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Trends in chemical concentrations in mussels and oysters collected along the US coast: Update to 2003

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Abstract

With data from the annual analyses of mussels and oysters collected in 1986–1993 from sites located throughout the coastal United States [O'Connor, T.P., 1996. Trends in chemical concentrations in mussels and oysters collected along the US coast from 1986 to 1993. Mar. Environ. Res. 41, 183–200] showed decreasing trends, on a national scale, for chemicals whose use has been banned or has greatly decreased and that concentrations of most other chemicals were neither increasing nor decreasing. With data through 2003 those conclusions still apply. National median concentrations of synthetic organic chemicals and cadmium continue to decrease. The added data show that concentrations of lindane and high molecular weight PAHs are also decreasing on a national scale. For metals other than cadmium and zinc (in mussels), the added data reveal trends at more sites than in 1993 but no additional national trends. However, the longer time series has revealed several local and regional trends. Published by Elsevier Ltd.

Keywords: Mussel Watch; Temporal trends; Chlorinated hydrocarbons; Butyltins; PAHs; Trace elements

1. Introduction

Following upon the US Mussel Watch Program sponsored by the Environmental Protection Agency in 1976–1979 (Goldberg et al., 1978), in 1986 the Mussel Watch Project within the National Status and Trends (NS&T) Program of the National Oceanic and

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Atmospheric Administration began to monitor spatial distributions and temporal trends of contaminant concentrations in coastal and estuarine regions of the United States. O'Connor (1996, 1998) summarized temporal trends through 1993 and 1996 as showing that concentrations of most metals and polycyclic aromatic hydrocarbons in mollusks displayed no overall national trends while concentrations of chlorinated hydrocarbons, butyltins, and cadmium revealed national decreasing trends. In this paper with data through 2003 we show that these conclusions remain almost the same. Chemicals with concentrations on a decreasing trend in 1993 remain on that trend. Concentrations of two chemicals, lindane (gamma isomer of hexachlorocyclohexane) and high molecular weight PAHs, not on a decrease through 1993 are now showing a decrease. No chemical concentrations are increasing on a national scale. Cadmium remains the only metal showing a national trend. The additional data reveal some trends in concentrations of metals within geographic groups of sites.

1.1. Sites

Mussels or oysters are collected at 292 sites distributed throughout the coastal United States and along the Great Lakes (Fig. 1). The sites are, on average, 20 km apart in estuaries and embayments and 100 km apart along open coastlines. They were selected with the intention of collecting samples that are representative of their surroundings. Small scale patches of contamination and known points of waste discharge have been avoided. Lauenstein et al. (1997) provided detailed descriptions of each Mussel Watch sampling site and an electronic file of site locations accompanies the chemical data file that is available upon request from the authors or on the Internet at http://nsandt.noaa.gov.

The numbers of sites occupied in each of the first 18 years of Mussel Watch were 145, 147, 174, 185, 214, 192, 195, 169, 150, 166, 129, 129, 136, 136, 134, 121, 133, and 114 in 1986–2003, respectively. A decrease began in 1994, with 1993 being a transition year, because sites began to be sampled on a biennial basis with collections at about half the sites in one year and the other half in the next. Additions of sites generally provided more spatial resolution or spatial coverage with expansions to Puerto Rico and the Great Lakes.
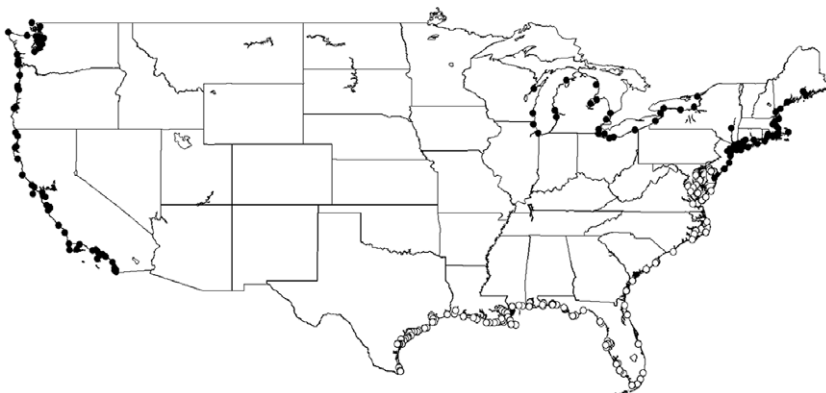

Fig. 1. NOAA Mussel Watch Sites; mussel sites (closed circles) oyster sites (open circles). There are sites (not shown) in Hawaii, Alaska, and in Puerto Rico.

1.2. Chemicals measured

The elements and compounds measured in the Mussel Watch Program and for which trends have been calculated are listed in Table 1. All are possible contaminants in the sense that their concentrations in the environment have been altered by human activities (Nriagu, 1989). The mere existence of the chlorinated organic compounds and butyltins indicates human activity. Polycyclic aromatic hydrocarbons (PAHs) are similar to metals in the sense that they occur naturally. They are found in fossil fuels such as coal and oil and are produced during the combustion of organic matter. Their environmental presence is also attributable to humans because they are released in the use and transportation of petroleum products and from a multitude of human activities, such as burning coal and wood or incinerating waste. The division of PAHs between the low-molecular weight (LMW) and high-molecular (HMW) weight compounds generally corresponds to the division between compounds entering the environment from petroleum and those with a pyrogenic source. Almost all the chemicals in Table 1 are also on the list of 127 priority pollutants created by the United States Environmental Protection Agency in the late 1970s (Keith and Teillard, 1979).

Six PAH compounds – acenaphthylene, 1,6,7-trimethylnaphthalene, benzo[*b*]- and benzo[*k*]fluoranthene, indeno[1,2,3-*cd*]pyrene, and benzo[*ghi*]perylene – were not measured until 1988. In this paper, trends for PAHs will be examined from 1988 to 2003. Also in 1988, the program began measuring the 18 PCB congeners listed in Table 1, while in 1986 and 1987 PCBs were quantified to level of chlorination. To maintain consistency over the years for trend analysis, the post-1987 PCB congener concentrations have been converted to total PCB concentrations, total PCB = 2 × (sum of 18 congeners) (O'Connor,

Table 1
Chemicals for which trends have been calculated

Elements
As, Cd, Cu, Hg, Ni, Pb, Se, Zn

Organic Compounds
Concentrations of individual compounds have been aggregated into groups.
tPCB = twice the sum of concentrations of 18 congeners PCB8, PCB18, PCB28, PCB44, PCB52, PCB66, PCB101, PCB105, PCB118, PCB128, PCB138, PCB153, PCB170, PCB180, PCB187, PCB195, PCB206, and PCB209
tDDT = 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
tCld = sum of concentrations of four compounds *alpha*-chlordane, *trans*-nonachlor, heptachlor, heptachlorepoxide
tDld = sum of concentrations of two compounds aldrin and dieldrin
Lind = the concentration of lindane, the gamma isomer of hexachlorocyclohexane
tBT = sum of concentrations of parent compound and metabolites monobutyltin, dibutyltin, and tributyltin [concentrations in terms of tin]
LMW = sum of concentrations of 12 2–3 ring polycyclic aromatic hydrocarbons naphthalene, 2-methylnaphthalene, 1-methylnaphthalene, biphenyl, 2,6-dimethylnaphthalene, acenaphthene, acenaphthylene, 1,6,7-trimethylnaphthalene, fluorene, phenanthrene, anthracene, 1-methylphenanthrene
HMW = sum of concentrations of 12 4–5 ring polycyclic aromatic hydrocarbons 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
tPAH = sum of concentrations of LMW and HMW compounds

1996). Lastly, systematic monitoring of tributyltin and its metabolites di- and mono-butyl tin did not begin until 1989, so trends can only be examined from that year.

The Mussel Watch Program includes chemicals beyond those listed in Table 1.

Some were so frequently not detected that no calculations of trends were made: Sn (35% frequency of no detectable concentration); endrin (71%), mirex (49%); and hexachlorobenzene (39%). Two trace elements, Sb and Tl, were so infrequently detected in 1986 and 1987 that they were not measured after 1987. Silver concentrations have not been used because of unresolved analytical problems with data since 2000. Chromium concentrations have not been used because extraction methods used with mollusk tissue fail to extract all of the element (O'Connor, 2002). Aluminum, Fe, and Mn data have not been used because their natural concentrations exceed any anthropogenic influence.

Mollusks have been analyzed for endosulfan, chlorpyrifos, chlorinated benzenes, pentachloroanisole, isomers of HCH (other than lindane), and forms of chlordane other than those listed in Table 1. However, the time series for those chemicals is less than the six-year minimum required for trend analysis.

1.3. Species collected

Since no single species of mollusk is common to all coasts, it has been necessary to collect eight 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* and *D. bugensis* at sites in the Great Lakes.

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 (O'Connor, 1994). Similarly, the two species of zebra mussel collected at Great Lakes sites are consider equivalent with regard to chemical concentration.

2. Methods

Mollusks are collected at marine sites in the months of November–March with each site visited within 30 days of a prescribed date. In the Great Lakes, collections are made in late August or early September. From 1986 to 1991, six separate composite samples of 20 oysters or 30 mussels were collected at each site. The small size of *Dreissena* spp. required that composite samples from the Great Lake sites consisted of up to 100 individuals. 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 preferred size ranges are 5–8 cm for mussels, 7–10 cm for *C. virginica*, 2.5–5 cm for *O. sandvicensis*. The size range of *Dreissena* spp. is limited and sample consisted of clumps of individuals. Mollusks are not shucked in the field. They are separated when found to be adhering to one another and scrubbed with a nylon or natural fiber brush to remove adhering detritus. Cleaned samples are then packed in ice or dry ice and shipped to the laboratory. Composite samples are prepared by homogenizing the soft parts of 30 mussels or 20 oysters.

All samples from the Gulf of Mexico Coast collected from 1986 to 1999 have been analyzed by scientists from the Geochemical and Environmental Research Group or the Department of Oceanography at Texas A&M University (TAMU) in College Station, TX. For 1986–1988, samples from California were analyzed by Scientific Applications International Corporation in LaJolla, CA. For 1986–1994 all other West Coast samples and all East Coast samples were analyzed by scientists from the Battelle Laboratories in Duxbury, MA and Sequim, WA. For 1994–1999 all samples from all coasts were analyzed by the Geochemical and Environmental Research Group of the Department of Oceanography at Texas A&M University in College Station, TX. Since 2000, all samples have been analyzed by TDI-Brooks in College Station, TX. Detailed descriptions of sampling and analytical protocols are available (Lauenstein and Cantillo, 1993a,b,c,d, 1998).

All laboratories, in all years, have used cold vapor atomic absorption for the analysis of Hg. TDI-Brooks International, Inc who has been performing the analyses since 2000 uses a modified version of EPA method 245.6. At Battelle, analyses for As, Cu, Se, and Zn have been made by X-ray fluorescence and those for Cd, Ni, and Pb by graphite furnace atomic absorption (GFAA). Beginning in 1992, inductively-coupled plasma with mass spectroscopy detection (ICP/MS) was used for Cd, Cr, Ni, and Pb. The analytical instruments used by SAIC and TAMU were GFAA for As, Cd, Ni, Pb, and Se; flame atomic absorption (FAA) for Zn; and both GFAA and FAA for Cu, depending on the concentration present. Since 2000 As and Se have been analyzed by atomic fluorescence spectrometry; As has also been analyzed by ICP-optical emission spectrometry (ICP/OEP). ICP/OEP is also used to determine Cu, Ni and Zn while ICP/MS is used for Cd, Ni and Pb.

Digestion procedures prior to elemental analysis varied to some extent among laboratories and years. Prior to 1990, Battelle used concentrated nitric and perchloric acids in a Teflon digestion bomb in a conventional oven. Since 1991, samples have been digested in nitric acid only, and heating has been by microwave irradiation. SAIC used conventional ovens and nitric acid. The Texas A&M Laboratory used nitric and perchloric acids with heating in a conventional oven. From 1991 to 1999, only nitric acid was used. The current analytical laboratory sequentially adds nitric acid and hydrogen peroxide to Teflon bombs to achieve sample dissolution of all but Hg samples.

Organic chemical extractions were similar among laboratories. After addition of internal standards and anhydrous sodium sulfate, the tissue was extracted three times with dichloromethane using a tissumizer. After concentration by solvent evaporation and exchange of dichloromethane with hexane, the tissue extract was fractionated by alumina:silica chromatography. The aromatic fraction eluted from the column with 1:1 pentane:dichloromethane was further purified by removing lipids through chromatography on a Sephadex column. Since 1988, this purification has been accomplished via an HPLC procedure developed by Krahn et al. (1988). Organic sample extraction since 2000 remains similar to that of previous years but Hydromatrix® is used to dry tissue samples and dichloromethane is the only solvent used in accelerated solvent extractor cells. Extracts

are purified using alumina/silica gel chromatography followed by high performance liquid chromatography.

Purified extracts were chromatographed on 30-m DB-5 fused silica capillary columns. Since 2000, a second column (DB-17HT) has also been used for chlorinated hydrocarbon confirmation. Chlorinated hydrocarbon concentrations were quantified using an electron capture detector (GC-ECD) by all laboratories in all years. For polycyclic aromatic hydrocarbons all analyses by SAIC in 1986–1988 (California and Hawaiian coasts) and those by Battelle in 1986 (East and Northwest Coasts) employed flame ionization detection (GC-FID). Battelle used mass spectrometry detection (GC-MSD) in full-scan mode in 1987. In all other cases the PAH analysis has been by GC-MSD in the selected ion mode (SIM).

For butyltin analyses, anhydrous homogenized tissue was extracted with troplone and hexane (East and West Coast samples) or troplone and dichloromethane (Gulf Coast samples). Current methods use either hexane or dichloromethane. The extracts underwent Grignard reactions by addition of hexylmagnesium bromide (Gulf Coast samples) and *n*-pentyl magnesium bromide (East and West Coast samples). The Gulf Coast method is currently in use for samples from all coasts. Florisil/silica gel or silica alumina column chromatography were used to separate the analytes. Quantization of Sn was performed using GC-FID. Since 2000 quantification of Sn has been performed using GC-flame photometric detection.

All laboratories annually participate in annual intercalibration exercise where common samples are analyzed by them, by other laboratories, and by a lead laboratory for the exercise. The lead laboratory for trace element intercalibrations has been the National Research Council of Canada and that for trace organic analysis has been the National Institute of Standards and Technology. Results from intercalibration exercises are published (Schantz et al., 2005; Willie, 2005).

2.1. Lowest detected concentrations and treatment of non-detected concentrations

For this analysis all analytical results showing no usable signal were treated as concentrations of zero. This can complicate trend detection because zeros will always be the lowest possible concentration, while non-zeros in other years could possibly indicate greater analytical sensitivity rather than higher concentrations. For the trace elements and the aggregated organic compounds for which trends have been calculated, zeros are infrequent and do not affect trend analysis. Mean concentrations for As, Cd, Cu, Hg, Ni, Pb, Se, and Zn among 2757 site/year combinations for 1986–2003 were undetected in 10 or fewer samples. Among 2782 mean concentrations of the aggregate groups of chlorinated organic compounds there are 481,153, 46, 15 and 10 zeros for lindane, tDieldrin, tChlordane, tPCB, and tDDT, respectively. Among 2488 mean tPAH concentrations since 1988, 22 are zero. Among 2289 tBT values since 1989, 261 are zero. Table 2 lists the lowest non-zero mean concentrations for each element and compound group.

3. Results

A trend is a statistically significant correlation between contaminant concentration and year. In this paper, Spearman correlations based on rankings of concentrations are used, because, unlike parametric correlations, they are free of assumptions about concentrations at a site being normally distributed with a common variance among sites. Moreover,

Table 2
Lowest non-zero concentration, per chemical, in data sets used to identify trends

As	0.91 (μg g^{-1})
Cd	0.050
Cu	0.50
Hg	0.010
Ni	0.081
Pb	0.020
Se	0.18
Zn	7.3
tCld	0.010 (ng g^{-1})
tDld	0.040
tDDT	0.16
Lind	0.02
tPCB	0.36
LMW	7.3
HMW	1.5
tPAH	7.3
tBT	0.27 ng of Sn g^{-1}

All concentrations given on a dry-weight basis.

because they are based on ranks rather than absolute concentrations, correlations are unaffected by particularly large between-year differences. Such differences, if they appear near the beginning or end of a temporal sequence, can force a parametric correlation to appear significant when, in fact, there is no consistent trend among the data as a whole. All calculations were performed with the SYSTAT package (SYSTAT, 2004). Calculations were made for all sites with at least six years of data. Through 2003 there are 246 sites with six or more years of data. The need for at least six years limited trend analysis through 1993 to 154 sites (O'Connor, 1996).

Table 3 lists all the 246 sites for which there are 6–15 years of data and indicates chemicals for which there is a 95% probability (Zar, 1984) of a correlation between concentration and year showing an increasing (I) or decreasing (D) trend. The sites are listed in a clockwise geographic sequence that moves from Maine through Alaska to Hawaii and through the Great Lakes. For LMW, HMW, and tPAHs, the numbers of years are two less than shown in the table (trends were calculated for the 221 sites with at least six years of tPAH data). Similarly, since tBT only began to be monitored in 1989 there are only 207 sites with six or more years of data.

The most common result in Table 3 is a lack of trends. Among the 4182 combinations of 17 chemicals at 246 sites there are only 108 increases and 830 decreases at the 95% level of confidence. Given a 5% probability of random data showing trends, there could be 105 increases and 105 decreases that are not real trends. Table 4 summarizing trends per chemical shows that most of the decreases are among the organic chemical groups, that very few increases were found for the organic chemicals, and that trends in metal concentrations were more or less evenly split between increases and decreases. Table 4 also lists the corresponding results for the period of 1986–1993.

Annual data have been aggregated to annual national medians in Table 5. In that aggregation only data from the commonly sampled species (*M. edulis*, *M. californianus*, and *C. virginica*) have been used and, because of species differences, concentrations in mussels have been separated from those in oysters for Cu and Zn (O'Connor, 1996). Since

Table 3
Site-by-site and per chemical listing of increasing or decreasing trends in chemical concentrations in mollusks

Site	General location	Specific location	State	sp	yrs	Lind	tCld	tDDT	tDld	tPCB	tTBT	LMW	HMW	tPAH	As	Cd	Cu	Hg	Ni	Pb	Se	Zn
PBPI	Penobscot Bay	Pickering Island	ME	me	12														D			
PBSI	Penobscot Bay	Sears Island	ME	me	14		D	D	D	D	d		D	D			D			d		
MSSP	Merriconeag Snd.	Stover Point	ME	me	12			d							i	I	d					
CAKP	Cape Arundel	Kennebunkport	ME	me	10	D	D				d				i							
CAGH	Cape Ann	Gap Head	MA	me	13		d		d								d			D	i	D
SHFP	Salem Harbor	Folger Point	MA	me	11		D	D		D	D	d	D	D	I	I				D		
MBNB	Massachusetts Bay	Nahant Bay	MA	me	8		D	D			D				I							
BHDI	Boston Harbor	Deer Island	MA	me	13	d	D	d	d		D	D		d			d	D		d		d
BHDB	Boston Harbor	Dorchester Bay	MA	me	12	D	D		D		D				i	D		i		D		D
BHHB	Boston Harbor	Hingham Bay	MA	me	13		D				D				I						i	
BHBI	Boston Harbor	Brewster Island	MA	me	14		D	D	d		D										I	
MBNR	Massachusetts Bay	North River	MA	me	7	D	D	D	D	D											d	
DBCI	Duxbury Bay	Clarks Island	MA	me	11	D	D	D	D	D							D					
CCNH	Cape Cod	Nauset Harbor	MA	me	10	D					d											
BBNI	Buzzards Bay	Naushon Island	MA	me	8						D					i		I				
BBWF	Buzzards Bay	West Falmouth	MA	me	8						D				d			I				I
BBCC	Buzzards Bay	Cape Cod Canal	MA	me	9						D				d	i		I	I	I		I
BBAR	Buzzards Bay	Angelica Rock	MA	me	13		D	d			D	d	D	d		D	d	I				I
BBRH	Buzzards Bay	Round Hill	MA	me	10		D		D		d						D	I			i	
BBGN	Buzzards Bay	Goosebury Neck	MA	me	13		d	D	D		d						D					
NBDI	Narragansett Bay	Dyer Island	RI	me	12												D		d	D		D
NBPI	Narragansett Bay	Patience Island	RI	me	11		D	D	D		d							d		d		D
NBDU	Narragansett Bay	Dutch Island	RI	me	13		D	D	D		d										I	
BIBI	Block Island Sound	Block Island	RI	me	13	D	D		D	D	D										I	
LICR	Long Island Sound	Connecticut River	CT	me	11		D	D			d		D	d								
LINH	Long Island Sound	New Haven	CT	me	13	D	D	D	D	D	d							d		d		
LIHR	Long Island Sound	Housatonic River	CT	me	12		D		D		D											
LISI	Long Island Sound	Sheffield Island	CT	me	10		D	D		D												

Code	Region	Site	State	Type	N	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
LIMR	Long Island Sound	Mamaroneck	NY	me	12	D	D	D		D	D	D	D	D					d			
LITN	Long Island Sound	Throgs Neck	NY	me	13	D	D	D		D	D		D	D				D		d	D	
LIHH	Long Island Sound	Hempstead Harbor	NY	me	13	D	D	D		D	D		D	D			D	D		d	D	
LIHU	Long Island Sound	Huntington Harbor	NY	me	12	D	D	D	D	D	D							d				
LIPJ	Long Island Sound	Port Jefferson	NY	me	13	D	D	D	D	D	D	D	D	D	I		I					
LIGB	Long Island	Gardiners Bay	NY	me	9		D								i	i						I
MBTH	Moriches Bay	Tuthill Point	NY	me	13		D	D		D		i								I	I	I
LIJI	Long Island	Jones Inlet	NY	me	7	D	D	d	d													
HRJB	Hudson/Raritan Est.	Jamaica Bay	NY	me	14		D	D		D			D					D		D		I
HRUB	Hudson/Raritan Est.	Upper Bay	NY	me	12		D	D	d	D	d		D				D	I	D	d	D	I
HRLB	Hudson/Raritan Est.	Lower Bay	NY	me	12	d	D	D		D	d							D		D		d
HRRB	Hudson/Raritan Est.	Raritan Bay	NY	me	8		d			D	d	D	I					D	D	D	d	D
NYSH	New York Bight	Sandy Hook	NJ	me	12		D	D		D	D		D	D		D						I
NYLB	New York Bight	Long Branch	NJ	me	12		D	D	D	D						i	D					i
NYSR	New York Bight	Shark River	NJ	me	14		D	D		D	d					I	D					
BIBL	Barnegat Inlet	Barnegat Light	NJ	me	10		D	D	D	D										I		I
AIAC	Absecon Inlet	Atlantic City	NJ	me	10		d	d				d										
DBCM	Delaware Bay	Cape May	NJ	me	10	D		d		d												D
DBFE	Delaware Bay	False Egg Is Point	NJ	cv	11	D	D	D											D			
DBBD	Delaware Bay	Ben Davis Point	NJ	cv	11	D	D	D	D	D						i			D			
DBAP	Delaware Bay	Arnolds Point	NJ	cv	10	D	D			D										I		
DBKI	Delaware Bay	Kelly Island	DE	cv	12	D	D		D	D			d						D		d	D
DBCH	Delaware Bay	Cape Henlopen	DE	me	10	D	D	D			D		D	d								
CBBO	Chesapeake Bay	Bodkin Point	MD	cv	8	D	D	D		D	D		D			d						
CBMP	Chesapeake Bay	Mountain Point Bar	MD	cv	11	D	D	D	D	D			D	D						i		d
CBHP	Chesapeake Bay	Hackett Point Bar	MD	cv	12	D	D	D		D			D	d					D			
CBCP	Chesapeake Bay	Choptank River	MD	cv	10	D	D	D			D		D						D			
CBHG	Chesapeake Bay	Hog Point	MD	cv	12	D	D	D	D	D	D										d	
PRSP	Potomac River	Swan Point	MD	cv	7		D	D														

Table 3 (*continued*)

Site	General location	Specific location	State	sp	yrs	Lind	tCld	tDDT	tDld	tPCB	tTBT	LMW	HMW	tPAH	As	Cd	Cu	Hg	Ni	Pb	Se	Zn
PRMC	Potomac River	Mattox Creek	VA	cv	7	d	D	D	D												d	
PRRP	Potomac River	Ragged Point	VA	cv	9			d												D		
RRRR	Rappahannock River	Ross Rock	VA	cv	10		D	D	D										I			d
CBDP	Chesapeake Bay	Dandy Point	VA	cv	13		D			D	D	D		D			I	d				i
CBJR	Chesapeake Bay	James River	VA	cv	8			d							d						D	
CBCC	Chesapeake Bay	Cape Charles	VA	cv	13	d	D		D		D											
CBCI	Chincoteague Bay	Chincoteague Inlet	VA	cv	13		D	D	D	D		D		d					I	i		
QIUB	Quinby Inlet	Upshur Bay	VA	cv	13		D	D	d	D					I		D			I	I	D
RSJC	Roanoke Sound	John Creek	NC	cv	13		D	D					d	d		D				D		
PSWB	Pamlico Sound	Wysocking Bay	NC	cv	13		D						i			D						
PSPR	Pamlico Sound	Pungo River	NC	cv	9	D										D	D			D		
PSNR	Pamlico Sound	Neuse River	NC	cv	10				d	i												
PSCH	Pamlico Sound	Cape Hatteras	NC	cv	9						D											
BIPI	Beaufort Inlet	Pivers Island	NC	cv	9		d	d			D			d			I					
CFBI	Cape Fear	Battery Island	NC	cv	14			d	D		d				d							
WBLB	Winyah Bay	Lower Bay	SC	cv	10	i														I		
SRNB	Santee River	North Bay	SC	cv	10															I		
CHFJ	Charleston Harbor	Fort Johnson	SC	cv	13	d	D	D	d		d									I		
CHSF	Charleston Harbor	Shutes Folly Island	SC	cv	12	D	D	D	D	D	D									i		
SRTI	Savannah River Est.	Tybee Island	GA	cv	13		D	D	D		d							I				
SSSI	Sapelo Sound	Sapelo Island	GA	cv	13		D	D	D													
ARWI	Altamaha River	Wolfe Island	GA	cv	10	i	d	d														
SJCB	St. Johns River	Chicopit Bay	FL	cv	13	D	D			D	D					i				d		d
MRCB	Matanzas River	Crescent Beach	FL	cv	13		d	d			D					d				i		
IRSR	Indian River	Sebastian River	FL	cv	11					I	D							I	I			I
NMML	North Miami	Maule Lake	FL	cv	11		D				D											
BBGC	Biscayne Bay	Goulds Canal	FL	cv	9											d						
FBJB	Florida Bay	Joe Bay	FL	cv	7				D							d	D					
FBFO	Florida Bay	Flamingo	FL	cv	7				D		D	D					I					
BHKF	Bahia Honda	Key FLorida	FL	cs	7				d							d		i				
PRBB	Puerto Rico	Bahia De Boqueron	PR	cr	7																	
PRBM	Puerto Rico	Bahia Montalva	PR	cr	6																	
PRBJ	Puerto Rico	Bahia De Jobos	PR	cr	7																	I

EVFU	Everglades	Faka Union Bay	FL	cv	13		d	d		D		D			d		d			d
RBHC	Rookery Bay	Henderson Creek	FL	cv	13		d	d		D				d		D				
NBNB	Naples Bay	Naples Bay	FL	cv	13	D	d	d	D	D				D	D	I	D			i
CBFM	Charlotte Harbor	Fort Meyers	FL	cv	10	D	D	D	d	d	d		d		d	D		D		
CBBI	Charlotte Harbor	Bird Island	FL	cv	15	D	D	D	D	D				D	D	I	D			I
TBCB	Tampa Bay	Cockroach Bay	FL	cv	12	D	D	d	D			D	d	i						i
TBHB	Tampa Bay	Hillsborough Bay	FL	cv	9													D		
TBKA	Tampa Bay	Knight Airport	FL	cv	10	D	D			D	D			I						
TBOT	Tampa Bay	Old Tampa Bay	FL	cv	10	D		D		D							D			
TBPB	Tampa Bay	Papys Bayou	FL	cv	13	D	D			D	D						D	D	D	D
TBMK	Tampa Bay	Mullet Key Bayou	FL	cv	13	D	D	D	d								d			
TBNP	Tampa Bay	Navarez Park	FL	cv	10	D	D			D	D	D					d			
CKBP	Cedar Key	Black Point	FL	cv	15	D	D	D				d			D	d				
AESP	Apalachee Bay	Spring Creek	FL	cv	11	D	D			D	D		D	D						
APCP	Apalachicola Bay	Cat Point Bar	FL	cv	13	D	D	D	D	D	D	D	D	D				I		
APDB	Apalachicola Bay	Dry Bar	FL	cv	13	D	D	D	D	D			D	d		D		i		
SAWB	St. Andrews Bay	Watson Bayou	FL	cv	13	d	D	D	D	D		D			i				d	
PCMP	Panama City	Municipal Pier	FL	cv	9		D	D			D	d		d	I				D	
PCLO	Panama City	Little Oyster Bar	FL	cv	11	D	D	d			D	D	D	d	D	I	d		i	D
CBSR	Choctawhatchee Bay	Off Santa Rosa	FL	cv	14		D	D	d		d	d	D	D				D	d	d
CBPP	Choctawhatchee Bay	Postil Point	FL	cv	14	D	D			D				d						
CBJB	Choctawhatchee Bay	Joes Bayou	FL	cv	11	D	D			D	d	D		D	d		i	d	d	
PBSP	Pensacola Bay	Sabine Point	FL	cv	6	D								I						d
PBIB	Pensacola Bay	Indian Bayou	FL	cv	11	D	D	D	D	D	d		D		d					
PBPH	Pensacola Bay	Public Harbor	FL	cv	11	d	D	D			D							d		
MBHI	Mobile Bay	Hollingers Is. Chan.	AL	cv	10	D							d	D						
MBCP	Mobile Bay	Cedar Point Reef	AL	cv	13	D	D	D	D	D	D		D	D						
MSPB	Mississippi Sound	Pascagoula Bay	MS	cv	13		D	D	D	D			D	d	D		d		d	

Table 3 (continued)

Site	General location	Specific location	State	sp	yrs	Lind	tCld	tDDT	tDld	tPCB	tTBT	LMW	HMW	tPAH	As	Cd	Cu	Hg	Ni	Pb	Se	Zn
MSBB	Mississippi Sound	Biloxi Bay	MS	cv	11	d	D	D	D	D	D						I						I
MSPC	Mississippi Sound	Pass Christian	MS	cv	12	D				d	D										i		
LPNO	Lake Pontchartrain	New Orleans	LA	cv	6						D											D	
LBGO	Lake Borgne	Gulf Outlet	LA	cv	7																		
LBMP	Lake Borgne	Malheureux Point	LA	cv	14	D	D	D	D	D													
BSBG	Breton Sound	Bay Gardene	LA	cv	13	D	D		D	d	D		d		D	i	I	I	I				I
BSSI	Breton Sound	Sable Island	LA	cv	13	D	d		d	d	D	d	D	D	d								
MRPL	Mississippi River	Pass A Loutre	LA	cv	9		D				D		D	D	d								
MRTP	Mississippi River	Tiger Pass	LA	cv	8		D											i					
BBMB	Barataria Bay	Middle Bank	LA	cv	13	D	D	d	d	d	D	d	D	D					I				D
BBSD	Barataria Bay	Bayou Saint Denis	LA	cv	12	D	D	D	d														
TBLF	Terrebonne Bay	Lake Felicity	LA	cv	13		D	D	D	d	d	d	D	d									d
TBLB	Terrebonne Bay	Lake Barre	LA	cv	12	d	D	D	D	D	D	d	D	D									
CLCL	Caillou Lake	Caillou Lake	LA	cv	14	D	D	D	D	d	d							I					
ABOB	Atchafalaya Bay	Oyster Bayou	LA	cv	13	D	D	D	D	D	D		D			d	d	d					d
VBSP	Vermilion Bay	Southwest Pass	LA	cv	14	D	D	D	D	D	D		d										
JHJH	Joseph Hrb. Bayou	Joseph Hrb. Bayou	LA	cv	13	D	D	D	D	D	D												
CLSJ	Calcasieu Lake	St. Johns Island	LA	cv	14	D	D	D	D	d													
CLLC	Calcasieu Lake	Lake Charles	LA	cv	12		D												i				
SLBB	Sabine Lake	Blue Buck Point	TX	cv	14	D	D	D	D		d												
GBHR	Galveston Bay	Hanna Reef	TX	cv	13	D	D		D		d		d						I				
GBSC	Galveston Bay	Ship Channel	TX	cv	10	D		D			D					i	D	D					
GBYC	Galveston Bay	Yacht Club	TX	cv	13	D					d					I	I	D					
GBTD	Galveston Bay	Todds Dump	TX	cv	14	D									i								
GBOB	Galveston Bay	Offatts Bayou	TX	cv	12			D			D						I						i
GBCR	Galveston Bay	Confederate Reef	TX	cv	13	D	D	D	d	D	D		d										
BRFS	Brazos River	Freeport Surfside	TX	cv	11																		
BRCL	Brazos River	Cedar Lakes	TX	cv	11																		
MBEM	Matagorda Bay	East Matagorda	TX	cv	13	D	D	D	D	d	D							D				D	D

Code	Bay	Location	State	Type	n																	
MBTP	Matagorda Bay	Tres Palacios Bay	TX	cv	13	D	D		d	D	D		d		D	D						
MBCB	Matagorda Bay	Carancahua Bay	TX	cv	9		D		D											D		
MBLR	Matagorda Bay	Lavaca River Mouth	TX	cv	12	d	d	D		d	D		D	d						I		
MBGP	Matagorda Bay	Gallinipper Point	TX	cv	13	D	D	d	D	d	D		d									
ESBD	Espiritu Santo	Bill Days Reef	TX	cv	9		d	d			D				D	D						
ESSP	Espiritu Santo	South Pass Reef	TX	cv	9	d	d		d		D											
SAMP	San Antonio Bay	Mosquito Point	TX	cv	8				d												D	
SAPP	San Antonio Bay	Panther Point Reef	TX	cv	11	d	D		D		d											
MBAR	Mesquite Bay	Ayres Reef	TX	cv	14	d	D	D	D	D										i		
ABLR	Aransas Bay	Long Reef	TX	cv	14	D	D	D	D			D		d	D					i		d
CBCR	Copano Bay	Copano Reef	TX	cv	12		D	D			D							D			D	
CCIC	Corpus Christi	Ingleside Cove	TX	cv	6	D	D															
CCNB	Corpus Christi	Nueces Bay	TX	cv	13	D	D	D	D	D		d								I		
LMAC	Lower Laguna Madre	Arroyo Colorado	TX	cv	6					D	d											
LMPI	Lower Laguna Madre	Port Isabel	TX	cv	6	D														I	D	
LMSB	Lower Laguna Madre	South Bay	TX	cv	13		D	D	D	D	D			d	D	D	D				D	d
IBNJ	Imperial Beach	North Jetty	CA	mc	13		D	D	D						D				D		D	D
SDCB	San Diego Bay	Coronado Bridge	CA	me	11										D	d	D			d		D
PLLH	Point Loma	Lighthouse	CA	mc	12			D		D	D								i			
SDHI	San Diego Bay	Harbor Island	CA	me	13		D	d			D		D	D		D				d	D	D
MBVB	Mission Bay	Ventura Bridge	CA	me	13		D	D		D	D			D	D		D			D	D	D
LJLJ	La Jolla	Point La Jolla	CA	mc	13	D	D					I	I	d			D			D		
OSBJ	Oceanside	Beach Jetty	CA	me	13					d	D	I		I							d	
SCBR	Santa Catalina Is.	Bird Rock	CA	mc	11	D									D						D	D
NBWJ	Newport Beach	Wedge Jetty	CA	mc	12						D			d						D		
ABWJ	Anaheim Bay	West Jetty	CA	mc	13		D	D	D		D			d	D	d	D			D		D
LBBW	Long Beach	Breakwater	CA	me	9						d		d	D	d							D
SPFP	San Pedro Harbor	Fishing Pier	CA	me	12		D	D		D	D	I			D		d			D		d
PVRP	Palos Verdes	Royal Palms Park	CA	mc	13	D	D	d	D	d					D		D		D	D	D	d
RBMJ	Redondo Beach	Municipal Jetty	CA	mc	9						d			d			D					D

Table 3 (*continued*)

Site	General location	Specific location	State	sp	yrs	Lind	tCld	tDDT	tDld	tPCB	tTBT	LMW	HMW	tPAH	As	Cd	Cu	Hg	Ni	Pb	Se	Zn
MDSJ	Marina Del Rey	South Jetty	CA	me	13	D					D						d	D		D		D
TBSM	Las Tunas Beach	Santa Monica Bay	CA	mc	9			d									D					
PDPD	Point Dume	Point Dume	CA	mc	13	D	D		D		D				D			D	d	d	D	D
SCFP	Santa Cruz Island	Fraser Point	CA	mc	11										D				d			
SBSB	Point S. Barbara	Point Santa Barbara	CA	mc	13	I	d				D				D							
PCPC	Point Conception	Point Conception	CA	mc	12										D						D	
SLSL	San Luis Obispo Bay	Point San Luis	CA	mc	13	D	D	D	D		D						D		D	D		d
SSSS	San Simeon Point	San Simeon Point	CA	mc	12	d		d			d				D		I					
PGLP	Pacific Grove	Lovers Point	CA	mc	14	d	D				d				D							
MBML	Monterey Bay	Moss Landing	CA	mc	12						D									D		d
MBES	Monterey Bay	Elkhorn Slough	CA	mc	8					D	d					i						
MBSC	Monterey Bay	Point Santa Cruz	CA	mc	14						D		I		D							d
SFSM	San Francisco Bay	San Mateo Bridge	CA	me	14		d		D	D	D	i					D	d	d			
SFDB	San Francisco Bay	Dumbarton Bridge	CA	me	14	d				D	D						D		d	i		
SFEM	San Francisco Bay	Emeryville	CA	me	13					d	D						d	D				
TBSR	Tomales Bay	Spengers Residence	CA	me	14												D					
BBBE	Bodega Bay	Entrance	CA	mc	14	d			D		d	I		i								
PALH	Point Arena	Lighthouse	CA	mc	14			D	D		D	I		i			I		i			
PDSC	Point Delgada	Shelter Cove	CA	mc	14	D			D			i	I	I			I		i			
HMBJ	Eureka	Humboldt Bay Jetty	CA	mc	13						d	i	i	I								
EUSB	Eureka	Samoa Bridge	CA	mc	9						D											
SGSG	Crescent	Point St. George	CA	mc	13	d		D				i										
CBCH	Coos Bay	Coos Head	OR	mc	13	D	d			D	D						D					
CBRP	Coos Bay	Russell Point	OR	me	13	D	D	D	d		D										I	
YBOP	Yaquina Bay	Oneatta Point	OR	me	14	d	D	D	D	D	D											
YHYH	Yaquina Bay	Yaquina Head	OR	mc	6					d								d				
YHFC	Yaquina Head	Foggerty Creek	OR	mc	7																	

Code	Site	Location	State	Type	N																			
TBHP	Tillamook Bay	Hobsonville Point	OR	me	14			d																D
CRSJ	Columbia River	South Jetty	OR	me	12																			d
WBNA	Willapa Bay	Nahcotta	WA	me	8												d		d					
GHWJ	Grays Harbor	Westport Jetty	WA	mc	13			d					D				I		d					D
JFCF	Juan de Fuca Strait	Cape FLattery	WA	mc	12	D							d			D								D
PSPA	Puget Sound	Port Angeles	WA	me	10				D				D					I				i		
PSPT	Puget Sound	Port Townsend	WA	me	9							D	D											I
PSHC	Puget Sound	Hood Canal	WA	me	10								D					i				I		I
SSBI	South Puget Sound	Budd Inlet	WA	me	13	D	d				d	i	d						D					D
CBTP	Commencement Bay	Tahlequah Point	WA	me	13								D					D				i		D
PSSS	Puget Sound	South Seattle	WA	me	9	D							D	I	I	I								D
EBDH	Elliott Bay	Duwamish Head	WA	me	6	d			D				D											
EBFR	Elliott Bay	Four-Mile Rock	WA	me	13													D		D		D		
SIWP	Sinclair Inlet	Waterman Point	WA	me	13								D					d	D			d	D	
WIPP	Whidbey Island	Possession Point	WA	me	13	D	d																	D
PSEH	Puget Sound	Everett Harbor	WA	me	7								d									I		
BBSM	Bellingham Bay	Squalicum Marina	WA	me	13	d	d						d	d						d		i		
PRPR	Point Roberts	Point Roberts	WA	me	12													d			i			
PVMC	Port Valdez	Mineral Creek Flats	AK	me	12															i				d
UISB	Unakwit Inlet	Siwash Bay	AK	me	12																			
HHKL	Honolulu Harbor	Keehi Lagoon	HI	os	11												d		i					
BPBP	Barbers Point	Barbers Point	HI	os	10				D								D		I			D		
GBBS	Green Bay	Bayshore Park	WI	ds	7				D	D	D	D				D			i					
LMMB	Lake Michigan	Milwaukee Bay	WI	ds	7				D	D	D	D	D						I			I		
LMNC	Lake Michigan	North Chicago	IL	ds	7				d		d						D		I			D		
LMHB	Lake Michigan	Holland Breakwater	MI	ds	7				D	D		D	d											
LMMU	Lake Michigan	Muskegon Breakwater	MI	ds	7				D	D		D									d			D
SBSR	Saginaw Bay	Saginaw River	MI	ds	7						D				d									
SBSP	Saginaw Bay	Sand Point	MI	ds	7					D	D				D	d	D						D	
LHBR	Lake Huron	Black River Canal	MI	ds	6				d		D					d						D		D
LSAB	Lake St. Clair	Anchor Bay	MI	ds	6				D								I	D	I					

Table 3 (*continued*)

Site	General location	Specific location	State	sp	yrs	Lind	tCld	tDDT	tDld	tPCB	tTBT	LMW	HMW	tPAH	As	Cd	Cu	Hg	Ni	Pb	Se	Zn
LESP	Lake Erie	Stony Point	MI	ds	7											D	I	d			D	
LERB	Lake Erie	Reno Beach	OH	ds	6				D							D			D		D	
SBPP	South Bass Island	Peach Orchard Point	OH	ds	7				d		d		D	d	d			d	D		D	D
LEOW	Lake Erie	Old Woman Creek	OH	ds	6	d	d	D	d													
LELR	Lake Erie	Lorain	OH	ds	6		D	D	d							D		d			D	
LEAB	Lake Erie	Ashtabula	OH	ds	6		d		d								I				D	
LEDK	Lake Erie	Dunkirk	NY	ds	6	D								D				D				
LOOC	Lake Ontario	Olcott	NY	ds	6	D	D	D								D			D			D
LORC	Lake Ontario	Roschester	NY	ds	6				D										D			
LOOS	Lake Ontario	Oswego	NY	ds	6				D						I							
LOCV	Lake Ontario	Cape Vincent	NY	ds	6	D	d	D					D			D				I		

"Site" is a code that is useful when accessing the data electronically. Species are me (*Mytilus edulis*), mc (*M. californianus*), cv (*Crassostrea virginica*), cr (*Crassostrea rhizophorae*), os (*Ostrea sandvicensis*), or ds (*Dreissena spp.*). "Yrs" is the number of years a site was sampled between 1986 and 2003. Chemicals are lindane (Lind), total chlordane (tCld), total DDT (tDDT), total Dieldrin (tDld), total PCB (tPCB), total butyltins (tBT), LMW, HMW, and tPAH all as defined in Table 1 and trace elements. Trends are shown as Increasing (I) or Decreasing (D) at the 95% level of confidence or n lower case "i" and "d" at the 90% level for Spearman correlations between concentration and year.

Table 4
Numbers of sites per chemical where there is a 95% level of confidence of increasing or decreasing trend in concentration in mollusks for the period 1986–1993 and 1983–2003

Chem	1986–1993		1986–2003	
	Inc.	Dec.	Inc.	Dec.
As	5	14	14	17
Cd	3	20	4	39
Cu	5	17	17	23
Hg	7	8	12	22
Ni	4	5	9	14
Pb	7	8	11	30
Se	2	11	16	22
Zn	6	7	6	31
tCld	0	43	1	133
tDDT	0	24	0	108
tDld	0	19	0	77
Lind			0	70
tPCB	0	26	1	75
tPAH	2	3	5	23
LMW			6	10
HMW			6	38
tBT	0	11	0	98

For the earlier and later periods there were 154 and 246 sites, respectively, with six or more years of data. The Spearman correlation coefficients required for 95% confidence with 6–15 years of data are decrease from 0.886 to 0.525 (Zar, 1984).

sites have been visited biennially since 1994, concentrations from pairs of years have been combined to form a single median for the pair (no site contributes data to both years in a pair). The 1986–1993 concentrations in Tables 5a and 5b are slightly different from those in the corresponding table in O'Connor (1996) because that table listed geometric means

Table 5a
Annual median concentrations (µg/g-dry) of metals in mollusks (*M. edulis*, *M. californianus*, and *C. virginica*) for sites sampled in at least six years from 1986 to 2003

Year	As	Cd	Cu(oys)	Cu(mus)	Hg	Ni	Pb	Se	Zn(oys)	Zn(mus)
1986	9.5	3.2	100	9.8	0.11	2.1	0.75	2.6	1700	140
1987	9.0	2.9	110	10.0	0.10	1.9	0.85	2.6	1800	130
1988	8.8	2.9	140	9.8	0.11	1.8	0.74	3.0	2400	130
1989	8.2	2.6	120	10.0	0.12	1.7	0.63	2.3	2300	120
1990	9.5	2.8	140	8.4	0.093	1.7	0.78	2.4	2400	130
1991	9.2	2.4	120	8.8	0.11	2.1	0.78	2.6	2000	130
1992	9.3	2.1	120	8.6	0.10	2.1	0.73	2.6	2200	120
1993	8.4	2.5	120	8.4	0.11	1.6	0.82	2.5	2100	120
1994/1995	9.4	2.1	120	8.6	0.10	1.7	0.90	3.0	2100	120
1996/1997	9.6	2.4	130	10.0	0.10	1.9	0.80	2.9	2000	120
1998/1999	8.1	2.5	130	9.6	0.11	2.2	0.98	2.6	2300	110
2000/2001	9.1	2.1	91	7.9	0.10	1.8	0.63	2.5	1600	110
2002/2003	8.8	2.1	120	8.0	0.10	1.8	0.77	2.4	2000	110
r_s(1986–2003)	−0.154	**−0.806**	−0.049	−0.555	−0.047	−0.105	−0.181	−0.038	−0.143	**−0.918**
r_s(1986–1993)	−0.192	**−0.898**	0.396	**−0.74**	−0.089	−0.282	0.036	−0.304	0.287	**−0.73**

Last two rows are Spearman rank correlation coefficients of concentration versus year for 1986–1993 and 1986–2002/2003, respectively, with bold type indicating significance at the 95% level.

Table 5b

Annual median concentrations (ng/g-dry with tBT in terms of ng of Sn g^{-1} dry weight) of organic compounds or compound groups in mollusks (*M. edulis*, *M. californianus*, and *C. virginica*) for sites sampled in at least six years from 1986 to 2003

Year	Lind	tCld	tDld	tDDT	tPCB	LMW	HMW	tPAH	tBT
1986	1.2	15	5.8	37	140				
1987	1.4	19	8.2	41	120				
1988	1.1	14	4.5	38	130	180	160	500	
1989	0.69	14	4.2	37	110	130	160	300	110
1990	2.5	13	3.4	30	100	110	110	250	73
1991	0.62	5.6	3.0	18	52	89	120	220	66
1992	0.84	5.9	3.6	24	68	120	110	240	47
1993	0.63	7.2	3.6	25	61	89	160	260	18
1994/1995	0.74	5.9	2.8	26	72	100	110	220	22
1996/1997	0.49	7.4	3.7	22	89	99	100	210	18
1998/1999	0.42	6.3	3.1	20	87	95	100	210	22
2000/2001	0.24	3.9	1.6	12	52	62	70	140	18
2002/2003	0.26	3.3	1.5	13	50	120	70	220	13
r_s(1986–2003)	**−0.852**	**−0.835**	**−0.828**	**−0.867**	**−0.798**	−0.464	**−0.909**	**−0.755**	**−0.892**
r_s(1986–1993)	−0.595	**−0.874**	**−0.778**	**−0.802**	**−0.905**	−0.714	−0.600	−0.6	**−1**

Last two rows are Spearman rank correlation coefficients of concentration versus year for 1986–1993 and 1986–2002/2003, respectively, with bold type indicating significance at the 95% level.

rather than medians. Concentrations are approximately log normally distributed on a national scale but using medians rather than geometric means forces no assumptions about the underlying distribution (O'Connor, 2002).

4. Discussion

4.1. Comparison between shorter and longer time series

For two reasons, site-by-site trends are much more common with data through 2003 than through 1993. First, there are more sites with 6 or more years of data. Secondly, the median number of years per site through 2003 was 12 while through 1993 the median was 8. The larger number of years decreased the Spearman correlation coefficient required for 95% confidence that a trend exists (0.591 for $n = 12$ versus 0.738 for $n = 8$) meaning that weaker trends become evident over longer time periods.

To a very large extent, results with annual national medians through 2003 were identical to those through 1993. Groups of organic chemicals with decreasing median concentrations through 1993 continued to show decrease through 2003. The national decrease in cadmium through 1993 is maintained through 2003 as is the decrease in median concentrations of zinc in mussels only. The decreasing trend in copper in mussels that appeared in data through 1993 is no longer sustained. The shift from geometric means to medians erased the trend in As through 1993 but left other trends unaffected.

Two chemicals showed trends in national medians in the longer but not the shorter time series. Lindane concentrations were not used in the earlier analysis because 27% of the concentrations in that period are zeros (no detectable signal). Detection has improved to the point where the longer dataset contains only 17% zeros. This is more than any other chemical for which trends were calculated but was deemed sufficiently low to proceed.

Total PAHs are the one chemical group showing a trend in the longer time series that was not evident earlier. The trend appears to be due to the HMW component of tPAH since HMW concentrations are trending down while the LMW concentrations are not showing a trend. The lack of tPAH trends in the earlier data may be a consequence of the tPAH time series being two years shorter than that for other chemicals. As explained above, in 1986 and 1987 the full suite of tPAH chemicals had not been measured.

4.2. National trends

All the monitored chlorinated hydrocarbons have been banned or heavily restricted for use in the United States. DDT was banned in 1972; PCB began being phased out in 1971 and further uses were banned in 1976; dieldrin (and aldrin) stopped being used in 1975. The agricultural use of chlordane was banned in 1983 and its use as a termiticide ended in 1988 (Shigenaka, 1990). After restrictions imposed in1983, lindane was used primarily for treating certain seeds and ornamental trees and as a dip for fleas and lice on pets, and livestock. Since 1993 further restrictions eliminated its use in flea dips and limited the types of seeds that could be treated (EPA, 2002). In 1988, the US Organotin Anti-Fouling Paint Control Act outlawed tributyltin application to boats of less than 25 m in length. It was not unexpected that concentrations of these chemicals would be decreasing through 1993. The only possibly unanticipated result after 10 more years is that all continue to show statistically significant decreasing trends. Increases are certainly not likely but at some point the concentrations must become too low for further decreases to be discerned above random noise in annual medians.

Among the three non-synthetic chemicals with decreasing trends in national median concentrations, PAHs, Cd, and Zn (in mussels), only the reason for Cd is clear. Fig. 2 based on data from Kelly et al. (2005) shows the dramatic decrease in Cd consumption that has occurred in the US since the mid 1960s when Cd was ubiquitously used as an anticorrosive coating on metals. The same figure shows no corresponding decrease in Zn consumption to parallel the observation here that Zn is decreasing at sites where mussels are sampled. Similarly, there is no data on the generation of high molecular weight PAHs that parallel the decreasing trend seen in mollusks. It is possible that decreases are the result of controls on Zn and PAH emissions from controlled sources such as industrial and municipal point sources. However, all the metals in Table 1 are also subject to controls yet show no trends.

4.3. Local and regional trends

Five percent of the calculated trends at 246 individual sites (Table 3) can be just random sequences of concentrations appearing as trends. There is no rigorous way to sort random sequences from real trends but when trends appear at geographically contiguous sites the likelihood improves that the trends are real. The great many decreasing trends for synthetic organic compounds at individual sites are just local examples of a national trend For metals and PAHs, however, the relatively few trends at individual sites may be of local, rather than national, interest. Table 6 lists areas where chemicals have shown common trends at three or more contiguous sites or two such trends if the third is a trend at the 90% level of confidence (see Table 3).

Three of the trends in Table 6 also appeared in results through 1993. However, there were four decreasing trends through 1993 were not sustained with more data; As in
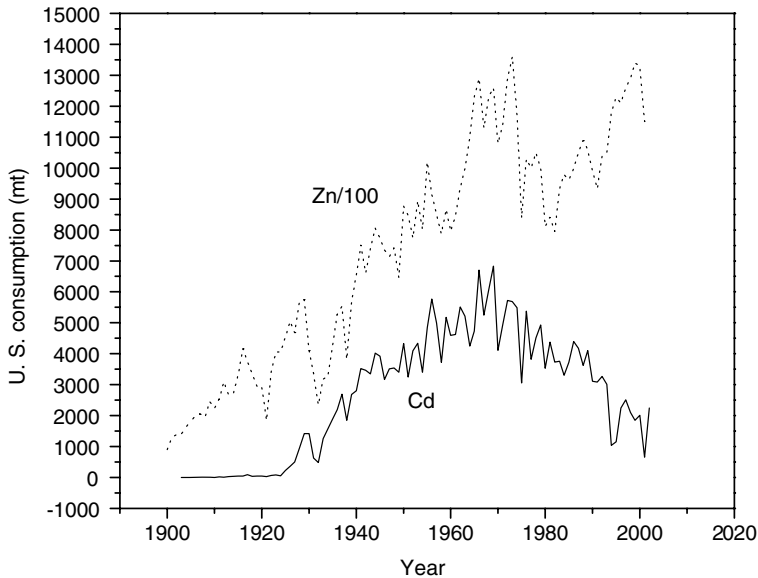
Fig. 2. Consumption of cadmium and zinc (mt per year) in the US (Kelly et al., 2005). Zn consumption has been divided by 100 for clarity.

southwest Florida, Se in northern California, and Cu and Hg at sites in the inner portion of Puget Sound, WA. In no case has a decreasing trend become an increase but the concentrations have simply failed to continue downward.

While we have no basis for identifying the natural concentrations of any chemical at any site, the existence of trends strongly implies a human influence. This follows from the fact that the myriad of natural factors affecting chemical concentrations in mollusks such as local mineralogy, salinity, runoff, and temperature (O'Connor, 1996) are unlikely to annually vary in a manner that would impose a linear trend on chemical concentrations in tissues.

Twenty-three of the 28 local or regional trends (not counting trends in LMW or HMW PAHs that are also trends in total PAH) in Table 6 are decreases. We cannot explain each one but local interests may take some satisfaction in that human actions may have contributed to the decreases. By the same token, the five increasing trends point to some local human activity. Among the increases, it should be pointed out that the tPAH concentrations at sites in Central California (increasing due to the LMW component) and the Pb concentrations at sites in South Carolina are near or less than national median concentrations for those chemicals (O'Connor, 2002). The increasing Se and Hg concentrations at sites in Buzzards Bay, MA, the increasing As at sites near Panama City, FL, and the increasing Se concentrations at sites in Puget Sound WA, do include concentrations above the national 85th percentile (O'Connor, 2002).

4.4. Other reports of trends

There are other reports of trends from periodic sampling of mollusks. Claisse and Beliaeff (2000) calculated temporal trends in chemical concentrations measured between 1979

Table 6
Areas where contiguous sites are showing common trends for metal or PAH concentrations

Chemical	Trend	Area
Arsenic	Increasing	NW Florida
	Decreasing	Southern to Central California
Cadmium	Decreasing	**Hudson/Raritan estuary and northern New Jersey**
	Decreasing	Pamlico Sound, NC
	Decreasing	Southwest Florida
	Decreasing	**Southern California**
	Decreasing	**San Francisco Bay, CA**
Copper	Decreasing	Buzzards Bay, MA
	Decreasing	**Western Long Island Sound**
	Decreasing	Southern California
Mercury	Increasing	Buzzards Bay
	Decreasing	Hudson/Raritan Estuary
	Decreasing	Southwest Florida
	Decreasing	Southern California
Lead	Decreasing	Hudson/Raritan Estuary
	Increasing	South Carolina
	Decreasing	**Southern California**
Selenium	Increasing	Buzzards Bay, MA
	Increasing	Outside Puget Sound, WA
	Decreasing	Lake Erie, OH
Zinc	Decreasing	Southern California
	Decreasing	Northwest Pacific Coast
	Decreasing	Puget Sound, WA
tPAH	Decreasing	Western Long Island Sound
	Decreasing	Northwest Florida–Louisiana
	Increasing	Central California
LMW	Decreasing	NW Florida
	Increasing	Central California
HMW	Decreasing	Western Long Island Sound
	Decreasing	Northern Chesapeake Bay, MD
	Decreasing	Northwest Florida to Louisiana

Areas are listed where three or more contiguous sites show trends at the 95% confidence level (upper case D or I in Table 3) or two or more at the 95% level if nearby sites are showing trends at the 90% level (lower case d or i). Areas that showed the same trends through 1993 are in bold type.

and 1999 in mussels and oysters at 102 sites along the coast of France. Mollusks were annually collected four times per year. The calculation of trends was based on a multiple linear regression that included the influence of season on concentration. Their results were similar to those listed in Table 4 in that; (1) for PCBs (only congener 153 measured), tDDT, lindane, and tPAH (six LMW- and 10 HMW-compounds) trends that did exist were only decreases (no increases), (2) Cd and Pb trends were overwhelmingly decreases with few increases and (3) Hg, Zn, and Cu decreases were about twice as common as increases. The only difference in the overall trends observed in France for 1979–1999 and those reported here for the US for 1986–2003 is that Pb decreased in France while not in the US. This is probably due the fact that the phasing-out of Pb in gasoline that

began in the US in the 1970s did not begin in France until the late 1980s. The latter decrease appears in the French mollusks collected over the 1979–1999 period but the corresponding decrease in the US was effectively over before The NOAA Mussel Watch began in 1986. Lauenstein et al. (1990) compared trace element results of the US Mussel Watch Program sponsored by the EPA in 1976–1978 (Goldberg et al., 1983) with results from NOAA Mussel Watch in 1986–1988. Mean concentrations over 1978–1980 were compared with 1986–1988 means at the 50 sites common to both programs. For Pb, 36 of the 50 comparisons showed the higher concentration in the 1970s; a result with less than a 1% chance of having occurred randomly. For Cd, Cu, Ni and Zn the number of differences between the two decades among the 50 sites did not indicate significant differences. Lauenstein (1995) secured archived samples collected in 1977 by the earlier Mussel Watch Program and arranged for the simultaneous analysis of organic chemicals in those samples along with samples collected by the NOAA program in 1992. This was done both because the earlier program did not measure the same organic chemicals and because analytical methods had changed considerably since the 1970s. At the 51 sites with data from 1977 and 1992, the 1977 samples had the higher concentrations of tDDT, tPCB, tChlordane, and tPAH 45, 38, 46, and 39 times, respectively, all of which are statistically significant.

Sediments cores that can be radiologically dated have provided information on long term trends in chemical contamination. While an exhaustive review of this topic is not appropriate in this paper, the results reported here on DDT and PCBs in mollusks are generally consistent with core data showing decreases that began in the 1970s. Valette-Silver (1993) summarized work showing maxima in DDT concentrations off southern California in the 1970s followed by a steady decrease; similarly PCB (arochlor) concentrations have decreased in sediments of the Hudson estuary (New York) since 1970. Venkatesan et al. (1999) reported maxima concentrations of DDT and PCB at depths corresponding to the 1970s for two cores taken from San Francisco Bay with subsequent decreasing concentrations. Santschi et al. (2001) also found 1970s maxima followed by decreases for PCBs in the Mississippi River Delta and Galveston Bay and for DDT in Galveston Bay and Tampa Bay. Santschi et al. (2001), however, did not observe a decrease in PCBs in their Tampa Bay core or a DDT decrease in the core from the Mississippi Delta. Van Metre and Mahler (2005) recently summarized their results from cores taken from 38 lakes in the United States. Testing for statistically significant linear trends most often led to the null result but for PCBs and DDT all the trends that were significant were decreases. Moreover, the median percentage differences over the 39 cores between concentrations of DDT and PCBs in period after 1990 were 69% and 49%, respectively, less than concentrations in core sections corresponding to 1965–1975. Van Metre and Mahler (2005), on the other hand, found as many increasing as decreasing trends for chlordane and a median increase of only 5% between post 1990 and 1965–1975 concentrations. They attributed their results with chlordane to the fact that chlordane use in the US was banned relatively recently compared to DDT and PCB.

While PAHs are not synthetic and their use cannot be banned, concentration trends would be expected to reflect changes in fossil fuel use and emission controls. The national median concentration of tPAH is decreasing in mollusks but there are many fewer tPAH decreases at individual Mussel Watch sites relative to synthetic organic compounds. PAH profiles in sediment cores have shown decreases, increases, and no discernable trends. Van Metre and Mahler (2005) usually did not find statistically significant linear trends in PAHs but, unlike the cases with synthetic compounds, the trends that were found were predom-

inately increases. They also noted that the median of comparisons between post 1990 concentrations and 1965–1975 concentrations was a 41% increase. Kannan et al. (2005) did not look for linear trends in 10 cores from Lake Michigan but, with one exception, did find lower PAH concentrations in the more recently deposited sediments than in core sections from the early 1990s, late 1980s, or 1970s. Santschi et al. (2001) found PAH maxima at depth in cores from Galveston Bay and Tampa Bay but with PAH concentrations at the top of the core from the Mississippi Delta only slightly less the maximum reached in the 1960s. Pereira et al. (1999) found PAH maxima in cores from San Francisco Bay at depths corresponding to the 1950s with only small concentration changes since then with, perhaps, a decrease to the core top. PAH concentration peaks have been attributed to the common use of coal in private homes but PAH changes since the 1980s can be decreases reflecting emission controls or increases reflecting growing populations and growing use of automobiles. Van Metre and Mahler (2005) attributed their increasing trends of PAH concentration to increasing urbanization and Kannan et al. (2005) attributed their one observation of increasing PAH to growing population pressure. There is a correlation between PAH concentrations in mollusks (O'Connor, 2002) and numbers of people residing within 20 km of Mussel Watch sites. Nonetheless the sites with increasing PAH trends reported here (Table 3) are not sites near population centers while some of the decreasing trends are in the general vicinity of so urban an area as New York City.

There are, to our knowledge, no reports of increasing trends since the 1980s of the trace elements discussed here (As, Cd, Cd, Hg, Ni, Pb, Se, or Zn) in sediment cores. The review of Valette-Silver (1993) showed that Pb, Zn, and Cu have decreased off Southern California as have Pb and Cu in the Hudson-Raritan Estuary but the decreases are relative to peaks in the 1960s with concentrations more or less constant since the 1980s. Santschi et al. (2001) report similar profiles for Pb, Cu, and Zn in cores from the Mississippi Delta, Galveston Bay and Tampa Bay. Cadmium concentrations also showed no trend since about 1980 in cores from two of the locations but were on a decreasing trend even since 1980 in the Mississippi Delta core. Hornberger et al. (1999) also found maxima for Pb, Cu, and Zn in cores from San Francisco Bay with slight indications of decreases since about 1980. Cadmium trends in San Francisco Bay cores are of particular interest because that is a location with decreasing trends in Cd concentrations in mussels (Table 3). van Geen and Luoma (1999) studied Cd profiles cores from one of the same cores examined by Hornberger et al. (1999). Consistent with the Cd trend in mollusks and with national use of Cd (Fig. 2), bulk concentrations of Cd decreased monotonically from about 40 cm downcore (about 1970) to the core top (1992). However, the Cd/Ca ratio in foraminifera in the cores did not show any trend over those same depths. Since the Cd/Ca ratio in foraminifera should be controlled by concentrations of dissolved Cd and Ca as foram tests grow, the authors interpreted their results to indicate no recent trend in dissolved Cd concentrations. However, since molluscan concentrations of chemicals are not controlled solely by dissolved concentrations, the observed decreasing trends in mussels (Table 3) and in concentrations in bulk sediments could both be reflecting decreased inputs of total Cd to San Francisco Bay.

5. Conclusion

The result of annually monitoring chemical concentrations in mussels and oysters in the coastal United States through 1993 was that, on a national scale, chlorinated hydrocarbons

and tributyltin, chemicals banned or greatly restricted for further use in the United States, were decreasing in concentration. Ten more years of monitoring have not changed that conclusion. Annual median concentrations in Cd and Zn (in mussels) also continued to show decreasing trends. National trends also appeared for lindane and total PAH that were not apparent earlier.

Relative to the large number of sites showing trends for concentrations of synthetic organic chemicals, there were relatively few sites with trends for metals and PAHs. Nonetheless more trends were identified among local or regional contiguous groups of sites than were found through 1993. Since most trends are decreasing and decreasing trends cannot persist indefinitely, it is not clear whether longer time series will yield significantly more information.

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