RESULTS FROM THE NOAA NATIONAL STATUS AND TRENDS PROGRAM ON DISTRIBUTION AND EFFECTS OF CHEMICAL CONTAMINATION IN THE COASTAL AND ESTUARINE UNITED STATES

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Abstract. The NOAA National Status and Trends (NS&T) Program has been monitoring chemical contamination in fish livers, and surface sediments since 1984 and in molluscan tissue and sediments since 1986. Data from fine-grained sediment at 175 sites are used to describe the spatial distribution of contamination throughout the coastal and estuarine United States. Highest levels are generally found in, and considered representative of, urban areas. It should be noted, however, that these levels are not as high as have been found near discharge pipes or in isolated industrial areas through other monitoring efforts. Dramatic biological responses, such as liver tumors in fish or apparently toxic contaminant levels in sediment, are found infrequently. Subtle biological changes, especially those that affect reproductive ability, are being sought. Data from three annual collections of mollusks have been used to identify early signals of temporal trends in contamination at NS&T sites.

Introduction

To assess effects of human activity on the quality of coastal and estuarine areas throughout the United States, the National Oceanic and Atmospheric Administration (NOAA) created the National Status and Trends (NS&T) Program to monitor spatial and temporal trends of chemical contamination and biological responses to that contamination. Beginning in 1984 and 1986, annual chemical analyses for trace elements and organic contaminants (Table I) were made on tissues of benthic fish and bivalve mollusks, respectively, collected from a network of sites (Figure 1) around the United States. On a less frequent basis, sediments from those sites were also collected and analyzed. Livers of fish from the annual collections undergo histologic examination for evidence of contaminant-induced damage and analyzed for contaminant-induced enzymatic activity. Each year, in selected areas, more intense monitoring is conducted to determine whether contamination has diminished the ability of adult fishes to reproduce, or of larval fishes to survive. Through bioassays, the presence of toxicity in sediments and water is measured. This paper reports some results on the spatial and temporal distribution of contamination and biological responses.

Methods

The samples and data are gathered through two programs. The Benthic Surveillance

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TABLE I

Chemicals and related parameters measured in the National Status and Trends Program

DDT and its metabolites	Polyaromatic hydrocarbons[b]	Major elements	
o,p'-DDD	*2-ring*	Al	Aluminum
p,p'-DDD	Biphenyl	Fe	Iron
o,p'-DDE	Naphthalene	Mn	Manganese
p,p'-DDE	1-Methylnaphthalene	Si	Silicon
o,p'-DDT	2-Methylnaphthalene		
p,p'-DDT	2,6-Dimethylnaphthalene		
	Acenaphthene	Trace elements	
Chlorinated hydrocarbons other than DDT and PCB	*3-ring*	Sb	Antimony
	Fluorene	As	Arsenic
	Phenanthrene	Cd	Cadmium
Aldrin	1-Methylphenanthrene	Cr	Chromium
Alpha-Chlordane	Anthracene	Cu	Copper
Trans-Nonachlor		Pb	Lead
Dieldrin	*4-ring*	Hg	Mercury
Heptachlor	Fluoranthene	Ni	Nickel
Heptachlor epoxide	Pyrene	Se	Selenium
Hexachlorobenzene	Benz(a)anthracene	Ag	Silver
Lindane (gamma-BHC)		SN	Tin
Mirex	*5-ring*	Zn	Zinc
	Chrysene		
	Benzo(a)pyrene		
	Benzo(e)pyrene		
	Perylene		
	Dibenz(a,h)anthracene		
Polychlorinated biphenyls[a]		Related parameters	
Dichlorobiphenyls		Lipid in tissure	
Trichlorobiphenyls		Grain size in sediment	
Tetrachlorobiphenyls		Total organic carbon in sediment	
Pentachlorobiphenyls		Coprostanol in sediments	
Hexachlorobiphenyls			
Heptachlorobiphenyls			
Octachlorobiphenyls			
Nonachlorobiphenyls			

[a] PCBs quantified to level of chlorination in 1984 through 1987. Beginning with samples collected in 1988, 18 individual compounds (congeners) have been quantified.

[b] Five PAH compounds added to the list of analytes in 1988; acenaphthylene, 2,3,5-trimethyl naphthalene, benzo(b) and benzo(k) fluoranthene, indeno(1,2,3-cd)pyrene, and benzo(g,h,i,)perylene.

Program began sampling fish and sediments in 1984. The Mussel Watch Program started sampling bivalve mollusks and sediments in 1986. The laboratories performing Benthic Surveillance activities have been, or are located at five sites: the NOAA National Marine Fisheries laboratories in Woods Hole, MA, Gloucester, MA, Beaufort, NC, Charleston, SC, and Seattle, WA. The Mussel Watch work is being performed at: the Battelle laboratories in Duxbury, MA, and in Sequim, WA, the Texas A&M University

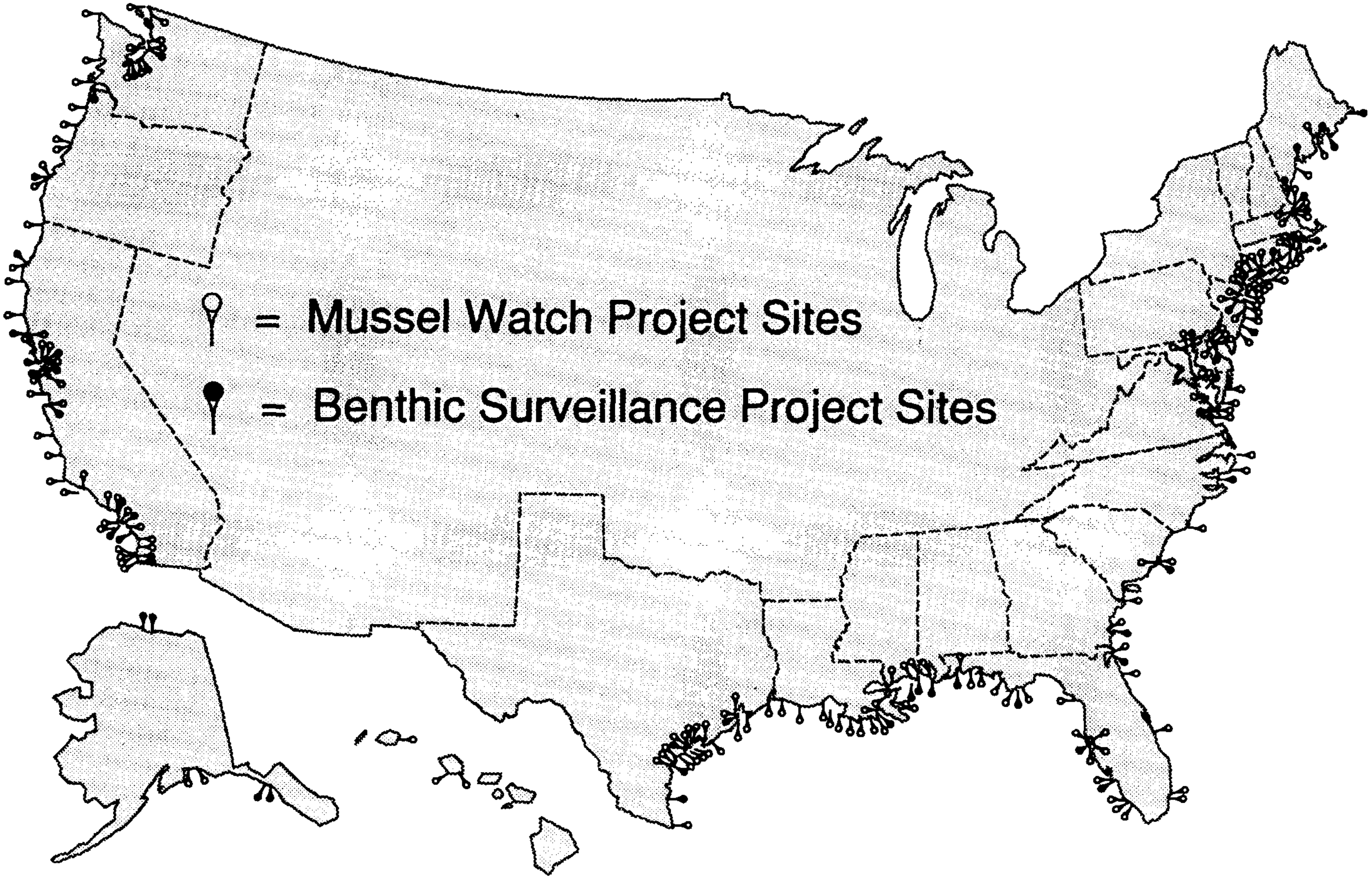

Fig. 1. Sites occupied by the Benthic Surveillance Project in 1984 through 1988 and by the Mussel Watch Project in 1986 through 1988. Larger scale maps and site designations are in NOAA (1988) and NOAA (1989).

Geochemical and Environmental Research Group in College Station, TX, and the LaJolla, CA laboratory of Scientific Applications International Corporation. The methods by which chlorinated compounds and polycyclic aromatic hydrocarbons have been extracted from sediments and subsequently analyzed have been based on those detailed in MacLeod *et al.* (1985) and Wade *et al.* (1988). Trace metal concentrations have been determined through atomic absorption analysis of total digests of tissue in nitric acid or sediment in hydroflouric acid. Intercalibration exercises continue as part of the NS&T Program. Results of organic chemical intercalibrations through 1987 are included in MacLeod (1989).

Shigenaka and Lauenstein (1987) provide a detailed listing of sampling protocols. Mussels *(Mytilus edulis)* in the size range of 5 to 8 cm have been taken at East Coast sites north of Delaware Bay. That species and the mussel, *Mytilus californianus*, have been collected on the West Coast. Seven to 10 cm long oysters (*Crassostrea virginica*) have been collected at Gulf Coast sites and at East Coast sites south of Delaware Bay. The oyster (*Ostrea sandvichensis*), 2.5 to 5 cm, has been sampled at the three sites in Hawaii. At all sites three composite samples (30 mussels or 20 oysters) have been collected in each year.

Sediment has been collected at three stations within each site, a station being anywhere within 500 m of a site center. In the Mussel Watch Project, if only sand could be found at a mollusk site, the sediment site could be as much as 2 km away. For Benthic Surveillance sampling in the Northeast, stations were up to about 5 km apart, so, in effect, a site could be 100 times larger than that for other NS&T sites. Sediment samples have been obtained with specially constructed box corers, Smith-MacIntyre bottom grabs, or Van Veen grab samplers. Subsamples for organic analyses have been surface skims from the top 3 cm (Benthic Surveillance) or 1 cm (Mussel Watch) of each grab or core. Composites have been made by mixing samples from three cores or grabs at each station.

At all NS&T sampling sites, three separate composite samples have been collected. For each year that a sediment, bivalve, or fish sample was collected at a site, the NS&T data base contains three separate values for the concentration of each contaminant. The NS&T data base now contains results of analyses of fish livers collected in 1984, 1985, and 1986, and bivalve mollusks collected in 1986 through 1988. Data are available from analyses of surface sediments collected in 1984 and 1985 in the Benthic Surveillance Project and in 1986 and 1987 in the Mussel Watch Project. These data have been summarized in a series of reports (NOAA, 1987; NOAA, 1988; and NOAA, 1989).

Site Selection

The primary criterion for site selection is the collection of samples from places that are representative of rather large areas and the avoidance of small-scale patches of contamination, or 'hot spots'. In particular, no sites are knowingly near waste discharge points. For the Mussel Watch Project a site also has to have a sufficiently large and robust population of mollusks to provide annual samples for an idefinite period. Lastly, when possible, Mussel Watch sites were chosen to coincide with sites from a previous monitoring program that sampled mollusks in 1976 through 1978 (Farrington *et al.*, 1983).

Sites are not uniformly distributed. Fully half of them are in urban areas (within 20 km of the centers of populations in excess of 100 000 people). This choice is based on the assumptions that contamination is higher, more likely to be causing biological effects, and more spatially variable in urban as opposed to rural areas. The same assumptions have led to sites being closer together in estuaries and embayments than along open coasts. Among the approximately 160 sites sampled in the Mussel Watch Project in 1986 and 1987, the mean distance between sites in enclosed areas was 23 km while, along coastlines it was 89 km. When analyses of sediment or mollusks from adjacent sites show very much higher concentrations at one site, a third site is sampled the subsequent year to test whether the high values are representative of the area.

Spatial Distribution of Contamination

Data from analyses of surface sediments have been used to define the spatial distribution of contamination. Contaminants are associated with particle surfaces and, under identical conditions, sand-sized particles have less contamination per unit weight of sediment than silt or clay. To account for this, the NS&T sediment data have been adjusted in two ways. First, no data for contaminants in sediment were used for comparisons among sites when the sediment contained more than 80% sand (particles with diameters $> 63\ \mu$). Secondly, and of less importance, contaminant levels in sediments containing less than 80% sand have been normalized by being divided by the fraction of sediment that is fine-grained (i.e., divided by a number between 0.20 and 1.00). That adjustment is the equivalent of considering sand to be only a diluent of sediment contamination. The exclusion of very sandy sediments acknowledges that some contamination may be associated with sand, but cannot be accounted for in this method of comparing among sites.

The results of the comparisons of 175 NS&T sites on the basis of concentrations of chemical contaminants in fine-grained sediments are presented in NOAA (1988). Mercury data in Figure 2 to exemplifies a log-normal distribution of mean concentrations (all contaminant concentrations are distributed in that manner). The mean concentrations at each site are based on all data collected (usually six samples minus any that were more than 80% sand) and variability has, therefore, been calculated. Generally, sites with similar concentrations have to be considered statistically equivalent. Because of that, because of the logarithmic distribution of the means, because the relative changes are largest among the highest concentrations, and because the higher concentrations are the more likely to be of biological significance, sites with the highest concentrations of each contaminant have been highlighted in Table II.

That table lists, in a clockwise geographic sequence, all sites sampled in 1984 through 1987. It shows which sites had concentrations among the highest 20 (approximately the upper 10%) concentrations of each contaminant. Most of the highest concentrations for any particular contaminant were found at the sites near Boston, New York, San Diego, Los Angeles, and Seattle. The association of higher levels of sediment contamination with highly populated areas is not an unexpected result. Nevertheless, it is important to note that these results are from what are considered representative sites and not isolated 'hot spots.'

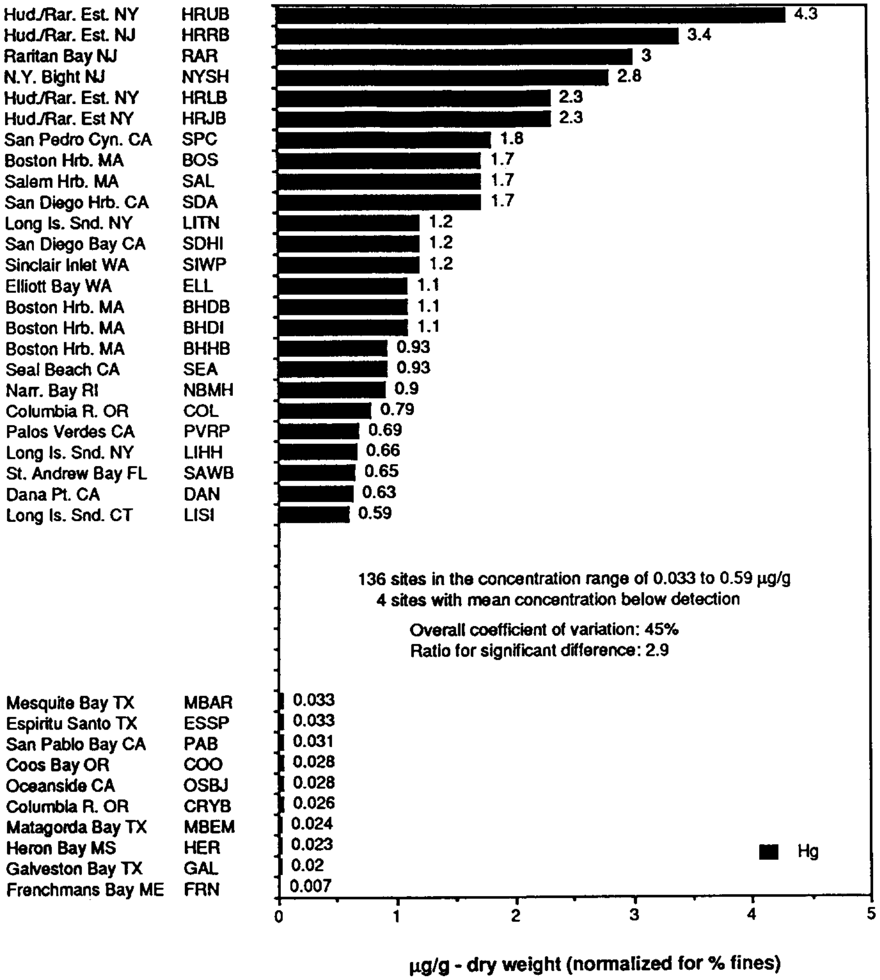

Fig. 2. Distribution of mean mercury concentrations at Benthic Surveillance sites (three-letter codes) sampled in 1984 and 1985, and at Mussel Watch sites (four-letter codes) sampled in 1986 and 1987 (NOAA, 1988). Means are based on all concentrations of all replicates regardless of year (usually a total of six replicates). Data have not been used for samples containing > 80% sand-sized (< 63 μ) particles. All raw data have been divided by the fraction of sediment in a sample which was fine-grained (< 63 μ). The overall coefficient of variation is the average of the coefficients for each site. The ratio for significant difference was estimated from a Least Significant Difference calculation on log-transformed data (the means in the figure and arithmetic means).

When objectives of a study have emphasized highly contaminated locations, levels of contamination have been found that exceed all the NS&T values. For example, Rodgerson *et al.* (1985) found levels of tPAH, tPCB, Cd, Cu, Pb, Zn in Black Rock Harbor in Bridgeport, CT, that are higher than all those found at NS&T sites. Levels of tPAH, Ag, Hg, and Pb in industrial waterways of Seattle and Tacoma have been found in excess of all NS&T concentrations (Malins *et al.*, 1982). Huggett *et al.* (1987) reported extremely high levels of tPAH in the Elizabeth River near Portsmouth, VA at a site where

TABLE II

NS&T sites sampled in 1984 through 1987, their locations, codes, total number (T) and identification of contaminants whose concentrations (normalized to fines fraction) ranked among the highest 20 concentrations (nfgs = no fine-grained sediment collected at the site).

Site location	Code[a]	T	Contaminant[b]
Machias, Bay, ME	MAC	0	
Frenchman Bay, ME	FRB	0	
Penobscot Bay, ME	PNB	0	
Penobscot Bay, ME	PBSI	0	
Penobscot Bay, ME	PBPI	1	tPAH
Casco Bay, ME	CAS	0	
Merrimack River, MA	MER	nfgs	
Salem Harbor, MA	SAL	12	Ag,Cd,Cr,Hg,Pb,Sb,Sn,Zn,tCHC,tDDT,tPCB,tPAH
Cape Ann, MA	CASI	3	Pb,Sb,Sn
Boston Harbor, MA	BHDI	10	Ag,Cr,Cu,Hg,Pb,Sb,Sn,tCHC,tPCB,tPAH
Boston Harbor, MA	BHDB	12	Ag,Cd,Cr,Cu,Hg,Pb,Sb,Sn,tCHC,tDDT,tPCB,tPAH
Boston Harbor, MA	BHHB	7	AG,Cr,Hg,Pb,Sb,Sn,tCHC
Boston Harbor, MA	BHBI	nfgs	
Boston Harbor, MA	BOS	12	Ag,Cd,Cr,Cu,Hg,Pb,Sb,Sn,Zn,tCHC,tPCB,tPAH
Buzzards Bay, MA	BBRH	1	tPCB
Buzzards Bay, MA	BBAR	4	Ag,tCHC,tPCB,tPAH
Buzzards Bay, MA	BBGN	0	
Buzzards Bay, MA	BUZ	0	
Narragansett Bay, RI	NBMH	2	Hg,Sn
Narragansett Bay, RI	NBCI	0	
Narragansett Bay, RI	NBDI	3	Sb,Sn,tPAH
Narragansett Bay, RI	NAR	2	Ag,Sn
Block Island, RI	BIBI	0	
E. Long Island Sound, CT	ELI	nfgs	
Long Island Sound, CT	LICR	0	
Long Island Sound, CT	LINH	nfgs	
Long Island Sound, CT	LIHR	nfgs	
Long Island Sound, CT	LISI	2	Cu,tPAH
W. Long Island Sound NY	WLI	2	Cu,Sn
Long Island Sound, NY	LIHU	0	
Long Island Sound, NY	LIPJ	nfgs	
Long Island Sound, NY	LIMR	3	Cd,Cu,Pb
Long Island Sound, NY	LIHH	6	Ag,Cd,Cu,Pb,Zn,tCHC
Long Island Sound, NY	LITN	11	Ag,Cd,Cu,Hg,Pb,Sn,Zn,tCHC,tDDT,tPCB,tPAH
Hudson/Raritan Est., NJ	HRJB	14	Ag,As,Cd,Cr,Cu,Hg,Pb,Sb,Sn,Zn,tCHC,tDDT, tPCB,tPAH
Hudson/Raritan Est., NJ	HRUB	8	Ag,As,Cu,Hg,Pb,Sb,Sn,tPAH
Hudson/Raritan Est., NJ	HRLB	14	Ag,As,Cd,Cr,Cu,Hg,Pb,Sb,Sn,Zn,tCHC,tDDT, tPCB,tPAH
Hudson/Raritan Est., NJ	HRRB	13	Ag,As,Cd,Cu,Hg,Pb,Sb,Sn,Zn,tCHC,tDDT,tPCB, tPAH
Raritan Bay, NJ	RAR	14	Ag,As,Cd,Cr,Cu,Hg,Pb,Sb,Sn,Zn,tCHC,tDDT,PCB, tPAH
New York Bight, NJ	NYSH	14	Ag,As,Cd,Cr,Cu,Hg,Pb,Sb,Sn,Zn,tCHC,tDDT, tPCB,tPAH
New York Bight, NJ	NYLB	nfgs	
New York Bight, NJ	NYSR	nfgs	

Table II (continued)

Site location	Code[a]	T	Contaminant[b]
New York Blight, NJ	NYSR	nfgs	
Morriches Bay, NY	MBTH	0	
Great Bay, NJ	GRB	0	
Delaware Bay, DE	DEL	2	tCHC,tPCB
Delaware Bay, DE	DBFE	0	
Delaware Bay, DE	DBBD	0	
Delaware Bay, DE	DBAP	0	
Delaware Bay, DE	DBKI	0	
Upper Ches. Bay, MD	UCB	2	Ni,Zn
Chesapeake Bay, MD	CBMP	1	Zn
Chesapeake Bay, MD	CBHP	0	
Chesapeake Bay, MD	CBHG	nfgs	
Mid. Chesapeake Bay, VA	MCB	0	
Chesapeake Bay, VA	CBIB	0	
Chesapeake Bay, VA	CBCC	0	
Chesapeake Bay, VA	CBDP	0	
Lower Ches. Bay, VA	LCB	0	
Chincoteague Bay, VA	CBCI	nfgs	
Quinby Inlet, VA	QIUB	0	
Roanoke Sound, NC	RSJC	nfgs	
Pamlico Sound, NC	PSWB	nfgs	
Pamlico Sound, NC	PAM	0	
Cape Fear, NC	CFBI	1	As
Charleston Harbor, SC	CHFJ	0	
Charleston Harbor, SC	CHSF	0	
Charleston Harbor, SC	CHS	0	
Savannah R. Estuary, GA	SRTI	0	
Sapelo Sound, GA	SSSI	nfgs	
Sapelo Is, GA	SAP	0	
St. Johns River, FL	SJCB	0	
St. Johns River, FL	SJR	1	tPCB
Matanzas River, FL	MRCB	nfgs	
Biscayne Bay, FL	BBPC	0	
Everglades, FL	EVFU	0	
Rookery Bay, FL	RBHC	o	
Naples Bay, FL	NBNB	0	
Charlotte Harbor, FL	CBBI	0	
Charlotte Harbor, FL	LOT	0	
Tampa Bay, FL	TAM	0	
Tampa Bay, FL	TBMK	1	tCHC
Tampa Bay, FL	TBCB	nfgs	
Tampa Bay, FL	TBHB	1	Pb
Tampa Bay, FL	TBPB	1	tPCB
Cedar Key, FL	CKBP	0	
Apalachicola Bay, FL	APCP	0	
Apalachicola Bay, FL	APDB	0	
Apalachicola Bay, FL	APA	0	
St. Andrew Bay, FL	SAWB	4	tCHC,tDDT,tPCB,tPAH
Choctawhatchee Bay, FL	CBSP	4	Pb,tCHC,tDDT,tPAH
Choctawhatchee Bay, FL	CBSR	1	As
Pensacola Bay, FL	PEN	0	

Table II (continued)

Site Location	Code[a]	T	Contaminant[b]
Pensacola Bay, FL	PBIB	0	
Mobile Bay, AL	MBCP	0	
Mobile Bay, AL	MOB	0	
Round Island, MS	ROU	0	
Heron Bay, MS	HER	0	
Mississippi Sound, MS	MSPB	0	
Mississippi Sound, MS	MSBB	1	tPAH
Mississippi Sound, MS	MSPC	0	
Mississippi Delta, LA	MRD	1	tCHC
Lake Borgne, LA	LBMP	0	
Breton Sound, LA	BSSI	0	
Breton Sound, LA	BSBG	0	
Barataria Bay, LA	BBSD	0	
Barataria Bay, LA	BBMB	0	
Barataria Bay, LA	BAB	0	
Terrebonne Bay, LA	TBLF	0	
Terrebonne Bay, LA	TBLB	0	
Caillou Lake, LA	CLCL	0	
Atchafalaya Bay, LA	ABOB	0	
Vermillion Bay, LA	VBSP	0	
Joseph Harbor Bayou, LA	JHJH	0	
Calcasieu Lake, LA	CLSJ	0	
Sabine Lake, LA	SLBB	0	
East Cote Blanche, LA	ECSP	nfgs	
Galveston Bay, TX	GBHR	0	
Galveston Bay, TX	GBYC	0	
Galveston Bay, TX	GBTD	0	
Galveston Bay, TX	GBCB	0	
Galveston Bay, TX	GAL	0	
Matagorda Bay, TX	MBEM	0	
Matagorda Bay, TX	MBTP	0	
Matagorda Bay, TX	MBGP	0	
Matagorda Bay, TX	MBLR	0	
Espiritu Santo, TX	ESSP	0	
Espiritu Santo, TX	ESBD	0	
San Antonio Bay, TX	SAMP	0	
San Antonio Bay, TX	SAPP	0	
San Antonio Bay, TX	SAB	0	
Mesquite Bay, TX	MBAR	0	
Copano Bay, TX	CBCR	0	
Aransas Bay, TX	ABLR	0	
Corpus Christi, TX	CCIC	0	
Corpus Christi, TX	CCNB	0	
Corpus Christi Bay, TX	CCB	0	
Lower Laguna Madre, TX	LMSB	0	
Lower Laguna Madre, TX	LLM	0	
Imperial Beach, CA	IBIB	nfgs	
San Diego Bay, CA	SDF	2	As,Cd
San Diego Bay, CA	SDHI	5	As,Cu,Hg,Pb,Zn
San Diego Harbor, CA	SDA	7	Cu,Hg,Pb,Sn,Zn,tPCB,tPAH
Point Loma, CA	PLLH	1	As

Table II (continued)

Site Location	Code[a]	T	Contaminant[b]
Mission Bay, CA	MBVB	nfgs	
La Jolla, CA	LJLJ	0	
Oceanside, CA	OSBJ	1	tDDT
Dana Point, CA	DAN	1	As
Newport Beach, CA	NBBC	1	tDDT
Anaheim Bay, CA	ABWJ	0	
Seal Beach, CA	SEA	1	Hg
Long Beach, CA	LNB	5	Cd,Pb,Zn,tCHC,tDDT
San Pedro Bay, CA	SPC	10	Ag,Cd,C,rCu,Hg,Ni,Sn,Zn,tDDT,tPCB
San Pedro Harbor, CA	SPFP	3	Cd,Cu,tDDT
Palos Verdes, CA	PVRP	9	Ag,Cd,Cr,Cu,Sn,Zn,tCHC,tDDT,tPCB
Santa Catalina Island, CA	SCBR	nfgs	
Santa Monica Bay, CA	SMB	nfgs	
Marina del Rey, CA	MDSJ	3	Ag,As,tDDT
Point Dume, CA	PDPD	2	As,tDDT
Santa Cruz Is. CA	SCFP	nfgs	
Point Santa Barbara, CA	SBSB	3	As,Cd,tDDT
Point Conception, CA	PCPC	nfgs	
San Luis Obispo Bay, CA	SLSL	nfgs	
San Simeon Point, CA	SSSS	nfgs	
Pacific Grove, CA	PGLP	nfgs	
Monterey Bay, CA	MBSC	0	
Monterey Bay, CA	MON	nfgs	
Southhampton Shoal, CA	SHS	nfgs	
Oakland Estuary, CA	OAK	1	Ni
Hunters Point, CA	HUN	2	Cr,Ni
San Francisco Bay, CA	SFDB	1	Ni
San Francisco Bay, CA	SFSM	1	Ni
San Francisco Bay, CA	SFEM	1	Ni
San Pablo Bay, CA	PAB	3	Cr,Ni,Zn
San Pablo Bay, CA	SPSM	3	As,Cr,Ni
San Pablo Bay, CA	SPSP	1	Ni
Tomales Bay, CA	TBSR	1	Ni
Bodega Bay, CA	BBBE	nfgs	
Bodega Bay, CA	BOD	nfgs	
Point Arena, CA	PAPA	nfgs	
Point Delgada, CA	PDSC	nfgs	
Humboldt Bay, CA	HMBJ	nfgs	
Humboldt Bay, CA	HMB	4	As,Cr,Ni,Sb
Point Saint George, OR	SGSG	nfgs	
Coos Bay, OR	COO	2	As,Cd
Coos Bay, OR	CBCH	3	As,Cr,Ni
Coos Bay, OR	CBRP	2	Cr,Ni
Yaguina Bay, OR	YBOP	0	
Yaquina Bay, OR	YHYH	2	Cr,Sb
Tillamook Bay, OR	TBHP	3	As,Cr,Ni
Columbia River, OR	CRYB	2	Sb,Zn
Columbia River, OR	COL	4	Ag,Cd,Hg,Zn
Grays Harbor, WA	GHWJ	nfgs	
Strait of Juan de Fuca WA	JFNB	1	Cr
South Puget Sound, WA	SSBI	1	Sb

Table II (continued)

Site location	Code[a]	T	Contaminant[b]
Nisqually Reach, WA	NIS	nfgs	
Commencement Bay, WA	COM	1	Ag
Commencement Bay, WA	CBBP	1	Sb
Elliott Bay, WA	EBFR	nfgs	
Elliott Bay, WA	ELL	8	Cd,Cu,Hg,Ni,Sb,Zn,tPCB,tPAH
Sinclair Inlet, WA	SIWP	3	Hg,Ni,Sb
Whidbey Island, WA	WIPP	0	
Bellingham Bay, WA	BBSM	1	Ni
Point Roberts, WA	PRPR	0	
Lutak Inlet, AK	LUT	0	
Nahku Bay, AK	NAH	nfgs	
Unakwit Inlet, AK	UISB	0	
Port Valdez, AK	PVMC	0	
Oliktok Point, AK	OLI	nfgs	
Prudhoe Bay, AK	END	1	Ni
Barbers Point, HI	BPBP	1	Ni
Honolulu Harbor, HI	HHKL	0	

[a] CODES are used here do differentiate among sites in the same general location. Specific site locations are indicated in NOAA (1988). Three and four letter codes designate Benthic Surveillance and Mussel Watch sites, respectively.

[b] The organic contaminants have been aggregated in classes: tDDT=sum of all DDT and its metabolites, tPCB=sum of all PCBs, tCHC=sum of all chlorinated organic compounds except tDDT and tPCB, tPAH=sum of all polynuclear aromatic hydrocarbons.

creosote had been discharged into the river. While sediments at the NS&T sites in Boston Harbor show high levels of tPAH, they are not as high as some of those reported by Shiaris and Jambard-Sweet (1986) who analyzed a similar suite of PAH compounds in sediments from around piers and other inner parts of Boston Harbor. There are many NS&T sites in Southern California, but with a single exception, none have high levels of tPAH in sediments (Table II). Nevertheless, high concentrations have been found near discharges and centers of industrial activity (Anderson and Gossett, 1987).

Sampling on a much finer spatial scale within industrial areas would yield much higher levels of contamination, but they would be of little spatial significance. The fact that NS&T sites in Table II that have high levels of contamination tend to appear in clusters substantiates their representativeness. These are not isolated, highly contaminated spots but, rather, are typical of their surroundings. However, the representativeness of some sites in Table II remains untested. The sites in Elliott Bay, WA, and St. Andrews Bay, FL, both showed high levels for several contaminants but there were no nearby sites against which to gauge their representativeness. Similarly, a site in Choctawatchee Bay, FL, showed high contaminant levels while another, in the same bay, did not. Those two sites are about 30 km apart. Data are being obtained from sediments taken within 10 km of all these sites to test whether or not they are representative.

Biological Effects

While chemical monitoring yields the distribution of contamination, it does not in itself indicate a biological consequence to the contamination. Two fundamentally different ways of approaching that issue are to examine indigenous species for evidence of response or, to conduct bioassay tests of the water or sediment at a site. Both are being employed in the NS&T Program. A number of studies on fish reproduction, biochemical changes in fish, and bioassays of sediment and water have been, or are now being conducted. All of those, however, are restricted to limited areas of the country. To date, the biological data from measurements that have been made around the country are from histological examinations of tissues from fish and mussels.

The existence of neoplastic tumors in feral fish is usually interpreted as a response to contamination (Susani, 1986). Complicating factors are that older fish will have a higher frequency of tumors than younger fish and, that even under identical exposures to contamination, different species of fish metabolize contaminants at different rates and are more likely than others to develop tumors (Varanasi *et al.*, 1987). The observations in the NS&T Program are not all on older fish and, because it is a national program, cannot be on a single species. Nevertheless, it is noteworthy that neoplasia in fish livers is usually not found. Among approximately 5600 fish that have been examined between 1984 and 1986, hepatic neoplasias (liver tumors) were found in only about 40, with more than half of those taken at two sites. The infrequent occurrence of liver tumors is related to the species examined, but is most probably due to liver tumors being an extreme response to contamination and the fact that NS&T sites are not located in known 'hot spots'.

Another extreme response to contamination is the fact that sediments can be found that are acutely toxic to test organisms. Independent of the NS&T program, some investigators, especially those working in Puget Sound, have compared their bioassay results with contaminant concentrations in the sediment being tested. Two such comparisons are summarized in Table III, along with the number of NS&T sites, where contaminant concentrations exceed the possible threshold levels.

The basis of thresholds is strictly empirical in that toxicity is not necessarily caused by the contaminant. The thresholds are just the highest concentrations of a contaminant in sediment that was not toxic. Since a future test may not find a sediment sample to be toxic even when it contains more than a threshold concentration for some contaminant, thresholds are subject to upward revision. The first set of data in Table III (Barrick *et al.*, 1988), implies that sediment toxicity due to PCBs or Hg might be found at about 20% of the NS&T sites. The PCBs and Hg thresholds in the second set of data (DeWitt *et al.*, 1988) would put fewer NS&T sites into the toxic category but, on the other hand, imply that more sites would be toxic because of other contaminants. The DeWitt *et al.* (1988) analysis is somewhat different than that of Barrick *et al.* (1988) because the values shown in Table III are concentrations in sediments where toxicity was observed but could not be distinguished from effects of particle size. Only at much higher concentrations were DeWitt *et al.* (1988) confident in assigning toxicity to chemical contamination.

It is possible, that at a small fraction of NS&T sites contaminant concentrations are

TABLE III

Numbers of sites (out of 175) where concentrations of contaminants in finegrained sediments exceed an estimated threshold of sediment toxicity

Contaminant[a]	Conc (μ g^{-1}-dry) at toxicity	# NS&T Sites in excess	
(AETs from Barrick *et al.* [1988])[b]			(%)
LMWPAH	5.2	4	(2)
HMWPAH	12	2	(1)
tPCB	0.13	37	(21)
As	93	0	(0)
Cd	6.7	2	(1)
Cu	390	0	(0)
Pb	530	0	(0)
Hg	0.41	38	(22)
Zn	960	0	(0)
(Amphipod test results of Dewitt *et al.* [1988])[c]			
tPAH	7.6	16	(9)
tPCB	0.26	26	(15)
Ag	0.56	52	(30)
As	25	25	(14)
Cu	138	16	(9)
Pb	140	15	(9)
Hg	1.4	10	(6)
Zn	200	46	(26)

[a] The organic contaminants have been aggregated in classes: LMWPAH and HMWPAH are low and high molecular weight polyaromatic hyrocarbons and are the sums of the concentrations of the 2 to 3-ring and 4 to 5-ring PAH compounds, respectively, listed in Table I, tPCB is the sum of all PCBs, and tPAH is the sum of all polynuclear aromatic hydrocarbons.

[b] First grouping contains the lowest of three bioassay based Apparent Effect Threshold (AET's) from Barrick *et al.* (1988). The three bioassyas were those based on amphipod survival, oyster embryo development, and bacterial bioluminescence.

[c] Second grouping contains the concentrations at which a decreased survival of amphipods toxicity could begin to be associated with contamination rather than natural factors (DeWitt *et al.*, 1988). The authors provide a second set of concentrations above which they are more confident that toxicity is related to chemical contamination. No NS&T sites have concentrations that exceed those higher levels for Hg, Pb, As, Cu, and Zn. Nine (5%) of the NS&T tPAH concentrations exceed the higher tPAH threshold. The second set of levels for tPCB and Ag are the same as the evels shown above because, in the sediments tested, toxicity increased without a corresponding increase in either contaminant concentration.

sufficiently concentrated to be toxic under some conditions. Certainly further work on sediment bioassays are needed if data like that in Table III are ever to allow more than approximate estimates of biological consequences to contamination. One of the reasons for including biological measurements in the NS&T Program is that it may never be possible to use chemical data alone to estimate biological effects. Several ongoing projects are seeking to find contaminant responses in fish that are less dramatic than liver tumors but may be more often found and of possible significance to the longevity or reproductive potential of fish. In that sense, the NS&T Program serves as an experimental matrix in which to find biological responses that are being caused by chemical contamination.

Temporal Trends in Molluscan Tissue Data

Because neither the same molluscan nor fish species can be collected throughout the country, tissue data cannot be used uncritically to describe the spatial distribution of contamination. The Mussel Watch Program uses two species of mussels and two of oysters (one of which is collected only at sites in Hawaii). Data are similar for both mussel species when they are collected at a single site. For organic contaminants it is probably acceptable to consider mussels and oysters as equivalent matrices, but for elemental analytes, especially silver, copper, and zinc, the two are not equivalent (NOAA, 1989). Comparisons among sites for fish liver data are more problematic than those based on mollusks because there are fewer sites (50 as opposed to 150), and more species (eight as opposed to three) in the Benthic Surveillance Program.

The correspondence between contaminant concentrations in sediments and bivalve mollusks can be quantified by examining data for 117 sites from which bivalves and fine-grained sediments were collected. Spearman Rank Correlations (Table IV) are a measure of concordance of rankings based on tissue data with those derived from sediment analyses. The negative correlations for siver, copper, and zinc are due to their being highly accumulated by oysters. Considering mussel and oyster data separately in Table IV shows a generally better correspondence between mussels and sediments than

TABLE IV

Spearman Ranked Correlation coefficients for arithmetic means of concentrations in sediments and mollusks at Mussel Watch sites where mollusks and fine-grained sediment were sampled in 1986 and 1987. All sediment data have been normalized for grain size. Correlation coefficients that are statistically different from zero at the 0.05 level of confidence are marked with an asterisk (*).

Contaminant[a]	All Bivalves (n=117)	Mussels (n=53)	Oysters (n=64)
Ag	-0.286*	0.587*	0.247
As	0.260*	0.238	0.333*
Cd	-0.175	0.049	-0.087
Cr	0.627*	0.565*	0.097
Cu	-0.513*	0.697*	0.102
Hg	0.430*	0.442*	0.242
Ni	0.427*	0.611*	0.406*
Pb	0.696*	0.750*	0.372*
Zn	-0.329*	0.253	0.509*
tPCB	0.681*	0.855*	0.372*
tDDT	0.696*	0.754*	0.644*
tChlordane	0.478*	0.567*	0.409*
Dieldrin	0.515*	0.690*	0.281*
LMWPAH	0.503*	0.550*	0.441*
HMWPAH	0.602*	0.611*	0.519*

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[a] Contaminant classes, tPCB, tDDT, LMWPAH, and HMWPAH are as defined in Tables II and III. tChlordane is the sum of four compounds alpha-chlordane, transnonachlor, heptachlor, and heptachlorepoxide and is dominated by first two. Dieldrin includes the aldrin concentration but averages more than 90% dieldrin.

oysters and sediments. Even for oysters, the correlations are often statistically significant, but are almost always lower than for mussels.

Data from analyses of bivalve mollusks are not the equivalent of sediment-derived data for determining spatial trends in contamination. On the other hand, species remain constant for each site and temporal trends at a site will be more readily revealed through mollusks than through sediments. The ability of mollusks to change contaminant levels in response to changes in their surroundings is attested to by their frequent use in transplant experiments or in tests of bioaccumulation (e.g., Roesijadi *et al.*, 1984; Pruell *et al.*, 1987). However, temporal resolution in sediments depends on rates of sedimentation and bioturbation. While neither is known at NS&T sites, it is not assumed that annual changes in contaminant levels will be manifest through annual collections of surface sediments.

For three years, mollusks have been collected at twelve-month intervals (in the late winter to early spring) at 132 sites. The molluscan data for each site have been examined for a statistically significant differences among the three years and a monotonic change (NOAA, 1989). Table V lists the number of times those two criteria were met for each contaminant. The generally more hints of trends among the elements than among the organic contaminants is due to the higher within-year variability for organic compounds. That variability diminishes the ability to detect significant differences between years. There are patterns in the data that beg explanation. For example, more trends are

TABLE V

Numbers of sites (out of 132) where mean contaminant concentrations in bivalve mollusks were significantly different (0.05 leve, Kruskal-Wallis test) among the years 1986, 1987, and 1988 and were monotonically increasing or decreasing

Contaminant	# sites Increasing	# sites Decreasing
Ag	11	13
As	6	8
Cd	4	17
Cr	8	7
Cu	10	12
Hg	10	4
Ni	7	9
Pb	5	1
Se	12	2
Zn	7	10
tDDT	4	8
tPCB	0	13[a]
tChlordane	2	9
Dieldrin	1	8
LMWPAH	5	6
HMWPAH	3	4

[a] The relatively high number of tPCB decreases relative to no increases may be an artifact of the 1988 analyses having been based on 18 PCB congeners, while the earlier were based on PCB concentrations at each level of chlorination (NOAA, 1989).

downward rather than upward, but for Pb, Hg, and Se, the reverse applies and there are geographic grouping among some of these possible trends. With only three years of data, however, explanations are premature. The apparent trends may disappear with one more set of data.

In the long term trends can be identified by correlating concentration with time. It will not be necessary that they be strictly monotonic. At this stage, with data for only three years, monotonicity is necessary to find even hints of trends. Three pairs of data per contaminant per site are all that exist and that makes trend identification very difficult. That difficulty, it should be noted, would not be alleviated if more time had lapsed between sample collections. For example, if sampling were done at five-year intervals, ten years would be needed to get three data points, and the identification of trends would be as tenuous as it is now with only three data points. The annual NS&T sampling is continuing and data will soon exist to test whether the hints of trends in the first three years are substantiated.

Conclusions

The spatial distribution of contamination, as exhibited through analyses of surface sediments, shows the higher levels to be characteristic of urban areas. More samples have been collected where unexpectedly high levels were found in rural areas or where high levels at one site were not substantiated by data from other sites. Higher levels of sediment contamination exist at discharge points and in isolated industrial locales, sites that do not qualify as representative NS&T sites. Among the NS&T sites frank biological responses to contamination, such as liver tumors in fish or possible sediment toxicity, have not been commonly found. Temporal trends in contaminant levels at NS&T sites are beginning to be drawn from annual analyses of mussels and oysters. At present, however, with data from only the first three years of the program, only hints of trends can be identified.

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