enfield gage and discharge was reported at both gages through 1996. The latter data set provided the opportunity to develop a regression equation relating West Enfield and Eddington discharges (Figure 1) and further, to match stage height and to the estimated discharge at Eddington (Figure 2). As noted by the USGS (Rantz 1982), stage-discharge relationships can change when/if the "control" cross section changes due to sedimentation or erosion. Cross sections under bedrock control tend to produce temporally stable stage-discharge relationships. The control at Eddington is thought to be reasonably stable as suggested by a plot of field-measured discharges between 1979 and 2012 (Figure 3). The power function fit to these data (Q=849xH<sup>1.65</sup>) is very similar to the function derived from the West Enfield-Eddington discharge function (Q= 1001xH<sup>1.60</sup>). For example, the maximum difference (e.g., 13% RPD at 2 feet) in predicted discharges by each equation occurs at the lowest stage heights and decreases to <5% at stage heights >10 feet. Thus, either of these relationships could have been used to estimate discharges for the purposes of this report. The West Enfield-Eddington function was selected for loading calculations. Note that discharge and gage height data reported by the USGS are in units of cubic feet per second and feet, respectively. Subsequently, we use metric units for discharge (m<sup>3</sup>/s) to facilitate loading calculations that use concentrations reported in metric units (mg/L, ng/L, µg/g).



Figure 1. Relationship of discharge at Eddington (1-day later) to West Endfield, 1991-1996.



Figure 2. Eddington discharge for 2006-2010 estimated using relationship developed from 1991-1996 data with daily discharge rates measured at both West Enfield and Eddington. Discharges estimated for Eddington from W. Enfield data are plotted here against gage height measured at Eddington.





http://waterdata.usgs.gov/nwis/measurements?site\_no=01036390&agency\_cd=USGS&format=html\_table

TSS concentration data were required to estimate particle loading and to calculate concentration of mercury on particles (referred to hereafter as particulate Hg, PTHg,  $\mu$ g/g dry wt.). Only a limited amount of TSS data are available but it was all collected by PRMS scientists at the same time as Hg data were collected, thus allowing calculation of particulate Hg. As discussed in the next paragraph additional TSS concentration values (daily) were estimated using *in-situ* turbidity measurements reported by the USGS. TSS was calibrated to turbidity using data for both collected between October 15, 2010 and November 24, 2010 (N=14).

Turbidity has been measured with an in-situ sensor installed and operated by the USGS at Eddington since 2007. This afforded the opportunity to predict TSS for any period when turbidity had been measured if a robust relationship could be established. As shown in Figure 4 a good relationship (TSS=1.70xTurbidity – 0.06) was found and we used it to predict daily values for TSS that were multiplied by discharge to generate daily and annual TSS loadings from August 16, 2007 through December 30, 2011. Note that in those cases where the daily turbidity value was reported as zero, a value of 0.1 mg/L was substituted. In addition, when no measured turbidity data were available the relationship between discharge and turbidity (Figure 5) was used to estimate a value. The latter relationship has a low r-squared but its use allowed filling of data gaps (overall ~370 days out of 1600) with best available estimates.







Figure 5. Relationship between discharge and turbidity used to generate estimate daily turbidity values only when no measured turbidity data were available.

Available analytical data for concentrations of total Hg, filter-passing Hg, TSS and particulate Hg in the Penobscot River near Veazie are given in Table 1, all collected under the auspices of the PRMS. Note that particulate Hg was calculated from measurements of total Hg, filter-passing Hg and TSS using the following equation:

Particulate Hg,  $\mu$ g/g dw = (Total Hg,ng/L – Filter-passing Hg, ng/L)/TSS, mg/L

Concentration data in this table have also been matched with stage height and discharge data for Eddington, calculated as described above. Discharge-weighted averages for TSS, total Hg and filter-passing Hg are also given as these values will be used subsequently to calculate fluxes by the "simple" method. A discharge-weighted average, C<sub>wgt</sub>, is calculated by first multiplying discharge, Q, by concentration, C, for each paired observation and then dividing the sum of these fluxes by the sum of discharges as follows:

$$C_{wgt} = \sum (Q_i \times C_i) / \sum Q_i$$

Discharge-weighted averages are useful for estimating solute and particulate loads because they "emphasize the composition of water during periods of high discharge" and because this average "may be thought of as representing the composition of water passing the sampling point during the period of the average.... if it had been collected and mixed in a large container" (Hem 1989). Obviously the robustness of this average is improved when it includes as wide a range of discharges as possible. For the Penobscot River analytical data the discharges sampled between 2004 and 2011 ranged over an order of magnitude (150 to 1500 m<sup>3</sup>/s) and averaged 618 m<sup>3</sup>/s, while the range of discharges that occurred during the main period of interest (August, 2007 through December, 2011) ranged from 126 to 3141 m<sup>3</sup>/s, with an average of 477 m<sup>3</sup>/s. While the very highest discharges that occurred were not sampled, discharges >1500 m<sup>3</sup>/s occurred only 2.7% of the time (43 out of 1578 days) during this period. Thus, the weighted averages shown at the bottom of Table 1 for TSS, total Hg and filter-passing Hg are judged to be reasonably robust for load calculations using the simple method. The weighted average TSS value shown in Table 1 was not used for the simple loading calculations because far better values (3.07 to 3.72 mg/L, N = 365) could be calculated from the measured turbidity and predicted TSS data. Note also that the weighted average for particulate Hg (µg/g dry wt.) was calculated in a different manner to preserve dimensional correctness when the value is used to calculate summary loading, e.g.,

## ∑(TSS x Q)<sub>annual</sub> x PTHg<sub>wgt</sub>

Relationships used to generate <u>daily</u> loading values for filter-passing, particulate and total Hg are shown in Figures 6 through 8, respectively. For filter-passing and total Hg the predicted concentrations were multiplied by the daily average discharge values to calculate loading and these were the summed for each period of interest. One estimate of particulate Hg loading was calculated as the difference between total and filter-passing Hg loadings. A second estimate was calculated by first multiplying the predicted daily particulate Hg concentration by the predicted daily TSS concentration and then

multiplying by discharge. These two estimates should be similar as they are derived from the same measurement data.

Table 1. Complete analytical data for mercury and TSS for Penobscot River at or above Veazie, 2004-2011. Note that Veazie-Upstream, Veazie-OV2 and Veazie-UV are all located immediately upstream of Veazie Dam while Veazie-Edd is located immediately downstream.								
StationID	Date	Gage Height (ft)	Discharge (m³/s)	Total Hg (ng/L)	Filter- Passing (Hg ng/L)	TSS (mg/L)	Particulate Hg (µg/g dry wt)	
Veazie-Edd	10/06/04	3.46	206.7	1.64	1.28	0.49	0.73	
Veazie-Edd	10/09/04	3.54	214.4	1.24	1.11	0.50	0.26	
Veazie-Edd	10/15/10	4.3	292.4	2.23	1.83	0.50	0.8	
Veazie-Edd	11/09/10	8.5	867.1	5.96	3.82	15.1	0.142	
Veazie-Edd	11/10/10	11	1308	6.26	4.79	22.5	0.065	
Veazie-Edd	11/11/10	12	1503	5.60	4.76	11.5	0.073	
Veazie-Edd	11/12/10	11	1308	5.05	3.95	5.67	0.194	
Veazie-Edd	11/13/10	10	1124	4.46	3.88	3.06	0.190	
Veazie-Edd	11/15/10	8	787.2	3.99	3.48	2.45	0.208	
Veazie-Edd	11/17/10	7.1	650.7	3.79	3.16	1.87	0.337	
Veazie-Edd	11/18/10	7.3	680.2	3.62	3.02	1.91	0.314	
Veazie-Edd	11/19/10	8.2	818.8	3.47	2.53	3.18	0.296	
Veazie-Edd	11/20/10	7.2	665.4	4.24	3.58	2.87	0.228	
Veazie-Edd	11/22/10	6.5	565.2	3.08	2.64	1.28	0.344	
Veazie-Edd	11/24/10	6	497.5	3.02	2.06	1.27	0.756	
Veazie-Edd	8/27/11	4.79	347.3	2.96	2.44	1.41	0.369	
Veazie-Edd	8/29/11	6.1	510.8	3.34	2.38	4.75	0.202	
Veazie-Edd	8/30/11	10.3	1180	6.01	2.63	17.7	0.191	
Veazie-Edd	9/02/11	7.15	658.0	4.94	4.04	9.22	0.098	
Veazie-OV2	8/3/06	4.3	292.4	2.94	2.26			
Veazie-OV2	9/11/06	3.5	213.4	2.67	2.55			
Veazie-OV2	10/3/06	3.9	246.1	2.12	1.74			
Veazie-OV2	10/23/06	8.7	903.2	5.71	3.58	10.2	0.21	
Veazie-OV2	5/31/07	5.2	397.1	3.29	2.56	1.64	0.45	

Table 1. Complete analytical data for mercury and TSS for Penobscot River at or above Veazie, 2004-2011. Note that Veazie-Upstream, Veazie-OV2 and Veazie-UV are all located immediately upstream of Veazie Dam while Veazie-Edd is located immediately downstream.								
StationID	Date	Gage Height (ft)	Discharge (m³/s)	Total Hg (ng/L)	Filter- Passing (Hg ng/L)	TSS (mg/L)	Particulate Hg (µg/g dry wt)	
Veazie-OV2	7/11/07	3.2	182.5	2.16	1.73	2.07	0.21	
Veazie- Upstream	10/15/10	4.3	292.4	2.52	1.82	1.08	0.648	
Veazie- Upstream	10/18/10	6.3	540.5	4.13	2.80	4.67	0.285	
Veazie-UV	8/19/08	4.8	348.5	2.87	2.59	0.78	0.36	
Veazie-UV	4/17/09	9.4	1015	3.34	2.81	1.89	0.28	
Veazie-UV	7/8/09	8.0	787.2	3.62	2.98	1.93	0.33	
Veazie-UV	9/3/09	3.6	220.2	1.82	1.69	0.50	0.25	
Veazie-UV	7/28/10	2.8	149.2	1.84	1.61	0.59	0.380	
Discharge-weighted means4.273.236.480.15 <sup>a</sup>								

<sup>a</sup> Value is mass-weighted PTHg and was calculated as  $\Sigma$  (TSS<sub>i</sub> x Q<sub>i</sub> x PTHg<sub>i</sub>)/ $\Sigma$  (TSS<sub>i</sub> x Q<sub>i</sub>)



Figure 6. Relationship between filter-passing Hg and discharge in Penobscot River near Veazie Dam. Function was used to generate daily values for filter-passing Hg loading.



Figure 7. Relationship between particulate mercury and discharge in Penobscot River near Veazie Dam. Function was used with estimated daily TSS values to generate daily values for particulate Hg loading.



Figure 8. Relationship between total mercury and discharge in Penobscot River near Veazie Dam. Function was used to generate daily values for total Hg loading.

Table 2 compares loads (kg/year) of particles and Hg for each period of interest, including as long-term averages, as calculated by the methods indicated. In general the agreement between loads calculated by different methods is reasonably good. For example, filter-passing Hg loads (41 to 54 kg/yr) calculated using the discharge-weighted mean from Table 1 (3.23 ng/L) varied by no more than 2 kg/yr from those (40 to 55 kg/yr) calculated using the sums of daily predicted loads. Similarly, total Hg loadings (55 to 72 kg/yr) calculated using the discharge-weighted average mean from Table 1 (4.27 ng/L) varied by no more than 7 kg/yr from those (50 to 69 kg/yr) calculated using sums of daily predicted values. Particulate Hg loads (8.0 to 13.4 kg/yr) calculated by summing daily products of discharge (m<sup>3</sup>/s) times TSS (mg/L) times particulate Hg (µg/g dry wt.) also compared well with loads (10 to 14 kg/yr) calculated as the difference between total and filtering-passing Hg loads. Results from the methods using daily sums and the long-term average discharge are recommended and will be carried forward in this chapter and elsewhere. These are:

Total Hg loading (kg/yr) = 50

Filter-passing Hg loading (kg/yr) = 40

Particulate Hg loading (kg/yr) = 8.7

by different methods.							
Variable	Method [Figure Number)	2008	2009	2010	2011	1979- 1996	
Discharge (m <sup>3</sup> x10 <sup>10</sup> )	∑(1001 x H <sup>1.60</sup> ) <sub>daily</sub> [2]	1.68	1.54	1.48	1.59	1.28	
TSS <sub>Qwgt</sub> (mg/L)	TSS <sub>annual load</sub> /Q <sub>annual</sub>	3.07	3.59	_ <sup>a</sup>	3.72	3.46	
TSS Load (kg/yr)	∑(TSS x Q) <sub>daily</sub>	51706	55340	<b>-</b> a	59148	44220	
FTHg (ng/L)	∑(FTHg x Q) <sub>daily</sub> /∑Q <sub>annual</sub>	3.3	3.0	3.0	3.1	3.1	
FTHg Load (kg/yr)	∑((0.002 x FTHg +1.51) <sub>daily</sub> x Q <sub>daily</sub> )[6]	55	46	45	49	40	
FTHg Load (kg/yr)	3.23 x Q <sub>annual</sub>	54	49	47	51	41	
PTHg <sub>wgt</sub> (μg/g dw)	$\sum_{A} (TSS x Q x PTHg)_{Aaily} / \sum (TSS x Q)_{Aaily}$	0.15	0.21	0.19	0.23	0.20	
PTHg Load (kg/yr)	∑(TSS x Q x PTHg) <sub>daily</sub>	8.0	11.5	12.4	13.4	8.7	
THg (ng/L)	∑(THg x Q) <sub>daily</sub> /∑Q <sub>annual</sub>	4.1	3.9	3.8	4.0	3.9	
THg Load (kg/yr)	∑((1.82 x InTHg <sub>daily</sub> -7.77) x Q <sub>daily</sub> )[8]	69	59	57	63	50	
THg Load (kg/yr)	4.27 x Q <sub>annual</sub>	72	66	63	68	55	
PTHg Load (kg/yr) <sup>b</sup>	THg Load – FTHg Load	14	13	12	14	10	

Table 2. Summary of mean concentrations (bold italics) and annual loads (kg) of particles (TSS) and Hg for Penobscot River near Veazie Dam as calculated by different methods.

<sup>a</sup> Insuffcient data. <sup>b</sup> Using difference in annual total loads estimated by summing daily loads.

## **Downstream Tributaries**

Available discharge and analytical data are for downstream tributaries tabulated in Table 3. No discharge data were available for times of sampling nor were any discharges measured or field-estimated by PRMS scientists. In lieu of actual discharge data we used estimated mean annual discharges calculated as described in Dudley (2004), or in the case of Kenduskeag Stream, the long-term average. The Dudley (2004) calculation employs watershed area (A, mi<sup>2</sup>) to estimate mean annual discharge (feet<sup>3</sup>/s) from this expression:

$$Q_{\text{mean cfs}} = (1.151 \text{ x } \text{A}^{0.991}) \text{ x } 1.9546$$

Tributary TSS loading was estimated using both the actual average TSS concentration (or a single value if that was all that was available) for each tributary and a fixed value (5 mg/L) for all tributaries. The latter method and value was chosen to estimate minimum TSS loadings given the bias towards overestimation inherent in using the actual data that included some very high TSS concentrations (and grand average of ~13 mg/L). Concentrations of TSS were multiplied by annual discharge to obtain loadings that were then summed to obtain combined tributary loading of TSS. Combined TSS loading calculated using actual data was 21 x 10<sup>6</sup> kg/yr was more than twice the minimum estimated loading of 9.7 x 10<sup>6</sup> kg/yr. In addition to estimating TSS loading for defined watersheds as just described we also estimated TSS loading from 53 mi<sup>2</sup> of river shoreline between Bangor and Bucksport. The mean annual discharge for this area (2.86 m<sup>3</sup>/s) was estimated using the Dudley (2004) method and then multiplied by 5 mg/L. The resulting total shoreline loading was 4.5 x  $10^5$  kg/yr and increased the grand total TSS loading by only 2 to 5% depending on which tributary estimate is used.

To estimate tributary loadings of Hg we averaged concentrations where more than one measurement of concentration was available, multiplied by discharge and then summed the loadings from all tributaries to obtain combined tributary loading for total and filterpassing Hg. Particulate Hg loading was calculated both from the difference between total (5.5 kg/yr) and filter-passing (3.3 kg/yr) and by multiplying the lower estimate of TSS loading (9.7 x  $10^6$  kg/yr) by mean particulate Hg concentration (0.20  $\mu$ g/g dry wt.). These values (2.2 and 1.9 kg/yr) agree reasonably well.

for tributary streams between Bangor and Bucksport.								
Station ID	Sample Date	Watershed Area (mi²)	Estimated Discharge (m³/s)ª	Total Hg (ng/L)	Filter- passing Hg (ng/L)	TSS (mg/L)	Particulate THg (µg/g)	
BO5	9/16/10	12.3	0.67	0.67	0.92	0.50		
Cove Br	9/16/10	22.6	1.21	0.79	0.75	0.50	0.08	
Cove Br	10/15/10	22.0		6.0	1.6	70	0.06	
Eaton Br	9/16/10	36.6	1.96	2.3	2.5	0.50		
Fells Br	9/16/10	27.6	1.49	1.5	1.2	0.99	0.32	
I-395 Br	9/16/10	-	-	0.82	0.65	0.50	0.33	
Kenduskeag Str	10/06/04		40.0	1.3	1.1	0.67	0.24	
Kenduskeag Str	10/09/04		1 <b>3.8</b> (measured)	1.2	0.99	0.80	0.25	
Kenduskeag Str	8/29/11			7.2	3.7	33	0.11	
Mill Cr	9/16/10	22.9	1.23	6.2	1.3	5.2	0.01	

Table 3. Complete analytical data and estimated annual discharges and combined loadings

Table 3. Complete analytical data and estimated annual discharges and combined loadingsfor tributary streams between Bangor and Bucksport.								
Station ID	Sample Date	Watershed Area (mi²)	Estimated Discharge (m³/s)ª	Total Hg (ng/L)	Filter- passing Hg (ng/L)	TSS (mg/L)	Particulate THg (μg/g)	
Penjajawoc Str	9/16/10	22.8	1.28	0.93	0.75	0.69	0.25	
Penjajawoc Str	10/15/10	23.0		8.1	1.8	121	0.05	
Reed's Br	9/16/10	5.88	0.32	24	1.2	21	1.2	
Sedgeunkedunk Str	10/06/04		2.17	1.3	0.92	4.5	0.08	
Sedgeunkedunk Str	10/09/04	40.5		1.2	0.86	3.2	0.11	
Sedgeunkedunk Str	9/16/10			1.2	1.1	0.50	0.10	
Souadabscook Str	10/06/04	393		1.2	1.2	0.66	0.08	
Souadabscook Str	10/09/04			1.5	1.1	0.80	0.48	
Souadabscook Str	9/16/10		20.6	1.1	0.85	0.63	0.32	
Souadabscook Str	10/15/10				5.5	1.8	64	0.06
Souadabscook Str	8/29/11			4.2	2.9	11	0.12	
North Marsh R	7/28/10		14.2	1.55	1.19	5.36	0.07	
North Marsh R	9/4/10	272		2.49	2.33	1.16	0.13	
North Marsh R	7/7/09		14.5	3.38	2.75	2.46	0.26	
North Marsh R	4/16/09			2.12	1.55	2.38	0.24	
South Marsh R	7/28/10			3.51	2.95	3.60	0.16	
South Marsh R	9/4/10	11 2	2.21	2.92	2.39	5.09	0.10	
South Marsh R	7/7/09	41.2		3.34	2.62	3.80	0.19	
South Marsh R	4/16/09			1.68	1.47	2.76	0.08	
Total Tributary Annual Loading (kg/yr)				5.5	3.3	9.7 x 10 <sup>6</sup>	1.9 <sup>b</sup>	

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<sup>a</sup> Values derived from watershed properties as described in Dudley (2004);<sup>b</sup> kg/yr=TSS<sub>total</sub> x [PTHg]<sub>ave</sub>

## Literature Cited

- Dudley, RW. 2004. Estimating monthly, annual, and low 7-day, 10-year stream flows for ungaged rivers in Maine. USGS Scientific Investigations Report 2004-5026. 22 p.
- Hem, J.D. 1989. *Study and interpretation of the chemical characteristics of natural water.* Third Edition, U.S. Geological Water-Supply Paper 2254.
- Rantz, S.E. 1982. *Measurement and computation of streamflow:Volume 1. Measurement of stage and discharge.* U.S. Geological Survey Water-Supply Paper 2175.