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Report to the Joint Standing Committee on Environment  
and Natural Resources  
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# Surface Water Ambient Toxics Monitoring Program 2013

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## Introduction

This 2013 Surface Water Ambient Toxic (SWAT) monitoring program final report is organized into an Executive Summary, introduction and 3 modules;

1. Marine and Estuarine,
2. Lakes,
3. Rivers and Streams.

The full report is available on DEP's website at

<http://www.maine.gov/dep/water/monitoring/toxics/swat/index.htm>

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Legislators: Senator Thomas Saviello, Energy and Natural Resources; Representative Windol Weaver, Marine Resources

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Collection of samples was conducted by the principal investigators and technical assistants listed (DEP staff unless otherwise specified). Chemical analyses were performed by AXYS Analytical Services, Sidney, British Columbia or other laboratories as listed in reports in individual sections.

The statutory deadline for this report is March 31 of each year. Due to the highly technical nature of the information collected and analyzed, it has never been possible to finalize the report by that deadline. Several factors resulted in the 2013 report being released in July. The most prominent factor was an equipment malfunction at one of the contracted labs which caused a ten week delay in analyses, resulting in some analytical data not being delivered to the Department until June 20<sup>th</sup>. A draft of the 2013 report was made available to the Technical Advisory Group at their July 2<sup>nd</sup>, 2014 meeting.

## Executive Summary

Maine's Surface Water Ambient Toxics (SWAT) monitoring program was established in 1993 (38 MRSA §420-B) and administered by the Department of Environmental Protection to determine the nature, scope and severity of toxic contamination in the surface waters and fisheries of the State. The authorizing statute states that program must be designed to comprehensively monitor the lakes, rivers and streams, and marine and estuarine waters of the State on an ongoing basis. The program must incorporate testing for suspected toxic contamination in biological tissue and sediment, may include testing of the water column and must include biomonitoring and the monitoring of the health of individual organisms that may serve as indicators of toxic contamination. This program must collect data sufficient to support assessment of the risks to human and ecological health posed by the direct and indirect discharge of toxic contaminants.

The Commissioner of the Department of Environmental Protection (DEP) must prepare a five-year conceptual work plan in addition to annual work plans which are each reviewed by a Technical Advisory Group (TAG). The TAG is composed of 10 individuals, made up of 2 each with scientific backgrounds representing five various interests (business, municipal, conservation, public health and academic) and 2 legislators.

The SWAT program is divided into four modules, 1) Marine and Estuarine, 2) Lakes, 3) Rivers and Streams, and 4) Special Studies. This annual report follows the goals of the 2009 five-year conceptual plan which are generally to continue to monitor previously identified and new toxic issues in the marine environment, lakes and ponds, and rivers and streams, including, among others, providing baseline data for use by the Department of Marine Resources for shellfish harvesting areas, providing fish and shellfish contaminants data to the Maine Center for Disease Control and Prevention (MCDC) for use in revising Maine's fish consumption advisories, and continuing biological assessment of rivers and streams attainment of Maine's Water Quality Standards.

This report more specifically presents the findings of the 2013 annual work plan recommended by the SWAT TAG in a meeting June 18, 2013. The 2013 work plan focused on monitoring of the first three modules only, including shellfish in known or suspected contaminated marine areas, freshwater fish for mercury in lakes, dioxins and PCBs in rivers and streams as requested by MCDC, biomonitoring of aquatic life in the Kennebec River basin, and monitoring of potentially toxic metals in Androscoggin River sediments. Following is a summary of key findings from the 2012 SWAT program for each of the three modules monitored this year.

### 1. MARINE AND ESTUARINE

#### General Approach:

- In 2013, blue mussel tissue from East End Beach, Portland, and Sears Island, Searsport, was analyzed for contaminants including metals, mercury, Polycyclic Aromatic Hydrocarbons (PAHs), Polychlorinated Biphenyls (PCBs), and Perfluorinated Compounds

(PFCs). In 2013, tissue from six additional blue mussel sites in the Rockland harbor area (Rockland and Owls Head) was analyzed for PCBs only.

- In 2013, softshell clam tissue from Mast Cove, Eliot, and Presumpscot River, Falmouth/Portland, was tested and reported with historical data from eight additional softshell clam sites sampled in 2004-05 and 2010-12. Clam tissue was analyzed for contaminants including metals, mercury, PAHs, PCBs, and PFCs in 2013.

#### Encouraging Results:

- PAH concentrations in mussel and clam tissues did not exceed the National Status and Trends (NS&T Musselwatch) nationwide 85<sup>th</sup> percentile at three of four sites tested, and those three were not considered to be elevated. PAH levels in Maine shellfish tend to be low when compared to the national average.
- PCB concentrations in mussel and clam tissues did not exceed the national (NS&T Musselwatch) 85<sup>th</sup> percentile at any site and were not considered to be elevated. PCB concentrations in mussel tissue and clam tissue from all but one site tested were below the Maine Center for Disease Control's (MCDC) cancer fish tissue action level (FTAL), indicating shellfish from all but one site remained safe for human consumption with regard to PCBs.
- Concentrations of 12 individual PFCs were below detection limits at one of two blue mussel sites and both of the two clam sites tested in 2013. Testing for PFCs, an emerging contaminant of concern, was new to the marine SWAT program in 2013.

#### Contaminants and Areas to Watch:

- Lead in mussel tissue exceeded the national (NS&T Musselwatch) 85<sup>th</sup> percentile concentration at two sites in 2013, East End Beach, Portland, and Rockland, resulting in these sites receiving an "elevated" designation. Of six spatial subsamples composing the mean lead concentration for Rockland, two of six geographic subsamples had a lead concentration which exceeded the MCDC's FTAL for lead in finfish. Lead in clam tissue in 2013 at Mast Cove, Eliot, and Presumpscot River, Falmouth/Portland, exceeded the MCDC FTAL for lead in finfish. These two sites, along with four clam sites sampled in previous years, are considered problematic for human shellfish consumption based on these lead concentrations.
- Mercury in mussel tissue exceeded the NS&T Musselwatch 85<sup>th</sup> percentile concentration at two of three sites tested in 2013, which resulted in assignment of an "elevated" classification. Mercury levels in all 2013 mussel and clam tissue were below the MCDC methylmercury developmental FTAL for finfish, indicating shellfish remained safe for human consumption with regard to mercury.

- PAH concentration in clam tissue from Presumpscot River was slightly above the national (NS&T Musselwatch) 85<sup>th</sup> percentile concentration, although in the lower end of the NS&T national mid-range classification of PAH in shellfish.
- The PFC perfluorooctane sulfonamide (PFOSA) was detected in all four spatial sub-samples of mussel tissue at East End Beach, Portland, while the 11 other PFCs for which testing was performed were all below the detection limit for those compounds.

## 2. LAKES

- Comparison of mercury concentrations in 23 inland lakes with historical data from the same lakes from the 1990s showed that, in 2013, concentrations increased in 4 lakes, decreased in 4 lakes and remained similar in 12 lakes. (There was no determination for 3 lakes due to limited historical data.) Aggregated data from 66 lakes collected from 2010 to 2013 show no clear regional or statewide trends either. The data were sent to the Maine Center for Disease Control and Prevention (ME-CDC) for use in reviewing the statewide Fish Consumption advisory.

## 3. RIVERS AND STREAMS

### Encouraging Results:

- Thirty-nine stations were assessed for the condition of the benthic macroinvertebrate community. Thirty of these thirty-nine stations attained the aquatic life standards of their assigned class.
- Dioxin concentrations in fish from many river stations continue to decline from previous levels. Although concentrations still exceed the Maine Center for Disease Control and Prevention's (ME-CDC) Fish Tissue Action Level (FTAL) for dioxin alone at many stations, concentrations are below a level that would require river specific fish consumption advisories more stringent than the statewide fish consumption advisory due to mercury. These results are currently being reviewed by ME-CDC for possible revision of the current river specific fish consumption advisories. Dioxin concentrations measured in white sucker from Gilead, Rumford, Gulf Island Pond at Auburn, and Lisbon on the Androscoggin River are slightly above the FTAL but lower than previous concentrations in white sucker from the same stations (to a lesser extent at Lisbon). Dioxin concentrations in white sucker from Kennebec River at Sidney are below the FTAL similar to those of recent years and show a declining trend. Dioxin concentrations in white sucker from the Penobscot River at Milford (Costigan) and in American eel at Orrington are below the FTAL and lower than in previous years, but the trend is less defined for the eels at Orrington. Dioxin concentrations in white sucker from the Salmon Falls River at South Berwick exceeded the FTAL but show a declining trend.

- Coplanar (dioxin-like) PCB concentrations in fish were lower in 2013 than in the 1990's at all stations sampled for dioxins. Coplanar PCBs added to dioxins resulted in an exceedance of the FTAL in white sucker from the Kennebec River at Augusta and in American eel from the Penobscot River at Orrington. Although coplanar PCB concentrations add to the exceedance of the dioxin FTAL caused by dioxin concentrations at the other stations, dioxin and coplanar PCB concentrations combined do not exceed a level that would require river specific fish consumption advisories more stringent than the statewide fish consumption advisory due to mercury at any station sampled in 2013.

#### Contaminants and Areas to Watch:

- Total PCBs exceeded the FTAL in fish from all same stations on the Androscoggin, Kennebec, Penobscot, and Salmon Falls rivers, except for white sucker from Milford on the Penobscot River which had concentrations below the FTAL. Total PCB concentrations were generally a little higher in white sucker from the Androscoggin River at Gilead, but similar in white sucker from Rumford, Gulf Island Pond at Auburn, and Lisbon compared to previous years. Total PCB concentration in white sucker from the Kennebec River at Augusta and Salmon Falls River at South Berwick were lower than in previous years, but levels in American eel from the Penobscot River at Orrington were intermediate those from previous years.

#### 4. SPECIAL STUDIES

##### *SPL (Soft Plastic Lures) Study*

This is a research study of the presence and bioaccumulation in fish of toxic plasticizers (such as phthalates, BPA) in soft plastic lures (SPLs) used by anglers. The project is divided into three tasks, including 1) a plasticizer identification study, 2) a bioavailability study, and 3) fish tissue analysis, all of which may require development of suitable methods. The study has just been initiated and preliminary results of task 1 document the presence of multiple phthalates and other organic constituents that differ among brands of SPLs that require more investigation.



# 1.0 MARINE MODULE

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## 1.1 INTRODUCTION

Maine's coastline lies within, and lends its name to, the Gulf of Maine, a diverse and productive ecosystem. The Maine coast and the larger Gulf of Maine provide economic opportunities including commercial fisheries, aquaculture, recreational fisheries, commerce via shipping, and a wide variety of tourism activities. Maine includes the urbanized areas of Portland and Bangor, and has experienced growth and increased development, especially in the southwestern portion of the state's coastline in recent years. With increased development, increases in chemical contaminants discharged to the marine environment may occur. Some contaminants can also become magnified as they move through the food chain, bioaccumulating at higher trophic levels and potentially causing impacts on the viability of marine species and ecosystem health, and causing concern about consequences to human health. All these reasons suggest that the monitoring of chemical contaminants is an important component of assessing the health of our marine environment in Maine.

### 1.1.1 Blue Mussels

Blue mussels (*Mytilus edulis*) have been used extensively by the SWAT program (since 1986) and other monitoring programs as an indicator of exposure of marine environments to chemical pollutants. Mussels are ubiquitous and readily collected across the coast of Maine, as well as across the entire Gulf of Maine. Published information about contaminants in mussels provides some historical context and allows comparisons between geographic areas and over time. Since blue mussels are consumed as food by humans, they can be used to understand potential human exposure to contaminants. Mussels are sessile, allowing attribution of their contaminant burdens to the environment where they were collected. Mussels filter large volumes of water as they feed, allowing them to concentrate many chemicals from the water column or sediments suspended in the water column. This allows detection of contaminants in mussel tissue that are sometimes found below detection limits in particulate matter, sediment, or water. Use of mussels also provides insight into the biologically available portion of contaminants, which may not readily be discerned from background sediment or water concentrations.

This report presents and summarizes contaminant data from the collection and analysis of blue mussel tissue collected in 2013 from three sites along the Maine coast. All mussel tissue samples were analyzed for heavy metals (including mercury) and polychlorinated biphenyls (PCBs), and a subset of two sites were analyzed for polycyclic aromatic hydrocarbons (PAHs) and perfluorinated compounds (PFCs). In order to provide comparability of results from these 2013 samples, blue mussel contaminant levels from the SWAT program are compared to blue mussel contaminant levels in other programs including the Gulfwatch program ("Gulfwatch": Gulf of Maine Council on the Marine Environment) and the National Status & Trends Mussel Watch Program ("NS&T": National Oceanographic and Atmospheric Administration). This analysis provides a regional and national context to the Maine SWAT data.

### 1.1.2 Softshell Clams

Like blue mussels, softshell clams (*Mya arenaria*) are consumed as food by humans and can be used to understand potential human exposure to contaminants. Clams are sessile, allowing attribution of their contaminant burdens to the environment where they were collected. Like mussels, clams filter large volumes of water as they feed, allowing them to concentrate many chemicals from the water column or sediments suspended in the water column. Softshell clam stations sampled by the SWAT program in recent years have been selected to characterize contaminant concentrations specifically in clam tissue, as opposed to blue mussel tissue which may or may not have been sampled previously in the same general area. Gulfwatch and SWAT softshell clam tissue contaminant data suggest that clams may have very different concentrations of some contaminants than blue mussel tissue taken from the same stations. This is an important point when considering the contaminant concentrations that humans are exposed to when consuming clams. Clam site selection for testing is typically driven by human consumption and exposure, and clams are used less in SWAT (or Gulfwatch) as a general environmental monitor or sentinel like the blue mussel.

This report presents and summarizes contaminant data from the collection and analysis of softshell clam tissue collected in 2013 from two sites on the Maine coast. Also presented are softshell clam contaminant data from ten additional sites sampled in 2004-05 and in 2010-12 by the SWAT program. Softshell clam tissue samples were analyzed for metals, mercury, PAHs, PCBs and PFCs. In order to provide comparability of results from the 2010-13 and 2004-05 samples, softshell clam contaminant concentrations from SWAT sampling are compared to contaminant concentrations from the Gulfwatch program to provide regional context.

The Maine Dept. of Marine Resources (Maine DMR) has asked Maine DEP to sample clams in areas currently closed to shellfish harvest, which usually is due to bacterial contamination that prevents safe consumption of the clams by humans. Some significant clam resources demonstrate improving bacterial indicator counts or may be candidates for additional work to reduce bacterial contamination in the vicinity of the resource. Without corresponding contaminant data from clam tissue to document safe human consumption, expenditure of resources to reduce bacterial contaminant sources might be premature if high contaminant concentrations are confirmed. Bacterial source clean up can then be targeted to clam resources that already have been documented as safe for human consumption from a contaminant concentration perspective. Like mussels, testing sites with low contaminant levels, which can only be determined post-sampling, still provides valuable data on background contaminant levels in clams and provides a context with which to compare more heavily contaminated sites.

## 1.2 METHODS

Sites sampled in recent years within the context of this program can be divided into three types based on the goals outlined above that drive the need for information. These types are: Spatial, Temporal, and Follow-Up sites. Sites that have never been sampled (or that have not been sampled for eight or more years), have been sampled for only one analyte

type, or have been sampled with no replication are classified as “Spatial” sites. The primary reason for sampling these sites is to provide data required to fill geographic gaps. Spatial sites enable a more complete picture of how contaminants vary across the Maine coastline, and provide screening data that can be used in assessing interest on testing these sites again in the future. Testing sites with low contaminant levels, which can only be determined post-sampling, still provides valuable data on background contaminant levels and provides a context with which to compare more heavily contaminated sites.

“Temporal” sites are locations where there is an interest in obtaining data to assess contaminant levels through time. These sites will be sampled on an accelerated schedule, with sampling occurring as often as biennially. More frequent data collection will provide more closely spaced data through time, which may permit trend analysis when sufficient data are acquired. Relatively few temporal sites will be sampled to minimize costs associated with repeated, higher frequency sampling.

“Follow-up” sites are those where previous SWAT contaminant levels (or results from another program like Gulfwatch) at the site or nearby indicate that additional sampling and analysis are warranted. Repeat sampling may occur at the same location in an attempt to confirm earlier results, or sampling of additional nearby sites might be used to determine local contaminant distribution. Follow-up sites may also occur in the Temporal or Spatial categories as well based on their historical sampling and data needs.

Resampling in subsequent years at Temporal or Follow-up sites does not occur at exact sub-site replicate coordinates sampled previously, but varies somewhat due to distribution and quantity of mussels available in the target size range from year to year. The slight spatial variation in sub-site replicates sampled provides additional information regarding patchiness of contaminants, and arithmetic means across all four sub-site replicates are used to compare between years.

### **1.2.1 Blue Mussels**

Blue mussel samples have been analyzed from more than 90 distinct locations sampled over the past 28 years. Blue mussels were collected from three sites during mid-October, 2013. All three of the mussel sites had been sampled previously as part of the SWAT program and are shown in Table 1.2.1.1. Follow up sampling in 2013 in the Rockland area included six spatial subsamples rather than the usual four, more closely spaced replicates or subsamples, typically used to construct a mean concentration. The six subsamples were widely spaced to assess the variability of PCBs in Rockland and Owls Head in a cost effective manner, and were compared to previous data collected from five different locations in Rockland. Because of this wide spacing, each subsample is listed with its individual coordinates in the Table 1.2.1.1. A map of the blue mussel sampling locations is provided in Figure 1.2.1.1.

Methodology of field collection, morphometric measurement, and laboratory preparation of mussel samples has been provided in previous SWAT reports and in the Gulfwatch field manual (Sowles et al. 1997) and will be reviewed here to familiarize the reader with

the general approaches used. SWAT mussel sampling is planned and conducted to control as much variability in data collected as possible. Variation in mussel shell size, seasonal timing of collections subsequent to spawning, location within the intertidal zone, and sample location were all minimized to reduce conflicting signals in the contaminant data.

**TABLE 1.2.1.1: 2013 SWAT Blue Mussel Sites**

<u>Site Name</u>	<u>Municipality</u>	<u>Station Code</u>	<u>West Longitude</u>	<u>North Latitude</u>	<u>Date Sampled</u>	<u>Site Type</u>
East End Beach	Portland	CBEEEE	-70.24071	43.66977	10/9/2013	T
Jameson Pt.	Rockland	PBRKCP	-69.08404	44.11506	10/11/2013	F, S
Owls Head	Owls Head	PBRKCP	-69.0455	44.09274	10/15/2013	F, S
Ocean Pursuits Boatyard	Rockland	PBRKCP	-69.10107	44.11413	10/15/2013	F, S
Town Landing	Rockland	PBRKCP	-69.10545	44.09225	10/15/2013	F, S
North of Breakwater	Rockland	PBRKCP	-69.07719	44.12457	10/11/2013	F, S
Owls Head Harbor	Owls Head	PBRKCP	-69.05296	44.08393	10/15/2013	F, S
Sears Island	Searsport	PBSIWS	-68.88906	44.45093	10/10/2013	F, S

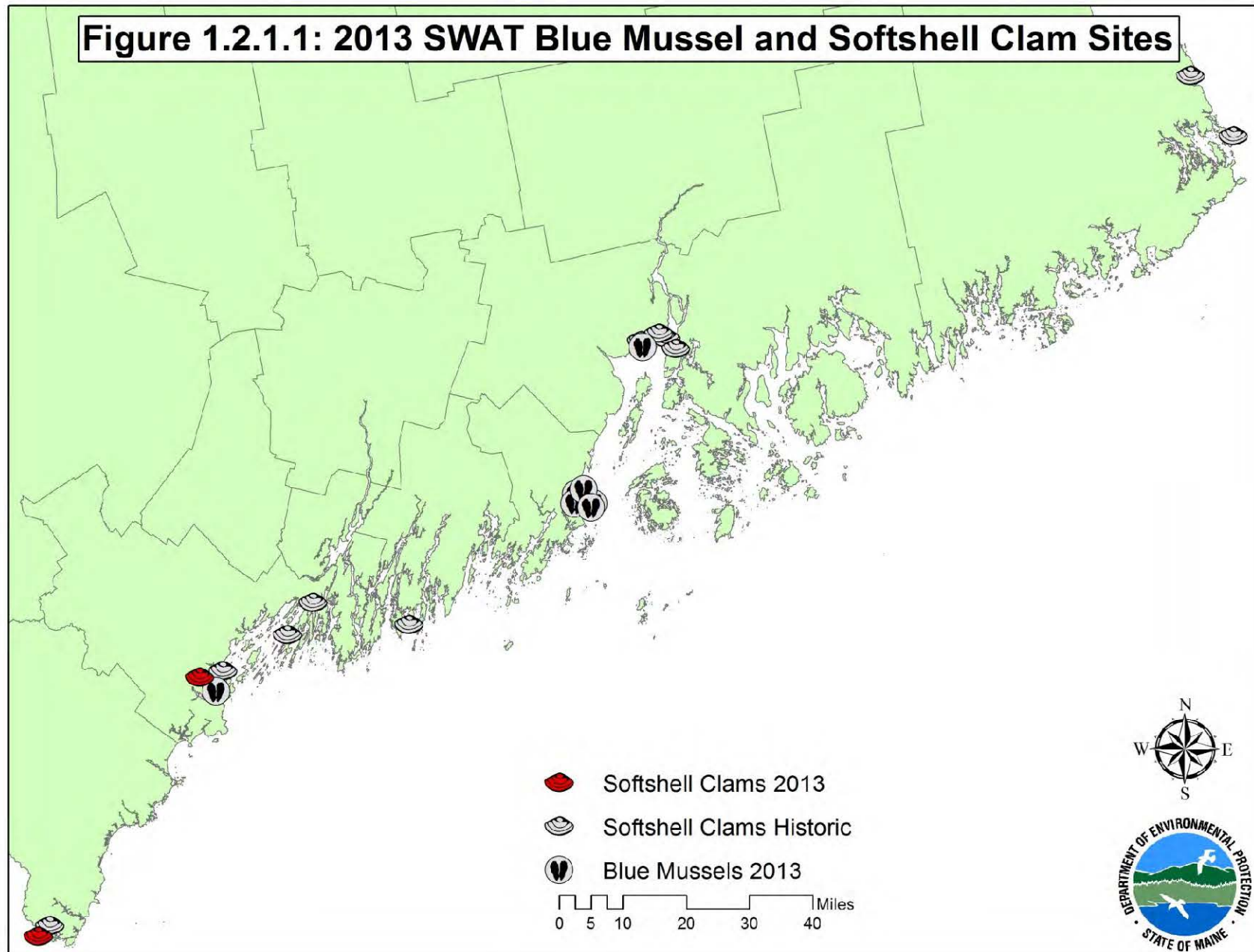
1 S = Spatial, T = Temporal, F = Follow Up

In order to characterize the contaminants present in a general area at the sampling site, mussels were collected along the shoreline from four distinct replicate areas (six for Rockland/Owls Head) whenever possible. Gauges were used to sort mussels by shell length in the field, and mussels within a size range of 50-60 mm were selected for analysis. For metals analysis, a minimum of 20 mussels were selected from within the target size range from each of the four intra-site locations (replicate) and placed in separate containers. For organics analysis including PAHs, PCBs, and PFCs, a minimum of 30 mussels were collected at each intra-site location. Replicates were washed in ambient sea water in a mesh or open bucket at the collection site to remove external debris and attached sediments. Mussel replicates were then transported to the laboratory in coolers (supplemented with ice packs in warmer weather). Mussels were not depurated prior to shucking to remove tissue for analysis.

Tissue sample processing was accomplished within 24 hours of field collections at all sites. At the laboratory, individual mussels were measured with calipers for length (anterior umbo to posterior growing edge) to the nearest 0.1 mm. Shell height and width (mm) and soft tissue wet weight (nearest 0.1 g) were also measured and recorded for ten mussels per replicate. All soft tissue was removed and combined with the soft tissue from mussels within the same replicate. Total soft tissue wet weights per replicate were recorded. Tissue composites were immediately placed in pre-cleaned glass jars and capped. Jars were pre-labeled and filled jars were stored at -5° C for up to two months until analysis.

Mussel tissues tested for PAHs, PCBs, and PFCs were analyzed by AXYS Analytical Services Ltd., Sidney, British Columbia. Mussel tissue tested for metals were analyzed by Pacific Northwest National Laboratory operated by Battelle, Sequim, Washington.

**Figure 1.2.1.1: 2013 SWAT Blue Mussel and Softshell Clam Sites**



### 1.2.2 Softshell Clams

Softshell clams were collected at two sites in 2013. These two locations were selected based on Maine DMR interest in potentially opening clam flats closed due to high bacterial levels. Both sites were sampled previously by SWAT, although one site was resampled on a wider geographic scale to include flats where much of the historic clam resource was thought to exist and the second was resampled to obtain newer data. In addition to the two softshell clam sites sampled in 2013, this report includes data from eight softshell clam sites sampled in 2010-12 and 2004-05 (Table 1.2.2.1; Figure 1.2.1.1). These data are included to provide a broader context for softshell clam contaminant concentrations across the state.

**TABLE 1.2.2.1: SWAT Softshell Clam Sites: 2004-05, 2010-13**

<u>Site Name</u>	<u>Municipality</u>	<u>Station Code</u>	<u>West Longitude</u>	<u>North Latitude</u>	<u>Date Sampled</u>	<u>Site Type<sup>1</sup></u>
Mast Cove	Eliot	PQMCMC	-70.8048	43.1210	11/9/2004	S
Mast Cove	Eliot	PQMCMC	-70.7981	43.1155	10/16/2013	S
Presumpscot R.	Falmouth/Portland	CBPRMT	-70.2460	43.6981	10/9/2012	S
Presumpscot R.	Falmouth/Portland	CBPRMT	-70.2543	43.6943	10/25/2013	S
Navy Pier	Harpswell	CBHWNP	-70.0136	43.7870	11/12/2004	S
Mare Brook	Brunswick	CBMBBH	-69.9334	43.8617	10/11/2012	S
Squirrel Island	Southport	MCBBSQ	-69.6290	43.8130	11/8/2004	S
Long Cove	Searsport	PBSTLC	-68.8938	44.4656	12/1/2005	S
Fort Point Cove	Stockton Springs	PBFPPF	-68.8150	44.4717	11/10/2005	S
Fort Point Cove	Stockton Springs	PBFPPF	-68.8372	44.4832	11/3/2011	F
Morse Cove	Penobscot/Castine	PBCAMC	-68.7835	44.4478	11/16/2010	S
Harris Cove	Eastport	PMHCHC	-66.9838	44.9171	11/9/2004	S
Mill Cove	Robbinston	PMSCMC	-67.1176	45.0580	11/29/2005	S

<sup>1</sup> S = Spatial, T = Temporal, F = Follow Up

Methodology of field collection, morphometric measurement, and laboratory preparation of mussel samples has been provided in previous SWAT reports and in the Gulfwatch field manual (Sowles et al. 1997) and any departures from that methodology in softshell clam sampling are noted in the following text. In order to characterize the contaminants present in a general area at the sampling station, softshell clams were collected from four distinct areas (replicates) along the shoreline at each site whenever possible. Clams at or above the commercial legal length of 2 inches (50.8 mm) were dug from each intra-site location. For metals analysis, a minimum of ten clams were selected from within the target size range from each of the four intra-site locations and placed in separate containers. For organics analysis, a minimum of 20 clams was collected at each intra-site location. Clams in these replicates were washed in ambient sea water in a mesh or open



bucket at the collection site to remove external debris and attached sediments. Clam replicates were then transported to the laboratory in coolers (supplemented with ice packs in warmer weather). Clams were not depurated prior to shucking to remove tissue for analysis.

Tissue sample processing was accomplished within 24 hours of field collections. At the laboratory, individual clams were measured with calipers for length (longest shell measurement perpendicular to a line extending from the umbo to the growing edge) to the nearest 0.1 mm. Shell height and width (mm) and soft tissue wet weight (nearest 0.1 g) were also measured and recorded for ten clams. All soft tissue was removed and combined with the soft tissue from the ten clams within the same replicate. Total soft tissue wet weights per ten clam replicate were recorded. For organics analysis (PAHs, PCBs, and PFCs), 10-20 clams were composited into a replicate to produce the requisite 100 grams of tissue required for the analyses.

Tissue composite samples for metals analyses included ten clams per replicate, and tissue composite samples for organics analyses included 10-20 clams per replicate. For both metals and organics, four replicates were collected per sampling station. Tissue composites were immediately placed in pre-cleaned glass jars and capped. Jars were pre-labeled and filled jars were stored at  $-5^{\circ}$  C for up to two months until analyses could be completed. Softshell clam tissues tested for PAHs, PCBs, PFCs, and organochlorinated pesticides in 2010-13 were analyzed by AXYS Analytical Services Ltd., Sidney, British Columbia, while clam tissues tested for metals in these same years were analyzed by Pacific Northwest National Laboratory operated by Battelle, Sequim, Washington. Clam tissues tested in 2004-05 for both the metals and organic contaminants were analyzed by Pace Analytical, Minneapolis, MN.

## **1.3 RESULTS AND DISCUSSION**

### **1.3.1 Metals**

#### **1.3.1.1 Blue Mussels**

Mussel tissue samples collected in 2013 were analyzed for 11 metals: Silver (Ag), aluminum (Al), arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), lead (Pb), mercury (Hg), nickel (Ni), and zinc (Zn). Results were compared to national NS&T (Kimbrough et al. 2008) and Gulf of Maine (Gulfwatch)(LeBlanc et al. 2009) blue mussel monitoring program data (collected through 2008, the most recent available) to place Maine SWAT data in a broader geographic context. From an environmental monitoring perspective, the concentration of an analyte in SWAT mussel tissue was considered elevated when that concentration exceeded the NS&T 85<sup>th</sup> percentile. This approach is consistent with the Gulfwatch program (LeBlanc et al. 2009).

##### **1.3.1.1.1 Silver (Ag)**

Silver was detected at all three sample locations visited in 2013 (Figure 1.3.1.1.1). Silver measured in mussels ranged from a low mean concentration of 0.041 ug/g dry wt.

at East End Beach, Portland, to a high mean concentration of 0.051 ug/g dry wt. at Rockland. Silver mean concentrations in 2013 SWAT mussels were also compared to the Gulfwatch median and 85<sup>th</sup> percentile concentrations. The mean concentration at all three sites exceeded the Gulfwatch median (0.037 ug/g dry wt.). None of the three mean concentrations exceeded the Gulfwatch 85<sup>th</sup> percentile (0.073 ug/g dry wt., Figure 1.3.1.1.1.1). Figure 1.3.1.1.1.2 compares the silver concentrations in 2013 SWAT blue mussel tissue to the NS&T median and 85<sup>th</sup> percentile. No tissue silver concentrations exceeded the NS&T median or 85<sup>th</sup> percentile, hence no sites were considered elevated for silver.

Higher silver concentrations in water and sediments coincide with municipal sewage discharge (Sanudo-Wilhelmy and Flegal 1992; Buchholtz ten Brink et al. 1997). The increasing use of silver, including nanosilver, in products like paints, caulking, and clothing makes monitoring silver of interest at present and in the future. Overall, silver concentrations in Maine mussels at sampled locations appear to be relatively low. The highest Gulfwatch values, which came from sites in Neponset River and Sandwich, Massachusetts, exceeded the NS&T median but fell short of the NS&T 85<sup>th</sup> percentile.

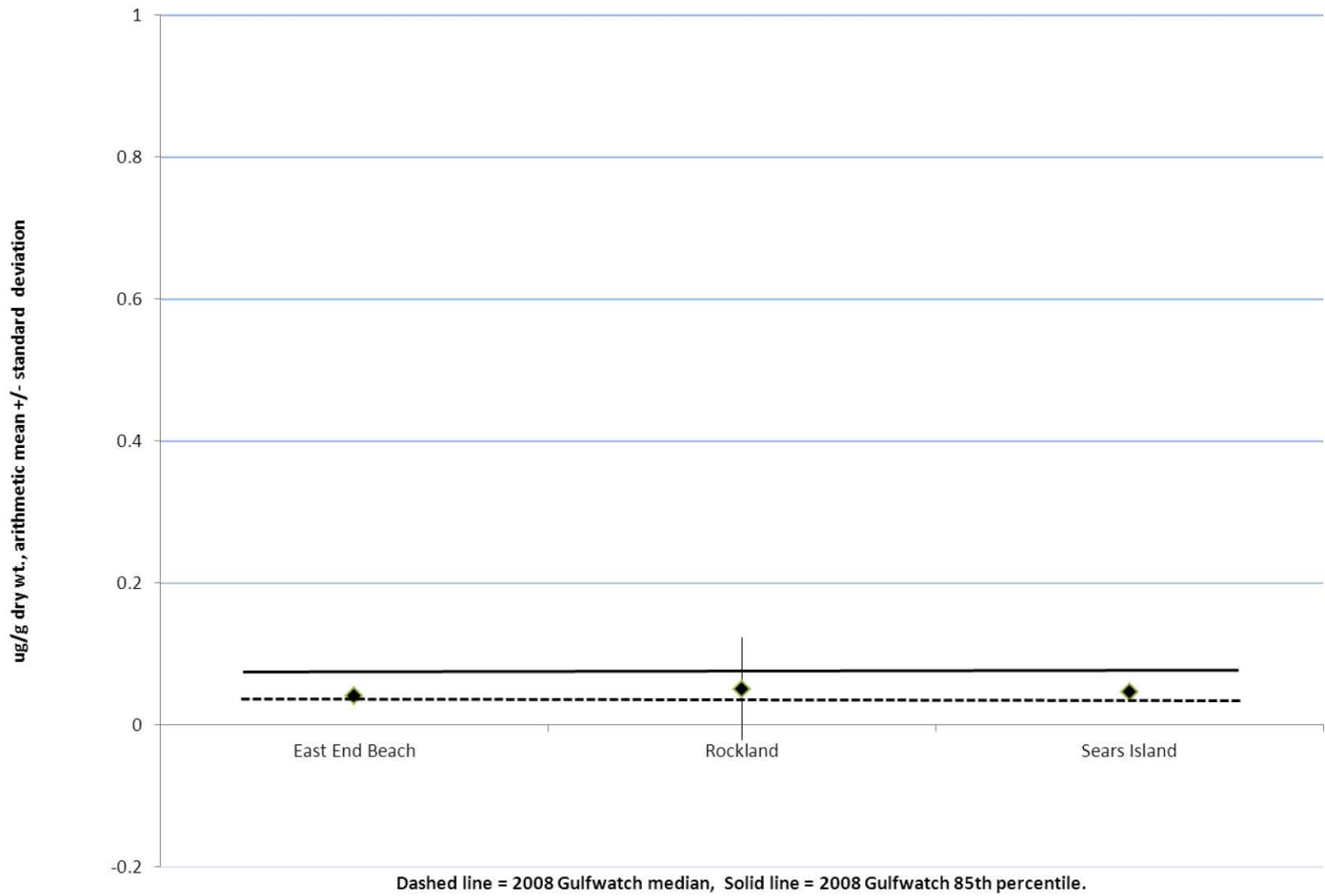
The Maine Center for Disease Control, Bureau of Health (MCDC) silver non-cancer fish tissue action level (FTAL) is 11 ug/g wet wt. (ppm) for non-commercially caught fish. The highest 2013 SWAT blue mussel tissue mean silver concentration, when expressed on a wet weight basis, is 0.011 ug/g wet wt. at Sears Island, Searsport. This concentration is three orders of magnitude below the 11 ug/g wet wt. FTAL.

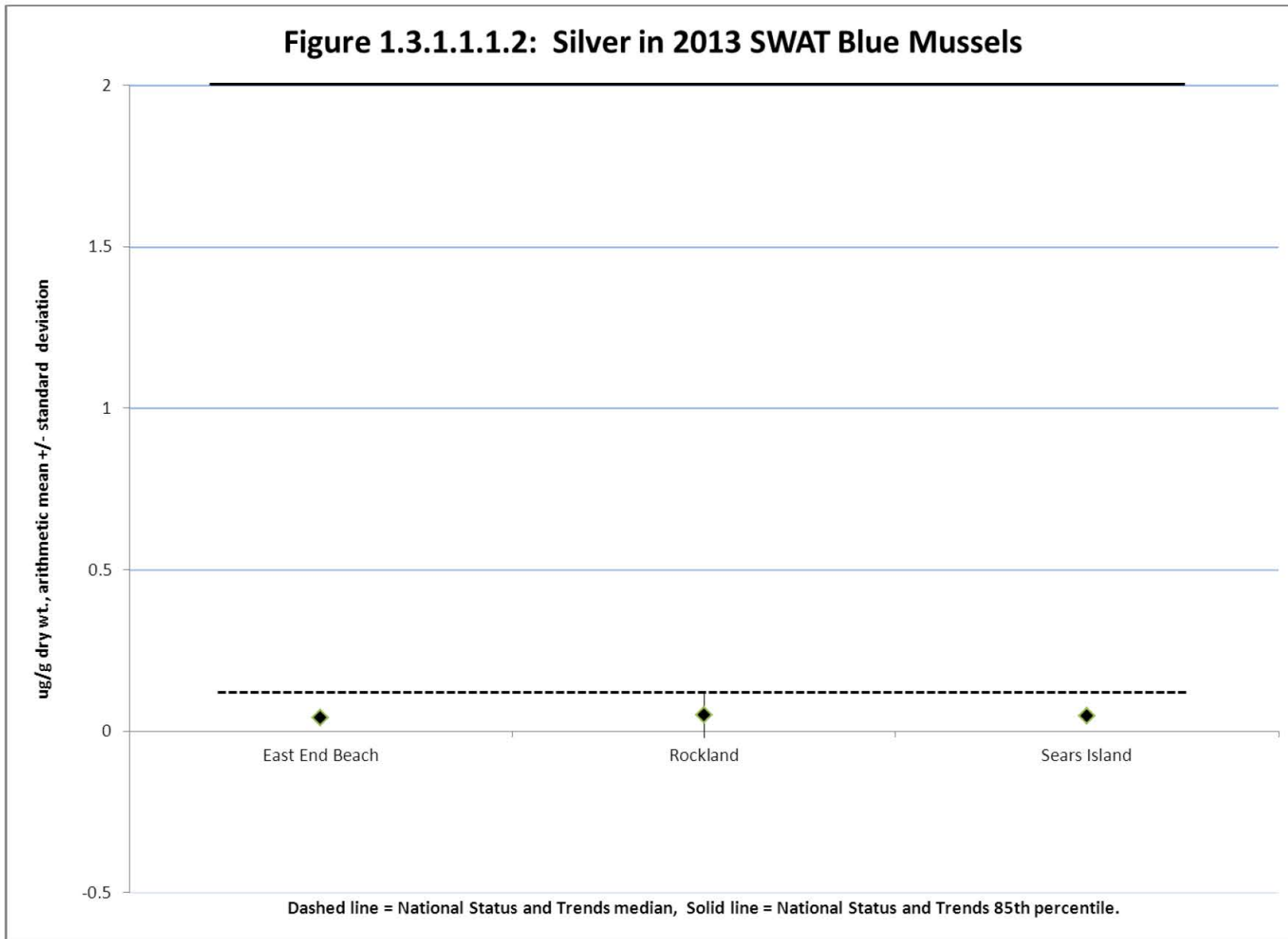
#### **1.3.1.1.2 Arsenic (As)**

Arsenic was detected at all three sample locations visited in 2013 (Figure 1.3.1.1.2.1). Arsenic levels measured in mussels ranged from a low mean concentration of 13.62 ug/g dry wt. at Sears Island, Searsport, to a high mean concentration of 18.37 ug/g dry wt. at Rockland. While Gulfwatch does not monitor arsenic concentrations, they are tracked regionally and nationally by NS&T. In blue mussels, NS&T considers 12-22 parts per million dry wt. (directly comparable to SWAT ug/g data) to be in the mid-range of three ranges of arsenic concentration nationally (Kimbrough et al. 2008). All three blue mussel sites had arsenic concentrations which fell into the mid-range of three NS&T ranges.

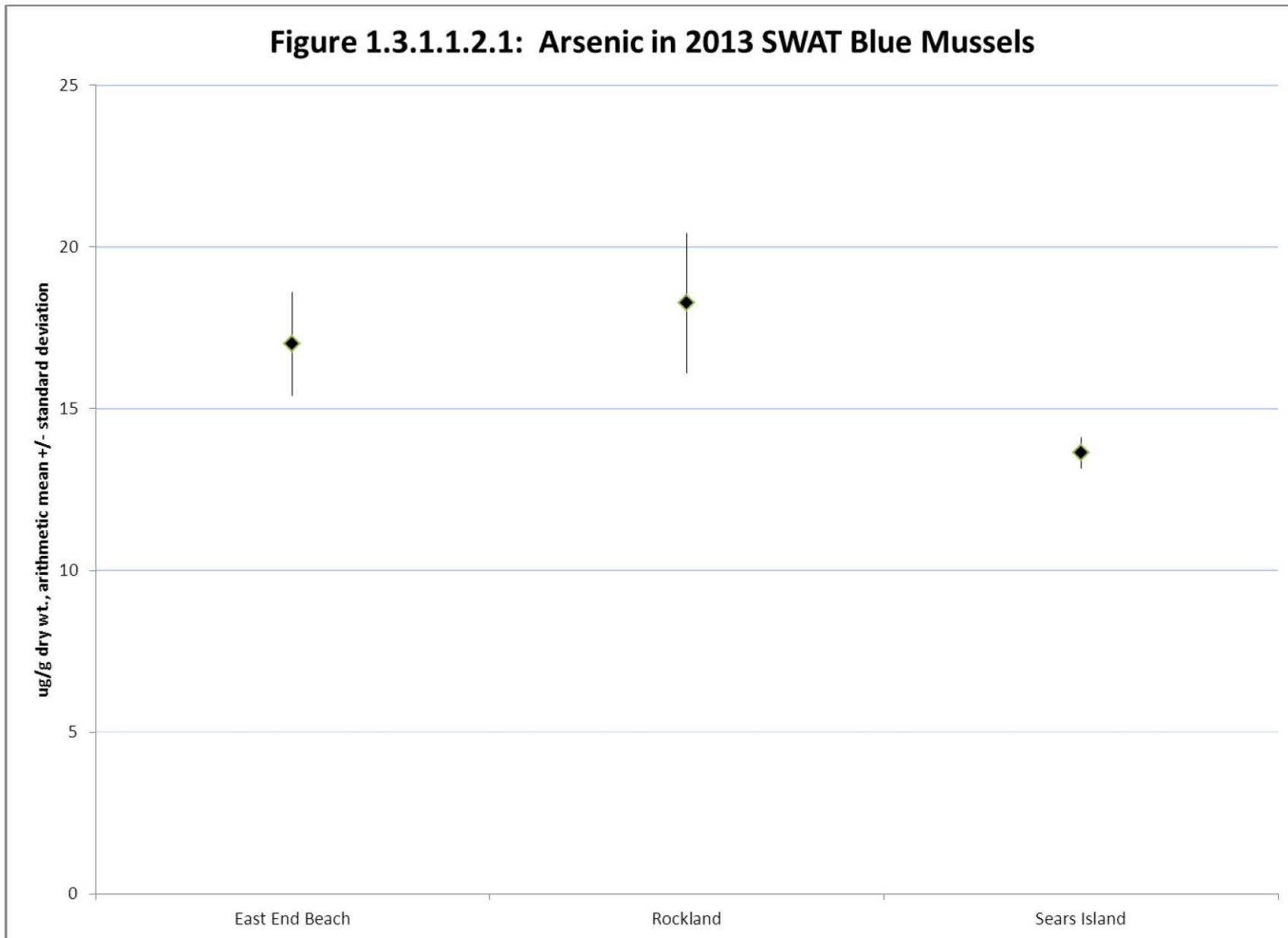
Nationally, the primary source for elevated levels of arsenic is crustal rock. Other than natural sources, industrial pollution can contribute arsenic to the environment from preserved wood, semiconductors, pesticides, defoliants, pigments, antifouling paints, and veterinary medicines. Atmospheric sources include smelting, fossil fuel combustion, power generation, and pesticide application (Kimbrough et al. 2008).

Figure 1.3.1.1.1: Silver in 2013 SWAT Blue Mussels





**Figure 1.3.1.1.2.1: Arsenic in 2013 SWAT Blue Mussels**



For non-commercially caught finfish, MCDC reports a cancer FTAL of 0.014 ppm and a non-cancer FTAL of 0.6 ppm, both for inorganic arsenic (the most toxic form). Most fish tissue data and the SWAT blue mussel tissue data are analyzed for total arsenic, not inorganic arsenic. MCDC uses FDA's 1993 assumption that 10% of total arsenic in finfish is inorganic arsenic. Using this assumption, approximate inorganic arsenic concentrations for SWAT blue mussels were calculated by dividing wet weight concentrations by a factor of 10. Therefore, 2013 SWAT blue mussel inorganic arsenic concentrations are estimated to range from 0.26 ug/g wet wt. to 0.31 ug/g wet wt. All three sites exceeded the MCDC cancer FTAL of 0.014 ug/g wet wt. (ppm).

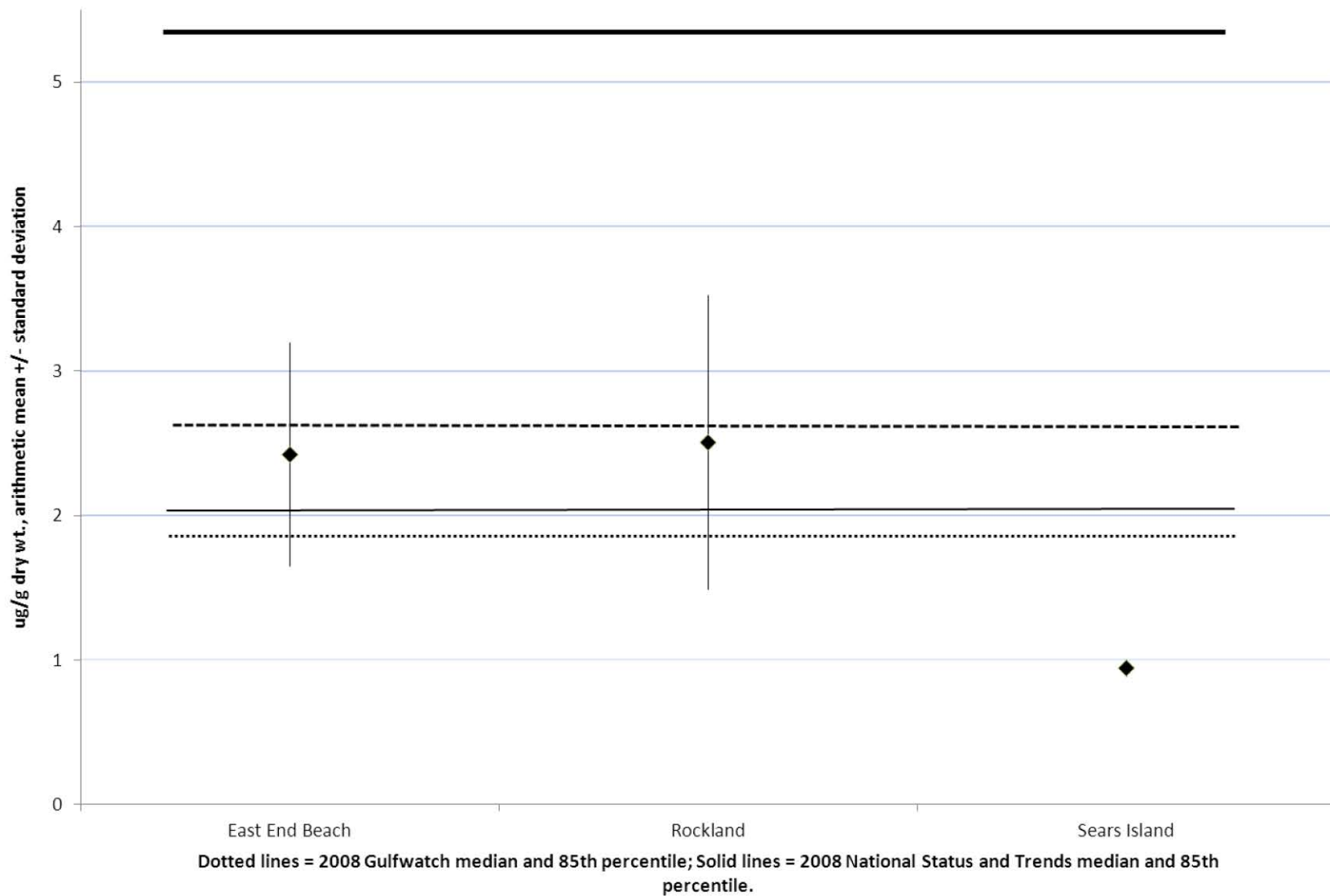
Comparing recent data from all 60 mussel sites sampled from 2007-13, calculated inorganic arsenic concentrations in SWAT blue mussel tissue ranged from a low of 0.11 ug/g wet wt. (Bar Harbor, 2007) to a high of 0.33 ug/g wet wt. (Turnip Island, Georgetown, 2012). All SWAT sites sampled from 2007-13 had calculated blue mussel tissue inorganic arsenic concentrations exceeding the MCDC cancer action level of 0.014 ug/g wet wt. (ppm). None of the three sites sampled in 2013 were calculated to have exceeded the MCDC non-cancer action level of 0.6 ug/g wet wt. (ppm) for inorganic arsenic. Similarly, none of the 57 mussel stations sampled from 2007-12 were calculated to have exceeded the MCDC non-cancer FTAL. The MCDC non-commercially caught finfish FTALs applied here assume an 8 oz. meal eaten by the consumer on a weekly basis. Maine SWAT data indicate that this 8 oz. meal size would translate to approximately 45-50 mussels per meal.

#### **1.3.1.1.3 Cadmium (Cd)**

Cadmium was detected at all three sample locations visited in 2013 (Figure 1.3.1.1.3.1). Cadmium levels measured in mussels ranged from a low mean concentration of 0.94 ug/g dry wt. at Sears Island, Searsport, to a high mean concentration of 2.51 ug/g dry wt. at Rockland. The cadmium concentration at Sears Island fell below the 2008 Gulfwatch median, with the concentrations at East End Beach and Rockland exceeding the Gulfwatch median but not the 85<sup>th</sup> percentile (Figure 1.3.1.1.3.1). Cadmium concentration at Sears Island fell below the 2008 NS&T median, with the concentrations at East End Beach and Rockland exceeding the NS&T median but not the 85<sup>th</sup> percentile (Figure 1.3.1.1.3.1) (Kimbrough et al. 2008). Since tissue cadmium concentrations did not exceed the NS&T 85<sup>th</sup> percentile, no sites were considered elevated for cadmium.

Cadmium originates from crustal elements as rocks weather and is transported seaward by rivers, which account for approximately half of worldwide cadmium sources. Cadmium is also released through forest fires and volcanic activity, with anthropogenic sources including manufacturing, fossil fuel combustion, and agriculture. Industrial sources include manufacture of batteries, plating, stabilizers, and nuclear power (Kimbrough et al. 2008).

**Figure 1.3.1.1.3.1: Cadmium in 2013 SWAT Blue Mussels**



From a human health perspective, the MCDC non-cancer FTAL for cadmium in non-commercially caught finfish is 2.2 ug/g wet wt. The FDA action level for clams, oysters, and mussels is 4 ppm wet wt. (Kimbrough et al. 2008). The highest scoring 2013 SWAT site, Rockland, had a mean cadmium concentration of 0.40 ug/g wet wt., which was below the MCDC and FDA action levels.

#### **1.3.1.1.4 Chromium (Cr)**

Chromium was detected at all three sites sampled in 2013 (Figure 1.3.1.1.4.1). Chromium levels measured in mussel tissue ranged from a low mean concentration of 0.57 ug/g dry wt. at Sears Island, Searsport, to a high mean concentration of 1.71 ug/g dry wt. at East End Beach, Portland. East End Beach exceeded the Gulfwatch median but not the 85<sup>th</sup> percentile, while the concentrations at the remaining two mussel sites did not exceed the Gulfwatch median (Figure 1.3.1.1.4.1). Figure 1.3.1.1.4.1 also depicts 2013 SWAT mussel chromium concentrations compared to the NS&T Mussel Watch median and 85<sup>th</sup> percentile concentrations. Two sites, East End Beach and Rockland, exceeded the NS&T median and no sites exceeded the NS&T national 85<sup>th</sup> percentile. No mussel sites were considered elevated for chromium.

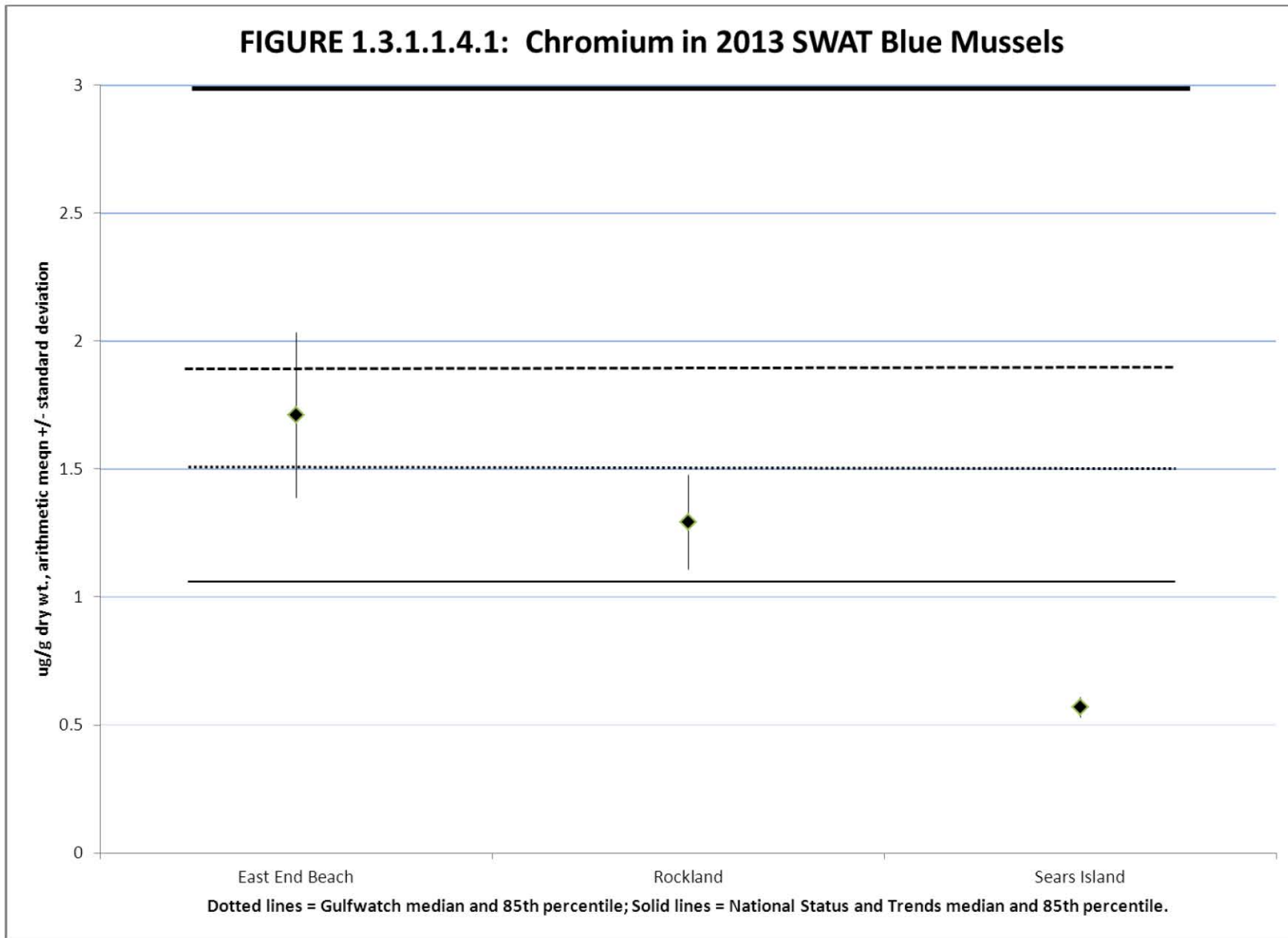
Natural sources of chromium include leaching from soil and rock into surface waters. Chromium is released from textile, electroplating, and leather tanning industries. Chromium is used extensively in tanning leather and was discharged with untreated tannery effluent during the last two centuries. Chromium persists in the marine environment in sediments near anthropogenic sources (Kimbrough et al. 2008).

From a human health perspective, the MCDC FTALs (7 ug/g cancer action level and 11 ug/g non-cancer action level) for chromium are based on chromium VI, and are not directly comparable to SWAT results, which measure total chromium (less toxic Cr III and more toxic Cr VI, combined).

#### **1.3.1.1.5 Copper (Cu)**

Copper was detected in samples taken at all three SWAT mussel sites visited in 2013 (Figure 1.3.1.1.5.1). Copper levels measured in mussels ranged from a low mean concentration of 4.20 ug/g dry wt. at Sears Island, Searsport, to a high mean concentration of 7.84 ug/g dry wt. at East End Beach, Portland, and at Rockland. Copper concentrations at East End Beach and Rockland exceeded the Gulfwatch median and 85<sup>th</sup> percentile (LeBlanc et al. 2009). Sears Island had a copper concentration below the Gulfwatch median. SWAT copper concentrations at all three sites sampled in 2013 fell below the NS&T median and 85<sup>th</sup> percentile (Figure 1.3.1.1.5.2) (Kimbrough et al. 2008). None of the three sites sampled in 2013 was considered elevated for copper.





Copper occurs naturally and is ubiquitous throughout the marine environment. Copper in trace amounts is considered to be an important nutrient for plant and animal growth. Heightened copper concentrations can occur due to anthropogenic sources including mining, agriculture, sewage sludge, antifouling paint, fungicides, wood preservatives, and brake pads. With the reduction of the use of chromated copper arsenate (CCA) wood preservative subsequent to being phased out by EPA, newer wood preservatives utilizing even higher levels of copper have come into use, including quaternary copper. Similarly, tributyltin marine bottom paint use was reduced in the 1980s, resulting in increased use of copper-based antifouling paints, and asbestos removal from brake pads has been offset by increased copper usage in brake pads (Kimbrough et al. 2008).

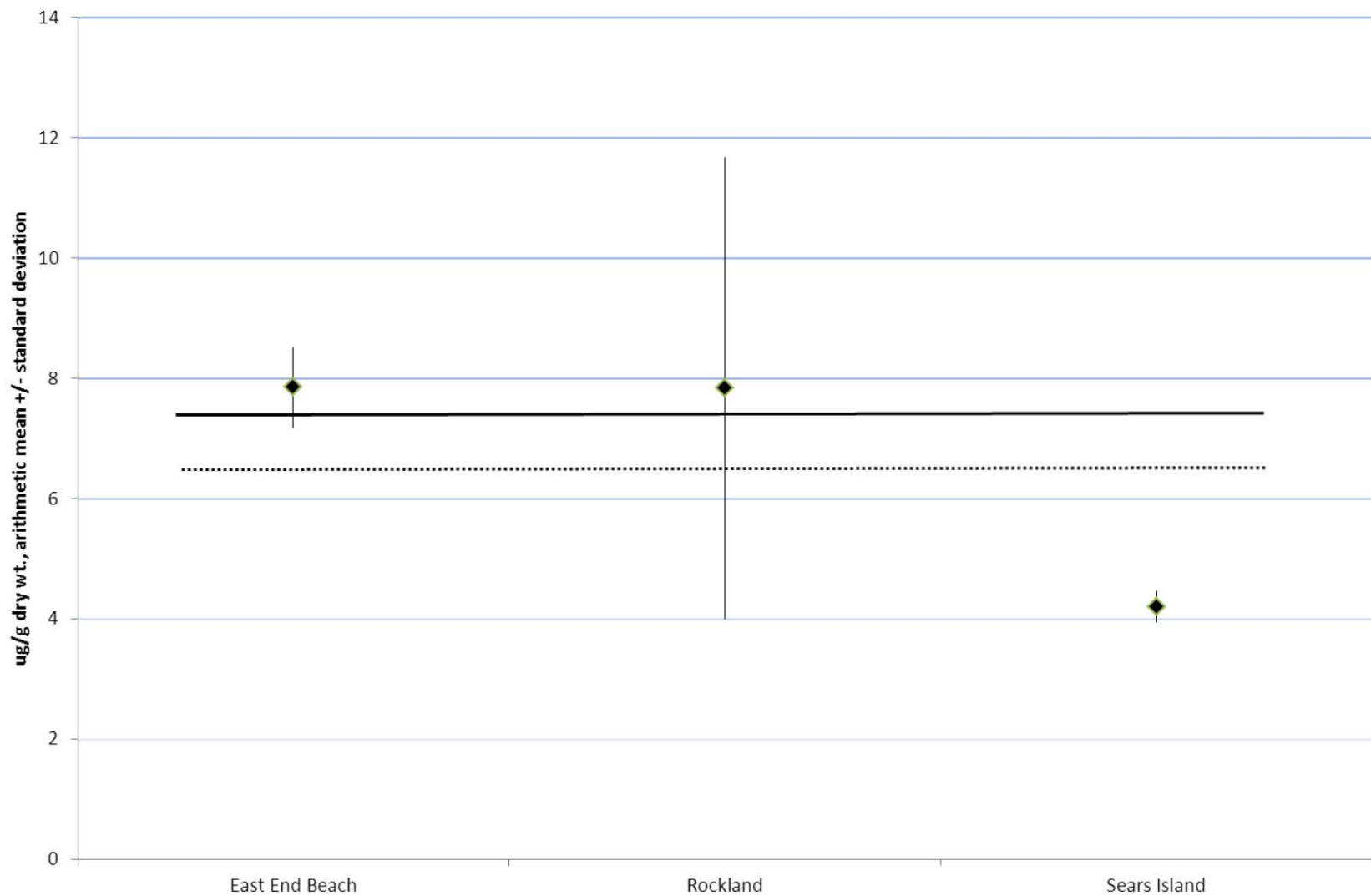
From a human health perspective, copper is not highly toxic to humans, though there are some chronic effects. There is neither a recommended FDA safety level for human consumption for copper in fish or shellfish (Kimbrough et al. 2008), nor does MCDC report a FTAL for copper in non-commercially caught sportfish.

#### **1.3.1.1.6 Iron (Fe) and Aluminum (Al)**

Iron was detected at all three SWAT blue mussel sites sampled in 2013 (Figure 1.3.1.1.6.1). Iron concentrations measured in mussels ranged from a low mean concentration of 138 ug/g dry wt. at Sears Island, Searsport, to a high mean concentration of 494 ug/g dry wt. at East End Beach, Portland. Iron concentrations at Rockland and Sears Island were below the Gulfwatch median, while the concentration at two East End Beach exceeded the Gulfwatch median. None of the sites sampled in 2013 exceeded the Gulfwatch 85<sup>th</sup> percentile. Figure 1.3.1.1.6.1 also shows a comparison of SWAT mean iron concentrations to NS&T median and 85<sup>th</sup> percentile iron concentrations. The iron concentration at one site, Sears Island, was below the NS&T national median, and the remaining two sites had iron concentrations between the NS&T median and 85<sup>th</sup> percentile. No site had an iron concentration in mussel tissue that exceeded the NS&T national 85<sup>th</sup> percentile and consequently, no site was considered elevated for iron.

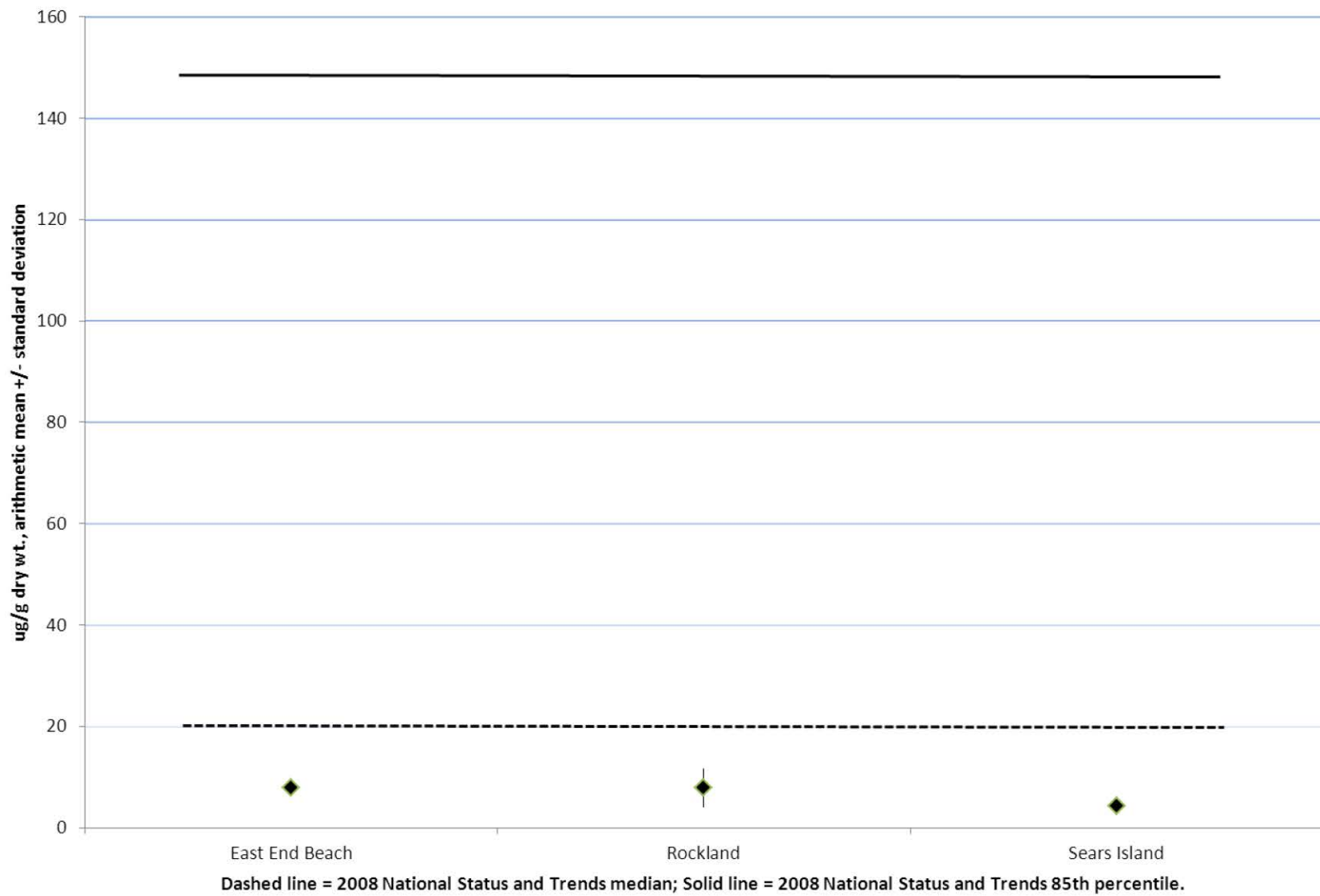
Aluminum concentrations detected in mussels ranged from a low mean concentration of 73 ug/g dry wt. at Sears Island, Searsport, to a high mean concentration of 284 ug/g dry wt. at East End Beach, Portland (Figure 1.3.1.1.6.2). Aluminum concentrations at all three sites were below the Gulfwatch median concentration (LeBlanc et al. 2009). Figure 1.3.1.1.6.2 also shows a comparison of SWAT mean aluminum concentrations to NS&T median and 85<sup>th</sup> percentile concentrations. The mean aluminum concentrations at two sites were below the NS&T median, while the mean concentration at one site, East End Beach, was between the NS&T median and 85<sup>th</sup> percentile concentrations. None of the sites was considered to have a mussel tissue concentration elevated in aluminum.

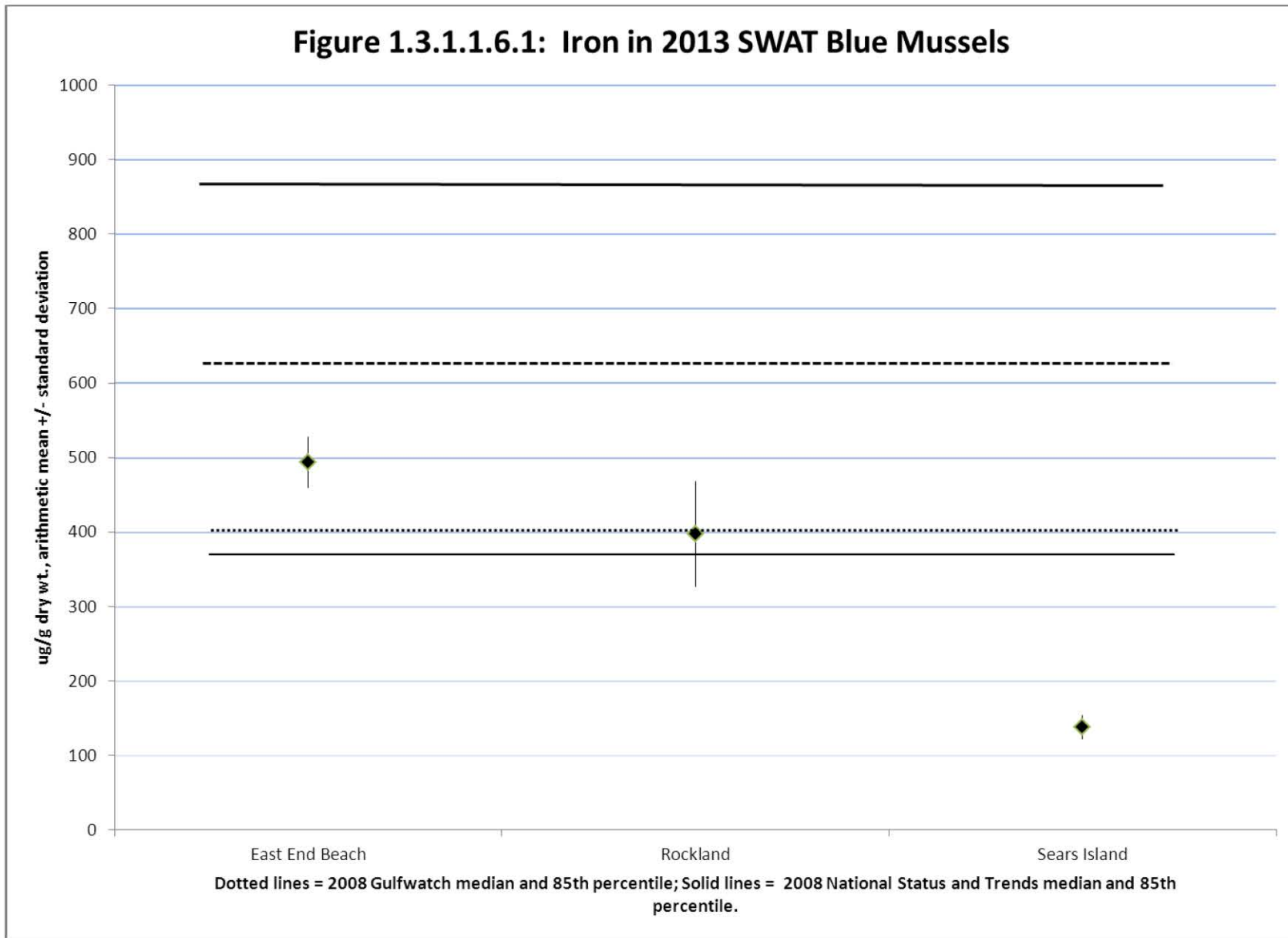
**Figure 1.3.1.1.5.1: Copper in 2013 SWAT Blue Mussels**



Dashed line = 2008 Gulfwatch median; Solid line = Gulfwatch 85th percentile.

**Figure 1.3.1.1.5.2: Copper in 2013 SWAT Blue Mussels**





High iron and aluminum concentrations are usually associated with the intake of high levels of suspended sediments by mussels at sampled sites, with both metals being common components of crustal rocks and coastal sediments. This correlation has also been shown with gut depuration experiments conducted as part of Gulfwatch monitoring in previous years, indicating that some of the iron and aluminum is associated with gut contents and not bioaccumulated loads (Leblanc, 2009). Monitoring for iron and aluminum provides an important reference to gauge sediment intake by mussels, allowing iron and aluminum levels to be referenced if other more toxic metals or contaminants are detected in mussel tissue.

From a human health perspective, MCDC does not report FTALs for iron and aluminum.

#### **1.3.1.1.7 Nickel (Ni)**

Nickel was detected at all three SWAT blue mussel sites visited in 2013 (Figure 1.3.1.1.7.1). Nickel levels measured in mussels ranged from a low mean concentration of 0.49 ug/g dry wt. at Sears Island, Searsport, to a high mean concentration of 1.53 ug/g dry wt. at Rockland. One site, Sears Island, had a nickel concentration below the Gulfwatch median, while the remaining two sites had concentrations that fell between the Gulfwatch median and 85<sup>th</sup> percentile (Figure 1.3.1.1.7.1). Figure 1.3.1.1.7.1 also compares 2013 SWAT blue mussel tissue nickel concentrations to NS&T median and 85<sup>th</sup> percentiles to place Maine data into a national context. Maine SWAT sites had nickel concentrations below the national median at all three sites. No 2013 SWAT nickel concentrations exceeded the NS&T 85<sup>th</sup> percentile, so no SWAT sites were considered to be elevated for nickel. Higher nickel concentrations are probably associated with sediment ingestion, similar to iron and aluminum concentrations.

Nickel occurs naturally in the environment and is an essential trace element to biological processes. Nickel from soil and weathering of rocks enters rivers and provides the largest source of nickel to coastal waters. Nickel occurs in stainless steel, nickel-cadmium batteries, pigments, computers, wire, coins, and is used in electroplating. Heightened nickel concentrations occur in the Great Lakes and speculation about sources centers on air deposition from a large nickel smelting operation in Ontario, Canada (Kimbrough et al. 2008).

Nickel is not thought to bioaccumulate in the food chain, however, nickel can be harmful to humans in large doses, inducing effects including bronchitis and even cancer from long term exposure (Kimbrough et al. 2008). The MCDC reports a non-cancer FTAL for nickel in non-commercially caught finfish of 43 ug/g wet weight (ppm), which is more conservative than the FDA action level for shellfish of 80 ug/g wet weight (ppm). The maximum mean concentration detected by SWAT in 2013 of 0.24 ug/g wet wt. (ppm) at Rockland is two orders of magnitude below the more conservative MCDC action level. MCDC does not report a cancer action level for nickel.

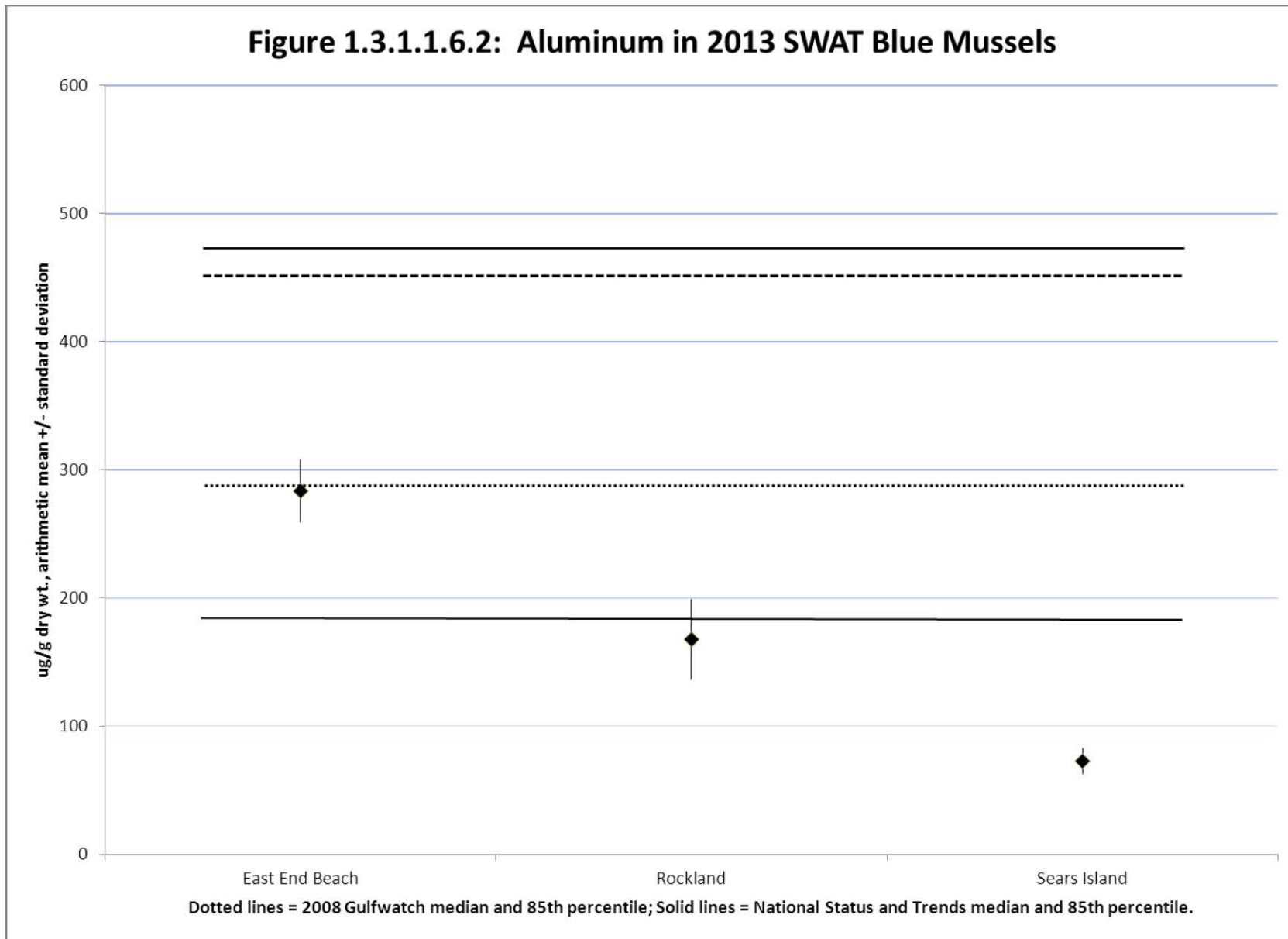
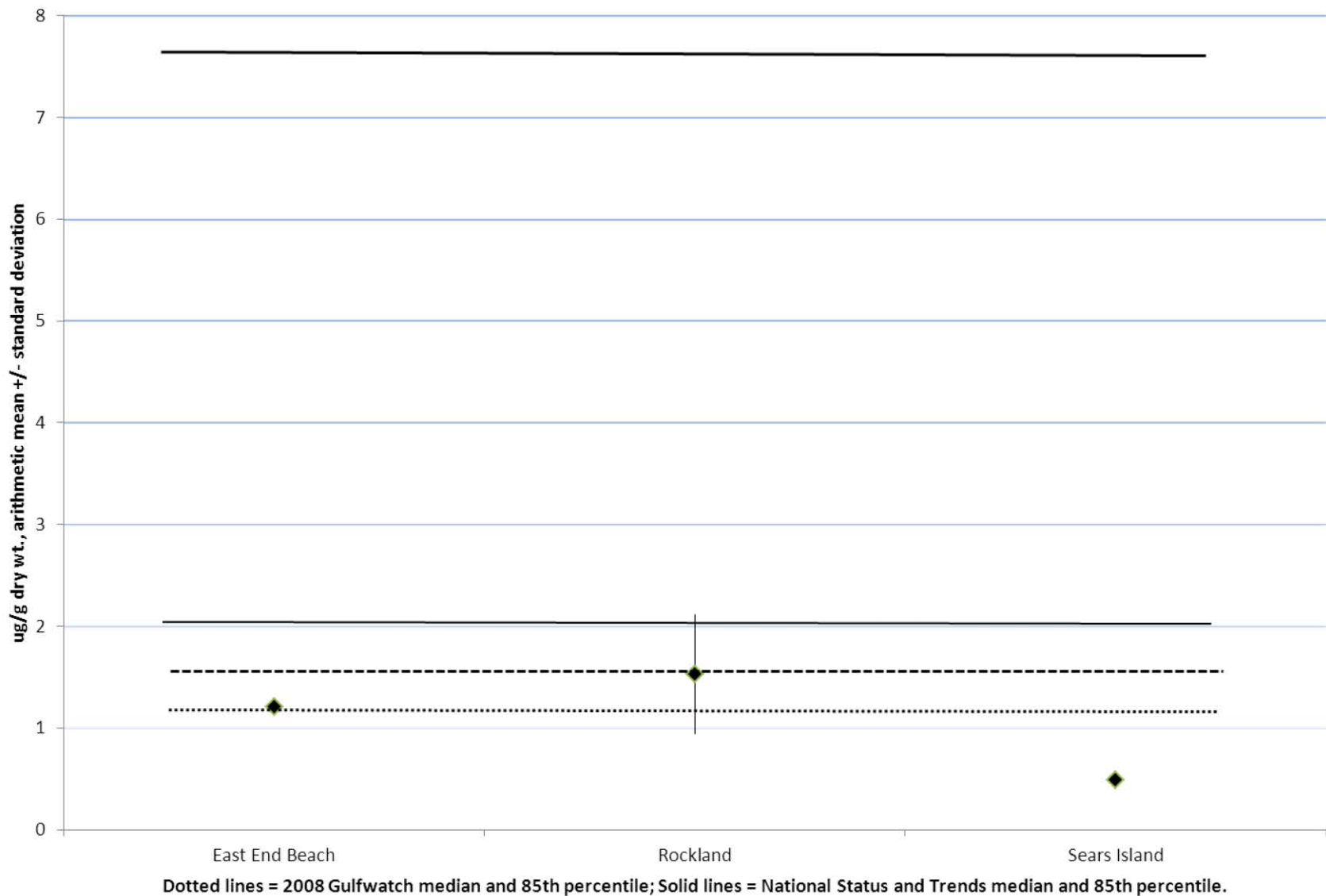


Figure 1.3.1.1.7.1: Nickel in 2013 SWAT Blue Mussels





### 1.3.1.1.8 Lead (Pb)

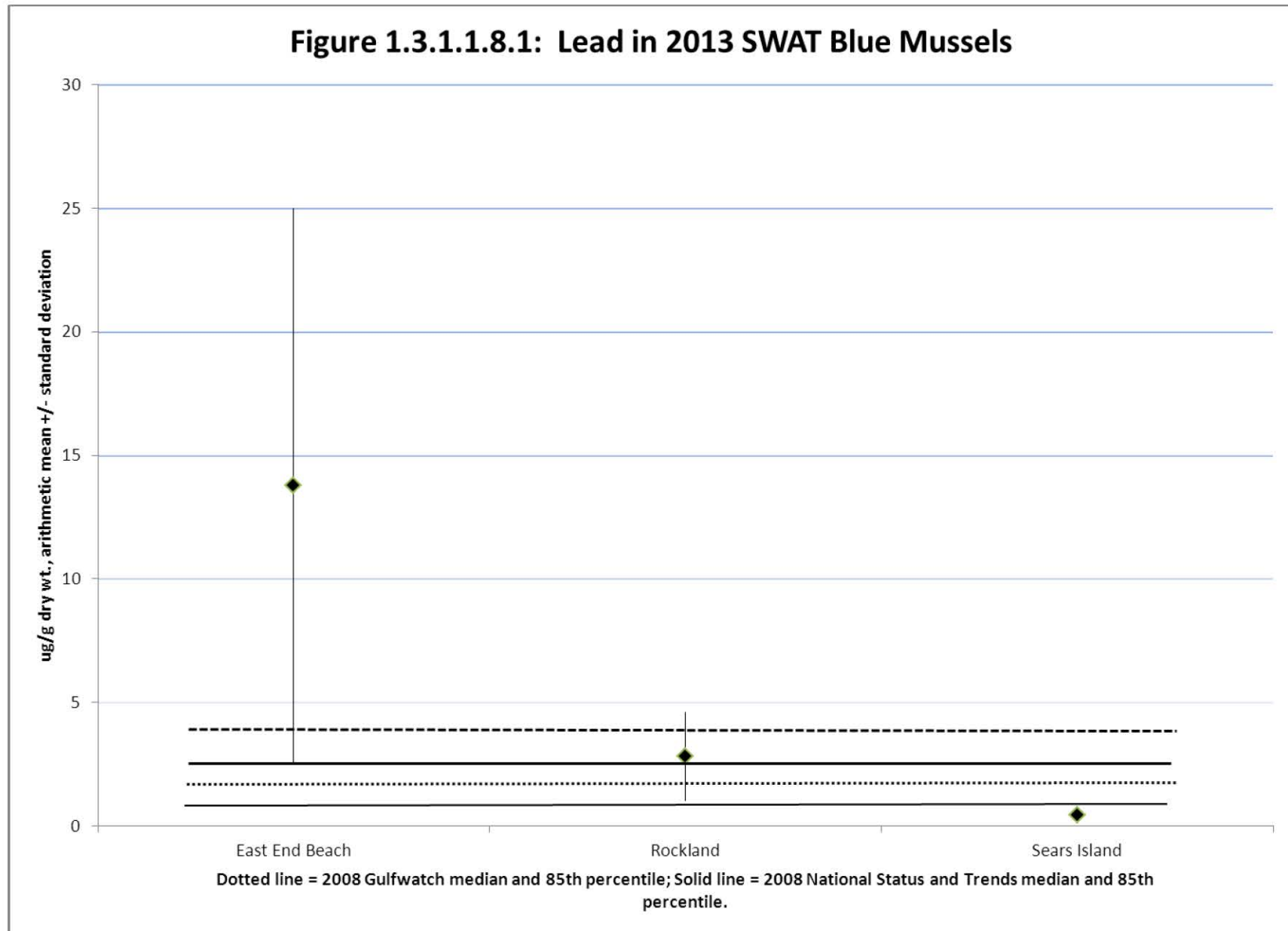
Lead was detected at all three SWAT blue mussel sites visited in 2013 (Figure 1.3.1.1.8.1). Lead levels measured in mussels ranged from a low mean concentration of 0.44 ug/g dry wt. at Sears Island, Searsport, to a high mean concentration of 13.77 ug/g dry wt. at East End Beach, Portland. Sears Island had a concentration less than the Gulfwatch median, Rockland had a lead concentration above the median and below the 85<sup>th</sup> percentile, and East End Beach had a concentration well above the Gulfwatch 85<sup>th</sup> percentile. Figure 1.3.1.1.8.1 also compares 2013 SWAT blue mussel lead tissue concentrations to NS&T median and 85<sup>th</sup> percentiles to place Maine data into a national context. Two SWAT sites had concentrations which exceeded the NS&T median, while Sears Island had a concentration below the median. Two of three SWAT sites, East End Beach, Portland, and Rockland, exceeded the NS&T 85<sup>th</sup> percentile for lead (2.61 ug/g dry wt.)(2008 NS&T data, latest available), and are considered elevated based on criteria in the SWAT and Gulfwatch programs. Rockland had a concentration of 2.82 ug/g dry wt., just over the NS&T 85<sup>th</sup> percentile.

Lead tissue concentrations from prior samples at Temporal Sites from East End Beach, Portland, and Sears Island, Searsport, were compared to 2013 concentrations (Figure 1.3.1.1.8.2). Lead concentrations at East End Beach appear to fluctuate somewhat from year to year, which is probably due to patchiness of contamination within the site. With four much more widely spaced years of data at Sears Island, lead concentrations appear to fluctuate somewhat but are much more stable than those at East End Beach. While more data will be required to demonstrate a consistent trend, other Maine sites with elevated lead levels and limited temporal data sampled in recent years suggest that concentrations are not increasing but have been relatively stable at sites statewide (and Gulf of Maine-wide in the Gulfwatch program supported by longer-term data sets).

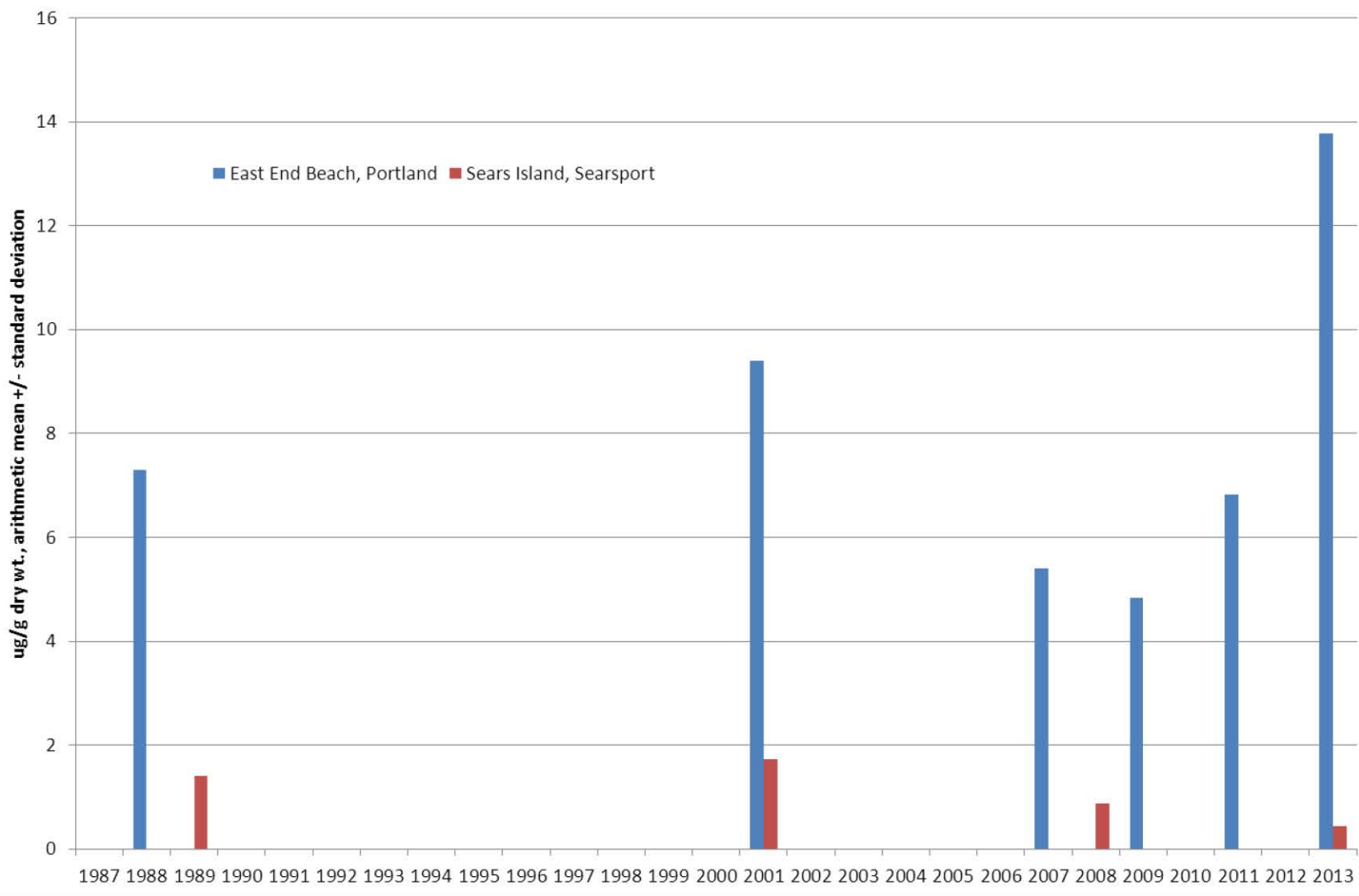
East End Beach, Portland, is located just north of the entrance to Portland Harbor and is located near a sewer outfall. In addition, several sewage treatment plants discharge to the north and south of the site. The upland area is densely settled residential development with a substantial amount of impervious surface that sheds storm water. Sears Island, Searsport, does not have the magnitude of development present in Portland harbor, but is influenced by commercial development, including a major shipping terminal nearby at Mack Point, Searsport across the harbor, and is located downstream in the Penobscot estuary from the greater Bangor/Brewer area and multiple upriver discharges. Repeated sampling at these sites should yield a more complete picture of trends in contaminants, including lead. Some inter-annual variability is to be expected especially with minor spatial differences between replicates. Contaminant patchiness may also be a factor in the variation in lead levels from year to year.

Lead occurs naturally in the earth's crust, however, global lead concentrations in the environment have increased in the last century due to the use of leaded gasoline. Reduction in lead loading through regulation of leaded gasoline and lead paints has occurred in recent decades. Elevated lead levels in the environment also occur due to

manufacturing, paints, lead solder, ammunition, plumbing, incineration and burning of fossil fuels. Lead loading in coastal waters is related to wastewater discharge, river



**Figure 1.3.1.1.8.2: Trend in Blue Mussel Tissue Lead Concentrations Through 2013**



runoff, atmospheric deposition, and natural weathering of crustal rock (Kimbrough et al. 2008).

From a human health perspective, the FDA action level for lead in clams, oysters, and mussels is 1.7 ug/g wet wt. (ppm) (Kimbrough et al 2008). The more conservative MCDC lead FTAL in non-commercially caught sportfish is 0.6 ug/g wet wt. (ppm), which is based on a blood lead concentration model. The highest mean concentration in the 2013 Maine SWAT mussel data, 2.1 ppm (ug/g) wet wt. at East End Beach, Portland, exceeds the MCDC lead FTAL. The mean lead concentrations of the remaining two sites sampled in 2013 did not exceed the MCDC FTAL for lead. However, due to the wide geographic spacing of six (more than the usual four) Rockland area subsamples in 2013, it is possible to determine that two of those subsamples exceeded the MCDC FTAL for lead, which were those located at the Ocean Pursuits Boatyard and the Town Landing, both in Rockland. The four additional subsamples in the broader Rockland Harbor area had lead concentrations below the MCDC FTAL for lead. These stations were located farther from the waterfront in Rockland and just inside the breakwater, north of the breakwater, on the northeastern end of Owls Head, and in Owls Head harbor.

Review of the 2007-13 SWAT blue mussel sampling data from 59 sites indicates that mean lead concentrations at eight sites equaled or exceeded the MCDC lead FTAL. Sites sampled in those years equaling or exceeding the MCDC FTAL for lead are:

Spring Point, S. Portland, 2007	0.6 ppm wet wt.
Spring Point, S. Portland, 2010	0.7 ppm wet wt.
Spring Point, S. Portland, 2012	0.6 ppm wet wt.
Middle Fore R., Portland, 2007	0.6 ppm wet wt.
East End Beach, Portland, 2007	0.8 ppm wet wt.
East End Beach, Portland, 2009	0.8 ppm wet wt.
East End Beach, Portland, 2011	0.9 ppm wet wt.
East End Beach, Portland, 2013	2.1 ppm wet wt.
Turnip Island, Georgetown, 2012	1.4 ppm wet wt.
Crockett Point, Rockland, 2007	1.1 ppm wet wt.
Crockett Point, Rockland, 2010	1.3 ppm wet wt.
Crockett Point, Rockland, 2011	1.1 ppm wet wt.
Ocean Pursuits Boat Yard, Rockland, 2013	0.6 ppm wet wt.
Town Landing, Rockland, 2013	0.9 ppm wet wt.
Camden Harbor, Camden, 2007	0.7 ppm wet wt.
Goose Falls, Brooksville, 2007	1.1 ppm wet wt.
Piscataqua River Back Channel, Kittery, 2008	0.6 ppm wet wt.

The MCDC lead FTAL is based on the consumer eating an 8 oz. meal. Maine SWAT data indicate that an 8 oz. meal would include approximately 45-50 blue mussels of the size tested by the SWAT program.

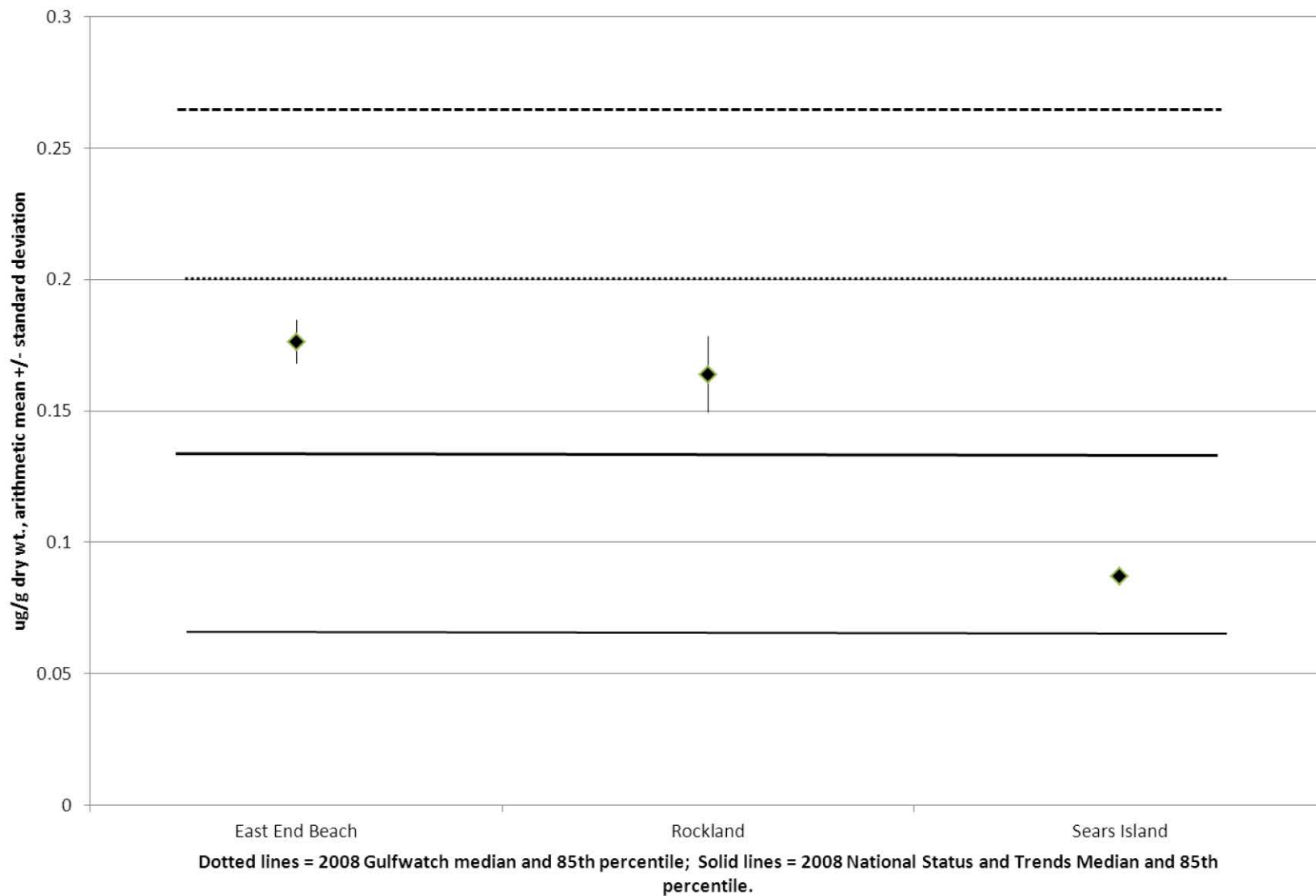
#### **1.3.1.1.9 Mercury (Hg)**

Mercury was detected in all three blue mussel sample locations visited in 2013 (Figure 1.3.1.1.9.1). Mercury levels measured in mussels ranged from a low mean concentration of 0.09  $\mu\text{g/g}$  dry wt. at Sears Island, Searsport, to a high mean concentration of 0.18  $\mu\text{g/g}$  dry wt. at East End Beach, Portland. None of the three mercury concentrations exceeded the 2008 Gulfwatch median, and all three also fell below the Gulfwatch 85<sup>th</sup> percentile concentration. Figure 1.3.1.1.9.1 also compares 2013 SWAT blue mussel mercury concentrations to NS&T Mussel Watch median and 85<sup>th</sup> percentile values. The reader should note that Gulfwatch median and 85<sup>th</sup> percentile values actually exceed NS&T Mussel Watch median and 85<sup>th</sup> percentile values, respectively, since the northeastern US has relatively high mercury levels due to air deposition of mercury from a wide range of sources in the Midwest US. Based on the Gulfwatch and SWAT criteria of “elevated” contaminants being those above the NS&T 85<sup>th</sup> percentile, two of three SWAT sites tested in 2013 would be considered elevated for mercury despite their more typical scores when compared to other northeast US samples from the Gulf of Maine. These two sites are East End Beach, Portland, and Rockland.

Mercury occurs naturally in the environment; however, elevated levels are associated with anthropogenic sources. United States sources of mercury to the air include coal fired electrical power generation, incinerators, mining, landfills, and sewage sludge (Kimbrough et al., 2008).

From a human health perspective, the developmental methylmercury FTAL (more protective) used by the MCDC is 0.2  $\mu\text{g/g}$  (ppm) wet wt. for non-commercially caught finfish (fish filet). This FTAL assumes an 8 oz. meal size is consumed weekly. Maine SWAT data uses a total mercury value, which is a more complete measure of mercury than the methylmercury concentration, but includes this more toxic form. The highest mean blue mussel total tissue mercury concentration measured in Maine in 2013 was 0.027  $\mu\text{g/g}$  wet wt. (ppm) at East End Beach, Portland. This compares favorably with the MCDC methylmercury developmental FTAL of 0.2 ppm, assuming a similar meal size and frequency. To consume approximately 8 oz. of blue mussel tissue the consumer would need to eat approximately 45-50 blue mussels based on the mean mass per mussel collected by the SWAT program.

**Figure 1.3.1.1.9.1: Mercury in 2013 SWAT Blue Mussels**



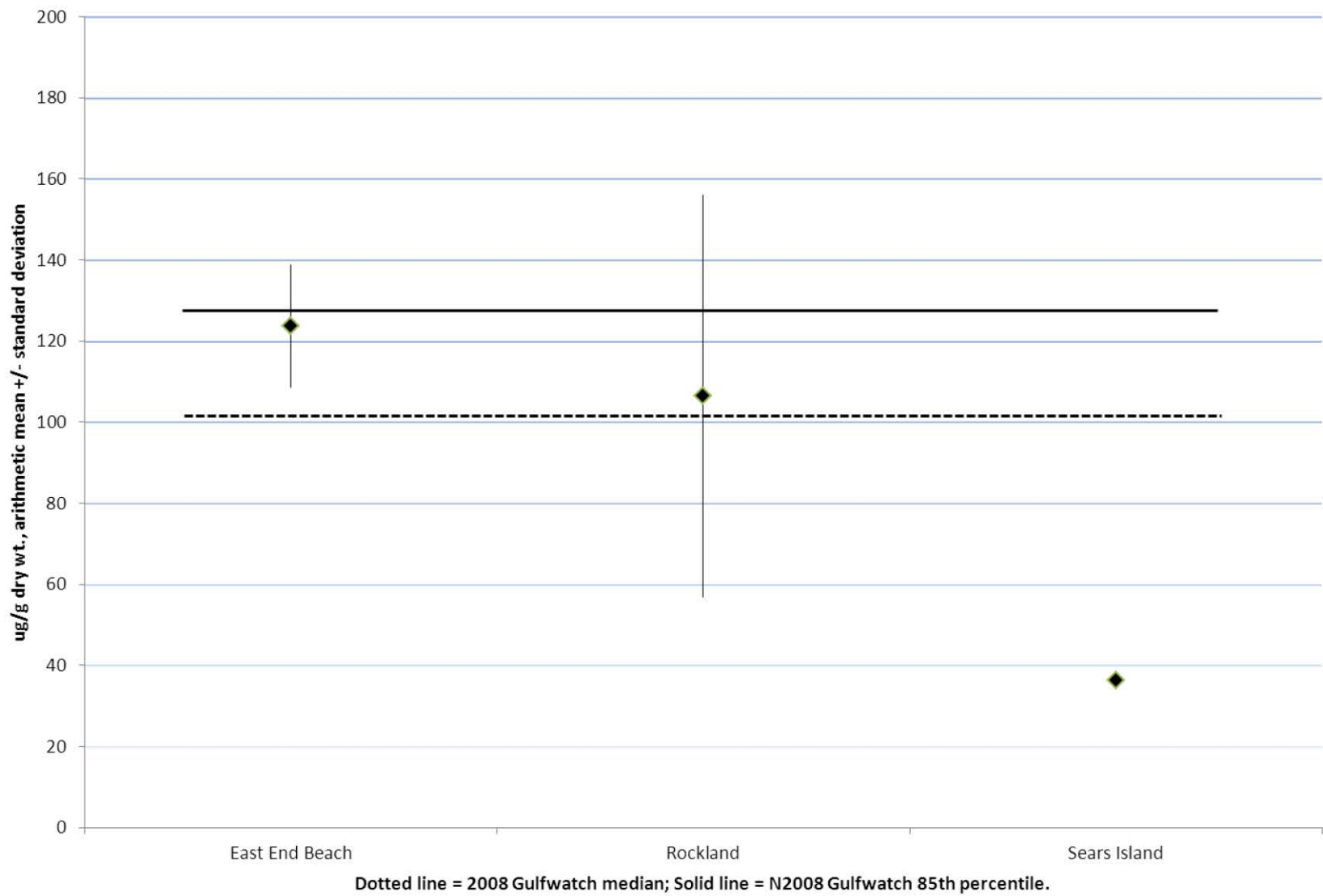
#### **1.3.1.1.10 Zinc (Zn)**

Zinc was detected in all three sample locations visited in 2013 (Figure 1.3.1.1.10.1). Zinc levels measured in mussels ranged from a low mean concentration of 36.3 ug/g dry wt. at Sears Island, Searsport, to a high mean concentration of 123.7 ug/g dry wt. at East End Beach, Portland. Two SWAT blue mussel tissue zinc concentrations exceeded the 2008 Gulfwatch median, though none exceeded the Gulfwatch 85<sup>th</sup> percentile. Figure 1.3.1.1.10.2 shows 2013 Maine SWAT blue mussel zinc concentrations were all below the NS&T Mussel Watch median and 85<sup>th</sup> percentile.

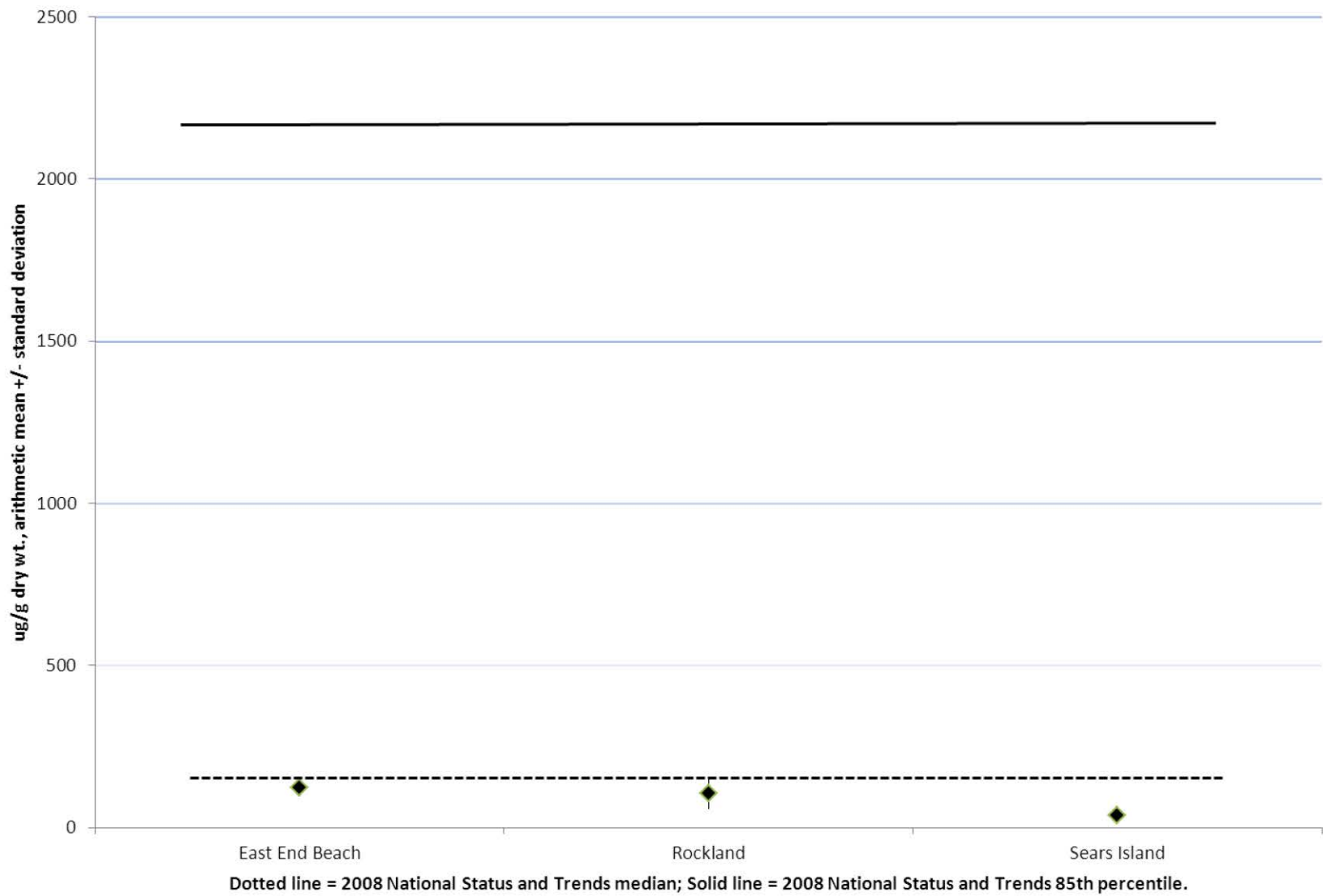
Zinc is widespread in its distribution but elevated levels primarily originate from a variety of human activities including vehicle tire wear, electroplating and galvanized metals, industrial wastes, and drainage from mining (Kimbrough et al. 2008). Though an essential nutrient at low levels, higher doses to humans can cause anemia or pancreatic and kidney damage. Since humans do not bioaccumulate zinc, health impacts are normally associated with high doses. From a human health perspective, MCDC reports a non-cancer FTAL for zinc of 648 ug/g wet wt. (ppm), which is higher than any wet wt. concentrations observed in SWAT blue mussel tissue. There is no recommended FDA safety level for zinc in fish (Kimbrough et al. 2008).



Figure 1.3.1.1.10.1: Zinc in 2013 SWAT Blue Mussels



**Figure 1.3.1.1.10.2: Zinc in 2013 SWAT Blue Mussels**



### 1.3.1.2 Softshell Clams

Two softshell clam sites were sampled in 2013: Mast Cove, part of the Piscataqua River in Eliot, and Presumpscot River, Falmouth/Portland. The samples were analyzed for 11 metals: Silver (Ag), aluminum (Al), arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), lead (Pb), mercury (Hg), nickel (Ni), and zinc (Zn). Results were compared to Gulf of Maine (Gulfwatch, see LeBlanc et al. 2009) softshell clam data to place Maine SWAT data set in a regional context.

#### 1.3.1.2.1 Silver (Ag)

Silver was detected at all sample locations visited historically (Figure 1.3.1.2.1.1). Silver measured in clams ranged from a low mean concentration of 0.13 ug/g dry wt. at Harris Cove, Eastport, to a high mean concentration of 2.39 ug/g dry wt. at Mast Cove, Eliot (2013). Silver mean concentrations at the two sites sampled in 2013, Presumpscot River, Falmouth/Portland, and Mast Cove, Eliot, were within the higher range of concentrations from previously sampled Maine clam sites. Silver mean concentrations in SWAT softshell clams were also compared to the Gulfwatch mean concentration for four sites sampled in 2008 (two in Maine and two in New Hampshire). The mean concentrations at Mast Cove, Eliot (2004 and 2013), and Presumpscot River, Falmouth/Portland (2012 only), exceeded the Gulfwatch mean (1.32 ug/g dry wt.). The silver concentration in clam tissue in Mast Cove appeared to be slightly higher than, though similar, to the 2004 concentration. The silver concentration in Presumpscot clam tissue appeared to be slightly lower than, though similar, to the 2012 concentration. Presumpscot clam tissues predominantly came from the eastern portion of the estuary in 2012, while the 2013 tissues originated from clams collected on the western portion of the estuary. Sampling was conducted in the western portion of the estuary in 2013 to include extensive flats that historically had a substantial clam resource and in which DMR had a strong interest. The differing concentrations may represent variability between the two shores, although significant overlap exists in concentrations between geographic subsamples. Variation from year to year may be an artifact of intra-site spatial variability (note length of whiskers representing standard deviation) or year to year variability.

Higher silver concentrations in water and sediments coincide with municipal sewage discharge (Sanudo-Wilhelmy and Flegal 1992; Buchholtz ten Brink et al. 1997). The increasing use of silver, including nanosilver, in products such as clothing, paints, and caulks, makes monitoring silver of interest at present and in the future. Silver concentrations in Maine softshell clams appear to be relatively low. The highest Gulfwatch values, which came from the two NH sites, were just over 2 ug/g dry wt., which is very similar to the Mast Cove, Eliot, SWAT site tissue concentration.

The Maine Center for Disease Control, Bureau of Health (MCDC) silver non-cancer fish tissue action level (FTAL) is 11 ug/g wet wt. (ppm) for non-commercially caught fish. The highest SWAT softshell clam tissue mean silver concentration, when expressed on a wet weight basis, is 0.43 ug/g wet wt. at Mast Cove, Eliot (2013). This concentration is over an order of magnitude below the 11 ug/g wet wt. FTAL, assuming the same meal size is applied.

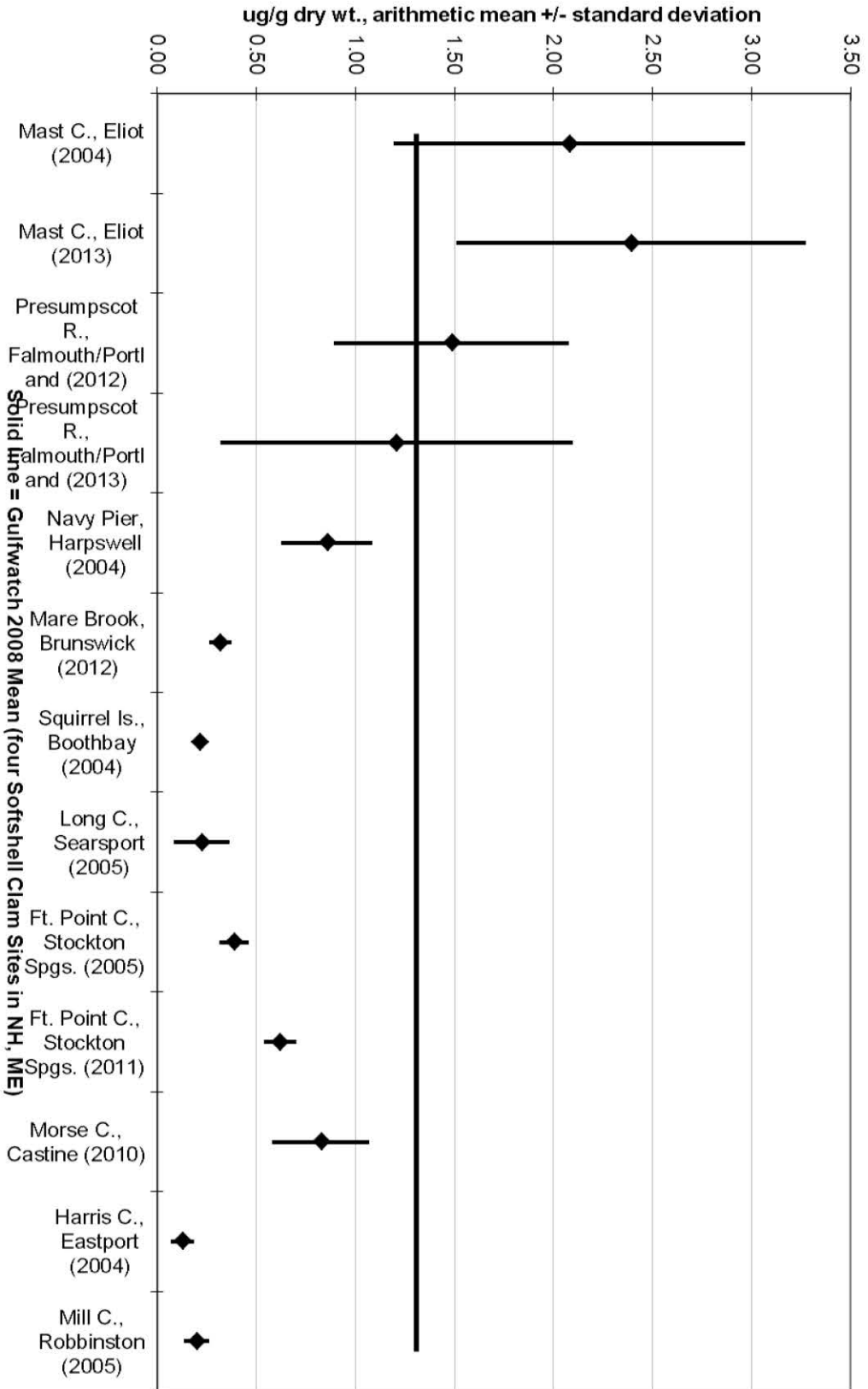


Figure 1.3.1.2.1.1: Silver in SWAT Softshell Clams

### 1.3.1.2.2 Arsenic (As)

Arsenic was detected in clam tissue at both Presumpscot River, Portland, and Mast Cove, Eliot in 2013. In previous years, analysis for arsenic was performed on clam tissues from only four sites: Presumpscot River, Portland, and Mare Brook, Brunswick (both in 2012), Fort Point Cove, Stockton Springs (2011), and Morse Cove, Castine (2010). Mean arsenic concentrations ranged from 9.97 ug/g dry wt. at Morse Cove (2010) to 92.95 ug/g dry wt. at Mare Brook, Brunswick (2012). Sites sampled in 2013 had mean arsenic concentrations in the middle of this range, 16.5 and 23.0 ug/g dry wt., at Mast Cove, Eliot, and Presumpscot River, Portland, respectively.

While Gulfwatch does not monitor arsenic in blue mussels or softshell clams in the Gulf of Maine, arsenic in mussels and oysters is tracked regionally and nationally by NS&T. In blue mussels, NS&T considers 5-11 parts per million dry wt. (directly comparable to SWAT ug/g data) to be in the lowest of three ranges of arsenic concentration within the region (Kimbrough et al. 2008). The mean arsenic concentration in softshell clams at Morse Cove (2010) fell into this range, while the mean arsenic concentrations at Mast Cove (2013), Presumpscot River (2012 and 2013), Fort Point Cove (2011) Morse Cove (2010) fell into the lower end of the middle range of NS&T arsenic concentrations (23-41 part per million dry wt., Kimbrough et al 2008). The mean arsenic concentration in softshell clams at Mare Brook, Brunswick (2012), 92.95 ug/g dry wt., fell above the high range used by NS&T regionally (23-41) and nationally (23-57 parts per million). The NS&T ranges are based on mussels or oysters as regionally available. However, it is of interest to give a point of comparison for Maine clam data. Higher concentrations at Mare Brook may be related to very fine grained sediments and sediment content in the clam gut, as clams are not depurated before tissue preparation for lab analysis.

Nationally, the primary source for elevated levels of arsenic is crustal rock. Other than natural sources, industrial pollution can contribute arsenic to the environment from preserved wood, semiconductors, pesticides, defoliants, pigments, antifouling paints, and veterinary medicines. Atmospheric sources include smelting, fossil fuel combustion, power generation, and pesticide application (Kimbrough et al. 2008).

For non-commercially caught finfish, MCDC reports a cancer FTAL of 0.014 ppm and a non-cancer FTAL of 0.6 ppm, both for inorganic arsenic (the most toxic form). Most fish tissue data, including the SWAT blue mussel tissue data, are analyzed for total arsenic, not inorganic arsenic. MCDC uses FDA's 1993 assumption that 10% of total arsenic in finfish is inorganic arsenic. Using this assumption, approximate inorganic arsenic concentrations for SWAT softshell clams were calculated by dividing wet weight concentrations by a factor of 10. Therefore, the Presumpscot River (2013) clam inorganic arsenic mean concentration is estimated to be 0.35 ug/g wet wt., and the Mast Cove (2013) clam inorganic arsenic mean concentration is estimated to be 0.30 ug/g wet wt. The same calculations yield Presumpscot River (2012) clam inorganic arsenic mean concentration estimated to be 0.23 ug/g wet wt., and the Mare Brook (2012) clam inorganic arsenic mean concentration estimated to be 1.56 ug/g wet wt. Historically, all six clam sites sampled for arsenic were calculated to exceed the MCDC cancer FTAL of 0.014 ug/g wet wt. (ppm). Note that all blue mussel sites sampled since arsenic data have

been recorded as part of the SWAT program also exceed the MCDC cancer FTAL. Only the Mare Brook, Brunswick (2012), estimated clam inorganic arsenic mean concentration (1.56 ug/g wet wt.) exceeds the MCDC non-cancer action level of 0.6 ug/g wet wt. (ppm) for inorganic arsenic. MCDC non-commercially caught finfish FTALs applied here assume an 8 oz. meal eaten by the consumer on a weekly basis.

#### **1.3.1.2.3 Cadmium (Cd)**

Cadmium was detected in tissue from all 13 clam locations (Figure 1.3.1.2.3.1). Cadmium levels measured in softshell clams ranged from a low mean concentration of 0.29 ug/g dry wt. at Mare Brook, Brunswick (2012), to a high mean concentration of 0.77 ug/g dry wt. at Mill Cove, Robbinston. Mill Cove, Robbinston, and Navy Pier, Harpswell, approached the 2008 Gulfwatch median, with all ten sites falling below that median. The mean cadmium concentration in the western portion of Presumpscot River sampled in 2013 appeared to be similar to the mean in the eastern portion of the river sampled in 2012, although there is a great deal of variability in samples comprising each mean. The mean concentration in Mast Cove in 2013 is higher than the previous cadmium mean obtained in 2004.

Cadmium originates from crustal elements as rocks weather and is transported seaward by rivers, which account for approximately half of worldwide cadmium sources. Cadmium is also released through forest fires and volcanic activity, with anthropogenic sources including manufacturing, fossil fuel combustion, and agriculture. Industrial sources include manufacture of batteries, plating, stabilizers, and nuclear power (Kimbrough et al. 2008).

From a human health perspective, the MCDC non-cancer FTAL for cadmium in non-commercially caught finfish is 2.2 ug/g wet wt. The FDA action level for clams, oysters, and mussels is 4 ppm wet wt. (Kimbrough et al. 2008). The highest scoring SWAT clam site, Mill Cove, Robbinston (2005), had a mean cadmium concentration of 0.088 ug/g wet wt., which was well below the MCDC and FDA action levels (4% of the more conservative MCDC non-cancer FTAL).

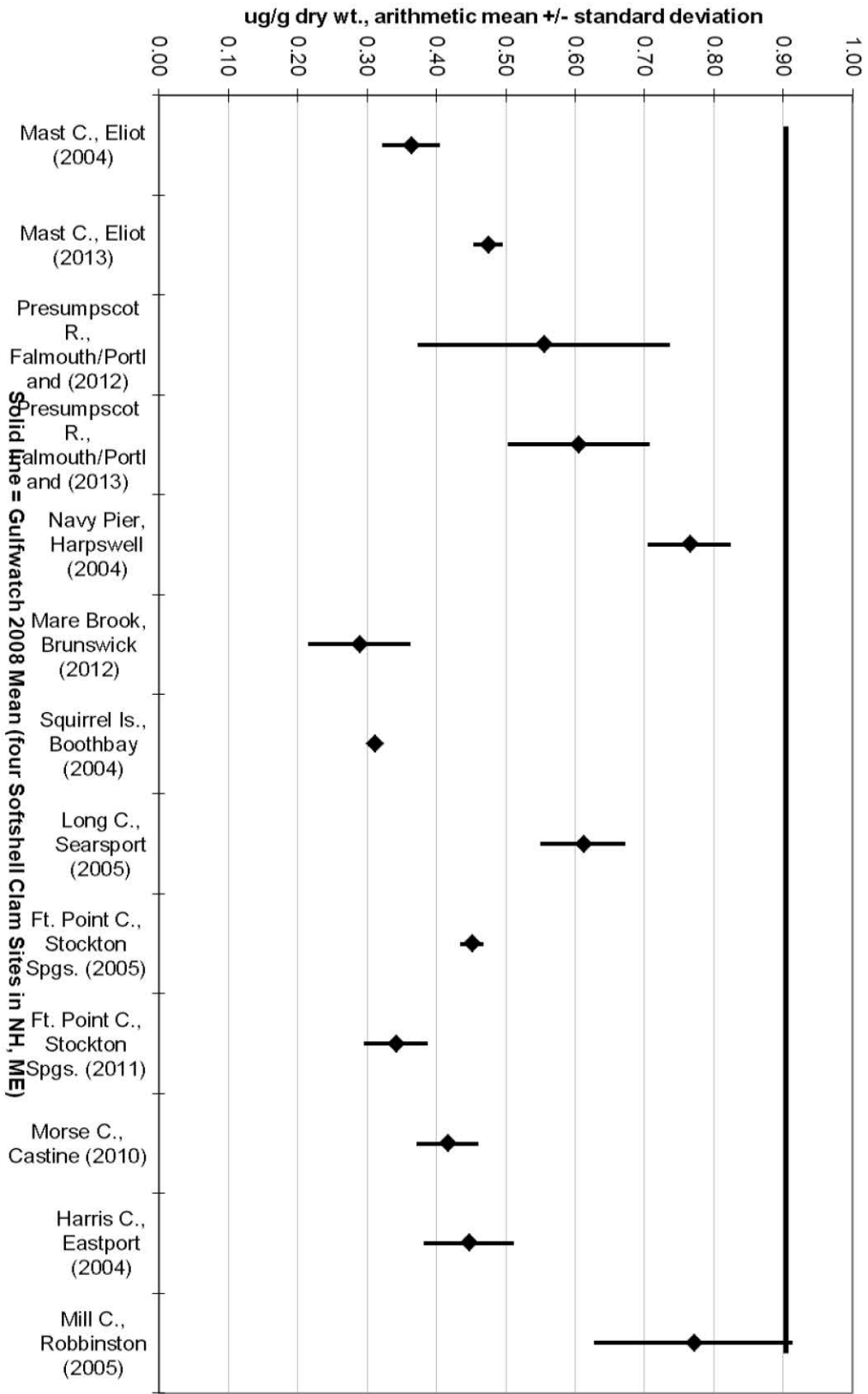


Figure 1.3.1.2.3.1: Cadmium in SWAT Softshell Clams

#### 1.3.1.2.4 Chromium (Cr)

Chromium was detected at all ten sites (Figure 1.3.1.2.4.1). Chromium levels measured in clam tissue ranged from a low mean concentration of 3.07 ug/g dry wt. at Fort Point Cove, Stockton Springs (2011), to a high mean concentration of 13.32 ug/g dry wt. at Mast Cove, Eliot (2004). Figure 1.3.1.2.4.1 depicts SWAT softshell clam chromium concentrations compared to the Gulfwatch 2008 mean concentration for four sites (two each in ME and NH). All but three clam sites, including the Presumpscot River in 2012, fell above the Gulfwatch 2008 mean. The mean concentration from the Presumpscot River in 2013 was higher than the 2012 mean, which may be due to 2013 samples coming from the western half of the estuary and 2012 samples coming from the eastern half. This variation also may be due to patchiness of contaminants. Mast Cove mean concentrations dropped noticeably from 2004 to 2013. The Fort Point Cove, Stockton Springs (2005), clam tissue chromium concentration was essentially the same as the Gulfwatch 2008 mean, while chromium concentrations appeared to be slightly lower in Fort Point Cove samples in 2011, falling below the Gulfwatch 2008 mean. The remaining sites were all above the Gulfwatch 2008 mean.

Natural sources of chromium include leaching from soil and rock into surface waters. Chromium is released from textile, electroplating, and leather tanning industries. Chromium is used extensively in tanning leather and was discharged with untreated tannery effluent during the last two centuries. Chromium persists in the marine environment in sediments near anthropogenic sources (Kimbrough et al. 2008).

From a human health perspective, the MCDC FTALs (7 ug/g cancer action level and 11 ug/g non-cancer action level) for chromium are based on chromium VI, and are not directly comparable to SWAT results, which are for total chromium.

#### 1.3.1.2.5 Copper (Cu)

Copper was detected in samples taken at all ten SWAT softshell clam sites (Figure 1.3.1.2.5.1). Copper levels measured in clam tissue ranged from a low mean concentration of 7.31 ug/g dry wt. at Long Cove, Searsport, to a high mean concentration of 28.5 ug/g dry wt. at Presumpscot River, Falmouth/Portland (2013). Copper concentrations in clam tissue at nine sites fell below the 2008 Gulfwatch mean, including Mast Cove when sampled in 2004 (LeBlanc et al. 2009). Presumpscot River exceeded the Gulfwatch 2008 mean in both 2012 and 2013, as did Mast Cove in 2013. The Mast Cove copper mean was higher in 2013 than in 2004. Differences in Presumpscot means may be attributed to east vs. west shores sampled in subsequent years, or patchiness in contaminants. In prior sampling, the copper concentration at Fort Point Cove appeared to have increased slightly in the 2011 clams compared to the 2005 clams.

Copper occurs naturally and is ubiquitous throughout the marine environment. Copper, in trace amounts, is considered to be an important nutrient for plant and animal growth. Heightened copper concentrations can occur due to anthropogenic sources, including mining, agriculture, sewage sludge, antifouling paint, fungicides, wood preservatives, and brake pads. With the reduction of the use of chromated copper arsenate (CCA) wood



preservative subsequent to being phased out by EPA regulations, newer wood preservatives utilizing even higher levels of copper have come into use, including quaternary copper. Similarly, tributyltin marine bottom paint use was reduced in the 1980s, resulting in increased use of copper-based antifouling paints, and asbestos removal from brake pads has been offset by increased copper usage in brake pads (Kimbrough et al. 2008).

From a human health perspective, copper is not highly toxic to humans, though there are some chronic effects. There is no recommended FDA safety level for human consumption for copper in fish or shellfish (Kimbrough et al. 2008), nor does MCDC report a FTAL for copper in non-commercially caught sportfish.

#### **1.3.1.2.6 Iron (Fe) and Aluminum (Al)**

Iron was detected at all ten SWAT softshell clam sites (Figure 1.3.1.2.6.1). Iron concentrations measured in clam tissue ranged from a low mean concentration of 1,370 ug/g dry wt. at Squirrel Island, Boothbay (2004), to a high mean concentration of 26,145 ug/g dry wt. at Presumpscot River, Portland (2013). Two SWAT sites had clam tissue iron concentrations that exceeded the 2008 Gulfwatch mean, including the Presumpscot River in 2013 and Mare Brook, Brunswick in 2012. Iron concentrations in Fort Point Cove clams in 2011 appeared to be similar or slightly lower than in 2005 (Figure 1.3.1.2.6.1).

Aluminum concentrations detected in clams ranged from a low mean concentration of 563 ug/g dry wt. at Squirrel Island, Boothbay (2004), to a high mean concentration of 5,760 ug/g dry wt. at Mare Brook, Brunswick (2012) (Figure 1.3.1.2.6.2). Clam tissue from four sites, Mare Brook, Presumpscot River (both in 2012 and 2013), and Mast Cove (2013), had aluminum concentrations exceeding the 2008 Gulfwatch mean concentration. Mast Cove had a higher aluminum concentration in 2013 than 2004, while samples from the two opposite banks of the Presumpscot estuary had similar aluminum concentrations in 2012 and 2013. Aluminum concentrations in Fort Point Cove clams in 2011 appeared to be similar or slightly higher than in 2005.

High iron and aluminum concentrations are usually associated with the intake of high levels of suspended sediments by mussels and clams at sampled sites, with the iron and aluminum being abundant crustal elements and therefore abundant in sediments. This correlation has also been shown with gut depuration experiments conducted as part of Gulfwatch monitoring in previous years, indicating that some of the iron and aluminum is associated with gut contents and not bioaccumulated loads (LeBlanc, 2009). Sediment loading in clam gut contents may be quite a bit higher than mussel gut loading, thus affecting aluminum and iron levels disproportionately in clam tissue concentrations since no depuration occurs prior to tissue removal.

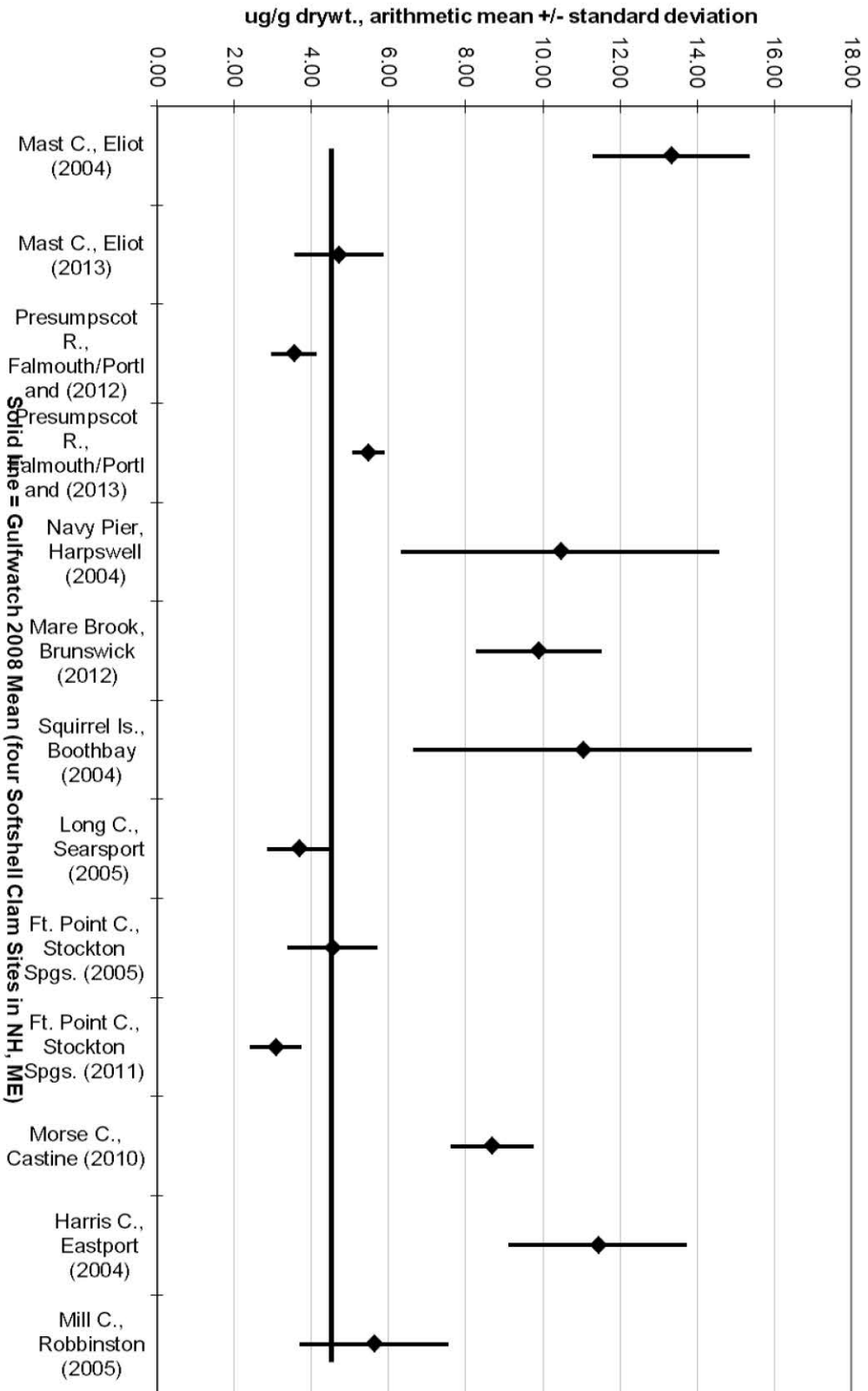


Figure 1.3.1.2.4.1: Chromium in SWAT Softshell Clams

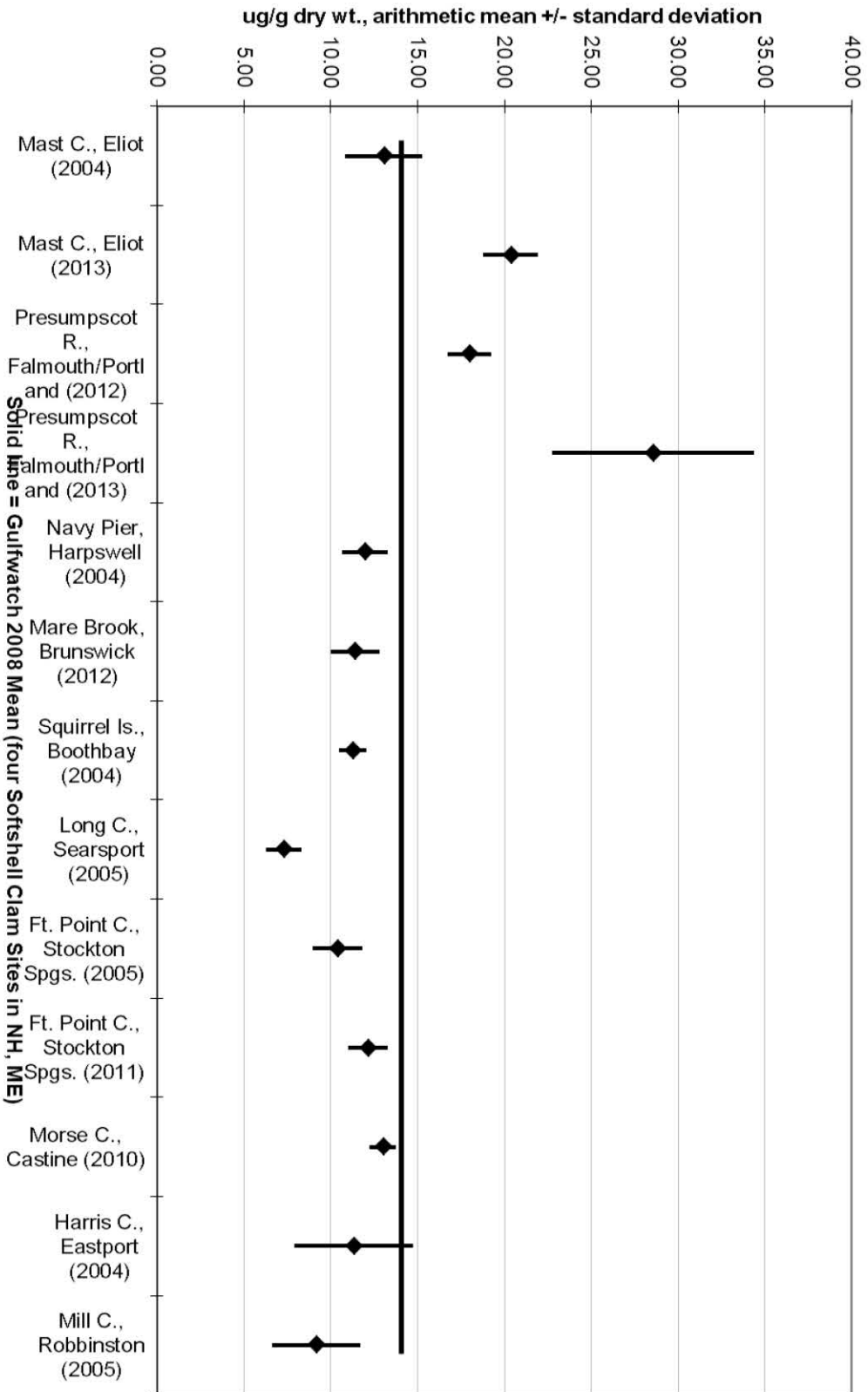


Figure 1.3.1.2.5.1: Copper in SWAT Softshell Clams

Monitoring for iron and aluminum provides an important reference to gauge sediment intake by clams, allowing iron and aluminum levels to be referenced if other more toxic metals or contaminants are detected in tissue. If iron and aluminum concentrations are high, it is likely that a fraction of the contaminant load can be traced back to high sediment intake with some contamination coming from sediment in clam gut contents, rather than bioaccumulated contaminants from within clam tissue.

From a human health perspective, MCDC does not report FTALs for iron and aluminum.

#### **1.3.1.2.7 Nickel (Ni)**

Nickel was detected in clam tissue at all ten SWAT softshell clam sites (Figure 1.3.1.2.7.1). Nickel levels measured in mussels ranged from a low mean concentration of 1.79 ug/g dry wt. at Fort Point Cove, Stockton Springs (2011), to a high mean concentration of 9.68 ug/g dry wt. at Mast Cove, Eliot (2004). Maine SWAT clam tissue nickel concentrations were all higher than the 2008 Gulfwatch clam mean except for 2012 Presumpscot River, 2011 Fort Point Cove, and 2013 Mast Cove, which were below the Gulfwatch mean. Mast Cove mean nickel concentration was lower in 2013 than 2004. Presumpscot nickel concentrations differed, with the 2013 concentrations from the western half of the estuary being higher and showing more variability than those from 2012 in the eastern half of the estuary. Higher nickel concentrations are probably associated with sediment ingestion, similar to iron and aluminum concentrations. The highest nickel concentration in the SWAT clam sites (Mast Cove, Eliot, 2004) was also found at the same site having the highest iron concentration, indicating sediment in the clam gut may be a contributing factor to nickel concentration in the samples.

Nickel occurs naturally in the environment and is an essential trace element to biological processes. Nickel from soil and weathering of rocks enters rivers and provides the largest source of nickel to coastal waters. Nickel occurs in stainless steel, nickel-cadmium batteries, pigments, computers, wire, coins, and is used in electroplating. Heightened nickel concentrations occur in the Great Lakes and speculation about sources centers on air deposition from a large nickel smelting operation in Ontario, Canada (Kimbrough et al. 2008).

Nickel is not thought to bioaccumulate in the food chain, however, nickel can be harmful to humans in large doses, inducing effects including bronchitis and even cancer from long term exposure (Kimbrough et al. 2008). The MCDC reports a non-cancer FTAL for nickel in non-commercially caught finfish of 43 ug/g wet weight (ppm), which is more conservative than the FDA action level for shellfish of 80 ug/g wet weight (ppm). The maximum mean concentration detected by SWAT in clam tissue is 1.5 ug/g wet wt.

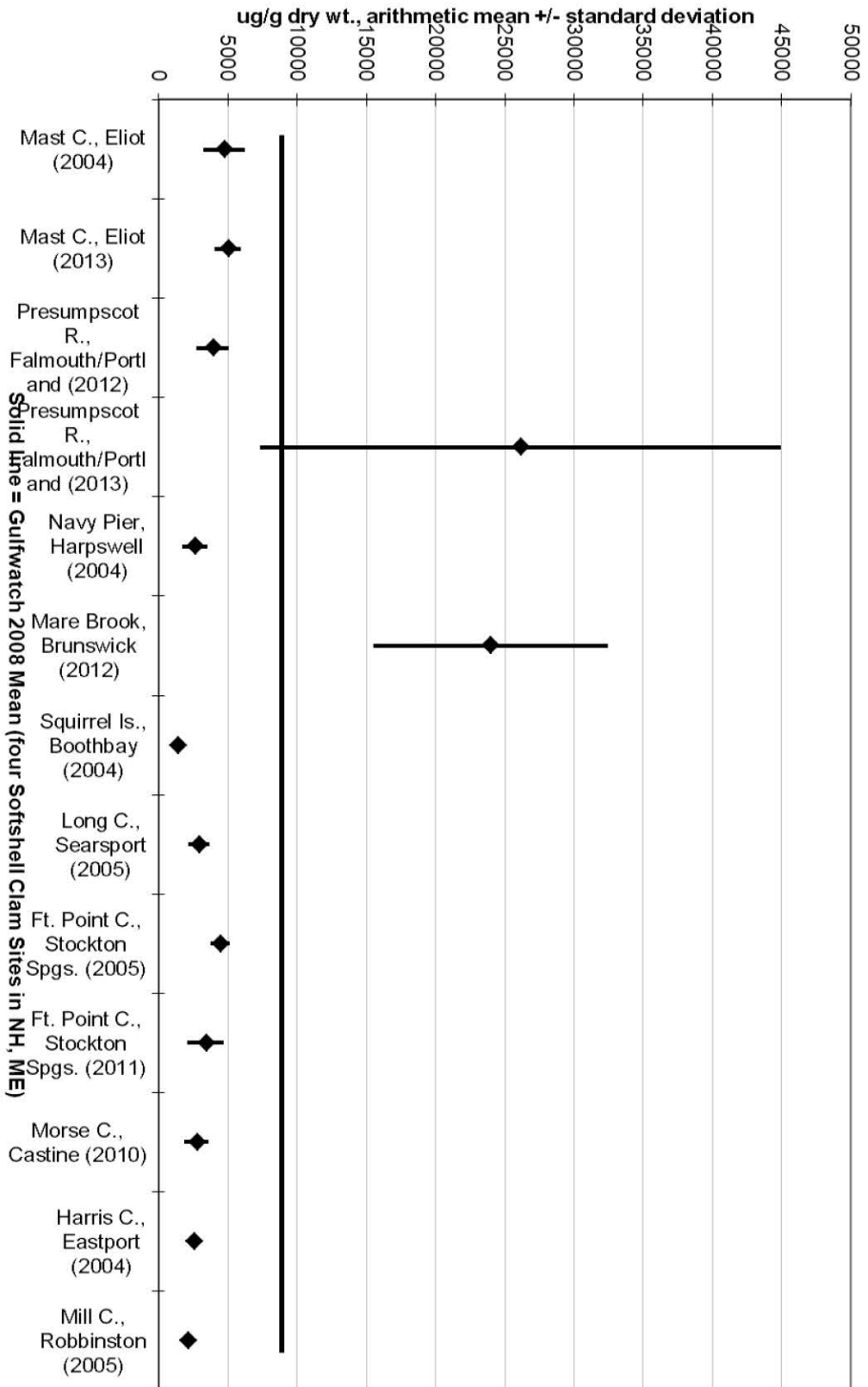


Figure 1.3.1.2.6.1: Iron in SWAT Softshell Clams

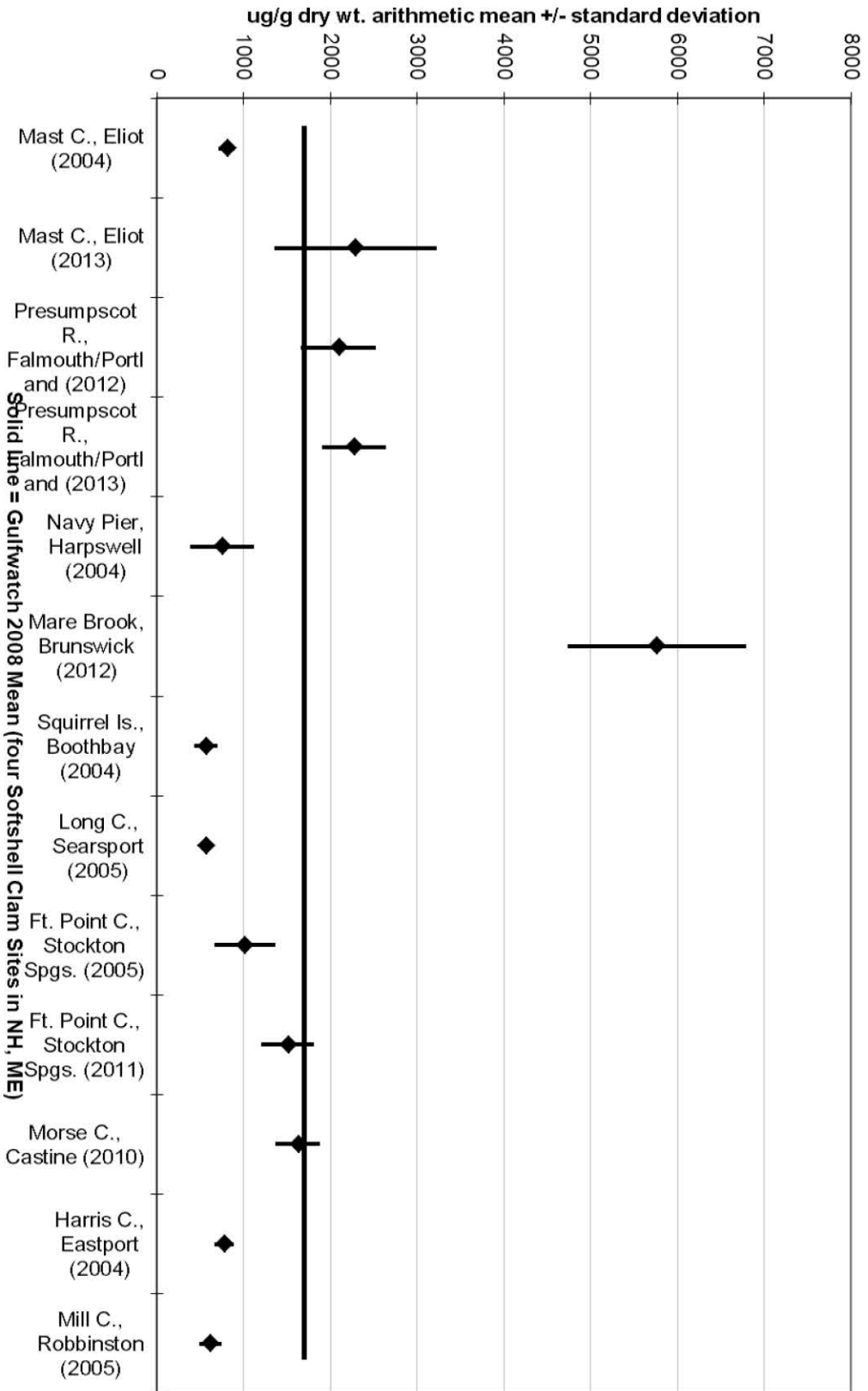


Figure 1.3.1.2.6.2: Aluminum in SWAT Softshell Clams

(ppm) at Mast Cove, Eliot, is an order of magnitude below the more conservative MCDC action level. MCDC does not report a cancer action level for nickel.

#### **1.3.1.2.8 Lead (Pb)**

Lead was detected at all ten SWAT softshell clam sites (Figure 1.3.1.2.8.1). Lead levels measured in clams ranged from a low mean concentration of 1.39 ug/g dry wt. at Navy Pier, Harpswell, to a high mean concentration of 10.73 ug/g dry wt. at Presumpscot River, Portland, 2013. Mean lead clam tissue concentrations at all but two SWAT sites fell below the 2008 Gulfwatch mean. Only Mare Brook and Presumpscot River (in 2013) had mean lead tissue concentrations exceeding the 2008 Gulfwatch mean. Lead concentrations at Mast Cove were similar or slightly higher in 2013 compared to 2004. The Presumpscot lead mean was higher in the western half of the estuary in 2013 compared to the eastern half of the estuary in 2012.

Lead occurs naturally in the earth's crust, however, global lead concentrations in the environment have increased in the last century due to the use of leaded gasoline. Reduction in lead loading through regulation of leaded gasoline and lead paints has occurred in recent decades. Elevated lead levels in the environment occur due to manufacturing, paints, lead solder, ammunition, plumbing, incineration and burning of fossil fuels. Lead loading in coastal waters is related to wastewater discharge, river runoff, atmospheric deposition, and natural weathering of crustal rock (Kimbrough 2008).

From a human health perspective, the FDA action level for lead in clams, oysters, and mussels is 1.7 ug/g wet wt. (ppm) (Kimbrough et al. 2008). The more conservative MCDC lead FTAL in non-commercially caught sportfish is 0.6 ug/g wet wt. (ppm), which is based on a blood lead concentration model. The highest mean concentration in the Maine SWAT softshell clam data is 1.65 ppm (ug/g) wet wt. in Presumpscot River (2013), followed by 1.413 ppm (ug/g) wet wt. at Mare Brook, Brunswick (2012), and then followed by 0.873 ppm wet wt. at Presumpscot River, Falmouth/Portland (2012). These three lead concentrations exceed the MCDC lead FTAL, as does Mast Cove in 2013 (0.871 ppm (ug/g) wet wt.), Harris Cove, Eastport (0.765 ppm (ug/g) wet wt.), and Fort Point Cove, Searsport (2005)(0.647 ug/g wet wt.). Mast Cove, Eliot, (0.597 ug/g wet wt.) is at the MCDC lead FTAL. The other five historic SWAT softshell clam sites fell below the more conservative MCDC lead FTAL, as did the 2011 Fort Point Cove clam tissue sample (0.52 ug/g wet wt.). One replicate of four at Fort Point Cove in 2011 scored 0.65 ug/g wet wt. indicating considerable variability in the lead tissue concentrations, with some falling on either side of the MCDC lead FTAL.

The MCDC FTAL is based on the consumer eating an 8 oz. meal. Maine SWAT data indicates that an 8 oz. meal would include approximately 21 softshell clams of the size tested by the SWAT program.

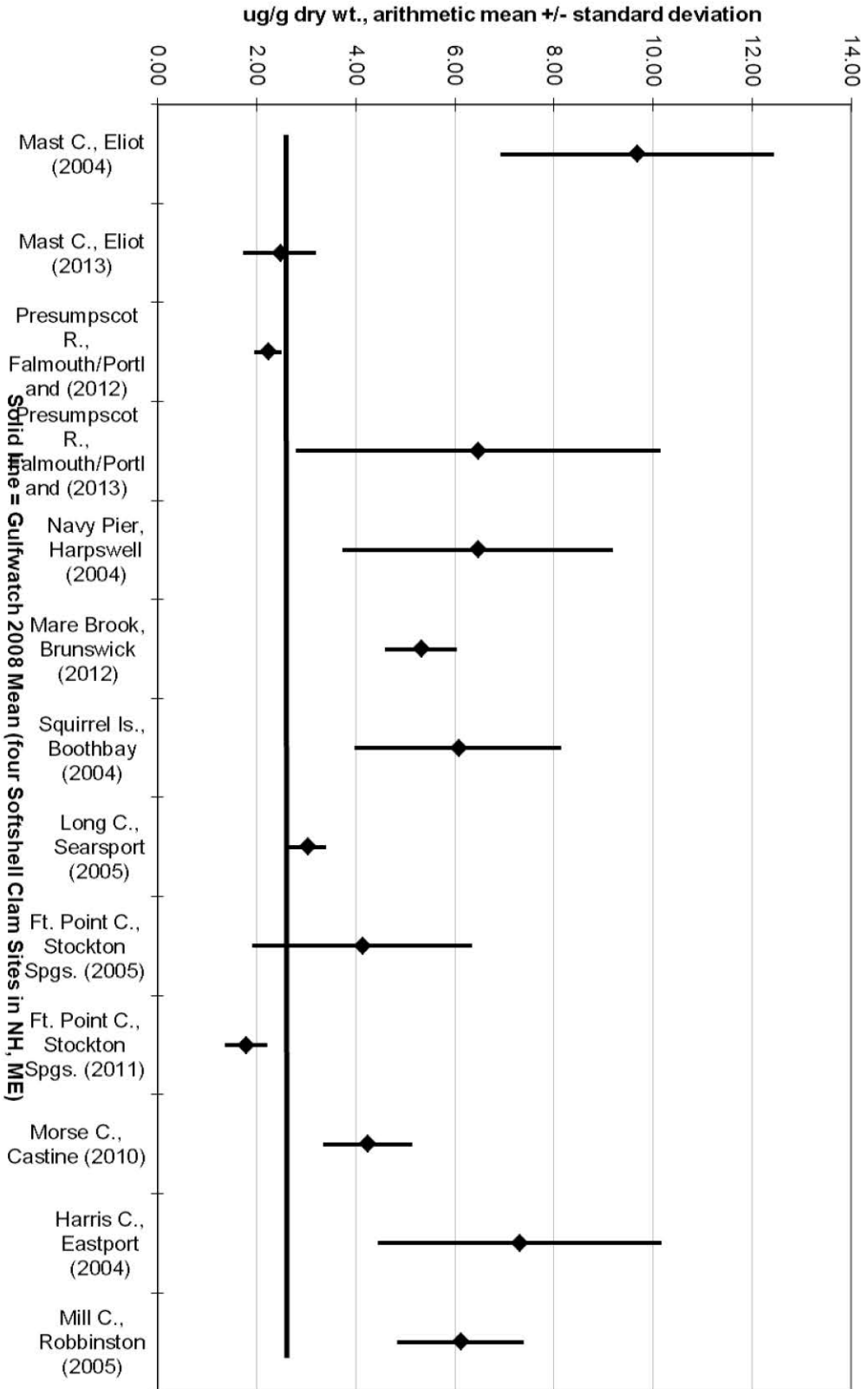


Figure 1.3.1.2.7.1: Nickel in SWAT Softshell Clams



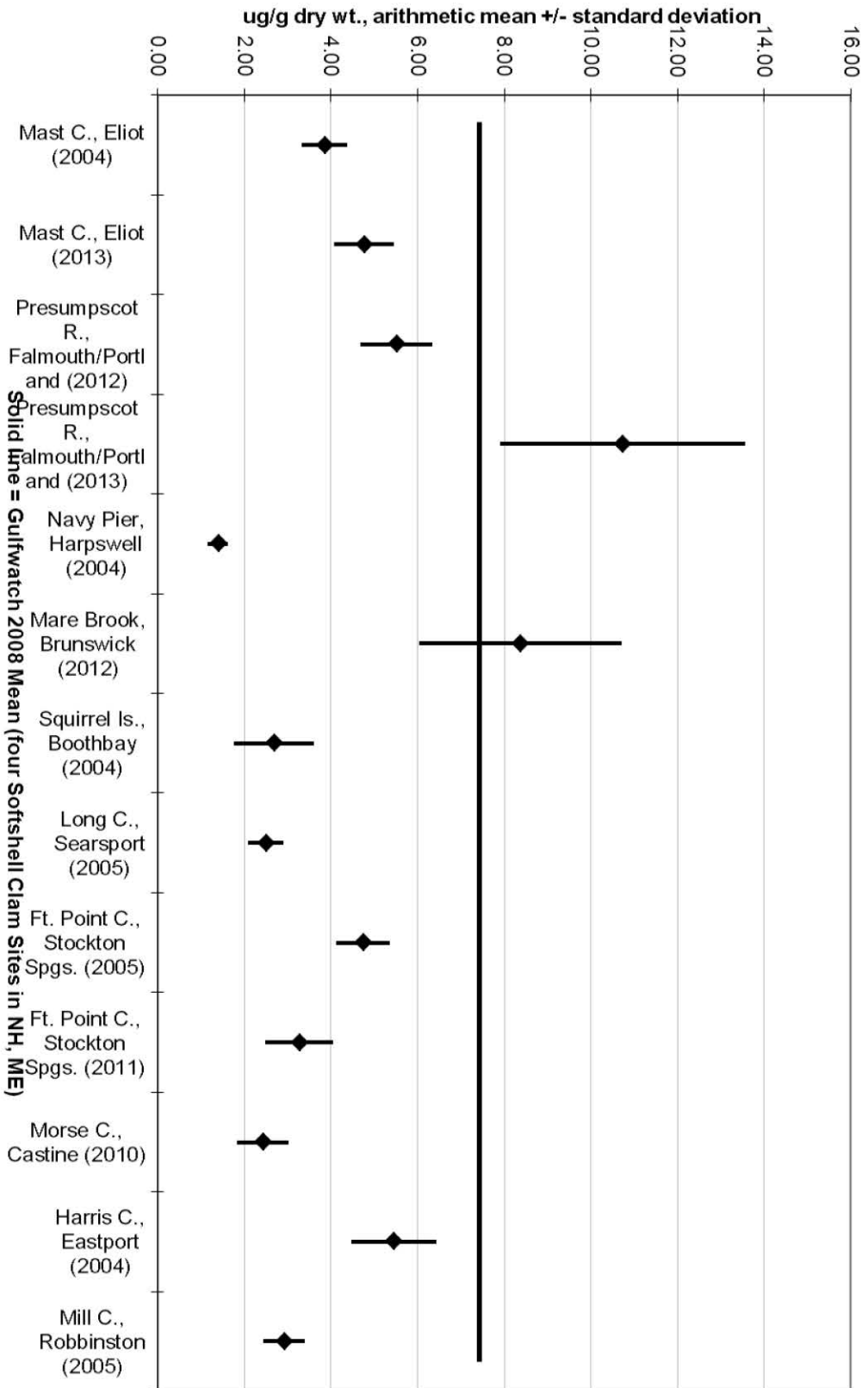


Figure 1.3.1.2.8.1: Lead in SWAT Softshell Clams

#### 1.3.1.2.9 Mercury (Hg)

Mercury was detected at all ten softshell clam locations (Figure 1.3.1.2.9.1). Mercury levels measured in clams ranged from a low mean concentration of 0.06  $\mu\text{g/g}$  dry wt. at Harris Cove, Eastport, to a high mean concentration of 0.64  $\mu\text{g/g}$  dry wt. at Fort Point Cove, Stockton Springs (2005). High mercury concentrations in a variety of matrices have been documented in the Penobscot and are likely associated with the chloralkali process employed at the former Holtrachem site. Five sites had clam tissue concentrations that exceeded the 2008 Gulfwatch mean: Presumpscot River, Falmouth/Portland (2012, 2013), Mast Cove, Eliot (2004, 2013), Long Cove, Searsport (2005), Fort Point Cove, Stockton Springs (2005, 2011), and Morse Cove, Castine (2010) (Figure 1.3.1.2.9.1).

Mercury occurs naturally in the environment; however elevated levels are associated with anthropogenic sources. United States sources of mercury to the air include coal fired electrical power generation, incinerators, mining, landfills, and sewage sludge (Kimbrough et al. 2008). From a human health perspective, the developmental methylmercury FTAL (more protective) used by the MCDC is 0.2  $\mu\text{g/g}$  (ppm) wet wt. for non-commercially caught finfish (fish filet). This FTAL assumes an 8 oz. meal size is consumed weekly. Maine SWAT data uses a total mercury value, which is a more complete measure of mercury than the methylmercury concentration, but includes this more toxic form. Total mercury is therefore a more protective measurement than methylmercury alone. The highest mean softshell clam total tissue mercury concentration measured by SWAT in this Maine data set was 0.088  $\mu\text{g/g}$  wet wt. (ppm) at Fort Point Cove, Stockton Springs in 2005 (note 2011 concentration appears to be somewhat lower, which may be due to patchiness of contaminants, sampling variability, or inter-annual variability). The 2005 concentration compares favorably with the MCDC methylmercury developmental FTAL of 0.2 ppm (less than half of the FTAL), assuming a similar meal size and frequency. To consume approximately 8oz. of tissue the consumer would need to eat approximately 21 softshell clams based on the mean mass per clam collected by the SWAT program.

#### 1.3.1.2.10 Zinc (Zn)

Zinc was detected at all ten clam sample locations (Figure 1.3.1.2.10.1). Zinc levels measured in clams ranged from a low mean concentration of 56.1  $\mu\text{g/g}$  dry wt. at Squirrel Island, Boothbay in 2004, to a high mean concentration of 89.9  $\mu\text{g/g}$  dry wt. at Presumpscot River, Falmouth/Portland, in 2012. Zinc concentrations were similar at Presumpscot River in 2012 and 2013, Mast Cove in 2004 and 2013, and Fort Point Cove in 2005 and 2011. All ten of the SWAT clam sites had zinc tissue concentrations that fell below the 2008 Gulfwatch mean.

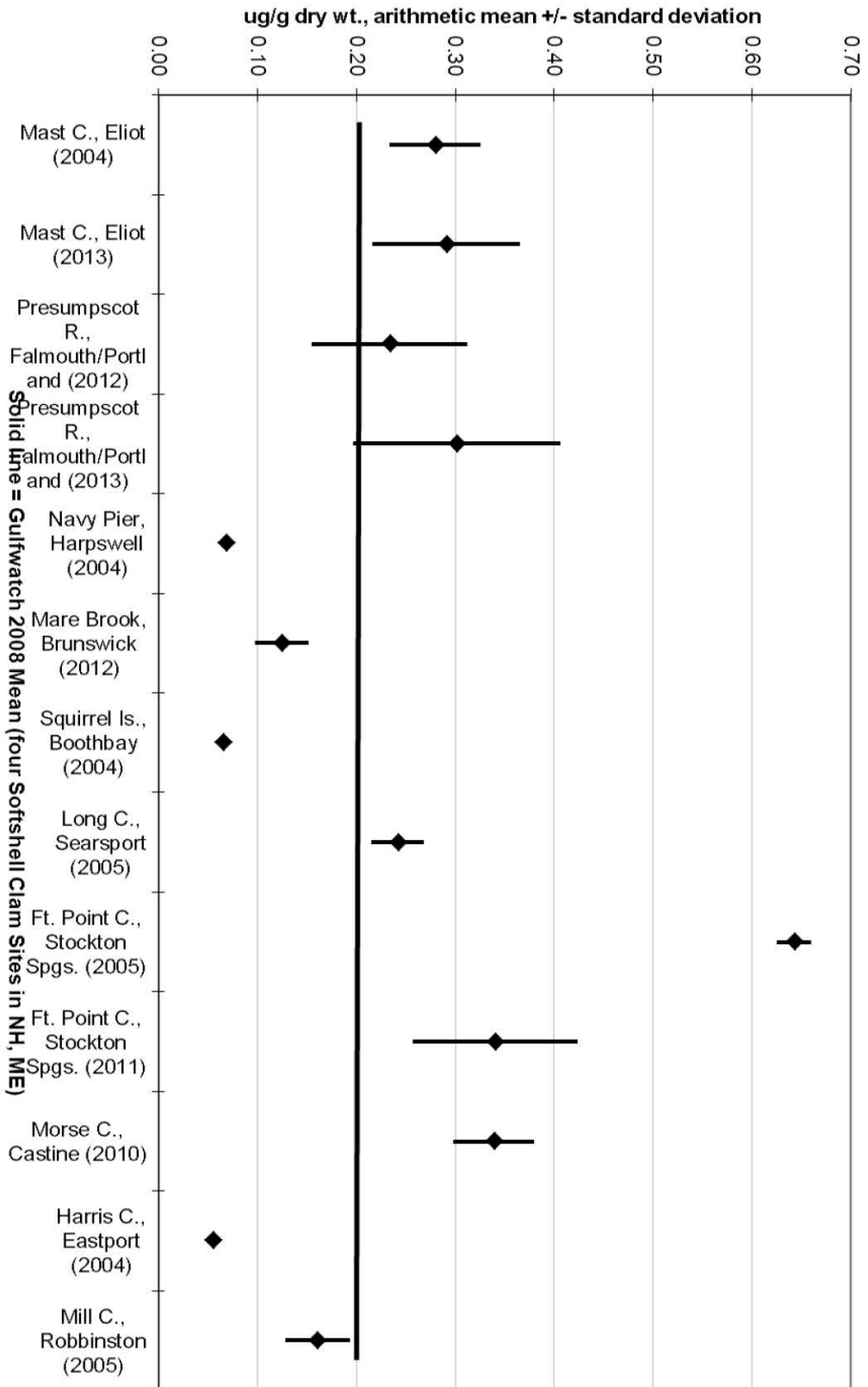


Figure 1.3.1.2.9.1: Mercury in SWAT Softshell Clams

Zinc is widespread in its distribution but elevated levels primarily originate from a variety of human activities including vehicle tire wear, electroplating and galvanized metals, industrial wastes, and drainage from mining (Kimbrough et al 2008). Though an essential nutrient at low levels, higher doses to humans can cause anemia or pancreatic and kidney damage. Since humans do not bioaccumulate zinc, health impacts are normally associated with high doses. From a human health perspective, MCDC reports a non-cancer FTAL for zinc of 648 ug/g wet wt. (ppm), which is more than an order of magnitude higher than any wet wt. concentrations observed in SWAT clam tissue. There is no recommended FDA safety level for zinc in fish (Kimbrough et al. 2008).

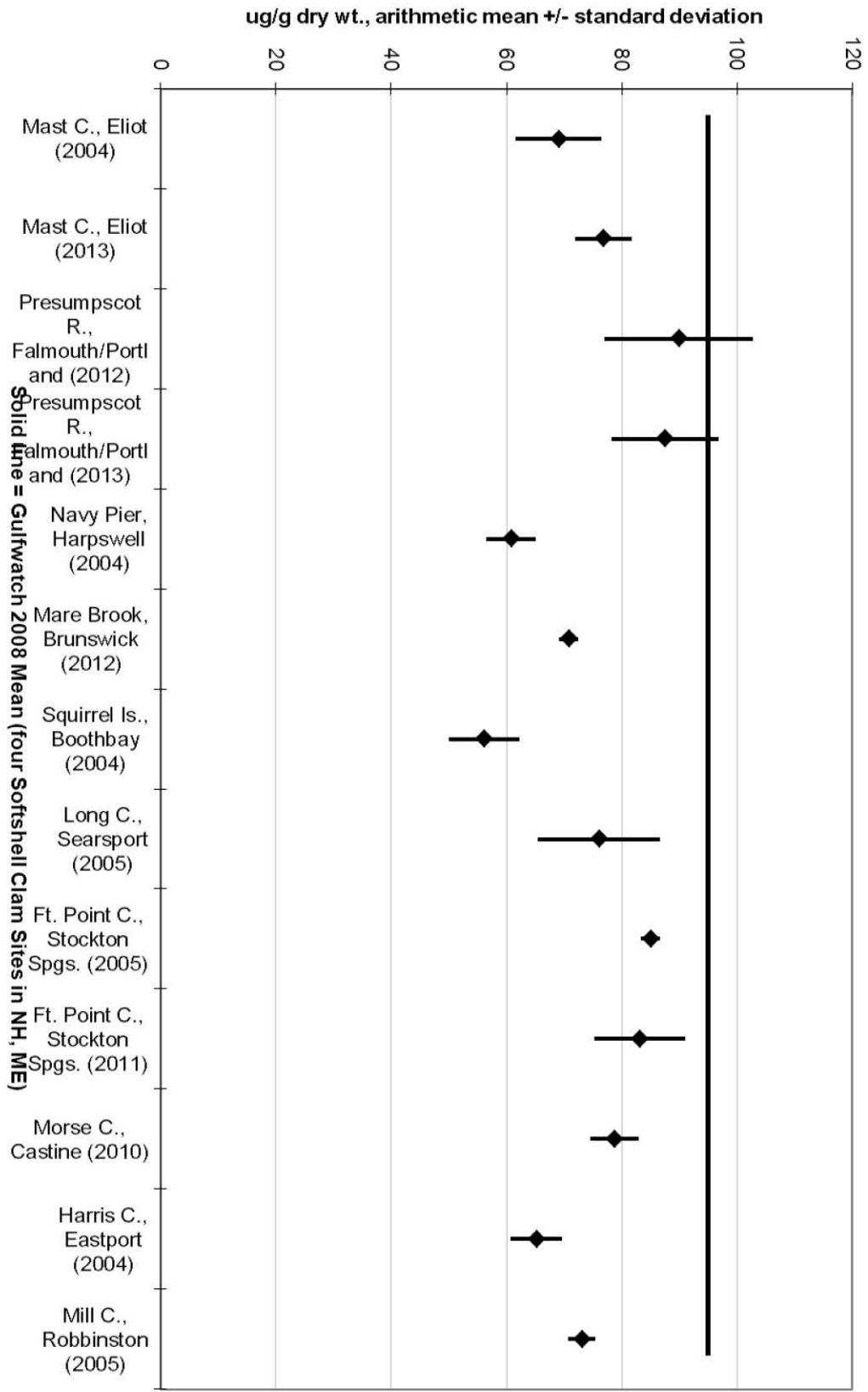


Figure 1.3.1.2.10.1: Zinc in SWAT Softshell Clams

### **1.3.2 Polycyclic Aromatic Compounds**

PAHs occur in elevated concentrations near petroleum manufacturing, creosote use, and burning wood (Kimbrough et al. 2008). Though there are natural sources, including forest fires and volcanoes, anthropogenic sources, including automobile emissions, home heating, and coal-fired power plants, contribute to elevated levels of PAHs. As their name implies, polycyclic aromatic hydrocarbons are made of fused benzene rings, fusion of which may occur during combustion. However, they also occur in uncombusted coal and oil. PAHs in the environment are primarily from forest fires, coal-fired power plants, automobile exhaust, and spilled oil (Kimbrough et al. 2008).

#### **1.3.2.1 Blue Mussels**

Results were compared to national (NS&T) (Kimbrough et al. 2008) and Gulf of Maine (Gulfwatch) (LeBlanc et al. 2009) blue mussel monitoring program data (when available) in an effort to place Maine SWAT data in a national and regional context, respectively.

The NS&T and Gulfwatch programs utilize a subset of PAHs, summing results from 19, 24 and 40 individual PAHs to construct groups of PAHs to assess overall PAH concentrations and to compare regional and national concentrations. Smaller subsets of PAHs were utilized historically as a substitute for more complete sets as a cost saving measure. This report utilizes the Maine SWAT blue mussel tissue PAH data generated by AXYS Analytical, which includes 75 individual and summed alkylated PAHs. To compare Maine results to the NS&T and Gulfwatch lists of 19 unsubstituted (non-alkylated) PAHs, this report sums 19 unsubstituted (non-alkylated) PAHs from 2012 SWAT data. The summation of 19 PAHs is also useful for comparison to SWAT PAH data sets prior to 2009, as previous SWAT data included only 24 individual PAHs.

Both the Gulfwatch and NS&T programs utilize a summation of 24 PAHs, which in addition to the 19 non-alkylated PAHs previously mentioned also includes some alkylated PAHs (C1, C2, C3 Naphthalene, and C1-Phenanthrene). The 2013 SWAT PAH data can also be used to generate a summation to compare to the Gulfwatch/NS&T summation of 40 PAHs, which includes even more alkylated PAHs. The corresponding SWAT data includes 39 PAHs, which is the closest approximation possible. The difference in the 40 PAH summation is the absence of C4-Flourenes in the SWAT data set. As a result, the SWAT summation includes 39 PAHs, rather than the 40 utilized in the Gulfwatch/NS&T programs. This difference is considered to be relatively minor, and with some caution in interpretation, still allows comparison of SWAT data to regional and national data sets.

SWAT 2013 PAH data include additional alkylated PAHs as well, with a total of 75 PAHs included. This number has also been totaled and is presented and discussed in this report as “total PAHs.” Comparisons to other summations of lesser numbers of PAHs reviewed above are included to illustrate the wider data set provided by the additional level of PAH analysis obtained for SWAT sites in recent years, including 2010-13. Alkylated PAHs are typically associated with pyrogenic sources, rather than the more petrogenic sources associated with non-alkylated PAHs.

Table 1.3.2.1.1, “Analyzed PAHs and PAH Summation Calculations” shows comparisons between Gulfwatch/NS&T summation lists and SWAT summation lists, and details differences between the lists with footnotes and notes in the right column of the table.

Figure 1.3.2.1.1 shows the summation of the 19 non-alkylated PAHs, 24 PAHs, and 40 PAHs compared to the summation of all 75 (“total”) PAHs (including many alkylated PAHs) at the two 2013 SWAT blue mussel sites, East End Beach, Portland, and Sears Island, Searsport.. Both the 19 summed non-alkylated PAHs and the total PAHs vary in a similar manner between sites, and the non-alkylated PAHs make up a small fraction of the total PAHs found at each site. The alkylated PAHs contribute the largest portion to the total PAHs, which is the difference between the sum of 19 PAHs and the total PAHs illustrated in Figure 1.3.2.1.1.

Total mean PAH concentrations were 1219 ng/g dry wt. at East End Beach, Portland, and 470 ng/g dry wt. at Sears Island, Searsport (Figure 1.3.2.1.1). The sum of 19 non-alkylated PAHs was 295 ng/g dry wt. at East End Beach and 87 ng/g dry wt. at Sears Island. The Gulfwatch program also utilized a summation of 24 PAHs in reports, the composition of which is outlined above. SWAT data were converted into this format and when 24 PAHs were summed, the mean concentrations for the sum of 24 PAHs were 366 ng/g dry wt. at East End Beach and 119 ng/g dry wt. at Sears Island (Figure 1.3.2.1.1).

Figure 1.3.2.1.1 also shows the summation of 40 PAHs compared to the summation of all 75 PAHs (Total PAHs) at the 2013 SWAT blue mussel sites. Both the 40 summed PAHs and the total PAHs vary in a similar manner between sites, but the sum of the 40 PAHs makes up the bulk of the total PAHs found at each site. The mean concentrations for the sum of 40 PAHs were 796 ng/g dry wt. at East End Beach and 246 ng/g dry wt. at Sears Island (Figure 1.3.2.1.1).

**TABLE 1.3.2.1.1: Analyzed PAHs and PAH Summation Calculations**

Gulfwatch, NS&amp;T, SWAT

Parameter	SWAT				Summations			Not Analyzed By Gulfwatch	Notes (See below list for more notes)
	2012- 13	2010- 11	2007- 08	2004- 05	ΣPAH19	ΣPAH24	ΣPAH40		
ACENAPHTHENE	x	x	x	x	x	x	x		
ACENAPHTHYLENE	x	x	x	x	x	x	x		
ANTHRACENE	x	x	x	x	x	x	x		
2-METHYLANTHRACENE	x	x						missing	
BENZ[A]ANTHRACENE	x	x	x	x	x	x	x		
DIBENZ(A,H)ANTHRACENE	x	x	x	x	x	x	x		
BIPHENYL	x	x	x	x	x	x	x		
BENZO[A]PYRENE	x	x	x	x	x	x	x		
BENZO(E)PYRENE	x	x	x	x	x	x	x		
7-METHYLBENZO[A]PYRENE	x	x						missing	
CHRYSENE	x	x	x	x	x	x	x		
1-METHYLCHRYSENE	x	x						missing	
5/6-METHYLCHRYSENE	x	x						missing	
5,9-DIMETHYLCHRYSENE	x	x						missing	
DIBENZOTHIOPHENE	x	x	1,2,3		x	x	x		
2,4-DIMETHYLDIBENZOTHIOPHENE	x	x						missing	
2/3-METHYLDIBENZOTHIOPHENES	x	x						missing	
FLUORANTHENE	x	x	x	x	x	x	x		
BENZO[B]FLUORANTHENES	x								SWAT split in 2012 from (B,J,K)
BENZO[J,K]FLUORANTHENES	x								SWAT split in 2012 from (B,J,K)
BENZO[B,J,K]FLUORANTHENES		x	x		x	x	x		in Gulfwatch list as BENZO[B]FLUORANTHENE BENZO[K]FLUORANTHENE
3-METHYLFLUORANTHENE/BENZO[A]FLUORENE	x	x							
FLUORENE	x	x	x	x	x	x	x		
2-METHYLFLUORENE	x	x						missing	
1,7-DIMETHYLFLUORENE	x	x						missing	



**TABLE 1.3.2.1.1: Analyzed PAHs and PAH Summation Calculations (continued)**  
 Gulfwatch, NS&T, SWAT

Parameter	SWAT				Summations			Not Analyzed By Gulfwatch	Notes (See below list for more notes)
	2012- 13	2010- 11	2007- 08	2004-05	ΣPAH19	ΣPAH24	ΣPAH40		
NAPHTHALENE	x	x	x	x	x	x	x		
1-METHYLNAPHTHALENE	x	x	x					missing	
2-METHYLNAPHTHALENE	x	x	x					missing	
1,2-DIMETHYLNAPHTHALENE	x	x						missing	
2,6-DIMETHYLNAPHTHALENE	x	x	x					missing	
2,3,5-TRIMETHYLNAPHTHALENE	x	x	x					missing	
2,3,6-TRIMETHYLNAPHTHALENE	x	x						missing	
1,4,6,7-TETRAMETHYLNAPHTHALENE	x	x						missing	
PERYLENE	x	x	x	x		x	x		
BENZO[GHI]PERYLENE	x	x	x	x	x	x	x		
PHENANTHRENE	x	x	x	x	x	x	x		
1-METHYLPHENANTHRENE	x	x	x					missing	
2-METHYLPHENANTHRENE	x	x						missing	
3-METHYLPHENANTHRENE	x	x						missing	
9/4-METHYLPHENANTHRENE	x	x						missing	
1,7-DIMETHYLPHENANTHRENE	x	x						missing	
1,8-DIMETHYLPHENANTHRENE	x	x						missing	
2,6-DIMETHYLPHENANTHRENE	x	x						missing	
3,6-DIMETHYLPHENANTHRENE	x	x						missing	
1,2,6-TRIMETHYLPHENANTHRENE	x	x						missing	
PYRENE	x	x	x	x	x	x	x		
INDENO[1,2,3-CD]PYRENE	x	x	x	x	x	x	x		

**TABLE 1.3.2.1.1: Analyzed PAHs and PAH Summation Calculations (continued)**

Gulfwatch, NS&amp;T, SWAT

Parameter	SWAT				SWAT Summations			Not Analyzed By Gulfwatch	Notes (See below list for more notes)
	2012-13	2010-11	2007-08	2004-05	ΣPAH19	ΣPAH24	ΣPAH40		
RETENE	x	x						missing	
C1-ACENAPHTHENES	x	x						missing	
C1-BENZO[A]ANTHRACENES/CHRYSENES	x	x	3				x		in Gulfwatch list as C1-CHRYSENE
C2-BENZO[A]ANTHRACENES/CHRYSENES	x	x	3				x		in Gulfwatch list as C2-CHRYSENE
C3-BENZO[A]ANTHRACENES/CHRYSENES	x	x	3				x		in Gulfwatch list as C3-CHRYSENE
C4-BENZO[A]ANTHRACENES/CHRYSENES	x	x	3				x		in Gulfwatch list as C4-CHRYSENE
C1-BENZOFLUORANTHENES/BENZOPYRENES	x	x						missing	
C2-BENZOFLUORANTHENES/BENZOPYRENES	x	x						missing	
C1-BIPHENYLS	x	x						missing	
C2-BIPHENYLS	x	x						missing	
C1-DIBENZOTHIOPHENES	x	x	3				x		
C2-DIBENZOTHIOPHENES	x	x	3				x		
C3-DIBENZOTHIOPHENES	x	x	3				x		
C4-DIBENZOTHIOPHENES	x	x						missing	
C1-FLUORANTHENES/PYRENES	x	x	3				x		
C2-FLUORANTHENES/PYRENES	x	x	3				x		
C3-FLUORANTHENES/PYRENES	x	x						missing	
C4-FLUORANTHENES/PYRENES	x	x						missing	
C1-FLUORENES	x	x	3				x		
C2-FLUORENES	x	x	3				x		
C3-FLUORENES	x	x	3				x		
C1-NAPHTHALENES	x	x	2,3			x	x		
C2-NAPHTHALENES	x	x	2,3			x	x		
C3-NAPHTHALENES	x	x	2,3			x	x		
C4-NAPHTHALENES	x	x						missing	

**TABLE 1.3.2.1.1: Analyzed PAHs and PAH Summation Calculations (continued)**  
 Gulfwatch, NS&T, SWAT

Parameter	SWAT				SWAT Summations			Not Analyzed By Gulfwatch	Notes (See below list for more notes)
	2012-13	2010-11	2007-08	2004-05	ΣPAH19	ΣPAH24	ΣPAH40		
C1-PHENANTHRENES/ANTHRACENES	x	x	2,3			x	x		in Gulfwatch list as C1-PHENANTHRENE
C2-PHENANTHRENES/ANTHRACENES	x	x	3				x		in Gulfwatch list as C2-PHENANTHRENE
C3-PHENANTHRENES/ANTHRACENES	x	x	3				x		in Gulfwatch list as C3-PHENANTHRENE
C4-PHENANTHRENES/ANTHRACENES	x	x	3				x		in Gulfwatch list as C4-PHENANTHRENE
C4-FLUORENES			3				x		Not analyzed by SWAT

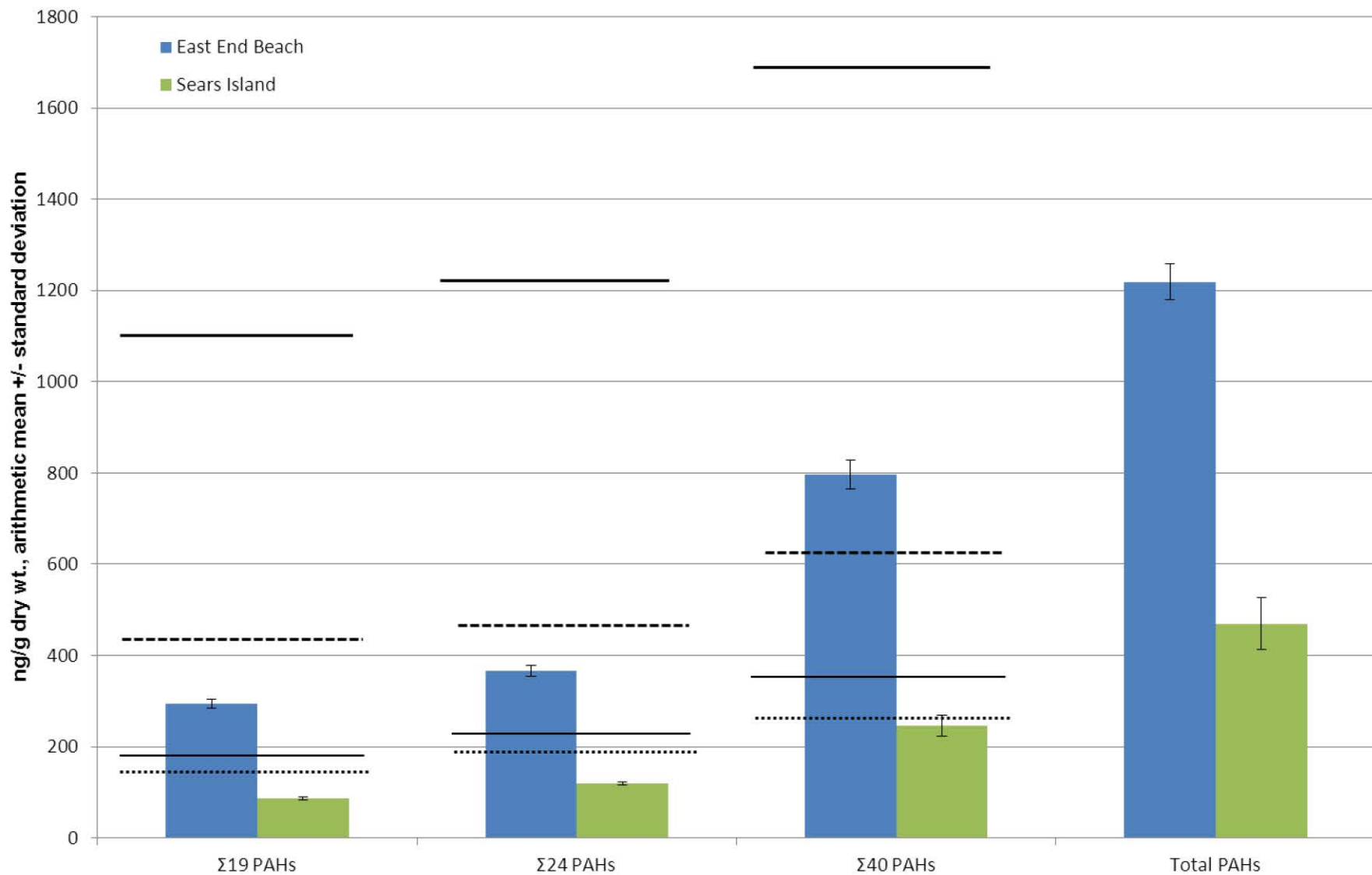
#### FOOTNOTES:

Prior to 2012: List of 'Sum PAH19' only has 18 compounds in it because we have BENZO[B]FLUORANTHENES and BENZO[K]FLUORANTHENES listed as one compound, BENZO[B,J,K]FLUORANTHENES; same applies to 'Sum PAH24' which has only 23 compounds. For 2012: List of 'Sum PAH19' has 19 compounds in it because we have BENZO[B]FLUORANTHENES and BENZO[J,K]FLUORANTHENES listed as two compounds; Same applies to 'Sum PAH24' which now has 24 compounds.

Prior to 2012: List of 'Sum PAH40' only has 38 compounds in it because we have BENZO[B]FLUORANTHENES and BENZO[K]FLUORANTHENES listed as one compound, BENZO[B,J,K]FLUORANTHENES and we do not have SWAT/AXYS data for C-4 FLUORENES (at bottom of above list). For 2012: List of 'Sum PAH40' has 39 compounds in it because we have BENZO[B]FLUORANTHENES and BENZO[J,K]FLUORANTHENES listed as two compounds, though we still do not have SWAT/AXYS data for C-4 FLUORENES (at bottom of above list)

In calculating the various summations, the approach used by SWAT is: Where SWAT has a slight variation from Gulfwatch in analytes, use the closest approximation to the Gulfwatch list as with the BENZO[B,J,K]FLUORANTHENES (prior to 2012), and the C1/2/3/4-BENZO[A]ANTHRACENES

**Figure 1.3.2.1.1: PAHs in Blue Mussels**



Dashed lines = 2008 Gulfwatch Median and 85th Percentile; Solid lines = National Status and Trends Median and 85th Percentile.

Figure 1.3.2.1.1 compares the sum of 19 PAHs at the SWAT blue mussel sites sampled in 2013 to the Gulfwatch 2008 median and 85<sup>th</sup> percentile results. The sum of 19 PAHs from mussel tissue at Sears Island was below the Gulfwatch median, while the sum of 19 PAHs from East End Beach exceeded the Gulfwatch median (154 ng/g dry weight) but did not exceed the Gulfwatch 85<sup>th</sup> percentile (429 ng/g dry weight). The summation of non-alkylated PAHs is useful for putting Maine data into a regional, Gulf of Maine context. Figure 1.3.2.1.1 also compares the sum of 19 non-alkylated PAHs at the 2013 SWAT blue mussel sites to recent NS&T median and 85<sup>th</sup> percentile for 19 summed non-alkylated PAHs (2008 data, the most recent available). Sears Island did not exceed the 2008 NS&T median of 180 ng/g (dry weight) for 19 summed non-alkylated PAHs, but East End Beach did. Neither of the SWAT mussel sites approached or exceeded the NS&T 85<sup>th</sup> percentile of 1,104 ng/g (dry weight) for 19 summed PAHs.

Figure 1.3.2.1.1 compares the sum of 24 PAHs at the SWAT blue mussel sites sampled in 2013, to the Gulfwatch 2008 median and 85<sup>th</sup> percentile results. Sears Island was below the Gulfwatch 2008 median of 198 ng/g (dry weight) for 24 summed PAHs. East End Beach exceeded the Gulfwatch median, but was below the Gulfwatch 85<sup>th</sup> percentile of 476 ng/g (dry weight) for 24 summed PAHs. The summation of these PAHs is useful for putting Maine data into a regional, Gulf of Maine context. Figure 1.3.2.1.1 also compares the sum of 24 PAHs at the 2013 SWAT blue mussel sites to recent NS&T median and 85<sup>th</sup> percentile for 24 summed PAHs (2008 data, the most recent available). Sears Island was below the NS&T 2008 median of 247 ng/g (dry weight) for 24 summed PAHs, while East End Beach exceeded it. Neither site approached or exceeded the NS&T 85<sup>th</sup> percentile of 1,216 ng/g (dry weight) for 24 summed PAHs.

Figure 1.3.2.1.1 compares the sum of 40 PAHs at the SWAT blue mussel sites sampled in 2013 to the Gulfwatch 2008 median and 85<sup>th</sup> percentile results. Sears Island had a sum of 40 PAHs below the Gulfwatch 2008 median of 260 ng/g (dry weight) for 40 summed PAHs, while the sum of 40 PAHs at East End Beach exceeded the Gulfwatch 85<sup>th</sup> percentile of 618 ng/g (dry weight) for 40 summed PAHs.

The differences between the SWAT list of PAHs and the Gulfwatch list of PAHs available for the sum of 40 PAHs may be part of the reason why the SWAT sum of 40 PAHs is comparably high to the Gulfwatch sum of 40 PAHs. As noted in Table 1.3.2.1.1, SWAT utilizes C1 through C4-Benzo[A]Anthracenes/Chrysenes, where Gulfwatch utilizes C1 through C4-Chrysenes. Similarly, SWAT utilizes C1 through C4-Phenanthrenes/Anthracenes, where Gulfwatch utilizes C1 through C4-Phenanthrenes. It is likely that the additional summations of C1 through C4-Benzo[A]Anthracenes plus C1 through C4-Anthracenes included in the SWAT data are pushing the SWAT sum of 40 PAHs higher than the Gulfwatch equivalents. This result cannot be avoided due to the composition of the SWAT data, but should be noted when viewing the comparison in Figure 1.3.2.1.1.

Figure 1.3.2.1.1 also compares the sum of 40 PAHs at the 2013 SWAT mussel sites to recent NS&T median and 85<sup>th</sup> percentile for 40 summed PAHs (2008 data, the most recent available). Sears Island had a sum of 40 PAHs below the NS&T 2008 median, while East End Beach exceeded the NS&T 2008 median of 353 ng/g (dry weight) for 40 summed PAHs. East End Beach exceeded the NS&T 85<sup>th</sup> percentile of 1,674 ng/g (dry weight) for 40 summed PAHs.

The differences between the SWAT list of PAHs and the NS&T list of PAHs available for the sum of 40 PAHs may contribute significantly to the relatively higher concentration apparent at East End Beach when compared to the NS&T (same as Gulfwatch) sum of 40 PAHs. These differences are explored in depth in a preceding paragraph. This result cannot be avoided due to the composition of the SWAT data, but should be noted when viewing the comparison in Figure 1.3.2.1.1.

For 2013 SWAT blue mussel sites, Figure 1.3.2.1.2 presents a graphic representation of selected PAHs expressed as a ratio. The equation used to derive the ratio is:

$$\text{Fluoranthene} + \text{Pyrene} / \Sigma(\text{Fluoranthene} + \text{Pyrene} + \text{C2-C4 Alkylphenanthrene})$$

This equation is utilized to show relative concentrations of non-alkylated to alkylated PAHs, which yields a ratio indicating that values <0.1 are interpreted as a petrogenic (unburned fuel or petroleum) source, while values >0.2 are interpreted as a pyrogenic (combusted fuel) source of PAHs. Both SWAT blue mussel sites tested in 2013, East End Beach, Portland, and Sears Island, Searsport, have ratios above the >0.2 mark, which would indicate a pyrogenic source of PAHs. The higher ratio in the two 2013 SWAT blue mussel sites may be attributed to the urbanized upland area (East End Beach, Portland, and Searsport that is downstream from Bangor/Brewer, major roads, etc.) with associated impervious surfaces and combusted hydrocarbon runoff or ship and boat emissions.

Toxicities of PAHs vary, with hundreds of compounds making up the pool of PAHs. Toxic responses in aquatic organisms may include reproductive inhibition, mutations, liver abnormalities, and even mortality. Exposure in the marine environment may be from spilled oil, boat exhaust, and runoff from urban areas. From a human health perspective, neither MCDC nor FDA have reported recommended safety levels for PAHs in fish or fish products (Kimbrough et al. 2008).

### 1.3.2.2 Softshell Clams

Results were compared to national (NS&T) (Kimbrough et al. 2008) shellfish data and Gulf of Maine (Gulfwatch) (LeBlanc et al. 2009) softshell clam data (when available) in an effort to place Maine SWAT data in a national and regional context, respectively. Differences in individual PAHs obtained from different laboratories and different years are described in depth in the previous section 1.3.2.1.. The same approach was utilized to develop lists of PAHs in clam tissues presented in this section. Comparisons were made to NS&T and Gulfwatch programs when data sets were available.

Table 1.3.2.1.1, “Analyzed PAHs and PAH Summation Calculations” shows comparisons between Gulfwatch/NS&T summation lists and SWAT summation lists, and details differences between the lists with footnotes and notes in the right column of the table.

Figure 1.3.2.2.1 shows the summation of the 19 non-alkylated PAHs at the two SWAT clam sites sampled in 2013: Mast Cove, Eliot, and Presumpscot River, Falmouth/Portland. The sum of 19 non-alkylated PAHs was 262 ng/g dry wt. at Mast Cove and 1223 ng/g dry wt. at Presumpscot River. Presumpscot River exceeded the 2008 Gulfwatch mean concentration for the sum of 19 non-alkylated PAHs (420 ng/g dry wt.), which was calculated for four sites (two in NH and two in ME).

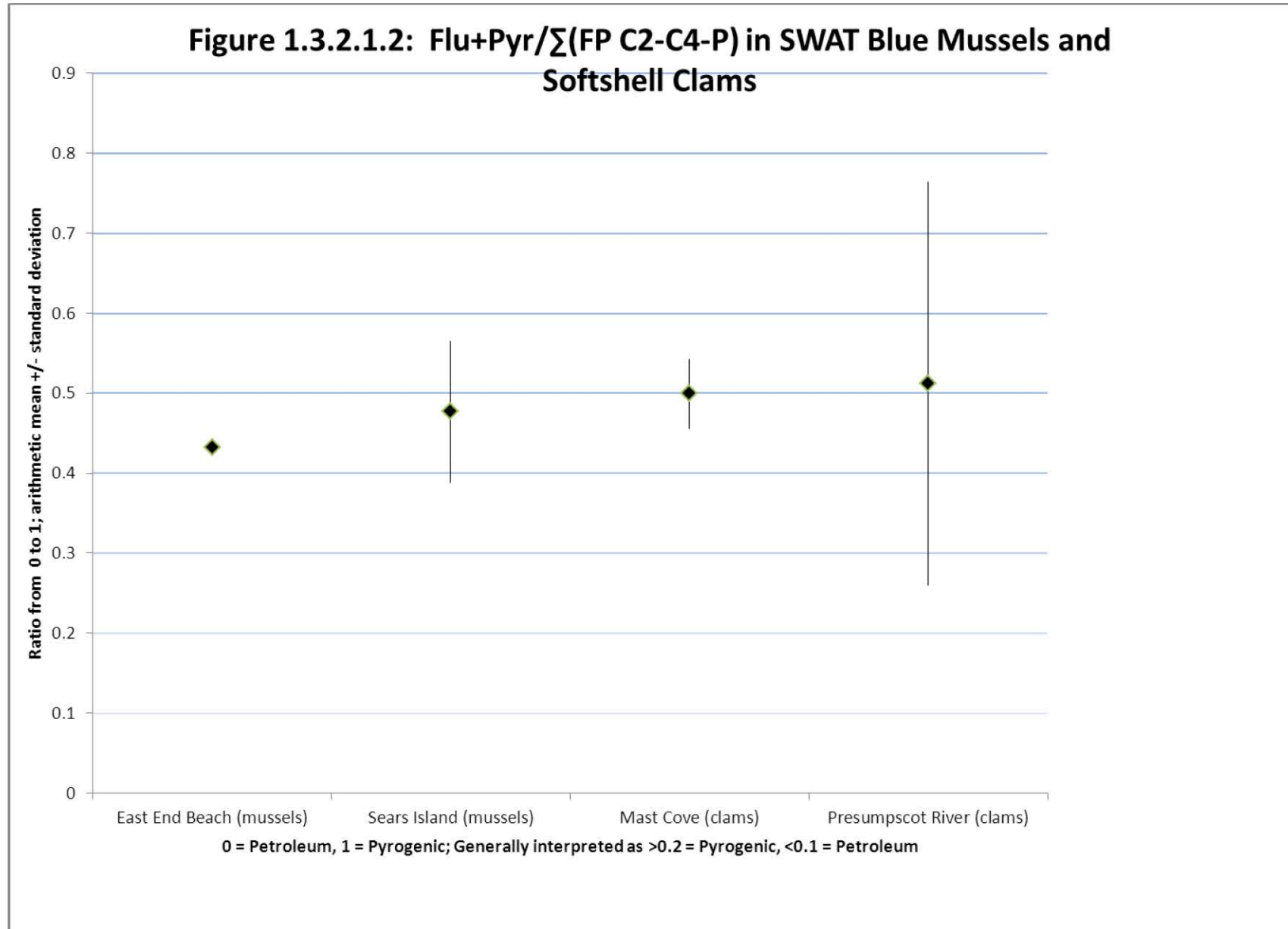
In addition to the summation for 19 non-alkylated PAHs, Figure 1.3.2.2.1 includes summations of 24, 40, and total PAHs. Summations of 24 PAHs at Presumpscot River exceeded the Gulfwatch mean, while the concentration in Mast Cove clam tissue did not. The sum of 40 PAHs concentration in Presumpscot River and Mast Cove clam tissues exceeded the 2008 Gulfwatch mean (four sites, two in NH and two in ME). The higher concentration of the sum of 40 PAHs in the Presumpscot clam tissue, falling above the Gulfwatch mean, may indicate a component of alkylated PAHs that is contributing to the higher summation concentration since more alkylated PAHs are included in broader summations. No summation of total PAHs is available for Gulfwatch data, so no mean can be calculated to present in Figure 1.3.2.2.1. Presumpscot River exceeded the 2008 NS&T 85<sup>th</sup> percentile mean concentration for the sum of 24 non-alkylated PAHs (1,216 ng/g dry wt.), which places the Presumpscot in an “elevated” status for PAHs in clam tissue. However, the Presumpscot concentration is just into the NS&T mid-range of 1,251 – 4,434 ng/g dry wt. and in general PAH concentrations are quite low in the upper Northeast segment of the nation in the NS&T dataset.

PAH results from Presumpscot River, Falmouth/Portland, and Mast Cove, Eliot, included the PAHs necessary to calculate the ratio used previously in the blue mussel PAH section to explore non-alkylated to alkylated PAHs. The equation used to derive the ratio is:

$$\text{Fluoranthene} + \text{Pyrene} / \Sigma(\text{Fluoranthene} + \text{Pyrene} + \text{C2-C4 Alkylphenanthrene})$$

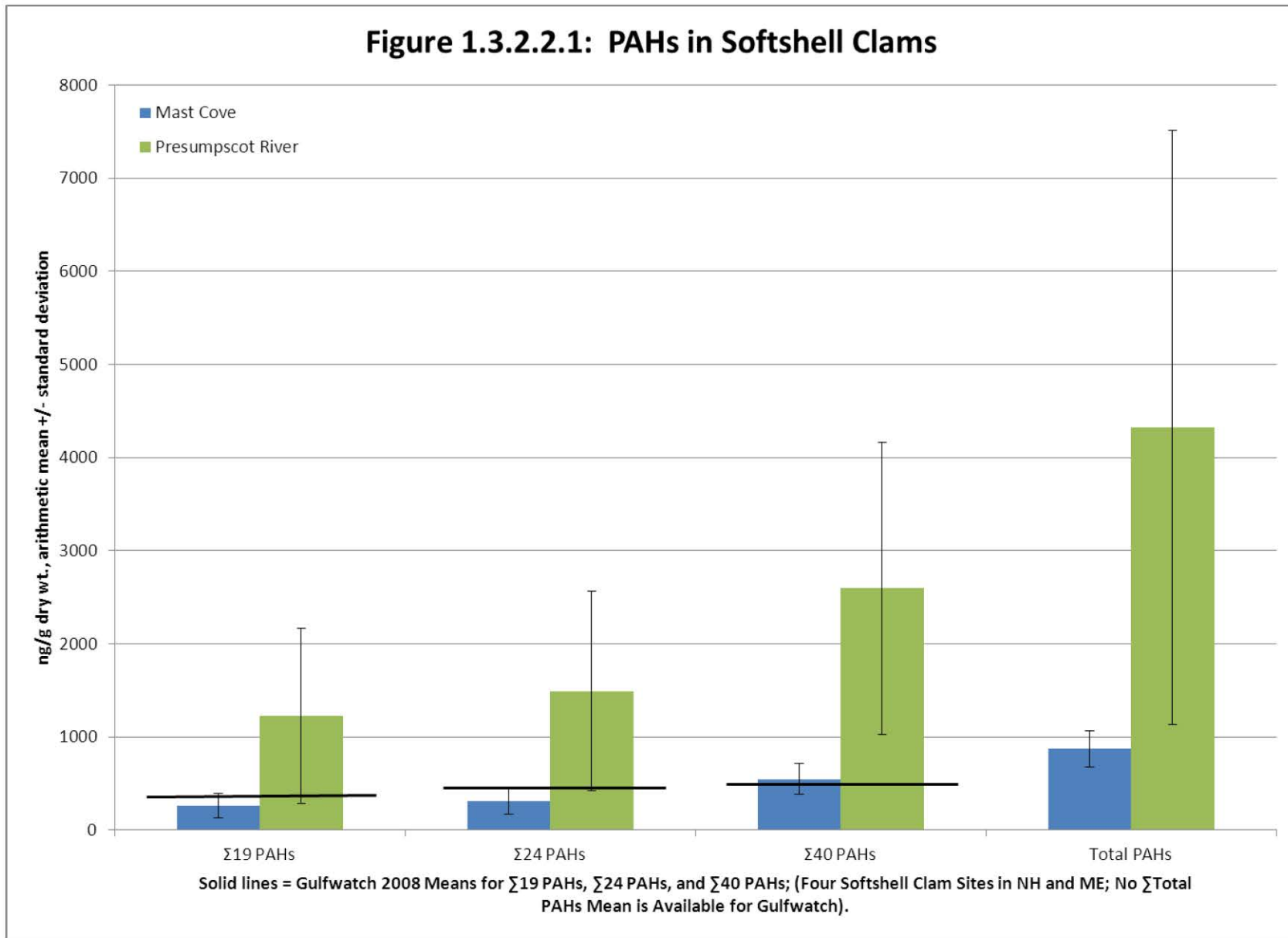
This equation is utilized to show relative concentrations of non-alkylated to alkylated PAHs, which yields a ratio indicating that values <0.1 are interpreted as a petrogenic (unburned fuel or petroleum) source, while values >0.2 are interpreted as a pyrogenic (combusted fuel) source of PAHs.

Since mussels (for PAH analysis) and clams were sampled only at two sites each in 2013, the calculated ratios for the two clam sites have been included in Figure 1.3.2.1.2. Both 2013 clam sites have pyrogenic ratios approximately 0.5 indicating the major PAH components present in clam tissue at these sites are from burnt fuel sources (Figure 1.3.2.1.2).





Toxicities of PAHs vary, with hundreds of compounds making up the pool of PAHs. Toxic responses in aquatic organisms may include reproductive inhibition, mutations, liver abnormalities, and even mortality. Exposure in the marine environment may be from spilled oil, boat exhaust, and runoff from urban areas. From a human health perspective, neither MCDC nor FDA have reported recommended safety levels for PAHs in fish or fish products (Kimbrough 2008).



### 1.3.3 Polychlorinated Biphenyls

PCBs (polychlorinated biphenyls) are synthetic organic compounds that consist of biphenyl with varying numbers of chlorine atoms. PCBs were manufactured from 1929 to 1977, though they were regulated in 1971 and new uses were banned in 1976. PCBs were used in electrical transformers and capacitors, and in lubricants and hydraulic fluids. They were also included in paints, adhesives, plasticizers, and flame retardants. Manufacturing of PCBs for flame retardants and lubricants was stopped in 1977. Current uses are in electrical equipment and transformers (Kimbrough et al. 2008).

#### 1.3.3.1 Blue Mussels

This report utilizes the Maine SWAT blue mussel tissue PCB data generated by AXYS Analytical, which includes 209 PCB congeners, some of which co-elute and are represented as combinations of PCB congeners. Co-elution refers to congeners that are collected together and then not separated during the detection/quantitation process on the gas chromatograph (GC) trace. The NS&T and Gulfwatch programs utilize a subset of PCBs, summing scores from 24 peaks on the gas chromatograph (GC) trace. The sum of these 24 GC peaks actually represents 31 PCB congeners since 7 of the 24 selected peaks contain two congeners each. These 31 summed PCB congeners will be called “Gulfwatch PCBs” or “NS&T PCBs” for the purposes of this report.

To compare Maine results to the NS&T and Gulfwatch PCBs, this report sums 35 congeners in the Maine SWAT PCB data, including 27 of 31 PCB congeners on the NS&T/Gulfwatch list, while including an additional 6 congeners that are not on the NS&T/Gulfwatch list. This difference is due to some congeners co-eluting differently or being summed differently at the various laboratories. These 35 summed congeners will be called “SWAT PCBs” for the purposes of this report.

Table 1.3.3.1.1 shows the list of PCB congeners used by NS&T and Gulfwatch compared to the list of PCB congeners reported by SWAT. Double numbers in the table represent co-elution or congeners that are quantified together within peaks on the GC output trace. Though the SWAT PCB and NS&T/Gulfwatch PCB congeners included in the summed lists are not identical, they are as close a comparison as possible. With some caution in data interpretation, this comparison may be used to place Maine SWAT blue mussel tissue PCB concentrations in a Gulf of Maine-wide and national perspective.

**TABLE 1.3.3.1.1: Comparison of 35 PCBs Summed for SWAT to 31 PCBs Summed for National Status & Trends and Gulfwatch.**

<u>SUM 35 PCBs</u> <u>“SWAT PCBs” List</u>	<u>SUM 31 PCBs</u> <u>“Gulfwatch, NS&amp;T PCBs”</u> <u>List</u>
PCB-5	PCB-8/5
PCB-8	PCB-18/15
PCB-15	PCB-29
PCB 18/30	PCB-50
PCB 26/29	PCB-28
PCB 20/28	PCB-52
PCB 50/53	PCB-44
PCB-52	PCB-66/95
PCB-66	PCB-101/90
PCB-77	PCB-87
PCB-90/101/113	PCB-77
PCB-118	PCB-118
PCB-126	PCB-153/132
PCB-132	PCB-105
PCB-153/168	PCB-138
PCB-169	PCB-126
PCB-187	PCB-187
PCB-170	PCB-128
PCB-190	PCB-180
PCB-128/166	PCB-169
PCB-195	PCB-170/190
PCB-208	PCB-195/208
PCB-180/193	PCB-206
PCB-206	PCB-209
PCB-209	
PCB-105	
<u>Unique to SWAT 35</u> <u>List</u>	<u>Unique to GW and</u> <u>NS&amp;T 31 List</u>
PCB-30	PCB-44
PCB-26	PCB-95
PCB-53	PCB-87
PCB-20	PCB-138
PCB-166	
PCB-193	

To compare what proportion of the total PCBs (209 congeners) the SWAT PCBs represent, Figure 1.3.3.1.1 shows the total PCBs next to the SWAT PCBs list used for comparison to Gulfwatch and NS&T data sets. Comparing the three mussel sites sampled for PCBs in 2013, the SWAT PCBs were 37.2%, 38.0%, and 38.4% of the total PCBs at Sears Island, Searsport, East End Beach, Portland, and Rockland, respectively. Total PCB concentrations were 59.4 ng/g dry wt. at East End Beach, 33.2 ng/g dry wt. at Rockland, and 20.2 ng/g dry wt. at Sears Island (Figure 1.3.3.1.1).

Figure 1.3.3.1.1 compares the SWAT PCBs at the 2013 SWAT mussel sites to Gulfwatch median and 85<sup>th</sup> percentile for 2008 PCB data, the most recent available. None of the three 2013 SWAT mussel sites exceeded the Gulfwatch 2008 median of 24.1 ng/g (dry weight), and consequently none of the sites tested in 2013 exceeded the Gulfwatch 85<sup>th</sup> percentile of 35.4 ng/g (dry weight) for Gulfwatch PCBs.

Figure 1.3.3.1.1 also compares the SWAT PCBs at the 2013 SWAT sites to NS&T (NS&T) median and 85<sup>th</sup> percentile 2008 PCB data, the most recent available. None of the three SWAT sites exceeded the NS&T 2008 median, 29.2 ng/g (dry weight), and so none of the three exceeded the NS&T national 85<sup>th</sup> percentile, 141 ng/g (dry weight).

In Figure 1.3.3.1.1, the standard deviation in blue mussel tissue total PCB concentration is high, which is due to the wider geographic spacing of the six spatial subsamples that constitute the mean. Toward gaining a better understanding of the spatial variability of the PCB tissue concentrations in and around Rockland harbor, six substations were sampled in 2013, all farther to the north and south from previously sampled substations in 2010 and 2011. Crockett Point was resampled in 2010 to confirm previous PCB concentrations, which were higher than many Maine coastal sites. Subsequently in 2011, four additional subsamples were collected: One to the north and three to the south of Crockett Point. With the wider spacing in 2011 (four substations) and 2013 (six substations), examination of the tissue concentration of each subsample yields more resolution to the spatial pattern of PCBs, which is lost when using the mean of all the distinct substations for each of these two years.

Figure 1.3.3.1.2 shows total PCB concentrations (non-detects = 0) at all sites sampled near Rockland from 2010 through 2013. Crockett Point, sampled in 2010, represents the mean of four replicates tested, while the remaining sites sampled in 2011 and 2013 represent individual results from composites of 20 mussels. Lower total PCB concentrations were evident at sites further from the center of the harbor and outside the harbor, as seen at North of Breakwater, Jameson Point, Owls Head, and Owls Head Harbor. In general, higher concentrations were noted at sites closer to the central harbor area and were highest at Crockett Point (2010) and Town Landing. Figure 1.3.3.1.3 shows the total PCB concentrations at the locations sampled in a map format. Again, higher total PCB concentrations are notable closer to the central portion of the harbor, while lower concentrations are predominantly toward more open water to the east.

The individual site SWAT PCBs ( $\Sigma 35$  PCBs) concentrations around Rockland were compared to the NS&T median and 85<sup>th</sup> percentile 2008 PCB data (Figure 1.3.3.1.4). One of six sites (Town Landing) sampled in 2013 exceeded the NS&T median, 29.2 ng/g (dry weight), along with the 2010 Crocket Point site, while none of four 2011 sites exceeded the NS&T median. None of the Rockland area SWAT sites approached or exceeded the NS&T 2008 national 85<sup>th</sup> percentile, 141 ng/g (dry wt.). The 2008 NS&T 85<sup>th</sup> percentile was 3.5 X higher than the highest scoring PCB site tested in the Rockland harbor area, Town Landing (2013). Gulfwatch median and 85<sup>th</sup> percentiles are also presented for comparison to SWAT sites in Figure 1.3.3.1.4. Some areas in southern New England have higher levels of PCBs than Maine waters but are still relatively cleaner than the lower Hudson River/Raritan Bay system, which is heavily contaminated from PCBs moving downriver from the upper Hudson (Kimbrough et al. 2008).

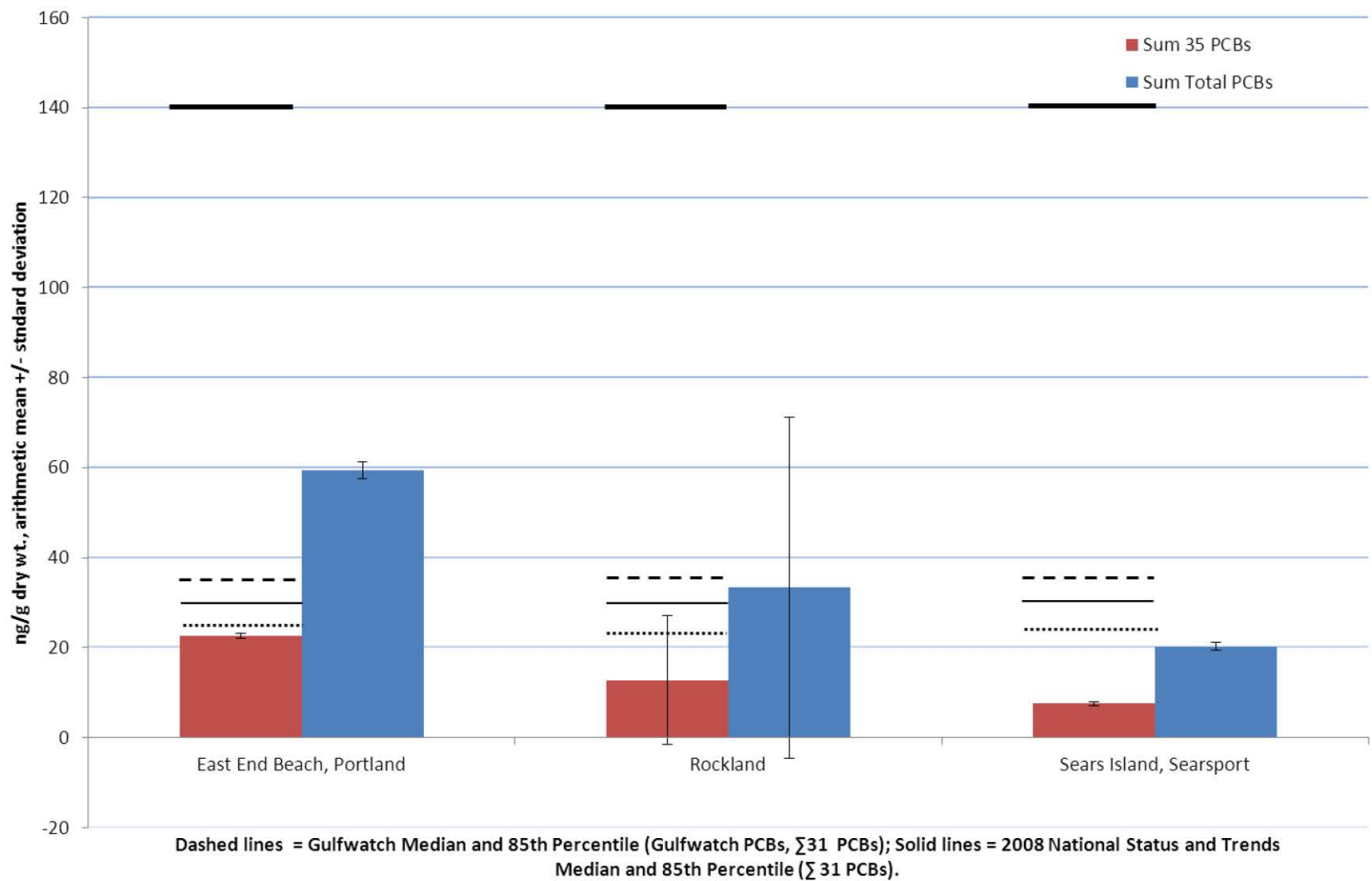
From a human health perspective, the MCDC cancer FTAL for total PCBs for non-commercially caught finfish is 11 ng/g wet wt. (ppb), while the MCDC non-cancer FTAL for total PCBs is 43 ng/g wet wt. (ppb). Of the six individual Rockland area total PCB concentrations from the 2013 sites, only Town Landing had a total PCB mean tissue concentration (15.6 ng/g wet wt.), which exceeded the 11 ng/g wet wt. MCDC cancer FTAL for total PCBs, the lower, more conservative of the two FTALs.

### 1.3.3.2 Softshell Clams

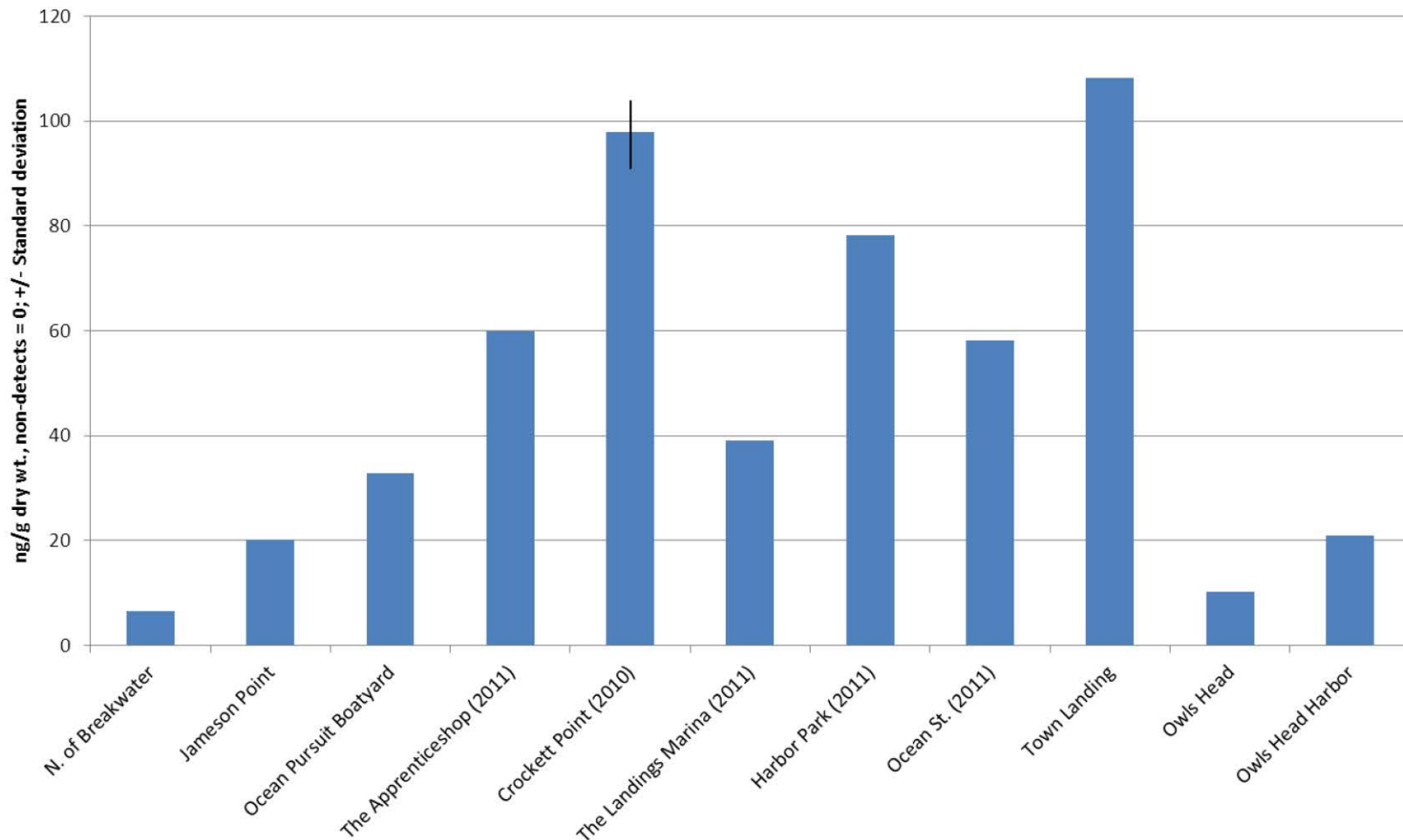
Softshell clams were tested for 209 PCBs from two sites in 2013, Presumpscot River, Falmouth/Portland, and Mast Cove, Eliot, and results compared to Gulf of Maine (Gulfwatch) (LeBlanc et al. 2009) softshell clam monitoring program data in an effort to place Maine SWAT data in a regional context. Summations of PCBs constructed for comparisons were previously discussed in Section 1.3.3.1. The same approach was utilized to construct clam PCB summations.

Table 1.3.3.1.1 shows the list of PCB congeners used by Gulfwatch compared to the list of PCB congeners reported by SWAT. Double numbers in the table represent co-elution or congeners that are quantified together within peaks on the GC output trace. Though the SWAT PCB and Gulfwatch PCB congeners included in the summed lists are not identical, they are as close a comparison as possible. With some caution in data interpretation, this comparison may be used to place Maine SWAT softshell clam tissue PCB concentrations in a Gulf of Maine-wide perspective.

**Figure 1.3.3.1.1.: SWAT PCBs ( $\Sigma 35$  PCBs) and Total PCBs in 2013 SWAT Blue Mussels**



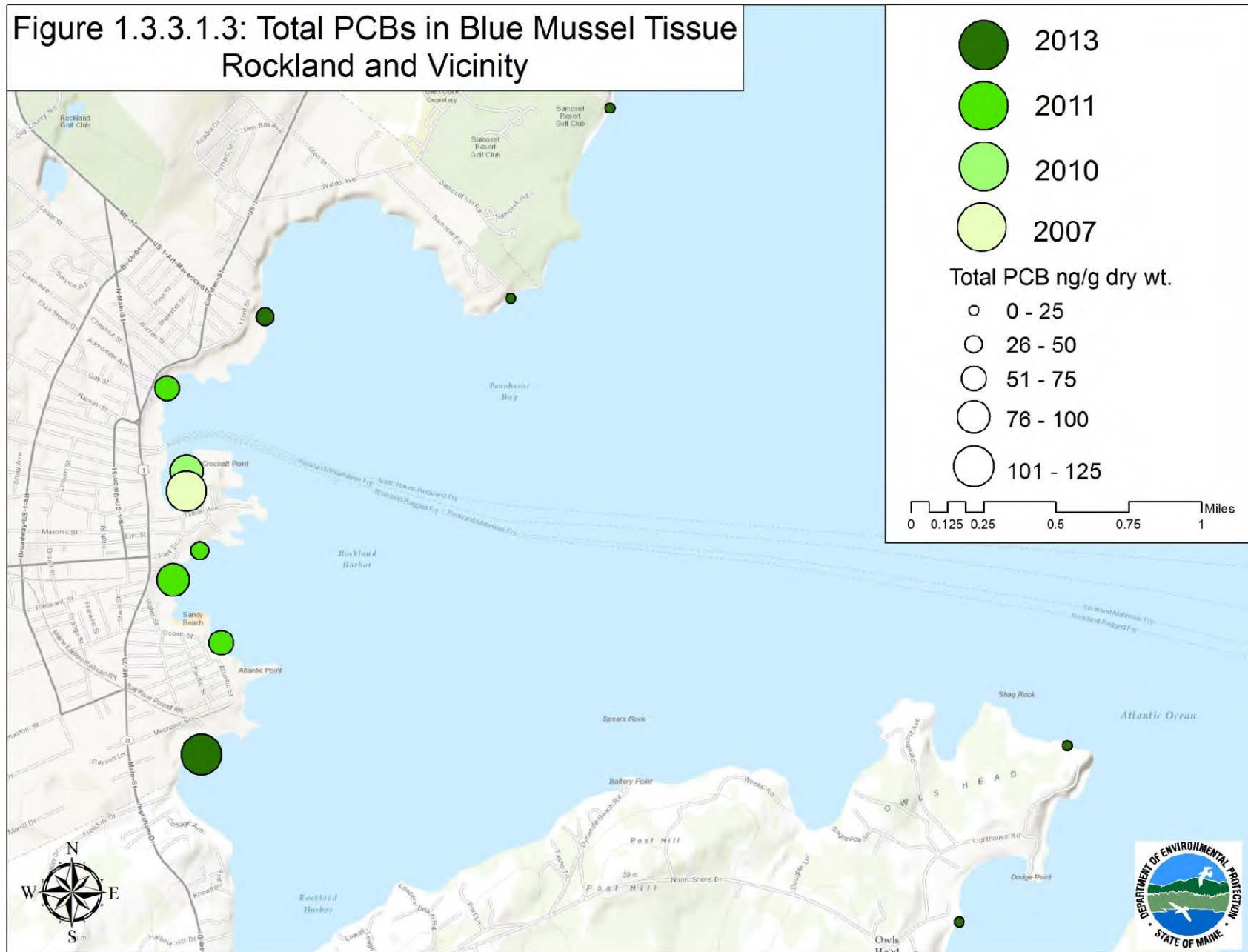
**Figure 1.3.3.1.2: Total PCBs in Blue Mussel Tissue at Locations Around Rockland Harbor - 2010 - 2013**



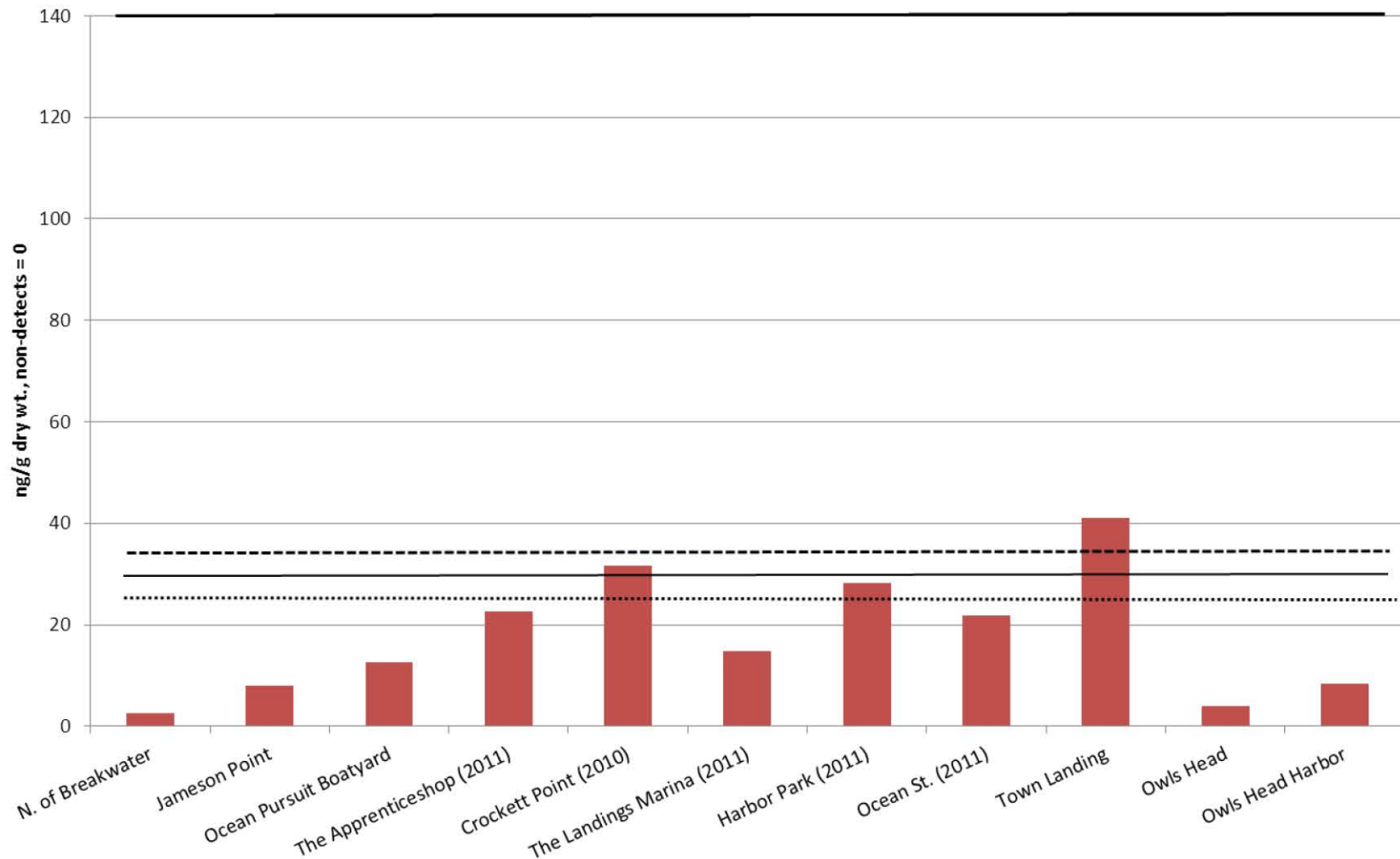
Sites : Left to right on X axis corresponds to north to south around Rockland harbor. Sites not marked with a year were sampled in 2013 (six sites; three most northerly and southerly).



Figure 1.3.3.1.3: Total PCBs in Blue Mussel Tissue  
Rockland and Vicinity



**Figure 1.3.3.1.4: SWAT PCBs ( $\Sigma 35$  PCBs) in Blue Mussel Tissue at Locations Around Rockland Harbor - 2010 - 2013**



Sites: Left to right on X-axis corresponds to north to south around Rockland harbor. Sites not marked with a year were sampled in 2013 (six sites; three most northerly and southerly). Dashed lines = Gulfwatch Median and 85th Percentile (Gulfwatch PCB)

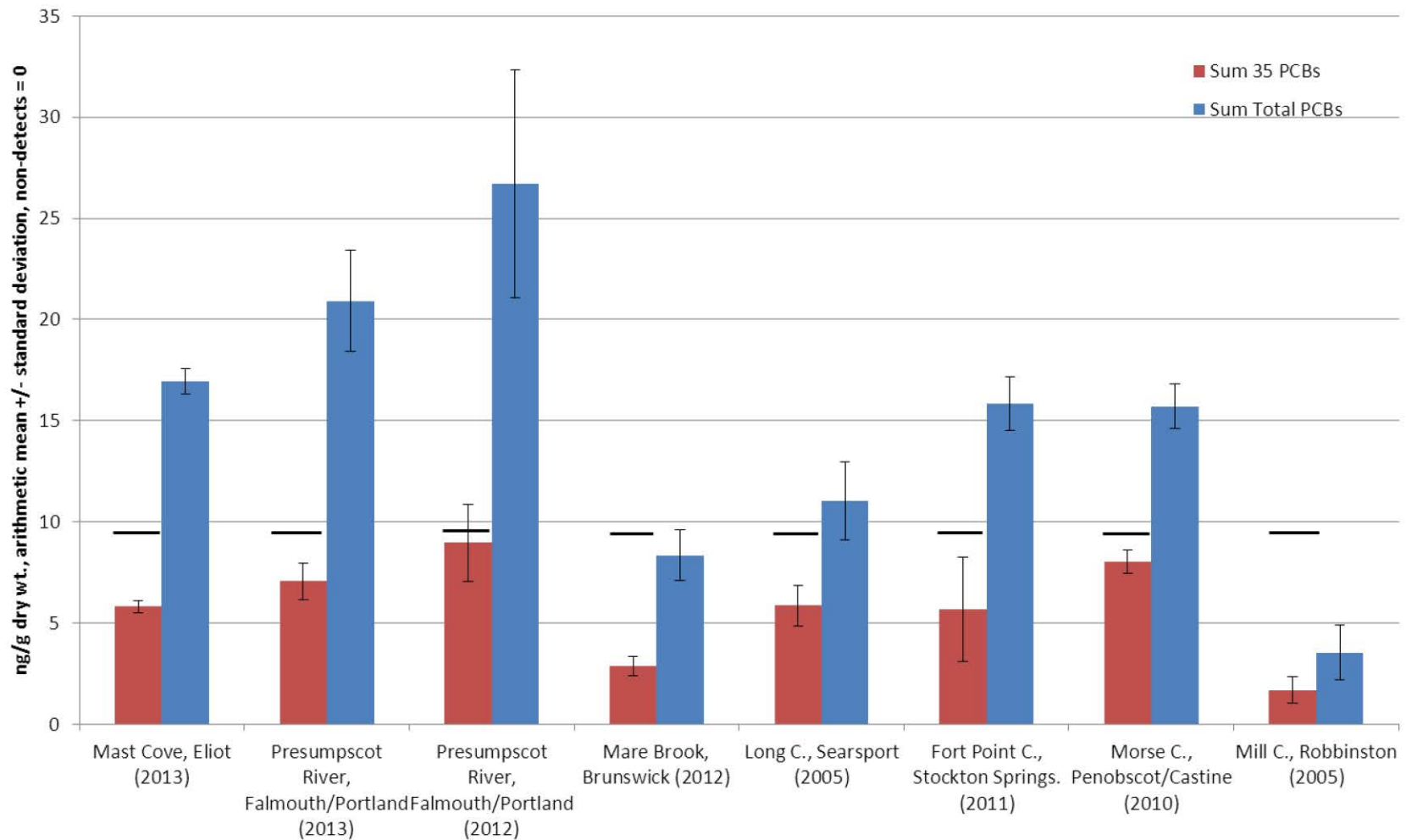
To compare what proportion of the total PCBs (209 congeners) the SWAT PCBs represent, Figure 1.3.3.2.1 shows the total PCBs next to the SWAT PCBs list used for comparison to Gulfwatch. Comparing the two 2013 clam sites, the SWAT PCBs were 33.7% and 34.2% of the total PCBs at Presumpscot River and Mast Cove, respectively. Total PCB concentrations were 17.0 ng/g dry wt. at Mast Cove, Eliot, and 20.9 ng/g dry wt. at Presumpscot River, Falmouth/Portland (Figure 1.3.3.2.1).

Mill Cove, Robbinston, and Long Cove, Stockton Springs, were sampled in 2005, and analyzed at a different lab than the 2010-12 clam tissues. The two early sites had much higher detection limits than those generated by the lab (AXYS Analytical) that analyzed the 2010-13 samples. In order to prevent the non-detects at Mill and Long Coves driving up the summations if non-detects were assigned a value of half the detection limit at the much higher detection limits used at the time of their analysis, all non-detects were assigned a value of zero for this figure and subsequent PCB analysis of the clam samples.

Figure 1.3.3.2.1 compares the SWAT PCBs at the seven recently sampled SWAT clam sites to a recent Gulfwatch clam site sampled in 2008 in New Hampshire. All seven SWAT clam site sums of 35 PCBs fell below the one Gulfwatch site concentration, though the 2012 Presumpscot summation was very close to the Gulfwatch summation (the 2013 Presumpscot summation was somewhat lower, but comparable). As noted above, comparison of 35 summed congeners from SWAT PCBs to 31 summed congeners from Gulfwatch PCBs is as close a comparison as possible due to differences in some PCBs co-eluting in different GC traces across laboratories. Gulfwatch non-detects were valued as half-detects, which will elevate the sum of 35 PCBs at North Mill Pond, NH, to some extent over the SWAT summations taken at non-detect valued at zero. Detection limits at the Gulfwatch site were lower than the older 2005 SWAT PCB analysis. Despite these differences, the summation of 35 SWAT congeners is useful for putting Maine data into a regional, Gulf of Maine context.

From a human health perspective, the MCDC cancer FTAL for total PCBs for non-commercially caught finfish is 11 ng/g wet wt. (ppb), while the MCDC non-cancer FTAL for total PCBs is 43 ng/g wet wt. (ppb). Of the seven SWAT clam sites sampled historically, the highest mean tissue concentration for total PCBs on a wet weight basis was 4.0 ng/g at Presumpscot River, Falmouth/Portland (2012), which was less than half of the MCDC cancer FTAL of 11 ng/g wet wt.

**Figure 1.3.3.2.1: Sum of 35 and Sum of Total PCBs in SWAT Softshell Clams**



Solid line = Gulfwatch 2008 softshell clam site at North Mill Pond, NH; three other 2008 Gulfwatch softshell clam sites had 0 summed PCB concentrations (sum 31 Gulfwatch PCBs).

### 1.3.4 Perfluorinated Compounds

Perfluorinated compounds or chemicals (PFCs) are organofluorine compounds that have fluorine substituted for all hydrogens where C-H bonds otherwise would occur in organic compounds. PFCs also have a functional group derived from the parent organic compound such that PFCs have properties of both fluorocarbons and the parent compound. The dual properties of PFCs make them useful in water, grease, and stain repellants (paper, fabric, and carpet treatments, notably Scotchgard by 3M), in the semiconductor industry, in firefighting foams, and as paint and other coating additives where flow is critical. Production of perfluorooctanesulfonyl fluoride related compounds, notably PFOSA (a sulfonamide), was terminated by 3M by 2003 but production overseas has continued or increased. While PFOSA was synthesized for use by industry, it is also created as a degradation byproduct of alkylated-perfluorooctanesulfonamides (which were used to treat paper, carpet, and fabric) through conversion into acetates and eventually to PFOSA.

Analysis for PFCs was suggested by the SWAT Technical Advisory Committee for inclusion in 2013 marine SWAT investigations. This report utilizes the Maine SWAT blue mussel tissue and softshell clam tissue PFC data generated by AXYS Analytical, which includes 12 compounds as presented in Table 1.3.4.1.1.

#### 1.3.4.1 Blue Mussels

Blue mussels were tested for PFCs from two sites in 2013, East End Beach, Portland, and Sears Island, Searsport. None of the twelve PFCs were detected in any of the four spatial replicates of mussel tissue collected at Sears Island. One PFC, perfluorooctane sulfonamide (PFOSA), was detected in all four spatial replicates of mussel tissue collected at East End Beach, Portland, while the 11 other PFCs were all below detection limits at East End Beach. Table 1.3.4.1.1 also shows the low and high values for the sample specific detection limits for the PFCs for which analyses were performed. In general, sample specific detection limits were approximately 0.5 to 6 parts per billion (ng/g) in mussel tissue. PFOSA levels detected in tissue from East End Beach ranged from 2.5 to 3.7 ng/g (ppb), with a mean of 3.2 ng/g dry wt.

#### 1.3.4.2 Softshell Clams

Softshell clams were tested for PFCs from two sites in 2013, Mast Cove, Eliot, and Presumpscot River, Portland/Falmouth. None of the twelve PFCs were detected in any of the four spatial replicates collected at each of the two sites. Table 1.3.4.1.1 shows the low and high values for the sample specific detection limits for the PFCs for which analyses were performed. In general, sample specific detection limits were approximately 0.50 to 7.50 parts per billion (ng/g) in clam tissue.

PFCs bioaccumulate and biomagnify through the food web. Testing of California *Mytilus spp.* tissue indicated >25% detection frequency for PFCs in samples and increasing concentrations with urbanization and proximity to stormwater discharge (Dodder, 2012). Total concentrations of PFCs ranged up to about 10 ppb, with some outliers above that range. Areas with mixed development topped out at total PFC concentrations of approximately 2 ng/g dry wt., while urban sites had higher total PFC concentrations approaching 9-10 ng/g dry wt. Two individual PFCs detected in the California study, PFDoDA and PFUnDA, had mean concentrations of 1.8 and 0.23 ng/g dry wt. respectively, which is roughly the same order of magnitude of the PFC

(PFOSA in the East End Beach, Maine, SWAT site) detected by SWAT in 2013 at one of four shellfish sites tested (Dodder, 2012). EPA has not released a fish tissue action level for PFCs.

**Table 1.3.4.1.1: LIST OF PERFLUORONATED COMPOUNDS AND THE RANGE OF SAMPLE SPECIFIC DETECTION LIMITS FOR 2013 SWAT BLUE MUSSELS AND SOFTSHELL CLAMS**

		<u>Non-Detects</u>			
		<u>Mussels</u>		<u>Clams</u>	
		<u>Low</u>	<u>High</u>	<u>Low</u>	<u>High</u>
PERFLUOROBUTANE SULFONATE	NG/G	0.8393	1.206	0.9881	1.498
PERFLUOROBUTANOATE	NG/G	4.196	6.031	4.941	7.488
PERFLUORODECANOATE	NG/G	0.4196	0.6031	0.4941	0.7488
PERFLUORODODECANOATE	NG/G	0.4196	0.6031	0.4941	0.7488
PERFLUOROHEPTANOATE	NG/G	0.4196	0.6031	0.4941	0.7488
PERFLUOROHEXANE SULFONATE	NG/G	0.8393	1.206	0.9881	1.498
PERFLUOROHEXANOATE	NG/G	0.4196	0.6031	0.4941	0.7488
PERFLUORONONANOATE	NG/G	1.049	1.507	1.236	1.872
PERFLUOROOCTANE SULFONATE	NG/G	2.098	3.015	2.47	3.744
PERFLUOROOCTANE SULFONAMIDE*	NG/G	0.5035	0.5865	0.5928	0.8986
PERFLUOROPENTANOATE	NG/G	0.4196	0.6031	0.4941	0.7488
PERFLUOROUNDECANOATE	NG/G	0.4196	0.6031	0.4941	0.7488

\* Non-detect values for mussels are from four spatial replicates at Sears Island, Searsport, as East End Beach, Portland/Falmouth had detects on all four spatial replicates.

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## 2.0 LAKES MODULE

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## 2.1. MERCURY TRENDS

### 2.1.1 Introduction

Mercury (Hg) is a global contaminant of major concern to both human and wildlife health due to its neurotoxicity. Increased loading of Hg from atmospheric deposition was documented in the continental U.S. in the early 1990s (Swain *et al.*, 1992), and current Hg loading is estimated to be three times pre-industrial deposition (Lindberg *et al.* 2007). In Maine, it is generally accepted that most Hg contained in atmospheric deposition can be traced to anthropogenic emissions from regional, local and global sources. Increased Hg loading translates to Hg contamination in fish in freshwater and coastal environments (Boesch *et al.*, 2001). US EPA recently found that approximately half (47%) of the freshwater fish sampled from 76,559 lakes across the US had mercury concentrations that exceeded the EPA's recommended safety level of 300 ppb (US EPA 2009). In the US, all 50 states and all the eastern Canadian provinces have established consumption advisories for marine and/or freshwater fish due to mercury (EPA, 2009; Environment Canada Website). These advisories affect 43% of the Nation's total lake acreage.

In Maine, fish tissue Hg concentrations documented in the early 1990s (Mower *et al.*, 1997), including notably high concentrations within Acadia National Park (Burgess, 1997), resulted in issuance of a statewide fish consumption advisory for all lakes and ponds in 1994 and all freshwaters in 1997 by the (now renamed) Maine Center for Disease Control and Prevention (ME-CDC). This advisory reduces recreational opportunity and a source of high quality protein for Maine residents. High Hg concentrations also pose a threat to wildlife including fish-eating birds and mammals; Acadia National Park and other sites in coastal Maine have had Hg values that exceed safety guidelines in bald eagles, tree swallows, and loons, among other species (Evers *et al.* 2005; Longcore *et al.* 2007). Although the source of the Hg in Maine is largely atmospheric deposition, Hg concentrations in biota are not uniform in lakes across Maine and are difficult to predict. Rather, Hg contamination of biota is highly variable and it is common for a lake containing fish with high Hg concentration to be located adjacent to another lake with relatively low fish Hg levels.

Increased controls on air emissions as a result of amendment of the US Clean Air Act since 1990 have resulted in decreases in atmospheric deposition of certain pollutants including mercury. Consistent with previous years' requests, additional data from fish from lakes were requested by ME-CDC in 2013 from as many of the lakes with historical data, such that current mercury levels can be readily compared to earlier mercury levels from similar locations in an attempt to determine any trending in the data sets. For trends, the following numbers of lakes have been resampled in the last few years for mercury in the same species of fish at least 10 years after initial sampling: 7 lakes in Aroostook County sampled from 1978-1984 and 2000-2001, 7 lakes from downwind of HoltraChem in Orrington sampled in 1996 and 2006, and 27, 9, and 13 lakes sampled statewide at least 10 years earlier and in 2010, 2011 and 2012 respectively. In 2013 a total of 23 lakes, with fish mercury data at least 10 years old, were sampled for white perch or smallmouth bass. Both species were collected from 2 of the lakes, McCurdy Pond in Bremen and Panther Pond in Raymond. A total of 13 of these lakes were sampled by the DEP Lakes Section in August for baseline trophic state conditions as part of continuing collaboration with researchers at the University of Maine to better understand ecological drivers that affect mercury concentrations in fish.

### 2.1.2 Methods.

A total of 10 fish from each lake were captured by gill nets and angling for individual analysis for mercury. Gill net sets were short as possible (usually checked every hour or so) to ensure capture of live fish and avoid oversampling of the lake. To avoid a potential bias due to desiccation, fish were captured and kept in a plastic bag on ice in a cooler until transfer to the lab where they processed immediately, stored in a refrigerator overnight for processing the next morning, or frozen for processing later. The fish were processed by 1) measuring and recording fish weights and lengths, 2) recording species, 3) using a scalpel, peeling a section of skin/scales from an area measuring approximately 1 cm × 4 cm from the side of the fish below or just in front of the dorsal fin, and then from the exposed muscle filet, obtaining three biopsy samples and placing them into labeled cryovials, and 4) removing a larger piece of filet and placing it into a larger labeled vial. Biopsy tissue samples generally weighed between 40 and 130 mg. These samples were frozen until analyzed at the Sawyer Environmental Chemistry and Research Laboratory at the University of Maine using a Direct Mercury Analyzer (DMA80) following EPA Method 7473.

### 2.1.3. Results and Discussion

In 2013, ten fish were collected from each lake, except for Clary Lake discussed in the Desiccation Study in the following section. Smallmouth bass were collected from 8 lakes, white perch from 13 lakes, and both species from 2 lakes (Table 2.1). Mean concentrations ranged from 0.19 ug/g to 1.09 ug/g. Concentrations exceeded ME-CDC's Fish Tissue Action Level (FTAL = 0.2 ug/g) for mercury in all lakes except Little (Lake St. George) Pond and only slightly exceeded the FTAL in McGrath Pond.

For trends analysis, given that mercury concentrations in fish vary with size and age, comparisons in mercury concentrations were made using ANCOVA with length as the covariate. The results show that there were 4 lakes where concentrations increased (denoted by a '+'), 4 lakes, including one (Panther Pond) with two species, where concentrations decreased (denoted by a '-'), and 12 lakes, including one (McCurdy Pond) with two species, where there was no change (denoted by a '0') from historical data. There were also two lakes (Branch Lake and Round Pond) where no comparison could be made with historical data since there were data from only one fish each from earlier dates. And there was one lake, Toddy Pond, where the comparison was with an ANOVA since there were no length data from the earlier date. Interestingly, while the mean mercury concentration in smallmouth bass from Panther Pond appeared much higher than the mean concentration in smallmouth bass collected in 2012, when adjusted for larger mean length in 2013, there was no significant difference between the years. These results were similar to those of previous three years combined with the data for all four years (2010-2013) combined showing increases in 11 lakes, decreases in 16 lakes, and no change in 42 lakes. Consequently, it appears that there is no statewide trend since the 1990s. In addition, there appears to be no discernable geographic pattern.

Comparison of concentrations among species showed differences among lakes. Concentrations in McCurdy Pond were higher in white perch than in smallmouth bass, but concentrations in Panther

Pond were higher in smallmouth bass. These differences were consistent in both recent and historical data for both lakes. Both lakes showed similar trends for both species, although concentrations in McCurdy Pond showed no trend, while concentrations in Panther Pond were lower in 2013 than historically. The data are sent to ME-CDC for use in evaluation of the statewide fish consumption advisory.

Table 2.1. Trends in mercury concentrations in fish from some Maine lakes from 2013 and 1990s (1993-2002).

LAKES	MIDAS	SPECIES	HG ug/g 2013	HG ug/g 1990s	HG % 2013-1990s	TREND +, -, 0	ANCOVA p-value	LENGTHS 2013-1990	COMMENT
BOTTLE L	4702	WHP	1.02	0.91	12	+	0.020	297-378	
BRANCH L	4328	WHP	0.90	0.26	246	?		335-254	1990s n=1
CHESUNCOOK L	0662	WHP	0.73	0.76	-4	-	0.006	193-219	NSD 13-96, SD 13-97
CUXABEXIS L	2892	WHP	1.09	1.51	-28	0	0.353	239-307	
ECHO L	5814	SMB	0.51	0.57	-11	-	0.000	389-329	
GRAHAM L	4350	SMB	0.89	0.73	22	0	0.524	358-290	
GREAT EAST L	3922	WHP	0.84	0.80	5	0	0.827	340-328	
GREEN L	4294	SMB	0.85	0.82	4	0	0.661	314-333	
KNIGHT P	3884	WHP	0.42	0.31	35	0	0.852	290-259	
LITTLE LAKE ST GEORGE P	7665	WHP	0.19	0.13	46	+	0.000	250-264	
MCCURDY P	5712	SMB	0.46	0.37	24	0	0.090	295-308	
MCCURDY P	5712	WHP	0.80	0.47	70	0	0.183	367-315	
MCGRATH P F	5348	WHP	0.26	0.26	0	0	0.077	257-213	
MEDUXNEKEAG L	1736	WHP	0.78	0.26	200	+	0.019	305-276	
NICKERSON L	1036	SMB	0.45	0.32	41	+	0.000	278-320	
PANTHER P	3694	SMB	0.52	0.72	-28	-	0.033	368-378	
PANTHER P	3694	WHP	0.43	0.62	-31	-	0.001	279-270	
PASSAGASSAWAUKEAG L	5496	WHP	0.60	0.55	9	0	0.570	352-321	
POCASSET L	3824	SMB	0.73	0.60	22	0	0.092	365-407	
ROUND P	5684	SMB	0.60	0.77	-22	?		319-360	1990s n=1
SEBAGO L	5786	WHP	0.79	0.65	22	0	0.247	291-293	
TAYLOR P	3750	WHP	0.50	0.62	-19	0	0.068	271-237	
TODDY P	4340	SMB	0.52	0.59	-12	0 <sup>a</sup>	0.356 <sup>a</sup>	315-???	no 1990 lengths
UPPER PLEASANT POND	5254	WHP	0.39	0.49	-20	-	0.044	245-236	
UPPER SHIN POND	2202	SMB	0.67	0.48	40	0	0.267	353-339	
ANCOVA with length as a covariate									
<sup>a</sup> ANOVA, no lengths available									

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## 2.2. DESICCATION STUDY

### 2.2.1 Introduction

In 2012, the mean mercury concentration in biopsy samples collected in the field from 30 white perch from Clary Lake were ~10% lower than the mean of biopsy samples collected from the same fish after being frozen. This was the opposite result from that found in a study in Massachusetts (Michael Hutcheson, Mass DEQ, personal communication), where contamination and or desiccation in the field was suspected as the cause of the higher levels from field biopsies. In the Clary Lake fish, desiccation during freezing may be the difference, although studies in 2010 (SWAT, 2010) indicated that desiccation from freezing was much less (1.8%). Nevertheless, to address the issue, in 2013, biopsies from 30 white perch from McGrath Pond were collected 1) in the field, 2) in the lab the same or next day on unfrozen fish, and 3) later in the lab from frozen fish. Biopsy samples were processed as described above in section 2.1.2.

### 2.2.2 Results

Mean mercury concentrations in biopsies taken from freshly captured fish from McGrath Pond in the lab were 2.8% higher than in biopsies collected at the pond the same day, but the difference was not statistically significant when compared by a one-tailed t-test (Table 2.2). Similarly, mean mercury concentrations in biopsies taken from the same fish after freezing for several days was 2.2% higher than in biopsies taken from freshly captured fish in the lab, but the difference was not also not statistically significant. Consequently, frozen fish had concentrations 5% higher than fresh caught fish sampled at the pond, although the difference was not statistically significant. This is slightly different than in 2012 where concentrations in frozen fish biopsies were ~10% higher than in biopsies collected in the field from Clary Lake. There is always some variability due to sampling and laboratory analysis also, but these results are systematic which suggests some other factor. Results from both years indicate that there is perhaps some loss of moisture with handling, time, and freezing such that the fixed amount of mercury in a sample divided by a partially dehydrated smaller mass of tissue results in a higher mercury concentration. That the magnitude of the difference is not consistent between lakes/years suggests additional comparisons should be made.

Table 2.2. Mercury concentrations in white perch from McGrath Pond sampled in 2013 at the Pond, Lab, and Frozen.

<b>ID</b>	<b>LENGTH</b> mm	<b>HG</b> ug/g <u>P</u> ond	<b>HG</b> ug/g <u>L</u> ab	<b>HG</b> ug/g <u>F</u> rozen	<b>HG</b> RPD L-P	<b>HG</b> RPD F-L	<b>HG</b> RPD F-P
WHP1	258	0.42	0.38	0.42	-8.4	8.0	-0.4
WHP2	252	0.20	0.21	0.19	5.4	-6.3	-0.9
WHP3	249	0.21	0.29	0.24	28.9	-16.7	12.3
WHP4	266	0.17	0.20	0.23	13.7	17.7	31.2
WHP5	264	0.26	0.27	0.32	2.1	18.6	20.7
WHP6	274	0.30	0.29	0.33	-5.1	13.9	8.8
WHP7	286	0.26	0.30	0.27	13.7	-11.8	1.9
WHP8	255	0.13	0.18	0.20	31.0	11.6	42.2
WHP9	277	0.41	0.44	0.48	5.0	9.1	14.1
WHP10	272	0.32	0.30	0.35	-6.9	15.8	8.9
WHP11	244	0.25	0.32	0.29	23.3	-8.1	15.2
WHP12	242	0.21	0.20	0.22	-6.9	13.2	6.3
WHP13	251	0.27	0.25	0.27	-7.0	5.2	-1.8
WHP14	245	0.31	0.28	0.35	-8.2	21.1	12.9
WHP15	251	0.16	0.19	0.17	14.1	-11.5	2.7
WHP16	251	0.19	0.22	0.23	14.2	7.1	21.2
WHP17	239	0.20	0.24	0.20	16.9	-16.0	0.9
WHP18	240	0.20	0.21	0.21	5.2	-2.7	2.4
WHP19	246	0.28	0.22	0.23	-26.7	7.3	-19.5
WHP20	264	0.31	0.30	0.25	-4.1	-15.0	-19.0
WHP21	240	0.19	0.24	0.20	21.2	-18.2	3.0
WHP22	241	0.25	0.24	0.25	-3.8	2.9	-0.9
WHP23	257	0.33	0.34	0.30	4.9	-12.8	-8.0
WHP24	243	0.21	0.20	0.23	-2.7	12.6	9.9
WHP25	258	0.29	0.31	0.31	8.0	0.5	8.5
WHP26	270	0.19	0.18	0.18	-4.0	0.9	-3.1
WHP27	270	0.42	0.33	0.37	-24.2	12.3	-12.0
WHP28	285	0.22	0.23	0.23	6.9	0.2	7.0
WHP29	250	0.22	0.23	0.24	3.4	3.0	6.3
WHP30	258	0.20	0.23	0.20	18.0	-18.0	0.0
<b>MEAN</b>		<b>0.25</b>	<b>0.26</b>	<b>0.27</b>	<b>2.8</b>	<b>2.2</b>	<b>5.0</b>
STD		0.08	0.06	0.07			
CV		0.30	0.24	0.28			
F-test p-value					0.316	0.376	0.907
t-test p-value					0.344	0.372	0.251
RPD= relative percent difference							

## 2.3 SELENIUM STUDY

### **Proposal to Assess the Potential for Mercury Toxicity Amelioration Through the Investigation of Selenium:Mercury Molar Ratios in Freshwater Fish**

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#### **Introduction**

Mercury has been shown to be pervasive through Maine's freshwater lakes and rivers, and recent studies have suggested that mercury is not trending downward or upward (SWAT 2010, 2011, 2012). The Maine Centers for Disease Control (CDC) has computed a fish tissue action level for mercury of 0.2 ppm wet weight. A majority of the fish tested from Maine's lakes and streams have mercury levels in excess of 0.2 ppm. As a result of the mercury levels in fish, the Maine CDC has issued a State-wide fish consumption advisory for fish caught from Maine rivers and lakes (<http://www.maine.gov/dhhs/eohp/fish/index.htm>).

Typically, the potential for mercury toxicity (in humans or biota) is assessed without information on co-occurring antagonists. However, the presence of selenium in the tissues of aquatic organisms, which as described below, has been shown to have potential mitigating, ameliorating, or antagonistic effects on mercury toxicity. Without addressing the relationship between mercury and selenium in tissues, potential ecological and human health risks to the presence of mercury may be overstated.

Selenium is essential for many normal metabolic processes in vertebrates. Amongst other functions, it acts as an antioxidant and is essential for normal thyroid hormone homeostasis and immunity. Dietary sources rich in selenium include marine fish. In addition to its essential biological functions, studies have shown that selenium can mitigate the toxicity of mercury in biota.

Since 1967, the peer-reviewed scientific literature has reported that selenium has an antagonistic (i.e., detoxifying) effect on mercury toxicity. Over the past four decades, studies have shown that selenium in the diets of fish, birds, and mammals, and in the water of aquatic receptors (algae, aquatic invertebrates, fish) also exposed to mercury can provide a protective effect on mercury induced toxicity of different target organs (Eisler, 1987; Cuvin-Aralar and Furness, 1991; Raymond and Ralston, 2004; Yang et al, 2008; Kahn & Wang, 2009; Peterson et al, 2009; Depew, 2012). The protectiveness provided by selenium has been suggested to ameliorate the potential toxic effects of mercury from consumption of fish and shellfish (Cabanero et al, 2007; Kahn & Wang, 2009; Peterson et al, 2009; Perugini et al, 2013).

A number of mechanisms have been proposed for this protective effect, although studies suggest that the primary interaction between mercury compounds, including methyl mercury, and biologically essential selenium results in an irreversible binding, or sequestration, of both mercury and selenium in the biological system (Berry and Ralston, 2008; Kahn & Wang, 2009; Ralston and Raymond,

2010). The molar ratio of mercury to selenium (Hg:Se) has been suggested as the most important factor to consider when assessing the potential antagonistic effects of selenium on mercury, although the interaction of Hg and Se species with sites on biological ligands is increasingly seen as being important (Cabanero et al, 2007). When Hg:Se molar ratios within the same tissue are less than 1.0, there is an excess of selenium. It has been suggested that a molar excess of selenium enables the irreversible binding of mercury, rendering it less toxic, while still maintaining adequate selenium levels to perform vital biological functions (Ralston and Raymond, 2004; Kahn & Wang, 2009; Peterson et al, 2009; Ralston and Raymond; 2010).

While research on the extent of mitigating effects from selenium:mercury interactions continue, we believe that getting a better understanding of the Hg:Se molar ratios that exist in Maine's freshwater fish will provide: 1) information that may be useful for assessing future human health risks from consumption of Maine's freshwater fish, 2) data that could be used to augment studies on potential ecological effects of mercury to piscivores, 3) information to assess the potential covariance of Hg:Se with respect to spatial and/or lake characteristics, and 4) baseline data against which future data can be compared.

### **Proposal**

To assess the potential for selenium:mercury binding in fish, we propose that tissue collected from freshwater fish already being caught and sampled for mercury (on behalf of CDC) also be analyzed for selenium. The combined mercury:selenium data set will be used to compute the molar ratios of Hg:Se within the fish.

DEP will be collecting freshwater fish from up to 26 lakes in 2013. It is our understanding that up to 10 fish per lake will be collected. Biopsy of fillet tissue will be collected by DEP for mercury analysis at the UM SECRL lab using the DMA 80. We propose that additional fillet tissue be sampled and submitted for total selenium analysis by the UM SECRL lab. For Se, the digestion method would be EPA 3052, since we are doing full dissolution; and the ICP method is EPA 200.7

To assess the potential for spatial and geologically dependent differences, we propose that 5 fish from 10 lakes (50 total) fillet samples be collected and analyzed for total selenium. Ideally, the lakes sampled would be spatially diverse and representative of different ecoregions within the state.

### **Results**

A total of ten white perch from each of five lakes or ponds where the fish were analyzed for mercury were also analyzed for selenium. The five ponds were scattered from York County to Washington County along the coast and inland (Table 2.3). Molar ratios of Hg:Se were all below 1, indicating a surplus of Se over Hg and potential mitigation of Hg toxicity. There was considerable variation in molar ratios that needs to be explored with respect to lake characteristics.



Table 2.3. Molar ratios of Hg:Se in white perch from five Maine lakes and ponds.

LAKE	TOWN	HG mole	SE mole	HG:SE mole/mole	HG:SE std
Branch Lake	Ellsworth	0.0045	0.0050	0.96	0.48
Knight Pond	S Berwick	0.0021	0.0032	0.68	0.20
Little (St George) Pond	Liberty	0.0010	0.0087	0.11	0.04
McCurdy Pond	Bremen	0.0040	0.0074	0.54	0.13
Taylor Pond	Auburn	0.0025	0.0071	0.36	0.10

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## 3.0 RIVERS AND STREAMS MODULE

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## 3.1 AMBIENT BIOLOGICAL MONITORING

### 3.1.1 Background

As part of the SWAT program, DEP's Biological Monitoring Unit evaluates benthic macroinvertebrate communities of Maine streams and rivers to determine if they are potentially impaired by toxic contamination. For reasons of comparability, a small number of unimpaired reference sites are also evaluated. Benthic macroinvertebrates are animals without backbones that can be seen with the naked eye and live on the stream bottom, such as mayflies, stoneflies, caddisflies, crayfish, snails, and leeches. In 2013, we evaluated the condition of 39 sample locations, primarily in the Androscoggin River basin.

The Biological Monitoring Unit uses a multivariate statistical model to analyze a benthic macroinvertebrate sample and predict if a waterbody is attaining the biological criteria associated with its statutory class (DEP Rule Chapter 579). If a waterbody does not meet minimum state aquatic life criteria, Class C, then the model class is predicted as Non-Attainment (NA). Classes AA and A are treated the same in the model. Final decisions on aquatic life attainment of a waterbody are made accounting for factors that may allow adjustments to the model outcome. This is called the final determination.

Table 3.1.1 summarizes the results of biological monitoring activities for the 2013 SWAT Program, sorted by waterbody name. Column headings of Table 3.1.1 are described below:

- *Station* – Since waterbodies are sometimes sampled in more than one location, each sampling location is assigned a unique “Station” number.
- *Log* – Each sample event is assigned a unique “Log” number.
- *Potential sources of pollution*
- *Statutory Class* – The state legislature has assigned a statutory class, either AA, A, B, or C, to every Maine stream and river. Class AA and A waterbodies shall support a “natural” biological community. Class B waterbodies shall not display “detrimental changes in the resident biological community”. Class C waterbodies shall “maintain the structure and function of the resident biological community”.
- *Final determination* – The final decision on aquatic life attainment of a waterbody; this decision accounts for factors that may allow adjustments to the model outcome. An ‘NA’ (Non-attainment) indicates that the sample did not meet the minimum Class C criteria. An ‘I’ (Indeterminate) indicates that a final decision could not be made based on the aquatic community collected.
- *Attains Class* – “Yes” is given if the final determination is equal to or exceeds the Statutory Class. A Class B stream, for example, would receive a “Yes” if its final determination was either A or B. “No” is given if a stream does not attain its Statutory Class. A Class B stream, for example, would receive a “No” if its final determination was either C or NA.
- *Probable Cause* – The probable cause column lists potential stressors to benthic macroinvertebrate communities, based on best professional judgment. In some cases, a probable cause may not be related to toxic pollution but instead to other factors.

Field and water chemistry data for each sampling event (where available) are given in Table 3.1.2 and 3.1.3, respectively. The data from tables 3.1.1 to 3.1.3 is also summarized in reports for each sampling event, known as Aquatic Life Classification Attainment Reports, which are available in electronic format with the web version of this report. Continuous water temperature data are given in Figure

3.1.1. The attainment history of sampling stations prior to 2013, where available, is given in Table 3.1.4.

For more information about the Biological Monitoring Unit, please e-mail us at [biome@maine.gov](mailto:biome@maine.gov) or visit our web site: <http://www.maine.gov/dep/water/monitoring/biomonitoring/>. The Data and Maps page of this website provides access to station information and available data via Google Earth.

### 3.1.2 Results Summary

The Biological Monitoring Unit concentrated its sampling in 2013 in the Androscoggin River basin. Thirty-nine stations were sampled under the SWAT Program (Table 3.1.1).

Thirty-nine stations have been analyzed for aquatic life attainment with thirty of these stations in attainment of their statutory class. Thirteen of these stations exceeded their assigned class. Three stations were called Indeterminate due to either a poor sample or an unusual taxa assemblage and one station did not meet statutory class due to a disturbed sample. No licensing / relicensing issues have been found in waterbodies sampled below municipalities or industries although a hatchery discharge may play a role in the enrichment of the aquatic community. The majority of streams that did not attain their statutory class were located on small urban and rural systems; summaries of these streams are found below.

#### Hart Brook – Lewiston Station 341

Hart Brook is a first order stream with a water quality goal of Class B. It flows southwest through a developed area and is crossed by I-95 before it enters the Androscoggin River. The benthic macroinvertebrate community is comprised mainly by tolerant midges and worms. The dominant taxa are the tolerant midges *Paratanytarsus* and *Dicrotendipes* which make up almost 60% of the community. Very few sensitive organisms are present in the community with only the two taxa *Cloen* (mayfly) and *Frenesia* (caddisfly) found. The stream did not meet the minimum Class C criteria for aquatic life. The specific conductance in the stream was very high at sample retrieval and the dissolved oxygen (DO) dropped to 4.7 mg/l (Table 3.1.2). In addition, Total Phosphorus and Total Dissolved Solids (TDS) were very high which are indicative of an urban system (Table 3.1.3). The stream banks were highly eroded indicating storm water issues. The stream has been listed on the 303(d) list and is part of a percent impervious cover TMDL. Hart Brook has not attained class since first being sampled in 1998 (Table 3.1.4).

#### Unnamed Stream – Lewiston Station 857

Unnamed Stream in Lewiston is a cold first order stream with a water quality goal of Class B. It flows from north to south through a highly developed area and passes adjacent to Lewiston's Municipal Landfill before entering the Androscoggin River. The stream did not attain the minimum Class C criteria for aquatic life. Although the invertebrate community was diverse with 61 different genera present there were very few sensitive organisms present. The dominant taxa in the community consisted of the tolerant midges *Polypedilum*, *Tanytarsus*, and *Paratendipes*, the worm *Limnodrilus*, and the tolerant non-insect *Caecidotea* (Isopods). The Nitrate + Nitrite as N level was high at .94 mg/l and the Total Dissolved Solids were elevated at 320 mg/l (Table 3.1.3). The stream channel is in poor condition. The banks are blown out and the stream channel is widened considerably. The wetted

width of the stream was only one third the bank full width at retrieval of the samplers. The stream did not attain class in 2008 (Table 3.1.4).

#### Halfmoon Stream – Thorndike Station 697

Halfmoon Stream is a third order stream which flows west to the town of Unity entering Sandy Stream and eventually Unity Pond. Above the Rt. 220 Bridge crossing in Thorndike, its water quality goal is Class A. The stream flows through a concentrated agricultural area consisting of large dairy farms. The station was sampled in 2003 and 2007 and met the Class A aquatic life criteria. In 2012, the macroinvertebrate community showed signs of enrichment with the Total Mean Abundance of the sample totaling over 1300 individuals. Although the Generic Richness was high, the total number of sensitive organisms was low compared to the total mean abundance and the most dominant taxa in the sample consisted of tolerant collector-filterers and scrapers (see SWAT 2012). The stream did not meet the Class A aquatic life criteria (Table 3.1.4). The stream was sampled again during the 2013 field season. The Total Mean Abundance of the sample totaled over 1700 individuals. The Generic Richness dropped to 29 taxa as compared to 70 taxa in 2012. Possible explanations for this significant drop in Generic Richness were the combination of a 3 foot change in water level due to a storm event in 2013 and the lack of algae attached to the rock bags during sample pick up. In 2012, dense mats of algae were present on the samplers. The Chironomidae and some mayfly taxa use the algae as a food source and as attachment sites. In addition, the Nitrate + Nitrite as N level was high at .62 mg/l (Table 3.1.3) indicating possible runoff from the agricultural fields. The stream did not meet the Class A aquatic life criteria. This high quality resource has shown a trend of increasing enrichment since 2003 and a follow up of sampling and, stream surveys to identify inputs to the system will need to be performed. In addition, Agricultural Best Management Practices will need to be instituted to maintain a Class A aquatic life community in the stream.

#### Unnamed Stream (Brunswick 2) - Brunswick Station 641

Unnamed Stream (Brunswick 2) is a cold first order stream with a water quality goal of Class B. The sample location is 17 meters downstream of River Road in a heavily developed area of Brunswick. The stream did not attain the minimum Class C criteria for aquatic life. The dominant taxa in the community consisted of the tolerant non-insect *Caecidotea* (Isopods), the tolerant midge *Tanytarsus*, and the tolerant worm *Limnodrilus*. Only one genus of mayfly (*Baetis*) was collected but in very low numbers. The stream banks were eroded and the stream channel was widened. Iron bacteria were observed at the deployment date. The specific conductance was high during the time of sampling (Table 3.1.2). Unnamed Stream (Brunswick 2) has not attained class since first being sampled in 2002 (Table 3.1.4) and is included in a Statewide % Impervious Cover TMDL.

**Table 3.1.1. 2013 SWAT Benthic Macroinvertebrate Biomonitoring Results**

Waterbody	Town	Station	Log	Potential sources of pollution <sup>1</sup>	Statutory Class/ Final Determination	Attains Class? <sup>2</sup>	Probable Cause
Androscoggin River	Bethel	355	2218	Municipal	B/A	Yes	
Aunt Hannah Brook	Dixfield	343	2212	Reference	B/A	Yes	
Bear River	Newry	866	2214	Reference	AA/A	Yes	
Bobbin Mill Brook	Auburn	357	2186	Urban NPS	B/A	Yes	
Bowley Brook	Weld	1003	2213	Reference	B/A	Yes	
Cupsuptic River	Upper Cupsuptic TWP	999	2201	Reference	AA/A	Yes	
East Branch Wesserunnett River	Athens	486	2198	Long Term Monitoring	B/A	Yes	
East Brook	Weld	1002	2211	Reference	B/A	Yes	
East Cathance Stream	Bowdoin	859	2205	Reference	B/B	Yes	
Frye Brook	Andover West Surplus	1000	2224	Reference	A/A	Yes	
Halfmoon Stream	Thorndike	697	2197	Agricultural NPS	A/B	No	Agricultural Runoff
Hart Brook	Lewiston	341	2187	Urban NPS	B/NA	No	NPS Toxics; Habitat
Kennebago River	Rangeley	868	2199	Reference	AA/A	Yes	
Little Androscoggin River	Oxford	1001	2206	Municipal	C/C	Yes	
Little Androscoggin River	Paris	79	2208	Municipal; Urban NPS	C/A	Yes	
Little Androscoggin River	Paris	43	2209	Urban NPS	C/A	Yes	
Martin Stream	Turner	693	2210	Agricultural NPS	B/A	Yes	
Merrill Brook	Newry	350	2215	NPS	A/A	Yes	
Nezinscot River	Turner	860	2207	NPS	B/A	Yes	
Rangeley River	Oquossoc	137	2200	Lake Outlet	A/B	No	Lake Outlet; Hatchery

<sup>1</sup> NPS, non-point source pollution.<sup>2</sup> This field is completed only for stations for which sampling results have been obtained as of the time of this report

**Table 3.1.1. 2013 SWAT Benthic Macroinvertebrate Biomonitoring Results (continued)**

Waterbody	Town	Station	Log	Potential sources of pollution <sup>1</sup>	Statutory Class/ Final Determination	Attains Class? <sup>2</sup>	Probable Cause
Sabattus River	Lisbon	170	2202	Urban NPS	B/I	Indeterminate- Generic Richness Low	Poor Sample
Sabattus River	Sabattus	359	2203	Agricultural NPS, Lake Outlet	C/B	Yes	
Sabattus River	Sabattus	629	2204	Lake Outlet, Municipal	C/C	Yes	
Sheepscot River	North Whitefield	74	2183	Long Term Monitoring	AA/A	Yes	
Stetson Brook	Lewiston	356	2189	Urban NPS	B/B	Yes	
Sunday River	Newry	444	2216	Reference	A/A	Yes	
Sunday River	Bethel	354	2219	NPS	A/A	Yes	
Swift River	Rumford	345	2220	NPS	B/A	Yes	
Swift River	Roxbury	346	2221	Reference	A/B	No	Disturbed Sample
Thompson Lake Outlet	Oxford	76	2193	Lake Outlet, Former Industrial Site	C/I	Indeterminate	Poor Sample
Unnamed Stream	Lewiston	857	2188	Municipal Landfill	B/NA	No	NPS toxics; In-Place Contamination; Habitat
Unnamed Stream #2	Brunswick	641	2190	Urban NPS	B/NA	No	NPS Toxics; Habitat
Unnamed Stream #2	Topsham	633	2191	Urban NPS	B/B	Yes	
Unnamed Stream #4	Topsham	634	2192	Urban NPS	B/I	Indeterminate	Unusual Taxa Assemblage
West Branch Nezinscot River	Sumner	664	2196	NPS	A/A	Yes	
West Branch Sheepscot River	China	268	2182	Long Term Monitoring	AA/A	Yes	
West Branch Ellis River	Andover	872	2222	Reference	A/A	Yes	
West Branch Ellis River	Andover	1004	2223	Reference	A/A	Yes	
Wild River	Gilead	103	2217	Reference	A/A	Yes	

<sup>1</sup> NPS, non-point source pollution.<sup>2</sup> This field is completed only for stations for which sampling results have been obtained as of the time of this report



**Table 3.1.2. 2013 SWAT Field Data**

Measurements were obtained using handheld electronic meters. Highlighted values are of concern or do not attain criteria.

Site	Station	Log	Sampler Deployment					Sampler Retrieval				
			Date	Temp	DO	SPC	pH	Date	Temp	DO	SPC	pH
				Deg C	mg/L	uS/cm	STU		Dec C	mg/L	uS/cm	STU
Androscoggin River	355	2218	7/24/13	21.5	7.9	31	6.14	8/21/13	22.3	8	37	6.32
Aunt Hannah Brook	343	2212	7/22/13	17.6	8.3	38	5.8	8/20/13	16.7	8.2	31	6.49
Bear River	866	2214	7/23/13	16.9	8.3	19	7.89	8/21/13	19.2	11.1	24	6.25
Bobbin Mill Brook	357	2186	7/9/13	24.1	8.3	69	6.67	8/6/13	23.2	8.2	74	6.77
Bowley Brook	1003	2213	7/22/13	18.6	8.4	28	6.4	8/20/13	18.5	9	28	6.35
Cupsuptic River	999	2201	7/17/13	20.9	7.8	18	6.76	8/14/13	16.8	8.5	17	6.41
East Branch Wesserunsett Stream	486	2198	7/19/13	23	8.1	63	7.19	8/13/13	19	8.1	36	7.24
East Brook	1002	2211	7/22/13	18.2	9	36	6.18	8/20/13	19.1	8.3	32	6.2
East Cathance Stream	859	2205	7/18/13	23.9	6	62	6.4	8/15/13	17.7	6.9	42	6.23
Frye Brook	1000	2224	7/25/13	15.6	10.4	13	6.3	8/26/13	16.3	8.8	13	5.86
Halfmoon Stream	697	2197	7/19/13	24.8	10.2	116	8.59	8/13/13	19.8	9.3	90	7.65
Hart Brook	341	2187	7/9/13	19.4	7.6	267	7	8/6/13	20.7	4.7	567	6.66
Kennebago River	868	2199	7/17/13	23.2	8.7	28	6.5	8/14/13	18.5	9.4	20	6.83
Little Androscoggin River	1001	2206	7/16/13	24.8	8	55	6.32	8/19/13	22.2	7.5	47	5.83
Little Androscoggin River	43	2209	7/16/13	22.1	7.9	53	6.3	8/19/13	18.1	7.6	57	6.38
Little Androscoggin River	79	2208	7/16/13	22.1	7.8	58	6.4	8/19/13	18.2	8.4	56	6.25
Martin Stream	693	2210	7/15/13	22.8	7.1	68	6.28	8/19/13	20.2	7.8	58	6.23
Merrill Brook	350	2215	7/23/13	16.4	8.8	60	6.44	8/21/13	16.3	7.7	44	6.3
Nezinscot River	860	2207	7/15/13	22.1	7.8	45	6.17	8/19/13	20.8	8.2	45	6.3
Rangeley River	137	2200	7/17/13	22.2	8.5	32	6.56	8/14/13	19.4	9.0	25	6.29

Temp = water temperature, DO = dissolved oxygen, SPC = specific conductance, pH= hydrogen ion concentration.

Table 3.1.2. 2013 SWAT Field Data (continued)

Site	Station	Log	Sampler Deployment					Sampler Retrieval				
			Date	Temp	DO	SPC	pH	Date	Temp	DO	SPC	pH
				Dec C	mg/L	uS/cm	STU		Dec C	mg/L	uS/cm	STU
Sabattus River	170	2202	7/18/13	27	7.3	145	6.9	8/15/13	21.4	7.6	85	7.21
Sabattus River	359	2203	7/18/13	27.4	6.8	128	6.8	8/15/13	21.9	8.5	73	7.22
Sabattus River	629	2204	7/18/13	27.1	7	88	6.8	8/15/13	23	8.9	69	8
Sheepscot River	74	2183	7/8/13	25	7.6	45	6.2	8/5/13	20.5	8.2	52	6.6
Stetson Brook	356	2189	7/9/13	21	7.7	77	6.58	8/6/13	19.4	8.8	79	7.23
Sunday River	354	2219	7/24/13	18.2	8.1	21	5.65	8/21/13	20.7	8.6	35	5.83
Sunday River	444	2216	7/23/13	17.4	8.5	16	7.48	8/21/13	19.5	9.1	15	5.75
Swift River	345	2220	7/25/13	17.3	9.3	28	6.05	8/26/13	19	8.5	38	5.98
Swift River	346	2221	7/25/13	15.9	8.5	16	5.62	8/26/13	17.8	8.8	19	5.45
Thompson Lake Outlet	76	2193	7/11/13	23.2	7.8	38	6.38	8/8/13	23.2	8.3	38	6.44
Unnamed Stream (Brunswick 2)	641	2190	7/10/13	15.8	8.1	393	6.65	8/7/13	16.3	8.8	345	6.18
Unnamed Stream (Lewiston)	857	2188	7/9/13	18.9	8.3	232	7.36	8/6/13	14.3	7.1	387	7.5
Unnamed Stream (Topsham 2)	633	2191	7/10/13	15.2	8.1	334	6.19	8/7/13	14	8.3	289	6.67
Unnamed Stream (Topsham 4)	634	2192	7/10/13	15.6	7.8	786	6.29	8/7/13	14.7	7.1	705	6.4
West Branch Ellis River	872	2222	7/25/13	17.3	9.3	26	6.56	8/26/13	16.1	8.6	55	6.35
West Branch Ellis River	1004	2223	7/25/13	18	8.6	19	6.52	8/26/13	16.2	9.1	34	6.15
West Branch Nezinscot River	664	2196	7/15/13	20.7	8	20	5.98	8/12/13	17.3	7.6	11	6.24
West Branch Sheepscot River	268	2182	7/8/13	22.3	8.4	63	6.94	8/5/13	20.6	8.5	58	6.82
Wild River	103	2217	7/24/13	18.5	8.5	9	5.42	8/21/13	18.4	8.8	15	6.26

Temp = water temperature, DO = dissolved oxygen, SPC = specific conductance, pH = hydrogen ion concentration.

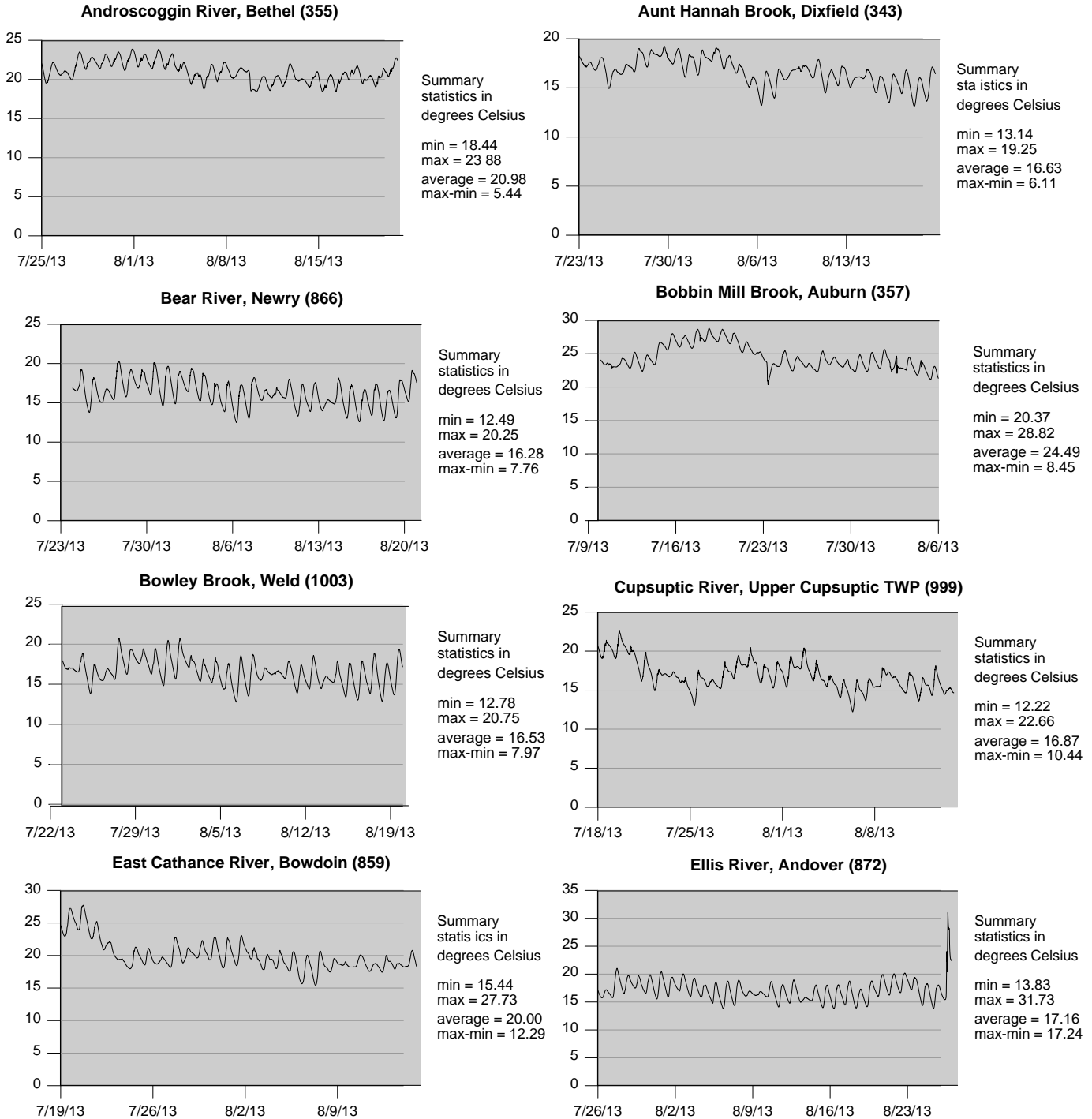
**Table 3.1.3. 2013 SWAT Water Chemistry Data**

Samples were analyzed by the Health & Environmental Testing Laboratory, Augusta, ME. Highlighted values indicate high results.

Waterbody	Station	Log	Sampling Date	DOC	NH <sub>3</sub> -N	TKN	NO <sub>2</sub> -NO <sub>3</sub> -N	SRP	Total P	TSS	TDS
				mg/L	mg/L	mg/L	mg/L	ug/L	mg/L	mg/L	mg/L
Androscoggin River	355	2218	8/21/13	3	<0.01	0.2	0.06	2	0.01	<2	41
Aunt Hannah Brook	343	2212	8/20/13	4.3	<0.01	0.2	0.05	5	0.017	<2	44
Bear River	866	2214	8/21/13	1.6	<0.01	0.1	0.05	3	0.005	<2	41
Bowley Brook	1003	2213	8/20/13	3.6	<0.01	0.2	0.01	3	0.01	<2	43
East Branch Wesserunsett Stream	486	2198	8/13/13	3.2	<0.01	0.2	0.01	1	0.008	<2	47
Halfmoon Stream	697	2197	8/13/13	2.6	0.01	0.2	0.62	7	0.019	<2	87
Hart Brook	341	2187	8/6/13	3.9	0.38	0.8	0.09	4	0.055	8.4	400
Little Androscoggin River	1001	2206	8/19/13	2.6	0.02	0.1	0.06	2	0.011	<2	43
Little Androscoggin River	79	2208	8/19/13	2.3	0.01	0.1	0.12	3	0.013	<2	56
Martin Stream	693	2210	8/19/13	2.9	0.01	0.2	0.25	2	0.019	3.8	53
Sabattus River	170	2202	8/15/13	3.4	<0.01	0.8	0.02	2	0.074	20	80
Sabattus River	629	2204	8/15/13	2.8	<0.01	0.9	<0.01	1	0.071	17	67
Sheepscot River	74	2183	8/5/13	4.8	0.01	0.3	0.02	2	0.013	<2	61
Stetson Brook	356	2189	8/6/13	6.4	0.01	0.5	0.04	7	0.031	3.4	96
Unnamed Stream (Lewiston)	857	2188	8/6/13	3.3	0.39	0.7	0.94	5	0.015	3	320
West Branch Nezinscot River	664	2196	8/12/13	4.8	<0.01	0.2	0.01	2	0.015	<2	39
West Branch Sheepscot River	268	2182	8/5/13	4.3	<0.01	0.3	0.02	1	0.012	<2	64

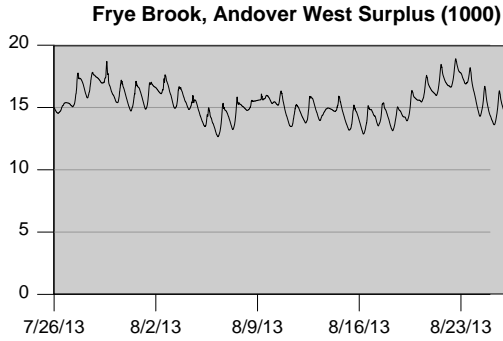
DOC = dissolved organic carbon, NH<sub>3</sub>-N = ammonia-nitrogen, TKN = total Kjeldahl-nitrogen, NO<sub>2</sub>-NO<sub>3</sub>-N = nitrite-nitrate-nitrogen, SRP = soluble reactive phosphorus (ortho-phosphate), Total P = total phosphorus, TSS = total suspended solids, TDS = total dissolved solids, "<" = constituent not detected at the reporting limit.

**Figure 3.1.1. 2013 In-Stream Continuous Temperature Data**  
**Please note: all data are in degrees Celsius**

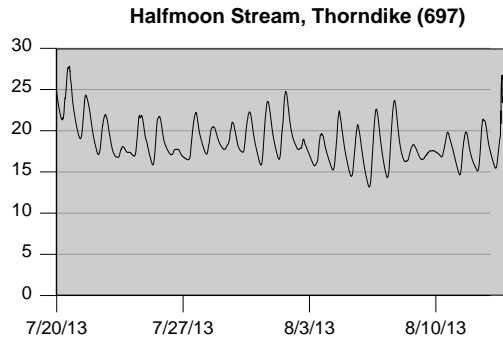


**Figure 3.1.1. 2013 In-Stream Continuous Temperature Data (continued)**

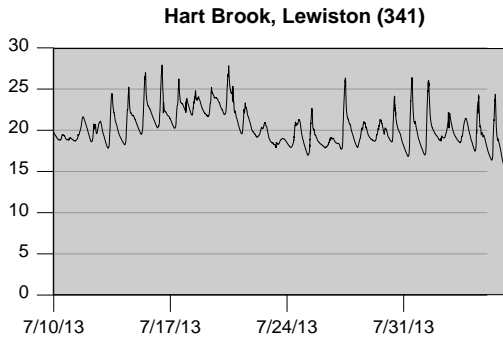
Please note: all data are in degrees Celsius



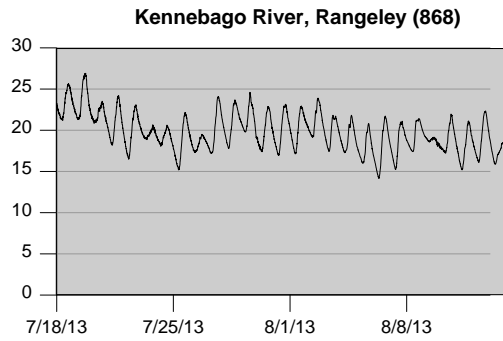
Summary statistics in degrees Celsius  
 min = 12.65  
 max = 18.91  
 average = 15.40  
 max-min = 6.26



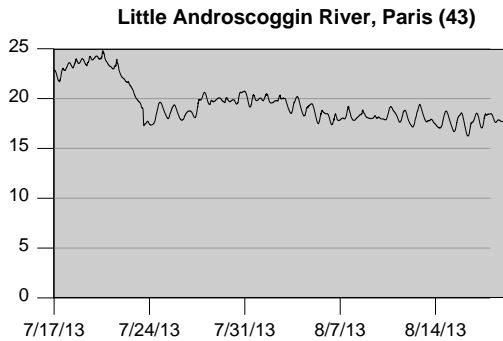
Summary statistics in degrees Celsius  
 min = 13.16  
 max = 27.85  
 average = 18.74  
 max-min = 14.69



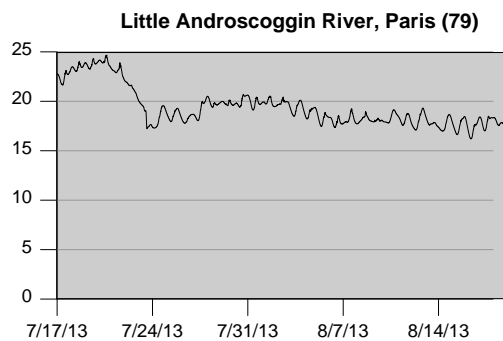
Summary statistics in degrees Celsius  
 min = 15.16  
 max = 27.92  
 average = 20.35  
 max-min = 12.01



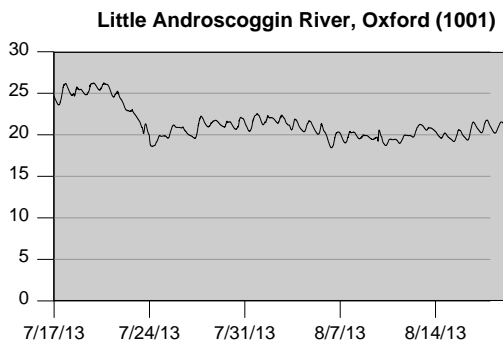
Summary statistics in degrees Celsius  
 min = 14.17  
 max = 26.92  
 average = 19.80  
 max-min = 12.75



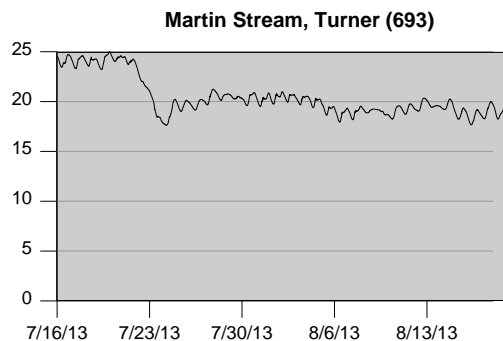
Summary statistics in degrees Celsius  
 min = 16.25  
 max = 24.80  
 average = 19.45  
 max-min = 8.55



Summary statistics in degrees Celsius  
 min = 16.20  
 max = 24.68  
 average = 19.39  
 max-min = 8.48



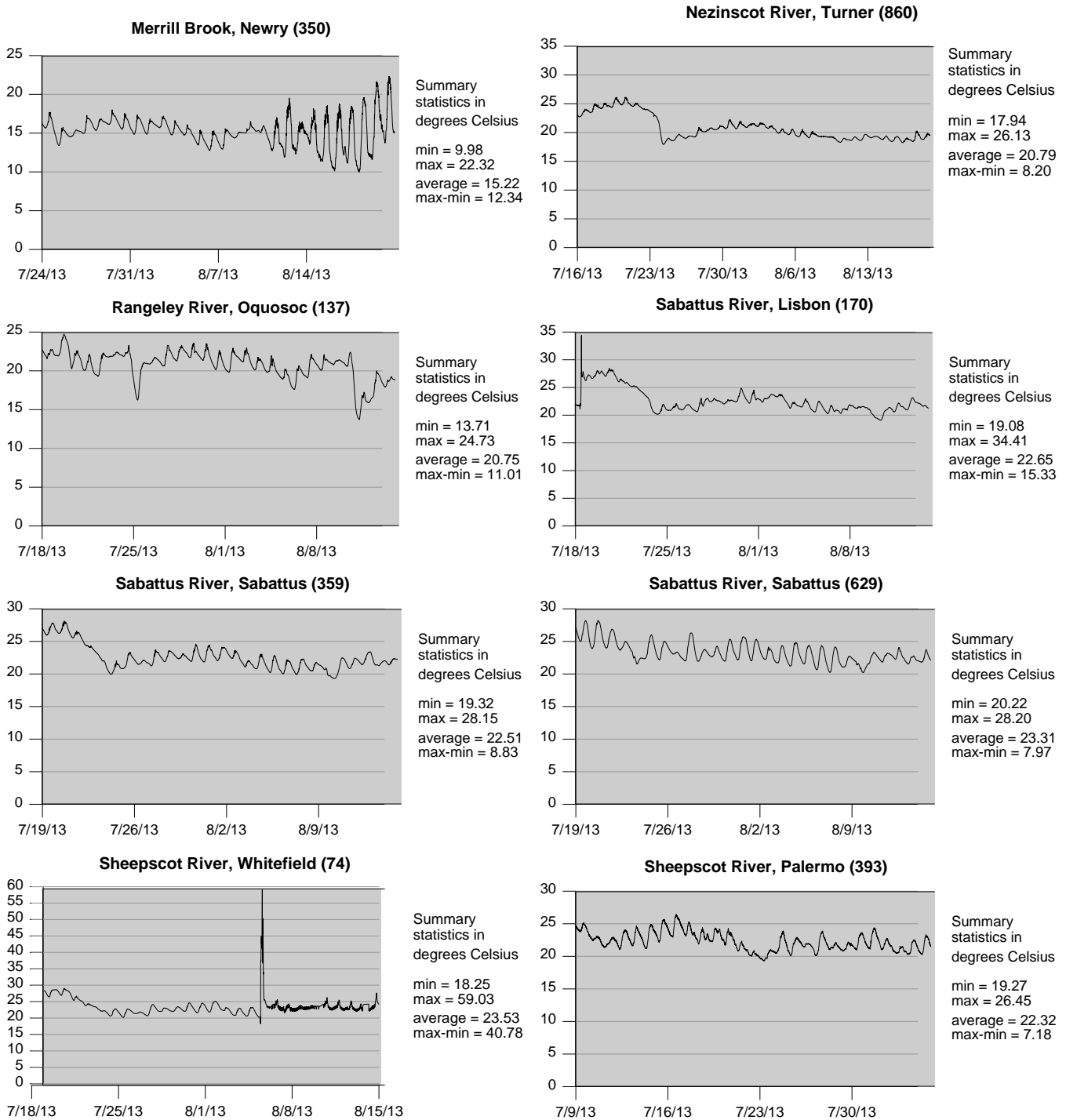
Summary statistics in degrees Celsius  
 min = 18.44  
 max = 26.26  
 average = 21.32  
 max-min = 7.82



Summary statistics in degrees Celsius  
 min = 17.63  
 max = 24.99  
 average = 20.43  
 max-min = 7.36

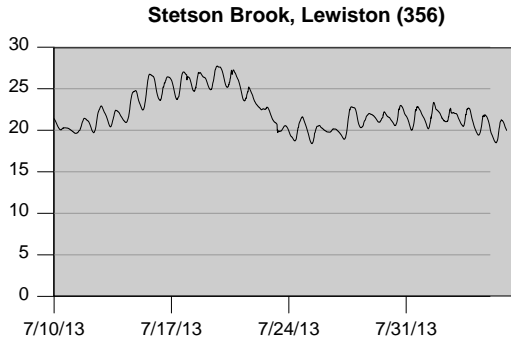
**Figure 3.1.1. 2013 In-Stream Continuous Temperature Data (continued)**

Please note: all data are in degrees Celsius

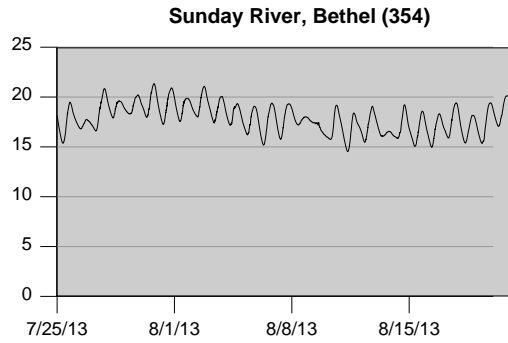


**Figure 3.1.1. 2013 In-Stream Continuous Temperature Data (continued)**

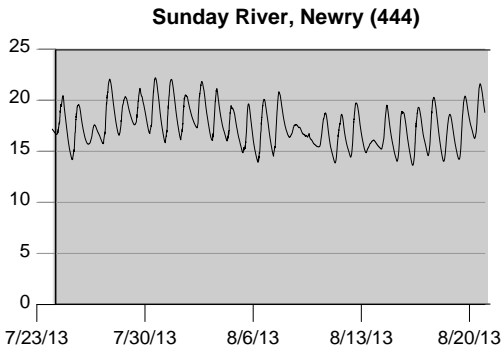
Please note: all data are in degrees Celsius



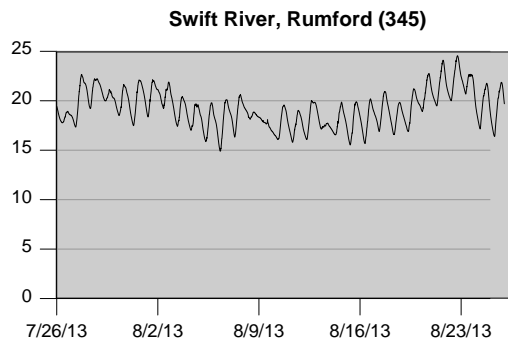
Summary statistics in degrees Celsius  
 min = 18.39  
 max = 27.73  
 average = 22.23  
 max-min = 9.34



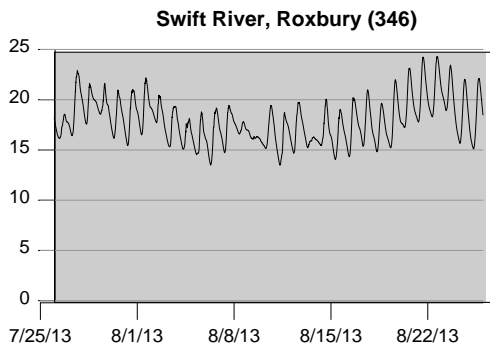
Summary statistics in degrees Celsius  
 min = 14.53  
 max = 21.34  
 average = 17.85  
 max-min = 6.81



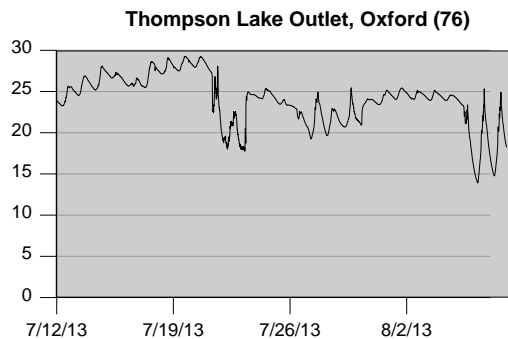
Summary statistics in degrees Celsius  
 min = 13.64  
 max = 22.20  
 average = 17.51  
 max-min = 8.56



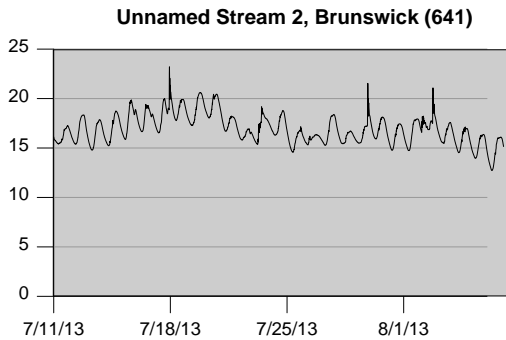
Summary statistics in degrees Celsius  
 min = 13.50  
 max = 24.29  
 average = 17.96  
 max-min = 10.80



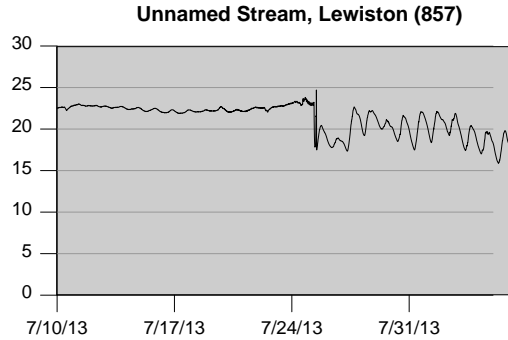
Summary statistics in degrees Celsius  
 min = 13.50  
 max = 24.29  
 average = 17.96  
 max-min = 10.79



Summary statistics in degrees Celsius  
 min = 13.90  
 max = 29.27  
 average = 23.99  
 max-min = 15.36



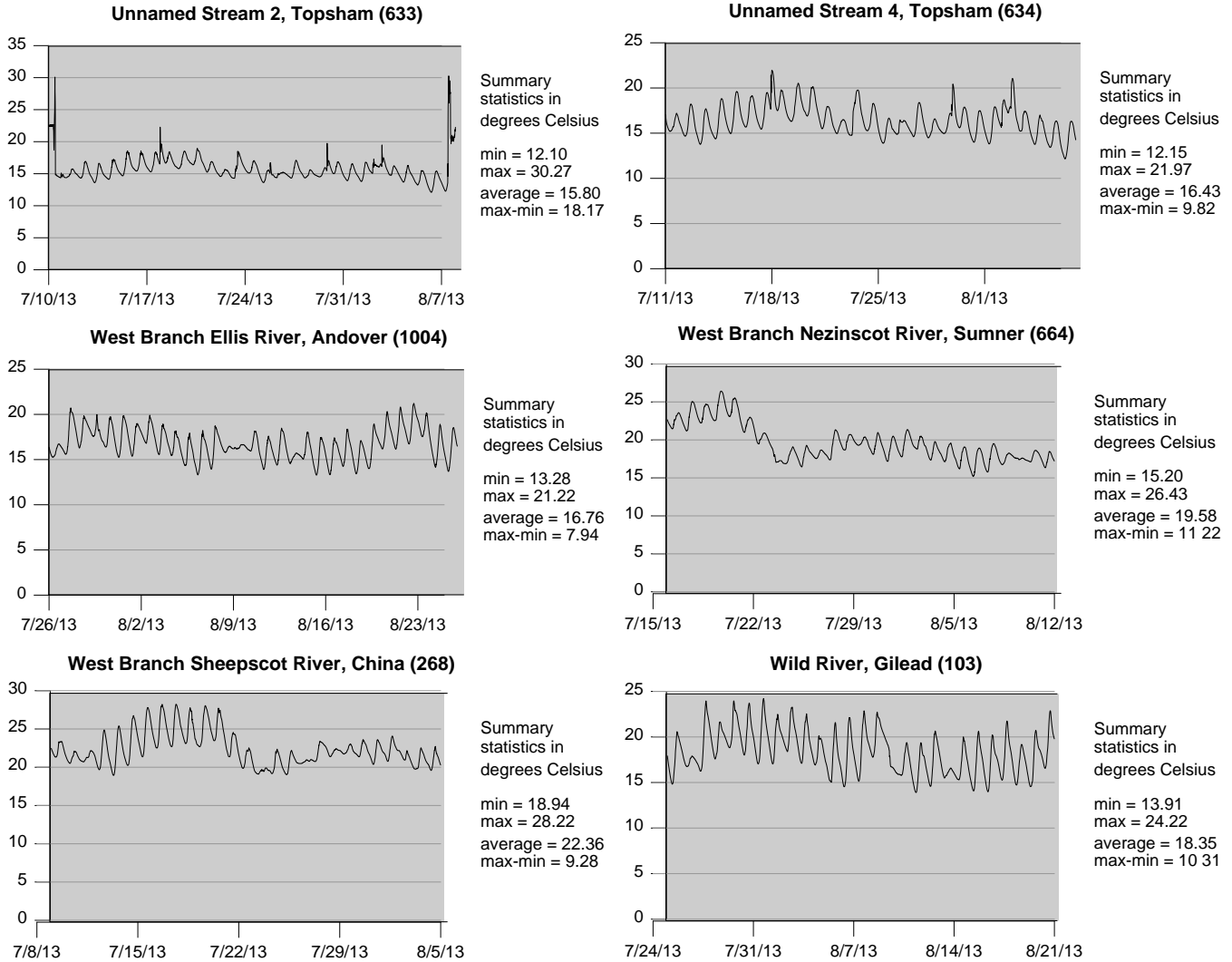
Summary statistics in degrees Celsius  
 min = 12.73  
 max = 23.21  
 average = 16.94  
 max-min = 10.48



Summary statistics in degrees Celsius  
 min = 15.87  
 max = 24.70  
 average = 21.29  
 max-min = 8.84

**Figure 3.1.1. 2012 In-Stream Continuous Temperature Data (continued)**

Please note: all data are in degrees Celsius



**3.1.3 Attainment History of Sampling Stations prior to 2013**

The table below provides the attainment history for sampling stations that have been sampled in the past.

**Table 3.1.4. Past Attainment History**

Waterbody	Station	Attained Class	Did not Attain Class	Indeterminate Result
Androscoggin River	355	1998, 2003, 2008		
Aunt Hannah Brook	343	1998, 2003, 2008		
Bear River	866	2008		
Bobbin Mill Brook	357	2003, 2008	1998	



<b>Waterbody</b>	<b>Station</b>	<b>Attained Class</b>	<b>Did not Attain Class</b>	<b>Indeterminate Result</b>
East Branch Wesserunsett Stream	486	2001, 2007, 2012		
East Cathance Stream	859			
Halfmoon Stream	697	2003, 2007	2012	
Hart Brook	341		1998, 2003, 2008	
Kennebago River	868	2008		
Little Androscoggin River	43	1998, 2003, 2008		
Little Androscoggin River	79	1998, 2003, 2008		
Martin Stream	693	2003, 2008		
Merrill Brook	350	1998, 2003, 2008		
Nezinscot River	860	2008		
Rangeley River	137	2006	2003	
Sabattus River	170	2010	1998, 2003, 2008	
Sabattus River	359	1998, 2008		
Sabattus River	629	2002, 2003, 2008		
Sheepscot River	74	1987 - 1990, 1992, 1995, 1996, 1998 - 2012	1984 - 1986, 1991, 1993, 1994, 1997	
Sheepscot River	393	2007	1999, 2006, 2012	
Stetson Brook	356	1998, 2008		
Sunday River	354	2000, 2003, 2008	1988	
Sunday River	444	2008	2000, 2003	
Swift River	345	1998, 2003,		
Swift River	346	1998, 2003		2008
Thompson Lake Outlet	76	1998, 2003		
Unnamed Stream (Brunswick 2)	641		2002, 2008	
Unnamed Stream (Lewiston)	857		2008	
Unnamed Stream (Topsham 2)	633	2006	2002, 2007	2008
Unnamed Stream (Topsham 4)	634	2002, 2006	2008	
West Branch Ellis River	872	2008		
West Branch Nezinscot River	664	2003, 2008		
West Branch Sheepscot River	268	1996 - 1999, 2001, 2002, 2005, 2007, 2009 - 2012	2000, 2003, 2004, 2006, 2008	1995
Wild River	103	1987, 2008		

## 3.2 FISH CONSUMPTION ADVISORIES

### 3.2.1 Dioxin and PCBs

#### 3.2.1.1 Introduction

Maine's Dioxin Monitoring Program (DMP), established in 1988, was merged with the Surface Water Ambient Toxics (SWAT) monitoring program in 2007 as 38 MRSA 420-B sub-§1-A for Dioxin monitoring. The goal of the monitoring is "to determine the nature of dioxin contamination in the waters and fisheries of the State" and to "determine the need for fish consumption advisories on affected waters". Charged with administration of the program, the Commissioner of the Department of Environmental Protection (DEP) is required to

- 1) Select a representative sample of wastewater treatment plant sludges from municipal wastewater treatment plants, bleached pulp mills or other sources. These facilities must be selected on the basis of known or likely dioxin contamination of their discharged effluent;
- 2) Sample and test the sludge of selected facilities of dioxin contamination at least once during each season of the year. The commissioner shall specify which congeners of dioxin will be analyzed;
- 3) At appropriate intervals, sample and test for dioxin contamination in selection of fish representative of those species present in the receiving waters of where there are consumption advisories for dioxin; Sufficient numbers of fish must be analyzed to provide a reasonable estimate of the level of contamination in the population of each waterbody affected; and
- 4) Assess the selected facilities of the costs of sample collection and analysis except that, if the selected facility is a publicly owned treatment works, the Commissioner may assess the primary industrial generator discharging effluent into the treatment facility if the generator is known or likely to be discharging dioxin into the treatment facility. Fees received under this subparagraph must be credited to the Maine Environmental Protection Fund. Payment of these fees is a condition of the discharge license issued pursuant to section 413 for continued operation of the selected facilities, except that if the selected facility is a publicly owned treatment works and the Commissioner assesses the fee on an industrial generator, payment of the fee is not a condition of the discharge license of the selected facility. The fees assessed under this subparagraph may not exceed a total of \$250,0000 in any fiscal year. The fees assessed under this subparagraph to facilities subject to section 420, subsection 2, paragraph I may not exceed a total of \$10,000 in any fiscal year.

The monitoring program is to be coordinated with other ongoing programs conducted by the Department, the Maine Center for Disease Control and Prevention (ME-CDC), US Environmental Protection Agency (EPA) and other federal agencies, or dischargers of wastewater. The proposed annual monitoring plan must be submitted to the Surface Water Ambient Toxics (SWAT) Technical Advisory Group (TAG), created under 38 MRSA section 420-B, for review and advice. The selected facilities must be notified of their inclusion in the proposed program at least 30 days prior to submittal to the TAG.

### 3.2.1.2 Program Design

Following attainment of the provisions of the 1997 Dioxin Law and elimination of the measurable discharge of dioxins (includes closely related furans) from the bleached kraft pulp and paper mills in 2003-2005, the Dioxin Monitoring Program is now focused on residual levels of dioxins from historic discharges and how they affect Maine's fish consumption advisories. This report contains the findings from the 2013 Dioxin Monitoring Program with respect to three objectives:

1. Human health assessment, Fish Consumption Advisories
2. Trend evaluation
3. 1997 Dioxin Law, Continued Compliance

This report also contains the (dioxin-like) coplanar polychlorinated biphenyl (PCB) data. Coplanar PCB data are included to show the total exposure to dioxin-like compounds from consumption of certain fish from several Maine rivers. The Environmental and Occupational Health Program (EOHP) of the Maine Center for Disease Control and Prevention (ME-CDC) uses both dioxins and coplanar PCB data, which have similar toxicity characteristics to dioxins, in order to make a complete assessment of the fish consumption advisories. Sources of the coplanar PCBs are not known, but likely include historic use and discharge in Maine, and long range transport and atmospheric deposition.

In January 2008, the ME-CDC issued a report titled 'Evaluation of the Health Implications of levels of Polychlorinated Dibenzo-p-Dioxins (dioxins) and Polychlorinated Dibenzofurans (furans) in Fish from Maine Rivers – 2008 Update'. In the report, ME-CDC adopted a new provisional Fish Tissue Action Level (FTAL) of 0.4 ppt, based on the same toxicity data for non-cancer effects used since 1990, but adjusted downward to account for substantial background exposure from other dietary foods. ME-CDC reviewed the data collected since their last review in 2003, i.e. 2004-2007 with respect to the new FTAL.

For 2009, ME-CDC did not request any monitoring, but did request monitoring in 2010, 2011, 2012 and 2013.

### 3.2.1.3 2013 monitoring requested by Maine Center for Disease Control and Prevention

#### I. Basis for Fish Sampling Requests

Suggestions for fish sampling are based on comparison of existing data with Fish Tissue Action Levels (FTALs) developed by the Environmental and Occupational Health Program (EOHP) of the Maine Center for Disease Control and Prevention (MECDC). FTALs are derived using a hazard index of one, or an increased cancer risk of 1 in 100,000, with a fish consumption rate of one 8 ounce meal per week. For dioxin and dioxin like compounds, the FTAL of 1.5 parts per trillion (ppt) is based on cancer risk for the general population.

The FTAL for dioxin is based on one meal per week, however the current statewide mercury advisory for the general population restricts intake of all fish species (except brook trout and landlocked salmon) to no more than two meals per month. A dioxin specific consumption advisory is necessary if the level of dioxins in fish present a hazard at the consumption limits already in place for mercury. Because the statewide advisory for mercury limits consumption to 2 meals per month, the level of dioxins and dioxin-like PCB congeners that would exceed the risk based consumption limit is doubled. In other words, a fish tissue level of 3.0 ppt dioxin equivalents, consumed on a 2 meal per month basis, will not exceed the risk based consumption limit for the general population.

The current statewide mercury fish consumption advisory restricts sensitive populations from any consumption of freshwater fish (except brook trout and landlocked salmon). Although sensitive populations are restricted from freshwater fish consumption, ME-CDC has developed a provisional FTAL for dioxins and dioxin-like compounds for non-cancer effects applicable to sensitive populations. The MECDC non-cancer FTAL for dioxin and dioxin-like compounds (0.4 ppt) incorporates an estimate of background dietary exposure. Further discussion of the derivation of this FTAL for sensitive populations can be found at <http://www.maine.gov/dhhs/mecdc/environmental-health/eohp/fish/index.htm>. US EPA updated the toxicity profile for dioxin in IRIS for 2,3,7,8-TCDD in 2012, and adopted a reference dose of 0.7 nanogram per kilogram body weight-day (ng/kg-day). This value is slightly lower than the reference dose used by Maine as the basis for the current non-cancer FTAL (1 ng/kg-day), and more importantly provides a much stronger scientific basis for this endpoint. However, MECDC does not intend to modify the non-cancer FTAL for dioxins until we have also reviewed background dietary exposure. The background dietary exposure used to develop the provisional FTAL was based on the data available in 2003. EPA is progressing toward completion of its comprehensive reassessment of dioxin exposure which will contain more recent exposure estimates and it is anticipated that the provisional FTAL for sensitive populations may be affected by changes in background exposure estimates. The current statewide mercury consumption advisory restricts women of child bearing age and children under age 8 from consuming any freshwater fish from Maine's inland waters, except for 1 meal per month of brook trout and landlocked salmon. Any change in the dioxin FTAL for non-cancer effects is not expected to impact fish advisories.

In reviewing the most recent fish tissue data, we have identified a few stations where total TEQ concentrations nearly equal or slightly exceed 3 ppt (the concentration that would result in a consumption advisory more restrictive than the current two meal per month statewide mercury advisory for the general population). In addition, recent data has demonstrated that fish tissue concentrations at several stations near this screening-level criteria have decreased. EPA guidance recommends that if contamination levels fall below a screening value, in this case the screening value is 3 ppt for the general population, they be confirmed in a subsequent sampling round (EPA Guidance for Assessing Chemical Contaminant Data for use in Fish Advisories Volume 1, 2000). Recommendations for sampling for the coming year are focused on further characterization of fish sampling stations that previously had borderline total TEQ concentrations, to support upcoming revisions to the river specific fish consumption advisories.

## 2. Fish Sampling Requests

The following is a list of waters and fish species for which additional data are needed for continued monitoring for fish consumption advisories.

### 1. Dioxin And PCB Data (Total PCBs or Dioxin-Like PCB Congeners) for the Major Rivers

Recommendations for sampling for dioxins and PCBs are provided below, by river. These requests are combined because we would like both dioxin and PCB data for every fish sampled. As in 2012, composite samples, with two samples being submitted for chemical analysis for any sampling location - fish species combination, are recommended. Analysis for total PCBs by the congener method is requested. Quantification of total PCBs by the congener method will provide measurement of both dioxin-like PCB congeners and total PCBs, both of which will be used to support revisions to the current fish advisories.

#### Androscoggin River

##### Gilead - white sucker

Dioxin TEQ in white suckers at Gilead, was approximately 8.6 ppt in 1996 and 5.8 ppt in 1997. There has been no subsequent sampling for dioxins at Gilead. ME-CDC requested DEP to sample white suckers at Gilead for dioxins and PCBs to demonstrate that the total dioxin TEQ levels are below the screening level of 3 ppt.

#### Rumford - white sucker

The 2001 sampling of white suckers at Rumford determined total TEQ to be approximately 4.9 ppt (dioxin TEQ approximately 4 ppt, PCB TEQ approximately 1 ppt). In 2002 the total dioxin TEQ was 1.9 ppt, ME-CDC requested DEP to confirm.

#### Auburn Gilead Island Pond (GIP) - white sucker

Dioxin TEQ in white suckers at GIP, decreased substantially between 2007 and 2008, from approximately 2.4 ppt to approximately 1.3 ppt, respectively. ME-CDC requested DEP to confirm this decrease with the 2013 sampling effort.

#### Lisbon- white sucker

The 1996 sampling of white suckers at Lisbon determined dioxin TEQ to be approximately 4.8 ppt. In 2007 the total dioxin TEQ was 1.0 ppt, ME-CDC requested DEP to confirm this decrease.

#### Kennebec River

##### Augusta- white sucker

Investigation of dioxins in white suckers collected in 1996 determined dioxin TEQ to be approximately 6.1 ppt. The 2009 sampling resulted in a PCB TEQ of approximately 0.5 ppt. ME-CDC requested DEP to determine whether or not the total TEQ for suckers at Augusta is below 3.0 ppt.

#### Penobscot River

##### Hamden and Orrington - eel

Eel have been collected from the Penobscot River at Hamden in 1996 and 2000 (dioxins only, TEQ 2.4 and 3.6 ppt, respectively), and at Orrington in 2001 with total TEQ 9.2 ppt (4.2ppt PCB TEQ, 4.9

ppt dioxin) and 2002, PCB only TEQ approximately 3.2ppt . These concentrations may warrant a species specific advisory. ME-CDC requested DEP to demonstrate that these are the current concentrations of dioxins and PCBs in eel in the Penobscot River.

#### Milford - white sucker

Investigation of dioxins in white suckers collected between 1997 and 2002 demonstrated total dioxin TEQ to be approximately from 3.7 to 5.4 ppt. No sampling has been done since 2002. Although the dioxin TEQ was slightly under 3 ppt, (approximately 2.7 ppt in both 2001 and 2002), the PCB contribution to the total TEQ at this sampling station has been substantial. ME-CDC requested DEP to determine that the dioxin total TEQ for suckers at Augusta is below 3.0 ppt.

#### Salmon Falls River

##### South Berwick - white sucker

White suckers collected in 1996 had a dioxin TEQ of approximately 8.7 ppt. This dropped to approximately 0.7 ppt in 2008. ME-CDC requested DEP to confirm the decrease in the dioxin total TEQ for suckers at South Berwick.

St. Croix River: No dioxin or PCB data were requested at this time.

Presumpscot River: No dioxin or PCB data were requested at this time.

Androscoggin Lake: No dioxin or PCB data were requested at this time.

#### DDT Data for the Aroostook County Rivers

No data were requested for DDT and its metabolites, DDE and DDD at this time.

### 3.2.1.4 Sampling Plan

In 2013, fish were collected by DEP by use of angling and gill nets. Fish were immediately euthanized, weighed and measured, rinsed in river water, wrapped in aluminum foil with the shiny side out, labeled, and placed in a cooler on ice for transport to DEP for secure storage in the freezer. Samples were transferred from DEP to the analytical laboratory for analysis using EPA method 1613b. All other procedures generally followed EPA's Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories, Volume I Fish Sampling and Analysis, 1993. Completed chain-of-custody field forms were kept in the freezer storage area for an inventory of samples at any time and an Excel spreadsheet documented final disposition of samples.

A total of 10 fish were targeted for collection and analysis at selected stations (Table 3.2.1). Skinless filets from all fish were analyzed for all 2378 substituted dioxins and furans and the 209 PCB congeners from which the TEQs (dioxin toxic equivalents (DTEhu) and coplanar PCB toxic equivalents CTEhu) were calculated. Sample costs were reduced from that of earlier years by combining the 10 fish into analysis of two composites of five fish for each station. Facilities with known or likely dioxin contamination of their discharged effluent, identified as a DMP facility, were assessed fees for the cost of chemical analysis of samples below their discharge. Analysis of other samples, identified as SWAT samples, was funded by DEP.

RIVER	STATION	FISH predators	FISH omnivores	SAMPLES N	DMP\$ PCDD/F	DMP\$ facility	SWAT\$ PCDD/F	SWAT\$ TCPB	SWAT\$ total	TOTAL COST
ANDROSCOGGIN	GILEAD		2C5 WHS	2			1540	900		2440
	RUMFORD		2C5 WHS	2	1540	RPC		900		2440
	GULF ISLAND POND		2C5 WHS	2	1540	VERSO		900		2440
	LISBON		2C5 WHS	2			1540	900	6680	2440
KENNEBEC	AUGUSTA		2C5 WHS	2	1540	SAPPI SOMERSET		900	900	2440
PENOBSCOT										
	MILFORD		2C5 WHS	2	1540	LINCOLN PAPER		900		2440
	HAMDEN		2C5 EEL	2	1540	RED SHIELD		900	1800	2440
SALMON FALLS	S BERWICK		2C5 WHS	2			1540	900	2440	2440
ANALYTICAL TOTAL				12	7700		4620	7200	11820	19520
misc supplies @ \$200 each					1000					
state indirect cost @ ~15%					1305					
TOTAL					10005					
legislative cap					10000					

An analytical issue is that of estimated maximum possible concentrations (EMPC). Some compounds, particularly hydroxydiphenyl ethers (DPEs), are coextracted with furans. Laboratory analysis has been modified to minimize these interferences, but some DPEs may remain. In the 2007 Dioxin Monitoring Program report, the Maine Center for Disease Control and Prevention calculated EMPCs as a detected value according to their policy for setting the fish consumption advisories. To be consistent for comparison with ME-CDC's FTAL, EMPCs were treated the same way in this report.

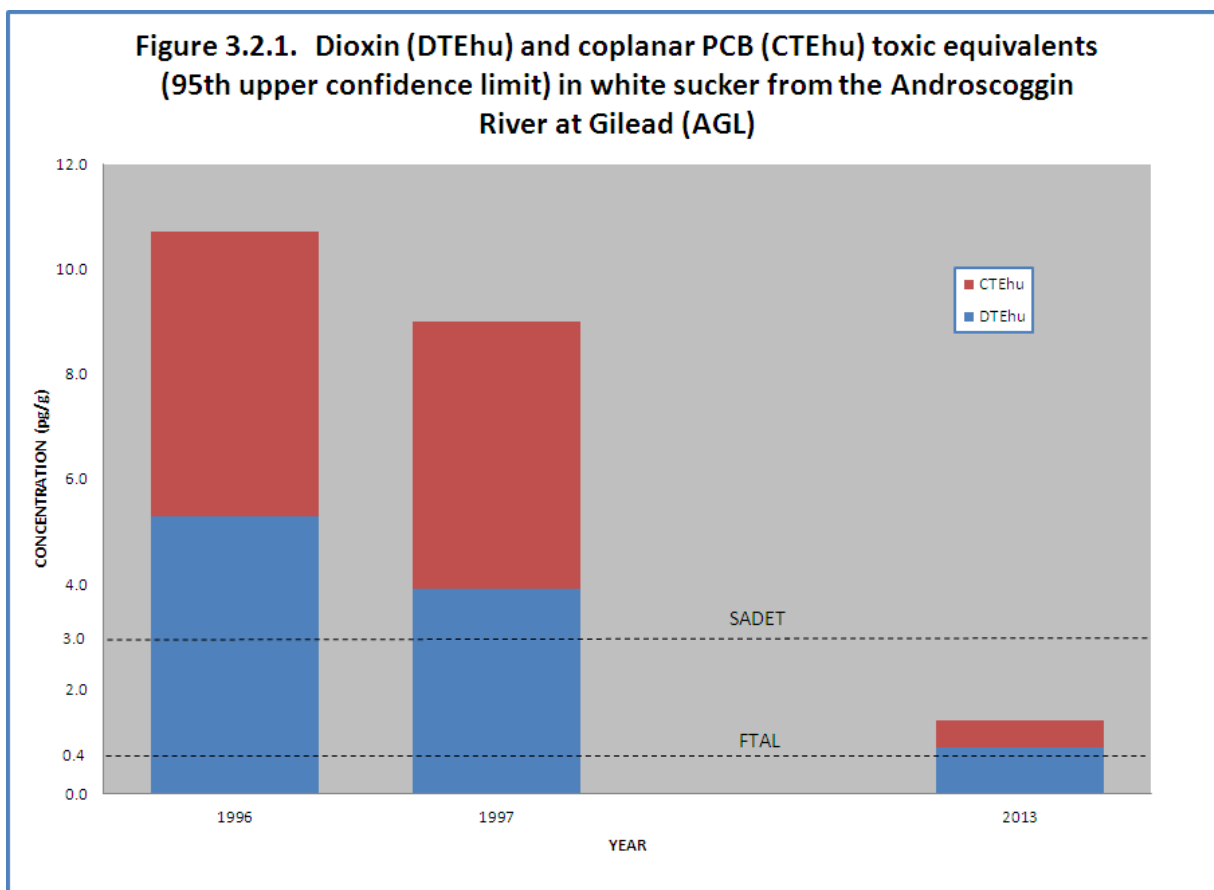
### 3.2.1.4 Results and Discussion

#### DIOXINS AND COPLANAR PCBS

##### *Androscoggin River*

##### Gilead

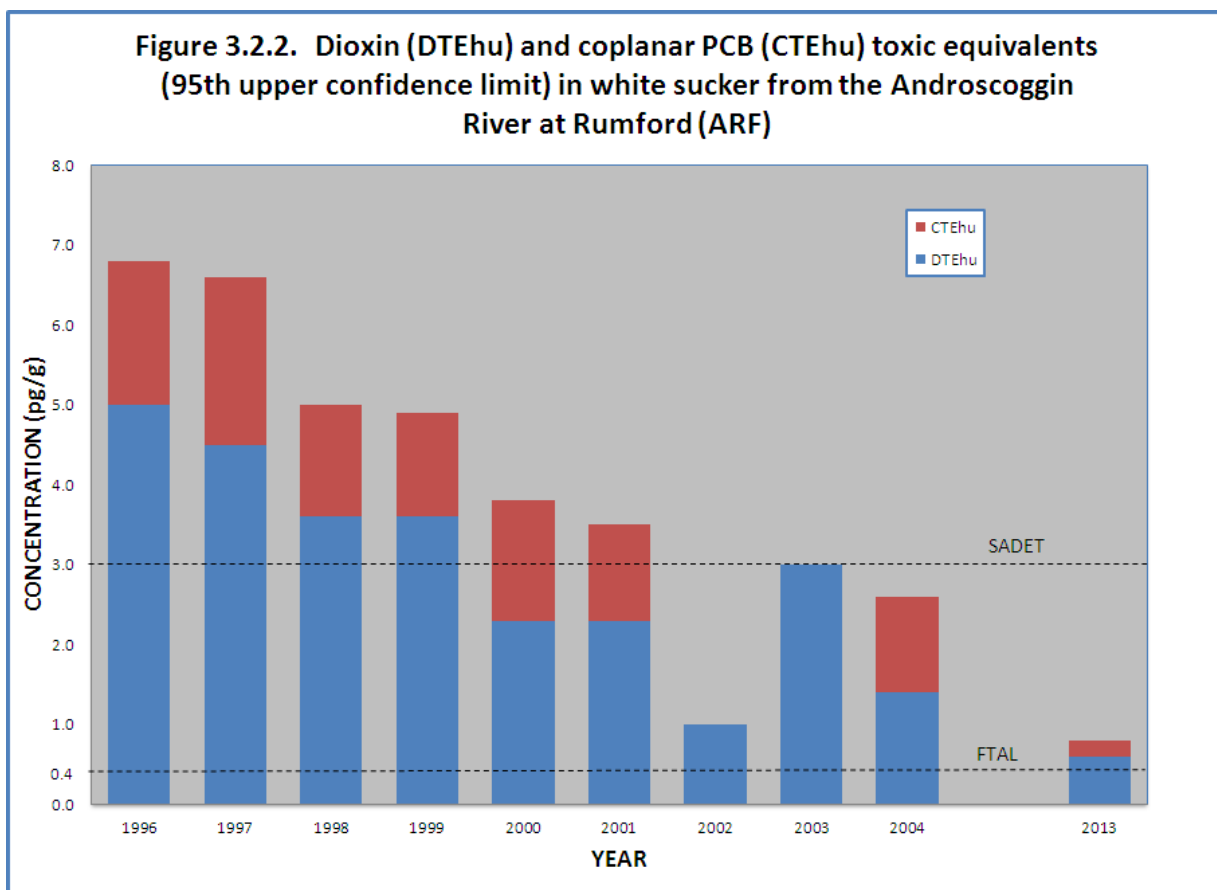
A total of ten white sucker (WHS) were successfully collected from the Androscoggin River at Gilead (AGL) for the first time since 1997. The dioxin toxic equivalent (DTEhu, calculated with non-detects at half of the detection limit as the upper 95<sup>th</sup> confidence level) concentration was slightly above the ME-CDC FTAL (0.4 pg/g) for dioxin-like compounds but below a statewide advisory dioxin equivalent threshold (SADET= 3 pg/g) for mercury (Figure 3.2.1). Concentrations of dioxin-like coplanar PCB toxic equivalents (CTEhu, calculated at the 95<sup>th</sup> upper confidence level with non-detects at ½ of the detection limit) added only a small amount to total toxic equivalents. Concentrations of both DTEhu and CTEhu are well below those from previous years for white sucker, consistent with a trend of declining concentrations in trout at this station since the late 1990s.





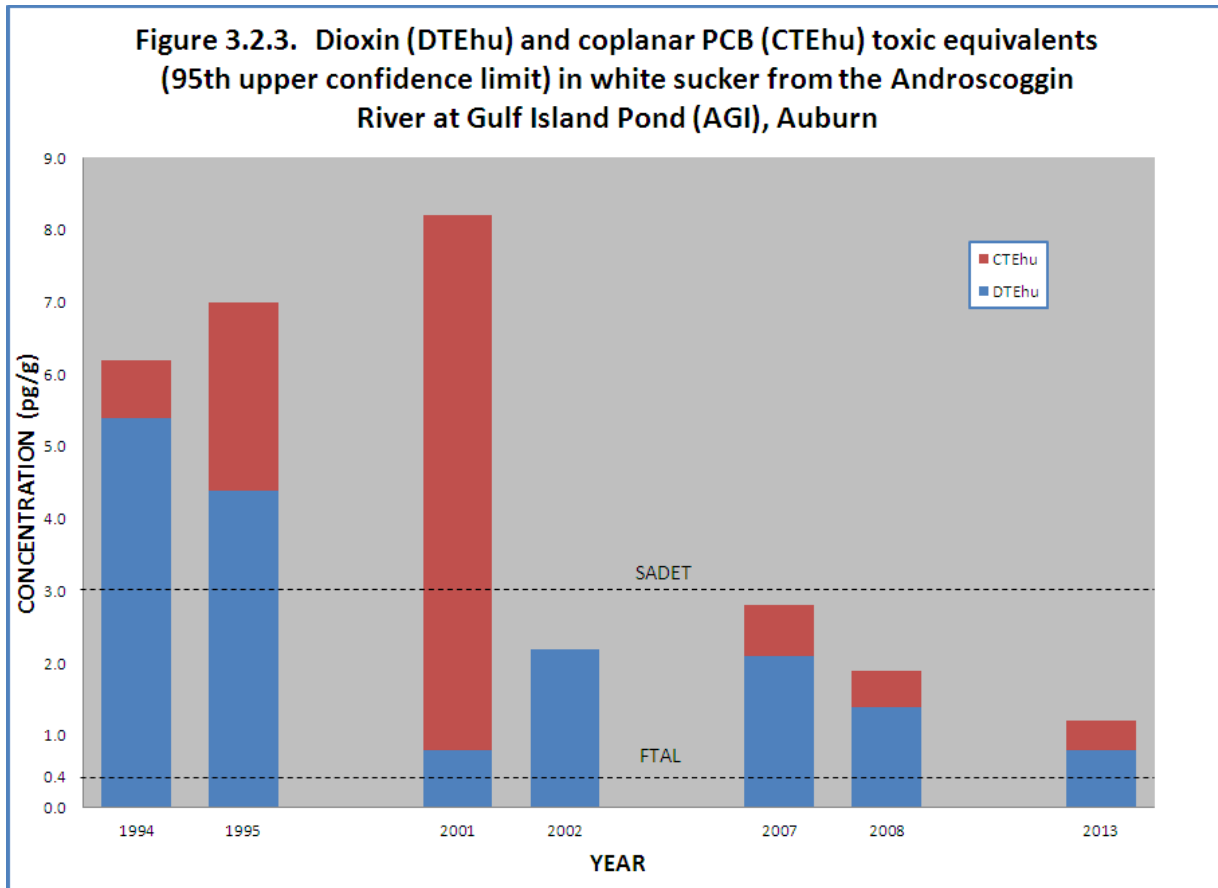
## Rumford

A total of ten white sucker (WHS) were successfully collected from the Androscoggin River below the Rumford Paper Company mill at Rumford (ARF). The dioxin toxic equivalent (DTEhu, calculated with non-detects at half of the detection limit as the upper 95<sup>th</sup> confidence level) concentration exceeded the ME-CDC FTAL for dioxin-like compounds, but was below a statewide advisory dioxin equivalent threshold (SADET= 3 pg/g) for mercury (Figure 3.2.2). Concentrations of coplanar PCB toxic equivalents (CTEhu, calculated at the 95<sup>th</sup> upper confidence level with non-detects at ½ of the detection limit) did not exceed the FTAL alone and the combination of the two groups of contaminants increased the exceedance of the FTAL very little. Concentrations of both DTEhu and CTEhu were well below those of previous years consistent with a declining trend.



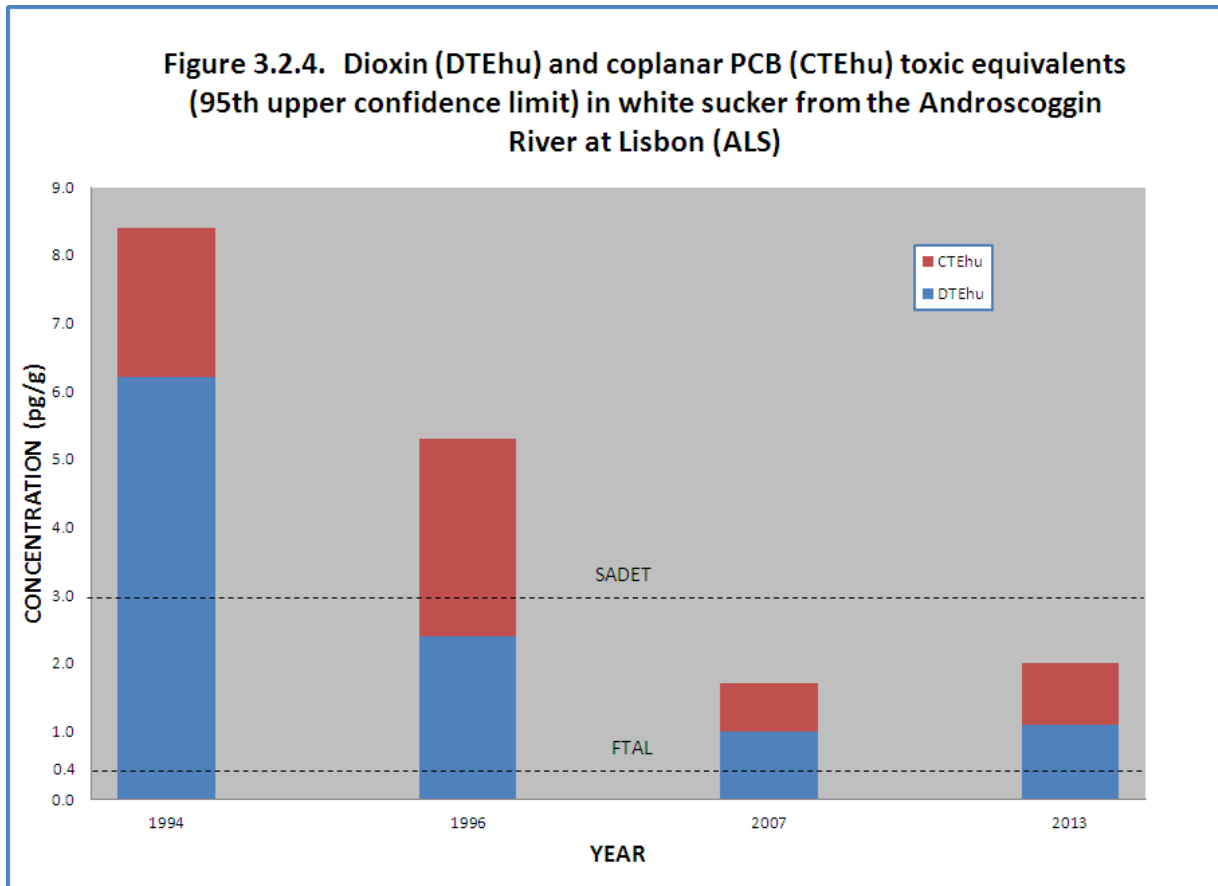
Gulf Island Pond, Auburn

A total of ten white sucker (WHS) were collected from the Androscoggin River in Gulf Island Pond (AGI) at Auburn. The DTEhu concentration exceeded the FTAL for dioxin-like compounds, but was below the SADET (Figure 3.2.3). The CTEhu concentration did not exceed the FTAL alone and the combination of the two groups of contaminants were well below the SADET. Concentrations of both DTEhu and CTEhu were below those of previous years showing a decline since the mid-1990s. The high CTEhu concentration in 2001 seems to be an anomaly.



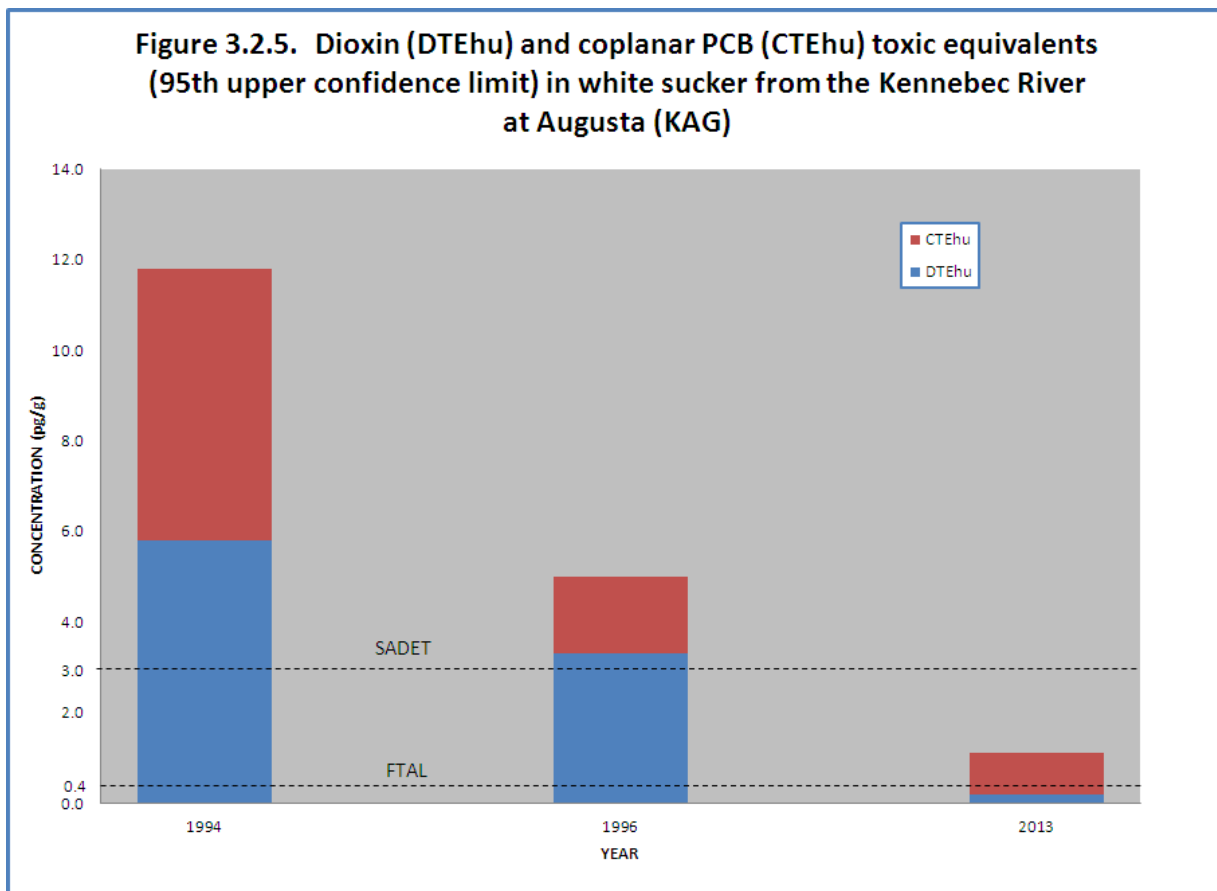
Lisbon

A total of ten white sucker (WHS) were collected from the Androscoggin River at Lisbon (ALS). The DTEhu concentration exceeded the FTAL for dioxin-like compounds, but was below the SADET (Figure 3.2.4). The CTEhu concentration also exceeded the FTAL alone but the combination of the two groups of contaminants remained below the SADET. Concentrations of both DTEhu and CTEhu were similar to those when last measured in 2007 but lower than those of previous years.



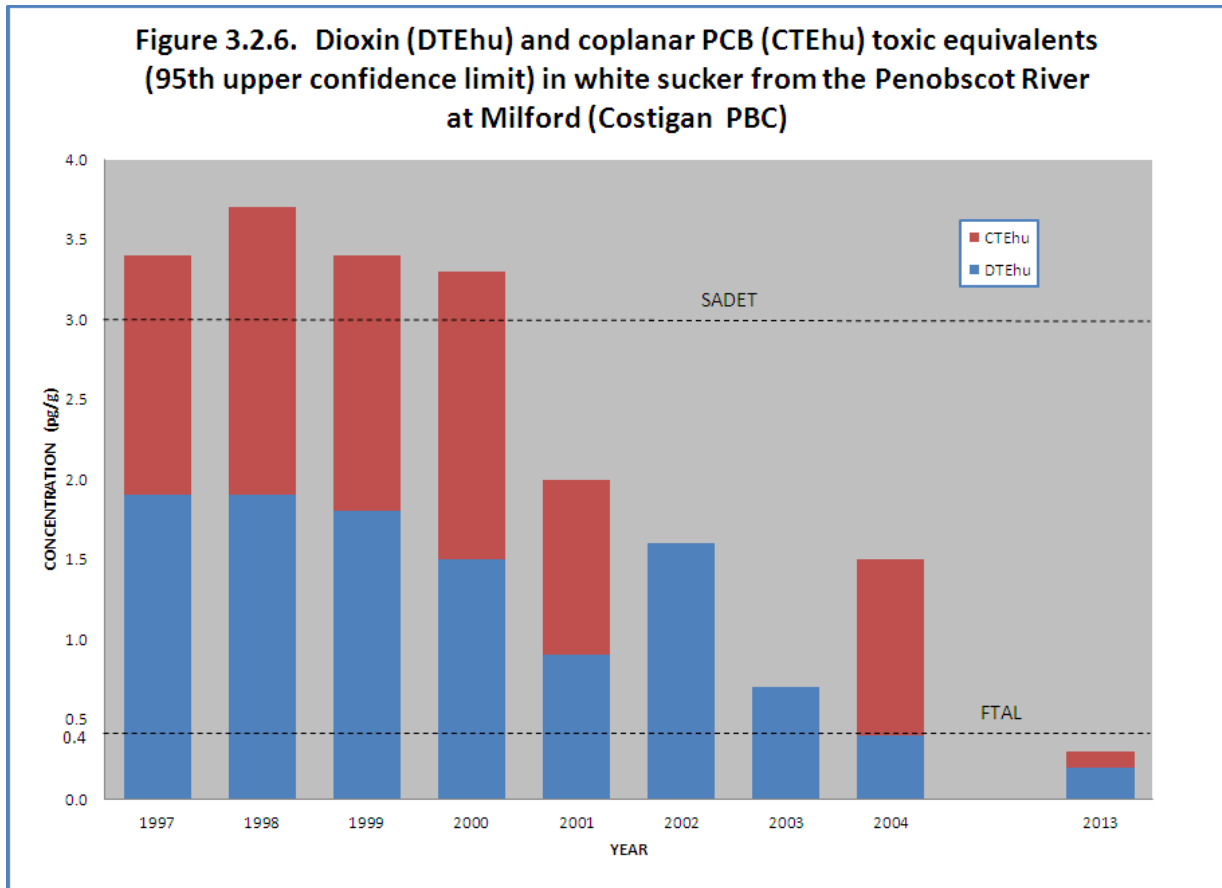
*Kennebec River*Augusta

A total of ten white sucker (WHS) were collected from the Kennebec River at Augusta (KAG). The DTEhu concentration was below the FTAL for dioxin-like compounds (Figure 3.2.5). The CTEhu concentration did exceed the FTAL alone but the combination of the two groups of contaminants remained well below the SADET. Concentrations of both DTEhu and CTEhu were well below those from earlier years.



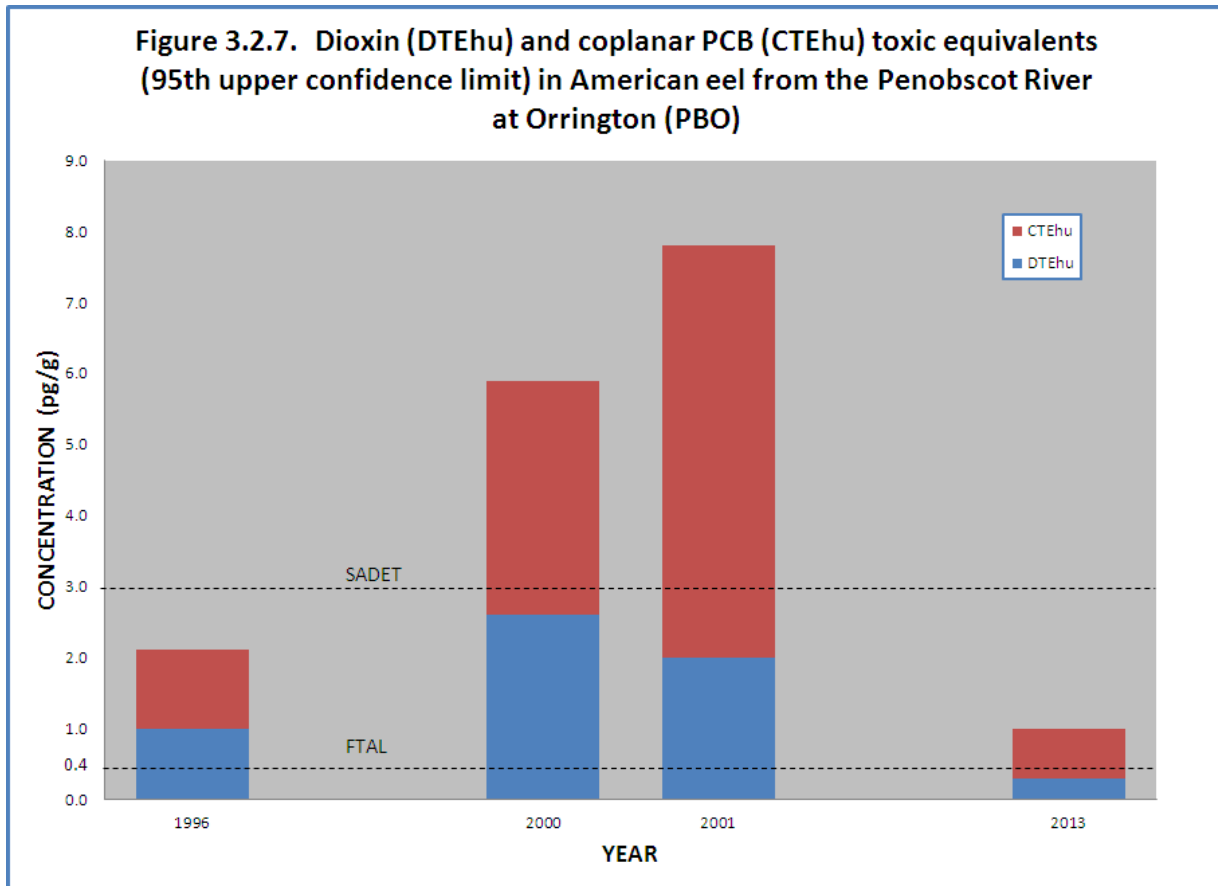
*Penobscot River*Milford

A total of ten white sucker (WHS) were collected from the Penobscot River at Milford (Costigan, PBC). The DTEhu concentration was below the FTAL for dioxin-like compounds (Figure 3.2.6). The CTEhu concentration was low. The sum of both DTEhu and CTEhu exceeded neither the FTAL nor the SADET. Both groups of contaminants documented a continuing declining trend.



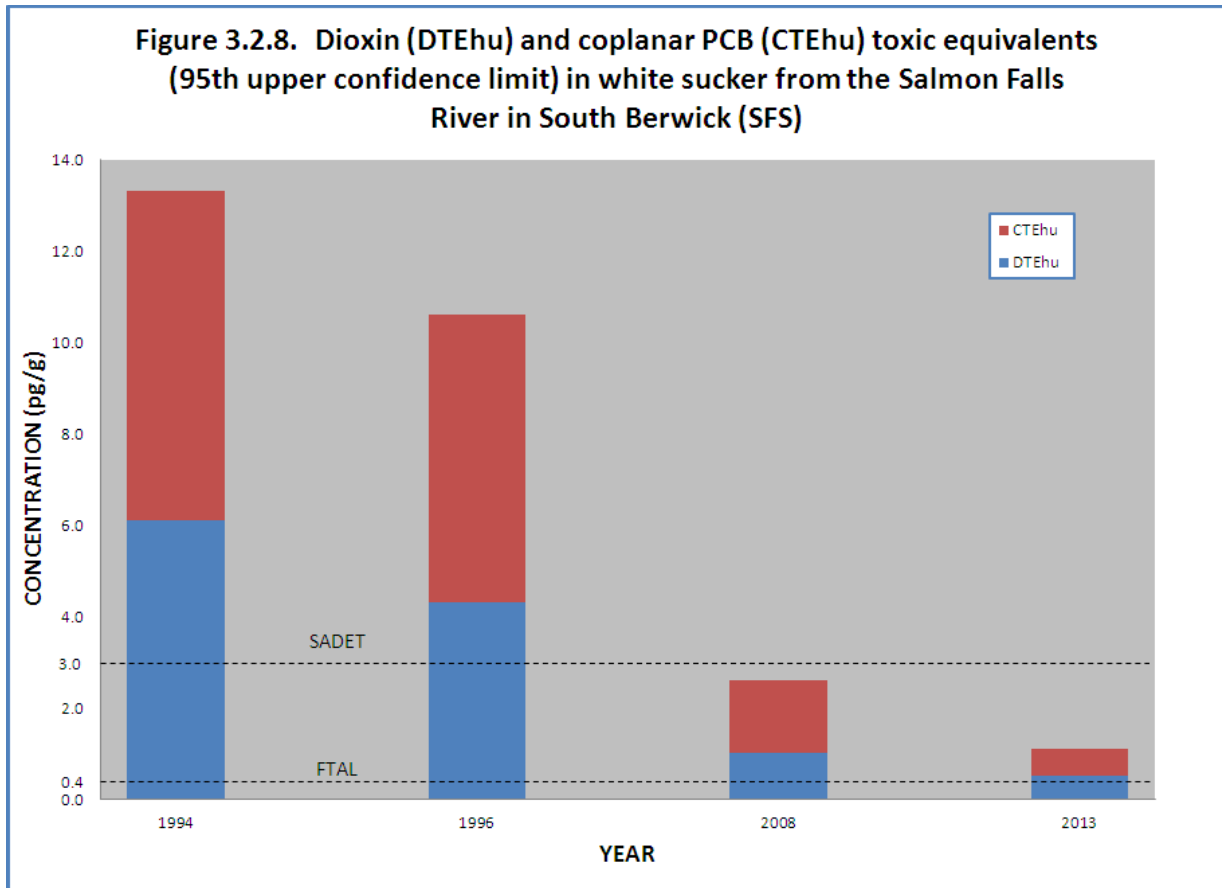
Orrington

A total of ten American eel (EEL) were collected from the Penobscot River at Orrington (PBO). The DTEhu concentration was below the FTAL for dioxin-like compounds (Figure 3.2.7). The CTEhu concentration alone exceeded the FTAL. The sum of both DTEhu and CTEhu exceeded neither the FTAL nor the SADET. Concentrations of both were similar to those in 1996 and much lower than those in 2000 and 2001.



*Salmon Falls River*South Berwick

A total of ten white sucker (WHS) were collected from the Salmon Falls River at South Berwick (SFS). The DTEhu concentration exceeded (barely) the FTAL for dioxin-like compounds (Figure 3.2.8). The CTEhu concentration also exceeded the FTAL. The sum of both DTEhu and CTEhu did not exceed SADET. Both groups of contaminants documented a continuing declining trend.



## TOTAL PCBs

*Androscoggin River*

In 2013, mean and maximum total PCB (TPCB) concentration in white sucker from Gilead were above the ME-CDC's FTAL of 11 ng/g and slightly higher than that in 2009 (Table 3.2.2). Mean and maximum TPCB concentrations in white sucker (WHS) below Rumford (ARF) were lower than at Gilead but exceeded the FTAL and were intermediate those of previous years when concentrations still exceeded the FTAL. Mean and maximum concentrations of TPCB in white sucker (WHS) from Auburn GIP (AGI), below Rumford, also exceeded the FTAL and were higher than those at Rumford and intermediate those of previous years. Mean and maximum concentrations of TPCB in white sucker (WHS) from Lisbon (ALS), below Jay, also exceeded the FTAL intermediate to those of previous years, and were similar to those at Gilead highest for the river. The variation in concentrations within species among all years may reflect differences among the four different labs that were used, although all data met quality assurance and control objectives. The variation may also be simply the natural variation in individuals and condition among years.

Table 3.2.2. Total PCBs in fish from the Androscoggin River, ng/g, mean and (max value where n=2 or 95th upper confidence level where n>2)

Year	Species	Gilead AGL	Rumford Pt ARP	Rumford ARF	Riley ARY	Jay ARJ	Livmore ALV	Livmore Fis ALF	AUBURN GIP AGI	Lisbon ALS	Androscoggin L ALW
2012	BKT	6 (8)									
2000	BNT	85									
1998	RBT	11									
2000	RBT	28									
2008	RBT	75 (86)									
2009	RBT	63 (73)									
1994	SMB			97		42	49		114	98	
1998	SMB		4 (4)	9 (12)	7 (8)		15 (19)		20 (26)	27 (30)	3.1
2000	SMB		10 (11)	21 (27)	15 (17)		38 (42)	27 (32)	29 (36)	52 (60)	
2001	SMB										15.5
2002	SMB		101	22	18		18		22	17	
2003	SMB						22	19			
2008	SMB								30 (35)		
2009	SMB		51 (65)						21 (24)		25 (25)
2010	SMB				47 (58)						
2011	SMB		66 (71)								
2012	SMB		59 (68)								
1994	WHS			80		129	39		114	145	
1996	WHS						31	58			
1998	WHS		17	21	24		33				5
2000	WHS						48	42			11.8
2001	WHS										
2008	WHS								80 (85)		
2009	WHS	61 (65)	36 (46)				71 (91)	40 (45)	31 (38)		32 (44)
2010	WHS				86 (110)						
2011	WHS				96 (104)		97 (110)				
2012	WHS		24 (28)		66 (84)		29 (38)				
2013	WHS	85 (93)		25 (25)					50 (57)	91 (94)	



## Kennebec River

In 2013, mean and maximum total PCB (TPCB) concentration in white sucker (WHS) from Augusta (KAG) exceeded the FTAL of 11 ng/g (Table 3.2.3). Concentrations were the lowest ever measured in white sucker at this station but still higher than measured upstream in previous years. Concentrations were only a little lower than those from 2009, both of which may be showing the beginning of a declining trend at this site.

Table 3.2.3. Total PCBs in fish from the Kennebec River, ng/g, mean and (max if n=2 or 95th upper confidence level if n>2)

Year		Norridgewock KNW	Skowhegan KSK	Fairfield KFF	Waterville KWV	Sidney KSD	Augusta KAG	Hallowell KRH	Gardiner KGD	Richmond KRD
1994	BNT			300						
1997	BNT			93 (107)			54.6 (70.9)			
1999	BNT						55 (71)			
2000	BNT	3				34 (45)				
2002	BNT	8		10						
2007	BNT	10 (14)		10 (14)						
2009	BNT			7 (7)						
1994	SMB			5		9	604			
1997	SMB	4	4 (5)	4 (5)		6 (7)	342 (357)			
1999	SMB						263 (323)		179 (227)	166
2000	SMB					32 (42)				
2002	SMB	2		2		20	111		47.5	
2006	SMB					8 (10)	83 (142)		51 (75)	
2007	SMB								52 (70)	44 (64)
2009	SMB			3 (4)		17 (22)	85 (100)			
2002	EEL									377
2005	SLT									46 (64)
2007	SLT							60 (83)		
2009	SLT								18 (20)	
1994	WHS			17		23	1354			
1996	WHS						850			
1997	WHS	7		54		12	831			
1999	WHS						708			
2009	WHS			5 (5)		46 (64)	91 (101)			
2011	WHS					26 (32)				
2012	WHS					64 (87)				
2013	WHS						67 (108)			

## Penobscot River

In 2013, mean and maximum total PCB (TPCB) concentrations in white sucker (WHS) from the Penobscot River at Milford (Costigan, PBC) did not exceed ME-CDC's FTAL of 11 ng/g (Table 3.2.4). There are no other TPCB for white sucker from this site, but the concentrations were lower than in white sucker from those at S Lincoln and Veazie in previous years. Coplanar PCBs were lower than in previous years in white sucker at this site showing a declining trend (Figure 3.2.6). Mean and maximum total PCBs in American eel at Bangor (Orrington, PBO) exceeded the FTAL and were near the lower end of the range of those of previous years (Table 3.2.4).

Table 3.2.4. Total PCBs in fish from the Penobscot River, ng/g, mean and (max value where n=2 or 95th upper confidence level where n&gt;2)

Year		Grindstone PBG	Medway PMD	Woodville PBW	Mattawamkeag PBM	S Lincoln PBL	Costigan PBC	Veazie PBV	Bangor PBO	Bucksport PBB
2000	ATS							19		
1996	EEL								37	
2000	EEL								253	
2002	EEL								98	
2013	EEL								45 (51)	
2007	SLT									27 (27)
1994	SMB					9		10		
1996	SMB	5		6						
2008	SMB			9		7 (8)				
2009	SMB			7 (10)		6 (9)		8 (12)		
2010	SMB	2 (2)	19 (22)							
1994	WHS					95		65		
1996	WHS	7		18						
2008	WHS			29 (30)		29 (49)				
2009	WHS			33 (35)		22 (26)		31 (32)		
2010	WHS	3 (5)	35 (44)							
2013	WHS						10 (11)			

## Salmon Falls River

In 2013, mean and maximum total PCB (TPCB) concentrations in white sucker (WHS) from the Penobscot River at Milford (Costigan, PBC) exceeded the ME-CDC's FTAL of 11 ng/g (Table 3.2.5). Concentrations were lowest of all years and may be showing a declining trend.

Table 3.2.5. Total PCBs in fish from the Salmon Falls River, ng/g. mean and (max value where n=2 or 95th upper confidence level where n>2)

Year		Acton SFA	Northeast P SFN	Spaulding P SFP	Berwick SFB	S. Berwick SFS
1994	SMB					91
1995		5 LMB				30 SMB
1997	SMB	5 (6)				75
2000	SMB					83 (100)
2002	SMB					110
2006	LMB			26 (49)		33 (44)
2007	LMB					47 (61)
2008	LMB					47 (59)
2009	LMB					42 (48)
1997	CHP					47 (53)
2002	WHP		23			
1994	WHS					576
1997	WHS					185
2008	WHS					115 (150)
2009	WHS					105 (113)
2013	WHS					72 (79)

## **4.0 SPECIAL STUDIES**

2.1 SOFT PLASTIC LURE FISH TOXICS BIOACCUMULATION STUDY 131

PRINCIPAL INVESTIGATORS

Dana DeGraff, DIFW

Dr. Lawrence Leblanc, UMO

## 4.0 SPECIAL STUDIES

### 4.1 SOFT PLASTIC LURES (SPL) STUDY

#### **Ingestion of Biodegradable and Non-Biodegradable Soft Plastic Lures by Salmonids Analysis of Plasticizer Concentrations in Edible Fish Tissue**

##### **Introduction**

Soft plastic lures (SPLs) are popular tackle among many sport fisheries in North America. In Maine, SPLs are used frequently in the bass fishery and are often lost to the aquatic environment when lines accidentally break, SPLs become hooked on underwater structures, or are lost when old or heavily used SPLs disengage from the line during casting. Discarded SPLs have been documented extensively in many Maine lakes (F. Brautigam and J. Seiders, pers. comm., Department of Inland Fisheries and Wildlife) and the ingestion of these SPLs by salmonids is a growing concern by anglers and fisheries managers (Danner et al. 2009). SPLs are produced by many manufacturers and are widely distributed among retailers. SPLs are highly variable in size, color, shape, scent, elasticity, and chemical constituency. Whereas the effects of ingested SPLs on brook trout (*Salvelinus fontinalis*; BKT) growth and condition factor has been documented (Danner et al. 2009), the bioaccumulation of chemicals leached from ingested SPLs has not. No data on the effects of chemicals leached from ingested SPLs were found from literature searches using search terms “fish health”, “plastic lure leachates”, “plastic lure toxicity” and “plastic lure leachates”.

Plasticizers, such as phthalates, or phthalate esters, are a low-molecular weight polymers; Di-*n*-butyl phthalate (DnBP) and di-(2-ethylhexyl)-phthalate (DEHP) are the major chemical constituents of plastics (Metcalf et al. 1973; Stalling et al. 1973; Chandra et al. 2012). Phthalates are frequently used in soft plastics and are used to render SPLs flexible (Danner et al. 2009; Johnson et al. 2009; Chandra et al. 2012). Phthalates represent 69% of plasticizer use in the United States, 92% in Western Europe, and 81% in Japan (Johnson et al. 2010). The widespread use of phthalate products globally has, within a few decades, resulted in the global contamination by this class of compounds (Bell, 1982). Phthalates may comprise 10-40% of the total weight of consumer products (Metcalf et al. 1973; Johnson et al. 2010), and likely comprise a substantial proportion of SPL weight based on the requirement of an SPL to be extremely flexible and “life like”.

Recently, fiber-reinforced and biodegradable SPLs have been developed to reduce the potential of SPL loss and spread of harmful chemicals into aquatic environments. The constituents of biodegradable SPLs are proprietary and not fully advertised and vary between manufacturers. Additionally, there are no established standards for what constitutes a biodegradable SPL and likely there is no, or very limited, information on the time period for SPLs to biodegrade. Some producers, such as Big Bite Baits, Inc., claim their product, Biobait<sup>®</sup>, is 100% biodegradable and made from all natural ingredients. However, a review of Biobait<sup>®</sup> quotes the manufacturer as saying that the SPL is a blend of plastic (i.e. 15% polyvinyl-chloride [PVC]) and natural plasticizers (i.e. 85% fish and vegetable oils; DeWitt, 2008). Even small percentages of PVC in SPLs are of concern; DEHP is the most common plasticizer in PVC formulation for many

consumer products (Metcalf et al. 1973; Carnevali et al. 2010). The harmful effects of phthalate esters on the environment and human health are well documented and summarized (Metcalf et al. 1973; Blount et al. 2000; Ghorpade et al. 2002; Duty et al. 2003; Lee et al. 2005; (Norman et al. 2005; Lithner, et al. 2009; Oehlmann et al. 2009).

Manufacturers are not currently required to list the ingredients of SPLs which makes evaluating the effects of discarded SPLs on aquatic biota difficult to determine. The negative effects of phthalates on both terrestrial and aquatic organisms have been documented in many studies however. For example, DEHP has been shown to have diverse biochemical effects in rats, rabbits and pigs, such as inhibition of cholesterologenesis in liver, testes, and adrenal gland, decreased plasma cholesterol levels, and increased fatty acid oxidation in liver mitochondria (Bell, 1982). In the aquatic environment, DEHP can bioaccumulate in a variety of plants and animals (Oehlmann et al. 2009). DEHP degrades very slowly in algae, *Daphnia* spp., mosquito larvae, snails, and clams; it closely resembles Dichloro-Diphenyl-Trichloroethane (DDT) in rate of uptake and storage in the lipids of plants and animals and is concentrated through food chains (Metcalf et al. 1973). Channel catfish (*Ictalurus punctatus*) exposed to 1 µg/l of DEHP for 24 hr resulted in tissue residues of 2.6 µg/g (Stalling et al. 1973). Exposure of early life-stages of Atlantic salmon (*Salmo salar*) to DEHP has been shown to interfere with gonad differentiation and cause intersex (ovo-testis) individuals (Norman et al. 2005). In recent studies, plastic leachates have caused acute toxic effects for *Daphnia magna*; of 15 different plastic types tested, PVC was one of two plastics that displayed toxicity in *D. magna* (Lithner et al. 2009). Environmental relevant doses have also been shown to affect vitellogenesis in zebrafish (*Danio rerio*) in the laboratory (Carnevali et al. 2010).

Phthalates are widespread in aquatic environments worldwide and fish are exposed to phthalates via water, food, and/or sediments, depending on their ecological niche (Oehlmann et al. 2009). In wild fish in the Netherlands, median [DEHP] ranged from 1.7 µg kg<sup>-1</sup> to 141 µg kg<sup>-1</sup> (wet wt.); however biotransformation of DEHP in fish appeared to be relatively fast (Peijnenburg and Struijs, 2006). Some biodegradable SPL manufacturers may have eliminated, or greatly reduced, phthalate use in their products; however some proportion of plastics like PVC, or more importantly phthalate esters found in plastics, may still be used in the production of SPLs which could continue to leach phthalates into the aquatic environment. Throughout the United States, fish consumption advisories are listed for various freshwater and marine fish species that contain chemicals that could cause human health risks, such as mercury, polychlorinated biphenyls (PCBs), and dioxins (USEPA 2012). Based on the chemical constituency of SPLs, the ubiquity of SPLs as discarded fishing tackle in Maine lakes and ponds, and the well documented environmental and human health impacts from phthalate esters, the purpose of this study is document the bioconcentration of phthalates into edible fish tissue resulting from ingestion of SPLs by hatchery and wild salmonids.

## Methodology

The study was divided into three tasks which may require development of suitable methods. The study will take until spring 2015 to complete.

### Task 1 Plasticizer Identification Study

Task one was to identify the most common plasticizers in biodegradable (e.g., Gulp® brand) and non-biodegradable (e.g., Z-Man® brand) SPLs. This work will be completed in spring 2014.

### Task 2 Bioavailability Study

Task two is to determine the potential bioavailability and uptake of toxic plasticizers in each type of SPL into fish muscle tissue. SPLs will be provided by DIFW. This work will be completed in late spring or early summer 2014.

### Task 3 Fish Tissue Analysis

Task three will be to determine fish muscle tissue concentrations of the most significant plasticizers identified in Tasks 1 and/or 2 after collaboration with DEP and DIFW. DIFW will conduct an SPL fish exposure study beginning in summer 2014 to be completed by fall 2014 with subsequent chemical analysis to be completed by spring 2015. DIFW will force feed SPLs to hatchery fish and have the muscle tissue sampled for toxic plasticizers at various sampling time intervals as shown below (Table 1). There will be 15 fish from each treatment and sampling interval to be sent to the analytical chemistry lab at the end of each sampling interval, except for the T=0 fish when there will be only fish from Treatment 3 to be collected at T=2 d. The lab will filet, homogenize, subsample, and combine the 15 fish from each time/treatment into 3 composites of 5 fish each, for a total of 39 tissue samples, prior to chemical analysis.

**Table 1. Number of fish sampled for each treatment group at each sampling interval.**

Sampling Interval (d)	Treatment 1 Biodegradable SPL (# Fish Sampled)	Treatment 2 Non-Biodegrade. SPL (# Fish Sampled)	Treatment 3 Control Group (# Fish Sampled)
<b>t = 0</b>	0	0	15
<b>7</b>	15	15	15
<b>30</b>	15	15	15
<b>60</b>	15	15	15
<b>120</b>	15	15	15

The study will be similar to that described by Danner et al. 2009. Study fish will be taken from lots which will be tested for pathogens of regulatory concern in Maine prior to commencing the study according to fish health inspection procedures. All study fish will be of the same strain, spawning group, and age. Study and control groups of spring yearling (SY) lake trout (LKT) will be kept in identical 4-foot diameter flow through tanks at the Maine Departments of Inland Fisheries and Wildlife (MDIFW) Governor Hill Fish Hatchery. Hatchery staff will provide fish feeding regimens and monitor photoperiod and water quality parameters including dissolved oxygen and water temperature.

Two study groups of 90 SY LKT each will be obtained from the MDIFW Governor Hill Fish Hatchery in June 2013. Fish will be anesthetized (using either MS-222 or Aqui-S 20e) and force-fed popular biodegradable (e.g., Gulp<sup>®</sup> brand; Group 1) and non-biodegradable (e.g., Z-Man<sup>®</sup> brand; Group 2) SPLs. A control group of 95 SY LKT will be kept in identical conditions as the study groups (Table 1). All study and control group fish will be fed a regular feed diet throughout the study period according to established hatchery regimens.

Fish samples will be obtained by euthanizing fish in an overdose of MS-222 or Aqui-S 20e. Fish will be processed and prepared at the MDIFW Fish Health Diagnostic Laboratory according to Maine Department of Environmental Protection, Surface Water Ambient Toxics (MDEP SWAT) protocols. Each fish will be measured for TL (mm) and wet weight (g) and placed on wet ice for immediate necropsy following methods outlined by Danner et al. 2009. One composite sample of 5 control fish will be collected at  $t = 0$  in order to screen for background [phthalate] in fish tissue. Following, triplicate composites of 5 fish each (15 fish total) will be collected from each of the study and control groups at specific time intervals (i.e. 7, 30, 60 and 120 d). All composite fish tissue samples will be analyzed for phthalates.

### Data Analysis

Data will be presented in graphical or tabular formats. Fish tissue [phthalate] will be directly compared between all laboratory study and control fish. The effects SPL ingestion on tissue [phthalate] between study groups for each sampling interval will be analyzed by one-way ANOVA. Additionally, effects of SPL ingestion on fish weight and length will be analyzed by one-way ANOVA. When effects are significant, a Tukey's *a posteriori* multiple range test will be used for comparisons. Correlations between SPL exposure time and [phthalate] in fish tissue will be made using the Pearson-type simple correlation model. [Phthalate] in fish tissue will be compared to established detection limits and exposure limits for human health risk. The results of study fish necropsies will be summarized and compared between study and control groups. General comparisons between field and laboratory [phthalate] data will be made when possible and/or appropriate.

### Results

Preliminary results of the Plasticizer Identification Study and Bioaccumulation Study are as reported below by Dr. Lawrence Leblanc, University of Maine, primarily for task 1.

Phthalate mixes 8060 and 8061 were purchased and individual phthalates were identified. Quantitative solutions were made up that included the following analytes:

- Dimethyl Phthalate (DMP)
- Diethyl Phthalate (DEP)
- Diisobutyl Phthalate
- Dibutyl Phthalate (DBP)
- Bis(2methoxyethyl)Phthalate
- Bis(4-methyl-2-pentyl)Phthalate



Bis(2-ethoxyethyl)Phthalate  
Diamyl Phthalate  
Di-n-hexyl Phthalate  
Benzo Butyl Phthalate (BBP)  
Hexyl-2ethylhexyl Phthalate  
bis(2-n-butoxyethyl) Phthalate  
Dicyclohexyl Phthalate  
Diethylhexyl Phthalate (DEHP)  
Dioctyl Phthalate (DOP)  
Dinonyl Phthalate

Using these standards, a cleanup column was devised using 2 grams of 100% activated silica gel (200-300 mesh) for the purpose of removing interfering matrix from extracts of the soft plastic lures. It was determined that an extract could be placed on the column, washed with 10 mL of 50:50 hexane:methylene chloride, prior to eluting the phthalate fraction, which came off the silica gel with 10 mL of 80:20 methylene chloride:ethyl acetate mixture.

Next three types of soft plastic lures (SPLs), purchased from sporting goods stores were extracted in order to identify major marker compounds for the fish feeding study. The brand names of the SPLs extracted to date are of the Zoom, Z-man and Yammamoto, none of which are advertised as being biodegradable. A fourth brand, Gulp is biodegradable and has yet to be analyzed. Samples were extracted using accelerated solvent extraction (Dionex ASE 200 system). Acetonitrile was used as the extraction solvent, as it has been shown to be a good solvent for extracting hydrophobic analytes, while leaving behind substantial amounts of background matrix. The samples were then re-extracted with solvents typically used for base-neutral extractions (a 50:50 methylene chloride:acetone mixture) to verify no analytes of interest were left behind. In addition 2 laboratory sample blanks were also processed.

Acetonitrile extracts were placed in a separatory funnel containing 150 mLs of MilliQ water, and then back-extracted with 3 x 75 mLs of hexane. Hexane extracts were combined, reduced in volume, and placed on the silica gel cleanup column described above. The water was then acidified and re-extracted with hexane.

Instrumental analysis for these preliminary trials was performed using a gas chromatography/mass spectroscopy system (Agilent 6890/5973 GC/MS system) using electron ionization. Using the standard mixes, a selected ion monitoring (SIM) method was developed to eliminate as much of the background matrix as possible, while retaining critical spectral information.

Results showed no phthalate compounds present in any of the sample lures. Instrumental traces from the mass spectrometry analyses revealed large unresolved “humps” with several partly resolved peaks. Pattern identification software identified many of these partly resolved peaks as dicarboxylic acid esters, which are phthalate esters. Similar results have been found in other studies examining residual phthalates in environmental samples (eg., Lin et al., 2003). These authors concluded these patterns reflect higher molecular weight phthalate isomeric mixtures,

and reported a method of quantification, based upon the carbon number of the straight or branched chain portion of the phthalate molecule, using LC/MS/MS (liquid chromatography/tandem mass spectroscopy). Similar results were found in the methylene chloride:acetone extracts. Based upon the intensity of the signal, the Zoom or the Gary Yamamoto brands were suggested as being the best for the feeding study, along with the Gulp (biodegradable) brand. Analysis of the Gulp lures is pending.

Analytical standards of higher molecular weight isomeric phthalate mixtures are commercially available. A number of these standards have been ordered and work is commencing on developing an LC/MS/MS instrumental procedure. If patterns match between isomeric standards and samples of fish lures, phthalates will be quantified as mentioned above, by separating isomers based upon the carbon number of the hydrocarbon chain moiety – i.e., the sum of C9 phthalates, C10 phthalates, etc. Laboratory blanks thus far showed small (1-10 ppb) concentrations of diethyl phthalate (DEP) and slightly higher (10-50 ppb) concentrations of diethylhexyl phthalate and dioctyl phthalate.

Preliminary desorption studies into distilled laboratory water are currently underway with Zoom, Yamamoto and Gulp brand lures. Samples have been placed in a covered 1L beaker, along with 800 mL of water and agitated gently for 3 days (72 hours). At that point the water is removed and extracted with 3 x 75 mL of hexane. Fresh water is added to the beaker and the worms are allowed to desorb for another 72 hours. A quick look at the Zoom sample did show measurable quantities of diethyl, dibutyl, diethylhexyl phthalates compounds (30, 160 and 220 ng/g, respectively), along with the same unresolved “hump” seen in the worm extracts. However comparisons to matched blanks have not yet been made.

Thus far I have not seen evidence of bisphenol A in the SPL extracts using GC/MS. However there is better chromatography and higher sensitivity using LC/MS/MS, which has yet to be performed.

#### Conclusions:

Based upon analyses of SPL extracts it appears that phthalates, if present, are in the form of isomeric mixtures of higher molecular weight than the phthalate compounds commonly analyzed in environmental samples and present on the EPA priority pollutant list. Next steps are adapting a published method for quantifying the higher molecular weight isomeric mixtures by LC/MS, and re-examining the SPL extracts as well as ongoing desorption trials of SPLs into water and tenax resin.

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