THE NEW HAMPSHIRE *GULFWATCH* PROGRAM: 1998

A PART OF THE EIGHTH YEAR OF THE GULF OF MAINE ENVIRONMENTAL MONITORING PLAN

July, 2000

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New Hampshire Department of Environmental Services
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INTRODUCTION

Rationale

The Gulf of Maine extends from Cape Sable, Nova Scotia, through New Brunswick, Maine, and New Hampshire to Cape Cod, Massachusetts, and includes the Bay of Fundy and Georges Bank. The combined primary productivity of seaweeds, salt marsh grasses, and phytoplankton make it one of the worlds most productive system that supports a vast array of animal species, including many species of invertebrates, fish, seabirds, and marine mammals, some of great commercial importance. Commercial fisheries are its principal income generating enterprises, although tourism is also a significant source of income to many small coastal communities, and the marine aquaculture industry is rapidly expanding. As coastal populations around the Gulf and its watersheds have increased, forests and agricultural lands have been converted to industrial and residential developments. Such changes in land use and increases in population have contributed to the deteriorating quality of sections of the coastal environment (Crawford and Sowles, 1992; Dow and Braasch, 1996). Inputs from non-point source and point source pollution are a significant threat to the nearshore environment of the Gulf (Crawford and Sowles, 1992; Dow and Braasch, 1996). Growth in industrial activity during the 20th century has resulted in a steady input of chemicals, either mobilized or synthesized by man, into the estuarine and coastal environments. Many of these chemicals are bioaccumulated to concentrations significantly above ambient levels. Furthermore, some of these environmental contaminants may also be present at toxic concentrations, and thus induce adverse biological effects.

In order to protect water quality and commercial uses in the Gulf of Maine, the *Agreement on the Conservation of the Marine Environment of the Gulf of Maine* was signed in December, 1989 by the premiers of Nova Scotia and New Brunswick, and the governors of Maine, New Hampshire and Massachusetts, establishing the Gulf of Maine Council on the Marine Environment. The overall mission of this council is to maintain and enhance the Gulfs' marine ecosystem, its natural resources and environmental quality.

To help meet the council's mission statement the Gulf of Maine Monitoring Committee was formed and charged with the development of the Gulf of Maine Environmental Monitoring Plan (Hayden, 1991). The Monitoring Plan is based on a mission statement provided by the Council:

> **It is the mission of the Gulf of Maine Environmental Quality Monitoring Program to provide environmental resource managers with information to support sustainable use of the Gulf and allow assessment and management of risk to public and environmental health from current and potential threats.**

Three monitoring goals were established to meet the mission statement:

(1) To provide information on the status, trends, and sources of risk to the marine environment in the Gulf of Maine;
(2) To provide information on the status, trends, and sources of marine based human health risks in the Gulf of Maine; and
(3) To provide appropriate and timely information to environmental and resource managers that will allow both efficient and effective management action and evaluation of such action.

In support of the mission, and to meet the desired goals, a project named Gulfwatch was established to measure Gulfwide chemical contamination. The Gulfwatch program was modeled after the NOAA National Status and Trends (NS&T) Mussel Watch program, and presently uses the blue mussel, *Mytilus edulis*, as an indicator for habitat exposure to organic and inorganic

contaminants. Bivalves like *M. edulis* have been used successfully as indicator organisms in environmental monitoring programs throughout the world (see NAS, 1980; NOAA, 1991; and Widdows and Donkin, 1992; Cantillo, 1998) to identify variation in chemical contaminants between sites, and contribute to the understanding of trends in coastal contamination (NOAA, 1991; O'Connor, 1992; O'Connor and Beliaeff, 1995; Widdows et al., 1995; Cantillo, 1998). The blue mussel was selected as the indicator organism for the Gulfwatch program for the following reasons:

(1) mussels are abundant within and across each of the 5 jurisdictions of the Gulfwatch Program and they are easy to collect and process;
(2) much is known about mussel biology and physiology;
(3) mussels are a commercially important food source and therefore a measurement of the extent of chemical contamination is of public health concern;
(4) mussels are sedentary, thereby eliminating the complications associated with the interpretation of results introduced by mobile species;
(5) mussels are suspension-feeders that pump large volumes of water and concentrate many chemicals in their tissues; therefore the presence of trace contamination is easier to document; and the measurement of chemicals in bivalve tissue provides an assessment of biologically available contamination that is not always apparent from the measurement of contamination in environmental compartments (water, sediment, and suspended particles).

Gulfwatch has taken two approaches to using marine mussels as bioindicators of anthropogenic contamination. The two approaches are use of transplanted mussels in cages to assess short-term contamination and sampling of native mussels. Native mussel sampling occurs in a subset of sites each year on a three year rotation while transplanting occurred only every three years at a few sites. Thus, both transplanted and native mussels sampled from areas adjacent to the transplant sites were analyzed for organic and inorganic contaminants (Crawford and Sowles, 1992) during 1991, 1992 and 1995. In 1993, 1994, 1996 and 1997, only indigenous mussels were sampled, although a greater number of sites were monitored compared to the years when mussels were transplanted (Chase et al., 1998; Chase et al., 1996a; Sowles et al, 1996). Sampling of native mussels provides an assessment of long-term exposure to bioavailable contaminants (on the order of months to a year). The 1998 sampling year followed the protocol for 1993-94 and 1996-97, sampling indigenous mussels from three to seven sites in each jurisdiction.

In addition to documenting the level of contaminants in mussel tissue, biological variables, including shell growth and condition index, have been measured as a means to determine the response of organisms to stress under different concentrations of contaminant burden. Growth is often one of the most sensitive measures of a contaminant's effect on an organism (Sheehan, 1984; Sheehan et al., 1984; Howells et al., 1990). However, growth measurements are dependent on protocols that are only used when transplanting of mussels occurs, thus growth could not be measured in 1998 on the indigenous mussels.

Condition Index (CI) has been used as an indicator of the physiological status of the mussels. It relates the tissue wet weight to shell volume and is a measure traditionally used by shellfishery biologists (Widdows, 1985). Because gonadal weight is a significant contributor to total body weight just prior to spawning, CI also reflects differences in the reproductive state of the sampled mussels. Since gonadal material tends to have low concentrations of metals (LaTouche and Mix, 1981), tissue metal concentrations may be reduced in mussels having a high CI due to ripened gonads. Organic contaminants, however, would tend to partition into both somatic and gonadal lipids, and may be less impacted by changes in CI that are due to the presence of ripe gametes. Since variable amounts of ripe gametes may be found in some mussel populations, even in late fall (Kimball, 1994), the relationship between CI and contaminant concentrations must be carefully considered. CI has been measured on mussel samples every year, including 1998.

After the success of the pilot studies in 1991 and 1992, it was recognized that there should be a broader, or Gulf-wide orientation of the program, in addition to known contaminated and reference sites within each jurisdiction. As such, a three year cycle was initiated in 1993 and the sample design was expanded (Jones et al., 1998). Native mussels were sampled in as many as seven new locations within each jurisdiction (state or province), where feasible, to increase the geographic coverage. However, one location in each jurisdiction was chosen as a baseline station, to be re-sampled every year. Increasing the number of sampling locaitons in each jurisdiction increased the chance of locating unforseen environmental contamination. The present program's three-year cycle, with indigenous mussels alone being sampled at 2-7 sites per jurisdiction, will be repeated for the remaining years of the present Gulfwatch Monitoring Plan. This sampling design allows the program investigators to assess both short-term and long-term contaminant exposures. The 1998 samples are the second sampling of sites previously sampled in 1995. The same sites will be re-sampled in 2001, the final year of the Plan.

New Hampshire Gulfwatch Program and Objectives

The goal of the Gulfwatch program in New Hampshire is to provide a more comprehensive assessment of toxic contaminant exposure, particularly oil, to biota in Seacoast waters. The specific objectives are:

1) to develop a baseline database for contaminant exposure concentrations for New Hampshire mussels in coordination with the broader Gulf of Maine Gulfwatch program;
2) to determine the impact and fate of spilled oil in the biota of the Great Bay Estuary;
3) to develop a petroleum contamination baseline to assist in damage assessment in the event of an oil spill;
4) to expand the geographical coverage of sampling sites to include mussels located in critical habitats areas of the Seacoast.

The New Hampshire Gulfwatch Program increases the frequency of sampling, resulting in a four-year schedule for New Hampshire sites that otherwise would be on a six-year schedule as part of the Gulf-wide program. The same protocols used in the Gulf-wide program have been adopted for all New Hampshire sampling and analysis. This expanded New Hampshire program is needed to answer pressing environmental questions, provide a baseline of data for assessing impacts of both source elimination efforts and accidental contaminant spills, and to take advantage of the ongoing Gulf-wide program that provides a critical regional perspective for interpretation of New Hampshire data.

METHODS

The 1998 Gulfwatch sample collection and analysis is the sixth year of the program's nine year sampling design (see Sowles et al., 1997). The 1998 sampling represents the third year of the second 3-year cycle. As such, some of the New Hampshire stations that were sampled in 1998 were the same stations sampled in 1995. Therefore, in addition to spatial analysis, temporal analysis can be performed on the contaminant concentrations for those sites. For other New Hampshire sites, 1998 is the first year of sampling. The New Hampshire Gulfwatch program includes six new sites in addition to six previously sampled sites.

1998 Sampling Locations

The stations sampled for 1998 are shown in Figure 1 and Table 1. Table 1 also shows the schedule for sampling at all sites over the planned four year program for New Hampshire. In 1998, three sites had been previously sampled, NHLH, NHDP and MECC, and three sites that were sampled for the first time: NHGP, NHSS and NHNM. MECC (Clark Cove) is a benchmark site that has been sampled each of the previous five years to enable trend analysis. Clark Cove is on Seavey Island, which is in either Maine or New Hampshire pending current court action. It is included as a New Hampshire site because it is located in the Great Bay/Piscataqua River watershed, and therefore most comparable to other sites in New Hampshire.

Field Procedures

Details regarding the mussel collection, measurement, and sample preparation are published in Sowles et al. (1997), however a summary of the procedures are given below. The mussels collected were intended to be *Mytilus edulis*. A similar species, *Mytilus trossulus*, was identified in some 1993 Bay of Fundy samples (Sowles et al., 1996). This species has a slower growth rate and attains a smaller maximum size compared to *M. edulis* (Bayne, 1976), and it has been shown that there are inter-specific differences in concentrations of certain metal (Cu, Ni, Pb, Hg and Zn) and organic (ΣPAH_{24}) contaminants (Mucklow, 1996). However, this has not been an issue in New Hampshire.

All field sampling was conducted in the fall of 1998 as outlined in Sowles et al. (1997). Collection times were set to avoid collecting during or shortly after periods when stormwater runoff and wave resuspension of bottom sediment could result in enhanced uptake and accumulation of sediment in the mussel gut. The presence of sediment in the mussels was suspected to be the cause of the elevated concentrations of some metals (iron, aluminum and associated metals: Lobel et al., 1991; Robinson et al., 1993) in previous reports (Sowles et al., 1994, 1996; Chase et al., 1996a, b, 1997).

Mussels were collected from four discrete areas within a segment of the shoreline that is representative of local water quality. Using a wooden gauge or a ruler to measrue length, 45-50 mussels of 50-60 mm shell length were collected. The mussels were cleaned of all sediment, epibiota, and other accretions in clean seawater from the collection site, placed in clean containers, then transported to the lab in coolers with ice packs. Prior to shucking, mussels were thoroughly rewashed to minimize tissue contamination from any remaining surface debris.

Laboratory Procedures

In the laboratory, individual mussel lengths, widths and heights (as defined by Seed, 1968) were determined to the nearest 0.1 mm using vernier calipers. Using plastic or stainless steel wedges, mussels were shucked directly into appropriately prepared Mason jars for metal and

Figure 1
Location of 1998 Gulfwatch Sampling Stations

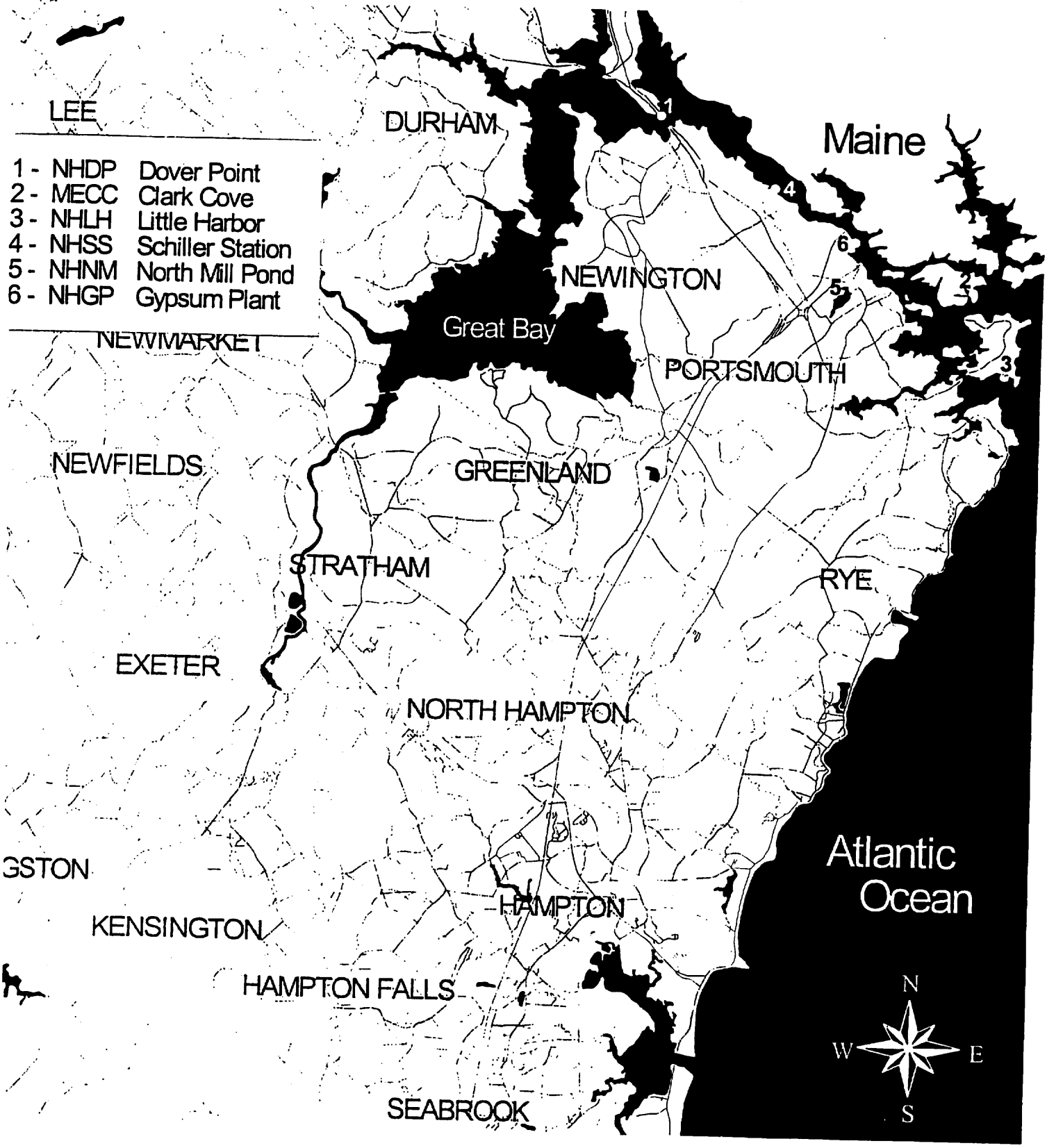


Table 1. New Hampshire *Gulfwatch* four-year sampling schedule.

Station: south to north	New sites	Station code	Latitude	Longitude	Year sampled 1998	1999	2000	2001
Hampton/Seabrook Estuary		NHHS	42°53.5'N	70°49.0'W		X		X
Rye Harbor		NHRH	43°00.0'N	70°44.4'W			X	
Little Harbor		NHLH	43°03.3'N	70°43.0'W	9/28/98			X
South Mill Pond	**	NHSM	43°04.5'N	70°45.6'W		X		X
Pierce Island	**	NHPI	43°04.3'N	70°44.6'W		X		X
Clark Cove		NHCC	43°04.4'N	70°43.4'W	9/25/98	X	X	X
NH Port Authority	**	NHPA	43°05.1'N	70°45.5'W		X		X
North Mill Pond	**	NHNM	43°04.5'N	70°45.6'W	9/28/98		X	
Gypsum plant	**	NHGP	43°05.1'N	70°45.5'W	9/28/98			
PSNH Schiller Station	**	NHSS	43°06.1'N	70°47.3'W	9/25/98		X	
Dover Point		NHDP	43°07.1"N	70°49.4"W	9/28/98	X	X	
Fox Point		NHFP	43°07.1'N	70°51.4'W		X		X

organic analysis, respectively (for details see Sowles et al., 1997). Composite samples (20 mussels/composite; 4 composites/station) were capped, labelled and stored at -15°C.

While a number of condition indices have been proposed over the years (Seed, 1968), the Gulfwatch Condition Index (CI) has been defined as:

CI = tissue wet weight (mg) / length (mm) * width (mm) * height (mm)

CI was determined on a minimum of 30 mussels.

Analytical Procedures

The analytical procedures used followed those reported for the previous years (Sowles et al., 1994, 1996; Chase et al., 1996a, b, 1997, 1998). Table 2 contains a summary of trace metal and organic compounds measured.

Metals

Inorganic contaminants were analyzed at the State of Maine Health and Environmental Testing Laboratory (Augusta, ME). Analyses for mercury were carried out on a sub-sample of 1 to 2 g of wet tissue and measured by cold vapor atomic absorption on a Perkin Elmer Model 503 atomic absorption spectrometer. Analyses for all other metals were conducted on 5 to 10 g of wet tissue dried at 100°C. Zinc and iron were measured by flame atomic absorption using a Perkin Elmer Model 1100 atomic absorption spectrometer. All remaining metals (Ag, Al, Cd, Cr, Cu, Ni and Pb) were run using Zeeman background corrected graphite furnace atomic absorption on a Varian Spectra AA 400. The analyte detection limits for the metals in µg/g dry weight are as follows; Ag, 0.1; Al, 4.0; Cd, 0.1; Cr, 0.2; Cu, 0.4; Fe, 4.0, Hg, 0.1, Ni, 0.4, Pb, 0.4; and Zn, 0.5.

Organics

Organic contaminants in mussel samples were analyzed at the Environment Canada, ECB Laboratory in Moncton, NB. The analyte detection limits ranged from 3.0 to 10 ng/g for aromatic hydrocarbons, from < 1.0 to 2.0 ng/g for PCB congeners, and from < 1.0 to 2.0 ng/g for chlorinated pesticides. Eighteen of the PCB congeners identified and quantified correspond to congeners analyzed by the National Oceanographic and Atmospheric Administration's (NOAA) National Status and Trends (NS&T) Program designated congeners. Other organic compounds selected for analysis are also consistent, for the most part, with NOAA National Status and Trends mussel monitoring (NOAA, 1989).

A description of the full analytical protocol and accompanying performance based QA/QC procedures are found in Sowles et al. (1997) and Jones et al. (1998). Tissue samples were extracted by homogenization with an organic solvent and a drying agent. Solvent extracts were obtained by vacuum filtration, and biomatrix interferences were separated from target analytes in extracts by size exclusion chromatography. Purified extracts were subjected to silica gel liquid chromatography which provided a non-polar PCB/chlorinated pesticides fraction and a polar chlorinated pesticide fraction. PCBs and pesticides were analyzed by High Resolution Dual Column Gas Chromatography/Electron Capture Detection (HRGC/ECD). Following PCB and pesticide analysis, the two fractions were combined and the resulting extract was analyzed for aromatic hydrocarbons by High Resolution Gas Chromatography/Mass spectrometry(HRGC/MS).

Mussel tissue was also analyzed for planar chlorobiphenyls (CB), polychlorinated dibenzodioxins (PCDD) and polychlorinated dibenzofurans (PCDF) at a subset of sites in 1998 that included North Mill Pond (NHNM) and Little Harbor (NHLH). Since 1996, the Gulfwatch program has analyzed mussels for these compounds in six different sites in New Hampshire. The

Table 2. Inorganic and organic contaminants analyzed in mussel tissues from the Seacoast of New Hampshire in 1998.

INORGANIC CONTAMINANTS

Metals

Ag, Al, Cd, Cr, Cu, Fe, Hg, Ni, Pb, Zn

ORGANIC CONTAMINANTS

Aromatic Hydrocarbons

Naphthalene
1-Methylnaphthalene
2-Methynaphthalene
Biphenyl
2,6-Dimethylnaphthalene
Acenaphthylene
Acenaphthene
2,3,5-Trimethylnaphthalene
Fluorene
Phenanthrene
Anthracene
1-Methylphenanthrene
Flouranthene
Pyrene
Benzo [a] anthracene
Chrysene
Benzo [b] flouranthrene
Benzo [k] flouranthrene
Benzo [a] pyrene
Benzo [e] pyrene
Perylene
Indeno [1,2,3-cd] pyrene
Dibenzo [a,h] anthracene
Benzo [g,h,i] perylene

Chlorinated Pesticides

Hexachlorobenzene (HCB)
gamma-hexachlorocyclohexane (HCH)
Heptachlor
Heptachlor epoxide
Aldrin
Mirex
cis-Chlordane
trans-Nonachlor
Dieldrin
Alpha-Endosulfan
beta-Endosulfan

DDT and Homologues

2,4'-DDE	4,4'-DDE
2,4'-DDD	4,4'-DDD
2,4'-DDT	4,4'-DDT

PCB Congeners

PCB 8, PCB 18, PCB 28, PCB 29, PCB 44, PCB 50, PCB 52, PCB 66, PCB 77, PCB 87, PCB 101, PCB 105, PCB 118, PCB 126, PCB 128, PCB 138, PCB 153, PCB 169, PCB 170, PCB 180, PCB 187, PCB 195, PCB 206, PCB 209

analyses were conducted at Axys Analytical Services Ltd, Sydney, BC.

Quality Assurances/Quality Control

Standard laboratory procedures for metals incorporated method blanks, spike matrix samples, duplicate samples, surrogate addition and standard oyster tissue (SRM 1566A). The method blanks were inserted: three at the beginning of the run, one at the end, and six at various intervals during the run. Duplicate samples and matrix spike recoveries were conducted on 15% of the samples.

The Moncton laboratory participated in the NIST Status and Trends Intercomparison Marine Sediment Exercise IV and Bivalve Homogenate Exercise. Internal quality control and method performance specifications are described in the Environment Canada Shellfish Surveillance Protocol (Sowles et al., 1997; Jones et al., 1998). The protocol includes mandatory QC measures with every sample batch including method blanks, spike matrix samples, duplicate samples, surrogate addition, and certified reference materials (SRM, 1974a). The protocol specifies the performance criteria relevant to method accuracy, precision, and detection limits and data reporting requirements for the analysis of organic contaminants in shellfish samples.

Bacterial Analyses

Both water and mussel tissue samples were analyzed for bacterial indicators of fecal contamination, including enterococci, *Escherichia coli* and fecal coliforms. Water samples were shaken vigorously for 30 sec before an aliquot was removed and filtered through a sterile 0.45 μm Gelman filter. Filters were placed on mE agar (Difco) for enterococci enumeration and mTEC media (Difco) for coliform and *E. coli* enumeration then incubated for 48 h at 41°C and 18-24 h at 44.5°C, respectively (U.S.E.P.A., 1986). Filters from mE plates were transferred onto EIA agar plates and incubated at room temperature for 20 minutes; colonies forming a dark precipitate were counted as positive. All yellow colonies on mTEC agar were counted as the fecal coliforms; fecal coliforms were further characterized by transferring the filters onto cellulose pads soaked with urea and counting the remaining yellow colonies as *Escherichia coli*.

Mussel samples were inoculated into a multiple tube fermentation (MPN) analysis series using an initial LT medium selective enrichment (Motes and Peeler, 1991). The MPN tubes were incubated at 35°C for 18-24 h at which time turbid tubes with gas production were considered indicative of a positive reaction. Samples from the positive tubes were transferred to EC/MUG tubes and incubated for 24 h at 44.5°C. Fecal coliforms were enumerated based on turbid, gas-positive tubes and *E. coli* enumerated based on fecal coliform positive tubes that fluoresced under UV light.

Statistical Methods

Data Analysis

All metal data were $\log_{10}$ transformed to correct for heterogeneity of variances. In several cases there were non-detectable (ND) data values. If all 4 replicates from a given site showed ND concentrations, the contaminant level was recorded as ND, but if at least one of the replicates was greater than the detection limit, then the other replicates were recorded as 1/2 the detection limit. Arithmetic means were used to summarize the results of replicate samples and are used in all subsequent tables and figures. In addition, geometric means were calculated for each metal for comparison with other data sets. The standard deviation (s) around the geometric mean (s_g) was calculated as:

$$s_g = \text{antilog}(s_l) = 10^{s_l} \quad (1)$$

where s_l = the standard deviation around the mean of the $\log_{10}$ transformed data (Snedecor and Cochran, 1967).

Total PAH (ΣPAH_{24}), total PCB (ΣPCB_{24}) and total pesticides ($\Sigma TPEST_{17}$) values were created from the sum of all individual compounds or congeners with values greater than the detection limit for the compound. Total DDT (ΣDDT_6) is the sum of o,p'-DDT and p,p'-DDT and homologues (o,p'-DDE; p,p'-DDE; o,p'-DDD; and p,p'-DDD). Organic variables in which all replicate measurements were below the detection limit were treated as zero. All data were $\log_{10}$ (x+1) transformed to correct for non-normality. Arithmetic means were used to summarize the results of replicate samples and are used in all subsequent tables and figures. In addition, geometric means were calculated for comparison with other data sets. The standard deviation around the geometric means were calculated as described (Eq. 1).

Spatial Analysis

Arithmetic means and standard deviations of all values for each metal and organic contaminant at each station were calculated. Arithmetic means were calculated because metals and organics at each station were normally distributed, with a few exceptions. Graphs of the mean concentrations (±SD) are presented for all stations sampled. Differences in metal and organic contaminant concentrations among sites were analyzed using one-way analysis of variance (ANOVA), followed by Tukey-Kramer multiple comparison test of means. A probability of ≤ 0.05 was chosen as the level of significance.

Temporal Analysis

Temporal analysis was performed on the 1998 sampling sites where sampling has occurred in past years (n=3 sites, n=3-6 years). Tissue contaminant concentrations at the sites (MECC, NHLH, NHDP) were analyzed for temporal trends using ANOVA. A probability of ≤ 0.05 was chosen as the level of significance.

RESULTS AND DISCUSSION

Field Operations and Logistics

Field collection proceeded as planned at all sites. Coordination between NHDES and UNH/JEL allowed for collection of mussels at all six sites within a three day span to minimize differences between sites that could result from time of collection.

Metal Contaminants

Table 3 contains the metal concentrations (arithmetic mean ± SD, μg/g dry weight) for mussels from all sites sampled in 1998. Metal concentrations for each of the composite samples (n=4) are provided in Appendix A. Overall metal concentrations for indigenous mussels are given as geometric means and standard deviations (Table 3). The concentration of Ag was below the detection limit (0.1 μg/g DW) in all of the tissue samples. The concentrations of all detectable metals ranged over relatively narrow concentrations, with lead having the greatest difference (1.9x) between the highest and lowest site means. In addition, no site had extremely high concentrations of any trace metal. Clark Cove (MECC) on Seavey Island had the highest Cr, Cu, Ni, Pb, Zn, Al and Fe concentrations, Dover Point (NHDP) had the highest Cd concentrations and the Schiller Station site (NHSS) had the highest Hg concentrations. The Gypsum Plant site (NHGP) had relatively low concentrations of all of the trace metals.

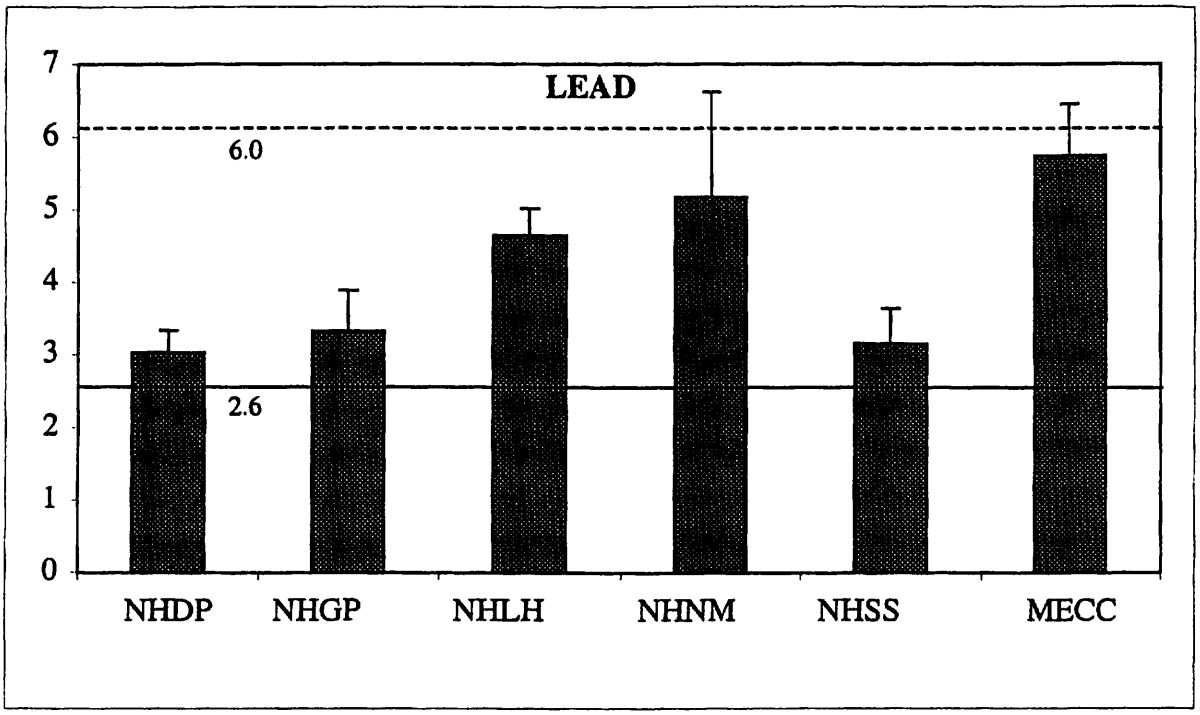
Comparisons of trace metal means were made in Table 3, and those followed by the same letters indicate no significant differences between sites. There were significant differences in mean concentrations between sites for all trace metals, except for Cr and Zn. The fact that MECC had the highest concentrations of Al and Fe suggests a higher degree of contamination of the tissue with inorganic sediments, which in turn may suggest that the observed elevated levels of some trace metals at this site may be caused by contaminated sediments (Robinson et al., 1993). However, statistical comparisons of means for Fe and Al concentrations show the concentrations at MECC were not significantly higher than most of the other sites. In contrast, the concentrations of Cu, Ni and Pb at MECC were significantly higher than two, one and three other sites, respectively. The Cd concentration at NHDP was significantly higher than at two other sites, while the concentration of Hg at NHSS was significantly higher than at three other sites. These results show that MECC, NHDP and NHSS have elevated concentrations of these trace metals compared to other sites in coastal New Hampshire, which may indicate sources of these contaminants close to these sites.

Spatial Variation in Metal Concentrations

Figures 2 through 5 show the concentrations of the metals measured in the tissue of *M. edulis* at the 1998 sampling stations, presented from north and up in the Great Bay Estuary to south into the mouth of Portsmouth Harbor and around to Little Harbor. In addition, the mean tissue metal concentrations at each of the Gulfwatch sites are compared with two "benchmark" values for each metal previously reported by Sowles (1993) from 23 Maine reference sites: (1) the arithmetic mean for each metal concentration (Maine Reference Mean or ME-RM); and (2) the arithmetic mean plus three standard deviations (Maine High Value or ME-HV; referred to by Sowles as the "anomalous value"). These Maine reference stations are located in areas where anthropogenic contamination should be low. Maine Reference concentrations are likely to be lower than those observed at several of the Gulfwatch stations.

Table 3. Tissue metal concentrations (µg/g dry weight; average ± SD) for New Hampshire Gulfwatch mussels in 1998. The geometric mean of all New Hampshire sites is given below. ANOVA of geometric means denoted by letters; same letter indicates no significant difference between means. n = 4 replicates per sample

Station	Ag	Al	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn
NHDP	ND	202±39^{ab}	2.80±0.28^{b}	2.95±0.06^{a}	6.06±0.69^{ab}	385±50^{ab}	0.97±0.05^{abc}	1.70±0.20^{ab}	3.02±0.31^{a}	130±14^{a}
NHGP	ND	175±49^{a}	1.92±0.52^{a}	2.08±0.56^{a}	4.70±1.27^{a}	358±103^{a}	0.86±0.09^{ab}	1.35±0.24^{ab}	3.32±0.56^{a}	111±25^{a}
NHLH	ND	162±31^{a}	2.42±0.10^{ab}	2.75±0.97^{a}	5.12±0.33^{a}	400±45^{ab}	1.00±0.05^{bc}	1.72±0.17^{ab}	4.65±0.37^{ab}	105±17^{a}
NHNM	ND	260±54^{ab}	1.98±0.37^{a}	2.32±0.43^{a}	6.55±0.60^{ab}	482±99^{ab}	0.79±0.12^{a}	1.24±0.20^{a}	5.18±1.45^{b}	135±21^{a}
NHSS	ND	192±34^{ab}	2.25±0.51^{ab}	2.30±0.18^{a}	6.12±0.49^{ab}	385±38^{ab}	1.08±0.10^{c}	1.45±0.24^{ab}	3.15±0.48^{a}	128±10^{a}
MECC	ND	298±64^{b}	2.08±0.13^{ab}	3.18±0.69^{a}	7.20±0.67^{b}	528±80^{b}	0.82±0.11^{ab}	2.32±1.08^{b}	5.75±0.70^{b}	135±24^{a}
Geometric mean	ND	206±1	2.20±1.24	2.52±1.28	5.85±1.23	413±1	0.91±1.16	1.57±1.32	4.00±1.35	122±1



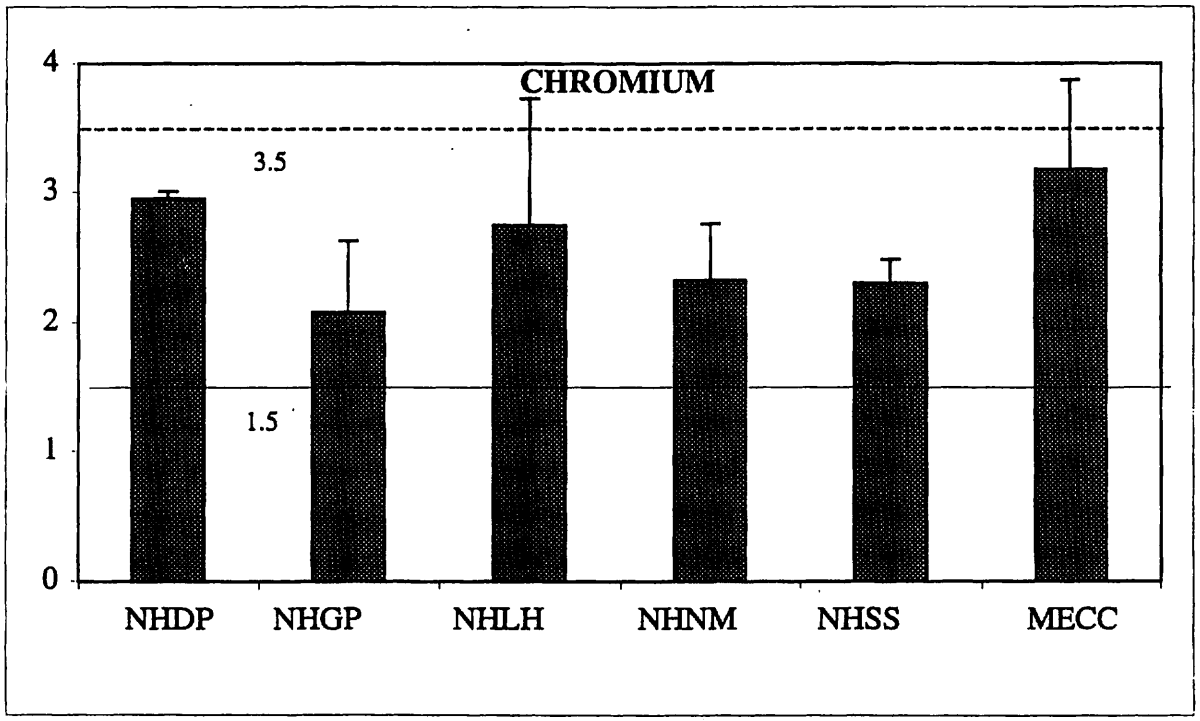
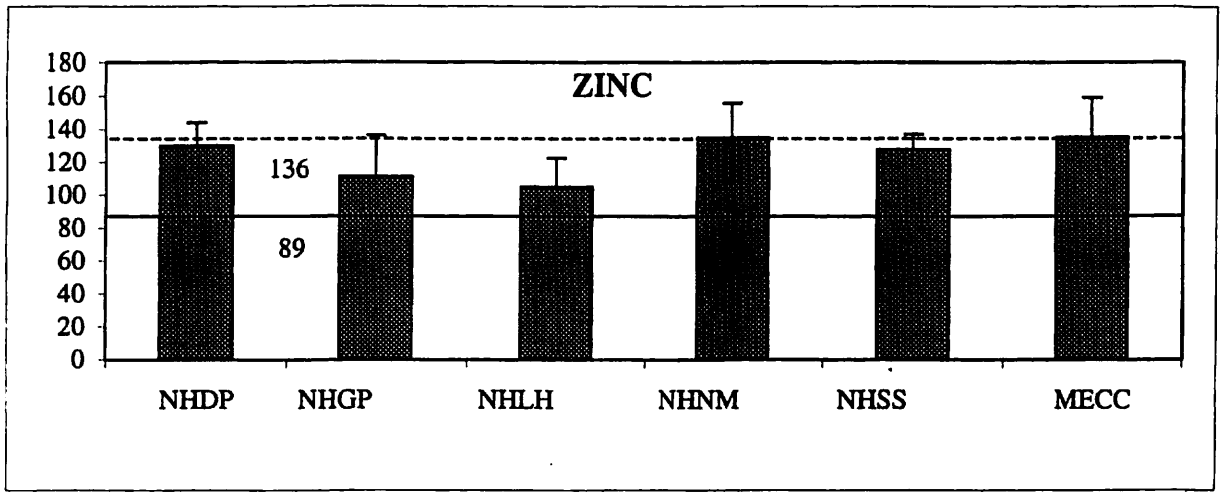
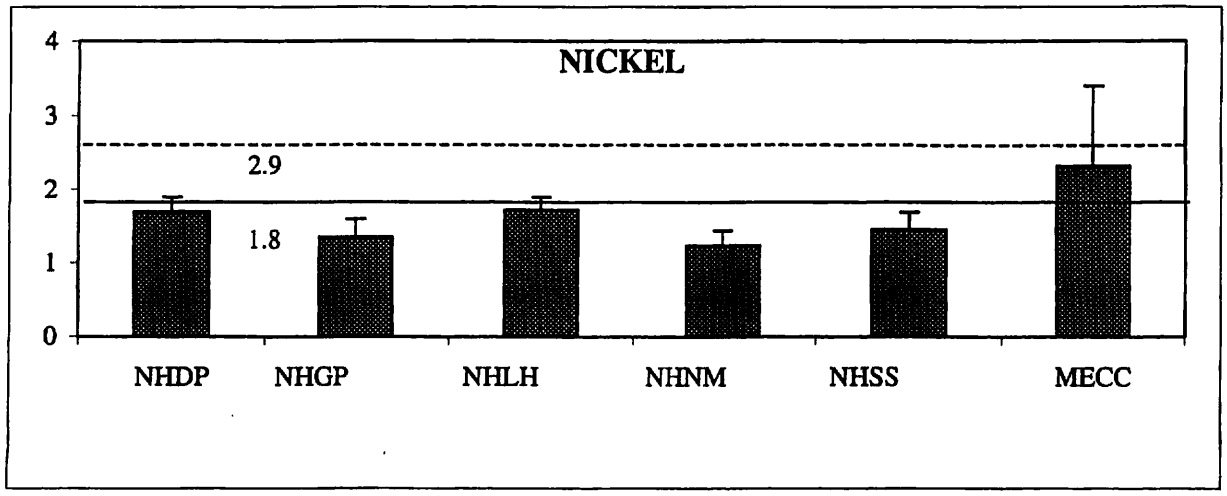


Figure 2. Distribution of lead and chromium tissue concentrations (arithmetic mean ± SD, µg/g DW) in mussels at the New Hampshire Gulfwatch stations in 1998. The reference mean, ME-RM (solid line) and the high value, ME-HV (dashed line) from the Maine reference data (Sowles, 1993) are shown for comparison.





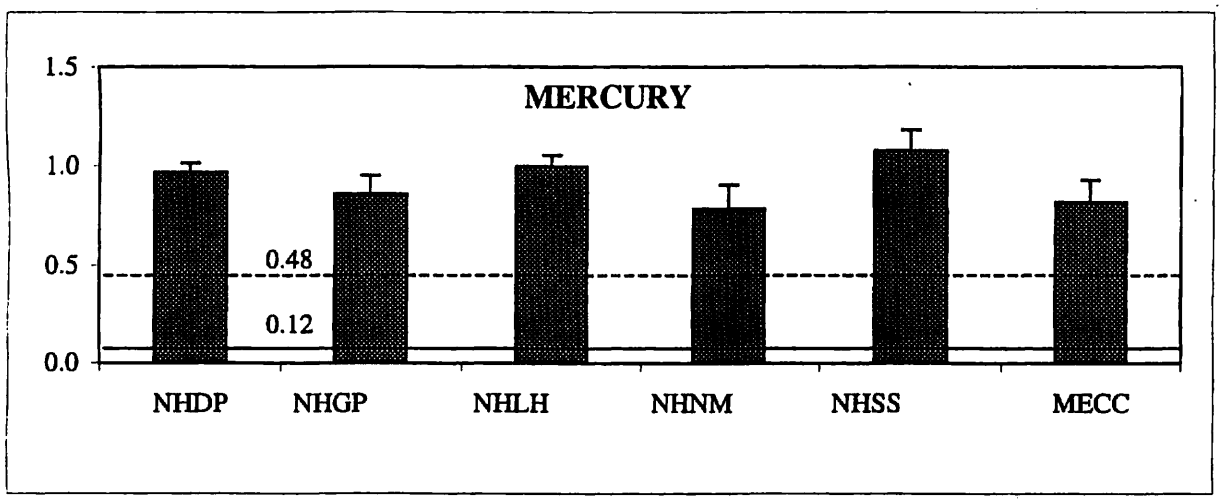


Figure 3. Distribution of zinc, nickel and mercury tissue concentrations (arithmetic mean ± SD, µg/g DW) in mussels at the New Hampshire Gulfwatch stations in 1998. The reference mean, ME-RM (solid line) and the high value, ME-HV (dashed line) from the Maine reference data (Sowles, 1993) are shown for comparison.

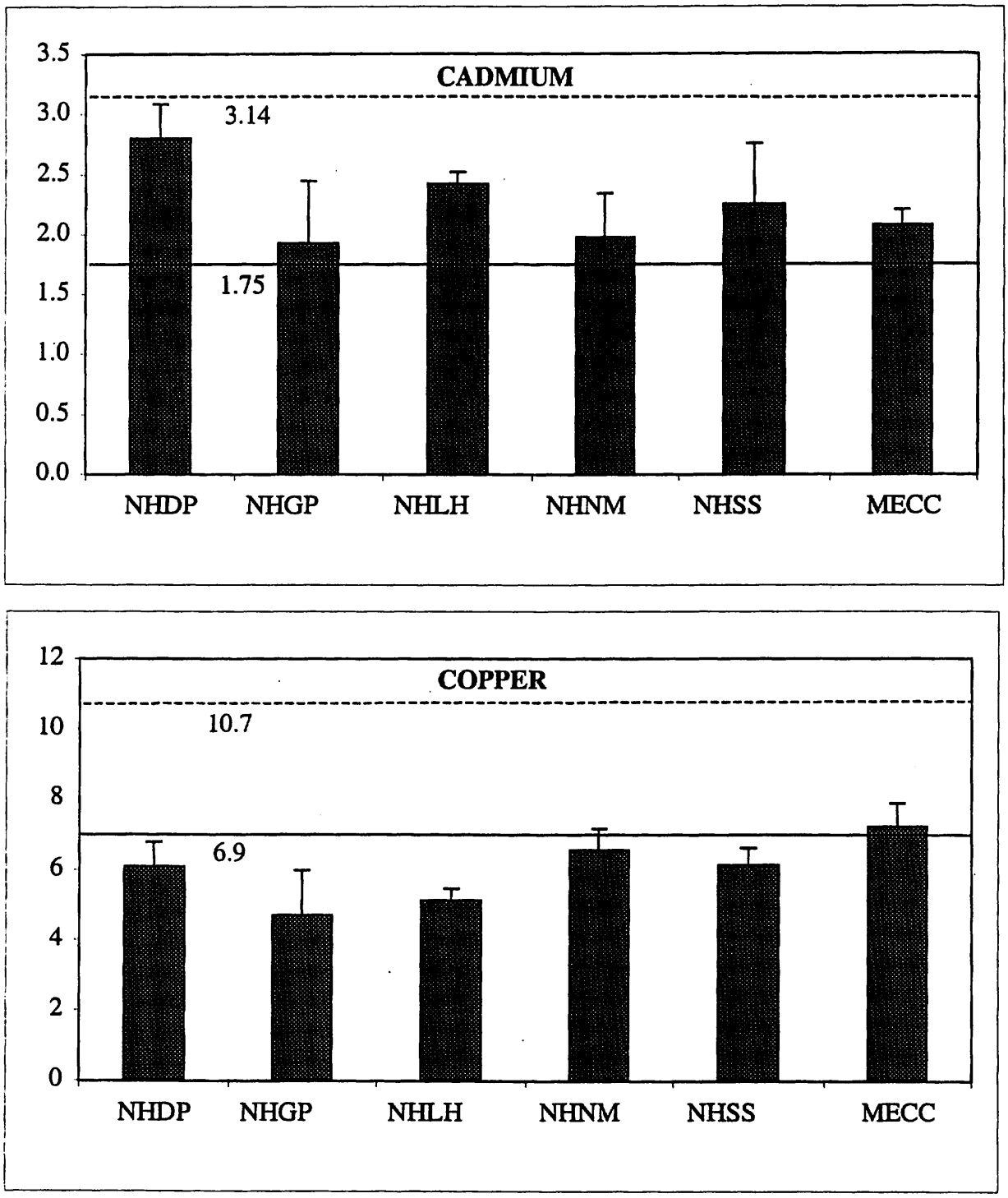
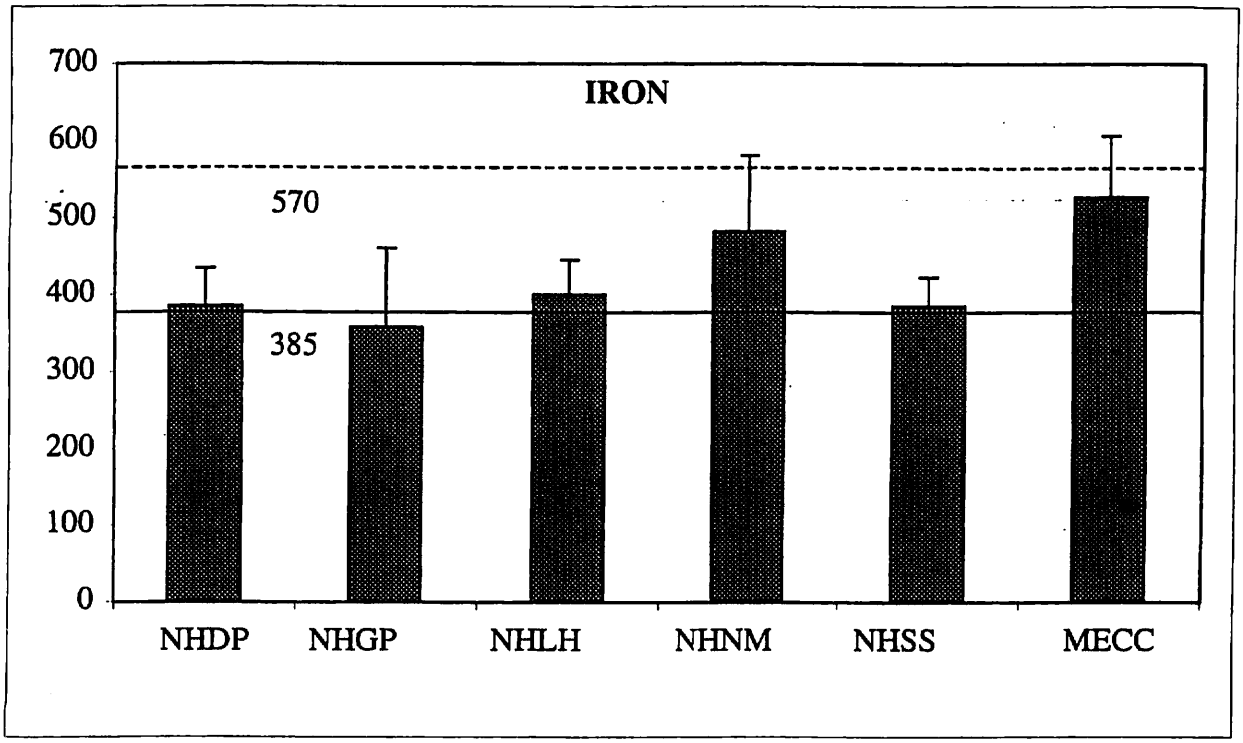


Figure 4. Distribution of cadmium and copper tissue concentrations (arithmetic mean ± SD, µg/g DW) in mussels at the New Hampshire Gulfwatch stations in 1998. The reference mean, ME-RM (solid line) and the high value, ME-HV (dashed line) from the Maine reference data (Sowles, 1993) are shown for comparison.



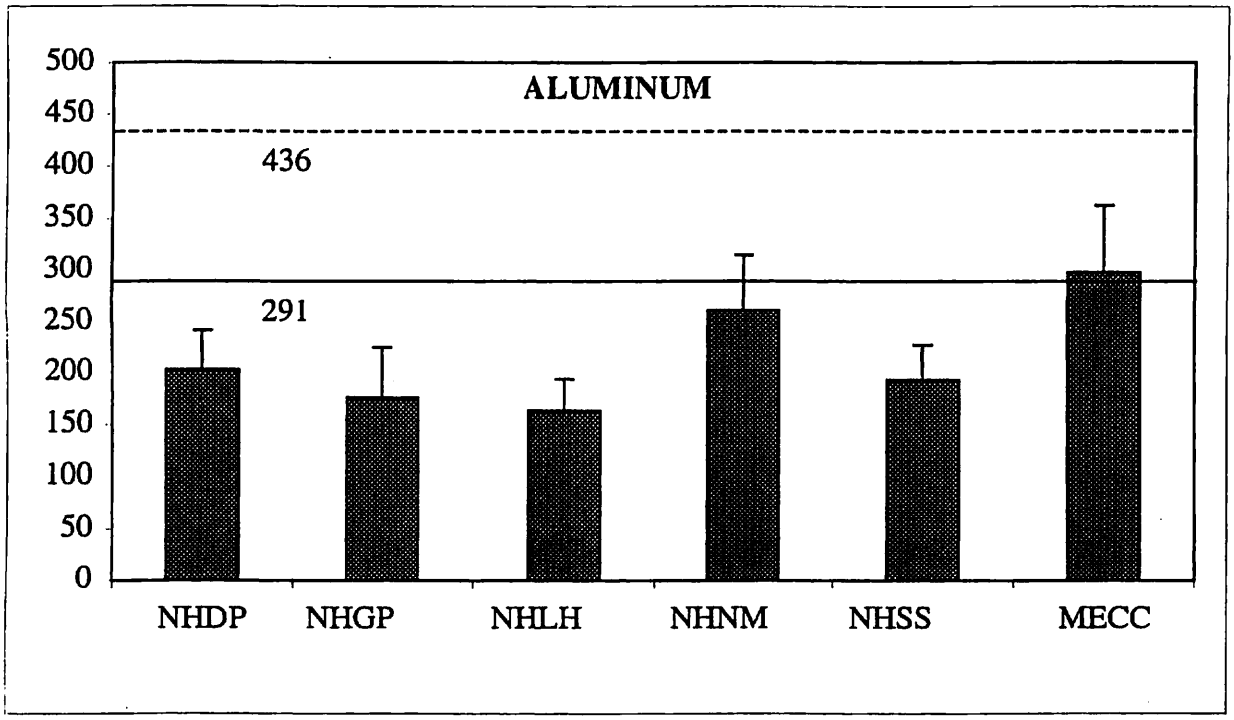


Figure 5. Distribution of iron and aluminum tissue concentrations (arithmetic mean ± SD, µg/g DW) in mussels at the New Hampshire Gulfwatch stations in 1998. The mean (solid line) and the high value (dashed line) from the NS&T data (O'Connor, 1992) are shown for comparison.

Silver (Ag)

Elevated silver exposure concentrations have been shown to coincide with regions receiving municipal sewage (Sanudo-Wlhelmy and Flegal, 1992; Buchholz ten Brink et al., 1996). Despite the presence of numerous municipal sewage sources in the Great Bay Estuary, mussel tissue concentrations of Ag were not detected (ND) at all six 6 sites examined in 1998. The detection limit was 0.1 μg/g dry weight, so all concentrations were below the Maine reference mean of 0.12 μg/g dry weight.

Lead (Pb)

The concentration of lead ranged from a value of 3.02 ± 0.31 μg/g dry weight (NHDP) to 5.75±0.70 μg/g dry weight (MECC) (Table 3, Figure 2). Mean concentrations of Pb in mussels from coastal regions generally range from 1 to 16 μg/g dry weight (Fowler, 1990). All six of the sites sampled in 1998 exceed the Maine reference concentration (ME-RM) of 2.6 ± 1.1 μg/g dry weight, but no site exceeded the ME-HV (6.00 μg/g dry weight). The close proximity to the Portsmouth Naval Shipyard (PNS) may account for elevated lead in the lower Great Bay Estuary, especially at MECC, which is located on Seavey Island. The Jamaica landfill and Defense Reutilization and Marketing Office (DRMO), located on Seavey Island, are sites of known sources of lead contamination to Portsmouth Harbor where waste plating sludge and lead batteries, respectively, were disposed and stored (NCCOSC, 1997). The potential for such sites to be sources of lead to estuarine biota was demonstrated in July, 1999, when significant amounts of contaminated soil, containing as much as 14.2 mg Pb/g soil (dry weight), along the shoreline of Seavey Island at the DRMO was discovered to be eroding into the Piscataqua River (NHDES, unpublished). The shoreline soil has been re-stabilized and the degree of exposure of estuarine biota to lead is being assessed. Results thus far show variable, yet high (199 μg/g DW) levels of lead to be present in some mussels from the shoreline at the DRMO, similar to levels (200-273 μg/g DW) measured in previous studies (NCCOSC, 1997).

Analysis of the concentrations of Pb in mussel tissue from the different sites (Table 3) showed that there were significant differences. Concentrations of Pb at MECC and NHNM were significantly higher than concentrations at NHSS, NHGP and NHDP. The higher Pb concentrations at NHNM may not be related to PNS sources, based on the significantly lower levels found at NHGP just upstream of the mouth of the North Mill Pond. Pb concentrations in the sediments of North Mill Pond are consistently >100 mg/g DW (ANMP, 1998). Sediments with Pb concentrations >100 mg/g DW are relatively common around Seavey Island in Portsmouth Harbor (Buchholtz ten Brink et al, 1997; NCCOSC, 1997), including some with concentrations ~14,000 mg/g DW (NHDES, unpublished). However, sediments elsewhere in the Great Bay Estuary are rarely >100 mgPb/g DW (Buchholtz ten Brink et al, 1997; NCCOSC, 1997). Thus, the elevated Pb concentrations in mussels from the North Mill Pond are probably associated with Pb-contaminated sediments within the pond.

Chromium (Cr)

The concentration of chromium exceeded the ME-RM (1.53 ± 0.66 μg/g dry weight) at all six 1998 sites in New Hampshire, although no site exceeded the ME-HV (3.51 μg/g dry weight). The lowest concentration was at NHGP (2.08 ± 0.56 μg/g dry weight) and the highest at MECC (3.18 ± 0.69 μg/g dry weight) (Table 3, Figure 2). Elevated concentrations at New Hampshire sites probably reflect historical tanning industry discharges (Capuzzo and Anderson, 1973; Jones et al., 1992). Analysis of the concentrations of Cr in mussel tissue from the different sites (Table 3) showed that there were no significant differences between sites. The elevated, yet relatively even distribution of Cr at sites in New Hampshire may reflect a dominance of historical contamination with little present-day source contributions.

Zinc (Zn)

Zinc concentrations generally reflect human activity associated with tire wear, galvanized materials and industrial discharges. All six sites had concentrations greater than the ME-RM (89 ± 16 μg/g dry weight). No site had concentrations greater than the ME-HV of 136 μg/g dry weight (Table 3, Figure 3). The lowest concentration of Zn measured was at NHLH (105±17 μg/g dry weight) and the highest concentrations were at MECC and NHNM (135 μg/g dry weight). Concentrations of zinc in bivalves in British estuaries often exceed 1000 μg/g dry weight, but many may be greater than 4000 μg/g dry weight in contaminated systems (Bryan et al., 1992). Analysis of the mussel tissue concentrations of Zn revealed that there were no significant differences among sites (Table 3).

Nickel (Ni)

The concentration of nickel ranged from 1.24 ± 0.20 μg/g dry weight at NHNM to 2.32 ± 1.08 μg/g dry weight at MECC (Table 3, Figure 3). MECC is the only site that exceeded the ME-RM of 1.8 ± 0.4 μg/g dry weight. Analysis of the mussel tissue concentrations of Ni revealed that there was a significant difference in Ni concentrations between NHNM and MECC (Table 3). There were no significant differences among all other sites.

Mercury (Hg)

The concentration of mercury in mussel tissue ranged from a value of 0.79 ± 0.12 μg/g dry weight at NHNM to 1.08 ± 0.10 μg/g dry weight at NHSS (Table 3, Figure 3). Mercury exceeded both the ME-RM of 0.12 ± 0.12 μg/g dry weight and the ME-HV of 0.48 μg/g dry weight at all sites. There are several known historical mercury sources in the Great Bay Estuary, including some that are suspected to be related to the Portsmouth Naval Shipyard (NCCOSC, 1997) and, especially, the PSNH Schiller Station on the Piscataqua River, where mercury steam was used from 1950 to 1968 (Nelson, 1986). Analysis of the mussel tissue concentrations of Hg revealed that there were significant differences in Hg concentrations between NHSS and all other sites except NHLH, and between NHLH and NHNM (Table 3).

In a review of the first five years of the Gulfwatch program tissue concentrations of Hg were discussed as being unusually high and a possible concern (Jones et al., 1998). Mean values of Hg in mussels (*Mytilus* spp.) from various coastal regions worldwide are about 0.1 to 0.4 μg/g dry weight (Kennish, 1996). Over half of all of the Gulfwatch sites sampled in 1997 exceeded the upper limit of this estimate range. Mytilids from some regions (e.g., northern Mediterranean and southwest Pacific) have Hg concentrations as high as 7.0 μg/g dry weight (Kennish, 1996).

Recent studies have shown that a mercury problem exists in freshwater systems of the northeast U.S. and maritime provinces of Canada (Welch, 1994; DiFranco et al., 1995; and Evers et al., 1996). About 47% of mercury deposition in the region originates from sources within the region, 30% from U.S. sources outside the region, and 23% from the global atmospheric reservoir (NESCAUM, 1998). On June 8, 1998, the New England governors and eastern Canadian premiers agreed to cut regional mercury emissions from power plants, incinerators, and other sources in half by the year 2003 (Boston Globe -6/9/98). However, until recently few coastal systems have been known to be affected by Hg pollution. Atmospheric mercury deposition measurements made at New Castle, NH, at the mouth of Portsmouth Harbor, showed ~ 8 ng/m^2 total mercury was deposited during 1998 (MDN, unpublished). The New Castle site, along with two other Maine coastal sites in Casco Bay and Acadia National Park, showed somewhat elevated total mercury atmospheric deposition compared to nearby, upstream inland sites. Other areas in the Gulf of Maine have elevated (5-20 μg/g) sediment mercury concentrations (Buchholtz ten Brink et al, 1997), including the Penobscot River near Orrington, where permitted and accidental discharges from the Holtra-Chem facility have resulted in sediments having much higher (>100 ppm) Hg concentrations (MEDEP, unpublished). Thus, data on mussel tissue mercury levels are important to help assess current contamination problems and the effects of discharge reduction

efforts in the future.

Cadmium (Cd)

Cadmium is widely used in industry for batteries, plating, stabilizers and as a neutron absorber in nuclear reactors. The concentration of cadmium in mussel tissue ranged from 1.92 ± 0.52 µg/g dry weight at NHGP to 2.80 ± 0.28 µg/g dry weight at NHDP (Table 3, Figure 4). Cd concentrations at all sites exceeded the ME-RM of 1.75 ± 0.46 µg/g dry weight but no values exceeded the ME-HV (3.14 µg/g dry weight). Mean concentrations of cadmium in mussels (*Mytilus* sp.) from several coastal regions world-wide range from approximately 1 to 5 µg/g dry weight (Fowler, 1990). The Cd concentration at NHDP was significantly higher than at NHGP and NHNM (Table 3).

Copper (Cu)

The level of copper in mussel tissue ranged from 4.70 ± 1.27 µg/g dry weight at NHGP to 7.20 ± 0.67 µg/g dry weight at MECC (Table 3, Figure 4). MECC was the only site to exceed the ME-RM (6.9 ± 1.6 µg/g dry weight), but it did not exceed the ME-HV (10.9 µg/g dry weight). Analysis of the mussel tissue level of Cu from the different sites (Table 3) showed that there were significant differences. Concentrations of Cu at MECC were significantly higher than at NHGP and NHLH.

Iron (Fe) and Aluminum (Al)

The concentration of iron in mussel tissue ranged from 358 ± 103 µg/g dry weight at NHGP to 528 ± 80 µg/g dry weight at MECC (Table 3, Figure 5). There were no reference values for Fe from Maine stations with which to compare our data, but comparisons could be made to NS&T values (Figure 6). All sites but NHGP had Fe concentrations greater than the NS&T mean, but no concentrations exceeded the NS&T high value. Analysis of the mussel tissue concentrations of Fe (Table 3) showed that the concentration at MECC was significantly higher than at NHGP.

The concentration of aluminum in mussel tissue ranged from 162 ± 31 µg/g dry weight at NHLH to 298 ± 64 µg/g dry weight at MECC (Table 3, Figure 5). There were no reference values for Al from Maine stations with which to compare data, but comparisons could be made to NS&T values (Figure 5). All sites but MECC had Fe concentrations less than the NS&T mean, and no concentrations exceeded the NS&T high value. Analysis of the level of Al in mussel tissue (Table 3) showed that the concentration at MECC was significantly higher than at NHGP and NHLH.

Comparisons with Previous Studies

Arithmetic mean concentrations for trace metals from New Hampshire sites previously sampled from 1991 to 1997 are presented in Table 4 to provide a basis for determining if concentrations measured at the 1998 sites differ from previously sampled locations. In general, the concentrations measured during 1998 were within the range of concentrations previously observed at other New Hampshire sites. The exception is Hg, where the concentrations at all 1998 sites exceeded the highest average (0.77 µg/g; NHDP 1994, 96, 97) for any previously sampled site.

Table 5 shows the overall average tissue trace metal concentrations from sites in New Hampshire during 1991-98 compared to; 1998 New Hampshire averages; average concentrations in *Mytilus edulis* samples from the NOAA National Status and Trends program (NS&T) Gulf of Maine sites (O'Connor, 1992); Maine reference sites (Sowles, 1993); and the background (lowest) concentrations found in a literature search for all New England and North Atlantic shelf sites (Metcalf and Eddy, 1995). Except for Cu and Al, all geometric means for trace metals were greater in New Hampshire Gulfwatch samples, both from the 1998 sites and from all sites, than in NOAA NS&T samples. Moreover, the level of Hg was much greater than the calculated "high value" (0.31 µg/g; geometric mean plus one standard deviation) for NOAA mussels. Similar results were observed in previous reports (Sowles et al., 1994, 1996; Chase et al., 1996a, b; 1997; 1998).

Table 4. Average mussel tissue trace metal concentrations (µg/g DW) at all Gulfwatch sites (1991-97) in New Hampshire and overall average for 1998 samples.

Location	Dover Point	Clark Cove	Shapleigh I.	Little Harbor	Odiorne Pt.	Rye Harbor	Hampton Hbr.	Overall	Overall
Site name	NHDP	MECC	NHSI	NHLH	NHOP	NHRH	NHHS	average	average
Sample years	1994,96,97	1991-97	1991-92	1991,92,95	1992	1994,97	1993,95,96	1991-97	1998
Ag	0.08	0.13	0.08	0.34	0.12	0.08	0.07	0.14	ND
Al	235	278	370	343	362	153	139	249	206
Cd	2.42	1.79	1.98	2.16	2.15	1.47	1.75	1.92	2.20
Cr	2.81	2.89	6.27	5.28	4.20	1.76	1.68	3.40	2.52
Cu	7.29	8.04	30.50	27.13		6.74	7.63	11.73	5.85
Fe	390	514	632	461	705	296	310	457	413
Hg	0.77	0.68	0.54	0.56	0.60	0.62	0.44	0.61	0.91
Ni	1.52	1.77	3.07	2.99	3.50	1.58	1.26	2.08	1.57
Pb	2.63	5.20	5.32	5.31	4.30	2.20	2.45	4.10	4.00
Zn	128	119	149	214	162	103	127	141	122
% solids	11.8	12.8	10.3	10.3	11.8	13.2	15.0	12.3	

Table 5. Average mussel tissue trace metal concentrations (µg/g DW; average±SD) at New Hampshire Gulfwatch sites (1991-98) and sites in the Gulf of Maine from other studies (*) compared to the USFDA (1993) Guideline levels.

	NH Gulfwatch all sites 1998	NH Gulfwatch all sites 1991-98	Gulf of Maine sites		ME reference sites	USFDA (1993)
			NOAA (1989)	Metcalf and Eddy (1995)	Sowles (1993)	
Ag	**<0.1**	0.14	0.22		0.12	
Al	**206**	244	203			
Cd	**2.20**	1.99	1.10	0.20	1.75	25
Cr	**2.52**	3.34	1.39	0.30	1.53	87
Cu	**5.85**	10.8	10.3	1.40	6.90	
Fe	**413**	454	312			
Hg	**0.91**	0.66	0.13	0.01	0.12	
Ni	**1.57**	2.06	1.18	0.30	1.80	533
Pb	**4.00**	4.16	2.97	0.60	2.60	11.5
Zn	**122**	138	92	3.70	89	

*Gulf of Maine sites from the NOAA/NS&T program (NOAA,1989), background concentrations at New England and North Atlantic shelf sites (Metcalf and Eddy, 1995), and Maine reference sites (Sowles, 1993).

This is surprising given that the NS&T program includes sites in relatively close proximity to known sources of contamination, as is the case for most of the 1998 New Hampshire sites. What is not surprising, because of relatively close proximity to industrial sources, is that all 1998 New Hampshire trace metal concentrations were much greater than the background concentrations from around the Gulf of Maine (Metcalf and Eddy, 1995), as well as being higher (except for Cu) than the Maine reference sites (Sowles, 1993).

Temporal Variation in Metal Concentrations

Sampling has occurred in up to five previous years at three of the sites included in the 1998 New Hampshire program (Table 6). MECC is considered a "benchmark" station in the Gulfwatch program and has been sampled each year since 1993. Both NHLH and NHDP have been sampled in two prior years. The ANOVA comparing metal contaminant concentrations showed significant differences between years at MECC except for Ag and Pb (Table 6). Concentrations of Cd, Cr, Hg, Ni, Zn and Fe in 1998 at MECC were significantly higher or not different than concentrations from previous years. Despite the relatively high concentrations in 1998 for most trace metals, there was no clear trend of increasing concentrations at MECC since 1993.

There were few obvious trends in concentrations of trace metals at the other previously sampled sites. At NHLH, only Hg showed a clear increasing trend since 1992, while Cu, Ni, Zn and Fe showed overall decreases in concentrations (Table 6). The concentrations of Cd, Cr, Pb and Al exhibited mixed trends since 1992. Even fewer trends were apparent at NHDP, where the time period of sampling was only five years (1994-1998). Since 1994, there have been steady, small decreases in Cu and Al concentrations at NHDP, but no clear trends for any of the other trace metals (Table 6).

Organic Contaminants

The total concentration of detectable polynuclear aromatic hydrocarbons (ΣPAH_{24}), polychlorinated biphenyls (ΣPCB_{24}) and organochlorine pesticides ($\Sigma TPEST_{17}$) measured in mussel tissue samples of indigenous mussels are presented in Table 7. In 1998, as in previous reports (Sowles et al., 1994, 1996; Chase et al., 1996a, b, 1997, 1998), ΣDDT_6 and its degenerative metabolites were the main contributors to total detectable pesticides. Because of the dominance of DDT and metabolites as part of the $\Sigma TPEST_{17}$, the concentrations of DDT and metabolites (ΣDDT_6) are presented separately from the total concentration of all other organochlorine pesticides ($\Sigma OPEST_{11}$) in Table 7. Individual analyte concentrations of each compound class are provided in Appendices B, C, and D. Analytes within each category of organic contaminant were detected at each site. There were much wider ranges in concentrations of organic compared to trace metal contaminants. This was in large part a function of the relatively high concentrations of organic contaminants in mussel tissue from NHNM. The widest range was observed for ΣDDT_6 concentrations, with the concentration at NHNM (62±9 ng/g DW) being >5 times higher than the next highest concentration and >12 times higher than the lowest concentration. The elevated concentrations of organic contaminants at NHNM suggests that sources of the compounds may have been or are still present in North Mill Pond. All of the organic contaminants measured are highly persistent in marine envirоonments, thus, it is not possible to state if their presence is associated with recent or historical sources. In contrast, concentrations of organic contaminants at NHLH were relatively low. NHLH had the lowest concentrations of any site for all organic contaminants except $\Sigma OPEST_{11}$. Concentrations of ΣPAH_{24}, ΣPCB_{24}, $\Sigma TPEST_{17}$, $\Sigma OPEST_{11}$ and ΣDDT_6 were relatively similar at the other four sites.

Table 6. Tissue metal concentrations (arithmetic mean ± standard deviation, µg/g dry weight) for Gulfwatch stations sampled in 1998 and in past years: 1993-1998. ANOVA of average concentrations denoted by letters; same letters indicate no significant differences among years.

Year	Ag	Cd	Cr	Cu	Pb	Hg	Ni	Zn	Al	Fe
MECC										
1993	0.10±0.05^{a}	2.38±0.27^{d}	3.31±1.28^{b}	7.51±0.87^{a}	5.35±2.18^{a}	0.74±0.06^{abc}	2.60±0.2^{c}	126±17^{b}	187±81^{a}	535±138^{b}
1994	ND	1.48±0.24^{a}	1.95±0.13^{a}	7.52±1.10^{a}	4.45±0.57^{a}	0.60±0.07^{ab}	1.30±0.28^{a}	96±6^{a}	163±17^{a}	372±56^{a}
1995	0.12±0.05^{a}	1.80±0.08^{bc}	3.32±0.82^{b}	9.92±1.41^{b}	6.05±0.68^{a}	0.56±0.13^{a}	1.65±0.17^{ab}	135±10^{b}	345±26^{b}	535±39^{b}
1996	0.01^{a}	1.72±0.19^{ab}	2.88±0.33^{ab}	8.22±0.61^{a}	5.10±0.48^{a}	0.86±0.31^{c}	1.42±0.13^{a}	112±5^{ab}	335±47^{b}	518±61^{b}
1997	0.02±0.05^{a}	1.55±0.31^{ab}	3.01±0.33^{ab}	7.00±1.18^{a}	5.06±1.07^{a}	0.66±0.06^{abc}	1.87±0.26^{ab}	124±24^{b}	428±57^{c}	611±112^{b}
1998	ND	2.08±0.13^{cd}	3.18±0.69^{b}	7.20±0.67^{a}	5.75±0.70^{a}	0.82±0.11^{bc}	2.32±1.08^{bc}	135±24^{b}	298±64^{b}	528±80^{b}
NHLH										
1992	0.05±0.01^{a}	Not done	4.83±1.82^{b}	Not done	4.88±0.80^{a}	0.55±0.08^{a}	3.22±0.17^{b}	202±52^{b}	287±83^{b}	535±54^{b}
1995	0.05±0.02^{a}	2.23±0.26^{a}	2.70±0.38^{a}	8.75±0.92^{b}	6.50±1.15^{b}	0.69±0.10^{b}	1.73±0.10^{a}	155±17^{ab}	350±56^{b}	510±122^{ab}
1998	ND	2.42±0.10^{a}	2.75±0.97^{a}	5.12±0.33^{a}	4.65±0.37^{a}	1.00±0.05^{c}	1.72±0.67^{a}	105±17^{a}	162±31^{a}	400±45^{a}
NHDP										
1994	0.01±0.00^{a}	3.05±0.33^{b}	3.13±0.22^{b}	7.88±0.88^{b}	3.40±0.28^{b}	0.83±0.03^{b}	1.65±0.19^{b}	145±21^{b}	238±28^{a}	455±58^{b}
1997	0.02±0.04^{a}	1.79±0.10^{a}	2.49±0.34^{a}	6.71±0.42^{a}	1.71±0.30^{a}	0.70±0.07^{a}	1.38±0.10^{a}	109±14^{a}	233±36^{a}	326±32^{a}
1998	ND	2.80±0.28^{b}	2.95±0.06^{b}	6.08±0.69^{a}	3.03±0.31^{b}	0.97±0.05^{c}	1.70±0.20^{b}	130±14^{ab}	202±39^{a}	385±50^{ab}

Table 7. Tissue organic contaminant concentrations (arithmetic mean ± SD, ng/g dry weight) from mussels collected from New Hampshire sites in 1998. ANOVA of concentrations denoted by letters; same letter indicates no significant difference among sites. Geometric mean (±SD) of concentrations for all sites is given below. n=4 replicates per sample

STATION	ΣPAH_{24}	ΣPCB_{24}	$\Sigma PEST_{17}$	ΣDDT_{6}	$\Sigma OPEST_{11}$
NHGP	164 ± 13^{b}	25.5 ± 1.5^{ab}	14.1 ± 1.9^{a}	9.63 ± 1.52^{a}	4.50 ± 0.82^{ab}
NHLH	78 ± 11^{a}	12.5 ± 1.7^{a}	10.2 ± 0.8^{a}	5.01 ± 0.46^{a}	5.16 ± 0.52^{ab}
NHSS	187 ± 46^{b}	30 ± 5^{bc}	14.6 ± 1.9^{a}	9.59 ± 1.25^{a}	5.00 ± 1.07^{ab}
NHDP	238 ± 29^{b}	32 ± 8^{bc}	16.1 ± 2.6^{a}	11.6 ± 2.8^{a}	4.55 ± 0.59^{ab}
NHNM	644 ± 55^{c}	65 ± 9^{d}	67 ± 10^{b}	62 ± 9^{b}	5.80 ± 1.28^{b}
MECC	200 ± 26^{b}	42 ± 7^{c}	15.4 ± 2.3^{a}	11.6 ± 2.0^{a}	3.78 ± 0.30^{a}
Geo. mean	210±2	31±2	18.7±1.9	13.0±2.3	4.7±1.2

Spatial Variation in Organic Concentrations

Figures 6, 7 and 8 show the concentration of ΣPAH_{24} (Figure 6), ΣPCB_{24} (Figure 7), and $\Sigma TPEST_{17}$ (Figure 8) measured in tissue of *M. edulis* in the 1998 New Hampshire sampling stations, presented from north to south, or from up in the Great Bay Estuary to the mouth of Portsmouth Harbor and around to Little Harbor. Concentrations of contaminants were plotted on a log scale and the geometric mean ± 1 SD has been added for comparison purposes. Concentrations above the geometric mean + 1 SD are considered high. Table 7 contains a summary of the geometric means for each jurisdiction, an overall geometric mean value for each organic contaminant at New Hampshire sites and the results of ANOVA of site geometric means.

Polyaromatic Hydrocarbons

Geometric means of the ΣPAH_{24} concentrations ranged from 78 ± 11 ng/g DW at NHLH to 644 ± 55 ng/g dry weight at NHNM. The concentration at NHNM was significantly greater than concentrations at all other sites, and the concentration at NHLH was significantly less than at all other sites (Table 7).

Geometric mean concentrations of ΣPAH_{24} at all but NHLH were as high as those reported from areas influenced by oil spills and municipal sewage outfall (148 ng/g in Rainio et al., 1986; 63-1060 ng/g in Kveseth et al., 1982), but none were as high as in industrialized areas affected by coking operations in Sydney Harbor, NS (1400-16,000 ng/g, in Environment Canada, 1986) or smelting operations in Saudafijord, Norway (5111 - 225,163 ng/g; in Bjorseth et al., 1979).

Figure 6 also shows the geometric mean of ΣPAH_{24} concentrations for all 1998 Gulfwatch sites. The New Hampshire sites exhibit relatively uniform concentrations relative to Gulf-wide geometric mean, with the exception of NHNM. The ΣPAH_{24} concentration at NHNM was the third highest in the 1998 Gulfwatch database, with only MEBB (1123 ng/g DW) and MAIH (3333 ng/g DW) having higher mean concentrations. MAIH is a site in Boston's Inner Harbor and has been subject to high levels of all types of contamination. Relatively uncontaminated mussels deployed in 1995 had ~1570 ng PAH/g DW after 60 days in cages at MAIH (Chase et al., 1996a). MEBB is a site in Boothbay Harbor, which had not been sampled since 1991, when no organic analyses were conducted. However, analysis of tissue samples showed mussels from MEBB to contain elevated levels of trace metals, especially Pb and Zn (Jones et al., 1998).

The possible source of the PAHs in North Mill Pond is not known. In contrast, mussels at NHDP were impacted by the 1996 *Provence* oil spill, yet ΣPAH_{24} concentrations have decreased since 1996 (Chase et al., 1997, 1998; see below) and are much lower than at NHNM in 1998. In addition, NHSS was chosen because of its close proximity to the Schiller Station oil terminal. Despite the potential for elevated PAH concentrations, the ΣPAH_{24} concentration at NHSS was much lower than at NHNM. Examination of the individual PAHs detected at NHNM reveals a marked dominance of higher molecular weight and non-alkylated PAHs (Table 8). This pattern was consistent for all 1998 New Hampshire sites (Appendix B), and suggests that the PAHs at NHNM may be from pyrogenic, as opposed to fresh petroleum, sources. The pattern also strongly suggests that the sources may be historical, or reflect past exposure. Lower molecular weight PAHs degrade faster (Shiaris, 1989) and are more mobile in the environment, and bivalves tend to metabolize and excrete higher molecular weight PAHs at slower rates (Widdows and Donkin, 1992). Sediments from sites in North Mill Pond, especially upstream sites, had ΣPAH_{17} concentrations ranging from <690 to 23,600 ng/g DW (ANMP, 1998). It is possible that PAH contaminated sediments from upstream sources could be taken up and accumulated by mussels at the downstream NHNM site, especially during high flow events at low tide.

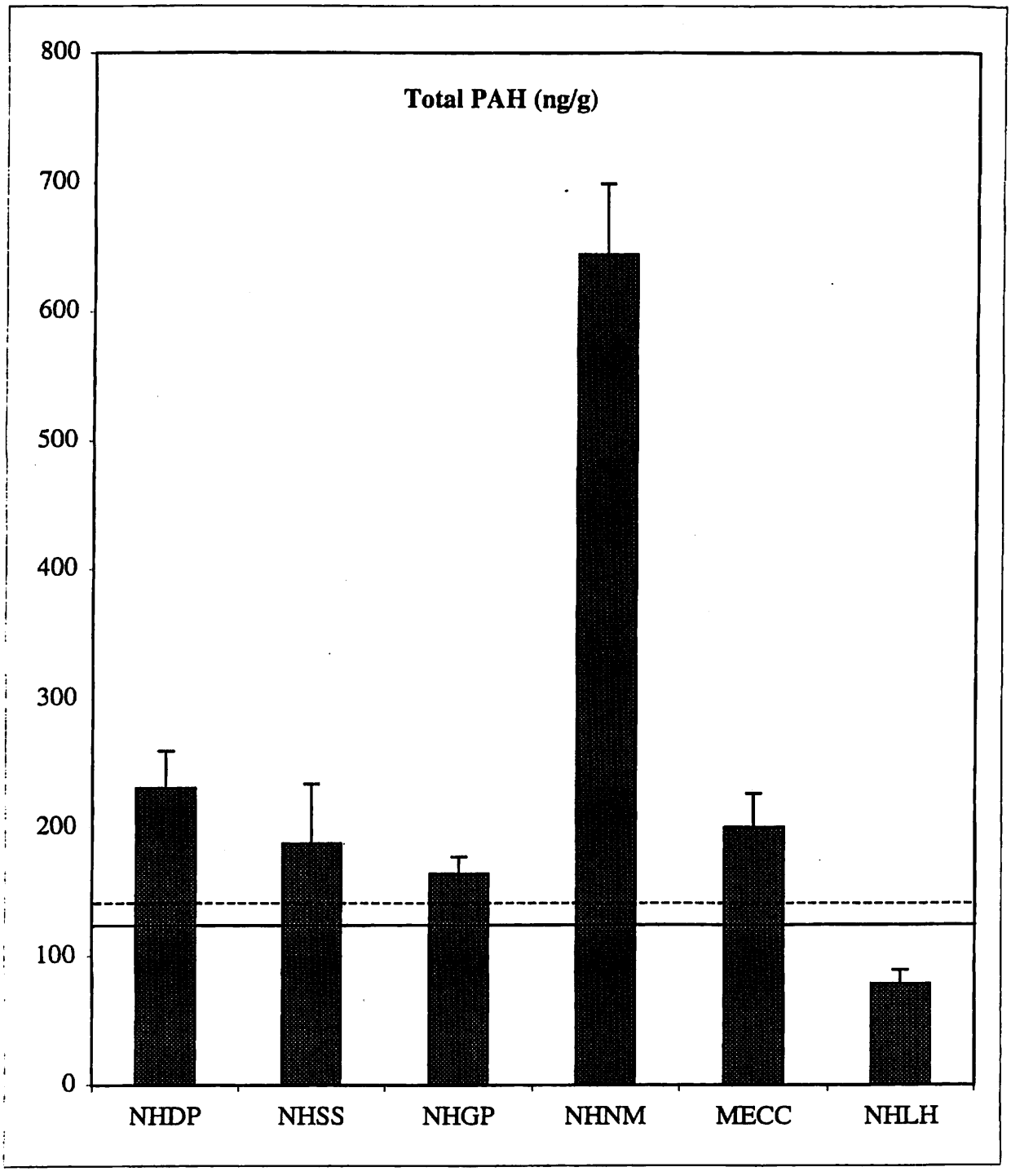


Figure 6. Distribution of total PAH tissue concentrations (arithmetic mean ± SD: ng/g DW) in mussels at New Hampshire Gulfwatch stations, 1998. Geometric mean (solid line) and one standard deviation (dashed line) of all Gulf of Maine stations, 1998.

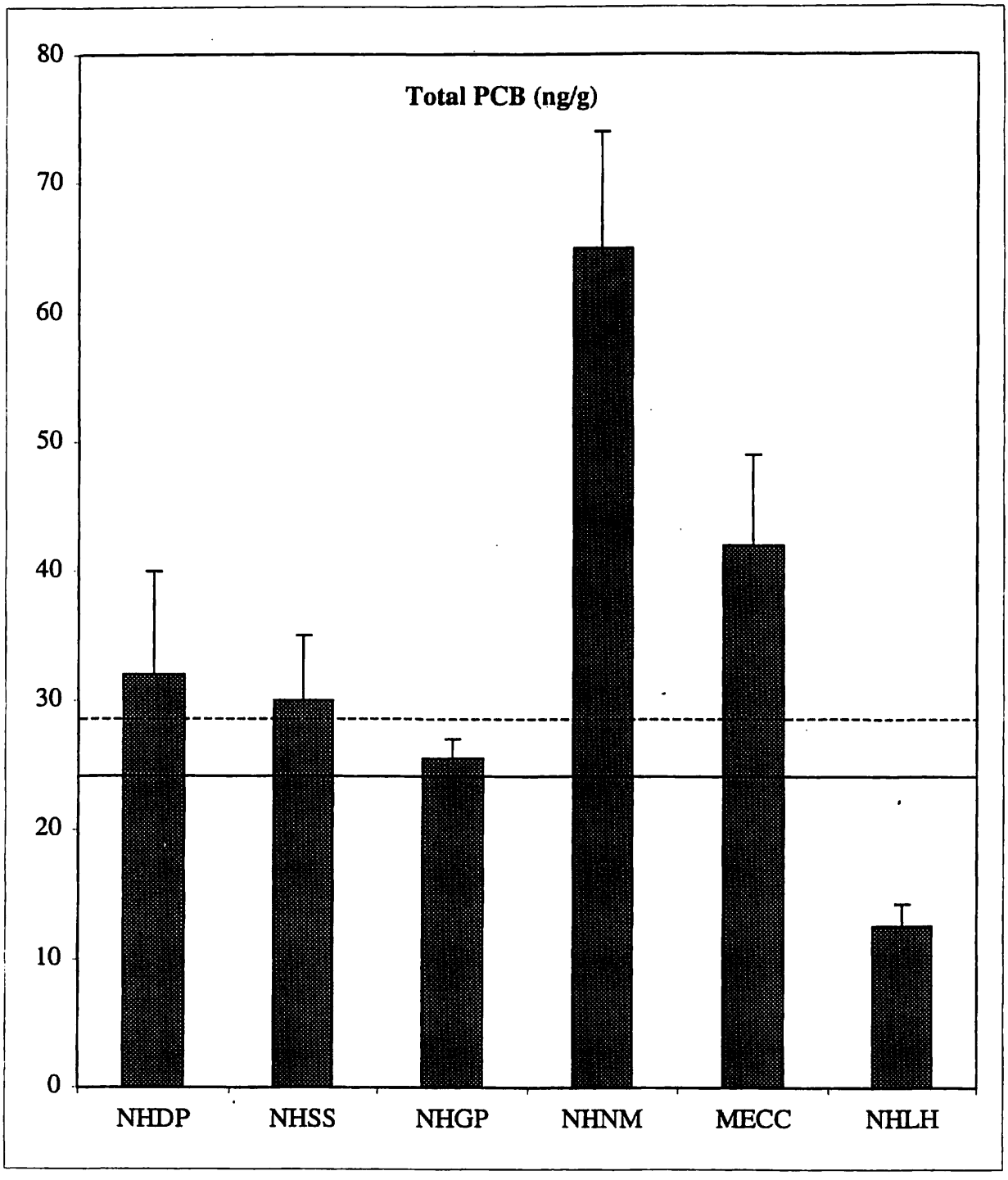


Figure 7. Distribution of total PCB tissue concentrations (arithmetic mean ± SD: ng/g DW) in mussels at New Hampshire Gulfwatch stations, 1998. Geometric mean (solid line) and one standard deviation (dashed line) of all Gulf of Maine stations, 1998.

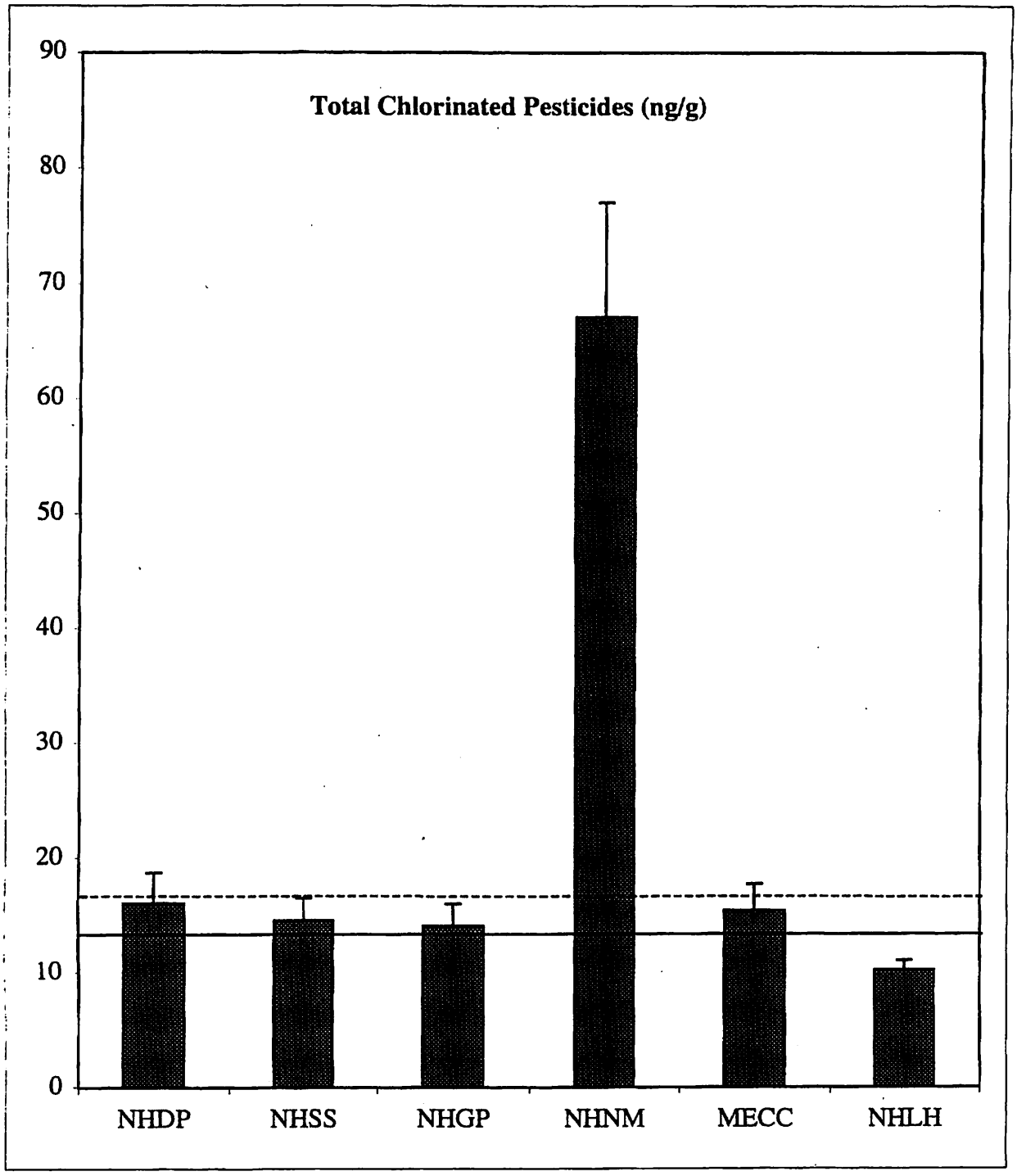


Figure 8. Distribution of total tissue concentrations (arithmetic mean ± SD: ng/g DW) of chlorinated pesticides in mussels at New Hampshire Gulfwatch stations, 1998. Geometric mean (solid line) and one standard deviation (dashed line) of all Gulf of Maine stations, 1998.

Table 8. *Mytilus edulis* tissue (1998) and sediment (1997; ANMP, 1998) concentrations of polyaromatic hydrocarbons (ng/g DW) at North Mill Pond.

PAHs are listed in order of increasing molecular weight.

	Mussel tissue					Sediment
Sample I.D.	**NHNM 1N**	**NHNM 1N duplicate**	**NHNM 2N**	**NHNM 3N**	**NHNM 4N**	**#1 NMP**
Naphthalene	<7	<7	<7	<7	<7	ND*
1-Methylnaphthalene	<8	<8	<8	<8	<8	-
2-Methylnaphthalene	<8	<8	<8	<8	<8	ND
Biphenyl	<6	<6	<6	<6	<6	-
2,6-Dimethylnaphthalene	<8	<8	<8	<8	<8	-
Acenaphthylene	<5	<5	<5	<5	<5	ND
Acenaphthene	<5	<5	<5	<5	<5	ND
2,3,5-Trimethylnaphthalene	<10	<10	<10	<10	<10	-
Fluorene	<6	<6	<6	<6	<6	ND
Phenanthrene	15	14	18	13	11	2700
Anthracene	<6	<6	6	<6	<6	540
1-Methylphenanthracene	<9	<9	<9	<9	<9	-
Fluoranthene	111	105	121	107	99	4600
Pyrene	103	96	113	101	92	4600
Benzo(a)Anthracene	42	39	46	41	36	1600
Chrysene	82	76	88	77	73	1800
Benzo(b)Fluoranthene	80	75	86	86	71	2400
Benzo(k)Fluoranthene	53	49	56	55	41	990
Benzo(e)Pyrene	71	66	79	73	65	-
Benzo(a)Pyrene	27	26	33	28	23	1800
Perylene	26	24	28	29	26	-
Indeno(1,2,3,4-cd)Pyrene	23	20	23	22	20	960
Dibenz(a,h)Anthracene	<4	<4	<4	<4	<4	710
Benzo(ghi)Perylene	25	22	24	24	18	920
Total	**658**	**611**	**721**	**656**	**575**	**23,620**

Site average = 645

*ND=Not detected; below detection limit.

Polychlorinated Biphenyls

The geometric means of ΣPCB_{24} ranged from 12.5 ± 1.7 ng/g DW at NHLH to 65 ± 9 ng/g DW at NHNM (Table 7). The ΣPCB_{24} concentration at NHNM was significantly greater than at all other sites. The concentration at MECC was significantly greater than at NHGP and NHLH, and the concentrations at NHDP and NHSS were significantly greater than at NHLH, which had concentrations significantly lower than at all other sites. The pattern of organic contaminant concentrations at New Hampshire sites was similar for both ΣPAH_{24} and ΣPCB_{24} with the exception of elevated ΣPCB_{24} at MECC and comparatively lower concentrations of ΣPCB_{24} at NHGP.

Figure 7 also shows the geometric mean of ΣPCB_{24} concentrations for all 1998 Gulfwatch sites. Except for NHLH and NHNM, the New Hampshire sites exhibit relatively uniform and somewhat elevated concentrations relative to the Gulf-wide geometric mean. The ΣPCB_{24} concentration at NHNM was the third highest in the 1998 Gulfwatch database, with only MAIH (741 ng/g DW) and MAPR (131 ng/g DW) having higher mean concentrations. This is similar to what was observed for ΣPAH_{24} concentrations, where MAIH also had the highest concentration and NHNM had the third highest concentration. As described previously, MAIH is a site in Boston's Inner Harbor and has been subject to high levels of all types of contamination. Relatively uncontaminated (~37 ng ΣPCB_{24}/g DW) mussels deployed in 1995 had ~361 ng PAH/g DW after 60 days in cages at MAIH (Chase et al., 1996a). MAPR is a site north of Boston Harbor. In 1995, ΣPCB_{24} concentrations for MAPR were the highest (131 ng/g DW) of any other indigenous mussels sampled (Chase et al., 1996a). As for PAHs, the source of the PCBs in North Mill Pond is not known. Analysis of sediments from North Mill Pond conducted on samples collected in 1997 showed no detectable PCBs, although detection limits (>2400 ng/g DW for seven Aroclors) were relatively high (ANMP, 1998). Sites in Portsmouth Harbor have had relatively high sediment PCB concentrations compared to upper estuary sites and other areas, except for Boston Harbor, around the Gulf of Maine (Buchholtz ten Brink et al, 1997).

Organochlorine Pesticides

The geometric means of $\Sigma TPEST_{17}$ ranged from 10.2 ± 0.8 ng/g DW at NHLH to 67 ± 10 ng/g DW at NHNM (Table 7). The $\Sigma TPEST_{17}$ concentration at NHNM was significantly higher than at all other sites; there were no signficant differences for the other five sites. The geometric means of ΣDDT_6 ranged from 5.01 ± 0.46 ng/g DW at NHLH to 62 ± 9 ng/g DW at NHNM (Table 7). As for significant differences among sites, the pattern was similar to that observed for $\Sigma TPEST_{17}$. The geometric means of $\Sigma OPEST_{11}$ only ranged from 3.78 ± 0.30 at MECC to 5.80 ± 1.28 at NHNM. The only significant difference in $\Sigma OPEST_{11}$ concentrations was between NHNM and MECC (Table 7).

Figure 8 also shows the geometric mean of ΣPCB_{24} concentrations for all 1998 Gulfwatch sites. The New Hampshire sites exhibit relatively uniform concentrations except for one site, NHNM, compared to the Gulf-wide geometric mean. The mean $\Sigma TPEST_{17}$ concentration at NHNM was the second highest in the 1998 Gulfwatch database, with only MAIH having a higher mean concentration, although NHNM, MEBB and NBCG all had concentrations between 60 and 70 ng/g DW. This is similar to what was observed for ΣPAH_{24} and ΣPCB_{24} concentrations, where MAIH also had the highest concentration and NHNM had a higher concentration than most other Gulf of Maine sites. Again, the $\Sigma TPEST_{17}$ concentration at MAIH for caged mussels were higher than all other sites in 1995 (Chase et al., 1996). As for PAHs and PCBs, the source of the organochlorine pesticides, particularly DDT and metabolites, in North Mill Pond is not known. Sediment analyses conducted on samples collected from North Mill Pond in 1997 showed no detectable concentrations of a similar set of chlorinated pesticides as used for Gulfwatch mussels (ANMP, 1998).

Comparisons with Previous Studies

Arithmetic mean concentrations for organic contaminants from New Hampshire sites, previously sampled from 1991 to 1997, are presented in Table 9 to provide a basis for determining if concentrations measured at the 1998 sites differ. The PCB concentrations measured during 1998 were within the range of concentrations previously observed at other New Hampshire sites. The average PAH concentration for all 1998 sites was higher than the average for all previous sites. The greatest difference was in chlorinated pesticide concentrations, where the concentration at NHNM far exceeded the highest average for any previously sampled site.

Table 9 also shows the overall average tissue trace metal concentrations from sites in New Hampshire during 1998, compared to the background (lowest) concentrations found in a literature search for all New England and North Atlantic shelf sites (Metcalf and Eddy, 1995). Because of the relatively close proximity to industrial sources, it is not surprising that all 1998 New Hampshire organic contaminant concentrations were much greater than the background concentrations from around the Gulf of Maine (Metcalf and Eddy, 1995).

Chlorobiphenyls and Polychlorinated Dibenzo Dioxins and Dibenzo Furans

Several non-ortho, mono-ortho and di-ortho PCB congeners and planar chlorobiphenyls (CBs) are known to be biologically active and have structural and toxic properties similar to highly toxic 2,3,7,8-terachlorodibenzodioxin (2,3,7,8-TCDD). Mussels from two New Hampshire Gulfwatch sites were analyzed for planar chlorobiphenyls (CBs) in 1998. Planar CB concentrations typically are found in the environment at lower levels than other co-occurring PCB congeners. Planar CB concentrations in mussels, therefore, were measured by GC-high resolution mass spectrometry. The analytical results obtained are generally lower than the method detection limits established for the standard list of Gulfwatch PCB congeners shown in Appendix C using typical NOAA NS&T Mussel Watch methods of clean-up/fractionation and analysis by GC-ECD.

Table 10a contains chlorobiphenyl (CB) concentrations of single composite mussels samples collected from the two New Hampshire Gulfwatch sites in 1998, NHLH and NHNM. The samples are part of a subset of the 1998 Gulfwatch sampling sites that are representative of several major riverine outflow locations in the Gulf of Maine. Concentrations of summed non-ortho, mono-ortho and di-ortho CBs in indigenous mussels for all 1998 Gulfwatch sites ranged from 175 to 34,394 pg/g wet weight (Chase et al., in prep). The highest concentration was measured in mussels at the Boston Inner Harbor site, MAIH, in Massachusetts, which, as described above, also has had the highest or one of the highest concentrations of each category of organic contaminant throughout the Gulfwatch program (Jones, 1998). The lowest concentrations were measured in mussels from two reference sites in Nova Scotia, NSAR and NSSC. Overall, Gulf-wide CB concentrations display a similar pattern of southerly increasing contamination that has been observed for other Gulfwatch organic contaminants in this and in past years, putting some New Hampshire mussels in the higher end of the concentration range characteristic of the southern Gulf (Chase et al., 1998).

In addition to planar CBs, polychlorinated dibenzodioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) were also measured in 1998 New Hampshire mussels. The results of these analyses are given in Table 10c. Concentrations of many of the different PCDD and PCDF in mussels were low and, in many cases, below the limits of detection. In no sample was a detectable concentration of the highly toxic 2,3,7,8-TCDD measured, with concentrations below detection limits for all other dioxin congener chlorinated in the 2,3,7,8 positions, with the exception of the less toxic 1,2,3,4,6,7,8 hepta- and octachloro congeners. Similar results have been observed in previous years for New Hampshire sites (Chase et al., 1997; 1998). On the other hand, low concentrations of 2,3,7,8-terachlorodibenzo(p) furan (2,3,7,8-TCDF) and other chlorinated TCDF congeners were detected in many samples, including both New Hampshire sites. Predominance of PCDF concentrations, particularly 2,3,7,8-TCDF relative to TCDD congener concentrations, can be indicative of pulp mill sources (Rappe et al., 1988) and/or of PCB

Table 9. Average mussel tissue organic contaminant concentrations (ng/g DW) at all Gulfwatch sites (1991-97) in New Hampshire and overall average for 1998 samples.

Location Site name Sample years	*Gulf of Maine Metcalf and Eddy (1995)	Fox Point NHFP 1996	Dover Point NHDP 1994,96,97	Clark Cove MECC 1991-97	Shapleigh I. NHSI 1991-92	Little Harbor NHLH 1991,92,95	Odiorne Pt. NHOP 1992	Rye Harbor NHRH 1994,97	Hampton Hbr. NHHS 1993,95,96	Overall average 1991-97	**NH Gulfwatch all sites 1998**
PCBs	0.01	78	40	49	51	32	32	8.8	9.6	35	**31**
PAHs	0.04	1355	250	160	378	174	129	70	71	163	**210**
Cl'd Pesticides	0.01	6.20	11.5	12.0	17.9	15.1	8.80	7.75	4.15	10.9	**18.7**

*Background concentrations at New England and North Atlantic shelf sites (Metcalf and Eddy, 1995).

Table 10

Table 10a. Non-, Mono-, and Di-ortho chlorobiphenyl concentrations (pg/g wet wt) in mussels at 1998 New Hampshire Gulfwatch sites.
Table 10b. Non-, Mono- and Di-ortho Chlorobiphenyl TEQs in Mussels at 1998 New Hampshire Gulfwatch sites.
Table 10c. Polychlorinated dibenzodioxin and dibenzofuran concentrations (pg/g wet wt) in mussels at 1998 NH Gulfwatch sites

Table 10a

Congener	NHLH	NHNM
	1998	1998
Non-ortho		
PCB-77	7.9	36
PCB-126	1.2	6
PCB-169	0.2	0.76
Mono-ortho		
PCB-105	110	420
PCB-114	10	20
PCB-118	290	1200
PCB-156	40	170
PCB-189	8	10
Di-ortho		
PCB-170	10	120
PCB-180	50	320
Total	**527**	**2303**

Table 10b

Congener	TEF	NHLH	NHNM
		1998	1998
Non-ortho			
PCB-77	0.0005	0.004	0.018
PCB-126	0.1	0.120	0.600
PCB-169	0.01	0.002	0.008
Mono-ortho			
PCB-105	0.0001	0.011	0.042
PCB-114	0.0005	0.005	0.010
PCB-118	0.0001	0.029	0.120
PCB-156	0.0005	0.020	0.085
PCB-189	0.0001	0.001	0.001
Di-ortho			
PCB-170	0.0001	0.001	0.012
PCB-180	0.00001	0.001	0.003
Total (pg/g wet wt)		**0.19**	**0.90**

Table 10c

Dioxins	**NHLH**	**NHNM**
	1998	1998
T4CDD - Total	5.3	12
2,3,7,8	<0.2	<0.2
P5CDD - Total	<0.4	0.6
1,2,3,7,8	<0.4	<0.4
H6CDD - Total	1.7	3.1
1,2,3,4,7,8	<0.6	<0.6
1,2,3,6,7,8	<0.6	<0.6
1,2,3,7,8,9	<0.6	<0.6
H7CDD - Total	5.7	7.3
1,2,3,4,6,7,8	2.4	2.9
O8CDD	15	16
Furans		
T4CDF - Total	1.9	7.1
2,3,7,8	0.5	1.3
P5CDF - Total	<0.4	2.6
1,2,3,7,8	<0.4	<0.4
2,3,4,7,8	<0.4	<0.4
H6CDF - Total	<0.6	0.9
1,2,3,4,7,8	<0.6	<0.6
1,2,3,6,7,8	<0.6	<0.6
2,3,4,6,7,8	<0.6	<0.6
1,2,3,7,8,9	<0.6	<0.6
H7CDF - Total	3.1	0.9
1,2,3,4,6,7,8	1.5	<0.7
1,2,3,4,7,8,9	<0.7	<0.7
O8CDF	1.0	0.8
TEQ -Total*	**0.10**	**0.18**

*TEQ=Toxic Equivalency Concentration. Calculated using WHO international Toxic Equivalency Factors (TEF).

contamination (Hutzinger et al 1974).

Since planar CBs, dioxins and furans share a similar mode of action, and their relative toxicities can be standardized through the use of toxic equivalency factors (TEF), toxic equivalency concentration (TEQs) can be calculated for CBs and for PCDDs/PCDFs in New Hampshire Gulfwatch samples. TEQs for the CB concentrations given in Table 10a are shown in Table 10b. TEQs were calculated using CB concentrations and the WHO interim toxic equivalency factors compiled by Alborg (Alborg at al. 1994). CB-derived TEQs in mussels from the 1998 sites ranged from 0.19 at NHLH to 0.9 pg/g wet weight at NHNM, compared to the Gulfwide concentration range of a high of 7.85 pg/g at Boston Inner Harbor (MAIH) to a low of 0.08 pg/g at NBCH (Chase et al., in preparation). A graphical representation of the CB-derived TEQ distribution in samples collected from New Hampshire Gulfwatch sites in 1996-1998 is shown in Figure 9. The spatial distribution of 1996-1998 New Hampshire Gulfwatch mussel PCDD/PCDF derived TEQs is also presented in Figure 9. PCDD/PCDF derived TEQs were calculated using PCDD/PCDF concentrations (Table 10) and established international toxic equivalency factors (NATO, 1988). PCDD/PCDF derived TEQs ranged from 0.1 at NHLH to 0.18 pg/g at NHNM, compared to the Gulf-wide concentration range that had a high of 1.01 pg/g at Boston Inner Harbor (MAIH) to a no detection at Damariscotta River in Maine (MEDM) (Chase et al., in preparation).

Total TEQs for both CBs and PCDD/PCDF for New Hampshire Gulfwatch samples from 1996 to 1998 are also shown in Figure 9. The range of total TEQs is 0.24 pg/g at Hampton-Seabrook Estuary (NHHS) to 2.2 pg/g at Dover Point (NHDP). The total TEQ for NHLH was the second lowest of the six sites sampled, while the total TEQ for NHNM was relatively high, being lower than only NHDP and NHRH. Interestingly, the greater contribution to total TEQs in mussels from NHNM and NHDP was due to planar CBs, while the greater contribution to total TEQs in samples from NHRH was due to PCDDs/PCDFs.

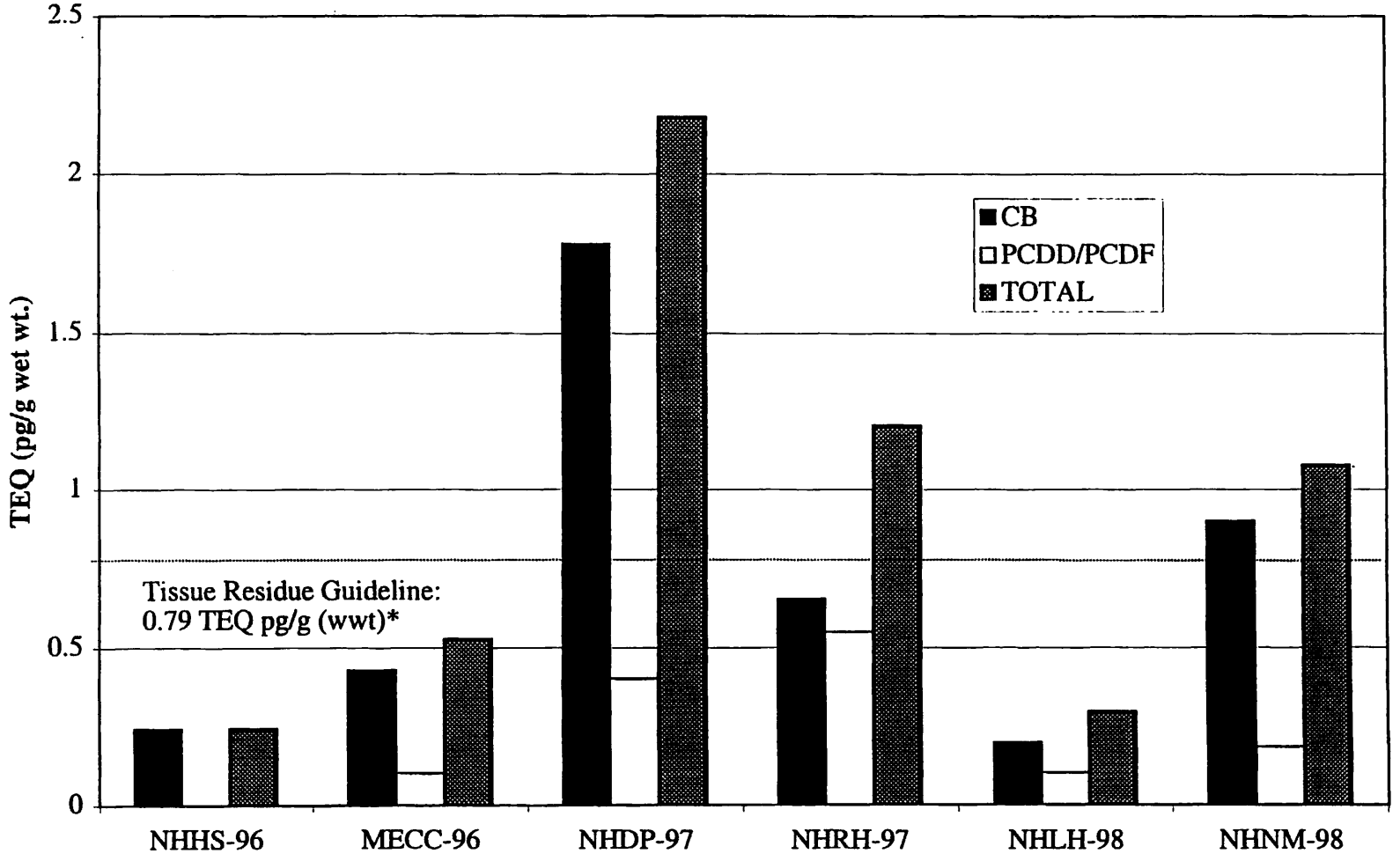
From a human health perspective, total toxic equivalency concentrations for 1998 Gulfwatch samples are well below the 20 pg/g 2,3,7,8-TCDD Canadian tolerance level for the consumption of seafood that is considered protective of human health (Health Canada, 1993). The highest total TEQ measured in all 1998 Gulfwatch mussel was 8.86 pg/g at Boston Inner Harbor (MAIH) (Chase et al., in preparation). A tissue reference concentration of 0.79 pg TEQ/g diet, that is considered protective of sensitive mammalian and avian species, is currently under development (Environment Canada, 1998) . From 1996 to 1998, New Hampshire mussels at MECC, NHDP, NHRH and NHNM exceeded this reference concentration (Figure 9).

Temporal Variation in Organic Concentrations

The ANOVA comparing organic contaminant concentrations at each of the 1998 New Hampshire sites, that have been sampled in previous years (MECC, NHLH, NHDP), showed significant differences between years at all three sites and for all three types of organic contaminants (Table 11). At MECC, the 1998 samples had significantly higher ΣPAH_{24} concentrations than all years except for 1996, significantly higher $\Sigma TPEST_{17}$ concentrations compared to 1996 and significantly lower ΣPCB_{24} concentrations than in 1993 and 1994.

At NHLH, concentrations of ΣPAH_{24}, ΣPCB_{24} and $\Sigma TPEST_{17}$ were all significantly lower than in 1992 but not different than in 1995 (Table 11), suggesting an overall decreasing trend for these contaminants at NHLH. A variety of trends were observed for 1998 mussel samples from NHDP (Table 10). The ΣPAH_{24} concentration was significantly lower than in 1997, but significantly higher than in 1994. The ΣPCB_{24} concentration was signficantly lower in 1998 than in 1997, and the $\Sigma TPEST_{17}$ concentration was significantly higher than in 1994.

Figure 9. Distribution of CB and PCDD/PCDF toxic equivalency concentrations (TEQs) in mussels at New Hampshire sites: 1996-98



* Canadian Tissue Reference Guideline for the protection of wildlife consumers of aquatic biota

Table 11. Tissue organic concentrations (arithmetic mean ± standard deviation, ng/g dry weight) for Gulfwatch stations sampled in 1998 and in past years: 1993-1998. ANOVA of average concentrations denoted by letters; same letters indicate no significant differences among years.

Year	PAH	PCB	Pest
MECC			
1993	154±47^{a}	70±11^{b}	11.1±5.3^{ab}
1994	137±10^{a}	67±5^{b}	12.5±1.3^{b}
1995	158±39^{ab}	35±10^{a}	13.8±1.0^{b}
1996	203±22^{c}	38±2^{a}	7.2±1.5^{a}
1997	147±19^{a}	37±8^{a}	15±5^{b}
1998	200±26^{bc}	42±8^{a}	16±2^{b}
NHLH			
1992	170±21^{b}	49±7^{b}	14.0±2.3^{b}
1995	67±23^{a}	7.8±4.8^{a}	10.4±2.5^{ab}
1998	78±11^{a}	12.5±1.9^{a}	10.0±0.7^{a}
NHDP			
1994	187±29^{a}	32±8^{a}	10.4±2.5^{b}
7/1996	658±77^{c}	66±9.6^{c}	2.3±0.5^{a}
10/1996	298±60^{b}	46±7.8^{b}	4.6±2.0^{a}
1997	266±22^{b}	55±12^{bc}	20±4^{d}
1998	238±17^{ab}	26±6^{a}	16±2^{c}

Effects of an Oil Spill in the Great Bay Estuary

On July 1, 1996, there was an oil spill from the vessel *Provence* into the Piscataqua River. Approximately 1,000 gallons of #6 fuel oil were dispersed with water currents into nearby areas of the Great Bay Estuary. Fuel oils are known to contain a variety of PAHs, especially 2 to 4-ring PAHs, although hundreds of organic compounds, including larger PAHs, are present in all crude oils (Kennish, 1996). The Gulfwatch station NHDP at Dover Point, located at the confluence of the Piscataqua River and Little Bay, approximately 2.5 miles upstream of the oil spill site, was sampled previously in 1994 and again in 1997. However, samples were also collected in response to the oil spill in July and October, 1996, to determine if contaminants from the spill were taken up by mussels, and the degree of elimination of the contaminants over time. The 1994 data serve as useful background information for assessing the degree of residual exposure of the 1996, 1997 and 1998 mussel tissue samples to the oil spill contaminants.

The PAH found in mussel tissue samples collected in 1994, on July 16, 1996 (16d), October 1, 1996 (3 mo.), in 1997 and in 1998 are illustrated in Figure 10 and summarized in Table 12. The PAH concentrations in the 1996/16d samples were much higher than in 1994 and compared to other sites in New Hampshire. Elevated levels of PAHs were also observed in samples of different shellfish (softshell clams, American oysters, blue mussels) collected at sites where the spilled oil collected compared to unimpacted sites (NHDES, unpublished).

The PAH found in mussel tissue samples collected in 1994, on July 16 (16d) and October 1 (3 mo.) of 1996 and in 1997 differed in individual and total PAH concentrations, patterns of PAHs and types of PAHs present. There were 13 different PAHs detected in the 16d samples, 11 in the 3 mo. and 1997 samples, 10 in the 1998 samples and 7 in the 1994 samples. Two low molecular weight (MW) alkylated PAHs, 2,3,5-trimethyl naphthalene and 1-methyl phenanthrene, detected in the 16d samples, were not detected in any later samples or in the 1994 samples. The four PAHs with the highest Mws, detected in 16d and 3 mo. and 1997 samples, were also not detected in the 1994 samples. However, only three of the four higher MW PAHs found in 16d, 3 mo. and 15 mo. samples were still detected in 1998. These patterns suggest that lower MW PAHs and alkylated naphthalenes were less available for uptake after the spill, or that they are eliminated from mussels more readily than the larger PAHs. Elimination rates are slower for higher MW PAHs in mussels (Livingstone and Pipe, 1992). The patterns also suggest that the higher MW PAHs from the spilled oil are more persistent. Weathering of PAHs in other oil spills have shown decreases in naphthalenes and greater stability of higher MW PAHs relative to other PAHs (Boehm et al., 1997; Brown et al., 1997).

All 13 PAHs detected in the 16d samples were present at higher concentrations than in all of the other samples, obviously a reflection of the recent exposure to PAHs from the oil spill. Concentrations of benzo(a)anthracene, benzo(a)pyrene and perylene were slightly (1-3 ng/g) higher in 1998, compared to 1997 samples, while concentrations of phenanthrene, fluoranthrene, pyrene, chrysene, benzo(b+k)fluoranthene and benzo(e)pyrene were present at lower (4-7 ng/g) concentrations than in the 1997 tissue. The average ΣPAH_{24} concentrations were 639, 298, 266, 238 and 187 ng/g DW for the 16d, 3 mo., 1997, 1998 and 1994 samples, respectively (Figure 11). Thus, the total PAH concentration has decreased greatly from 1996 and less drastically from 1997 to 1998. Nearly the same number of PAHs are still detectable, including three of the four higher MW PAHs that were not present before the oil spill. However, even those are now present at relatively low concentrations close to detection limits.

Acceptable Levels and Standards of Mussel Contamination

Limited information is available on observed human health effects of consumption of chemically-contaminated shellfish. While there may be limited epidemiological documented effects, laboratory assays and isolated occurrences of acute human poisonings are responsible for

Figure 10. PAH concentrations in mussel tissue from Dover Point, NH, before (1994), 16 days, and 3, 15 and 27 months after an oil spill on July 1, 1996

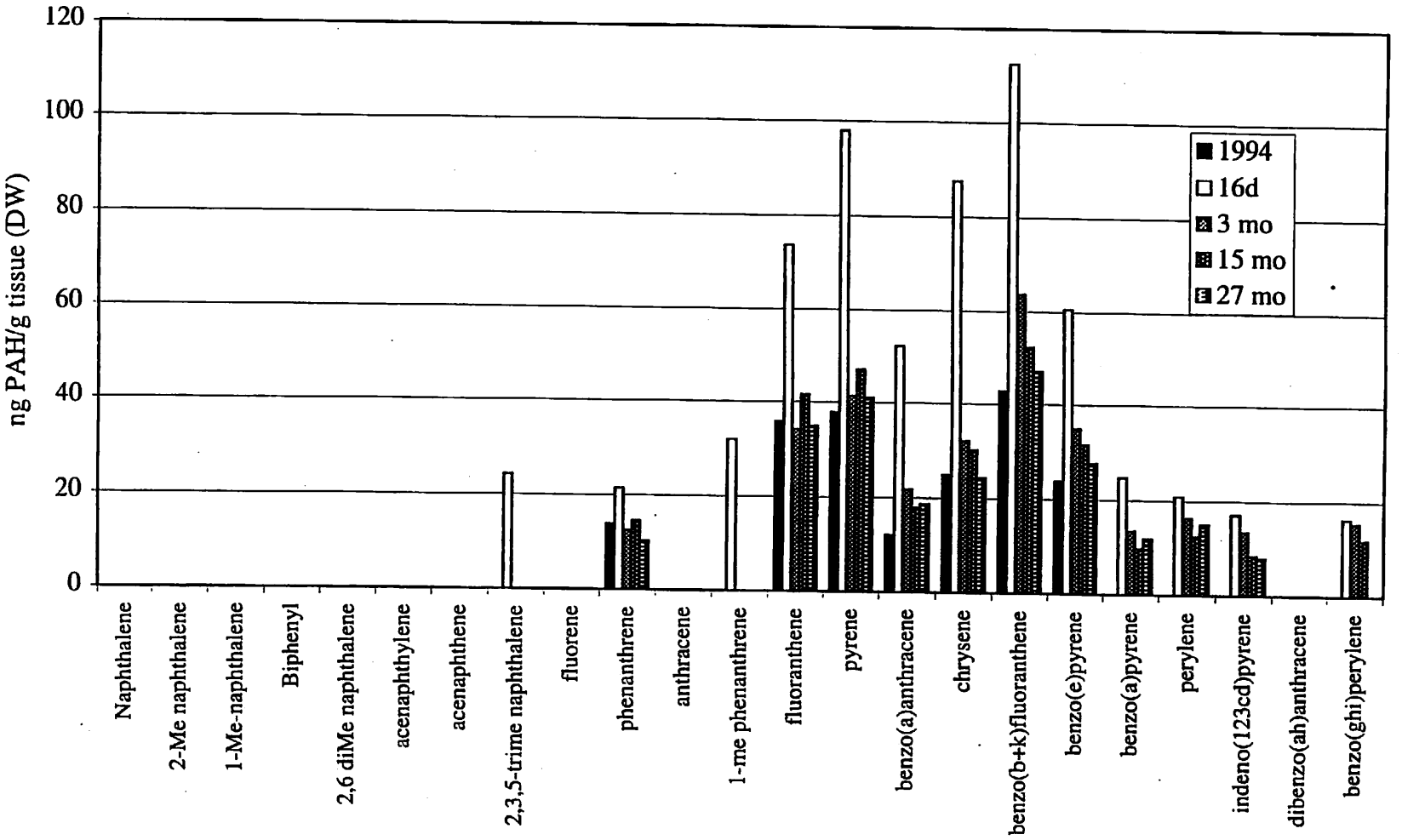
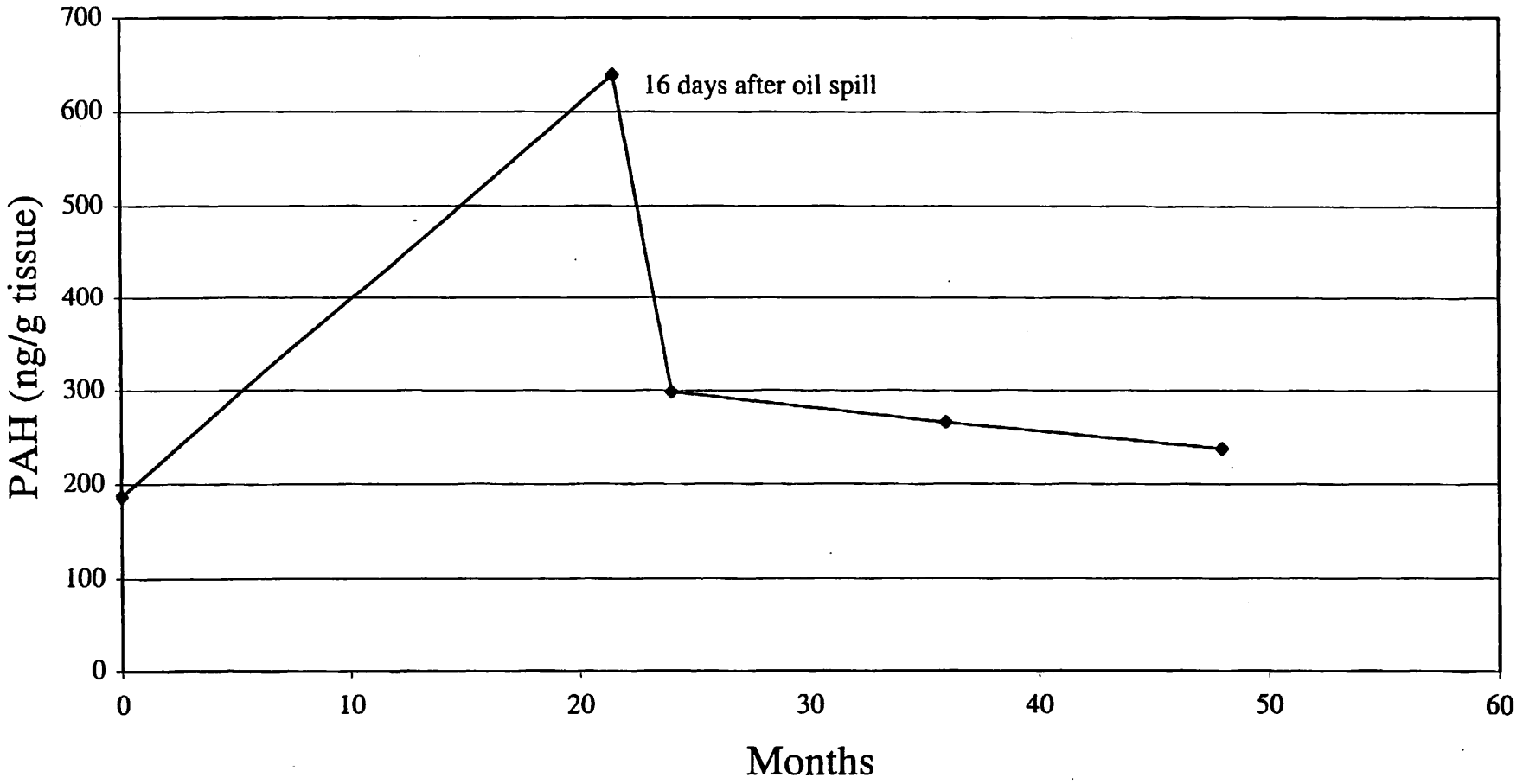


Table 12. Tissue concentrations (ng/g DW) of polyaromatic hydrocarbons in *Mytilus edulis* at Dover Point in Great Bay Estuary in 1994 (NHDP-1994) and 16 days (NHDP-16d), 3 months (NHDP-3 mo.), 15 months (NHDP-15 mo.) and 27 months (NHDP-27 mo.) after a 1996 oil spill.

PAH	NHDP-1994	1996 NHDP-16d	1996 NHDP-3 mo.	1997 NHDP-15 mo.	1998 NHDP-27 mo.	1997-98 change	Detection limit (1998)
naphthalene	<30	<30	<30	<8.2	<7		7
2-Me naphthalene	<30	<30	<30	<9	<8		
1-Me-naphthalene	<30	<30	<30	<8.9	<8		8
biphenyl	<20	<20	<20	<6.1	<6		6
2,6 diMe naphthalene	<20	<20	<20	<8.8	<8		8
acenaphthylene	<10	<10	<10	<5.9	<5		5
acenaphthene	<10	<10	<10	<6.4	<5		5
2,3,5-trime naphthalene	<20	24	<20	<9.8	<10		10
fluorene	<10	<10	<10	<5.1	<6		6
phenanthrene	14	21	13	15	10	-	9
anthracene	<10	<10	<10	<5.1	<6		6
1-me phenanthrene	<10	32	<10	<8	<9		9
fluoranthene	36	74	34	42	35	-	9
pyrene	38	98	41	47	41	-	15
benzo(a)anthracene	12	52	22	18	19	+	9
chrysene	25	88	32	30	24	-	9
benzo(b+k)fluoranthene	43	113	64	52	47	-	12, 7
benzo(e)pyrene	24	60	35	32	28	-	12
benzo(a)pyrene	<10	25	14	10	12	+	8
perylene	<10	21	16	12	15	+	9
indeno(123cd)pyrene	<10	17	14	8	8	=	3
dibenzo(ah)anthracene	<10	<10	<10	<3.6	<4		4
benzo(ghi)perylene	<10	16	15	12	<13		13
TOTAL	**187**	**639**	**298**	**266**	**238**	-28	

Figure 11. Effect of an oil spill (July, 1996) on PAH contamination in blue mussels at Dover Point, NH.



the focus of attention on human health impacts from eating chemically contaminated marine fish and shellfish. In New Hampshire, there are currently human consumption advisories for Hg and PCBs (NHEP, 2000; NHDES, 1998). The advisory for Hg is based on elevated Hg levels in inland lakes and rivers and is for all freshwater fish. For marine waters, there is a consumption advisory for both lobsters and bluefish based on elevated levels of PCBs. The PCB advisories for bluefish and lobsters are based on studies done in 1987 and 1991, respectively.

Published tolerance or action levels for PAHs in commercial marine species are not available in Canada or in the United States. In marine areas where PAH contamination may be a human health concern, closure of commercial fisheries as a result of high contamination levels has been dealt with on a case by case basis. In general, most concentrations reported in the literature are on a wet weight basis in contrast to Gulfwatch dry weight values. To facilitate general comparisons with Gulfwatch values, an average moisture content of 85% has been applied to wet weight health values to derive dry weight equivalents. All reported organic concentrations are within acceptable concentrations for those compounds that have established FDA Action Limits in fish and shellfish. PCB concentrations found in New Hampshire Gulfwatch mussels (Table 10) are less than the action level of 13 ppm dry weight or 2 ppm wet weight (USFDA, 1990; CSSP, 1992), with NHNM having the highest concentrations of PCBs in mussels, 0.19 ± 0.065 ppm dry weight, during the 1998 survey. The action level for the pesticides dieldrin, aldrin, chlordane, heptachlor, and heptachlor epoxide is 2.0 ppm dry weight, or 0.3 ppm wet weight (USFDA, 1990). Only dieldrin and chlordane were detected in the 1998 mussel survey, but at concentrations barely above detection limits which are orders of magnitude below the action levels. The total DDT concentrations found are several orders-of-magnitude below the action level of 33 ppm dry weight or 5 ppm wet weight (USFDA,1990; CSSP, 1992). NHNM had the highest level, 0.062 ± 0.01 ppm dry weight, in 1998. Canadian limits for agricultural chemicals exclusive of DDT are 0.67 ppm dry weight or 0.1 ppm wet weight.

Admissible levels of methyl mercury, expressed as mercury, are less than 6.7 ppm dry weight, or 1 ppm wet weight in the United States (USFDA, 1990), and less than 3.3 ppm dry weight, or 0.5 ppm wet weight in Canada (CSSP, 1992). The highest concentration of mercury found in the 1998 New Hampshire Gulfwatch study was 1.08 ± 0.10 ppm dry weight, at the Schiller Station, New Hampshire, which is high but still well below both federal action concentrations.

A series of FDA "Guidance Documents" (USFDA, 1993) for cadmium, chromium, lead and nickel has been released in the United States to complement the FDA Mercury Action Level. These "alert" levels, however, are guidelines and by themselves do not warrant the issuance of health advisories. In Table 13, guidance concentrations are reported on both wet weight and dry weight bases and are compared to the highest observed concentration in any single replicate analyzed in the 1998 Gulfwatch Program. All nickel, chromium and cadmium concentrations in 1998 New Hampshire Gulfwatch mussels were well below the guideline values. However, Pb concentrations were all >2.5 µg/g DW and the highest concentration was 6.8 µg/g DW at MECC. These concentrations are near to the FDA guideline alert level of 11.5 µg/g DW and are thus of concern.

Table 13. A comparison of United States Food and Drug Administration guidelines for various metals with the Gulfwatch results.

Metal	Guideline (Wet weight)	Guideline (dry weight)	Highest Observed 1998 Gulfwatch value (dry weight)	Location
Cadmium	3.7 µg/g	25 µg/g	3.0 µg/g	Dover Point, NH
Chromium	13 µg/g	87 µg/g	4.2 µg/g	Little Harbor, NH
Lead	1.7 µg/g	11.5 µg/g	6.8 µg/g	Clark Cove, NH/ME
Nickel	80 µg/g	533 µg/g	2.0 µg/g	Dover Point, NH

The U.S. EPA has promulgated a series of "screening values" for three metals (Cd, Hg, Se), 11 organochlorine compounds, one chlorophenoxy herbicide, total PCBs and dioxins/dibenzofurans (EPA, 1993) which were derived using human health risk assessment procedures. The promulgated values are based on several exposure assumptions (70 kg man, an average consumption rate of 6.5 g/day), and either the most current Reference Dose (RfD) values for non-carcinogens or the most recent Slope Factor (SF) plus an acceptable lifetime cancer risk of 1×10^{-5} for the carcinogenic compounds listed. Exceedances of any of the screening values is meant to trigger a more in-depth assessment of actual human health risk. Applying these screening values to the Gulfwatch data provides yet another index of possible human health concern.

Mean concentrations of Cd, Hg and $\sum DDT_6$ at all 1998 Gulfwatch stations are well below the EPA Screening Values (EPA, 1993). The Screening Value for the $\sum PCB_{24}$ is notably low (0.01 µg/g wet weight or approximately 0.07 µg/g dry weight; EPA, 1993). The mean $\sum PCB_{24}$ concentration (0.065 µg/g DW) at NHNM nearly exceeded this value. This station should therefore be examined in more detail to determine potential sources of these contaminants. In the past there has generally been two or fewer Gulfwatch sites each year that have exceeded this screening value.

Morphometric Comparison

Table 14 contains a summary of the morphological measurements [length (mm), height (mm), width (mm), wet weight (g) and condition index (CI)] for indigenous mussels collected at each site.

Shell Morphology

All mussels collected were within the recommended length range of 50 - 60 mm. The mean shell length at the six New Hampshire sites was 54.6 mm (Table 14). There was very little difference in average length for different sites, with a range of 53.9 to 55.6 mm for all mussels collected. For the 40 mussels used to determine condition index, the range in size was 53.4 to 55.5 mm for average mussel length, 21.4 to 22.5 mm for mussel width and 26.7 to 29.6 mm for mussel height. No one site had the smallest or the largest values for the three dimensions.

Condition Index and Wet Weight

Condition indices (CI) of mussels collected in 1998 are shown in Table 14. The average CI for all sites in New Hampshire was 0.145. The CI ranged from a value of 0.123 at NHLH to 0.169 at NHNM. In past years, the New Hampshire CI values have been below the Gulf-wide mean (Chase et al., 1998). The average wet weights (g) of mussels collected in 1998 are shown in Table 14. Comparisons of CI with wet weight and CI with mussel height (Table 14) reveal similar patterns of variation. There was a significant correlation between wet weight and the mussel height for the sites ($p<0.05$).

Water Quality Measurements

As part of the New Hampshire Gulfwatch program, water samples were also collected at the time of sampling at each site to allow for determination of the degree of fecal contamination. Of the four sites where water was analyzed, only North Mill Pond had fecal coliform concentrations that exceeded the State guideline for approved shellfish waters of 14 FC/100 ml (Table 15). The State standard for marine recreational waters of 35 enterococci/100 ml was not exceeded at any site. Comparison of bacterial contaminants in mussel tissue to water samples from the same sites showed a close relationship in concentrations between shellfish and water. These results suggest

Table 14. Average shell dimensions, tissue weight and condition indices for mussel samples from 1998 New Hampshire Gulfwatch sites.

Site name	Location	Sample date	Condition index mg/mm3	Wet weight g	Condition index mussels (n=40) Length mm	Height mm	Width mm	All mussels (n=160) Length mm
MECC	Clark Cove	9/25/98	0.155	5.50	55.4	29.6	21.4	55.0
NHDP	Dover Point	9/28/98	0.138	4.29	55.5	26.7	22.0	53.9
NHGP	Gypsum Plant	9/28/98	0.139	4.82	54.0	28.1	22.1	54.5
NHLH	Little Harbor	9/28/98	0.123	4.50	53.4	27.9	22.5	54.2
NHNM	North Mill Pond	9/28/98	0.169	5.91	54.4	28.7	22.3	54.5
NHSS	Schiller Station	9/25/98	0.143	5.12	55.0	28.7	22.3	55.6
	Average		**0.145**	**5.02**	**54.6**	**28.3**	**22.1**	**54.6**

Table 15. Fecal bacterial concentrations in water and mussel tissue samples from the 1998 New Hampshire Gulfwatch sites.

Site name	Location	Sample date	Water samples		
			Fecal coliforms cfu/100 ml	*E. coli* cfu/100 ml	Enterococci cfu/100 ml
MECC	Clark Cove	9/25/1998	-	-	-
NHSS	PSNH	9/25/1998	-	-	-
NHLH	Little Harbor	9/28/1998	<1	<1	<1
NHNM	North Mill Pond	9/28/1998	24	23	1
NHDP	Dover Point	9/28/1998	13	13	7
NHGP	Gypsum plant	9/28/1998	8	5	1

Site name	Location	Sample date	Mussel tissue	
			Fecal coliforms MPN/100 g	*E. coli* MPN/100 g
MECC	Clark Cove	9/25/1998	400	<200
NHSS	PSNH	9/25/1998	900	210
NHLH	Little Harbor	9/28/1998	80	20
NHNM	North Mill Pond	9/28/1998	900	500
NHDP	Dover Point	9/28/1998	300	110
NHGP	Gypsum plant	9/28/1998	80	40

that NHNM and to a lesser extent the other sites may be exposed to relatively recent pollution associated with fecal contamination, such as sewage and contaminated stormwater pipe effluent.

CONCLUSIONS

The 1998 New Hampshire Gulfwatch program represents the initiation of an expansion of the Gulf of Maine-wide program. The results suggest that re-scaling the program to a more spatially-intensive level was successful for addressing local issue such as assessing impacts of suspected contaminant sources, oil spills and potential chronic exposure to petroleum products. The specific conclusions are listed as follows:

•Comparison of data from different New Hampshire sites showed evidence that suspected sources affected the spatial distributions of some trace metals.

The 1998 New Hampshire results suggest both point and nonpoint sources of contaminants. Mercury concentrations are relatively high throughout the lower Great Bay Estuary, probably as a result of atmospheric deposition. Chromium concentrations are also elevated throughout the study area, with no site having significantly higher levels than other sites. This suggests that chromium from historical sources have become evenly distributed. The results also suggest that the Schiller Station, which had the highest mussel tissue mercury concentrations, is or has been a source for mercury. Mussels from the Gypsum Plant site had relatively low levels of most contaminants despite being located in the midst of all the other sites.

•The spatial distribution of contaminants in New Hampshire mussels showed North Mill Pond to have the highest concentrations of all three classes of organic contaminants.

•The concentrations of organic contaminants at North Mill Pond were much lower than both human health limits and levels observed from Boston's Inner Harbor site.

The sources of organic contaminants in North Mill Pond are not known. It is a concern that levels of PCBs, PAHs and chlorinated pesticides in mussels from this site are higher than other sites in New Hampshire. However, levels are much lower than other sites in the Gulf of Maine, including from Boston's Inner Harbor in 1998. Thus, the Gulf-wide program, conducted using the same procedures as the New Hampshire program, provides a useful comparative reference for interpreting results from local studies.

•Consistent temporal trends for contaminant concentrations are not yet apparent .

•PAH concentrations indicated that no gross contamination is occurring from oil terminal areas. However, levels are elevated compared to much of the rest of the Gulf of Maine.

PAH concentrations may be used as an indication of oil pollution. Levels are elevated in New Hampshire sites compared to areas north and east of New Hampshire. However, levels at the sites in closest proximity to the oil terminals on the Piscataqua River were not higher than levels at other New Hampshire sites. This and the profile of individual PAHs detected throughout New Hampshire suggest that the higher molecular weight PAHs in mussels are either from historical contamination or from pyrogenic sources.

•The impact of the oil spill from the *Provence* in 1996 has been significantly diminished.

Mussel samples have been collected from Dover Point, which was directly affected by the 1996 *Provence* oil spill, each year since the spill. PAH concentrations have steadily decreased since the spill and are nearly back to concentrations detected in 1994, before the spill.

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APPENDIX A. Tissue concentrations of trace metals in *Mytilus edulis* : 1998

[µg/g dry weight; mean and standard deviation (SD)]

New Hampshire

STATION	Ag	Al	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Solid
NHDP 1N	ND 0.1	150	3.00	2.90	5.60	330	1.02	1.60	3.00	130	11.0
NHDP 2N	ND 0.1	200	2.80	3.00	7.10	370	0.92	1.60	3.30	150	11.6
NHDP 3N	ND 0.1	220	3.00	3.00	5.80	390	0.99	2.00	3.20	120	11.5
NHDP 4N	ND 0.1	240	2.40	2.90	5.80	450	0.94	1.60	2.60	120	12.4
Mean	ND 0.1	203	2.80	2.95	6.08	385	0.97	1.70	3.03	130	11.6
SD	-	39	0.28	0.06	0.69	50	0.05	0.20	0.31	14	0.6
NHGP1N	ND 0.1	180	2.30	2.10	5.40	360	0.83	1.50	3.70	120	12.2
NHGP2N	ND 0.1	180	2.30	2.30	5.20	380	0.78	1.50	3.50	130	11.9
NHGP3N	ND 0.1	110	1.20	1.30	2.80	220	0.99	0.99	2.50	74	11.7
NHGP4N	ND 0.1	230	1.90	2.60	5.40	470	0.84	1.40	3.60	120	12.7
Mean	ND 0.1	175	1.93	2.08	4.70	358	0.86	1.35	3.33	111	12.1
SD	-	49	0.52	0.56	1.27	103	0.09	0.24	0.56	25	0.4
NHLH1N	ND 0.1	160	2.50	2.30	5.30	400	1.01	1.70	4.30	110	10.9
NHLH2N	ND 0.1	180	2.50	2.40	4.90	410	1.07	1.80	5.10	110	10.6
NHLH3N	ND 0.1	190	2.40	4.20	4.80	450	0.95	1.90	4.80	120	10.8
NHLH4N	ND 0.1	120	2.30	2.10	5.50	340	0.96	1.50	4.40	80	11.1
Mean	ND 0.1	163	2.43	2.75	5.13	400	1.00	1.73	4.65	105	10.9
SD	-	31	0.10	0.97	0.33	45	0.05	0.17	0.37	17	0.2
NHNM1N	ND 0.1	300	2.40	2.70	7.10	570	0.83	1.40	6.80	160	14.1
NHNM2N	ND 0.1	280	2.00	2.40	6.60	500	0.76	1.30	5.50	140	15.1
NHNM3N	ND 0.1	280	2.00	2.50	6.80	520	0.92	1.30	5.10	130	14.0
NHNM4N	ND 0.1	180	1.50	1.70	5.70	340	0.64	0.94	3.30	110	16.4
Mean	ND 0.1	260	1.98	2.33	6.55	483	0.79	1.24	5.18	135	14.9
SD	-	54	0.37	0.43	0.60	99	0.12	0.20	1.45	21	1.1
NHSS 1N	ND 0.1	240	2.40	2.50	6.60	440	1.08	1.40	3.20	130	12.7
NHSS 2N	ND 0.1	190	1.90	2.20	6.00	380	0.98	1.30	2.70	140	13.9
NHSS 3N	ND 0.1	180	1.80	2.10	5.50	360	1.03	1.30	2.90	120	13.6
NHSS 4N	ND 0.1	160	2.90	2.40	6.40	360	1.22	1.80	3.80	120	11.5
Mean	ND 0.1	193	2.25	2.30	6.13	385	1.08	1.45	3.15	128	12.9
SD	-	34	0.51	0.18	0.49	38	0.10	0.24	0.48	10	1.1
MECC 4N	ND 0.1	280	2.20	3.00	7.00	540	0.82	1.80	6.60	140	11.6
MECC 3N	ND 0.1	280	2.10	2.80	8.10	500	0.97	3.90	5.70	150	11.6
MECC 2N	ND 0.1	240	2.10	2.70	7.20	440	0.77	1.50	4.90	100	12.9
MECC 1N	ND 0.1	390	1.90	4.20	6.50	630	0.71	2.10	5.80	150	12.4
Mean	ND 0.1	298	2.08	3.18	7.20	528	0.82	2.33	5.75	135	12.1
SD	-	64	0.13	0.69	0.67	80	0.11	1.08	0.70	24	0.6

APPENDIX B. Tissue concentrations (ng/g DW) of polyaromatic hydrocarbons in *Mytilus edulis* : 1998.

PAH	NHGP1N	NHGP2N	NHGP3N	NHGP4N	NHLH1N	NHLH2N	NHLH3N	NHLH4N	NHSS1N	NHSS2N	NHSS3N	NHSS4N	NHSS4N
Naphthalene	<7	<7	<7	<7	8.8	7.1	<7	<7	<7	<7	<7	<7	<7
1-Methylnaphthalene	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
2-Methylnaphthalene	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Biphenyl	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6
2,6-Dimethylnaphthalene	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Acenaphthylene	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
Acenaphthene	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
2,3,5-Trimethylnaphthalene	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Fluorene	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6
Phenanthrene	9.7	<9	9.6	<9	<9	<9	<9	<9	9.2	<9	<9	13	<9
Anthracene	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6
1-Methylphenanthracene	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Fluoranthene	37	34	34	33	26	22	26	25	48	38	36	38	31
Pyrene	36	30	34	31	21	19	21	20	48	36	37	36	29
Benzo(a)Anthracene	11	11	11	11	<9	<9	<9	<9	20	11	12	12	10
Chrysene	22	20	20	19	15	13	14	14	29	19	21	22	19
Benzo(b)Fluoranthene	14	16	14	16	<12	<12	<12	<12	27	13	16	15	13
Benzo(k)Fluoranthene	16	14	14	14	11	8.4	9.5	10	24	13	16	16	14
Benzo(e)Pyrene	21	20	18	21	12	<12	<12	12	30	20	22	20	18
Benzo(a)Pyrene	9.3	9.5	9.7	9.7	<8	<8	<8	<8	13	8.2	9.0	9.1	7.9
Perylene	<9	<9	<9	<9	<9	<9	<9	<9	14	<9	9.6	9.0	<9
Indeno(1,2,3,4-cd)Pyrene	6.1	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3
Dibenz(a,h)Anthracene	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4
Benzo(ghi)Perylene	<13	<13	<13	<13	<13	<13	<13	<13	<13	<13	<13	<13	<13
Total	**182**	**155**	**165**	**156**	**93**	**69**	**70**	**82**	**263**	**160**	**178**	**190**	**142**
Surrogate Recovery %													
Naphthalene-d8	86	76	86	77	77	92	83	80	76	93	82	72	84
Acenaphthene-d10	105	80	88	78	76	89	80	77	73	89	80	75	83
Phenanthrene-d10	117	101	105	103	95	104	95	94	92	103	93	96	103
Fluoranthene-d10	132	114	113	116	110	115	113	115	114	116	108	112	117
Chrysene-d12	128	116	110	116	110	116	116	123	121	113	110	113	113
Benzo(a)pyrene-d12	118	100	91	102	95	90	96	112	102	92	85	89	89
Benzo(g,h,i)perylene-d12	104	121	110	123	115	112	116	133	124	111	116	117	115

APPENDIX B. Tissue concentrations (ng/g DW) of polyaromatic hydrocarbons in *Mytilus edulis* : 1998.

PAH	NHDP1N	NHDP2N	NHDP3N	NHDP4N	NHNM1N	NHNM 1N	NHNM2N	NHNM3N	NHNM4N	MECC1N	MECC2N	MECC3N	MECC4N
Naphthalene	<7	<7	<7	<7	<7	<7	<7	<7	<7	<7	<7	<7	<7
1-Methylnaphthalene	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
2-Methylnaphthalene	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Biphenyl	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6
2,6-Dimethylnaphthalene	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8	<8
Acenaphthylene	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
Acenaphthene	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5
2,3,5-Trimethylnaphthalene	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Fluorene	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6	<6
Phenanthrene	10	<9	<9	<9	15	14	18	13	11	<9	12	10	9.4
Anthracene	<6	<6	<6	<6	<6	<6	6	<6	<6	<6	<6	<6	<6
1-Methylphenanthracene	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9	<9
Fluoranthene	30	35	34	41	111	105	121	107	99	37	49	36	40
Pyrene	35	42	40	47	103	96	113	101	92	34	44	32	37
Benzo(a)Anthracene	16	17	18	24	42	39	46	41	36	11	12	12	12
Chrysene	21	24	23	29	82	76	88	77	73	22	28	21	25
Benzo(b)Fluoranthene	20	22	26	36	80	75	86	86	71	10	17	13	16
Benzo(k)Fluoranthene	19	19	21	25	53	49	56	55	41	18	19	13	17
Benzo(e)Pyrene	24	25	29	33	71	66	79	73	65	20	24	16	21
Benzo(a)Pyrene	11	10	12	14	27	26	33	28	23	9.0	9.2	9.3	9.2
Perylene	14	15	14	16	26	24	28	29	26	9.5	11	11	10
Indeno(1,2,3,4-cd)Pyrene	7	8	9	8	23	20	23	22	20	6.1	7.1	7.9	7.1
Dibenz(a,h)Anthracene	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4	<4
Benzo(ghi)Perylene	<13	<13	<13	<13	25	22	24	24	18	<13	<13	<13	<13
Total	**207**	**217**	**226**	**272**	**658**	**611**	**721**	**656**	**575**	**176**	**233**	**183**	**206**
Surrogate Recovery %													
Naphthalene-d8	90	74	64	79	72	86	89	78	54	50	64	79	52
Acenaphthene-d10	95	84	73	80	80	90	95	91	64	72	87	95	68
Phenanthrene-d10	103	99	91	90	94	101	103	100	78	85	100	104	85
Fluoranthene-d10	121	119	109	109	119	121	118	116	99	105	112	112	110
Chrysene-d12	113	106	99	104	118	120	115	111	91	106	104	101	111
Benzo(a)pyrene-d12	99	87	93	99	112	117	113	107	99	90	90	101	103
Benzo(g,h,i)perylene-d12	101	105	110	100	119	114	108	110	101	84	87	82	90

APPENDIX C. Tissue concentrations of polychlorinated biphenyls in *Mytilus edulis* : 1998.

(ng/g dry weight)

PCB Congener	NHLH1N	NHLH2N	NHLH3N	NHLH4N	NHSS1N	NHSS2N	NHSS3N	NHSS4N	NHSS4N duplicate	NHGP1N	NHGP2N	NHGP3N	NHGP4N
8;5	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
18;15	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
29	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
28	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
50	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
52	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
44	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
66;95	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
101;90	1.5	1.6	<1.5	<1.5	4.8	3.6	4.6	3.7	3.3	3.6	3.2	3.6	3.3
87	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
77	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
118	2	2.1	1.7	1.9	5.3	4	5	4.1	3.8	4	3.8	4	3.7
153;132	4.7	5	4	4.6	11	8.6	9.4	8.3	7.8	7.9	7.3	8.1	7.4
105	<1	<1	<1	<1	1.4	<1	1.3	<1	<1	<1	<1	<1	<1
138	3.8	3.9	3.3	3.6	8.8	6.7	7.9	6.5	6.1	6.5	6.2	6.7	6.1
126	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
187	1.6	1.6	1.4	1.6	3.7	2.9	3	2.7	2.5	2.6	2.4	2.6	2.5
128	<1	<1	<1	<1	1.8	1.4	1.6	1.4	1.4	1.4	1.4	1.5	1.3
180	<1	<1	<1	<1	1.1	<1	1.1	1	<1	<1	<1	1	<1
169	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
170;190	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
195;208	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
206	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
209	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
Total	14	14	10	12	38	27	34	28	25	26	24	28	24
Surrogate Recovery %													
103	88	93	82	87	83	87	82	86	91	84	84	78	82
198	76	80	72	79	82	81	81	82	84	84	84	82	83

APPENDIX C. Tissue concentrations of polychlorinated biphenyls in *Mytilus edulis* : 1998.

(ng/g dry weight)

PCB Congener	NHDP1N	NHDP2N	NHDP3N	NHDP4N	NHNM1N	NHNM1N duplicate	NHNM2N	NHNM3N	NHNM4N	MECC1N	MECC2N	MECC3N	MECC4N
8;5	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
18;15	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
29	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
28	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
50	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
52	<2	<2	<2	<2	2.1	2	2.8	2.7	3.6	<2	<2	<2	<2
44	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
66;95	<2	<2	<2	<2	<2	<2	2.4	2.2	3.1	<2	<2	<2	<2
101;90	2.9	4.6	4.1	5.6	6.9	6.6	9.1	8.9	9.5	4.8	4.8	4.4	5.6
87	<1.5	<1.5	<1.5	<1.5	2.1	2.1	2.8	2.7	2.8	<1.5	<1.5	<1.5	2.1
77	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
118	3	5	4.3	5.7	7.1	7.4	9.4	9.2	9.5	5.1	5.0	4.6	6.2
153;132	7.1	11	9.6	13	14	14	17	18	17	13	12	10	14
105	<1	1.5	1.3	1.6	2.3	2.3	3.3	3	3.5	2.4	1.7	1.6	4.7
138	5.9	8.4	7.4	11	12	13	15	15	15	11	10	8	12
126	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
187	2.6	3.2	3	4	4.7	4.6	5.3	5.4	5.1	4.9	4.6	3.9	5.2
128	<1	<1	<1	1	1.8	1.8	2	2.1	2.1	1.1	<1	<1	1.1
180	<1	<1	<1	<1	2	1.9	2.1	2.1	2.1	2.0	1.7	1.5	1.7
169	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
170;190	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
195;208	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
206	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
209	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
Total	22	34	30	42	55	56	71	71	73	44	40	34	52
Surrogate Recovery %													
103	99	101	89	104	99	89	86	80	OT ADDE	117	100	114	103
198	91	104	95	110	109	86	87	80	OT ADDE	112	97	96	111

APPENDIX D. Tissue concentrations of chlorinated pesticides in *Mytilus edulis* : 1998.

(ng/g dry weight)

Pesticide	NHLH1N	NHLH2N	NHLH3N	NHLH4N	NHSS1N	NHSS2N	NHSS3N	NHSS4N	NHSS4N duplicate	NHGP1N	NHGP2N	NHGP3N	NHGP4N
HCB	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2
g-HCH	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2
Heptachlor	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Aldrin	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
Hepta Epoxide	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2
cis-Chlordane	2.12	2.48	2.1	2.11	2.84	2.05	2.36	2.16	2.1	2.16	2.18	2.52	2.15
trans-Nonachlor	1.1	1.25	1.14	1.34	1.62	1.15	1.35	1.27	1.28	1.27	1.17	1.38	1.25
a-endosulfan	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2
o,p'-DDE	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2
p,p'-DDE	3.12	3.38	2.73	3.14	5.82	5.23	6.16	5.51	6.26	5.33	4.95	5.5	5.15
Dieldrin	1.4	2.1	1.7	1.8	1.5	1.3	2.1	1.9	<1.2	1.2	1.2	1.5	<1.2
o,p'-DDD	<1	<1	<1	<1	1.38	1.03	1.5	<1	1.16	2.72	<1	1.4	1.24
o,p'-DDT	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2
p,p'-DDD	2.07	2.04	1.63	1.92	3.67	2.61	2.91	2.31	2.38	3.28	2.71	3.15	3.1
b-Endosulfan	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
p,p'-DDT	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Mirex	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
Total	9.8	11	9.3	10	17	13	16	13	13	16	12	15	13
Surrogate Recovery %													
g-Chlordene	102	93	80	90	90	93	83	83	87	110	104	109	115

APPENDIX D. Tissue concentrations of chlorinated pesticides in *Mytilus edulis* : 1998.

(ng/g dry weight)

Pesticide	NHDP1N	NHDP2N	NHDP3N	NHDP4N	NHNM1N	NHNM1N duplicate	NHNM2N	NHNM3N	NHNM4N	MECC1N	MECC2N	MECC3N	MECC4N
HCB	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2
g-HCH	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2
Heptachlor	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Aldrin	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
Hepta Epoxide	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2
cis-Chlordane	2	2.1	1	1.2	2.7	1.7	2.7	2.4	3.1	2.3	2.4	2.1	2.6
trans-Nonachlor	1.4	1.5	1.5	1.6	2.3	2	2.3	2.4	2.4	1.4	1.5	1.3	1.5
a-endosulfan	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2
o,p'-DDE	<1.2	<1.2	<1.2	<1.2	1.8	1.9	2.1	2.3	2.8	<1.2	<1.2	<1.2	<1.2
p,p'-DDE	5.2	7.1	6.3	7.7	14	13	17	17	18	5.6	5.7	5.2	6.6
Dieldrin	1.5	1.6	1.5	1.3	1.9	<1.2	1.6	1.5	<1.2	<1.2	<1.2	<1.2	<1.2
o,p'-DDD	<1	<1	1	1.4	10	8.2	12	11	8.3	<1	1.1	1.2	1.1
o,p'-DDT	1.31	1.56	1.3	1.9	2.6	2.4	2.7	2.9	2.9	<1.2	1.4	<1.2	1.3
p,p'-DDD	<1.5	1.8	2.2	2.1	25	20	31	29	36	4.2	4.3	3.7	5
b-Endosulfan	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
p,p'-DDT	1.3	1.3	1.3	1.6	3.6	2.8	2.7	3	1.9	<1	<1	<1	<1
Mirex	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5	<1.5
Total	13	17	16	19	64	52	74	72	75	14	16	14	18
Surrogate Recovery %													
g-Chlordene	99	115	87	97	126	104	not added	81	120	118	121	109	123