

PENOBSCOT RIVER MERCURY STUDY

Chapter 15

Temporal trends of total and methyl mercury in surface sediments, 2006-2010

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1. Penobscot River Mercury Study

1 SUMMARY

The purpose of this chapter is to present data on total mercury (Hg) concentrations in surface sediments in the contaminated zone of the Penobscot River over the period 2006 – 2010, to determine whether statistically significant declines are evident. Six different classes of sediments were sampled: subtidal (Fort Point Cove), intertidal, wetland – high elevation, wetland – medium elevation, wetland – low elevation, and wetland – mudflats. It was concluded that total Hg concentrations were generally stable. There were no significant changes over time at any of the subtidal sites, or at any of the wetland high elevation, medium elevation or low elevation sites. One out of seven intertidal sites showed a significant increase in total Hg whereas two out of six wetland – mudflat elevation sites showed significant decreases. Methyl Hg concentrations were also generally stable over the period sampled, but more variable than total Hg, with some tendency towards decreases. One site out of five subtidal sites showed a significant decrease in methyl Hg, two sites out of seven intertidal sites showed decreases, one site out of six medium elevation wetland sites showed a decrease, one site out of six low elevation wetland sites showed a decrease and two sites out of six mudflat elevation wetland sites showed a decrease. Also, at high elevation wetland sites, two sites showed increases and one site showed a decrease. Thus, as predicted by sediment core studies and by modeling of the system, declines in total Hg concentrations may be taking place but not at a pace discernible within our five year sample period. Methyl Hg concentrations are subject to short term changes in environmental conditions, such as temperature and supply of organic matter to methylating bacteria, and this was reflected by more variability in methyl Hg concentrations at individual sites, as compared to total Hg. However, methyl Hg concentrations continued to reflect, overall, those of total Hg concentrations.

2 INTRODUCTION

The purpose of the work described in this chapter was to determine whether there have been declines in the concentrations of total mercury (Hg) in surface sediments in the contaminated zone of the Penobscot River over the period 2006 – 2010. At other sites contaminated by chlor-alkali plants, it is commonly observed that Hg in surface sediments decrease during the later years of plant operation and after plants have been closed (see Munthe et al. 2007 for a review). Also, we have observed, in deep sediment cores (Phase I Report, Chapters 6 and 7 of this report) that Hg concentrations tend to decrease towards the surface at many contaminated sites in the Penobscot system. Further, modeling of the Penobscot system (Chapter 15 of this report) predicts steady, if slow, declines in the concentration of total Hg in surface sediments. It is therefore expected that Hg in surface sediments at sites in the lower river and bay would decrease over time and analyses presented here were used to determine whether such decreases were statistically evident from 2006 – 2010. A secondary objective of the chapter was to examine methyl Hg concentrations in the same surface sediments to determine whether they tend to follow total Hg concentrations.

The Penobscot River Mercury Study began in 2006 and 2007 with geographic surveys to define the extent of contamination in the system. Various other studies on Hg in sediments were also conducted in 2008 and 2009; many of the same sites were sampled. In 2010, a dedicated sampling program was put in place to build on the data collected at various sites in previous years. This chapter examines relevant data from 2006 to 2009 and compares them to the results of sampling conducted in 2010. Sampling was also conducted in 2012 but these data are not yet available for inclusion in this analysis.

3 DATA EXAMINED IN THIS ANALYSIS

Dedicated sampling was carried out in August 2010 to define recent concentrations of total Hg and methyl Hg in surface sediments of Fort Point Cove (Sites E01-1, E01-2, E01-3, E01-4 and E01-5), at selected intertidal sites (OV1, OV4 (reference sites), OB1, OB5, ES2, ES4, ES13), and at selected wetland sites (four elevations at each of the following sites: W63, W17, W21, and W25). E01 sites were sampled in 2007 as part of the geographic survey of subtidal sediments (see Phase I report). Intertidal sites were sampled in 2006 and 2007 as part of the original geographic surveys of intertidal sites (Samplings I, II, III, IV, V, and VI – see Phase I Update report). Intertidal sites were sampled in 2008 and 2009 to define seasonal trends in methyl Hg production (see Chapter 5). Wetlands were sampled in 2007 as part of the geographic survey of Hg in contaminated wetlands (see Phase I Update report). They were also sampled in 2008 and 2009 to define seasonal trends in methyl Hg production (See Chapter 5). W63 was not sampled in 2007. All sampling in 2010 followed the same methods as used in previous years (2006, 2007, 2008, and 2009). Surface sediments (0-3 cm) only were sampled. Samples were taken by hand at wetland and intertidal sites and by Van Veen dredge at subtidal sites. All samples for Hg analysis were frozen in the field, on dry ice, within one minutes of being exposed to air. All analysis methods for total Hg and methyl Hg followed standard methods previously described in the Phase I and Phase I Update report. All raw data are shown in Appendices 12d-1, 12d-2, and 12d-3.

Temporal trends were examined statistically, on a site-by-site basis using analysis of variance with an independent model and pooled variance, with year as the independent variable. Because it was not expected that total Hg concentrations would vary seasonally, concentrations derived from samples taken at different times of the year were used directly. Because it was possible that methyl Hg concentrations varied with time of the year (see Chapter 12 of this report), statistical tests were first conducted to determine whether there was any seasonal influence on methyl Hg concentrations. Sampling across years was grouped into 11 collection periods, from May 11-12 to October 20-22 and Analysis of Variance on % methyl Hg by reach was performed with further subgroups for Mendall Marsh and ES South. There were no statistically significant seasonal differences in % methyl Hg in Brewer to Orrington (BO), Orrington to Bucksport (OB), Mendall Marsh or ES North. There were statistically significant seasonal differences in ES South, however % methyl Hg did not differ consistently according to season, so it was concluded that there were no statistically significant seasonal trends. Similarly, sites in the Old Town to Veazie (OV) reach also had statistically significant differences, but no consistent trends. Therefore, sample date was not used a control variable for these analyses. The results of ANOVA for individual sites for total Hg and methyl Hg concentrations at the six classes of sites are shown in Appendix 4. All analyses tested for significant effects among years.

4 RESULTS

4.1 Subtidal Sites (Transect E01)

At all subtidal sites, there were no obvious declines in total Hg in surface sediments (Figure 15-1). Concentrations in 2010 were as high or higher as concentrations seen in 2007, the first year of sampling at all five E01 sites. There was no statistically significant temporal trend in total Hg concentrations at any of these sites. Total Hg concentrations did vary consistently among years at the E01 sites: concentrations increased from 2007 to 2008, decreased to 2009 and then increased again to 2010. This pattern was presumably due to some physical process that affected all of Fort Point Cove, such as rates of sedimentation or the lateral redistribution of surface sediments.

Methyl Hg at most subtidal sites also did not decline significantly over the period 2007 – 2010 (Figure 15-1). At one site (E01-4), there was a significant decline in methyl Hg concentration ($p=0.045$) (Appendix 15-4). Based on visual examination of % methyl Hg concentrations over time, there appeared to be a general trend at all sites toward lower percent methyl Hg concentrations over time (Figure 15-1), resulting in lower percent methyl Hg values in all 2010 subtidal samples as compared to the first year of sampling in 2007.

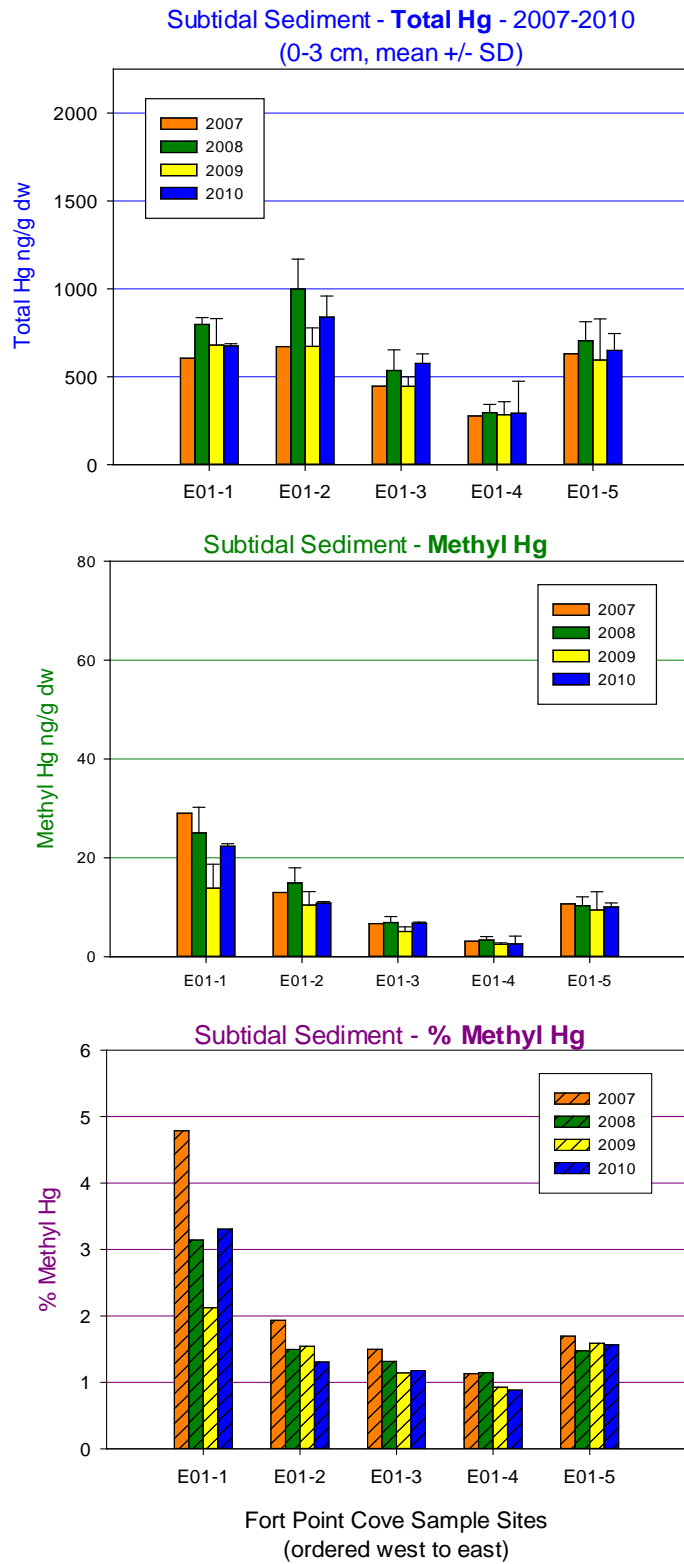


Figure 15-1. Mean concentrations of total Hg (+/- 1 SD), methyl Hg (+/- 1 SD) and % methyl Hg in surface (0-3 cm) sediments at five E01 sites in Fort Point Cove, 2007 to 2010.

4.2 Intertidal Sites

Total Hg concentrations in surface sediments at intertidal sites were quite variable and there was no consistent pattern among the sites. At most sites, there were no significant trend at intertidal sites over the period 2006 – 2010 (Figure 15-2; Appendix 15-4). However, at ES13, at the southern tip of Verona Island, there was a significant increase in total Hg concentrations ($p=0.009$).

Methyl Hg concentrations at most of the intertidal sites also did not change significantly over the period 2006-2010. Significant declines were found at two of the seven sites (Figure 15-2; Appendix 15-4). ES04 ($P = 0.003$) and OV1 ($P = 0.001$; a reference site) but the extremely low concentrations at both sites reduce the importance of these findings.

Percent methyl Hg at intertidal sites varied among years with no consistent trend. Declines in percent methyl Hg were apparent at ES04 and OV1, sites where significant declines in methyl Hg concentrations were found (Figure 15-2). Again, site OV1 was a reference site.

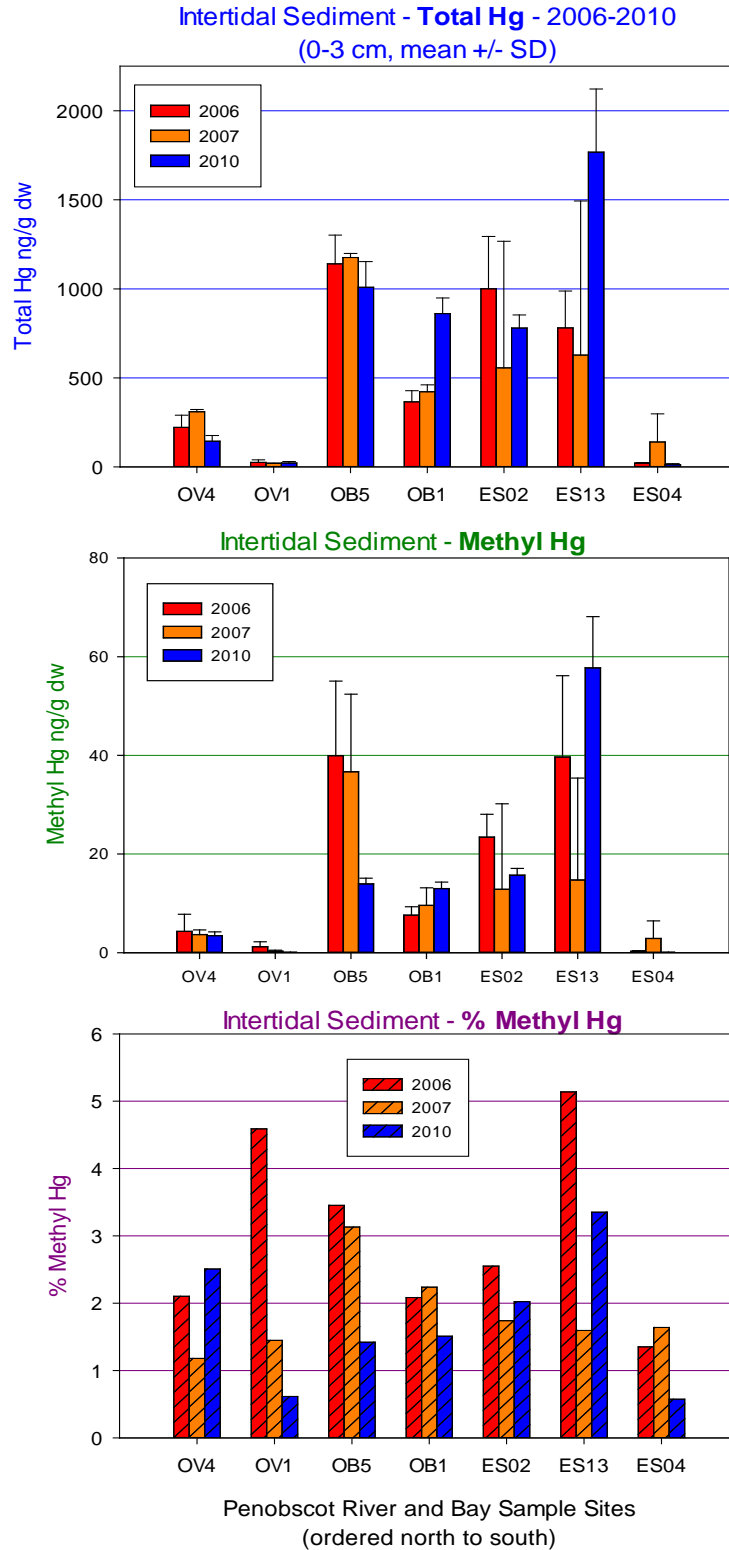


Figure 15-2. Mean concentrations of total Hg (+/- 1 SD), methyl Hg (+/- 1 SD), and % methyl Hg in surface (0-3 cm) sediments at OV1, OV4, OB1, OB5, ES2, ES4, intertidal sites in the Penobscot system, 2006 to 2010.

4.3 Wetland Sites (High Elevations)

At the six high elevation sites sampled, there was no statistically significant trend in total Hg concentrations detected (Figure 15-3, Appendix 15-4). Some sites appeared to show modest increases in total Hg over the sample period, whereas others seemed to show modest decreases, but none of these changes were statistically significant.

Methyl Hg concentrations showed significant, though conflicting, trends at three of the six sites (Figure 3; Appendix 4). There were significant increases in methyl Hg at W25 ($p = 0.001$) and W26 ($P = 0.003$) and a significant decrease at W63 ($p = 0.04$; Appendix 15-4). Percent methyl Hg varied within sites, but there was no consistent pattern among the sites sampled.

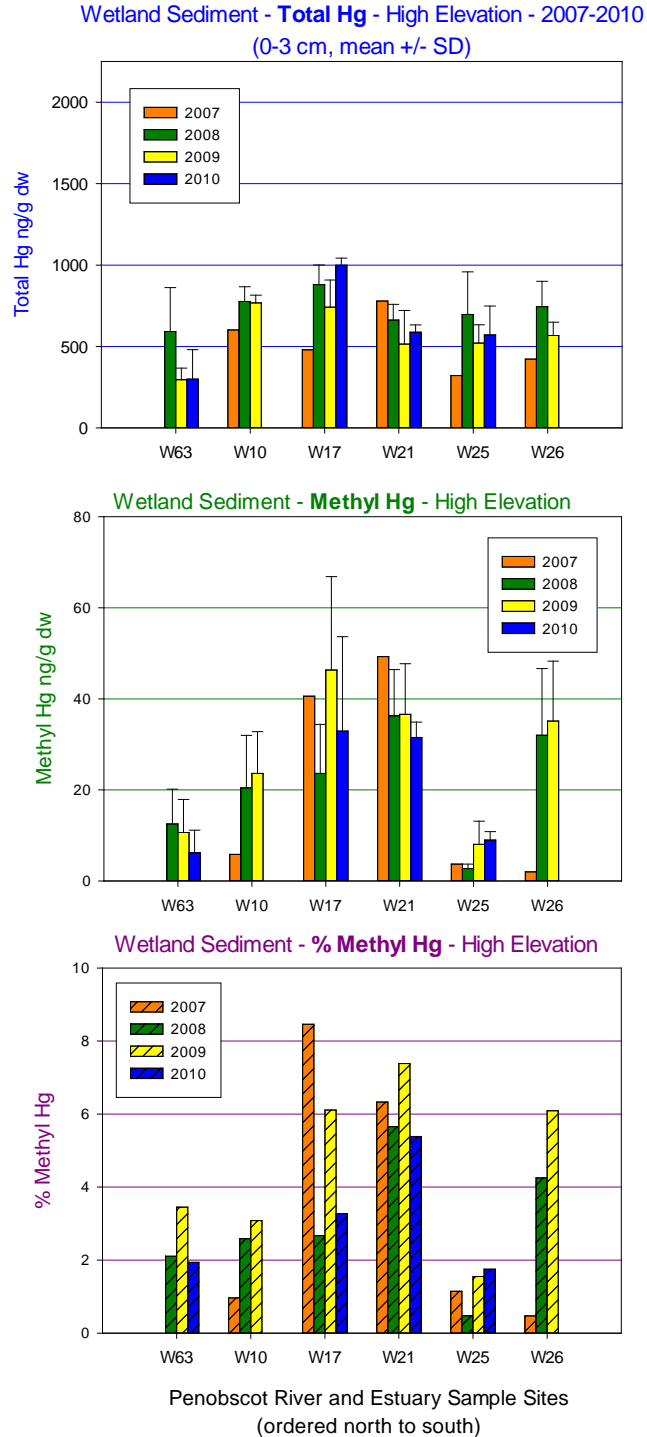
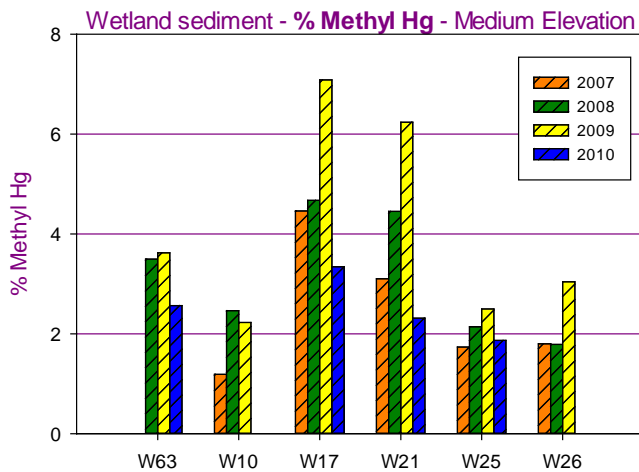
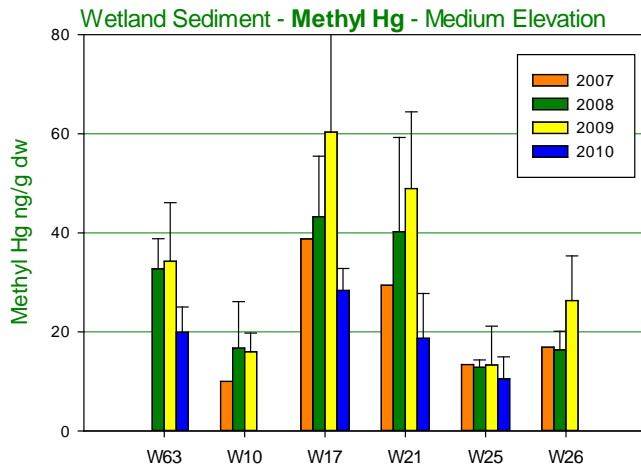
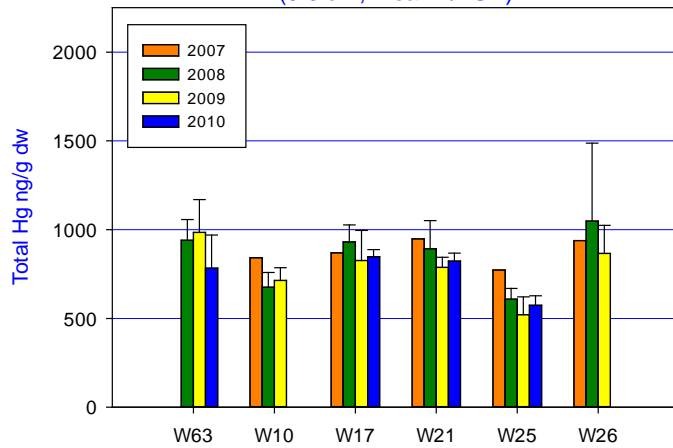


Figure 15-3. Mean concentrations of total Hg (+/- 1 SD), methyl Hg (+/- 1 SD), and % methyl Hg in surface (0-3 cm) sediments at high elevation wetland sites W63, W17, W21, and W25, 2007 or 2008 to 2010. Sites are shown in North to South order. Four elevations were sampled at each site: High, Medium, Low and Mudflat.

4.4 Wetland Sites (Medium Elevations)

Total Hg in the surface sediments at all of the medium elevation wetland sites did not show statistically significant changes over the sampling period (Figure 15-4; Appendix 15-4). Methyl Hg showed a significant decrease at one individual site (W21; $p = 0.038$). The net effect on % methyl Hg was a variable mix of increases and declines (Figure 15-4).

Wetland Sediment - **Total Hg** - Medium Elevation - 2007-2010
(0-3 cm, mean +/- SD)



Penobscot River and Estuary Sample Sites
(ordered north to south)

Figure 15-4. Mean concentrations of total Hg (+/- 1 SD), methyl Hg (+/- 1 SD), and % methyl Hg in surface (0-3 cm) sediments at medium elevation wetland sites W63, W17, W21, and W25, 2007 or 2008 to 2010. Sites are shown in North to South order. Four elevations were sampled at each site: High, Medium, Low and Mudflat.

4.5 Wetland Sites (Low Elevations)

At the low elevation sites, there were no statistically significant changes in total Hg concentrations in surface sediments, as was found for the high and medium elevation sites (Figure 15-5; Appendix 15-4). Methyl Hg concentrations declined significantly at one site (W25; $P = 0.011$), while concentrations at the other five sites had no temporal trend. Percent methyl Hg values showed no consistent temporal trends over the sites sampled (Figure 15-5).

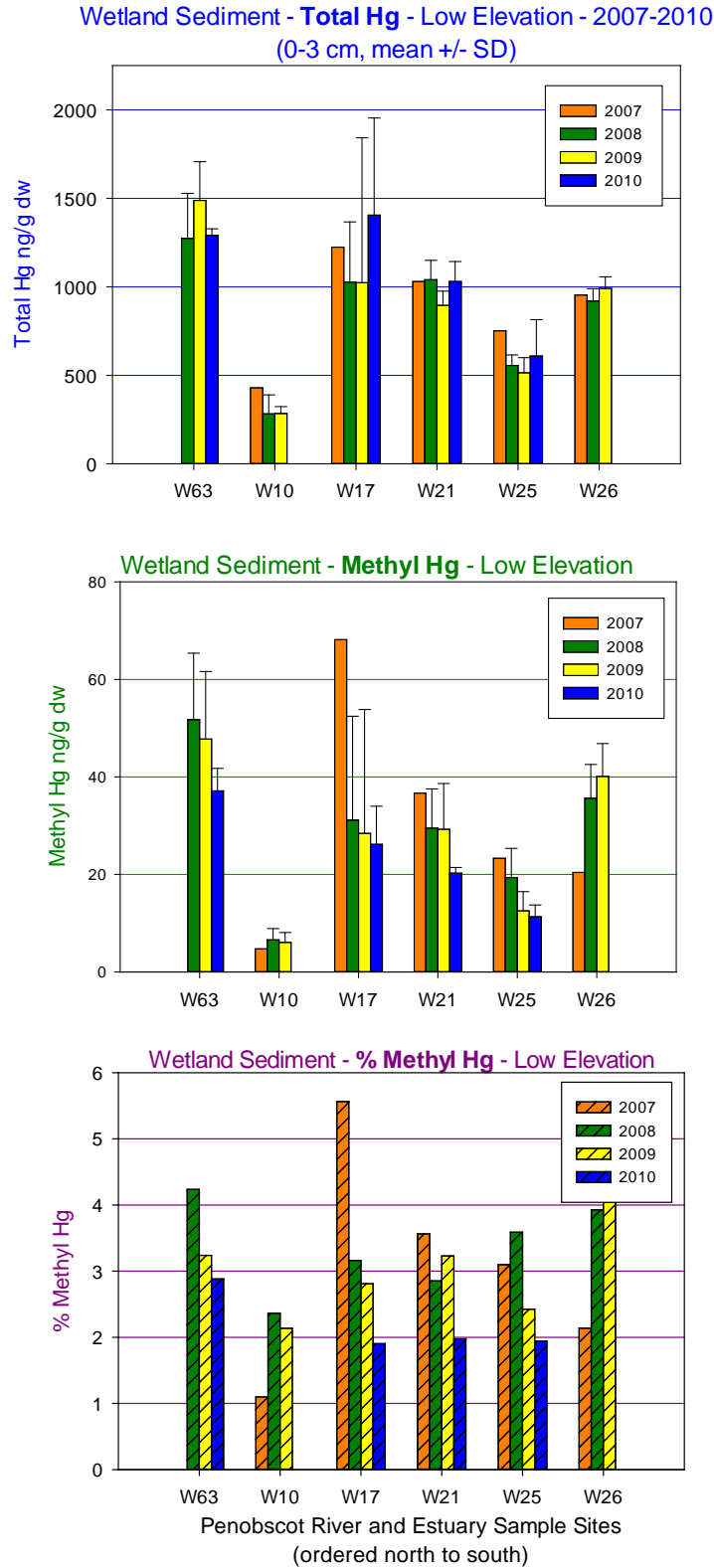


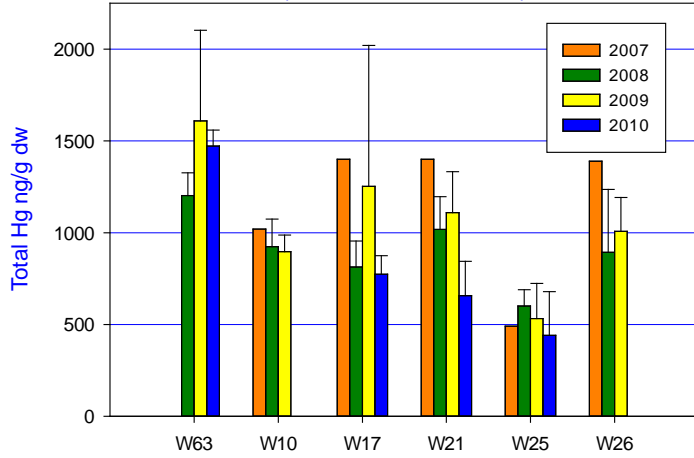
Figure 15-5. Mean concentrations of total Hg (+/- 1 SD), methyl Hg (+/- 1 SD), and % methyl Hg in surface (0-3 cm) sediments at low elevation wetland sites W63, W17, W21, and W25, 2007 or 2008 to 2010. Sites are shown in North to South order. Four elevations were sampled at each site: High, Medium, Low and Mudflat.

4.6 Wetland Sites (Mudflats)

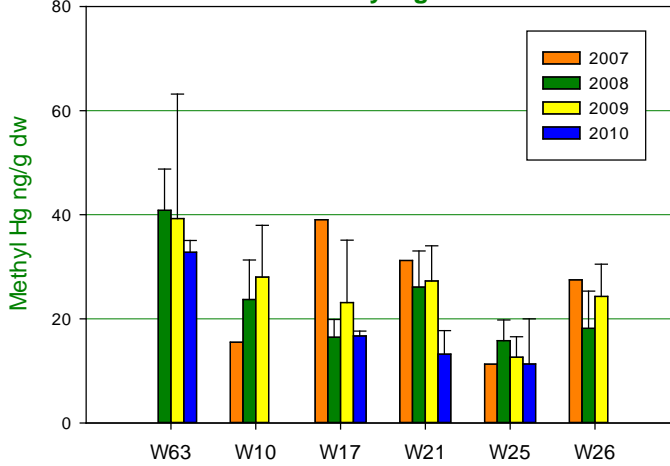
At mudflat elevations, two of the six sites sampled had significant declines in total Hg concentrations (W21, $P = 0.016$ and W25, $P = 0.044$). Methyl Hg also declined significantly at the same two sites W21 ($P = 0.011$) and W25 ($P = 0.036$). Thus, the declines in methyl Hg at these two sites were related to decreases in total Hg concentrations. Percent methyl Hg appeared to be variable at the six different sites sampled, with no noticeable overall trend.

The mudflat sites were similar to the intertidal sites presented above, although mudflat sites are adjacent to wetlands and intertidal sites are not. However, trends in total and methyl Hg concentrations at these two classes of sites were the somewhat different; total Hg at one intertidal site (ES13) showed an increase but decreases were present at two mudflat sites (W21 and W25). Methyl Hg declined at two intertidal sites (OV1 and ES04) and two mudflat sites showed decreases at W21 and W25, although as noted above, decreases in methyl Hg at the two mudflat sites were probably related to decreases in total Hg.

Wetland Sediment - **Total Hg** - Mudflat Elevation - 2007-2010
(0-3 cm, mean +/- SD)



Wetland Sediment - **Methyl Hg** - Mudflat Elevation



Wetland Sediment - **% Methyl Hg** - Mudflat Elevation

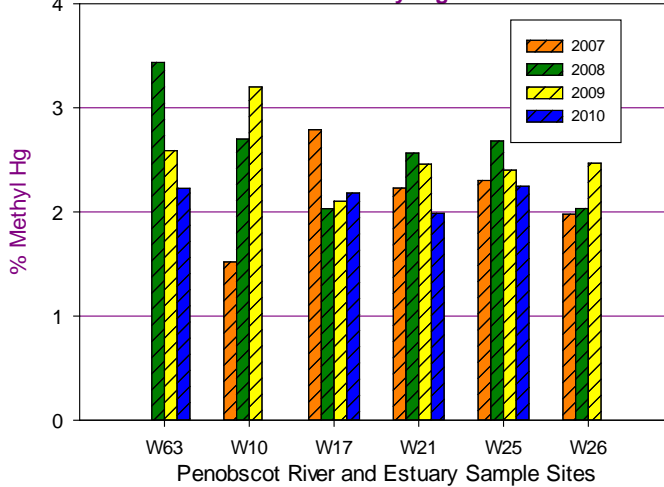


Figure 15-6. Mean concentrations of total Hg (+/- 1 SD), methyl Hg (+/- 1 SD), and % methyl Hg in surface (0-3 cm) sediments at mudflat (intertidal) elevations wetland sites W63, W17, W21, and W25, 2007 or 2008 to 2010. Four elevations were sampled at each site: High, Medium, Low and Mudflat.

5 DISCUSSION

Our primary hypothesis was that there would be declines in total Hg concentrations in surface sediments of the contaminated zone (Brewer to Fort Point) of the Penobscot River over the period 2006 – 2010. We found little support for this hypothesis in that there were no significant decreases in total Hg at the majority of sites sampled in any of the six classes of sediments that were examined. Changes at individual sites showed little in the way of consistent trends in total Hg as well: only one individual site (ES13 Intertidal) showed a significant increase in total Hg and only two individual sites (W21 and W25 Wetland Mudflat) showed a significant decrease in total Hg. There was a fairly large amount of inter-year variation at most sites and this variability is probably due to small-scale spatial variability (Krabbenhoft et al. 2007). The monitoring plan for the Penobscot system has taken this variability into account (see Chapter 13 of this report). Power analysis has indicated that assuming a half-time for recovery of 20 years, sampling every second year, and a monitoring period of 12 years, a minimum power of detection of 0.8 at most sites would result from collecting n=3 replicates at subtidal sites, n=5 replicates at intertidal sites, and n=4 replicates at wetland sites. We may see more statistically significant declines after the analysis of samples taken in 2012.

Similar to the finding that total Hg was stable at most sites, methyl Hg also did not show significant changes, on an overall basis, in the six classes of sediments sampled. Certainly, methyl Hg was more variable than total Hg with some tendency for decreasing concentrations: eight individual sites showed significant decreases in methyl Hg whereas two individual sites showed significant increases in methyl Hg. Two of the declines in methyl Hg would appear to be related to decreases in total Hg as it has been shown that, within sediment types, methyl Hg is closely related to total Hg in the Penobscot system (see Chapter 12 of this report). This greater variability in methyl Hg concentrations as compared to total Hg concentrations is to be expected (Krabbenhoft et al. 2007). Even though the supply of inorganic Hg on particles to the surface layers of sediments may not have diminished significantly over the period 2006 to 2010 at most sites (keeping total Hg relatively stable), the conditions for Hg methylation or the balance between Hg methylation and methyl Hg demethylation are more likely to have undergone changes, especially on a site to site basis. Because so many factors can affect methylation and demethylation of Hg, such as temperature, supply of organic C, the supply of sulfate which can be influenced by river flows, dissolved organic matter, and others, it is not surprising that methyl Hg concentrations were more variable than total Hg. However, it is difficult to monitor for all of the parameters that affect methyl Hg concentrations (see Chapter 12 of this report) and therefore it is difficult to ascribe differences in methyl Hg concentrations to any particular factor.

It would be unrealistic to expect a lock-step relationship between methyl Hg in surface sediments, the source of Hg for many species of biota, and Hg in biota because of the complicated system that transfers methyl Hg from sediments and porewater to the bottom of the food chain. There are many factors that can affect Hg in biota, including transfer efficiencies, growth rates, feeding patterns and excretion rates (Sandheinrich and Wiener 2011). However, there would appear to be a general correspondence

between temporal trends in methyl Hg concentrations in surface sediments and temporal trends in Hg in biota. This should be especially true for animals tied to sediment-based food webs (e.g. tomcod, flounder and eels – see Chapter 16 of this report). Methyl Hg in surface sediments did not, in general, show trends over time as was also generally the case for Hg in eels, tomcod, and flounder (see Chapter 14 of this report). Also, birds that inhabit wetland areas have also shown little in the way of temporal trends (see Chapter 14 of this report), mirroring the general lack of trend of methyl Hg in the surface sediments of wetlands, as shown here.

Demonstrating that Hg in biota is often related to Hg in surface sediments, there were a number of significant correlations between the two (Table 15-1). Significant relationships were found for six of the nine biota species tested (mussels, sand worms, periwinkles, eels, smelt and flounder). The significant relationships for mussels and smelt were not expected because they feed on pelagically-based food chains. There were no significant relationships for soft-shelled clams, tomcod and mummichogs. The latter two are surprising because of indications that they receive their Hg from sediment-based food webs (see Chapter 16 of this report). Figures 12d-7 and 12d-8 provide two examples of these correlations. These observations do not prove cause-effect relationships, perhaps indicating general correlations between levels of Hg contamination in the Penobscot system and Hg in biota. However, they do strengthen the hypothesized link between Hg in surface sediments and Hg in biota (See Chapter 16 of this report). They also support the view that if total and methyl Hg in sediments can be lowered by remediation actions, then Hg in biota will follow.

Table 15-1: Details of correlations between Hg (methyl or total) in sediment and Hg (methyl or total) in various species of biota in the Penobscot system. Hg data for sediments and biota was from sampling in 2006-7 in Phase I of the Study. The strongest correlation, depending on total Hg or methyl Hg in sediment or biota is given. Asterisks indicate $p < 0.05$. r^2 values given only when p values were less than 0.05.

SPECIES	ln X (sediment)	ln Y (biota)	p value	r^2
SHELLFISH				
Blue mussel	Methyl Hg	Total Hg	0.005*	0.65
Soft-shelled clam	Methyl Hg	Total Hg	0.133	-
INVERTEBRATES				
Sand worm	Methyl Hg	Total Hg	0.000*	0.69
Periwinkles	Methyl Hg	Total Hg	0.000*	0.79
FISHES				
American eel	Total Hg	Total Hg	0.013*	0.74

Table 15-1: Details of correlations between Hg (methyl or total) in sediment and Hg (methyl or total) in various species of biota in the Penobscot system. Hg data for sediments and biota was from sampling in 2006-7 in Phase I of the Study. The strongest correlation, depending on total Hg or methyl Hg in sediment or biota is given. Asterisks indicate $p < 0.05$. r^2 values given only when p values were less than 0.05.

SPECIES	ln X (sediment)	ln Y (biota)	p value	r^2
Rainbow smelt	Methyl Hg	Total Hg	0.035*	0.54
Winter flounder	Methyl Hg	Total Hg	0.021*	0.87
Tomcod	Methyl Hg	Total Hg	0.117	-
Mummichog	Total Hg	Total Hg	0.279	-

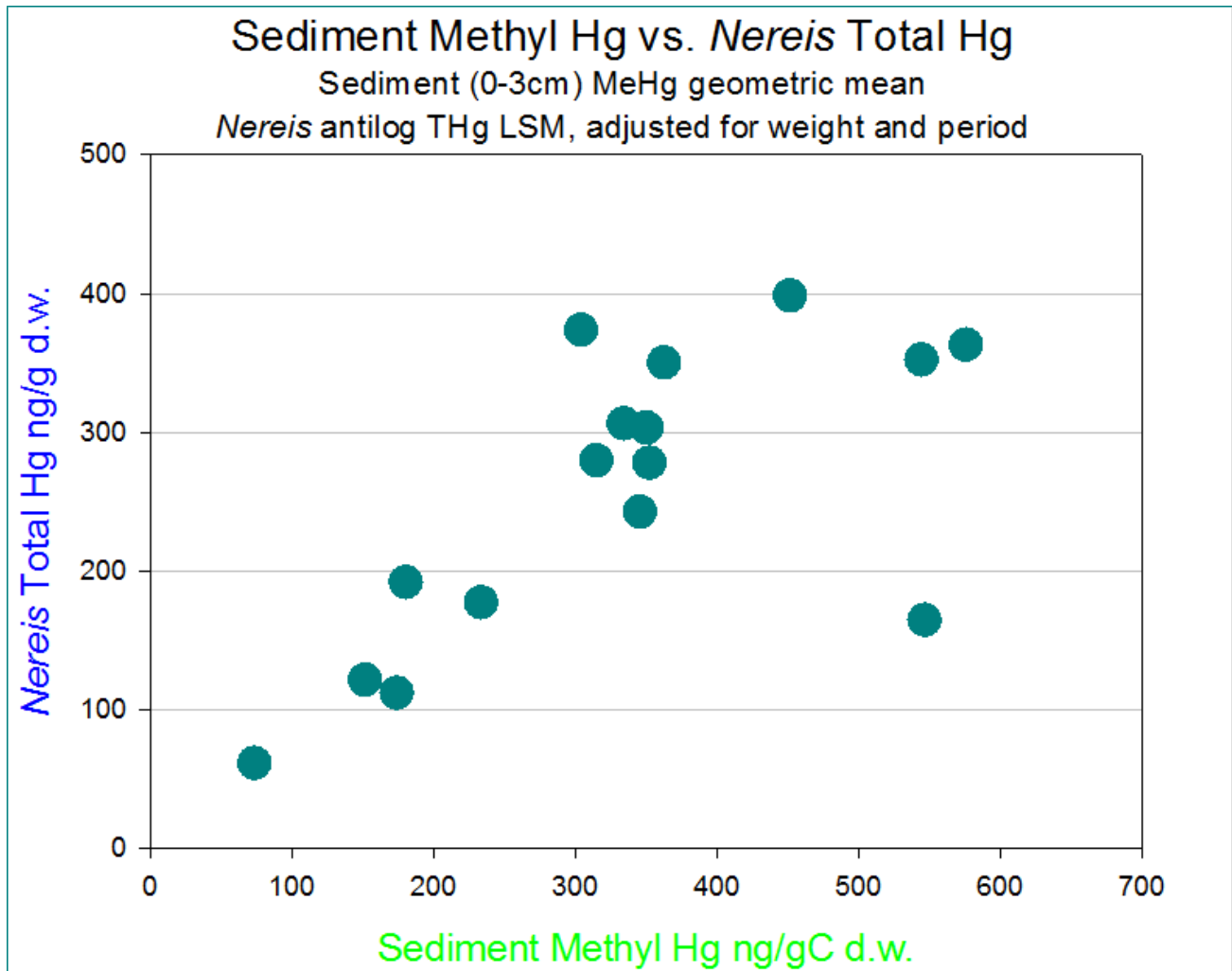


Figure 15-7. Relationship between total Hg in *Nereis* and methyl Hg in surficial (0-3 cm) sediments, based on data from 2006-7 (Phase I of the Study). See Table 1 for statistical details.

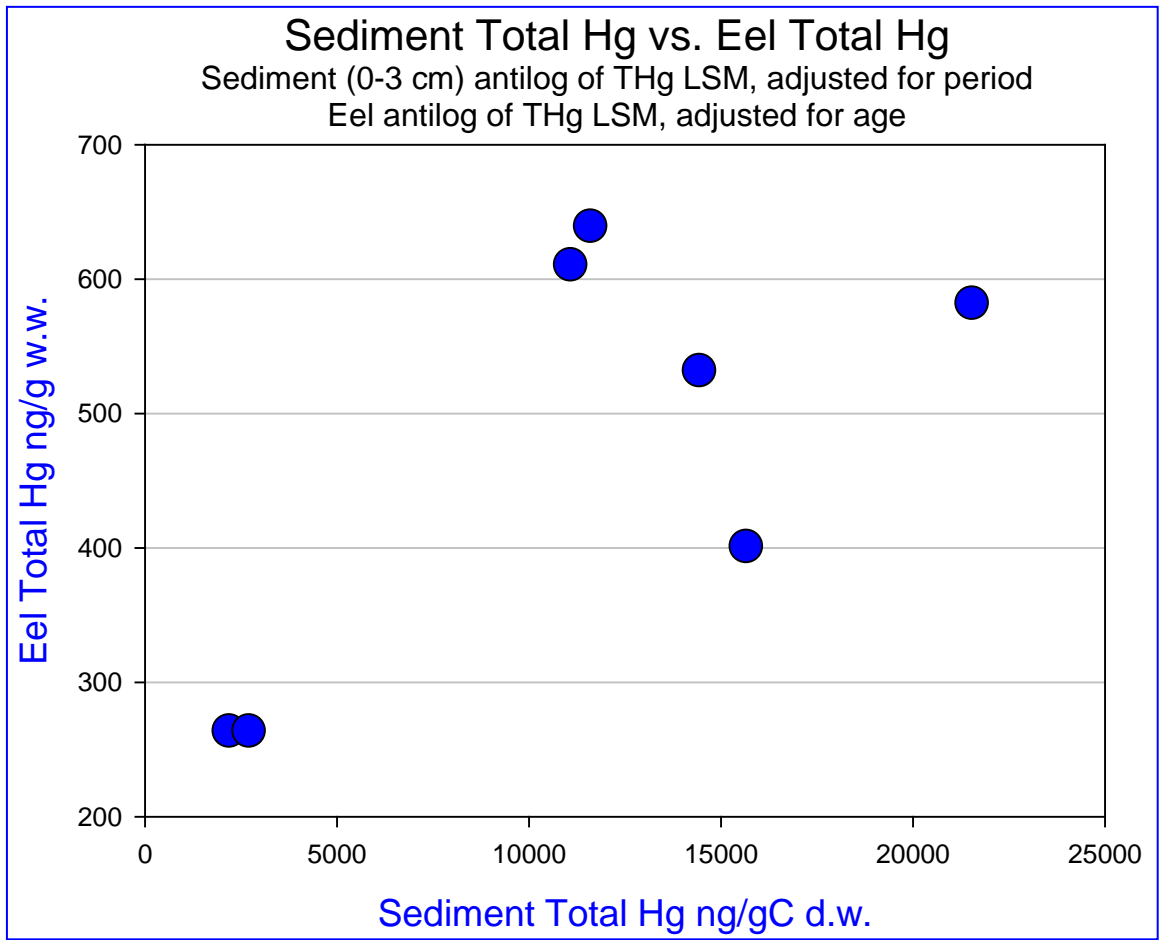


Figure 15-8. Relationship between total Hg in American eels and total Hg in surficial (0-3 cm) sediments, based on data from 2006-7 (Phase I of the Study). See Table 15-1 for statistical details.

6 ACKNOWLEDGEMENTS

Staff at Normandeau Associates, Inc. carried out most of the sediment sampling over the five years of study. In particular, we thank M. Bowen, C. Francis, S. Lee, K. Payne, R. Simmons, and E. Sobo. M. Bowen of Normandeau Associates is thanked for her efficient management oversight of field sampling. Statistical advice was provided by J. Siegrist of Applied BioMathematics. Most laboratory analyses were carried out at Flett Research Ltd, with particular thanks to R. Flett and D. Gilbert. J. Wiener (University of Wisconsin – La Crosse) provided insightful comments that strengthened the chapter.

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APPENDIX 15-1:

Raw data for total Hg concentrations in surface (0-3 cm) sediments in Fort Point Cove (the E01 transect). All concentrations are given as ng/g d.w. Data from 2007 are from the geographic survey of Hg in estuary sediments (see Phase I Report). Data from 2008 and 2009 are from seasonal survey of Hg in estuary sediments (see Chapter 12 of this report). Data from 2010 are from sampling done to define temporal trends in Hg in sediments (see methods section in this chapter). Multiple observations for the same date are from replicate samples.

E01-1	THg	MeHg
8/17/2007	606	29
7/23/2008	864	30.5
8/6/2008	810	26.9
8/20/2008	760	29.2
9/3/2008	830	19.5
9/18/2008	788	25.75
9/30/2008	760	27.1
10/20/2008	777	16.4
5/12/2009	672	9.52
6/3/2009	931	8.74
6/25/2009	654	17.7
7/15/2009	458	10.7
8/5/2009	671	20.4
9/4/2009	699	16.1
8/23/2010	690	22.9
8/23/2010	672	21.9
8/23/2010	665	22.2

E01-2	THg	MeHg
8/17/2007	672	13
7/23/2008	1030	13.3
8/6/2008	980	14
8/20/2008	1260	20.5
9/3/2008	1160	15.7
9/18/2008	927	16.1
9/30/2008	885	14.3
10/20/2008	754	10.6
6/3/2009	773	14.7
6/25/2009	734.5	10.4
7/15/2009	479	6.89
8/5/2009	639	11.3
9/4/2009	728	11.2
8/23/2010	960	10.7
8/23/2010	840	11.2
8/23/2010	720	10.6

E01-3	THg	MeHg
8/17/2007	447	6.69
7/23/2008	369	5.79
8/6/2008	684.5	6.565
8/20/2008	542.5	8.61
9/3/2008	564	7.07
9/18/2008	674.5	8.52
9/30/2008	456	5.88
10/20/2008	462	5.92
5/12/2009	433	5.59
6/3/2009	476.5	4.78
6/25/2009	500	5.25
7/15/2009	360	3.77
8/5/2009	493	6.49
9/4/2009	414	4.74
8/23/2010	535	6.58
8/23/2010	556	6.63
8/23/2010	638	7.03

E01-4	THg	MeHg
8/17/2007	278	3.14
7/23/2008	268	3.07
8/6/2008	369	4.14
8/20/2008	253	3.81
9/3/2008	324	3.66
9/18/2008	334	2.9
9/30/2008	279.5	3.83
10/20/2008	244	2.17
5/12/2009	225	2.43
6/3/2009	425	2.3
6/25/2009	286	2.64
7/15/2009	252	3.05
8/5/2009	231	2.32
9/4/2009	289	2.31
8/23/2010	143	1.285
8/23/2010	495	4.33
8/23/2010	241	2.095

E01-5	THg	MeHg
8/17/2007	631	10.7
7/23/2008	720	10.3
8/6/2008	655	9.72
8/20/2008	606	11.7
9/3/2008	604	7.7
9/18/2008	744	8.88
9/30/2008	689	10.85
10/20/2008	918	13.1
5/12/2009	273	3.71
6/3/2009	973	13.3
6/25/2009	643	12.1
7/15/2009	544	11.6
8/5/2009	671	9.39
9/4/2009	472	6.52
8/23/2010	695	11
8/23/2010	716	9.63
8/23/2010	540	9.515

APPENDIX 5-2:

Raw data for total Hg concentrations in surface (0-3 cm) sediments at intertidal sites. All concentrations are given as ng/g dry.wt. Data from 2006 are from the geographic survey of Hg in intertidal sediments (Sampling I, II, III and IV) (see Phase I Report). Data from 2007 are from the geographic survey of Hg in intertidal sediments (Sampling V and VI) (see Phase I Report). Data from 2010 are from sampling done to define temporal trends in Hg in sediments (see methods section in this chapter). Multiple observations from the same date are from replicate samples.

ES02	THg	MeHg
5/29/2007	993.0	27.8
7/10/2007	1059.0	25.1
8/1/2006	886.2	27.9
9/6/2006	1059.5	25.4
9/27/2006	680.8	23.3
10/22/2006	1374.9	17.1
8/26/2010	785.0	13.7
8/26/2010	717.0	16.7
8/26/2010	695.0	15.3
8/26/2010	846.0	15.6
8/26/2010	858.0	17.2

ES04	THg	MeHg
8/1/2006	24.8	0.2
9/6/2006	17.5	0.5
9/27/2006	22.3	0.2
10/22/2006	20.7	0.2
5/29/2007	251.6	5.4
7/10/2007	29.3	0.3
8/23/2010	11.6	0.1
8/23/2010	12.7	0.0
8/23/2010	9.2	0.0
8/23/2010	8.3	0.0
8/23/2010	23.0	0.2

ES13	THg	MeHg
8/1/2006	702.7	36.3
9/6/2006	693.2	56.5
9/27/2006	640.1	18.3
10/22/2006	1087.9	47.6
5/29/2007	17.4	0.1
7/10/2007	1239.6	29.3
8/26/2010	2390.0	55.2
8/26/2010	1640.0	46.6
8/26/2010	1550.0	51.2
8/26/2010	1710.0	73.1
8/26/2010	1550.0	62.6

OB1	THg	MeHg
8/1/2006	417.1	9.9
9/6/2006	347.8	5.9
9/27/2006	283.8	6.8
10/22/2006	412.6	7.8
5/29/2007	449.9	12.1
7/10/2007	394.4	7.0
8/24/2010	883.0	12.5
8/24/2010	797.0	12.8
8/24/2010	922.0	13.3
8/24/2010	958.0	15.0
8/24/2010	745.0	11.4

OB5	THg	MeHg
8/1/2006	1091.7	41.2
9/6/2006	1161.9	27.1
9/27/2006	961.0	30.7
10/22/2006	1347.7	60.8
5/29/2007	1159.6	47.8
7/10/2007	1191.7	25.5
8/25/2010	1050.0	13.2
8/25/2010	978.0	13.8
8/25/2010	784.5	15.9
8/25/2010	1050.0	13.0
8/25/2010	1180.0	13.9

OV1	THg	MeHg
8/1/2006	11.5	0.8
9/6/2006	44.4	2.7
9/27/2006	27.7	0.8
10/22/2006	20.9	0.5
5/29/2007	19.6	0.2
7/10/2007	21.4	0.4
8/24/2010	7.6	0.1
8/24/2010	24.0	0.1
8/24/2010	30.7	0.1
8/24/2010	25.0	0.1
8/24/2010	21.7	0.1

OV4	THg	MeHg
8/1/2006	168.5	1.4
9/6/2006	166.9	8.5
9/27/2006	309.4	5.9
10/22/2006	246.5	1.5
5/29/2007	302.2	3.0
7/10/2007	319.0	4.3
8/26/2010	103.0	2.9
8/26/2010	179.0	2.7
8/26/2010	173.0	3.3
8/26/2010	141.0	3.8
8/26/2010	127.0	4.6

APPENDIX 15-3:

Raw data for temporal analysis of total mercury concentrations in wetland sediments. All values are for surface sediments (0-3 cm) in ng/g dry wt. Data from 2007 are from the geographic survey of Hg in wetland sediments (see Phase I Report). Data from 2008 and 2009 are from sampling to define seasonal trends in methyl Hg production (see Chapter 12 of this report). Data from 2010 are from sampling done to define temporal trends in Hg in sediments (see methods section in this chapter). Multiple observations from the same date are from replicate samples.

W63 High	THg	MeHg
7/22/2008	727.00	12.40
8/4/2008	1,030.00	25.20
8/20/2008	371.00	6.45
9/3/2008	603.00	20.85
9/16/2008	420.00	9.15
9/30/2008	752.00	8.45
10/21/2008	242.00	5.07
5/12/2009	320.00	4.20
6/2/2009	319.00	19.20
6/24/2009	214.00	3.57
7/16/2009	299.00	7.10
8/4/2009	221.00	9.73
9/1/2009	405.00	19.90
8/25/2010	297.00	6.50
8/25/2010	124.00	2.35
8/25/2010	231.00	2.97
8/25/2010	549.00	13.05

W63 Medium	THg	MeHg
7/22/2008	948	27.8
8/4/2008	1030	35.45
8/20/2008	832	32.6
9/3/2008	1100	44.7
9/16/2008	828	32.9
9/30/2008	1030	27.3
10/21/2008	817	28.5
5/12/2009	1030	37.5
6/2/2009	944	14.6
6/24/2009	644	37
7/16/2009	1020	51.3
8/4/2009	1090	32.9
9/1/2009	1180	32.35
8/25/2010	978	23.2
8/25/2010	902.5	21.6
8/25/2010	591	12.3
8/25/2010	660	22.5

W63 Low	THg	MeHg
7/22/2008	1130	62.15
8/4/2008	1610	52.5
8/20/2008	1100	70
9/3/2008	1400	52
9/16/2008	962	54
9/30/2008	1580	44.7
10/21/2008	1135	26.9
5/12/2009	1550	46
6/2/2009	1200	30.5
6/24/2009	1320	70.4
7/16/2009	1780	53.7
8/4/2009	1410	37.3
9/1/2009	1670	48.8
8/25/2010	1260	43.2
8/25/2010	1315	38
8/25/2010	1330	32.5
8/25/2010	1260	34.9

W63 Mudflat	THg	MeHg
7/22/2008	1420	46.3
8/4/2008	1090	41.1
8/20/2008	1100	43.75
9/3/2008	1280	36.7
9/16/2008	1090	53.4
9/30/2008	1250	35.4
10/21/2008	1190	29.1
5/12/2009	935	28.65
6/2/2009	1160	28.65
6/24/2009	1720	25.5
7/16/2009	1650	86.2
8/4/2009	1900	42.6
9/1/2009	2290	23.9
8/25/2010	1450	33.2
8/25/2010	1360	29.6
8/25/2010	1550	33.5
8/25/2010	1530	34.85

W10 High	THg	MeHg
8/17/2007	602	5.82
5/12/2008	823	16
7/22/2008	763	2.87
8/4/2008	645	21.25
8/20/2008	871	37.3
9/3/2008	786	16
9/16/2008	892.5	26.4
9/30/2008	797	28.2
10/21/2008	685	11.1
6/2/2009	819	22.2
6/24/2009	698	14.5
7/16/2009	749	26.1
8/4/2009	762	40.1
9/1/2009	759	22.8

W10 Medium	THg	MeHg
8/17/2007	842	10
5/12/2008	721	18.8
7/22/2008	737	15
8/4/2008	587	10.2
8/20/2008	724	37.3
9/3/2008	805	12.4
9/16/2008	612	17
9/30/2008	676	10.8
10/21/2008	594	14.2
6/2/2009	689	9.99
6/24/2009	605	13.9
7/16/2009	830	20.8
8/4/2009	722	16
9/1/2009	715.5	16.3

W10 Low	THg	MeHg
8/17/2007	430	4.72
5/12/2008	293	8.05
7/22/2008	353.5	9.76
8/4/2008	476	8.48
8/20/2008	237	6.42
9/3/2008	217	7.48
9/16/2008	148	2.81
9/30/2008	255	4.62
10/21/2008	296	6.37
6/2/2009	254	7.91
6/24/2009	240	4.19
7/16/2009	263	3.44
8/4/2009	317	7.56
9/1/2009	341	5.17

W10 Mudflat	THg	MeHg
8/17/2007	1020	15.5
5/12/2008	871	33.6
7/22/2008	980	22.5
8/4/2008	768	23.5
8/20/2008	819	35.6
9/3/2008	831	29.5
9/16/2008	1030	20.4
9/30/2008	1190	11.2
10/21/2008	855	23.3
6/2/2009	824	27.6
6/24/2009	1060	20.1
7/16/2009	822	45.2
8/4/2009	938	20.9
9/1/2009	869	20.6

W17 High	THg	MeHg
8/18/2007	480	40.6
7/22/2008	928	29.7
8/4/2008	720	18.6
8/20/2008	818	33.5
9/3/2008	1100	37.9
9/18/2008	838	10.9
9/30/2008	933	10.4
10/21/2008	824	24.2
5/12/2009	834	43.5
6/2/2009	802	37.6
6/24/2009	791	59.55
7/15/2009	865	79.5
8/4/2009	747	37.4
9/2/2009	411	20.6
8/24/2010	974	19.2
8/24/2010	1020	63.8
8/24/2010	1050	25.3
8/24/2010	953	23.4

W17 Medium	THg	MeHg
8/18/2007	870	38.8
7/22/2008	939	46.7
8/4/2008	937	43.7
8/20/2008	929	68.7
9/3/2008	884	34.2
9/18/2008	909	39.6
9/30/2008	1120	36.5
10/21/2008	802	33.5
5/12/2009	1055	95.5
6/2/2009	908	91.5
6/24/2009	840.5	54.8
7/15/2009	803	56.6
8/4/2009	816	31.9
9/2/2009	537	31.7
8/24/2010	819	27.5
8/24/2010	826	31.4
8/24/2010	843	22.4
8/24/2010	906	32.15

W17 Low	THg	MeHg
8/18/2007	1225	68.15
7/22/2008	413	6.81
8/4/2008	1540	16.8
8/20/2008	996	48.3
9/3/2008	908	69.9
9/18/2008	1220	29.1
9/30/2008	1050	26.5
10/21/2008	1060	20.4
5/12/2009	305	12.9
6/2/2009	785	12.4
6/24/2009	1410	40.7
7/15/2009	565	17.6
8/4/2009	566	11.6
9/2/2009	2510	75.2
8/24/2010	921	18.7
8/24/2010	2190	37.1
8/24/2010	1320	25.4
8/24/2010	1190	23.4

W17 Mudflat	THg	MeHg
8/18/2007	1400	39.05
7/22/2008	507	10.8
8/4/2008	856.5	17.9
8/20/2008	906	20.4
9/3/2008	921.5	18.8
9/18/2008	790	18.2
9/30/2008	853	16
10/21/2008	860.5	13.2
5/12/2009	658	17
6/2/2009	708	27.4
6/24/2009	1290	9.44
7/15/2009	752	13.9
8/4/2009	1440	28.8
9/2/2009	2670	42.1
8/24/2010	739	16
8/24/2010	894	18.05
8/24/2010	656	16.6
8/24/2010	806.5	16.2

W21 High	THg	MeHg
8/22/2007	779	49.3
7/23/2008	837.5	28.6
8/5/2008	595	47.2
8/20/2008	755	36.15
9/3/2008	654	21.1
9/18/2008	608	30
9/30/2008	600	46.4
10/21/2008	589	44.5
5/12/2009	908	46.5
6/3/2009	396	22
6/25/2009	432	40.7
7/15/2009	569.5	49.4
8/4/2009	341	25.4
9/2/2009	441	35.6
8/26/2010	652	29.3
8/26/2010	564	36.3
8/26/2010	553	28.7
8/26/2010	583	31.6

W21 Medium	THg	MeHg
8/22/2007	948	29.4
7/23/2008	1110	37.4
8/5/2008	666	17.6
8/20/2008	1000	73.3
9/3/2008	885	57.1
9/18/2008	786	38.8
9/30/2008	785	27
10/21/2008	1020	30.5
5/12/2009	775	49.2
6/3/2009	859	25.35
6/25/2009	839	65
7/15/2009	796	64.4
8/4/2009	752	52.4
9/2/2009	703.5	37.3
8/26/2010	793	21.6
8/26/2010	826	11.725
8/26/2010	787	30.2
8/26/2010	885	11.25

W21 Low	THg	MeHg
8/22/2007	1030	36.7
7/23/2008	1040	29.1
8/5/2008	944	28.4
8/20/2008	1030	39.4
9/3/2008	1240	37.8
9/18/2008	903	32.2
9/30/2008	1100	16.25
10/21/2008	1030	23.15
5/12/2009	892	30.2
6/3/2009	840.5	15.9
6/25/2009	893	38
7/15/2009	1050	41.5
8/4/2009	872	24.9
9/2/2009	823	24.95
8/26/2010	942	18.7
8/26/2010	1190	21.5
8/26/2010	1030	20.6
8/26/2010	958	20.2

W21 Mudflat	THg	MeHg
8/22/2007	1400	31.2
7/23/2008	959	29.5
8/5/2008	962	21.35
8/20/2008	1100	38.1
9/3/2008	1340	29.7
9/18/2008	890	24.7
9/30/2008	1090	22.2
10/21/2008	789	17.1
5/12/2009	1120	33.2
6/3/2009	866	19.2
6/25/2009	1220	30.3
7/15/2009	1450	35.8
8/4/2009	863	23.5
9/2/2009	1140	21.6
8/26/2010	849	18.7
8/26/2010	402	7.71
8/26/2010	691	14
8/26/2010	689	12.4

W25 High	THg	MeHg
8/19/2007	322	3.69
7/23/2008	1050	1.34
8/5/2008	567	2.02
8/18/2008	819	2.66
9/4/2008	546	3.02
9/17/2008	1010	2.12
9/29/2008	487	4.26
10/22/2008	399	3.55
5/11/2009	507	1.86
6/3/2009	606	4.75
6/25/2009	507	15.1
7/16/2009	678	12.75
8/5/2009	481	8.38
8/31/2009	350	5.37
8/24/2010	682	9.43
8/24/2010	403	10
8/24/2010	761	6.18
8/24/2010	440	10.2

W25 Medium	THg	MeHg
8/19/2007	773	13.4
7/23/2008	711	11.9
8/5/2008	569	14
8/18/2008	682	10.4
9/4/2008	576	14.1
9/17/2008	578.5	11.9
9/29/2008	573	14.4
10/22/2008	572	13.2
5/11/2009	529	12.6
6/3/2009	572	7.29
6/25/2009	481	7.94
7/16/2009	642	18
8/5/2009	558	26.8
8/31/2009	349	7.17
8/24/2010	622	6.07
8/24/2010	499	11.15
8/24/2010	598	8.48
8/24/2010	579	16.4

W25 Low	THg	MeHg
8/19/2007	752	23.3
7/23/2008	646	20
8/5/2008	542	24.4
8/18/2008	446	29.4
9/4/2008	565	15
9/17/2008	559	13.5
9/29/2008	558	19.6
10/22/2008	577	13.5
5/11/2009	527	16.4
6/3/2009	621	12.5
6/25/2009	511	11.2
7/16/2009	480	7.91
8/5/2009	576	17.9
8/31/2009	373	9.22
8/24/2010	528	8.47
8/24/2010	895	13.2
8/24/2010	602	13.4
8/24/2010	415	10.2

W25 Mudflat	THg	MeHg
8/19/2007	491	11.3
7/23/2008	489	14.4
8/5/2008	528	18.4
8/18/2008	598	16.7
9/4/2008	770	17.4
9/17/2008	586	17.9
9/29/2008	604	18.5
10/22/2008	630	7.38
5/11/2009	518	12.4
6/3/2009	398	10.4
6/25/2009	305	7.4
7/16/2009	506	11.3
8/5/2009	859	18.3
8/31/2009	609.75	15.9
8/24/2010	133	1.78
8/24/2010	380	7.26
8/24/2010	582.75	14.4
8/24/2010	668	21.8

W26 High	THg	MeHg
8/23/2007	423	1.995
7/23/2008	903	25.2
8/5/2008	693	35.5
8/18/2008	907	62.8
9/4/2008	575	26.8
9/17/2008	559	17.8
9/30/2008	682	24.5
10/22/2008	897	31.3
5/11/2009	640	53.1
6/3/2009	439	24.2
6/25/2009	645	34.9
7/16/2009	611	37.2
8/5/2009	504	16.9
8/31/2009	565	44.5

W26 Medium	THg	MeHg
8/23/2007	939	16.9
7/23/2008	926	16.9
8/5/2008	2020	10.27
8/18/2008	998.5	16.8
9/4/2008	872.5	14.7
9/17/2008	756	16.9
9/30/2008	781	22.9
10/22/2008	986	16.2
5/11/2009	860	34.5
6/3/2009	825	22
6/25/2009	686	26.9
7/16/2009	930	33.1
8/5/2009	1140	31
8/31/2009	755	10.3

W26 Low	THg	MeHg
8/23/2007	954	20.4
7/23/2008	865	41.1
8/5/2008	1010	32.2
8/18/2008	901	38.1
9/4/2008	1030	24
9/17/2008	870	45.7
9/30/2008	891	34.2
10/22/2008	876	34.1
5/11/2009	1030	45.3
6/3/2009	920	37.1
6/25/2009	974	38.6
7/16/2009	1100	42.7
8/5/2009	986	47.9
8/31/2009	939	29.1

W26 Mudflat	THg	MeHg
8/23/2007	1390	27.5
7/23/2008	1340	28.2
8/5/2008	992	19.5
8/18/2008	671	16.9
9/4/2008	1360	26.45
9/17/2008	515	9.66
9/30/2008	656	10.6
10/22/2008	719	15.8
5/11/2009	1190	28.1
6/3/2009	1150	13.6
6/25/2009	1150	31.1
7/16/2009	976	27.5
8/5/2009	813	23.5
8/31/2009	768	21.9

APPENDIX 15-4:

Results of ANOVA statistical analysis of data for total and methyl Hg in surface sediments 2006 – 2010. All analyses test for an effect of year. Level of significance used was $p=0.05$. Only significant test results shown; all sites not listed were not significant.

Sites	Parameter	Site	p value	Comment
E01 transect	Total Hg	All	n/a	No sites significant
	Methyl Hg	E01-4	0.045	Significant decrease
Intertidal	Total Hg	ES13	0.009	Significant increase
	Methyl Hg	OV1	0.001	Significant decrease
		ES04	0.003	Significant decrease
Wetlands – high elevation	Total Hg	All	n/a	No sites significant
	Methyl Hg	W25	0.001	Significant increase
		W26	0.003	Significant increase
		W63	0.04	Significant decrease
Wetlands – medium elevation	Total Hg	All	n/a	No sites significant
	Methyl Hg	W21	0.038	Significant decrease
Wetlands – low elevation	Total Hg	All	n/a	No sites significant
	Methyl Hg	W25	0.011	Significant decrease
Wetlands - mudflats	Total Hg	W21	0.016	Significant decrease
		W25	0.044	Significant decrease
	Methyl Hg	W21	0.011	Significant decrease
		W25	0.036	Significant decrease