Report to the Joint Standing Committee on Environment and Natural Resources 126<sup>th</sup> Legislature, First Session

# Surface Water Ambient Toxics Monitoring Program 2012

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#### Introduction

This 2012 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 <a href="http://www.maine.gov/dep/water/monitoring/toxics/swat/index.htm">http://www.maine.gov/dep/water/monitoring/toxics/swat/index.htm</a>

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#### **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 fiveyear 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 2012 annual work plan recommended by the SWAT TAG in a meeting July 3, 2012. The 2012 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 2012, blue mussel tissue from Scarborough River, Scarborough, and Spring Point, South Portland, was analyzed for contaminants including metals, mercury, Polycyclic Aromatic Hydrocarbons (PAHs), Polychlorinated Biphenyls (PCBs), and organochlorinated pesticides. In 2012, tissue from four additional blue mussel sites in the Sheepscot estuary was analyzed for metals and mercury only.

• In 2012, softshell clam tissue from Presumpscot River, Falmouth/Portland, and Mare Brook (Harpswell Cove), Brunswick, was tested and reported with historical data from eight additional softshell clam sites sampled in 2004-05 and 2010-11. Clam tissue was analyzed for contaminants including metals, mercury, PAHs, PCBs, and organochlorinated pesticides.

#### 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 any site and 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 and clam tissue were below the Maine Center for Disease Control's (MCDC) cancer fish tissue action level (FTAL), indicating shellfish remained safe for human consumption with regard to PCBs.
- Organochlorinated pesticide concentrations in mussel and clam tissue were low at Maine sites compared to national (NS&T Musselwatch) data, and pesticide levels were safely below MCDC FTAL values, indicating shellfish remained safe for human consumption with regard to pesticides.

#### 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 2012, Spring Point, South Portland, and Turnip Island, Georgetown, resulting in these sites receiving an "elevated" designation. Lead concentrations at these two sites also exceeded the MCDC"s FTAL for lead in finfish. Lead in clam tissue in 2012 at Mare Brook, Brunswick, and Presumpscot River, Falmouth/Portland, exceeded the MCDC FTAL for lead in finfish. These four sites 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 five of six sites tested in 2012, which resulted in assignment of an "elevated" classification. Mercury levels in 2012 mussel and clam tissue were below the MCDC methylmercury developmental FTAL for finfish, indicating shellfish remained safe for human consumption with regard to mercury.

- Mercury in Sheepscot estuary blue mussel tissue is elevated over Maine coast background levels by a factor of 2.5 on average.
- Cadmium in mussel tissue exceeded the national (NS&T Musselwatch) 85<sup>th</sup> percentile concentration at one site, Turnip Island, Georgetown, in 2012, which resulted in assignment of an "elevated" classification. Cadmium levels in 2012 mussel and clam tissue, including Turnip Island, were below the MCDC FTAL for cadmium in finfish, indicating shellfish remained safe for human consumption with regard to cadmium.

#### 2. LAKES

#### Encouraging Results:

• Although limited data from previous studies indicated that mercury concentrations in fish from thirteen coastal lakes might be higher than historical levels in fish from inland lakes, concentrations were, in fact, similar to those from inland lakes, except for Round Pond, Hodgdon Pond and Seal Cove Pond in Acadia National Park, where concentrations were much higher than in other coastal and inland lakes, as was the case in the mid-1990s for these two lakes.

#### Contaminants and Areas to Watch:

• Comparison of mercury concentrations in twelve inland lakes with historical data from the same lakes from the 1990s showed that, in 2012, concentrations increased in one lake, decreased in five lakes and remained similar in six lakes; this is unlike previous years when the number of lakes with increased and decreased concentrations was generally equal. Aggregated data from forty-six lakes collected from 2010 to 2012 show no clear trends, however. 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:

- Forty-four stations were assessed for the condition of the benthic macroinvertebrate community. Thirty-five of these stations attained the aquatic life standards of their assigned class.
- Evaluation of sediments from impoundments below pulp and paper mills on the Androscoggin River showed little evidence of toxicity from heavy metals.

- 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 brook trout from Gilead on the Androscoggin River are below the FTAL and lower than previous concentrations in rainbow trout from the same station. Concentrations in smallmouth bass at Rumford Point and in white sucker at Rumford Point above Rumford, Riley and Livermore above and below Jay still exceed the FTAL however, although concentrations are lower than previous years at Rumford Point and Livermore. Dioxin concentrations in filet of American shad from Waterville on the Kennebec River exceeded the FTAL but concentrations in roe did not. Dioxin concentrations in white sucker from Kennebec River at Sidney are below the FTAL similar to those of 2011. Dioxin concentrations in Sebasticook Lake still exceed the FTAL and are higher than in 2011.
- Coplanar (dioxin-like) PCB concentrations in fish were lower in 2012 than in the 1990's at all stations sampled. Although coplanar PCB concentrations add to the exceedance of the dioxin FTAL, concentrations 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 most stations sampled in 2012. Coplanar PCBs added to dioxins resulted in an exceedance of the FTAL for the American shad roe at Waterville and white sucker from the Kennebec River at Sidney, and white sucker from Sebasticook Lake. Coplanar PCB concentrations were detected and increased the exceedance of the FTAL in fish at all other stations except the brook trout at Gilead. The sum of coplanar PCBs and dioxins exceeded a Statewide Advisory Dioxin Equivalent Threshold, which would require an additional advisory beyond the Statewide Fish Consumption Advisory due to mercury, for white sucker from Riley on the Androscoggin River and from Sebasticook Lake.

#### Contaminants and Areas to Watch:

- Total PCB concentrations were generally a little lower on the Androscoggin and higher on the Kennebec compared to previous years. Total PCBs exceeded the FTAL in all fish from the same stations sampled for dioxin except for brook trout from Gilead on the Androscoggin River which were below the FTAL and lower than previous levels in brown trout and rainbow trout from this stations. Total PCB concentrations in American Shad filet was well above the FTAL while those in shad roe only slightly exceeded the FTAL.
- Contaminant levels were measured in resident fish and freshwater mussel species at above and below the two dams scheduled to be removed as part of the Penobscot River Restoration project. The results provide a baseline for contaminant studies, following removal of the dams, to document any changes in contaminant levels as a result of movement of any contaminated sediment that has accumulated in the impoundments over the years.

## **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 up 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 here in Maine.

#### 1.1.1 Blue Mussels

Blue mussels 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.

Blue mussels have been a long term focus of the marine SWAT sampling efforts over the years and were included again in the SWAT program in 2012. This report presents and summarizes contaminant data from the collection and analysis of blue mussel (*Mytilus edulis*) tissue collected in 2012 from six sites along the Maine coast. All mussel tissue samples were analyzed for heavy metals (including mercury), and a subset of two sites were analyzed for polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), and organochlorinated pesticides. In order to provide comparability of results from these 2012 samples, blue mussel contaminant levels from the SWAT program are compared to blue mussel contaminant levels in other programs including the Gulfwatch program (Gulf of Maine Council on the Marine Environment) and the National Status & Trends Mussel Watch Program (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 2012 from two sites on the Maine coast. Also presented are softshell clam contaminant data from eight additional sites sampled in 2004-05 and in 2010-11 by the SWAT program. Softshell clam tissue samples were analyzed for metals, mercury, PAHs, PCBs, and organochlorinated pesticides. In order to provide comparability of results from the 2010-11 and 2004-05 samples, softshell clam contaminant concentrations from SWAT sampling are compared to contaminant concentrations in 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 have improving bacterial trends 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 report 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 a long time), 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, spatial needs. This gives a more complete picture of how contaminants vary across the Maine coastline, and provides 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 sites where there is an interest in obtaining data to assess contaminants 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 replicate 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 subsite 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 for toxics as part of the SWAT program since 1986, with over 90 distinct locations sampled in the past 27 years. Sampling stations are selected to meet one or more of three goals: 1) Provide spatial coverage of the Maine coast; 2) provide data to determine temporal patterns or trend; and 3) provide more focused results to assess problems documented by earlier sampling and analyses. Early sampling efforts sometimes took a screening approach, included only metals analyses, or sometimes included only one replicate, which provides no information to assess variability of contaminants within site but does reduce cost.

Blue mussels were collected from six sites in 2012. Two of the six mussel sites had been sampled previously as part of the SWAT program. Names and locations of blue mussel collection sites for 2012 are presented in Table 1.2.1.1. This table presents sites by name and includes municipality, latitude and longitude, and the site selection type: spatial, temporal, or follow-up. 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: 2012 SWAT Blue Mussel Sites						
		<u>Station</u>	<u>West</u>	<u>North</u>	<b>Date</b>	Site
<u>Site Name</u>	<b>Municipality</b>	Code	Longitude	Latitude	<b>Sampled</b>	Type <sup>1</sup>
Scarborough River	Scarborough	SCSRRR	-70.34413	43.55424	9/12/2012	Т
Spring Point	South Portland	CBSPSP	-70.22765	43.6506	9/25/2012	Т
Beal Island	Georgetown	MCSHBL	-69.73498	43.88208	10/24/2012	F, S
Turnip Island	Georgetown	MCSHTU	-69.70726	43.83356	10/25/2012	F, S
Pratts Island	Southport	MCSHPR	-69.68412	43.80978	10/25/2012	F, S
Boston Island	Southport	MCSHBO	-69.68092	43.8519	10/22/2012	F, S
S = Spatial, T = Temporal, F = Follow-up						

Sampling occurred from mid-September through October and sampling dates are included for specific sites in Table 1.2.1.1. In order to characterize the contaminants present in a general area at the sampling station, mussels were collected from four distinct areas (replicates) along the shoreline at each site 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 and placed in separate containers. For organics analysis, 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, width (in 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  $-5^{\circ}$  C for up to 1 to 2 months until analysis.

Mussel tissues tested for PAHs, PCBs, and organochlorinated pesticides (four sites in 2012) 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.



#### **1.2.2 Softshell Clams**

Softshell clams were collected at two sites in 2012. Softshell clams were sampled at the request of the Maine DMR to assess contaminants present in tissue at two locations where there is interest in opening clam flats closed due to high bacterial levels.

In addition to the two softshell clam sites sampled in 2012, this report includes data from eight softshell clam sites sampled in 2010-11 and 2004-05. These data are included to provide a broader context for softshell clam contaminant concentrations across the state. The data for the two sites sampled in 2012, Presumpscot River, Falmouth/Portland and Mare Brook, Brunswick, and the eight sites sampled previously are presented in Table 1.2.2.1, and include municipality and latitude and longitude. The location of the softshell clam sampling stations is presented in Figure 1.2.1.1.

TABLE 1.2.2.1: SWAT Softshell Clam Sites: 2004-05, 2010-12						
		<b>Station</b>	West	<u>North</u>	Date	<u>Site</u>
<u>Site Name</u>	<u>Municipality</u>	<u>Code</u>	Longitud	Latitude	<u>Sampled</u>	<b>Type</b> <sup>1</sup>
Mast Cove	Eliot	PQMCMC	-70.8048	43.1210	11/9/2004	S
Presumpscot Rive	Falmouth/Portlan	CBPRMT	-70.2460	43.6981	10/9/2012	S
Navy Pier	Harpswell	CBHWNF	-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	PBFPFP	-68.8150	44.4717	11/10/200	S
Fort Point Cove	Stockton Springs	PBFPFP	-68.8372	44.4832	11/3/2011	F
Morse Cove	Penobscot/Castin	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	11/29/200	S
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 below.

Sampling typically occurred from mid-October through mid-November and the specific sampling dates are included in Table 1.2.2.1. 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 were 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, width (in 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, 10-20 clams were composited into a replicate to produce the requisite 100 grams of tissue required for the scheduled analyses.

Tissue composite samples for metals analyses included ten clams per composite sample or replicate, and tissue composite samples for organics analyses included 10-20 clams per composite sample or 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° C for up to one to two months until analyses could be completed. Softshell clam tissues tested for PAHs, PCBs, and organochlorinated pesticides in 2010-12 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 2012 were analyzed by Battelle Marine Sciences Laboratory, Sequim, WA. The samples were analyzed for 11 metals: Silver (Ag), aluminum (Al), arsenic (Ar), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), lead (Pb), mercury (Hg), nickel (Ni), and zinc (Zn). Results were compared to national (NOAA National Status & Trends Mussel Watch (NS&T), see Kimbrough et al. 2008) and Gulf of Maine (Gulfwatch, see 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 in all six sample locations visited in 2012 (Figure 1.3.1.1.1). Silver detected in mussels ranged from a low mean concentration of 0.047 ug/g dry wt. at Spring Point, South Portland, to a high mean concentration of 0.627 ug/g dry wt. at Turnip Island, Georgetown. Silver mean concentrations in 2012 SWAT mussels were also compared to the

Gulfwatch median and  $85^{\text{th}}$  percentile concentrations. The mean concentration at all six sites exceeded the Gulfwatch median (0.037 ug/g dry wt.). Mean concentrations exceeded the Gulfwatch  $85^{\text{th}}$  percentile (0.073 ug/g dry wt.) at two sites (Figure 1.3.1.1.1).

Figure 1.3.1.1.1.2 compares the silver concentrations in 2012 SWAT blue mussel tissue to the NS&T median and 85<sup>th</sup> percentile. The mean silver concentration at Turnip Island, Georgetown, exceeded the NS&T median. No tissue silver concentrations exceeded the NS&T 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 Nano silver, 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 SWAT tissue silver concentration from Turnip Island, Georgetown, in 2012 exceeded these Massachusetts tissue silver concentrations, but was still far below 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 2012 SWAT blue mussel tissue mean silver concentration, when expressed on a wet weight basis, is 0.102 ug/g wet wt. at Turnip Island, Georgetown. This concentration is two orders of magnitude below the 11 ug/g wet wt. FTAL

#### 1.3.1.1.2 Arsenic (As)

Arsenic was detected in all six sample locations visited in 2012 (Figure 1.3.1.1.2.1). Arsenic levels detected in mussels ranged from a low mean concentration of 12.93 ug/g dry wt. at Scarborough River, Scarborough, to a high mean concentration of 20.33 ug/g dry wt. at Turnip Island, Georgetown. 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 six 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).

For non-commercially caught finfish, MCDC reports a cancer FTAL of 0.014 ppm and a noncancer 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, 2012 SWAT blue mussel inorganic arsenic concentrations are estimated to range from 0.19 ug/g wet wt. to 0.33 ug/g wet wt. All six sites exceeded the MCDC cancer FTAL of 0.014 ug/g wet wt. (ppm).

Comparing recent data from all 57 mussel sites sampled from 2007-12, 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 2007-12 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 six sites sampled in 2012 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 51 mussel stations sampled from 2007-11 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 indicates 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 in all six sample locations visited in 2012 (Figure 1.3.1.1.3.1). Cadmium levels detected in mussels ranged from a low mean concentration of 1.70 ug/g dry wt. at Spring Point, South Portland, to a high mean concentration of 9.91 ug/g dry wt. at Turnip Island, Georgetown. The cadmium concentration at Spring Point fell below the 2008 Gulfwatch median, with the concentrations at Scarborough River, and Pratts Island and Boston Island (Southport) exceeding the Gulfwatch median. The remaining two sites exceeded the Gulfwatch median and 85<sup>th</sup> percentile (Figure 1.3.1.1.3.1).

Cadmium concentrations at Spring Point, South Portland, and Boston Island, Southport, fell below the NS&T median, with the concentrations at three sites falling between the NS&T median and 85<sup>th</sup> percentile. One site, Turnip Island, Georgetown, had a cadmium concentration which exceeded the NS&T national 85<sup>th</sup> percentile, which was considered elevated (Figure 1.3.1.1.3.1) (Kimbrough et al. 2008).



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 naturally 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 noncommercially 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 2012 SWAT site, Turnip Island, Georgetown, had a mean cadmium concentration of 1.60 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 six sites sampled in 2012 (Figure 1.3.1.1.4.1). Chromium levels detected in mussel tissue ranged from a low mean concentration of 0.76 ug/g dry wt. at Turnip Island, Georgetown, to a high mean concentration of 2.93 ug/g dry wt. at Beal Island, Georgetown. Beal Island, Georgetown, exceeded both the Gulfwatch median and 85<sup>th</sup> percentile, while the concentrations at the remaining five mussel sites did not exceed the Gulfwatch median (Figure 1.3.1.1.4.1).

Figure 1.3.1.1.4.1 also depicts 2012 SWAT mussel chromium concentrations compared to the NS&T Mussel Watch median and 85<sup>th</sup> percentile concentrations. Three sites, Spring Point, South Portland, Beal Island, Georgetown, and Boston Island, Southport, exceeded the NS&T median, and Beal Island approached, but did not exceed, the NS&T national 85<sup>th</sup> percentile.

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 six SWAT mussel sites visited in 2012 (Figure 1.3.1.1.5.1). Copper levels detected in mussels ranged from a low mean concentration of 5.95 ug/g dry wt. at Scarborough River, Scarborough, to a high mean concentration of 40.9 ug/g dry wt. at Turnip Island, Georgetown. Copper concentrations at Spring Point, South Portland, and Beal Island and Turnip Island, Georgetown, exceeded the Gulfwatch median, while only Turnip Island exceeded the Gulfwatch 85<sup>th</sup> percentile (LeBlanc et al. 2009). SWAT copper concentrations at five sites sampled in 2012 fell below the NS&T median, with Turnip Island exceeding the median but falling well below the NS&T 85<sup>th</sup> percentile as shown in Figure 1.3.1.1.5.2 (Kimbrough et al. 2008).

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 1980's; 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 in all six SWAT blue mussel sites sampled in 2012 (Figure 1.3.1.1.6.1). Iron concentrations detected in mussels ranged from a low mean concentration of 126 ug/g dry wt. at Pratts Island, Southport, to a high mean concentration of 1055 ug/g dry wt. at Beal Island, Georgetown. Iron concentrations at three sites were below the Gulfwatch median, while concentrations at two sites exceeded the Gulfwatch median. One site, Beal Island, 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. Iron concentrations at two sites were below the NS&T national median, and three sites had iron concentrations between the NS&T median and 85<sup>th</sup> percentile. Only the iron concentration in mussel tissue from Beal Island, Georgetown, exceeded the NS&T national 85<sup>th</sup> percentile.







Aluminum concentrations detected in mussels ranged from a low mean concentration of 65 ug/g dry wt. at Pratts Island, Southport, to a high mean concentration of 477 ug/g dry wt. at Beal Island, Georgetown (Figure 1.3.1.1.6.2). Aluminum concentrations at four sites were below the Gulfwatch median concentration, while the aluminum concentration at Spring Point, South Portland, was between the Gulfwatch median and 85<sup>th</sup> percentile concentrations. One site, Beal Island, Georgetown, exceeded the Gulfwatch 85<sup>th</sup> percentile (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 mean concentrations at three sites were between the NS&T median and 85<sup>th</sup> percentile concentrations. One site, Beal Island, Georgetown, had a mean aluminum concentration that exceeded the NS&T national 85<sup>th</sup> percentile.

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. 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 six SWAT blue mussel sites visited in 2012 (Figure 1.3.1.1.7.1). Nickel levels detected in mussels ranged from a low mean concentration of 0.91 ug/g dry wt. at Pratts Island, Southport, to a high mean concentration of 7.06 ug/g dry wt. at Turnip Island, Georgetown. Three sites had nickel concentrations below the Gulfwatch median, while one site fell between the Gulfwatch median and 85<sup>th</sup> percentile. The remaining two sites, Beal Island, Georgetown, and Turnip Island, Georgetown, had nickel concentrations exceeding the Gulfwatch 85<sup>th</sup> percentile (Figure 1.3.1.1.7.1).

Figure 1.3.1.1.7.1 also compares 2012 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 distributed below the national median at five sites, with only Turnip Island, Georgetown, exceeding the national median. No 2012 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 2012 of 1.14 ug/g wet wt. (ppm) at Turnip Island, Georgetown, is an order of magnitude below the more conservative MCDC action level. MCDC does not report a cancer action level for nickel.

#### 1.3.1.1.8 Lead (Pb)

Lead was detected in all six SWAT blue mussel sites visited in 2012 (Figure 1.3.1.1.8.1). Lead levels detected in mussels ranged from a low mean concentration of 1.09 ug/g dry wt. at Pratts Island, Southport, to a high mean concentration of 8.53 ug/g dry wt. at Turnip Island, Georgetown. Two sites had concentrations less than the Gulfwatch median, with Beal Island, Georgetown, and Boston Island, Southport, lead concentrations at the median and just above the median, respectively. Spring Point, South Portland lead concentration just exceeded the Gulfwatch 85<sup>th</sup> percentile while the concentration at Turnip Island, Georgetown was twice as high.

Figure 1.3.1.1.8.1 also compares 2012 SWAT blue mussel lead tissue concentrations to NS&T median and 85<sup>th</sup> percentiles to place Maine data into a national context. All six SWAT sites exceeded the NS&T median. Two of six SWAT sites, Spring Point, South Portland, and Turnip Island, Georgetown, 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.

Lead tissue concentrations from prior samples at Temporal Sites from Spring Point, South Portland, and Scarborough River, Scarborough, were compared to 2012 concentrations (Figure 1.3.1.1.8.2). Lead concentrations at Spring Point appear to fluctuate somewhat from year to year, which is probably due to patchiness of contamination within the site. With only two years of data at Scarborough River, lead concentrations appear to be relatively stable. 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).





Spring Point, South Portland, is located just south of the entrance to Portland Harbor and is located near an oil shipping pier. The upland area is densely settled residential development. Several sewage treatment plants discharge upstream in the harbor. Scarborough River does not have the heavy industry present in Portland harbor, but is influenced by residential and commercial development in the watershed, which includes large areas of impervious cover, golf courses, and segments of I-95 and Rte. 1. Due to the proximity of the Scarborough River to coastal development and because of its unique and important ecological functions, it has been chosen to be sampled more frequently to enable assessment of trends in contaminants. As a result, 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 sub-site 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 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 concentrations in the 2012 Maine SWAT mussel data, 0.6 ppm (ug/g) wet wt. at Spring Point, South Portland, and 1.4 ppm (ug/g) at Turnip Island, Georgetown, exceed the MCDC lead FTAL. The remaining four sites sampled in 2012 did not exceed the MCDC FTAL for lead.

Review of the 2007-12 SWAT blue mussel sampling data from 59 sites indicates that mean lead concentrations at seven 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.
Turnip Island, Georgetown, 2012	1.4 ppm wet wt.

Crockett Point, Rockland, 2007 Crockett Point, Rockland, 2010 Crockett Point, Rockland, 2011	<ul><li>1.1 ppm wet wt.</li><li>1.3 ppm wet wt.</li><li>1.1 ppm wet wt.</li></ul>
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 six blue mussel sample locations visited in 2012 (Figure 1.3.1.1.9.1). Mercury levels detected in mussels ranged from a low mean concentration of 0.12  $\mu$ g/g dry wt. at Scarborough River to a high mean concentration of 0.59  $\mu$ g/g dry wt. at Turnip Island, Georgetown. Only mercury concentrations at Scarborough River and Pratts Island, Southport, did not exceed the 2008 Gulfwatch median, while Spring Point, South Portland, fell between the Gulfwatch median and 85<sup>th</sup> percentile concentrations. The remaining three sites all exceeded the Gulfwatch 85<sup>th</sup> percentile and were all located in the Sheepscot River estuary.

Figure 1.3.1.1.9.1 also compares 2012 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, five of six SWAT sites tested in 2012 would be considered elevated for mercury despite their more typical scores when compared to other northeast US samples from the Gulf of Maine.

Four blue mussel sites sampled in 2012 were within the Sheepscot River estuary, a follow-up to eight stations sampled in 2009 and six in 2011. The more intensive sampling in the Sheepscot was initiated subsequent to the detection of higher mercury concentrations in blue mussel tissue in the Wiscasset area in 2008. Two of the follow-up stations sampled in 2012 also showed higher mercury concentrations, similar to those detected in 2009 and 2011, but at additional locations within the Sheepscot estuary. Figure 1.3.1.1.9.2 shows the 2009, 2011, and 2012 Sheepscot estuary mercury concentrations in blue mussel tissue in contrast to mercury concentrations from blue mussel tissue from a wide range of sites along the Maine coast sampled from 2007-12. Tissue mercury concentrations in the Sheepscot range from 0.15 to 0.59 ug/g dry wt.




# Figure 1.3.1.1.9.2: Mercury in SWAT Blue Mussel Tissue Across the Maine Coast and Sheepscot River Estuary

61 Stations 2007-12 - West to East; Large Diamonds= 2012 Sheepscot Estuary Sample Points Triangles = 2011 Sheepscot Estuary Sample Points Circles = 2009 Sheepscot Estuary Sample Points

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with the lowest tissue concentration found at the most southeasterly site, Pratts Island, Southport (2012). Tissue concentrations with the exception of Whittum Island (2009), Boston Island and Pratts Island (both 2012), fell between 0.4 to 0.6 ug/g dry wt. Figure 1.3.1.1.9.2 shows that most Maine blue mussel tissue mercury concentrations fall below 0.3 ug/g dry wt., with a few sites having concentrations above 0.3, including the Piscataqua River and Penobscot River (Sandy Point), sites with documented industrial histories.

Figure 1.3.1.1.9.3 shows that the lowest concentration found in the Sheepscot, 0.15 ug/g dry wt. at Pratts Island, Southport, is the most southerly site shown on the map. In 2009, a control site outside the Sheepscot, just to the east in the Damariscotta River estuary at Goose Ledge, revealed mean mussel tissue concentration of 0.14 ug/g dry wt. (Figure 1.3.1.1.9.3). The location of the Mason Station, an historic coal (and subsequently oil) fired electrical power generation station, is also shown.

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 ug/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 blue mussel total tissue mercury concentration measured in Maine in 2012 was 0.095  $\mu$ g/g wet wt. (ppm) at Turnip Island, Southport, in the Sheepscot Estuary. 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.



#### 1.3.1.1.10 Zinc (Zn)

Zinc was detected in all six sample locations visited in 2012 (Figure 1.3.1.1.10.1). Zinc levels detected in mussels ranged from a low mean concentration of 75.5 ug/g dry wt. at Pratts Island, Southport, to a high mean concentration of 994.8 ug/g dry wt. at Turnip Island, Georgetown. The SWAT blue mussel tissue zinc concentrations fell around the 2008 Gulfwatch median, with only Turnip Island exceeding the Gulfwatch 85<sup>th</sup> percentile.

Figure 1.3.1.1.10.2 shows 2012 Maine SWAT blue mussel zinc concentrations were all below the NS&T Mussel Watch median except Turnip Island, Georgetown, which did not exceed the NS&T 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).

#### 1.3.1.2 Softshell Clams

Two softshell clam sites were sampled in 2012: Presumpscot River, Falmouth/Portland, and Mare Brook, Brunswick, which is the upper portion of Harpswell Cove. These two clam resources were sampled at the request of Maine DMR to determine concentrations of toxic contaminants in clam tissue. Results from the two sites sampled in 2012 are compared to previous results from samples collected from eight other clam sites. Softshell clam tissue samples collected in 2010-12 were analyzed by Battelle Marine Sciences Laboratory, Sequim, WA. Clam tissues from 2004-05 were analyzed by Pace Analytical Services, Minneapolis, MN. For purposes of this report, data was compared directly between the two labs. The samples were analyzed for 11 metals: Silver (Ag), aluminum (Al), arsenic (Ar), 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 in all ten sample locations visited historically (Figure 1.3.1.2.1.1). Silver detected 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 2.08 ug/g dry wt. at Mast Cove, Eliot. Silver mean concentrations at the two sites sampled in 2012, Presumpscot River, Falmouth/Portland, and Mare Brook, Brunswick, were within the higher and lower ends of the range of concentrations from previously sampled Maine clam sites. Silver mean concentrations in SWAT softshell clams were also compared to the Gulfwatch median concentration for four sites sampled in







#### Figure 1.3.1.2.1.1: Silver in SWAT Softshell Clams

2008 (two in Maine and two in New Hampshire). The mean concentrations at Mast Cove, Eliot (2004), and Presumpscot River, Falmouth/Portland (2012), exceeded the Gulfwatch median (1.32 ug/g dry wt.). The silver concentration in clam tissue in Fort Point Cove appeared to be slightly higher in 2011 than in 2005, which may be an artifact of intra-site spatial variability 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 Nano silver, 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 0.32 ug/g wet wt. at Mast Cove, Eliot (2004). This concentration is over an order of magnitude below the 11 ug/g wet wt. FTAL, assuming the same meal size is applied.

#### 1.3.1.2.2 Arsenic (As)

Arsenic was detected in clam tissue at both Presumpscot River, Falmouth/Portland, and Mare Brook, Brunswick (2012). Two previous sites, Fort Point Cove, Stockton Springs (2011), and Morse Cove, Castine (2010), were the only softshell clam sites where tissue was tested for arsenic. 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).

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 fell into this range, while the mean arsenic concentrations at Fort Point Cove and Presumpscot River 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, 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 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 noncancer 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 (2012) clam inorganic arsenic mean concentration is estimated to be 0.23 ug/g wet wt., and the Mare Brook (2012) clam inorganic arsenic mean concentration is estimated to be 1.56 ug/g wet wt. Both clam sites sampled in 2012, along with the two previously sampled clam sites for which there is arsenic data, 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, estimated clam inorganic arsenic mean concentration (1.56 ug/g wet wt.) exceeds the MCDC non-cancer action level of 0.6 ug/gwet 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 ten clam locations sampled historically (Figure 1.3.1.2.3.1). Cadmium levels detected 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. In earlier sampling, Fort Point Cove cadmium concentrations appeared to be slightly lower in the 2011 clam tissues than the previously sampled 2005 tissues.

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 naturally 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 noncommercially 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 noncancer FTAL).





#### **1.3.1.2.4 Chromium (Cr)**

Chromium was detected at all ten sites sampled historically (Figure 1.3.1.2.4.1). Chromium levels detected 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 median concentration for four sites (two each in ME and NH). All but three clam sites, including the Presumpscot River (2012), fell above the Gulfwatch 2008 median. The Fort Point Cove, Stockton Springs (2005), clam tissue chromium concentrations appeared to be slightly lower in Fort Point Cove samples in 2011, falling below the Gulfwatch 2008 median. The remaining seven sites, including Mare Brook, Brunswick (2012), were all above the Gulfwatch 2008 median.

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 visited historically (Figure 1.3.1.2.5.1). Copper levels detected 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 18 ug/g dry wt. at Presumpscot River, Falmouth/Portland (2012). Copper concentrations in clam tissue at nine sites fell below the 2008 Gulfwatch median, including Mare Brook, Brunswick, sampled in 2012 (LeBlanc et al. 2009). Only Presumpscot River exceeded the Gulfwatch 2008 median. 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).



Figure 1.3.1.2.4.1: Chromium in SWAT Softshell Clams





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 in all ten SWAT softshell clam sites sampled historically (Figure 1.3.1.2.6.1). Iron concentrations detected in clam tissue ranged from a low mean concentration of 1,370 ug/g dry wt. at Squirrel Island, Boothbay, to a high mean concentration of 4,712 ug/g dry wt. at Mast Cove, Eliot. No SWAT sites had clam tissue iron concentrations that approached or exceeded the 2008 Gulfwatch median, including the Presumpscot River, Falmouth/Portland, and Mare Brook, Brunswick sites sampled 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, to a high mean concentration of 5,760 ug/g dry wt. at Mare Brook, Brunswick (Figure 1.3.1.2.6.2). Only clam tissue from two sites, Mare Brook and Presumpscot River (both sampled in 2012), had aluminum concentrations exceeding the 2008 Gulfwatch mean concentration. 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. 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.

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 mussel tissue.

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



#### Figure 1.3.1.2.6.1: Iron in SWAT Softshell Clams





#### 1.3.1.2.7 Nickel (Ni)

Nickel was detected in clam tissue at all ten SWAT softshell clam sites visited historically (Figure 1.3.1.2.7.1). Nickel levels detected 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 median except for the 2012 Presumpscot River concentration and the 2011 Fort Point Cove concentration, which were below the Gulfwatch median. The 2011 Fort Point Cove concentration appeared to be somewhat lower than the 2005 concentration.

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) 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. (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 in all ten SWAT softshell clam sites visited historically (Figure 1.3.1.2.8.1). Lead levels detected 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 8.37 ug/g dry wt. at Mare Brook, Brunswick. Mean lead clam tissue concentrations at nine of ten SWAT sites fell below the 2008 Gulfwatch median. Only the Mare Brook mean lead tissue concentration exceeded the 2008 Gulfwatch median. Lead concentrations at Fort Point Cove appeared to be slightly lower in 2011 than in 2005.

Lead occurs naturally in the earth's crust, however, global lead concentrations in the environmental 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



Figure 1.3.1.2.7.1: Nickel in SWAT Softshell Clams

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Figure 1.3.1.2.8.1: Lead in SWAT Softshell Clams

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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 1.413 ppm (ug/g) wet wt. at Mare Brook, Brunswick (2012), followed by 0.873 ppm wet wt. at Presumpscot River, Falmouth/Portland (2012). Both of these lead concentrations exceed the MCDC lead FTAL, as does 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.

#### 1.3.1.2.9 Mercury (Hg)

Mercury was detected in all ten softshell clam sample locations sampled historically (Figure 1.3.1.2.9.1). Mercury levels detected in clams ranged from a low mean concentration of 0.06  $\mu$ g/g dry wt. at Harris Cove, Eastport, to a high mean concentration of 0.64  $\mu$ 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 Holtrachem site. Five sites had clam tissue concentrations that exceeded the 2008 Gulfwatch mean: Presumpscot River, Falmouth/Portland (2012), and Mast Cove, Eliot (2004); 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 ug/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  $\mu$ 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 in all ten clam sample locations (Figure 1.3.1.2.10.1). Zinc levels detected in clams ranged from a low mean concentration of 56.1 ug/g dry wt. at Squirrel Island, Boothbay, to a high mean concentration of 89.9 ug/g dry wt. at Presumpscot River, Falmouth/Portland, in 2012. Zinc concentrations appeared to be very similar at Fort Point Cove in 2005 and 2011. All ten of the SWAT clam sites had zinc tissue concentrations that fell below the 2008 Gulfwatch median.

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).





#### 1.3.2 PAHs

PAHs occur in elevated concentrations near petroleum manufacturing, creosote use, and wood burning (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 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 (NOAA National Status & Trends, see Kimbrough et al. 2008) and Gulf of Maine (Gulfwatch, see 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. This comparison may be used to place Maine SWAT blue mussel tissue PAH concentrations in a Gulf of Maine-wide and national perspective. 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 Napthalene, and C1-Phenanthrene). The 2012 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 2012 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 2010-12. 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. The table details the PAHs included in summations including 19, 24, and 40 PAHs, and includes a complete list of all PAHs for which results were obtained in 2012 (SWAT data, 75 PAHs described above).

Figure 1.3.2.1.1 shows the summation of the 19 non-alkylated PAHs, the summation of 24 PAHs, and the summation of 40 PAHs compared to the summation of all 75 PAHs (including many alkylated PAHs) and known as total PAHs at the two blue mussel sites sampled by SWAT in 2012, Scarborough River, Scarborough, and Spring Point, South Portland.. Both the 19 summed non-alkylated PAHs and the total PAHs vary in a similar manner between sites, but through viewing the figure it is clear that 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 on the graph in the figure.

Total mean PAH concentrations were 457 ng/g dry wt. at Scarborough River, Scarborough, and 1,292 ng/g dry wt. at Spring Point, South Portland (Figure 1.3.2.1.1). The sum of 19 non-alkylated PAHs was 85 ng/g dry wt. at Scarborough River and 347 ng/g dry wt. at Spring Point.

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 128 ng/g dry wt. at Scarborough River and 429 ng/g dry wt. at Spring Point (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 2012 SWAT blue mussel sites. Both the 40 summed PAHs and the total PAHs vary in a similar manner between sites, but through viewing the figure it is clear that 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 377 ng/g dry wt. at Scarborough River and 1,064 ng/g dry wt. at Spring Point (Figure 1.3.2.1.1).

TABLE 1.3.2.1.1:	Analyzed PAHs and PAH Su	ummation Calculations
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		S	WAT		Gulfwa	atch, NS	&T, SWA	T Summations		
Parameter	2012	2010-11	2007-08	2004-05	ΣΡΑΗ19	ΣΡΑΗ24	ΣΡΑΗ40	Not Analyzed By Gulfwatch	Notes (See below list for more notes)	
ACENAPHTHENE	x	x	x	x	x	x	x			
ACENAPHTHYLENE	х	х	х	х	х	х	х			
ANTHRACENE	х	х	х	х	х	х	х			
2-METHYLANTHRACENE	х	х						missing		
BENZ[A]ANTHRACENE	х	х	х	х	х	х	х			
DIBENZ(A,H)ANTHRACENE	х	x	х	х	х	x	x			
BIPHENYL	х	x	х	х	х	x	x			
BENZO[A]PYRENE	х	х	х	х	х	x	х			
BENZO(E)PYRENE	х	x	x	x	х	x	x			
7-METHYLBENZO[A]PYRENE	x	x						missing		
CHRYSENE	х	x	x	x	x	x	x			
1-METHYLCHRYSENE	x	x						missing		
5/6-METHYLCHRYSENE	x	x						missing		
5,9-DIMETHYLCHRYSENE	х	x						missing		
DIBENZOTHIOPHENE	x	x	1,2,3		x	x	x			
2,4-DIMETHYLDIBENZOTHIOPHENE	x	х						missing		
2/3-METHYLDIBENZOTHIOPHENES	х	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 and BENZO[K]FLUORANTHENE	
3-METHYLFLUORANTHENE/BENZO[A]FLUORENE	x	x								
FLUORENE	х	х	х	х	x	x	х			
2-METHYLFLUORENE	x	x						missing		

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

		S	WAT		Gulfwatch, NS&T, SWAT Summations				
Parameter	2012	2010-11	2007-08	2004-05	ΣΡΑΗ19	ΣΡΑΗ24	ΣΡΑΗ40	Not Analyzed By Gulfwatch	Notes (See below list for more notes)
1,7-DIMETHYLFLUORENE	x	x						missing	
NAPHTHALENE	x	х	x	x	x	х	х		
1-METHYLNAPHTHALENE	x	x	x					missing	
2-METHYLNAPHTHALENE	x	х	x					missing	
1,2-DIMETHYLNAPHTHALENE	x	х						missing	
2,6-DIMETHYLNAPHTHALENE	x	x	x					missing	
2,3,5-TRIMETHYLNAPHTHALENE	x	x	x					missing	
2,3,6-TRIMETHYLNAPHTHALENE	x	х						missing	
1,4,6,7-TETRAMETHYLNAPHTHALENE	x	x						missing	
PERYLENE	x	х	x	x		х	х		
BENZO[GHI]PERYLENE	x	x	x	х	x	х	x		
PHENANTHRENE	x	x	x	x	x	x	x		
1-METHYLPHENANTHRENE	x	х	x					missing	
2-METHYLPHENANTHRENE	x	x						missing	
3-METHYLPHENANTHRENE	x	x						missing	
9/4-METHYLPHENANTHRENE	x	х						missing	
1,7-DIMETHYLPHENANTHRENE	x	х						missing	
1,8-DIMETHYLPHENANTHRENE	x	x						missing	
2,6-DIMETHYLPHENANTHRENE	x	х						missing	
3,6-DIMETHYLPHENANTHRENE	x	x						missing	
1,2,6-TRIMETHYLPHENANTHRENE	x	x						missing	
PYRENE	x	x	x	x	x	х	x		
INDENO[1.2.3-CD]PYRENE	x	x	x	x	x	x	x		

		:	ions						
Parameter	2012	2010-11	2007-08	2004-05	ΣΡΑΗ19	ΣΡΑΗ24	ΣΡΑΗ40	Not Analyzed By Gulfwatch	Notes (See below list for more notes)
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				х		
C3-FLUORENES	x	x	3				x		
C1-NAPHTHALENES	x	x	2,3			x	х		
C2-NAPHTHALENES	х	x	2,3			х	х		
C3-NAPHTHALENES	x	x	2,3			х	х		
C4-NAPHTHALENES	x	х						missing	

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

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

		ns							
Parameter	2012	2010-11	2007-08	2004-05	ΣΡΑΗ19	ΣΡΑΗ24	ΣΡΑΗ40	Not Analyzed By Gulfwatch	Notes (See below list for more notes)
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				х		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 ' 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 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 compares the sum of 19 PAHs at the SWAT blue mussel sites sampled in 2012 to the Gulfwatch 2008 median and 85<sup>th</sup> percentile results. The sum of 19 PAHs from mussel tissue in Scarborough River was below the Gulfwatch median, while the sum of 19 PAHs from Spring Point 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 2012 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). Scarborough River did not exceed the 2008 NS&T median of 180 ng/g (dry weight) for 19 summed non-alkylated PAHs, but Spring Point 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 2012, to the Gulfwatch 2008 median and 85<sup>th</sup> percentile results. Scarborough River was below the Gulfwatch 2008 median of 198 ng/g (dry weight) for 24 summed PAHs. Spring Point 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 2012 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). Scarborough River was below the NS&T 2008 median of 247 ng/g (dry weight) for 24 summed PAHs, while Spring Point 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 2012 to the Gulfwatch 2008 median and 85<sup>th</sup> percentile results. Scarborough River had a sum of 40 PAHs between the Gulfwatch 2008 median of 260 ng/g (dry weight) for 40 summed PAHs and the Gulfwatch 85<sup>th</sup> percentile of 618 ng/g (dry weight) for 40 summed PAHs, while the sum of 40 PAHs at Spring Point exceeded the Gulfwatch 85<sup>th</sup> percentile 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-Phananthrenes/Anthracenes, where Gulfwatch utilizes C1 through C4-Phananthrenes. 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 1.3.2.1.4.

Figure 1.3.2.1.1 also compares the sum of 40 PAHs at the 2012 SWAT mussel sites to recent NS&T median and 85<sup>th</sup> percentile for 40 summed PAHs (2008 data, the most recent available). Both sites had a sum of 40 PAHs which exceeded the NS&T 2008 median of 353 ng/g (dry weight) for 40 summed PAHs. Neither of the SWAT sites approached or 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 Spring Point 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 2012 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:

Fluoranthene + Pyrene/ $\Sigma$ (Flouranthene + Pyrene + 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 2012, Scarborough River, Scarborough, and Spring Point, South Portland, have ratios above the <0.1 mark, which would indicate a petrogenic source of PAHs. The higher ratio in the two 2012 SWAT blue mussel sites (both in the greater Portland area) may be attributed to the urbanized upland area (Portland, major roads, etc.) with associated impervious surfaces and combusted hydrocarbon runoff or to 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 reproduction 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 (NOAA National Status & Trends, see Kimbrough et al. 2008) shellfish data and Gulf of Maine (Gulfwatch, see 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, Blue Mussels. 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 and to place Maine SWAT data in wider geographic context.

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. Table 1.3.2.1.1 details the PAHs included in summations and includes a complete list of all PAHs for which results were obtained in the different years sampled.



Figure 1.3.2.2.1 shows the summation of the 19 non-alkylated PAHs at the two SWAT clam sites sampled in 2002: Presumpscot River, Falmouth/Portland, and Mare Brook, Brunswick. The sum of 19 non-alkylated PAHs was 137 ng/g dry wt. at Mare Brook and 248 ng/g dry wt. at Presumpscot River. Neither of the SWAT clam sites exceeded the 2008 Gulfwatch mean concentration for the sum of 19 non-alkylated PAHs, 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. The sum of 40 PAHs concentration in Presumpscot River clam tissue exceeded the 2008 Gulfwatch median (four sites, two in NH and two in ME), though the concentration in Mare Brook clam tissue did not. Summations of 24 PAHs at both sites did not exceed the Gulfwatch mean. 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.

PAH results from Presumpscot River, Falmouth/Portland, and Mare Brook, Brunswick, 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:

Fluoranthene + Pyrene/ $\Sigma$ (Flouranthene + Pyrene + 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 only two sites each in 2012, the calculated ratios for the two clam sites have been included in Figure 1.3.2.1.5, which also depicts the calculated ratios for the blue mussel sites sampled in 2012. Both 2012 clam sites have pyrogenic ratios, between 0.2 and 0.3, indicating the major PAHs components present in clam tissue at these sites is 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 reproduction 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).



## Figure 1.3.2.2.1: PAHs in Softshell Clams

Solid lines = Gulfwatch 2008 Means for  $\sum$ 19 PAHs,  $\sum$ 24 PAHs, and  $\sum$ 40 PAHs; (Four softshell Clam Sites in NH and ME; No  $\sum$ Total PAHs Mean is Available for Gulfwatch).
# 1.3.3 PCBs

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, with the SWAT 35 congener list 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 the co-elution issue, since some congeners are co-eluting differently or are summed together differently at the various laboratories used. 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 coelution 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 completely 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.

<u>SUM 35 PCBs</u> "SWAT PCBs" List	<u>SUM 31 PCBs</u> <u>"Gulfwatch, NS&amp;T PCBs"</u>
	List
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	

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

Unique to SWAT 35 List	Unique to GW and NS&T List
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 other data sets like Gulfwatch and NS&T Musselwatch. Comparing the two mussel sites sampled for PCBs in 2012, the SWAT PCBs were 38.6% to 38.3% of the total PCBs at Scarborough River, Scarborough, and Presumpscot River, Falmouth/Portland, respectively. The relationship between total PCBs and the 2012 SWAT PCBs subset can be noted. Total PCB concentrations were 17.6 ng/g dry wt. at Scarborough River and 57.5 ng/g dry wt. at Spring Point (Figure 1.3.3.1.1).

Figure 1.3.3.1.1 compares the SWAT PCBs at the 2012 SWAT mussel sites to recent Gulfwatch median and 85<sup>th</sup> percentile for 2008 PCB data, the most recent available. Neither Scarborough River nor Spring Point exceeded the Gulfwatch 2008 median of 24.1 ng/g (dry weight), and consequently neither of the sites tested in 2012 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 2012 SWAT sites to recent NS&T (NS&T) median and 85<sup>th</sup> percentile 2008 PCB data, the most recent available. Neither of the two SWAT sites exceeded the NS&T 2008 median, 29.2 ng/g (dry weight), and so neither exceeded the NS&T national 85<sup>th</sup> percentile, 141 ng/g (dry weight). The 2008 NS&T 85<sup>th</sup> percentile was approximately 7 X higher than the highest scoring PCB site tested by SWAT in Maine in 2012, Spring Point, South Portland (220.3 ng/g, dry weight). 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 noncommercially 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 two SWAT blue mussel sites sampled in 2012, Spring Point had a total PCB mean tissue concentration of 8.7 ng/g wet wt., while Scarborough River had a total PCB mean tissue concentration of 2.7 ng/g wet wt. Neither site had a tissue concentration exceeding 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 2012, Presumpscot River, Falmouth/Portland, and Mare Brook, Brunswick, and results compared to Gulf of Maine (Gulfwatch, see LeBlanc et al. 2009) softshell clam monitoring program data in an effort to place Maine SWAT data in a national and 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 NS&T/Gulfwatch PCB congeners included in the summed lists are not completely

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.



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 2012 clam sites, the SWAT PCBs were 33.4% and 33.8% of the total PCBs at Presumpscot River and Mare Brook, respectively. Total PCB concentrations were 8.3 ng/g dry wt. at Mare Brook, Brunswick, and 26.7 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 newer lab (AXYS Analytical) that analyzed the 2010-12 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 six recently sampled SWAT clam sites to a recent Gulfwatch clam site sampled in 2008 (the most recent available). All six SWAT clam site sums of 35 PCBs fell below the one Gulfwatch site concentration, though the Presumpscot summation was very close to the Gulfwatch summation. 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 noncommercially 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 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.



## 1.3.4 Pesticides

#### 1.3.4.1 Blue Mussels

Blue mussels were tested for pesticides from two sites in 2012. Organochlorinated pesticide results were compared to national (NOAA NS&T, see Kimbrough et al. 2008) and Gulf of Maine (Gulfwatch, see 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. To allow comparison to other NS&T Mussel Watch program work, summations of SWAT data were completed for: DDDs, DDEs, and DDTs; chlordanes; and dieldrins. Methodology was consistent with that used by NS&T in constructing summations of these pesticide compound groups. Use of these summations assists in putting Maine SWAT data into a national context.

The NS&T and Gulfwatch programs utilize a summation of 21 organochlorinated pesticides to look at general pesticide concentrations. SWAT pesticide laboratory results include these 21 organochlorinated pesticides and several more. Table 1.3.4.1.1 shows the Gulfwatch list of 21 organochlorinated pesticides (also used by NS&T Mussel Watch Program) and as well shows additional pesticides included in SWAT results. To allow direct comparison to Gulfwatch and NS&T results summing 21 organochlorinated pesticides, SWAT data were summed for the same 21 organochlorinated pesticides.

		SWA]	Γ
	<b>Gulfwatch Chlorinated</b>		
	Pesticides	2010-2012	2005
<b>Organochlorines</b>			
ALDRIN	Х	Х	Х
ALPHA-BHC	Х	Х	
BETA-BHC		Х	
DELTA-BHC		Х	
GAMMA-BHC (LINDANE)	Х	Х	Х
CAPTAN		Х	
ALPHA-CHLORDANE (cis-	Х	Х	Х
CHLORDANE)			
GAMMA-CHLORDANE	х	Х	Х
CHLOROTHALONIL		Х	
DACTHAL		Х	
2,4'-DDD	Х	Х	Х
4,4'-DDD	Х	Х	Х
2,4'-DDE	х	Х	Х
4,4'-DDE	Х	Х	Х
2,4'-DDT	Х	Х	Х
4,4'-DDT	Х	Х	Х
DIELDRIN	Х	Х	Х

# Table 1.3.4.1.1: Pesticides Utilized in SWAT Blue Mussel and Softshell Clam Analysis

ENDOSULFAN Ι (α-ENDOSULFAN)	Х	X	х
ENDOSULFAN ΙΙ (β-ENDOSULFAN	Х	Х	х
ENDOSULFAN SULFATE		Х	
ENDRIN	Х	Х	х
ENDRIN KETONE		Х	
HEPTACHLOR	Х	Х	х
HEPTACHLOR EPOXIDE	Х	Х	х
HEXACHLOROBENZENE	Х	Х	х
METHOXYCHLOR	Х	Х	
MIREX	Х	Х	х
CIS-NONACHLOR		Х	
TRANS-NONACHLOR	Х	Х	х
OCTACHLOROSTYRENE		Х	
OXYCHLORDANE		Х	
PERTHANE		Х	
QUINTOZENE		Х	
TECNAZENE		Х	
ENDRIN ALDEHYDE			
Total Number	21	34	19

#### 1.3.4.1.1 ΣDDTs

The summations of DDDs, DDEs, and DDTs, (six compounds total, called  $\Sigma$ DDTs in this report) were calculated from SWAT data and mean concentrations were 6.8 ng/g dry wt. at Scarborough River, Scarborough, and 13.8 ng/g dry wt. at Spring Point, South Portland. The NS&T Mussel Watch considers  $\Sigma$ DDTs scores between 0 and 112 ng/g dry wt. in blue mussel tissue to be "low" (groupings include low, moderate, and high) on a national scale. This is consistent with all mussel sites sampled Maine coast-wide over the past four years, with all sites falling into the low end of the low NS&T grouping.

 $\Sigma$ DDTs are in the low range in blue mussels throughout the northeast, with higher scores occurring in oysters in the Gulf of Mexico and in mussels on the southwest coast of California. Highest concentrations are generally found near historic DDT manufacturing plants. DDT was banned in the US in 1972, after widespread use as a pesticide. DDT is persistent in the environment and also is hydrophobic, leading to DDT bioaccumulating in organisms. DDT concentrations in shellfish are decreasing across US sampling stations (Kimbrough et al. 2008).

From a human health perspective, MCDC reports a cancer DDT FTAL of 64 ng/g wet wt. (ppb) and a non-cancer DDT FTAL of 1,080 ng/g wet wt. The MCDC DDT FTALs are based on the summation of DDDs, DDEs, and DDTs, (six compounds total, called  $\Sigma$ DDTs in this report), except that they are expressed on a wet tissue weight basis rather than a dry weight basis used in the SWAT monitoring segments of this report. When converted to wet weight, the highest 2012 SWAT blue mussel tissue DDT concentration was 1.1 ng/g wet wt., which is <2% of the more conservative MCDC cancer FTAL of 64 ng/g wet wt.

## **1.3.4.1.2** ΣChlordanes

The summations of alpha-chlordane, heptachlor, trans-nonachlor, and heptachlor epoxide (four compounds total, called  $\Sigma$ Chlordanes in this report) were calculated from SWAT data and mean concentrations were 1.1 ng/g dry wt. at Scarborough River and 1.5 ng/g dry wt. at Spring Point. NS&T considers  $\Sigma$ Chlordanes scores between 0 and 8 ng/g dry wt. in blue mussel tissue to be "low" (groupings include low, moderate, and high) on a national scale. Both sites sampled in 2012 fall in the lowest quarter of that low category (Kimbrough et al. 2008).

 $\Sigma$ Chlordanes are in the low range in blue mussels throughout much of the northeast US, with a few exceptions in urbanized areas like Boston or New York City. Highest concentrations are generally found near areas of historic agricultural use or in urban areas from termite control applications (Kimbrough et al. 2008). Chlordane, one of the cyclodiene organic pesticides, is a mixture of more than fifty compounds, but is predominantly made up of alpha- and gamma-chlordane, heptachlor, and nonachlor. The NS&T and this SWAT summation capture three of these compounds, plus one transformation product (heptachlor epoxide). Chlordane was used from roughly 1948 through 1983 in agriculture, when it was banned. Chlordane was also the primary insecticide for termite control underground. All uses were banned in 1988 (Kimbrough et al. 2008). NS&T Mussel Watch reported that Chlordane was one of the most ubiquitous contaminants measured by that program.  $\Sigma$ Chlordanes concentrations in shellfish are decreasing across US sampling stations (Kimbrough et al. 2008).

The MCDC reports a cancer and non-cancer FTALs for chlordane/nonachlor (summation of alpha-chlordane, gamma-chlordane, and trans-nonachlor) and heptachlor epoxide. MCDC reports a cancer FTAL of 17 ng/g wet wt. and a non-cancer FTAL of 130 ng/g wet wt. for chlordane/nonachlor. The 2012 SWAT blue mussel tissue data, when summed in the same manner, shows the highest mean concentration recorded to be 0.18 ng/g wet wt., which is 1% of the 17 ng/g cancer FTAL. MCDC reports a cancer FTAL of 2.4 ng/g wet wt. and a non-cancer FTAL 28 ng/g wet wt. for heptachlor epoxide. The highest mean value for heptachlor epoxide in 2012 SWAT blue mussel tissue was 0.031 ng/g wet wt., which is 1.3% of the 2.4 ng/g wet wt. cancer FTAL.

## 1.3.4.1.3 ΣDieldrins

The summations of aldrin and dieldrin (two compounds total, called  $\Sigma$ Dieldrins in this report) were calculated from SWAT data and mean concentrations were 0.58 ng/g dry wt. at Scarborough River and 0.31 ng/g dry wt. at Spring Point. NS&T Mussel Watch considers  $\Sigma$ Dieldrins scores between 0 and 8 ng/g dry wt. in blue mussel tissue to be "low" (groupings include low, moderate, and high) on a national scale, with both of the 2012 SWAT sites falling in the bottom of the low category (Kimbrough et al. 2008).

 $\Sigma$ Dieldrins are in the low range in blue mussels throughout most of the northeast US. Nationally, the highest concentrations are generally found near areas of historic pesticide use and manufacturing (Kimbrough et al. 2008). Dieldrin and aldrin were used as insecticides through the 1960s for the control of termites and on crops. All uses were suspended in 1970, but use as a termite insecticide was allowed again from 1972 through 1989, when use was again cancelled. Aldrin and dieldrin are carcinogenic in animals, and are thought to be in humans (Kimbrough et al. 2008).

From a human health perspective, MCDC reports cancer and non-cancer FTALs for dieldrin and separately for aldrin. MCDC reports a cancer FTAL of 1.4 ng/g wet wt. and a non-cancer FTAL of 108 ng/g wet wt. for dieldrin. The highest dieldrin mean concentration in blue mussel tissue in 2012 SWAT data was 0.07 ng/g wet wt., which is 5.2% of the MCDC cancer FTAL. MCDC reports a cancer FTAL of 1.3 ng/g wet wt. and a non-cancer FTAL of 65 ng/g wet wt. for aldrin. The highest aldrin mean concentration in blue mussel tissue in 2012 SWAT data was 0.033 ng/g wet wt., which is 2.5% of the MCDC cancer FTAL.

#### 1.3.4.1.4 Σ21 Organochlorines

The summations of 21 organochlorine pesticides (as noted in Table 1.3.4.1.1 and called  $\Sigma 21$  Pesticides in this report) were calculated from SWAT data and mean concentrations were 10.6 ng/g dry wt. at Scarborough River and 18.2 ng/g dry wt. at Spring Point. Both 2012 SWAT sites exceeded the Gulfwatch 2008 median, 9.9 ng/g (dry weight) for  $\Sigma 21$  Pesticides. Spring Point exceeded the Gulfwatch 85<sup>th</sup> percentile, 14.3 ng/g (dry weight), for  $\Sigma 21$  Pesticides. Neither of the SWAT sites tested exceeded the NS&T 2008 median, 22.9 ng/g (dry weight) for  $\Sigma 21$  Pesticides. Neither of the NS&T 85<sup>th</sup> percentile of 128 ng/g (dry weight) for  $\Sigma 21$  Pesticides.

From a human health perspective, the MCDC reports cancer and/or non-cancer FTALs for several individual chlorinated pesticides which fall under the heading of the  $\Sigma 21$  Pesticides discussed above. To compare the FTALs to SWAT data, the individual pesticide data have been expressed on wet weight basis and matched to the corresponding MCDC FTAL.

For hexachlorobenzene, MCDC reports a cancer FTAL of 14 ng/g wet wt. and a non-cancer FTAL of 1,728 ng/g wet wt. The highest mean hexachlorobenzene concentration in blue mussel tissue detected by SWAT in 2012 was 0.071 ng/g wet wt., which is 0.5% of the more protective MCDC cancer FTAL.

For heptachlor, MCDC reports a cancer FTAL of 5 ng/g wet wt. and a non-cancer FTAL of 1,080 ng/g wet wt. The highest mean heptachlor concentration in blue mussel tissue detected by SWAT in 2012 was 0.068 ng/g wet wt., which was 1.4% of the more protective MCDC cancer FTAL.

For mirex, MCDC reports a non-cancer FTAL of 432 ng/g wet wt. MCDC does not report a cancer FTAL for mirex. The highest mean mirex concentration in blue mussel tissue detected by SWAT in 2012 was 0.155 ng/g wet wt., which was 0.04% of the MCDC non-cancer FTAL.

For lindane, MCDC reports a cancer FTAL of 17 ng/g wet wt. and a non-cancer FTAL of 648 ng/g wet wt. The highest mean lindane concentration in blue mussel tissue detected by SWAT in 2012 was 0.175 ng/g wet wt., which was 1.0% of the more protective MCDC cancer FTAL.

For endosulfan (summation of endosulfan I and II), MCDC reports a non-cancer FTAL of 12,963 ng/g wet wt. MCDC does not report a cancer FTAL for endosulfan. The highest mean endosulfan concentration in blue mussel tissue detected by SWAT in 2012 was 0.111 ng/g wet wt., which was 0.0009% of the MCDC non-cancer FTAL.

# 1.3.4.2 Softshell Clams

Softshell clam tissue sampled from Presumpscot River, Falmouth/Portland, and Mare Brook, Brunswick, was analyzed for pesticides. Organochlorinated pesticide results were compared to national (NOAA NS&T, see Kimbrough et al 2008) and Gulf of Maine (Gulfwatch, see LeBlanc et al. 2009) softshell clam data (when available) in an effort to place Maine SWAT data in a national and regional context, respectively. To allow comparison to other NS&T Mussel Watch program work, summations of SWAT data were completed for: DDDs, DDEs, and DDTs; chlordanes; and dieldrins. Methodology was consistent with that used by NS&T in constructing summations of these pesticide compound groups. Use of these summations assists in putting Maine SWAT clam tissue data into a national context.

The summations of 21 organochlorinated pesticides utilized by the NS&T and Gulfwatch programs were discussed previously in Section 1.3.4.1 and Table 1.3.4.1.1 in that section shows the Gulfwatch list of 21 organochlorinated pesticides (also used by NS&T Mussel Watch Program) and additional pesticides included in SWAT results. To allow direct comparison to Gulfwatch and NS&T results summing 21 organochlorinated pesticides, SWAT data were summed for the same 21 organochlorinated pesticides.

# 1.3.4.2.1 ΣDDTs

The summation of DDDs, DDEs, and DDTs, (six compounds total, called  $\Sigma$ DDTs in this report) is presented in Figure 1.3.4.2.1.1 for the two 2012 SWAT softshell clam sites, and for four historical clam sites (2005, 2010-11).  $\Sigma$ DDTs ranged from a low mean concentration of 3.2 ng/g dry wt. at Mare Brook, Brunswick (2012), to a high mean concentration of 8.7 ng/g dry wt. at Presumpscot River, Falmouth/Portland (2012).  $\Sigma$ DDTs at Fort Point Cove appear to have decreased slightly or remained consistent from 2005 to 2011 (5.1 to 4.6 ng/g dry wt.). The NS&T Mussel Watch considers  $\Sigma$ DDTs scores between 0 and 112 ng/g dry wt. in blue mussel tissue to be "low" (groupings include low, moderate, and high) on a national scale. The NS&T Mussel Watch considers  $\Sigma$ DDTs scores between 1 and 34 ng/g dry wt. in oyster tissue to be "low" (groupings include low, moderate, and high) on a national scale. Since no NS&T clam tissue data is available, comparison to these mussel and oyster groupings suggests that all six SWAT clam sites appear to have  $\Sigma$ DDTs concentrations that lie within the low category of these two ranges.

 $\Sigma$ DDTs are also in the low range in blue mussels throughout the northeast, with higher scores occurring in oysters in the Gulf of Mexico and in mussels on the southwest coast of California. Highest concentrations are generally found near historic DDT manufacturing plants. DDT was banned in the US in 1972, after widespread use as a pesticide. DDT is persistent in the environment and also is hydrophobic, leading to DDT bioaccumulating in organisms. DDT concentrations in shellfish are decreasing across US sampling stations (Kimbrough et al. 2008).

From a human health perspective, MCDC reports a cancer DDT FTAL of 64 ng/g wet wt. (ppb) and a non-cancer DDT FTAL of 1,080 ng/g wet wt. The MCDC DDT FTALs are



based on the summation of DDDs, DDEs, and DDTs, (six compounds total, called  $\Sigma$ DDTs in this report), except that they are expressed on a wet tissue weight basis rather than a dry weight basis used in the SWAT monitoring segments of this report. When converted to wet weight, the highest SWAT softshell clam tissue DDT concentration was 1.39 ng/g wet wt., which is 2.2% of the more conservative MCDC cancer FTAL of 64 ng/g wet wt.

# 1.3.4.2.2 ΣChlordanes

The summation of alpha-chlordane, heptachlor, trans-nonachlor, and heptachlor epoxide (four compounds total, called  $\Sigma$ Chlordanes in this report) was determined from SWAT data and is presented in Figure 1.3.4.2.2.1.  $\Sigma$ Chlordanes ranged from a low mean concentration of 0.5 ng/g dry wt. at Mare Brook (2012), to a high mean concentration of 3.1 ng/g dry wt. at Mill Cove, Robbinston (2005). The large standard deviation in the  $\Sigma$ Chlordanes used to calculate the mean results from one of three samples having a much higher concentration, which influenced the mean as well. NS&T considers  $\Sigma$ Chlordanes scores between 0 and 8 ng/g dry wt. (in blue mussel tissue) to be "low" (groupings include low, moderate, and high) on a national scale. NS&T considers  $\Sigma$ Chlordanes scores between 0 and 7 ng/g dry wt. (in oyster tissue) to be "low" (groupings include low, moderate, Since no NS&T clam tissue data is available, comparison to these mussel and oyster groupings suggests that all six SWAT clam sites appear to have  $\Sigma$ Chlordane concentrations that lie within the lower half of the low grouping of these two species (Kimbrough et al. 2008). Chlordanes are discussed in more detail in the previous Section 1.3.4.1.2, including their geographic distribution, composition, historic usage, and recent trends.

The MCDC reports cancer and non-cancer FTALs for chlordane/nonachlor (summation of alpha-chlordane, gamma-chlordane, and trans-nonachlor) and heptachlor epoxide. MCDC reports a cancer FTAL of 17 ng/g wet wt. and a non-cancer FTAL of 130 ng/g wet wt. for chlordane/nonachlor. The SWAT clam tissue data, when summed in the same manner, shows the highest mean concentration recorded at a site to be 0.24 ng/g wet wt., which is 1.4% of the 17 ng/g cancer FTAL. MCDC reports a cancer FTAL of 2.4 ng/g wet wt. and a non-cancer FTAL 28 ng/g wet wt. for heptachlor epoxide. The highest mean value for heptachlor epoxide in SWAT clam tissue was 0.165 ng/g wet wt., which is 6.5% of the 2.4 ng/g wet wt. cancer FTAL.

## 1.3.4.2.3 ΣDieldrins

The summation of aldrin and dieldrin (two compounds total, called  $\Sigma$ Dieldrins in this report) was determined from SWAT clam tissue data and is presented in Figure 1.3.4.2.3.1.  $\Sigma$ Dieldrins ranged from a low mean concentration of 0.23 ng/g dry wt. at Mare Brook, Brunswick (2012) to a high mean concentration of 0.61 ng/g dry wt. at Mill Cove, Robbinston (2005). The large standard deviation in the  $\Sigma$ Dieldrins used to calculate the mean results from one of three samples having a much higher concentration, which influenced the mean as well. NS&T Mussel Watch considers  $\Sigma$ Dieldrins scores between 0 and 8 ng/g dry wt. in blue mussel tissue to be "low" (groupings include low, moderate, and high) on a national scale. NS&T Mussel Watch considers  $\Sigma$ Dieldrins scores between 0 and 5 ng/g dry wt. in oyster tissue to be "low" (groupings include low, include low, include low,





moderate, and high) on a national scale. Since no NS&T clam tissue data is available, comparison to these mussel and oyster groupings suggests that all six SWAT clam sites appear to have  $\Sigma$ Dieldrins concentrations that lie within the lower third of the low grouping of these two species (Kimbrough et al. 2008). The geographic distribution, historic usage, and recent trends in dieldrins are discussed in Section 1.3.4.1.3 in relation to blue mussels.

From a human health perspective, MCDC reports cancer and non-cancer FTALs for dieldrin and separately for aldrin. MCDC reports a cancer FTAL of 1.4 ng/g wet wt. and a non-cancer FTAL of 108 ng/g wet wt. for dieldrin. The highest dieldrin mean concentration in SWAT softshell clam tissue was 0.073 ng/g wet wt., which is 5.2% of the MCDC cancer FTAL. MCDC reports a cancer FTAL of 1.3 ng/g wet wt. and a non-cancer FTAL of 65 ng/g wet wt. for aldrin. The highest aldrin mean concentration in clam tissue data was 0.028 ng/g wet wt., which is 2% of the MCDC cancer FTAL.

## 1.3.4.2.4 Σ21 Organochlorines

The summation of 21 organochlorine pesticides (as noted in Table 1.3.4.1.1) was determined from SWAT softshell clam data and is presented in Figure 1.3.4.2.4.1 (called  $\Sigma$ 21 Pesticides in this report).  $\Sigma$ 21 Pesticides ranged from a low mean concentration of 6.27 ng/g dry wt. at Mare Brook, Brunswick (2012), to a high mean concentration of 13.71 ng/g dry wt. at Fort Point Cove, Stockton Springs (2005). Figure 1.3.4.2.4.1 compares the sum of  $\Sigma$ 21 Pesticides at the SWAT clam sites to a recent Gulfwatch median at four clam sites (2008 data, the most recent available). All seven of the SWAT mean concentrations (six sites) exceeded the Gulfwatch 2008 median, 4.2 ng/g (dry weight) for  $\Sigma$ 21 Pesticides. No Gulfwatch 85<sup>th</sup> percentile was calculated since only four Gulfwatch clam sites comprised the entire 2008 data set. Of note is the highest scoring Gulfwatch clam site, with  $\Sigma$ 21 Pesticides of 12.0 ng/g (dry weight) at North Mill Pond, NH, which was quite similar to the highest Maine SWAT clam site.

Of the seven SWAT clam mean concentrations, none exceeded the NS&T 2008 median, 22.9 ng/g (dry weight), for  $\Sigma 21$  Pesticides in shellfish, and consequently none of the SWAT sites exceeded the NS&T 85<sup>th</sup> percentile of 128 ng/g (dry weight) for  $\Sigma 21$  Pesticides.

From a human health perspective, the MCDC reports cancer and/or non-cancer FTALs for several individual chlorinated pesticides which fall under the heading of the  $\Sigma 21$  Pesticides discussed above. To compare the FTALs to SWAT data, the individual pesticide data has been expressed on wet weight basis and matched to the corresponding MCDC FTAL.

For hexachlorobenzene, MCDC reports a cancer FTAL of 14 ng/g wet wt. and a non-cancer FTAL of 1,728 ng/g wet wt. The highest mean hexachlorobenzene concentration in softshell clam tissue detected by SWAT was 0.89 ng/g wet wt., which is 6.4% of the more protective MCDC cancer FTAL.



For heptachlor, MCDC reports a cancer FTAL of 5 ng/g wet wt. and a non-cancer FTAL of 1,080 ng/g wet wt. The highest mean heptachlor concentration in SWAT clam tissue was 0.22 ng/g wet wt., which was 4.4% of the more protective MCDC cancer FTAL.

For mirex, MCDC reports a non-cancer FTAL of 432 ng/g wet wt. MCDC does not report a cancer FTAL for mirex. The highest mean mirex concentration in SWAT clam tissue was 0.174 ng/g wet wt., which was 0.04% of the MCDC non-cancer FTAL. All mirex results from clam tissue analyzed yielded non-detects at the detection limits utilized by the two labs, with concentrations noted above based on estimated values at the half the detection limits.

For lindane, MCDC reports a cancer FTAL of 17 ng/g wet wt. and a non-cancer FTAL of 648 ng/g wet wt. The highest mean lindane concentration in SWAT clam tissue was 0.32 ng/g wet wt., which was 1.8% of the more protective MCDC cancer FTAL.

For endosulfan (summation of endosulfan I and II), MCDC reports a non-cancer FTAL of 12,963 ng/g wet wt. MCDC does not report a cancer FTAL for endosulfan. The highest mean endosulfan concentration in SWAT clam tissue was 0.128 ng/g wet wt., which was 0.001% of the MCDC non-cancer FTAL. All endosulfan results from clam tissue analyzed yielded non-detects at the detection limits utilized by the two labs, with concentrations noted above based on estimated values at the half the detection limits.

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# 2.0 LAKES MODULE

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# 2.1 MERCURY IN COASTAL LAKES

**2.1.1** Background. 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 (DiFranco *et al.*, 1995), including notably high concentrations within Acadia National Park (Burgess, 1997), resulted in a statewide fish consumption advisory for all lakes and ponds in 1994 and all freshwaters in 1997. 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; Kahl *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 concentration to be located adjacent to another lake with relatively low fish Hg levels.

Currently, Maine's Department of Environmental Protection is collaborating with researchers at the University of Maine to better characterize Hg concentrations in fish and understand ecological drivers that result in spatial heterogeneity. This effort includes an updated assessment of fish tissue contamination in Maine lakes - many of which have not been sampled since the original advisories were issued – and rigorous analysis and state specific modeling of linkages among lake trophic status, of Hg body burden in freshwater fish, and of other relevant watershed characteristics. Results of the ongoing work and final model will be shared with toxicologists at the Maine Center for Disease Control (ME-CDC), for consideration as they move forward with advisory evaluations. Preliminary sampling of a few lakes in close proximity to the Maine coast indicated the Hg concentrations were particularly high in both lake water and fish tissue (Bacon, unpubl. data). Enriched Hg deposition in fog as documented on Grand Manan Island just northeast of the Maine-New Brunswick border is a potential mechanism that could explain high concentrations of Hg on the Maine coast (Ritchie et al. 2006). In addition, Krabbenhoft et al. (2011) have developed a model that predicts sensitivity to Hg methylation based on geochemical parameters such as dissolved organic carbon and sulfate, which are elevated in many systems along the Maine coast. These factors could combine to make coastal systems, in particular, Acadia National Park, among the most sensitive to methylation and bioaccumulation of Hg in fish.

Considerable research at Acadia National Park (ANP) on Mount Desert Island (MDI), Maine, has focused on Hg over the past few decades (Vaux et. al., 2008). Notably, neither fish nor lake water has been analyzed for Hg on MDI since the original study in the mid-1990s (Burgess, 1997), despite stream and wetland studies indicating elevated Hg concentrations in surface waters throughout the Park (Figure 1) and extremely high concentrations of Hg in biota: amphibian Hg in Acadia exceeded that at an acidified site (Bank et al. 2005); and tree swallow Hg exceeded that at a Superfund site (Longcore et al. 2007). This proposed research would support fish tissue, water, and sediment sampling and Hg and MeHg analyses from 15 lakes located within approximately 30 km of the Maine Coast, with at least half located on Mount Desert Island (MDI), to allow for comparison to Hg datasets collected from stream watersheds on MDI and update Acadia National Park data regarding Hg in fish for the first 2010 and Nelson et al. 2007). time since the mid-1990s. This sampling will be



Figure 1. Total Hg concentrations in streams on the Mount Desert Island section of Acadia National Park (from Flanagan

done according to Maine DEP protocols to assure that the results will be useable by both DEP and ME-CDC. These data will be utilized in conjunction with existing data (e.g., landscape attributes, lake chemistry and morphometry), to develop a model component for coastal proximity that could improve predictions of Hg contamination levels in Maine fish. These results will be shared with the ME-CDC.

2.1.2 Objectives. This research project will assess the relative importance of proximity to the ocean (coastal proximity) as one factor that aids in prediction of Hg body burdens in fish. Ongoing research by our group and other investigators has already identified several controls on Hg in lake water or fish. For example, Hg concentration in fish is inversely proportional to lake productivity (trophic status), with fish in eutrophic lakes possessing a lower Hg body burden, via a process termed 'biodilution' (Adams et al., 2009; Dittman and Driscoll, 2009; Chen and Folt, 2005; Chen et al, 2005). Phosphorus (P) concentration is a control on lake productivity, and is largely controlled by landscape factors and sediment geochemistry. Percent wetland cover, land cover type (e.g., agriculture, forest type), lake geomorphology, and lake water chemistry (e.g., DOC, sulfate, pH) have been implicated as controls on Hg concentrations in lake water or fish (Kamman et al. 2004, Simonin et al. 2008). However, to our knowledge, no published studies have focused on the importance of coastal proximity.

Consequently, modeling of fish tissue Hg contamination in Maine is currently being pursued with close evaluation of the determinants of lake trophic status in concert with Hg concentrations (in water, fish tissue, and sediment) and watershed characteristics. Watershed characteristics are hypothesized to control (1) the flux of Hg from the watershed to the lake, and (2) lake productivity, and thereby Hg accumulation in the food chain. This modeling project suggests that coastal proximity is an additional factor worthy of consideration.

We have used the existing DEP and UMaine databases to identify relationships among watershed attributes, lake trophic state, and fish Hg burden to build a predictive model. Our preliminary statistical analysis from 92 lakes in Maine suggests that dissolved organic carbon (DOC) concentration, lake maximum depth and Secchi depth positively influence fish tissue Hg concentrations and alkalinity negatively influences fish tissue Hg concentrations. DOC transports Hg from the watershed to the lake. Wetlands are the most important source of DOC to lakes and are known as 'hotspots' for the highly bioaccumulative methylmercury (MeHg) production (Hall *et al.*, 2008). Therefore, high DOC lakes that are connected to wetlands tend to have higher concentrations of MeHg.

The larger research project to which this work would contribute also identified data gaps and targeted lakes from which data has been collected in 2010 and 2011. Preliminary data indicate that coastal lakes could have higher concentrations of fish tissue Hg as well as higher concentrations of Hg and methylmercury (MeHg) in lake water; however, there are not enough coastal lakes in the dataset to confirm this statistically. This project would identify 15 candidate lakes within 30 km of the Maine coast for sampling during field season 2012. During the spring/summer of 2012, fish tissue samples will be collected for total Hg analysis. In addition, epilimnetic lake water will be collected and analyzed for Hg, MeHg, total P, base cations, inorganic anions, DOC, ANC, pH, and chlorophyll. In addition, sediment from each lake will be collected and analyzed for Hg, Fe, Al and P. We will analyze data in late 2012 and early 2013 using a number of approaches including coastal/non-coastal group comparisons, regression analysis (Hg concentration versus distance from coast), and multivariate techniques to look at coastal distance along with P and all the other variables. These results will be presented at the 2013 Maine Water Conference.

The objectives of this project were as follows:

- Determine total Hg levels in fish tissue, and total Hg and MeHg concentrations in lake water and sediments in 15 lakes located within approximately 30 km of the Maine coast;
- Determine the improvement in model predictions when a coastal proximity component is added to models for Hg contamination in fish tissue;
- Share results of the project and final Maine model with Maine Center for Disease Control (CDC) toxicologists to use at their discretion

**2.1.3** Methods. During early 2012, using the Department of Inland Fisheries and Wildlife's expertise, data, maps, and GIS, we determined which lakes within  $\leq$  30 km of the Maine coast have viable populations of warm-water and long-lived cold-water fish species. From these we identified a subset of 15 lakes to visit during 2012. Due to restricted access fish were collected from only 13 study lakes by DEP staff using gill nets and angling. We collected 10 fish from each lake to obtain a statistically representative number of samples (for example, Power analysis suggests 6-10 fish per lake suffice for t-tests and ANOVA with power>0.8) except for Lower Hadlock Pond and Somes Pond where fewer were collected.

Fish were captured by gill nets and angling. 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 were frozen. At a later date, 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  $1 \text{ cm} \times 4 \text{ cm}$  from the right 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) using Method 7473.

Lake sediment samples, water samples and physical measures were obtained during the late summer stratification period (August and early September). As part of an associated project funded by the University of Maine, Secchi transparency depth, and temperature/dissolved oxygen profiles were obtained from each lake. Water samples were collected from the epilimnion of each lake using an integrated tube sampler, lowering the device to the bottom of the epilimnion (or one meter from the lake bottom, in the case of shallow unstratified lakes) as determined by temperature/dissolved oxygen profiles. Water samples were collected for the following analytes: DOC, TP, chlorophyll A, alkalinity, anions, cations, pH, Fe and other important metals. Samples were obtained using DEP SOPs and measured using the DEP-approved techniques at the Sawyer Environmental Chemistry Laboratory at UMaine.

Total Hg and MeHg samples were obtained from subsurface water grabs immediately upon arrival at the station, while the boat is still moving forward after the engine has been stopped or paddling has ceased to minimize the risk of contamination. Water total Hg and MeHg were analyzed at Brooks Rand labs using EPA Methods 1631 and 1630, respectively. Care was taken to collect Hg water samples using established 'clean' protocols (EPA method 1669).

Three sediment samples were collected from the profundal zone of each lake (where most of P and Hg are mobilized) using a Hongve-type gravity corer, composited, and analyzed for Hg, Al, Fe, and P. Immediately after collection, sediments were frozen and stored until shortly before analysis, which is still pending. Hg will be analyzed in the top 5 cm of the sediment using the DMA80, and the latter three elements will be analyzed following a sequential chemical extraction scheme employed previously by our group (Lake *et al.*, 2007). The proposed sequential extraction procedure is modification of the procedure first employed by Psenner et al. (1984) and includes 0.11 M NaHCO<sub>3</sub> and 0.11 M NaS<sub>2</sub>O<sub>4</sub> for 0.5 hr at 40°C to extract reducible metal hydroxides and associated P followed by 1 M NaOH for 16 hr at 25°C to extract organic material, Al(OH)<sub>3(S)</sub> and associated P. This sequential extraction will be applied to the top 2 cm and 8-10 cm fractions of the sediment column. Our previous work and those of others have shown that, during summer anoxia, most of the released P is from the top 2 cm of sediment. The 8-10 cm interval is more likely the long-term net picture of processes, minus recent elevated atmospheric Hg delivery. The depths between 0-2 and 8-10 also contribute to P release. However, P distribution in these depths is likely influenced by transient changes in the redox boundary on a seasonal basis. Metals will be analyzed using the ICP-OES, and P will be analyzed colorimetrically using the molybdate blue method. Loss-on-ignition (LOI) analysis will be performed as a proxy for the sediment organic carbon content.

Results will be assembled into a database that links the water, sediment, and fish and chemistry data with landscape variables extracted from GIS data layers including geologic watershed attributes, distance to the ocean, elevation, vegetation characteristics, and aspect with respect to prevailing winds. Statistical analysis of the data using SYSTAT is likely to involve step-wise regression and principal component analysis and other standard tests as needed. The Spatial Analyst module within the GIS software ArcMap will be used to model spatial variables that contribute to fish Hg as necessary.

# 2.1.4 Results

T 11 0 1 M

Mercury concentrations in water from 2010-2012 have been measured but the data have not yet been analyzed and will be reported at a later date as part of a PhD dissertation along with other water quality and sediment data.

# 2.1.4.1 Coastal Lakes

Mercury concentrations in fish from all of the coastal lakes and ponds sampled exceeded the ME-CDC's fish tissue action level (FTAL=0.2 ug/g) (Table 2.1). Concentrations were no higher than in fish from most inland lakes and ponds for any species previously sampled, except for Round Pond, Hodgdon Pond and Seal Cove Pond, where concentrations were generally higher than in similar species inland, as was the case in the 1990s for the latter two. Concentrations were similar in brook trout (BKT) and landlocked salmon (LLS) from Echo Lake, but were significantly different between chain pickerel (PKL) and white perch (WHP) in Seal Cove Pond, and between smallmouth bass (SMB) and brook trout in Somes Pond. There were no historical data for the same species for any water except Seal Cove, where results from 2012 for PKL were not significantly different from those from the 1990s. In 2012, the mean concentration in white perch from Seal Cove Pond appeared higher than the level in 1994, but no statistical comparison can be made since there was only one white perch captured in 1994. Concentrations were of the same order of magnitude for both years however.

Table 2.1. Mercury conce	entrations in f	fish from coa	astal Maine	e Lakes fro	m 2012 and 1	990s			
LAKES			2012	1990s	2012-1990s	TREND	ANCOVA	1990s	1990s
	MIDAS	SPECIES	HG ug/g	HG ug/g	%	+,-,0	p-value	SPECIES	HG ug/g
Eagle L	4606	LKT	0.62						
Echo L	4624	BKT	0.38						
Echo L	4624	LLS	0.43						
Hamilton L	8577	LMB	0.45						
Hodgdon P	4628	PKL	1.41					SMB	1.93
Jordan P	4608	LKT	0.26						
Little Round P	4618	PKL	0.74						
Long	0477	BKT	0.18						
Long (Great) P	4622	SMB	0.67						
Lower Hadlock P	4610	BKT	0.32						
Round P	4620	SMB	1.56						
Seal Cove P	4630	PKL	0.52	0.66	-21	0	0.524		
Seal Cove P	4630	WHP	1.38	1.02	35	? n=1 1994			
Somes P	4614	SMB	0.91						
Somes P	4614	BKT	0.08						
Upper Hadlock P	4612	BKT	0.32						

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## **2.2. MERCURY TRENDS**

#### **2.1.1 Introduction**

As in 2010 and 2011, ME-CDC requested resampling of fish from lakes from DEP's REMAP study or other study with previous data, such that current mercury levels can be readily compared to historical 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 previously resampled 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 26 lakes and 10 lakes, sampled statewide in the 1990s, resampled in 2010 and 2011 respectively . In 2012, 2 of the 15 target coastal lakes that had been previously sampled were resampled. In addition 12 additional lakes, with fish mercury data at least 10 years old were resampled.

#### 2.1.2. Methods

The methods for collection and processing of fish samples were similar to that for coastal lakes described above. In addition, to evaluate any potential issue with the location and timing of biopsy sampling, biopsies from 30 white perch from Clary Lake were taken in the field immediately after fish collection and later in the lab after being frozen for several days.

#### 2.1.3. Results and Discussion

In 2012, 10 fish were collected from each lake, except for Clary Lake discussed above. Smallmouth bass were collected from 6 lakes, white perch from 5 lakes, and both species from 1 lake (Table 2.2). Mean concentrations ranged from 0.11 ug/g to 1.2 ug/g. Concentrations exceeded ME-CDC's Fish Tissue Action Level (FTAL = 0.2 ug/g) for mercury in all lakes except Lovejoy Pond and Sabattus Pond and only slightly exceeded the FTAL in Panther Pond and Webber 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 was one lake (Androscoggin Lake) where concentrations increased (denoted by a '+'), five lakes where concentrations decreased (denoted by a '-'), and six lakes where there was no change (denoted by a '0') from historical data. These findings are a bit different from those of 2011 when there were 3 lakes where concentrations were significantly higher in 2011 than in the 1990s, 2 lakes where concentrations were lower, and 2 lakes where there was no difference. The 2012 results are also different than those of 2010, when concentrations were higher in fish from 16 lakes and lower in 10 lakes than concentrations in the 1990s. Nevertheless, from all three years, there appears to be no statewide trend during the last 20 years.

Comparison of concentrations in fish from 2012 with concentrations in other species from the 1990s demonstrates some differences among species (Table 2.2). Concentrations were often relatively similar in multiple species, corroborating a lake's status as a high, medium, or low mercury lake. The data are sent to ME-CDC for use in evaluation of the statewide fish consumption advisory.

Mean mercury concentrations in biopsies taken in the field from freshly captured fish from Clary Lake and from frozen fish in the lab were not significantly different when compared by a two-tailed t-test with log transformed data (p=0.069). However, the mean concentration of fresh biopsies was significantly lower (~10%) than the mean concentration of frozen lab biopsies in a one-tailed t-test with log transformed data (p=0.034) (Table 2.2). This was somewhat unexpected as a study in Massachusetts indicated a potential for contamination and desiccation when taking biopsies in the field, both of which could result in higher concentrations from field collected biopsies. Nevertheless, there is still a possibility that desiccation during freezing is significant. In a previous study in 2010, we found an average 5% higher concentration calculated from frozen biopsies compared to frozen biopsies whose weight had been adjusted to account for losses due to desiccation, loss of tissue during handling, and use of different balances. In the same fish, the average concentration of biopsies was only 1.8% higher than the average concentration of fish filets analyzed at a commercial lab, both of which had been frozen. Although the differences in the 2010 study were not considered significant, the latest results suggest that sampling time and place may be an issue and that the issue should be revisited. We will propose to compare field and lab collected biopsies again this year.

Table 2.2. Mercury trends	in fish from	some Maine	e Lakes fro	om 2012 at	nd 1990s				
LAKES			2012	1990s	2012-1990s	TREND	ANCOVA	1990s	1990s
	MIDAS	SPECIES	HG ug/g	HG ug/g	%	+,-,0	p-value	SPECIES	HG ug/g
Anasagunticook L	3604	SMB	1.2	0.61	90	0	0.268		
Androscoggin L	3836	WHP	0.77	0.39	97	+	0.000	SMB, PKL	0.71, 0.71
Clary L field	5382	WHP	0.38						
Clary L frozen lab	5382	WHP	0.43	0.36	19	0	0.163		
Lovejoy P	5176	WHP	0.16	0.42	-62	-	0.000		
Moose P	3134	SMB	0.45	0.90	-50	-	0.000	WHP	0.43
Panther P	3694	SMB	0.21	0.72	-71	-	0.001	WHP	0.62
Pemadumcook L	0982	WHP	0.82	0.74	11	0	0.494		
Pitcher P	4848	SMB	0.56	0.70	-20	-	0.000		
Round P	3818	SMB	0.64	0.39	64	0	0.707		
Round P	3818	WHP	0.64						
Sabattus P	3796	WHP	0.11	0.14	-21	-	0.016	NPK	0.14
South Branch L	2144	SMB	0.43	0.47	-9	0	0.918		
Webber P	5408	SMB	0.21	0.58	-64	0	0.907	WHP	0.24
ANCOVA with length as o	ovariate								

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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 2012, we evaluated the condition of 44 sample locations, primarily in the Kennebec 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 2012 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 2012, where available, is given in Table 3.1.4.

For more information about the Biological Monitoring Unit, please e-mail us at <u>biome@maine.gov</u> or visit our web site: <u>http://www.maine.gov/dep/water/monitoring/biomonitoring/index.htm</u>. 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 2012 in the Kennebec River basin. Forty-four stations were sampled under the SWAT Program (Table 3.1.1).

All stations have been analyzed for aquatic life attainment with thirty-five stations in attainment of their statutory class. Twelve of these stations exceeded their assigned class. One station was called Indeterminate due to low numbers. No licensing/relicensing issues have been found in waterbodies sampled below municipalities or industries. The streams that did not attain their statutory class were located on small urban, residential, and rural systems; summaries on these streams are found below.

#### Bond Brook – Unnamed Tributary – Augusta Station 489

The Unnamed Tributary to Bond Brook is a first order stream with a water quality goal of Class B. It originates on the western side of the Kennebec River in a highly developed area and the stream is crossed by I-95 before it enters Bond Brook. The benthic macroinvertebrate community is comprised mainly by tolerant midges and worms. Very few sensitive organisms are present in the community. The stream did not meet minimum Class C criteria for aquatic life. The specific conductance (Table 3.1.2) in the stream was very high and the stream banks were highly eroded. The stream has been listed on the 303(d) list and is part of a percent impervious cover TMDL.

#### Fish Brook – Fairfield Station 422

Fish Brook is a small first order stream in Fairfield which flows south adjacent to an old landfill site and then flows through a concentrated agricultural area before entering Messalonskee Stream above Waterville. Its water quality goal is Class B. Fish Brook did not attain the minimum Class C aquatic life criteria. The aquatic macroinvertebrate community consisted of a low number of organisms with Total Mean Abundance and Generic Richness barely meeting minimum criteria. Sensitive taxa were present in very low numbers. The dominant organisms in the community were made up of tolerant midges and worms indicating a possible toxic response to a stressor. Specific conductance (Table 3.1.2) was high and the water was very cloudy during deployment and retrieval. In addition, there was beaver activity noted upstream which may have contributed to the high total suspended and dissolved solids (Table 3.1.4). Station 422 failed to attain the minimum Class C aquatic life criteria in 2000 and 2001 (Table 3.1.3).

#### 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 and during sample retrieval a considerable amount of filamentous algae was observed. The macroinvertebrate community was

diverse with Richness counts totaling 70 different genera. However, the macroinvertebrate community showed signs of enrichment with the Total Mean Abundance of the sample totaling over 1300 individuals. 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. In addition, the Nitrate + Nitrite as N level was high at .84 mg/l (Table 3.1.3) indicating possible runoff from the agricultural fields. Although the macroinvertebrate community appears to be border line Class A this high quality resource should be observed and evaluated in the near future. Agricultural Best Management Practices may be required in order to maintain a Class A aquatic life community.

#### Riggs Brook – Augusta Station 599

Riggs Brook is a second order stream with a water quality goal of Class B. The stream flows north through a large agricultural and residential area before entering the east side of the Kennebec River. Riggs Brook does not attain the Class B aquatic life criteria. The stream was greatly enriched with filamentous algae covering the bottom and the Total Mean Abundance of the sample totaling 1604 organisms. There were very few sensitive organisms present although the Richness count totaled over 50 different genera. The most dominant taxa in the sample consisted of tolerant collector-filterers. Total Phosphorus was .043 mg/l (Table 3.1.3) indicating possible runoff from the agricultural and residential area surrounding the watershed.

#### Whitney Brook – Augusta Station 601

Whitney Brook is a small, first order stream that flows through a heavily developed residential area on the east side of the Kennebec River before entering the Kennebec River near the Civies Steel site. Its water quality goal is Class B. Whitney Brook did not attain the minimum Class C criteria for aquatic life. There were very few sensitive organisms present in the community. The dominant organism was the tolerant Caecidotea (Isopods) which made up 41% of the community. Specific conductance (Table 3.1.2) was very high and the water had a milky appearance which is common in an urban system. The stream banks were eroded and the stream channel widened indicating storm water surges. The stream has been listed on the 303(d) list and is part of a percent impervious cover TMDL. The stream did not attain the minimum Class C criteria in 2007 (Table 3.1.4).

Waterbody	Town	Station	Log	Potential sources of pollution <sup>1</sup>	Statutory Class/ Final Determina- tion	Attains Class? <sup>2</sup>	Probable Cause
Aroostook River	Caribou	370	2133	Agricultural NPS	C/A	Yes	
Aroostook River	Presque Isle	594	2134	Agricultural NPS	B/A	Yes	
Aroostook River	Presque Isle	595	2135	Agricultural NPS	C/A	Yes	
Bond Brook	Augusta	30	2092	Urban NPS	B/B/	Yes	
Bond Brook	Augusta	597	2093	Urban NPS	B/B/	Yes	
Bond Brook - Unnamed Tributary	Augusta	489	2095	Urban NPS	B/NA/	No	NPS Toxics; Habitat
Burgess Brook	Lily Bay TWP	834	2127	Reference	A//A	Yes	
Burgess Brook	Lily Bay TWP	835	2128	Reference	A/B	No	Low DO (natural)
Burnham Brook	Big Moose TWP	869	2132	Reference	A/A	Yes	
Carlton Stream	Тгоу	839	2124	NPS	B//C	No	Community response to upstream Carleton Bog
Carrabassett River	Kingfield	16	2117	Municipal	A/A	Yes	
Carrying Place Stream	Carrying Place TWP	768	2118	Forestry	A/A	Yes	
China Lake Outlet Stream	Winslow	604	2102	NPS	B/B	Yes	
Ducktrap River	Lincolnville	626	2096	Reference	AA/A	Yes	
East Branch Sebasticook River	Corinna	194	2115	In-Place Contamination	C/C	Yes	
East Branch Wesserunsett Stream	Athens	486	2113	Reference	B/A	Yes	
Fish Brook	Fairfield	422	2103	Agricultural NPS; In-Place Contamination	B/NA	No	Agricultural runoff; Habitat; In-Place Contamination?
Halfmoon Stream	Thorndike	697	2126	Agricultural NPS	A/B	No	Agricultural runoff
Hardy Brook	Madrid TWP	769	2120	Forestry	A//A	Yes	
Higgins Brook	Wellington	767	2114	Reference	B/A	Yes	
Kennebec River	Madison	22	2112	Municipal / Industrial	B/A	Yes	
Kennebec River	Benton	196	2107	Municipal / Industrial	C/A	Yes	
Kennebec River	Bingham	636	2119	Municipal	A/A/	Yes	

# Table 3.1.1. 2012 SWAT Benthic Macroinvertebrate Biomonitoring Results

Kennedy Brook	Augusta	620	2100	Urban NPS	B/C/	No	Samplers disturbed.
							Resample

<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

Waterbody	Town	Station	Log	Potential sources of pollution <sup>1</sup>	Statutory Class/ Final Determina- tion	Attains Class? <sup>2</sup>	Probable Cause
Lily Bay Brook	Lily Bay TWP	844	2129	Reference	A/A	Yes	
Madawaska Brook	Palmyra	637	2108	NPS	B/C	No	Community response to upstream Madawaska Bog
Martin Stream (Dixmont)	Dixmont	756	2125	Agricultural NPS	A/A	Yes	
North Brook	Lily Bay TWP	841	2130	Reference	A/A	Yes	
Orbeton Stream	Madrid TWP	840	2121	Reference	A//A	Yes	
Passagassawakeag River	Belfast	430	2097	NPS	B/B	Yes	
Riggs Brook	Augusta	599	2099	Agricultural NPS	B/C	No	Agricultural Runoff; Landfill
Sandy River	Phillips	17	2122	Reference	AA/	Yes	
Sandy River	Farmington	572	2110	Municipal / Agric NPS	B/A	Yes	
Sandy River	New Sharon	617	2111	Municipal / Agric NPS	B/A	Yes	
Sebasticook River	Benton	299	2106	Municipal / NPS	C/B	Yes	
Sheepscot River	Whitefield	74	2089	Long Term Monitoring	AA/A	Yes	
South Branch Carrabassett River	Carrabassett Valley	836	2116	NPS/Ski Area	AA/A	Yes	
Stone Brook	Augusta	944	2094	Urban NPS	B/B	Yes	
Togus Stream	Pittston	612	2101	Municipal / NPS	B/A	Yes	
Unnamed Stream (Big Moose TWP)	Big Moose TWP	870	2131	Reference	A/I	Indeterm inate	Low Numbers due to beaver activity.
West Branch Sebasticook River	Pittsfield	27	2109	Municipal / Industrial	C/B	Yes	
West Branch Sheepscot River	China	268	2088	Long Term Monitoring	AA/A	Yes	
Whitney Brook	Augusta	601	2098	Urban NPS	B/NA	No	NPS Toxics; Habitat
Wilson Stream	Wilton	34	2123	NPS	C/A/	Yes	

Table 3.1.1. 2012 SWAT	<b>Benthic Macroinve</b>	rtebrate Biomonit	oring Results	(continued)
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<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. 2012 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	pН	Date	Temp	DO	SPC	pН
				Deg C	mg/L	uS/cm	STU		Dec C	mg/L	uS/cm	STU
Aroostook River	370	2133	7/30/12	28.8	12.2	177	9.22	8/28/12	22.1	8.2	167	8.10
Aroostook River	594	2134	7/31/12	25.6	8.6	102	7.67	8/27/12	27.7	8.5	95	8.20
Aroostook River	595	2135	7/30/12	29.0	12.1	156	9.01	8/27/12	26.9	10.2	122	8.78
Bond Brook	30	2092	7/9/12	20.2	9.3	195	8.23	8/7/12	22.0	8.5	217	8.06
Bond Brook	597	2093	7/9/12	21.4	9.4	279	8.00	8/7/12	20.7	9.3	324	8.01
Bond Brook -Unnamed Tributary	489	2095	7/9/12	15.6	8.4	1175	7.63	8/7/12	16.5	8.3	1361	7.87
Burgess Brook	834	2127	7/25/12	17.0	8.0	59	6.65	8/23/12	16.2	8.5	31	6.72
Burgess Brook	835	2128	7/25/12	21.0	4.3	64	5.84	8/23/12	18.7	5.5	24	5.97
Burnham Brook	869	2132	7/25/12	23.8	7.7	115	7.56	8/23/12	21.0	7.8	115	6.37
Carlton Stream	839	2124	7/19/12	25.7	7.1	68	6.85	8/22/12	24.6	8.1	75	7.48
Carrabassett River	16	2117	7/24/12	23.8	7.9	78	7.26	8/20/12	18.3	9.2	75	7.58
Carrying Place Stream	768	2118	7/24/12	19.1	7.1	68	7.17	8/20/12	18.2	8.9	25	7.35
China Lake Outlet Stream	604	2102	7/12/12	21.7	8.6	113	7.56	8/13/12	22.8	6.2	141	6.80
Ducktrap River	626	2096	7/10/12	20.2	7.7	46	6.81	8/8/12	20.7	6.8	85	6.45
East Branch Sebasticook River	194	2115	7/18/12	27.3	7.2	173	7.57	8/16/12	23.2	6.7	185	8.03
East Branch Wesserunsett Stream	486	2113	7/18/12	21.6	8.0	92	7.67	8/16/12	19.1	8.2	85	8.24
Fish Brook	422	2103	7/12/12	19.1	8.0	345	7.58	8/13/12	19.0	8.0	380	7.87
Halfmoon Stream	697	2126	7/19/12	18.4	9.6	136	7.76	8/22/12	19.1	9.2	158	6.51
Hardy Brook	769	2120	7/23/12	20.3	8.6	55	7.47	8/21/12	16.6	8.7	57	7.38
Higgins Brook	767	2114	7/18/12	22.6	8.9	79	7.77	8/16/12	18.3	8.3	78	7.08

Temp = water temperature, DO = dissolved oxygen, SPC = specific conductance, pH= hydrogen ion concentration.
# Table 3.1.2. 2012 SWAT Field Data (continued)

Site	Station	Log	Sampler Deployment				Sampler Retrieval					
			Date	Temp	DO	SPC	pН	Date	Temp	DO	SPC	pН
				Dec C	mg/L	uS/cm	STU		Dec C	mg/L	uS/cm	STU
Kennebec River	22	2112	7/17/12	23.6	8.1	48	6.89	8/15/12	23.4	8.3	68	7.44
Kennebec River	196	2107	7/16/12	27.8	9.1	78	7.41	8/14/12	24.7	7.4	73	7.20
Kennebec River	636	2119	7/24/12	21.5	8.1	60	6.61	8/20/12	19.0	8.3	47	6.95
Kennedy Brook	620	2100	7/11/12	16.9	8.6	636	7.84	8/9/12	17.8	8.9	713	8.92
Lily Bay Brook	844	2129	7/25/12	14.4	9.3	59	7.00	8/23/12	15.6	8.9	27	6.83
Madawaska Brook	637	2108	7/16/12	27.1	6.4	143	7.18	8/14/12	22.5	6.3	188	7.56
Martin Stream (Dixmont)	756	2125	7/19/12	22.2	8.7	138	7.85	8/22/12	21.4	9.7	162	7.83
North Brook	841	2130	7/25/12	17.0	8.9	70	6.78	8/23/12	15.7	8.9	62	7.86
Orbeton Stream	840	2121	7/23/12	18.1	8.9	49	7.32	8/21/12	19.0	8.4	55	7.44
Passagassawakeag River	430	2097	7/10/12	22.1	8.4	52	7.54	8/8/12	23.8	8.3	90	7.26
Riggs Brook	599	2099	7/11/12	23.8	8.4	137	7.77	8/9/12	24.6	8.4	251	7.52
Sandy River	17	2122	7/23/12	21.5	8.8	50	7.26	8/21/12	20.8	8.8	32	7.34
Sandy River	572	2110	7/17/12	24.8	7.6	74	6.97	8/15/12	22.6	7.9	76	6.76
Sandy River	617	2111	7/17/12	25.7	7.6	79	7.22	8/15/12	23.6	7.6	81	7.10
Sebasticook River	299	2106	7/16/12	28.0	8.4	101	6.76	8/14/12	27.2	7.9	99	7.79
Sheepscot River	74	2089	7/5/12	23.3	7.7	46	6.99	8/6/12	25.3	6.8	106	7.11
South Branch Carrabassett River	836	2116	7/24/12	19.7	8.3	54	6.99	8/20/12	17.8	8.4	56	6.73
Stone Brook	944	2094	7/9/12	20.9	8.2	332	7.88	8/7/12	23.3	7.4	585	7.88
Togus Stream	612	2101	7/11/12	23.2	8.6	71	7.08	8/9/12	26.0	8.5	121	7.92
Unnamed Stream (Big Moose Twp)	870	2131	7/25/12	18.1	7.3	66	7.23	8/23/12	16.1	8.6	74	6.85
West Branch Sebasticook River	27	2109	7/16/12	27.6	7.6	88	6.72	8/14/12	26.9	11.3	129	7.74
West Branch Sheepscot River	268	2088	7/5/12	22.6	8.2	63	7.49	8/6/12	22.1	7.8	118	7.41
Whitney Brook	601	2098	7/11/12	18.6	8.7	488	7.6	8/9/12	19.4	7.4	667	7.41
Wilson Stream	34	2123	7/23/12	23.7	8.2	61	7.08	8/21/12	20.6	7.8	57	6.75

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

#### Table 3.1.3. 2012 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
Aroostook River	370	2133	8/28/2012	4.5	0.02	0.3	0.31	4	0.024	<2	90
Aroostook River	594	2134	8/27/2012			0.3	0.02	1	0.015		
Aroostook River	595	2135	8/27/2012	5.1	0.01	0.4	0.09	1	0.020	<2	68
Bond Brook	30	2092	8/7/2012	2.3	0.01	0.2	< 0.01	2	0.012	2.1	120
Bond Brook – Unnamed Tributary	489	2095	8/7/2012	2.8	0.02	0.3	0.64	5	0.008	<2	940
Carlton Stream	839	2124	8/22/2012	10.0	0.01	0.6	0.02	3	0.015	<2	62
Carrabassett River	16	2117	8/20/2012	4.0	< 0.01	0.2	0.04	2	0.005	<2	44
Carrying Place Stream	768	2118	8/20/2012	6.4	< 0.01	0.3	0.03	2	0.009	<2	37
Fish Brook	422	2103	8/13/2012	2.5	0.02	0.3	0.17	6	0.034	9.1	230
Halfmoon Stream	697	2126	8/22/2012	2.8	0.01	0.2	0.84	3	0.015	<2	79
Kennebec River	196	2107	8/14/2012	5.1	0.02	0.3	0.10	12	0.027	<2	44
Kennedy Brook	620	2100	8/9/2012	1.7	0.01	0.1	0.51	10	0.012	<2	450
Martin Stream (Dixmont)	756	2125	8/22/2012	5.5	0.01	0.3	0.03	2	0.017	<2	91
Passagassawakeag River	430	2097	8/8/2012	4.6	0.01	0.3	0.07	5	0.024	<2	50
Riggs Brook	599	2099	8/9/2012	6.7	0.02	0.5	0.09	10	0.043	<2	130
Sandy River	617	2111	8/15/2012	4.2	0.02	0.3	0.07	2	0.017	2.3	50
Sandy River	17	2122	8/21/2012	2.9	< 0.01	0.2	0.05	3	0.005	<2	40
Sheepscot River	74	2089	8/6/2012	4.9	0.01	0.4	0.01	2	0.012	<2	54
Stone Brook	944	2094	8/7/2012	4.4	0.01	0.4	0.07	3	0.013	<2	360
West Branch Sebasticook River	27	2109	8/14/2012	5.3	0.01	0.4	0.04	2	0.013	<2	59
West Branch Sheepscot River	268	2088	8/6/2012	4.8	0.01	0.4	0.10	2	0.013	<2	63

 $DOC = dissolved organic carbon, NH_3-N = ammonia-nitrogen, TKN = total Kjeldahl-nitrogen, NO_2-NO_3-N = nitrite-nitrate-nitrogen, SRP = soluble reactive phosphorus (ortho-phosphate), Total P = total phosphorus, TSS = total suspended solids, TDS = total dissolved solids, "<" = constituent not detected at the reporting limit.$ 



#### Figure 3.1.1. 2012 In-Stream Continuous Temperature Data Please note: all data are in degrees Celsius











# 3.1.3 Attainment History of Sampling Stations prior to 2012

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	
Aroostook River	370	1999, 2009			
Aroostook River	594	2001, 2009			
Aroostook River	595	2001, 2009			
Bond Brook	30	1983, 1991, 1992, 1997, 2002, 2007			
Bond Brook	597	2002, 2007			
Bond Brook – Unnamed Tributary	489		2001, 2002, 2007		
Burgess Brook	834	2007			
Burgess Brook	835		2007		
Burnham Brook	869		2008		
Carlton Stream	839		2007		
Carrabassett River	16	1983, 1997, 2007			
Carrying Place Stream	768	2004, 2007			
China Lake Outlet Stream	604	2002, 2007			
Ducktrap River	626	2007	2002		
East Branch Sebasticook River	194	1993, 1997, 2003, 2007	1994		
East Branch Wesserunsett Stream	486	2001, 2007			
Fish Brook	422		2000, 2001		
Halfmoon Stream	697	2003, 2007			
Hardy Brook	769	2004, 2007			
Higgins Brook	767	2004			
Kennebec River	22	1983, 1997, 2007			
Kennebec River	196	1992, 1997, 2007			
Kennebec River	636	2002, 2007			
Kennedy Brook	620	2002	2004, 2007		
Lily Bay Brook	844	2007			
Madawaska Brook	637		2002		
Martin Stream (Dixmont)	756		2005, 2006, 2007	2004	
Orbeton Stream	840	2007			
Passagassawakeag River	430	2000, 2007			
Riggs Brook	599		2007		

Waterbody	Station	Attained Class	Did not Attain Class	Indeterminate Result	
Sandy River	17	1983, 2000, 2007			
Sandy River	572	2007	2000		
Sandy River	617	2007			
Sebasticook River	299	1997, 2002, 2007			
Sheepscot River	74	1987-1990, 1992, 1995, 1996, 1998-2011	1984-1986, 1991, 1993, 1994, 1997		
South Branch Carrabassett River	836	2007			
Togus Stream	612	2002, 2007			
Unnamed Stream (Big Moose TWP)	870	2008			
West Branch Sebasticook River	27	1983, 2002, 2007			
West Branch Sheepscot River	268	1996-1999, 2001, 2002, 2005, 2007, 2009-2011	2000, 2003, 2004, 2006, 2008	1995 (low numbers)	
Whitney Brook	601		2007		
Wilson Stream	34	1983, 1991, 1997, 2007			

# Table 3.1.4. Past Attainment History (continued)

# **3.2 FISH CONSUMPTION ADVISORIES**

#### 3.2.1 Dioxin and coplanar 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 2012 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 are 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 pptr, 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 and 2012.

# 2012 monitoring requested by Maine Center for Disease Control and Prevention

Suggestions for fish sampling were 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 (ME-CDC) (provided at http://www.maine.gov/dhhs/eohp/fish/index.htm). For dioxins (DTE) and dioxin-like PCB congeners (CTE), a fish tissue level of 3.0 parts per trillion (ppt) was used to evaluate the need for additional data. This value reflects the level of dioxins and dioxin-like compounds, normalized to dioxin toxic equivalents (TEQ = DTE + CTE) that can be consumed twice per month without exceeding the cancer-based FTAL of 1.5 ppt for this class of chemicals. 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. Therefore, 3 ppt is the concentration threshold corresponding to the current state-wide mercury advice for species other than brook trout and landlocked salmon (no consumption for sensitive populations; 2 meals per month for the general population). Brook trout and landlocked salmon have not been species of interest on rivers where dioxin sampling is performed. This value is referred to as the mercury Statewide Advisory Dioxin Equivalent Threshold (SADET= 3 pg/g) in this report.

ME-CDC has also developed a provisional FTAL for dioxins and dioxin-like compounds for noncancer effects applicable to sensitive populations, which considers background dietary exposure (0.4 ppt). 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 . ME-CDC is aware that US EPA has recently updated the toxicity profile for dioxin in IRIS for 2,3,7,8-TCDD and adopted a reference dose of  $0.7 \times 1^{-9}$  mg/kg-day. This value is slightly lower than the reference dose used by Maine as the basis for the current non-cancer FTAL (1 x 1<sup>-9</sup> mg/kg-day), and more importantly provides a much stronger scientific basis for this endpoint. However, ME-CDC does not intend to modify the non-cancer FTAL for dioxins until it has 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. As the statewide mercury consumption advisory restricts women of child bearing age and children under age 8 for 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 noncancer effects is not expected to impact fish advisories.

In reviewing the most recent fish tissue data, ME-CDC has identified a few stations where total TEQ (or dioxinconcentrations nearly equal or slightly exceed 3 ppt, the concentration that would result in a consumption advisory more restrictive than the current advisory. In addition, recent data have demonstrated that fish tissue concentrations at several stations which were near the criteria have decreased. EPA guidance recommends that if contamination levels fall below decision criteria, they be confirmed in a subsequent sampling round. Recommendations for sampling for 2012 were focused on further characterization of fish sampling stations that previously had borderline total TEQ concentrations, to support upcoming revisions to the fish advisories.

# **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. The requests are listed in order of preference.

# 1. Dioxin Data for the Major Rivers

Recommendations for further sampling for dioxins are provided below, by river. As in 2011, composite samples, with two samples (of 5 fish each) being submitted for chemical analysis for any sampling location - fish species combination, are recommended.

# Androscoggin River

#### Gilead

Brook trout. While there are no data for brook trout in the major rivers, brook trout are commonly caught in some, most notably the Androscoggin River at Gilead, where concentrations of dioxins

have been historically elevated in brown trout and rainbow trout. Brook trout will be collected at Gilead for dioxin analysis.

White sucker. Dioxin DTE in white suckers at Gilead, decreased substantially between 2007 and 2008, from approximately 2.4 ppt to approximately 1.3 ppt, respectively. ME-CDC would like to confirm this decrease with the 2012 sampling effort.

# Rumford Point

Smallmouth bass. The 2011 sampling of smallmouth bass at Rumford Point determined total TEQ to be approximately 1.1 ppt (dioxin DTE approximately 0.8 ppt, PCB CTE approximately 0.3 ppt). This is a decrease from previous values (2004 total TEQ of 2.98 ppt). ME-CDC would like to confirm this result with a subsequent sampling round.

#### Riley

White sucker. Previous values for total TEQ in white sucker at Riley, estimated in 2007, were approximately 3.01 ppt. The 2011 sampling effort resulted in total TEQ of approximately 2.3 ppt (dioxin DTE approximately 1.8 ppt, PCB CTE approximately 0.5 ppt). ME-CDC would like to confirm this decrease with the 2012 sampling.

#### Livermore Falls

White sucker. Total TEQ in white sucker at Livermore Falls estimated from the 2007 sample collection was approximately 3.47 ppt. The 2011 sampling effort resulted in total TEQ of approximately 2.4 ppt (dioxin DTE approximately 1.9 ppt, PCB CTE approximately 0.5 ppt). ME-CDC would like to confirm this decrease with the 2012 sampling.

#### Kennebec at Augusta or Sidney

White sucker. Investigation of dioxins in white suckers collected in 1995 determined total TEQ to be approximately 5 ppt, the dioxin DTE approximately 3 ppt. A PCB CTE was determined in 2009 (0.5-0.6 ppt), however no corresponding dioxin analysis was conducted. The 2011 sampling resulted in a total TEQ of approximately 0.4 ppt. (dioxin DTE approximately 0.16 ppt, PCB CTE approximately 0.2 ppt). ME-CDC would like to confirm this with a subsequent sampling round.

#### Sebasticook River in Newport

White sucker. Based on data collected on 2010, the total TEQ concentration for white sucker at Newport slightly exceeded 3 ppt (3.08 ppt). The 2011 sampling effort resulted in a total TEQ of approximately 2.1 ppt (dioxin DTE approximately 1.3 ppt, PCB CTE approximately 0.8 ppt). ME-CDC would like to confirm this decrease with the 2012 sampling effort.

**Penobscot River:** No dioxin data were requested at this time. **St. Croix River:** No dioxin data were requested at this time. **Presumpscot River:** No dioxin were requested at this time. **Salmon Falls River:** No dioxin were requested at this time. **Androscoggin Lake:** No dioxin were requested at this time.

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

Recommendations for sampling for PCBs are provided below, by river. As in 2011, 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**: As previously stated, stations and fish species that have had total TEQs at or above 3 ppt include:

- (1) Brook trout PCB data for Gilead do not exist and are requested.
- (2) Smallmouth bass at Rumford Point (2004 total TEQ of 2.98 ppt, 2011 total TEQ approximately 2.4);

(3) White sucker at Gilead, (total TEQ approximately 2.8 ppt in 2007, approximately 1.7 ppt in 2008;

(4) White sucker at Livermore Falls (2007 and 2011 total TEQ 3.47 and 2.4 ppt respectively);

(5) White sucker at Riley (2007 and 2011 total TEQ 3.01 and 2.3 ppt respectively).

The contribution of dioxin-like PCBs to the total TEQ is 30% or less at each of these stations. Data for dioxin-like PCB congeners for the identified species at each of these stations is requested to confirm that total TEQ concentrations remain stable or have further decreased.

**Kennebec River:** As stated above, previous data for dioxins in white suckers were collected in 1995, with total TEQ approximately 5 ppt, dioxin DTE approximately 3 ppt. In 2009, PCB CTE was approximately 0.5-0.6 ppt, however no corresponding dioxin data was collected. The 2011 sampling resulted in a total TEQ of approximately 0.4 ppt. (dioxin DTE approximately 0.16 ppt, PCB CTE approximately 0.2 ppt). ME-CDC would like to confirm this with a subsequent sampling round. Because both PCB and dioxin data are required to calculate the total TEQ, ME-CDC requests that additional sampling be conducted to confirm the decrease in total TEQ.

**Sebasticook River**: As stated above, the total TEQ concentration for white sucker at Newport, based on data collected in 2010, slightly exceeded 3 ppt (3.1 ppt). The 2011 sampling effort resulted in a total TEQ of approximately 2.1 ppt (dioxin DTE approximately 1.3 ppt, PCB CTE approximately 0.8 ppt). ME-CDC would like to confirm this decrease with the 2012 sampling effort.

**St. Croix River:** No PCB data are requested at this time. **Penobscot River:** No PCB data are requested at this time. **Presumpscot River:** No PCB data are requested at this time. **Salmon Falls River:** No PCB data are requested at this time. **Androscoggin Lake:** No PCB data are requested at this time.

#### 3.2.1.3 Sampling Plan

In 2012 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.

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.

2012 Dioxin and Cop	blanar PCB Sample Wo	orkplan			
RIVER	STATION	DMP	DMP	SWAT	SWAT
		PCDD/F	facility	PCDD/F	CPCB
ANDROSCOGGIN	GILEAD			2C5 BKT	2C5 BKT
				2C5 WHS	2C5 WHS
	RUMFORD POINT			2C5 SMB	2C5 SMB
	RILEY	2C5 WHS	RUMFORD PAPER CO		2C5 WHS
	LIVERMORE	2C5 WHS	VERSO PAPER CO		2C5 WHS
KENNEBEC	SIDNEY	2C5 WHS	SD WARREN SOMERSET		2C5 WHS
SEBASTICOOK					
EAST BRANCH	NEWPORT			2C5 WHS	2C5 WHS
2C5 = 2 composites	of 5 fish each				
BKT = brook trout, S	MB= smallmouth bass,	WHS= white	e sucker		
DMP = Dioxin Monito	ring Program funds,				
SWAT = SWAT prog	ram funds				
PCDD/F= dioxins and	d furans,				
CPCB = coplanar PC	Bs				

#### 3.2.1.4 Results and Discussion

#### Androscoggin River

#### Gilead

For the first time at this station, a total of ten brook trout (BKT) were successfully collected from the Androscoggin River at Gilead (AGL). 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 below the ME-CDC FTAL (0.4 pg/g) for dioxin-like compounds and a statewide advisory dioxin equivalent threshold (SADET= 3 pg/g) for mercury (Figure 3.2.1). Concentrations of coplanar PCB toxic equivalents (CTEhu, calculated at the 95<sup>th</sup> upper confidence level with non-detects at  $\frac{1}{2}$  of the detection limit) added only a small amount to total dioxin-like compounds. Concentrations of both DTEhu and CTEhu are well below those from previous years for rainbow trout, consistent with a trend of declining concentrations since the late 1990s.



# Rumford Point

A total of ten smallmouth bass (SMB) were successfully collected from the Androscoggin River at Rumford Point (ARP). 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  $\frac{1}{2}$  of the detection limit) did exceed the FTAL alone and the combination of the two groups of contaminants increased the exceedance of the FTAL. Concentrations of both DTEhu and CTEhu were generally similar to those of 2011, well below those from 1995 and slightly lower than those of most recent years.



A total of ten white sucker (WHS) were collected from the Androscoggin River at Rumford Point (ARP). The DTEhu concentration slightly exceeded the FTAL for dioxin-like compounds, but was well below the SADET (Figure 3.2.3). The CTEhu concentration added a small amount to the exceedance of the FTAL. Concentrations of both DTEhu and CTEhu are well below those from previous years continuing the trend of declining concentrations since the 1990s.



# <u>Riley</u>

A total of ten white sucker (WHS) were collected from the Androscoggin River at Riley (ARY). The DTEhu concentration exceeded the FTAL for dioxin-like compounds, but was below the SADET (Figure 3.2.4). The CTEhu concentration did exceed the FTAL alone and the combination of the two groups of contaminants exceeded the SADET. Concentrations of both DTEhu and CTEhu are well below those from 1997 and generally similar to those of since 2006.



#### Livermore

A total of ten white sucker (WHS) were collected from the Androscoggin River at Livermore (ALV). The DTEhu concentration exceeded the FTAL for dioxin-like compounds, but was below the SADET (Figure 3.2.5). The CTEhu concentration did not exceed the FTAL alone but the combination of the two groups of contaminants slightly increased the exceedance of the FTAL. Concentrations of both DTEhu and CTEhu were lower than those of all previous years.



#### Kennebec River

#### Waterville

A total of ten American shad (AMS) were collected from the Kennebec River at Waterville (KWV). Both roe (gonad) of females and filet (muscle tissue) of both males and females were analyzed. The DTEhu concentration in filet exceeded the FTAL for dioxin-like compounds whereas the concentration in roe did not (Figure 3.2.6). The CTEhu concentration in filets did exceed the FTAL alone; the concentration in roe did not exceed the FTAL alone, but the combination of the two groups of contaminants resulted in a slight exceedance of the FTAL. The combination of both DTEhu and CTEhu were well below the SADET.



#### Sidney

A total of ten white sucker (WHS) were collected from the Kennebec River at Sidney (KSD). The DTEhu concentration was below the FTAL for dioxin-like compounds (Figure 3.2.7). The CTEhu concentration did not exceed the FTAL alone but the combination of the two groups of contaminants resulted in a slight exceedance of the FTAL although remained well below the SADET. Concentrations of both DTEhu and CTEhu were similar to those of 2011 and well below those from 1994 and 1995.



#### Sebasticook Lake

A total of ten white sucker (WHS) were collected from Sebasticook Lake at Newport (SLN). The lake is approximately 3 miles downstream of a former textile mill and Superfund site on the East Branch of the Sebasticook River in Corinna and a half mile below an historical dioxin sample station (SEN) and popular fishing spot at the County Road bridge. The DTEhu concentration exceeded the FTAL for dioxin-like compounds, but was below the SADET (Figure 3.2.8). The CTEhu concentration did exceed the FTAL alone and the combination of the two groups of contaminants exceeded the SADET. The sum of both DTEhu and CTEhu was intermediate those of the only two previous years.



# 3.2.2 Total PCBs

# **3.2.2.1 Introduction**

ME-CDC's requests were discussed in the section above regarding coplanar PCBs.

#### 3.2.2.2 Results and Discussion

#### Androscoggin River

The mean total PCB (TPCB) concentration in brook trout from Gilead were below the ME-CDC's FTAL of 11 ng/g in 2012 and lower than concentrations in rainbow trout and brown trout from previous years (Table 3.2.1). Mean and maximum TPCB concentrations in smallmouth bass (SMB) at Rumford Point (ARP), below Berlin, NH, exceeded the FTAL in 2012 similar to those in 2009 and 2011; mean and maximum concentrations in white sucker were intermediate those of previous years but still exceeded the FTAL. Mean and maximum concentrations of TPCB in white sucker (WHS) from Riley (ARY), below Rumford, also exceeded the FTAL although were lower than in 2010 and 2011 all of which were higher than previous years. Mean and maximum concentrations of TPCB in white sucker (WHS) from Livermore (ALV), below Jay, also exceeded the FTAL but was much lower than 2009 and 2011 similar to those from earlier years and much lower than at Riley. The variation in concentrations within species among all years may reflect differences among the four different labs that were used, although all data meet quality assurance and control objectives, or simply the natural variation in individuals and condition among years.

Table 3.2	.1. Total F	PCBs in fish	n from the And	oscoggin Riv	ver, ng/g. mea	an and (max	value where	n=2 or 95th u	pper confiden	ce level wher
Year	Species	Gilead	Rumford Pt	Rumford	Riley	Jay	Livermore	Livermore FIs	AUBURN GIP	Lisbon
		AGL	ARP	ARF	ARÝ	ARJ	ALV	ALF	AGI	ALS
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)
2000	SMB		10 (11)	21 (27)	15 (17)		38 (42)	27 (32)	29 (36)	52 (60)
2001	SMB									
2002	SMB		101	22	18		18		22	17
2003	SMB						22	19		
2008	SMB								30 (35)	
2009	SMB		51 (65)						21 (24)	
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			
2000	WHS						48	42		
2001	WHS									
2008	WHS								80 (85)	
2009	WHS	61 (65)	36 (46)				71 (91)	40 (45)	31 (38)	
2010	WHS				86 (110)					
2011	WHS				96 (104)		97 (110)			
2012	WHS		24 (28)		66 (84)		29 (38)			

#### Kennebec River

Mean and maximum total PCB (TPCB) concentration in American shad filet (AMS-F) from Waterville (KWV) exceeded the FTAL of 11 ng/g in 2012, while concentrations in shad roe (AMS-R) was much lower slightly above the FTAL (Table 3.2.2). Mean and maximum TPCB concentrations in white sucker (WHS) at Sidney (KSD) below Fairfield, exceeded ME-CDC's FTAL. Concentrations were the highest ever measured at this station although much lower than measured downstream at Augusta in previous years. The variation in concentrations within species among all years may reflect differences among the four different labs that were used, although all data meet quality assurance and control objectives, or simply the natural variation in individuals and condition among years.

Year		Norridgewock	Skowhegan	Fairfield	Waterville	Sidney	Augusta	Hallowell	Gardiner	Richmond
		KNW	KSK	KFF	KVVV	KSD	KAG	KRH	KGD	KRD
4004	DUT			200						
1994	BNT			300			54.0 (70.0)			
1997	BNI			93 (107)			54.6 (70.9)			
1999	BNI	_					55 (71)			
2000	BNI	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			0 (0)		26 (32)	0. (101)			
2012	WHS					64 (87)				
2012						04 (07)				
2012	AMS-E				27 (31)					
2012	AMS.P				12 (13)					
2012	AMS-R				1Z (13)					

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

#### Sebasticook Lake

Mean and maximum total PCB (TPCB) concentrations in white sucker (WHS) from Sebasticook Lake (SLN) at Newport exceeded ME-CDC's FTAL of 11 ng/g in 2012 (Table 3.2.3). Both mean and maximum concentrations were similar to those of 2010 and 2011, the only other years for which there are data. This station is below a former textile mill on the on the East Branch of the Sebasticook River at Corinna which became a Superfund site that has been remediated for other contaminants. Concentrations from other years are higher downstream below the confluence with the West Branch at Burnham in recent years.

Table 3.2.3. Total PCBs in fish from the Sebasticook River, ng/g									
Year	Species	E Br Newport	Sebasticook L	W Br Palmyra	Burnham	Winslow			
		SEN	SLN	SWP	SBN				
1994	SMB			9					
1997	LMB	3		4	3	6			
2009	LMB	31 (36)		4 (5)	39 (41) SMB				
2010	LMB	38 (40)			54 (63) SMB				
1997	WHP	4							
1997	WHS			6	7	14			
2009	WHS			7 (8)	70 (77)				
2010	WHS		60 (67)		113 (133)				
2011	WHS		49 (49)						
2012	WHS		60 (61)						

#### **3.3** ASSESSMENT OF ENVIRONMENTAL CONDITIONS ON THE PENOBSCOT RIVER (MAINE) PRIOR TO DAM REMOVAL: CHARACERIZATION OF CONTAMINANTS IN FISH AND BIVALVES

# Principal Investigator:

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# **Project Synopsis**

This report provides preliminary data on profiles of selected contaminants in a subset of archived samples of resident Penobscot River (Maine) biota collected in 2008, prior to removal of the Great Works and Veazie dams. Contaminant concentrations were measured in populations of resident juvenile common shiners (Luxilus cornutus) and Elliptio sp. mussels from two sites in the Penobscot River: the Costigan boat launch (upstream of the Great Works and Milford dams), and the Eddington Salmon Club (downstream of the Veazie dam). Total mercury (tHg) levels were slightly, but significantly, higher in Costigan mussels  $(0.750 \pm 0.112 \text{ ug/g dry weight})$  relative to Costigan common shiners (0.483  $\pm$  0.047), Eddington fish (0.396  $\pm$  0.050) and Eddington mussels (0.520  $\pm$ 0.086) (mean  $\pm$  SD, n=3 pools of 9-10 individuals). There was no difference in tHg content of fish between the two sites, or between the fish and mussels from the Eddington site. Methods were optimized for measuring trace levels of organic contaminants in tissue using negative ion chemical ionization mass spectrometry and electron impact mass spectrometry. Tissue concentrations of organic contaminants were quite low. Organochlorine pesticides were not detected in any samples. A small subset of PAH analytes was observed in mussels. In mussels, total PCB concentrations ranged from  $188.6 \pm 179.4$  ng/g dry weight (Costigan) to  $272.0 \pm 210.3$  ng/g dry wt (Eddington), and for common shiners from  $66.1 \pm 48.2$  (Costigan) to  $58.0 \pm 33.7$  ng/g dry wt. (Eddington) (mean  $\pm$ SD, n=2-3 pools of 9-10 individuals). There were no site differences in PCB content for either species.

Pre-dam removal baseline data are essential for assessment of post-dam removal effects on contaminant mobilization, transport and potential chemical trophic transfer, for Penobscot Indian Nation (PIN) communities, and for local, state and federal agencies involved in Northeast diadromous species restoration focused on Penobscot River populations.

# Statement of the Problem and Justification

The Penobscot River is the second largest river system in New England, draining 8,570 square miles of northern and central Maine. An agreement to remove two dams, Great Works and Veazie, and improve fish passage on others, was reached between the Pennsylvania Power and Light Corporation (PPL, the power utility), conservation groups, the Penobscot Indian Nation (PIN), the State of Maine, and the U.S. Department of the Interior to restore declining runs of salmon and other sea-run fish. Removal of the Great Works dam was completed in November 2012, with Veazie dam designated for removal in 2013.

Dam removal can result in the movement of contaminants. Sediments behind and downstream of dams can be mobilized, transported, and re-deposited with concomitant release to overlying waters

of sediment-associated pollutants (Simpson, Apte et al. 1998; Latimer, Davis et al. 1999). Contaminants released from resuspended sediments include persistent, toxic, legacy chemicals known to be in the Penobscot River (polychlorinated biphenyls (PCBs), polynuclear aromatic hydrocarbons (PAHs), mercury (Hg), toxic metals, and polybrominated diphenyl ethers (PBDEs), among others) (Simpson, Apte et al. 1998; Latimer, Davis et al. 1999). Dam removal will also greatly improve the passage of migratory fish which could act as contaminant vectors, transporting pollutants up/down stream in the form of chemical body burdens acquired in-river, along the coast or at sea. Contaminants are of critical importance in the Northeast USA where the only extant population of migratory Atlantic salmon (*Salmo salar*) is in Maine and is listed as endangered (NRC 2004).

Information on contaminants in Penobscot River fish is confined to data collected by the Maine Department of Environmental Protection (MDEP), the PIN and the U.S. Fish and Wildlife Service (USFWS) on highly mobile adults whose body burdens represent an integrated signal over many river miles and many years. To evaluate year-to-year variability in local contaminant conditions, and to facilitate identification of hot-spots and unexpected changes in chemical signals (e.g. spills, runoff) in the vicinity of the dams, it is critical that contaminant profiles be determined for non-migratory, resident sentinel species, including bivalves and fish with limited home-ranges whose body burdens reflect short-term changes in local exposure to bioavailable toxicants, including mercury.

This study links to ongoing restoration efforts in which multi-million dollar investments have been made by federal and state resource agencies, non-governmental organizations, and the private sector for river surveys, fish migration studies, dam removal, state-wide culvert improvements, hatchery rearing, and Atlantic salmon egg, fry and smolt stocking in listed rivers. In-kind support of this study has been provided since 2008 by MDEP (fish collection) and the PIN (collection permits) for the collection and archiving of fish and mussel samples. Funding was provided by USGS in the initial phase of the project in 2007 for purchase of collection equipment, freezer storage, and site reconnaissance.

The objective of this study was to obtain preliminary data on pre-removal baseline contaminant profiles in resident juvenile fish and bivalve populations up and down stream of the Great Works and Veazie dams. Chemicals were chosen for analysis to represent important contaminant classes, and not to identify potential sources of these contaminants.

# Approach

# Overview

This project measured persistent, bioavailable toxicants in a subset of archived samples of *Elliptio* mussels and resident juvenile (1-2 y old) common shiners (*Luxilus cornutus*) collected upstream of Great Works dam (Costigan boat ramp area, 45° 00'46.24"N/68°38'27.77"W) and down-stream of the Veazie dam (Eddington Salmon club area, 44°49'27.58"N/68°41'41.40"W). *Elliptio* species, like other mussels, rapidly accumulate metals and hydrophobic contaminants, metabolize these compounds very little, and release them very slowly (Beckvar, Salazar et al. 2000; Thorsen, Forestier et al. 2004). These characteristics, together with their non-mobile life history, make them

excellent sentinels of bioavailable chemicals in their habitats. Common shiners are classified as benthopelagic invertivores (Ontario Freshwater Fishes Life History Database). Fish accumulate and retain halogenated hydrocarbons, such as PCBs (Rubinstein, Gilliam et al. 1984), but rapidly metabolize and clear PAHs (Elskus and Stegeman 1989). For this reason PCBs and halogenated pesticides, but not PAHs, were evaluated in fish in this study.

Removal of the Great Works dam began in June 2012 and was completed in November 2012. Archived samples were collected nearly every year from 2007 - 2012; this study measured a subset of these from 2008, three composites of 9-10 mussels and three composites of 10 fish, from each site for a total of 12 samples. These measurements provide preliminary pre-removal information on contaminant profiles for PCBs, chlorinated pesticides and total mercury for resident fish and mussels, and polynuclear aromatic hydrocarbon (PAH) profiles for mussels, informing strategies for funding analysis of the remaining samples and providing a baseline against which future measurements of contaminants in non-migratory resident fish and mussel biota can be measured. Archiving of resident fish and mussels for chemical analysis will resume post-dam removal (2013 or 2014).

# **Organism** collection

Juvenile common shiners (*Luxilus cornutus*) (1.4-8.4 g wet) were collected by boat- seine with the Maine Department of Environmental Protection. Permission was obtained each year from the Penobscot Indian Nation (Indian Island, Old Town, Maine) for collections in the Costigan boat ramp area. Fish were maintained alive in aerated buckets until euthanized within 3 hours of collection with an overdose of tricane methanesulfonate (MS222). Wet weight and fork-length were measured and fish were wrapped in acetone-rinsed aluminum foil and immediately frozen at -20°C. *Elliptio* mussels (5.1 - 8.6 cm) were collected by wading using a glass bottom 'view' bucket. Mussels were maintained on ice for transport to the laboratory, placed in bags and frozen at -20 C.

# Tissue preparation

Fish heads and tails were removed and the remaining carcasses homogenized using a stainless steel Brinkmann Polytron<sup>TM</sup>. Ten fish were homogenized per composite and 3 composites made per site. Mussels were thawed, drained of excess water, minced with stainless steel scalpels, and homogenized as described for the fish. Nine to ten mussels were homogenized per composite, 3 composites per site. Homogenates were divided into aliquots for analysis of mercury and organics and immediately frozen at -20 C.

# Mercury Analysis

Homogenates for mercury analysis were transferred frozen, on ice, to the Sawyer Environmental Chemistry Research Laboratory (SECRL) at the University of Maine, Orono. Homogenates were analyzed by SECRL using atomic absorption spectrometry on a Milestone DMA-80 Direct Mercury Analyzer. All analyses were accompanied by appropriate QA/QC samples, including analyzing 10% of the samples in duplicate, 10% of the samples for matrix spikes, procedural blanks, standards, and mercury reference materials. Sample reference material recovery ranged from 96.3-103.1%. Reported values are not corrected for recovery.

# **Organics** Analysis

Homogenates for organics analysis were freeze-dried, pulverized with a mortar and pestle, and extracted using a modification of the pressurized liquid extraction method of Gomez-Ariza et al. (Gomez-Ariza, Bujalance et al. 2002). Briefly, 1 g of freeze-dried tissue was suspended in diatomaceous earth, layered over a bed of 100% activated alumina and 100% activated florisil in a stainless steel cartridge and capped with fine sand. O-terphenyl and 2,2', 3,4,5,6'hexachlorobiphenyl (CB#143) were added as surrogate standards and the homogenates extracted under pressure (1500 psi) and heat (50 C) in an Accelerated Solvent Extraction (ASE<sup>TM</sup>) system using a mixture of hexane/dichloromethane (80/20). Extracts were volume reduced using a Rotary Evaporator<sup>TM</sup> with gentle heating in a water bath (30 C) and brought to a final volume of 0.5 mL under a gentle stream of high purity nitrogen gas. Prior to injection, each sample was spiked with internal standards (d10 phenanthrene and octachloronaphthalene) to determine recoveries. Recoveries ranged from 58 - 111 % ( $78 \pm 16\%$ , mean  $\pm$  SD) for PCBs and OCs, 82 - 109 % ( $93 \pm$ 10 %) for PAHs. Reported values are not corrected for recovery. Alkylated PAHs were not chosen for analysis as the intent of this study was to evaluate baseline levels of important contaminants, not to identify potential sources of those contaminants. Procedural blanks did not contain any detectable analytes for OCs. A single PAH analyte, phenanthrene, was found in the procedural blanks at a level of 2 ng/g dry weight, and was not subtracted from the sample values. Low levels of PCB congeners detected in the procedural blanks were subtracted from the samples. Instrument detection limits were 0.4 ng/g dry weight for PCBs, 0.5 ng/g dry weight (2 - 4 ring) and 2.5 ng/g dry weight (5-6)ring) for PAHs, 5 ng/g for DDTs and 10 ng/g for OCs.

We employed negative ion chemical ionization mass spectrometry (NCI-MS) for detection of OCs and PCB congeners, a technique which offers greater selectivity and higher sensitivity than traditional electron impact (EI) mass spectrometry for these kinds of (electrophilic) compounds. PAHs were analyzed using EI mass spectrometry. For NCI-MS, samples were injected (2 uL, splitless) onto a 30 m DB-5 (J&W Scientific) column with high purity methane as the reagent gas and helium as the carrier gas. For EI-MS, samples were injected (2 uL, splitless) onto a 30 m DB-5 (J&W Scientific) column with helium as the carrier gas. Response factors and relative response factors were derived from five point standard curves for PCB, PAH, and OC analytes and used to calculate analyte concentrations. A list of the analytes is presented in Table 1.

# **Statistics**

Site and species differences were evaluated using two-tailed Student t-test at a significance level of p < 0.05.

Polynuclear aromatic hydrocarbons	Organochlorine pesticides	Polychlorinated biphenyls		
naphthalene	aldrin	IUPAC#		
2-methylnaphthalene	alpha-BHC	1	2	monochlorobiphenyl
acenaphthylene	beta-BHC	5	2,3	dichlorobiphenyl
acenaphthene	delta-BHC	18	2,2',5	trichlorobiphenyl
fluorene	gamma-BHC	31	2,4',5	trichlorobiphenyl
phenanthrene	4,4'-DDT	44	2,2',3,5'	tetrachlorobiphenyl
anthracene	4,4'-DDE	52	2,2',5,5'	tetrachlorobiphenyl
fluoranthene	4,4'-DDD	66	2,3,4,4'	tetrachlorobiphenyl
pyrene	2,4'-DDT	101	2,2',4,5,5'	pentachlorobiphenyl
benz[a]anthracene	2,4'-DDE	87	2,2',3,4,5'	pentachlorobiphenyl
chrysene	2,4'-DDD	110	2,3,3',4',6	pentachlorobiphenyl
benzo[b]fluoranthene	dieldrin	151	2,2',3,5,5',6	hexachlorobiphenyl
benzo[k]fluoranthene	endosulfan I	153	2,2',4,4',5,5'	hexachlorobiphenyl
benzo[a]pyrene	endosulfan II	141	2,2',3,4,5,5'	hexachlorobiphenyl
indeno[1,2,3,c,d]pyrene	endosulfan sulfate	138	2,2',3,4,4',5	hexachlorobiphenyl
dibenz[a,h]anthracene	endrin	187	2,2',3,4',5,5',6	heptachlorobiphenyl
benzo[g,h,i]perylene	endrin aldehyde	183	2,2',3,4,4',5',6	heptachlorobiphenyl
	heptachlor	180	2,2',3,4,4',5,5'	heptachlorobiphenyl
	heptachlor epoxide-isomer B	170	2,2',3,3',4,4',5	heptachlorobiphenyl
	methoxychlor	206	2,2',3,3',4,4',5,5',6	nonachlorobiphenyl

	Table 1.	Chemical	contaminants	analyzed	for in	fish and	mussel	tissues.
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# Results

#### Mercury

Total mercury (tHg) levels in composite mussel samples ranged from 0.43 to 0.86 ug/g dry wt, and in composite fish from 0.36 to 0.52 ug/g dry wt. (Figure 1). Mean total mercury (tHg) levels (Figure 2) were slightly, but significantly, higher in Costigan mussels ( $0.750 \pm 0.112$  ug/g dry weight) relative to Costigan common shiners ( $0.483 \pm 0.047$ ) (p=0.0185), and Eddington mussels ( $0.520 \pm 0.086$ ) (p=0.047). There was no difference in tHg content of fish between the two sites, Costigan common shiners ( $0.483 \pm 0.047$ ) and Eddington common shiners ( $0.396 \pm 0.050$ ) (p=0.096), or between fish and mussels from the Eddington site (p=0.096).

# **Organics**

Tissue concentrations of organic contaminants were quite low. Organochlorine pesticides were not detected in any samples. A small subset of PAH analytes was observed in mussels (Figure 3). PCB congeners were consistently detected in all samples (Figure 4), with composite values ranging from 29.3 to 121.8 ng/g in composite fish and 11.2 to 448.4 ng/g in composite mussels (Figure 5). Mean total PCB concentrations ranged from 58.0 to 66.1 ng/g dry weight for common shiners and from 188.6 to 272.0 ng/g dry weight for mussels (Figure 6). There were no site differences in total PCB tissue concentration for either species.



Figure 1. Total mercury concentrations (ug/g dry weight) of 3 individual composite samples, each comprised of a single pool of 10 fish and 9-10 mussels, for common shiners (S) and *Elliptio* mussels (M) collected in 2008 from sites upstream (C=Costigan) and downstream (E=Eddington) of the dams identified for removal in the Penobscot River.



Figure 2. Mean total mercury levels (ug/g dry wt) in the composites of common shiners (S) and *Elliptio* mussels (M) from Costigan (C) and Eddington (E) depicted in Figure 1. Values represent mean  $\pm$  SD, n = 3 composites of 9-10 individuals per composite. \*Significantly different from all other samples at p <0.05.



Figure 3. Polynuclear aromatic hydrocarbon analytes (ng/g dry weight) in composite samples of *Elliptio* mussels from Costigan and Eddingt collected in 2008. Values are ng/g dry weight.


Figure 4. Tissue concentrations of PCB congeners listed by IUPAC number (Ballschmiter and Zell 1980) in composite samples of common shiners and *Elliptio* mussels from Costigan and Eddington collected in 2008. Values are ng/g dry weight.

Costigan fish and mussels, Eddington fish and mussels



Figure 5. Total PCB concentrations (ng/g dry weight) of 3 individual composite samples, each comprised of 10 fish and 9-10 mussels, for common shiners (S) and *Elliptio* mussels (M) collected in 2008 from Costigan (C) and Eddington (E).



Figure 6. Mean total PCB (ng/g dry weight) of composite samples of common shiners (S) and *Elliptio* mussels (M) from Costigan (C) and Eddington (E). Values are means  $\pm$  SD, n = 3 composites of 9-10 individuals.

#### Summary and Discussion

Mercury and organic contaminants were measured in composite, whole body homogenates of resident juvenile common shiners and resident *Elliptio* mussels collected in 2008 from sites in the Penobscot River upstream and downstream of two dams prior to their removal: the Great Works and Veazie dams. Total mercury levels were slightly, but significantly, higher in mussels from the upstream, Costigan site, relative to common shiners from this site, and also higher than both common shiners and mussels from the downstream Eddington site. There was no difference in tHg content of fish between the two sites, or between the fish and mussels from the Eddington site. Mercury levels in mussels from the Costigan and Eddington sites ranged from 0.43 to 0.86 ug/g dry wt (430 to 860 ng/g dry wt). These concentrations are similar to levels reported in *Elliptio complanata* deployed along a gradient near a Superfund site in the Sudbury River, MA, whose tHg tissue concentrations ranged from 340 to 950 ng/g dry weight (Beckvar, Salazar et al. 2000). The authors of that study emphasize the difficulty of finding mussels with low mercury concentrations, even from sites considered pristine and/or with no known local sources of mercury (Beckvar, Salazar et al. 2000).

On a wet weight basis, mercury values in Costigan and Eddington common shiners  $(0.095 \pm 0.003 \text{ and} 0.088 \pm 0.016 \text{ ug/g}$  wet weight whole fish, respectively) are somewhat lower than those found in juvenile fish of a non-migratory species, *F. heteroclitus*, collected in the Penobscot River from sites downstream from a mercury point source (0.136 - 0.241 ug tHg/g wet wt fillet in fish from Soudabascook to Old Pier). However, levels in the Costigan and Eddington shiners are ~ 6 times higher than those of *F. heteroclitus* collected from Little River (0.015 ± 0.008 ug/g wet fillet), a reference site in the Wells National Estuarine Research Reserve, Wells, Maine (Elskus 2012). It is important to note that the common shiner mercury measurements in the present study are not directly comparable to the *F. heteroclitus* mercury values as the former were evaluated on fillets. Mercury levels in Penobscot River common shiners from Eddington and Costigan are below those shown to have adverse effects in juvenile/adult fish (0.200 ug/g wet) (Wiener, Sandheinrich et al. 2012).

Polynuclear aromatic hydrocarbons were found in trace amounts in mussels from Eddington and Costigan, and showed no site-related differences in either relative concentration or profiles of contaminants. In keeping with the likelihood of steric hindrance preventing passage across biological membranes of PAHs with log Kows> 6 (Thorsen, Forestier et al. 2004), chrysene (log Kow =5.86, Thorsen, Forestier et al. 2004) was the highest molecular weight PAH we observed in mussel tissue. Organochlorine pesticides were not detected in fish or mussels from either site.

In contrast to PAHs and OCs, several PCB congeners were detected. Total PCB concentrations in mussels ranged from 11.2 to 448.4 ng/g dry weight; there were no site differences. In comparison to the only other dataset on PCBs in freshwater mussels in Maine that the author is aware of, total PCB concentrations ranged from 2.7 to 188 ng/g dry wt in the freshwater mussel, *Elliptio complanata*, deployed in cages at several sites in the Kennebec river for 53 days in the year 2000 (MDEP 2002). In other studies, total PCB concentrations ranging from 4.2 to 179 ng/g wet weight (approximately 35 to 1,520 ng/g dry weight assuming a wet/dry ratio of 8.5) were reported for the freshwater zebra mussel, *Dreissena polymorpha*, from sites with different degrees and types of pollution (Voets, Talloen et al. 2006).

Total PCBs in common shiners ranged from 29.3 to 121.8 ng/g dry weight (5.8 to 22.9 ng/g wet weight); there were no site differences. These values are in the same range as total PCB values reported in other fish species from Maine rivers (MDEP 2010) (Mierzykowski 2007). Total PCBs have been reported in adult smallmouth bass, American eel, Atlantic salmon, rainbow smelt and white suckers from a variety of locations throughout the Penobscot River by the Maine Department of Environmental Protection Agency as part of their Surface Water Ambient Toxics Monitoring program. For fish collected in 2008 from the Penobscot River, total PCBs ranged from 7 ng/g wet wt (S. Lincoln) to 9 ng/g wet wt (Woodville) in smallmouth bass, and from 49 to 29 ng/g wet wt in white suckers from S. Lincoln and Woodville, respectively (MDEP 2010). For white suckers collected from rivers in the distinct population segment of the Atlantic salmon, total PCBs in whole body homogenates were reported as 17 - 61 ng/g wet wt (Denny's river), 7 and 18 ng/g wet weight (Pleasant river), and a mean of 5 ng/g wet weight (East Machias river) (Mierzykowski 2007), which are in line with those reported by National monitoring programs (see Table 5 in (Mierzykowski 2007).

## Benefits

Contaminant profiles in resident biota indicate which contaminants are bioavailable in the Penobscot system and relevant to migrating fish, resident species, chemical trophic transfer, ecosystem health and human health. These data are also critically important for native peoples in this area (PIN) who use aquatic plants and animals for food and cultural activities. Eleven diadromous species migrate in the Penobscot River system, all potential carriers of contaminants up/down stream post-dam removal. Comparing up/down stream contaminant profiles in resident, non-migratory species pre- and post-dam removal may influence decisions on which organisms and contaminants to monitor, and guide hypothesis development to elucidate contaminant dynamics in the river. The Penobscot River dam removal project has Congressional support, is lauded as an example of grass-roots river restoration efforts in the US; contaminant work in this river increases visibility in additional aspects of the Penobscot River and diadromous species restoration in the Northeast region and nationwide. USGS cooperators and partners (PIN, US Fish and Wildlife Service (USFWS), NOAA Fisheries, Maine Department of Environmental Protection (MDEP), and Maine Department of Marine Resources (MDMR) have provided both financial and in-kind support for Penobscot River restoration projects, including sample collection and archiving for this project.

The results of this project have relevance to projects targeting removal of low-head, run-of-the river dams, such as the Penobscot dams, as the vast majority of studies published to date are on large scale hydrodams, conditions so different from low-head dam removals that comparisons, and thus predictions, are tenuous at best.

Disclaimer: Any use of trade, product, or firm names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

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## **3.4 ANDROSCOGGIN RIVER SEDIMENTS**

### 3.4.1 Introduction

As required by EPA, Maine has adopted ambient water quality criteria (AWQC) for toxic pollutants by rule at CMA 06-096 Chapter 584 to be used in calculation of effluent limits for dischargers (see http://www.maine.gov/sos/cec/rules/06/chaps06.htm). AWQC for heavy metals are expressed as total metal, even though the most bioavailable and toxic specie are ionic forms. Use of total metal provides some margin of safety for various uncertainty factors. The AWQC are developed according to EPA guidelines to include toxicity data, usually from laboratory studies, from 8 families of aquatic organisms. Chapter 584 also allows development of site specific criteria (SSC) generally following EPA guidance with additional requirements specified in the rule. The development of SSC is usually initiated by a discharger with the recognition that for heavy metals, some is bound to particles and therefore not bioavailable or toxic; their goal is an increase in the amount of total metal allowed to be discharged. One concern is that an increase in total metal discharged to a waterbody may result in an increased in sediment concentrations and have negative effects on aquatic organisms in the sediment that is not evaluated by AWQC, which addresses only water column toxicity. According to EPA (2005)

Toxic contaminants in bottom sediments of the nation's lakes, rivers, wetlands, and coastal waters create the potential for continued environmental degradation even where water column contaminant levels meet applicable water quality standards. In addition, contaminated sediments can lead to water quality impacts, even when direct discharges to the receiving water have ceased.

The AWQC for many metals are functions of hardness. Most Maine rivers that receive discharges have relatively soft water. Consequently, DEP uses a default value for total hardness of 20 mg/l CaCO3 for calculation of AWQC for those metals. One method for development of SSC for metals is to use a site-specific total hardness value. To address potential seasonal variability, DEP requires total hardness to be measured at least monthly over a period of a year. Although increased hardness reduces the amount of dissolved metal responsible for toxicity in the water column, total loading of metals to the receiving water system might not be reduced, but in fact be increased. Much of the increased metal could precipitate or sorb to particles and settle out to the sediments downstream, where it could be toxic to benthic organisms. To account for this uncertainty and provide a margin of safety, DEP has required measurement of site-specific hardness upstream of discharges. Dischargers have requested that hardness be measured downstream sample location, but agreed that states could make their own determination.

Another method for developing SSC for copper is Use of EPA's BLM (Biotic Ligand Model). The BLM uses measurements of 10 water quality variables (temperature, alkalinity, calcium, magnesium, sodium, sulfate, potassium, chloride, dissolved organic carbon, and pH) to calculate the bioavailable fraction of copper in the water column which would then be used as the basis for the permit limit. There are draft BLMs for other metals as well. The BLM also raises concern for the potential effects of increased loading of total copper to the sediments.

Recently, DEP developed DETOX, a mass balance based waste load allocation model for toxic pollutants, which allocates waste loads of specific pollutants among all the dischargers to a waterbody to ensure that the AWQC are not exceeded at any location in the waterbody. Application of DETOX to the Androscoggin River identified the discharge of several heavy metals from multiple dischargers that would result in exceedance of the AWQC in at least one reach of the river. The dischargers have inquired about development of SSC for these metals using downstream site-specific total hardness and or the BLM. To address the sediment toxicity issue, DEP proposes to sample sediments in the river in 2012.

In order to address sediment contamination and the effects on aquatic biota, there are several methods that have been used. Bulk sediments may be analyzed for total metal and results compared to SQC or guidelines (MacDonald et al. 2000). Total metals are not always biologically available, however, and have not always been predictive of sediment toxicity. It is the dissolved metal (and to a much lesser extent some other metal compounds such as metal hydroxides) that account for toxicity. To better address bioavailability and toxicity, EPA suggests use of the Equilibrium Partitioning (EqP) approach, and development of Equilibrium partitioning Sediment Benchmarks (ESBs) (EPA, 2005). Two approaches are proposed for establishment of ESBs.

One is measurement of sediment pore water and comparison to AWQC. In this approach, sediments containing heavy metals should not cause direct toxicity to benthic organisms if the sum of the dissolved interstitial water concentrations for each of the metals ( $\sum M_{i,d}$ ) divided by their respective Water Quality Criteria (WQC) Final Chronic Value (FCV) is  $\leq 1.0$ . Sampling sediment interstitial water for metals is not a routine procedure. The least invasive technique employs a diffusion sampler that has cavities covered with a filter membrane. The sampler is inserted into the sediment and the concentrations on either side of the membrane equilibrate. Because the sampler is removed after equilibration, the concentrations of metals inside the sampler should be equal to the concentrations of freely-dissolved metals in the interstitial water. The time required for equilibration, typically several days, depends on the size of the filter membrane and the geometry of the cavity. An alternative technique for separating interstitial water is to obtain an undisturbed sediment sample as a whole sediment or core that can be sliced for vertical resolution, filter or centrifuge the sample, and then filter the resultant interstitial water twice. For anaerobic sediments, this must be done in a nitrogen atmosphere to prevent precipitation of iron hydroxide, which would scavenge the metals and yield artificially low dissolved concentrations of metals (EPA, 2005).

Another method is use of the ratio of simultaneously extracted metals and acid volatile sulfides (SEM-AVS), which has been used to predict the combined sediment toxicity of copper (Cu), cadmium (Cd), nickel (Ni), lead (Pb), silver (Ag), and zinc (Zn) (Ankley et al. 1996; Berry et al. 1996). Research indicates that the amount of AVS present in sediment limits the metal bioavailability and subsequent toxicity in sediments. Sulfide is an important binding component in modeling metal sorption in sediments (Morse et al. 1987). In the presence of excess sulfide, most of the reactive metal will form insoluble metal sulfides. The AVS is one part of the ratio of available sulfide to the SEM metals and allows the partitioning of free aqueous phase metal and solid phase metal in sediments. Silver (Ag) and the five divalent metals (Cd, Ni, Cu, Pb and Zn) form metal sulfide complexes. If the molar ratios of the SEMs are greater than that of the AVS, the excess fraction of the metals may be considered to have a high potential for bioavailability. For divalent metals, one mole of SEM will react with one

mole of AVS, whereas for silver one mole of SEM will bind two moles of Ag. These metals should not cause direct toxicity to benthic organisms if the  $\Sigma$ SEM-AVS is  $\leq 0.0$ .

Uncertainty bounds on  $\sum$ SEM-AVS and ( $\sum$ SEMAVS)/ $f_{OC}$  can be used to identify sediments where toxicity, because of these metals, is unlikely, uncertain, or likely. If the  $\sum$ SEM-AVS is >0.0 or  $\sum M_{i,d}$  divided by their respective FCVs is >1.0, effects may occur with increasing severity as the degree of exceedance increases. Toxicity may be estimated from the excess SEM normalized to the fraction of organic carbon. Toxicity is likely when the normalized excess SEM ( $\sum$ SEM-AVS)/fOC>3000 umol/g OC, uncertain between 130 and 3000 umol/g OC, and unlikely <130 umol/g<sub>oc</sub> (EPA, 2005).

In addition to varying with depth, AVS can vary seasonally. For example, in systems where overlying water contains appreciable oxygen during cold-weather months, AVS tends to decrease, presumably because of a constant rate of oxidation of the AVS linked to a decrease in its generation by sulfate-reducing bacteria. Because of potential temporal and spatial variability of AVS, it appears that the way to avoid possible underestimation of metal bioavailability is to sample the biologically "active" zone of sediments at times when AVS might be expected to be present at low concentrations. It is recommended that, at a minimum, AVS and SEM measurements be made using samples of the surficial (0 to 2 cm) sediments during the period from November to early May. (EPA, 2005)

# 3.4.2 Methods

Targeted fine grained sediments were scarce and confined to small areas along in coves and along the banks in the Virginia impoundment at Rumford Point (ARP), below Rumford in the Riley impoundment (ARY), and below Jay in the Otis impoundment at Livermore (ALV), but were predominant in Gulf Island Pond (AGI) below both mills. Three sediment samples were collected from the river from the first depositional zone (impoundment) at each of these stations in November 2012. Sediments were collected with a pre-cleaned Eckman dredge and the top 2 cm were collected from the center of the dredge to avoid contact with it. Each sediment sample was divided into two sealed airtight clean glass jars, one of which was refrigerated for analysis for grain size and percent moisture, and one of which was frozen for analysis of SEM-AVS, total metals, and total organic carbon. Enough sediment was collected to almost fill each jar but allow for expansion upon freezing. Samples were sent to the US Department of Energy's Pacific Northwest National Laboratory operated by Battelle to be analyzed. At the lab sampling of the stored sediment from the middle of the jar limited potential effects of oxidation on AVS.

## 3.4.3 Results and Discussion

Since contaminants such as heavy metals sorb to fine grained sediments, to determine if appropriate sediments were sampled or were even present at each station, sediments from each sample was analyzed for grain size. The potential toxicity of the sediments was evaluated by two methods, 1) comparison of the concentrations of metals in the bulk sample to sediment quality guidelines (MacDonald et al 2000) and (2) AVS-SEM.

## Grain Size Analysis

Grain size distribution was relatively similar for the three samples at each station except for ALV-3, below the mill in Jay, which had less sand and more silt than the other two samples at ALV (Table 3.4.1). There were measurable differences among stations with coarser sediments at the upper three stations than in Gulf Island Pond. Although the abundance of sand at the upper three stations limits the binding of metals, these were the finest grained sediments that could be found there, and therefore are representative of the likelihood of retention of heavy metals at these stations.

Table 3.4.1. Grain size of				
	GRAVEL	SAND	SILT	CLAY
RUMFORD POINT				
ARP 1	0	64	34	2
ARP 2	0	54	37	2
ARP 3	0	64	35	2
MEAN	0	61	35	2
RILEY				
ARY 1	0	75	23	1
ARY 2	0	78	25	3
ARY 3	0	73	24	1
MEAN	0	75	24	2
LIVERMORE				
ALV 1	0	43	52	4
ALV 2	1	46	49	2
ALV 3	0	27	67	5
MEAN	0	39	56	4
GULF ISLAND POND				
AGI 1	0	7	73	16
AGI 2	0	7	78	16
AGI 3	0	5	77	16
MEAN	0	7	76	16

## Bulk Sample Chemistry

Concentrations of five metals (Cd, Cu, Ni, Pb, and Zn) were compared to threshold effects concentrations (TEC) and probable effects concentrations (PEC) determined by consensus of leading researchers in the field (MacDonald et al. 2000). TECs are concentrations of metals in bulk sediments below which there were negative effects in less than 25% of sediments in studies from the literature and are unlikely to cause adverse effects on aquatic biota. PECs are concentrations in bulk sediments above which there were effects in more than 75% of the sediments studied and above which adverse effects are probable. The results showed that the concentration of Cd was below the TEC for the upper three impoundments and only slightly exceeded the TEC in Gulf Island Pond; concentrations were well below the PEC at all four stations (Table 3.4.2).

able 3.4.2. Total metals	in Androscoggin River se	diments 2012 (ug/g dw)			
	Cd	Cu	Ni	Pb	Zn
TEC ug/g dw	0.99	31.6	22.7	35.8	121
PEC ua/a dw	4.98	149	48.6	128	459
RUMFORD POINT					
ARP 1F	0.142	11.6	11.9	8.54	53.1
RP 2F	0.187	12.6	12.3	9.22	56.3
ARP 3F	0.192	12.0	12.5	9.53	59.4
MEAN	0.174	12.1	12.2	9.10	56.3
RILEY					
ARY 1F	0.359	14.7	14.5	14.7	78.2
ARY 2F	0.292	12.2	12.5	11.0	61.1
ARY 3F	0.535	19.0	18.6	19.3	99.1
MEAN	0.395	15.3	15.2	15.0	79.5
IVERMORE					
ALV 1F	0.185	10.1	11.0	7.67	63.1
ALV 2F	0.195	9.82	11.0	8.43	61.4
ALV 3F	0.195	10.0	10.5	7.63	62.8
MEAN	0.192	10.0	10.8	7.91	62.4
GULF ISLAND POND					
AGI 1F	1.10	26.6	25.5	30.1	164
AGI 2F	1.02	25.3	24.8	29.0	156
AGI 2FDUP	1.01	25.6	25.0	28.7	158
AGI 3F	1.00	25.4	24.8	28.6	156
MEAN	1.04	25.8	25.1	29.2	159

Concentrations of Cu and Pb were below the TEC at all four stations although highest in Gulf Island Pond. Concentrations of Ni and Zn were below the TEC at the three upper stations but exceeded it in Gulf Island Pond. Higher concentration of all metals in Gulf Island Pond may reflect cumulative loading from upstream sources and opportunity for deposition as evidenced by a greater percent of silts and clay. That there is no trend toward increased concentrations of metals from Rumford Point to Livermore may simply be due to the shallow impoundments which reduce deposition of fines. In any event, these results suggest no toxicity for Cd, Cu, and Pb, with a slight possibility of toxicity from Ni and Zn, but do not account for bioavailability of these two metals.

### SEM-AVS

The sum of simultaneously extracted metals minus acid volatile sulfides ( $\sum$ SEM-AVS) was negative for all but one sample (ALV-3) (Table 3.4.3) demonstrating that all metal would be bound to AVS and not bioavailable to exert toxicity. Mean ( $\sum$ SEM-AVS) for all four stations also indicated the lack of toxicity. For ALV-3 the ( $\sum$ SEM-AVS) normalized to organic carbon ( $\sum$ SEM-AVS)/fOC was well below the threshold where toxicity is unlikely (<130 umol/g<sub>oc</sub>). As was the case for total metals, SEM were highest in Gulf Island Pond, but so was AVS. The result is that ( $\sum$ SEM-AVS) and ( $\sum$ SEM-AVS)/fOC are not highest at Gulf Island Pond and there appears to be no trend of increase in either of these among the four impoundments. Regardless, these results show that metal concentrations are quite low and unlikely bioavailable to cause toxicity in the sediments.

Table 3.4.3. Simultane	eously extracted meta	als - acid volatile sul	fides (SEM-AVS) in s	ediments from four				
impoundments on the Androscoggin River, 2012								
SAMPLE ID	Σ SEM (µmole/g DW)	AVS (µmole/g DW)	Σ SEM-AVS (µmole/g DW)	<sup>1</sup> fOC	(SEM-AVS)/fOC (µmole/g <sub>oc</sub> )			
RUMFORD POINT								
ARP 1F	0.364	0.717	-0.353	0.013	-26.5			
ARP 1F (DUP)	0.361	0.698	-0.337	NA				
ARP 2F	0.404	0.478	-0.073	0.016	-4.68			
ARP 3F	0.389	0.555	-0.166	0.014	-11.8			
MEAN	0.385	0.580	-0.195	0.014	-14.3			
RILEY								
ARY 1F	0.768	2.53	-1.764	0.013	-132			
ARY 2F	0.834	3.10	-2.262	0.018	-124			
ARY 3F	1.06	10.1	-9.066	0.011	-795			
MEAN	0.888	5.25	-4.36	0.014	-350			
LIVERMORE								
ALV 1F	0.396	0.882	-0.486	0.035	-13.8			
ALV 2F	0.500	0.640	-0.140	0.039	-3.60			
ALV 3F	0.423	0.277	0.147	0.047	3.12			
MEAN	0.440	0.600	-0.160	0.014	-4.77			
GULF ISLAND POND								
AGI 1F	2.14	7.04	-4.901	0.060	-81.1			
AGI 2F	2.16	4.59	-2.435	0.058	-42.3			
AGI 3F	2.15	3.90	-1.746	0.059	-29.6			
MEAN	2.15	5.18	-3.03	0.014	-51.0			
<sup>1</sup> fOC = fraction of organ	nic carbon							

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