PENOBSCOT RIVER MERCURY STUDY

Chapter 17

Background concentrations of mercury in central Maine estuaries

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1. Penobscot River Mercury Study

1 SUMMARY

The purpose of this study was to determine background concentrations of mercury (Hg) in sediments and mussels in estuaries in central Maine. To accomplish this, sampling was carried out in three estuaries that were thought to be relatively unaffected by point sources. Sediments were sampled in the intertidal and subtidal zones, and mussels were sampled in the intertidal. Results were compared to those obtained previously for reaches of the Penobscot out of the direct influence of the HoltraChem site. Total Hg in the deep layers of a core taken in Fort Point Cove was about 20 nanograms per gram dry weight (ng/g dry wt.), a concentration representing the pre-industrial background level or natural abundance of Hg for the area. Present-day surface sediments in the Narraguagus and St. George estuaries and the East Branch of the Penobscot River were 28 to 51 ng/g dry wt.; these levels encompass the modern regional background concentrations, which include anthropogenic Hg derived from atmospheric deposition. The National Oceanic and Atmoshpheric Administration (NOAA) considers levels below 51 ng/g to be at background. Surface sediments in the Sheepscot estuary and in the Old Town – Veazie (OV) reach of the Penobscot ranged from 78 to 145 ng/g, notably higher than the modern regional background. The overall mean for all these surface sediments at sites where Hg contamination was derived primarily from atmospheric deposition, rather than from direct industrial discharges to surface watershed locations was 55 ng/g, much less than the mean of about 800 ng/g in the contaminated zone of the Penobscot. Mussels in the St. George and Narraguagus estuaries had total Hg concentrations of 100 to 200 ng/g dry wt. as compared to 500 to 700 ng/g in the more contaminated Sheepscot estuary and 500 to 1,200 ng/g in the contaminated zone of the Penobscot. In conclusion, it would appear that the modern regional background level for total Hg in surface sediments is about 55 ng/g dry wt.

2 INTRODUCTION

In contemplating any sort of active remediation of the contaminated parts of the Penobscot River and Bay, or in assessing the pace of natural attenuation, it will be important to know the regional background of mercury (Hg) contamination. All natural systems contain Hg at low but measureable concentrations. These concentrations vary from region to region and site to site and are here referred to as *regional background concentrations*. Regional background Hg is a mixture of Hg from natural sources (including volcanoes, degassing of Hg from fresh and salt waters, and weathering of the earth's crust) and human sources (including burning of fossil fuels, especially coal, chemical processes, waste incineration, artisanal gold mining, and dental uses). It would be unrealistic to attempt to remediate any contaminated system below the regional background Hg concentration. The purpose of the work described in this section was to define the regional background concentration of Hg in the general region of the lower Penobscot River. It draws on data collected specifically for this purpose, in Phase II of the study in 2009, and on other relevant data collected as part of Phase I of the Study.

3 METHODS

Sampling to attempt to define regional background concentration of Hg in the general area of the Penobscot drainage basin was conducted in the fall of 2009. This sampling was conducted to supplement data that had already been collected on Hg in sediments from the Penobscot system (upstream of the zone of contamination), in Penobscot Bay far enough south to be outside of the area of contamination, from sediments found in deep sedimentary strata that were deposited before the river and bay were contaminated, and in the St. George estuary, sampled in 2007 as a reference area. Intertidal sediments and mussels and subtidal sediments were sampled in three estuaries in 2009: St. George, Narraguagus, and Sheepscot. The St. George system was known from previous limited sampling to have low concentrations of Hg. The Narraguagus is located in a relatively unpopulated and undeveloped area of Maine and was therefore expected to have similarly low concentrations of Hg. The Sheepscot did not have a known history of Hg contamination but is in a more populated and developed area of Maine.

Sites in the intertidal zone of the three estuaries were sampled for surficial sediments (0-3 cm) and for mussels. Both were sampled by hand. Sites in the subtidal zone were sampled, using a Van Veen dredge, for surficial sediments (also 0-3 cm). All sites were chosen at random. If a particular intertidal or subtidal site was unsuitable (no soft sediments), alternate sites, also chosen at random, were sampled. Three sites were sampled at each estuary site. Sediments were frozen on dry ice within 1 minute of exposure to air to prevent any degradation of methyl Hg. Sediments and mussels were analysed for total Hg and methyl Hg by the standard methods used for this study, which were subjected to our quality assurance/quality control (QA/QC) program. Raw data are given in Appendix tables 14-1 to 14-3.

Data from sampling conducted in 2009 were compared to data from subtidal sediments in the St. George estuary sampled in 2007, and to estimates of background concentrations in the Penobscot from profundal sediments in Fort Point Cove taken in 2007. They were also compared to sediments sampled in southern Penobscot Bay and to sediments in the Penobscot upstream of any known sources of contamination in the East Branch (EB) of the Penobscot and upstream of HoltraChem in the Old Town to Veazie (OV) reach. These samples were collected in 2006 and 2007.

4 RESULTS AND DISCUSSION

4.1 Sediments

Total Hg concentrations were lowest, on average, in the deepest layers of the D-01 core taken from Fort Point Cove, in Penobscot Bay. Hg in the four deepest depths from the core (55-75 cm) were very similar (varying only from 18.3 to 19.4 ng/g dry wt.), were much lower than concentrations in all shallower depths sampled in the core, and showed no variation with depth (Figure 17-1). We consider these concentrations to represent natural, pre-industrial sediment concentrations of total Hg in the area of Penobscot Bay and the central coast of Maine.

Surface sediments sampled from the St. George and Narraguagus estuaries were slightly higher than those in the deep sedimentary strata of Penobscot Bay, with means varying from 28 to 51 ng/g dry wt. (Figure 17-2; Appendix Table 17-1). The East Branch of the Penobscot and sediments taken from the transect furthest south in Penobscot Bay (E-05) had concentrations similar to the St. George sediments, with means of 36 and 50 ng/g dry wt., respectively. The concentrations at all of these sites, with means ranging from 28 to 51 ng/g total Hg, represent modern regional background levels of Hg in sediments in the area of central Maine. NOAA (2004) considers that sediments with less than 51 ng/g dry wt. are indicative of background conditions. Background concentrations of total Hg in sediments in a river floodplain and in estuarine sediments were about 20-45 ng/g dry wt. (Leigh 1994; Gobeil and Cossa 1993), similar to the NOAA range and to the range of concentrations at reference sites in central Maine.

Sediments in the Sheepscot River estuary and those from the Penobscot between Old Town and Veazie (our OV sampling reach) were higher in total Hg, with means ranging from 78 to 145 ng/g dry wt. (Figure 17-2). These areas are apparently slightly contaminated with Hg; however, the source(s) are unknown. These concentrations are above regional background levels (NOAA 2004; Gobeil and Cossa 1993; Leigh 1994). Concentrations in these slightly contaminated areas (78 to 145 ng/g) are lower than those in the more heavily contaminated zone of the Penobscot River and Bay in which surface sediments usually averaged from about 600 to 1,000 ng/g dry wt.

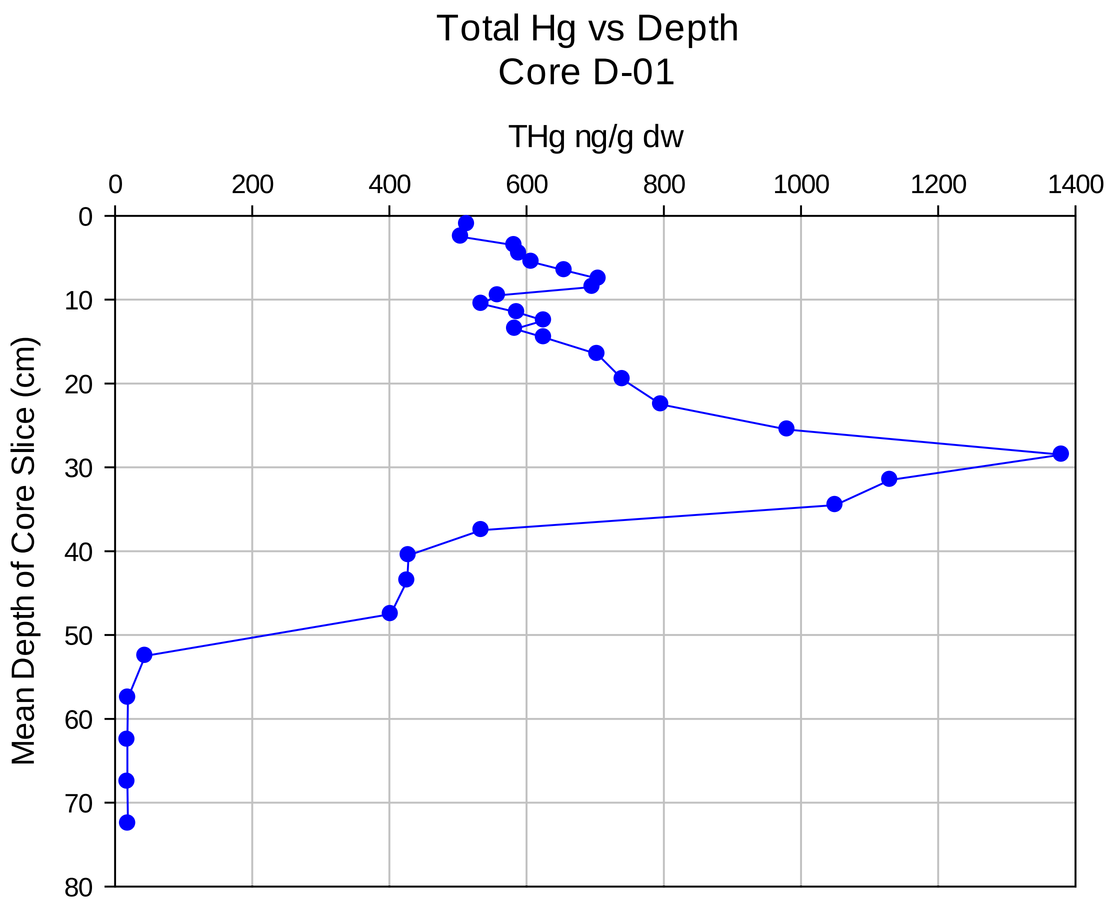

Figure 17-1. Total Hg (ng/g dry wt.) in sedimentary strata in relation to depth in the D-01 piston core taken from Fort Point Cove in August 2007. Taken from Phase I Update report.

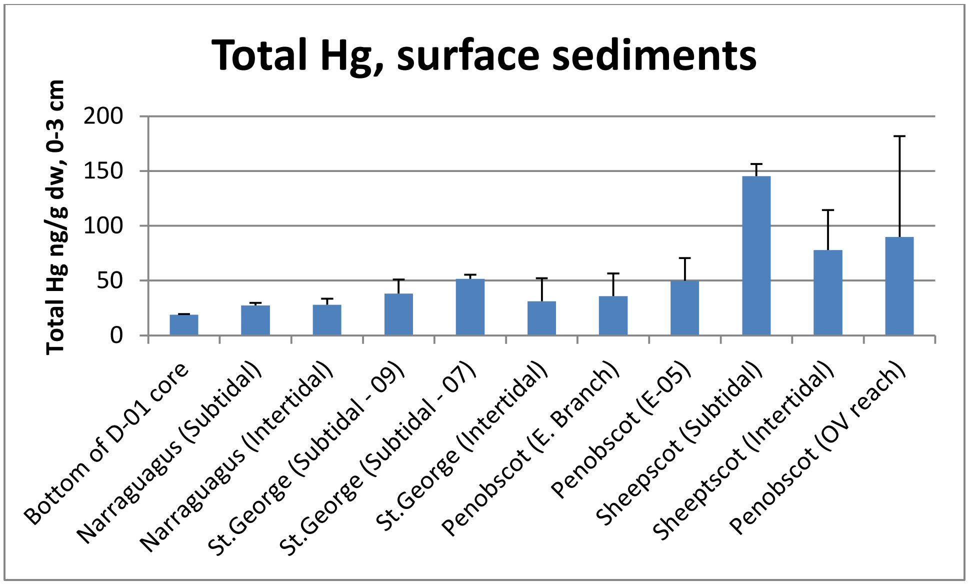

Figure 17-2. Total Hg concentrations in surface sediments (0-3 cm) at various reference or background sites in the Penobscot and adjacent systems, Maine. Error bars are one standard deviation. See Appendix Table 17-1 for raw data and sampling locations. D-01 core was taken in Fort Point Cove in 2007; mean is of the bottom four slices of the core (55-75 cm depths) (see Phase I Update Report). Samples from the Narraguagus, St. George and Sheepscot estuaries were taken in October 2009 except for St. George subtidal samples taken in 2007 (Transect E-08). Penobscot E-05 samples (sub-tidal surface sediments) were also taken in 2007. The E-05 samples were the most southerly profundal samples taken in Penobscot Bay. Penobscot East Branch (EB) and OV (Old Town-Veazie) (inter-tidal surface sediments) were sampled in 2006 and 2007 during the six sampling rounds of Phase I of the Study.

The results of this component of the Study provide evidence that the regional background concentration for modern surface sediments in the central coast of Maine ranges from 30 to 150 ng/g dry wt. and averages about 55 ng/g dry wt.

The sediments sampled in these background areas had methyl Hg concentrations that were closely and positively related to total Hg concentrations, as has been seen in the contaminated zone of the Penobscot system (Figure 17-3). The average percent methyl Hg in surface sediments in background areas was about 1%, which is similar to the percent methyl Hg seen in sediments in the marine portion of Penobscot Bay.

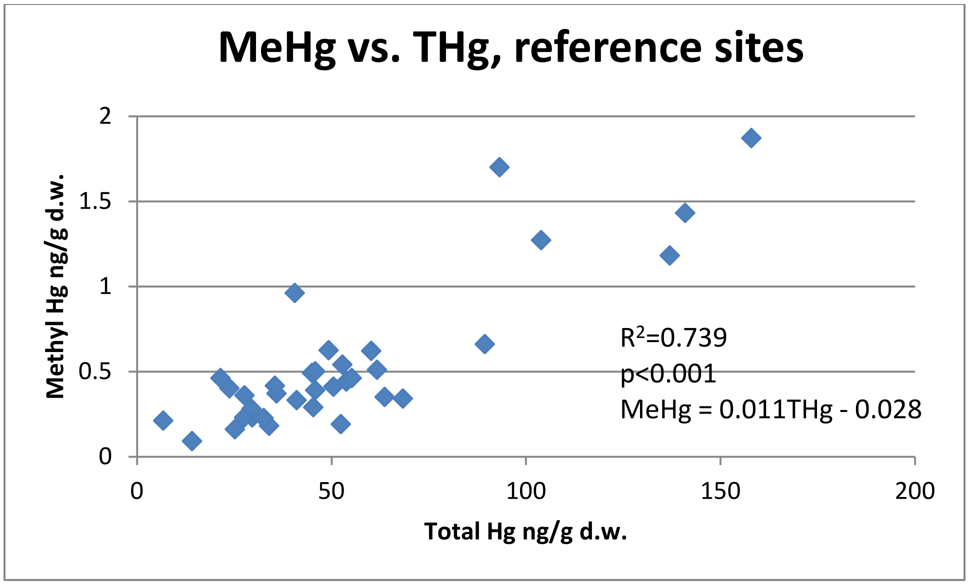

Figure 17-3. Methyl Hg versus total Hg in surface (0-3 cm) sediments in various reference areas sampled on the Central Maine coast, including Outer Penobscot Bay.

4.2 Mussels

Total Hg in mussels from the three reference estuaries varied about 7-fold among the 9 sites sampled (Figure 17-4). The lowest levels were in the St. George and Narraguagus estuaries, which had mean total Hg concentrations of about 100 to 200 ng/g dry wt. Mussels in the Sheepscot estuary, like sediment concentrations, were notably higher, averaging about 500 to 700 ng/g dry wt. Mean concentrations in mussels at contaminated sites in the upper part of Penobscot Bay varied from about 500 to 1,200 ng/g. Hg in mussels in the Narraguagus and St. George were much lower than at contaminated sites in the upper part of Penobscot Bay. Mussel concentrations from the Sheepscot estuary overlapped with concentrations in upper Penobscot Bay and were higher than in mussels in the outer part of Penobscot Bay. Reported total Hg concentrations in mussels from other reference areas in North America and Europe range from 70 to 400 ng/g dry wt. (Bourget and Cossa 1976; Flegal et al 1981; Beiras et al 2002). Concentrations in the St. George and Narraguagus estuaries were within the range of these reference sites reported in the literature whereas those from the Sheepscot and the Penobscot estuaries were notably higher.

There were significant positive relationships between Hg in mussels and Hg in sediments for the three reference estuaries sampled. The correlations between all combinations of total Hg and methyl Hg in mussels and sediment were statistically significant, similar to what has been seen for contaminated sites in the Penobscot system (see Section on relationships between Hg in biota and Hg in sediments). One example, the relationship between total Hg in mussels and total Hg in sediments, is shown in Figure 17-5. In that case, total Hg in sediments explained about 70% of the variation in total Hg in mussels.

Overall, the percent methyl Hg in mussels at reference sites varied from about 20% to 55%. It was lower in the Sheepscot system (20% to 30%) where total Hg was highest, and higher in the St. George (30% t0 35%) and Narraguagus (45% to 55%) estuaries where total Hg was lower than in the Sheepscot (Figure 17-6). Percent methyl Hg in mussels sampled from various sites in the Penobscot during 2006 averaged 32% to 43%.

4.3 Interpretation of results

We have established a baseline for Hg in sediments that is very robust because it depends not only on reference systems outside the Penobscot but also on background within the Penobscot (both geographically and with time, i.e. before industrialization). These two different approaches to the determination of background concentrations agreed closely in that reference areas of the Penobscot (i.e. East Branch) were similar to less contaminated reference systems (i.e. St. George and Narraguagus) whereas some contaminated areas in the Penobscot (Old Town-Veazie) were similar to a slightly contaminated reference estuary (Sheepscot). Concentrations of mercury in mussels in the less contaminated reference estuary overlapped with the less contaminated sites in the Penobscot system (Phase I Report). The background and pre-impact concentrations established by this study, will, along with other possible criteria for determining harm to the system (such as human consumption limits and concentrations known to cause impacts on wildlife) provide reference levels for possible remediation targets to be considered for the Penobscot system.

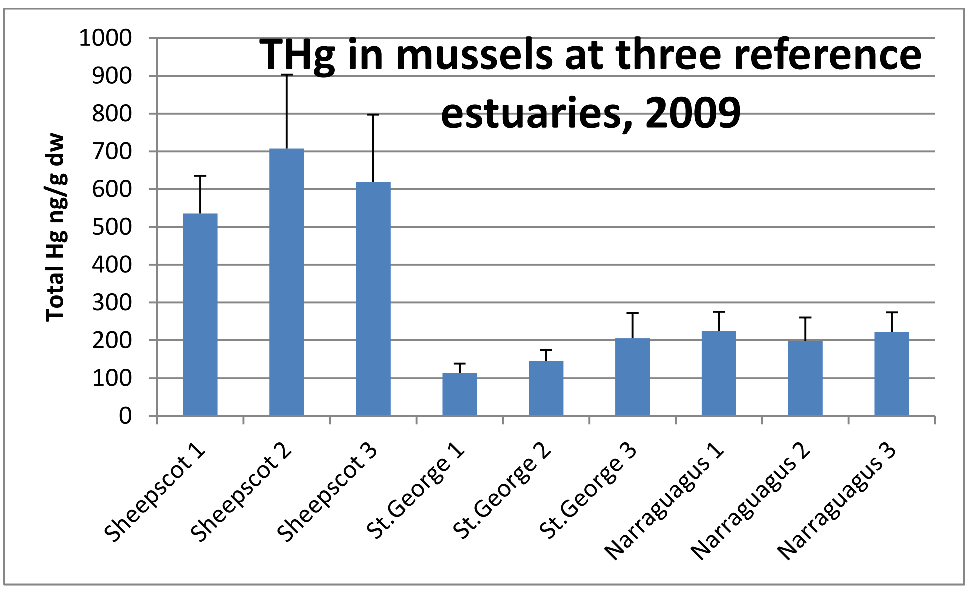

Figure 17-4. Mean total Hg concentrations (ng/g dry wt.) in the soft tissue of mussels from three sites in each of three reference estuaries (Sheepscot, St. George and Narraguagus), 2009. Error bars are one standard deviation. Sample sizes are 20 for all sites. See Appendix Tables 14-2 and 14-3 for summary of raw data.

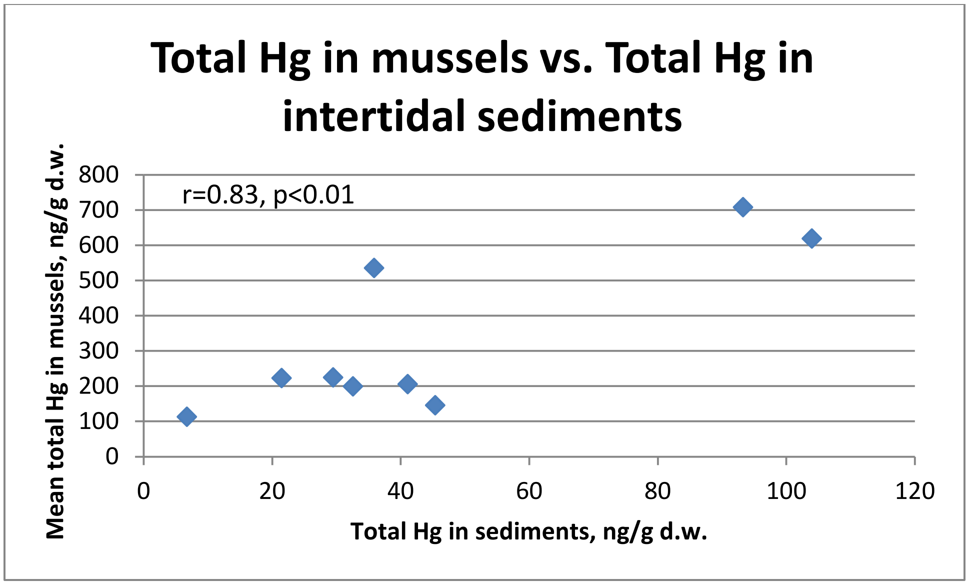

Figure 17-5. Relationship between total Hg in mussels and total Hg in surface (0-3 cm) sediments in the intertidal zone of 9 sites in 3 reference estuaries (St. George, Narraguagus, and Sheepscot), 2009.

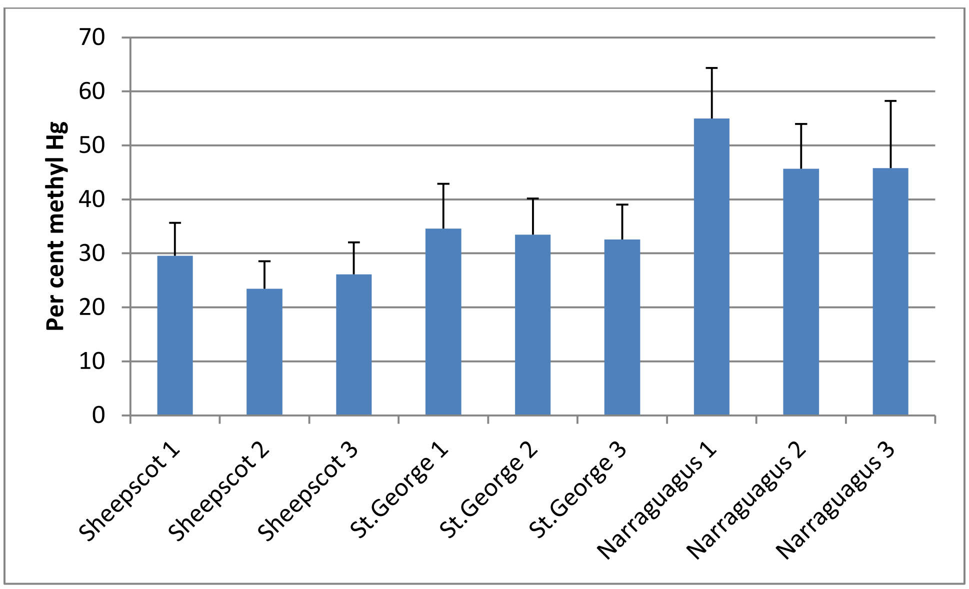

Figure 17-6. Percent methyl Hg in the soft tissue of mussels at three sites in each of three reference estuaries, 2009. See Appendix Table 17-3 for summary of raw data and sampling sites.

5 ACKNOWLEDGEMENTS

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6 REFERENCES

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APPENDIX 17-1:

Raw data for total Hg and methyl Hg in surface (0-3 cm) sediments in various reference (background) sites in the Penboscot and adjacent systems, Maine.

SITE	Site Latitude	Site Longitude	Date Sampled	Total Hg ng/g dry wt.	Methyl Hg ng/g dry wt.*
SC1-Subtidal	43 58 42.6	69 39 44.7	October 14, 2009	141.0	1.43
SC2-Subtidal	44 55 35.9	69 40 37.2	October 14, 2009	158.0	1.87
SC3-Subtidal	43 59 07.6	69 39 10.7	October 14, 2009	137.0	1.18
SG1-Subtidal	43 56 57	69 16 52	October 13, 2009	27.7	0.36
SG2-Subtidal	44 02 01	69 12 08	October 13, 2009	34.0	0.18
SG3-Subtidal	44 01 14	69 12 48	October 13, 2009	52.5	0.19
NG1-Subtidal	44 32 14	67 52 20	October 1, 2009	29.7	0.23
NG2-Subtidal	44 32 04	67 52 21	October 1, 2009	25.2	0.16
NG3-Subtidal	44 31 15	67 51 28	October 1, 2009	27.6	0.23
SC1-Intertidal	43 55 36.2	69 40 40.0	October 14, 2009	35.9	0.37
SC2-Intertidal	43 56 39.3	69 40 13,9	October 14, 2009	93.3	1.70
SC3-Intertidal	43 58 15.5	69 79 41.0	October 14, 2009	104.0	1.27
SG1-Intertidal	44 01 24.6	69 12 48.5	October 13, 2009	6.8	0.21
SG2-Intertidal	44 01 45.6	69 12 33.1	October 13, 2009	45.4	0.29
SG3-Intertidal	44 02 55.5	69 11 45.8	October 13, 2009	41.1	0.33
NG1-Intertidal	44 32 10.8	67 52 09.6	October 1, 2009	29.5	0.28
NG2-Intertidal	44 31 56	67 52 17.4	October 1, 2009	32.6	0.23
NG3-Intertidal	44 31 42	67 51 34.5	October 1, 2009	21.5	0.46
E05-01	44.11487	69.05456	August 22, 2007	61.8	0.15
E05-02	44.11474	69.02993	August 22, 2007	60.3	0.17
E05-03	44.11441	68.99982	August 22, 2007	89.5	0.17
E05-04	44.11593	68.97286	August 22, 2007	50.6	0.09

SITE	Site Latitude	Site Longitude	Date Sampled	Total Hg ng/g dry wt.	Methyl Hg ng/g dry wt.*
E05-05	44.11391	68.92167	August 22, 2007	68.4	0.10
E05-06	44.11424	68.77984	August 22, 2007	63.7	0.10
E05-07	44.11404	68.75247	August 22, 2007	45.9	0.12
E05-08	44.11494	68.7243	August 22, 2007	14.2	0.06
E05-10	44.11748	68.67036	August 22, 2007	45.1	0.11
E05-11	44.1146	68.63982	August 22, 2007	40.6	0.16
E05-12	44.11406	68.6141	August 22, 2007	23.8	0.09
E05-13	44.11327	68.58661	August 22, 2007	35.5	0.10
E08-01	43.94913	69.28264	August 27, 2007	49.3	0.21
E08-02	43.9736	69.26436	August 27, 2007	52.9	0.16
E08-03	44.00802	69.22604	August 27, 2007	53.9	0.14
E08-04	44.03767	69.19842	August 27, 2007	55.3	0.16
E08-05	44.06709	69.18121	August 27, 2007	45.9	0.22
EB-1	45.64917	-68.55359	I II III IV V VI 2006-2007	23.2	0.47
EB-2	45.66003	-68.5602	I II III IV V VI 2006-2007	20.0	0.89
EB-3	45.65209	-68.55207	I II III IV V VI 2006-2007	27.8	0.75
EB-4	45.62107	-68.5375	I II III IV V VI 2006-2007	71.0	1.33
EB-5	45.6652	-68.56596	I II III IV V VI 2006-2007	36.7	1.22
OV-1	44.8564	-68.67973	I II III IV V VI 2006-2007	24.3	0.89
OV-2	44.83901	-68.70116	I II III IV V VI 2006-2007	65.9	1.41
OV-3	44.8484	-68.69355	I II III IV V VI 2006-2007	51.2	1.72
OV-4	44.8776	-68.6728	I II III IV V VI 2006-2007	252.1	4.10
OV-5	44.84783	-68.69498	I II III IV V VI 2006-2007	55.9	0.85
D-01 55-60	44.48233	68.8087	August 20, 2007	19.1	n.d.
D-01 60-65	44.48233	68.8087	August 20, 2007	18.7	n.d.
D-01 65-70	44.48233	68.8087	August 20, 2007	18.3	n.d.
D-01 70-75	44.48233	68.8087	August 20, 2007	19.4	n.d.

SC=Sheepscot; NG=Narraguagus; SG=St. George

*Methyl Hg concentrations from sampling periods I, II, III, and IV (2006) for EB and OV sites were adjusted to the equivalent for results obtained from the distillation method. Concentrations from the extraction method were multiplied by two. See Phase I Update Report and the QA/QC chapter of this report.

APPENDIX 17-2:

Summary of size data for blue mussels collected at intertidal reference sites, 2009.

Site	n	Date Sampled	Range wet wwt. g	Mean wt. g	SD wt.
SC1	20	October 14, 2009	1.65-13.09	4.65	2.64
SC2	20	October 14, 2009	1.48-8.16	4.77	2.01
SC3	20	October 14, 2009	2.34-16.47	5.45	3.16
SG1	20	October 13, 2009	2.60-9.66	5.39	1.98
SG2	20	October 13, 2009	3.48-11.08	6.62	2.26
SG3	20	October 13, 2009	2.50-8.38	4.80	1.62
NG1	20	October 1, 2009	0.48-2.12	1.25	0.48
NG2	20	October 1, 2009	0.84-6.01	2.24	1.28
NG3	20	October 1, 2009	1.12-7.11	3.14	1.57

SC=Sheepscot; SG=St. George; NG=Narraguagus. Weight refers to fresh weight of soft tissue of individual mussels.

See Appendix Table 17-1 for geographic coordinates of intertidal sampling sites where mussels were collected.

APPENDIX 17-3:

Summary of raw data for total Hg and methyl Hg for blue mussels sampled at reference sites in 2009.

Site	Range total Hg ng/g dry wt.	Mean total Hg ng/g dry wt.	SD total Hg	Range methyl Hg ng/g dry wt.	Mean methyl Hg ng/g dry wt.	SD methyl Hg
SC1	406-758	539.79	100.01	109-192	153.83	22.11
SC2	396-1340	714.50	195.67	113-211	157.85	24.26
SC3	343-1070	618.80	178.47	112-229	154.15	28.79
SG1	64.1-156	112.82	25.72	17.7-55.6	38.00	9.74
SG2	109-214	145.63	29.39	33.1-63.8	47.64	8.74
SG3	122-419	205.63	66.71	42.0-117	65.09	18.06
NG1	174-367	224.83	50.72	94-189	121.39	23.72
NG2	111-258	198.66	61.82	58.9-135	87.84	19.81
NG3	133-325	222.53	51.84	57.2-136	98.67	21.99

See Appendix Table 17-2 for sample sizes, sampling dates and data on fresh weight of mussels.