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A Special NOAA 20th Anniversary Report

Coastal Environmental Quality in the United States, 1990

Chemical Contamination in Sediment and Tissues



U. S. Department of Commerce
National Oceanic and Atmospheric Administration
National Ocean Service

National Status and Trends Program

Since 1984, the Office of Oceanography and Marine Assessment has monitored, through its National Status and Trends (NS&T) Program, the concentrations of toxic organic compounds and trace metals in bottom-feeding fish, shellfish, and sediments at almost 300 coastal and estuarine locations throughout the United States. The objective of the program is to determine the status and long-term trends of toxic contamination in these important areas. Samples collected annually through the program are analyzed to determine levels of synthetic chlorinated compounds (e.g., DDTs), polychlorinated biphenyls (PCBs), polynuclear aromatic hydrocarbons (PAHs), and toxic trace metals (e.g., mercury and lead). NOAA's NS&T Program is the first to use a uniform set of techniques to measure coastal and estuarine environmental quality over relatively large space and time scales. A "specimen bank" of samples taken each year at about 10% of the sites is maintained at the National Institute of Standards and Technology for future, retrospective analyses. A related program of directed research is examining the relationships between contaminant exposures and indicators of biological responses in fish and shellfish (i.e. bioeffects) in areas that are shown by the NS&T monitoring results to have high levels of toxic chemicals.

This report, based on six years of results from the NS&T Program and other monitoring efforts, describes the spatial extent and severity of chemical contamination and changes in concentrations of contaminants over the last decade.

Additional information on NOAA's NS&T Program and related activities is available from: Thomas P. O'Connor, Coastal and Estuarine Assessment Branch, Ocean Assessments Division, Office of Oceanography and Marine Assessment, National Ocean Service, National Oceanic and Atmospheric Administration, 6001 Executive Boulevard, Rockville, MD 20852.

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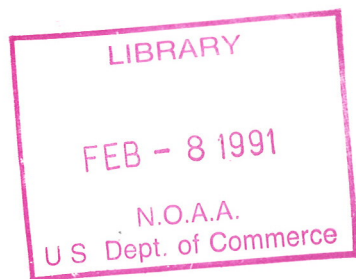
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Thomas P. O'Connor
Manager

National Status and Trends Program



Coastal and Estuarine Assessment Branch
Ocean Assessments Division
Office of Oceanography and Marine Assessment
National Ocean Service
National Oceanic and Atmospheric Administration
Rockville, Maryland 20852

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Introduction

Reports of beaches being closed, trash washing ashore, prohibitions on shell-fishing, health warnings to seafood consumers, waste discharges to the sea, ocean dumping, and habitat losses have aroused considerable public concern about the quality of the coastal environment in the United States. To assess the effects of human activities on the quality of coastal and estuarine areas throughout the Nation, the National Oceanic and Atmospheric Administration (NOAA) created the National Status and Trends (NS&T) Program to monitor trends of chemical contamination in space and time and to determine biological responses to that contamination. Since 1984, annual chemical analyses for trace metals and organic contaminants, e.g., pesticides, have been made on surface sediments, on livers of benthic fish, and on whole soft-parts of mussels and oysters collected from a network of almost 300 sites around the U.S.

The need for this type of national monitoring of ambient environmental quality was recently emphasized by the U.S. National Research Council. Its report (NRC, 1990) indicated that the United States annually spends more than \$130 million on coastal environmental monitoring, but that most is devoted to compliance monitoring, i.e., testing wastewaters and other materials prior to discharge or to performing measurements prescribed by regulation very near to discharge points. Since compliance monitoring, by design, covers very small spatial scales and short time periods, programs such as NOAA's NS&T Program are required to focus on wider

public concerns about the long-term effects of coastal pollution throughout the U.S.

This report, based on six years of results from the NS&T Program and other monitoring efforts, describes the spatial extent and severity of chemical contamination and changes in concentrations of contaminants over the last decade. While conclusions are always subject to new information, it appears that, on a national scale, high and biologically significant concentrations of contaminants measured in the NS&T Program are limited primarily to urbanized estuaries. In addition, levels of those contaminants have, in general, begun to decrease in the coastal U.S.

Sampling Sites

NOAA's NS&T Program is designed to describe national, rather than local, distributions of contamination. Since its inception, the primary criterion for NS&T site selection has been the collection of samples from places that are "representative" of large coastal areas and the avoidance of small-scale patches of contamination, or "hot spots." In particular, no sites were knowingly selected near waste discharge points. For the "Mussel Watch" component of the NS&T Program, a site also has to have a sufficiently large and robust population of mussels or oysters to provide annual samples for an indefinite period.

NOAA sampling sites are not uniformly distributed. Almost half of them are in urban estuaries, within 10 miles of the centers of populations in excess of 100,000 people. This choice is based on assumptions that contamination is

higher, more likely to be causing biological effects, and more spatially variable in urban, as opposed to rural, coastal areas. The same assumptions have led to sites being closer together in estuaries and bays than along open coasts.

Contaminants Measured

The NS&T Program monitors concentrations of trace metals and organic compounds in sediment and in tissues. Except for most chlorinated organic compounds — DDT and PCB, for example, whose existence is due to human activity — some concentration of chemicals in sediments and organisms is natural and would be present even in the absence of human activity. Only sediments and tissues that contain chemical concentrations in excess of natural levels are considered to be "contaminated."

The exact line demarcating natural concentrations from contamination is not easily drawn because it depends on many local and regional conditions. This report highlights NS&T sites where concentrations are highest and, while cases exist where high concentrations might be natural, for the most part, they are due to discharges to coastal waters from human activities.

Data on concentrations of the seven trace metals and four groups of organic compounds listed in **Table 1** are used in this report to describe the status and trends of contamination in the coastal and estuarine areas of the U.S. Concentrations of all of these chemicals can serve as indicators of human activity. While the metals all have different uses, they can be categorized as chemicals whose discharge to the environment

has been enhanced through industrialization.

The groups of organic compounds cannot be categorized so generally. Two of those groups, total DDT (tDDT) and chlordane (tCdane), are chlorinated pesticides. Use of DDT in the United States was banned in 1970. The use of chlordane on crops and ornamental plants was first restricted in 1974. Its major use as a termiticide came under severe restriction in 1988.

Polychlorinated biphenyls (tPCB) are a mixture of compounds based on the biphenyl molecule chlorinated to various extents. It was first used in 1929 for a number of industrial purposes. Its high heat capacity and low dielectric constant were exploited for its major use in electrical transformers and capacitors. Its use in the United States began to be phased out in 1971 and it has been banned in new devices since 1976.

All of these banned compounds — tDDT, tCdane, and tPCB — continue to be used in other countries and still exist in the environment. In the United States, chlordane is still in the ground as a termiticide, PCB-containing devices are still in use, and DDT, while no longer used, remains in the environment because (like chlordane and PCB) of its resistance to degradation. The pesticide DDT is metabolized to DDE and DDD in the environment, but the tDDT group of compounds resists further degradation.

Polycyclic aromatic hydrocarbons (PAHs) are like metals in the sense that they are not synthetic but occur naturally. They are found in fossil fuels such as coal and oil. Their existence, though,

Table 1. *Chemicals measured in NOAA's National Status and Trends Program.*

Trace Metals

Cadmium (Cd)
Chromium (Cr)
Copper (Cu)
Lead (Pb)
Mercury (Hg)
Silver (Ag)
Zinc (Zn)

Organic Compounds

Total DDT (tDDT)

The sum of concentrations of DDT (dichloro-diphenyl-trichloroethane) and its metabolites DDE (dichloro-diphenyl-trichloroethylene) and DDD (dichloro-diphenyl-dichloroethylene).

Total chlordane (tCdane)

The sum of concentrations of two major constituents of chlordane mixtures: alpha-chlordane and trans-nonachlor.

Total polychlorinated biphenyls (tPCB)

The sum of the concentrations of di-, tri-, tetra-, penta-, hexa-, hepta-, octa-, and nona-chlorobiphenyls. Since 1988, the equivalent tPCB has been calculated from the sum of concentrations of 18 individual PCB congeners.

Total polycyclic aromatic hydrocarbons (tPAH)

The sum of concentrations of 18 PAH compounds: six 2-ring compounds (biphenyl, naphthalene, 1-methylnaphthalene, 2-methylnaphthalene, 2,6-dimethylnaphthalene, and acenaphthene); four 3-ring compounds (flourene, phenanthrene, 1-methylphenanthrene, and anthracene); three 4-ring compounds (flouranthene, pyrene, and benz(a)anthracene); and five 5-ring compounds (chrysene, benzo(a)pyrene, benzo(e)pyrene, perylene, and dibenz(a,h)anthracene).

is also attributable to human activity because they are produced when organic matter is burned. A multitude of human activities, from burning coal and wood to incineration of wastes, create PAH compounds in excess of those that would exist naturally. Often, the lower molecular weight compounds (2- and 3-ring

compounds in Table 1) are classified separately from the higher-weight compounds (4- and 5-rings) because the lower-weight compounds have a higher association with petroleum and the higher-weight compounds with combustion products. Since high concentrations of both types of compounds tend to be

found in the same locations, all PAH compounds have been combined into a single group for this report.

All of these trace metals and groups of organic compounds can be acutely or chronically toxic to marine life and even to people under some conditions. Those conditions include the total concentration of chemical that is biologically available and the ability of each species to accommodate increased chemical exposure. An important aspect of the NS&T Program is to determine the distribution of locations where contamination is of biological consequence.

Nationwide Distribution of Contaminants in Sediment

The nationwide results from NOAA analyses of surface sediments can be used to define the spatial distribution of contamination. Before simply using the data, however, it is important to know that contaminants are associated with particle surfaces. 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 less than 63 microns). Secondly, contaminant levels in sediments containing less than 80% sand have been adjusted by dividing 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 dilutant of sediment contamination. The exclusion of very sandy sediments acknowledges that some contamination may be associated with

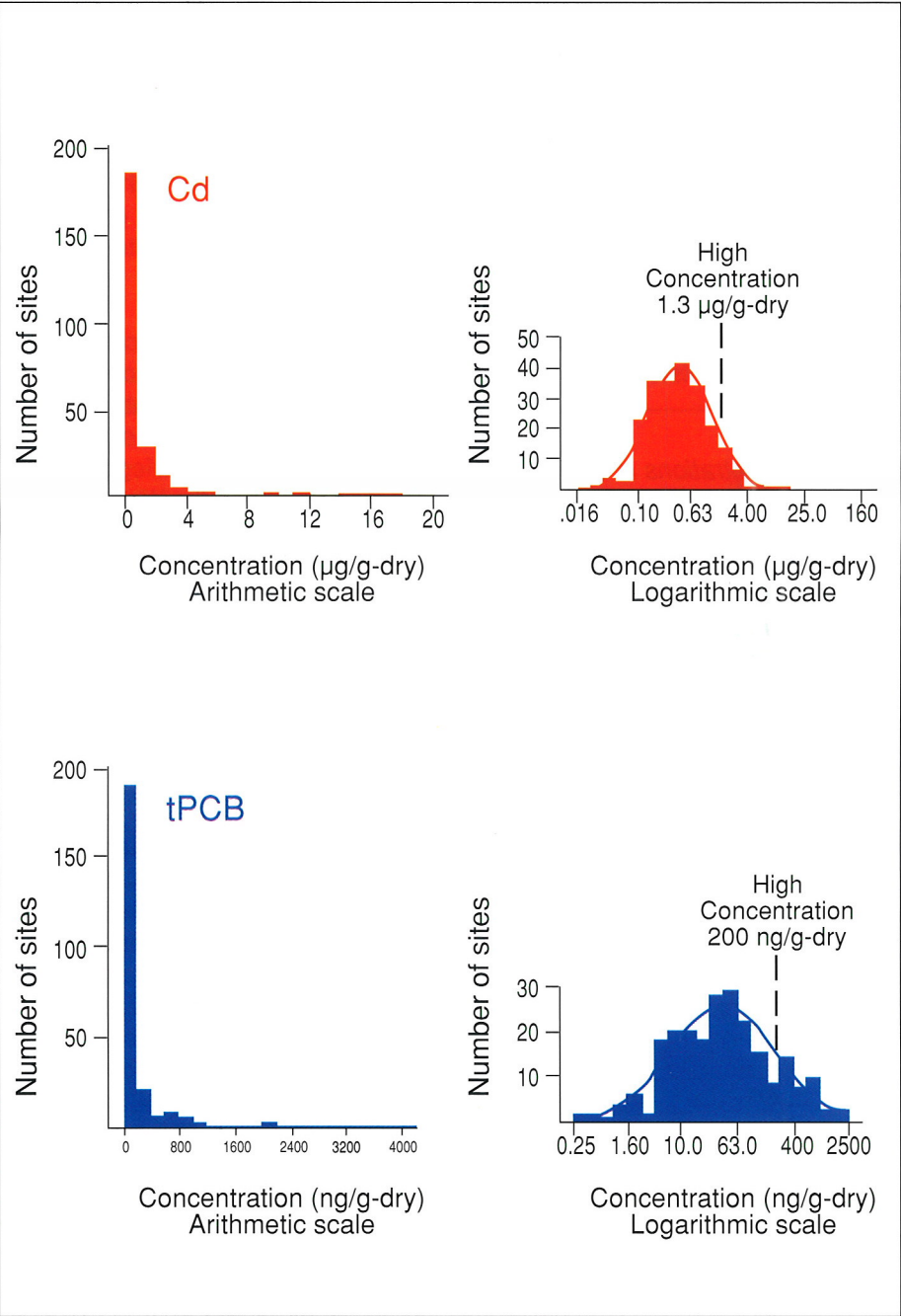
sand, but cannot be accounted for in this method of comparing among sites.

The NS&T results of sediment analyses have already been reported (NOAA, 1988). Those data, plus new data from NOAA Mussel Watch sites occupied in 1988 and 1989 and NOAA Benthic Surveillance data from 1986, provide information on chemical concentrations in sediments at 287 sites. Fine-grained sediment was collected at 232 of those sites. Each time sediments were sampled at a site, three separate samples of surface sediment were collected. The overall total number of samples per site varies somewhat depending on how often a site was sampled and on the availability of fine-grained sediment, but in general, the mean concentrations used in this report are based on analyses of six samples at each site.

Data on cadmium and tPCB are used in **Figure 1** to show that, for both trace metals and organic chemicals, concentrations are distributed in such a way that there are few high concentrations that stand out from the rest. When distributions of concentrations are highly skewed toward the lower concentrations, it is useful to examine the distributions of the logarithms of the data. As exemplified in Figure 1, when the logarithms of the concentrations are plotted, the distributions become bell-shaped. In statistical analyses such distributions are referred to as normal distributions and, in this case because logarithms were required, the distributions are "log-normal."

The advantage of the fact that the concentrations are log-normally distributed is that it allows a statistically objective

Figure 1. *Distributions of Cadmium and tPCB concentrations in sediment on arithmetic and logarithmic scales*



and convenient definition of "high" concentrations as those whose logarithmic value is more than the mean plus one standard deviation of the logarithms for all concentrations. In practice, because we are dealing with normal distributions, about 17% of all the concentrations for each chemical will fall into the "high" category. For cadmium and tPCB, for example, the "high" concentrations correspond to 1.3 µg/g (dry) and 200 ng/g, respectively, as shown in Figure 1. For those and the rest of the chemicals being used in this report, the "high" concentrations are listed in Table 2.

Table 2. *Concentrations in sediment that are defined as "high" for NS&T sites. Concentrations are in units of µg/g (dry) for trace metals and ng/g (dry) for groups of organic compounds.*

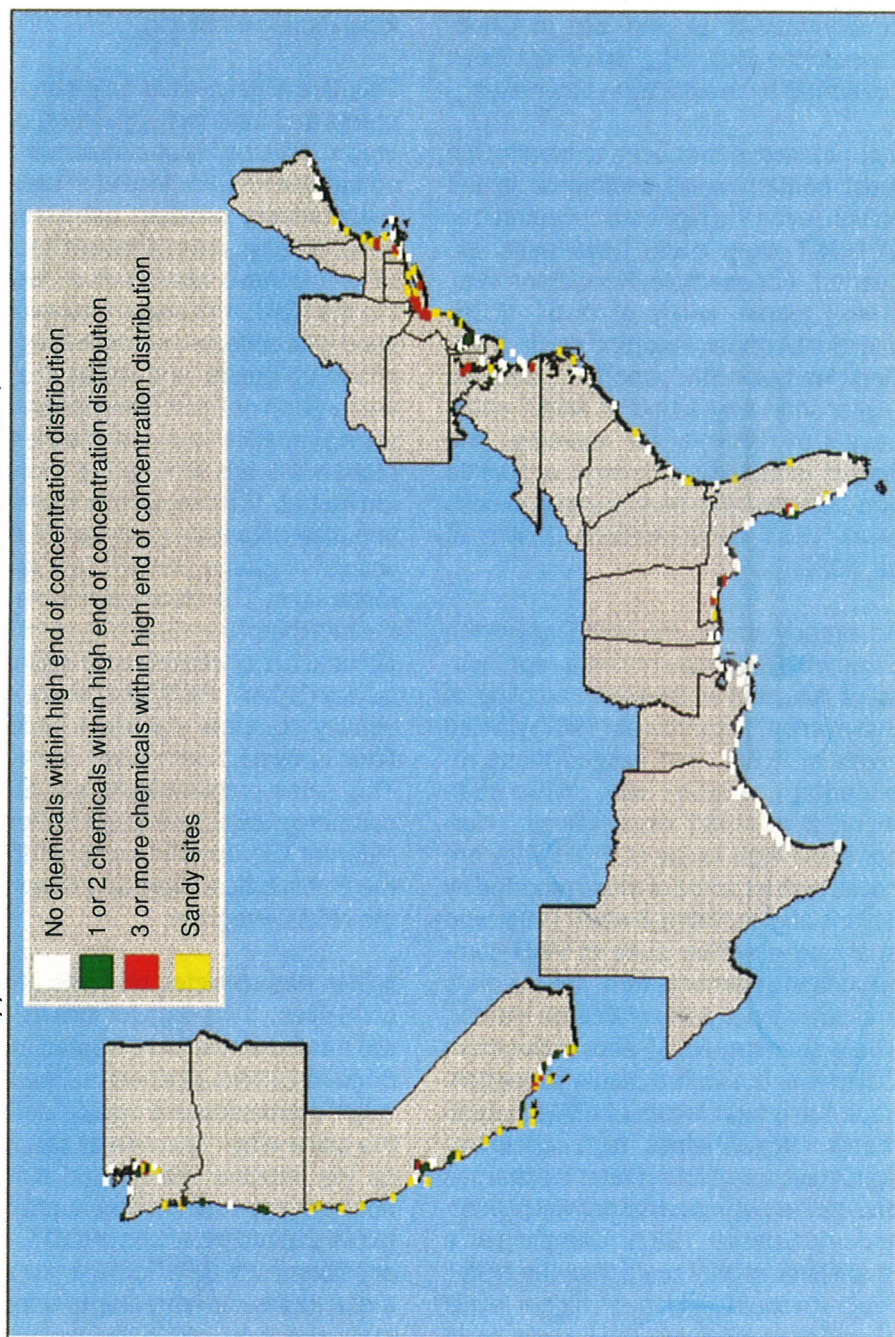
| <u>Chemical</u> | <u>High concentration</u> |
|-----------------|-------------------------------|
| Cd | 1.3 |
| Cr | 230 |
| Cu | 87 |
| Pb | 87 |
| Hg | 0.51 |
| Ag | 1.2 |
| Zn | 280 |
| tDDT | 40 |
| tCdane | 5.5 |
| tPCB | 200 |
| tPAH | 3900 |

That definition of "high" in a listing of all the NS&T sites can be used to indicate which ones have sediments with "high" concentrations of each chemical. The **Appendix** lists, in clockwise geographic sequence from Maine to Hawaii, all sites sampled in 1984 through 1989. It also indicates which chemicals, if any, had concentrations in the "high" range. That information is displayed graphically in **Figure 2** where NS&T sites are shown on a map of the U.S.

On a national scale, particularly for sites with three or more high concentrations, it is clear that contamination is associated with urbanized areas of the Northeast states; near San Diego, Los Angeles, and Seattle on the West Coast; and, except at a few sites, relatively rare in the Southeast and along the Gulf of Mexico Coast. The association of higher levels of sediment contamination with highly populated areas is not a surprising result. Nevertheless, it is important to note that these results come from sites that are considered to be "representative."

However, some NS&T sites may not be representative. One could doubt that the NS&T site in the Elizabeth River, VA, is typical of the southern end of Chesapeake Bay, because no chemicals are at high concentrations in sediments at other sites in that area. Sediments from sites near Los Angeles, the one off Palos Verdes and one in Santa Monica Bay, were taken within about a mile of the ends of discharge pipes from major sewage treatment plants. In those cases concentrations are very high, but other sites in the area, away from major discharges, also have sediments with high concentrations. The high concentrations in St. Andrews Bay, FL, were unex-

Figure 2. Geographic distribution of NS&T sites highlighting those with contaminants at high concentrations in sediments (see Appendix for more detailed information on individual sites)



pected but are confirmed by data from other sites near Panama City. The high concentrations at one site in Choc-tawhatchee Bay, FL, have not been confirmed by results from other sites.

If our objective had been to identify the most contaminated sediments in the Nation, even the highest concentrations at NS&T sites could have been exceeded. For example, Rodgers et al. (1985) found levels of tPAH, tPCB, cadmium, copper, lead, and zinc in Black Rock Harbor in Bridgeport, CT, that are higher than any found at NS&T sites. Levels of tPAH, silver, mercury, and lead in industrial waterways of Seattle and Tacoma have been found in excess of all NS&T concentrations (Malins et al., 1982).

As already stated, the NS&T site in the Elizabeth River may not be a representative site, but a detailed sampling of that river by Huggett et al. (1987) yielded levels of tPAH that exceed those reported at any NS&T site. While sediments at all 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. Nevertheless, high concentrations have been found near discharges and centers of industrial activity (Anderson and Gossett, 1987). Sampling on a much finer spatial scale than the NS&T Program could yield much higher levels of contamination, but they would be of little spatial significance from a national perspective. However, this fact illus-

trates the need for more detailed monitoring programs in selected areas for local decision-making.

Fifty-five sites yielded only sandy sediments and have not, therefore, contributed to defining the spatial distribution of contamination. For the most part this is not a severe problem because, on a national scale, those sites are near other sites that have fine-grained sediment. On the California coast, however, only sand was collected at most sites on the offshore islands and along the rocky shore north of Point Conception. There are no samples to reveal whether any of these sites are among the more contaminated. We can, with some reservation, use data from analyses of mussel tissues to gauge contaminant levels at these sites. The reservations are due to the fact that mussels and oysters do not accumulate chemicals with equal proficiency (NOAA, 1989), and so we cannot usually compare chemical concentrations in oysters with those in mussels. This same problem limits our ability to compare results across the Nation from different fish species sampled through the Benthic Surveillance component of the NS&T Program.

NOAA has collected mussels at a total of 96 sites. If the data for those 96 sites are treated as we have treated the data from the 232 fine-grained sediment sites, "high" concentrations can be defined in the same way. Mussels at sandy sites on the offshore islands and along the northern coast of California often contain high concentrations of tDDT. This is consistent with data from the few sites in that area that did have fine-grained sediment and is related to the major historical discharges of DDT from production facilities in Los Angeles. Except for

DDT, however, mussels at these sites are not highly contaminated. If fine-grained sediment had been available, it is unlikely that their chemical concentrations would be among the high concentrations.

NS&T sites along the stretch of coast from Point Conception, CA, to the Hood Canal, WA, provide an example of sediment concentrations that are high due to natural, rather than human, causes. Levels of human population and industrialization are low, but a few metals appear at high concentrations. Chromium concentrations in sediments are high, most likely because minerals forming the bedrock of that region are also enriched in chromium (USGS, 1981). High chromium concentrations in sediments elsewhere in the nation can be attributed to human activity and considered to be contamination, but in the Northwest chromium is naturally high and is not a contaminant. This point will become important as we discuss levels of chemicals in sediments that have been found to produce biological responses.

Biological Effects of Contaminants

A crucially important aspect of chemical contamination is whether or not it is causing any biological effects. One approach to that question is to collect and examine organisms living at each site. The other is to expose test organisms to sediment samples and use the organism's response as a measure of toxicity. Both methods are being used in NOAA's NS&T Program.

The presence of tumors in fish is usually interpreted as a response to contamina-

tion (Susani, 1986), and livers of fish collected at all NS&T sites have been examined for them. They were found in only 36 of the approximately 5,600 fish examined between 1984 and 1986. Fourteen of those were found in winter flounder in Boston Harbor, MA, and five in English sole in Elliott Bay, WA. These two areas do appear in the Appendix and Figure 2 as having sediments with high concentrations of chemicals. Two fish with liver tumors were found at each of four other sites: Raritan Bay, NJ; Great Bay, NJ; Bodega Bay, CA; and Commencement Bay, WA. Two of those sites, Great Bay and Bodega Bay, have not shown evidence of chemical contamination. So, while there is a general connection between contamination and liver tumors in fish, tumors are not always found in places where contamination is high, and they are occasionally found in what might be considered uncontaminated areas.

Complicating factors include the facts that older fish 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, the focus cannot be on a single species, i.e., no single species of fish is found at all sites. The infrequent occurrence of liver tumors is related to the species examined, but is due also to liver tumors being an extreme response to contamination and the fact that NS&T sites are not isolated areas of extreme contamination.

Recent work by Long and Morgan (1990)

allows us to extrapolate from chemical data describing the nationwide distribution of contamination to give us a sense of the distribution of sediment toxicity. They reviewed 150 scientific papers and reports on sediment toxicity and found 85 with data on both biological response and chemical concentrations in the sediment. They examined all of this information and found chemical concentrations, listed in **Table 3**, that correspond to concentrations above which effects were frequently observed. Since the values in Table 3 (except for chromium) are all greater than those defined in this report as "high" concentrations, it follows that sediment toxicity will be found at fewer sites than are highlighted in the Appendix and Figure 2. The dichotomy between "high" and "possibly biologically effective" concentrations is actually somewhat larger because the sediment data used by Long and Morgan were not adjusted for their sand content. The middle column in Table 3 shows that the "high" NS&T concentrations would have been about 40% lower if the data had not been adjusted.

As noted in Table 3, the exception of chromium is probably due to its naturally high levels in the Northwest combined with the fact that many tested sediments have been collected in that region. As Long and Morgan indicated, chromium probably was not causing toxicity in any of the tested sediments.

This extrapolation from chemistry to biological effect cannot be accepted unequivocally. The exhaustive compilation of reports that Long and Morgan used included bioassays based on different organisms, different test procedures, and different indicators of effect (usually death). The list of reports in-

cluded studies where chemicals were added to sediments, cases where sediments were tested as taken from the field, and cases where toxicity was estimated from calculations of chemical concentrations in the pore water of sediments. Finally, it included reports where sediment quality was judged on the basis of the species of organisms found (or not found) living in association with it. All of these differences make comparisons precarious. Nevertheless, the conclusion that sediment toxicity is not widespread is consistent with results from NS&T studies that tested sediment toxicity.

Scott (1989) collected sediment at NS&T sites in western Long Island Sound and in the Hudson/Raritan estuary that are listed in the Appendix as having sediment with high levels of contamination. In a standard bioassay based on 10-day survival of amphipods these sediments proved nontoxic. It may be that toxic sediments are found in only very localized and highly contaminated places. For example, Rodgerson et al. (1985), using that same amphipod test, did find toxicity in Black Rock Harbor near Bridgeport, CT, a site already mentioned as being highly contaminated.

While the NS&T sediment data do not indicate high levels of contamination in San Francisco Bay, Chapman et al. (1987) and Long and Buchman (1989) did find toxic responses to sediments taken from Islais Waterway and the inner parts of Oakland Harbor, two highly industrialized locations within San Francisco Bay. Swartz et al. (1982) found a range of toxic responses within the industrial waterways of Tacoma, WA, off Commencement Bay. There is no question that sediment toxicity exists in

Table 3. Comparison of "high" concentrations with concentrations found by Long and Morgan (1990) to usually coincide with a toxic response by most species.

| <u>Chemical</u> | <u>"High"</u> concentrations | | <u>Possible biological effect concentrations^c</u> |
|-----------------|---------------------------------|-------------------------------|--|
| | <u>adjusted^a</u> | <u>unadjusted^b</u> | |
| Cd | 1.3 | 0.72 | 9 |
| Cr | 230 | 135 | 145 ^d |
| Cu | 87 | 55 | 390 |
| Pb | 87 | 52 | 110 |
| Hg | 0.51 | 0.30 | 1.3 |
| Ag | 1.2 | 0.74 | 2.2 |
| Zn | 280 | 172 | 270 |
| | | | |
| tDDT | 40 | 24 | 350 |
| tPAH | 3900 | 2400 | 35000 |
| tCdane | 5.5 | 3.2 | 6 |
| tPCB | 200 | 120 | 400 |

^a High concentrations from Table 2, units are µg/g-dry for trace metals and ng/g-dry for groups of organic compounds

^b High concentrations derived, like those in Table 2, from lognormal distributions of concentrations but without adjusting those concentrations for their sand content. (As in the original calculation, all samples with more than 80% sand were excluded from consideration.)

^c Median concentrations in the compilation of Long and Morgan (1990) and defined by them as concentrations that would probably cause an adverse response in most benthic organisms

^d Chromium concentrations of 145 µg/g and higher are common in the Northwest because of mineralogy. It is probable that the concentration of 145 µg/g is the median of the tested values because so many of the tested sediments were collected in the Northwest. Long and Morgan (1990) concluded that, in tested sediments, Cr was probably not exerting any biological response at any concentration.

some places. However, sediment toxicity is not usually found at NS&T sites because they are chosen to be representative of more than isolated locations.

It appears that, on a national scale, biological effects are restricted to extremely contaminated and spatially limited locales. That result is, of course, a welcome assessment of the status of coastal contamination. However, it is very important to continue to test for possible effects, especially non-lethal ones.

Ongoing projects at the NMFS Environmental Conservation Division in Seattle 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 the species. For example, while there is not yet a nationwide set of data, livers of fish are being biochemically examined for the activity of enzymes produced to metabolize organic contaminants. Direct measurements of the reproductive success of fish are being made in selected locations.

The NS&T Program serves as an experimental matrix in which to find biological responses that are being caused by chemical contamination. Research into effects of contaminants is not restricted to NS&T sites. In fact, samples are deliberately collected in very contaminated areas to test whether the hypothesized response occurs under extreme conditions. Once it is known that a response occurs under these conditions, it will be possible to incorporate the measurement into NOAA's national monitoring program.

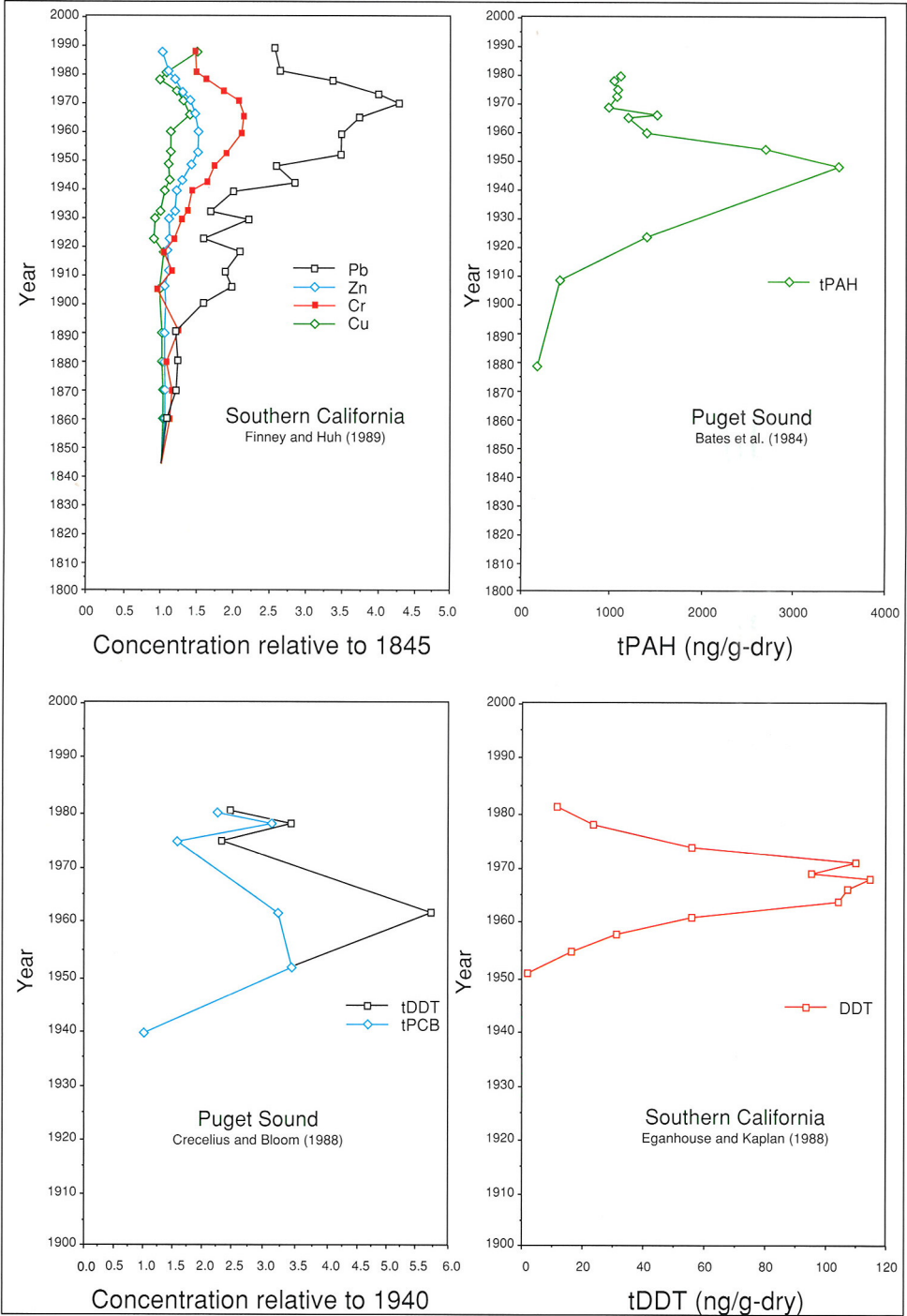
Temporal Trends of Contaminants in Sediments

Sediment data can be used to describe the spatial distribution of contamination, but periodic analyses of surface sediments do not reveal temporal trends. Organisms on the seafloor mix newly arrived particles into the existing sediment. When the NS&T Program samples the upper one to two centimeters of sediment, we do not know how much time has been integrated into that sample. It could be, for example, that the upper 10 cm of sediment are all mixed together and that sediment is delivered slowly at that site. Quite possibly, a chemical concentration of the surface represents a 20-year average. Getting chronological data from sediments requires a knowledge of rates of particle deposition and rates of sediment mixing.

It is possible, under certain circumstances, to find sediment that can be specially examined so that layers in vertical sections can be associated with a sequence of years. Generally, the objective is to apply radiological dating techniques to sediments that are not overly disturbed by biological activity and have sufficiently rapid rates of sedimentation. The NS&T Program has begun investigations in this field, but there are data collected in other programs that allow an important general observation about trends in contamination.

Figure 3 contains data extracted from a number of scientific papers. The important point is that contaminant concentrations in sediments, except possibly for copper, have begun to decrease

Figure 3. Chronological profiles of chemical concentrations in sediment cores



since the late 1970s or earlier. This point needs to be substantiated with more data from sediment cores. The data in Figure 3 were chosen because the chronologies extend to at least 1980. There are many reports where the chronologies end in the 1970s. In these latter cases, contaminant concentrations that had been increasing since the industrial revolution appear to have stopped increasing, but evidence of decreases is lacking. The reason for studying new cores is to verify or refute the hypothesis that contamination has been decreasing over the last decade.

Temporal Trends of Contaminants in Molluscan Tissue

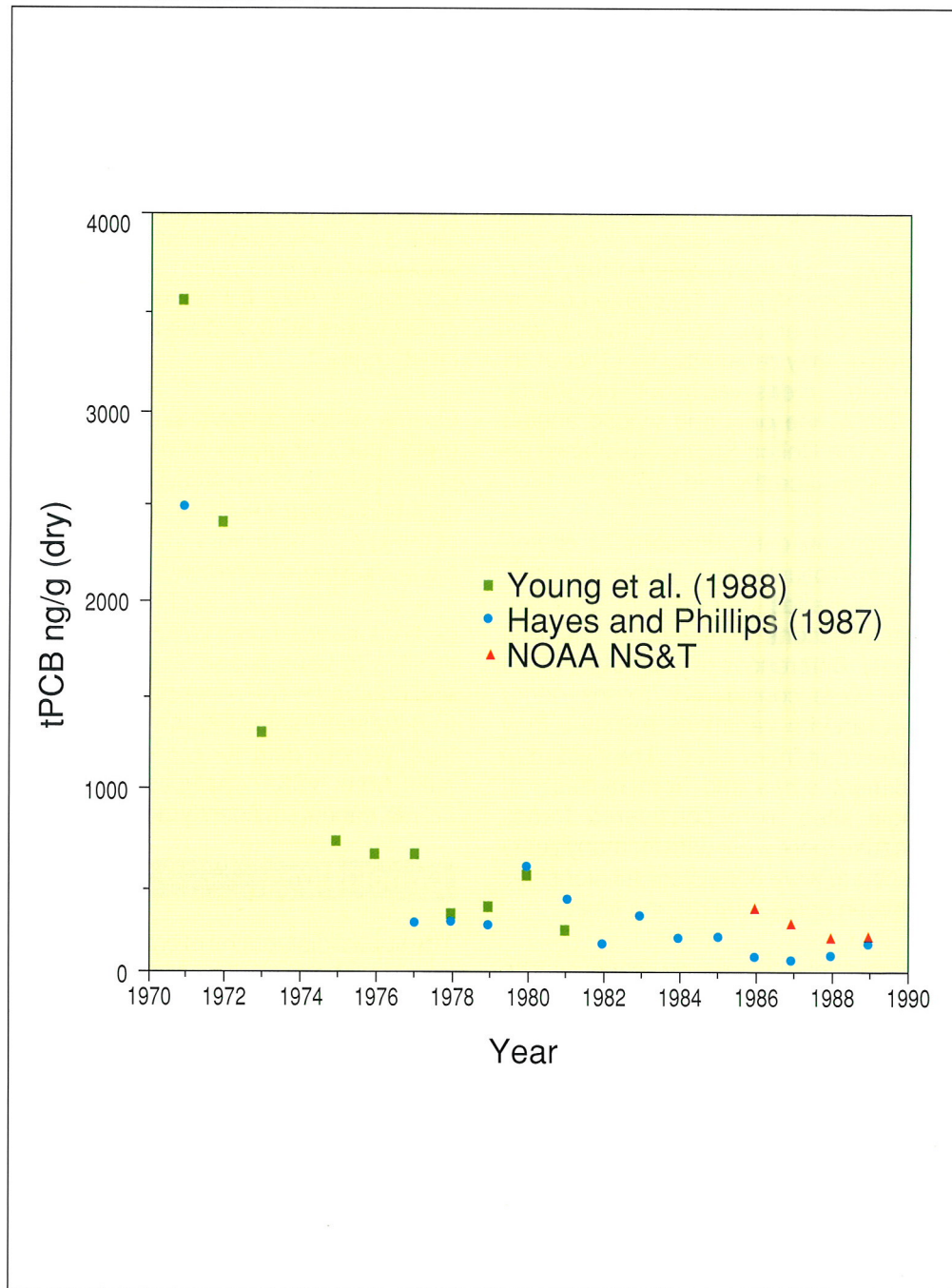
Except under special circumstances, one cannot expect samples of surface sediment collected at a single site to differ on an annual basis even if contaminant inputs change. Mussels and oysters, on the other hand, can change their contaminant levels in response to changes in their surroundings (Roesijadi et al., 1984; Pruell et al., 1987). This and the fact that they are immobile makes them ideal for monitoring changes in chemical concentrations in the coastal environment.

Figure 4 is an 18-year record of tPCB in mussels at the NS&T site in Royal Palms Park on the Palos Verdes coast of Los Angeles. It shows a dramatic decrease that began in 1971 when the U.S. began to phase out PCB use. The record has been constructed by combining three sets of data from three separate programs. Sericano et al. (1990) have combined data from diverse sources to show similarly dramatic historical changes in the average tDDT concen-

tration in oysters in the Gulf of Mexico. In that case as well as in Figure 4, the major decreases occurred in the early 1970s. Unlike trends found in sediment cores, where all sections would have been analyzed by a single laboratory, trends from annual collections of mollusks have had to be based on data from several sources. As explained by Stout (1986), analytical artifacts must be suspected because chemical techniques for quantifying organic compounds in environmental samples have improved dramatically over the same period of the apparent decreases. Nevertheless, the timing and the large magnitude of the decreases lends credibility to the argument that major decreases have occurred in concentrations of now-banned chlorinated compounds.

Decadal trends in trace metal contamination have been sought by comparing NS&T data of 1986 through 1988 with data from analyses of mussels and oysters collected in 1976 through 1978 by a previous "mussel watch" program (Goldberg et al., 1983) sponsored by the U.S. Environmental Protection Agency (EPA). Statistically, since the earlier program collected a single sample each year, it was necessary to aggregate the three years of data for each decade. With that aggregated data it was possible to estimate differences in trace metal concentrations in mollusks at the 50 sites that were common to both programs. **Figure 5** shows the 50 site locations and demonstrates the overwhelming (39 out of 50) dominance of decadal decreases in lead. That excess of decreases over increases or lack of difference is itself statistically sufficient to declare a national decrease in lead concentrations since the late 1970s, a result consistent with the phase-out of

Figure 4. Concentrations of tPCB in mussels collected at Palos Verdes State Park since 1971



leaded gasoline.

For copper, cadmium, silver, and zinc the directions of change were not overwhelmingly in a single direction. However, for cadmium, there were 12 sites where the 1970s data were statistically different from the NS&T data, and in 11 of those cases the 1970s concentrations were higher. Conversely, for 18 of the 22 sites where copper concentrations were statistically different they were lower in the 1970s. As pointed out by Lauenstein et al. (1990), the copper increase may reflect the fact that of all the metals measured in both programs, copper is the only one whose annual use in the United States has shown an increase since the mid-1970s.

Changes in concentrations of chemicals in mollusks at a single site may indicate trends, but they could be random variations unrelated to chemical inputs. Differences over a 10-year time span were considered trends rather than random variations because they occurred at many sites. The data from sediment cores and from mussels at single sites were considered trends because there were data for many years and there were consistent relationships between concentration and time.

Changes over short time scales may be found in data on mussels and oysters that have been collected by NOAA at 12-month intervals (in the late winter to early spring) at NS&T sites since 1986. This is the beginning of what will become an increasingly valuable set of data. At the early stages, however, it is difficult to separate changes that signify trends from those that do not.

The first three years of concentration

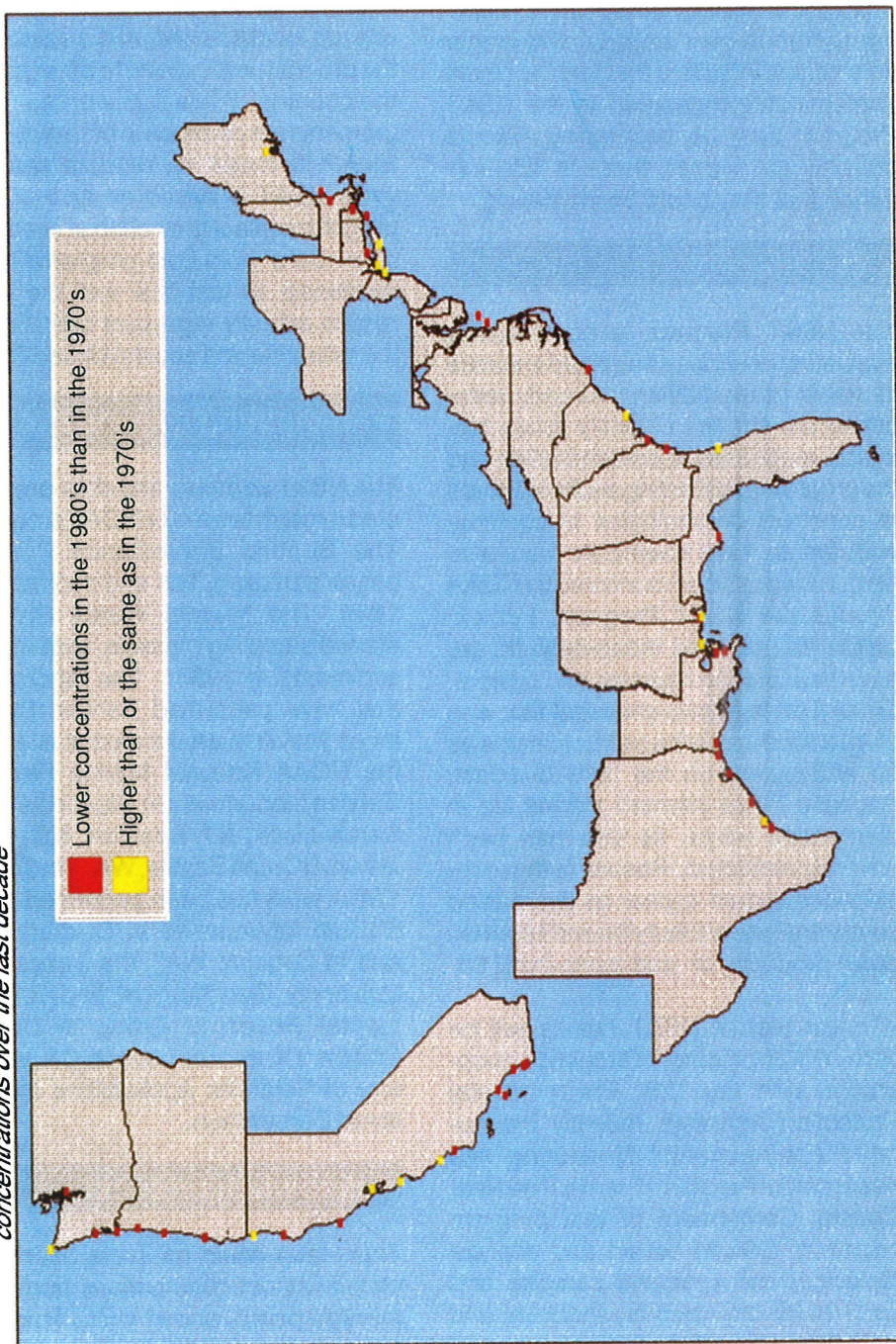
data for 132 sites have been examined for cases where there were statistically significant differences and where the concentration in the middle year was not the highest or lowest (NOAA, 1989). This restriction was imposed so that possible trends could only be those cases where concentrations had continuously increased or decreased over the three years. Even with that restriction, trends cannot be confidently identified at single sites with only three years of data. Some confidence may be associated with cases where groups of sites show similar changes.

On the whole, concentrations of each trace metal or group of organic compounds changed in a statistically significant and uniform fashion at about 10% of the sites. There were a few groups of sites showing a common change. For example, cadmium and chlordane concentrations were decreasing in mussels at sites in Long Island Sound; copper and mercury were increasing at sites in the Hudson/Raritan Estuary. Nevertheless, without data for more years, results from NS&T analyses, by themselves, cannot confidently identify trends.

Conclusions

The spatial distribution of contamination throughout the coastal United States, revealed through analyses of surface sediments, shows the higher levels to be characteristic of urbanized estuaries. Those high levels, however, are generally lower than those expected to cause sediment toxicity, and among the NS&T sites, biological responses to contamination, such as liver tumors in fish or sediment toxicity, have not been commonly found. Temporal trends in contaminant levels at NS&T sites are

Figure 5. Map of sites used for NS&T and previous mussel watch projects and directions of changes in lead concentrations over the last decade



beginning to be drawn from annual analyses of mussels and oysters. Looking at trends over decadal and longer time scales indicates that levels of most contaminants measured in the NS&T Program may be decreasing. Except possibly for copper, there is little evidence that they could be increasing.

New Directions

The NS&T Program is continuously evaluating new ideas that could enhance its value to the Nation. Already mentioned are efforts to identify subtle biological responses to contamination and the program of collecting sediment cores to construct chronologies of contaminant inputs. New chemicals have also been added to those monitored by NOAA through the NS&T Program. For example, tri-, di-, and mono-butyl tin are now measured in mussels and oysters. Tributyl tin is a toxic chemical that was often added to marine paints to serve as an anti-foulant on the hulls of boats. Because it has harmed marine life in unintended ways, its use has been severely restricted. Results of those restrictions should appear as decreasing concentrations of tributyltin and its breakdown products, di- and mono-butyl tin.

A major test of NS&T results will be derived from strong interagency cooperation with the U.S. Environmental Protection Agency's recently inaugurated Environmental Monitoring and Assessment Program (EMAP). The Near Coastal Component of that program began in 1990 to collect and analyze biological and sediment samples and perform toxicity tests on sediment and water from about 200 randomly selected estuarine sites between Chesapeake Bay and Cape Cod. For the next few

years that sampling intensity will be repeated at another set of randomly chosen locations and, at the same time, the program will expand to other parts of the country. While many of the measurements are common to both programs, EMAP is based on random sampling while NS&T is based on annual sampling throughout the nation at fixed locations. Data from both programs will be examined to test how well the NS&T results actually represent conditions in the estuarine and coastal United States.

Participating Organizations

The NS&T samples and data are gathered through two major NOAA programs. The Benthic Surveillance Program began sampling fish and sediments in 1984. The Mussel Watch Program started sampling mussels, oysters, and sediments in 1986. The laboratories that have performed Benthic Surveillance activities are located at five sites: the NOAA National Marine Fisheries Service laboratories in Gloucester, MA; Sandy Hook, NJ; Beaufort, NC; Charleston, SC; and Seattle, WA. The Mussel Watch work has been performed at the Battelle laboratories in Duxbury, MA, and in Sequim, WA; the Texas A&M University Geochemical and Environmental Research Group in College Station, TX; and the LaJolla, CA, laboratory of Scientific Applications International Corporation.

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This report could not have been completed without contributions from innumerable people and organizations. The several laboratories responsible for collection and analysis of samples have been mentioned. Members of the staff

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Appendix

The appendix lists all NS&T sites sampled in the Benthic Surveillance Program from 1984-1986 and the Mussel Watch Program from 1986-1989. Benthic Surveillance sites are those with only a general site name. Mussel Watch sites are given both a general and specific site designation. If all sediment

samples from a site contained more than 80% sand-sized particles, that site is indicated to be sandy and chemical data from it have not been used when comparing among sites. The last columns indicate which chemical concentrations, if any, at a site exceeded the "high" concentrations listed in Table 2.

Errata. Four sites listed as "sandy" should have been listed as follows:

[illegible]

Appendix. NS&T Sites: Benthic Surveillance 1984-86; Mussel Watch 1986-89

| General Site Name | Specific Site Name | St. | Type | Cd | Cr | Cu | Pb | Hg | Ag | Zn | tDDT | tCdane | tPCB | tPAH |
|-----------------------|--------------------|-----|-------|----|----|----|----|----|----|----|------|--------|------|------|
| NORTH ATLANTIC | | | | | | | | | | | | | | |
| Machias Bay | | ME | | | | | | | | | | | | |
| Frenchmans Bay | | ME | | | | | | | | | | | | |
| Penobscot Bay | | ME | | | | | | | | | | | | |
| Penobscot Bay | Sears Island | ME | | | | | | | | | | | | |
| Penobscot Bay | Pickering Island | ME | | | | | | | | | | | | • |
| Merriconeag Sound | Stover Point | ME | Sandy | | | | | | | | | | | |
| Casco Bay | | ME | | | | | | | | | | | | |
| Cape Arundel | Kennebunkport | ME | Sandy | | | | | | | | | | | |
| Merrimac River | | MA | Sandy | | | | | | | | | | | |
| Cape Ann | Gap Head | MA | | | | | • | | | | | | | • |
| Salem Harbor | Folger Point | MA | Sandy | | | | | | | | | | | |
| Salem Harbor | | MA | | • | • | • | • | • | • | • | • | • | • | • |
| Boston Harbor | Deer Island | MA | | • | • | • | • | • | • | | | • | • | • |
| Boston Harbor | Dorchester Bay | MA | | • | • | • | • | • | • | | • | • | • | • |
| Boston Harbor | Hingham Bay | MA | | • | • | • | • | • | • | | | • | • | |
| Boston Harbor | Brewster Island | MA | Sandy | | | | | | | | | | | |
| Boston Harbor | | MA | | • | • | • | • | • | • | • | • | • | • | • |
| Quincy Bay | | MA | | | • | • | • | • | • | | • | • | • | • |
| Duxbury Bay | Clarks Island | MA | Sandy | | | | | | | | | | | |

| General Site Name | Specific Site Name | St. | Type | Cd | Cr | Cu | Pb | Hg | Ag | Zn | tDDT | tCdane | tPCB | tPAH |
|------------------------|--------------------|-----|-------|----|----|----|----|----|----|----|------|--------|------|------|
| MID-ATLANTIC | | | | | | | | | | | | | | |
| Cape Cod | Nauset Harbor | MA | | | | | | | | | | | | |
| Buzzards Bay | Cape Cod Canal | MA | | | | | | | | | | | | |
| Buzzards Bay | Round Hill | MA | | | | | | | | | | | • | |
| Buzzards Bay | Angelica Rock | MA | | | | | • | | • | | | | • | • |
| Buzzards Bay | Goosebury Neck | MA | | | | | | | | | | | | |
| Buzzards Bay | | MA | | | | | | | | | | | • | |
| Narragansett Bay | Mount Hope Bay | RI | | | | • | • | • | • | | | | | |
| Narragansett Bay | Patience Is. | RI | Sandy | | | | | | | | | | | |
| Narragansett Bay | Dyer Island | RI | | | | | • | | • | | | | | • |
| Narragansett Bay | Dutch Island | RI | | | | | | | | | | | | |
| Narragansett Bay | | RI | | | | • | • | | • | | | | • | |
| Block Island | Block Island | RI | | | | | | | | | | | | |
| Long Island | Gardiners Bay | NY | Sandy | | | | | | | | | | | |
| East Long Island Sound | | CT | Sandy | | | | | | | | | | | |
| Long Island Sound | Connecticut River | CT | | | | | | | | | | | • | • |
| Long Island Sound | New Haven | CT | Sandy | | | | | | | | | | | |
| Long Island Sound | Housatonic River | CT | Sandy | | | | | | | | | | | |
| Long Island Sound | Sheffield Island | CT | | • | | • | • | • | • | • | | | | • |
| West Long Island Sound | | NY | | | | | • | • | • | • | | | • | • |
| Long Island Sound | Huntington Harbor | NY | | • | | • | • | | • | • | | | | |
| Long Island Sound | Port Jefferson | NY | Sandy | | | | | | | | | | | |
| Long Island Sound | Mamaroneck | NY | | • | | • | • | • | • | • | | • | | • |
| Long Island Sound | Hempstead Harbor | NY | | • | | • | • | • | • | • | • | • | • | • |
| Long Island Sound | Throgs Neck | NY | | • | | • | • | • | • | • | • | • | • | • |
| Moriches Bay | Tuthill Point | NY | | | | | • | • | • | | • | | | |

| General Site Name | Specific Site Name | St. | Type | Cd | Cr | Cu | Pb | Hg | Ag | Zn | tDDT | tCdane | tPCB | tPAH |
|------------------------|---------------------|-----|-------|----|----|----|----|----|----|----|------|--------|------|------|
| Hudson/Raritan Estuary | Jamaica Bay | NY | | • | • | | • | • | • | • | • | • | • | • |
| Hudson/Raritan Estuary | Upper Bay | NY | | • | | • | • | • | • | • | | | • | • |
| Hudson/Raritan Estuary | Lower Bay | NY | | • | • | • | • | • | • | • | • | • | • | • |
| Hudson/Raritan Estuary | Raritan Bay | NJ | | • | • | • | • | • | • | • | • | • | • | • |
| Raritan Bay | | NJ | | • | • | • | • | • | • | • | • | • | • | • |
| New York Bight | Sandy Hook | NJ | | • | • | • | • | • | • | • | • | • | • | • |
| New York Bight | Long Branch | NJ | Sandy | | | | | | | | | | | |
| New York Bight | Shark River | NJ | Sandy | | | | | | | | | | | |
| Barnegat Inlet | Barnegat Light | NJ | Sandy | | | | | | | | | | | |
| Great Bay | | NJ | | | | | | • | | | | | | |
| Absecon Inlet | Atlantic City | NJ | Sandy | | | | | | | | | | | |
| Delaware Bay | Cape May | NJ | | | | | | | • | • | | | | |
| Delaware Bay | | DE | | | | | | | | | | • | • | |
| Delaware Bay | False Egg Island Pt | NJ | | | | | | | | | • | | | |
| Delaware Bay | Ben Davis Pt. Shl. | NJ | | | | | | | | | | | | |
| Delaware Bay | Arnolds Pt. Shoal | DE | | | | | | | | | | | | |
| Delaware Bay | Hope Creek | NJ | | | | | • | | | • | | | | |
| Delaware Bay | Woodland Beach | DE | | | | | | | | • | | | | |
| Delaware Bay | Kelly Island | DE | | | | | | | | | | | | |
| Delaware Bay | Cape Henlopen | MD | | | | | | | | | | | | |
| Baltimore Harbor | | MD | | • | • | • | • | • | • | • | | • | • | • |
| Upper Chesapeake Bay | | MD | | | | | | | | • | | | • | • |
| Chesapeake Bay | Bodkin Point | MD | | | | | | | | • | | | • | • |
| Chesapeake Bay | Mountain Point Bar | MD | | | | | | | | • | | | | • |
| Chesapeake Bay | Hackett Point Bar | MD | | | | | | | | • | | | | • |
| Chesapeake Bay | Choptank River | MD | | | | | | | | | | | | |

| General Site Name | Specific Site Name | St. | Type | Cd | Cr | Cu | Pb | Hg | Ag | Zn | tDDT | tCdane | tPCB | tPAH |
|------------------------|--------------------|-----|-------|----|----|----|----|----|----|----|------|--------|------|------|
| Savannah River Estuary | Tybee Island | GA | | | | | | | | | | | | |
| Sapelo Sound | Sapelo Island | GA | Sandy | | | | | | | | | | | |
| Sapelo Island | | GA | | | | | | | | | | | | |
| Altamaha River | Wolfe Island | GA | Sandy | | | | | | | | | | | |
| St. Johns River | Chicopit Bay | FL | | | | | | | | | | | | |
| St. Johns River | | FL | | | | | | | | | | | | • |
| Matanzas River | Crescent Beach | FL | Sandy | | | | | | | | | | | |
| Indian River | Sebastian River | FL | Sandy | | | | | | | | | | | |
| North Miami | Maule Lake | FL | | | | | | | | | | | | |
| Biscayne Bay | Princeton Canal | FL | | • | | | | | | | | | | |
| GULF OF MEXICO | | | | | | | | | | | | | | |
| Everglades | Faka Union Bay | FL | | | | | | | | | | | | |
| Rookery Bay | Henderson Creek | FL | | | | | | | | | | | | |
| Naples Bay | Naples Bay | FL | | | | | | | | | | | | |
| Charlotte Harbor | Bird Island | FL | | | | | | | | | | | | |
| Charlotte Harbor | Fort Meyers | FL | Sandy | | | | | | | | | | | |
| Charlotte Harbor | | FL | | | | | | | | | | | | |
| Tampa Bay | | FL | | | | | | | | | | | | |
| Tampa Bay | Mullet Key Bayou | FL | | | | | | | | | | • | | |
| Tampa Bay | Cockroach Bay | FL | Sandy | | | | | | | | | | | |
| Tampa Bay | Navarro Park | FL | | • | | | | | | | | • | | |
| Tampa Bay | Hillsborough Bay | FL | | • | | | • | | | | | | • | |
| Tampa Bay | Papys Bayou | FL | | | | | | | | | • | • | • | |
| Tampa Bay | O'Knight Airport | FL | | | | | • | | | | • | • | • | |
| Tampa Bay | Old Tampa Bay | FL | | | • | | | | | | | | | |

| General Site Name | Specific Site Name | St. | Type | Cd | Cr | Cu | Pb | Hg | Ag | Zn | tDDT | tCdane | tPCB | tPAH |
|---------------------|--------------------|-----|------|----|----|----|----|----|----|----|------|--------|------|------|
| Breton Sound | Bay Garderne | LA | | | | | | | | | | | | |
| Breton Sound | Sable Island | LA | | | | | | | | | | | | |
| Mississippi River | Tiger Pass | LA | | | | | | | | | | | | |
| Mississippi River | Pass a Loutre | LA | | | | | | | | | | | | |
| Barataria Bay | Bayou Saint Denis | LA | | | | | | | | | | | | |
| Barataria Bay | Turtle Bay | LA | | | | | | | | | | | | |
| Barataria Bay | Middle Bank | LA | | | | | | | | | | | | |
| Barataria Bay | | LA | | | | | | | | | | | | |
| Terrebonne Bay | Lake Felicity | LA | | | | | | | | | | | | |
| Terrebonne Bay | Lake Barre | LA | | | | | | | | | | | | |
| Caillou Lake | Caillou Lake | LA | | | | | | | | | | | | |
| Atchafalaya Bay | Oyster Bayou | LA | | | | | | | | | | | | |
| Vermillion Bay | Southwest Pass | LA | | | | | | | | | | | | |
| Joseph Harbor Bayou | Joseph Harbor Bay | LA | | | | | | | | | | | | |
| Calcasieu Lake | Lake Charles | LA | | | | | | | | | | | | |
| Calcasieu Lake | St. Johns Island | LA | | | | | | | | | | | | |
| Sabine Lake | Blue Buck Point | TX | | | | | | | | | | | | |
| East Cote Blanche | South Point | LA | | | | | | | | | | | | |
| Galveston Bay | Hanna Reef | TX | | | | | | | | | | | | |
| Galveston Bay | Ship Channel | TX | | | | | | | | | | | | |
| Galveston Bay | Yacht Club | TX | | | | | | | | | | | | |
| Galveston Bay | Todd's Dump | TX | | | | | | | | | | | | |
| Galveston Bay | Confederate Reef | TX | | | | | | | | | | | | |
| Galveston Bay | Offats Bayou | TX | | | | | | | | • | • | | | |
| Galveston Bay | | TX | | | | | | | | | | | | |
| Brazos River | Ferrport Surfside | TX | | | | | | | | | | | | |

| General Site Name | Specific Site Name | St. | Type | Cd | Cr | Cu | Pb | Hg | Ag | Zn | tDDT | tCdane | tPCB | tPAH |
|-----------------------|--------------------|-----|-------|----|----|----|----|----|----|----|------|--------|------|------|
| PACIFIC | | | | | | | | | | | | | | |
| Imperial Beach | North Jetty | CA | Sandy | | | | | | | | | | | |
| Outside San Diego Bay | | CA | | • | | | | | | | | | | |
| South San Diego Bay | | CA | | • | | • | • | • | • | • | | • | • | • |
| San Diego Bay | Coronado Bridge | CA | Sandy | | | | | | | | | | | |
| San Diego Bay | Harbor Island | CA | | • | | • | • | • | • | • | | | • | |
| North San Diego Bay | | | | | | • | • | • | • | • | | | • | • |
| Point Loma | Lighthouse | CA | | | | | | | | | • | | | |
| Mission Bay | Ventura Bridge | CA | Sandy | | | | | | | | | | | |
| La Jolla | Point La Jolla | CA | | | | | | | | | | | | |
| Oceanside | Beach Jetty | CA | | | | | | | | | • | | | |
| Dana Point | | CA | | | | | | | | | | | | |
| Newport Beach | Wedge Jetty | CA | | | | | | | | | • | | | |
| Anaheim Bay | West Jetty | CA | | | | | | | | | • | | | |
| Seal Beach | | CA | | | | | | • | | | | | | |
| Long Beach | | CA | | • | | • | • | | | • | • | • | • | |
| San Pedro Bay | | CA | | | | • | | | | | • | | • | |
| San Pedro Canyon | | CA | | • | • | • | | • | • | • | • | | • | |
| San Pedro Harbor | Fishing Pier | CA | | • | | • | | | | | • | | • | |
| Palos Verdes | Royal Palms Park | CA | | • | • | • | | • | • | • | • | • | • | |
| Santa Catalina Island | Bird Rock | CA | Sandy | | | | | | | | | | | |
| West Santa Monica Bay | | CA | | • | • | • | • | • | • | • | • | • | • | |
| East Santa Monica Bay | | CA | Sandy | | | | | | | | | | | |
| Marina Del Ray | South Jetty | CA | | | | | | | • | | • | | | |
| Point Dume | Point Dume | CA | | • | | | | | • | | • | | | |

[illegible]

| General Site Name | Specific Site Name | St. | Type | Cd | Cr | Cu | Pb | Hg | Ag | Zn | tDDT | tCdane | tPCB | tPAH |
|--------------------------|--------------------|-----|-------|----|----|----|----|----|----|----|------|--------|------|------|
| Humboldt Bay | | CA | | | • | | | | | | | | | |
| Klamath River | Flint Rock Head | CA | Sandy | | | | | | | | | | | |
| Point St. George | Point St. George | OR | Sandy | | | | | | | | | | | |
| Coos Bay | | OR | | • | | | | | | | | | | |
| Coos Bay | Coos Head | OR | | • | • | | | | | | | | | |
| Coos Bay | Russell Point | OR | | | • | | | | | | | | | |
| Yaquina Bay | Oneata Point | OR | | | | | | | | | | | | |
| Yaquina Head | Sally's Slough | OR | | • | • | | | | | | | | | |
| Tillamook Bay | Hobsonville Point | OR | | | • | • | | | | | | | | |
| Columbia River | Youngs Bay | OR | | | | | | | | • | | | | |
| Young's Bay | | OR | | | | | | | | | | | | |
| Columbia River | | OR | | • | | | | • | | • | | | | |
| Columbia River | North Jetty | WA | Sandy | | | | | | | | | | | |
| Gray's Harbor | Westport Jetty | WA | Sandy | | | | | | | | | | | |
| Straight of Juan de Fuca | Neah Bay | WA | | | • | | | | | | | | | |
| South Puget Sound | Budd Inlet | WA | | | | | | | | | | | | |
| Nisqually Reach | | WA | Sandy | | | | | | | | | | | |
| Commencement Bay | | WA | | | | | | | | | | | | |
| Commencement Bay | Brown's Point | WA | | | | | | | | | | | | |
| Puget Sound | South Seattle | WA | | | • | | | | | | | | | |
| Elliott Bay | Four-Mile Rock | WA | Sandy | | | | | | | | | | | • |
| Elliott Bay | | WA | | • | | • | • | • | | • | | | • | |
| Sinclair Inlet | Waterman Point | WA | | | • | • | • | • | • | | | | | |
| Puget Sound | Hood Canal | WA | | | • | | | | | • | | | | |
| Whidbey Island | Possession Point | WA | | | | | | | | | | | | |
| Puget Sound | Everett Harbor | WA | | | • | | | | | | | | | |

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