SURFACE WATER AMBIENT TOXICS MONITORING PROGRAM

2010

FINAL REPORT



DIVISION OF ENVIRONMENTAL ASSESSMENT MAINE DEPARTMENT OF ENVIRONMENTAL PROTECTION AUGUSTA, MAINE 04333

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INTRODUCTION

This 2010 Surface Water Ambient Toxic (SWAT) monitoring program final report is organized into an Executive Summary (with introduction and table of contents) and 4 modules, 1) Marine and Estuarine, 2) Lakes, 3) Rivers and Streams.

The full report is available on DEP's website at http://www.maine.gov/dep/blwq/docmonitoring/swat/index.htm

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Chemical analyses were performed by AXYS Analytical Services, Sidney, British Columbia or other laboratories as listed in reports in individual sections.

EXECUTIVE SUMMARY

Maine's Surface Water Ambient Toxics (SWAT) monitoring program was established in 1993 (38 MRSA §420-B) 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 5-year conceptual workplan in addition to annual workplans 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 4 modules, 1) Marine and Estuarine, 2) Lakes, 3) Rivers and Streams, and 4) Special Studies. This annual report follows the outline of the 2010 workplan recommended by the SWAT TAG in a meeting June 23, 2010. Following is a summary of key findings from the 2010 SWAT program for each module.

1. MARINE AND ESTUARINE

- In 2010, blue mussel tissue from Spring Point, South Portland, Crockett Point, Rockland, and Wadsworth Cove, Castine was analyzed for contaminants including metals, mercury, Polycyclic Aromatic Hydrocarbons (PAHs), Polychlorinated Biphenyls (PCBs), and organochlorinated pesticides.
- In 2010, softshell clam tissue from Morse Cove, Castine was tested and reported with data from seven other softshell clam sites sampled in 2004 and 2005. Clam tissue from selected sites was analyzed for contaminants including metals, mercury, PAHs, PCBs, and organochlorinated pesticides.
- In 2010, American lobster hepatopancreas and muscle tissues from 19 sites along the Maine coast were analyzed for PAHs, coplanar PCBs, and dioxins and furans. The U.S. Environmental Protection Agency (EPA), through the National Coastal Condition Assessment (NCCA), will be analyzing the same lobster tissues for metals, mercury, PCBs, and organochlorinated pesticides.
- Lead in mussel tissue exceeded the National Status and Trends (NS&T) Musselwatch 85th percentile concentration at Spring Point, South Portland, and Crockett Point, Rockland, both of which also exceeded the Maine Center for Disease Control's (MCDC) fish tissue action level (FTAL) for lead in finfish. Lead in clam tissue at Harris Cove, Eastport, Fort Point

Cove, Searsport, and Mast Cove, Eliot exceeded the MCDC fish tissue action level (FTAL) for lead in finfish.

- Mercury in mussel tissue exceeded the NS&T Musselwatch 85th percentile concentration at Spring Point, South Portland, Crockett Point, Rockland, and Wadsworth Cove, Castine, which resulted in assignment of an "elevated" classification. Mercury levels in mussel tissue at these sites were below the more conservative MCDC methylmercury developmental FTAL for finfish. Mercury in clam tissue also was below the MCDC methylmercury developmental FTAL for finfish.
- Chromium in mussel tissue exceeded the NS&T Musselwatch 85th percentile concentration at Spring Point, South Portland, which resulted in assignment of an "elevated" classification.
- PAHs in mussel and clam tissues did not exceed the NS&T Musselwatch 85th percentile and were not considered to be elevated.
- PCB concentrations in mussel tissue at Crockett Point, Rockland exceeded the MCDC cancer FTAL, which is consistent with elevated concentrations detected in 2007 at the same site. PCB concentrations in clam tissue were below the MCDC cancer FTAL.
- Organochlorinated pesticide concentrations in mussel and clam tissue were low at Maine sites compared to NS&T Musselwatch data, and pesticide levels were safely below MCDC FTAL values.
- Dioxin, furan, and coplanar PCB concentrations remain very high in lobster hepatopancreas (also known as tomalley), indicating the need to continue the MCDC advisory against consumption of lobster hepatopancreas. Concentrations of these contaminants in lobster muscle tissue remain very low in comparison to tomalley and to the MCDC FTAL, indicating lobster meat is still safe to eat.

2. LAKES

• Fish from 45 lakes were sampled and analyzed for mercury concentrations by a new quicker less expensive method using the Direct Mercury Analyzer 80 at the Sawyer Environmental Research and Chemistry Lab at the University of Maine in Orono. The results compared favorably with those from a subset of 22 lakes analyzed by a commercial lab using a more conventional method. There was no trend for fish from 26 lakes comparing 2010 results with those from the 1990s. The data were sent to the Maine Center for Disease Control and Prevention (MCDC) for use in reviewing the statewide Fish Consumption advisory.

3. RIVERS AND STREAMS

• Forty-two stations were assessed for the condition of the benthic macroinvertebrate community. Results have been received to date (June 10, 2011) for forty-one stations.

Twenty-six of these forty-one stations attained the aquatic life standards of their assigned class.

- Dioxin concentrations measured in fish from the West Branch of the Sebasticook River were lower than when last measured, but still exceed MCDC's Fish Tissue Action Level (FTAL). The dioxin concentrations in fish from the St. Croix River above Woodland are below the FTAL; dioxin-like coplanar PCBs were not measured in 2010, but in previous years the addition of the PCBs resulted in an exceedance of the FTAL.
- Fish captured from the Androscoggin River, Penobscot River below Millinocket, Presumpscot River in Gorham and Westbrook, and Sebasticook River at Newport and Burnham exceeded MCDC's FTAL for dioxins.
- A study of the Little Androscoggin River below the wastewater discharge from South Paris found no evidence of toxicity of the sediments to bottom dwelling organisms or of the river water to fish species, despite repeated exceedances of the copper limit in their discharge permit.
- A project funded at the University of Maine developed a new non-lethal method of detecting exposure of fish to organic contaminants using fish scales.

1.0 MARINE MODULE

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1.1. INTRODUCTION

This work is funded by the Maine Coastal and Inland Surface Oil Clean-up Fund, which is administered by the Bureau of Remediation and Waste Management in the DEP.

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 a long term focus of the marine SWAT sampling efforts over the years and were included again in the SWAT program this year. This report presents and summarizes contaminant data from the collection and analysis of blue mussel (*Mytilus edulis*) tissue collected in 2010 from three sites along the Maine coast. Mussel tissue samples were analyzed for heavy metals (including mercury), polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyl (PCBs), and organochlorinated pesticides. In order to provide comparability of results from these 2010 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 Mussel Watch Program (National Oceanographic and Atmospheric Administration). This analysis provides a regional and national context to the Maine SWAT data.

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. It also gives insight into the biologically available portion of contaminants, which may not readily be discerned from background sediment or water concentrations.

1.1.2 Softshell Clams

This report presents and summarizes contaminant data from the collection and analysis of softshell clam (*Mya arenaria*) tissue collected in 2010 from one site on the Maine coast. Also presented are softshell clam contaminant data from seven additional sites sampled in 2004-05 by the SWAT program. Softshell clam tissue samples from selected sites were analyzed for metals, mercury, PAHs, PCBs, and organochlorinated pesticides. In order to provide comparability of results from the 2010 and 2004-05 samples, softshell clam contaminant concentrations from SWAT sampling are compared to contaminant concentrations in the Gulfwatch program. This analysis provides some regional context to the Maine SWAT clam data.

Gulfwatch, primarily a blue mussel sampling program, also has some limited clam tissue contaminant data, the most recent collected at two sites in New Hampshire and two sites in Maine in 2008. Like blue mussels, softshell clams 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 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.

The Maine Dept. of Marine Resources 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.

1.1.3 American Lobster

This report presents data from American lobster (*Homerus americanus*) tissues collected in 2010. Lobsters were collected as part of EPA's National Coastal Condition Assessment (NCCA), which also provides data on water column parameters, sediment chemistry, and benthic community structure. In most states participating in NCCA, finfish are collected and used for fish tissue

contaminant analysis as part of the program. Some New England states have elected to collect lobster to fulfill the fish tissue portion of the NCCA, as Maine did in the last NCCA sampling effort in 2006 (and prior).

Lobster was selected to provide information concerning the quality of the benthic environment and because Maine has a fish consumption advisory on lobster hepatopancreas (tomalley) tissue. As predators and scavengers of benthic infauna and detritus on the sea bottom, lobsters ingest toxic contaminants and bioaccumulate those contaminants in their body tissues. Lobsters are ubiquitous along the Maine coast, allowing collections to take place along the entire coast and facilitating geographic comparisons. The lobster fishery is Maine's premier fishery, with the highest landed value of any commercial fishery in the state. In addition, Maine lobstermen strive to provide a high quality product and determining and assuring the quality of this product is of importance to the future sustainability of the fishery. This project builds upon early work done by DEP in 1994-1996 on contaminants in lobster tissues at several locations, and on previous sampling of lobster by NCCA in 2005-06 at many more locations along the Maine coast.

1.2 METHODS

Sites sampled in recent years within the context of this report can be divided in three types, based on the goals outlined above that drive the need for information from each site. 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 better, 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, fine scale contaminant distribution. Follow Up sites may also occur in the Temporal or Spatial categories as well, based on their historical sampling and data needs at the site.

1.2.1 Blue Mussels

Blue mussel samples have been analyzed for toxics as part of the SWAT program since 1986, with over 80 distinct locations sampled in the past 25 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 costs.

Blue mussels were collected from three sites in 2010. Two of the three mussel sites had been sampled previously as part of the SWAT program, with Spring Point, South Portland, and Crockett Point, Rockland, having been sampled three years prior in 2007. Wadsworth Cove, Castine, had not been sampled previously as part of SWAT. Names and locations of blue mussel collection sites for 2010 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, 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 site location were all minimized to reduce conflicting signals in the contaminant data.

Sampling occurred from mid-October to mid-November 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 intrasite 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.

TABLE 1.2.1.1: 2010 SWAT Blue Mussel Sites

		Station	West	North	Date	Site
Site Name	Municipality	Code	Longitude	Latitude	Sampled	Type1
Spring Point	S. Portland	CBSPSP	-70.2276	43.65053	10/14/2010	Т
Crockett Point	Rockland	PBRKCP	-69.1065	44.10641	10/18/2010	F, T
Wadsworth Cove	Castine	PBCAWA	-68.10963	44.40367	11/16/2010	S
¹ $S = Spatial, T = T$	Cemporal, F = Foll	ow Up				

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° C for up to 1 to 2 months until analysis.

Mussels tested for PAHs from three sites in 2010 were analyzed by AXYS Analytical Services Ltd., Sidney, British Columbia. Mussel tissue samples were analyzed for PAHs using modified EPA Method 8270/1625.

1.2.2 Softshell Clams

Softshell clams were slated for collection at two sites in 2010, but only one was successfully collected. Blue mussels were used to generate data at the site where no clams could be collected, Wadsworth Cove in Castine. Softshell clams were collected at Morse Cove, Penobscot/Castine, the only site sampled successfully for clams in 2010. Neither site had been sampled previously for blue mussels or softshell clams. In addition to the softshell clam site sampled in 2010 this report includes data from seven softshell clam sites sampled in 2004-05. This data is included to provide a broader look at softshell clam contaminant concentrations across the state and to present the Morse Cove data in a statewide context. The data from Morse Cove and the seven other 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 the previous mussel station map, 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, 1997) and any departures that methodology in softshell clam sampling will be noted below.

Sampling typically occurred in mid-November and the specific sampling date is 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.



		Station	West	North	Date	Site
Site Name	Municipality	Code	Longitude	Latitude	Sampled	Type ¹
Mast Cove	Eliot	PQMCMC	-70.8048	43.1210	11/9/2004	S
Navy Pier	Harpswell	CBHWNP	-70.0136	43.7870	11/12/2004	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	11/10/2005	S
Morse Cove	Penobscot/Castine	PBCAMC	-68.7835	44.4478	11/16/2010	S
Harris Cove	Eastport	PMHCHC	-66.9838	44.9171	11/9/2004	S
Mill Cove	Robbinston	PMSCMC	-67.1176	45.0580	11/29/2005	S
¹ S - Spatial T -	Temporal E - Follow	, I In				

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

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

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, 20 clams were composited into a replicate.

Tissue composite samples for metals analyses included ten clams per composite sample or replicate, with four replicates collected per sampling station. Tissue composite samples for organics analyses included 20 clams per composite sample or replicate, with four replicates 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 clams tested for PAHs in 2010 were analyzed by AXYS Analytical Services Ltd., Sidney, British Columbia. Clam tissue analyzed in 2004-05 was analyzed by Pace Analytical, Minneapolis, MN. Clam tissue samples were analyzed for PAHs using modified EPA Method 8270/1625.

1.2.3 American Lobster

The NCCA uses a probabilistic sampling design that selects stations in a random fashion via a computer program. Thus, the stations are not selected due to proximity to pollution sources or known features of any type. Of 43 stations selected to represent the Maine coast in 2010 as part of NCCA, Maine DEP attempted to obtain lobster samples from as many stations as possible; however, many of the 43 stations were not suitable for lobster collection, either lacking suitable habitat, being too shallow or too far offshore, or exhibiting salinity too low for lobster habitation. Lobsters were collected successfully at 21 of the 43 stations in the design.

Sampling occurred in late summer/early autumn before lobsters migrated to deeper, offshore waters. Up to seven lobsters were purchased directly from lobstermen, who agreed to provide lobsters from within the vicinity of the sampling station after the location was provided via use of a map or from station coordinates. Seven lobsters per station were collected to provide adequate tissue (specifically the mass-limited hepatopancreas tissue) for achieving laboratory detection limits. Smaller, legal lobsters that just recruited to the fishery (approximately 1 to 1.25 lbs. or 0.45 to

0.57kg) were selected for inclusion in the composite samples at each station. Since smaller lobsters are thought to have a reduced home range of perhaps less than 1-2 km, using these lobsters in late summer would limit the integration of contaminants to a more localized area within that home range. Lobsters were individually double wrapped in aluminum foil, labeled with station ID and individual lobster number, placed in a polyethylene bag, and frozen live at -20°C.

Lobsters were dissected by DEP staff into hepatopancreas and muscle tissues. Whenever possible, lobster samples were composites of seven individual animals, though one sample contained six lobsters. EPA, as part of the NCCA program and through a contracted laboratory, analyzed lobster hepatopancreas and muscle tissues for mercury, heavy metals, non-alkylated PAHs, pesticides, and PCBs. As part of the SWAT program, DEP provided additional analysis of lobster hepatopancreas and muscle tissues for additional PAHs including alkylated PAHs, dioxins, furans, and coplanar PCBs. This report includes data analysis for DEP SWAT-funded suites of the analytes listed above, from 19 of 21 stations collected along the Maine coast in 2010. Insufficient SWAT funds resulted in lobsters from two sites not being analyzed.

Table 1.2.3.1 includes coordinates and sampling dates for the 19 SWAT-analyzed lobster stations. Figures 1.2.3.1 (western Maine), 1.2.3.2 (mid-coast Maine), and 1.2.3.3 (Downeast Maine) show the location of 19 SWAT-analyzed lobster collection stations. Lobster tissues tested for PAHs in 2010 were analyzed by AXYS Analytical Services Ltd., Sidney, British Columbia, using modified EPA Method 8270/1625. Additional data stemming from the EPA analysis for mercury, heavy metals, non-alkylated PAHs, pesticides, and PCBs were not available as the lab work was not completed by the time of this report. These data will be acquired by DEP for future reference and for inclusion in the DEP database.

Location	Station Code	West Longitude	North Latitude	Date Sampled
Luckse Sound, Casco Bay	NCCA10-1017	70.20406	43.62466	11/16/2010
Cousins Island Sound, Casco Bay	NCCA10-1021	70.16395	43.71207	11/16/2010
Broad Sound, Casco Bay	NCCA10-1020	70.07681	43.69880	11/2/2010
W. Pond Is., Casco Bay	NCCA10-1016	69.98100	43.73903	10/16/2010
E. Pond Is., Casco Bay	NCCA10-1018	69.95756	43.73462	11/2/2010
Pemaquid Point, Muscongus Bay	NCCA10-1067	69.49613	43.83977	11/9/2010
Muscongus Sound	NCCA10-1063	69.44983	43.92972	11/9/2010
Belfast Bay	NCCA10-1054	68.91670	44.41312	10/7/2010
W. Isle Au Haut, IAH Bay	NCCA10-1057	68.66315	44.05863	10/4/2010
Merchant Row	NCCA10-2050	68.63407	44.11128	9/23/2010
Eggemoggin Reach	NCCA10-1069	68.65338	44.26565	10/22/2010
W. Swans Is., Jericho Bay	NCCA10-1084	68.51483	44.13697	10/5/2010
Flye Is., Blue Hill Bay	NCCA10-1065	68.50157	44.25105	10/14/2010
S. Bartlett Is., Blue Hill Bay	NCCA10-1082	68.43315	44.31983	9/20/2010
Narraguagus Bay	NCCA10-1072	67.84495	44.49680	9/24/2010
Pleasant Bay	NCCA10-1068	67.74645	44.49827	10/20/2010
W. Great Wass Is., Western Bay	NCCA10-1083	67.62112	44.47708	9/29/2010
Machias Bay	NCCA10-1064	67.34738	44.61583	9/21/2010
Eastport, Cobscook Bay	NCCA10-1060	66.97215	44.89848	11/16/2010

Table 1.2.3.1: 2010 SWAT Lobster Stations







1.3 RESULTS AND DISCUSSION

1.3.1 Metals

1.3.1.1 Blue Mussels

Mussel tissue samples collected in 2010 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, 2008) and Gulf of Maine (Gulfwatch, see LeBlanc, 2009) blue mussel monitoring program data to place Maine SWAT data in a national and regional context. From an environmental monitoring perspective, the concentration of an analyte in SWAT mussel tissue was considered elevated when that concentration exceeded the national NS&T 85th percentile. This approach is consistent with the Gulfwatch program (LeBlanc, 2009).

1.3.1.1.1 Silver (Ag)

Silver was detected in all three sample locations visited in 2010. Silver detected in mussels ranged from a low mean concentration of 0.022 ug/g dry wt. at Crockett Point, Rockland, to a high mean concentration of 0.047 ug/g dry wt. at Wadsworth Cove, Castine (Figure 1.3.1.1.1.1). Silver mean concentrations in 2010 SWAT mussels were also compared to the Gulfwatch median and 85th percentile concentrations. The mean concentration at Wadsworth Cove, Castine, exceeded the Gulfwatch median (0.037 ug/g dry wt.). None of the SWAT mean concentrations approached or exceeded the Gulfwatch 85th percentile (0.073 ug/g dry wt., Figure 1.3.1.1.1).

Figure 1.3.1.1.1.2 compares the silver concentrations in 2010 SWAT blue mussel tissue to the NS&T national median and national 85th percentile. Silver mean concentrations at all three SWAT sites fell below the NS&T national median and NS&T 85th 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). Silver concentrations in Maine mussels 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 85th percentile. Increasing use of silver, including nanosilver, in products like paints, caulking, and clothing makes monitoring silver of interest at present and in the future.

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 2010 SWAT blue mussel tissue mean silver concentration, when expressed on a wet weight basis, is 0.009 ug/g wet wt. at Wadsworth Cove, Castine. This concentration is three orders of magnitude below the 11 ug/g wet wt. FTAL.



Figure 1.3.1.1.1.1: Silver in 2010 SWAT Blue Mussels



Figure 1.3.1.1.1.2: Silver in 2010 SWAT Blue Mussels

1.3.1.1.2 Arsenic (As)

Arsenic was detected in all three sample locations visited in 2010. Arsenic levels detected in mussels ranged from a low mean concentration of 7.93 ug/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 14.83 ug/g dry wt. at Spring Point, South Portland (Figure 1.3.1.1.2.1). While Gulfwatch does not monitor arsenic concentrations, they are 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 (Kimbrough, 2008). Spring Point, South Portland, and Crockett Point, Rockland, show levels above 11 mg/kg dry wt.) for arsenic. The remaining site, Wadsworth Cove, Castine, falls in the lowest range delineated by NS&T, below 11 ug/g (ppm) dry wt.

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, 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, SWAT blue mussel data were transformed to inorganic arsenic by dividing wet weight concentrations by a factor of 10. Therefore, 2010 SWAT blue mussel inorganic arsenic concentrations are estimated to range from 0.14 ug/g wet wt. to 0.21 ug/g wet wt. All three sites exceeded the MCDC cancer FTAL of 0.014 ug/g wet wt. (ppm).

Comparing recent data from all 42 mussel sites sampled from 2007-09, 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 0.23 ug/g wet wt. (Scarborough R., 2008). All 42 SWAT sites sampled from 2007-09 had blue mussel tissue inorganic arsenic concentrations exceeding the MCDC cancer action level of 0.014 ug/g wet wt. (ppm). None of the three sites sampled in 2010 exceeded the MCDC non-cancer action level of 0.6 ug/g wet wt. (ppm) for inorganic arsenic. Similarly, none of the 42 mussel stations sampled from 2007-09 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.



Figure 1.3.1.1.2.1: Arsenic in 2010 SWAT Blue Mussels

1.3.1.1.3 Cadmium (Cd)

Cadmium was detected in all three sample locations visited in 2010. Cadmium levels detected in mussels ranged from a low mean concentration of 1.24 ug/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 2.18 ug/g dry wt. at Spring Point, South Portland (Figure 1.3.1.1.3.1). All three sites had concentrations comparable to the 2008 Gulfwatch median, with two falling below that median. Only Spring Point, South Portland, exceeded the Gulfwatch median. No sites exceeded the Gulfwatch 85th percentile (Figure 1.3.1.1.3.1).

Cadmium concentrations at one site, Spring Point, South Portland, exceeded the NS&T national median (Figure 1.3.1.1.3.1) (Kimbrough, 2008). None of the three SWAT sites sampled in 2010 had cadmium concentrations approaching or exceeding the NS&T national 85th percentile.

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, 2008).

From a human health perspective, the MCDC non-cancer FTAL for cadmium in non-commercially caught finfish is 2.2 ug/g wet wt. The FDA action level for clams, oysters, and mussels is 4 ppm wet wt. (Kimbrough, 2008). The highest scoring 2010 SWAT site, Spring Point, South Portland, had a mean cadmium concentration of 0.27 ug/g wet wt., which was well below the MCDC and FDA action levels (12% of the more conservative MCDC non-cancer FTAL).



Figure 1.3.1.1.3.1: Cadmium in 2010 SWAT Blue Mussels

1.3.1.1.4 Chromium (Cr)

Chromium was detected at all three sites sampled in 2010. Chromium levels detected in mussel tissue ranged from a low mean concentration of 2.62 ug/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 3.49 ug/g dry wt. at Spring Point, South Portland (Figure 1.3.1.1.4.1). All three SWAT site concentrations exceeded the Gulfwatch median and the Gulfwatch 85th percentile (Figure 1.3.1.1.4.1).

Figure 1.3.1.1.4.1 also depicts 2010 SWAT mussel chromium concentrations compared to the NS&T Mussel Watch national median and 85th percentile concentrations. All three SWAT sites fell above the NS&T national median, while one site, Spring Point, South Portland, exceeded the NS&T national 85th percentile and was considered elevated for chromium (Kimbrough, 2008).

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.

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

1.3.1.1.5 Copper (Cu)

Copper was detected in samples taken at all three SWAT mussel sites visited in 2010. Copper levels detected in mussels ranged from a low mean concentration of 5.55 ug/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 9.21 ug/g dry wt. at Crockett Point, Rockland (Figure 1.3.1.1.5.1). Copper concentrations exceeded the Gulfwatch median and Gulfwatch 85th percentile at two sites, Spring Point, South Portland, and Crockett Point, Rockland (LeBlanc, 2009). SWAT copper concentrations at all three sites sampled in 2010 fell below the NS&T national median, as shown in Figure 1.3.1.1.5.2 (Kimbrough, 2008).

Copper occurs naturally in the environment and is ubiquitous, including in 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 due to its phase out by EPA, newer wood preservatives utilizing even higher levels of copper have come into use, including quaternary copper. Similarly, tributyltin marine bottom paint use was reduced in the 1980s, resulting in increased use of copper based antifouling paints and asbestos removal from brake pads has been offset by increased copper usage in brake pads (Kimbrough, 2008).

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



Figure 1.3.1.1.4.1: Chromium in 2010 SWAT Blue Mussels



Figure 1.3.1.1.5.1: Copper in 2010 SWAT Blue Mussels





Figure 1.3.1.1.5.2: Copper in 2010 SWAT Blue Mussels

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

1.3.1.1.6 Iron (Fe) and Aluminum (Al)

Iron was detected in all three SWAT blue mussel sites sampled in 2010. Iron concentrations detected in mussels ranged from a low mean concentration of 514 ug/g dry wt. at Crockett Point, Rockland, to a high mean concentration of 750 ug/g dry wt. at Spring Point, South Portland as shown in Figure 1.3.1.1.6.1. Iron concentrations at all three sites exceeded the Gulfwatch median, while only Spring Point, South Portland, exceeded the Gulfwatch 85th percentile. Figure 1.3.1.1.6.1 also shows a comparison of SWAT mean iron concentrations to NS&T national median and 85th percentile iron concentrations. Iron concentrations at all three sites exceeded the NS&T national median, though none of the three sites exceeded the NS&T national 85th percentile.

Aluminum concentrations detected in mussels ranged from a low mean concentration of 308 ug/g dry wt. at Crockett Point, Rockland, to a high mean concentration of 539 ug/g dry wt. at Spring Point, South Portland (Figure 1.3.1.1.6.2). Aluminum concentrations at all three sites exceeded the Gulfwatch median concentration, while only Spring Point, South Portland, exceeded the Gulfwatch 85th percentile concentration (LeBlanc, 2009). Figure 1.3.1.1.6.2 also shows a comparison of SWAT mean aluminum concentrations to NS&T national median and 85th percentile iron concentrations. Mean aluminum concentrations at three sites exceeded the NS&T national median, while only Spring Point, South Portland, exceeded the NS&T national median, while only Spring Point, South Portland, exceeded the NS&T national median.

Spring Point, South Portland, appeared to have relatively high levels of iron and aluminum, even compared to Gulfwatch results from 2008, the most recent year available (LeBlanc, 2009). 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. Sediment loading in the gut therefore may have some effect on the Spring Point iron and aluminum tissue concentrations.

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. 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 mussel gut contents, rather than bioaccumulated contaminants from mussel tissue.

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



Figure 1.3.1.1.6.1: Iron in 2010 SWAT Blue Mussels



Figure 1.3.1.1.6.2: Aluminum in 2010 SWAT Blue Mussels

1.3.1.1.7 Nickel (Ni)

Nickel was detected at all three SWAT blue mussel sites visited in 2010. Nickel levels detected in mussels ranged from a low mean concentration of 1.37 ug/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 2.85 ug/g dry wt. at Spring Point, South Portland (Figure 1.3.1.1.7.1). Maine concentrations were all higher than the Gulfwatch median, with Spring Point, South Portland, and Crockett Point, Rockland, falling above the Gulfwatch 85th percentile.

Figure 1.3.1.1.7.1 also compares 2010 SWAT blue mussel tissue nickel concentrations to NS&T national median and national 85th percentiles to place Maine data into a national context. Maine SWAT sites had nickel concentrations distributed about the national median, with only Spring Point, South Portland, exceeding the national median. No 2010 SWAT nickel concentrations exceeded the NS&T national 85th percentile, and 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. The highest nickel concentration in the 2010 SWAT sites (Spring Point, South Portland) was also found at the same site having the highest iron and aluminum concentrations indicating sediment in the mussel 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, 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, 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 2010 of 0.35 ug/g wet wt. (ppm) at Spring Point, South Portland, is two orders of magnitude below the more conservative MCDC action level. MCDC does not report a cancer action level for nickel.



Figure 1.3.1.1.7.1: Nickel in 2010 SWAT Blue Mussels

1.3.1.1.8 Lead (Pb)

Lead was detected in all three SWAT blue mussel sites visited in 2010. Lead levels detected in mussels ranged from a low mean concentration of 0.89 ug/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 7.47 ug/g dry wt. at Crockett Point, Rockland (Figure 1.3.1.1.8.1). Only Wadsworth Cove, Castine, was less than the Gulfwatch median, with Spring Point, South Portland, and Crockett Point, Rockland, falling not only above the median but also above the Gulfwatch 85th percentile.

Figure 1.3.1.1.8.1 also compares 2010 SWAT blue mussel lead tissue concentrations to NS&T national median and national 85th percentiles to place Maine data into a national context. Crockett Point, Rockland, and Spring Point, South Portland, exceeded the NS&T national median, while Wadsworth Cove, Castine fell very near (but just below) the national median. The two higher sites at Crockett Point and Spring Point exceeded the NS&T national 85th 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. Prior samples from both Spring Point and Crockett Point taken in 2007 both show somewhat lower levels of lead in blue mussel tissue (Table 1.3.1.1.8.1). Other Maine sites with elevated lead levels 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).

Table 1.3.1.1.8.1 Lead Trends in Blue Mussels at Two Sites

Year	Spring Point, S. Portland	Crockett Point, Rockland
2007	4.36 ug/g dry wt.	5.49 ug/g dry wt.
2010	5.95 ug/g dry wt.	7.47 ug/g dry wt.

Spring Point, South Portland, is located just outside Portland Harbor and is located down current on an outgoing tide from the highly developed harbor. Major sewage treatment facilities from Portland and South Portland enter into Portland Harbor/Fore River. The oil terminal for the Portland pipeline is located just across from the Spring Point sampling station, which is also adjacent to a commercial marina/boat yard. Crockett Point, Rockland, is located in a busy commercial fishing port, and on a peninsula that has a history of commercial development. Due to the proximity of these sites to coastal development, Spring Point and Crockett Point have been selected 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, and contaminant patchiness may also be a factor in the variation in lead levels from year.

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 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, 2008). The more conservative MCDC lead FTAL in noncommercially 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 2010 Maine SWAT data, 1.27 ppm (ug/g) wet wt. at Crockett Point, Rockland, exceeds the MCDC lead FTAL, as does Spring Point, South Portland (0.73 ug/g wet wt.). Wadsworth Cove, Castine, did not exceed the MCDC FTAL for lead.

Review of the 2007-09 SWAT blue mussel sampling data from 42 sites indicates that mean lead concentrations at eight sites equaled or exceeded the MCDC lead FTAL. Sites sampled in those years equaling or exceeding the MCDC FTAL for lead are:

Spring Point, S. Portland, 2007	0.6 ppm wet wt.
Middle Fore R., Portland, 2007	0.6 ppm wet wt.
East End Beach, Portland, 2007	0.8 ppm wet wt.
Crockett Point, Rockland, 2007	1.1 ppm wet wt.
Camden Harbor, Camden, 2007	0.7 ppm wet wt.
Goose Falls, Brooksville, 2007	1.1 ppm wet wt.
Piscataqua River Back Channel, Kittery, 2008	0.6 ppm wet wt.
East End Beach, Portland, 2009	0.8 ppm wet wt.

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



Figure 1.3.1.1.8.1: Lead in 2010 SWAT Blue Mussels

1.3.1.1.9 Mercury (Hg)

Mercury was detected in all three blue mussel sample locations visited in 2010. Mercury levels detected in mussels ranged from a low mean concentration of 0.14 μ g/g dry wt. at Crockett Point, Rockland, to a high mean concentration of 0.22 μ g/g dry wt. at Spring Point, South Portland (Figure 1.3.1.1.9.1). Only Spring Point, South Portland, exceeded the 2008 Gulfwatch median, and none of the sites tested in 2010 exceeded the Gulfwatch 85th percentile.

Figure 1.3.1.1.9.1 also compares 2010 SWAT blue mussel mercury concentrations to NS&T Mussel Watch national median and 85th percentile values. The reader should note that Gulfwatch median and 85th percentile values actually exceed NS&T Mussel Watch median and 85th 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 national 85th percentile, all the SWAT sites tested in 2010 would be considered elevated for mercury, despite their more typical scores when compared to other northeast US samples from the Gulf of Maine.

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, 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 2010 was 0.027 μ g/g wet wt. (ppm) at Spring Point, South Portland. This compares favorably with the MCDC methylmercury developmental FTAL of 0.2 ppm, assuming a similar meal size and frequency. To consume approximately 8 oz. of blue mussel tissue the consumer would need to eat approximately 45-50 blue mussels based on the mean mass per mussel collected by the SWAT program.



Figure 1.3.1.1.9.1: Mercury in 2010 SWAT Blue Mussels

1.3.1.1.10 Zinc (Zn)

Zinc was detected in all three sample locations visited in 2010. Zinc levels detected in mussels ranged from a low mean concentration of 55.8 ug/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 141.5 ug/g dry wt. at Spring Point, South Portland (Figure 1.3.1.1.10.1). The SWAT blue mussel tissue zinc concentrations at Spring Point, South Portland, and Crockett Point, Rockland, exceeded both the 2008 Gulfwatch median and the Gulfwatch 85th percentile. Wadsworth Cove, Castine, had a zinc concentration well below the Gulfwatch median.

Figure 1.3.1.1.10.2 shows 2010 Maine SWAT blue mussel zinc concentrations were all below the NS&T Mussel Watch national median, and so it follows that all SWAT concentrations also fell below the NS&T 85th 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, 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 much higher than any wet wt. concentrations observed in SWAT blue mussel tissue. There is no recommended FDA safety level for zinc in fish (Kimbrough, 2008).



Figure 1.3.1.1.10.1: Zinc in 2010 SWAT Blue Mussels

Dashed line = 2008 Gulfwatch Median; Solid line = 2008 Gulfwatch 85th Percentile.

Figure 1.3.1.1.10.2: Zinc in 2010 SWAT Blue Mussels



1.3.1.2 Softshell Clams

Softshell clam tissue samples collected in 2010 were analyzed by Battelle Marine Sciences Laboratory, Sequim, WA. Clam tissues from 2004-05 were analyzed by Pace Analytical Services, Minneapolis, MN. 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, 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 eight sample locations visited. 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 (Figure 1.3.1.2.1.1). Silver mean concentrations in SWAT softshell clams were also compared to the Gulfwatch median concentration for four sites sampled in 2008 (two in Maine and two in New Hampshire). The mean concentration at Mast Cove, Eliot, exceeded the Gulfwatch median (1.32 ug/g dry wt.).

Higher silver concentrations in water and sediments coincide with municipal sewage discharge (Sanudo-Wilhelmy and Flegal, 1992; Buchholtz ten Brink et al., 1997). 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. Increasing use of silver, including nanosilver, in products such as clothing and paints and caulks, makes monitoring silver of interest at present and in the future.

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. 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 at Morse Cove, Castine, the only site tested in 2010 and the only softshell clam site where tissue was tested for arsenic. This was not part of the metals data provided by the previous SWAT contract laboratory when clam sampling was completed in 2004-05. The mean arsenic concentration in Morse Cove clams was 9.97 ug/g dry wt. While Gulfwatch does not monitor arsenic in blue mussels 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 (Kimbrough, 2008). The mean arsenic level in softshell clams at Morse Cove fell into this range, although as noted this is for mussels/oysters. However, it is of interest to give a point of comparison for Maine clam data.

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, 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, 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, SWAT softshell clam data were transformed to inorganic arsenic by dividing wet weight concentrations by a factor of 10. Therefore, the Morse Cove clam inorganic arsenic mean concentration is estimated to be 0.16 ug/g wet wt., which exceeds the MCDC cancer FTAL of 0.014 ug/g wet wt. (ppm). Note that all blue mussel sites sampled since arsenic data has been recorded as part of the SWAT program also exceed the MCDC cancer FTAL. The Morse Cove estimated mean inorganic arsenic concentration does not exceed the MCDC non-cancer action level of 0.6 ug/g wet wt. (ppm) for inorganic arsenic. MCDC non-commercially caught finfish FTALs applied here assume an 8 oz. meal eaten by the consumer on a weekly basis.

1.3.1.2.3 Cadmium (Cd)

Cadmium was detected in tissue from all eight clam locations visited. Cadmium levels detected in softshell clams ranged from a low mean concentration of 0.31 ug/g dry wt. at Squirrel Island, Southport, to a high mean concentration of 0.77 ug/g dry wt. at Mill Cove, Robbinston (Figure 1.3.1.2.3.1). Only Mill Cove and Navy Pier, Harpswell, approached the 2008 Gulfwatch median, with all eight sites falling below that median. 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, 2008).

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



Figure 1.3.1.2.1.1: Silver in SWAT Softshell Clams



Figure 1.3.1.2.3.1: Cadmium in SWAT Softshell Clams

1.3.1.2.4 Chromium (Cr)

Chromium was detected at all eight sites sampled. Chromium levels detected in clam tissue ranged from a low mean concentration of 3.67 ug/g dry wt. at Long Cove, Searsport, to a high mean concentration of 13.32 ug/g dry wt. at Mast Cove, Eliot (Figure 1.3.1.2.4.1).

Figure 1.3.1.2.4 depicts SWAT softshell clam chromium concentrations compared to the Gulfwatch 2008 median concentration for four sites (two each in ME and NH). All clam sites but one, Long Cove, Searsport, fell above the Gulfwatch 2008 median. The Fort Point Cove, Stockton Springs, clam tissue chromium concentration was essentially the same as the Gulfwatch 2008 median.

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, 2008).

From a human health perspective, the MCDC FTALs (7 ug/g cancer action level and 11 ug/g noncancer 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 eight SWAT softshell clam sites visited. 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 13.07 ug/g dry wt. at Mast Cove, Eliot (Figure 1.3.1.2.5.1). Copper concentrations in clam tissue at all eight sites fell below the 2008 Gulfwatch median (LeBlanc, 2009).

Copper occurs naturally in the environment and is ubiquitous, including in 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 due to its phase 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, 2008).

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



Figure 1.3.1.2.4.1: Chromium in SWAT Softshell Clams



Figure 1.3.1.2.5.1: Copper in SWAT Softshell Clams

1.3.1.2.6 Iron (Fe) and Aluminum (Al)

Iron was detected in all eight SWAT softshell clam sites. 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 as shown in Figure 1.3.1.2.6.1. No SWAT sites had clam tissue iron concentrations that exceeded the 2008 Gulfwatch median (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 1,623 ug/g dry wt. at Morse Cove, Castine (Figure 1.3.1.2.6.2). None of the clam tissue from the eight sites had aluminum concentrations exceeding the 2008 Gulfwatch mean concentration.

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



Figure 1.3.1.2.6.2: Aluminum in SWAT Softshell Clams

1.3.1.2.7 Nickel (Ni)

Nickel was detected in clam tissue at all eight SWAT softshell clam sites visited. Nickel levels detected in mussels ranged from a low mean concentration of 3.01 ug/g dry wt. at Long Cove, Searsport, to a high mean concentration of 9.68 ug/g dry wt. at Mast Cove, Eliot (Figure 1.3.1.2.7.1). Maine concentrations were all higher than the 2008 Gulfwatch clam median.

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, 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, 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 eight SWAT softshell clam sites visited. 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 5.45 ug/g dry wt. at Harris Cove, Eastport (Figure 1.3.1.2.8.1). Mean lead clam tissue concentrations at all eight SWAT sites fell below the 2008 Gulfwatch median.

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 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).



Figure 1.3.1.2.7.1: Nickel in SWAT Softshell Clams



Figure 1.3.1.2.8.1: Lead in SWAT Softshell Clams

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, 2008). The more conservative MCDC lead FTAL in noncommercially 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, 0.765 ppm (ug/g) wet wt. at Harris Cove, Eastport, exceeds the MCDC lead FTAL, as does Fort Point Cove, Searsport (0.647 ug/g wet wt.). Mast Cove, Eliot, (0.597 ug/g wet wt.) is at the MCDC lead FTAL. The other five SWAT softshell clam sites fell below the more conservative 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 eight softshell clam sample locations visited. Mercury levels detected in clams ranged from a low mean concentration of 0.06 μ g/g dry wt. at Harris Cove, Eastport, to a high mean concentration of 0.64 μ g/g dry wt. at Fort Point Cove, Stockton Springs (Figure 1.3.1.2.9.1). Four sites had clam tissue concentrations that exceeded the 2008 Gulfwatch mean: Mast Cove, Eliot; Long Cove, Searsport; Fort Point Cove, Stockton Springs; and Morse Cove, Castine (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, 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 μ g/g wet wt. (ppm) at Fort Point Cove, Stockton Springs. 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 21 softshell clams based on the mean mass per clam collected by the SWAT program.



Figure 1.3.1.2.9.1: Mercury in SWAT Softshell Clams

1.3.1.2.10 Zinc (Zn)

Zinc was detected in all eight clam sample locations. 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 85.0 ug/g dry wt. at Fort Point Cove, Stockton Springs (Figure 1.3.1.2.10.1). All eight of the SWAT clam sites had zinc tissue concentrations that fell below the 2008 Gulfwatch median.

Zinc is a 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, 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, 2008).



Figure 1.3.1.2.10.1: Zinc in SWAT Softshell Clams

1.3.2 PAHs

1.3.2.1 Blue Mussels

When available, results were compared to national (NOAA National Status & Trends, see Kimbrough, 2008) and Gulf of Maine (Gulfwatch, see LeBlanc, 2009) blue mussel monitoring program data (when available) in an effort to place Maine SWAT data in a national and regional context.

The NS&T and the 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. This report utilizes the Maine SWAT blue mussel tissue PAH data generated by AXYS Analytical, which includes 74 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 18 unsubstituted (non-alkylated) PAHs from 2010 SWAT The difference in one PAH counted is because SWAT results include data. BENZO[B,J,K]FLUORANTHENES, while the Gulfwatch and NS&T results include both BENZO[B]FLUORANTHENES and BENZO[K]FLUORANTHENES individually. This slight difference is not considered to be important in comparing overall summary concentrations of PAHS Though the sum of 18 PAHs in SWAT and 19 PAHs in for purposes of this report. NS&T/Gulfwatch 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 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). Due to the previously outlined difference regarding BENZO[B,J,K]FLUORANTHENES, the SWAT PAH summation used to compare to the Gulfwatch/NS&T summation of 24 PAHs actually contains 23 PAHs for SWAT.

The 2010 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 38 PAHs, which is the closest approximation possible. As noted previously, one discrepancy is the BENZO[B,J,K]FLUORANTHENES. The second difference in the 40 PAH summation is the absence of C4-Flourenes in the SWAT data set. As a result, the SWAT summation includes 38 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 2010 PAH data includes additional alkylated PAHs as well, with a total of 74 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. 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. It 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 2010 (SWAT data, 74 PAHs described above).

Figure 1.3.2.1.1 shows the summation of the 19 non-alkylated PAHs compared to the summation of all 74 PAHs (including many alkylated PAHs) at the three blue mussel sites sampled by SWAT in 2010. 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 two data series illustrated on the graph in the figure. The sum of 19 non-alkylated PAHs varied from 17% (Wadsworth Cove, Castine) to 30% (Crockett Point, Rockland) of total PAHs (74) across the 3 SWAT sites.

Total PAH concentrations ranged from a low mean concentration of 287 ng/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 1,756 ng/g dry wt. at Crockett Point, Rockland (Figure 1.3.2.1.1). Spring Point, South Portland (1,514 ng/g dry wt.), had the second highest total PAH concentrations of the three sites sampled. The sum of 19 non-alkylated PAHs varied from a concentration of 49 ng/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 523 ng/g dry wt. at Crockett Point, Rockland.

TABLE 1.3.2.1.1: Analyzed PAHs and PAH Summation Calculations

	SWAT Gulfwatch, NS&T, SWAT Summations						
Parameter	2004-05	2010	Sum PAH1	Sum PAH2	Sum PAH4	Not Analyzed	Notes (See below list for more notes)
ACENAPHTHENE	Х	Х	Х	Х	Х		
ACENAPHTHYLENE	Х	Х	Х	Х	Х		
ANTHRACENE	Х	Х	Х	Х	Х		
2-METHYLANTHRACENE		Х				missing	
BENZ[A]ANTHRACENE	Х	Х	Х	Х	Х		
DIBENZ(A,H)ANTHRACENE	Х	Х	Х	Х	Х		
BIPHENYL	Х	Х	х	Х	Х		
BENZO[A]PYRENE	Х	Х	х	Х	Х		
BENZO(E)PYRENE	Х	Х	Х	Х	Х		
7-METHYLBENZO[A]PYRENE		Х				missing	
CHRYSENE	Х	Х	х	Х	Х		
1-METHYLCHRYSENE		Х				missing	
5/6-METHYLCHRYSENE		Х				missing	
5,9-DIMETHYLCHRYSENE		Х				missing	
DIBENZOTHIOPHENE		Х	Х	Х	Х		
2,4-DIMETHYLDIBENZOTHIOPHENE		Х				missing	
2/3-METHYLDIBENZOTHIOPHENES		Х				missing	
FLUORANTHENE	Х	Х	х	Х	Х		
BENZO[B,J,K]FLUORANTHENES		х	x	x	x		in Gulfwatch list as BENZO[B]FLUORANTHENE and BENZO[K]FLUORANTHENE
3-METHYLFLUORANTHENE/BENZO[A]FLUORENE		Х					
FLUORENE	Х	Х	х	Х	Х		
2-METHYLFLUORENE		Х				missing	
1,7-DIMETHYLFLUORENE		х				missing	

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

SWAT Gulfwatch, NS&T, SWAT Summations							
Parameter	2004-05	2010	Sum PAH1	Sum PAH2	Sum PAH4	Not Analyzed	Notes (See below list for more notes)
NAPHTHALENE	Х	Х	Х	Х	х		
1-METHYLNAPHTHALENE		х				missing	
2-METHYLNAPHTHALENE		Х				missing	
1,2-DIMETHYLNAPHTHALENE		х				missing	
2,6-DIMETHYLNAPHTHALENE		Х				missing	
2,3,5-TRIMETHYLNAPHTHALENE		х				missing	
2,3,6-TRIMETHYLNAPHTHALENE		х				missing	
1,4,6,7-TETRAMETHYLNAPHTHALENE		Х				missing	
PERYLENE	х	х		х	х		
BENZO[GHI]PERYLENE	х	х	х	х	х		
PHENANTHRENE	х	Х	Х	Х	х		
1-METHYLPHENANTHRENE		х				missing	
2-METHYLPHENANTHRENE		х				missing	
3-METHYLPHENANTHRENE		х				missing	
9/4-METHYLPHENANTHRENE		Х				missing	
1,7-DIMETHYLPHENANTHRENE		х				missing	
1,8-DIMETHYLPHENANTHRENE		х				missing	
2,6-DIMETHYLPHENANTHRENE		Х				missing	
3,6-DIMETHYLPHENANTHRENE		х				missing	
1,2,6-TRIMETHYLPHENANTHRENE		х				missing	
PYRENE	х	Х	х	х	х		
INDENO[1,2,3-CD]PYRENE	х	х	х	x	x		

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

	SWAT Gulfwatch, NS&T, SWAT Summations						
Parameter	2004-05	2010	Sum PAH1	Sum PAH2	Sum PAH4	Not Analyzed	Notes (See below list for more notes)
RETENE		х				missing	
C1-ACENAPHTHENES		х				missing	
C1-BENZO[A]ANTHRACENES/CHRYSENES		Х			Х		in Gulfwatch list as C1-CHRYSENE
C2-BENZO[A]ANTHRACENES/CHRYSENES		Х			Х		in Gulfwatch list as C2-CHRYSENE
C3-BENZO[A]ANTHRACENES/CHRYSENES		х			Х		in Gulfwatch list as C3-CHRYSENE
C4-BENZO[A]ANTHRACENES/CHRYSENES		х			Х		in Gulfwatch list as C4-CHRYSENE
C1-BENZOFLUORANTHENES/BENZOPYRENES		Х				missing	
C2-BENZOFLUORANTHENES/BENZOPYRENES		х				missing	
C1-BIPHENYLS		х				missing	
C2-BIPHENYLS		х				missing	
C1-DIBENZOTHIOPHENES		х			х		
C2-DIBENZOTHIOPHENES		х			х		
C3-DIBENZOTHIOPHENES		х			х		
C4-DIBENZOTHIOPHENES		х				missing	
C1-FLUORANTHENES/PYRENES		х			х		
C2-FLUORANTHENES/PYRENES		х			х		
C3-FLUORANTHENES/PYRENES		х				missing	
C4-FLUORANTHENES/PYRENES		х				missing	
C1-FLUORENES		х			х		
C2-FLUORENES		х			х		
C3-FLUORENES		х			х		
C1-NAPHTHALENES		х		х	Х		
C2-NAPHTHALENES		Х		Х	Х		
C3-NAPHTHALENES		Х		Х	Х		
C4-NAPHTHALENES		х				missing	

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

	SV	/AT	Gulfw	atch, NS&T	', SWAT Su	mmations	
Parameter	2004-05	2010	Sum PAH1	Sum PAH2	Sum PAH4	Not Analyzed	Notes (See below list for more notes)
C1-PHENANTHRENES/ANTHRACENES		х		х	х		in Gulfwatch list as C1-PHENANTHRENE
C2-PHENANTHRENES/ANTHRACENES		Х			х		in Gulfwatch list as C2-PHENANTHRENE
C3-PHENANTHRENES/ANTHRACENES		Х			х		in Gulfwatch list as C3-PHENANTHRENE
C4-PHENANTHRENES/ANTHRACENES		х			х		in Gulfwatch list as C4-PHENANTHRENE
C4-FLUORENES					х		Not analyzed by SWAT

FOOTNOTES:

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

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)

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



Figure 1.3.2.1.1: Sum of 19 PAHs and Total PAHs at 2010 SWAT Blue Mussel Sites

Figure 1.3.2.1.2 presents the sum of 19 PAHs across the SWAT blue mussel sites sampled in 2010, and compares these results with Gulfwatch 2008 median and 85th percentile results. Of the three SWAT sites tested in 2010, Crockett Point, Rockland, and Spring Point, South Portland, exceeded the Gulfwatch 2008 median of 154 ng/g (dry weight) for 19 summed PAHs. Only one site, Crockett Point, Rockland, exceeded the Gulfwatch 85th percentile of 429 ng/g (dry weight) for 19 summed PAHs. The remaining site at Spring Point, South Portland, was essentially the same as the Gulfwatch 85th percentile concentration. Despite the use of 18 summed PAHs (SWAT) to compare to 19 summed PAHs utilized in the Gulfwatch program, the summation of non-alkylated PAHs is useful for putting Maine data into a regional, Gulf of Maine context.

Figure 1.3.2.1.2 also compares the sum of 19 non-alkylated PAHs at the 2010 SWAT sites to recent NS&T median and 85th percentile for 19 summed non-alkylated PAHs (2008 data, the most recent available). Of the three SWAT sites tested in 2010, Crockett Cove, Rockland, and Spring Point, South Portland, exceeded the 2008 NS&T national median of 180 ng/g (dry weight) for 19 summed non-alkylated PAHs. None of the three SWAT mussel sites approached or exceeded the NS&T national 85th percentile of 1,104 ng/g (dry weight) for 19 summed PAHs.

The Gulfwatch program also utilized a summation of 24 PAHs in reports, the composition of which is outlined above. SWAT data was converted into this format and when 24 PAHs were summed, 2010 SWAT mean concentrations ranged from 25% (Wadsworth Cove, Castine) to 36% (Crockett Point, Rockland) of total PAHs (74) across the three SWAT mussel sites.

The mean concentrations for the sum of 24 PAHs ranged from a low mean concentration of 73 ng/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 641ng/g dry wt. at Crockett Point, Rockland (Figure 1.3.2.1.3). Figure 1.3.2.1.4 presents the sum of 24 PAHs across the SWAT blue mussel sites sampled in 2010, and compares these results with Gulfwatch 2008 median and 85th percentile results. Of the three SWAT sites tested in 2010, Crockett Point, Rockland, and Spring Point, South Portland, are the two sites that exceeded the Gulfwatch 2008 median of 198 ng/g (dry weight) for 24 summed PAHs. Only one site, Crockett Point, Rockland, also exceeded the Gulfwatch 85th percentile of 476 ng/g (dry weight) for 24 summed PAHs, though Spring Point, South Portland was essentially the same as the Gulfwatch 85th percentile. Wadsworth Cove, Castine, was well below the Gulfwatch median concentration of 198 ng/g (dry weight) for 24 summed PAHs. Despite the use of 23 summed PAHs (SWAT) to compare to 24 summer PAHs utilized in the Gulfwatch program, the summation of these PAHs is useful for putting Maine data into a regional, Gulf of Maine context.

Figure 1.3.2.1.4 also compares the sum of 24 PAHs at the 2010 SWAT sites to recent NS&T median and 85th percentile for 24 summed PAHs (2008 data, the most recent available). Of the three SWAT sites tested in 2010, Crockett Point, Rockland, and Spring Point, South Portland, exceeded the NS&T national 2008 median of 247 ng/g (dry weight) for 24 summed PAHs. None of the 2010 SWAT sites approached or exceeded the NS&T national 85th percentile of 1,216 ng/g (dry weight) for 24 summed PAHs.



Figure 1.3.2.1.2: Sum of 19 PAHs in 2010 SWAT Blue Mussels



Figure 1.3.2.1.3: Sum of 24 PAHs and Sum of Total PAHs at 2010 SWAT Blue Mussel Sites



Figure 1.3.2.1.4: Sum of 24 PAHs in 2010 SWAT Blue Mussels

Figure 1.3.2.1.5 shows the summation of 40 PAHs compared to the summation of all 74 PAHs at the three blue mussel sites sampled by SWAT in 2010. 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 sum of 40 PAHs varied from 76% (Wadsworth Cove, Castine) to 81% (Crockett Point, Rockland) of total PAHs (74) across the three SWAT sites. The mean concentrations for the sum of 40 PAHs ranged from a low mean concentration of 217 ng/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 1,417 ng/g dry wt. at Crockett Point, Rockland (Figure 1.3.2.1.5).

Figure 1.3.2.1.6 presents the sum of 40 PAHs across the SWAT blue mussel sites sampled in 2010, and compares these results with Gulfwatch 2008 median and 85th percentile results. Of the three SWAT sites tested in 2010, two exceeded the Gulfwatch 2008 median of 260 ng/g (dry weight) for 40 summed PAHs. Two sites also exceeded the Gulfwatch 85th percentile of 618 ng/g (dry weight) for 40 summed PAHs. The differences between the SWAT list of PAHs and the Gulfwatch list of PAHs available for the sum of 40 PAHs may be part of the reason why the SWAT sum of 40 PAHs is comparably high to the Gulfwatch sum of 40 PAHs. As noted in Table 1.3.2.1.1, SWAT utilizes C1 through C4-Benzo[A]Anthracenes/Chrysenes, where Gulfwatch utilizes C1 through C4-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 exact 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 1.3.2.1.6.

Figure 1.3.2.1.6 also compares the sum of 40 PAHs at the 2010 SWAT sites to recent NS&T median and 85th percentile for 40 summed PAHs (2008 data, the most recent available). Of the three SWAT sites tested in 2010, two sites (Crockett Point, Rockland, and Spring Point, South Portland), exceeded the NS&T national 2008 median of 353 ng/g (dry weight) for 40 summed PAHs. None of the three sites exceeded the NS&T national 85th 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 high concentrations apparent in two of the SWAT sites when compared to the NS&T (same as Gulfwatch) sum of 40 PAHs. These differences are explored in depth in the 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 1.3.2.1.6.


Figure 1.3.2.1.5: Sum of 40 PAHs and Total PAHs at 2010 SWAT Blue Mussel Sites



Figure 1.3.2.1.6: Sum of 40 PAHs in 2010 SWAT Blue Mussels

For 2010 SWAT blue mussel sites, Figure 1.3.2.1.7 presents a graphic representation of selected PAHs expressed as a ratio. The equation used to derive the ratio is:

Fluoranthene + Pyrene/ Σ (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.

Of the three SWAT blue mussel sites tested in 2010, Wadsworth Cove, Castine had the lowest ratio calculated, 0.33. Even this lowest ratio site did not fall below the <0.1 mark, which would indicate a petrogenic source of PAHs. It is of interest to note the proximity of the Spring Point, South Portland site to the oil terminal for the pipeline located just north of the sampling station. Despite this proximity to nearly daily tanker unloading, PAHs sources here appear to be fairly strongly pyrogenic and may be attributed to the urbanized upland area (Portland and South Portland, with associated impervious surfaces and combusted hydrocarbon runoff) or to ship and boat emissions. Wadsworth Cove, Castine, with its lower ratio of 0.33, may be somewhat less exposed to sources of pyrogenic PAHs, being geographically removed from larger urban areas.

PAHs occur in elevated concentrations near petroleum manufacturing, creosote use, and wood burning (Kimbrough, 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, 2008).

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 1.3.2.1.7: Flu+Pyr/Sum(FP C2-C4-P) in SWAT 2010 Blue Mussels and Softshell Clams

0 = Petroleum, 1 = Pyrogenic; Generally Interpreted as >.2 = Pyrogenic, <.1 = Petroleum

1.3.2.2. Softshell Clams

Results were compared to national (NOAA National Status & Trends, see Kimbrough, 2008) shellfish data and Gulf of Maine (Gulfwatch, see LeBlanc, 2009) softshell clam data (when available) in an effort to place Maine SWAT data in a national and regional context.

Differences in individual PAHs obtained from different laboratories and different years are described in depth in the previous section 1.3.2.1, PAHs in 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" which also was presented in the previous section 1.3.2.1, 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. It 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 eight SWAT clam sites. Sum of 19 non-alkylated PAHs ranged from a low mean concentration of 88 ng/g dry wt. at Long Cove, Searsport, to a high mean concentration of 319 ng/g dry wt. at Morse Cove, Castine. No 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).

Due to the lesser number of PAHs for which lab analysis was conducted in 2004-05, no summations for 24 or 40 PAHs are available for clam tissues collected in these years. Due to the larger number of PAHs from lab analysis in 2010, all summations can be constructed for Morse Cove, Castine, which was sampled in 2010. Figure 1.3.2.2.2 includes summations for 19 non-alkylated PAHs, as well as summations of 24, 40, and total PAHs. The sum of 40 PAHs concentration in Morse Cove clam tissue exceeded the 2008 Gulfwatch median (four sites, two in NH and two in ME), though the summations of 19 and 24 PAHs did not exceed the mean. This may indicate a component of 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 1.3.2.2.2.

Only PAH results from Morse Cove, Castine (2010), included the PAHs necessary to calculate the ratio used previously to explore non-alkylated to alkylated PAHs. Sites sampled in 2004-05 were not analyzed for all necessary PAHs to complete this ratio calculation. The equation used to derive the ratio is:

Fluoranthene + Pyrene/ Σ (Flouranthene + Pyrene + C2-C4 Alkylphenanthrene)



Figure 1.3.2.2.1: Sum of 19 PAHs in SWAT Softshell Clams

Solid line = Gulfwatch 2008 Mean (four Softshell Clam Sites in NH, ME)



Figure 1.3.2.2.2: PAHs in Morse Cove, Penobscot/Castine, Softshell Clams

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 only Morse Cove, Castine, had the necessary PAH data to calculate the ratio, it has been included in Figure 1.3.2.1.7, which also depicts the calculated ratios for the blue mussel sites sampled in 2010 and is in the previous section of the report, section 1.3.2.1. Morse Cove appears to have a predominantly pyrogenic PAH signature, scoring between 0.3 and 0.4 in Figure 1.3.2.1.7.

PAHs occur in elevated concentrations near petroleum manufacturing, creosote use, and wood burning (Kimbrough, 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, 2008).

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).

1.3.2.3. American Lobster

Summations of 19 and total PAHs are described in depth in Section 1.3.2.1, PAHs in Blue Mussels. The same approach was utilized to develop lists of PAHs in lobster tissues presented in this section. Table 1.3.2.1.1, "Analyzed PAHs and PAH Summation Calculations" which also was presented in the previous section 1.3.2.1, shows SWAT PAH summation lists and includes a complete list of all PAHs for which results were obtained.

Figure 1.3.2.3.1 shows the summation of 19 non-alkylated PAHs compared to total PAHs in lobster muscle tissue at the 19 SWAT lobster stations sampled in 2010. Sum of 19 non-alkylated PAHs ranged from a low mean concentration of 12 ng/g dry wt. at Pleasant Bay to a high mean concentration of 121 ng/g dry wt. at Luckse Sound, Casco Bay. Total PAHs ranged from a low mean concentration of 85 ng/g dry wt. at Great Wass Island, Western Bay, to a high mean concentration of 402 ng/g dry wt. at Eastport, Cobscook Bay.

Figure 1.3.2.3.1 also demonstrates the small percentage of 74 total PAHs comprised by the Sum of 19 non-alkylated PAHs. Luckse Sound, Casco Bay, appears to exhibit a somewhat higher percentage of non-alkylated PAHs than many of the other sites. This may indicate more petrogenic sources of PAHs near the site, which is just outside Portland Harbor.

Figure 1.3.2.3.2 shows the summation of 19 non-alkylated PAHs compared to total PAHs in lobster hepatopancreas at the 19 SWAT lobster stations sampled in 2010. Sum of 19 non-alkylated PAHs ranged from a low mean concentration of 64 ng/g dry wt. at Flye Island, Blue Hill Bay, to a high

mean concentration of 1,447 ng/g dry wt. at Luckse Sound, Casco Bay. Total PAHs ranged from a low mean concentration of 317 ng/g dry wt. at Pleasant Bay to a high mean concentration of 4,021 ng/g dry wt. at Luckse Sound, Casco Bay. Figure 1.3.2.3.2 also shows the higher percentage of non-alkylated PAHs present in lobster tissues in Luckse Sound, Casco Bay.

Figure 1.3.2.3.3 compares the sum of 19 non-alkylated PAHs in both lobster muscle and hepatopancreas tissues. The sum of 19 PAHs averages 7 times higher in hepatopancreas tissue than in muscle tissue, with the difference varying from 4 to 12 times higher in hepatopancreas at different stations.

Figure 1.3.2.3.4 compares total PAHs in both lobster muscle and hepatopancreas tissues. Total PAHs average 5 times higher in hepatopancreas tissue than in muscle tissue, with the difference varying fom 3 to 10 times higher in hepatopancreas at different stations. Both figures demonstrate the partitioning of PAHs into the hepatopancreas, which serves as the lobster's liver and pancreas and sequesters organic contaminants within the lobster. It has a high lipid content, which contributes to its high PAH content since PAHs (as organic contaminants) are lipophilic.

Lobster tissue PAH data was used to calculate the ratio used previously to explore the relationship between non-alkylated and alkylated PAHs. All 19 lobster stations were included since the necessary PAHs were available to complete this ratio calculation. The equation used to derive the ratio is:

Fluoranthene + Pyrene/ Σ (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.







Figure 1.3.2.3.2: Sum of 19 PAHs and Total PAHs in SWAT Lobster Hepatopancreas



Figure 1.3.2.3.3: Sum of 19 PAHs in SWAT Lobster Muscle and Hepatopancreas



Figure 1.3.2.3.4: Sum of Total PAHs in SWAT Lobster Muscle and Hepatopancreas

Figure 1.3.2.3.5 depicts the calculated ratios for lobster muscle tissue. Belfast Bay; Eggemoggin Reach; West Swans Island, Jericho Bay; and Eastport all show a ratio of less than 0.1, and appear to have a predominantly petrogenic PAH signal. Nine stations had ratios above 0.2, indicating a primarily pyrogenic PAH signal, including all five Casco Bay sites. Luckse Sound had the highest ratio at 0.6. It is of interest to note the proximity of the Luckse Sound station to the tanker traffic proceeding to the oil terminal for the pipeline located in Portland Harbor. Despite this proximity to nearly daily tanker traffic, PAH sources here appear to be dominated by pyrogenic sources and may be attributed to the urbanized upland area (Portland and South Portland, with associated impervious surfaces and combusted hydrocarbon runoff) or to ship and boat emissions. The four stations with ratios below 0.1 may be somewhat less exposed to sources of pyrogenic PAHs, being geographically removed from larger urban areas.

Figure 1.3.2.3.6 shows the calculated ratios for lobster hepatopancreas tissue. Only West Swans Island, Jericho Bay, had a ratio below 0.2. All other stations exhibit a ratio suggesting more pyrogenic sources of PAHs. This is a different pattern than exhibited by the muscle tissue, and may indicate sequestration of alkylated PAHs by the hepatopancreas tissue at many stations where muscle meat shows less alkylated PAHs as expressed in this ratio calculation.

PAHs occur in elevated concentrations near petroleum manufacturing, creosote use, and wood burning (Kimbrough, 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, 2008).

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 1.3.2.3.5: Flu+Pyr/Sum(FP C2-C4-P) in SWAT Lobster Muscle

^{0 =} Petroleum, 1 = Pyrogenic; Generally Interpreted as >.2 = Pyrogenic, <.1 = Petroleum.



Figure 1.3.2.3.6: Flu+Pyr/Sum(FP C2-C4-P) in SWAT Lobster Hepatopancreas

0 = Petroleum, 1 = Pyrogenic; Generally Interpreted as >.2 = Pyrogenic, <.1 = Petroleum.

1.3.3 PCBs

1.3.3.1 Blue Mussels

Blue mussels were tested for PCBs from all three sites where mussels were collected in 2010 and were analyzed by AXYS Analytical Services Ltd., Sidney, British Columbia. Mussel tissue samples were analyzed for 209 PCBs using EPA Method 1668A. Results were compared to national (NOAA National Status & Trends, see Kimbrough, 2008) and Gulf of Maine (Gulfwatch, see LeBlanc, 2009) blue mussel monitoring program data (when available) in an effort to place Maine SWAT data in a national and regional context.

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

To compare what proportion of the total PCBs (209 congeners) the SWAT PCBs represent, Figure 1.3.3.1.1 shows both the total PCBs next to the SWAT PCBs list used for comparison to other data sets like Gulfwatch and NS&T Musselwatch.Comparing the three mussel sites sampled in 2010, the SWAT PCBs ranged from 36% to 39% of the total PCBs. The close relationship between total PCBs and the SWAT PCBs subset for 2010 can easily be noted in Figure 1.3.3.1.1.Total PCB concentrations ranged from a low mean concentration of 25.6 ng/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 98.1 ng/g dry wt. at Crockett Point, Rockland (Figure 1.3.3.1.1). Spring Point, South Portland, had a concentration of total PCBs of 70 ng/g dry wt. (Figure 1.3.3.1.1).

Figure 1.3.3.1.2 compares the SWAT PCBs at the 2010 SWAT mussel sites to recent Gulfwatch median and 85th percentile for Gulfwatch PCBs (2008 data, the most recent available). Of the three SWAT sites, Crockett Point, Rockland, and Spring Point, South Portland, exceeded the Gulfwatch

2008 median of 24.1 ng/g (dry weight) for Gulfwatch PCBs. None of the three sites tested in 2010 exceeded the Gulfwatch 85th percentile of 35.4 ng/g (dry weight) for Gulfwatch PCBs, though Crockett Point, Rockland, was comparable to the Gulfwatch 85th percentile with a concentration of 35.2 ng/g dry wt. Wadsworth Cove, Castine, had a SWAT PCB concentration below the Gulfwatch PCB 2008 median. 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. Despite these differences, the summation of 35 SWAT congeners is useful for putting Maine data into a regional, Gulf of Maine context.

Figure 1.3.3.1.2 also compares the SWAT PCBs at the 2010 SWAT sites to recent NS&T (NS&T) median and 85th percentile for NS&T PCBs (2008 data, the most recent available). Of the three SWAT sites, only Crockett Point, Rockland, exceeded the NS&T 2008 national median, 29.2 ng/g (dry weight), for NS&T PCBs. However, Crockett Point and Spring Point, South Portland, were very close to the NS&T national median. No 2010 SWAT mussel sites approached or exceeded the NS&T national 85th percentile, 141 ng/g (dry weight), for NS&T PCBs. The 2008 NS&T national 85th percentile was approximately 4 X higher than the highest scoring PCB site tested by SWAT in Maine in 2010, Crockett Point, Rockland (35.2 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, 2008).

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 electrical equipment and transformers (Kimbrough, 2008).

From a human health perspective, the MCDC cancer FTAL for total PCBs for non-commercially caught finfish is 11 ng/g wet wt. (ppb), while the MCDC non-cancer FTAL for total PCBs is 43 ng/g wet wt. (ppb). Of the three SWAT blue mussel sites sampled in 2010, one site had total PCB mean tissue concentrations equal to or exceeding the MCDC cancer FTAL of 11 ng/g wet wt. This site was:

Crockett Point, Rockland, 2010 Crockett Point, Rockland, 2007 15 ng/g wet wt. 23 ng/g wet wt.

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

SUM 35 PCBs	SUM 31 PCBs	
"SWAT PCBs" List	"Gulfwatch, NS&T PCBs"	
	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		
Unique to SWAT 35 List	Unique to GW and	
	NS&T 31 List	
PCB-30	PCB-44	
PCB-26	PCB-95	

PCB-53	
PCB-20	
PCB-166	
PCB-193	

Unique to O w and
NS&T 31 List
PCB-44
PCB-95
PCB-87
PCB-138



Figure 1.3.3.1.1: SWAT PCBs (Sum of 35 PCBs) and Total PCBs at 2010 SWAT Blue Mussel Sites



Figure 1.3.3.1.2: SWAT PCBs (Sum of 35 PCBs) in 2010 SWAT Blue Mussels

ashed lines = 2008 Gulfwatch Median and 85th Percentile (Gulfwatch PCBs, Sum 31); Solid lines = 2008 National Status and Trends Median and 85th Percentile (Sum 31). None of the three SWAT blue mussel sites sampled had total PCB concentrations approaching the MCDC non-cancer FTAL of 43 ng/g wet wt.

When Crockett Point, Rockland, was tested previously in 2007, the mean concentration at the site was 23 ng/g wet wt., indicating some inter-annual variability in total PCB concentration. This variability may be a result of patchiness and heterogeneity within site, or differences between sampling years. Not enough information is available to suggest a trend in the PCB concentrations, but future sampling will be undertaken to better understand PCBs levels at Crockett Point and its vicinity.

1.3.3.2 Softshell Clams

Softshell clams were tested for PCBs from three sites: Long Cove, Searsport; Morse Cove, Castine; and Mill Cove, Robbinston. Morse Cove samples (collected in 2010) were analyzed by AXYS Analytical Services Ltd., Sidney, British Columbia. Long Cove and Mill Cove (collected in 2005) were analyzed by Pace Analytical, Minneapolis, MN. Clam tissue samples were analyzed for 209 PCBs using EPA Method 1668A. Results were compared to Gulf of Maine (Gulfwatch, see LeBlanc, 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 in the blue mussel PCB section. The same approach was utilized to construct clam PCB summations and will not be repeated in this section.

Table 1.3.3.1.1 (in Section 1.3.3.1) 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 blue mussel 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 both the total PCBs next to the SWAT PCBs list used for comparison to Gulfwatch. Comparing the three clam sites sampled in 2010, the SWAT PCBs ranged from 47% to 53% of the total PCBs. The close relationship between total PCBs and the SWAT PCBs subset for 2010 clam tissue can easily be noted in Figure 1.3.3.2.1. Total PCB concentrations ranged from a low mean concentration of 3.5 ng/g dry wt. at Mill Cove, Robbinston, to a high mean concentration of 15.7 ng/g dry wt. at Morse Cove, Castine (Figure 1.3.3.2.1).



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

Mill Cove, Robbinston, and Long Cove, Stockton Springs, were sampled in 2005, and analyzed at a different lab than the Morse Cove, Castine, clam tissue from 2010. These two sites had much higher detection limits than those generated by the newer lab (Axys Analytical) that worked up the recent Morse Cove sample. 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 1.3.3.2.2 compares the SWAT PCBs at the 2010 SWAT clam sites to a recent Gulfwatch clam site sampled in 2008 (the most recent available). All three SWAT clam site sums of 35 PCBs fell below the one Gulfwatch site mean. 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.

Summations of 35 PCBs at the SWAT clam sites all fall below recent NS&T (NS&T) median (29.2 ng/g, dry weight) and 85th percentile (141 ng/g, dry weight) for NS&T PCBs (2008 data, the most recent available). This was not included in Figure 1.3.3.2.2 due to the massive scale differences between the two data sets. The 2008 NS&T national 85th percentile was approximately 9 times higher than the highest scoring clam PCB site tested by SWAT in Maine in 2010.

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 electrical equipment and transformers (Kimbrough, 2008).

From a human health perspective, the MCDC cancer FTAL for total PCBs for non-commercially caught finfish is 11 ng/g wet wt. (ppb), while the MCDC non-cancer FTAL for total PCBs is 43 ng/g wet wt. (ppb). Of the three SWAT clam sites sampled, the highest mean tissue concentration for total PCBs on a wet weight basis was 2.6 ng/g at Morse Cove, Castine, which was approximately one fourth of the MCDC cancer FTAL of 11 ng/g wet wt.



Figure 1.3.3.2.2: Sum 21 PCBs in SWAT Softshell Clams

1.3.4. Pesticides

1.3.4.1 Blue Mussels

Blue mussels were tested for pesticides from all three sites where mussels were collected in 2010 and tissues were analyzed by AXYS Analytical Services Ltd., Sidney, British Columbia. Mussel tissue samples were analyzed for organochlorinated pesticides (modified EPA Method 8081/EPA 1625) using gas chromatograph/mass spectrometer (GC/MS). Organochlorinated pesticide results were compared to national (NOAA NS&T, see Kimbrough, 2008) and Gulf of Maine (Gulfwatch, see LeBlanc, 2009) blue mussel monitoring program data (when available) in an effort to place Maine SWAT data in a national and regional 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.

To allow comparison to other NS&T Mussel Watch program work, summations of SWAT data were completed for: Dichlorodiphenyldichloroethanes (DDDs), dichlorodiphenyldichloroethylenes (DDEs), and dichlorodiphenyltrichloroethanes (DDTs), chlordanes; and dieldrins. Methodology was consistent with that used by Musselwatch in constructing summations of these pesticide compound groups. Use of these summations assists in putting Maine SWAT data into a national context.

1.3.4.1.1 ΣDDTs

The summation of DDDs, DDEs, and DDTs, (six compounds total, called Σ DDTs in this report) is presented in Figure 1.3.4.1.1.1 for the three 2010 SWAT mussel sites. Σ DDTs ranged from a low mean concentration of 6.8 ng/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 14.2 ng/g dry wt. at Crockett Point, Rockland. The NS&T Mussel Watch considers Σ 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, with all three SWAT sites sampled in 2010 falling in the low category of that range. This is consistent with all mussel sites sampled Maine coast-wide over the past four years, with all falling into the low end of the low NS&T grouping.

 Σ 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, 2008).

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

 Analysis

Organochlorines	Gulfwatch Chlorinated Pesticides	SWAT	
	(Sum 21)		
		2010	2005
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-CHLORDANE)	Х	х	х
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
ENDOSULFAN Ι (α-ENDOSULFAN)	Х	х	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			
	21	34	19



Figure 1.3.4.1.1.1: Sum of DDDs, DDEs, and DDTs in 2010 SWAT Blue Mussels

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 Σ 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 2010 SWAT blue mussel tissue DDT concentration was 2.2 ng/g wet wt., which is 3.4% of the more conservative MCDC cancer FTAL of 64 ng/g wet wt.

1.3.4.1.2 ΣChlordanes

The summation of alpha-chlordane, heptachlor, trans-nonachlor, and heptachlor epoxide (four compounds total, called Σ chlordanes in this report) was determined from SWAT data and is presented in Figure 1.3.4.1.2.1. Σ chlordanes ranged from a low mean concentration of 1.2 ng/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 2.0 ng/g dry wt. at Crockett Point, Rockland. NS&T considers Σ 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. All three sites sampled in 2010 fall in the lowest quarter of that low category (Kimbrough, 2008).

Σchlordanes 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, 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 our 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 under ground. All uses were banned in 1988 (Kimbrough, 2008). NS&T Mussel Watch reported that Chlordane was one of the most ubiquitous contaminants measured by that program. Σchlordanes concentrations in shellfish are decreasing across US sampling stations (Kimbrough, 2008).

The MCDC reports a cancer and non-cancer FTALs for chlordane/nonachlor (summation of alphachlordane, 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 2010 SWAT blue mussel tissue data, when summed in the same manner, shows the highest mean concentration recorded to be 0.31 ng/g wet wt., which is 1.8% 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 2010 SWAT blue mussel tissue was 0.027 ng/g wet wt., which is 1.1% of the 2.4 ng/g wet wt. cancer FTAL.



Figure 1.3.4.1.2.1: Sum of Chlordanes in 2010 SWAT Blue Mussels

1.3.4.1.3 ΣDieldrins

The summation of aldrin and dieldrin (two compounds total, called Σ dieldrins in this report) was determined from SWAT data and is presented in Figure 1.3.4.1.3.1. Σ dieldrins ranged from a low mean concentration of 0.44 ng/g dry wt. at Spring Point, South Portland, to a high mean concentration of 0.90 ng/g dry wt. at Crockett Point, Rockland. NS&T Mussel Watch considers Σ 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 all three of the 2010 SWAT sites falling in the bottom of the low category (Kimbrough, 2008).

 Σ 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, 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, 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 2010 SWAT data was 0.12 ng/g wet wt., which is 8.6% 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 2010 SWAT data was 0.0018 ng/g wet wt., which is 0.1% of the MCDC cancer FTAL.

1.3.4.1.4 Σ21 Organochlorines

The summation of 21 organochlorine pesticides (as noted in Table 1.3.4.1.1) was determined from SWAT data and is presented in Figure 1.3.4.1.4.1 (21 compounds total, called Σ 21 Pesticides in this report). Σ 21 Pesticides ranged from a low mean concentration of 10.4 ng/g dry wt. at Wadsworth Cove, Castine, to a high mean concentration of 19.1 ng/g dry wt. at Crockett Point, Rockland. Figure 1.3.4.1.4 compares the sum of Σ 21 Pesticides at the 2010 SWAT sites to recent Gulfwatch median and 85th percentile for Σ 21 Pesticides (2008 data, the most recent available). All three of the 2010 SWAT sites exceeded the Gulfwatch 2008 median, 9.9 ng/g (dry weight) for Σ 21 Pesticides, although Wadsworth Cove, Castine, was very close to the Gulfwatch median. Two sites exceeded the Gulfwatch 85th percentile, 14.3 ng/g (dry weight), for Σ 21 Pesticides (Spring Point, South Portland, and Crockett Point, Rockland). The remaining station at Wadsworth Cove, Castine, fell below the Gulfwatch 85th percentile concentration for Σ 21 Pesticides.

Figure 1.3.4.1.4.1 also compares the sum of $\Sigma 21$ Pesticides at the 2010 SWAT sites to recent NS&T Mussel Watch median and 85th percentile for $\Sigma 21$ Pesticides (2008 data, the most recent available). Of the three SWAT sites tested, none exceeded the NS&T 2008 median, 22.9 ng/g (dry weight), for $\Sigma 21$ Pesticides. None of the three sites sampled in 2010 approached or exceeded the NS&T 85th percentile of 128 ng/g (dry weight) for $\Sigma 21$ Pesticides.



Figure 1.3.4.1.3.1: Sum of Dieldrins in 2010 SWAT Blue Mussels

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 blue mussel tissue detected by SWAT in 2010 was 0.048 ng/g wet wt., which is 0.3% 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 2010 was 0.032 ng/g wet wt., which was 0.6% 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 2010 was 0.052 ng/g wet wt., which was 0.01% 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 2010 was 0.045 ng/g wet wt., which was 0.3% 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 2010 was 0.034 ng/g wet wt., which was 0.0003% of the MCDC non-cancer FTAL.



Figure 1.3.4.1.4.1: Sum of 21 Chlorinated Pesticides in 2010 SWAT Blue Mussels

1.3.4.2 Softshell Clams

Softshell clams from Morse Cove, Castine (2010), were tested for pesticides and tissues were analyzed by AXYS Analytical Services Ltd., Sidney, British Columbia. Softshell clams tested for pesticides in 2005 (Long Cove, Searsport; Fort Point Cove, Stockton Springs; and Mill Cove, Robbinston) were analyzed at Pace Analytical, Minneapolis, MN. Clam tissue samples were analyzed for organochlorinates (modified EPA Method 8081/EPA 1625) using GC/MS. Organochlorinated pesticide results were compared to national (NOAA NS&T, see Kimbrough, 2008) and Gulf of Maine (Gulfwatch, see LeBlanc, 2009) blue mussel monitoring program data (when available) in an effort to place Maine SWAT data in a national and regional context.

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 Mussel Watch in constructing summations of these pesticide compound groups. Use of these summations assists in putting Maine SWAT 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 Σ DDTs in this report) is presented in Figure 1.3.4.2.1.1 for the four SWAT softshell clam sites. Σ DDTs ranged from a low mean concentration of 4.1 ng/g dry wt. at Mill Cove, Robbinston, to a high mean concentration of 5.2 ng/g dry wt. at Long Cove, Searsport. The NS&T Mussel Watch considers Σ 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, with all four SWAT clam sites falling in the low category of that range.

 Σ 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, 2008).



Figure 1.3.4.2.1.1: Sum of DDDs, DDEs, and DDTs in SWAT Softshell Clams
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 Σ 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 0.87 ng/g wet wt., which is 1.4% 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 Σ chlordanes in this report) was determined from SWAT data and is presented in Figure 1.3.4.2.2.1. Σ chlordanes ranged from a low mean concentration of 0.8 ng/g dry wt. at Morse Cove, Castine, to a high mean concentration of 3.1 ng/g dry wt. at Mill Cove, Robbinston. NS&T considers Σ 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. All four SWAT clam sites sampled fall in the lower half of that low category (Kimbrough, 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 a cancer and non-cancer FTALs for chlordane/nonachlor (summation of alphachlordane, 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 to be 0.47 ng/g wet wt., which is 2.8% 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 0.037 ng/g wet wt., which is 1.5% of the 2.4 ng/g wet wt. cancer FTAL.



Figure 1.3.4.2.2.1: Sum of Chlordanes in SWAT Softshell Clams

1.3.4.2.3 ΣDieldrins

The summation of aldrin and dieldrin (two compounds total, called Σ dieldrins in this report) was determined from SWAT clam tissue data and is presented in Figure 1.3.4.2.3.1. Σ dieldrins ranged from a low mean concentration of 0.40 ng/g dry wt. at Morse Cove, Castine, to a high mean concentration of 0.61 ng/g dry wt. at Mill Cove, Robbinston. NS&T Mussel Watch considers Σ 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 all three of the 2010 SWAT softshell clam sites falling in the bottom of the low category (Kimbrough, 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 0.055 ng/g wet wt., which is 3.9% 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 2010 SWAT data was 0.021 ng/g wet wt., which is 1.6% 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 (21 compounds total, called Σ 21 Pesticides in this report). Σ 21 Pesticides ranged from a low mean concentration of 7.65 ng/g dry wt. at Morse Cove, Castine, to a high mean concentration of 13.71 ng/g dry wt. at Fort Point Cove, Stockton Springs. Figure 1.3.4.2.4.1 compares the sum of Σ 21 Pesticides at the SWAT clam sites to a recent Gulfwatch median at four clam sites (2008 data, the most recent available). All four of the SWAT sites exceeded the Gulfwatch 2008 median, 4.2 ng/g (dry weight) for Σ 21 Pesticides. No Gulfwatch 85th 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 Σ 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 four SWAT clam sites tested, none exceeded the NS&T 2008 median, 22.9 ng/g (dry weight), for Σ 21 Pesticides in shellfish, and none of the SWAT sites approached or exceeded the NS&T 85th percentile of 128 ng/g (dry weight) for Σ 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 Σ 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.



Figure 1.3.4.2.3.1: Sum of Dieldrins in SWAT Softshell Clams



Figure 1.3.4.2.4.1: Sum of 21 Chlorinated Pesticides in SWAT Softshell Clams

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 0.6 ng/g wet wt., which is 4.3% 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 0.13 ng/g wet wt., which was 0.03% 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.

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 0.16 ng/g wet wt., which was 0.9% 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 0.083 ng/g wet wt., which was 0.0006% 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.

1.3.5. Dioxins, Furans, Coplanar PCBs

1.3.5.1. American Lobster

Lobster tissues from 19 NCCA/SWAT stations were tested for dioxins, furans, and coplanar PCBs by AXYS Analytical Services Ltd., Sidney, British Columbia. Dioxin/furan analysis was completed using EPA Method 1613B and GC/HRMS. Coplanar PCB congener analysis was completed using EPA Method 1668A and GC/HRMS. Lobster hepatopancreas and muscle tissues were analyzed. Table 1.3.5.1.1 presents the dioxins, furans, and coplanar PCBs for which analyses were completed.

Tissue concentrations of the individual compounds determined in the laboratory were multiplied by their toxic equivalencies and summed to construct CTEs and DTEs, coplanar and dioxin toxic equivalencies. Compounds with non-detects were assigned a concentration value at half the detection limit, which were then used to calculate the CTEs and DTEs. CTEs and DTEs were calculated on a wet weight basis for the lobster tissues, as they are principally used from a human health perspective to assess risk associated with human dietary intake of these compounds in food. Since the food, lobster meat, is eaten in a wet weight form, the CTEs and DTEs were calculated in the wet weight format to provide the best prediction of CTE and DTE intake.

Table 1.3.5.1.1:	SWAT Dioxins, Furar	is and Coplanar PCBs
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Furans and Dioxins	Coplanar PCBs
2,3,7,8-TCDF	PCB-77
1,2,3,7,8-PECDF	PCB-81
2,3,4,7,8-PECDF	PCB-126
1,2,3,4,7,8-HXCDF	PCB-105
1,2,3,6,7,8-HXCDF	PCB-114
2,3,4,6,7,8-HXCDF	PCB-118
1,2,3,7,8,9-HXCDF	PCB-123
1,2,3,4,6,7,8-HPCDF	PCB-156/157
1,2,3,4,7,8,9-HPCDF	PCB-167
OCDF	PCB-169
2,3,7,8-TCDD	PCB-189
1,2,3,7,8-PECDD	
1,2,3,4,7,8-HXCDD	
1,2,3,6,7,8-HXCDD	
1,2,3,7,8,9-HXCDD	
1,2,3,4,6,7,8-HPCDD	
OCDD	

Figure 1.3.5.1.1 shows CTEs and DTEs calculated for lobster hepatopancreas tissue at the 19 lobster stations sampled in 2010. The stations are organized from left to right in west to east order along the Maine coast. Summed (dioxin) DTEs comprise the base of each bar (shown in blue), with the top of each bar comprised of the summed (coplanar PCB) CTEs (shown in purple). Since the toxicities of the dioxins and furans and the coplanar PCBs are additive, the DTEs and CTEs are shown in one bar that adds their toxicity for comparison to the fish tissue action level.

Summed DTEs/CTEs in hepatopancreas ranged from 4.88 to 28.03 pg/g at South Bartlett Island, Blue Hill Bay, and Cousins Island Sound, Casco Bay, respectively. The Maine CDC has produced a fish tissue action level (FTAL) for dioxins, furans and coplanar PCBs (DTEs/CTEs) for recreationally caught finfish fillet. Maine CDC has recommended that this 0.4 pg/g (or parts per trillion) FTAL be utilized to compare lobster tissues to determine acceptability for human consumption. Consistent with prior hepatopancreas data, the 2010 data confirms that consumption of hepatopancreas should be avoided as all stations sampled had DTEs/CTEs well above the 0.4 pg/g FTAL. This FTAL assumes a meal size of 8 ounces (227 g), which may be quite high for hepatopancreas (requiring consumption of hepatopancreas from many lobsters in one meal). However, DTEs/CTEs in hepatopancreas are so high that recalculating the risk assessment with a smaller meal size will not change the recommendation that hepatopancreas consumption should be avoided.

Figure 1.3.5.1.2 shows CTEs and DTEs calculated for lobster muscle tissue at the 19 lobster stations sampled in 2010. The stations are organized from left to right in west to east order along the Maine coast. Summed (dioxin) DTEs comprise the base of each bar (shown in blue), with the top of each bar comprised of the summed (coplanar PCB) CTEs (shown in purple). Since the toxicities of the

dioxins and furans and the coplanar PCBs are additive, the DTEs and CTEs are shown in one bar that adds their toxicity for comparison to the fish tissue action level.

Summed DTEs/CTEs in muscle ranged from 0.096 to 0.188 pg/g at South Bartlett Island, Blue Hill Bay, and Belfast Bay, respectively. Even the highest of these levels is less than half of the 0.4 pg/g FTAL, indicating that lobster muscle tissue contains considerably lower concentrations of DTEs/CTEs when compared to hepatopancreas and that lobster muscle tissue is safe to eat.

Hepatopancreas tissue appears to have a higher percentage of its overall toxicity contributed by CTEs (from coplanar PCBs) than does the muscle tissue. This may be due to sequestration of the coplanar PCBs in the hepatopancreas. Both DTEs and CTEs in hepatopancreas appeared to be higher in Casco Bay stations than on other areas of the coast. In muscle tissue, Casco Bay sites appeared to have DTEs that are similar to those from the rest of the Maine coast, while some Casco Bay muscle tissue CTEs appear to slightly higher than coast-wide muscle CTEs.



Figure 1.3.5.1.1: Dioxin and Coplanar PCB DTEs and CTEs (Toxic Equivalencies) in SWAT Lobster Hepatopancreas



Figure 1.3.5.1.2: Dioxin and Coplanar PCB DTEs and CTEs (Toxic Equivalencies) in SWAT Lobster Muscle

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2.0 LAKES MODULE

2.1 MERCURY IN LAKE FISH PRINCIPAL INVESTIGATORS

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2.1 MERCURY IN LAKE FISH

2.1.1 Introduction

Fish were collected from Maine lakes to meet the objectives of the Maine Center for Disease Control and Prevention (MCDC) and DEP. Lakes and species to be sampled were selected so that all samples will be useful for both agencies.

As requested by MCDC, sampling for the coming year was geared primarily toward filling data gaps identified in the sampling network and establishing a current baseline data set for mercury. Data were also requested from as many of the lakes sampled as part of EPA's REMAP study in 1992-93 such that current mercury levels can also be compared to historical mercury levels from similar locations in an attempt to determine any trending in the data sets.

Secondly, DEP, in collaboration with researchers at the University of Maine, collected fish from lakes to try to determine factors that influence mercury accumulation in fish. From the literature the most important factors that may affect mercury concentrations in fish seem to be drivers of lake trophic status (i.e., nitrogen, phosphorus, iron, aluminum, pH), lake mercury concentrations, and watershed characteristics. From the list of REMAP lakes and other lakes with mercury concentrations in fish from 10 years ago or longer, DEP selected lakes whose characteristics included combinations of the range of variables that are correlated with these factors. Maximum depth, average dissolved organic carbon (DOC), and epilimnetic total phosphorus (TP) concentrations were trisected yielding 27 possible combinations. Some of these factor combinations did not have representation in the existing dataset of 216 lakes for which we had filet mercury, DOC and TP data, so we targeted other white perch lakes for which we had DOC and TP data. From the resulting set of 443 lakes, we selected 102 lakes to be sampled and prioritized them according to fish tissue sampling history. With limited staff time for sampling, we targeted half of the lakes for 2010, and expect to complete the sampling in 2011. Lakes having white perch were of particular interest because this species is the most common one for which we have data.

From each lake, 10 fish were collected. Species included the species previously collected if possible and white perch, black bass, chain pickerel, lake trout, or yellow perch in order of preference. These were the species having the highest mercury concentration in the REMAP data set and are, for the most part, game fish caught and eaten by Maine anglers. A possible exception is yellow perch, which are not consumed by many anglers, but are common, are consumed by common loons and other piscivorous birds, and have been used in ecological risk analysis in Maine and the rest of New England. Water quality and sediment data were collected by DEP's lakes staff during the August 'baseline' period.

Studies in 2008 and 2009 were directed towards investigation of the use of biopsy samples of fish tissue analyzed for mercury by the Direct Mercury Analyzer (DMA80) housed at the Sawyer Environmental Research and Chemistry Lab (SERCL) at the University of Maine. The benefits include lower cost, ability to work with a Maine lab, and potentially non-lethal sampling of the fish in the future. Filets from the same fish were also analyzed by our commercial lab, AXYS Analytical Services. When the results were compared, there was no significant difference (p<0.05) in mean concentrations for all three groups of ten fish each between labs, as reported in the 2009

SWAT report. The mean concentration in the group with the highest mercury levels, the smallmouth bass from the Androscoggin River (0.74 ug/g) from AXYS, however, appeared lower than in the SERCL analyzed fish (0.88 ug/g). The reason for the discrepancy may be that of partial desiccation of the SERCL samples. Fish may loose moisture during processing and freezing such that the reported concentration may not be an accurate wet weight concentration, but rather a partially dehydrated and unknown fish concentration. The biopsy samples may be even more prone to desiccation than whole fish or filets. Biopsy samples are collected with a 4 mm diameter biopsy punch typically collecting <100 mg of tissue. Consequently, there is a relatively large surface to volume ratio that promotes even greater desiccation of the biopsy sample.

To avoid this problem, we proposed weighing biopsies from fish soon after capture before they could dry significantly, and then freezing them in cryovials. We would use these weights to calculate the final wet weight concentrations, thereby avoiding the effects of any weight loss due to dessication. One issue with this approach was to deteremine what percent, if any, of mercury may be left adsorbed to the cryovial when it was emptied into the quartz boat used in the DMA 80. Another issue was whether the new cryovials contained any mercury that would be leached and transferred to the quartz boat. To answer these questions, SERCL conducted a study described in the next section.

2.1.2. SERCL Study

To determine the amount of any adsorbtion or leaching from the vials and desiccation of fish tissue, SERCL conducted the following study.

- 1. Collected a total of 10 replicate biopsy samples from the same fish and freeze in separate cryovials.
- 2. Removed the biopsies from each tube, plus any liquid that drips out easily.
- 3. Added 2 ml of 2% bromine monochloride (preservative / oxidant) to all 10 vials and 10 new emptied vials to capture available mercury.
- 4. Diluted 1 ml to 50 ml (required for analysis) leaves 1 ml for a rerun if needed.
- 5. Analyzed all 20 vials and biopsy samples. Detection limit was 50 pg of Hg in the vial.
- 6. Calculated % mercury left adsorbed to vial.

The result was that the 10 replicate biopsies had low variance (CV=1.65%) (Table 2.1.1). The amount of mercury leached from 10 new vials was very low (average 0.06%). The tissue mercury left in the vials was also very low (average 0.47 %). Consequently, it appeared the issue of desiccation could be surmounted.

Replicate #	Hg leached from blank vials (ng)	Hg leached from blank vials % of fish Hg	Fish Sample ID	Hg left in used vials (ng)	Hg in biopsy samples (ng)	tissue Hg left in vials (ng)	tissue Hg left in vials % of fish Hg	Hg fish concentration ug/g
	0.474	0.45		0 705	445.0	0.700	0.00	4.040
1	0.171	0.15	WHP 11-1	0.785	115.2	0.726	0.63	1.019
2	0.222	0.27	WHP 11-2	0.828	82.8	0.769	0.93	1.010
3	0.047	0.05	WHP 11-3	0.364	98.4	0.305	0.31	1.036
4	0.036	0.04	WHP 11-4	0.486	102.2	0.427	0.42	1.033
5	0.021	0.04	WHP 11-5	0.206	57.5	0.147	0.25	1.027
6	0.046	0.06	WHP 11-6	0.206	77.2	0.147	0.19	1.016
7	0.013	0.01	WHP 11-7	0.630	191.3	0.571	0.30	1.034
8	0.010	0.01	WHP 11-8	0.449	118.1	0.390	0.33	0.984
9	0.015	0.01	WHP 11-9	1.228	140.8	1.169	0.83	1.043
10	0.012	0.00	WHP 11-10	1.486	269.8	1.427	0.53	1.026
MEAN STD SE CV %	0.059	0.06		0.667	125.3	0.608	0.47	1.023 0.017 0.005 1.65

Table 2.1.1. SERCL study results

2.1.3. Methods

In 2010, 50 samples of 10 fish from a total of 45 lakes (for all but 3 lakes where N<10) were analyzed at SERCL by the DMA80. From these lakes, 22 samples of 10 fish from a total of 19 lakes were also sent to AXYS for analysis of mercury concentrations in filets for comparison to the biopsy samples. To avoid a potential bias due to desiccation, fish were captured and kept in a plastic bag on ice or the refrigerator until biopsy samples were collected and weighed the same day or the next morning. The biopsy samples were then frozen in 2 ml cryovials. At the lab the contents of the cryovial were transferred to the DMA 80 quartz sample boat, where the entire mass of mercury was measured. The intent was that the original wet weight concentration would then be calculated from the mass of mercury and original wet weight. However, since a small amount of tissue was left behind in the some of the cryovials, the lab reweighed the tissue and used the new wet weight to calculate results.

2.1.4. Results

Although calculation of mercury concentrations based on the weight of the tissue removed from the cryovial does not account for desiccation, the mean relative percent difference (RPD) between mercury concentrations in fish analyzed by the two labs was only -1.8% (Table 2.1.2), which includes desiccation, tissue left behind in the cryovial, and lab variability. The maximum RPD for all lake means (22%) was well within the standard quality assurance limit (30%). If desiccation was significant, then mercury concentrations would be higher in the SERCL results than in the AXYS results. In fact, mean concentrations were higher in the SERCL samples only in 9 lake/species samples, while lower in 7 lake/species samples and the same as AXYS in 6 lake/species samples, suggesting no bias due to desiccation. Furthermore, when the SERCL mercury concentrations were adjusted for sample weight loss attributed to desiccation, there was poorer correspondence with

Table 2.1.2. Mercury concentrations in fish from Maine lakes and Ponds in 1990s and 2010

LAKES	MIDAS	SPECIES	1990s HG ug/g	2010 AXYS HG ug/g	2010 SERCL HG ug/g	RPD AXYS/SERCL	2010-1990s %
Russell P	2022	BKT	00		0.15		
Ben Annis P	2282	BLC	0.18	0.19	0.19	0.0	6
Hermon P	2286	BLC		0.37	0.41	-10.3	
Cobbosseecontee L	5236	BNT	0.29		0.21		-28
Dutton P	4872	LMB			0.48		
Little Medomak P	5694	LMB		0.40	0.37	7.8	
North P	5690	LMB		0.61	0.58	5.0	
Pease P	5198	LMB	0.36		0.48	0.0	33
Meddybemps I	177	SMB	0.32	0.65	0.65	0.0	103
Pleasant River I	1210	SMB	0.01	0.00	1.35	0.0	100
Raymond P	3690	SMB			0.44		
Sheenscot P	4896	SMB	1 25	1 28	1.30	-16	4
Alamoosook I	4336	WHP	1.20	1.20	0.72	1.0	7
Cedar I	2004		0.01	0.83	0.72	_1 7	_1
China I	5448		0.31	0.00	0.07	10.5	- - _18
	5236		0.22	0.20	0.10	10.5	-10
	3626		0.00		0.40		-33
Crystal P	3452		0.41		0.50		57
Damariscotta I	5400				0.53		
	5296				0.53		
	5360				0.50		
	5349	WHP	0.40	0.50	0.14	0.0	04
	3712	WHP	0.48	0.58	0.58	0.0	21
Great Moose L	2590	WHP	0.66	0.60	0.71	-16.8	8
Hermon P	2286	WHP	0.07	0.46	0.46	0.0	07
	3734	WHP	0.67	0.00	0.49	40.4	-27
Horseshoe L	4788	WHP	0.80	0.98	1.11	-12.4	39
	4322	WHP	0.77		0.79		3
Kingsbury P	262	WHP			0.73		
Lermond P	4800	WHP	• • -		0.35		
Little Sebago L	3/14	WHP	0.35		0.36		3
Lobster L	2948	WHP			1.02		
Meddybemps L	1//	WHP		0.70	0.88	-22.8	
Nicatous L	4/66	WHP	0.59	0.95	0.97	-2.1	64
Pattee P	5458	WHP	0.38	0.55	0.53	3.7	39
Pease P	5198	WHP			0.29		
Pennesseewasee L	3434	WHP		0.51	0.51	0.0	
Rocky P	4330	WHP	0.68		0.37		-46
Salmon L	5352	WHP	0.31		0.21		-32
Sebois L	954	WHP		0.76	0.75	1.3	
Sennebec P	5682	WHP	0.41		0.60		46
Sheepscot P	4896	WHP		1.48	1.50	-1.3	
Threemile P	5416	WHP	0.50	0.38	0.35	8.2	-30
Togus P	9931	WHP	0.15		0.25		67
Upper Middle Branch P	4492	WHP	0.91		0.94		3
Webber P	5408	WHP	0.24	0.20	0.20	0.0	-17
Webber P	4857	WHP			0.68		
West Grand L	1150	WHP		0.90	0.99	-9.5	
Wilson L	3920	WHP	0.56		0.59		5
Woodbury P	5240	WHP		0.44	0.42	4.7	
Little Purgatory P	5250	YLP	0.23		0.20		-13
					MEAN	-1.8	

fish at least 15mm longer than those from other time period

AXYS mercury concentrations than the concentrations reported by SERCL.

The concentrations of mercury exceeded MCDC's fish tissue action level (FTAL=0.20 ug/g) for mercury in the means for fish in all but 6 lakes (Figure 2.1.2). Four of these lakes were for small brook trout from Russell Pond, black crappie from Ben Annis Pond, white perch from China Lake, and yellow perch from Little Purgatory Pond. White perch from the other two lakes, East Pond and Webber Pond, were larger, but both ponds have algal blooms which generally results in lower mercury concentrations in fish.

For trends analysis, in 2010, we collected the same species of fish from 26 lakes that had been previously sampled at least 10 years ago in 1990s. There appeared to be no overall trend of statewide, since, of those 26 lakes, mean fish concentrations appeared lower in 10 lakes but higher in 16 lakes (Table 2.1.2). Differences from 2010 to 1990s ranged from -46% to 105% and some would be too small to be significant. Of those 10 lakes indicating a decline in 2010, fish sizes were smaller (by at least 15 mm) in 4 lakes, which might explain the lower concentrations in 2010, but to the contrary were in fact higher in 3 lakes and no different in 3 lakes. Of the 16 lakes indicating an increase in 2010, fish sizes were larger (by at least 15 mm) in 5 lakes, which might explain the higher concentrations in 2010, but to the contrary were in fact lower in 4 lakes and no different in 7 lakes. Although fish sizes explain some of the differences in mercury concentrations between 1990s and 2010, there were more lakes where sizes do not explain differences between time periods. Differences, therefore, are due to other factors including random variance and small sample sizes.

Even though there were differences between time periods, data from the two labs in 2010 tracked the 1990s data confirming lakes and species with high concentrations and those with low concentrations. Nevertheless, there was some variability in mercury levels in fish among the lakes and ponds. In 2010, DEP began a complementary multi-year project investigating factors, such as mercury levels in water and sediments, lake trophic status, lake and watershed hydrologic and morphometric characteristics, which may influence mercury levels in fish.

3.0 RIVERS AND STREAMS MODULE

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3.1 AMBIENT BIOLOGICAL MONITORING

3.1.1 Background

As part of the SWAT program, DEP's Biological Monitoring Unit evaluates benthic macroinvertebrate communities of Maine streams and rivers to determine if they are potentially impaired by toxic contamination. For reasons of comparability, a small number of unimpaired reference sites are also evaluated. Benthic macroinvertebrates are animals without backbones that can be seen with the naked eye and live on the stream bottom, such as mayflies, stoneflies, caddisflies, crayfish, snails, and leeches. In 2010, we evaluated the condition of 42 sample locations, primarily in the Southern Maine 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 2010 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' (Indeterminant) 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 2010, 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.state.me.us/dep/blwq/docmonitoring/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 2010 in Southern Maine. Forty-two stations were sampled under the SWAT Program (Table 3.1.1).

At this time (June 10, 2011), forty-one stations have been analyzed for aquatic life attainment with twenty-six stations in attainment of their statutory class. In fact, nine stations exceeded their assigned class. So far 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 systems; summaries on these streams are found below.

Birch Stream – Bangor Station 312

Birch Stream is located below Bangor International Airport and the Airport Mall and much of the headwaters of the stream have been altered through the years. The stream did not attain the minimum Class C criteria for aquatic life. The stream supported a good number of organisms with a moderate number of different taxa. However, sensitive taxa were present in very low numbers. Eighty percent of the community consisted of tolerant snails. Temperature, specific conductance, and total dissolved solids were all high and indicative of an urban system (Table 3.1.2). Deicer continues to impact the benthic community as well as an altered hydrology which has affected the habitat and stream bank stability. The stream bottom condition has improved as a result of efforts to contain the deicer. Sewage fungus was not present in 2010. The stream has been listed on our 303(d) list and is a designated TMDL stream; it has not attained class since first being sampled in 1997 (Table 3.1.4).

Frost Gulley – Freeport Station 303

Frost Gulley is unique as it is one of a few systems in southern Maine whose statutory class is Class A. It attained Class A aquatic life criteria in 3 of 4 previous sampling events (see Table 3.1.4). In 2010, sensitive organisms were present but were not as diverse as you would typically see in a Class A system. The stream banks show some erosion indicating some altered hydrology in the system. We will continue to monitor the stream in the future.

Goosefare Brook – Saco Station 48

The station is below the Great Heath in Saco which is highly acidic during certain times of the year and its discharge may negatively influence the community. In addition, the flow in the system was not measurable.

Goosefare Brook – Saco Station 271

The number of organisms present in the sample was very low and a determination of class attainment could not be made. However, specific conductance was very high (Table 3.1.2) indicating possible urban NPS. The station will be resampled in the near future to determine if the low numbers found were due to a specific stressor.

Kimball Brook – South Portland Station 795

The number of organisms present in the sample was very low and there were no sensitive organisms present. The stream was covered by an iron floc and had a distinct sewage smell. Conductivity was very high (Table 3.1.2) at sampler retrieval. Although the sample did not have minimum numbers to run through the model it was determined using Professional Judgment that the stream did not attain the minimum Class C criteria for aquatic life.

Long Creek – South Portland Stations 409, 411, 414, 415, and 752

These stations did not attain the minimum Class C criteria for aquatic life. Generic diversity and the numbers of sensitive taxa present were very low at all locations. The top three in dominant taxa in all stations were very tolerant organisms that made up between 40 and 80% of the communities sampled. The stream banks were heavily cut and there was a great deal of accumulation of debris and sand in the system indicating altered hydrology. Specific conductance was very high at all stations (Table 3.1.2) indicating possible urban NPS.

Red Brook – Scarborough Station 219

The number of organisms collected in the sample was very low and a determination of class attainment could not be made. Flow was very low during the sampling period. The station will be sampled again in the near future.

South Branch Long Creek – South Portland Station 753

This station did not meet the minimum Class C criteria for aquatic life. The number of sensitive taxa were very low and over 70% of the community was made up of three taxa of tolerant midge larvae. Specific conductance was very high (Table 3.1.2) indicating possible urban NPS.

Tannery – Gorham Station 474

This station did not attain the minimum Class C criteria for aquatic life. Generic diversity and number of sensitive taxa were very low. Over 40% of the aquatic community consisted of a tolerant caddisfly. The habitat was degraded (stream banks were eroded and the stream bottom was covered with silt) indicating altered hydrology. The stream banks were eroded and the bottom of the stream was covered with silt. The stream attained Class B aquatic life criteria in 2000 and 2005 (Table 3.1.4). Follow up sampling will occur to verify that Class B criteria are not met.

Trout Brook – South Portland Station 675

This station did not attain the minimum Class C criteria for aquatic life. Generic diversity and numbers of sensitive taxa were very low. The specific conductance during sampling was very high and further investigation will be undertaken to determine if this is due to salt intrusion from ground water. Specific conductance at a lower station in the watershed is known to be affected by tidal fluctuations.

West Brook – Biddeford Station 797

The diversity of sensitive taxa was low in West Brook although the Generic Richness of the community was high. The temperature at the time of sample placement was high and specific conductance somewhat elevated (Table 3.1.2). This station is located below Wilcox Pond and the outlet discharge of the pond is probably reducing the number of sensitive taxa present due to high temperatures. West Brook attained Class B aquatic life criteria in 2005 (Table 3.1.4).

Waterbody	Town	Station	Log	Potential sources of pollution ¹	Statutory Class/ Final Determina- tion	Attains Class? ²	Probable Cause ¹
Androscoggin River	Brunswick	954	1956	Industrial / Municipal	C / B	Yes	
Androscoggin River	Brunswick	955	1977	Industrial / Municipal	C / C	Yes	
Androscoggin River	Brunswick	956	1978	Industrial / Municipal	C / C	Yes	
Back Brook	Limington	107	1961	Reference	B / A	Yes	
Birch Stream	Bangor	312	1923	Urban NPS / Airport deicer	B/NA	No	NPS Toxics
Branch Brook	Sanford	106	1952	NPS / Airport	A / A	Yes	
Brown Brook	Limerick	445	1965	Urban NPS / Impoundment	B / B	Yes	
Frost Gulley	Freeport	303	1920	Urban NPS	A / B	No	NPS?
Frost Gulley	Freeport	304	1921	Urban NPS	A / A	Yes	
Goosefare Brook	Saco	48	1958	Control	B / C	No	Natural – Great Heath
Goosefare Brook	Saco	271	1959	Urban NPS / In Place Contamination	B / I	No	Low Numbers - Resample
Great Works River	North Berwick	439	1934	NPS	B / B	Yes	
Kennebunk River	Kennebunk	270	1974	Urban NPS / Turnpike	B / B	Yes	
Kimball Brook	South Portland	795	1927	Urban NPS	C / NA	No	NPS Toxics / Habitat
Little Ossippee River	Limington	446	1964	Industrial / Urban NPS / Impoundment	B / B	Yes	
Little Ossippee River	Limerick	447	1963	Reference	B / A	Yes	
Little River	Lebanon	440	1936	NPS	B / A	Yes	
Long Creek	Westbrook	411	1942	Urban NPS	B / NA	No	NPS Toxics / Habitat
Long Creek	South Portland	570	1944	Urban NPS	C / C	Yes	

Table 3.1.1. 2010 SWAT Benthic Macroinvertebrate Biomonitoring Results

¹ NPS, non-point source pollution.

² 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 ¹	Statutory Class/ Final Determina- tion	Attains Class? ²	Probable Cause ¹
Long Creek	South Portland	752	1932	Urban NPS	C / NA	No	NPS Toxics / Habitat
Long Creek	South Portland	415	1926	Urban NPS	C / NA	No	NPS Toxics / Habitat
Long Creek (Blanchette Brook)	Westbrook	409	1943	Urban NPS	B / NA	No	NPS Toxics / Habitat
Long Creek (North branch)	South Portland	414	1933	Urban NPS	C / NA	No	NPS Toxics / Habitat
Merriland River	Wells	436	1975	Reference	A / A	Yes	
Merriland River	Wells	437	1976	NPS	A / A	Yes	
Mill Brook	Porter	698	1962	Reference	B / A	Yes	
Mousam River	Sanford	259	1951	Urban NPS / Landfill	C / C	Yes	
Mousam River	Sanford	390	1954	Control	C / B	Yes	
Mousam River	Sanford	391	1955	Municipal	C / B	Yes	
Presumpscot River	Westbrook	72	1929	Municipal / Industrial / Urban NPS	C / C	Yes	
Red Brook	Scarborough	219	1968	Landfill / NPS	C / I	No	Low numbers - Resample
Red Brook	South Portland	412	1930	NPS	C / C	Yes	
Red Brook	Scarborough	413	1967	NPS	C / C	Yes	
Salmon Falls River	Berwick	52	1935	Municipal			
Sheepscot River	Whitefield	74	1919	Long Term Monitoring	AA / A	Yes	
South Branch Long Creek	South Portland	753	1925	Urban NPS	C / NA	No	NPS Toxics/ Habitat
Tannery Brook	Gorham	474	1970	NPS	B / NA	No	NPS Toxics/ Habitat
Tannery Brook	Gorham	562	1969	NPS	B / A	Yes	
Thacher Brook	Biddeford	451	1960	Urban NPS	B / A	Yes	
Trout Brook	South Portland	675	1928	Urban NPS	C / NA	No	NPS Toxics/Salt intrusion
West Branch Sheepscot River	China	268	1918	Long Term Monitoring	AA / A	Yes	
West Brook	Biddeford	797	1957	Urban NPS	B/C	No	

Table 3.1.1. 2010 SWAT Benthic Macroinvertebrate Biomonitoring Results (continued)

¹ NPS, non-point source pollution.
 ² 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. 2010 SWAT Field Data

Waterbody	Station	Sampler D	Sampler Deployment					Sampler Retrieval			
		Date	Temp*	\mathbf{DO}^*	SPC [*]	pН	Date	Temp*	\mathbf{DO}^*	SPC [*]	pН
			Deg C	mg/L	uS/cm	STU		Deg C	mg/L	uS/cm	STU
Androscoggin River	954	7/27/2010	25	7.2	90		8/24/2010	22.3	7.3	79	
Androscoggin River	955	7/27/2010	24.8	8.4	95		9/8/2010	22.4	8.1	104	
Androscoggin River	956	7/27/2010	25.2	7.9	92		9/8/2010	22.8	7.9	103	
Back Brook	107	7/26/2010	16.8	8.3	58		8/25/2010	14.4	8.5	53	6.33
Birch Stream	312	7/9/2010	25.7	9.8	540		8/12/2010	20.7	8.6	450	7.96
Branch Brook	106	7/21/2010	17.4	7.9	69		8/23/2010	15.4	7.8	65	6.72
Brown Brook	445	7/26/2010	25.7	7.9	72		8/25/2010	20.4	6.9	57	6.85
Frost Gulley Brook	303	7/8/2010	20.6	8.3	275		8/11/2010	19.4	8.2	265	7.41
Frost Gulley Brook	304	7/8/2010	21.1	8.3	312		8/11/2010	19.9	8.3	305	7.22
Goosefare Brook	48	7/22/2010	20.1	7.1	87		8/24/2010	16.3	6.4	136	6.65
Goosefare Brook	271	7/22/2010	20.6	6.7	397		8/24/2010	16.1	7.3	565	6.81
Great Works River	439	7/14/2010	22	8.1	81		8/17/2010	21.7	8.3	101	6.95
Kennebunk River	270	7/28/2010	24.6	10	85	8.03	8/31/2010	22.5	9.4	95	7.43
Kimball Brook	795	7/12/2010	23	7.4	266		8/13/2010	21.1	6.8	410	
Little Ossippee River	446	7/26/2010	25.2	7.5	71		8/25/2010	21.1	6.5	59	6.7
Little Ossippee River	447	7/26/2010	22.1	9.3	63		8/25/2010	17.1	8.6	51	7.25
Little River	440	7/14/2010	21.7	7.9	71		8/17/2010	21.4	8.2	89	6.98
Long Creek	411	7/19/2010	23.7	6.52	600	7.48	8/18/2010	20.7	6.38	209	7.6
Long Creek	570	7/22/2010	22.2	7.82	265		8/18/2010	21.2	7.77	265	7.53
Long Creek	752	7/19/2010	21.8	6.84	750	7.3	8/16/2010	19.3	6.99	370	7.5
Long Creek	415	7/12/2010	21.5	6.5	490		8/13/2010	19.5	6.7	576	6.88
Long Creek (Blanchette Brook)	409	7/19/2010	22.1	7.29	857	7.55	8/18/2010	19.7	7.28	963	7.41
Long Creek (North branch)	414	7/19/2010	21.9	7.91	1245	7.37	8/16/2010	19.4	6.86	725	7.49

* Temp = water temperature; DO = dissolved oxygen; SPC = specific conductance.

Waterbody	Station	Sampler D	Sampler Deployment					Sampler Retrieval				
		Date	Temp	DO	SPC	pН	Date	Temp	DO	SPC	pН	
			Deg C	mg/L	uS/cm	STU		Deg C	mg/L	uS/cm	STU	
Merriland River	436	7/28/2010	21.8	7.4	57	6.37	8/31/2010	20.9	8	57	5.81	
Merriland River	437	7/28/2010	21.7	8.1	63	6.57	8/31/2010	20.9	8.2	64	6.07	
Mill Brook	698	7/26/2010	22.6	8.5	36		8/25/2010	18	7.9	29	7.04	
Mousam River	259	7/30/2010	25.5	8.7	92	6.82	8/23/2010	21.6	8.1	78	6.7	
Mousam River	390	7/21/2010	26.3	8.4	98		8/23/2010	20.6	7.8	70	6.51	
Mousam River	391	7/21/2010	25.8	7.6	133		8/23/2010	20.2	8.2	106	6.86	
Presumpscot River	72	7/13/2010	26.8	8.2	68		8/16/2010	24.2	7.9	66	6.98	
Red Brook	219	7/27/2010	18.5	8.3	137		8/30/2010	18.3	8.6	128	6.58	
Red Brook	412	7/19/2010	21.1	7.73	501	7.16	8/16/2010	18.1	7.47	623	7.08	
Red Brook	413	7/27/2010	18.8	8.2	243		8/30/2010	19.8	7.2	269	6.83	
Salmon Falls River	52	7/14/2010	25.8	7.8	111		8/17/2010	25.7	7.9	134	7.08	
Sheepscot River	74	7/7/2010	26.7	6.7	50		8/10/2010	23.8	6.8	67	6.78	
South Branch Long Creek	753	7/12/2010	23.1	6.7	1082		8/13/2010	19.7	6.6	1172	7.4	
Tannery Brook	474	7/27/2010	22.5	6.7	221		8/30/2010	18.6	6.1	186	6.73	
Tannery Brook	562	7/27/2010	19.4	8.1	375		8/30/2010	17.5	7.4	344	7.42	
Trout Brook	451	7/22/2010	22.2	7.8	197		8/24/2010	18.7	8.2	185	7.47	
Trout Brook	675	7/12/2010	14.8	5.5	749		8/13/2010	14	5.2	975	6.37	
West Branch Sheepscot River	268	7/7/2010	25.7	7.9	65		8/10/2010	22.1	7.9	78	7.19	
West Brook	797	7/22/2010	25.1	7.1	148		8/24/2010	18	7	201	6.83	

Table 3.1.2. 2010 SWAT Field Data (continued)

*

Table 3.1.3. 2010 SWAT Water Chemistry Data

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

Waterbody	Log	Sampling Date	DOC	NH ₃ -N	TKN	NO ₂ - NO ₃ -N	SRP	Total P	TSS	TDS
			mg/L	mg/L	mg/L	mg/L	ug/L	mg/L	mg/L	mg/L
Back Brook	1961	8/25/2010	1.2	< 0.01	0.14	0.140	3	0.017	9.6	45
Birch Stream	1923	8/12/2010	2.5	0.03	0.30	0.260		0.020	2.2	270
Branch Brook	1952	8/23/2010	1.2	< 0.01	0.10	~0.164	1	0.005	<2	54
Frost Gulley Brook	1920	8/11/2010	2.2	< 0.01	0.20	0.280		0.024	<2	180
Frost Gulley Brook	1921	8/11/2010	2.2	< 0.01	0.20	0.280		0.027	<2	200
Goosefare Brook	1958	8/24/2010	1.6	0.02	0.24	0.250	3	0.013	<2	94
Goosefare Brook	1959	8/24/2010	2.2	0.07	0.30	0.200	2	0.015	<2	330
Great Works River	1934	8/17/2010	2.7	0.01	0.30	0.040	1	0.016	<2	83
Kennebunk River	1974	8/10/2010	5.8	0.01	~0.40	0.030	4	0.034	3.1	73
Little Ossippee River	1963	8/25/2010	2	< 0.01	0.10	0.020	1	0.009	<2	42
Little Ossippee River	1964	8/25/2010	3.7	< 0.01	0.30	< 0.010	<1	0.013	<2	65
Little River	1936	8/17/2010	3.7	< 0.01	0.30	0.100	1	0.015	<2	82
Merriland River	1975	8/31/2010	16	0.01	0.60	0.030	3	0.025	<2	75
Merriland River	1976	8/31/2010	15	0.01	0.70	0.030	2	0.030	<2	82
Mousam River	1951	8/23/2010	2.1	0.03	0.20	~0.030	<1	0.011	<2	55
Salmon Falls River	1935	8/17/2010	3	0.02	0.20	0.470	35	0.071	<2	110
Sheepscot River	1919	8/10/2010	3.8	< 0.01	0.30	< 0.010	<1	0.011	<2	41
Thacher Brook	1960	8/24/2010	1.8	< 0.01	0.20	0.110	16	0.027	4.1	110
West Branch Sheepscot River	1918	8/10/2010	3.6	< 0.01	0.30	0.050	<1	0.010	<2	53

 $DOC = dissolved organic carbon, NH_3-N = ammonia-nitrogen, TKN = total Kjeldahl-nitrogen, NO_2-NO_3-N = nitrite-nitrate-nitrogen, SRP = soluble reactive phosphorus (ortho-phosphate), Total P = total phosphorus, TSS = total suspended solids, and TDS = total dissolved solids.$

Figure 3.1.1. 2010 In-Stream Temperature Data











3.1.3 Attainment History of Sampling Stations Prior to 2010

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

Waterbody	Station	Attained Class	Did not Attain Class	Indeterminate Result
Back Brook	107	1987, 2005		
Birch Stream	212		1997,1999, 2001,	
Blich Stream	512		2003-2007	
Branch Brook	106	1987	2000, 2005	
Brown Brook	445		2005	2000 (low numbers)
Frost Gulley	303	1997, 2000, 2005	1998	
Frost Gulley	304	2000	1997, 1998, 2005	
Goosefare Brook	48	1984, 1986, 1994, 1998, 2000	1995, 2005	
Goosefare Brook	271	2005	1995, 1998, 2000	
Great Works River	439	2000, 2005		
Kennebunk River	270	1995, 2000	2005	
Kimball Brook	795		2005	
Little Ossippee River	446		2000, 2005	
Little Ossippee River	447	2000, 2005		
Little River	440	2000, 2005		
Long Creek	411		1999	
Long Creek	752		2004	
Long Creek	415	1999		
Long Creek (Blanchette Brook)	409		1999	
Long Creek (North branch)	414		1999	
Merriland River	436	2000, 2005		
Merriland River	437	2000, 2005		
Mill Brook	698	2003		
Mousam River	259	1995, 1999, 2005		
Mousam River	390	1999		
Mousam River	391	1999		
Presumpscot River	72	2005	1984, 1994-1996	
Red Brook	219	1994, 2005	1999	
Red Brook	412	1999		
Red Brook	413	2007		1999 (low numbers)
Salmon Falls River	52	2005	1984, 1991, 1992, 1995	
Sheepscot River	74	1985, 1987-1990, 1992, 1995, 1996, 1998-2009	1984, 1986, 1991, 1993, 1994, 1997	

Table 3.1.4. Past Attainment History

Waterbody	Station	Attained Class	Did not Attain Class	Indeterminate Result
South Branch Long Creek	753		2004	
Tannery Brook	474	2000, 2005		
Tannery Brook	562	2000, 2005		
Thacher Brook	451	2000, 2005		
Trout Brook	675		2003-2005	
West Branch Sheepscot River	268	1996-1999, 2001, 2002, 2005, 2007, 2009	2000, 2003, 2004, 2006, 2008	1995 (low numbers)
West Brook	797		2005	

 Table 3.1.4 – Past Attainment History (continued)

3.2 FISH CONSUMPTION ADVISORIES

Although there is a statewide Fish Consumption Advisory for all freshwaters based on mercury, there are more restrictive advisories for certain rivers and streams due to other contaminants. Based on a review of the fish sampling data by the Maine Center for Disease Control and Prevention (MCDC) conducted through 2009, dioxin, coplanar PCB and total dioxin toxic equivalents (TEQ) concentrations, which were responsible for many advisories on rivers and streams, had decreased significantly over the previous 10 to 15 years. The remaining contaminants of primary concern for continued fish consumption advice more restrictive than the mercury advice are DDT and total PCBs. As for DDT, the three new locations tested in 2009 along with the 2009 testing at seven previously sampled rivers and streams demonstrate concentrations above the FTAL of 64 ppb, indicating that DDT is affecting a number of rivers and streams in Aroostook County. Based on the data collected through 2010, sufficient data are now available to begin evaluation of revisions to the fish advisories for the next (2012) fishing season.

Available data, including those collected in 2009, indicated the continued presence of levels significantly above the PCB fish tissue action level (FTAL) of 11 ppb at some locations. Consequently, MCDC recommendations for sampling for 2010 were geared primarily toward filling data gaps identified in the sampling network for dioxins and/or total PCBs. *Though limited dioxin monitoring was requested in 2010 to fill identified data gaps, additional future sampling will be requested beginning in 2011 as part of a periodic monitoring program to confirm that total TEQ levels are either stable or continuing to trend downward.*

No analytical data for striped bass or bluefish were requested for 2010. Data for striped bass and bluefish will be requested in the future as part of a plan to periodically monitor contaminant levels over 3 to 5 year intervals to support the striped bass and bluefish consumption advisory that was revised in 2009.

In 2010 fish were collected by DEP by use of angling and gill nets. Chain of custody forms were used. Fish were immediately killed, 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. All samples from other agencies were transferred to DEP and fully documented with chain of custody forms. 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.

3.2.1 Dioxin in Fish Tissue

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, 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 2010 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 Maine Center for Disease Control (MCDC) 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 Maine Center for Disease Control and Prevention (MCDC) issued the latest 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', which can be seen at <u>http://www.maine.gov/dep/blwq/docmonitoring/dioxin/index.htm</u>. In the report, MCDC adopted a new 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. MCDC reviewed the data collected since their last review in 2003, i.e. 2004-2007 with respect to the new FTAL.

For 2009, MCDC did not request any monitoring, but made the following statement:

Dioxin monitoring, though not requested this year for the major rivers, will be requested in future years as part of a periodic monitoring program to confirm that total TEQ levels are either stable or continuing to trend downward

For 2010, MCDC made the following request to gather data to allow a review of the fish consumption advisories by the 2011 open water fishing season:

Though limited dioxin monitoring is requested this year to fill identified data gaps, future sampling will be requested beginning next year as part of a periodic monitoring program to confirm that total *TEQ* levels are either stable or continuing to trend downward.

Dioxin Data for the Major Rivers

As stated previously, requested dioxin sampling is based on identified data gaps. Because dioxins, in general, are higher in white sucker than game fish but game fish have been the focus of previous
sampling, white sucker data are requested to determine whether separate advice for white sucker, more restrictive than for other game fish, is required. Recommendations for further sampling for dioxins are provided below, by river.

Kennebec River: White suckers have not been sampled for dioxins at Augusta or Sidney since 1995. Total TEQ concentrations were approximately 5 parts per trillion (ppt) at that time with the dioxin TEQ approaching 3 ppt. Sampling conducted in 2009 for coplanar PCBs indicates that the PCB TEQ in white sucker in the Augusta/Sidney reach of the river has decreased significantly and now is on the order of 0.5-0.6 ppt. Lacking companion dioxin data precludes a conclusion that total TEQ concentrations have now fallen below 3 ppt. The lack of recent dioxin TEQ data for white sucker is identified as a data gap and analysis for dioxins is requested for white sucker at either Augusta or Sidney to fill this data gap.

Sebasticook River: Total TEQ data are available for white sucker only at one location (Burnham; total TEQ ~0.8 ppt in 2008) within the last 10 years. Total TEQ data for the west branch of the river (Palmyra) indicate levels greater than 4 ppt in 1996. No dioxin data for white sucker are available from the east branch of the river. Therefore, dioxin data for white sucker are requested from Palmyra and Newport (if possible) to fill these data gap.

St. Croix River: The most recent total TEQ data available for white sucker were collected in 1999. Total PCB data collected for white sucker at Baring indicate that more conservative advice than the mercury advice is necessary downstream of this sampling location, while data at Woodland indicate that the mercury advice is sufficiently protective. Therefore, to confirm that the mercury advice is sufficiently protective at Woodland, upstream of Baring, dioxin data for white sucker are requested from Woodland to fill this data gap.

3.2.1.3 Sampling Plan

In 2010, a total of 10 white suckers 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. Sample costs were reduced from that of previous years by 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 MCDC's FTAL, EMPCs were treated the same way in this report.

Table 3.2.1. 2010 DMP/SWAT dioxin and coplanar PCB samples.

RIVER	STATION	DMP PCDD/F samples	DMP facility	SWAT PCDD/F samples	SWAT CPCB samples
KENNEBEC R	SIDNEY	2C5 WHS	SAPPI SOMERSET		2C5 WHS
ST CROIX R	WOODLAND			2C5 WHS	2C5 WHS
SEBASTICOOK R EAST BRANCH WEST BRANCH	NEWPORT PALMYRA	2C5 WHS	HARTLAND	2C5 WHS	2C5 WHS 2C5 WHS
TOTAL		4		4	8

2C5 WHS = 2 composites of 5 white sucker each

DMP = Dioxin Monitoring Program; SWAT = Surface Water Ambient Toxics monitoring program PCDD/F=dioxins and furans; CPCB = coplanar PCBs

3.2.1.4 Results and Discussion

White sucker were not successfully collected from the Kennebec River at Sidney. White sucker were collected from the East Branch of the Sebasticook River above the County Road Bridge below Corinna in Newport (SEN), from the West Branch of the Sebasticook River below Hartland at Palmyra (SWP), and from the St. Croix River above the mill in Woodland.

At SEN in 2010, concentrations of dioxin toxic equivalents (DTEhu, calculated at the 95th upper confidence level with non-detects at ½ of the detection limit) exceeded the MCDC FTAL for dioxin like compounds by a large amount (Figure 3.2.1). Similarly concentrations of coplanar PCB toxic equivalents (CTEhu, calculated at the 95th upper confidence level with non-detects at ½ of the detection limit) also exceeded the FTAL alone and increased the exceedance for the combination of the two groups of contaminants. There are no historical data for white sucker at this location, which is below a Superfund site at a former textile mill that has been extensively remediated for other compounds that may have been precursors for dioxin. There is an extensive wetland with much deposition of fine grained organic sediments at this sample site.

At SWP in 2010 concentrations of DTEhu also exceeded the MCDC FTAL for dioxin like compounds by a large amount (Figure 3.2.1). Concentrations were somewhat lower than in the only previous sample in 1996. CTEhu also exceeded the FTAL in 1996 but were not sampled in 2010.

At SCW in 2010, concentrations of DTEhu were slightly below the FTAL and previous levels from 1997-1999 (Figure 3.2.1). CTEhu were not measured in 2010, but exceeded the FTAL, although were declining, in previous years.



Figure 3.2.1 Dioxin (DTEhu) and coplanar PCB (CTEhu) toxic equivalents(upper 95th CL) in white sucker from the Sebasticook River East Br at Newport (SEN) & West Br at Palmyra (SWP), & St Croix River at Woodland (SCW) in 2010 compared to historical data

3.2.2 PCBs in Fish Tissue

3.2.2.1 Introduction

Total PCB Data for the Major Rivers

As stated previously, total PCBs are a limiting factor for revision of the fish consumption advice for the major rivers to advice consistent with that based on state-wide mercury concentrations in fish. In 2010, MCDC requested PCB sampling primarily based on identified data gaps. Recommendations for further sampling for total PCBs are provided below, by river. Two composite samples were recommended for each sampling location - fish species combination. Analysis for total PCBs by the congener method was requested.

Androscoggin River: Monitoring data for total PCBs indicates that more protective advice is needed upstream of Rumford with less restrictive advice indicated downstream of Jay. The Riley sampling station, located between Jay and Rumford, has not been sampled for total PCBs since 2002. Sampling data was requested at this station to determine whether this reach of the river should be included within the area subject to the more restrictive or less restrictive advice. Therefore, bass and white sucker data at Riley for total PCBs by the congener method was requested.

Penobscot River: No total PCB data are available for bass or white sucker upstream of Woodville. It is possible that less restrictive advice could be recommended upstream of Woodville. Therefore, total PCBs in both bass and white sucker were recommended at Medway on the West Branch and Grindstone on the East Branch.

Sebasticook River: Total PCB data were collected in bass and white sucker in 2008. These data indicate that more restrictive advice is necessary for the main stem and East Branch of the river. Therefore, a second year of sampling was requested for bass and white sucker at Burnham and Newport. These data were recommended for collection prior to changing the fish consumption advice for this river.

Presumpscot River: Data collected for total PCBs in 2009 indicate levels exceeding the total PCB FTAL. The mercury advice is not protective given the levels of total PCBs detected in this single round of sampling. Therefore, a second year of sampling was requested for bass and white sucker at Westbrook and Windham prior to changing the fish consumption advice for this river. Since a new station at Gorham, sampled for other metrics in 2008 and 2009, showed some level of impact, this station was also included for sampling in 2010.

3.2.2.2 Results and Discussion

A total of 10 fish were captured at each station and analyzed as two composites of skinless filets from each of five fish for all 209 or coeluting PCB congeners via EPA method 1668. Total PCBs were then calculated as the sum of congeners and coeluting groups. From these data, coplanar PCBs were also calculated but, as there were no accompanying dioxin and furan data, total dioxin toxic equivalents are not available and the coplanar data are, therefore, not shown.

Androscoggin River

Mean total PCB (TPCB) concentration in smallmouth bass at Riley exceeded MCDC's FTAL of 11 ng/g in 2010 (Table 3.2.2.1). Both mean and maximum concentrations in 2010 were much higher than in three previous years. Mean and maximum concentrations in white sucker were even higher than in bass and greatly exceed the FTAL also. Concentrations in white sucker were higher than in the only previous year with data. Concentrations in both species were among the highest for all stations on the river. The variation in concentrations within species among all years may reflect differences among the four different labs that were used, although all data meet quality assurance and control objectives, or simply the natural variation in individuals and condition among years.

Table 3.2.2.1. Total PCBs in fish from the Androscoggin River, ng/g, mean and (max value where n=2 or 95th upper confidence level where n>2)						/here n>2)				
Year	Species	Gilead	Rumford Pt	Rumford	Riley	Jay	Livermore/Jay	Livermore FIs	AUBURN GIP	Lisbon
		AGL	ARP	ARF	ARY	ARJ	ALV	ALF	AGI	ALS
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)					
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)					

Penobscot River

Mean total PCB (TPCB) concentration in both smallmouth bass and white sucker from Grindstone on the East Branch of the Penobscot River were well below MCDC's FTAL of 11 ng/g in 2010 (Table 3.2.2.2). Both mean and maximum concentrations in 2010 were slightly lower than in 1996, the only previous year with data. Mean and maximum concentrations in bass from Medway on the West Branch of the river were much higher and exceeded the FTAL. Concentrations in white sucker were higher than in bass and also exceeded the FTAL. Medway is below the towns of Millinocket and East Millinocket where there are discharges from municipal sewage treatment plants and pulp and paper mills and urban runoff.

Table 3.2.2.2.	2. Total PCBs in fish from the Penobscot River, ng/g. mean and (max value where n=2 or 95th upper confidence level where n>2)									
Year		Grindstone PBG	Medway PMD	Woodville PBW	Mattawamkeag PBM	S Lincoln PBL	Costigan PBC	Veazie PBV	Bangor PBO	Bucksport PBB
2000	ATS							19		
1996	EEL								37	
2000	EEL								253	
2002	EEL								98	
2007	SLT									27 (27)
1994	SMB					9		10		
1996	SMB	5		6						
2008	SMB			9		7 (8)				
2009	SMB			7 (10)		6 (9)		8 (12)		
2010	SMB	2 (2)	19 (22)							
1994	WHS					95		65		
1996	WHS	7		18						
2008	WHS			29 (30)		29 (49)				
2009	WHS			33 (35)		22 (26)		31 (32)		
2010	WHS	3 (5)	35 (44)							

Presumpscot River

Mean and maximimun total PCB (TPCB) concentrations in smallmouth bass at Windham in 2010 were well below MCDC's FTAL of 11 ng/g and similar to that of 1994, the only other year with data (Table 3.2.2.3). The mean and maximum concentrations in white sucker in 2010 were higher than in bass and near the FTAL but lower than in 1994. Concentrations in both smallmouth bass and white sucker at Gorham greatly exceeded the FTAL and concentrations from 2009. Concentrations in smallmouth bass and white sucker at Westbrook exceeded the FTAL and those from 2009, although concentrations in white sucker were lower than those in 1994. Concentrations were lowest at Windham, where there are no point source discharges or significant urban runoff. Concentrations were highest at Gorham, where there is urban runoff. Although there is no known current point source discharge, the sample station is an impoundment adjacent to an old industrial mill complex, which may have left a legacy of PCB contamination in the river. Interestingly, concentrations were somewhat lower at Westbrook, where there are current sewage treatment plant and paper mill discharges as well as urban runoff.

Figure 3.2.2.3. Total PCBs in fish from the Presumpscot River, ng/g.								
mean and (max value where n=2 or 95th upper confidence level where n>2)								
Year	Species	Windham PWD	Gorham PGO	Westbrook PWB				
1994	SMB	8		8				
2009	SMB		41 (48)	26 (27)				
2010	SMB	6 (7)	109 (131)	51 (51)				
1994	WHS	25		128				
2009	WHS		40 (44)	41 (42)				
2010	WHS	12 (13)	216 (257)	103 (109)				

Sebasticook River

Mean and maximum total PCB (TPCB) concentrations in largemouth bass from the East Branch of the Sebasticook River at Newport exceeded MCDC's FTAL of 11 ng/g in 2010 (Table 3.2.2.1). Both mean and maximum concentrations in 2010 were similar to that of 2009 but higher than in 1997. Mean and maximum concentrations in white sucker were even higher than in bass and greatly exceed the FTAL also. This station is below a former textile mill that became a Superfund site that has been remediated for other contaminants. These concentrations are no higher than at many other stations in Maine with no history of industrial point sources of contaminants. Concentrations in both smallmouth bass and white sucker at Burnham on the main stem of the river exceeded the FTAL and those of previous years. They were also higher than those from the East Branch at Newport. This station is below the confluence of the East with the West Branch, where there is a tannery in Hartland, and also below the towns of Newport and Pittsfield with their sewage treatment plant discharges and urban runoff.

Figure 3.2.2.4.					
mean and (ma>					
Year	Species	E Br Newport SEN	W Br Palmyra SWP	Burnham SBN	Winslow
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)	

3.3. LITTLE ANDROSCOGGIN RIVER

3.3.1. Sediments

As required by EPA, Maine has adopted ambient water quality criteria (AWQC) for toxic pollutants, by rule at CMA 06-096 Chapter 584 (http://www.maine.gov/sos/cec/rules/06/chaps06.htm), to be used in calculation of effluent limits for dischargers. AWQC for heavy metals are expressed as total metal, even though the most bioavailable and toxic species 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. One concern is that an increase in total metal discharged to a waterbody may have negative effects on aquatic organisms in the sediment that is not evaluated by AWQC, which addresses only water-column toxicity.

DEP received a request from Paris Utilities District (PUD), which discharges treated domestic wastewater from its facility in South Paris, to develop a SSC for copper. PUD has not been able to meet its Maine Pollution Discharge Elimination System (MPDES) permit limit for copper, which is based on AWQC. PUD requested to use EPA's BLM (Biotic Ligand Model) for development of the SSC. 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 heavy metal to be used as the basis for a SSC. Out of concern for the potential effects of increased loading of total copper to the sediments, DEP proposed to conduct a study of the effects of current discharge on sediment biota.

There are several methods that have been used to address effects of sediment on aquatic biota. Sediments may be analyzed for total metal and results compared to sediment quality criteria (SQC) or guidelines (MacDonald et al. 2000). EPA has no specific SQC for copper and in the absence suggests use of the Equilibrium Partitioning approach, where concentrations are measured in sediment pore water and compared to AWQC (EPA, 2002). DEP was to collect sediments and pore water above and below the PUD discharge to address the issue for the SSC being developed in 2010. In researching methods for collection of pore water, it became apparent that it would not be easy to avoid aeration of the sample and potentially change the chemistry of the pore water. Another approach biomonitoring of the benthic organisms by conducting surveys of biological communities or sediment toxicity tests with aquatic organisms such as the amphipod *Hyalella azteca* or *Chironomus tentans* for freshwater. An alternate method suggested by EPA is use of AVS/SEM.

Acid volatile sulfide (AVS) has been used to predict the sediment toxicity of copper (Cu), cadmium (Cd), nickel (Ni), lead (Pb) and zinc (Zn) (Ankley et al. 1996; Berry et al. 1996). This relationship results from the volatilization of AVS present in sediment and simultaneous release of previously sulfide bound metals (SEM). As a metal sulfide complex, research indicates that metals are not available for uptake by benthic organisms. Under this assumption, 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/SEM becomes an indicator 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. 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.

Sediment samples were collected from the Little Androscoggin in August at two locations, above the dam at Rt 117 approximately 1 mile upstream of the PUD discharge and at Rt 26 approximately 3 miles below the PUD discharge and 2.5 miles below the Norway STP discharge. Due to questionable holding times, samples were collected again in October. Samples were collected at Route 117 from a shallow (~ 2 m) impoundment. Most of the substrate of the river in this reach is sand and there were few depositional areas; consequently the Rt26 sample station was in a side eddy in a shallow (0.2 m) run. Samples were collected by use of a clean Ekman Dredge and plastic spoon by carefully sampling sediment that was not in contact with the dredge from the top ~ 2 cm in the center of the dredge. Samples were collected in pre-cleaned wide mouth glass jars and taken to the lab where they were immediately frozen. Samples were shipped to Batelle Pacific Northwest National Lab for analysis for AVS/SEM, total organic carbon, and grain size. Samples of benthic macroinvertebrates were also collected at the same sites via the Ekman dredge.

Sediment samples were extracted and analyzed for AVS in accordance with Battelle SOP MSL-C-001. This procedure is based on a peer-reviewed, published procedure for the analysis of AVS in sediment and dissolved sulfide in aqueous samples, adopted from a draft USEPA Method (Allen et al. 1991). In this method, sulfide in the sample is converted to hydrogen sulfide by the addition of hydrochloric acid at room temperature. The hydrogen sulfide (H₂S) is purged from the sample by an inert gas and trapped in a sodium hydroxide (NaOH) solution. With the addition of a mixed-diamine reagent (MDR), the sulfide is converted to methylene blue and measured on a spectrometer. AVS results were reported in units of µmole/g on a dry-weight basis.

The SEM extracts were analyzed for all other metals by Inductively Coupled Plasma- Mass Spectrometry (ICP-MS) in accordance with Battelle SOP MSL-I-022, Determination of Elements in Aqueous and Digestate Samples by ICP/MS. The analysis guidelines for this procedure are adapted from USEPA Method 1638 Determination of Trace Elements in Ambient Waters by Inductively Coupled Plasma- Mass Spectrometry. The SEM metal solution concentrations were determined in units of μ g/L and then converted to μ g SEM/g of sediment extracted for AVS. These data were further converted to μ mole/g for each SEM metal.

Results

The potential toxicity of the sediments was evaluated by three methods, 1) comparison of the concentrations of metals in the bulk sample to sediment quality guidelines (MacDonald et al 2000), 2) AVS/SEM, and 3) biomonitoring of macroinvertebrate communities at the two stations.

Bulk 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 total concentrations of all five metals were well below the TEC at both stations (Table 3.3.1). While SEM metals may be only a fraction of total metals (as low as 13% for copper, Warren Boothman personal communication, EPA NHEERL, AED, Narragansett, RI), adjusted total copper concentrations would be below the PEC at both stations. In these samples bioavailability was not an issue.

AVS/SEM

Simultaneously extracted metals (SEM) were generally similar at both Rt117 and Rt26 (Table 3.3.1). Relative percent difference between duplicates was within the data quality objective (25%) except for copper (33-35%). Acid volatile sulfides (AVS) were significantly greater than SEM at Rt26 (AVS/SEM>1) indicating that the metals were bound and not bioavailable to cause toxicity. To the contrary, AVS were much lower than SEM at the upstream station at Rt117 (AVS/SEM<1), suggesting that some metal may be bioavailable for toxicity. Zinc appeared to be the metal with the largest effect, but the concentration was still well below the TEC and total zinc would be well below the PEC, as was the case for all the metals.

Total organic carbon was slightly lower and grain size was generally more coarse at Rt117 reflecting the absence of the discharges from PUD and Norway and urban runoff from both South Paris and Norway. These results are consistent with the lower AVS at this site. The higher AVS at Rt26 was likely due to the discharges and urban runoff.

Table 3.3.1. Acid volatile sulfides-simultaneously extracted metals (AVS-SEM), total organic carbon (TOC), and grain size (GS)										
in sediments from the Little Androscoggin River above (Rt117) and below (Rt 26) the Norway and Paris Utilities District										
sewage treatment plant disch	arges.									
METRIC	RT 117	RT 117	RT 26	RT 26	TEC1	PEC1	RT 117	RT 117	RT26	RT26
	ug/g	ug/g	ug/g	ug/g	ug/g	ug/g	umole/g	umole/g	umole/g	umole/g
		DOI		DOI				001		001
AVS							0.487	0.465	1.33	1.66
SEM Cd	0.321	0.317	0.221	0.250	0.99	4.98	0.00285	0.00282	0.00197	0.00223
SEM Cu	4.39	6.11	6.92	9.83	31.6	149	0.0690	0.0962	0.109	0.155
SEM Ni	1.38	1.48	1.45	1.73	22.7	48.6	0.0234	0.0252	0.0247	0.0294
SEM Pb	10.6	10.2	18.3	20.2	35.8	128	0.0513	0.0493	0.0882	0.0977
SEM Zn	38.2	39.2	26.2	26.7	121	459	0.584	0.600	0.401	0.409
SEM SUM							0.731	0.774	0.625	0.693
AVS/SEM							0.7	0.6	2.1	2.4
% TOC	5.39		6.78							
% SOLIDS	43.6	43.6	46.2	46.2						
GRAIN SIZE %										
GRAVEL, MED	0.19		0.97							
GRAVEL FINE	2.32		1.07							
SAND, VERY COARSE	7.00		2.67							
SAND, COARSE	6.26		2.93							
SAND, MEDIUM	8.98		3.43							
SAND, FINE	39.1		24.7							
SAND, VERY FINE	11.8		18.8							
SILT	18.7		38.8							
CLAY	1.11		0.62							
% TOTAL WT RECOVERED	95.5		94.0							
% SOLIDS	32.1		30.4							
1750 0 0 0 0	–				=		<u> </u>			
' IEC= threshold effect concentration, PEC = probable effect concentration+A5. From MacDonald et al. 2000										

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3.3.2. Biomonitoring

It was difficult finding organic sediments to sample at these two stations and the sediment samples were not large. Consequently, the results should be considered essentially qualitative. Analysis of the benthic communities from the dredge samples reflect differences among habitats at the two stations. LAR117 was in an impoundment with much decaying leaves and deep organic sediments, while LAR 26 is a cove in a free flowing reach with some aquatic macrophytes and a shallow (perhaps seasonal) organic layer over sand. There were slightly different community structures for the two stations, validated by reasonable duplication for the two sampling periods (Table 3.3.2). The abundance of benthic macroinvertebrates was greater at LAR26 than at LAR117, likely reflecting the influence of the discharges. There were more burrowing mayflies (Ephemeridae) at LAR117, perhaps due to deeper sediments there. Sialidae (alderfly larvae) prefer standing water with soft organic substrates with lots of organic matter such as dead leaves, such as occurs at LAR117, rather than flowing water as occurred at LAR26. Amphipoda typically fill a shredder functional feeding niche and were also more abundant at LAR117. Nevertheless, even though these data are quite limited, there was no indication that the communities at either station were impacted to the extent of not attaining the classification of the river. This finding is congruent with the results of biomonitoring of macroinvertebrate communities at the surface of the substrate using rock basket artificial substrates from previous years, which indicated attainment of the criteria for the classification of the river

Taxon	LAR26 8/13/2010	LAR26 10/14/2010	LAR117 8/13/2010	LAR117 10/14/2010
Elmidae Tabanidae Chironomidae Gastropoda Pelecypoda Oligochaeta Ceratopogonid Tipulidae	15 1 226 5 26 13 2	2 2 54 12 8 7 10	1 1 75 4 2 1	14 1
Chaorboridae Psychodidae Amphipoda Sialidae Ephemeridae Libellulidae	1	1	7 1 1	7 4 6

Table 3.3.2. Number of macroinvertebrates in Little Androscoggin River dredge samples, 2010.

3.3.3. Fish Populations

While biomonitoring of macroinvertebrate benthic communities is one method of assessing any impact of point source discharges of municipal or industrial wastewater on aquatic life, the macroinvertebrate community is only one component of aquatic community. Often, other groups, such as fish are more sensitive to some toxic pollutants as shown by species mean acute values calculated by EPA for the development of AWQC. In addition to heavy metals, organic chemicals may affect aquatic communities. Organic endocrine disrupting chemicals (EDCs), such as nonyl phenol (NP), the human birth control estrogen ethinylestradiol (EE2), and other synthetic compounds have been shown in the lab to disrupt the reproductive pathway of fish populations at concentrations found in some streams with municipal sewage treatment plant (STP) discharges (Jobling et al. 2006). Concentrations ranging from 1% to 20% STP effluent have been shown to disrupt reproductive development in fish (Dube et al. 2003; Liney et al. 2006; Iwanowicz et al. 2009).

While DEP has stressor based laboratory programs to assess the potential effects of STP discharges on fish in Maine rivers, those programs are able to detect only relatively gross impacts and may not detect the impact of EDCs. Effects based biological field studies may be able to detect more subtle but important impacts from EDCs or other factors. DEP intends to conduct field studies to investigate those rivers most likely to be impacted first, i.e. those below dischargers with the lowest dilution and highest receiving water concentration (RWC) of STP wastewaters. The RWC of the PUD discharge is 20% at critical (7Q10) low river flow. To evaluate the potential impact of the PUD discharge in general and or the exceedance of the AWQC for copper specifically, DEP conducted a fish assemblage study of the Little Androscoggin River in August 2010.

Three stations were selected to be studied, 1) LARa, immediately above the PUD discharge, 2) LARb, immediately below the PUD discharge but above the Norway discharge \sim 0.5 miles below, and 3) LAR26, the Rt 26 crossing \sim 2.5 miles below the Norway discharge. The habitats were all

generally similar alternating runs and riffles, although the substrate varied somewhat (Figure 3.3.2). Norway holds its effluent and does not discharge from mid June to end of August each summer. The year 2010 offered a good opportunity for the study as it was a relatively dry warm year, and, consequently, during the study river flow was low (10-12 cfs) approaching 7Q10 (4 cfs). Fish were sampled by use of a small beach seine in targeted riffle and run habitats. Consequently, density estimates could not be made.

The results show a total of eight species present (Figure 3.3.1), although anglers report catching brook trout (Salvelinus fontinalis) as well. The most common species were fallfish (Semotilus corporalis), common shiner (Luxilus cornutus) and white sucker (Catostomus commersoni) although the order varied among stations. Fallfish were most abundant at all three stations but diminished slightly at the two stations below the discharges. At LARb common shiners replaced some of the fallfish in abundance, whereas at LAR26 white sucker replaced some of the fallfish.

Table 3.3.3. Habitats at Little Androscoggin fish sampling stations, 2010

METRIC	LARa	LARb	LAR26
WIDTH m	5-7	5-6	6-9
DEPTH m	0.3	0.4	0.2
VELOCITY m/s	0.3	0.3	0.3
% COBBLE	50	20	0
% GRAVEL	25	40	10
% SAND	25	40	90
ALGAE	10%	15%	0%





Fulton's condition factor (K), weight divided by length cubed, is a measure of fitness of fish. Examination of condition factors for the three most common species and largemouth bass showed some significant (p<0.05) differences (Table 3.3.4). Only one white sucker was captured at LARa and consequently no comparison was made between that station and LARb. Common shiners had higher K below the PUD discharge than either of stations upstream or downstream. This result parallels the increase in relative abundance of common shiners at LARb. On the contrary, there was no difference in K for fallfish among all the stations or in white sucker for the lower two stations, despite changes in relative abundance among these stations.

SPECIES	LARa	LARb	LARb	LAR26	LAR26
	K	K	р	K	р
BND		0.87		0.79	
CHP		0.54			
CS	0.72	0.82*	0.003	0.73*	0.000
FF	0.82	0.83	0.248	0.82	0.680
LMB	1.13	1.2	0.169	1.06*	0.030
SMB	1.26	1.45		1.21	
WHS	0.85	0.94		0.93	0.553
YLP				0.88	

Table 3.3.4. Condition factor (K) in fish from three stations in the Little Androscoggin River, 2010

* = significally different from upstream station in a t-test

These differences in species composition and K may reflect the subtle effects of the discharges. LARa has no upstream discharges and little urban runoff, while LARb has the PUD discharge and some urban runoff, and LAR26 has an additional discharge from Norway at some times of the year as well as additional urban runoff. The differences in relative abundance and K among the three species may be a result of the different types and abundance of nutritional resources at the different sites. White sucker are bottom feeders and the increase at LAR26 may reflect a response to increased organic sediments and food supply therein below the two STPs and urban runoff. Common shiners and fallfish primarily feed on the surface. Fallfish prefer clear gravel bottom streams, which may explain their maximum abundance at LARa, which is above all the discharges and urban runoff and has the clearest water. Despite these small differences in species composition and K, there was no indication of large scale impacts below the PUD discharge.

PUD has submitted a report with a requested site-specific criterion for copper for the Little Androscoggin River developed using the Biotic Ligand Model. DEP will evaluate the request in light of the sediment issue discussed above.

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3.4. FISH SCALES AS NON-LETHAL BIOSENSORS OF SURFACE WATER CONTAMINANTS

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Abstract

There is great need for non-lethal, biologically relevant screening tools for assessing the effects of surface water contaminants on threatened or endangered fish species. Typical screening procedures are highly invasive or lethal to the fish. Recent studies show that fish scales biochemically respond to a range of contaminants, including toxic metals, organic compounds, and endocrine disruptors. Fish scales can be collected rapidly and non-lethally from fish, and are regularly collected by state and federal agencies, such as NOAA, DMR, and IFW. The overall goal of our work is to evaluate fish scales as non-lethal biosensors for pharmaceuticals, organic contaminants, endocrine disruptors, metals, and contaminant mixtures. We used cytochrome P4501A (CYP1A) as a biomarker for exposure to pharmaceuticals and organic contaminants (Meyers, 2009; van der Oost et al. 2003; Quiros et al. 2007) and will use estrogen receptor (ER) as a biomarker for endocrine disruptors (van der Oost et al. 2003; Pinto et al. 2009), and metallothionein (MT) as a biomarker for toxic metals (van der Oost et al. 2003; Suzuki et al. 2004); each of these biomarkers is inducible in fish scales (Meyers, 2009, Suzuki et al. 2003; Suzuki et al. 2004; Quiros et al. 2007; Pinto et al. 2009; Suzuki et al. 2009). Our hypotheses were that 1) biomarkers in fish scales can be used as contaminant biosensors and 2) these biomarkers are reliable in contaminant mixtures. We used Atlantic salmon (Salmo salar) as the test species. Atlantic salmon are an endangered diadromous fish, the smolts and adults of which migrate through polluted areas. Our project objectives were to 1) establish protocols for fish scale collection, storage, and biochemical analyses, and 2) evaluate biomarker induction in scales of S. salar exposed to contaminants. We have determined that the pollutant biomarker, cytochrome P4501A (CYP1A)) is 1) inducible in scales of Atlantic salmon (Salmo salar) parr aqueously exposed to polychlorinated biphenyls and polynuclear aromatic hydrocarbons, 2) is expressed in the epidermal covering of these scales as well as in scale osteocytes, and 3) is reliably and sensitively measured using RNA isolated from fish scales that can be stored indefinitely after scale collection. We optimized isolation of high quality RNA from fish scales for use in quantitative (real-time) reverse-transcriptase polymerase chain reaction (qRT-PCR) assays to detect fish exposure and biochemical response to chlorinated and polynuclear aromatic hydrocarbon contaminants. The strength of the scale assay is that it provides an in vivo biosensor of fish response (e.g. scales from wild-caught or caged fish can be used as first pass sensors to evaluate water quality in a given habitat). This technique could be used with any scale-bearing fish species. Screening for contaminants using fish scales provides a rapid, inexpensive, non-lethal and biologically relevant first pass indicator of water quality for sensing the presence of bioactive chemicals in surface water and the exposure to such compounds by endangered and threatened fish species.

Approach

Fish: Source and Maintenance

Juvenile land-locked Atlantic salmon (*Salmo salar*) were obtained from Grand Lake Stream hatchery (Maine) and maintained at the University of Maine Aquaculture Research Center. Fish were held in flow-through tanks at seasonally appropriate temperatures and light cycles and fed daily with trout chow provided by the hatchery. Fish were anesthetized with buffered MS222 prior to removal of scales. MS222 does not affect the activity of biotransformation enzymes in fish, including CYP1A (Kolanczyk et al. 2003). The ability to store scales indefinitely in RNALaterTM for molecular assays (e.g. RT-PCR) makes scales ideal for field-collection.

Fish Exposure

To induce CYP1A expression, we exposed Atlantic salmon parr to the organic contaminants 3,4,3',4',5-pentachlorobiphenyl (PCB126) and β -naphthoflavone (β NF). In each experiment, fish were exposed to aqueous doses of PCB126 (0.3, 3.27 and 32.7 ppb), β NF (330 ppb), vehicle (40 ppb acetone) or no treatment for 48 hours using 3 to 5 fish per dose, depending on the experiment.

Endpoints

We used three endpoints to evaluate CYP1A in fish scales: enzyme activity, protein location, and gene (mRNA) expression. Catalytic analysis of scale CYP1A is the most rapid way to determine if the induction of CYP1A is successful prior to conducting more time consuming analyses. To confirm the findings of Jen Meyers, a previous student in Dr. Elskus's lab, and to confirm the induction of CYP1A enzyme activity in our fish, we used the ethoxyresorufin-o-deethylase (EROD) assay (Meyers, 2009), with 15 scales per well, 3 wells per fish in a 96-well plate format, using 7-ethoxyresorufin as the substrate, essentially as described (Elskus et al. 1999).

To determine whether CYP1A is expressed in fish scale osteocytes or the epidermal covering of the scale, we used immunohistochemistry (IHC) analysis. We collected three 2 cm³ skin samples and one small snip of liver tissue, fixed these in formalin and had them paraffin embedded and sectioned (5 micron thick) by the University of Maine's Aquatic Animal Health Diagnostics Laboratory. We performed the IHC analyses at the Woods Hole Oceanographic Institution in the laboratory of Dr. John Stegeman under the supervision of Bruce Woodin. Three sets of slides were cut, and stained with hematoxylin and eosin (to establish tissue architecture), probed with MOPC 31 (non-specific antibody), or probed with MAb 1-12-3 (CYP1A specific antibody), essentially as described (Kloepper-Sams et al. 1987; Elskus et al. 1999). The H&E stained tissue layers were identified using Amin et al. 1992. The anti-body probed sections were examined under a microscope and stained tissues assigned scores for "Occurrence" (0-3) and "Intensity" (0-5) of staining, with final scores calculated as "Occurrence X Intensity" (0-15), as described (Elskus et al. 1999).

To measure scale CYP1A gene (mRNA) expression, we used quantitative real time polymerase chain reaction (qRT-PCR). Scales removed from treated fish were placed in RNAlaterTM, which allows indefinite storage at -20 C. We extracted RNA from the scales (30+ scales per sample per fish) and reverse transcribed the RNA to cDNA. We identified *S. salar* specific primers and optimized qRT-PCR parameters for *S. salar* (CYP1A and β -actin primer concentration and cDNA template concentration). Amplified scale CYP1A cDNA was quantified using the method of Pfaffl (Pfaffl 2001).

Results

Scale Architecture

The scales are arranged in an imbrication pattern, each partially encased in an epidermal 'pocket' that is partially retained when the scale is removed (Fig 1). It is within this epidermal covering, rather than in the scale bone itself, that we believe the CYP1A protein is expressed, based on our IHC results (see below).

CYP1A catalytic activity

As expected, CYP1A enzyme activity was induced over controls in the scales of β NF- and PCB126-treated fish, with a strong dose-response evident in PCB126-treated fish (Fig 3). This established CYP1A response in the fish scales, allowing us to proceed with IHC and qPCR analyses.

CYP1A protein location

CYP1A protein staining was present in all skin tissue layers (epidermis, scale, dermis, fat, and muscle) and in the liver of β NF- and PCB126-treated fish (Figs 2, 4). From these results, it appeared that CYP1A is expressed in the epidermal covering of the scale, rather than in scale bone cells. Although we observed staining in the vicinity of the scale bone in some samples, we believed this to be a staining artifact, due to physical capture of stain precipitate by the circuli of the scales, rather than staining of CYP1A protein in living osteocytes. We re-ran the IHC assays on new sections from these blocks at WHOI in March 2011 and discovered that, in fact, the scale osteocytes are expressing CYP1A. Thus the signal we observed and measured earlier was not an artifact but was indeed true staining.

Scale CYP1A mRNA expression

We identified 30 as a sufficient number of scales for extracting good quality, high yield scale RNA. An earlier paper (Quiros et al. 2007) used 1 - 3 scales per fish, however our trials with fewer scales (1 - 10) produced low yield, poor quality RNA (data not shown).

Scale CYP1A gene expression was elevated in all treated fish relative to the controls, with a strong dose-response in scales of PCB126 treated fish (Figs 4 & 5).

Discussion

Scale CYP1A enzyme activity is readily inducible in PCB126- and β NF-treated fish and is easily measured through EROD, but ultimately EROD is not a field friendly assay. Meyers (2009) reported rapid (hours) and sustained (34 days) induction in scales from *S. salar* exposed to PCB126 for 24 hours. We confirmed rapid induction of scale CYP1A activity by organic contaminants. Unfortunately, the need for rapid analysis of enzyme activity (within 24 h of scale collection, Meyers, 2009) makes scale CYP1A enzyme activity unsuitable for field-collected samples.

CYP1A protein in scales is likely located in the epidermal tissue surrounding the scales and not in the scales themselves. The aryl hydrocarbon receptor (AhR), which regulates CYP1A induction, is expressed in the osteocytes of mouse bone cells (Wejheden et al. 2010), but AhR expression in fish scale osteocytes is unexplored. We observed unmistakable CYP1A staining in scale osteocytes, indicating that like mouse bone, it is likely AhR is expressed in fish scale osteocytes as well. Work in fathead minnows identified CYP1A expression in chondroid cells (hyaline cartilage), which are

associated with skeletal formation {Iwata, 2000 #20299; Lindstromseppa, 1994 #20306}, but we know of no other work investigating CYP1A or AhR expression in fish bone cells.

We demonstrated that CYP1A gene expression in scales is strongly induced by organic contaminants. Scale CYP1A mRNA expression differed 1,500-4,000 fold from controls in juvenile salmon aqueously exposed for 48 hours to 330 ppb β NF using 30 scales/sample. In the only other comparable study published to date, scale CYP1A mRNA expression was induced 10 to 50 fold over controls in goldfish (*Carassius auratus*) injected intraperitoneally with 50 mg β NF/kg (50,000 ppb) with signal measured 8-72 h post-exposure using 1-3 scales/sample (Quiros et al. 2007). The difference between these outcomes could be due to differences between species, routes of exposure, exposure time, dose, number of scales extracted, and quality of the RNA extracted. Our work is the first to report scale CYP1A mRNA induction by a chlorinated inducer (PCB126), with strong doseresponse to this toxicant.

Our work demonstrates that scale CYP1A mRNA is a sensitive indicator of fish exposure to organic contaminants that is non-lethal, field-friendly (i.e. scales can be stored indefinitely prior to processing), and easily measured. We are continuing this project and will next evaluate CYP1A mRNA expression in scales of Atlantic salmon exposed to the pharmaceutical fluoxetine (Prozac). To evaluate scales as biomarkers for other prominent contaminant classes we will evaluate scale biomarkers for endocrine disruptors (estrogen receptor (ER) mRNA, Pinto et al. 2009) and toxic metals (metallothionein (MT) mRNA, Suzuki et al. 2004)). Because fish are exposed to mixtures of contaminants, which can alter the expression of biomarkers in various ways, we will evaluate mixture effects on scale CYP1A, ER, and MT mRNA expression. We will evaluate our mixture results based on published interactions that indicate metals can inhibit ER mRNA expression (Suzuki et al. 2004) and estrogens can inhibit CYP1A mRNA expression (Navas et al. 2001). As chemical-specific biomarkers do not currently exist for any pollutant, our aim is to detect inappropriate up-regulation of genes by different classes of chemicals as an indicator of potential water quality issues. We selected endpoints based on their low expression levels in un-impacted fish: ER mRNA levels are vanishingly low in male and juvenile fish not exposed to estrogens, MT levels are low in fish not exposed to exogenous metals, and CYP1A is nearly non-detectable except in fish exposed to certain organic contaminants. In isolation, these endpoints are not indicative of toxic effects. Rather, induction of these biochemical responses points to potential problems with water quality, triggering in-depth water chemistry analysis targeting the chemical classes inducing these endpoints. Scale biosensors are not meant to identify contaminant concentrations, except in a relative sense (absence/presence, low/high concentrations). While biochemical responses to toxicants may differ among tissues, the strength of any sensor lies in its ability to respond to the chemicals of interest.

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Figure 1. Light microscope (left) and scanning electron microscope (right) images of *S. salar* scale (left) and skin sample (right). Black line indicates extent of epidermal covering left on scale after removal. Images courtesy of E. Kelly, University of Maine.



Figure 2. Histology sections of skin from a BNF-exposed fish. Red circles indicate CYP1A protein staining



Figure 3. Representative CYP1A activity profiles of *S. salar* scales (15 scales/well; 3 wells/fish) from fish exposed for 48 hours to vehicle (acetone), PCB126 (3.27 or 32.7 ppb) or BNF (330 ppb) in the water. Y-axis is relative fluorescence units x 1000.



Figure 4. Relative CYP1A protein expression in liver and skin layers of contaminant-exposed fish. Staining was observed in epidermis (red circle) with faint staining in scale osteocytes (**). Mean IHC score +/- SE (n=3). The IHC assay was repeated in March 2011 with much stronger staining in all tissues; those sections are currently being scored.



Figure 5. CYP1A mRNA expression fold induction in treated fish relative to controls. CYP1A mRNA values normalized to β -actin mRNA using the Pfaffl (2001) method. Mean +/- SE (n=5 fish).