

PENOBSCOT RIVER MERCURY STUDY

Chapter 12

Factors controlling methyl mercury production in intertidal, wetland and bay sediments in the Penobscot estuary

**Submitted to Judge John Woodcock
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1 SUMMARY

This work was carried out to determine the effect of temperature, seasonality, periodic flooding, total mercury (Hg) concentration, and geographic location on methyl Hg production in the surface sediments of the Penobscot system.

Evidence was found for a spring pulse of methyl Hg production in tidally flooded sediments in the Penobscot system, both in the project data and in the data obtained by the Smithsonian Environmental Research Center (SERC) (Dr. Gilmour) (Chapter 11—Figure 11-2.33). However, in wetland sediments, spring was not always the period of highest methylation activity. In the deeper subtidal sites in Penobscot Bay, that are not directly tidally influenced, a spring pulse was not observed.

Regarding evidence for seasonality in methyl Hg production, results for intertidal sediments from 2006-2007 indicated higher methyl Hg concentrations in late summer/early autumn. However, in 2008 and 2009, only half of the wetland sites studied showed this. High methylation activity in late summer/early fall was only seen in one of the two sampling years (2008). This may have been related to higher July temperatures in 2008. Subtidal sites in Penobscot Bay did show a seasonal pattern of lower methyl Hg values in spring, higher in summer, and lower in fall. Only high elevation sites exhibited a consistently positive, but weak, relationship with sediment surface temperature.

There was only weak evidence for an effect of high spring tides, which are tides that flood the higher elevations of wetlands periodically (not just in the spring). This was evident at some sites but not others; regular patterns might have been clearer with more frequent sampling.

Higher methyl Hg concentrations at sites with intermediate salinity, observed previously, were evident in the results of this study. The differences were most evident at the higher elevations. The same patterns were seen in the SERC data (Chapter 11 of this report).

Because of the differences in methylation rates from one habitat to another, and from one season to another, it can appear in a large data set that methyl Hg is not related primarily to total Hg. However, when individual habitats were considered singly, relationships between methyl Hg and total Hg were clearly evident.

The results of this study have two implications for remediation:

1. Concern over the season of sediment removal, if it were recommended, would appear to be minimal.
2. Any remediation method that decreased total Hg in surface sediments should lower methyl Hg in those sediments.

2 INTRODUCTION

The purpose of this work was to determine the effect of some of the many factors known to affect mercury (Hg) methylation, including temperature, seasonality, periodic flooding, total Hg concentration, and geographic location, in surface sediments in the Penobscot. The proportion of the Hg that is methyl Hg (% methyl Hg) in surficial sediments was used as a proxy for the net methylation balance in these sediments because the proportion of total Hg that is in the methyl form is a good indicator of the relative rate of production of methyl Hg (Krabbenhoft et al. 2007). Seasonality of methyl Hg production is often observed in northern systems because rates of methylation of Hg are controlled in part by temperature (Callister and Winfrey 1986; Bodaly et al. 1993) and other factors, including sulfate supply (Krabbenhoft et al. 1998). This temperature dependence is to be expected because Hg methylation is mediated by activity of certain bacterial species, and generally bacterial metabolic rates are linked positively with temperature. Sediments in a river in Wisconsin showed a peak of methylation activity in the summer, although this was thought to be unrelated to temperature (Callister and Winfrey 1986). However, some sediment sites in this Wisconsin river showed little seasonality in methylation activity (Callister and Winfrey 1986). Korthals and Winfrey (1987) and Ramlal et al. (1993) found that rates of methylation in northern lake sediments peaked in the warm summer months. Bloom et al. (2004) studied sediments in the Venice Lagoon and found that methyl Hg concentrations were 3 times higher in spring and summer than in winter. However, in Lavaca Bay, Texas, sediments, Bloom et al. (1999) found peaks in %MeHg in sediments and porewater in the early spring, with lower values at other times of the year.

We have previously shown for the Penobscot River, that the percent of total Hg that is methyl Hg showed a seasonality in about a third of the intertidal sediment sites (Phase I Update Report, Appendix 3), with % methyl Hg being about 50% higher in August and early September as compared to other times of the year. In this part of the Study, we sampled a number of selected sites in the Penobscot Bay and at wetlands in the lower river to examine the seasonality of methyl Hg production. We also examined the importance of temperature, high spring tides, geographic location, and total mercury concentration in affecting methyl mercury production.

3 SITES

Two categories of sites were sampled in the Penobscot estuary. One category of sites was located in Penobscot Bay (Fort Point Cove) and were the same sites sampled previously in Phase I of the Study, that is, five sites in the northern part of Penobscot Bay along the transect designated E01 (Table 12-1). This transect is located in Fort Point Cove in upper Penobscot Bay (exact locations are shown on a map in the Phase I Update report). These sites were sampled 13 times in 2008 and 2009.

Table 12-1: Sites in upper Penobscot Bay (Fort Point Cove; Transect E-01). Mean values of total Hg, methyl Hg and % methyl Hg were calculated from all 13 sampling dates to provide an indication of differences among the sites.

Site	Latitude (N)	Longitude (W)	Total Hg ng/g dry wt., mean +/- 1 SD	Methyl Hg ng/g dry wt., mean +/- 1 SD	% methyl Hg mean +/- 1 SD
E01-1	44.48209	68.82766	744+/-118	20 +/- 8	2.7 +/- 0.9
E01-2	44.4822	68.81854	849 +/-219	13 +/- 4	1.5 +/- 0.2
E01-3	44.48138	68.80859	495 +/-101	6 +/- 1	1.2 +/- 0.2
E01-4	44.48163	68.79856	291+/-58	3 +/-1	1.0 +/- 0.3
E01-5	44.48299	68.78791	655+/-178	10 +/-3	1.5 +/- 0.3

A second category of sites were wetland sites (Tables 12-2a and 12-2b). These sites were a subset of those examined as part of the geographic survey of wetlands to determine the extent of contamination of wetlands in Phase I of the Study (Appendix 2). A total of six wetlands were sampled. Two were chosen to be representative of wetlands in the predominantly freshwater zone of the lower river (W63 and W10), which had relatively low % methyl Hg in the 2007 survey. Two wetland sites were intended to be representative of the transition zone of the lower river between freshwater and saline conditions (W17 and W21), which had relatively high % methyl Hg in the 2007 survey. And, two wetland sites were intended to be representative of more saline conditions in upper Penobscot Bay (W25 and W26), which had relatively low % methyl Hg in the survey conducted in 2007. This comparison of wetlands was therefore to examine whether methyl Hg production and its seasonality was different in different zones of the lower river and upper estuary. The locations of these sites are shown in Figure 7 of the Phase I Report (Appendix 2 of Chapter 1). These sites were also sampled 13 times in 2008 and 2009.

Table 12-2a: Location and general description of wetland sites.

Site	Latitude (N)	Longitude (S)	Salinity	Description
W63	44 42 31.6	68 50 19.7	More freshwater	South of Orrington, near McGinn Rd, east side of Penobscot River
W10	44 41 52.6	68 50 16.7	More freshwater	South of Orrington, near Bald Hill Cove, west side of Penobscot River
W17	44 36 56.3	68 51 25.6	Intermediate	South of Winterport, west

Site	Latitude (N)	Longitude (S)	Salinity	Description
				side of Penobscot River
W21	44 34 51.7	68 51 20.2	Intermediate	Mendall Marsh, west side of Marsh River
W25	44 33 40.3	68 45 58	More saline	South of Bucksport, East Channel around Verona Island, on east side of river
W26	44 33 47.4	68 44 46.8	More saline	Orland River, west side

Site	Total Hg ng/g dry wt. mean +/- 1 SD	Methyl Hg ng/g dry wt. mean +/- 1 SD	% methyl Hg ng/g dry wt., mean +/- 1 SD
W63 High	456 +/- 249	12 +/- 7	2.7 +/- 1.5
W63 Medium	961 +/- 147	33 +/- 9	3.6 +/- 1.1
W63 Low	1373 +/- 255	50 +/- 13	3.8 +/- 1.4
W63 Mudflat	1390 +/- 392	40 +/- 16	3.0 +/- 1.2
W10 High	773 +/- 71	22 +/- 10	2.8 +/- 1.2
W10 Medium	694 +/- 77	16 +/- 7	2.4 +/- 0.9
W10 Low	284 +/- 79	6 +/- 2	2.3 +/- 0.7
W 10 Mudflat	912 +/- 121	26 +/- 9	2.9 +/- 1.2
W17 High	816 +/- 155	34 +/- 19	4.3 +/- 2.3
W17 Medium	883 +/- 140	51 +/- 22	5.8 +/- 2.1
W17 Low	1025 +/- 581	30 +/- 22	3.0 +/- 1.8
W17 Mudflat	1016 +/- 555	20 +/- 9	2.1 +/- 0.7
W21 High	594 +/- 169	36 +/- 10	6.5 +/- 2.0
W21 Medium	844 +/- 130	44 +/- 17	5.3 +/- 2.0
W21 Low	974 +/- 120	29 +/- 8	3.0 +/- 0.8
W21 Mudflat	1061 +/- 197	27 +/- 7	2.5 +/- 0.5

Site	Total Hg ng/g dry wt. mean +/- 1 SD	Methyl Hg ng/g dry wt. mean +/- 1 SD	% methyl Hg ng/g dry wt., mean +/- 1 SD
W25 High	616 +/- 218	5 +/- 4	1.0 +/- 0.8
W25 Medium	569 +/- 90	13 +/- 5	2.3 +/- 0.9
W25 Low	537 +/- 72	16 +/- 6	3.1 +/- 1.3
W25 Mudflat	569 +/- 143	14 +/- 4	2.6 +/- 0.6
W26 High	663 +/- 153	33 +/- 14	5.1 +/- 1.8
W26 Medium	964 +/- 340	21 +/- 8	2.4 +/- 1.1
W26 Low	953 +/- 75	38 +/- 7	4.0 +/- 0.8
W26 Mudflat	946 +/- 277	21 +/- 7	2.2 +/- 0.5

Site W21 is located within Mendall Marsh. Other sites, located in upper Mendall Marsh near site W21 were also sampled, in August 2009, to examine the effects of high spring tides that are the only tides that flood the upper marsh surface. These sites are given in Table 12-2c. Six sites were sampled on three different dates - August 13; August 21; and August 24, 2009. Samples from August 13th were taken after a prolonged period of no tidal flooding – the previous high spring tide above 12 feet was July 26th, 18 days previously. The next high spring tide above 12 feet was August 17th, or 22 days after the previous flooding spring tide. Sampling on August 21st was four days after the first tide in August above 12 feet and sampling on August 24th was 7 days after the first tide above 12 feet. Light Detection and Ranging (LiDAR) analysis of the elevation of Mendall Marsh has shown that the whole main platform of the upper marsh is flooded during tides of 12 feet or higher.

Site	Latitude N	Longitude W	Total Hg ng/g d.w., mean +/- 1 s.d.	Methyl Hg ng/g d.w., mean +/- 1 s.d.	% methyl Hg, mean +/- 1. s.d.
W21-Upper-West-A	44 34.851	68 51.691	218 +/- 78	21 +/- 6	10.0 +/- 2.2
W21-Upper-West-B	44 34.849	68 51.697	550 +/- 208	5 +/- 2	1.0 +/- 0.2
W21-Upper-Central-A	44 34.821	68 51.663	205 +/- 71	15 +/- 2	7.8 +/- 2.7
W21-Upper-Central-B	44 34.817	68 51.663	264 +/- 51	26 +/- 7	9.8 +/- 2.5
W21-Upper-East-A	44 34.840	68 51.517	488 +/- 27	39 +/- 8	8.0 +/- 1.4
W21-Upper-East-B	44 34.851	68 51.466	742 +/- 72	20 +/- 7	2.7 +/- 0.8

4 METHODS

Penobscot Bay (Fort Point Cove) sediments were sampled using a Van Veen dredge, as previously described (Phase I Update Report). Only the top 3 cm of sediments were retained for analysis. Wetland sediments were sampled by hand at four elevations at each site, as previously described: intertidal, low, medium and high. Again, only the top 3 cm of sediment was sampled. Sediment to be analyzed for methyl Hg was frozen on dry ice within one minute of being exposed to air to prevent the degradation of methyl Hg. Analyses of total Hg and methyl Hg in sediments were performed by Flett Research by standard methods (refer to general methods section).

Sampling periods for Bay and wetland sediments were as follows:

1. Late July 2008 (July 22 and 23).
2. Early August 2008 (August 4, 5, 6).
3. Late August 2008 (August 18 and 20).
4. Early September 2008 (September 3 and 4).
5. Mid September 2008 (September 16, 17, 18).
6. Late September 2008 (September 29 and 30).
7. Late October 2008 (October 20, 21, 22).
8. Mid May 2009 (May 11 and 12).
9. Early June 2009 (June 2 and 3).
10. Late June 2009 (June 24 and 25).
11. Mid July 2009 (July 15 and 16).
12. Early August 2009 (August 4 and 5).
13. Early September 2009 (August 31, September 1, 2, 4).

The raw data from all sample sites and dates are given in Appendices 1-5.

4.1 Seasonality of Methyl Hg in Penobscot Bay Sediments

Some consistent differences were observed among the five sites sampled. Although all sites in the Bay sediment transect were located in Fort Point Cove, site E01-1 (the most westerly location) usually had higher % methyl Hg (generally 2-3 %, Figure 12-1) than the other sites. The other four sites were generally similar to each other with regard to % methyl Hg (means of 1–1.5 %, Figure 12-1).

Site E01-1 showed marked seasonality (Figure 12-1), compared to the other 4 sites, which showed only slight patterns (E01-4 and E01-5), or no patterns at all (E01-2 and E01-3). The most “typical” seasonal pattern was seen in at E01-1 in 2009, when % methyl Hg increased from early June to mid-summer, with peak values for % methyl Hg occurring in mid-July or early August (Figure 12-1). Percent methyl Hg increased from early June to mid-summer and then decreased to early September. In 2009, there were also some fall peaks in % methyl Hg (Figure 12-1). Thus, in the Penobscot Bay sediments sampled, seasonal patterns were evident at some but not all the sites sampled in this transect. Percent methyl Hg values were similar in May 2009 as compared to the previous fall, indicating no “spring pulse” of methyl Hg production in the Bay sediments.

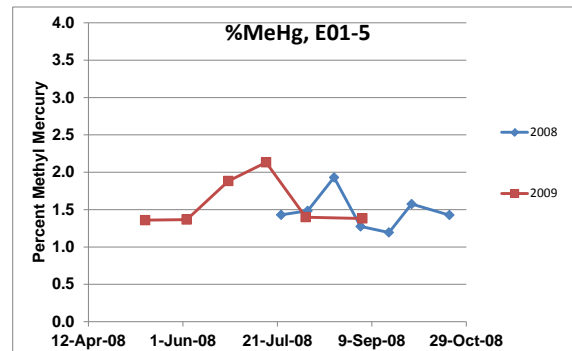
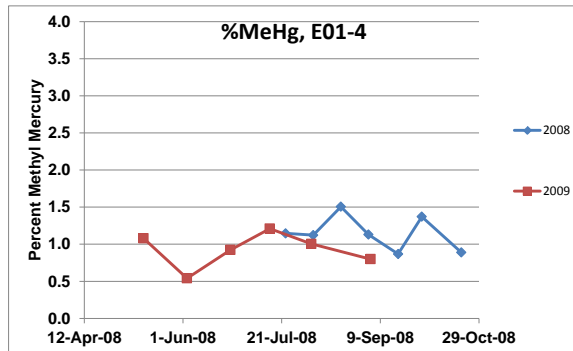
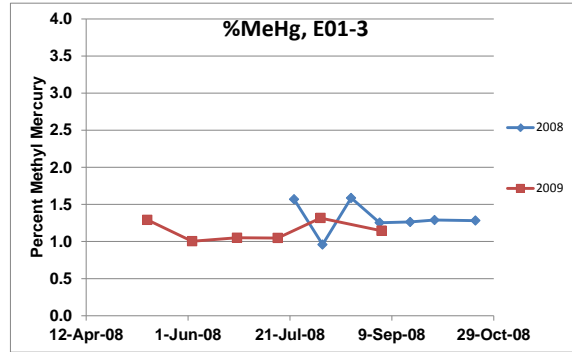
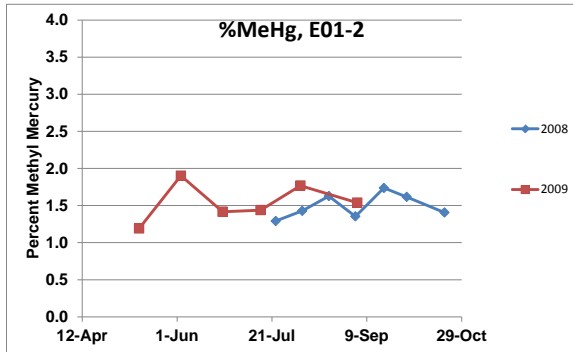
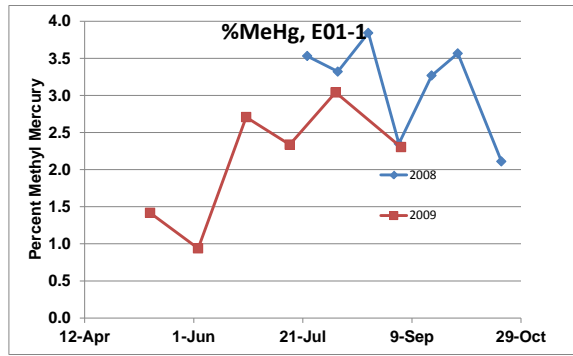


Figure 12-1. Percent methyl Hg in surficial sediments (0-3 cm) in Penobscot Bay (Fort Point Cove) at sites along the E01 transect, 2008 and 2009. See Appendix 1 for sample dates and raw data.

4.2 Seasonality of Methyl Hg in Penobscot Wetland Sediments

4.2.1 High Elevation

Percent methyl Hg values at wetland high elevation sites in the spring of 2009 were quite variable compared to those seen in the fall of 2008. At some sites, % methyl Hg was higher in the spring, at some sites it was lower, and at some sites it was similar to the previous fall (Figure 12-2). Therefore, there is little evidence of a spring pulse of methyl Hg production in high elevation wetland sediments, as has been concluded previously for intertidal sediments.

There were noticeable temporal patterns in % methyl Hg at many high elevation wetland sediment sites in the two sampling years, but they were not completely consistent over the two years (Figure 12-2). Percent methyl Hg was highest in late August or early September at three of the six sites in 2008, although there was little consistent pattern at the other three sites. In 2009, % methyl Hg showed peaks from late June to early August at four of the six sites, but little pattern at W26 (Orland River) and W63 (the lowest salinity site). Thus, high elevation wetland sites generally showed a seasonal pattern in both years with peaks usually occurring in late summer. Two of the sites (W63 and W26) also showed high % methyl Hg in the spring (Figure 12-2).

In addition to the “high elevation” sites sampled in this study, some sites in the “upper Marsh” were sampled for seasonality by SERC. The term “upper marsh” means the marsh platform, at sites further back from the river edge. These sites are not higher in elevation than the “high” elevation sites referred to in this section, but are most appropriately discussed here, because the “high” sites are the furthest back from the river’s edge. These upper marsh sites all showed a clear maximum of % methyl Hg in April (Chapter 11).



Figure 12-2. Percent methyl Hg in surficial sediments (0-3 cm) at high elevation locations at six wetland sites sampled in 2008 and 2009. Panels are shown in order from north to south. See Appendix 2 for sample dates and raw data.

4.2.2 Medium elevation sites

Percent methyl Hg values at three of the six wetland medium elevation sites in the spring of 2009 were noticeably higher as compared to those seen in the fall of 2008. Spring values were higher at both sites with intermediate salinity (W17 and W21) and at the site in the Orland River (W26). At the other three sites, % methyl Hg in the spring was similar to the % methyl Hg in the previous fall (Figure 12-3). These sites were the wetlands further upstream in more freshwater conditions (W63 and W10) and the site further downstream in the river in more saline conditions (W25). Therefore, there was a spring pulse of methyl Hg production in medium elevation wetland sediments at half the sites, especially in the zone of intermediate salinity, as has been observed previously for intertidal sediments.

As for high elevation sites, there were noticeable temporal patterns in % methyl Hg at many medium elevation wetland sediment sites in the two sampling years (Figure 12-3).

Percent methyl Hg was highest in late August or early September at four of the six sites in 2008 (the four upstream sites, W63, W10, W17 and W21). However, there was little consistent pattern at the other two sites that were located further downstream. In 2009, % methyl Hg showed highest values between mid-May and mid-July at four of the six sites, but less regular patterns than in 2008. The peak at W25 occurred in early August, and there was little evidence of seasonality at W10. Thus, many of the medium elevation wetland sites showed a seasonal pattern in both years, with peaks occurring in late summer in one year and early to mid-summer in the other year.

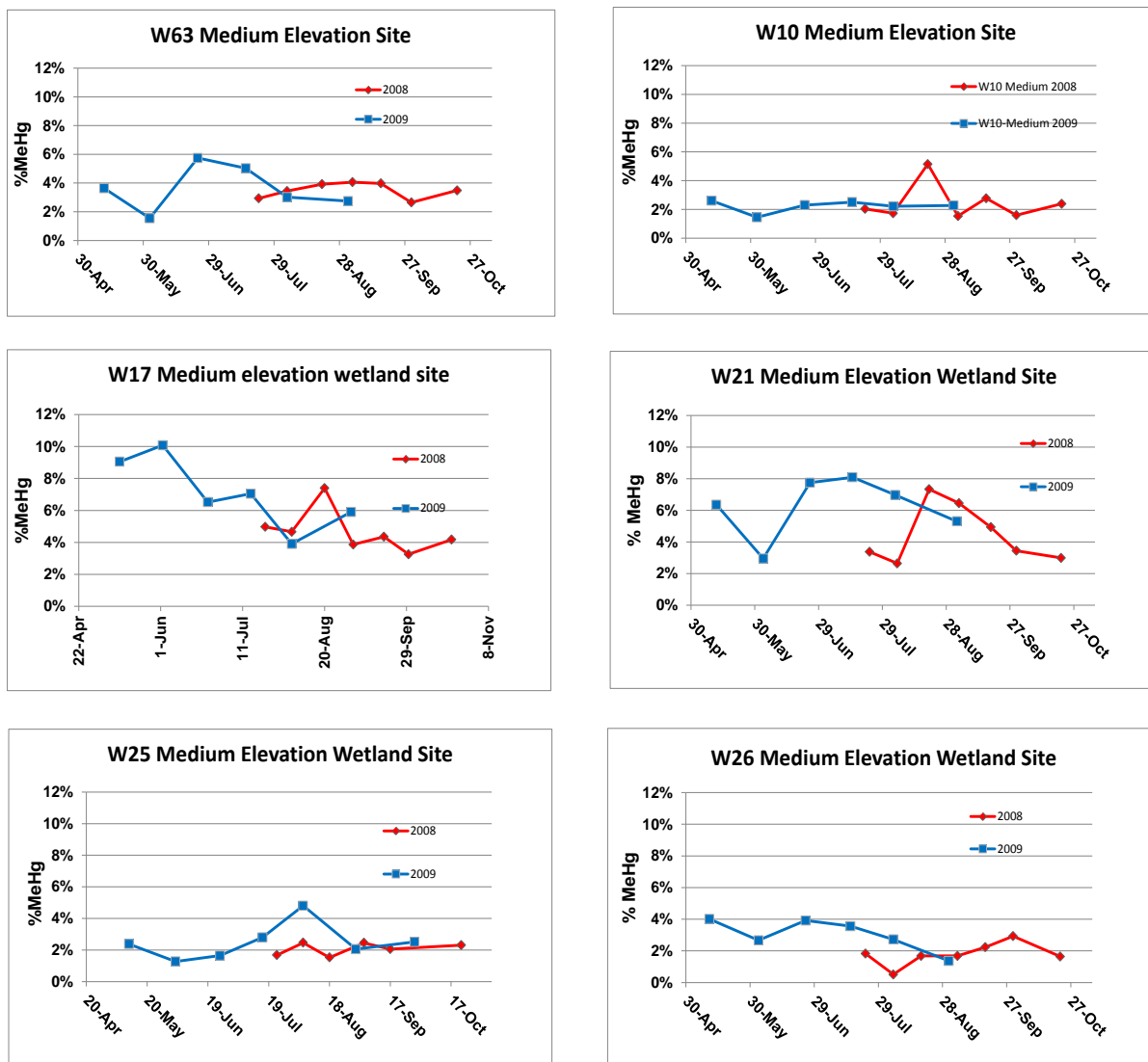


Figure 12-3. Percent methyl Hg in surficial sediments (0-3 cm) at medium elevation locations at six wetland sites sampled in 2008 and 2009. Panels are shown in order from north to south. See Appendix 2 for sample dates and raw data.

4.2.3 Low elevations

Percent methyl Hg was higher in the spring of 2009 than in the fall of 2008 at all sites, indicating an early spring burst of methylation, before the sites were first sampled in mid

May 2009. The % methyl Hg was about 2% at five of the sites at the end of October 2008, and increased to 3% to 4% by the first spring sampling in 2009 (Figure 12-4). At W26, the increase was only from 4% to 4.4%. Increases in % methyl Hg were most pronounced at the two sites with intermediate salinities (W17 and W21) where % methyl Hg increased by 50% to 120%, than in the two most freshwater sites (W63 and W10) where % methyl Hg increased by 25% to 28% or at the two most saline sites (W25 and W26) where % methyl Hg increased only by 13% to 33%. So, a spring pulse of methylation occurred more strongly at sites with intermediate salt concentrations, and these are also the sites that have the highest % methyl Hg in absolute terms.



Figure 11-b4. Percent methyl Hg in surficial sediments (0-3 cm) at low elevation locations at six wetland sites sampled in 2008 and 2009. Panels are shown in order from north to south. See Appendix 2 for sample dates and raw data.

In 2008, three of the low elevation sites showed peaks in % methyl Hg in surficial sediments in mid-August to early September (W10, W17, and W25). The other three sites showed irregular patterns with time during 2008 (Figure 12-4). The presence or lack of obvious seasonal trend was not associated with geographic location, as one

wetland in each of the three geographic areas showed a pattern and the other did not (Figure 12-4).

In contrast to 2008, in 2009, there was very little evidence of any seasonal pattern in % methyl Hg concentrations at low elevations at any of the sites sampled (Figure 12-4).

4.2.4 Intertidal mudflat elevations

At intertidal elevations, % methyl Hg values were higher in the spring of 2009 as compared the previous fall at all sites, indicating an early spring (before mid-May) small to large pulse of Hg methylation, depending on the site (Figure 12-5). Increases between the previous fall and mid-May ranged from 7% to over 100%. These increases were similar in magnitude to low and intermediate elevation site increases, when they occurred.

In 2008, seasonal patterns were hard to discern. This is in contrast to what was observed in 2006-2007. At W63, % methyl Hg was highest in mid-September. At W10, % methyl Hg was highest in late August, as expected, and with a regular pattern of increasing values until late August and declines later in the year, except for a jump in mid-October. At W25, there was little variation in % methyl Hg from July to September and a decline October. At the two remaining sites (W21 and W26), little seasonal pattern was evident.

In 2009, there also appeared to be little in the way of seasonal patterns. At the two most upstream sites, W63 and W10, % methyl Hg was highest in mid-July, but with no regular seasonally-based pattern on either side of those peaks. At W17, the next downstream site, % methyl Hg was highest in early June, with no evident seasonal pattern. At the three more downstream sites there was little variation in % methyl Hg and no indication of a seasonally-based pattern.

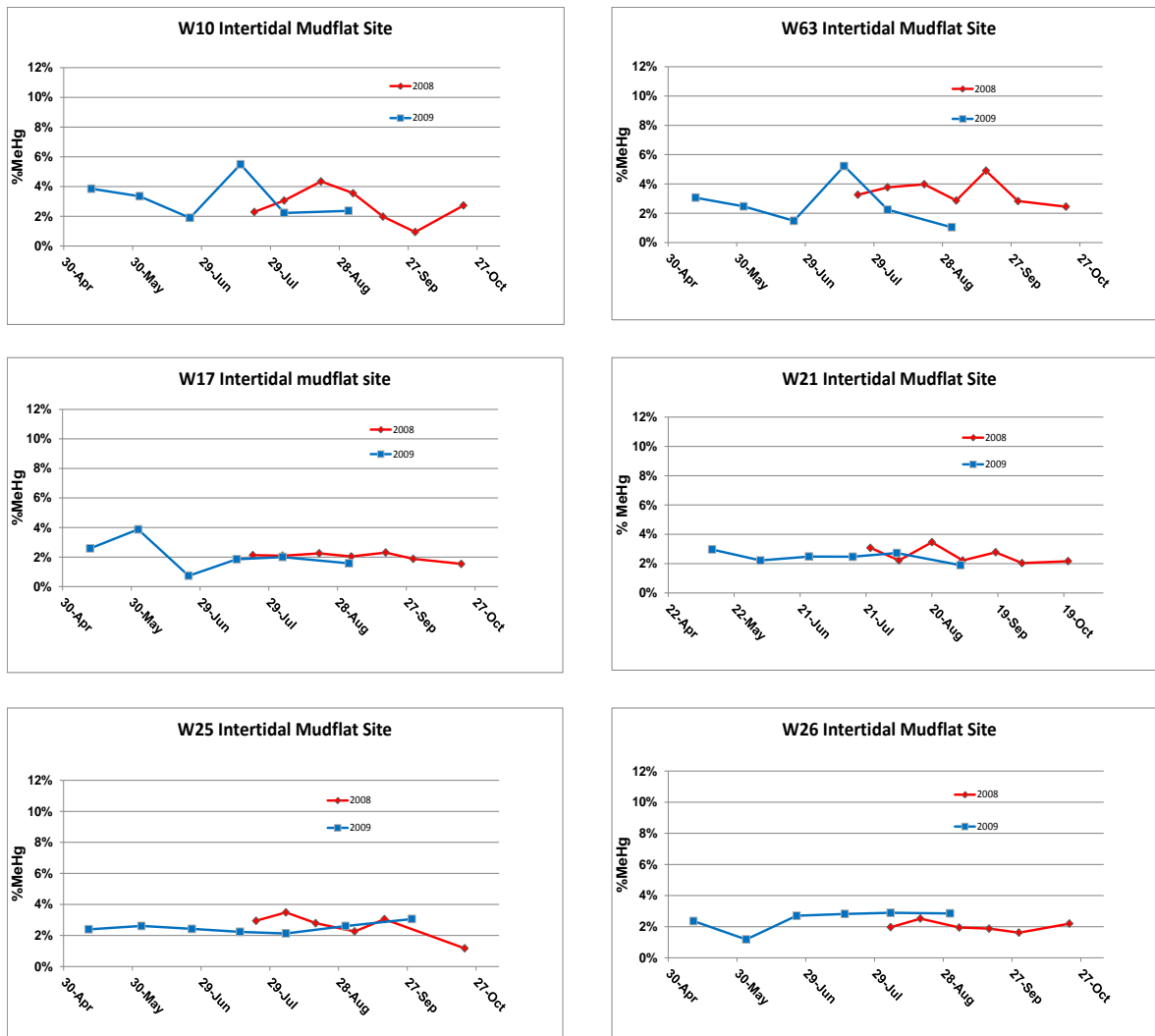


Figure 5. Percent methyl Hg in surficial sediments (0-3 cm) at intertidal mudflat elevation locations at six wetland sites sampled in 2008 and 2009. Panels are shown in order from north to south. See Appendix 2 for sample dates and raw data.

4.3 Summary—Seasonality

Spring pulses in methylation were seen clearly at the SERC upper marsh sites in Mendall Marsh, but were less often seen in the riverine wetlands, or near the river's edge in Mendall Marsh. Near the river, the intertidal sites most often showed a spring pulse (6 out of 6 sites), followed by the low and medium elevations (3 out of 6 sites) and the high elevations (2 out of 6 sites). At other elevations, the most common pattern was a lack of a spring pulse. Thus, while it often occurred, the spring pulse pattern was not universal.

Overall temporal patterns in % methyl Hg were not consistent from year to year, nor were the patterns the same at all the sites within one year. A late summer peak was one of the most common patterns seen, but another common pattern was that of very little predictability from one month to another.

4.3.1 Effect of Spring Tides

At high elevation sites in the six wetlands sampled, there were no noticeable increases in % methyl Hg values in surface wetland sediments related to the occurrence of very high spring tides that would have flooded these elevations. In 2008, spring tides occurred within 3 days of sampling conducted in early August, early September, mid-September, and late October and two weeks or more before sampling carried out in mid-August and late July. If these high spring tides had triggered a significant production of methyl Hg, higher values at sampling times days after high tides should have been obvious, but a visual examination of the data (Figure 12-2) indicated that this was not the case. In 2009, spring tides occurred on July 26, and Aug 17. Similarly, no obvious effects of very high spring tides that occurred on six sampling dates in 2009 were observed (Figure 12-2).

There was also no obvious influence of very high spring tides on % methyl Hg at mid elevation sites in 2008 or in 2009 (Figure 12-3).

In contrast to the apparent lack of an effect of spring tide flooding on periodically sampled wetland sites, discussed above, sampling carried out at high marsh sites in Mendall Marsh specifically to investigate the effects of spring tides did seem to show an effect, but only at half of the sites sampled (Figure 12-6). Spring tides appeared to have a stimulating effect on Hg methylation at three of the six sites sampled, but not at the other three. Percent methyl Hg at Upper West A and Upper Central B rose from 8% to over 12 % during the spring tides, and at Upper East A from about 7% to about 10%. At Upper West B and Central A, % methyl Hg increased after the first high spring tides, but declined after further flooding from spring tides. In contrast, % methyl Hg declined at Upper East B during the spring tides. There were no consistent patterns comparing the pairs of sites from adjacent areas. There would appear to be some evidence from these data for a stimulatory effect of spring tide flooding on Hg methylation, but a more thorough study, including pore water sampling, would be required to make a more definitive conclusion.

Another observation on the high marsh sites is that there were large differences among sites in the proportion of total Hg that was methylated (Figure 12-6), and these differences did not appear to be related to location or habitat type. Four of the six sites sampled had consistently high % methyl Hg values (6% to 12 %) whereas two sites had consistently lower % methyl Hg (1% to 3 %). Both sites that had lower % methyl Hg were adjacent to, and similar, to sites that had high % methyl Hg. Thus, Upper West A and B, sites that are close geographically and similar with regard to vegetation and water conditions (both sites were quite wet with small potholes filled with water and both had a sulfidic smell), nevertheless had very different methylating conditions. Also, Upper East A and B, also close to together and similar (both sites were grassy, flat, fairly dry and had no sulfidic smells), had noticeably different % methyl Hg values. The two Central sites were fairly similar in % methyl Hg values, despite the fact that they were quite different. The Central A site was in cattails and had no sulfidic smell, whereas the Central B site was grassy, adjacent to cattails and did have a sulfidic smell). Thus, obvious similarities in some general site characteristics were not sufficient to distinguish differences that were seen in methylating activities.

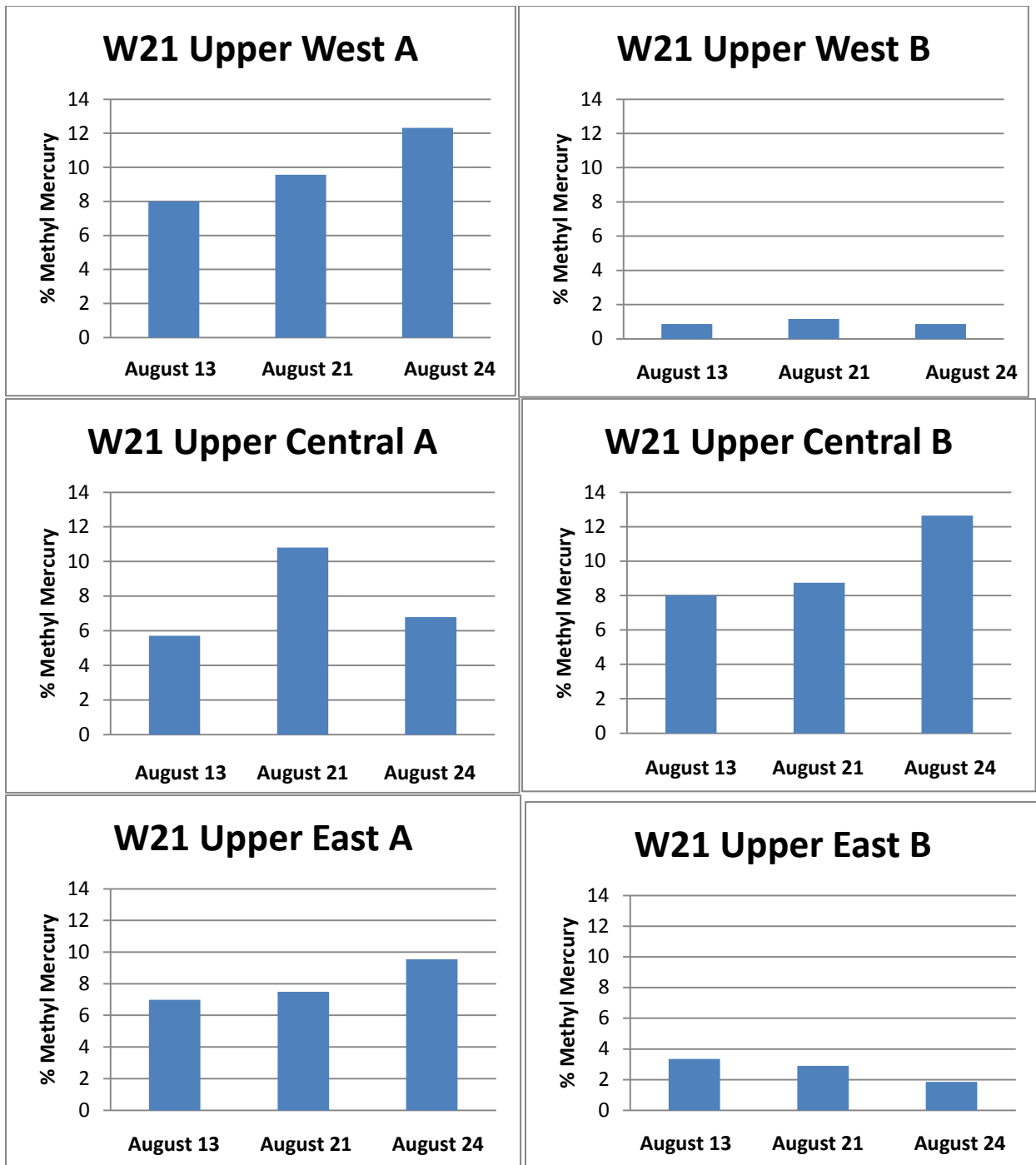


Figure 6. Percent methyl Hg at six upper marsh sites in Mendall Marsh, August 2009. Spring tides occurred on July 26, but there were no spring tides for the next 18 days before sampling on August 13. Four days of spring tides preceded the sampling on August 21, and 7 days of spring tides preceded the sampling on August 24.

4.3.2 Effect of Temperature

Temperature is an obvious factor to examine when looking for seasonal patterns in any biological activity. An overall plot of data from all sites and dates during 2008 and 2009, for % methyl Hg and sediment temperature, did not show any relationship (Figure 12-7).

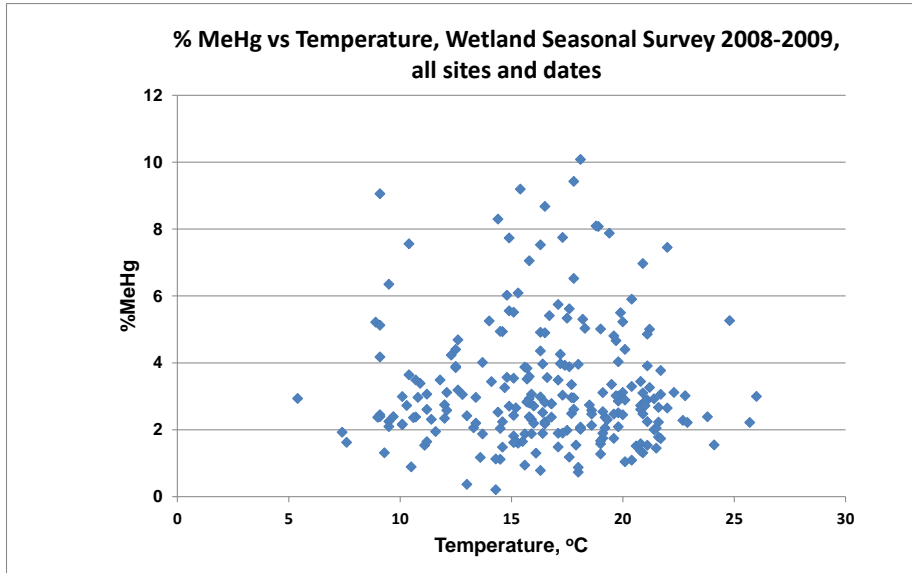
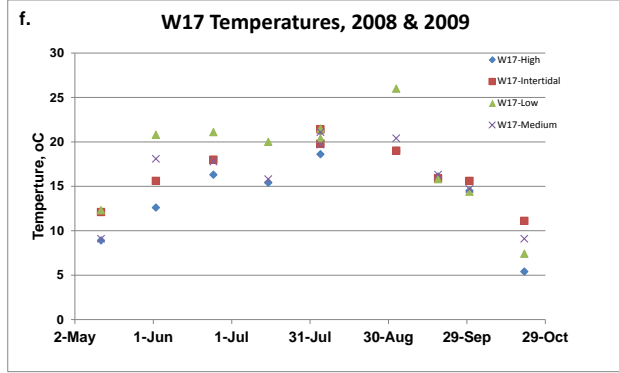
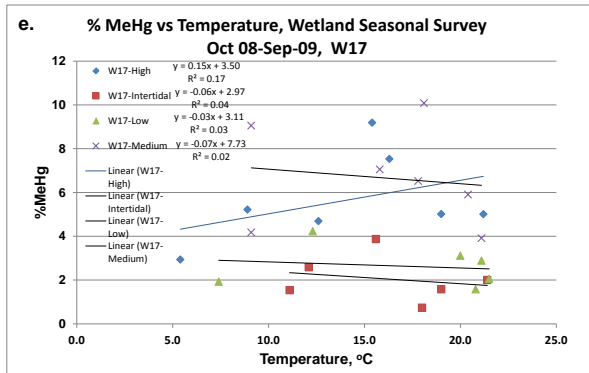
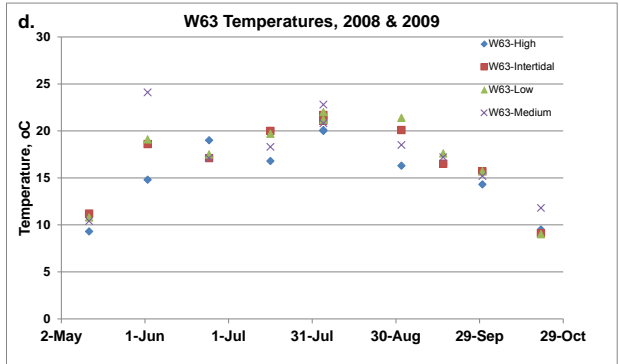
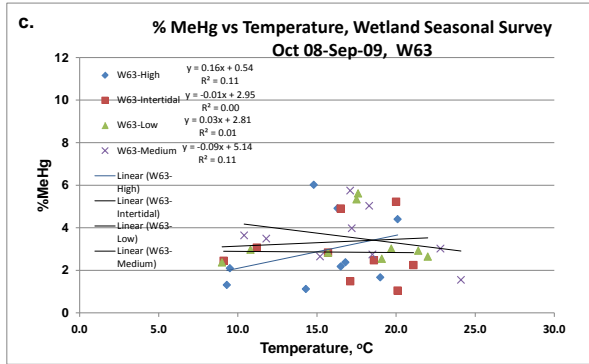
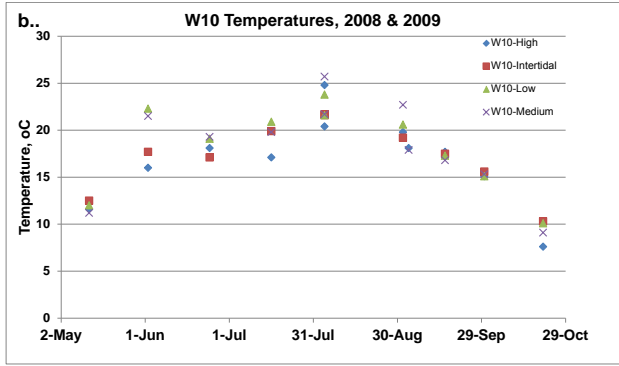
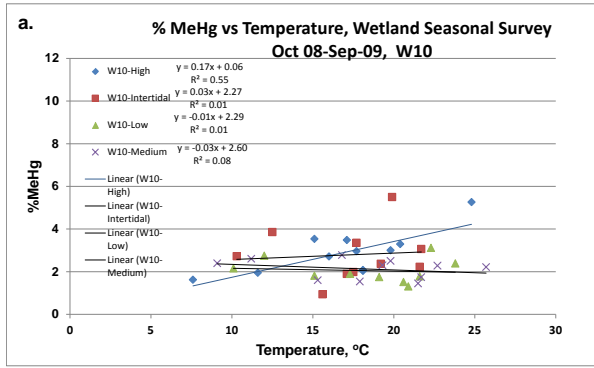


Figure 12-7. Percent methyl Hg in wetland sediments versus surface sediment temperature at sites W63, W10, W17, W21, W25 and W26, at all sampling elevations (High, Medium, Low and Intertidal), during 2008 and 2009. Each site was sampled 10 times.

Surface sediment temperatures at each wetland site, for all elevations, showed the typical increase from spring to midsummer, and decrease into autumn (Figure 12-8 b,d,f,h,j,l).

When measurements were separated by elevation, % methyl Hg had a consistently positive relationship with temperature at the High elevation sites, but not at the other elevations (Figure 12-8 a,c,e,g,i,k). The only other positive relationships were at the medium and low elevation sites in W21. These relationships were not very strong, indicating that surface sediment temperature was only one of the variables affecting methylation activity.



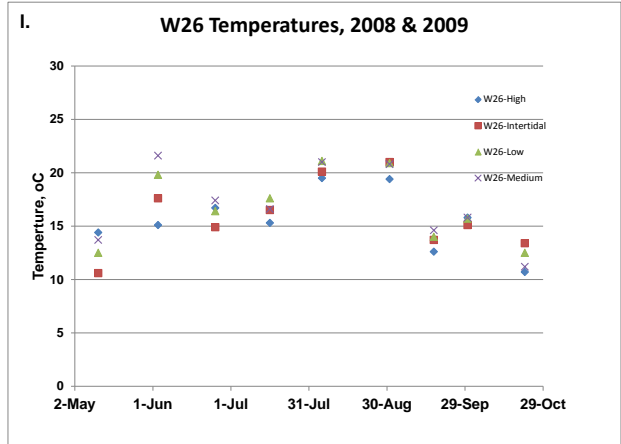
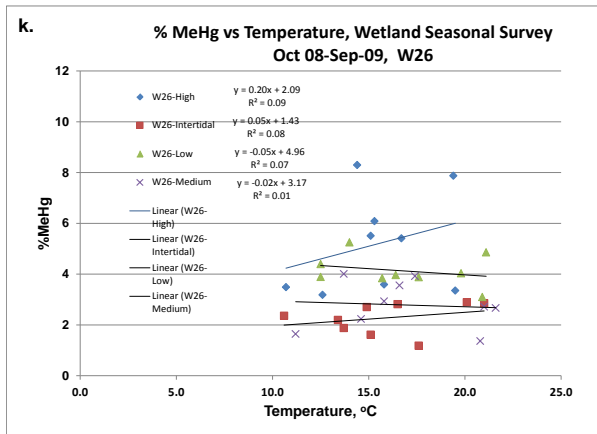
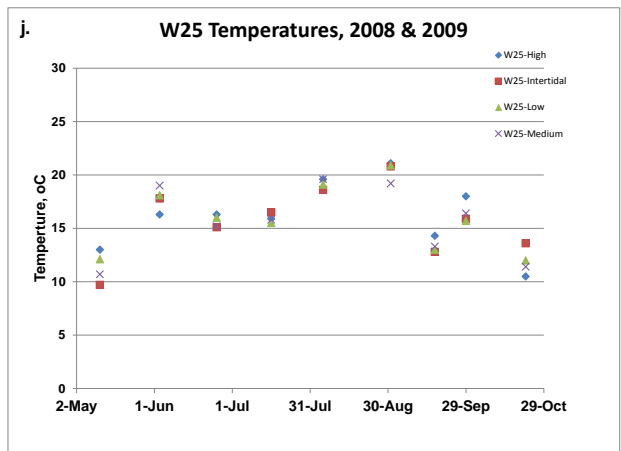
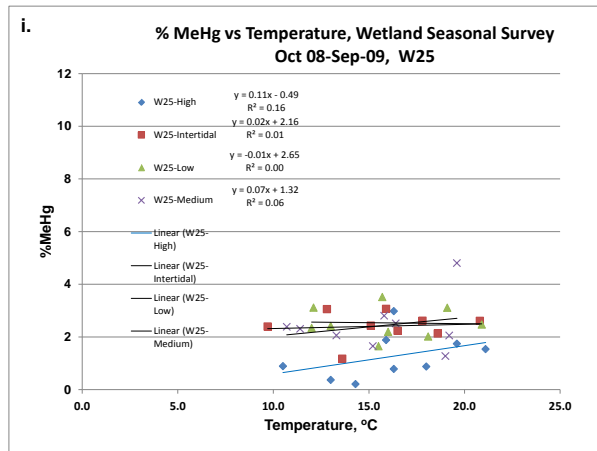
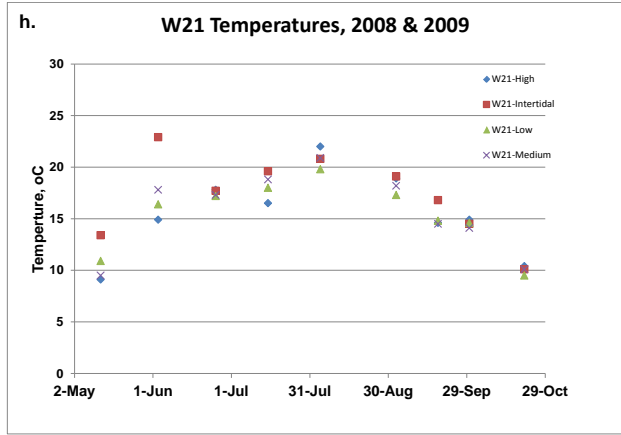
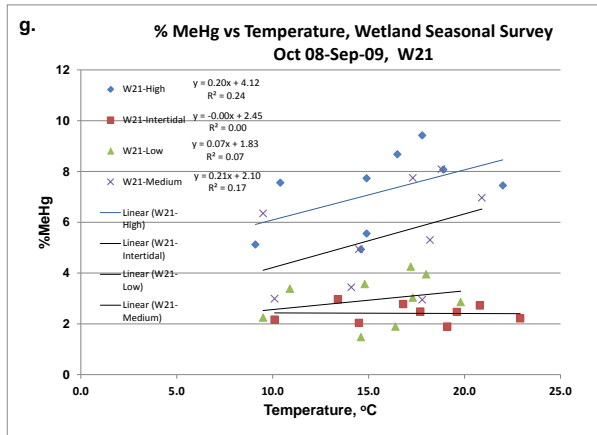


Figure 12-8. Percent methyl Hg plotted against surface sediment temperature, and sediment temperatures measured at each site and elevation during 2008 and 2009.

4.4 Geographic Trends

4.4.1 High Elevation

Based on results from 2007, where higher % methyl Hg values were observed in sites located in the transitional area between upstream freshwater sites and downstream

saltwater sites, it was expected that higher % methyl Hg would be seen at sites W17 and W21. This was certainly the case in 2009, when W21 and W17 were noticeably higher than sites further upstream or downstream, tending to confirm results from 2007 (Figure 12-2). However, in 2008, this geographic pattern was not so clear. W21 had, by far, the highest % methyl Hg values of all other sites, but W17 was lower and similar to the two freshwater sites. The most saline site (W25) had by far the lowest % methyl Hg values, as expected. Also, W63 and W10, the most freshwater sites, had relatively low % methyl Hg as expected (Figure 12-2). However, W26 (Orland River) was also quite high. Year-to-year differences in average salinity (and other chemical factors associated with salinity, such as sulfate concentrations) may be responsible for these contrasting findings. For example, in years where there is more rainfall, some estuarine sites might have lower average salinities than other sites which are not as much influenced by freshwater runoff.

Geographic trends were also observed in medium elevation sites (Figure 12-3). As expected, % methyl Hg was generally higher (but not always) at the W17 and W21 transitional sites. The Orland River site (W26) also tended to be higher in % methyl Hg, but it may experience lower salinities than was expected.

Geographic trends in % methyl Hg were not evident at low elevation sites (Figure 12-4).

There were also no obvious geographic trends evident in the data at intertidal sites (Figure 12-5).

4.5 Effect of Total Mercury Concentration.

In the Penobscot estuary, there is a strong positive relationship between methyl Hg and total Hg when looking across a broad range of wetland sites or intertidal sites (Phase I Report, Appendix 2). These relationships were observed in sampling a large number of sites at the same time of year. In the data set in this chapter, each site was sampled 13 times over two years, so it would be useful to examine this dataset to see whether there are positive relationships between methyl and total Hg concentrations at different times of the year. This is important when trying to predict what will happen in the future as total mercury concentrations in the Penobscot estuary decline. It should be noted, however, that the range of total Hg concentrations in this data set is smaller than in the larger data set.

In the Penobscot Bay sediments, there was a fairly large range of total Hg concentrations over the five sites in the transect. When all the data were used (all 5 sites, all 13 sampling dates), differences in total Hg explained 52% of the variance in methyl Hg concentrations (Figure 12-9). When each date was examined separately (see examples in Figure 12-10a and 12-10b), the variance was similar or greater on five dates, and the variance was less on eight dates. Thus, on a single date, when the influence of other conditions is likely to be more similar at all sites, the influence of total Hg concentration is more apparent (Figure 12-10a and 12-10b) than when all dates are combined (Figure 12-9).

Interestingly, even though so many different dates were sampled, the slopes of the regressions of methyl Hg concentrations vs total Hg concentrations were 0.02 (6 dates), 0.03 (6 dates) and 0.01 (1 date). This is another way to look at the strength of methylation activity, i.e., a steeper slope is an indication of higher activity. The fact that the slopes were almost always either 0.2 or 0.3 indicates that the contribution of methylation activity in determining methyl Hg concentrations was fairly similar from one date to another.

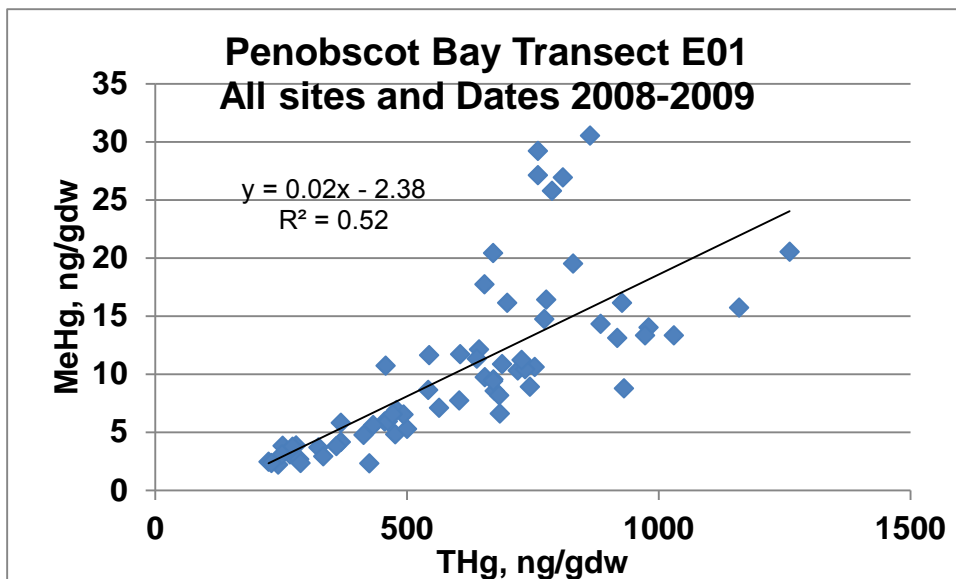


Figure 12-9. Methyl Hg vs total Hg concentrations at five sites in Penobscot Bay (Fort Point Cove), sampled on 13 different dates in 2008 and 2009.

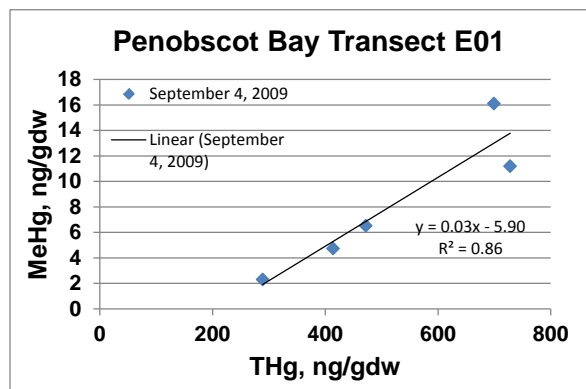
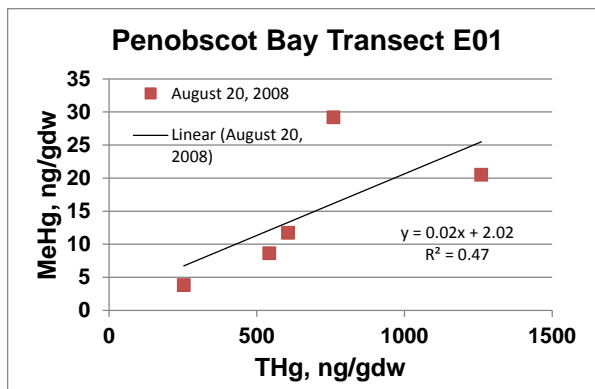


Figure 12-10a and 12-10b. Methyl Hg concentrations versus total Hg concentrations in Penobscot Bay (Fort Point Cove) sediments sampled on August 30, 2008 and September 4, 2009.

The same approach can be taken with the wetland data. If all dates and sites (6 wetlands with 4 elevations in each wetland) are used to look at the relationship between methyl Hg concentrations and total Hg concentrations, the relationship is not strong (Figure 12-11). Looking at this relationship on a single date, and further separating it into elevations on that date (Figure 12-12a and 12-12b), often improved the amount of variation in methyl Hg concentrations that could be explained by total Hg concentrations

(Table 12-3). Improvements were most often seen in the data from the medium, low and intertidal elevations, and not the high elevations, indicating that at high elevations, factors other than the total Hg concentration must have been more important.

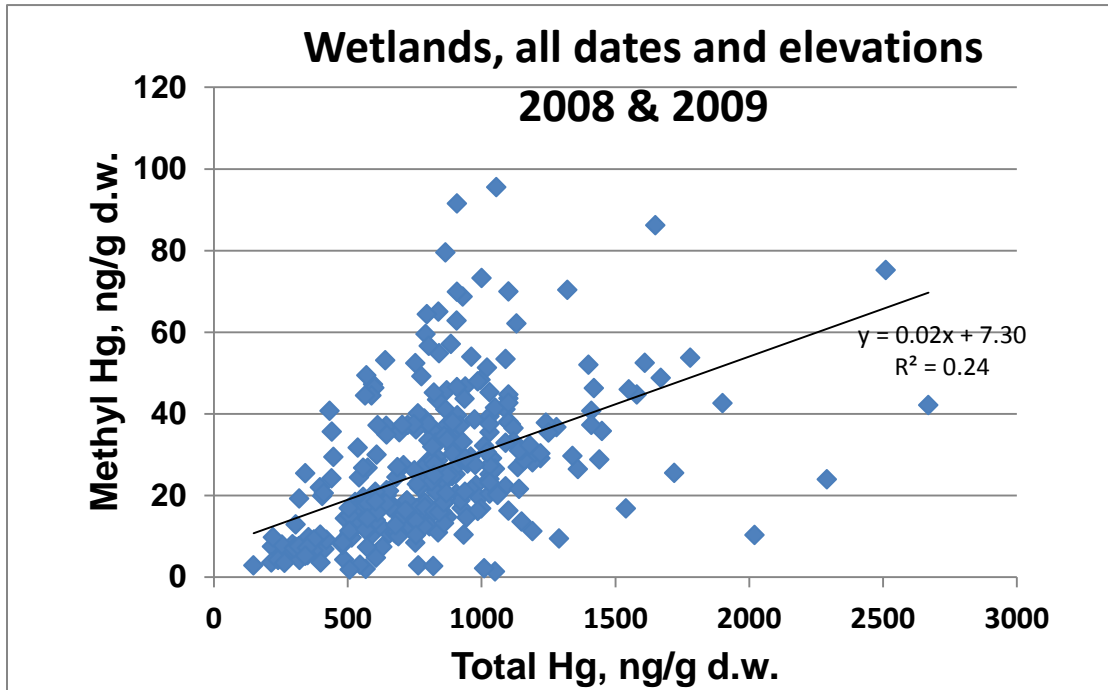


Figure 12-11. Methyl Hg concentrations versus total mercury concentrations in wetland sediments sampled on 13 dates in 2008 and 2009. Six wetlands were sampled at four elevations each.

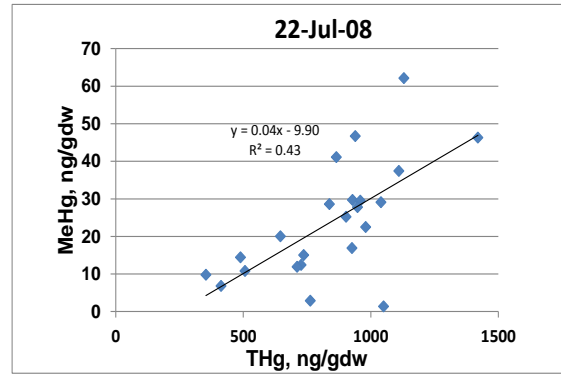
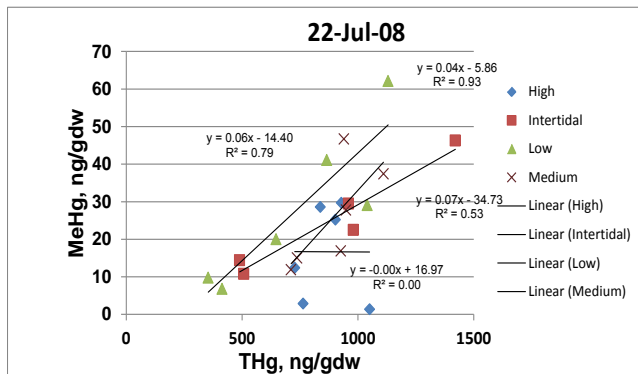


Figure 12-12a and 12-12b. Methyl Hg concentrations versus total Hg concentrations in six wetlands, sampled at four elevations each on July 22, 2008. In the first panel, each elevation is plotted separately, and in the second panel, all elevations are plotted together.

Table 12-3: Results for regression of methyl Hg concentrations versus total Hg concentrations on sampling dates in 2008. The slope of the linear regression, and the regression coefficient are given for all samples taken on each date, and for samples taken at each elevation on the same date.

	n = 24		n = 6		n = 6		n = 6		n = 6	
	All elevations		High		Medium		Low		Intertidal	
	slope	r ²	slope	r ²	slope	r ²	slope	r ²	slope	r ²
Aug 17/07	0.03	0.31	0.09	0.43	0.067	0.16	0.07	0.79	0.025	0.77
Jul 22/08	0.04	0.43	0	0	0.068	0.53	0.06	0.79	0.035	0.93
Aug 4/08	0.0007	0.05	0.03	0.04	0	0.007	0.02	0.34	0.025	0.33
Aug 18/08	0.06	0.38	0.07	0.37	0.1	0.27	0.05	0.77	0.05	0.7
Sep 3/08	0.03	0.31	0.04	0.48	0.07	0.37	0.04	0.43	0.02	0.39
Sep 16/08	0.035	0.33	-0.01	0.05	0.08	0.75	0.04	0.55	0.05	0.56
Sep 29/08	0.02	0.25	-0.01	0.01	0.04	0.74	0.025	0.68	0.025	0.38
Oct 21/08	0.025	0.33	0.04	0.34	0.03	0.28	0.02	0.6	0.035	0.76

5 DISCUSSION AND SUMMARY

The data collected for the Penobscot Bay sediment transect and the wetland sites can be used to address five hypotheses:

1. There is a spring pulse of methylation activity in Penobscot sediments and wetlands.
2. Methylation activity in sediments and wetlands is related to seasonal changes in temperature.
3. High spring tides that flood the marsh platform triggered an increase in methylation activity.
4. Certain geographic patterns are more important than seasonal variation in determining the level of methylation activity in different wetlands.
5. Total mercury concentration is an important factor in determining methyl Hg concentrations within each habitat in Penobscot Bay and wetland sediments.

1. This study does provide evidence for a spring pulse of methyl Hg production in tidally flooded sediments in the Penobscot system. Of 24 wetland sites, 13 showed strong

increases in % methyl Hg in the spring of 2009, compared to fall of 2008, 4 sites showed small increases, and the remaining 5 had no increase. Strong increases were most often observed at low and intertidal sites, and not as often at medium and high elevation sites. So, this effect seems to be strongest at elevations that are low enough to be flooded regularly twice each day and weaker at higher elevations in relation to the tides. Results presented in Chapter 11 of this report, based on sampling the marsh platform of Mendall Marsh, confirm these results. A possible explanation is that in aquatic environments, over winter, decomposition releases nutrients (nitrogen and phosphorus) that contribute to algal blooms, or to new plant growth in the spring when day length increases. These blooms or growth then provide fresh organic material to sediment or soils, which provides substrates for microbial activity. In the Venice Lagoon, Italy, Bloom et al. (2004) saw concentrations of methyl Hg in sediments in the spring and summer that were 3 times higher than in the winter and in Lavaca Bay, Texas, Bloom et al. (1999) observed that concentrations of methyl Hg in sediments in the spring that were 3-4 times those in the winter, and in pore water that were 20-30 times those in the winter. At these very warm locations (Texas and Italy), summer temperatures perhaps exceed the optimum for Hg methylation. The strong increases seen in the Penobscot wetland sites (whole sediment, not pore water) were 2-3 times, while other increases were smaller than this.

It should be noted that the spring pulses seen in the Penobscot wetland sediments were not always the period of highest methylation activity. For most of the sites presented here, there were also peaks in activity later in the year, in the summer or early fall. In the sites on the upper marsh platform, further away from the river, the spring pulses were usually the highest time of methylation activity (Chapter 11).

None of the sediments at sites in the E01 transect in Penobscot Bay exhibited a spring pulse.

2. Previous work on intertidal sediments suggested that warm temperatures in later summer and early fall might be a factor in increased methylation activity observed at this time of year. We previously demonstrated, in 2006 and 2007 that % methyl Hg was about 50% higher in August and early September compared to other times of the year in many intertidal sediments (Phase I Update Report). This pattern was assumed to be due to temperature, with the usual “lag” seen at other temperate sites (e.g. Ramlal et al. 1993). The differences seen were modest compared to locations located in warmer climates. Also, seasonal trends in methyl Hg in intertidal sediments were not observed in 2006-2007 at sites in the lower Bay, where methyl Hg was fairly uniformly low at all sites and at all times of the year.

Based on our observations on seasonal trends in intertidal sediments in 2006-2007, we expected to see similar trends in intertidal elevation sediments in 2008 and 2009 and further, we hypothesized that seasonal trends with methyl Hg peaking in late summer at higher elevations in Penobscot wetlands. With 24 sites and 2 years of data, however, there were only 12 site-years that showed peaks at this time of year. Further, only high elevation sites exhibited a consistently positive, but weak, relationship with sediment surface temperature. Thus, temperature did not appear to be a factor at medium, low and intertidal elevation sites in the wetlands, and was a minor factor at high elevations.

High methylation activity in late summer/early fall was more often seen in 2008 than in 2009. In 2008, peak % methyl Hg was often seen in late August or early September, especially in sites at W10 and W17. Three of the intertidal sites had this pattern. However, in 2009, there was little evidence of regular seasonal patterns at any of the sites or elevations. Why the difference between the two years? Temperatures were quite similar between the two years (and generally very similar to mean monthly temperatures), although July 2008 (mean monthly temperature 21°C) was noticeably warmer than July 2009 (18°C). Maybe this higher July temperature in 2008 was just high enough to stimulate methylating bacteria to produce higher % methyl Hg values in August and early September. Also, temperature is not the only factor that can influence microbial activity and that could vary from year to year.

A site that showed a “typical” seasonal pattern of lower values in spring, higher in summer, and lower in fall was the E01-1 site.

3. We also hypothesized that methyl Hg might be affected by very high spring tides, with methylation increasing just after flooding of sediments at higher elevations that had been dry for some period. This may be analogous to the flooding of terrestrial soils in new reservoirs (Bodaly et al. 2004). Very high spring tides which flood high and mid-elevation sites appeared to affect methyl Hg production at some sites but not at others. There was a high amount of variation in % methyl Hg in general at the high elevation sites, and this large degree of temporal variation could have obscured the impact of flooding caused by spring tides. Three of the high marsh sites in Mendall Marsh that were sampled before and after high spring tides did show an apparent effect of periodic flooding on methyl Hg production.

4. The geographic pattern observed in the 2007 survey of wetlands (see Phase I report, Appendix x), with wetlands located in the zone of intermediate salinity having the highest methylation activity, was seen again in this 2008-2009 data set, as well as in the data set collected by SERC (Chapter 11). The differences were most evident at the higher elevations. The high elevation site in the Orland River was also relatively high, even though its salinity was presumably at the higher end of values in this estuary, perhaps indicating that this site is more intermediate in salinity than assumed from its location.

5. The importance of total mercury concentrations in determining methyl Hg concentrations in surficial sediments is substantiated by this data set, but some details of this relationship are important to note. On the one hand, if all other conditions are equal, we expect that more methyl Hg will be produced when total Hg is higher. On the other hand, when comparing a large data set that includes different sampling times or different types of methylation habitats, other factors will vary, and it may appear that total Hg concentration is a less important factor. That is, when data from a single habitat type is examined, e.g., one elevation level in the wetlands, the chances are greater that there will be a good statistical relationship between methyl Hg and total Hg. This is not as likely when a wide variety of sampling dates and habitats are examined together, because conditions other than total Hg are much more variable. Overall, however, these data suggest that if total Hg could be decreased in all habitats, then methyl Hg would

also decrease, but the specific amount by which it decreases would be different in different habitats, and at different times of the year.

Implications for remediation: If sediment removal (dredging) was recommended as a remediation approach for the Penobscot, sediment removal might release more methyl Hg if it was done in late August/early September when methyl Hg concentrations in sediments were generally higher than at other times of the year. However, this would be more of a concern if seasonal trends were more pronounced than they were observed to be. Also, it would appear that noticeable seasonal trends (i.e. higher values for methyl Hg in later summer) do not occur in every year. So, the concern over the season of sediment removal, if it were recommended, should be minimal. However, sediment removal might be best accomplished during cooler months. Geographic location could be a consideration, however, with areas with intermediate salinity showing the greatest seasonal trends, and often the higher % methyl Hg concentrations.

Remediation approaches that lead to decrease in total mercury concentrations in Penobscot wetlands and sediments should result in lower methyl Hg concentrations. This conclusion is suggested by the positive relationships between methyl Hg and total Hg in a variety of types of surface sediments.

6 REFERENCES

- Bloom, N.S., L.M. Moretto, P. Scopece, P. Ugo. 2004. Seasonal cycling of mercury and monomethyl mercury in the Venice Lagoon (Italy). *Marine Chemistry*. 91: 85-99.
- Bloom, N.S., G.A. Gill, S. Cappellino, C. Dobbs, L. McShea, C. Driscoll, R. Mason, J. Rudd. 1999. Speciation and cycling of mercury in Lavaca Bay, Texas, sediments. *Environmental Science & Technology*. 33: 7-13.
- Bodaly, R.A., J.W.M. Rudd, R.J.P. Fudge, and C.A. Kelly. 1993. Mercury concentrations in fish related to size of remote Canadian shield lakes. *Canadian Journal of Fisheries and Aquatic Sciences*. 50:980-987.
- Bodaly, R.A. and 14 co-authors. 2004. Experimenting with hydroelectric reservoirs. *Environmental Science & Technology*. 38:347A-352A.
- Callister, S.M. and M.R. Winfrey. 1986. Microbial methylation of mercury in upper Wisconsin river sediments. *Water, Air and Soil Pollution*. 29:453-465.
- Korthals, E.T. and M.R. Winfrey. 1987. Seasonal and spatial variations in mercury methylation and demethylation in an oligotrophic lake. *Applied and Environmental Microbiology*. 53:2397-2404.
- Krabbenhoft, D.P., C.C. Gilmour, J.M. Benoit, C.L. Babiarz, A.W. Andren, J.P. Hurley. 1998. Methyl mercury dynamics in littoral sediments of a temperate seepage lake. *Canadian Journal of Fisheries and Aquatic Sciences*. 55:835-844.
- Krabbenhoft, D., D. Engstrom, C. Gilmour, R. Harris, J. Hurley, R. Mason. 2007. Monitoring and evaluating trends in sediment and water indicators, pp. 47-86, in *Ecosystem responses to mercury contamination: Indicators of Change*, R Harris, DP Krabbenhoft, R Mason, MW Murray, R Reash, T Saltman (eds.), CRC Press, Boca Raton, FL.
- Ramlal, P.S., C.A. Kelly, J.W.M. Rudd, A. Furutani. 1993. Sites of methyl mercury production in remote Canadian Shield lakes. *Canadian Journal of Fisheries and Aquatic Sciences*. 50:972-979.

APPENDIX 12-1:

Raw data for surficial (0-3 cm) sediment samples taken from Penobscot Bay at five sites along transect E01 (Fort Point Cove), 2008 and 2009. The locations of the sites are shown on Figure 7 in the Phase I Update Report and the geographic coordinates for the sites are shown in Table 12-1 (above).

Site	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
E01-1	July 23, 2008	864	30.50	3.53
E01-2	July 23, 2008	1030	13.30	1.29
E01-3	July 23, 2008	369	5.79	1.57
E01-4	July 23, 2008	268	3.07	1.15
E01-5	July 23, 2008	720	10.30	1.43
E01-1	August 6, 2008	810	26.90	3.32
E01-2	August 6, 2008	980	14.00	1.43
E01-3	August 6, 2008	685	6.57	0.96
E01-4	August 6, 2008	369	4.14	1.12
E01-5	August 6, 2008	655	9.72	1.48
E01-1	August 20, 2008	760	29.20	3.84
E01-2	August 20, 2008	1260	20.50	1.63
E01-3	August 20, 2008	543	8.61	1.59
E01-4	August 20, 2008	253	3.81	1.51
E01-5	August 20, 2008	606	11.70	1.93
E01-1	September 3, 2008	830	19.50	2.35
E01-2	September 3, 2008	1160	15.70	1.35
E01-3	September 3, 2008	564	7.07	1.25
E01-4	September 3, 2008	324	3.66	1.13
E01-5	September 3, 2008	604	7.70	1.27
E01-1	September 18, 2008	788	25.75	3.27

Site	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
E01-2	September 18, 2008	927	16.10	1.74
E01-3	September 18, 2008	675	8.52	1.26
E01-4	September 18, 2008	334	2.90	0.87
E01-5	September 18, 2008	744	8.88	1.19
E01-1	September 30, 2008	760	27.10	3.57
E01-2	September 30, 2008	885	14.30	1.62
E01-3	September 30, 2008	456	5.88	1.29
E01-4	September 30, 2008	280	3.83	1.37
E01-5	September 30, 2008	689	10.85	1.57
E01-1	October 20, 2008	777	16.40	2.11
E01-2	October 20, 2008	754	10.60	1.41
E01-3	October 20, 2008	462	5.92	1.28
E01-4	October 20, 2008	244	2.17	0.89
E01-5	October 20, 2008	918	13.10	1.43
E01-1	May 12, 2009	672	9.52	1.42
E01-2	May 12, 2009	683	8.14	1.19
E01-3	May 12, 2009	433	5.59	1.29
E01-4	May 12, 2009	225	2.43	1.08
E01-5	May 12, 2009	273	3.71	1.36
E01-1	June 3, 2009	931	8.74	0.94
E01-2	June 3, 2009	773	14.70	1.90
E01-3	June 3, 2009	477	4.78	1.00
E01-4	June 3, 2009	425	2.30	0.54
E01-5	June 3, 2009	973	13.30	1.37

Site	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
E01-1	June 25, 2009	654	17.70	2.71
E01-2	June 25, 2009	735	10.40	1.42
E01-3	June 25, 2009	500	5.25	1.05
E01-4	June 25, 2009	286	2.64	0.92
E01-5	June 25, 2009	643	12.10	1.88
E01-1	July 15, 2009	458	10.70	2.34
E01-2	July 15, 2009	479	6.89	1.44
E01-3	July 15, 2009	360	3.77	1.05
E01-4	July 15, 2009	252	3.05	1.21
E01-5	July 15, 2009	544	11.60	2.13
E01-1	August 5, 2009	671	20.40	3.04
E01-2	August 5, 2009	639	11.30	1.77
E01-3	August 5, 2009	493	6.49	1.32
E01-4	August 5, 2009	231	2.32	1.00
E01-5	August 5, 2009	671	9.39	1.40
E01-1	September 4, 2009	699	16.10	2.30
E01-2	September 4, 2009	728	11.20	1.54
E01-3	September 4, 2009	414	4.74	1.14
E01-4	September 4, 2009	289	2.31	0.80
E01-5	September 4, 2009	472	6.52	1.38

APPENDIX 12-2:

Raw data for total Hg and methyl Hg in surficial (0-3 cm) sediments at four elevations at six wetland sites, 2008, 2009. Geographic coordinates for these sites are given in Table 12-2a (above).

Site & Elevation	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
W63-High	July 22, 2008	727	12.4	1.71
W63-Medium	July 22, 2008	948	27.8	2.93
W63-Low	July 22, 2008	1130	62.15	5.50
W63-Intertidal	July 22, 2008	1420	46.3	3.26
W10-High	July 22, 2008	763	2.87	0.38
W10-Medium	July 22, 2008	737	15	2.04
W10-Low	July 22, 2008	353.5	9.76	2.76
W10-Intertidal	July 22, 2008	980	22.5	2.30
W17-High	July 22, 2008	928	29.7	3.20
W17-Medium	July 22, 2008	939	46.7	4.97
W17-Low	July 22, 2008	413	6.81	1.65
W17-Intertidal	July 22, 2008	507	10.8	2.13
W21-High	July 23, 2008	837.5	28.6	3.41
W21-Medium	July 23, 2008	1110	37.4	3.37
W21-Low	July 23, 2008	1040	29.1	2.80
W21-Intertidal	July 23, 2008	959	29.5	3.08
W25-High	July 23, 2008	1050	1.34	0.13
W25-Medium	July 23, 2008	711	11.9	1.67
W25-Low	July 23, 2008	646	20	3.10
W25-Intertidal	July 23, 2008	489	14.4	2.94
W26-High	July 23, 2008	903	25.2	2.79
W26-Medium	July 23, 2008	926	16.9	1.83

Site & Elevation	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
W26-Low	July 23, 2008	865	41.1	4.75
W26-Intertidal	July 23, 2008	1340	28.2	2.10
W63-High	August 4, 2008	1030	25.2	2.45
W63-Medium	August 4, 2008	1030	35.45	3.44
W63-Low	August 4, 2008	1610	52.5	3.26
W63-Intertidal	August 4, 2008	1090	41.1	3.77
W10-High	August 4, 2008	645	21.25	3.29
W10-Medium	August 4, 2008	587	10.2	1.74
W10-Low	August 4, 2008	476	8.48	1.78
W10-Intertidal	August 4, 2008	768	23.5	3.06
W17-High	August 4, 2008	720	18.6	2.58
W17-Medium	August 4, 2008	937	43.7	4.66
W17-Low	August 4, 2008	1540	16.8	1.09
W17-Intertidal	August 4, 2008	856.5	17.9	2.09
W21-High	August 5, 2008	595	47.2	7.93
W21-Medium	August 5, 2008	666	17.6	2.64
W21-Low	August 5, 2008	944	28.4	3.01
W21-Intertidal	August 5, 2008	962	21.35	2.22
W25-High	August 5, 2008	567	2.02	0.36
W25-Medium	August 5, 2008	569	14	2.46
W25-Low	August 5, 2008	542	24.4	4.50
W25-Intertidal	August 5, 2008	528	18.4	3.48
W26-High	August 5, 2008	693	35.5	5.12
W26-Medium	August 5, 2008	2020	10.27	0.51
W26-Low	August 5, 2008	1010	32.2	3.19
W26-Intertidal	August 5, 2008	992	19.5	1.97
W63-High	August 20, 2008	371	6.45	1.74
W63-Medium	August 20, 2008	832	32.6	3.92

Site & Elevation	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
W63-Low	August 20, 2008	1100	70	6.36
W63-Intertidal	August 20, 2008	1100	43.75	3.98
W10-High	August 20, 2008	871	37.3	4.28
W10-Medium	August 20, 2008	724	37.3	5.15
W10-Low	August 20, 2008	237	6.42	2.71
W10-Intertidal	August 20, 2008	819	35.6	4.35
W17-High	August 20, 2008	818	33.5	4.10
W17-Medium	August 20, 2008	929	68.7	7.40
W17-Low	August 20, 2008	996	48.3	4.85
W17-Intertidal	August 20, 2008	906	20.4	2.25
W21-High	August 20, 2008	755	36.15	4.79
W21-Medium	August 20, 2008	1000	73.3	7.33
W21-Low	August 20, 2008	1030	39.4	3.83
W21-Intertidal	August 20, 2008	1100	38.1	3.46
W25-High	August 18, 2008	819	2.66	0.32
W25-Medium	August 18, 2008	682	10.4	1.52
W25-Low	August 18, 2008	446	29.4	6.59
W25-Intertidal	August 18, 2008	598	16.7	2.79
W26-High	August 18, 2008	907	62.8	6.92
W26-Medium	August 18, 2008	998.5	16.8	1.68
W26-Low	August 18, 2008	901	38.1	4.23
W26-Intertidal	August 18, 2008	671	16.9	2.52
W63-High	September 3, 2008	603	20.85	3.46
W63-Medium	September 3, 2008	1100	44.7	4.06
W63-Low	September 3, 2008	1400	52	3.71
W63-Intertidal	September 3, 2008	1280	36.7	2.87
W10-High	September 3, 2008	786	16	2.04
W10-Medium	September 3, 2008	805	12.4	1.54

Site & Elevation	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
W10-Low	September 3, 2008	217	7.48	3.45
W10-Intertidal	September 3, 2008	831	29.5	3.55
W17-High	September 3, 2008	1100	37.9	3.45
W17-Medium	September 3, 2008	884	34.2	3.87
W17-Low	September 3, 2008	908	69.9	7.70
W17-Intertidal	September 3, 2008	921.5	18.8	2.04
W21-High	September 3, 2008	654	21.1	3.23
W21-Medium	September 3, 2008	885	57.1	6.45
W21-Low	September 3, 2008	1240	37.8	3.05
W21-Intertidal	September 3, 2008	1340	29.7	2.22
W25-High	September 4, 2008	546	3.02	0.55
W25-Medium	September 4, 2008	576	14.1	2.45
W25-Low	September 4, 2008	565	15	2.65
W25-Intertidal	September 4, 2008	770	17.4	2.26
W26-High	September 4, 2008	575	26.8	4.66
W26-Medium	September 4, 2008	872.5	14.7	1.68
W26-Low	September 4, 2008	1030	24	2.33
W26-Intertidal	September 4, 2008	1360	26.45	1.94
W63-High	September 16, 2008	420	9.15	2.18
W63-Medium	September 16, 2008	828	32.9	3.97
W63-Low	September 16, 2008	962	54	5.61
W63-Intertidal	September 16, 2008	1090	53.4	4.90
W10-High	September 16, 2008	892.5	26.4	2.96
W10-Medium	September 16, 2008	612	17	2.78
W10-Low	September 16,	148	2.81	1.90

Site & Elevation	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
	2008			
W10-Intertidal	September 16, 2008	1030	20.4	1.98
W17-High	September 18, 2008	838	10.9	1.30
W17-Medium	September 18, 2008	909	39.6	4.36
W17-Low	September 18, 2008	1220	29.1	2.39
W17-Intertidal	September 18, 2008	790	18.2	2.30
W21-High	September 18, 2008	608	30	4.93
W21-Medium	September 18, 2008	786	38.8	4.94
W21-Low	September 18, 2008	903	32.2	3.57
W21-Intertidal	September 18, 2008	890	24.7	2.78
W25-High	September 17, 2008	1010	2.12	0.21
W25-Medium	September 17, 2008	578.5	11.9	2.06
W25-Low	September 17, 2008	559	13.5	2.42
W25-Intertidal	September 17, 2008	586	17.9	3.05
W26-High	September 17, 2008	559	17.8	3.18
W26-Medium	September 17, 2008	756	16.9	2.24
W26-Low	September 17, 2008	870	45.7	5.25
W26-Intertidal	September 17, 2008	515	9.66	1.88

Site & Elevation	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
W63-High	September 30, 2008	752	8.45	1.12
W63-Medium	September 30, 2008	1030	27.3	2.65
W63-Low	September 30, 2008	1580	44.7	2.83
W63-Intertidal	September 30, 2008	1250	35.4	2.83
W10-High	September 30, 2008	797	28.2	3.54
W10-Medium	September 30, 2008	676	10.8	1.60
W10-Low	September 30, 2008	255	4.62	1.81
W10-Intertidal	September 30, 2008	1190	11.2	0.94
W17-High	September 30, 2008	933	10.4	1.11
W17-Medium	September 30, 2008	1120	36.5	3.26
W17-Low	September 30, 2008	1050	26.5	2.52
W17-Intertidal	September 30, 2008	853	16	1.88
W21-High	September 30, 2008	600	46.4	7.73
W21-Medium	September 30, 2008	785	27	3.44
W21-Low	September 30, 2008	1100	16.25	1.48
W21-Intertidal	September 30, 2008	1090	22.2	2.04
W25-High	September 29, 2008	487	4.26	0.87
W25-Medium	September 29, 2008	573	14.4	2.51

Site & Elevation	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
W25-Low	September 29, 2008	558	19.6	3.51
W25-Intertidal	September 29, 2008	604	18.5	3.06
W26-High	September 30, 2008	682	24.5	3.59
W26-Medium	September 30, 2008	781	22.9	2.93
W26-Low	September 30, 2008	891	34.2	3.84
W26-Intertidal	September 30, 2008	656	10.6	1.62
W63-High	October 21, 2008	242	5.07	2.10
W63-Medium	October 21, 2008	817	28.5	3.49
W63-Low	October 21, 2008	1135	26.9	2.37
W63-Intertidal	October 21, 2008	1190	29.1	2.45
W10-High	October 21, 2008	685	11.1	1.62
W10-Medium	October 21, 2008	594	14.2	2.39
W10-Low	October 21, 2008	296	6.37	2.15
W10-Intertidal	October 21, 2008	855	23.3	2.73
W17-High	October 21, 2008	824	24.2	2.94
W17-Medium	October 21, 2008	802	33.5	4.18
W17-Low	October 21, 2008	1060	20.4	1.92
W17-Intertidal	October 21, 2008	860.5	13.2	1.53
W21-High	October 21, 2008	589	44.5	7.56
W21-Medium	October 21, 2008	1020	30.5	2.99
W21-Low	October 21, 2008	1030	23.15	2.25
W21-Intertidal	October 21, 2008	789	17.1	2.17
W25-High	October 22, 2008	399	3.55	0.89
W25-Medium	October 22, 2008	572	13.2	2.31
W25-Low	October 22, 2008	577	13.5	2.34

Site & Elevation	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
W25-Intertidal	October 22, 2008	630	7.38	1.17
W26-High	October 22, 2008	897	31.3	3.49
W26-Medium	October 22, 2008	986	16.2	1.64
W26-Low	October 22, 2008	876	34.1	3.89
W26-Intertidal	October 22, 2008	719	15.8	2.20
W63-High	May 12, 2009	320	4.2	1.31
W63-Medium	May 12, 2009	1030	37.5	3.64
W63-Low	May 12, 2009	1550	46	2.97
W63-Intertidal	May 12, 2009	935	28.65	3.06
W10-High	May 12, 2009	823	16	1.94
W10-Medium	May 12, 2009	721	18.8	2.61
W10-Low	May 12, 2009	293	8.05	2.75
W10-Intertidal	May 12, 2009	871	33.6	3.86
W17-High	May 12, 2009	834	43.5	5.22
W17-Medium	May 12, 2009	1055	95.5	9.05
W17-Low	May 12, 2009	305	12.9	4.23
W17-Intertidal	May 12, 2009	658	17	2.58
W21-High	May 12, 2009	908	46.5	5.12
W21-Medium	May 12, 2009	775	49.2	6.35
W21-Low	May 12, 2009	892	30.2	3.39
W21-Intertidal	May 12, 2009	1120	33.2	2.96
W25-High	May 11, 2009	507	1.86	0.37
W25-Medium	May 11, 2009	529	12.6	2.38
W25-Low	May 11, 2009	527	16.4	3.11
W25-Intertidal	May 11, 2009	518	12.4	2.39
W26-High	May 11, 2009	640	53.1	8.30
W26-Medium	May 11, 2009	860	34.5	4.01
W26-Low	May 11, 2009	1030	45.3	4.40

Site & Elevation	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
W26-Intertidal	May 11, 2009	1190	28.1	2.36
W63-High	June 2, 2009	319	19.2	6.02
W63-Medium	June 2, 2009	944	14.6	1.55
W63-Low	June 2, 2009	1200	30.5	2.54
W63-Intertidal	June 2, 2009	1160	28.65	2.47
W10-High	June 2, 2009	819	22.2	2.71
W10-Medium	June 2, 2009	689	9.99	1.45
W10-Low	June 2, 2009	254	7.91	3.11
W10-Intertidal	June 2, 2009	824	27.6	3.35
W17-High	June 2, 2009	802	37.6	4.69
W17-Medium	June 2, 2009	908	91.5	10.08
W17-Low	June 2, 2009	785	12.4	1.58
W17-Intertidal	June 2, 2009	708	27.4	3.87
W21-High	June 3, 2009	396	22	5.56
W21-Medium	June 3, 2009	859	25.35	2.95
W21-Low	June 3, 2009	840.5	15.9	1.89
W21-Intertidal	June 3, 2009	866	19.2	2.22
W25-High	June 3, 2009	606	4.75	0.78
W25-Medium	June 3, 2009	572	7.29	1.27
W25-Low	June 3, 2009	621	12.5	2.01
W25-Intertidal	June 3, 2009	398	10.4	2.61
W26-High	June 3, 2009	439	24.2	5.51
W26-Medium	June 3, 2009	825	22	2.67
W26-Low	June 3, 2009	920	37.1	4.03
W26-Intertidal	June 3, 2009	1150	13.6	1.18
W63-High	June 24, 2009	214	3.57	1.67
W63-Medium	June 24, 2009	644	37	5.75
W63-Low	June 24, 2009	1320	70.4	5.33

Site & Elevation	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
W63-Intertidal	June 24, 2009	1720	25.5	1.48
W10-High	June 24, 2009	698	14.5	2.08
W10-Medium	June 24, 2009	605	13.9	2.30
W10-Low	June 24, 2009	240	4.19	1.75
W10-Intertidal	June 24, 2009	1060	20.1	1.90
W17-High	June 24, 2009	791	59.55	7.53
W17-Medium	June 24, 2009	840.5	54.8	6.52
W17-Low	June 24, 2009	1410	40.7	2.89
W17-Intertidal	June 24, 2009	1290	9.44	0.73
W21-High	June 25, 2009	432	40.7	9.42
W21-Medium	June 25, 2009	839	65	7.75
W21-Low	June 25, 2009	893	38	4.26
W21-Intertidal	June 25, 2009	1220	30.3	2.48
W25-High	June 25, 2009	507	15.1	2.98
W25-Medium	June 25, 2009	481	7.94	1.65
W25-Low	June 25, 2009	511	11.2	2.19
W25-Intertidal	June 25, 2009	305	7.4	2.43
W26-High	June 25, 2009	645	34.9	5.41
W26-Medium	June 25, 2009	686	26.9	3.92
W26-Low	June 25, 2009	974	38.6	3.96
W26-Intertidal	June 25, 2009	1150	31.1	2.70
W63-High	July 16, 2009	299	7.1	2.37
W63-Medium	July 16, 2009	1020	51.3	5.03
W63-Low	July 16, 2009	1780	53.7	3.02
W63-Intertidal	July 16, 2009	1650	86.2	5.22
W10-High	July 16, 2009	749	26.1	3.48
W10-Medium	July 16, 2009	830	20.8	2.51
W10-Low	July 16, 2009	263	3.44	1.31

Site & Elevation	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
W10-Intertidal	July 16, 2009	822	45.2	5.50
W17-High	July 15, 2009	865	79.5	9.19
W17-Medium	July 15, 2009	803	56.6	7.05
W17-Low	July 15, 2009	565	17.6	3.12
W17-Intertidal	July 15, 2009	752	13.9	1.85
W21-High	July 15, 2009	569.5	49.4	8.67
W21-Medium	July 15, 2009	796	64.4	8.09
W21-Low	July 15, 2009	1050	41.5	3.95
W21-Intertidal	July 15, 2009	1450	35.8	2.47
W25-High	July 16, 2009	678	12.75	1.88
W25-Medium	July 16, 2009	642	18	2.80
W25-Low	July 16, 2009	480	7.91	1.65
W25-Intertidal	July 16, 2009	506	11.3	2.23
W26-High	July 16, 2009	611	37.2	6.09
W26-Medium	July 16, 2009	930	33.1	3.56
W26-Low	July 16, 2009	1100	42.7	3.88
W26-Intertidal	July 16, 2009	976	27.5	2.82
W63-High	August 4, 2009	221	9.73	4.40
W63-Medium	August 4, 2009	1090	32.9	3.02
W63-Low	August 4, 2009	1410	37.3	2.65
W63-Intertidal	August 4, 2009	1900	42.6	2.24
W10-High	August 4, 2009	762	40.1	5.26
W10-Medium	August 4, 2009	722	16	2.22
W10-Low	August 4, 2009	317	7.56	2.38
W10-Intertidal	August 4, 2009	938	20.9	2.23
W17-High	August 4, 2009	747	37.4	5.01
W17-Medium	August 4, 2009	816	31.9	3.91
W17-Low	August 4, 2009	566	11.6	2.05

Site & Elevation	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
W17-Intertidal	August 4, 2009	1440	28.8	2.00
W21-High	August 4, 2009	341	25.4	7.45
W21-Medium	August 4, 2009	752	52.4	6.97
W21-Low	August 4, 2009	872	24.9	2.86
W21-Intertidal	August 4, 2009	863	23.5	2.72
W25-High	August 5, 2009	481	8.38	1.74
W25-Medium	August 5, 2009	558	26.8	4.80
W25-Low	August 5, 2009	576	17.9	3.11
W25-Intertidal	August 5, 2009	859	18.3	2.13
W26-High	August 5, 2009	504	16.9	3.35
W26-Medium	August 5, 2009	1140	31	2.72
W26-Low	August 5, 2009	986	47.9	4.86
W26-Intertidal	August 5, 2009	813	23.5	2.89
W63-High	September 1, 2009	405	19.9	4.91
W63-Medium	September 1, 2009	1180	32.35	2.74
W63-Low	September 1, 2009	1670	48.8	2.92
W63-Intertidal	September 1, 2009	2290	23.9	1.04
W10-High	September 1, 2009	759	22.8	3.00
W10-Medium	September 1, 2009	715.5	16.3	2.28
W10-Low	September 1, 2009	341	5.17	1.52
W10-Intertidal	September 1, 2009	869	20.6	2.37
W17-High	September 2, 2009	411	20.6	5.01
W17-Medium	September 2, 2009	537	31.7	5.90
W17-Low	September 2, 2009	2510	75.2	3.00
W17-Intertidal	September 2, 2009	2670	42.1	1.58
W21-High	September 2, 2009	441	35.6	8.07
W21-Medium	September 2, 2009	703.5	37.3	5.30
W21-Low	September 2, 2009	823	24.95	3.03

Site & Elevation	Date	Total Hg (ng/g dry wt.)	Methyl Hg (ng/g dry wt.)	% methyl Hg
W21-Intertidal	September 2, 2009	1140	21.6	1.89
W25-High	August 31, 2009	350	5.37	1.53
W25-Medium	August 31, 2009	349	7.17	2.05
W25-Low	August 31, 2009	373	9.22	2.47
W25-Intertidal	August 31, 2009	609.75	15.9	2.61
W26-High	August 31, 2009	565	44.5	7.88
W26-Medium	August 31, 2009	755	10.3	1.36
W26-Low	August 31, 2009	939	29.1	3.10
W26-Intertidal	August 31, 2009	768	21.9	2.85

APPENDIX 12-3:

Raw data for sampling at six sites on the upper marsh of Mendall Marsh in relation to spring tides, August 13, 2009. All samples are the top 3 cm of sediment. August 13, 2009 was preceded by 18 days of tides less than 12 feet (MSL). August 21, 2009 was after four days of spring tides exceeding 12 feet and August 24, 2009 was after seven days of tides exceeding 12 feet.

Site	Latitude N	Longitude W	Date	Total Hg ng/g wet wt.	Total Hg ng/g dry wt.	Methyl Hg ng/g wet wt.	Methyl Hg ng/g dry wt.	% methyl Hg
W21- Upper- West-A	44 34.851	68 51.691	8/13/2009	30.4	304	2.43	24.3	7.99
W21- Upper- West-B	44 34.849	68 51.697	8/13/2009	82.2	781	0.715	6.79	0.87
W21- Upper- Central- A	44 34.821	68 51.663	8/13/2009	32.9	245	1.88	14	5.71
W21- Upper- Central- B	44 34.817	68 51.663	8/13/2009	35.2	222	2.82	17.8	8.02
W21- Upper- East-A	44 34.840	68 51.517	8/13/2009	104	497	7.26	34.7	6.98
W21- Upper- East-B	44 34.851	68 51.466	8/13/2009	304	807	10.2	27	3.35

APPENDIX 12-4:

Raw data for sampling at six sites on the upper marsh of Mendall Marsh in relation to spring tides, August 21, 2009. All samples are the top 3 cm of sediment. August 13, 2009 was preceded by 18 days of tides less than 12 feet (MSL). August 21, 2009 was after four days of spring tides exceeding 12 feet and August 24, 2009 was after seven days of tides exceeding 12 feet.

Site	Date	Total Hg ng/g wet wt.	Total Hg ng/g dry wt.	Methyl Hg ng/g wet wt.	Methyl Hg ng/g dry wt.	% Methyl Hg
W21- Upper- West-A	8/21/2009	19.5	151.5	1.87	14.5	9.57
W21- Upper- West-B	8/21/2009	50.9	489	0.595	5.69	1.16
W21- Upper- Central-A	8/21/2009	16	123	1.73	13.3	10.81
W21- Upper- Central-B	8/21/2009	41.9	320	3.67	28	8.75
W21- Upper- East-A	8/21/2009	92.5	458	6.925	34.3	7.49
W21- Upper- East-B	8/21/2009	235	664	6.79	19.2	2.89

APPENDIX 12-5:

Raw data for sampling at six sites on the upper marsh of Mendall Marsh in relation to spring tides, August 24, 2009. All samples are the top 3 cm of sediment. August 13, 2009 was preceded by 18 days of tides less than 12 feet (MSL). August 21, 2009 was after four days of spring tides exceeding 12 feet and August 24, 2009 was after seven days of tides exceeding 12 feet.

Site	Date	Total Hg ng/g wet wt.	Total Hg ng/g dry wt.	Methyl Hg ng/g wet wt.	Methyl Hg ng/g dry wt.	% methyl Hg
W21- Upper- West-A	8/24/2009	24.3	198	2.99	24.4	12.32
W21- Upper- West-B	8/24/2009	44.9	379	0.385	3.255	0.86
W21- Upper- Central-A	8/24/2009	28.5	246	1.94	16.7	6.79
W21- Upper- Central-B	8/24/2009	36.7	249	4.64	31.5	12.65
W21- Upper- East-A	8/24/2009	107	509	10.2	48.55	9.54
W21- Upper- East-B	8/24/2009	297.5	754.5	5.545	14.05	1.86