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National distribution of chemical concentrations in mussels and oysters in the USA

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Abstract

Since 1986 the NOAA National Status and Trends (NS&T) Program Mussel Watch has monitored concentrations of trace chemicals in the coastal United States by sampling mussels, oysters, and sediment. The sediment data have been used to define the status or geographic distribution of chemical concentrations (Daskalakis, K. D., & O'Connor, T. P (1995). Distribution of chemical contamination in coastal and estuarine sediments. *Marine Environmental Research 40*, 381–398) and the molluscan data have provided an estimate of temporal trends (O'Connor, T. P. (1996). Trends in chemical concentrations in mussels and oysters collected along the US coast from 1986 to 1993. *Marine Environmental Research 41*,183-200, O'Connor, T. P. (1998). Mussel Watch results from 1986 to 1996. *Marine Pollution Bulletin 37*, 14–19). This paper centers on chemical concentrations in mollusks at 263 sites around the United States. It provides perspective on concentration ranges and on geographic distributions. For most organic chemicals and lead, concentrations vary in proportion to numbers of people living near a site. For elements, other than lead, high concentrations in mollusks can be due more to natural factors than to human activity. Concentrations of PAHs in tissues of mussels from urban areas are in a range reported to exert biological responses. Published by Elsevier Science Ltd.

Keywords: Mussel watch; Tissue analysis; Chlorinated hydrocarbons; Organotins; Polyaromatic hydrocarbons; Trace elements; Biological responses

1. Introduction

The US National Oceanic and Atmospheric Administration (NOAA) created the National Status and Trends (NS&T) Program to assess the influence of human

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activities on the quality of coastal and estuarine areas. The NS&T Mussel Watch Project began in 1986 to monitor spatial and temporal trends of chemical contamination by chemically analyzing mussels and oysters collected at fixed sites throughout the coastal United States. Butler (1973) inaugurated such wide-scale chemical monitoring through analyses of fish and mollusks collected in 1965–1972, when the main concern was possible contamination of seafood with the commonly used chlorinated pesticides. The Goldberg et al. (1978) nationwide project of 1976–1978 introduced the term "Mussel Watch" for a program using mollusks as sentinel organisms to monitoring chemical contamination. Mollusks serve that purpose because they concentrate chemicals from their surroundings making the chemical analyses simpler and less prone to error than that for water, they stay in a single location rather than swim around, and they are fairly resistant to chemical contamination and can be found living in areas where less hardy species may be absent. The NS&T Mussel Watch follows those earlier programs in its annual sampling of indigenous mollusks. This paper centers on statistical and geographic distributions of the observed chemical concentrations.

2. Chemicals monitored

The elements and groups of organic compounds listed in Table 1 are the subject of this report. The elements are all potential contaminants in the sense that their concentrations in the environment have been altered by human activities (Nriagu, 1989). Total DDT (ΣDDT), total chlordane (ΣChlordane), and total dieldrin (ΣDieldrin) are chlorinated pesticides. All uses of DDT and dieldrin were banned in the United States in the 1970s. Chlordane use on US crops ended in 1983, and its use for termite control effectively ended in 1988 (Shigenaka, 1990). Polychlorinated biphenyls (ΣPCB) are chlorinated compounds first used in the 1920s for a number of industrial purposes. Their high heat capacities and low dielectric constants were ideal for use in electrical transformers and capacitors. PCB use in the United States began being phased out in 1971, and a ban on new uses took effect in 1976. Large changes in concentrations of ΣDDT and ΣPCB were seen at some locations in the 1970s following bans on their further use (Mearns, Matta, Simecek-Beatty, Shigenaka, & Wert, 1988). The compounds are still found in tissues and sediments because PCB-containing devices are still in use, chlordane remains in the ground as a termiticide, and ΣDDT remains in the environment due to its resistance to degradation (Woodwell, Craig, & Johnson, 1971). The pesticide DDT is metabolized to DDE and DDD in the environment, but those compounds degrade very slowly under environmental conditions. The three butyltin compounds, aggregated as ΣBT, are found in mollusks because tributyltin (TBT) has been used as an antifouling agent in the paint commonly used on boats and some underwater marine facilities. Its use on vessels under 75 feet long was banned in 1988 by the US Organotin Anti-Fouling Paint Act. For all the chlorinated hydrocarbons and ΣBT, NS&T Mussel Watch results have shown statistically that annually measured concentrations are decreasing at a great many individual sites while increasing at few or none and that annual

Table 1
Chemicals measured in mollusks

Elements
Ag, Al, As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Se, Zn

Concentrations of individual organic compounds have been aggregated into groups.

ΣPCB = twice the sum of concentrations of eighteen congeners
PCB8, PCB18, PCB28, PCB44, PCB52, PCB66, PCB101, PCB105, PCB118, PCB128, PCB138, PCB153, PCB170, PCB180, PCB187, PCB195, PCB206, and PCB209

ΣDDT = sum of concentrations of *ortho* and *para* forms of parent and metabolites 2,4′DDE; 4,4′DDE; 2,4′DDD; 4,4′DDD; 2,4′DDT; and 4,4′DDT

ΣChlordane = sum of concentrations of four compounds
alpha-chlordane, *trans*-nonachlor, heptachlor, heptachlorepoxide

ΣDieldrin = sum of concentrations of two compounds
aldrin and dieldrin

ΣBT = sum of concentrations of parent compound and metabolites
monobutyltin, dibutyltin, and tributyltin [concentrations in terms of tin]

ΣLMW = sum of concentrations of 12 two-and three ring PAH compounds
naphthalene, 2-methylnaphthalene, 1-methylnaphthalene, biphenyl,
2,6-dimethylnaphthalene, acenaphthene, acenaphthylene,
1,6,7-trimethylnaphthalene, fluorene, phenanthrene, 1-methylphenanthrene and anthracene

ΣHMW = sum of concentrations of 12 four-and more-ring PAH compounds, fluoranthene, pyrene, benz[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*e*]pyrene, benzo[*a*]pyrene, perylene, dibenzanthracene, indeno[1,2,3-*cd*], pyrene, and benzo[*ghi*]perylene

ΣPAH = LMW + HMW (sum of 24 PAH compound concentrations)

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national median concentrations are decreasing in the coastal United States (O'Connor, 1996, 1998)

Polycyclic aromatic hydrocarbons (PAHs) are like elements in that they occur naturally. They are found in fossil fuels such as coal and oil and are produced when organic matter burns. Nonetheless, a multitude of human activities, from coal and wood burning to waste incineration, create PAH compounds in excess of those that would exist naturally. In addition, human production, transport, and use of oil release more PAHs to the environment, on a globally averaged basis, than does natural seepage (NRC, 1985). Because they are relatively more concentrated in oil than in combustion products, the low molecular-weight (LMW) 2- and 3-ring PAH compounds (Table 1) can be classified separately from the higher molecular-weight (HMW) 4- and 5-ring PAH compounds.

All of these trace metals and groups of organic compounds can be acutely or chronically toxic to marine life and to humans under some conditions. On the other hand, while the elements arsenic, copper, nickel, selenium, and zinc can be toxic at high concentrations, they are also essential in small quantities to the maintenance of life (Nielsen, 1988). Except for cadmium which is showing a decreasing trend, concentrations of elements in mollusks are not revealing temporal trends (O'Connor, 1996, 1998).

In recent years, the Mussel Watch project has added currently used pesticides and chlorinated hydrocarbons to its list of regularly measured monitored chemicals. National distributions of those compounds have been reported by Wade, Sericano, Gardinali, Wolff, and Chambers, (1998). Also, while not regularly measured, the Project has also generated a national distribution of molluscan concentrations of dioxins and furans (Gardinali & Wade, 1998).

3. Sampling sites

The NS&T Mussel Watch Project is national in scale and sampling sites should be representative of large areas rather than the small-scale patches of contamination commonly referred to as "hot spots". To this end, no sites were knowingly selected near waste discharge points. Furthermore, since the Mussel Watch Project is based on analyzing indigenous mussels and oysters, a site must support a sufficient population of these mollusks to provide annual samples.

NS&T sampling sites are not uniformly distributed along the coast. Within estuaries and embayments, they average about 20 km apart, while along open coastlines the average separation is 70 km. Almost half of the sites were selected in waters near urban areas, within 20 km of population centers in excess of 100 000 people. This choice was based on the assumptions that chemical contamination is higher, more likely to cause biological effects, and more spatially variable in these waters than in rural areas. The listing in Table 2 includes the 263 sites that have been sampled since 1990 and through 1998. Lauenstein, Cantillo, Kokkinakis, Frew, Jobling, and Fay, (1997) provide a full description of each site.

4. Species collected

Since no single species of mollusk is common to all coasts, it has been necessary to collect seven different ones: the blue mussel *Mytilus edulis* on the East Coast from Maine to Cape May, NJ; the American oyster *Crassostrea virginica* from Delaware Bay southward and throughout the Gulf of Mexico; the mussels *M. edulis* and *M. californianus* on the West Coast; the oyster *Ostrea sandvicensis* in Hawaii; the smooth-edged jewel box *Chama sinuosa at* the one site in the Florida Keys; the mangrove oyster *Crassostrea rhizophorae* in Puerto Rico; and the zebra mussels *Dreissena polymorpha* at sites in the Great Lakes.

The multitude of species complicates comparisons among sites, because tissues of different species can have different chemical concentrations even if the surrounding environments are identical. At sites in Long Island Sound where it was

Table 2
15th, 50th, and 85th percentiles concentrations among 214 sites measured in 1990 (except for Cr where concentration is 85th percentile of 1996 to 1998 site means, see text)

Chemical	15th pctl.	50th pctl.	85th pctl.
As	6.0 μg g^{-1} (dry)	9.5 μg g^{-1} (dry)	16 μg g^{-1} (dry)
Cd	1.2	2.8	5.9
Cr	0.41	1.4	4.7
Hg	0.04	0.10	0.23
Ni	0.82	1.7	3.1
Se	1.9	2.4	3.4
Pb	0.38	0.79	3.0
ΣChlordane	6.2 ng g^{-1}(dry)	13 ng g^{-1}(dry)	34 ng g^{-1}(dry)
ΣDieldrin	nd	3.5	8.5
ΣDDT	12	31	140
ΣPCB	25	110	420
ΣLMW	48	110	320
ΣHMW	24	120	770
ΣPAH	77	230	1100
ΣBT	9 ng of Sn g^{-1}(dry)	86 ng of Sn g^{-1}(dry)	300 ng of Sn g^{-1}(dry)
*Among oyster sites (*n = *107)*			
Ag	0.69 μg g^{-1}(dry)	2.5 μg g^{-1}(dry)	6.7 μg g^{-1}(dry)
Cu	63	150	360
Zn	1100	2400	5100
*Among mussel sites (*n = *107)*			
Ag	0.04 μg g^{-1}(dry)	0.12 μg g^{-1}(dry)	0.75 μg g^{-1}(dry)
Cu	6.9	8.6	12
Zn	86	130	200

possible to sample both mussels and oysters, *M. edulis* and *C. virginica*, three trace elements, Ag, Cu and Zn, were enriched in oysters by more than a factor of 10 relative to mussels. For other elements and for organic compounds no strong species-effect was observed. Similarly, at a site off the Columbia River two species of mussels, *M. edulis* and *M. californianus*, were sampled. In that case there were no important concentration differences for any element or organic compound (O'Connor, 1994).

There has been some discussion among malacologists over whether the West Coast organism called *M. edulis* is actually *M. galloprovincialis* in California and *M. trossulus* towards the north. In fact, the three species may be strains of a single *Mytilus* species (Seed, 1992). Given this uncertainty, the mussels collected at the Columbia River site may have been *M. trossulus* or even *M. galloprovincialis* instead of *M. edulis*. However, the lack of concentration differences between two *Mytilus* species at that site has been taken to validate comparisons among all marine mussels collected in the program.

5. Methods

Mollusks are shucked and whole soft parts are analyzed for their chemical content. Mollusks are collected at marine sites in the months of November through March with each site visited within 30 days of a prescribed date. In the Great Lakes, collections are made in August. From 1986 through 1991, six separate composite samples of 20 oysters or 30 mussels were collected at each site. Three composites were homogenized for trace element analyses and three for trace organic analyses. That protocol still applies whenever a site is sampled for the first time. Otherwise, since 1992 only two composite samples have been collected; one for organic and one for trace element analysis. The annual concentrations used in this report are the arithmetic means of three concentrations measured prior to 1992 or the single value measured since then. All data are available on the Internet at http://ccma-server.nos.noaa.gov. The detailed methods for sampling and chemical analysis are contained in National Status and Trends Methods Documents (Lauenstein & Cantillo, 1993a, 1993b, 1993c, 1993d, 1998).

O'Connor (1998) explained how Ag concentrations were often not detectable in mussels by the Instrumental Neutron Activation Analysis (INAA) method used since 1995. Mussel samples from those years have been reanalyzed by Graphite Furnace Atomic Absorption following sample digestion in excess HCl as required by Daskalakis, O'Connor, and Crecilius (1997). The data on the Internet now include the revise Ag concentrations. The revised data do not alter the conclusion of O'Connor (1998) that Ag concentrations are decreasing in mussels (not in oysters). O'Connor (1998) dismissed Cr concentrations in his trend analysis because, it was said, Cr is concentrated in entrained particles rather than mollusk tissue and was not thoroughly dissolved by methods used for tissue digestion. Since 1996 all Cr analyses have been by INAA so dissolution is not an issue, and Cr concentrations measured since then are used in this paper. Among all chemicals, Cr data are the most sensitive to entrained particles because its concentration in particles is more than 100 times its tissue concentration.

6. Summary statistics and comparisons among sites

Chemical concentrations in mollusks are the resultant of chemical intake via food and water and discharge via excretion and depuration into water. When the various rates, assimilation efficiencies, and partitioning of chemicals among and within dissolved and particulate phases are known, it is possible to quantitatively equate chemical concentrations in mussels to concentrations in their surroundings (Wang, Fisher, & Luoma, 1996). However, so many factors affect rates and assimilation efficiencies that it is generally not possible to back-calculate from a chemical's concentration in a mollusk to its concentration in the mollusk's surroundings. Nonetheless, when a molluscan concentration at a site is high on a national scale it is valid to conclude that the chemical's concentration is also relatively high in the water at that site.

As exemplified in Fig. 1a–d for ΣPCB, ΣHMW, Hg, and Cd, respectively, the national distributions of concentrations are approximately lognormal. As was done with NS&T sediment data (Daskalakis & O'Connor, 1995), results could be summarized as geometric means plus or minus the standard deviations. However, rather than force a mathematical distribution function, central tendencies and variations are tabulated simply in Table 2 as the 15th, 50th, and 85th percentiles among 214 site means sites measured in 1990 at marine sites. That year was chosen as the base for comparison because it was the year in which the most sites were sampled. Among the 238 marine sites only 24 were not sampled until 1991 or later. The numbers in Table 2 would change little if they were percentiles of site means calculated over all years. Nonetheless, because some chemical concentrations are decreasing (O'Connor, 1998), concentrations from one individual year are used as a basis for comparisons. For chromium where only data since 1996 are used, the percentiles in Table 2 are from the distribution of site mean concentrations aggregated over 1996, 1997, and 1998 (i.e. 1 or 2 years per site).

The 85th percentile concentrations in Table 2 are a basis for identifying sites with "high" concentrations. All chemicals and marine sites are listed in Table 3 with site/concentration pairs indicated with an "X" if the concentration was at or above the 85th percentile in at least half the sampled years since 1990. The latter condition avoids any site categorizations based on a single atypical concentration. While they are listed, caution is in order for sites sampled only once or twice and put in a "high" category on the basis of one measurement. An alternative to Table 3 could be readily constructed using 15th percentiles as "low" concentrations. However it is the "highs" that might be thought to be subject to human control or of biological consequence.

7. Concentrations above public health advisories

The intent of the Mussel Watch Project is to monitor the status and trends of coastal contamination, regardless of whether chemical concentrations present a hazard to marine biota or to human consumers of seafood. Naturally, though, there is interest in hazard potential. The US Food and Drug Administration (FDA) prohibits the interstate shipment and sale of seafood containing more than specified limiting concentrations for Hg and certain chlorinated hydrocarbons. There are also FDA guidelines suggesting that mollusks not be consumed if concentration limits are exceeded for Cr, Ni, Pb, Cd, and As. For As, the FDA limit is for the inorganic form of the element and the FDA assumption is that inorganic As is 10% of the total As in marine organisms. In all cases FDA limits are wet-weight based concentrations. Among the 4000 mussel and oyster samples analyzed in the Mussel Watch Program, the mean and median wet weight percentages have both been 88%. Multiplying by 7.5 to convert wet-weight based limits to a dry-weight basis (assuming 85% wet weight) indicates that no mollusks collected in any year exceeded the FDA limit or guideline for Hg, Cr, Ni, or As. Except for ΣPCB at the Angelica Rock site in Buzzards Bay, MA in 1989, none of the chlorinated hydrocarbon limits

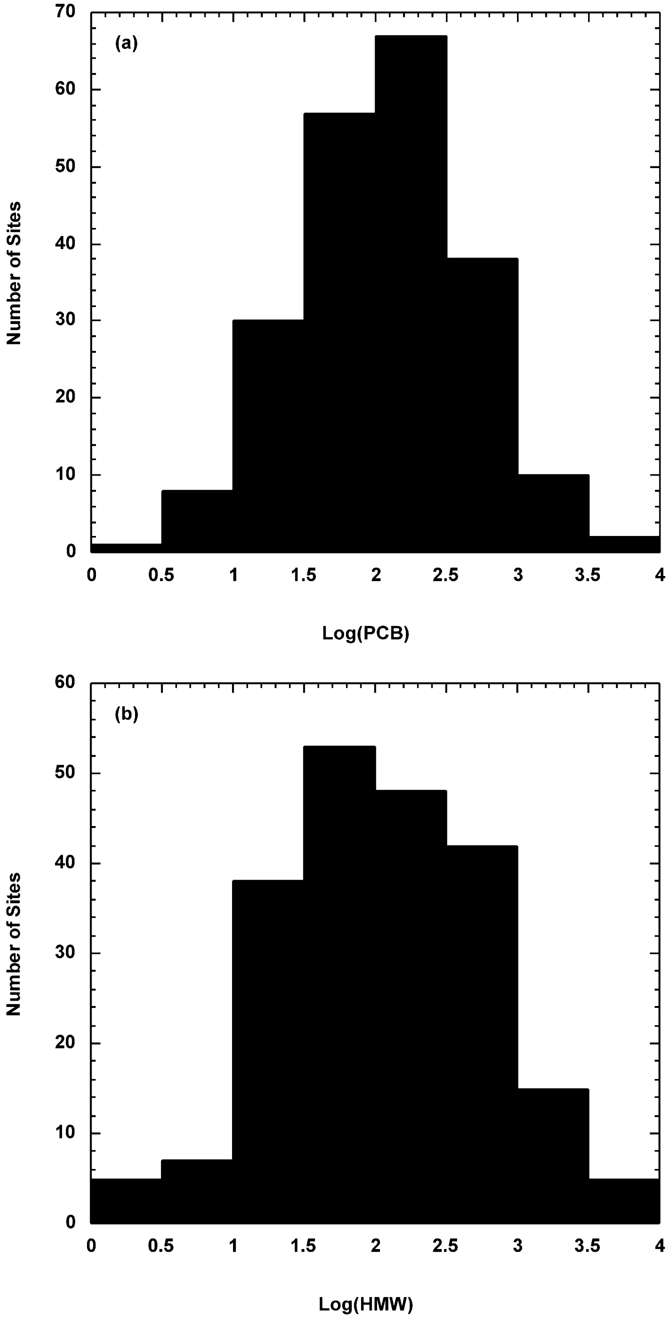


Fig. 1. (a–d) Histograms of logarithms of 1990 site mean concentrations of (a) ΣPCB, (b) ΣHMW, (c) Hg, and (d) Cd.

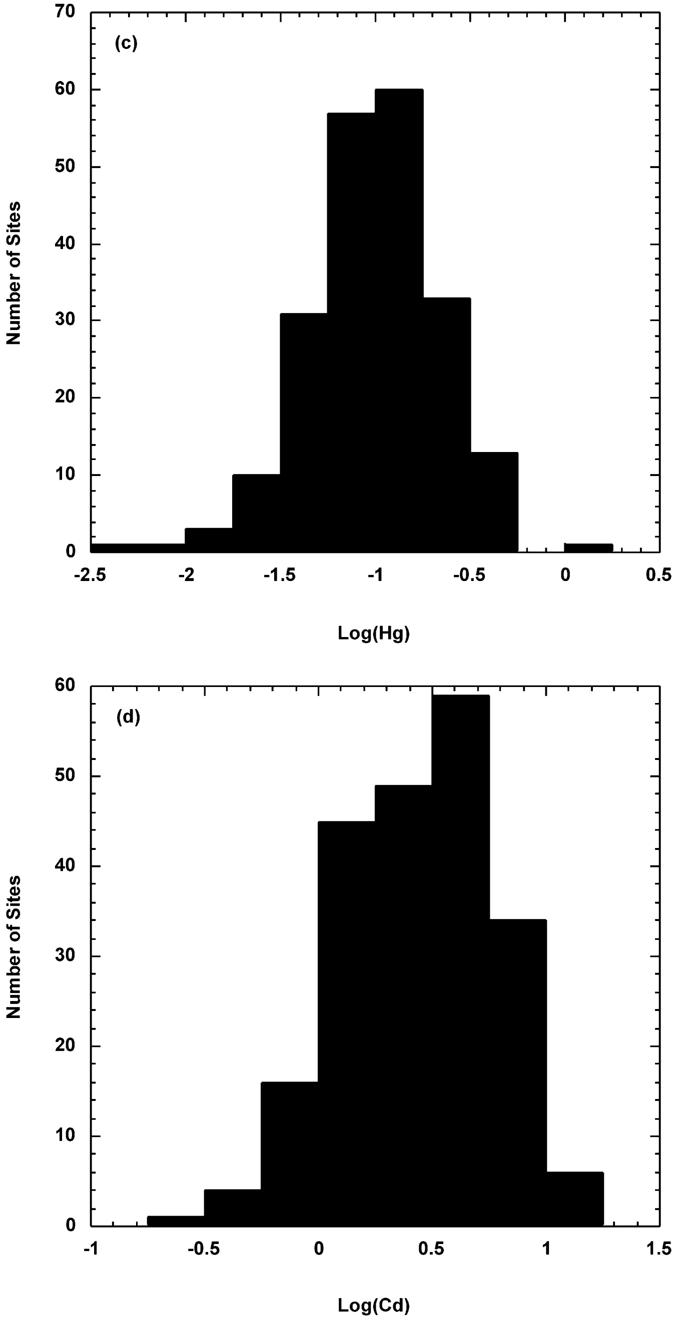


Fig. 1. (*Continued*).

Table 3
Site/chemicalcombinations (x) where concentration in molluscs (Sp = Species) was "high" in at least half the years samples since 1990 (years)[a]

Site	General location	Specific location	ST	Sp	Years	Ag	As	Cd	Cu	Cr	Hg	Ni	Pb	Se	Zn	tDield	tCdane	tDDT	tPCB	tLMW	tHMW	tPAH	tBT
PBPI	Penobscot Bay	Pickering Island	ME	me	6																		
PBSI	Penobscot Bay	Sears Island	ME	me	7						x												
MSSP	Merriconeag Snd.	Stover Point	ME	me	7																		
CAKP	Cape Arundel	Kennebunkport	ME	me	6																		
GBDP	Great Bay	Dover Point	NH	me	1				x	x	x	x	x	x									
CAGH	Cape Ann	Gap Head	MA	me	7		x				x												
SHFP	Salem Harbor	Folger Point	MA	me	6					x	x		x										
MBNB	Massachusetts Bay	Nahant Bay	MA	me	5						x		x										
BHDI	Boston Harbor	Deer Island	MA	me	6					x	x		x						x	x	x	x	
BHDB	Boston Harbor	Dorchester Bay	MA	me	5				x	x	x		x						x	x	x	x	
BHHB	Boston Harbor	Hingham Bay	MA	me	6	x				x	x		x			x			x				
BHBI	Boston Harbor	Brewster Island	MA	me	7						x		x						x				
MBNR	Massachusetts Bay	North River	MA	me	4						x		x										
DBCI	Duxbury Bay	Clarks Island	MA	me	7								x										
CCNH	Cape Cod	Nauset Harbor	MA	me	6																		
BBNI	Buzzards Bay	Naushon Island	MA	me	6		x							x					x				
BBWF	Buzzards Bay	West Falmouth	MA	me	6		x												x				
BBCC	Buzzards Bay	Cape Cod Canal	MA	me	6		x																
BBAP	Buzzards Bay	Angelica Point	MA	me	1									x					x				
BBAR	Buzzards Bay	Angelica Rock	MA	me	7		x						x						x				
BBRH	Buzzards Bay	Round Hill	MA	me	4		x												x				
BBGN	Buzzards Bay	Goosebury Neck	MA	me	7																		
NBDI	Narragansett Bay	Dyer Island	RI	me	5											x							
NBPI	Narragansett Bay	Patience Island	RI	me	7											x			x				
NBDU	Narragansett Bay	Dutch Island	RI	me	7																		
BIBI	Block Island Sound	Block Island	RI	me	7																		
LICR	Long Island Sound	Connecticut River	CT	me	6				x										x				
LINH	Long Island Sound	New Haven	CT	me	7														x				
LIHR	Long Island Sound	Housatonic River	CT	me	6				x							x			x				
LISI	Long Island Sound	Sheffield Island	CT	me	5											x							
LIMR	Long Island Sound	Mamaroneck	NY	me	6				x				x			x	x		x				
LITN	Long Island Sound	Throgs Neck	NY	me	7								x			x	x		x		x	x	
LIHH	Long Island Sound	Hempstead Harbor	NY	me	7											x							
LIHU	Long Island Sound	Huntington Harbor	NY	me	6																		
LIPJ	Long Island Sound	Port Jefferson	NY	me	7																		

Table 3 (*continued*)

Site	General location	Specific location	ST	Sp	Years	Ag	As	Cd	Cu	Cr	Hg	Ni	Pb	Se	Zn	tDield	tCdane	tDDT	tPCB	tLMW	tHMW	tPAH	tBT
LIGB	Long Island	Gardiners Bay	NY	me	6																		
MBTH	Moriches Bay	Tuthill Point	NY	me	7						x		x	x									
LIFI	Long Island	Fire Island Inlet	NY	me	3				x		x			x									
LIJI	Long Island	Jones Inlet	NY	me	5						x												x
HRJB	Hudson/Raritan Estuary	Jamaica Bay	NY	me	6				x		x		x			x			x	x	x	x	
HRUB	Hudson/Raritan Estuary	Upper Bay	NY	me	5	x			x		x	x	x	x	x	x		x	x	x	x	x	
HRLB	Hudson/Raritan Estuary	Lower Bay	NY	me	4	x			x			x	x	x		x	x	x	x		x	x	
HRRB	Hudson/Raritan Estuary	Raritan Bay	NJ	me	4	x			x	x	x	x	x	x	x	x	x	x	x		x	x	
NYSH	New York Bight	Sandy Hook	NJ	me	5				x		x	x	x			x		x	x		x	x	
NYLB	New York Bight	Long Branch	NJ	me	5				x	x		x	x	x		x			x		x	x	
NYSR	New York Bight	Shark River	NJ	me	7					x	x		x						x	x	x	x	
BIBL	Barnegat Inlet	Barnegat Light	NJ	me	6				x		x			x									
AIAC	Absecon Inlet	Atlantic City	NJ	me	6					x	x		x	x									
DBCM	Delaware Bay	Cape May	NJ	me	7									x									
DBFE	Delaware Bay	False Egg Island Point	NJ	cv	6							x			x								
DBBD	Delaware Bay	Ben Davis Point Shoal	NJ	cv	5	x						x		x	x	x							
DBAP	Delaware Bay	Arnolds Point Shoal	NJ	cv	6	x		x	x			x			x	x		x					
DBKI	Delaware Bay	Kelly Island	DE	cv	6	x						x				x							
DBCH	Delaware Bay	Cape Henlopen	DE	me	7									x									
CBBO	Chesapeake Bay	Bodkin Point	MD	cv	4	x		x	x			x		x	x	x							x
CBMP	Chesapeake Bay	Mountain Point Bar	MD	cv	6	x		x	x			x			x								
CBHP	Chesapeake Bay	Hackett Point Bar	MD	cv	6							x											
CBCP	Chesapeake Bay	Choptank River	MD	cv	6							x											
CBHG	Chesapeake Bay	Hog Point	MD	cv	6							x											
PRSP	Potomac River	Swan Point	MD	cv	3							x											
PRMC	Potomac River	Mattox Creek	VA	cv	4																		
PRRP	Potomac River	Ragged Point	VA	cv	6							x											
RRRR	Rappahannock River	Ross Rock	VA	cv	6			x															
CBDP	Chesapeake Bay	Dandy Point	VA	cv	6															x			
CBJR	Chesapeake Bay	James River	VA	cv	4	x		x	x			x		x	x								x
CBCC	Chesapeake Bay	Cape Charles	VA	cv	6																		
CBCI	Chincoteague Bay	Chincoteague Inlet	VA	cv	7																		
QIUB	Quinby Inlet	Upshur Bay	VA	cv	7	x																	
RSJC	Roanoke Sound	John Creek	NC	cv	6																		

(*Table continued on next page*)

Table 3 (*continued*)

Site	General location	Specific location	ST	Sp	Years	Ag	As	Cd	Cu	Cr	Hg	Ni	Pb	Se	Zn	tDield	tCdane	tDDT	tPCB	tLMW	tHMW	tPAH	tBT
PSWB	Pamlico Sound	Wysocking Bay	NC	cv	6							x											
PSPR	Pamlico Sound	Pungo River	NC	cv	5																		
PSNR	Pamlico Sound	Neuse River	NC	cv	6																		
PSCH	Pamlico Sound	Cape Hatteras	NC	cv	7		x																
BIPI	Beaufort Inlet	Pivers Island	NC	cv	7		x													x		x	
CFBI	Cape Fear	Battery Island	NC	cv	8		x																
WBLB	Winyah Bay	Lower Bay	SC	cv	7		x																
SRNB	Santee River	North Bay	SC	cv	7		x							x									
CHFJ	Charleston Harbor	Fort Johnson	SC	cv	7		x																
CHSF	Charleston Harbor	Shutes Folly Island	SC	cv	6		x																
SRTI	Savannah River Estuary	Tybee Island	GA	cv	6		x																
SSSI	Sapelo Sound	Sapelo Island	GA	cv	6		x																
ARWI	Altamaha River	Wolfe Island	GA	cv	6		x							x									
SJCB	St. Johns River	Chicopit Bay	FL	cv	6		x							x									
MRCB	Matanzas River	Crescent Beach	FL	cv	6		x																
IRSR	Indian River	Sebastian River	FL	cv	6															x			
NMML	North Miami	Maule Lake	FL	cv	6				x						x								x
BBGC	Biscayne Bay	Goulds Canal	FL	cv	6																		
FBJB	Florida Bay	Joe Bay	FL	cv	5						x												
FBFO	Florida Bay	Flamingo	FL	cv	5				x											x	x	x	
BHKF	Bahia Honda	Key FLorida	FL	cs	5		x					x											
PRBB	Puerto Rico	Bahia De Boqueron	PR	cr	5																		
PRBM	Puerto Rico	Bahia Montalva	PR	cr	4									x									
PRBJ	Puerto Rico	Bahia De Jobos	PR	cr	5																		
EVFU	Everglades	Faka Union Bay	FL	cv	7																		
RBHC	Rookery Bay	Henderson Creek	FL	cv	7																		
NBNB	Naples Bay	Naples Bay	FL	cv	7				x														
CBFM	Charlotte Harbor	Fort Meyers	FL	cv	6																		x
CBBI	Charlotte Harbor	Bird Island	FL	cv	7																		
TBCB	Tampa Bay	Cockroach Bay	FL	cv	6						x						x						
TBHB	Tampa Bay	Hillsborough Bay	FL	cv	4															x	x	x	x
TBKA	Tampa Bay	Peter O. Knight Airport	FL	cv	7												x		x				x
TBOT	Tampa Bay	Old Tampa Bay	FL	cv	6						x				x								
TBPB	Tampa Bay	Papys Bayou	FL	cv	7						x												
TBMK	Tampa Bay	Mullet Key Bayou	FL	cv	7																		
TBNP	Tampa Bay	Navarez Park	FL	cv	7		x										x						

Table 3 (*continued*)

Site	General location	Specific location	ST	Sp	Years	Ag	As	Cd	Cu	Cr	Hg	Ni	Pb	Se	Zn	tDield	tCdane	tDDT	tPCB	tLMW	tHMW	tPAH	tBT
CKBP	Cedar Key	Black Point	FL	cv	7		x																
AESP	Apalachee Bay	Spring Creek	FL	cv	7		x																
APCP	Apalachicola Bay	Cat Point Bar	FL	cv	6																		
APDB	Apalachicola Bay	Dry Bar	FL	cv	6																		
SAWB	St. Andrews Bay	Watson Bayou	FL	cv	6		x		x											x	x	x	
PCMP	Panama City	Municipal Pier	FL	cv	4		x		x											x	x	x	x
PCLO	Panama City	Little Oyster Bar	FL	cv	7													x					
CBSR	Choctawhatchee Bay	Off Santa Rosa	FL	cv	8									x									
CBBL	Choctawhatchee Bay	Bens Lake	FL	cv	3						x			x		x		x					
CBPP	Choctawhatchee Bay	Postil Point	FL	cv	8						x		x	x				x					
CBBB	Choctawhatchee Bay	Boggy Bayou	FL	cv	3	x								x		x							
CBJB	Choctawhatchee Bay	Joes Bayou	FL	cv	8				x		x			x	x					x	x	x	x
PBSP	Pensacola Bay	Sabine Point	FL	cv	4																		
PBIB	Pensacola Bay	Indian Bayou	FL	cv	5																		
PBPH	Pensacola Bay	Public Harbor	FL	cv	7									x		x							
MBDR	Mobile Bay	Dog River	AL	cv	3	x		x	x			x			x			x		x	x		
MBHI	Mobile Bay	Hollingers Is. Chan.	AL	cv	7	x		x	x						x			x					
MBCP	Mobile Bay	Cedar Point Reef	AL	cv	7																		
MSPB	Mississippi Sound	Pascagoula Bay	MS	cv	7																		
MSBB	Mississippi Sound	Biloxi Bay	MS	cv	5										x		x			x			
MSPC	Mississippi Sound	Pass Christian	MS	cv	6			x															
LPNO	Lake Pontchartrain	New Orleans	LA	cv	4	x		x	x			x			x	x	x			x			
LBGO	Lake Borgne	Gulf Outlet	LA	cv	4			x				x			x								
LBMP	Lake Borgne	Malheureux Point	LA	cv	8			x						x									
BSBG	Breton Sound	Bay Gardene	LA	cv	7																		
BSSI	Breton Sound	Sable Island	LA	cv	7			x				x				x							
MRPL	Mississippi River	Pass A Loutre	LA	cv	6			x	x			x				x							
MRTP	Mississippi River	Tiger Pass	LA	cv	6			x						x		x							
BBMB	Barataria Bay	Middle Bank	LA	cv	6															x			x
BBSD	Barataria Bay	Bayou Saint Denis	LA	cv	5																		
TBLF	Terrebonne Bay	Lake Felicity	LA	cv	7																		
TBLB	Terrebonne Bay	Lake Barre	LA	cv	6																		
CLCL	Caillou Lake	Caillou Lake	LA	cv	7																		
ABOB	Atchafalaya Bay	Oyster Bayou	LA	cv	6			x															

(*Table continued on next page*)

Table 3 (*continued*)

Site	General location	Specific location	ST	Sp	Years	Ag	As	Cd	Cu	Cr	Hg	Ni	Pb	Se	Zn	tDield	tCdane	tDDT	tPCB	tLMW	tHMW	tPAH	tBT
VBSP	Vermilion Bay	Southwest Pass	LA	cv	7			x	x					x									
JHJH	Joseph Harbor Bayou	Joseph Harbor Bayou	LA	cv	6									x									
CLSJ	Calcasieu Lake	St. Johns Island	LA	cv	7									x									
CLLC	Calcasieu Lake	Lake Charles	LA	cv	7																		
SLBB	Sabine Lake	Blue Buck Point	TX	cv	7									x									
GBHR	Galveston Bay	Hanna Reef	TX	cv	6			x						x									
GBSC	Galveston Bay	Ship Channel	TX	cv	5			x						x		x	x		x		x	x	x
GBYC	Galveston Bay	Yacht Club	TX	cv	6											x	x						x
GBTD	Galveston Bay	Todds Dump	TX	cv	7																		
GBOB	Galveston Bay	Offatts Bayou	TX	cv	7											x	x						
GBCR	Galveston Bay	Confederate Reef	TX	cv	6	x								x									
BRFS	Brazos River	Freeport Surfside	TX	cv	6									x				x			x		
BRCL	Brazos River	Cedar Lakes	TX	cv	7																		
MBEM	Matagorda Bay	East Matagorda	TX	cv	6			x						x									
MBTP	Matagorda Bay	Tres Palacios Bay	TX	cv	6									x									
MBCB	Matagorda Bay	Carancahua Bay	TX	cv	5																		
MBLR	Matagorda Bay	Lavaca River Mouth	TX	cv	6						x	x		x									
MBGP	Matagorda Bay	Gallinipper Point	TX	cv	7					x	x	x		x									
ESBD	Espiritu Santo	Bill Days Reef	TX	cv	6																		
ESSP	Espiritu Santo	South Pass Reef	TX	cv	5									x									
SAMP	San Antonio Bay	Mosquito Point	TX	cv	4									x									
SAPP	San Antonio Bay	Panther Point Reef	TX	cv	7									x									
MBAR	Mesquite Bay	Ayres Reef	TX	cv	7									x									
ABLR	Aransas Bay	Long Reef	TX	cv	7																		
CBCR	Copano Bay	Copano Reef	TX	cv	5			x															
CCIC	Corpus Christi	Ingleside Cove	TX	cv	3																		x
CCNB	Corpus Christi	Nueces Bay	TX	cv	6									x									
CCBH	Corpus Christi	Boat Harbor	TX	cv	3			x							x					x			
LMAC	Lower Laguna Madre	Arroyo Colorado	TX	cv	4		x																
LMPI	Lower Laguna Madre	Port Isabel	TX	cv	3																		
LMSB	Lower Laguna Madre	South Bay	TX	cv	7																		
IBNJ	Imperial Beach	North Jetty	CA	mc	6	x				x								x					
SDCB	San Diego Bay	Coronado Bridge	CA	me	7				x	x					x				x		x	x	x
PLLH	Point Loma	Lighthouse	CA	mc	5	x	x		x		x												
SDHI	San Diego Bay	Harbor Island	CA	me	6				x						x				x	x	x	x	x
MBVB	Mission Bay	Ventura Bridge	CA	me	6																		

Table 3 (*continued*)

Site	General location	Specific location	ST	Sp	Years	Ag	As	Cd	Cu	Cr	Hg	Ni	Pb	Se	Zn	tDield	tCdane	tDDT	tPCB	tLMW	tHMW	tPAH	tBT
LJLJ	La Jolla	Point La Jolla	CA	mc	6	x	x																
OSBJ	Oceanside	Beach Jetty	CA	me	6													x					
SCBR	Santa Catalina Island	Bird Rock	CA	mc	6																		
NBWJ	Newport Beach	Wedge Jetty	CA	mc	6	x							x					x					
ABWJ	Anaheim Bay	West Jetty	CA	mc	7								x			x	x	x					
LBBW	Long Beach	Breakwater	CA	me	7								x			x		x					
SPFP	San Pedro Harbor	Fishing Pier	CA	me	6				x									x	x	x	x	x	x
PVRP	Palos Verdes	Royal Palms State Park	CA	mc	7	x	x								x			x					
RBMJ	Redondo Beach	Municipal Jetty	CA	mc	7	x							x	x				x					
MDSJ	Marina Del Rey	South Jetty	CA	me	7								x	x		x	x	x					
TBSM	Las Tunas Beach	Santa Monica Bay	CA	mc	7	x				x			x										
PDPD	Point Dume	Point Dume	CA	mc	7	x																	
SCFP	Santa Cruz Island	Fraser Point	CA	mc	6		x																
SBSB	Point S. Barbara	Point Santa Barbara	CA	mc	7	x				x													
PCPC	Point Conception	Point Conception	CA	mc	6																		
SLSL	San Luis Obispo Bay	Point San Luis	CA	mc	7																		
SSSS	San Simeon Point	San Simeon Point	CA	mc	6		x	x		x	x	x											
PGLP	Pacific Grove	Lovers Point	CA	mc	7								x			x							
MBML	Monterey Bay	Moss Landing	CA	mc	8					x		x				x		x					
MBES	Monterey Bay	Elkhorn Slough	CA	mc	4					x						x		x					
MBSC	Monterey Bay	Point Santa Cruz	CA	mc	7											x							
SFSM	San Francisco Bay	San Mateo Bridge	CA	me	7						x	x		x		x			x				
SFDB	San Francisco Bay	Dumbarton Bridge	CA	me	7						x	x		x		x			x				
SFEM	San Francisco Bay	Emeryville	CA	me	7			x		x	x	x	x	x	x	x		x	x	x	x	x	
TBSR	Tomales Bay	Spengers Residence	CA	me	7					x		x		x									
BBBE	Bodega Bay	Bodega Bay Entrance	CA	mc	7			x															
PALH	Point Arena	Lighthouse	CA	mc	7			x				x											
PDSC	Point Delgada	Shelter Cove	CA	mc	7			x															
HMBJ	Eureka	Humboldt Bay Jetty	CA	mc	6					x													
EUSB	Eureka	Samoa Bridge	CA	mc	6				x	x		x											
SGSG	Crescent	Point St. George	CA	mc	6		x	x		x		x											
CBCH	Coos Bay	Coos Head	OR	mc	7																		
CBRP	Coos Bay	Russell Point	OR	me	7					x		x			x								
YBOP	Yaquina Bay	Oneatta Point	OR	me	7					x													

(*Table continued on next page*)

Table 3 (*continued*)

Site	General location	Specific location	ST	Sp	Years	Ag	As	Cd	Cu	Cr	Hg	Ni	Pb	Se	Zn	tDield	tCdane	tDDT	tPCB	tLMW	tHMW	tPAH	tBT
YHYH	Yaquina Bay	Yaquina Head	OR	mc	2							x											
YHFC	Yaquina Head	Foggerty Creek	OR	mc	4					x													
TBHP	Tillamook Bay	Hobsonville Point	OR	me	7																		
CRSJ	Columbia River	South Jetty	OR	me	6					x													
CRNJ	Columbia River	North Jetty	WA	me	1																		
WBNA	Willapa Bay	Nahcotta	WA	me	6					x		x		x									
GHWJ	Grays Harbor	Westport Jetty	WA	mc	7					x						x							
JFCF	Juan de Fuca Strait	Cape FLattery	WA	mc	7																		
PSPA	Puget Sound	Port Angeles	WA	me	7																		
PSPT	Puget Sound	Port Townsend	WA	me	7															x	x	x	
PSHC	Puget Sound	Hood Canal	WA	me	7																		
SSBI	South Puget Sound	Budd Inlet	WA	me	7																		
CBTP	Commencement Bay	Tahlequah Point	WA	me	7															x	x	x	
PSSS	Puget Sound	South Seattle	WA	me	6															x	x	x	
EBDH	Elliott Bay	Duwamish Head	WA	me	4															x	x	x	
EBFR	Elliott Bay	Four-Mile Rock	WA	me	7										x					x	x	x	
SIWP	Sinclair Inlet	Waterman Point	WA	me	7																		
WIPP	Whidbey Island	Possession Point	WA	me	7							x											
PSEH	Puget Sound	Everett Harbor	WA	me	4					x		x	x							x	x	x	
BBSM	Bellingham Bay	Squalicum Marina Jetty	WA	me	7					x		x			x					x	x	x	
PRPR	Point Roberts	Point Roberts	WA	me	6					x		x											
KTMP	Ketchikan	Mountain Point	AK	me	2				x					x									
NBES	Nahku Bay	East Side	AK	me	2				x				x	x									
PVMC	Port Valdez	Mineral Creek FLats	AK	me	6					x				x									
UISB	Unakwit Inlet	Siwash Bay	AK	me	6									x									
CIHS	Cook Inlet	Homer Spit	AK	me	2				x														
HHKL	Honolulu Harbor	Keehi Lagoon	HI	os	6	x			x	x	x	x	x	x		x	x			x	x	x	x
HHKB	Hawaii	Kaneohe Bay	HI	os	2	x	x		x							x	x						
BPBP	Barbers Point	Barbers Point Harbor	HI	os	5		x		x			x											x

[a] Site codes in forst column of use when reviewing data on Internet (see text).

were exceeded. The limit for Cd (for humans eating shellfish at the 90th percentile consumption rate) was exceeded in 1991 at the site on Lake Ponchartrain in New Orleans, LA. In several years, mollusks at many of the sites listed as having "high" Pb in Table 3 had Pb concentrations that exceeded the 6 ug/g-dry guideline for children consuming mollusks at the 90th percentile rate. Fewer sites or years had Pb in excess of the 11 ug/g limit for children consuming at the mean rate or pregnant woman consuming at the 90th percentile rate. None exceed guidelines for adult consumption.

8. Concentrations affecting mollusks

The Mussel Watch Project has not included any biochemical measures of response to chemical contamination. Histological slides were made of gonadal tissue to quantify the reproductive stage of the organisms and neoplasia and parasitic infestations were noted. Kim, Powell, Wade, Presley, and Sericano, (1998) could not connect any of the parasite observations with tissue concentrations of chemicals. Hillman et al. (1992) did note that the ΣPAH body burdens at the 18 sites where neoplasia was found in *M. edulis* were among the higher body burdens, but ΣPAH body burdens were higher at sites without neoplasia. Moreover, in a detailed correlative and experimental examination Krishnakumar, Casillas, Snider, Kagley, and Varanasi (1999) could not establish that hemic neoplasia, endemic to mussels in Puget Sound, is induced or promoted by PAHs or PCBs.

Jarvinen and Ankley (1999) have compiled a large dataset of chemical body burdens and corresponding measures of effect on survival, growth, and reproduction of aquatic organisms. The maximum concentrations measured in the Mussel Watch program for Cd, Hg, Pb, and Ni, are an order of magnitude below the lowest concentrations associated with those effects in mollusks. That also applies for Cu and Zn in mussels. For tributyltin the only listed body burden with an effect was about 30% above the highest tributyltin concentration (3000 ng/g-dry as Sn) measured in Mussel Watch. There are no data for Cu and Zn effects on oysters and no data for effects on any mollusks associated with body burdens of As, Se, Ag, or any of the organic chemicals in Table 1. Field collected mollusks obviously do not harbor lethal residues of chemicals but it is noted that the higher of the observed molluscan concentrations of organic chemicals are of the order of 1 nM/g-wet which is 1000× lower than μM/g critical body residues of nonpolar organics associated with death of marine organisms (Kane Driscoll, Schaffner, & Dickhut, 1998).

Widdows, Donkin, Evans, Page, and Salkeld (1995) measured hydrocarbon concentrations in and Scope for Growth of *M. edulis* held in cages along hydrocarbon gradients in waters of the United Kingdom. When all their data are viewed as a whole, effects began to appear at body burdens of about 100 ng/g-wet of 2- and 3-ring petroleum hydrocarbons. Their chemical methods did not discriminate among different LMW compounds and comparisons with Mussel watch concentrations are only approximations but 100 ng/g-wet corresponds to about 750 ng/-dry Σ LMW.

This is well above the Mussel Watch "high" concentration (Table 2) but has been exceeded at 31 sites in at least 1 year since 1990.

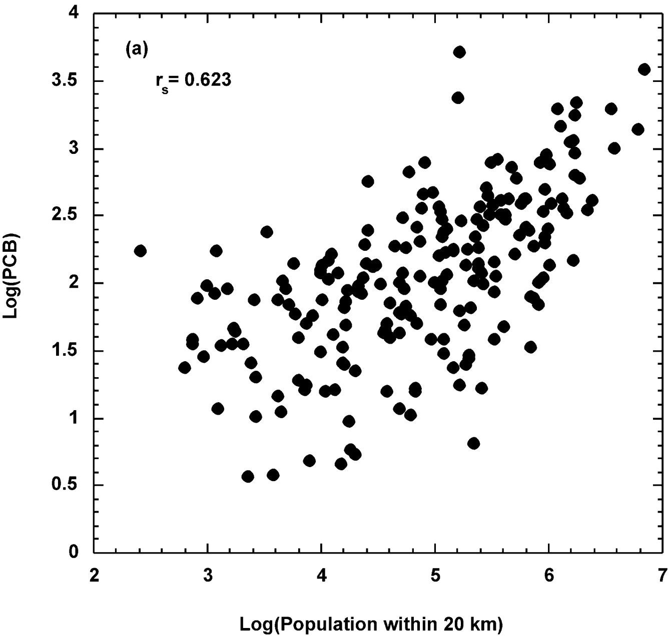
Krishnakumar, Casillas, and Varanasi (1994) associated alterations in lysosomes of digestive cells in mussels with body burdens of PAHs and PCBs. In their case PAH compounds were measured just as they are in the NS&T Program. (Their ΣLMW and ΣHMW totals included 5 and 1, respectively, fewer compounds than the totals used in this paper but overall differences are slight.) At the four sites of their nine where no lysosomal alterations were found, ΣLMW and ΣHMW concentrations in *M. edulis* were 80 and 90 ng/g-dry or less. Mollusks at about half of the NS&T sites have ΣLMW and ΣHMW concentrations below those levels. The five sites where alterations were found included one with ΣLMW and ΣHMW concentrations above the highest ever measured at a Mussel Watch site. However, at the fourth most contaminated of the five affected sites, the ΣLMW and ΣHMW concentrations were 820 and 320 ng/g, respectively. Concentrations close to those defining "high" in Table 2. The lowest concentrations among the five affected sites were 100 (ΣLMW) and 120 (ΣHMW), close to the median Mussel Watch concentrations and little different from the concentrations in unaffected mussels. The ΣPCB tissue concentrations, like those of the PAHs, were near or above the Mussel Watch "high" at the four most contaminated sites. Also like PAHs, ΣPCB concentrations at one affected and four unaffected sites were near or below the Mussel Watch median. Among other chemicals measured by Krishnakumar et al. (1994) concentrations were generally less than the Mussel Watch "highs" and not consistently elevated among affected mussels relative to normal mussels.

It does appear that PAHs and possibly PCBs at concentrations found in molluscan tissues at Mussel Watch sites in urban areas could be affecting the organisms.

9. Human influence on concentrations

Concentrations of chemicals are evidence of contamination only if they would be lower in the absence of any human influence. There are fairly strong connections between human population density and chemical concentrations in oysters and mussels for ΣChlordane, ΣDDT, ΣPCB, Σbutyltin, ΣHMW, and lead where Spearman correlation coefficients are greater than 0.5 (Fig. 2; Table 4). The connection with urban areas for ΣPCB, and lead was also noted in the earlier Mussel Watch project (Goldberg, Koide, Hodge, Flegal, & Martin, 1983) is not surprising. ΣChlordane, ΣDDT, ΣPCB, and Σbutyltin, are synthetic chemicals whose concentrations would be zero in the absence of man. While ΣHMW and lead would always be found in mollusks, their present concentrations are due almost entirely to human actions. For ΣDieldrin, ΣLMW and the elements Ag, Hg, and Zn the national-scale correlations (Table 4) are low but more than 40% of the "high" concentrations are found among the 15% of marine sites with 800 000 or more people living within 20 km.

While one might expect high concentrations of all chemicals to be due to humans and to cluster at sites near population centers, high concentrations of four elements



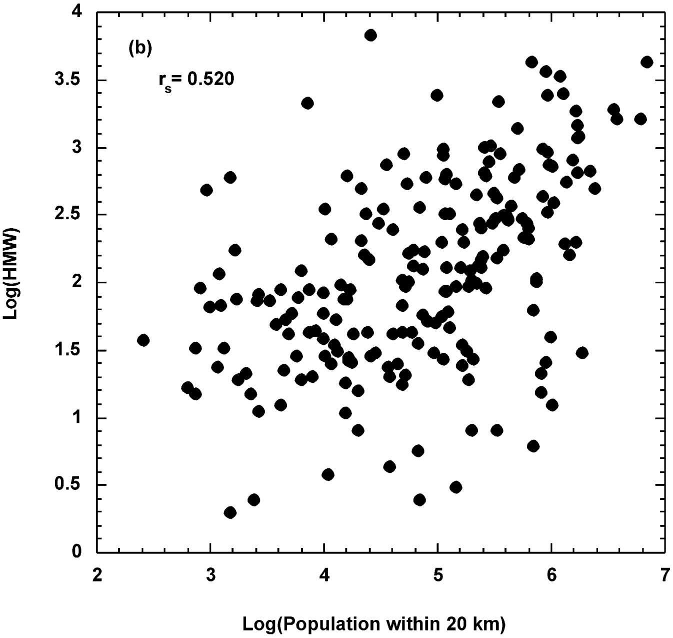


Fig. 2. (a–d) 1990 site mean concentrations of (a) ΣPCB, (b) ΣHMW, (c) Hg, and (d) Cd versus population within 20 km of each site. Log-log plot provides separation among points and, for that reason, data are not shown for four sites with <100 people residing within 20 km. The four plots span the range of Spearman correlations (Table 4) based on ranks not actual concentration or population values.

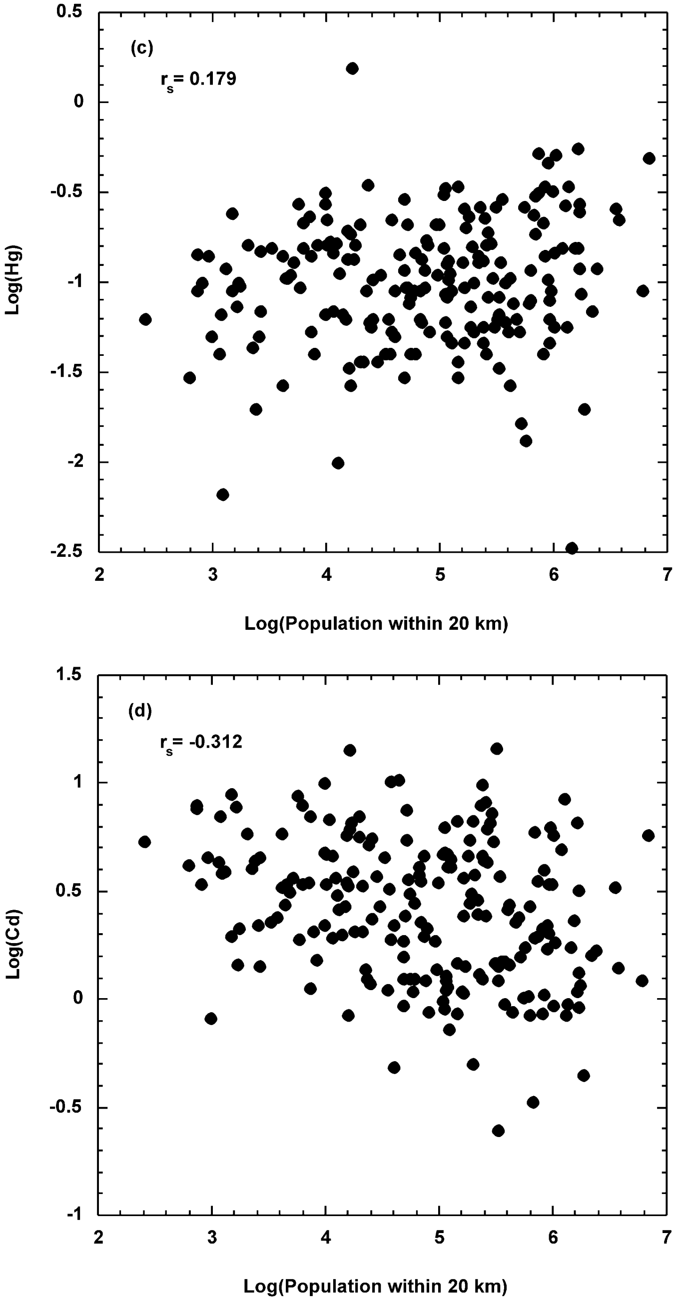


Fig. 2. (*continued*).

Table 4
Spearman correlation coefficients between molluscan concentrations and population for 1990 (n = number of sites = 214)[a]

Chemical	Spearman correlation coefficient (r_s)
ΣPCB	0.623
Pb	0.598
ΣBT	0.585
ΣChlordane	0.598
ΣDDT	0.553
ΣHMW	0.520
Zn(oyster)	0.486
Ag (mussel)	0.458
ΣPAH	0.473
Cu (mussel)	0.288
ΣLMW	0.252
Cu (oyster)	0.193
Cr[b]	0.181
Hg	0.179
Zn(mussel)	0.174
ΣDieldrin	0.153
Ag (oyster)	0.044
As	−0.024
Ni	−0.107
Se	−0.140
Cd	−0.312

[a] Population is the number of people living within 20 km of each site as per 1990 US Census. (With so large an "n", any coefficient with an absolute value greater than 0.135 is "significant" at the 0.05 level.)

[b] Cr concentrations in correlation are site means for 1996–1998.

are associated with rural areas. Less than 20% of the Se and Ni highs and less than 10% of the As and Cd highs are at the more populated sites.

10. Naturally high concentrations

For each element there is a minimum concentration, possibly site-specific, that is set entirely by natural factors. Separating this baseline level from the measured concentration would leave the remainder attributable to human activity. Scanes and Roach (1999) made this separation with data on elemental concentrations in oysters from 20 estuaries in New South Wales, Australia. On the basis of both cluster and principal component analyses, the concentrations in oysters separated into two groups; those from estuaries in rural watersheds and from those in populated areas. The mean concentrations in rural areas were taken to be baseline or natural levels. No such clear-cut separation exists in the NS&T national data (even when mussels and oysters are considered separately). Chemicals with concentrations that correlated with population generally correlated with one another. But for all the trace elements, except lead, there is such considerable overlap of concentrations between rural and urban sites that statistical tests fail to identify what could be called a "natural" group.

In fact, rather than revealing contamination, the high concentrations for As, Cd, Ni, and Se may represent entirely natural conditions. It is certainly possible for human activities in rural areas to cause mollusks at isolated sites to have high concentrations. One example is high mercury in oysters from parts of Matagorda Bay, Texas, attributable to extreme concentrations of mercury in sediments from past discharges of a chlor-alkali plant (Holmes, 1977). Another is the high concentrations of several chemicals in Choctawhatchee Bay, Florida in the vicinity of Eglin Air Force Base. However, when groups of sites in particular regions rather than individual sites show high concentrations, a natural source is more likely. Some natural sources for these four elements have been identified. The high As concentrations in the Southeast are probably due to phosphate deposits in that region (Vallette-Silver, Riedel, Crecilius, Windom, Smith, & Dolvin, 1999). The serpentine rocks of the Northwest (USGS, 1981) are a source of both the Cr and Ni that are found at high levels in mollusks along that coast. Serpentine outcrops in the Mid-Atlantic were a source of the rock itself and of its associated Cr in colonial America (Mountford, 1997) and may be the source for the high Ni in oysters of that region. As noted earlier by Goldberg et al. (1983) and by Farrington, Goldberg, Risebrough, Martin, and Bowen (1983), the high Cd concentrations in mussels of Northern California may reflect the facts that the coastal water in that region upwells from the deep Pacific and that deep ocean waters are naturally enriched in Cd relative to surface waters. What remain unknown are the reasons for high Cd and Se concentrations in oysters at sites in Louisiana and Texas.

11. Great Lake sites

Mollusks sampled at three sites in Hawaii, three in Puerto Rico, one in the Florida Keys, and all sites on the Great Lakes may not accumulate chemicals to extents that are comparable with the *Mytilus* sp. or the *C. virginica* collected at all other sites. For organic contaminants, species differences may be unimportant, because marine mussels and oysters collected from common sites show common concentrations. For metals, though, it is important to be aware that an indication in Table 3 of high concentrations at these sites could be a species effect. Except for Se at one, no chemicals are high at the Puerto Rico sites. The Florida Keys site is high for As and Ni, the first of which is typically high in the southeast. There is a human influence at the Hawaiian sites as evident by high concentrations of some organic compounds. Nonetheless, the high metals at the Hawaiian sites may reflect a species-effect, especially for Cu because it is high at all three.

In 1992, NOAA began to sample zebra mussels at freshwater sites. Concentration percentiles for 25 zebra mussel sites are in Table 5 and comparisons with marine "highs" are in Table 6. One of those 25 sites is in a NOAA Estuarine Reserve in the tidal portion of the Hudson River, the rest are on the Great Lakes. They are listed separately from marine mollusks because, while the 85th percentiles of concentrations

Table 5
15th, 50th, and 85th percentiles concentrations among zebra mussels collected at Great Lakes 25 sites in 1992–1998

Chemical	15th pctl.	50th pctl.	85th pctl.
Ag	0.019 μg g^{-1}(dry)	0.04 μg g^{-1}(dry)	0.066 μg g^{-1}(dry)
As	6.2	7.7	9.8
Cd	2.6	5.6	8.4
Cu	14	23	34
Cr	4.2	7.1	10
Hg	0.041	0.058	0.082
Ni	18	26	33
Se	3.5	5.2	7.9
Pb	0.77	2.0	4.6
Zn	98	120	160
ΣChlordane	4.2 ng g^{-1}(dry)	9.9 ng g^{-1}(dry)	18 ng g^{-1}(dry)
ΣDieldrin	4.2	8.4	14
ΣDDT	9.8	30	96
ΣPCB	110	370	1200
ΣLMW	160	230	340
ΣHMW	110	290	470
ΣPAH	270	580	650
ΣBT	35 ng of Sn g^{-1}(dry)	100 ng of Sn g^{-1}(dry)	240 ng of Sn g^{-1}(dry)

at 214 marine sites is a convenient basis for comparison, the highs in Table 6 may have more to do with the chemistry of freshwater and the physiology of zebra mussels than differences in metal concentrations between Great Lakes and coastal marine waters. Ni is high at all the zebra mussel sites, Cu at 22, Se at 21, Cr at 17, and Cd at 11. Conversely Ag is high at no site and Zn at only one. These differences are also evident in Table 4 where the 15th percentile concentrations for Cu, Ni, and Se and the medians for Cr and Cd in zebra mussels are close to or exceed the 85th percentile concentrations among marine sites. These strong tendencies are probably more reflective of the mussel species than the environment

Roditi and Fisher (1999) have measured rates and efficiencies for metal uptake of Ag, Cd, Cr, and Se by zebra mussels and compared them with blue mussels. Consistent with a propensity for high concentrations among zebra mussels, the efficiencies of uptake for Cd and Cr from both water and food are higher for zebra mussels than for blue mussels. There are no high Ag concentrations among zebra mussels and, compared to blue mussels, its assimilation from food was found to be relatively low, while its efficiency of uptake from the dissolved phase was slightly greater. The high selenium concentrations are inconsistent with the results of Roditi and Fisher (1999) who found no difference between species with regard to uptake from water but that blue mussels had higher efficiencies of uptake from food. There are no comparative data for accumulation of the other metals, but it is clear that species differences preclude extrapolating the observed differences in metal

Table 6
Site/chemical combinations (x) where concentration in Zebra mussel (Great Lakes and Hudson River sites; Sp = Species) was "high" in at least half the years sampled since 1990 (years)[a]

Site	General location	Specific location	ST	Sp	Years	Ag	As	Cd	Cu	Cr	Hg	Ni	Pb	Se	Zn	tDield	tCdane	tDDT	tPCB	tLMW	tHMW	tPAH	tBT
GBBS	Green Bay	Bayshore Park	WI	dp	4				x			x							x				
LMMB	Lake Michigan	Milwaukee Bay	WI	dp	4				x	x		x		x	x	x	x	x	x		x	x	x
LMNC	Lake Michigan	North Chicago	IL	dp	4			x	x	x		x	x	x		x		x	x				
LMCB	Lake Michigan	Calumet Breakwater	IN	dp	3			x	x	x		x	x	x									
LMHB	Lake Michigan	Holland Breakwater	MI	dp	4				x	x		x				x	x	x	x				
LMMU	Lake Michigan	Muskegon Breakwater	MI	dp	4				x	x		x		x		x	x	x	x				
TBLL	Traverse Bay	Leelanau Park	MI	dp	2			x	x	x		x		x		x							
LHTB	Lake Huron	Thunder Bay	MI	dp	2			x	x			x		x		x							
SBSR	Saginaw Bay	Saginaw River	MI	dp	5				x			x		x					x				
SBSP	Saginaw Bay	Sand Point	MI	dp	5				x			x		x									
LHBR	Lake Huron	Black River Canal	MI	dp	5			x	x	x		x		x		x							
LSAB	Lake St. Clair	Anchor Bay	MI	dp	4				x	x		x		x		x				x			
LESP	Lake Erie	Stony Point	MI	dp	4			x	x	x		x	x	x					x		x		x
LERB	Lake Erie	Reno Beach	OH	dp	4			x	x	x		x		x		x							
SBPP	South Bass Island	Peach Orchard Point	OH	dp	4			x	x	x		x		x		x			x				
LEOW	Lake Erie	Old Woman Creek	OH	dp	3				x			x	x			x				x			
LELR	Lake Erie	Lorain	OH	dp	3			x	x	x		x		x		x			x	x			
LEAB	Lake Erie	Ashtabula	OH	dp	3			x	x	x		x		x					x	x			
LEDK	Lake Erie	Dunkirk	NY	dp	3				x			x		x					x				
NRNF	Niagara River	Niagara Falls	NY	dp	2				x		x	x	x	x					x	x	x	x	x
LOOC	Lake Ontario	Olcott	NY	dp	3			x		x		x		x		x			x				
LORC	Lake Ontario	Rochester	NY	dp	3					x		x		x									
LOOS	Lake Ontario	Oswego	NY	dp	3				x	x		x		x									
LOCV	Lake Ontario	Cape Vincent	NY	dp	3							x		x						x			
HRCI	Hudson River	Cruger Island	NY	dp	2				x	x		x	x						x	x			

[a] Site codes in first column of use when reviewing data on Internet (see text).

concentrations between zebra- and marine-mussels to differences in metal availability in freshwaters and coastal marine waters.

12. Conclusions

While the purpose of the Mussel Watch Project is to monitor temporal trends in coastal contamination, the data also define concentration distributions and give some indication of when highs are due to nature rather than man. These distributions are a basis that any program can use to judge whether its observed concentrations are at all unusual. However, except for synthetic organic chemicals, PAHs and lead, it should never be assumed that a high concentration is necessarily unnatural and subject to human control. Tissue concentrations in urban areas of two groups of organic chemicals, PAHs and PCBs, are in a range that may be causing lysosomal alterations the mollusks.

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