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PCB AND CHLORINATED PESTICIDE CONTAMINATION IN U.S. FISH AND SHELLFISH: A HISTORICAL ASSESSMENT REPORT

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ABSTRACT. Geographic and long-term trends of PCB and chlorinated pesticide contamination were examined in U.S. coastal fish and shellfish using existing historical data. The study identified 35,000 samples from over 300 projects, surveys, and monitoring programs. The 35,000 samples represent over 540 species collected between 1940 and 1985.

Despite many sources of variability (tissue type, species diversity, season, size, age, sex, sample size, and analytical techniques) trends are evident when at least some of these factors are taken into account and when data encompass long-time intervals, such as decades.

Using data from 13 national and seminational programs, involving 12,000 records or one-third of the total data base, it was possible to map regional and nationwide patterns of fish or shellfish contamination for polychlorinated biphenyls (PCBs), DDT, dieldrin, and, to a limited extent, other pesticides during three data-rich periods: 1965-72, 1976-77, and 1984. Using both federal and locally generated data, it was also possible to discover trends in specific bays for specific chemicals and target species. Small-scale geographic patterns, long-term (20-year) declines and short- term variations of PCB, DDT, and dieldrin contamination were apparent in the Chesapeake Bay, San Francisco Bay, and in the Arroyo Colorado adjacent to southern Laguna Madre, Texas. Past surveys were nationally comprehensive for bivalves but not necessarily for flatfish--two groups of organisms monitored in NOAA's NS&T Program. Finally, it was possible to develop a 15- to 20-year history of pesticide and PCB contamination in stocks of several commercially-important species: striped bass and Atlantic and Gulf menhaden.

Decreasing DDT contamination was clearly evident on a nationwide basis during the 1970s. Decreasing contamination by PCBs, dieldrin, and toxaphene was evident for selected species and sites. Dramatic nationwide declines of DDT contamination and local declines of PCB contamination have already occurred and the annual rate of declines are now small. Toxaphene, DCPA (dacthal), endosulfan, endrin, and pentachlorophenl (PCP) should be resurveyed on a national basis to confirm that past or recent hot spots are localized and that previously identified declines have continued. Mirex, aldrin, HCB, methcxyclor, heptachlor, and perhaps lindane may have once been important estuarine contaminants, but this is not currently the case.

Additional monitoring should focus on edible tissues (such as muscle) of larger coastal predatory fish and on poorly surveyed areas such as the Florida east coast, Oregon, Alaska, and Hawaii and high seas fishes. A nationally centralized and easily accessible PCB and pesticide data base should be completed and used to receive and process new data from local, state, and federal programs on a continuing basis.

1.0 INTRODUCTION

Polychlorinated biphenyls (PCBs) and chlorinated pesticides comprise a few of the thousands of synthetic organic chemicals that have been used widely throughout the United States for nearly half a century. All are toxic to some degree to invertebrates, fish, birds, mammals, and humans. Some are carcinogenic (cancer promoting). Although they have been released into the environment for decades, PCBs and most of the chlorinated pesticides have either been banned or severely limited in use during the past 15 years. Nevertheless, they persist and are still found in terrestrial, aquatic, and marine plants and animals throughout the United States and the world. By virtue of their toxicity, carcinogenicity, and continued occurrence, PCBs and pesticides such as DDT, chlordane, and toxaphene, are prominent and pivotal factors in decisions concerning the clean up of dumpsites, disposal of contaminated dredged bottom sediments, the treatment of municipal waste, and the closure or posting of fishery grounds and fishery resources (Figure 1.1).

What has been happening to these chemicals in the coastal zone? Have they declined nationally, if not locally, in response to the controls? Unfortunately, it is very difficult to find answers to these questions. No national survey has been mounted to document trends continually over the past two decades. There have been occasional national surveys and these chemicals have been routinely measured in hundreds of regional and local monitoring programs, but with the exception of PCBs (Stout, 1987), these data have not been brought together to develop a unified picture of the history of PCB and pesticide contamination of marine life of the U.S. sea coasts.

This report begins this long needed synthesis of nationwide trends of PCB and chlorinated pesticides contamination of U.S. coastal fish and shellfish. It is based on the results of a nationwide search for data from published reports and unpublished records on file with various federal, state, and local agencies (Mearns, 1986; NOAA, 1986).

The intent of the report is to provide interested readers with a general view of the abundance and distribution of existing data, how it can be reassembled to learn more about what has been happening with these chemicals in U.S. marine organisms over the past several decades, and how monitoring might be more clearly focused to allow for continued assessment of trends. Specifically, this report:

1. Describes what chemicals have been looked for, how much data actually exists for each, and how it is distributed among locations, species, and types of tissues.

2. Shows how different data sets can be reassembled to develop contaminant histories at several levels of concern including national, regional, local, and for specific resource species.

3. Provides guidance for future monitoring by identifying contaminants, locations, species, and tissues that need additional assessment.

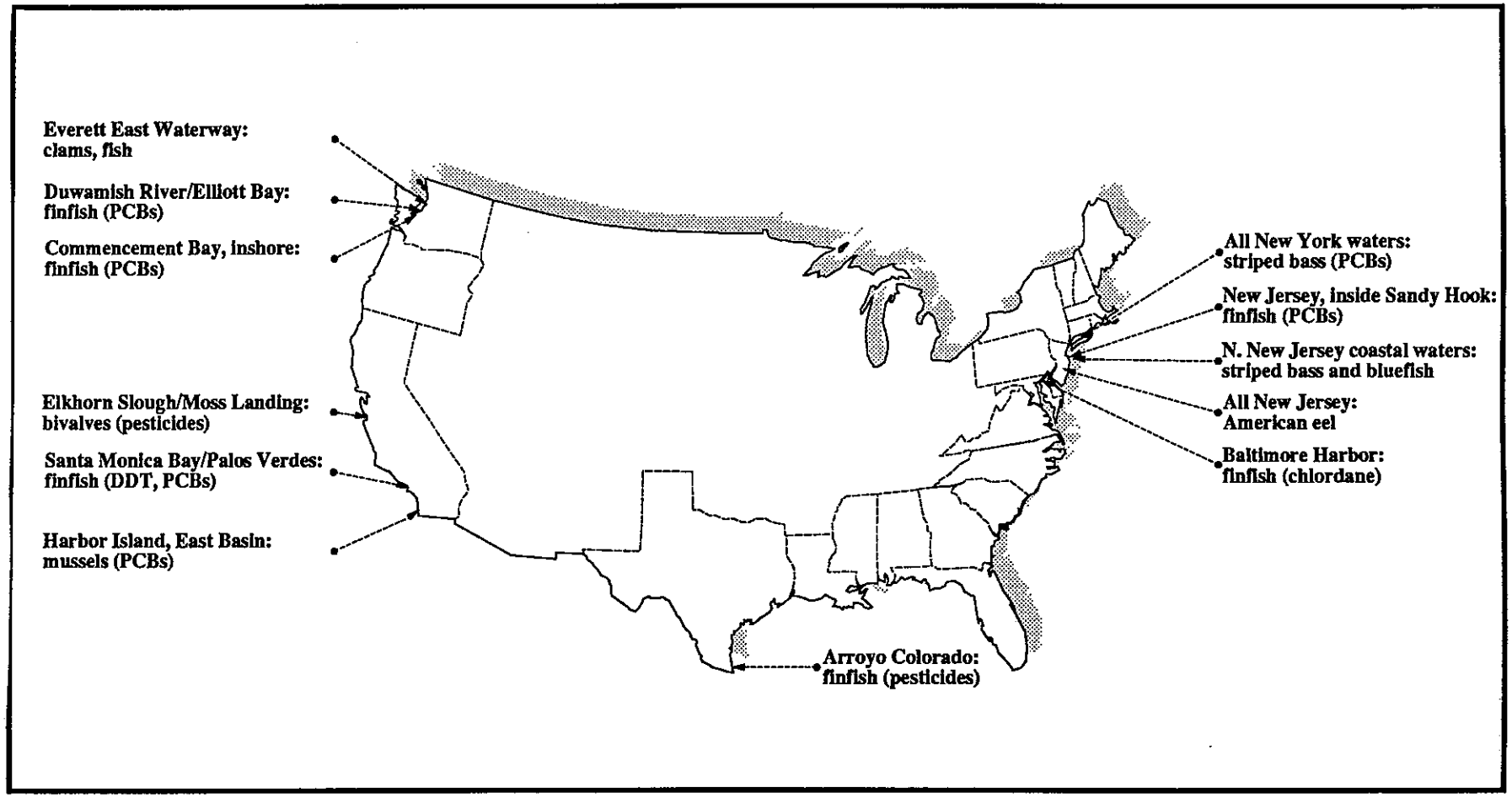


Figure 1.1. U.S. coastal sites sustaining marine fishery advisories due to unacceptable PCB or chlorinated pesticide contamination, 1980-87. Some advisories may be discontinued .

This report is principally a descriptive, not a statistical, effort. It is intended to provide the interested reader with a reminder of past monitoring and sampling efforts, with an overview of general results of published and unpublished survey data; and with identification of obvious, order-of-magnitude, geographical patterns, and long-term trends. In short, it is intended to serve as a provocative framework for further analysis and for design of future monitoring or survey efforts that could complete our knowledge of trends.

2.0 METHODS

To prepare this report, it was necessary to conduct a nationwide data search, acquire appropriate data sets, organize them into readily accessible files, and reassemble acceptable records into comparable units of time, geography, species, and tissue types. These points are outlined in more detail below.

2.1 Project Scope

Contaminants selected for review included PCBs and seven classes of historically important chlorinated pesticides. These are: (a) DDT and structurally related chemicals such as methoxyclor, (b) the cyclodiene pesticides including chemicals such as the chlordanes, endosulfan, and dieldrin; (c) the hexachlorocyclohexane insecticides (HCHs), principally lindane; (d) the hexachlorocyclopentadiene pesticides, mainly kepone and mirex; (e) chlorinated camphenes, namely toxaphene; (f) the carboxylic acid herbicides, such as 2,4-D and dacthal; and (g) chlorinated benzenes and phenols such as the wood-preservative pentachlorophenol (PCP). Together, these classes represent nearly 50 specific chemicals such as dieldrin or chemical mixtures such as the PCBs and toxaphene.

The project focused geographically on marine and estuarine organisms of the Arctic, North Pacific, and North Atlantic oceans, the Chukchi, Bering, and Carribean seas, and the Gulf of Mexico with emphasis on U.S. waters within the U.S. 200 mile Exclusive Economic Zone (EEZ), those within the jurisdiction of all coastal states (24), and those within the territorial waters of Puerto Rico and the U.S. Virgin Islands. Further, data were accepted from samples taken in all marine provinces (including intertidal, coastal shelf, slope, and oceanic basin regions) and all ecological zones epi-, meso-, and bathy-pelagic, and benthic).

The study was limited to fish and shellfish. Fish were defined to include hagfish, lampreys, sharks, and rays as well as all bony fishes. "Shellfish" were defined as all conspicuous invertebrates encountered but with emphasis on echinoderms (starfish, sea urchins, sea cucumbers), mollusks (snails, clams, oysters, mussels, squid, octopus), and crustaceans (shrimp, crab, lobster). "Estuarine" species were defined to include any species taken from tidal waters including both anadromous and catadromous species.

The data used were not limited by type of tissue analyzed (i.e., whole, muscle, liver, gonad, etc.), by collection method, or by chemical extraction and analytical methods.

2.2 Data Collection and Processing

A nationwide search for data meeting the criteria described above was carried out by several teams of OAD scientists and technicians during 1984-85. The objective was to collect original data if it had been omitted from other published reports (NOAA, 1986).

Table 2.1. List of target chemical residues and their use and occurrence. Residues are listed as reported in literature.*

Residue	Use and Occurrence
Polychlorinated biphenyls (PCBs)	Dielectric fluid in capacitors; transformer fluid;
Total PCB	libricants; hydraulic fluids; plasticizers; cutting oil
Aroclor 1016	extenders; carbonless paper. Banned in 1976. Total is
Aroclor 1242	either sum of chlorination mixtures (Aroclors) or
Aroclor 1248	number.
Aroclor 1254	
Aroclor 1260	
PCBs by chlorination number (2-10)	
DDT and structurally-related chemicals	
DDE (o-p and p-p')	Insecticides; DDT metabolites
DDD (TDE; o-p, and p-p')	Insecticides; DDT metabolites
DDT (o-p and p-p')	Insecticides; parents of DDD/DDE
Total DDT	Sum of parent and metabolites
Kelthane (Dicofol)	Acaracide; parent of DDE
Methoxychlor	Insecticide
Cyclodiene pesticides	
Technical chlordane	Insecticide; mix of constituents
Chlordane (trans- and cis-)	Insecticides; major constituents of technical chlordane
Nonachlor (trans- and cis-)	Insecticides; minor constituents of technical chlordane
Oxychlordane	Chlordane metabolite
Heptachlor	Insecticide; minor constituent of technical chlordane
Heptachlor epoxide	Metabolite of heptachlor
Endosulfan (I and II)	Insecticides; 7:3 mixture of stereoisomers
Endosulfan sulfonate	Metabolite of endosulfans
Aldrin	Insecticide
Dieldrin	Insecticide; Aldrin metabolite
Endrin	Insecticide
Hexachlorocyclohexane insecticides	
a-BHC	Constituent of BHC insecticide mix
Y-BHC (Lindane)	Insecticide; BHC constituent
Hexachlorocyclopentadiene pesticides	
Kepone	Acaricide, larvicide, fungicide, ant bait
Mirex	Insecticide (fire ant control)
Chlorinated camphenes	
Toxaphene	Insecticide (cotton)
Carboxylic Acid Derivatives	
2,4-D (2,4-DEP)	Weed herbicide (in cereals)
2,4,5-T	Woody plant herbicide
DCPA (dacthal)	Pre-emergence weed herbicide
Chlorinated benzenes and phenols	
HCB (hexachlorobenzene)	Fungicide
PCP (pentachlorophenol)	Wood preservative

* List modified from Schmitt et al. (1985). Classification of pesticides according to Buchel (1983).

Data and auxiliary information on sampling and analytical procedures were actively collected in any available form including published and unpublished reports, memos and other documents, raw data sheets, magnetic tapes, and diskettes.

Data concerning contaminant concentrations, sample and sampling characteristics, and analytical methods were extracted into a common format and then entered into the desk-top computer data base management system specifically developed for this purpose (Buchman, 1986; NOAA, 1986).

2.3 Data Analysis

Several guidelines were used to sort, combine, and analyze the contaminant data.

Data were always sorted by taxonomic categories and tissue types or product to eliminate the known major sources of variability. PCBs and chlorinated pesticides were very often more concentrated in fish than in shellfish (such as bivalve mollusks) and often more concentrated in whole fish, fish liver, and meal than in fish muscle tissue. Tissue categories distinguished for this report were: whole organisms (bivalves and fish separately), muscle (flesh), liver tissue, gonad tissue, fish meal, and fish oil.

Unless otherwise given, contaminant concentrations were transformed to a wet weight (ww) basis, in keeping with units used in seafood and predator protection quality criteria (see below). Data originally provided in dry weight (dw) were converted to wet weight concentrations by multiplying given dry content, expressed as a fraction, by given dry weight chemical concentration. When dry content (or its reciprocal, moisture content) was not given, wet weight concentrations were computed by multiplying dry weight concentrations by 0.2 for fish tissues and 0.1 for soft tissues of bivalve mollusks.

Sites with replicate or repeated samples taken during a survey period (e.g., a year or longer) were characterized by averaging all encountered values using the arithmetic mean. Frequently, contaminants did not occur at concentrations above specified detection limits. When concentrations were at or below detection, the specified limits were halved and the resulting values included in computation of these site means.

In addition to characterizing individual sampling sites, it was also of interest to characterize results of whole surveys or data sets in a simple fashion.

Whole surveys or data sets with many sites spanning large geographic areas (such as the entire U.S. coastline) were characterized using median (50th percentile) of all site mean concentrations. In most contaminant surveys, a few sites with very high concentrations ("hot spots") can seriously elevate regional averages and give misleading results about the majority of the sites. The median minimized this effect.

2.4 Data Presentation

The principal products of analyses done for this report were maps and graphs designed to reveal the spatial and temporal variations of contaminant levels and the abundance or poverty of data at various localities and time periods. Individual or site mean concentrations were generally plotted as bars using scales that encompassed most of the data points. However, attempts were made to use uniform scales wherever possible to encourage direct comparisons. Caution should be exercised since this was not always possible.

All data are presented in units of parts per million wet weight (ppm ww) in keeping with older historical conventions and to help in evaluating significance of concentrations relative to existing and proposed seafood and predator protection quality criteria (see below). Parts per million is equivalent to milligrams per kilogram (mg/kg) and micrograms per gram (ug/g). It is currently popular to report tissue concentrations in units of parts per billion (ppb) or in micrograms per kilogram (ug/kg). These can be computed by multiplying the data presented here by 1,000.

More specific details of data presentation and analysis are presented with the findings in each of the following chapters.

2.5 Comparison to Criteria

It is useful and important to compare encountered contaminant concentration with levels of concern. Unfortunately, no criteria have been proposed that indicate tissue concentrations that are hazardous to organisms bearing those concentrations (although there is an abundance of research with which to make some judgments). However, there do exist a suite of enforceable "action limits" and proposed criteria for the protection of human and wildlife consumers of organisms contaminated with PCBs or chlorinated pesticides. The only criteria that are federally enforced are the U.S. Food and Drug Administration (FDA) "action limits," used to prevent interstate sale of contaminated seafood. The action-limit values range from 0.1 ppm ww for mirex to 5.0 ppm ww for DDT (Table 2.2). Other nations have similar, equivalent, or more restrictive action limits (Nauen, 1983).

There also exist proposed criteria for protecting humans and/or wildlife from effects of pesticides and PCBs, including numerical concentration values for U.S. FDA-designated "warning" levels of excessive contamination of shellfish. These values, proposed by the National Shellfish Sanitation Program (NSSP) range from 0.03 ppm ww for chlordane to 1.5 ppm ww for total DDT (tDDT) National Academy of Sciences (NAS, 1974). Further, NAS (1974) also recommended numerical criteria concerning warning level concentrations of contaminants in fish for protection of predatory wildlife. For freshwater fish, these concentrations ranged from 0.1 ppm ww for nine pesticides to 1.0 ppm ww for DDT (Table 2.2). For protecting predators of marine fish, even more restrictive criteria were proposed by NAS (1974): these range from 0.005 ppm ww for the sum of dieldrin, aldrin, endrin, and heptachlor epoxide to 0.05 ppm ww for tDDT (Table 2.2). These values have not been adopted as regulatory criteria.

In addition, models now exist for computing incremental risk of cancer in humans given specified seafood consumption rates and contaminant concentrations (Tetra Tech, 1985). All these existing U.S. FDA action limits and proposed tissue concentration criteria are for the protection of human or wildlife consumers of marine and aquatic animals, and not, to our knowledge, for the protection of the health of fish or invertebrates themselves.

Table 2.2 Summary of action limits and proposed criteria (in ppm ww) for PCBs and pesticides in fish or shellfish.

Chemical	FDA Action Limit	NSSP Shell-fish	Predator Protection Levels NAS[d] Aquatic Wildlife	NAS[e] Marine Wildlife
Aldrin	0.30	0.20[a]	0.10[b]	f
Dieldrin	0.30	0.20[a]	0.10[b]	f
Endrin	0.30	0.20[a]	0.10[b]	f
DDT				
DDE Sum	5.0	1.5	1.0	0.05
DDD (TDE)				
Chlordane	0.3	0.03	0.10[b]	0.05
Heptachlor	0.3	0.20[a]	0.10[b]	0.05
Heptachlor epoxide		0.20[a]		
Lindane		0.20	0.10[b][f]	0.05
BHC (other than lindane)		0.20		
Methoxychlor		0.20		0.05
Endosulfan			0.10[b]	0.05
Mirex	0.1			0.05
Kepone	0.4 crabs 0.3 fish, shellfish			
Toxaphene	5.0		0.10[b]	0.05
HCB			0.1	0.05
2,4-D		0.50		
PCBs	2.0		0.5[c]	0.5[g]
TCCD				

a. "Alert" level if combined value of the five pesticides exceeds 0.20 ppm ww; shellfish bed should be closed if combined values exceed 0.25 ppm ww. (NAS, 1974, p. 37).
b. Singly or in combination with others listed. (NAS, 1974, p. 186).
c. NAS, 1974, p. 177.
d. In whole fish. (NAS, 1974, p. 18).
e. Homogenate of at least 25 fish of appropriate size and species. (NAS, 1974, pp. 226-227).
f. Sum of these four pesticides should not exceed 0.005 ppm ww. (NAS, 1974, p. 227).
g. Add all Aroclors for total. (NAS, 1974, p. 226).

3.0 ABUNDANCE AND DISTRIBUTION OF DATA

The analysis of trends using existing data is necessarily dependent on the abundance and distribution of data over time and among species, tissue types, sampling localities, and laboratories using different analytical methods. This study identified at least 300 separate surveys in which over 35,000 samples of fishes and invertebrates have been analyzed for chlorinated pesticides and PCBs. Over the 48-year period, from 1940 to the present, these surveys have provided over 125,000 individual data points.

The review we present necessarily reflects the bias of geographic location of sites and quantities of samples taken. A previous review (OAD, 1986) reported half of its samples as coming from the East Coast (Table 3.1), with the largest numbers of samples coming from coastal waters of California, New York, Texas, and North Carolina (Table 3.1). Since that review, the total number of records received for Massachusetts, New York, Maryland, and Hawaii have grown by nearly 10,000 and will be compiled in a subsequent inventory. Over 200 individual bays, estuaries, and other distinct embayments are represented (partial list presented in Figure 3.1). Bays of the East (Atlantic) Coast appear to be represented by more samples than those of the West (Pacific) and Gulf of Mexico coasts, perhaps by a factor of 2 to 5. Based on information on file in the OAD digitized data base, the most heavily sampled embayments are the Chesapeake Bay (4,411 samples), the Palos Verdes Peninsula outfall area near Los Angeles (1,404), Long Island Sound (1,261), Georgia estuaries (collectively 1,105), San Francisco Bay (930), the New York Bight (843), and the Hudson River-Raritan Bay estuarine region (812) (Figure 3.1). Least sampled embayments include Biscayne Bay near Miami, Florida, and embayments from Central Oregon through Northern California.

For 43 estuaries and bays sampled as part of NOAA's current National Status and Trends (NS&T) Program, historical sample sizes range from 5 (Salem Harbor) to 4,411 (Chesapeake Bay). A complete account of all embayments sampled requires further analysis of all local and regional data.

The acquired data included information on more than 35 chlorinated pesticides, including all isomers and metabolites as well as PCB data quantified as Aroclor groups, as total PCB (tPCB) and by chlorination number. Over 90 percent of 25,000 extracted and digitized records contain data on tDDT or on one or more of its metabolites. More than half of the records contain data on PCBs. Over one-quarter of the records contain data on aldrin, chlordane, dieldrin, endrin, heptachlor, lindane, methoxyclor, mirex, and toxaphene or were subject to dedicated analyses for these chemicals. Chemicals represented in approximately 10 percent or less (2,000 records) of the samples include nonachlor, oxychlordane, kepone, HCB, BHCs (other than lindane), endosulfan, dioxins, dibenzoforans, and the herbicides dacthal (DCPA), 2,4-D, and 2,4,5-T. Thus, large amounts of data (thousands of records) are available for more than half of the pesticides.

Archived data exist for at least 540 species of invertebrates and fishes including over 100 species of echinoderms, annelids, anthropods, mollusks. and other invertebrates and representatives of over 100 families of sharks, rays, and bony fishes. Indeed, nearly one-fourth of all identified coastal fish species of the United States have been examined for chlorinated pesticides

Table 3.1. Distribution of data provided by 217 surveys of PCBs and chlorinated pesticides in U.S. marine and estuarine fish and shellfish.

Region/State	Years	Estimated Numbers of Surveys	Samples	Sites
Atlantic				
Maine	65-84	6	400	20-25
New Hampshire	--	0	0	--
Rhode Island	69-84	11	200	20-25
Massachusetts	70-86	17	1,000	30-40
Connecticut	70-84	14	450	15-20
New York	66-86	34	3,300	100
New Jersey	66-84	30	900+	40-50
Delaware	66-84	11	300	15-20
Maryland	66-84	16	400	50-60
Virginia	65-84	14	950	40-50
North Carolina	66-84	15	1,500	40-50
South Carolina	66-84	11	800	40-50
Georgia	66-84	17	800	20-30
Regional Total	65-86	--	11,000	420-500
Gulf and Florida				
Florida	64-84	20	1,000	30-50
Alabama	68-84	11	78	15-20
Mississippi	65-84	12	600	25-30
Louisiana	68-84	12	300	25-30
Texas	65-84	15	1,700	40-50
Regional Total	65-84	--	3,700	135-180
Pacific Region				
Alaska	65-82	8	190	40-50
British Columbia	72-84	11	260	30-50
Washington	65-84	29	1,600	60-80
Oregon	67-84	12	360	35-50
California	40-85	66	5,000	150-200
Baja Calif. (Mex.)	49-84	15	200	30-40
Hawaii	65-84	14	450	40-50
Regional Total	40-84	--	8,060	385-520
GRAND TOTAL	40-85	217	22,760	940-1,200

Modified from OAD, 1986.

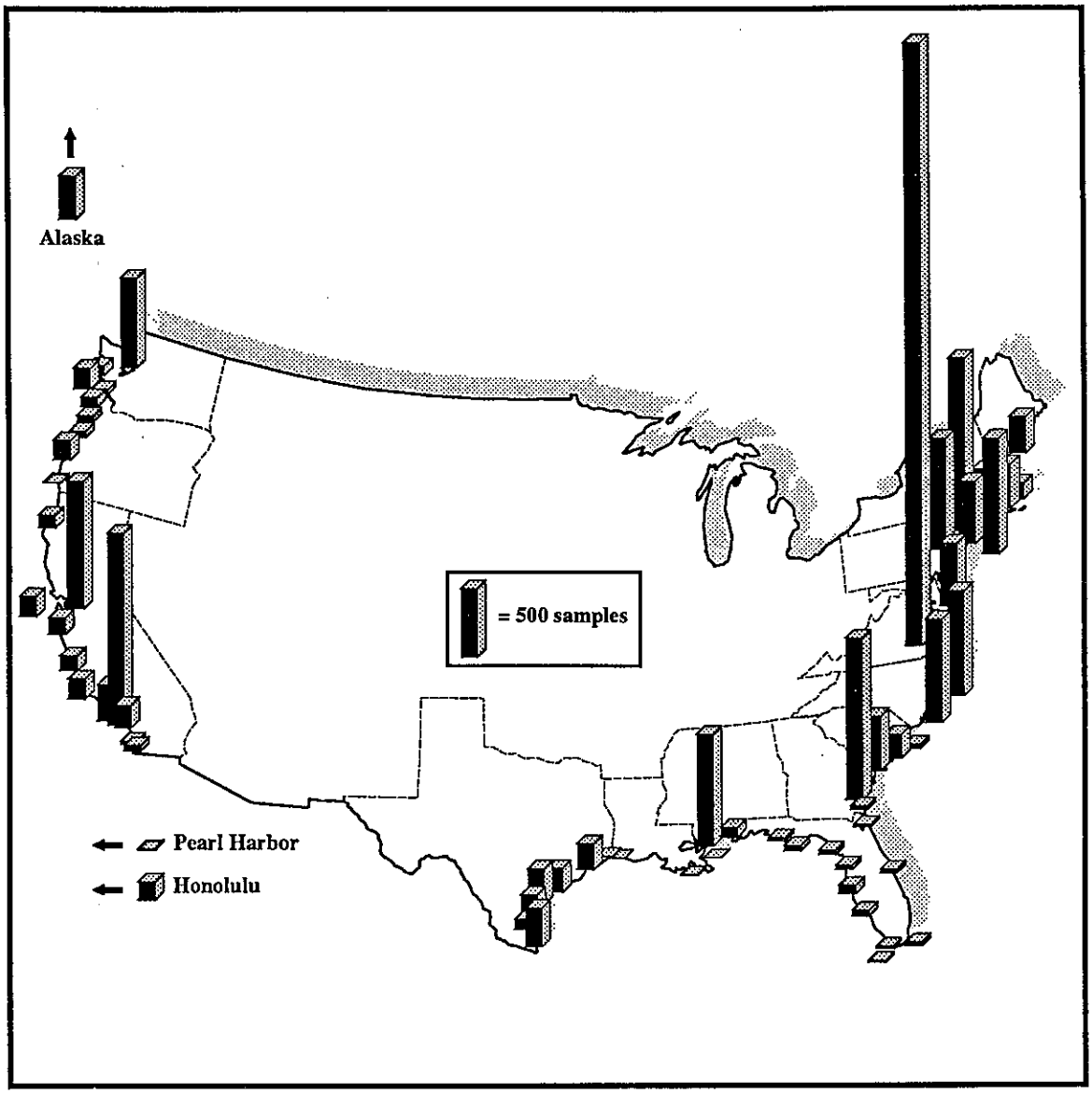


Figure 3.1. Geographic distribution of numbers of samples analyzed for PCBs and/or chlorinated pesticides, 1940 to 1985. Based on data residing in OAD database system, June 1987.

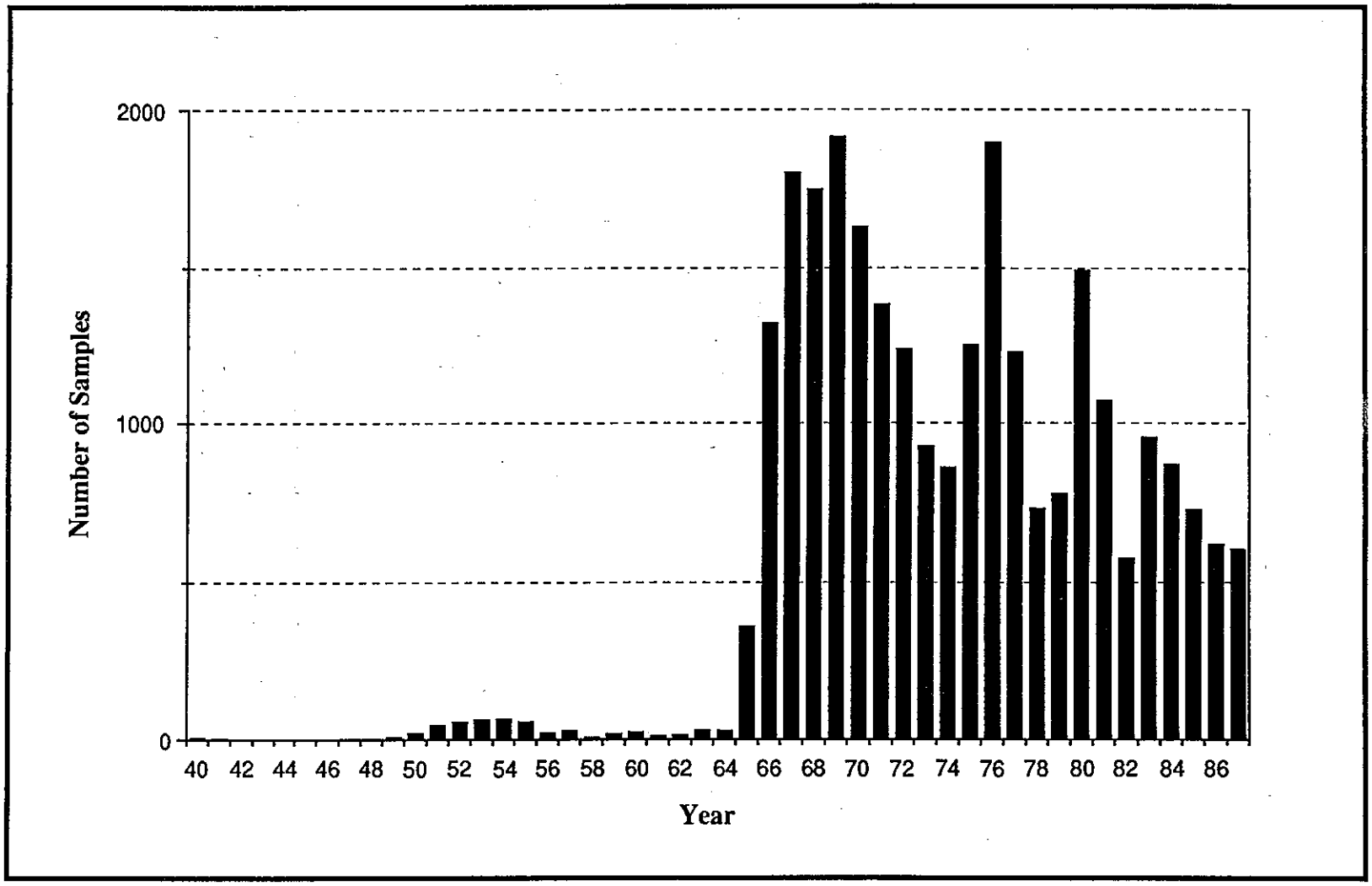


Figure 3.2 Numbers of samples by year, 1940 to 1985, in OAD historical data base.

and for PCBs. Most of the sampling however, has been done with bivalves and fish, as we reported above for West Coast data (Matta et al., 1986).

The acquired data included a broad range of sampling frequencies and intervals that may be useful in determining spatial and temporal variability. Samples reported by single laboratories were taken at various frequencies--monthly, quarterly, semiannually, annually, and interannually--for periods of up to a decade. For example, many of the sites sampled during the NPMP during 1965-72 were recorded monthly during periods of 3 to nearly 5 years (Butler, 1973). Monthly sampling data also exist for fish (Smith and Cole, 1970; Cross and Hose, 1986).

Most of the data were generated by one type of gas chromatography column. About half of these were accompanied by specific information on the use of standards and intercalibration; only a small fraction came with information on internal calibration. Most of the data (96 percent) were generated using packed columns; only 4 percent of the digitized data were generated using capillary columns. Both column types were used in only two of the data sets. Nearly half the samples were generated by surveys or programs that explicitly involved intercalibration with other laboratories.

4.0 NATIONAL OVERVIEW

This chapter reviews geographic and long-term trends of PCB and chlorinated pesticide contamination at the national level. The review identifies sources of nationwide monitoring data and uses these to build cases for large-scale, long-term trends in fish and shellfish. The results indicate: (1) that some contaminants such as PCBs, DDT, chlordane, and dieldrin are or have been nationally widespread, (2) that dramatic nationwide declines have occurred for DDT but not necessarily the others, (3) that knowledge is very poor about the current levels of several such as dacthal (DCPA), endosulfan, toxaphene, and pentachlorophenol (PCP), and (4) that several such as mirex, HCB, and lindane do not currently appear to be important contaminants on a nationwide basis.

Nationwide trends result from a comparison of data collected at about the same time from the three principal U.S. sea coasts--Atlantic, Gulf of Mexico, and Pacific. Two main potential sources of data for evaluating national trends are (1) dedicated synoptic national surveys and (2) local or regional (state) programs producing data which could be assembled to form a nationwide data base. The preferred approach, which we used here, is to use data from nationally synoptic surveys produced using uniform procedures.

Fortunately, several surveys have been undertaken that singly or in combination qualify as national surveys (Table 4.1). During the past two decades (since 1965), these 14 surveys have produced over 12,000 records or one-third of the total identified data base.

These national surveys began with the 1965-72 Estuarine Mollusk Program of the NPMP which measured pesticides monthly in oysters, clams, and mussels from at least 180 sites in 15 coastal states (Butler, 1973). Eighty-seven sites were resampled twice in 1977 (Butler et al., 1978). Several large-scale programs overlapped the NPMP effort, with particular focus on fish (Stout, 1980; Stout and Beezhold, 1981; Stout et al., 1981). Beginning in 1972, the NPMP began a 5-year surveillance of pesticides in whole juvenile estuarine fish at up to 144 sites nationwide, including Alaska, Hawaii, Puerto Rico, and the Virgin Islands (Butler and Schutzmann, 1978). At this time (1976) the Environmental Protection Agency (EPA) launched the EPA U.S. Mussel Watch which conducted two annual surveys for PCB and DDE at 100 sites nationwide (Farrington et al., 1982). During 1979-81, NOAA/NMFS conducted one nationwide survey for PCBs (Gadbois and Maney, 1982). No nationwide surveys were conducted from this time until NOAA's NS&TP began the Benthic Surveillance Project in 1984 and NOAA Mussel Watch in 1986. However, there were surveys of reduced scale through the 1970s (e.g., Markin et al., 1974; Butler, 1978; ERCO, 1980 and 1981; Sherwood, 1982).

Together these surveys form a mosaic of nationwide sampling over the two-decade period, 1965-1986. However, sampling effort has not been evenly distributed across time, geographic locations, species, and tissue types. For example, most of the national sampling effort, in terms of numbers of samples, was concentrated in the late 1960s and early 1970s as a result of over 8,000 samples analyzed by the NPMP (Figure 4.1). National surveillance sampling intensity has actually decreased since the mid 1970s (Figure 4.1).

Table 4.1 Characteristics of 14 national and seminational U.S. monitoring and surveillance programs.

YEARS	PROGRAM (AND SPONSOR)	FREQUENCY	ORGANISM(S)	SUBSTRATE	# OF STATES	# OF SITES	# OF SAMPLES	REFERENCE
1965-72	NPMP[a]-Estuarine (EPA)	M[b]	Bivalves	W[c]	15	180	7,843	(1)
1967-76 P[d]	CHCS in NE Pacific (NMFS)	I	Fish, Shellfish	M	5	85	338	(2)
1969-77 A,G	CHCS in Menhaden (NMFS)	A	Menhaden	W, M, P	12	10	691	(3)*
1973-75 A,G	CHCS in Atlantic Fish (NMFS)	O	Fish	M	6	9	70	(4)*
1971 A, G	Mirex in seafood (USDA)	O	Fish, Shellfish	W	8	9	70	(5)
1972-76	NPMP-Estuarine Fish (EPA)	Q	Juvenile fish	W	21	144	1,831	(6)
1976-77	CEMP (NOAA/EPA)	O	Fish	M, L	10	11	565	(7)+
1976-77 P,A	Comp. Three Areas (NOAA)	O	Fish	M, L, B	4	7	287	(8)+
1976-78	US Mussel Watch (EPA)	A	Bivalves	W	18	100	240	(9)
1977	NPMP-Estuarine (EPA)	S	Bivalves	W	15	87	147	(10)
1979-81	PCBs in finfish (NMFS)	O	Fish	M	10	13	72	(11)
1980 A,G	GAS I and II (NOAA)	O	Fish, crustacea	M, L	11	67	204	(12)
1984-present	NS&T/Benthic Survey (NOAA)	A	Fish	L	18	50	450	(13)
1985-present	NS&T/Mussel Watch (NOAA)	A	Bivalves	W	18	100	300	(13)

a NPMP = National Pesticide Monitoring Program; CEMP = Cooperative Estuarine Monitoring Program; NS&T = National Status and Trends Program

b M = Monthly; Q = Quarterly; S = Semiannually; A = Annually; O = Once

c W - Whole animal; M = Muscle or flesh; L = Liver; B = Brain

d P = Pacific, A = Atlantic, G = Gulf of Mexico

Reference numbers: (1) Butler, 1973; (2) Stout and Beezhold, 1981; (3) Stout et al., 1981; (4) Stout, 1980; (5) Markin et al., 1974; (6) Butler and Schutzmann, 1978; (7) Butler, 1978; (8) Sherwood, 1982; (9) Farrington et al., 1982; (10) Butler et al., 1978; (11) Gadbois and Maney, 1982; (12) ERCO, 1980 and 1981; (13) NOAA, 1987a,b; Malins et al., 1986; Zdanowicz et al., 1986, and Hanson et al., 1986.

* Combined data from reference surveys 3 and 4 resulted in national (three-coast) survey effort.

+ Combined data from reference surveys 7 and 8 resulted in national (three-coast) survey effort.

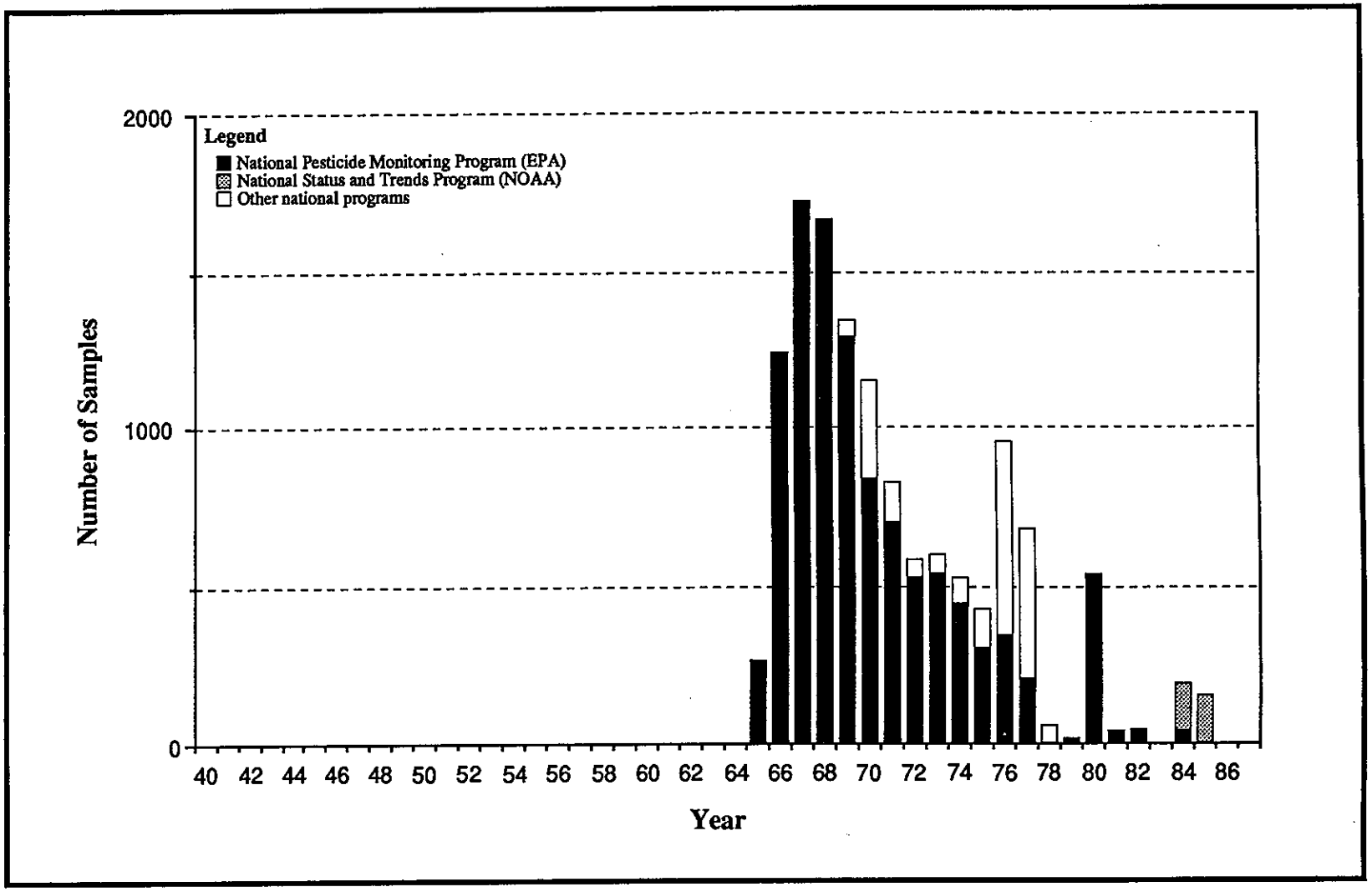


Figure 4.1. Numbers of samples by year, 1940-85, from national and other comprehensive surveys for PCBs and/or chlorinated pesticides in U.S. fish and shellfish.

The surveys are also not directly comparable because each program sampled different combinations of sites, species, and tissues. Even data generated within a single program are not all geographically comparable because the same species were not sampled at all sites (although in many cases related species were). Further, exact sampling sites for some historical programs have not all been identified beyond specific embayments. Finally, during this period, analytical techniques evolved through more sensitive chemical detection limits and increased ability to identify and resolve specific chemicals some of which (PCBs) previously interfered with the identification and quantification of other chemicals.

Despite these problems, it is still possible to reconstruct at least a partial history and geography of several chemicals, most notably PCBs, DDT, dieldrin, and the chlordanes. To do this, we segregated records first by general types of organisms (e.g., bivalve mollusks vs fish), second by tissue type (whole organism, muscle or flesh, liver, oil and meal), and third by performing laboratory, and then made geographic and temporal comparisons.

In the review that follows, sufficient detail is provided so that the reader can judge how we attempted reconstructions of national patterns and trends. Each chemical is reviewed separately beginning with PCBs. For each chemical, the review begins with an estimate of the amount of data encountered and archived, a brief history of sampling, and the ranges of encountered concentrations. This is followed by a more detailed review of trends in national surveys by substrate or tissue type. In surveys where all three sea coasts are sampled, a grand national median is computed. Where two or more grand means are available across a several year period, a non-statistical case is made for a nationwide trend of increasing, decreasing, or unchanging concentrations. In some cases, regional (single coast) medians were calculated and compared. Finally, where possible, we provided additional perspective by contrasting coastal values with similar data from inland freshwater fish programs, especially those of the NPMP (e.g., Schmitt et al., 19856).

4.1 PCBs

PCBs have occurred in marine life in all coastal areas sampled since the early 1970s and have been particularly high in organisms from urban embayments of the Northeast, the Northwest, and Southern California. They have declined in concentration at or near known industrial sources and other "hot spots", but it is not clear there has been any large scale nationwide decline over the past decade. Finally, there is some evidence that on a national scale, PCB concentrations have been lower in estuarine fish than in inland freshwater fish. Nationwide trends of PCBs in coastal marine life are reviewed in more detail below as well as in Chapter 6.

The digitized data base achieved for this report contains over 11,000 records for PCBs, acquired from all national surveys and from over 75 local surveys, some of which are described below.

PCBs have been contaminants of the marine environment for nearly five decades and of marine fish for at least three and a half decades. Analysis of dated sediment cores confirms that PCBs occurred in several marine ecosystems as early as the late 1930s (Hom et al., 1974, Santa Barbara basin; Dexter et al., 1985, Puget Sound) and Aroclor 1254 was detected in museum specimens of southern California mid-water fishes collected as early as 1951 (MacGregor, 1974). However, PCBs were first quantified in U.S. Pacific marine organisms by Risebrough (1969) based on 1965 collections of coastal fish. PCBs were detected in oysters collected near Pensacola in 1969 (Duke et al., 1970) and in menhaden also collected in 1969 from several Atlantic and Gulf of Mexico sites (Stout et al., 1981).

Concentrations of tPCB (the sum of all Aroclors detected) in fish muscle ranged from below detection to 730.0 ppm ww in an American eel collected from New Bedford Harbor, Massachusetts, in 1979 (Massachusetts Division of Marine Fisheries, 1986). Concentrations of tPCB in fish livers range from below detection to 162.1 ppm ww in an English sole collected from off the Palos Verdes Peninsula near Los Angeles in 1977 (unpublished data, Los Angeles County Sanitation Districts (LACSD)). PCB concentrations in menhaden fish meal and oil have ranged as high as 1.46 ppm ww and 19.5 ppm ww, respectively (from mid-Atlantic processing plant samples collected in 1972, reported by Stout et al., 1981).

4.1.1 PCBs in Bivalve Mollusks

Data from four national surveys have sampled and analyzed PCBs in bivalves since 1970. Results of the 1970-72 and 1977 NPMP (Butler, 1973; Butler et al., 1978) and the U.S. EPA Mussel Watch Program (Farrington et al., 1982) and the 1986 NOAA Mussel Watch Program (NOAA, 1987b) establish that the most contaminated sites occurred along the Northeast Coast and in Southern California harbors. The grand national median for the PCB mixture, Aroclor 1242 in the 1976 EPA U.S. Mussel Watch Survey (86 sites) was 0.009 ppm ww with a range of 0.0008 to 2.09. Although not strictly comparable because of analytical, site, and species differences, preliminary calculations indicate that the grand national median of total PCBs in the 1986 NOAA Mussel Watch Survey (144 sites) was 0.015 ppm ww (range 0.0009 to 0.68 ppm ww). The values are comparable and suggest no great change over the past decade. However, the former survey measured one (Aroclor 1254) of several possible components of PCBs; whereas, the latter measured total (chlorination number). This difference, as well as others, seriously complicates a comparison of these data. All but one value (2.09 ppm ww) from these two national surveys were below the proposed FDA alert action limit of 2.0 ppm ww. However, some, principally from the Northeast, were above the predator protection level of 0.5 ppm ww proposed by NAS (1974) for fish (see Table 2.2). Temporal trends of PCBs in bivalves are reviewed in more detail in Chapter 6.

4.1.2 PCBs in Whole Fish

Concentrations of PCBs are generally higher in fish than in bivalve mollusks. Total PCB concentrations in whole estuarine or coastal fish from national programs have ranged as high as 6.59 ppm ww in a 1972 sample of menhaden collected from a New Jersey processing plant (Table 4.2; Stout et al., 1981). However, other data suggest the national coastal average has been slightly below 0.1 ppm ww.

Table 4.2. Summary by site* of maximum concentrations of tPCBs in fish collected during national** monitoring surveys, 1969–1984.

LOCATION	CONCENTRATION (in ppm ww)	SPECIES	SAMPLING PERIOD	NO./ TOTAL NO.	DATA SOURCE
WHOLE FISH					
Connecticut River, CT	1.1	Cunner	1972–74	1/4c	Butler and Schutzman, 1978
New Jersey area, NJ	6.6	Atlantic menhaden	1969	1/1c	Stout et al., 1980
	3.3	Atlantic menhaden	1971	1/1c	Stout et al., 1980
	5.7	Atlantic menhaden	1972	5/5c	Stout et al., 1980
	2.4	Atlantic menhaden	1973	3.5c	Stout et al., 1980
	1.5	Atlantic menhaded	1974	2/4c	Stout et al., 1980
	1.7	Atlantic menhaden	1976	3.4c	Stout et al., 1981
Delaware Bay, DE	1.2	Unidentified fish	1971	ND/2c	Markin et al., 1974
	4.3	White perch	1972–76	5/9c	Butler and Schutzmann, 1978
	1.2	Bay anchovy	1973–76	4/8c	Butler and Schutzmann, 1978
Chesapeake & Delaware Canal, DE	4.5	White perch	1972–73	2/2c	Butler and Schutzmann, 1978
	3.0	Alewife	1972–73	1/2/c	Butler and Schutzmann, 1978
Patapsco River, MD	1.1	White perch	1972–76	1/15c	Butler and Schutzmann, 1978
James River, VA	2.6	Striped bass	1975	2/2c	Butler and Schutzmann, 1978
North Carolina, NC	3.6	Atlantic menhaden	1972	1/12c	Stout, unpub. data
	1.9	Atlantic menhaden	1976	2/8c	Stout et al., 1980
Duwamish River, WA	4.9	English sole	1972–76	14/16c	Butler and Schutzman, 1978
	3.4	Pacific staghorn sculpin	1972–76	3/16c	Butler and Schutzmann, 1978
Elliot Bay, WA	1.7	Dover sole	1975	1/1c	Stout and Beezhold, 1981

*Listing includes only sites that produced at least one sample exceeding 1.0 ppm ww PCBs in whole fish; in fish muscle, flesh, or filet; or in fish liver tissue.

**"National" refers to synoptic surveys described in Table 4.1.

Table 4.2, continued

LOCATION	CONCENTRATION	SPECIES	SAMPLING PERIOD	NO./ TOTAL NO.	DATA SOURCE
MUSCLE, FLESH, OR FILLET					
Hudson River, NY	2.1	Striped bass	1979–81	5/5c	Gadbois and Maney, 1983[2]
	22.0	White perch	1979–81	5/5c	Gadbois and Maney, 1983
Sandy Hook Bay, NJ	1.2	Bluefish	1979–81	1/1c	Gadbois and Maney, 1983
New York Bight Apex, NY	3.6	Striped bass	1979–81	ND/4c	Godbois and Maney, 1983
Florida Keys, FL	1.8	King mackerel	1975	1/2s	Stout, 1980
Duwamish River, WA	2.1	Starry flounder	1977	1/24s	Sherwood, 1982
Coos River, OR	1.9	Striped bass	1979–81	ND/28c	Gadbois and Maney, 1983
Sacramento River, CA	4.0	Striped bass	1979–81	ND/11c	Gadbois and Maney, 1983
	2.9	Striped bass	1973	3/10s	Stout and Beezhold, 1984
Palos Verdes, CA	2.4	Dover sole	1976	13/24s	Sherwood, 1982
Koko Head, Oahu, HI	2.2[3]	Goatfish	1970	1/1c	Stout and Beezhold, 1981
Hawaiian waters, HI	1.0	Yellowfin tuna	1972	1/1c	Stout and Beezhold, 1981
LIVER					
Boston Bay, MA	3.1	Winter flounder	1984	3/3c	OAD, 1984
Western Long Island Sound NY	6.4	Bluefish	1976	6/410c	Butler, 1978
	2.2	Striped bass	1976	5/10s	Butler, 1978
Sandy Hook Bay, NJ	9.9	Winter flounder	1977	23/23s	Sherwood, 1982
New York Bight Apex, NY/NJ	9.3	Winter flounder	1977	14/15s	Sherwood, 1982
Great Bay, NJ	2.4	Winter flounder	1977	5/10	Sherwood, 1982

Table 4.2, continued

LOCATION	CONCENTRATION	SPECIES	SAMPLING PERIOD	NO./ TOTAL NO.	DATA SOURCE
Rappahonock River, VA	1.4	Striped bass	1973-75	1/6c	Butler and Schutzmann, 1978
Lower Chesapeake, VA	1.3	Bluefish	1976	1/2c	Butler, 1978
	1.3	Summer flounder	1976	2/cc	Butler, 1978
Virginia coastal shelf	1.0	Butterfish	1980	1/1c	ERCO, 1981
South Texas coastal shelf	2.3	Rough scad	1980	1/1c	ERCO, 1981
Straits of Georgia, WA	1.3	Dover sole	1976	1/10x	ERCO, 1981
	1.2	Pacific cod	1976	2/10s	ERCO, 1981
	3.0	Pacific hake	1976	5/10s	ERCO, 1981
	1.3	Spiny dogfish	1976	3/10s	ERCO, 1981
	2.1	Walleye pollack	1976	1/10s	ERCO, 1981
Duwamish River, WA	160.0	Starry flounder	1977	24/24s	Sherwood, 1982
Elliot Bay, WA	6.3	Flathead sole	1984	1/1c	OAD, 1987; Malins et al., 1986
	2.8	English sole	1984	3/3c	OAD, 1987; Malins et al., 1986
Commencement Bay, WA	2.0	English sole	1984	3/3c	OAD, 1987; Malins et al., 1986
Nisqually Reach, WA	1.1	Starry flounder	1977	1/10s	Sherwood, 1982
Hunter's Point, San Francisco, CA	3.4	Starry flounder	1984	3/3c	OAD 1987; Malins et al., 1986
Southampton Shoal, SF Bay CA	1.5	Starry Flouder	1984	1/2c	OAD 1987; Malins et al., 1986
Oakland, CA	2.7	White croaker	1984	3/3c	OAD 1987; Malins et al., 1986
Santa Monica Bay, CA	2.2	Hornyhead turbot	1984	3/3c	OAD 1987, Malins et al., 1986
San Pedro Canyon, CA	3.0	Hornyhead turbot	1984	3/3c	OAD 1987, Malins et al., 1986
Palos Verdes, CA	56.0	Dover sole	1976	23/24s	Sherwood, 1982
Seal Beach, CA	4.4	White croaker	1984	3/3c	OAD 1987; malins et al., 1986
San Diego Harbor, CA	4.2	Diamond trubet	1984	1/1c	OAD 1987; Malins et al., 1986

1 Based on original data supporting these documents except as noted
2 Based on data in document
3 Fillet with skin

The only nationwide survey to systematically measure PCBs in whole fish was the 1972-76 juvenile estuarine fish monitoring program of the NPMP (Figure 4.2; Butler and Schutzmann, 1978). In this survey, PCBs were detected in 331 samples, or 22 percent of the 1,524 composite samples. The 144-station median was below 0.10 ppm ww. More than half of the sites (58 percent) produced whole fish samples at or below the detection limit of 0.05 ppm ww. No site produced a time-averaged mean above the current FDA action limit of 2.0 ppm ww. However, samples from the Chesapeake-Delaware canal showed a mean PCB concentration of 1.9 ppm ww; five other sites showed individual values above 1.0 ppm ww (Table 4.2). There were no sites in the Gulf of Mexico, the Caribbean territories, Hawaii, or Alaska that indicated PCB levels above 1.0 ppm ww in juvenile whole fish composite samples. However, in the Arroyo Colorado of southern Texas, fish were so contaminated with toxaphene, (tens of ppm ww) DDT, and other chlorinated pesticides that PCB concentrations could not be determined (Butler and Schutzmann, 1978). The 1972-76 median of less than 0.10 ppm ww for all 144 NPMP sites was substantially lower than the 99-site whole-fish geometric mean of 0.88 ppm ww (range 0.01 to 70.98 ppm ww) from the 1976-77 NPMP survey of whole inland U.S. freshwater fish (Schmitt et al., 1985), suggesting that at about that time, on a nationwide basis, juvenile estuarine fish were about one-tenth as contaminated with PCB as freshwater fish (size and age varies).

Whole coastal or estuarine fish have not been monitored on a national basis since 1976-77. As a result, it is not possible to document whether levels have since changed for this substrate.

4.1.3 PCBs in Fish Muscle or Flesh

Since 1976, PCBs have been more frequently measured on a nationwide basis in fish muscle and liver than in whole fish. Three synoptic national surveys during 1976-1981 (Butler, 1978; Gadbois and Maney, 1982; Sherwood, 1982) included muscle tissue. The highest reported concentration was 22.0 ppm ww in a muscle composite of white perch collected in 1980 from the Hudson River (Gadbois and Maney, 1982). However, more typical (average) concentrations from these surveys were on the order of 0.1 to 0.2 ppm ww.

In 1976-77, PCBs were measured in both muscle and liver tissue of several dozen bottom dwelling and pelagic fish from 19 sites as part of the NOAA/EPA Cooperative Estuarine Monitoring Program (CEMP) (Butler, 1978) and as part of a NOAA-Southern California Coastal Water Research Project (SCCWRP) survey in three widely separated coastal regions (Sherwood, 1982). These two surveys form a national mosaic for three coasts. Most data on muscle tissue concentrations (90 percent) from the CEMP surveys showed PCB levels below the rather high detection limit of 0.4 ppm ww (sum of four aroclor detection limits of 0.1 ppm ww). The range of time-averaged site means was from less than 0.4 for many sites to 0.78 ppm ww in samples of southern flounder from the Savannah River, Georgia (Figure 4.3). In contrast, almost all of the NOAA-SCCWRP flatfish muscle concentrations were above the detection limit of about 0.004 ppm ww, ranging from a site mean of 0.01 ppm ww in Dover sole from Dana Point, California, to a maximum site mean of 1.11 ppm ww in Dover sole from the Palos Verdes outfall site near Los Angeles. Maximum muscle or flesh composite concentrations are reviewed in Table 4.2.

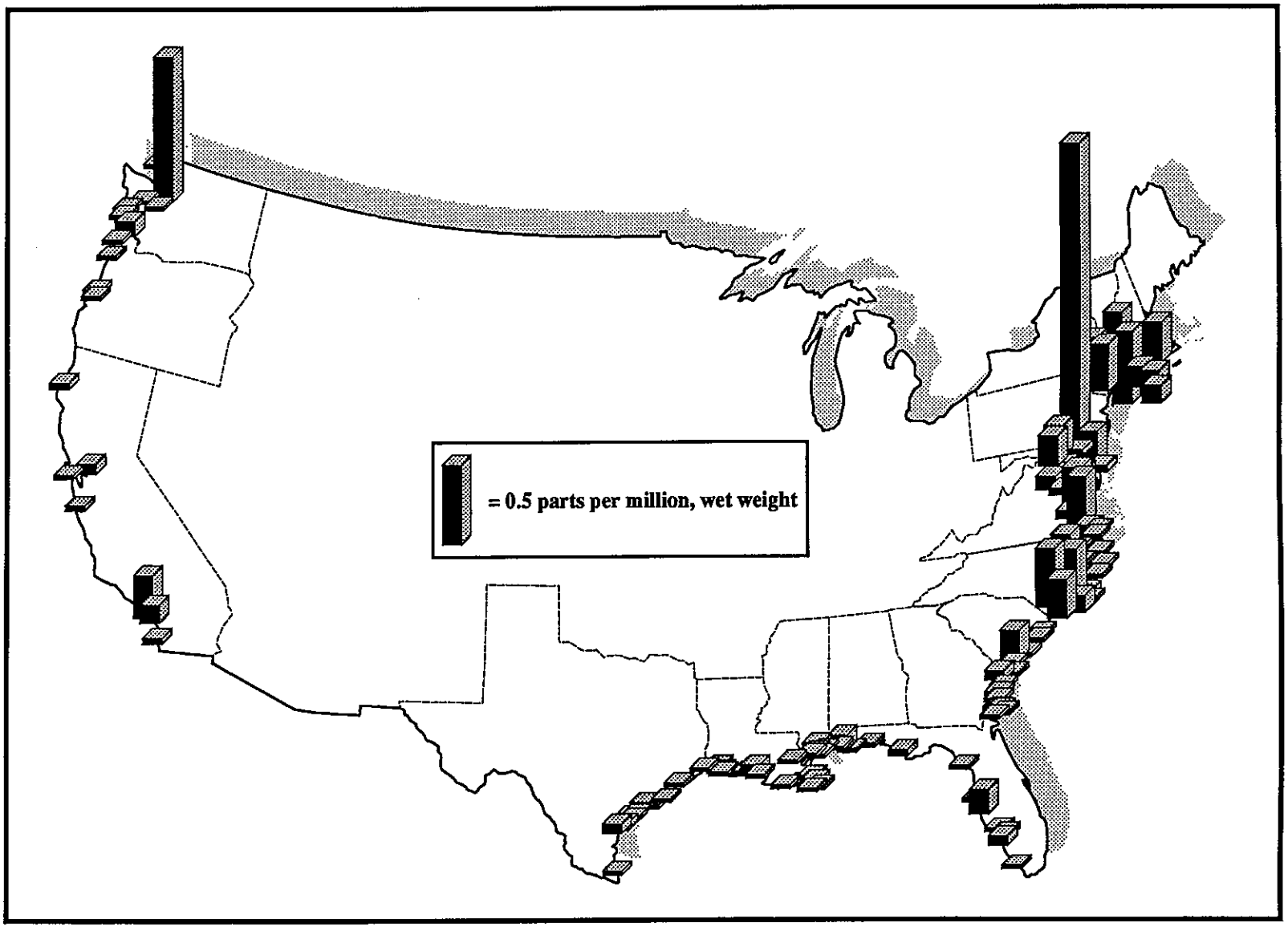


Figure 4.2. Aroclor 1254 (PCB) concentrations in juvenile estuarine fish, 1972-76. Bar represents mean of all composites for all species collected at a site. Sample sizes vary. Computed from original data supporting Butler and Schutzmann (1978).

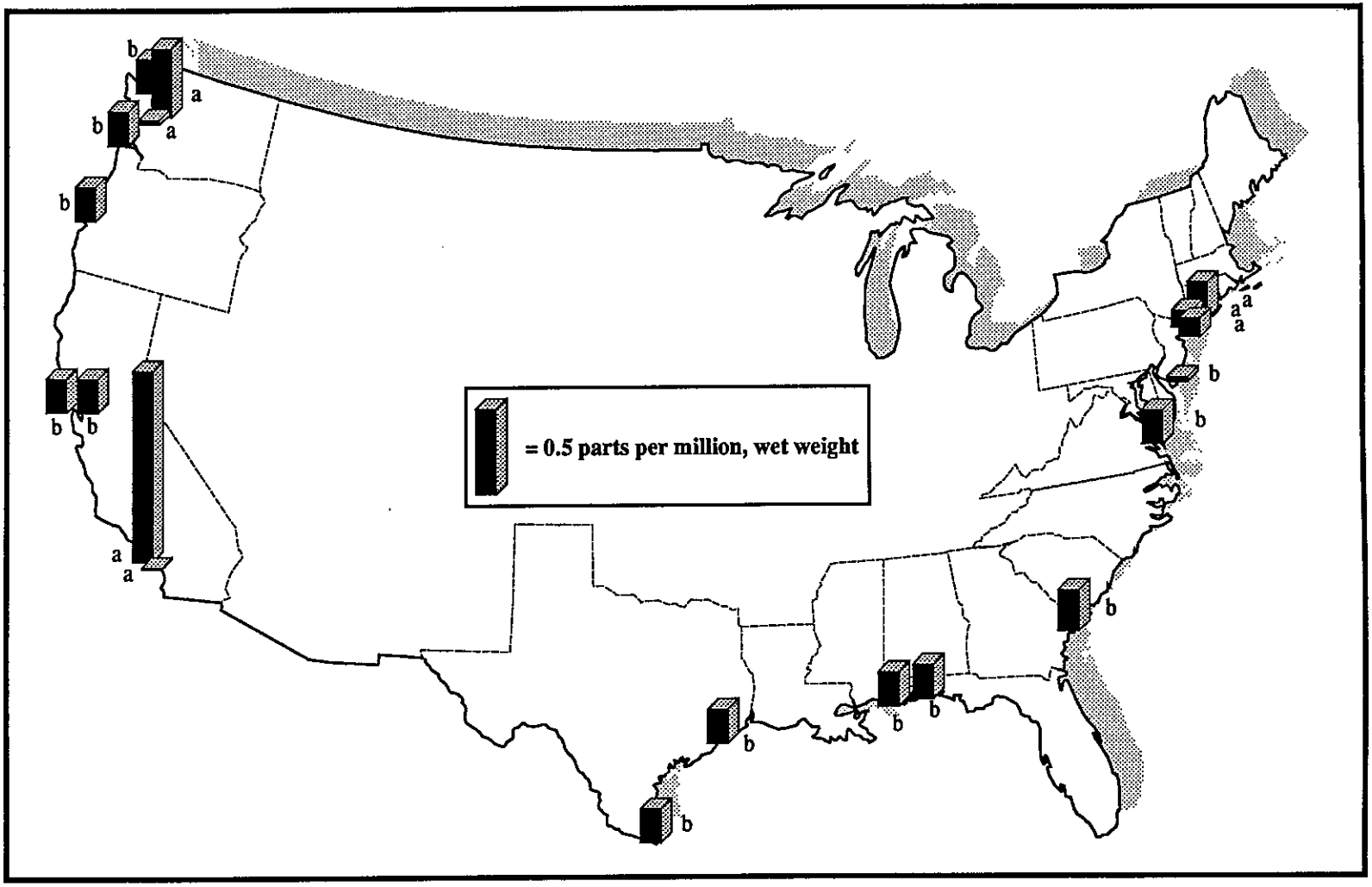


Figure 4.3. Total PCBs (Aroclor) in muscle of coastal and estuarine fish from 19 sites sampled in 1976 and 1977. Bar represents mean of all individuals from all species collected at a site. Compiled from original data supporting (a) Sherwood (1982); or (b) Butler (1978). Half detection limit for (a) was 0.001 ppm ww, and for (b) 0.2 ppm ww.

Gadbois and Maney (1982) reported on 1979-80 sampling of fillets of a mix of pelagic and nearshore predatory fish from sites in 15 coastal and estuarine areas, several of which were close to one another (Figure 4.4). Of the 70 samples, 63 contained PCBs, with concentrations ranging as high as 22.0 ppm ww in Hudson River white perch. Sites producing the highest mean concentrations (for all species combined) were the New York Bight Apex (1.1 ppm ww) and East Bay, near Panama City, Florida (0.42 ppm ww). Sites producing fish with the lowest PCB concentrations were at Catalina Island, offshore of Los Angeles (less than 0.04 ppm ww); Chandeleur Sound, east of New Orleans, Louisiana (0.05 ppm ww in 3 species); and San Luis Pass at the entrance to Galveston Bay, Texas (0.09 ppm ww in 13 species). Additional data on PCBs in fish muscle tissue are reviewed in Chapters 5 to 7. Additional maximum concentrations are cited in Table 4.2.

4.1.4 PCBs in Fish Liver

PCB concentrations are generally much higher in liver than in muscle tissue. The highest concentration of total PCBs in any fish liver sample from a national program was 160.0 ppm ww in a starry flounder collected from the Duwamish River near Seattle in 1977 (from Sherwood, 1982). However, a more typical concentration was about 0.6 ppm ww (median from all site means of Sherwood, 1982 and Butler, 1978, collectively).

In the CEMP survey (Butler, 1978), the time-averaged, site-mean liver PCB concentration was highest in fish from Pensacola Bay, Florida (0.83 ppm ww). Site-mean concentrations were similar in fishes from western Long Island Sound, lower Chesapeake Bay, and the southern Strait of Georgia sites (0.62-0.64 ppm ww). Lowest concentrations were encountered in samples from other Gulf of Mexico sites and from two Oregon estuaries (Sherwood, 1982) (Figure 4.5). In the 1976-77 NOAA-SCCWRP survey, site means for fish liver were highest in Dover sole from the Palos Verdes outfall area (18.6 ppm ww), in starry flounder from the Duwamish River near Seattle (26.7 ppm ww), in winter flounder from Sandy Hook Bay, New Jersey (5.14 ppm ww), and the New York Bight Apex (4.87 ppm ww; Figure 4.5). In contrast, flatfish livers from regionally compatible reference sites (Great Bay, New Jersey; Nisqually Reach, Washington; and Dana Point, California) produced site-average concentrations similar to values reported from the CEMP survey (range 0.07-1.40 vs. less than 0.4 to 0.83). This suggests that very high liver concentrations (i.e., greater than 10.00 ppm ww) could be obtained through concentrated sampling at "hot spots" and that lower detection limits (less than 0.01 ppm ww) could better reveal the extent of low concentrations at nonurban reference sites.

Data are currently available on PCB concentrations in livers of nearshore and benthic fish from 42 Pacific and U.S. Northeast coast sites sampled during the 1984 NS&T Benthic Surveillance Project. The combined Pacific-Northeast median PCB concentration was 0.58 ppm ww a value nearly indistinguishable from the median of 0.6 from the 1976-77 surveys cited above. Area means ranged from 0.13 ppm ww in white croaker (Bodega Bay, California) to 5.45 in a mix of hornyhead turbot and barred sand bass from a site in San Diego Harbor, California. Other areas with sites producing high PCB (in ppm ww) concentrations in fish liver were Boston Harbor (2.62); Elliott Bay near Seattle, Washington (4.23); Commencement Bay near Tacoma, Washington (2.30);

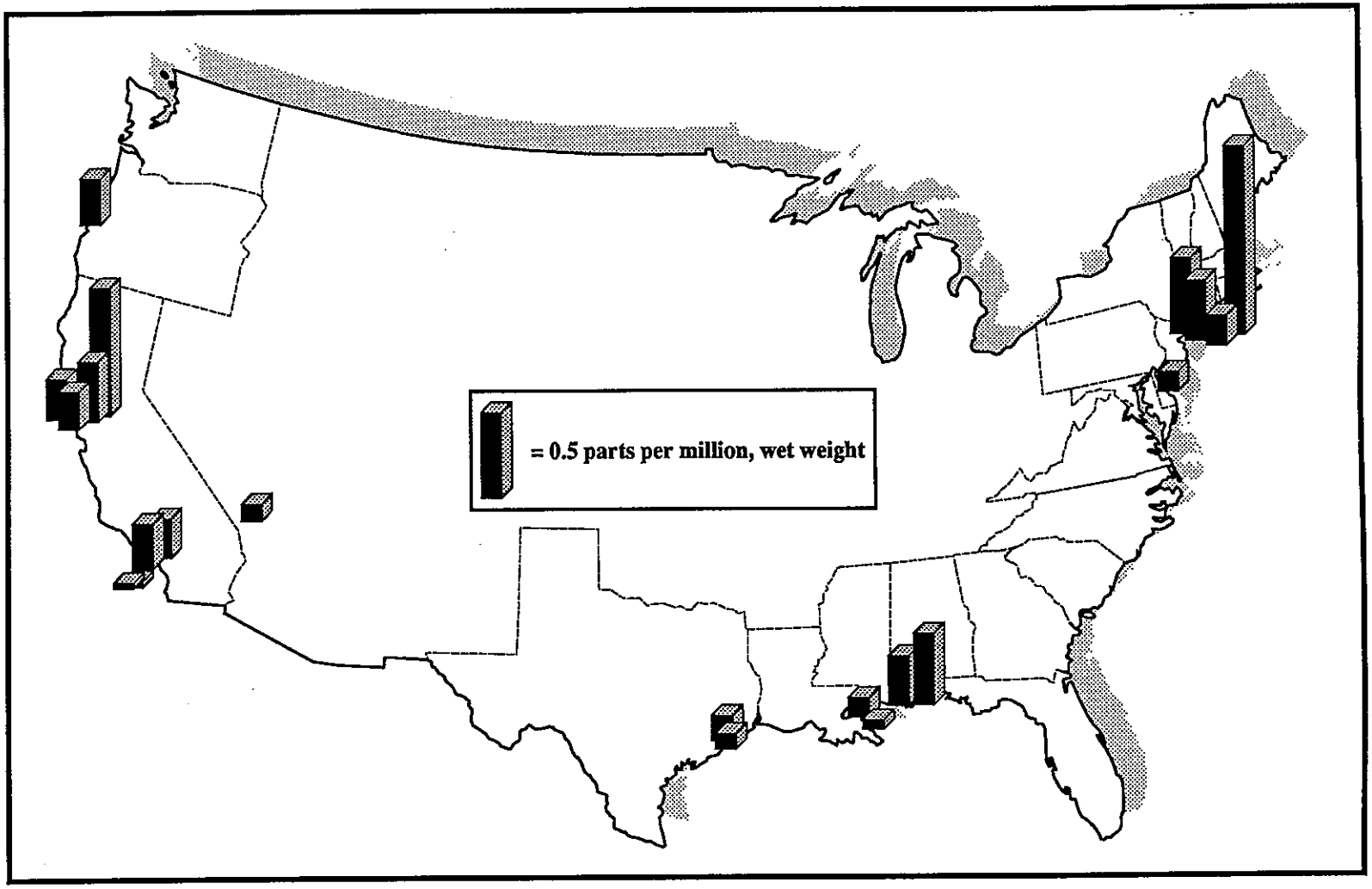


Figure 4.4 Total PCBs (Aroclor) in muscle of coastal and ocean fish sampled 1979-81. Data from Gadbois and Maney (1982). Bar represents mean of all species means at each site.

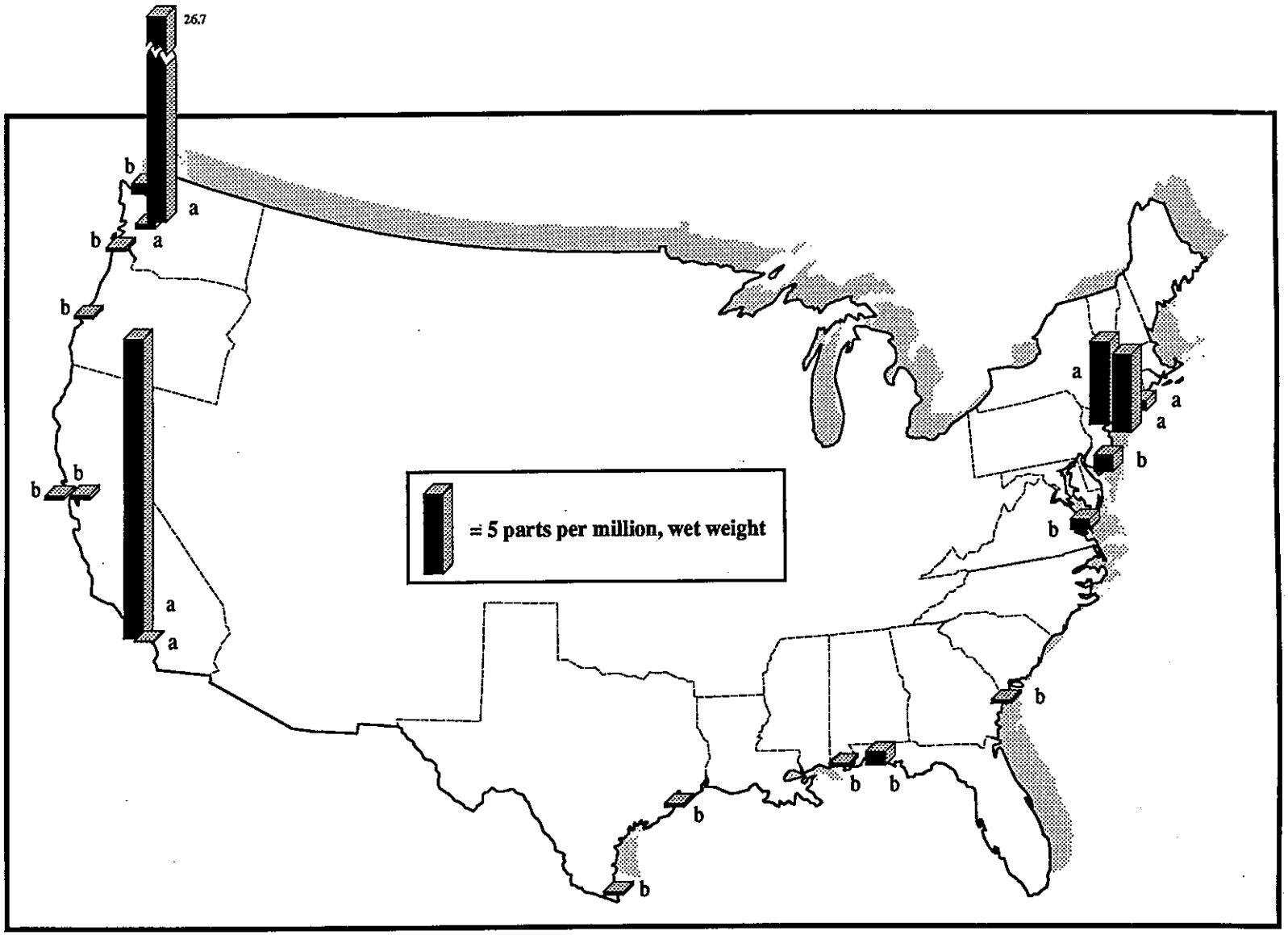


Figure 4.5. Total PCBs (Aroclor) in liver of coastal and estuarine fish from 19 sites sampled in 1976 and 1977. Bar represents mean of all individual values for all species collected at a site. Computed from original data supporting (a) Sherwood (1982); and (b) Butler (1978). Compare with muscle values shown in Figure 4.3 (note 10 X scale change).

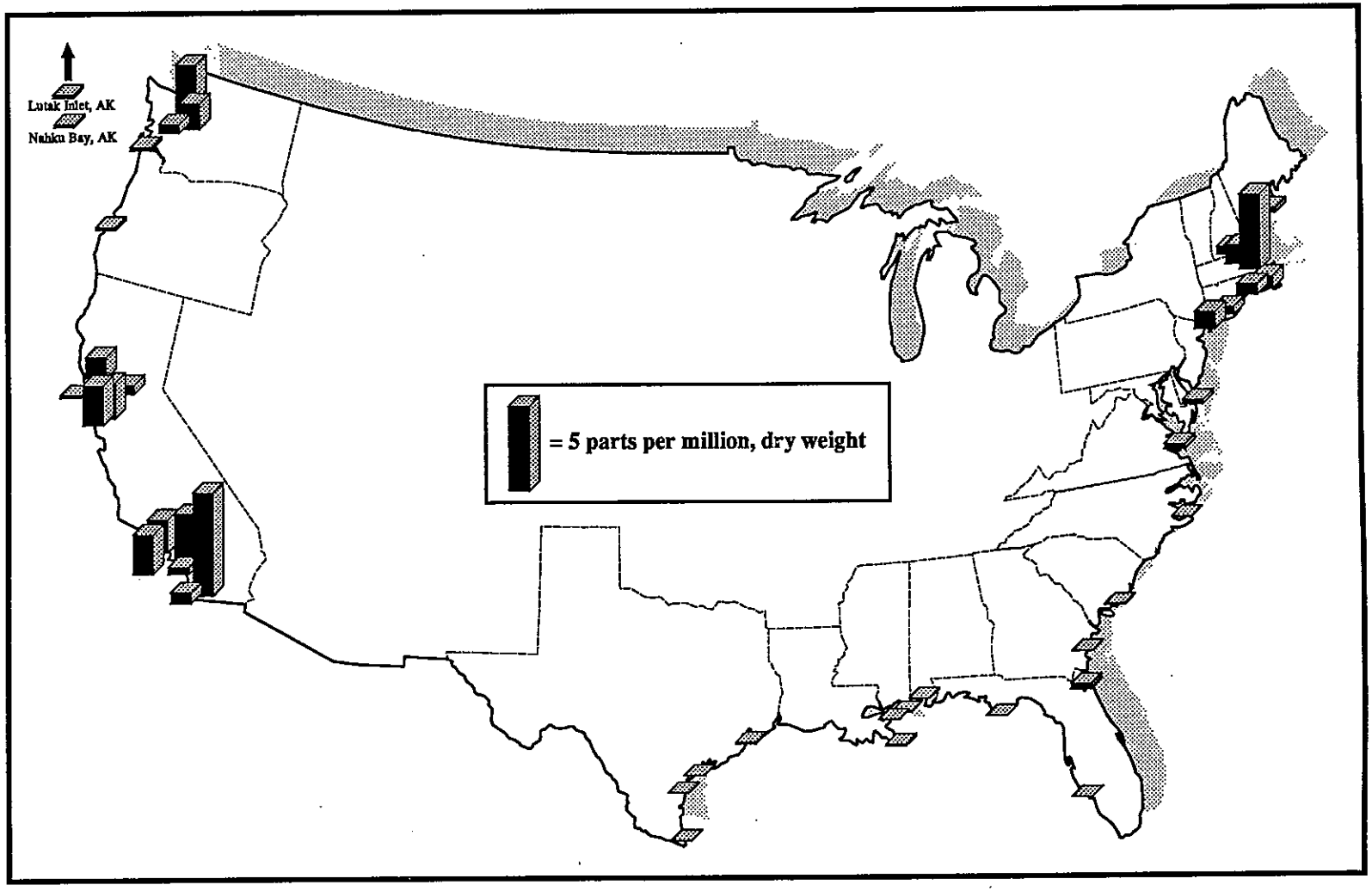


Figure 4.6. Total PCBs (chlorination number) in liver tissue of estuarine fish composites collected at 42 sites in 1984. Computed from original data for the NOAA National Status and Trends Benthic Surveillance Project, supporting OAD (1987), Malins et al. (1986), and Hansen et al. (1986). Bar represents mean of 1 to 5 composite values (approximately 30 fish per composite). Species differ among sites (see OAD, 1987).

three sites in south San Francisco Bay (1.23 to 2.30); and three sites along the Los Angeles County coast, California (1.93 to 4.92) (Figure 4.6).

Have PCB concentrations in fish liver changed over the past two decades? One possible comparison is for tPCBs in liver measured in 1976-77 both by the CEMP surveys (Butler, 1978) and by the NOAA-SCCWRP survey (Sherwood, 1982), and in 1984 using NOAA NS&T Benthic Surveillance Project data (NOAA, 1987a,b; Zdanowicz et al., 1986 and Malins et al., 1986). At least nine areas were common in both sampling periods (Table 4.3). The nine-area 1976-77 fish liver PCB median was 0.31 ppm ww with a range of less than 0.2 to 18.6 ppm ww. The nine-area 1984 median was slightly higher at 0.49 ppm ww with a range of 0.15 to 4.23 ppm ww (Table 4.3). The comparison may or may not indicate a nationwide increase in PCB contamination of coastal and estuarine fish livers, but it clearly does not indicate a substantial decline. Inspection of individual pairs of data indicate the possibility of substantial increases in some areas (e.g., south San Francisco Bay and Dana Point, California), substantial decreases in others (i.e., Palos Verdes-San Pedro Canyon, Lower Chesapeake Bay or Duwamish River-Elliott Bay), and relatively little change elsewhere. However, the question simply cannot be answered without a more penetrating analysis of existing data and, most likely, without renewed sampling of previously sampled sites and species, especially in areas away from known PCB hot spots.

4.1.5 PCB Summary

Thus far, the collected available data from nationwide, large-scale sampling efforts confirm that PCBs have occurred in fish and shellfish from all estuaries sampled, including, presumably remote or at least nonindustrialized sites in Alaska (Malins et al., 1986), the Virgin Islands (Butler and Schutzmann, 1978), and the Hawaiian Islands (Stout and Beezhold, 1980). Further, these data indicate the the highest concentrations have occurred in fishes from urban embayments on the Pacific and East coasts and near Pensacola, Florida; concentrations generally have been lower in tissues of fishes from the southeastern and Gulf of Mexico estuaries.

As described in Chapters 5, 6, and 7 declining PCB concentrations have been clearly recorded for Atlantic menhaden and striped bass in the New York-Hudson River area, and for mussels and flatfish near the Palos Verdes in Los Angeles County, and for bivalves near Pensacola, Florida. However, all three of these regions were at or near known major discharges of PCBs that were subject to source control.

National trends in PCBs are not yet clear because few species and tissue types have been consistently resampled using the same methods and analytes over long periods (a decade or more). On the basis of available comparable data, it is evident that there has been no dramatic national change, or at most a minor decline, in PCB contamination of fish and shellfish over the past 10 to 15 years.

Table 4.3 Comparison of average PCB concentrations in livers of nearshore and estuarine fish from nine areas occupied by national surveys 1976-77 and in 1984.

Area		tPCB, ppm ww 1976-77	1984
Western Long Island Sound		0.62[a]	0.81b[c]
Lower Chesapeake Bay, Virginia		0.62[a]	0.28[b]
Duwamish River/Elliott Bay, Washington		26.7[c]	4.23[d]
Nisqually Reach, Washington		0.31[c]	0.49[d]
Columbia River, Oregon		0.24[a]	0.20[d]
Coos Bay, Oregon	(less than)	0.20[a]	0.15[d]
Southern San Francisco Bay, California		0.22[a]	1.23-2.30[d]
Palos Verdes/San Pedro Canyon, California		18.63[c]	2.27[d]
Dana Point, California		0.07[c]	0.38[d]
Median		0.31	0.49
Range: minimum	(less than)	0.20	0.15
maximum		26.7	4.23

[a] From original data supporting Butler (1978)

[b] From original data supporting Zdanowicz et al. (1986) and OAD (1987)

[c] From original data supporting Sherwood (1982)

[d] From original data supporting Malins et al. (1986) and OAD (1987)

This conclusion is consistent with observations in inland fish. Schmitt et al. (1985) report that the geometic mean of tPCBs in freshwater fish from 99 inland United States sites decreased by less than a factor of 2, or from 0.88 ppm ww in the 1976-77 sampling season to 0.53 ppm ww in the 1980-81 sampling season.

4.2 DDT

DDT compounds have been the most widespread and frequently sampled chlorinated hydrocarbons and, unlike PCBs, levels of DDT have declined dramatically over the past 15 years in U.S. coastal fish and shellfish--perhaps by as much as 100-fold on a national basis.

The OAD digitized data base contains over 20,000 entries for DDT, or its metabolites, which have been measured in hundreds of species of fishes and invertebrates from United States and adjacent marine and estuarine waters. DDT has been detected in samples from every coastal state, at many offshore and deep-water sites, and from nearly every estuary.

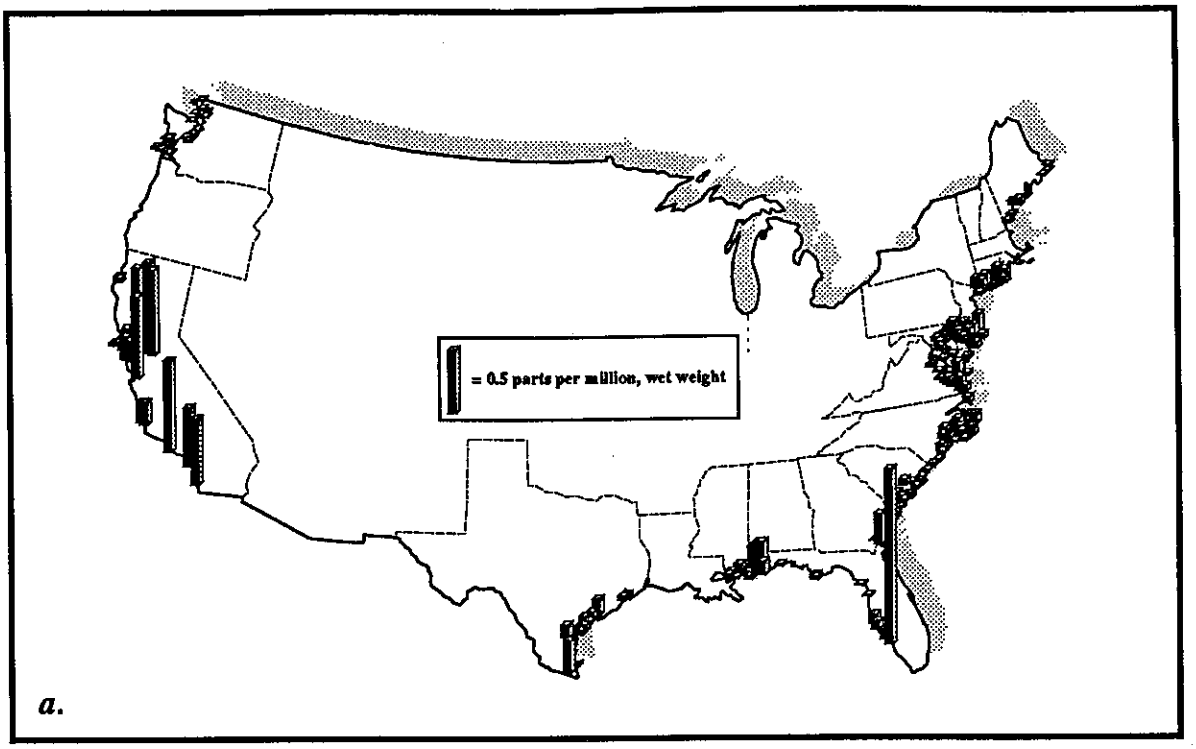
It is difficult to determine exactly when DDT was first reported from U.S. marine ecosystems. A survey of chlorinated pesticides in oysters from six Southern States was conducted in 1964-65 (Bugg et al., 1967; Rowe et al., 1970). DDT, dieldrin, toxaphene, and lindane were reported in various marine species that may have been collected in 1964 from Suisun Bay near San Francisco, California, (Bailey and Hannum, 1967) and in other California marine bivalves and anadromous fish collected sometime between 1963 and 1965 (Keith and Hunt, 1966). However, based on the retrospective analyses of museum fish specimens as reported by MacGregor (1973), there is no question that DDT metabolites were present in U.S. Pacific marine fish as early as the late 1940s.

In general, concentrations of tDDT in fish muscle ranged from below detection to a maximum of 437 ppm ww from a 1974 sample of Dover sole from the Palos Verdes Peninsula, California (LACSD, unpublished data); details of other significant national data are reported below. By comparison, the FDA action limit is 5.0 ppm ww and the NAS (1974) recommended criterion for predator protection is 0.05 ppm ww. The highest reported level of DDT in fish liver was 1,589 ppm ww from an English sole collected from the Palos Verdes Peninsula in April 1977 (SCCWRP, unpublished data).

4.2.1 DDT Trends in Bivalve Mollusks

DDT metabolites were found in 63 percent of the 8,095 samples of oysters, clams, and mussels from 180 sites sampled during the NPMP (Butler, 1973). Time-averaged site mean concentrations of tDDT ranged from below detection to a maximum of 1.4 ppm ww at Iona Point in southeast Florida (Figure 4.7a). The 180-site median was 0.024 ppm ww (range $\leq$ 0.0075 to 1.4). Six sites, five in California and one in Florida, had time-averaged site means exceeding 0.5 ppm ww (Figure 4.7a).

The maximum tDDT concentration from an individual composite was 5.4 ppm ww from a sample of eastern oyster at Iona Point. Seven additional sites, six in California and one in Texas, produced individual bivalve composites containing in excess of 1.0 ppm ww.



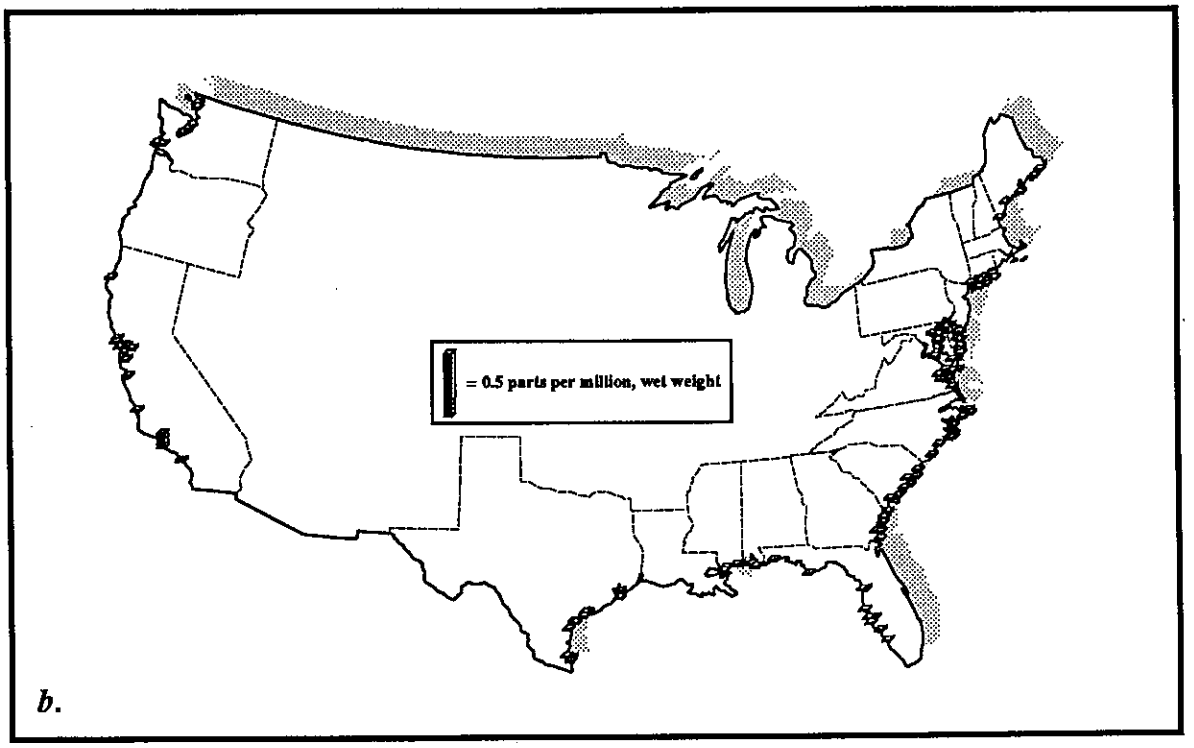


Figure 4.7. Total DDT in estuarine bivalves from 89 estuaries sampled in (a) 1965-72; and (b) 1977. Computed from original data supporting Butler (1973), and Butler et al. (1978). Bar represents mean of (generally) two composite values at each site.

When the NPMP survey resampled all sites in 1977 (Butler et al., 1978), tDDT concentrations had fallen below the 0.01 ppm ww detection limit everywhere except at the Point Mugu Lagoon site near Oxnard in Southern California (0.12 ppm ww) and at six sites in upper Delaware Bay which averaged 0.033 ppm ww; (Butler et al., 1978). Thus there was a dramatic nationwide decline in bivalve DDT contamination during the early 1970s. The magnitude of this nationwide decline and the persistence of the "hot spots" is apparent from a comparison of Figures 4.7a and b.

During 1976-78, the EPA Mussel Watch also analyzed DDE (not tDDT) in bivalves from 80 of their nationwide sites. These data (Figure 4.8) are in agreement with those of the 1977 NPMP (Figure 4.7) in that the same remaining "hot spots" were identified. In addition, the sensitivity of the EPA survey was much higher (i.e., detection limits were lower). As a result, many of the sites listed, as "below detection" by Butler, et al. (1978) actually yielded values on the order of 0.001 to 0.010 ppm ww for DDE.

Preliminary analyses from the 1986 NOAA NS&T Mussel Watch (NOAA, 1987) indicate that the national grand median of 145 sites was about 0.003 ppm ww for tDDT (range approximately 0.00002 to 0.111), a value approximately 10 percent of the national median of .024 from the 1965-72 estuarine mollusk survey. Thus, DDT concentrations in U.S. bivalves on a national basis have declined nearly an order of magnitude during the past two decades, and most of that apparently occurred prior to 1976.

4.2.2 DDT Trends in Whole Fish

DDT metabolites were also measured in whole animal composites of juvenile estuarine fish during the 1972-76 hiatus in the NPMP shellfish monitoring. In addition, the estuarine fish survey included states and territories not otherwise sampled in the shellfish program: Hawaii, Alaska, Oregon, Connecticut, Rhode Island, Louisiana, Puerto Rico, and the Virgin Islands. Conversely, Massachusetts was excluded from the fish program. The grand national median was 0.014 with a range of 0.01 to 2.1 ppm ww. In general, geographic patterns for tDDT in estuarine fish were similar to those for the shellfish--that is, the highest time-averaged site concentrations (in this case, greater than 0.5 ppm ww) were at sites in the Northeast (Delaware Bay and the Chesapeake-Delaware canal) and in the Southwest (Southern California) (Figure 4.9). However, there were some notable differences between the data sets. Juvenile fish from the Caloosahatchee River, Florida produced fish containing low DDT concentrations typical of much of the coast at that time (less than 0.1 ppm ww). This observation is important because the site is near Iona Point, which contained the most contaminated oysters in previous years (see above). In contrast, two sites near southern Laguna Madre, Texas (near Port Harlingen and Arroyo City, respectively) had the distinction of producing the highest and second highest site-averaged tDDT in the nation (2.1 and 1.7 ppm ww, respectively) (Figure 4.9); whereas during the 1965-72 period, shellfish from at or near these sites were only marginally contaminated with DDT (by less than a factor of 10 above the coastal average). Further, none of the newly-added states or territories lead to the identification of additional prominent DDT "hot spots." However,

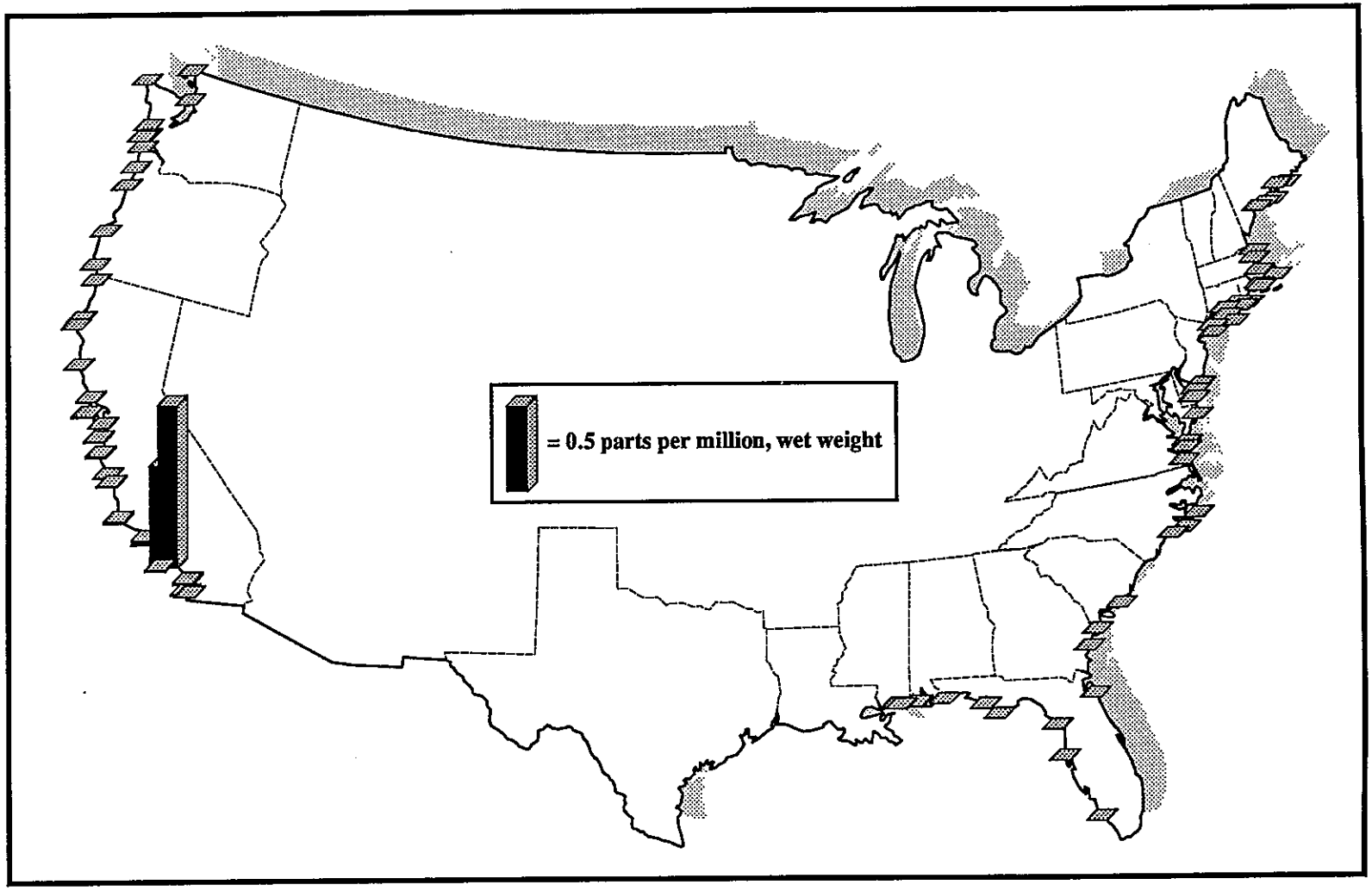


Figure 4.8. DDE in bivalve mollusks during the 1976 U.S. EPA Mussel Watch. Computed from dry weight values in Farrington et al. (1982) and moisture content given in Goldberg et al. (1978), and in original data supporting Farrington et al. (1982). Bar represents mean of duplicate composite samples. Species vary among sites.

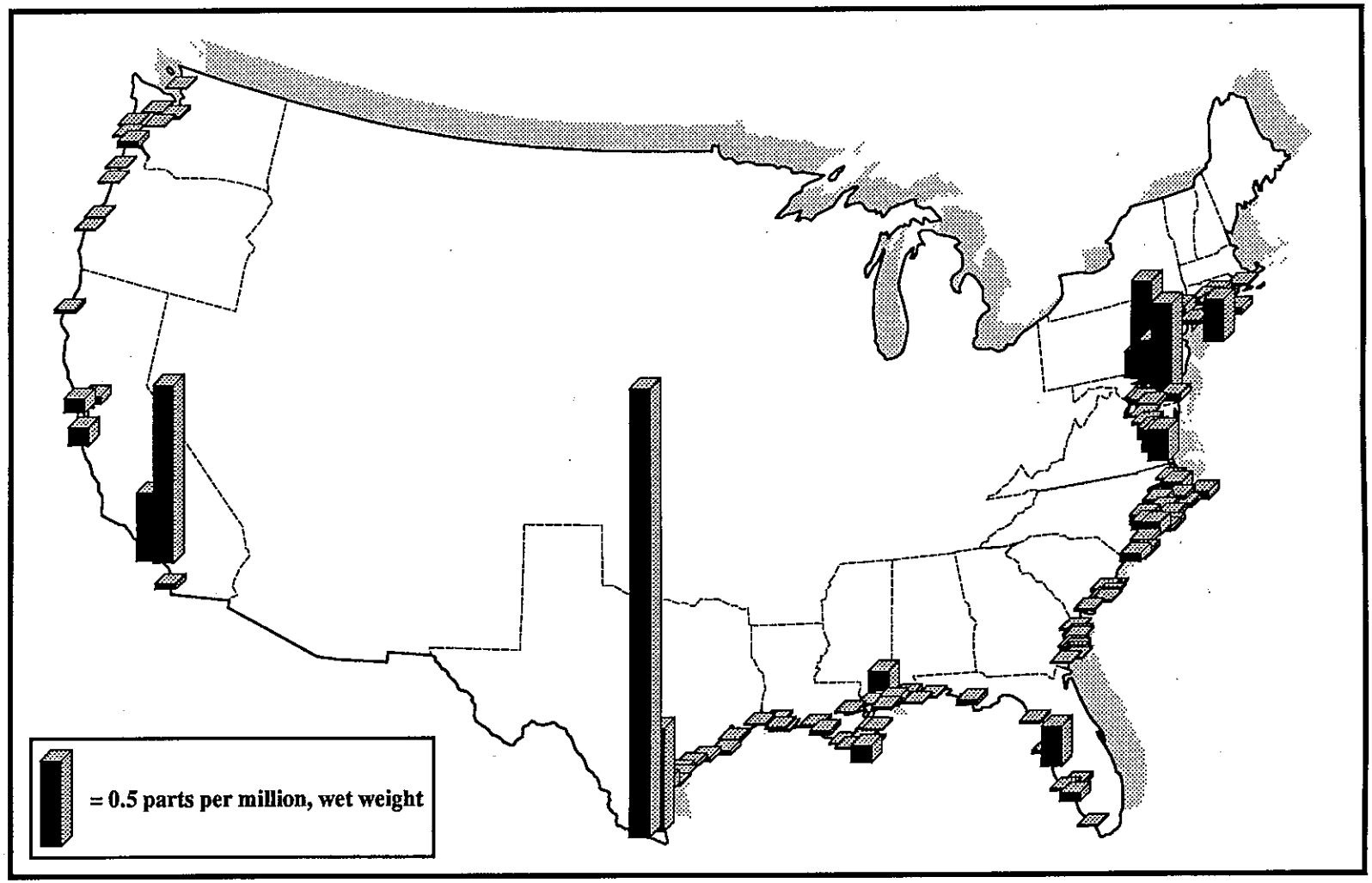


Figure 4.9. Total DDT in whole juvenile estuarine fish, 1972-1976. Bar represents mean of all composites for all species collected at a site. Sample sizes and numbers vary. Computed from original data supporting Butler and Schutzmann (1978).

marginal concentrations of DDT were found in juvenile estuarine fish from the Columbia River Estuary and Coos Bay, Oregon; from sites at or near Mayaguez, Punta Congreos, and Rio de la Plata, Puerto Rico; from three sites along the western Louisiana shoreline (but not in the delta region); from Narragansett Bay, Rhode Island; and from four Connecticut sites.

Concentrations in the juvenile whole fish survey were lower than those observed in whole menhaden samples independently analyzed by Stout (1981). During the period 1969-71, mean levels in menhaden, a pelagic fish of the Atlantic and Gulf coasts, ranged from less than or equal to 0.1 ppm ww in whole fish from Florida to about 1.2 ppm ww in fish from New Jersey (Figure 4.10a); Gulf of Mexico menhaden, landed at plants in Louisiana and Alabama averaged an intermediate value of 0.4 ppm ww at this time. Stout continued sampling through 1979 when levels everywhere were largely below 0.1 ppm ww except for whole menhaden delivered to North Carolina plants (0.5 to 1.0 ppm ww). Data through 1975 are shown in Figure 4.10.

4.2.3 DDT in Fish Muscle or Flesh

Stout and co-authors sampled coastal and nearshore marine fish muscle tissue at various localities along the central and southern East Coast, the Gulf of Mexico, in the northeast Pacific from the Bering Sea south along the Canadian and U.S. Pacific Coast, and Hawaii (Stout, 1980; Stout et al., 1981; Stout and Beezhold, 1981). During the decade 1969-79, these were the only nationwide surveys of DDT in marine fish flesh prior to the ban and controls of 1971-72. At that time (1969-71) tDDT concentrations in Pacific coastal and offshore fish were astonishingly high and appeared to increase nearly 10,000-fold from north (Bering Sea and Gulf of Alaska, (equal to 0.010 ppm ww) to south (Southern California, maximum 76.3 ppm ww; Figure 4.10a). An area-weighted compilation produces a Pacific coastal fish muscle median of 0.71 ppm ww. Highest concentrations occurred in muscle of rockfish and sablefish from Santa Monica Bay, California, where maximum composite concentrations ranged from 20 to 75 ppm ww or 4 to 15 times above the FDA action limit of 5.0 ppm ww. Other samples from here and elsewhere also above the FDA action limit were: Pacific bonito from off Ventura, California (8.1 ppm ww); sablefish from offshore of the Los Angeles Orange County area (7.5 to 9.3 ppm ww); and collections of sablefish from off Santa Cruz, California, in the Monterey Bay area (mean 8.9 ppm ww). Fish bearing flesh concentrations within a factor of 10 of the FDA action limit (i.e., greater than or equal to 0.500 ppm ww) were reported in all coastal and shelf catches sampled from off Grays Harbor, Washington south to the United States-Mexico border (a distance of 1,500 kilometers (km) and offshore 150 km) to Cortez Bank off the United States-Mexico international boundary.

We noted that fishes from Northwest Coast bays and returning salmon caught from bays and rivers, contained lower concentrations of DDT than did resident offshore fish (i.e., Puget Sound and Grays Harbor fish greater than or equal to 0.13 ppm ww), which supports an independent observation by Risebrough (1969) that Pacific coastal and ocean fish were more contaminated than bay and estuarine fish (including those in San Francisco Bay).

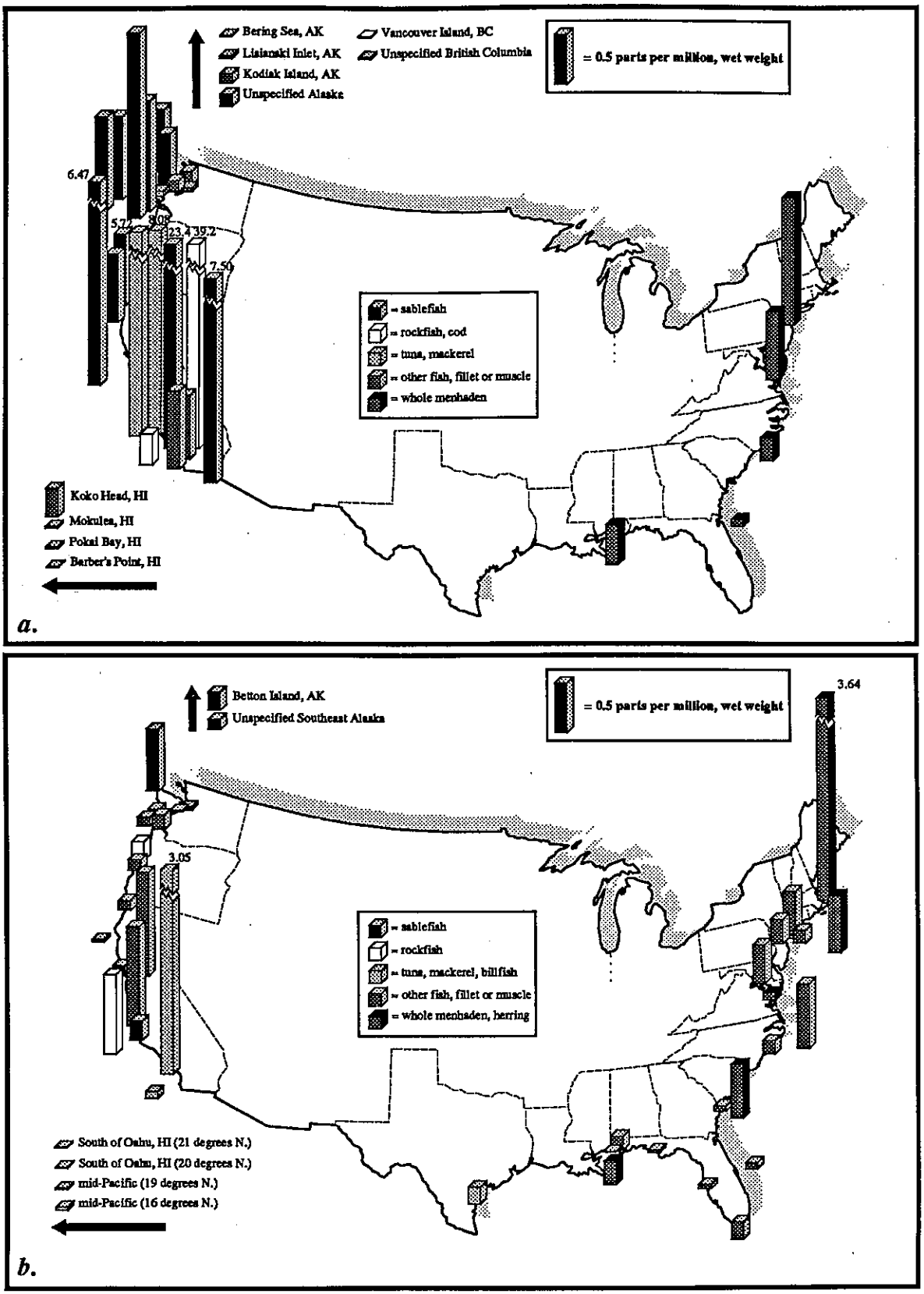


Figure 4.10. Total DDT in fillets (muscle of many species) or whole fish (menhaden) for (a) 1969-71, and (b) 1972-75. Bar represents given mean or mean computed from original data. Compiled from original data supporting Stout (1980); Stout and Beezhold (1981); and Stout et al. (1981).

Stout continued sampling following the ban and control of DDT in 1971. DDT in Pacific fish appeared to decline such that between 1972 and 1975 flesh concentrations ranged from 0.01 ppm ww in salmon returning to Puget Sound to 4.54 ppm ww in fishes from near Los Angeles Harbor; an exception was a value of 6.94 ppm ww in striped bass from the Sacramento River near Clarksburg, California. An area-weighted compilation produces a Pacific coastal median of 0.16 ppm ww or one-fifth the 1969-71 median of 0.71 ppm ww (Figure 4.10b). Within the Southern California area, no mean concentrations exceeded the 5.0 ppm ww FDA action limit; however, it was still possible to catch fish with flesh values within a factor of 10 of the limit (greater than or equal to 0.5 ppm ww). Further, fish bearing DDT concentrations above 0.1 ppm ww were, for the first time, recovered from Gulf of Alaska commercial catches.

During this period (1972-75), a special survey of pesticides in mackerels, groupers, and other coastal fish was carried out at 10 sites between Beaufort, North Carolina and Aransas Pass, near Galveston, Texas (Stout, 1981, Figure 4.10b). DDT concentrations were largely below 0.3 ppm ww. The 10-area median was 0.08 ppm ww or about half the Pacific area-weighted median of 0.16 ppm ww. Stout also provided unpublished data suggesting equivalent or slightly higher concentrations occur in muscle of Chesapeake striped bass (0.4 ppm ww), menhaden from Sandy Hook, New Jersey (0.5 ppm ww), and in Atlantic herring from an undescribed site off Massachusetts (3.6 ppm ww) (Virginia Stout, unpublished data). An area-weighted compilation of all these 1972-1975 data from Stout's collections (31 areas) produces a nation-wide coastal fish muscle median of 0.11 ppw. This is about five times higher than the median of 0.02 ppm ww compiled for tDDT in whole juvenile estuarine fish analyzed by the NPMP (Butler and Schutzmann, 1978) during about the same time.

During 1976-77, DDT was measured in muscle of fish from 19 sites as part of the NOAA/EPA CEMP (Butler, 1978) and as part of the NOAA-SCCWRP surveys cited above (Sherwood, 1982). Sites with the highest concentrations were the Palos Verdes Peninsula near San Pedro (17.4 ppm ww in Dover sole), San Francisco Bay (0.044 ppm ww in four species), and western Long Island Sound (0.022 ppm ww in four species) (Figure 4.11). Together these two data sets yield a 19-area 1976-77 median of 0.013 ppm ww. This is one-tenth the presumably comparable median of 0.11 derived from Stout's 1972-75 fish muscle data allowing one to at least hypothesize that there was a dramatic nation-wide decline of DDT in coastal and estuarine fish muscle during the mid 1970s. However, the hypothesis must be tempered with the fact that each survey used a different mix of species and sizes of fish. As seen in following chapters, specific time series data sets indicate there is no question declines in DDT contamination occurred in many areas on the Atlantic, Gulf, and Pacific coasts.

4.2.4 DDT in Fish Liver

Liver generally contains higher concentrations of DDT than muscle tissue; for example, in the 1976-77 CEMP surveys, median tDDT concentrations were 13 times higher in liver than in muscle tissue (0.147 vs. 0.011 ppm ww). Liver was the target tissue for determining trace contaminant levels in fish in the NS&T Benthic Surveillance Project. It was also a target tissue of the CEMP and NOAA-SCCWRP surveys of 1976-77 (Butler, 1978 and Sherwood, 1982),

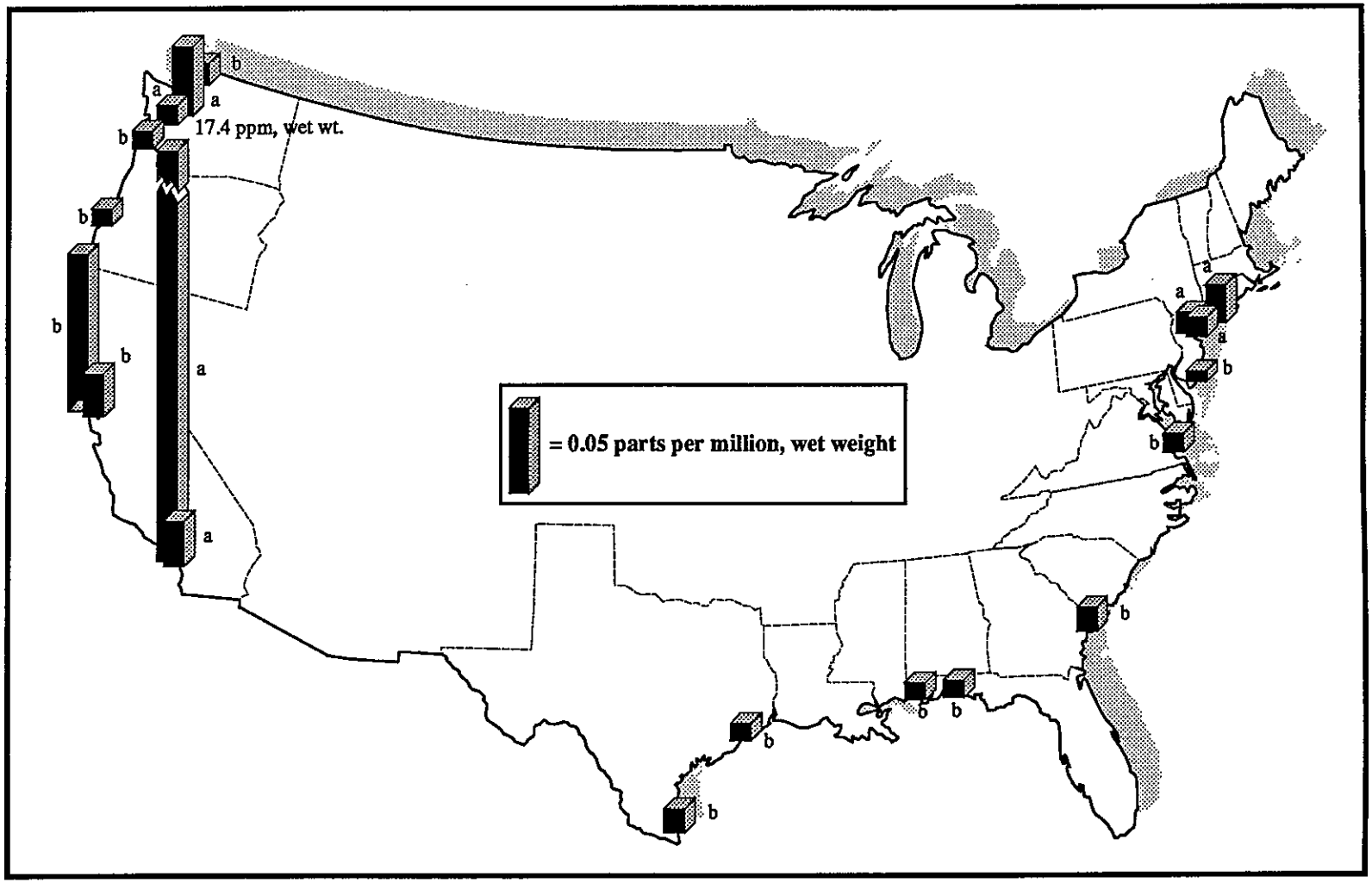


Figure 4.11. Total DDT in muscle of coastal and estuarine fish from 19 sites sampled in 1976 and 1977. Bar represents mean of all individuals for all species collected at a site. Computed from original data supporting (a) Sherwood (1982), or (b) Butler (1978).

one regional survey of Pacific fishes (Duke and Wilson, 1971) conducted by a laboratory also involved in national CEMP surveys (EPA, Gulf Breeze, Florida), and of NOAA's Benthic Surveillance Project beginning in 1984.

The CEMP liver site-averaged means ranged from 0.02 ppm ww in fish from the Savannah River, Georgia (15 samples, 4 species), to 0.97 ppm ww in fishes from San Francisco Bay (Figure 4.12). The 13-site median was 0.11 ppm ww. The maximum individual or composite concentration was 2.66 ppm ww in liver from a spotted sea trout from southern Laguna Madre, Texas. Complimenting this is data from the NOAA-SCCWRP study (Sherwood, 1982) which produced DDT concentrations ranging from 0.05 ppm ww in livers of starry flounder from the Nisqually River area of southern Puget Sound to 326.6 ppm ww in livers of Dover sole from the Palos Verdes Peninsula near Los Angeles (also plotted in Figure 4.12). The seven-area median was 0.66 ppm ww. Together, these two national surveys occupied 19 areas, yielding an overall fish liver tDDT median of 0.22 ppm ww.

Total DDT was also measured in 1984 collections of nearshore and estuarine fishes from 42 sites nationwide, as part of the NOAA Benthic Surveillance Project. Concentrations ranged from 0.003 ppm ww from a composite of Atlantic croaker from Heron Bay, Louisiana to a maximum site-average of 5.97 for hornyhead turbot from San Pedro Submarine Canyon near Los Angeles, California (Figure 4.13). The 42-area median was 0.054 ppm ww of about one-quarter of the median from livers in the 1976-77 CEMP and NOAA-SCCWRP studies.

For 13 areas for which there is potentially comparable data from both the combined 1976-77 CEMP/NOAA-SCCWRP data and the 1984 NS&T data, the 1976-77 data produce a median of 0.22 ppm ww whereas the 1984 data produce a median of 0.06 ppm ww. This comparison further supports the hypothesis that, on a nationwide basis, by 1984 DDT in coastal fish liver declined to about one-quarter of 1976-77 concentrations. Further, both agree on identification of the estuary with the most contaminated fish (San Francisco Bay) and on the two estuaries with the least contaminated fish (Coos Bay, Oregon, and Galveston Bay, Texas); otherwise, site ranking is not exactly comparable.

One rather remarkable comparison remains. Duke and Wilson (1971) from the same laboratory that also conducted the NPMP, reported concentrations of DDT in fish collected in 1970 only along the Pacific Coast. In this study, liver concentrations ranged 10,000 fold from 0.02 ppm ww in fish from the Gulf of Alaska to 1,026 for rockfish from Santa Monica Bay. The median for Santa Monica Bay fish was 349 ppm ww and for Oregon and Northern California fish, about 1.0 ppm ww (Figure 4.14). A 12-area-weighted fish median (Washington to San Diego) is 2.7 ppm ww. For eight areas for which there is potentially comparable data 14 years later--from the 1984 NS&T surveys--the 1970 median is 5.7 ppm ww whereas the 1984 median is 0.23 ppm ww or nearly one-twentieth of the potential historical peak in DDT contamination. A general hypothesis, if not conclusion, from these comparisons is that not only have concentrations in Pacific fish declined, they have declined dramatically from astonishingly high concentrations in the past.

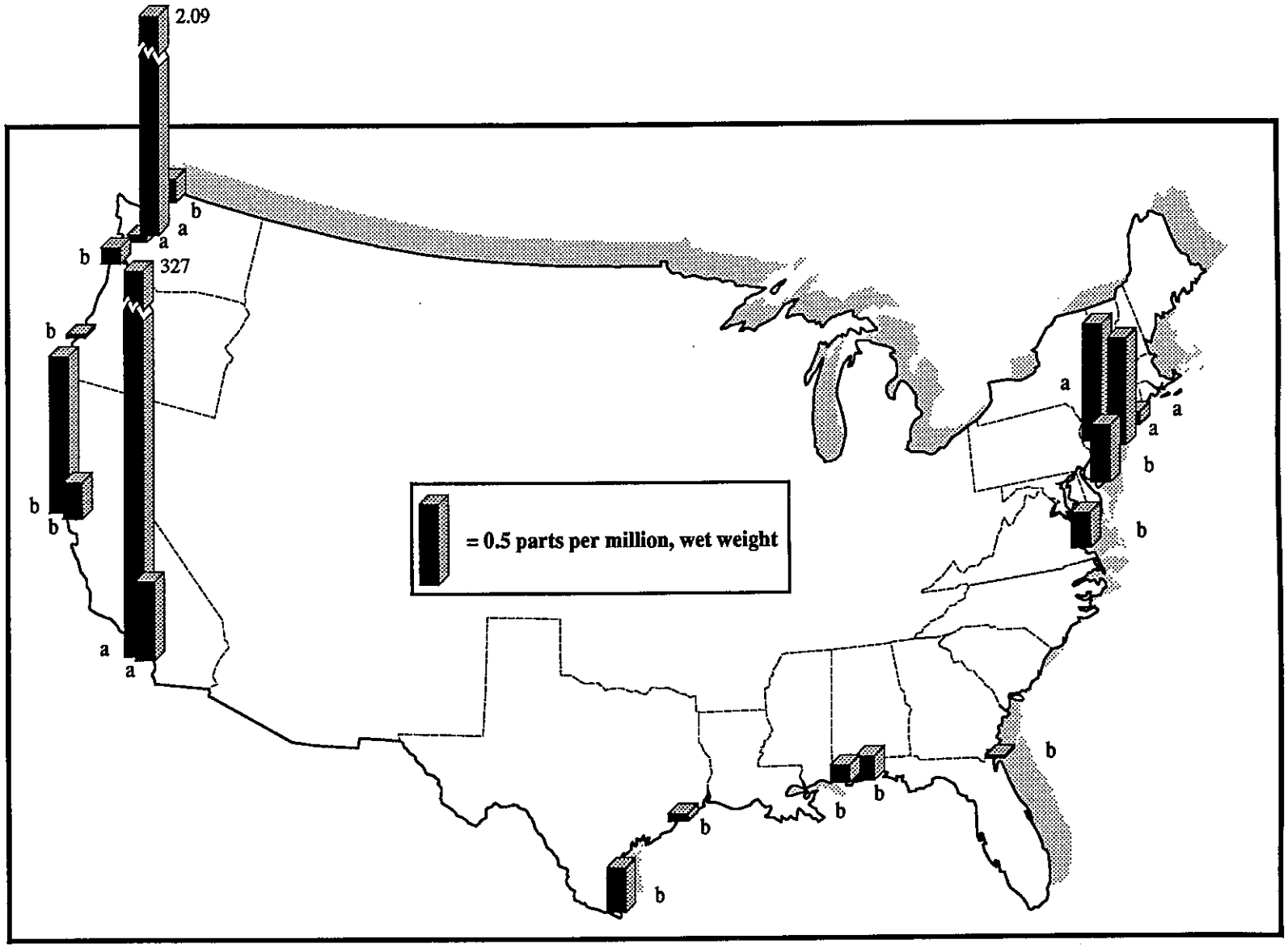


Figure 4.12. Total DDT in liver of coastal and estuarine fish from 19 sites sampled in 1976 and 1977. Bar represents mean of all individual or composite values for all species collected at a site. Compiled from original data supporting (a) Sherwood (1982), and (b) Butler (1978).

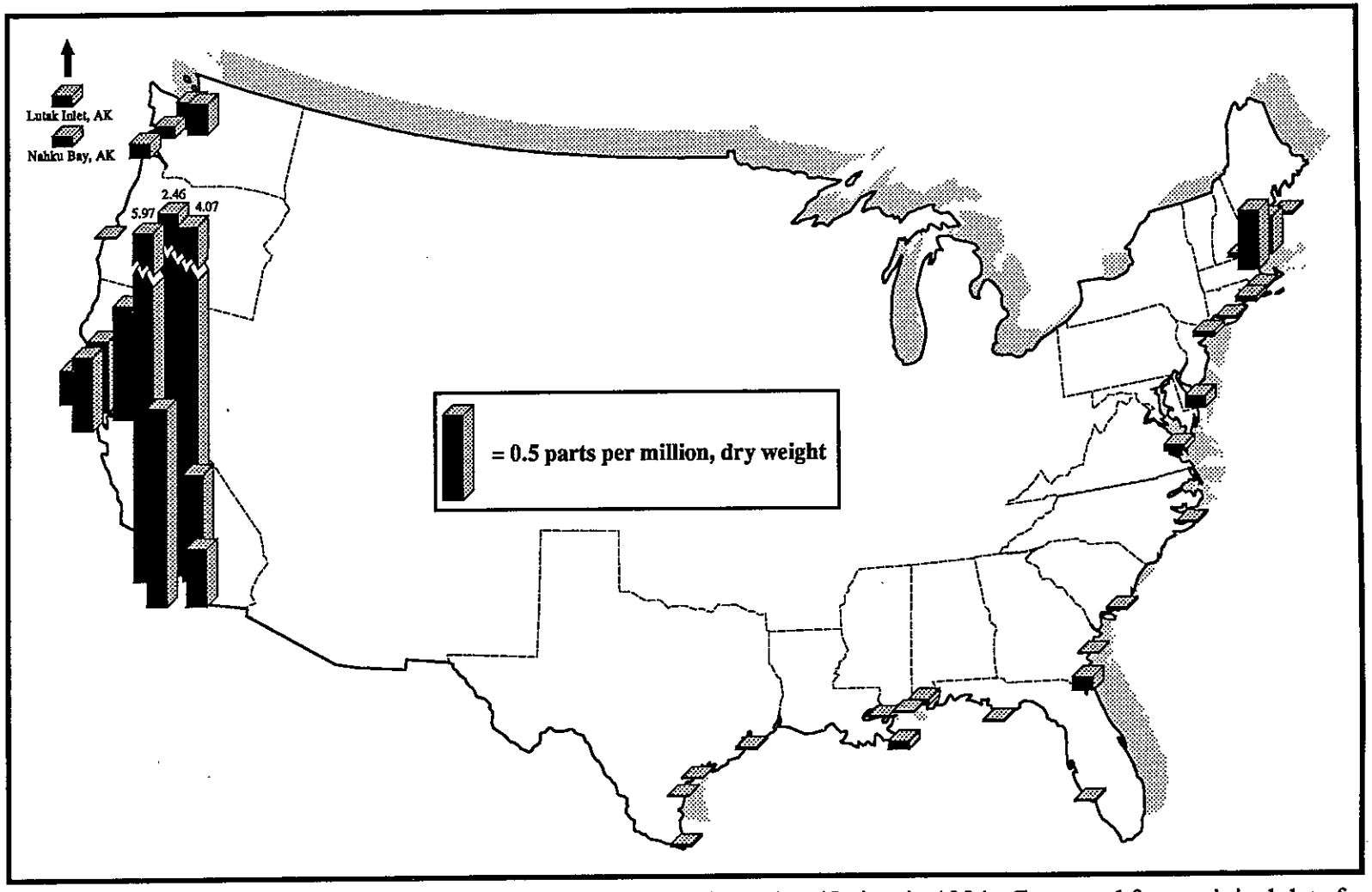


Figure 4.13. Total DDT in liver of estuarine fish composites collected at 42 sites in 1984. Computed from original data for the 1984 NOAA National Status and Trends Program, Benthic Surveillance Project supporting OAD (1987), Malins et al. (1986), and Hansen et al. (1986). Bar represents mean of 1 to 5 composite values (approximately 30 fish per composite). Species differ among sites. Compare to Figure 4.12.

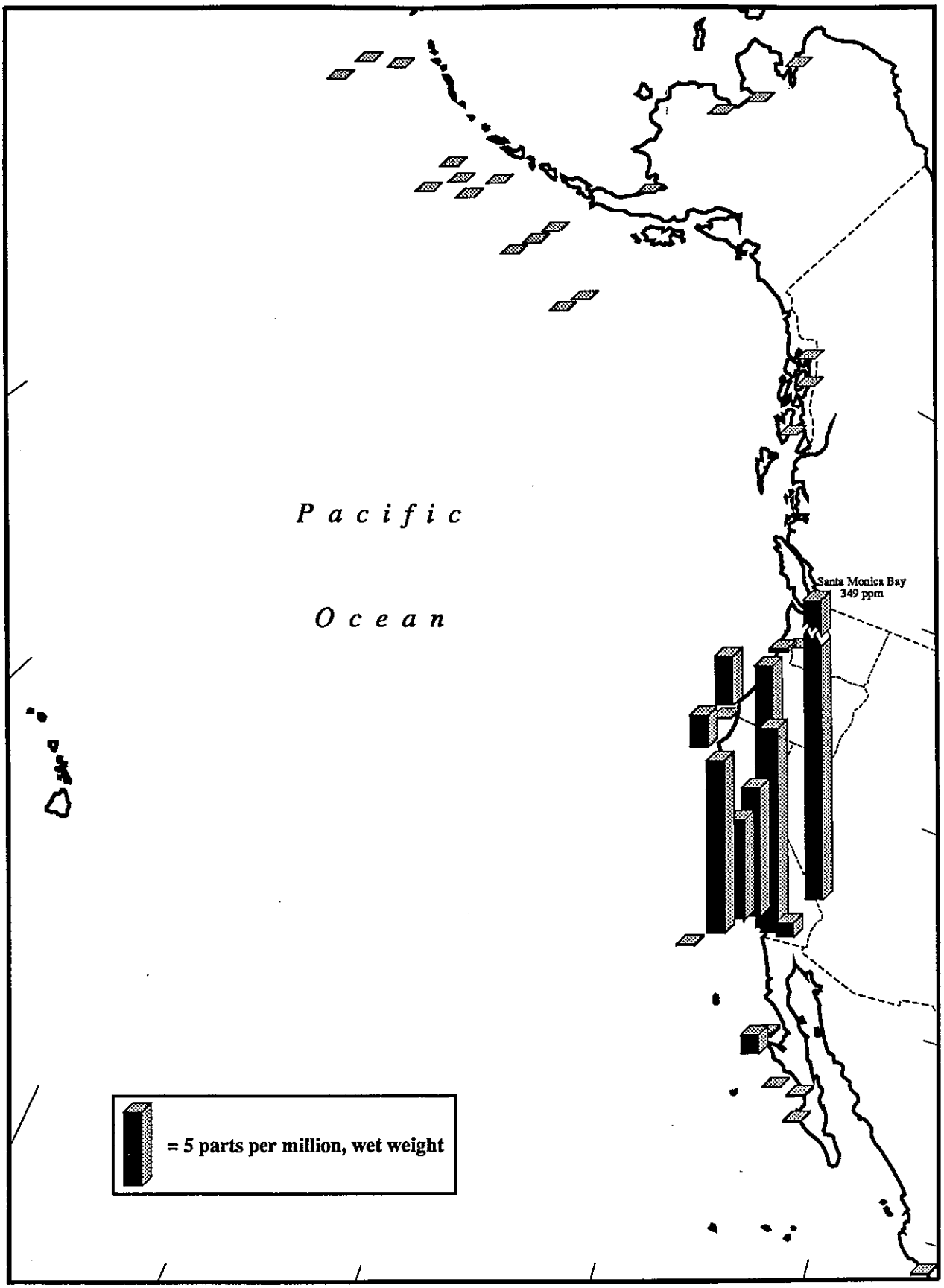


Figure 4.14. Total DDT in livers of fish from various sites in the northeast Pacific Ocean during 1970. Bar represents given value and mean. Species vary among sites. From Duke and Wilson (1978). Compare to Figures 4.12 and 4.13.

Duke and Wilson (1972) also included liver DDT data for Pacific coastal and ocean fish from the remote peninsula of Baja California, Mexico. Site-averaged liver DDT concentrations ranged from 0.10 ppm ww in barred sand bass and ocean whitefish from Bahia Magdalena to 1.28 ppm ww in Pacific mackerel taken near Isla Navidad (both off the central part of the Peninsula). Conversely, no DDT residues were detectable in livers of salmon from the northern Gulf of Alaska (Figure 4.14). The Baja California seven-area median was 0.435 ppm ww tDDT ww, a value twice as high as the current U.S. Pacific nearshore fish liver median of 0.23 ppm ww (above).

4.2.5 DDT Summary

DDT was a prevalent and widespread contaminant in U.S. estuarine bivalves and coastal fish during the late-1960s and early 1970s. The most contaminated regions were Southern California, Mobile Bay, Central California, and San Francisco Bay where shellfish were 6 to 20 times as contaminated as a national coastal shellfish grand median of about 0.024 ppm ww. In addition, an apparently isolated and short-lived "hot spot" occurred in Florida. There also appeared to be a broad low-level gradient of DDT contamination along the U.S. East Coast radiating north and south of the Delaware Bay region and perhaps, secondarily from New York (Long Island). Washington and Massachusetts had the greatest number of sites free of DDT contamination while, with notable exceptions, levels in estuarine bivalves along the Gulf of Mexico Coast were at or below the national median. By 1977, combined evidence from two bivalve monitoring programs and an estuarine fish survey indicated that on a national basis concentrations had already declined 10-fold with three coastal DDT epicenters remaining: near Los Angeles in Southern California, upper Delaware Bay, and southern Laguna Madre, Texas. Finally, based on preliminary comparisons of fish liver data and 1986 bivalve data, it is apparent that concentrations declined dramatically in the mid-1970s and continued to decline into the mid-1980s, perhaps again by a factor of 4 to 5. Thus, one can infer a decline of 80- to 100-fold in DDT concentrations on a nationwide basis between 1965-72 and 1984-86 (Table 4.4).

4.3 DDT Analogues: Methoxychlor and Kelthane (Dicofol)

With several exceptions, methoxychlor, a once widely-used DDT analog, has not been reported in measurable concentrations in any of over 10,000 samples screened. One exception is a value of approximately 0.06 ppm ww in freshwater mussels collected in December 1985 in a slough draining into Monterey Bay north of Moss Landing, California (Stephenson et al., 1986). The second is a 1974 occurrence of methoxyclor (0.017-0.06 ppm ww) in oysters from several sites in the lower Chesapeake Virginia State Health Department (VSHD).

Most of these samples (over 9,000) were from the NPMP surveys conducted between 1965 and 1977 with detection limits of 0.01 ppm ww. No national surveys have attempted to document methoxychlor since then. On the other hand, methoxychlor was detected in fish in low concentrations (less than or equal to 0.2 ppm ww) at 32 percent of the nationwide inland fish stations sampled during 1980-81 fish surveys by NPMP (Schmitt et al., 1985). Highest

Table 4.4 Median or geometric mean DDT concentrations for several national survey events

ORGANISM:	tDDT or DDE, ppm ww by sampling period			
SUBSTRATE	1965-72	1972-75	1976-77	1984-86
Bivalves	0.024[1]		0.01[2]/ 0.001[3]	.003[10]
Fish, whole juvenile		0.014[4]	ND	ND
Fish, muscle		0.110[5]	0.012[6]	
Fish, liver			0.220[6]	0.054[7]
Fish, whole F.W.	0.7 - 1.1[8]	0.4 - 0.6[8]	0.370[9]	

[1] From Butler (1973); median of 8180-site means composited from 7,839 samples.
[2] From original data supporting Butler et al. (1978); median of 89-site means composited from 188 samples.
[3] From Farrington et al. (1981); median of 80-site values or site means.
[4] From original data supporting Butler and Schutzmann (1978); median of 144-site means composited from 1,524 composites.
[5] From original data supporting Stout and Beezhold (1981) and Stout (1980); median of area- or site-means from samples.
[6] From original data supporting Butler (1978) and Sherwood (1982); median of 19-site means from samples.
[7] From original data supporting Zdanowicz et al. (1986), Malins et al. (1986), and Hanson et al. (1986); median of 42-site medians from 126 composites.
[8] From Schmitt et al. (1983)
[9] From Schmitt et al. (1985).
[10] Preliminary calculations from NOAA (1987)

concentrations occurred in fish at two sites in the lower Mississippi River Valley and at four sites in the upper Great Lakes. Thus, a decade ago, methoxychlor was a demonstrable contaminant in fish from inland areas, but not along the coast. Presumably it is not now a coastal contaminant, but there has been no recent sampling done to confirm this.

We have not yet reviewed records of kelthane, also known as dicofol.

4.4 Cyclodiene Pesticides: Dieldrin, Aldrin, and Endrin

Dieldrin, aldrin, and endrin are closely related cyclodiene pesticides. Each have been used directly as crop insecticides. However, because aldrin is rapidly metabolized to dieldrin (Schoor, 1981) it can only be expected to occur immediately after application. Of the three, endrin is the most toxic. In fact, it is the most toxic of all the chlorinated hydrocarbon pesticides and was responsible for huge basin-wide fish kills in the lower Mississippi Valley during the early 1960s (Graham, 1970).

4.4.1 Dieldrin

Dieldrin has historically been the most frequently detected cyclodiene pesticide in coastal fish and shellfish. Over 15,000 samples have been analyzed for dieldrin (over 10,000 by the NPMP alone). Concentrations in muscle tissue ranged from below detection (many as low as 0.005 ppm ww) to a maximum of 1.56 ppm ww in a 1970 sample of milkfish from the Ali Wai Canal, Honolulu, Hawaii (Schultz, 1971). Dieldrin contamination in fish near Honolulu was noteworthy. In addition to the milkfish, tarpon from Honolulu had levels ranging from 0.36 to 1.56 ppm ww or from 1 to 5 times the FDA action limit (Schultz, 1971). Two rockfish collected from off the Palos Verdes Peninsula, California, also had excessive muscle dieldrin concentrations (about 0.7 ppm ww). Intermediate concentrations were also found in coastal collections of menhaden in the mid-Atlantic region during the early 1970s (Stout et al., 1981; also see Chapter 6). Salmon from Coos Bay, Oregon, also contained dieldrin (Claeys et al., 1975).

Dieldrin was second only to DDT in frequency of detection in the 1965-72 NPMP shellfish surveys, occurring in 15 percent of all samples and from all 15 states sampled (Butler, 1973; Figure 4.15). In no case, however, was the FDA action limit of 0.3 ppm ww exceeded, but individual sites in New York, South Carolina, Georgia, Texas, and Washington approached this limit (0.08 to 0.23 ppm ww). When NPMP resurveyed bivalve mollusks in 1977, no samples at any of the 87 sites exceeded the detection limit of 0.01 ppm ww (Butler et al., 1978). Dieldrin was not again monitored in bivalves on a national basis until the 1986 NOAA NS&T Mussel Watch. These new data have not yet been examined.

Dieldrin occurred above the 0.01 ppm ww detection limit in juvenile estuarine fish from 17 percent or 25 of 144 sites sampled by the NPMP between 1972 and 1978. Areas yielding positive samples included several sites each in the Chesapeake Bay and southwest Florida, a site in Mississippi Sound, and in the Arroyo Colorado, Texas. In no case was the FDA action limit exceeded,

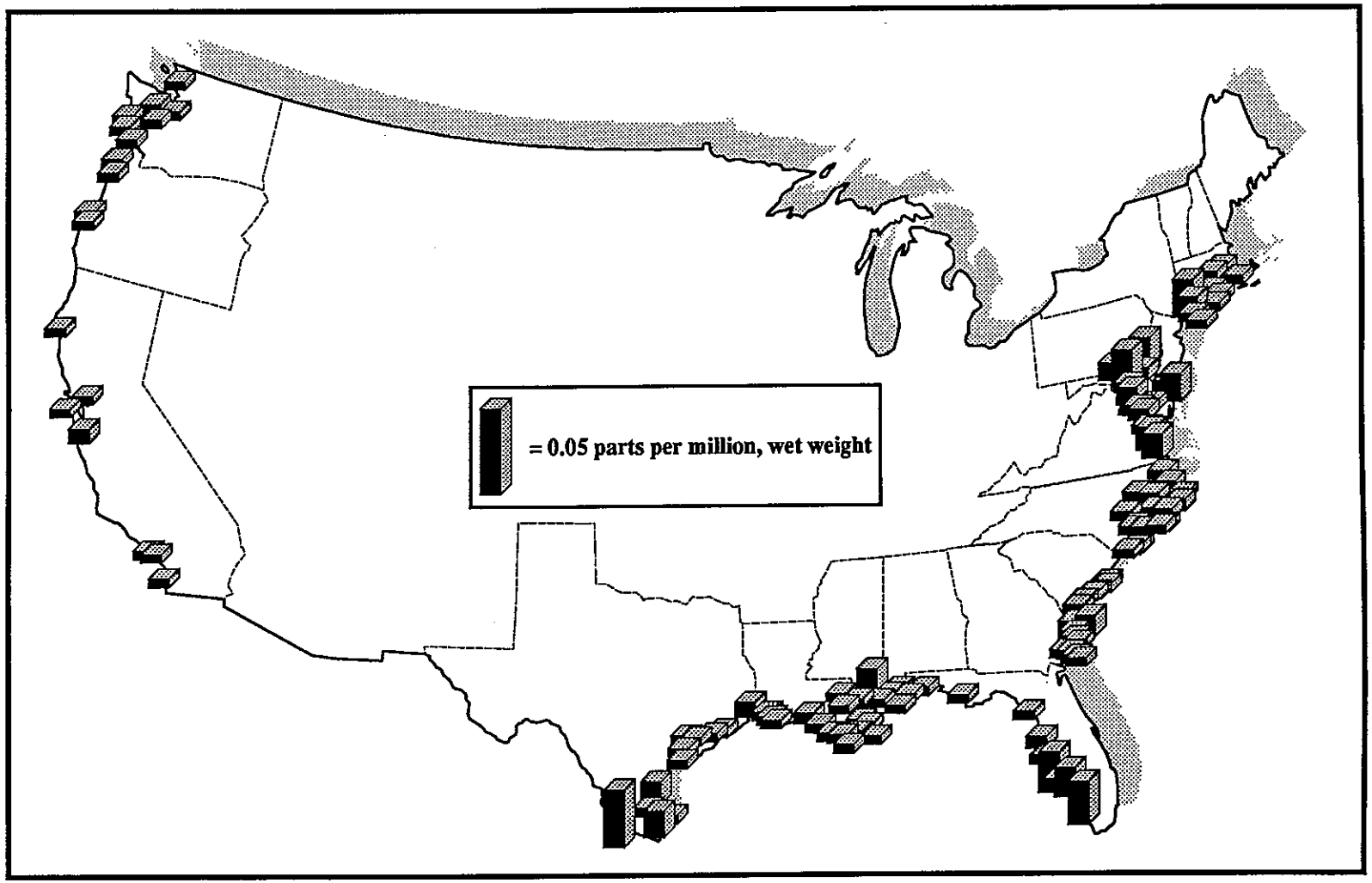


Figure 4.15. Dieldrin in juvenile estuarine fish, 1972-1976. Bar represents mean of all composites for all species collected at a site. Sample sizes and numbers vary. Computed from original data supporting Butler and Schutzmann (1978).

but it was approached, within a factor of 3, or 0.1 ppm ww, in individual samples at two sites in the Chesapeake Bay (Patapsco and York Rivers) and in fish from the Arroyo Colorado at Harlingen, Texas. Since this type of survey has not been repeated, it is not possible to judge if dieldrin has since declined in whole fish on a nationwide basis. See Chapter 7 for additional details.

Dieldrin was also a common contaminant in the 1984 NS&T Benthic Surveillance Project fish liver survey, occurring in fish livers at 28 or 58 percent of the sites at concentrations above 0.001 ppm ww. The range was less than 0.001 to a high of 0.104 ppm ww in liver of winter flounder from a site in Salem Harbor, Massachusetts (in Hanson et al., 1986; Figure 4.16). The next highest concentrations occurred in livers of starry flounder and white croaker from two collection sites in San Francisco Bay (0.09 ppm ww as computed from data supporting Malins et al., 1986). Some of the cleanest fish with respect to dieldrin were in urbanized embayments such as San Diego Harbor (less than 0.003 ppm ww) and Elliott Bay, Puget Sound (less than 0.01 ppm ww). Intermediate concentrations occurred in fish from St. Johns River, Florida; the Mississippi River Delta; San Pablo Bay, California; and Boston Harbor.

Dieldrin was the second most frequently documented pesticide in the 1980-81 NPMP whole composites of inland freshwater fish national surveys, occurring at 75 percent of the sites nationwide; nevertheless, this represented a significant decrease from previous years and compare to an incidence of 17 percent in estuarine fish during the 1972-76 NPMP survey. Mean concentrations in whole freshwater fish on a nationwide basis declined only slightly from 0.05 ppm ww in 1976-77 to 0.04 ppm ww in 1980-81, but maximum concentrations over this time decreased from 5.0 to 0.72 ppm ww, a trend due primarily to declines in Hawaiian stream fishes (Schmitt et al., 1985).

In summary, dieldrin was a common nationwide contaminant of inland and estuarine fishes. Definitive declines in concentration can be confirmed for inland sites and some marine sites. There is limited evidence to suggest that dieldrin contamination has also declined in coastal and estuarine shellfish on a nationwide basis, but further examination of data is required to determine if this also represents a nationwide trend in the marine fish. Finally, it is hypothesized that, nationally, marine and estuarine fish have been considerably less contaminated than inland fish.

4.4.2 Aldrin

Over 10,000 samples of fish and invertebrates have been analyzed for aldrin (approximately 8,000 of these by the NPMP). None of the thousands of shellfish samples from the 1965-72 or the 1977 NPMP survey contained aldrin above a detection limit of 0.01 ppm ww (Butler, 1977 and Butler et al., 1978). Nor was it reported in any of the 1972-76 NPMP estuarine fish samples from 144 sites nationwide (Butler and Schutzmann, 1978). In other surveys, concentrations in fish muscle tissue ranged from below detection levels (often as low as 0.005 ppm ww) to a maximum of 0.248 ppm ww tentatively identified in a 1975 sample of a Dover sole from the Palos Verdes Peninsula,

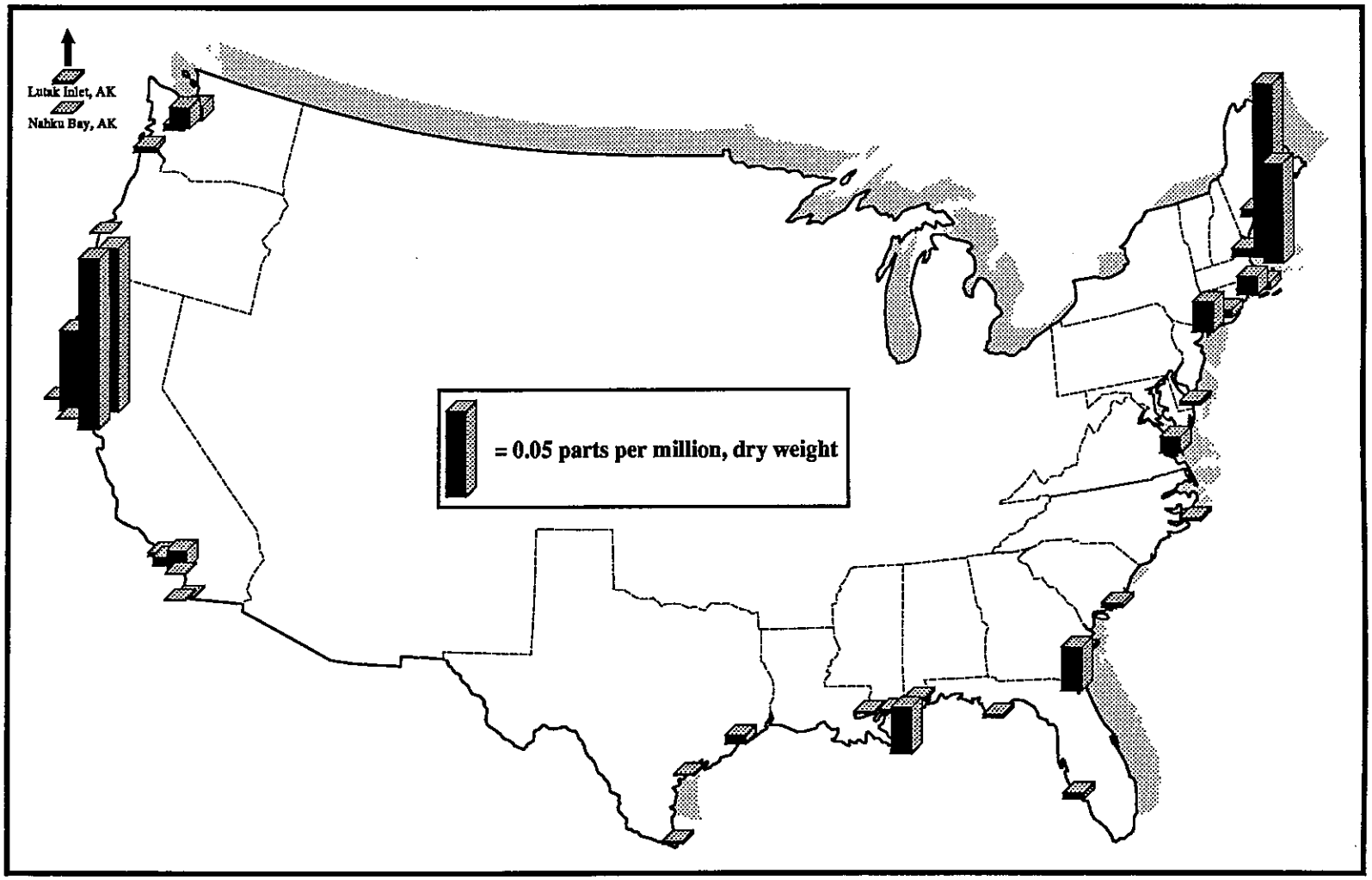


Figure 4.16. Total dieldrin in liver of estuarine fish composites collected at 42 sites in 1984. Computed from original data for the 1984 NOAA National Status and Trends Program, Benthic Surveillance Project, supporting OAD (1987), Malins et al. (1986), and Hansen et al. (1986). Bar represents mean of 1 to 5 composite values (approximately 30 fish per composite). Species differ among sites (see OAD, 1987).

California (LACSD, unpublished data). This value is close to the FDA action limit of 0.3 ppm ww. None of 46 striped bass from San Francisco Bay, the Hudson River, or Coos Bay contained aldrin above detection limits of 0.05 ppm ww (unpublished data, National Marine Fisheries Service (NMFS), Tiburon, California). Aldrin was generally at or below 0.001 ppm ww in livers of estuarine fish from the southeast, Gulf of Mexico, and Pacific Coast 1984 NS&T Benthic Surveillance Project collection sites; the highest was 0.009 ppm ww in livers of spot from Sapelo Sound, Georgia (data supporting Hanson et al., 1986). Because aldrin is rapidly metabolized to dieldrin (Schoor, 1981) and because no aldrin has been used in U.S. agriculture since 1974, it should not be surprising that aldrin has been rarely detected. Based on available evidence, it is possible to conclude that since dieldrin contamination has decreased, so has aldrin.

4.4.3 Endrin

Endrin has clearly been an important pesticide in some Gulf of Mexico and Southeastern U.S. coastal areas and in two California bays.

Over 10,000 samples have been analyzed for endrin (over 8,000 by the NPMP). Concentrations in muscle tissue ranged from below detection levels (often as low as 0.005 ppm ww) to a maximum of 0.131 ppm ww in a sample of spotted sea trout from Port Harlingen, Texas, in 1973 (Butler and Schulzmann, 1978). Concentrations in whole juvenile sea trout from this region have ranged to 0.66 ppm ww. A maximum endrin concentration of 0.26 ppm ww was also reported for whole Gulf menhaden from a site in Mississippi Sound (Stout, 1980). These values, all from mid-1970s fish collections, approach or exceed the FDA action limit of 0.3 ppm ww. Other sites producing fish with measurable endrin concentrations included Raritan Bay, the lower Chesapeake Bay, several sites in North Carolina, and St. Andrews Bay, Florida. The California Mussel Watch reported endrin at concentrations of approximately 0.007 to 0.028 ppm ww in mussels from Elkhorn Slough, Monterey, California in 1981 (Ladd et al., 1984).

Endrin occurred in bivalve mollusks from several sites in 2 of the 15 states monitored by the NPMP in 1965-72 (Butler, 1973). Samples containing greater than the 0.01 ppm ww detection limit came from southern San Antonio Bay, Neuces Bay, and the Arroyo Colorado, Texas; from several central California sites including the Sacramento-San Joaquin river mouths, Coyote Point (San Francisco Bay), a site in Drakes Estero, and in Elkhorn Slough (on Monterey Bay). Endrin was not detected at these or any other sites during the NPMP resurvey in 1977 (Butler et al., 1978). During the 1972-76 NPMP juvenile estuarine fish surveys, endrin occurred in fish from 2 of the 144 sites, both in the Arroyo Colorado near Port Harlingen, Texas (see above). Endrin has not been a target chemical of any national coastal survey subsequent to the NPMP.

Inland, endrin was detected in whole fish from 22 percent of sites sampled across the United States in 1980-81 by the NPMP (Schmitt et al., 1985). The maximum concentration was 0.3 ppm ww and the geometric mean less than 0.01 ppm ww, with highest concentrations in fish from the south and southeast (Cotton Belt) and Lake Michigan. Nationwide concentrations declined between 1976-77 and 1978-79, then increased in 1980-81 (Schmitt et al., 1985).

In summary, endrin appears to have been a contaminant of marine and estuarine fishes principally of the Gulf of Mexico region and secondarily from mid- and south-Atlantic sites. It is clear that concentrations in estuarine mollusks declined during the early 1970s, but it is not clear what is happening in fish or in any organisms since the mid-1970s. To confirm that endrin is no longer a significant coastal contaminant, it should be included in any future nationwide sampling program.

4.5 Cyclodiene Pesticides: Chlordane and Heptachlor Compounds

Chlordane compounds have been contaminants of fish from several estuaries for many years and are among several pesticides that contaminanted coastal fishes of Hawaii as well as the U.S. mainland. While concentrations do not appear to be increasing, neither are they decreasing dramatically.

Technical chlordane is a mixture of related cyclopentadiene chemicals including two isomers of chlordane (trans-, or alpha; and cis-, or beta) two of nonaclor (trans- and cis-) and heptachlor. Oxychlordane is a metabolite of chlordane and nonachlor and heptachlor epoxide a metabolite of heptachlor. Heptachlor has also been used as a pesticide separate from technical chlordane. Although chlordane itself was subject to use limitations during the 1970s it remained in use for termite control through 1986.

As noted by Schmitt et al. (1985) assessment of chlordane trends is confusing because it is not always clear which of these compounds were measured and how a "total" chlordane value is derived. It is recommended they be reported separately.

4.5.1 Chlordane, Nonachlor, and Oxychlordane

Over 12,000 samples have been analyzed for one or more of the true chlordane compounds--chlordane, nonachlor, or the chlordane metabolite, oxychlordane. Past estuarine and coastal epicenters of chlordane contamination included the Hudson River; the Patapsco River near Baltimore; Arroyo Colorado, Texas; and three sites in the Hawaiian Islands. The highest concentration of trans- or a-chlordane in whole fish was 0.71 ppm ww in alewife caught in the Patapsco River in 1973 (original data supporting Butler and Schutzmann, 1978). Other fish from this area since 1973 contained whole body concentrations in the range 0.1 to 0.35 ppm ww. By comparison, the FDA action limit is 0.3 ppm ww. The highest concentration in liver was approximately 0.2 ppm ww from a composite of white croaker caught in 1984 adjacent to the Los Angeles River (Malins et al., 1986). The highest concentrations in the striped bass were 0.54 ppm ww from fish collected from the Hudson River in 1982 and 0.17 ppm ww in fish collected in 1978 from San Joaquin River (J. Whipple, NMFS, Tiburon, California, original data). Marine fish from three sites in Kauai, Hawaii (Captain Cook Landing, Lihue Bay, and Nawiliwili Harbor) contained concentrations ranging from 0.24 to 0.34 ppm ww (Butler and Schutzmann data, 1978). Finally, a collection of spotted sea trout from Arroyo City, Texas yielded whole fish concentration of 0.19 ppm ww in 1976 (original data supporting Butler and Schutzmann data, 1978). Since 1980, at least two seafood advisories were issued due to chlordane contamination, one in the Paquonock River, Connecticut (Shultz, 1981) and one for the Patapsco River near Baltimore, Maryland (Garreis and Murphy, 1986).

It is nearly impossible to determine nationwide trends in chlordane contamination from national surveys either because concentrations were below detection or because chlordane was not searched for. Chlordane did not occur above the detection limit of 0.01 ppm ww in any of the over 8,000 samples analyzed during the 1965-72 or 1977 NPMP estuarine bivalve monitoring activities (Butler, 1973 and Butler et al. 1978). Further, chlordane was not a target chemical of bivalve monitoring again until the 1986 NOAA NS&T Mussel Watch Project.

By contrast, chlordane was discovered in at least 39 samples of whole juvenile fish from five states during the 1972-76 NPMP surveys (original data supporting Butler and Schutzmann, 1978), where the highest concentration was 0.34 ppm ww in a sample of Iao from Nawiliwili Harbor on the Hawaiian Island of Kauai. Concentration in fish at other sites in Alabama, Maryland, New York, and Texas are shown in Figure 4.17. Chlordanes were not found in any liver or muscle of fish from the 1976 19-site CEMP survey (Butler, 1978). To the best of our knowledge, chlordanes were not looked for in any other concurrent or subsequent national or seminational survey prior to the NOAA NS&T Benthic Surveillance Project.

Two chlordane compounds, alpha-chlordane and trans-nonachlor, were measured in livers of fish from 48 NS&T site collections made by NOAA in 1984 (NOAA, 1987). Using detection limits lower than those used in earlier national surveys of the 1970s, at least one of these compounds was detected in 95 percent of the site collections. Concentrations of total chlordane, defined here as the sum of the two target compounds, averaged 0.038 ppm ww with a median of 0.013 ppm ww; the range was from (0.001 to a high of 0.213 ppm ww in liver composites of diamond turbot from a site in San Diego Harbor. Total chlordane was nearly equally as high (0.01 ppm ww) in livers of fishes from off Seal Beach, California (0.212 ppm ww); in Salem Harbor Massachusetts (0.121 ppm ww); in a second species in San Diego Harbor (0.117 ppm ww);and at Hunters Point, San Francisco Bay (0.108 ppm ww).

Of the two chlordane compounds measured in fish liver in the 1984 NS&T Program, trans-nonachlor was generally higher in concentration (mean, 0.024 ppm ww). This is consistent with national data from inland U.S. whole freshwater fish monitored in 1980-81 by NPMP (Schmitt et al., 1985). In that survey, chlordane compounds were ubiquitous, occurring at individual compound averages of 0.01 to 0.04 ppm ww and trans-nonachlor was nearly always higher than any other chlordane and measured at more sites (85 percent). In addition, while there was no obvious change in total chlordane concentrations between 1976-77 and 1978-79 surveys, there was a slight decline by 1980-81. The highest whole fish concentrations were in fish from a site near Honolulu, Hawaii (to 0.77 ppm ww, alpha-chlordane alone). Other sites with high concentrations were in the Great Lakes, the Ohio River Valley, lower Mississippi River Valley, and the Cape Fear River, North Carolina.

Chlordane compounds have frequently occurred in shellfish from other local and regional surveys, such as the California Mussel Watch (Hayes and Phillips, 1985). Chlordane was second only to DDT and PCBs in abundance in 1981-82 samples of marine life from the Gulf of Alaska and Bering Sea (Kawano et al., 1986) and, in contrast to DDT and PCBs, increased in concentration

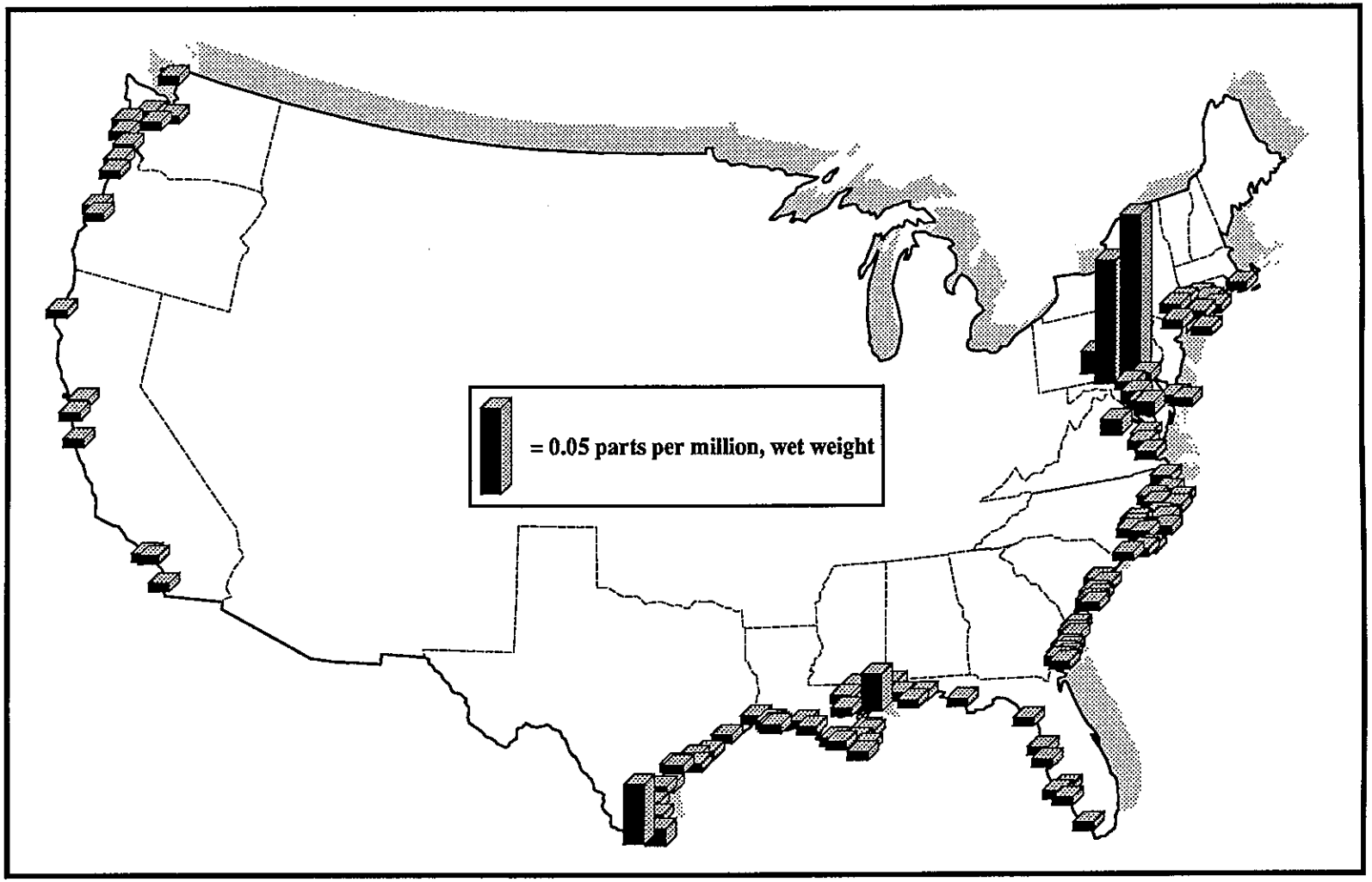


Figure 4.17. Chlordane in juvenile estuarine fish, 1972-1976. Bar represents mean of all composites for all species collected at a site. Sample sizes and numbers vary. Computed from original data supporting Butler and Schutzmann (1978). Chlordane is presumed to be total chlordane (see reference).

during 1971-82 in fishes of the Baltic Sea (Moilianen, et al., 1982). Chlordane compounds should receive continued scrutiny in national and regional marine monitoring programs. A more penetrating review of local and regional survey data is needed to clarify the status of chlordane.

4.5.2 Heptachlor and Heptachlor Epoxide

Heptachlor, and its metabolite heptachlor epoxide, have been looked for in over 12,000 samples (mainly those from the NPMP). The highest heptachlor concentrations in marine organisms have been recorded from two sites, the Weweanic River Estuary in Massachusetts and, tentatively, the Palos Verdes Peninsula in southern California. The highest concentration encountered to date was 1.55 ppm ww in a winter flounder collected in 1967 from the Weweanic estuary (Smith and Cole, 1970); other fish from this site had concentrations approaching 1.0 ppm ww. A striped bass collected in 1977 from the upper Hudson River, near Troy, had a concentration of 0.26 ppm ww (R. Sloan, New York Department of Environmental Conservation (NYDEC), 1986). Finally, several specimens of three species of fish collected from the Palos Verdes Peninsula between 1971 and 1975 had tentatively identified heptachlor concentrations ranging from 0.12 to 0.17 ppm ww (LACSD).

In contrast, heptachlor epoxide occurred in high concentrations more frequently than heptachlor in samples taken during the 1960s and 1970s. Of numerous marine fish sampled off the Palos Verdes Peninsula between 1971 and 1972, 37 had tentatively identified heptachlor epoxide concentration above 0.1 ppm ww; the highest was 0.59 ppm ww in muscle of a Dover sole collected from a depth of 60 meters near the Los Angeles County outfall in 1975 (LACSD). The maximum concentration in the Weweanic estuary winter flounder from Smith and Cole's 1967 collections was similar--0.56 ppm ww. In contrast, heptachlor was well below 0.01 ppm ww in livers of fish from the 1984 NS&T Benthic Surveillance Project survey.

Like chlordane and nonachlor, heptachlor and heptachlor epoxide did not occur above the 0.01 ppm ww detection limits in any of over 8,000 shellfish samples analyzed between 1965 and 1977 during the NPMP (Butler, 1973 and original data supporting Butler et al. 1978). Nor was heptachlor recorded in any fish from the 1972-76 NPMP surveys or the 1976 11-site CEMP program (original data supporting Butler, 1978). Further, neither heptachlor nor heptachlor-epoxide was specifically looked for in any other national or seminational survey prior to the 1984 NOAA NS&T Benthic Surveillance Project. In that program, heptachlor and heptachlor epoxide concentrations in livers of fish were largely below a detection limit on the order of 0.001 ppm ww. Mean concentration was 0.001 ppm ww for heptachlor and 0.002 ppm ww for heptachlor epoxide; maximum in both cases was 0.006 ppm ww.

In contrast, heptachlor (total) occurred in 39 percent of the NPMP inland fish samples at concentrations above 0.01 ppm ww in the 1980-81 NPMP surveys (Schmitt et al., 1985). The maximum concentration was 0.27 ppm ww. Further, there was a slight decline in inland fish between 1978-79 and 1980-81, but a slight rise just prior to that.

Based on the foregoing, it appears that heptachlor may have been a significant contaminant in several areas (Weweanic Estuary, Massachusetts and Palos Verdes, California) but not on a nationwide basis. Further, heptachlor does not currently appear to be a prominent contaminant of marine fish anywhere, including the Los Angeles area.

4.6 Cyclodiene Pesticides: Endosulfan

Endosulfan compounds may be significant contaminants of estuarine organisms in the few places that have been carefully sampled in recent years, but data is clearly insufficient to judge the current nationwide situation.

Endosulfan compounds (endosulfan I, endosulfan II and the metabolite, endosulfan sulfate) have been sought in over 4,500 samples of marine and estuarine fishes and invertebrates but detected in very few. The highest concentration of total endosulfan reported in marine shellfish is about 1.4 ppm ww in a 1983-84 sample of bay mussels from Moss Landing, Monterey County, California (reported as 7,200 ppb dw, Ali et al., 1984). The highest confirmed concentration of endosulfin in marine fish was 0.05 ppm ww in liver of a 1983 sample of fringehead sculpin from Elkhorn Slough near the same Moss Landing site (Ali et al., 1984).

It is probably impossible to make a long-term nationwide assessment of endosulfan in coastal and estuarine organisms. The only national or otherwise comprehensive survey explicitly including endosulfan as a target chemical, was the 1972-76 NPMP estuarine fish program (Butler and Schutzmann, 1978). It was not reported above the detection limit of 0.02 ppm ww from any of the nearly 1,600 samples.

Besides the Elkhorn Slough samples, endosulfan was detected in fish from an Oregon estuary (Claeys et al., 1975) and in a Pacific Coast estuary in Mexico (Rosales et al., 1982).

Fishes and invertebrates from sloughs and bays of coastal Monterey County, California, were notably contaminated with endosulfan during surveys conducted in 1980-81 (Ali et al., 1984) and contamination of mussels from Elkhorn Slough continued through 1984 (Hayes and Phillips, 1986). Concentrations in Elkhorn Slough mussels ranged up to 0.7 ppm ww. Freshwater fishes from the nearby Salinas River contained concentrations as high as 1.2 ppm ww. In 1983, seven species of marine and estuarine fishes contained total endosulfan concentrations ranging from 0.021 to 0.052 ppm ww (Ali et al., 1984). Several other California sites also produced mussels containing endosulfan: Trinidad Head, Bodega Head, four San Francisco Bay sites, Bolinas Lagoon, Pacific Grove, Newport Bay, Port Hueneme, and Santa Cruz. The last two sites, plus the Elkhorn Slough site, also experienced increasing endosulfan concentrations between 1979 and 1981 and continued contamination occurred into 1986 (Stephenson et al., 1986).

The experience in Elkhorn Slough and the Salinas River drainage of central California (as reviewed in Ali et al., 1984) suggests that endosulfan contamination deserves more attention in estuaries near agricultural drainage areas.

4.7 Cyclodiene Pesticides: Lindane and BHCs

Lindane, or gamma-BHC (also known as gamma-HCH) has been looked for in nearly 12,000 marine or estuarine fish and invertebrate samples but occurred above detection (0.01 ppm ww) in few of these. All three NPMP activities included scans for lindane, but reported none above the 0.01 ppm ww detection limit in over 8,000 samples of shellfish or over 1,500 samples of fish. In the 1984 NS&T Benthic Surveillance Project survey, lindane occurred in 47 percent of the fish liver site collections at concentrations above 0.001 ppm ww. The nationwide average was about 0.002 ppm ww and the highest mean concentration was 0.014 ppm ww in livers of Atlantic croaker from a site in the Chesapeake Bay; the other 20 lindane-positive collections produced concentrations ranging from less than 0.001 to 0.005 ppm ww.

Lindane was looked for and detected in about 90 samples of fishes and invertebrates from the Palos Verdes Peninsula and Catalina Island sampled by the LACSD; highest concentration was 0.15 ppm ww in liver of a Dover sole from the Palos Verdes Peninsula. Lindane was also detected in 28, or 44 percent, of 64 California mussel watch samples in 1980-81 through 1985-86 surveys; most of these were from San Francisco Bay and the Los Angeles area, at concentrations slightly exceeding 0.001 ppm ww (e.g., Stephenson et al., 1986). Lindane was documented in 16 percent of the 1980-81 NPMP inland fish samples at concentrations exceeding 0.01 ppm ww (Schmitt et al., 1985). Highest concentrations were in freshwater fish from a stream in Honolulu, Hawaii, in fish from Lake Mead (Colorado River), and several Great Lakes sites.

Based on all these observations, it is possible lindane was perhaps once a common contaminant in livers of estuarine and coastal fish at concentrations exceeding 0.01 ppm ww, at least in California and Hawaii. The low levels observed in fish livers during the 1984 NOAA NS&T Benthic Surveillance Project suggest that there is no nationwide lindane contamination today.

We have not fully examined data for other BHC isomers. Alpha-BHC was more prevalent than lindane and occurred at higher concentrations in the 1980-81 NPMP inland U.S. fish surveys (Schmitt et al., 1985) It may be important to look for these other isomers on a national basis in marine and estuarine organisms. Further, it may be important to more carefully examine marine fish from the Honolulu area.

4.8 Chlorinated Benzenes and Phenols

4.8.1 HCB (Hexachlorobenzene)

Hexachlorobenzene, a formerly ubiquitous fungicide, has been sampled in relatively few U.S. marine or estuarine surveys/organisms (less than 700). The highest reported concentration was about 0.7 ppm ww in liver of English sole collected in 1979 from the Hylebos Waterway in Commencement Bay, Washington (Malins et al., 1980). Measurable, but lower, concentrations were also reported in fish and shellfish from the New York Bight (MacLeod et al., 1981); Galveston Bay, Texas (Murray et al., 1980 and 1981); Upper Chesapeake Bay; the Palos Verdes Peninsula, California (Gossett et al., 1983); and near the Santa Monica Bay outfalls in California (Young and Gossett, 1980).

HCB was not included as a target chemical in the NPMP surveys or any other national or seminational survey prior to the NOAA NS&T Benthic Surveillance Project beginning in 1984. As a result, it is impossible to reconstruct a past geography or long-term trend. Livers of fish from the 48 1984 NS&T Benthic Surveillance Project site collections had a median concentration of 0.0013 ppm ww HCB and a range of 0.001 to 0.037 ppm ww. About 60 percent of the liver samples contained HCB above 0.001 ppm ww, but only 4.5 percent exceeded 0.01 ppm ww. Highest concentrations occurred at one site each in Commencement Bay (0.037 ppm ww), the Nisqually Reach (0.01 ppm ww) and Elliott Bay (0.007 ppm ww) all in Puget Sound, Washington; and in fish from Casco Bay, Maine (0.004 ppm ww).

These concentrations are similar to levels of HCB reported by Young and Gossett (1980) from livers of flatfish collected near two Southern California outfall sites, 1976-1978: mean values in fish near the outfall sites ranged from 0.024 to 0.060 ppm ww compared to a value of 0.0006 ppm at a coastal control site. In contrast, muscle concentrations in nine seafood organisms near one outfall were barely detectable in fish and shellfish (0.0006).

Schmitt et al. (1985) detected HCB at concentrations above 0.01 ppm ww in 24 percent of the samples of whole fish from the 1980-81 inland U.S. NPMP freshwater fish survey; highest concentrations (0.12 to 0.13 ppm ww) occurred in whole fish from the Tombigbee River, Alabama and from the Mississippi River at a site in Louisiana. There was also evidence of a nationwide decline in HCB contamination since 1976-77 surveys.

In summary, it does not appear that HCB is currently an important national contaminant in estuarine or marine fish relative to inland fish, but it may be in industrial waterways.

4.8.2 PCP (Pentachlorophenol)

PCP, a wood-preservative, slimicide, metabolite of the fungicide HCB and carrier of carcinogenic dioxins (PCDDs), has not been surveyed in marine and estuarine fish on a national scale. A few site-intensive studies have included PCP and detected it in fish from several embayments. Concentrations in organisms from San Luis Pass, Texas ranged from 0.002 in blue crab to 0.017 ppm ww in brown shrimp (Murray et al., 1981). Adjacent Galveston bay oysters contained PCP concentrations ranging from 0.003 to 0.008 ppm ww (Murray, 1980). Maximum concentrations in animals from sites in Puget Sound, Washington were 0.003 to 0.008 ppm ww in clams from Eagle Harbor, site of a wood-treatment operation (Yale et al., 1984). Finally, PCP has recently been documented from native and transplanted mussels from the northern part of Humbolt Bay (Hayes et al., 1986).

PCA, pentachloro-anisole, a metabolite of PCP, was detected in 24 percent of the whole fish samples in the 1980-81 NPMP surveys of the interior United States (Schmitt et al, 1985). Concentrations were generally low (0.02 ppm ww) but the highest were ten times any observed in marine fish studies: 0.03 to 0.07 ppm ww in fish from the Raritan River (New Jersey), Cape Fear River (North Carolina), Penobscott River (Maine), Mississippi River (Louisiana), and Willamette River (Oregon). It is possible that a truly

nationwide survey of PCP and related chlorophenols would identify additional estuarine contamination. This may be particularly important since PCP is a known major source of dioxin.

4.8.3 Other Chlorobenzenes

Di- and tri-chlorinated benzenes have been looked for in marine fish from several waste discharge sites in southern California (Young and Gossett, 1980; Gossett et al., 1983) and other related monocylic chlorinated aromatic hydrocarbons have been reported in striped bass from the San Francisco Bay-Delta region by Whipple et al. (1983). Dichlorobenzenes were not detected in livers of flatfish near two Southern California sewage discharge sites, but 1,2,4-trichlorobenzene was measured at a mean concentration of 0.026 ppm ww from one of the four test sites (Young and Gossett, 1980).

Some of these chemicals cause tainting. Several are used as space deodorants and in public urinals. Further, several were once used to control invertebrate pests in oyster beds (Loosanoff et al., 1960). All are now among the EPA priority pollutants. None were included in any past national or semi-national survey. Since they have been detected in a few local surveys, it may be worth including them in a future national survey.

4.9 Mirex and Kepone

Mirex, an ant poison, was once widely thought to be a serious contaminant of Southeast U.S. estuarine organisms but subsequent surveys do not suggest that was the case recently or in the past.

Mirex and kepone (also known as chlordecone) are two closely-related pesticides that were commonly used to control ants, especially in the Southeast (mirex).

4.9.1 Mirex

Over 10,000 samples of shellfish and fish have been screened for mirex. Most of these were from the NPMP and CEMP surveys (Butler, 1973; Butler, 1978; Butler et al., 1978, and Butler and Schutzmann, 1978). In all cases, mirex did not occur above the detection limits of 0.01 ppm ww in shellfish and adult fish or 0.05 ppm ww in juvenile estuarine fish.

The highest concentration in fish muscle was 0.06 ppm ww from a window-pane flounder from the New York Bight collected in 1978 (MacLeod et al., 1979). The highest concentration in bivalves was 0.54 ppm ww from eastern oysters collected at Fort Johnson, South Carolina in 1969 (Butler, 1973). Markin et al. (1975) suggested that reports of widespread coastal mirex contamination prior to 1970 may have been due to previously unrecognized PCB Aroclor 1260 peaks; in fact, in their 1971 survey of pesticides in a semi-national survey from Delaware to Louisiana, Markin et al. found mirex in only 9 of 77 seafood samples at concentrations ranging from 0.007 to 0.024 ppm ww; all were from around Savannah, Georgia. Mirex was also measured in bivalves from only nine sites in the 1965-72 NPMP surveys and all of these were from South Carolina (Butler, 1973). When all NPMP sites were re-surveyed in 1977, mirex was everywhere below the 0.01 ppm ww detection limit (Butler et al., 1978).

Mirex was not searched for again, on a national basis, until the 1984 NOAA Benthic Surveillance Project. In that survey, concentrations in fish livers at 44 sites ranged from less than 0.001 ppm ww to a maximum of 0.003 ppm ww in a liver composite of spot from the Appalachicola River, Florida (original data supporting OAD, 1986).

Mirex was measured in inland fish NPMP surveys for the first time during the 1980-81 surveys and occurred at 18 percent of the stations mainly in the Great Lakes and southeast; the maximum concentrations occurred in spotted sucker from the Altamaha River, Georgia (0.21 ppm ww) and bowfin from the Savannah River (0.12 ppm ww).

In summary, it is not certain that mirex was a widespread estuarine pesticide prior to 1970, but is is clear both concentrations were low and distribution limited mainly to the South Carolina-Georgia area in the early 1970s and that traces can still be found in estuarine fish from that area. Today, mirex does not appear to be an important national estuarine contaminant.

4.9.2 Kepone (Chlordecone)

Since its discovery in the James River in 1973, kepone has been looked for and found in thousands of samples of fish, crab, and oysters from the lower James River and in adjacent areas of lower Chesapeake Bay, Virginia. Intensive local surveys and resulting trends have been described in various review articles such as Bender and Hugget, 1980. At this time, we have only scanned the James River data (Virginia Institute of Marine Sciences (VIMS)) which indicate that by the mid-1980s, concentrations in oysters were generally below 0.1 ppm ww and in fish and crabs on the order of 0.2 to 0.8 ppm ww. Concentrations in some fish had exceeded 7.0 ppm ww in the mid-1970s and levels above 1.0 ppm ww were common.

Despite its discovery in the James, which resulted from an illicit discharge (Bender and Hugget, 1980), kepone was not included as a target chemical in any post-1973 national or seminational survey. State monitoring programs in Maryland and North Carolina included kepone. We have not reviewed these data in detail but do note that kepone was detected at low concentrations, 0.01 ppm ww in flesh of some sport fishes taken near inlets in North Carolina in 1976 (North Carolina Department of Natural Resources and Community Development, personal communication).

Presumably, kepone remains an important contaminant only in the lower James River area of Virginia. However, since no one has apparently conducted a nationwide survey of kepone, it is not certain if kepone has not occurred in other estuaries. A national survey would resolve this uncertainty.

4.10 Toxaphene

In addition to wide use as an insecticide, toxaphene has also been used in the past by fish and wildlife agencies to control trash fish in lakes (Eisler and Jacknow, 1985). It has been an important contaminant of stream and lake fishes, but its importance as an estuarine contaminant has apparently been ignored. The following review suggests it may be very important.

Toxaphene, a mix of chlorinated camphenes, has been measured in over 12,000 samples but was consistently present above detection limits (historically about 0.25 ppm ww) only in a few regions, notably southern Georgia and southern Laguna Madre, Texas. Secondary occurrences have been reported for fishes from the San Francisco Bay-Delta area; from Moss Landing, Mississippi; from East Bay, Los Angeles; and from Oso Bay, Texas (original data supporting Butler and Schutzmann, 1978); and curiously, in mussels from Hedionda Lagoon, San Diego County, California (Butler, 1973 and Cohen et al., 1982). The highest concentration in muscle was 35.6 ppm ww in a mullet and in a goatfish collected from the Back River near Brunswick, Georgia in 1971; the highest concentration in whole fish was 236.0 ppm ww in a sample of anchovy (silverside) collected from the nearby Little River, Georgia in 1970. Elsewhere, concentrations of toxaphene in muscle tissue ranged from below detection levels (0.05 to 0.25 ppm ww) to a maximum of 8.57 ppm ww in a sample of alligator gar from Arroyo City, Texas in 1974 (from NPMP data, Butler and Schutzmann, 1978).

During the NPMP mollusk surveys of 1965-72, toxaphene was below the 0.250 ppm ww quantification limit at all but 8 of the 89 sites surveyed (Figure 4.18). However, at the contaminated sites in Georgia, Texas, and California, concentrations were high: maxima were 54 ppm ww in oysters from Terry Creek near Brunswick, Georgia; 11.0 ppm ww in bay mussels from Hedionda Lagoon, San Diego County, California; and an undetermined concentration in oysters from the mouth of the Arroyo Colorado, Texas (Butler, 1973). When all sites were resampled in 1977, concentrations everywhere were below detection. However, during the 1972-76 NPMP estuarine fish surveys, toxaphene was a prevalent contaminant of fish (whole, liver, and muscle) in the Arroyo Colorado, but below the detection limit of 0.05 ppm ww everywhere else except as noted above (Butler and Schutzmann, 1978).

Since that time, toxaphene has not been a target chemical of nationwide estuarine or coastal surveys. However, this does not mean it has disappeared. Inland, Schmitt et al. (1985) reported a steady increase in toxaphene contamination of freshwater fish of the United States through the 1970s following the ban on DDT use. A plateau was reached between 1978-79 and 1980-81 surveys when the chemical was recorded above a detection limit of 0.01 ppm ww in nearly 88 percent of the stations and at a maximum concentration of 21 ppm ww and a geometric national mean of 0.27 ppm ww, which is close to the historical detection limit used in the estuarine surveys. Highest concentrations were in whole fish of the Mississippi River, the Great Lakes, and the Cape Fear River, North Carolina. In addition, the State of Virginia recorded toxaphene (1.3 - 2.6 ppm ww) in oysters from the eastern shore of the Chesapeake in 1977 (VSHD, unpublished data). It was not identified thereafter.

Therefore, it is possible toxaphene remained a significant contaminant at some estuarine sites into the 1980s and may even have increased locally and on a national basis. To underscore this point, we further review three notable local cases.

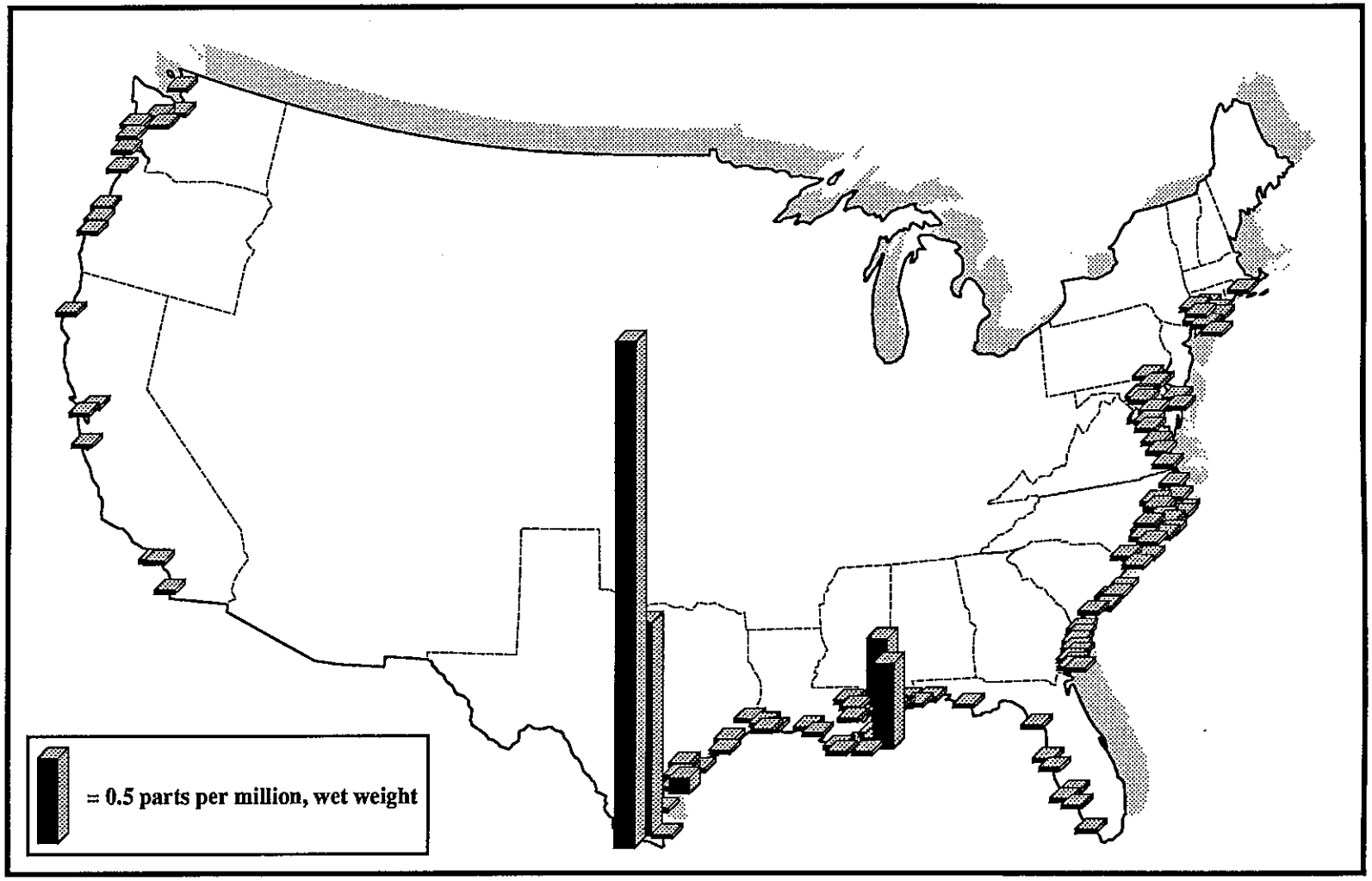


Figure 4.18. Toxaphene in juvenile estuarine fish, 1972-76. Bar represents mean of all composites for all species collected at a site. Sample sizes and numbers vary. Computed from original data supporting Butler and Schutzmann (1978).

In the first case, marine life in at least seven estuaries surrounding Brunswick, Georgia were heavily contaminated with toxaphene from wastes discharged by a pesticide plant during the late 1960s and early 1970s. Toxaphene was first noticed in estuarine shellfish from four NPMP monitoring stations in southern Georgia in 1967 (Butler, 1973). In follow-up surveys, Reimold and Durant (1971 and 1972) and Reimold (1974) reported concentrations well in excess of 1 ppm ww, and often above 10 ppm ww, in hundreds of specimens of fish (over 20 species), mollusks (3 species), and crustaceans (4 species). Sampling was terminated in early 1974 following a substantial decline in concentrations accompanying source control. Nevertheless, when last monitored, many organisms remained contaminated in the ppm ww range.

The second region of high toxaphene contamination was the Arroyo Colorado draining into southern Laguna Madre, Texas. Between 1974 and 1976 muscle tissue of spotted sea trout from Port Harlingen and Arroyo City, Texas contained concentrations as high as 5.0 ppm ww; the highest concentration in muscle was 8.6 ppm ww in an alligator gar collected from near Arroyo City in 1974. The highest concentration in liver was 23.6 ppm ww in a spotted sea trout from Port Harlingen; gonads had concentrations as high as 8.2 ppm ww. Whole menhaden also experienced concentrations as high as 8.0 ppm ww in 1974. Toxaphene contamination of the adjacent Rio Grande Valley drainage fish continued through 1979 (White et al., 1983). See Chapter 5 for a further review of this situation.

The third region of high toxaphene contamination was the delta inland of San Francisco Bay. In 1978 and 1980 striped bass had gonad concentrations in the range of 1.1 to 1.9 ppm ww (Whipple, personal communication, 1985). Inland at this time, toxaphene in freshwater fish approached 4.0 ppm ww (Cohen et al., 1982). By 1985, the California Mussel Watch had recorded chronic occurrences in mussels from Richmond in San Francisco Bay at levels approaching 0.2 ppm ww (Stephenson et al., 1986). Further, Stephenson et al. also reported high levels (up to 2.0 ppm ww) in freshwater bivalves from the Salinas River-Elkhorn Slough area near Monterey Bay and lower levels at Mugu Lagoon near Oxnard, California. Finally, a 1986 California State Water Quality Assessment Report concluded that "Toxaphene leads the list of organic chemicals found at elevated concentrations in 1984" (Lavenda, 1986).

In summary, it is possible that toxaphene neither was or is an important estuarine contaminant on a nationwide basis. However, in contrast to most other chlorinated pesticides, there is evidence from inland national surveys and from past and recent local surveys in several states, that toxaphene concentrations in near-tidal waters may have been increasing into the 1980s. Because of this, and since there has been no nationwide estuarine sampling since 1976-77, toxaphene is a prime candidate for a renewed coastal and estuarine nationwide survey. At a minimum, historically sampled sites in at least three states--Georgia, California, and Texas--should be resampled to determine what has happened at known past toxaphene "hot spots".

4.11 Carboxylic Acid Herbicides (DCPA, 2,4-D and 2,4,5-T)

With the exception of the 1965-76 NPMP shellfish and fish surveys, there has been no national effort to document the occurrence of chlorinated herbicides in U.S. marine or estuarine fish and shellfish. We have not yet completed a review of existing data. However, it is clear that the most notable case of chlorinated herbicide contamination is for DCPA (dacthal) in fishes of the Arroyo Colorado and adjacent areas in southern Texas (Miller and Gomes, 1974). Trends in this area are treated more fully in Chapter 5.

The herbicide 2,4-D, was documented in northern Chesapeake Bay oysters in 1979 and 1981 (EPA, 1982) and there is one confirmed occurrence in Alaska.

4.12 Summary and Conclusion

It is clear from the first part of this chapter that PCBs, DDT, dieldrin, and chlordane have been ubiquitous contaminants. National "hot spots" were also indicated. "Hot spots" and temporal trends for the remaining chemicals are not as clear and, in some cases, require further analysis. Keeping this caution in mind, we suggest that most urban coastal areas have been subject to contamination by at least one chemical group. A more detailed summary and conclusion appear in Chapter 8.

5.0 TRENDS IN SELECTED BAYS AND ESTUARIES

The previous chapter reviewed patterns of contamination in fish and shellfish on a national scale and demonstrated the sharp regional and long-term gradients that have existed for PCBs and several chlorinated pesticides. What has been happening within and among specific bays and estuaries? Specifically, how can contamination trends in embayments and estuaries be characterized so that comparisons can be made among them? Is contamination uniform across an embayment and changing uniformly over time, or are sharp gradients the rule? If gradients prevail, how does one represent "average" conditions so that real differences in contamination can be fairly compared among embayments and estuaries?

The answers to these questions are vital to agencies required to set priorities for waste management and estuarine rehabilitation. This chapter provides some insight. Specifically, it briefly describes some spatial, taxonomic, and temporal contamination trends in three embayments that range in scale from the very large to rather small: the Chesapeake Bay, San Francisco Bay, and the Arroyo Colorado near Laguna Madre in southern Texas.

5.1 Chesapeake Bay

Chesapeake Bay is the most heavily sampled United States estuary for chlorinated pesticides in fish and shellfish. The Bay is a 300-km long, shallow estuarine system composed of the central main stem and seven major tributaries or "segments" (James River, Rappahanock River, etc.). The bay opens at its southern (Virginia) end to the Atlantic Ocean and is separated from the ocean by the Delmarva Peninsula. Major urban centers adjacent to the main stem or its segments include Baltimore, Maryland, on the Patapsco River; Washington, DC, on the Potomac River; and Norfolk, Virginia, a major naval operations center near the junction of the entrance Hampton Roads and the Elizabeth River and, to the west, the James River. Farms ring the bay between the major urban areas. The Susquehanna River, at the far north, drains most of western Pennsylvania and adjacent areas in other states nearby. The Chesapeake and Delaware Canal connects the heads of the Chesapeake and Delaware Bays.

At least 38 surveys measured or monitored chlorinated pesticides and PCBs in Chesapeake Bay biota from 1965 to 1986. The two largest contributions to the existing data base are from the State of Maryland (1974 to present, e.g., Eisenberg et al., 1981 and annual data reports such as Allison and Butler, 1982 and special reports such as Garreis and Murphy, 1986); the NPMP (Butler, 1973; Butler and Schutzmann, 1978, and Butler et al., 1978) and kepone surveys conducted by VIMS (as reviewed by Bender and Huggett, 1984); and the State of Virginia (e.g., VSHD unpublished data, 1974-85). These, coupled with data from other surveys, yield over 4,400 samples involving 20 species of fish and 6 species of invertebrates.

The largest amount of data for Chesapeake Bay is for American oysters. On average, over 100 samples per year have been analyzed since 1965. As shown in Figure 5.1, sampling intensity for the three largest federal and state bay-wide data sources has been bimodal, with peak sampling occurring in

LONG TERM MONITORING OF DDT CONTAMINATION IN AMERICAN OYSTERS FROM CHESAPEAKE BAY

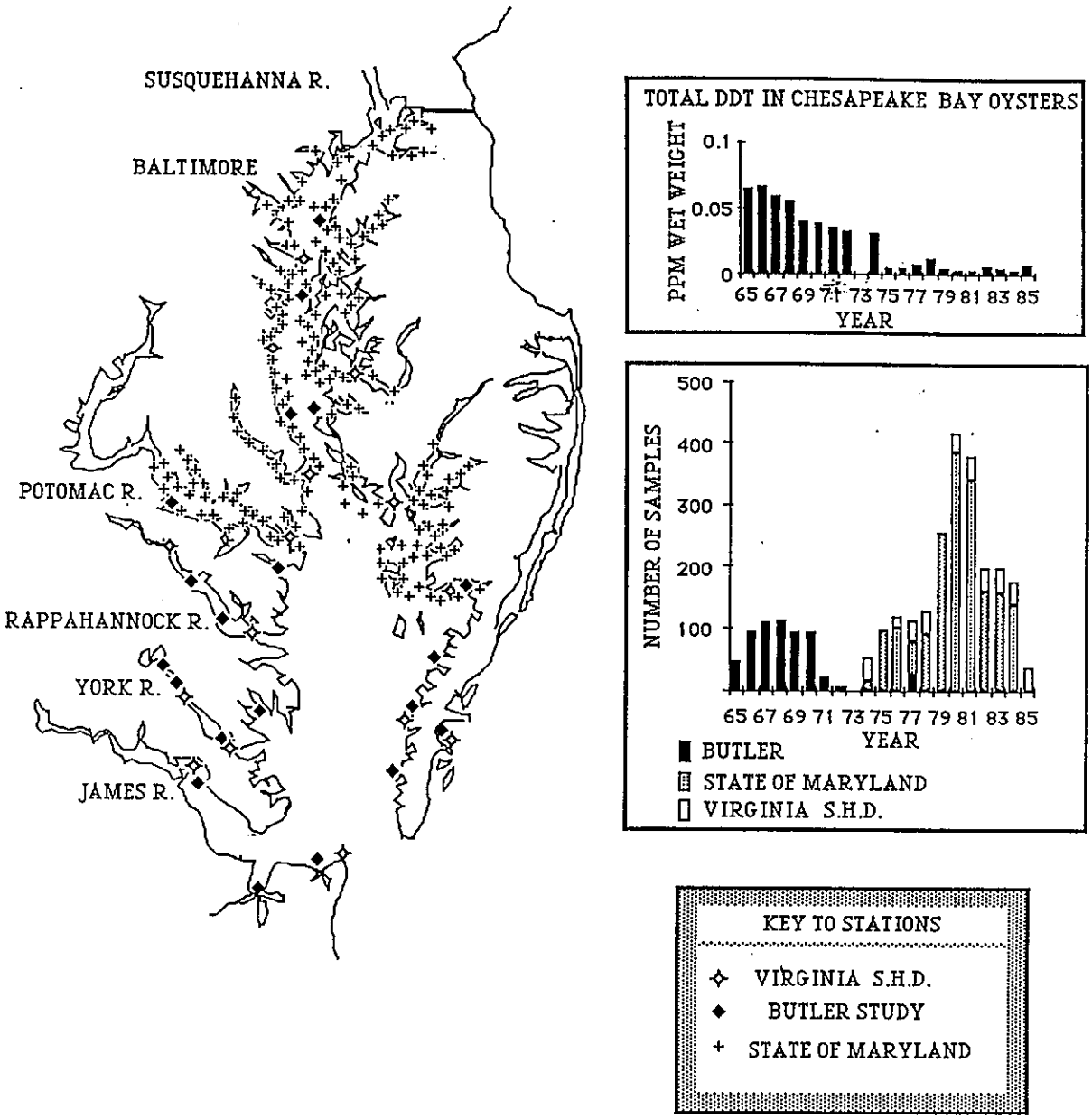


Figure 5.1. Overview of federal (NPMP) and state monitoring sites, sampling effort, and Bay-wide oyster DDT trends in the Chesapeake Bay, 1965-1985.

1967-68 (NPMP-Butler) and again in 1980-81 (State of Maryland). Also, as shown in Figure 5.1, sampling by the State of Maryland has been extensive through the upper bay whereas sampling by the NPMP (Butler) and the VSHD was concentrated at a few specific sites, but also over a long period of time (12 and 11 years, respectively).

Data confirming the history of PCB contamination in Chesapeake organisms can be traced to 1971, when the NPMP survey first recorded concentrations as high as 2.8 ppm ww in oysters from the Elizabeth River. No PCBs occurred above the NPMP detection limit of 0.05 ppm ww when NPMP revisited the sites in 1977. However, continuing from the NPMP shellfish study, and using lower detection limits, the VSHD identified PCB residues in Chesapeake oysters from various sites through 1982. Since 1974, the State of Maryland has also been involved in long-term contamination studies of Chesapeake biota, including oysters. Like the Virginia study, PCB residues were identified in Maryland shellfish after 1978. Mean PCB levels ranged from 0.15 ppm ww to just over 0.5 ppm ww in American oysters from Baltimore Harbor (Figure 5.2) and near the mouth of the Chesapeake and Delaware Canal from 1978 to 1985 (not shown). These sampling stations, which are near major urban and industrial areas, were not previously sampled by NPMP.

DDT was found in most bivalve mollusk samples taken by the NPMP between 1965 and 1977 (Butler, 1973 and original data supporting Butler et al., 1978). When all oyster data are combined (top panel, Figure 5.1) there emerges a general bay-wide trend of decreasing tDDT concentrations from a mean peak of 0.07 ppm ww in 1966 to concentrations below 0.005 (lower state detection limit) in the early 1980s. When data for some estuarine segments (Potomac, Rappahannock, etc.) are examined separately, variations on this bay-wide trend emerge. First, DDT concentration at an eastern Bay site (Pocomoke) and two western bay sites (Potomac and Rappahannock) began low (e.g., less than .03) and either declined or remained low (Figure 5.3). In contrast, concentrations at southern (Virginia) sites (James and Elizabeth Rivers) began relatively high (greater than or equal to 0.100 ppm ww) and then declined while another site (near Baltimore) was high in the late 1970s and early 1980s. Thus, the Bay-wide trend is largely driven by sites such as those in the southern Chesapeake (Virginia), but was relatively unaffected by recent higher values in oysters from sites near Baltimore.

DDT did not occur above a detection limit of 0.01 ppm ww at any Chesapeake Bay site when the NPMP re-sampled oysters in 1977 (Butler et al., 1978). However, the State of Maryland did find DDT residues at concentrations close to 0.01 ppm ww after 1978. The VSHD also identified similar low levels in oysters at southern bay sites in the fall of 1985.

Although PCBs and chlorinated pesticides have been measured in at least 25 other species in Chesapeake Bay, few offer a long-term record similar to that of the oysters. As shown in Figure 5.4, data from blue crab and spot, a small croaker, suggest that at any point in time, concentrations of DDT and PCBs were higher in crab and fish by a factor of 4 to 5 than in oysters presumably due to biomagnification. Therefore, these would be excellent species for continued monitoring to confirm that contamination is decreasing.

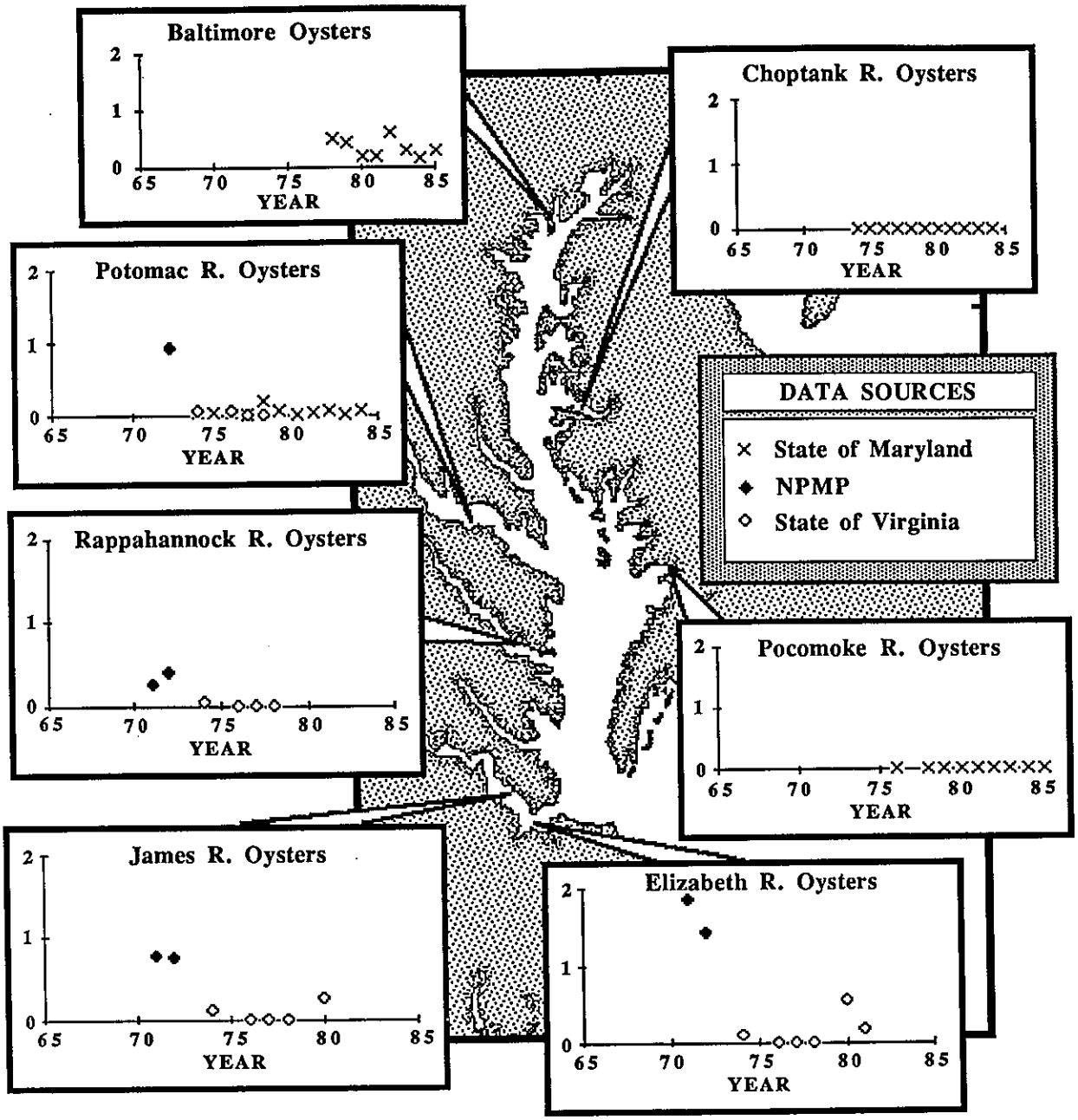


Figure 5.2. Annual variations of tPCBs (Aroclor, ppm ww) in oysters from seven Chesapeake Bay estuarine segments, 1971-85. Mean concentrations computed from data in Butler, 1973; Butler et al., 1978; and in original data provided by the States of Maryland and Virginia.

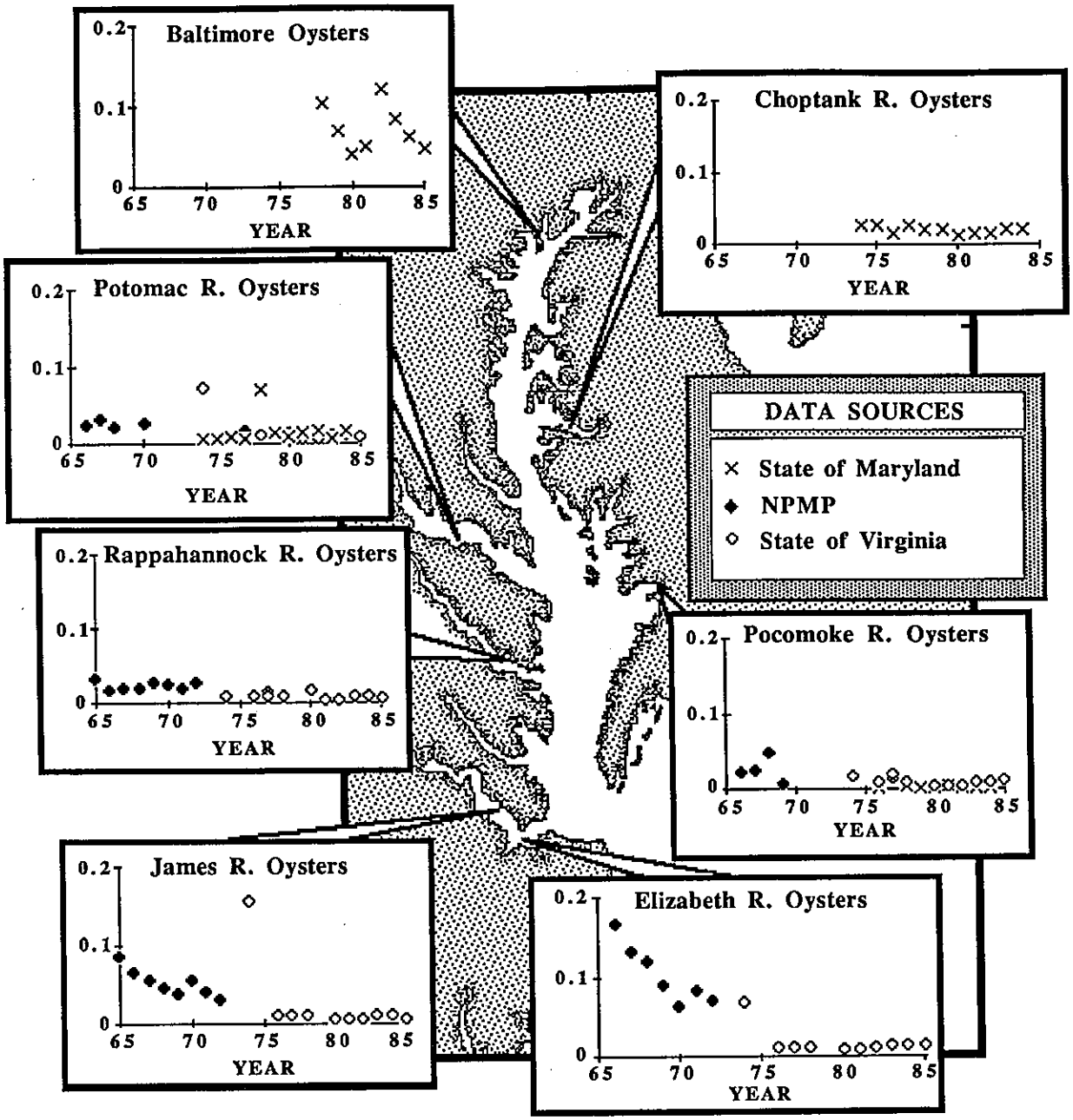


Figure 5.3. Annual variations of tDDT in oysters from seven Chesapeake Bay estuarine segments, 1965-85. Mean concentrations computed from data in Butler, 1973; Butler et al., 1978; and in original data provided by the States of Maryland and Virginia.

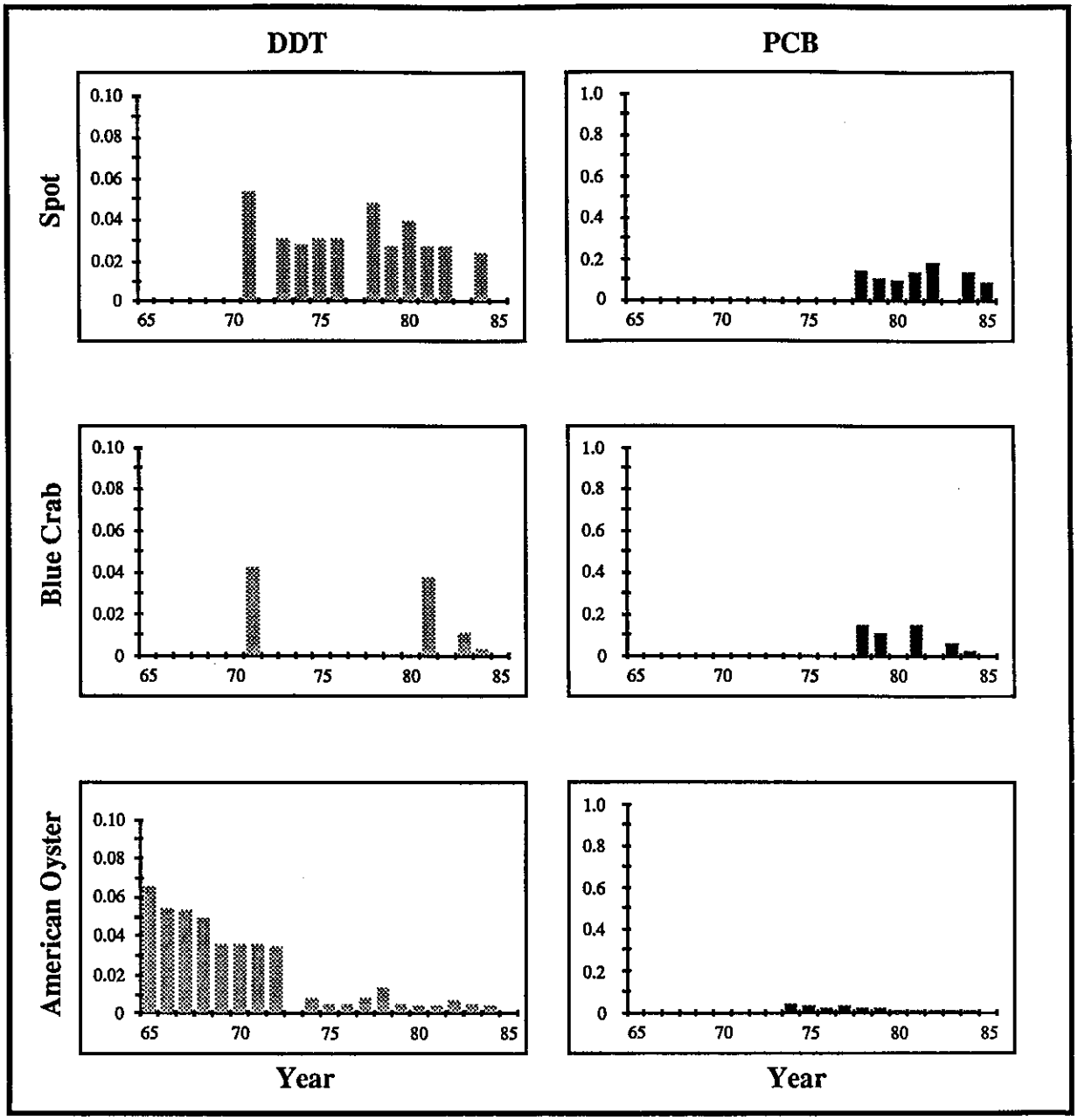


Figure 5.4. Comparison of annual variations of DDT and PCB contamination in all samples of spot, blue crabs, and oysters from the Chesapeake Bay, 1965-85. Based on data as cited previously (Figures 5.2 and 5.3). Units in parts per million, wet weight.

Dieldrin was quantified in Chesapeake oysters by Butler from 1965 to 1977, with the highest level of 0.040 ppm ww being recorded in 1967 from James River oysters. Elsewhere since 1971, dieldrin levels have been below 0.015 ppm ww in all areas of the Bay. Dieldrin was identified in James River oysters by the State of Maryland in 1974, but was never detected thereafter.

Aldrin did not occur in any oysters sampled in the NPMP between 1965 and 1977, but was identified in one sample of oysters from the Chester River in 1974 by the State of Maryland. It has not been detected by any other survey since then.

In the first year of shellfish monitoring by VSHD (1974), heptachlor epoxide (0.03 ppm ww) and methoxychlor (0.017 - 0.06 ppm ww) were detected in several sites but subsequent sampling efforts failed to identify these compounds. In 1977, toxaphene was quantified in oysters (1.3 - 2.6 ppm ww) from the Chesapeake's eastern shore but was not identified thereafter. Lindane (0.01 - 0.02 ppm ww) and alpha-BHC (0.01 - 0.06 ppm ww) were identified only during their fall 1980 sampling period. In the spring of 1981 heptachlor was identified (0.01 ppm ww) on the eastern shore of the Chesapeake. In the spring of 1982, total chlordane was identified at every site monitored with concentrations ranging from 0.01 ppm ww to 0.10 ppm ww. The Hampton Roads area contained oysters with the highest chlordane levels.

Kepone levels were quantified in oysters and other organisms from the Chesapeake by the VSHD from 1975 to 1985. James River kepone levels were the highest found in any part of the Bay. In 1975, the mean value of kepone in James River oysters was 0.221 ppm ww; since then levels have steadily declined so that by 1985, the mean concentration level was 0.022 ppm ww. The FDA action level for kepone is 0.3 ppm ww.

The chlorinated herbicides atrazine and 2,4-D were the most consistently detected herbicides in the Susquehanna and Potomac Rivers during 1979 and 1981 (EPA, 1982).

In summary, several chlorinated hydrocarbons concentrations in biota from Chesapeake Bay, although high in previous decades, have decreased. Pesticide and PCB contamination of oysters has largely been confined to the southern shore (Hampton Roads-James River region), the northwest (Patapsco River at Baltimore), and the far northern reaches including the Susquehanna River and especially the Chesapeake-Delaware Canal. In contrast, eastern shore biota have been relatively free of DDT and PCB but not of other pesticides such as toxaphene, heptaclor, and chlordane. Therefore, it is not possible to characterize the Chesapeake with one or a few sites. Industrial, urban, and rural segments all need to be included for an adequate assessment.

5.2 San Francisco Bay

San Francisco Bay is a 100-km long estuarine system in central California. The northern third of the system, San Pablo Bay, receives inflow directly from two major rivers draining the major inland agriculture regions of the Sacramento and San Joaquin Valleys. Three rivers flow into the Bay through the intermediate Delta region and Suisun Bay. Secondary flows come from the Petaluma and Napa Rivers draining the nation's major wine growing

area and from runoff and sewage from the growing suburban population of Contra Costa County. Seaward, San Pablo bay connects with the central and deeper segment of San Francisco Bay where there is a strong bottom water inflow. To the south, the largest segment in aereal extent, south San Francisco Bay, receives comparatively little river flow, but is nearly completely surrounded by the major urban areas of (clockwise) Berkeley, Oakland, San Jose, other peninsula cities, and the city of San Francisco.

San Francisco Bay was previously cited as the estuary yielding the earliest samples (1964) of fish for DDT, dieldrin, toxaphene, and lindane analysis (Chapter 4). Evidence was also presented suggesting that PCB concentrations in fish livers may have increased since 1976 (Table 4.3) Bailey and Hannun (1967) further documented a not-unexpected 100-fold gradient of pesticide concentration in Bay water, increasing from the mouth at the Golden Gate northeast and upstream through San Pablo and Suisun Bays and into the Sacramento and San Joaquin Rivers.

The San Francisco Bay Delta complex was sampled by at least 15 surveys between 1965 and 1984. These include the NPMP, the CEMP, the Striped Bass Health Index Program (Whipple et al., 1982), the California Mussel Watch Program (Ladd et al., 1984) and 11 local surveys. A total of more than 900 samples from 15 species of fish and 10 shellfish species have been analyzed for pesticide or PCB residues.

Although many species have been sampled in San Francisco Bay, very few have been sampled repeatedly to produce a rich source of temporal trend data. The NPMP sampled Pacific oysters monthly between 1966 and 1972 at Point San Quentin and Olympia oysters at Coyote Point: the Coyote Point station was resampled using bay mussels in 1975, 1976, and 1980 by the State of California.

A mosaic of selected contaminant trends is shown in Figure 5.5. In general, PCB concentrations were highest in fish and shellfish in southern San Francisco Bay and lowest in San Pablo Bay; further, concentrations in bivalves were increasing prior to 1981 and decreasing since then (Matta, in preparation).

Total PCB levels were analyzed in six species of fish or shellfish by the NPMP and the CEMP in 1977. Concentrations were below detection limit of 0.1 ppm ww in bay mussels, Asiatic clams, ribbed horse mussels, and the flesh of striped bass and American shad. Total PCBs were below detection in all but one sample of spiny dogfish flesh (0.161 ppm ww) Inspection of data from the California Mussel Watch Program shows increasing PCB concentration through 1981 and 1982 and then declining concentrations since then at most sites (Ladd at al., 1984).

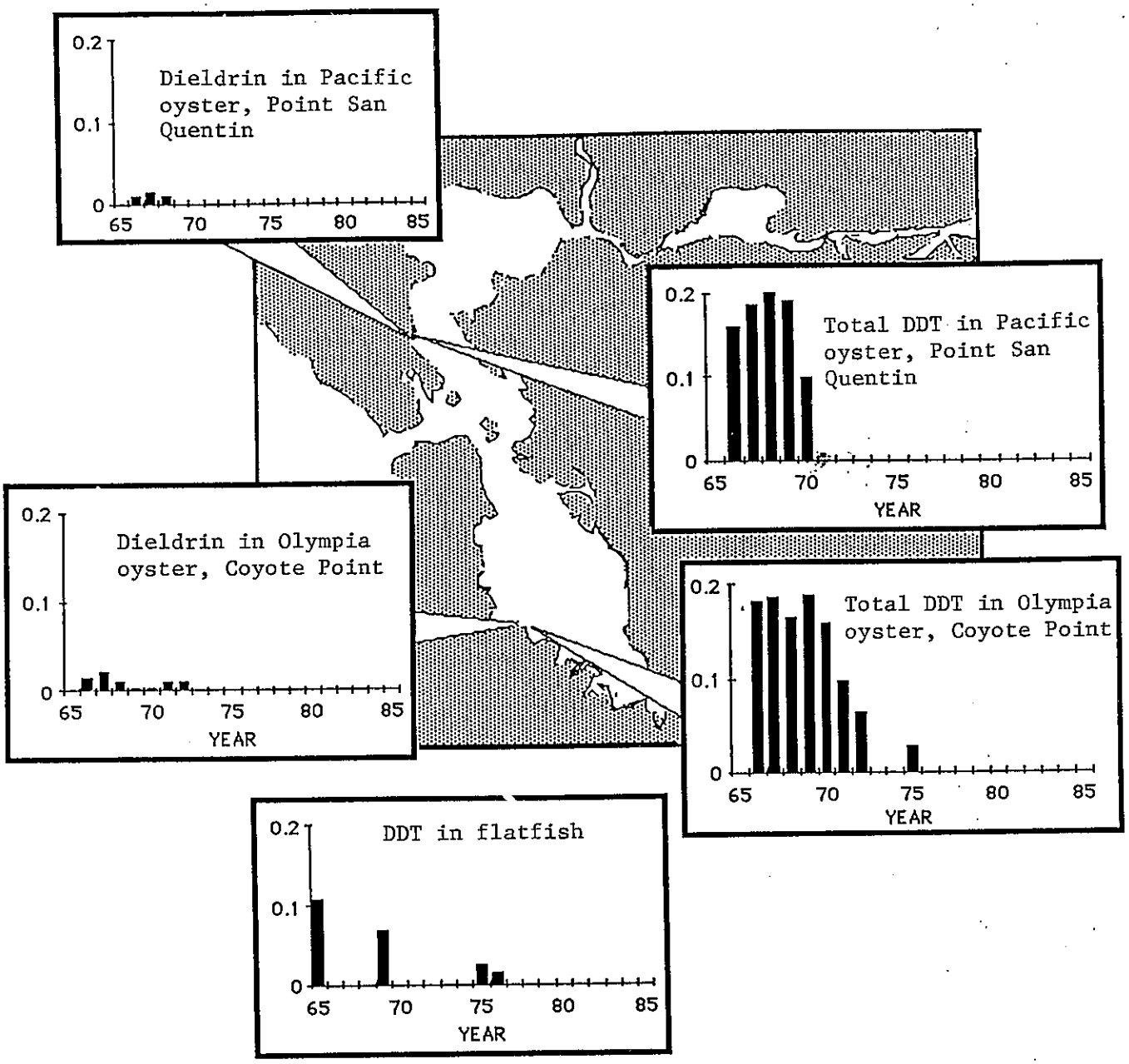


Figure 5.5. Annual variations in concentrations of dieldrin and DDT from selected historical San Francisco Bay data sets. Based on Butler, 1978; Butler et al., 1978; and other data sets.

In general, DDT concentrations in fish and shellfish have been highest in the Delta Region, along the east shore of San Pablo Bay and in south San Francisco Bay and lowest in the main body and along the north shore of San Pablo Bay (Long, et al., in preparation).

The ensemble of data, now being prepared for a special NOAA report (Long et al.) suggest that there was a strong decline in DDT contamination in fish and bivalves in all segments of the Bay between 1969 and 1977 followed by a plateau, and in one case, an increase. That increase, through the early 1980s, occurred at Albany Hill east of Richmond and north of Berkeley. Using deployed mussels, the California Mussel Watch Program has since identified a DDT hot spot near Richmond Harbor and associated the high DDT concentration in this area with high DDT concentrations in soil surrounding a nearby pesticide packaging plant (Hayes and Phillips, 1986).

Three species of flatfish have been sampled in San Francisco Bay, yielding trend information between 1965 and 1976. Levels of tDDT in English sole, starry flounder, and Pacific sanddab (muscle or whole fish) have declined since they were first sampled in 1965 (Figure 5.5).

Dieldrin levels in bivalve or fish samples were measured by the NPMP between 1966 and 1977. Mean dieldrin levels in Olympia oysters have been less than 0.025 ppm ww since 1966, and declined to less than the detection limit (0.005 ppm ww) by 1969. Muscle tissue of three species of fish (American shad, striped bass, and spiny dogfish) were analyzed in 1977 by NPMP. Levels of dieldrin in all samples were below detection (less than 0.01 ppm ww), except in one sample of spiny dogfish (0.014 ppm ww). Dieldrin levels in San Francisco Bay fish or shellfish have never exceeded 0.1 ppm ww. The maximum value (0.096 ppm ww) was detected in a sample of striped bass gonads from the San Joaquin River in 1978 (J. Whipple, original data, personal communication).

As noted previously in Chapter 4, the recent 1984 NS&T Benthic Surveillance Project data confirms the continued occurrence of DDT, PCBs, chlordane, heptachlor, and dieldrin in livers of flatfish and croakers in San Francisco Bay (Malins et al., 1986 and OAD, 1987). In particular, concentrations of PCBs were on the order of 1-3 ppm ww in these samples but since there is little comparable historical data, it is difficult to judge whether these represent an increase, decrease, or no change over previous years. A more detailed analysis is in progress (Long et al., in preparation).

5.3 Arroyo Colorado and the Lower Laguna Madre

The Arroyo Colorado and adjacent lower Laguna Madre was previously cited as an area yielding some of the most DDT- and toxaphene-contaminated fish in the United States (Chapter 4). Closer scrutiny reveals that past contamination was most likely restricted to the inland ship channel--the Arroyo Colorado--rather than the entire estuary and that many pesticides were involved in contamination of fish and shellfish.

Laguna Madre is a 200-km, shallow lagoon system of the south Texas coast. It is separated from the Gulf of Mexico by Padre Island, the longest barrier island in the United States. The southern third of the Laguna (Figure 5.6) drains croplands surrounding Harlingen and Brownsville and is

adjacent to, but separate from, the Rio Grande River Valley. The principal drainage, the Arroyo Colorado enters Laguna Madre about midway between two passes through Padre Island and nearly borders the north end of the Atacosa Wildlife Refuge. The Arroyo is maintained as a dredged ship channel and harbors a saltwater fauna 30 km inland to Harlingen.

The importance of the Laguna Madre-Arroyo Colorado system lies in the intensity of pesticide use. Dicks (1982) indicated that the four counties bordering on the Arroyo accounted for one third of the total Texas pesticide use in 1979. Further, flow in the Arroyo itself, is "sustained from irrigation return flow, urban runoff, and treated municipal sewage."

Nearly 400 samples of oysters and 14 species of estuarine fish have been sampled from the Laguna Madre-Arroyo Colorado area since 1965. Included are data for whole oysters and fish (principally Gulf menhaden) as well as separate tissues, principally liver, gonad, and flesh of spotted sea trout. Principal contaminants documented include DDD, DDT, DDE, toxaphene, dieldrin, endrin, and the pre-emergence herbicide, DCPA (or dacthal). Principal data sources included the NPMP (Butler, 1973; Butler and Schutzmann, 1978, and Butler et al., 1978); The Texas Parks and Wildlife (now Texas Water Commission; Childress, 1971 and Rogers, personal communication, 1987), the CEMP (Butler, 1978) and special surveys of DCPA (Miller and Gomes, 1974 and White et al., 1983).

Data-rich periods include 1969 and 1973-74 (Figure 5.6). Most of the samples were taken in the Arroyo Colorado and principally near the saltwater head at Harlingen and near the entrance at Arroyo City. Of 119 samples that might be attributable to collections from the Laguna proper, the exact location of 24 (from Butler, 1978) remains uncertain and another 84 were analyzed only for DCPA (Miller and Gomes, 1974). Only eight samples of fish, including six from the 1984 NOAA NS&T Benthic Surveillance Project, have definitely been collected from the main body of Laguna Madre and analyzed for more than one chlorinated hydrocarbon. Thus the principal data is from the Arroyo.

At least eight organochlorine pesticides--DDT, DDE, DDD, toxaphene, dieldrin, chlordane, endrin, and DCPA (dacthal)--have been repeatedly measured in tissues of fish from the Arroyo by the NPMP; in addition, unpublished data (P. Butler, personal communication, 1985) indicates some fish contained measurable concentrations of five non-organochlorine pesticides including ethion, trithion, ethyl parathion, methyl parathion, and DEF. In short, Arroyo fish populations have experienced a surprising variety of contaminants.

It is not yet clear whether PCBs have been a significant contaminant in estuarine fish of this region. Interference by the extremely high concentrations of DDT and toxaphene in the past have apparently precluded the possibility of documenting concentrations in the range of urban values (e.g., 0.2 ppm ww). Livers of spotted sea trout from 1977 collections in the Laguna Madre (site unspecified) contained 0.7 ppm ww PCBs but other species from the same collections did not have concentrations exceeding the rather high 0.2 ppm ww detection limit used by the CEMP.

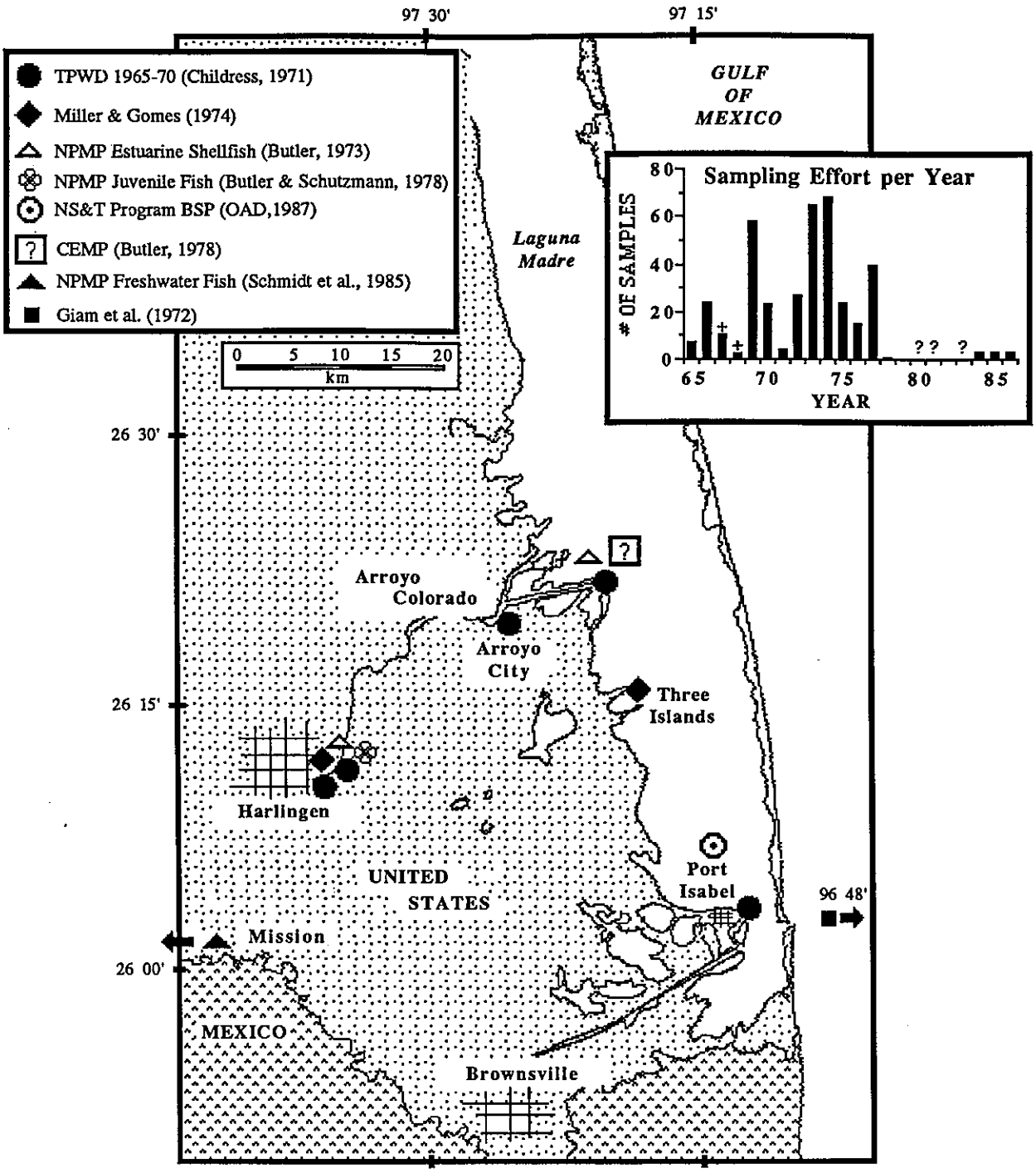


Figure 5.6. Overview of pesticide and PCB monitoring activity in the Arroyo Colorado and Laguna Madre area, Texas, 1965-86.

The possibility of substantial pesticide contamination gradient down the Arroyo is supported by DDT measurements made principally during 1967-71. During 1967, tDDT was at least an order of magnitude higher in oysters from Harlingen (0.257 ppm ww) than in those from a site at the entrance (0.024 ppm ww). In 1969, whole fish (species undescribed) presented nearly a 20-fold gradient from 2.09 to 0.120 ppm ww between these two points. During the same year, there was a four-fold gradient for DDT in whole Gulf menhaden along the upper two-thirds of the Arroyo between Harlingen (1.25 ppm ww) and Arroyo City (0.497 ppm ww) (Figure 5.7a). The gradient may have continued into and down the Laguna Madre as suggested by a 10-fold difference in egg DDT concentrations between Arroyo City (2.32 ppm ww) and Port Isabel (0.22 ppm ww) and a 72-fold difference in liver DDT between the same localities (3.96 ppm ww vs. 0.055 ppm ww). Another major drop in concentration was evident from a comparison of all whole fish data with a value of 0.027 ppm ww reported for a flatfish collected 40 km offshore in 1971 by Giam et al. (1972). Finally, of all liver DDT data, the lowest mean concentration was 0.014 ppm ww from six composites collected in Laguna Madre in 1984 by the NOAA Benthic Surveillance Project (Hanson et al., 1986).

Both temporal as well as spatial trends can be reconstructed for DDT and three other insecticides: toxaphene, dieldrin, and endrin. As shown in Figure 5.7a, DDT in oysters doubled from 0.14 ppm ww in 1965 to 0.29 ppm ww in 1971 whereas DDT in whole fish generally declined from a range of 2.0 to 5.0 ppm ww tDDT at Harlingen between 1969 and 1977 but appeared to have increased from 0.3 to about 1.0 ppm ww at Arroyo City. By comparison, tDDT in gizzard shad upstream in the Rio Grande River decreased from above 10.0 ppm ww in the late 1960s to values approaching 1.0 ppm ww by 1980-81 (from Schmitt et al., 1983 and 1985). Collectively, these data suggest a region-wide rise and then slow fall of DDT contamination between 1965 and 1980.

Toxaphene occurred in higher concentrations in whole fish from Harlingen (maximum mean over 4.0 ppm ww) than in those from Arroyo City (maximum mean about 2.5 ppm ww) and also appeared to reach peak concentrations in the Arroyo about 1973 (Figure 5.7b). In contrast, toxaphene originally occurred in low concentrations in gizzard shad from the adjacent Rio Grande River and were increasing through 1980-81 (Schmitt et al., 1985). Contamination continued through 1979 resulting in a seafood advisory (White et al., 1983).

Dieldrin and endrin were reported sporatically in whole Arroyo fish. Mean concentrations over the the period 1969 through 1976 were low and similar at both the Harlingen and Arroyo City sites, but maximum concentrations were an order of magnitude higher at Harlingen (Figure 5.8a and b). Dieldrin was reported more frequently yielding higher mean concentrations at Harlingen than at Arroyo City (Figure 5.8a). Dieldrin was also higher in Gulf menhaden from Harlingen than from gizzard shad at Mission on the Rio Grande.

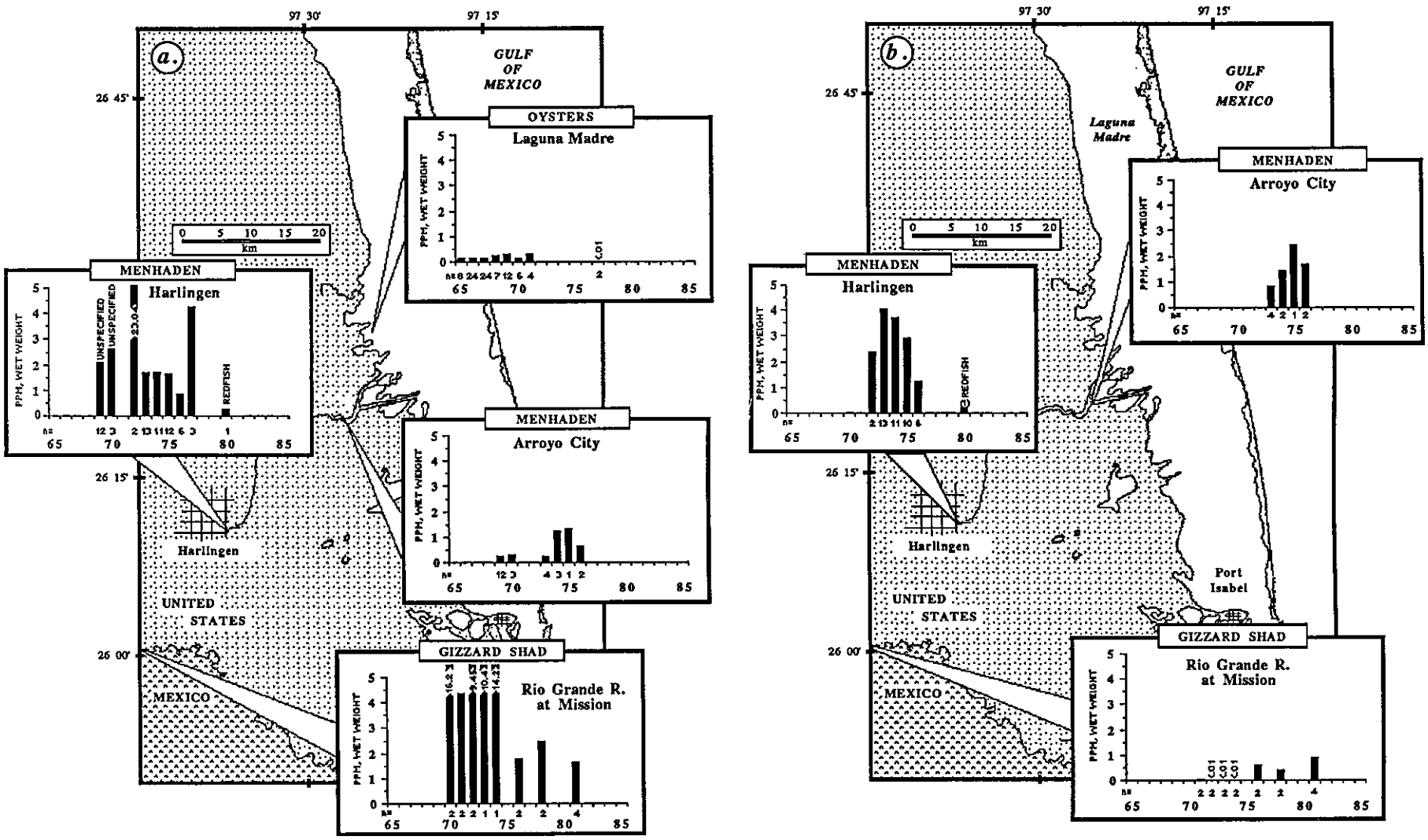


Figure 5.7. Annual variation of pesticides in oysters, whole menhaden, and gizzard shad from several sites in the Arroyo Colorado and Rio Grande systems of Texas. (a) DDT, 1965-81; (b) Toxaphene, 1971-81.

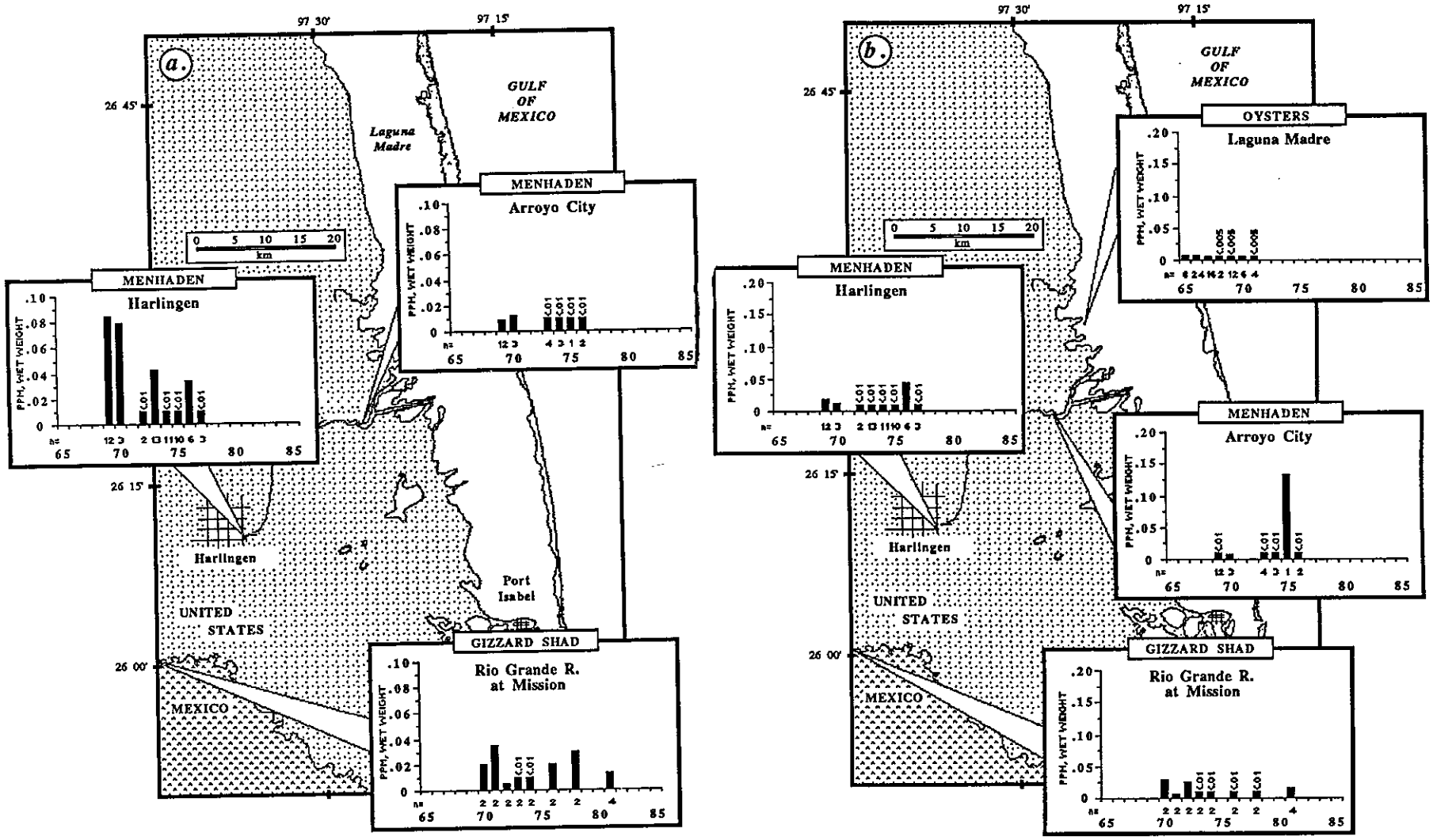


Figure 5.8. Annual variation of pesticide in oysters, whole menhaden, and gizzard shad from several sites in the Arroyo Colorado and Rio Grande River systems, Texas; (a) dieldrin, 1967-81; (b) endrin, 1965-81.

During Arroyo Colorado fish surveys in 1971, Miller and Gomes (1974) identified a chromatographic peak, previously thought to be heptachlor, as DCPA, a pre-emergent herbicide. Subsequent sampling of fish from Laguna Madre and the Arroyo indicated that DCPA was "a frequent, although intermittent, chlorinated hydrocarbon contaminant of the aquatic and estuarine environments of the lower Rio Grande Valley." Occurrence was patchy in livers of fish of the Arroyo, but all three liver samples from the Laguna Madre (Three Islands) yielded a mean DCPA concentration of 0.422 ppm ww (Figure 5.9). A plot of maximum values and frequencies suggests a declining occurrence since 1972 in the Arroyo-Laguna area in general, but this is far from a firm conclusion (Figure 5.10).

In summary, estuarine fish and oysters inhabiting the confined waters of the Arroyo Colorado have been heavily contaminated with toxaphene and DDT in the past, but it is not certain that high levels also occurred in biota of the Laguna Madre proper. There is evidence of a gradient. There is also evidence of extreme persistence following presumed termination of use or production of these chemicals in this region, at least in the Arroyo. Renewed sampling in the Arroyo and new sampling in the Laguna would help confirm the apparent declining concentrations of toxaphene and DDT, serve as a check on other heavily used pesticides, and reduce the uncertainty that the Laguna and adjacent Atacosa Wildlife Refuge is adequately protected.

5.4 Summary and Conclusion

Several lessons emerge from this brief analysis of three bays. First, there is a large amount of historical data. Indeed, there appears to be more historical than recent data to the extent that previously detected trends have not been reconfirmed in recent years because previously monitored species, tissues and sites have not been resampled. The exception appears to be the Chesapeake which recent investigators, by choice or chance, have continued monitoring historically sampled species at or near previously sampled sites. Elsewhere, there remains the challenge of resampling previously tested species and reoccupying old monitoring sites.

Second, it seems clear that one or a few sites in bays as large as the Chesapeake or San Francisco Bays are inadequate to characterize the bay. There is a good chance that data from a few sites can severely bias conclusions about the bay as a whole. Each bay has "hot spots" and relatively uncontaminated regions so that conclusions that one bay is more contaminated than another are unjustified unless demonstrated by a geographically synoptic survey.

Third, this review underscores the value of local monitoring and surveillance programs and helps place them in a national perspective. Past national programs, such as NPMP, provided a framework for comparison and helped identify areas where there might be emerging or declining pollution hot spots. If continued, programs such as NPMP or NS&T Program also provide a long-term reference point for judging long-term trends. However, they do not necessarily provide the detail required to identify problematic sources. In the cases cited here, and in many others, local monitoring provided the necessary detail.

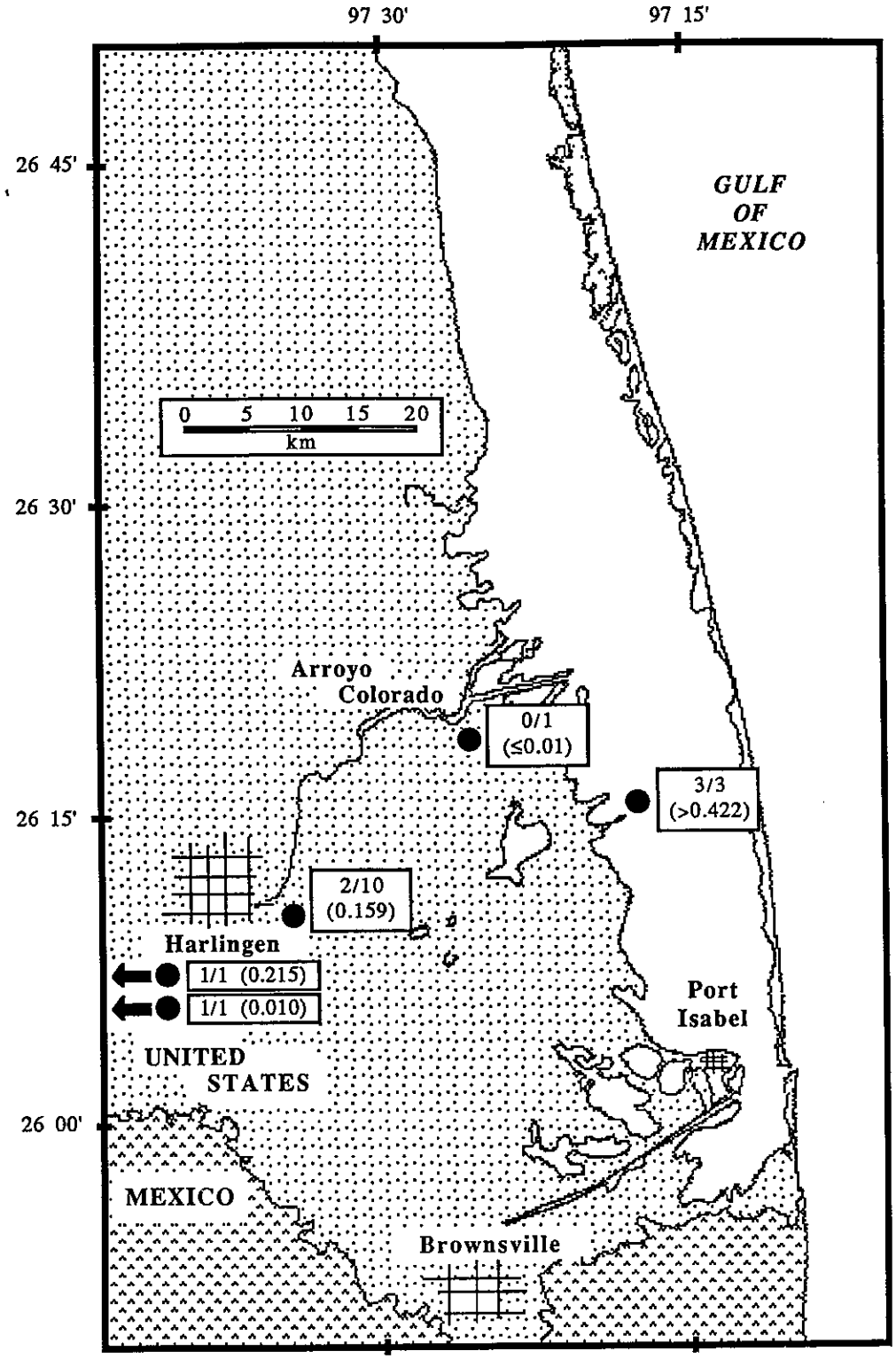


Figure 5.9. Frequency of occurrence (fraction) and maximum concentrations (ppm ww) of the herbicide DCPA (dacthal) in marine and freshwater fish of the Arroyo Colorado system, Texas, 1973. From data in Miller and Gomes (1974) and original data supporting Butler (1978).

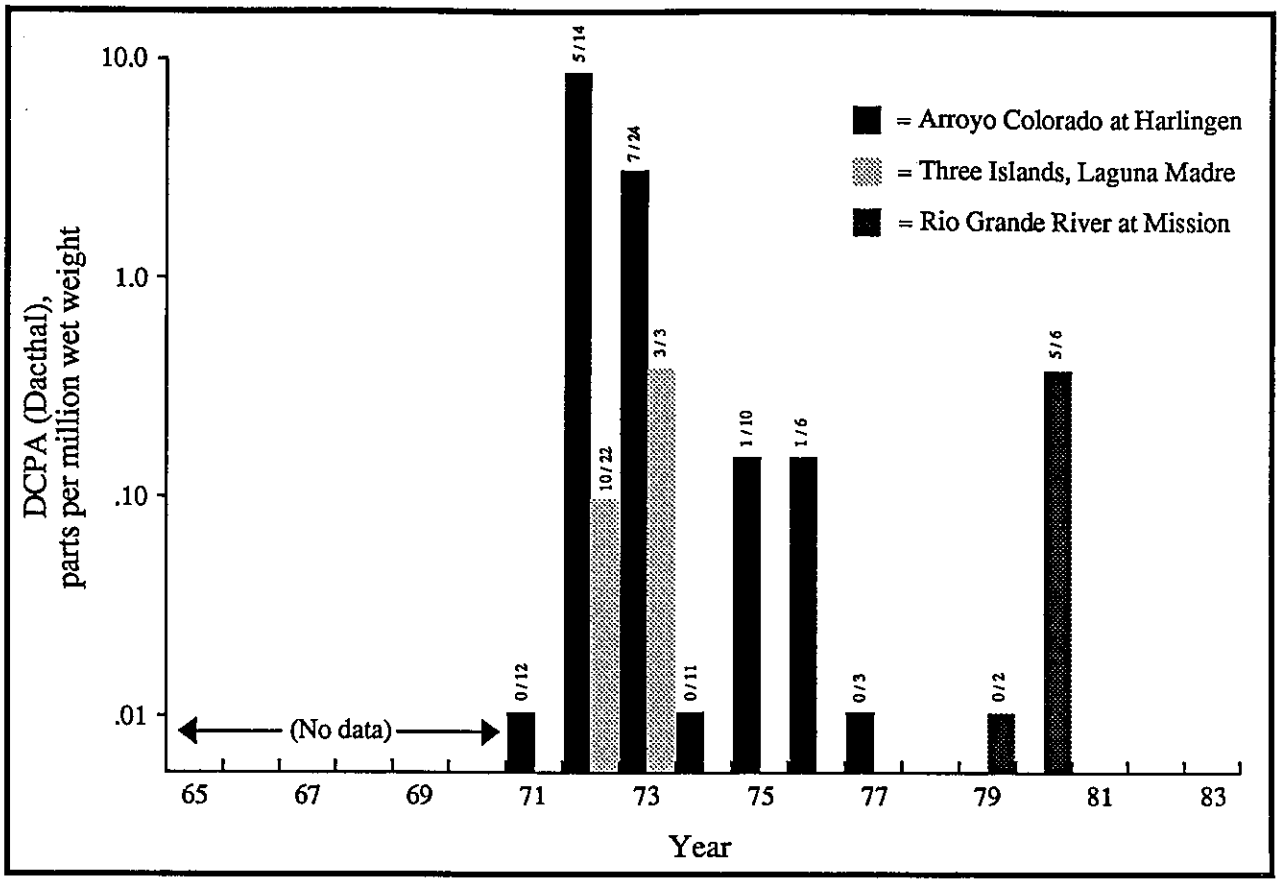


Figure 5.10. Trends in maximum concentrations and frequencies of occurrence (fraction) of DCPA (dacthal) in fishes of the Arroyo Colorado, Rio Grande River, and southern Laguna Madre, Texas, 1971-80. Based on applicable data in references cited in Figure 5.6. Note log scale.

Finally, there were additional signs in these examples of successful connections between nationwide surveys and local monitoring. For example, it appears that the State of Virginia (VIMS) continued monitoring NPMP sites long after that program concluded, thus providing additional detail and confirmation that would have been missing or otherwise extremely difficult to reconstruct from unlinked monitoring networks.

6.0 PCBs IN FLATFISH AND BIVALVES

This chapter explores the extent to which local, state, and other monitoring data may help amplify or explain trends derived from national survey efforts. The chapter focuses exclusively on one class of chemicals--PCBs--and on two groups of organisms--bivalve mollusks and flatfish--frequently used in national pollutant monitoring surveys.

Both geographic and temporal trends are explored with emphasis on the ability of aggregate local monitoring data to help resolve whether PCB contamination has been increasing, decreasing, or remaining unchanged on a national basis. The results of this analysis support the conclusion stated in Section 4.2 that dramatic declines of PCB contamination have occurred in several localities and regions, that it is difficult to judge trends on a national level, and that there remains an abundance of local and regional data that can be further explored for trends in more areas than covered in this limited review.

6.1 Trends in Bivalves

Bivalves (oysters, mussels, and clams) support large recreational and commercial fisheries in many areas. They have been popular choices for monitoring since 1965, mainly due to their sessile state and ability to filter and concentrate contaminants from water. Table 6.1 shows species and data sources combined to produce the nationwide trends reviewed below.

6.1.1 National Overview

The NPMP screened only a few samples for PCBs between 1971 and 1972 (Butler, 1973). PCBs were detected in Chesapeake Bay oysters in 1971 and 1972 with a maximum concentration of about 2.8 ppm ww found in the Elizabeth River. This may be the highest concentration reported in a bivalve. NPMP also analyzed PCBs in oyster samples from Escambia Bay, near Pensacola, Florida, in 1970, 1971, and 1972. The mean level of total PCBs in this bay was 0.165 ppm ww.

During the 1977 NEMP national resurvey, (Butler and Schutzmann, 1978)PCB concentrations had so decreased that the detection limit (0.05 ppm ww) was no longer appropriate.

The EPA Mussel Watch was the first program to synoptically document PCBs in bivalves at low detection limits on a national basis (Goldberg et al., 1978; Figure 6.1). The median concentration of Aroclor 1254 for 83 sites was 0.009 ppm ww. Highest levels (greater than 1.0 ppm ww) of PCBs in individual samples of bivalves in 1976 were in mussels from San Pedro Harbor, California. Despite this, levels of total PCBs in bivalves from West Coast sites were more often below 0.01 ww than those from Northeastern sites, where levels often exceeded 0.02 ww (Figure 6.1). The highest concentration of PCB 1254 along the Atlantic Coast was about 0.108 ppm ww in mussels from a 1976 collection at Manhasset Neck on Long Island Sound, New York. Comparably high concentrations were also reported from Boston Harbor (0.09 ppm ww), Narragansett Bay (0.03 ppm ww), and Rockaway (0.07 ppm ww). Only three sites were

sampled in 1976 for PCBs in the Gulf of Mexico, with oyster concentration ranging from 0.01 ppm ww at Pensacola, Florida, to 0.037 ppm ww at Biloxi, Mississippi. Similar patterns were found in 1977 (Farrington et al., 1981).

Table 6.1. Common names of bivalve species and data sources for trends in PCB residues.

COMMON NAME	DATA SOURCE
American oyster	1, 2, 3, 6, 7, 8, 9
Asiatic clam	1, 2, 4
Bay mussel	1, 2, 3, 4, 8
Coastal mussel	1, 3, 4, 5
Hard clam (Northern quaheq	1, 2, 8, 9
Northern horse mussel	1
Olympia oyster	1
Pacific oyster	1,2
Ribbed mussel	1,2
Soft-shell clam	1, 2, 9

1. Butler, 1973
2. Butler et al., 1978
3. Farrington et al., 1982
4. Hayes and Phillips, 1987 (and previous California State Mussel Watch reports)
5. Young et al. (in press)
6. Wilson and Forester, 1978
7. Eisenberg and Topping, 1981 (and subsequent Maryland State monitoring reports)
8. Hatch et al., 1981
9. Kolek and Cuervals, 1981

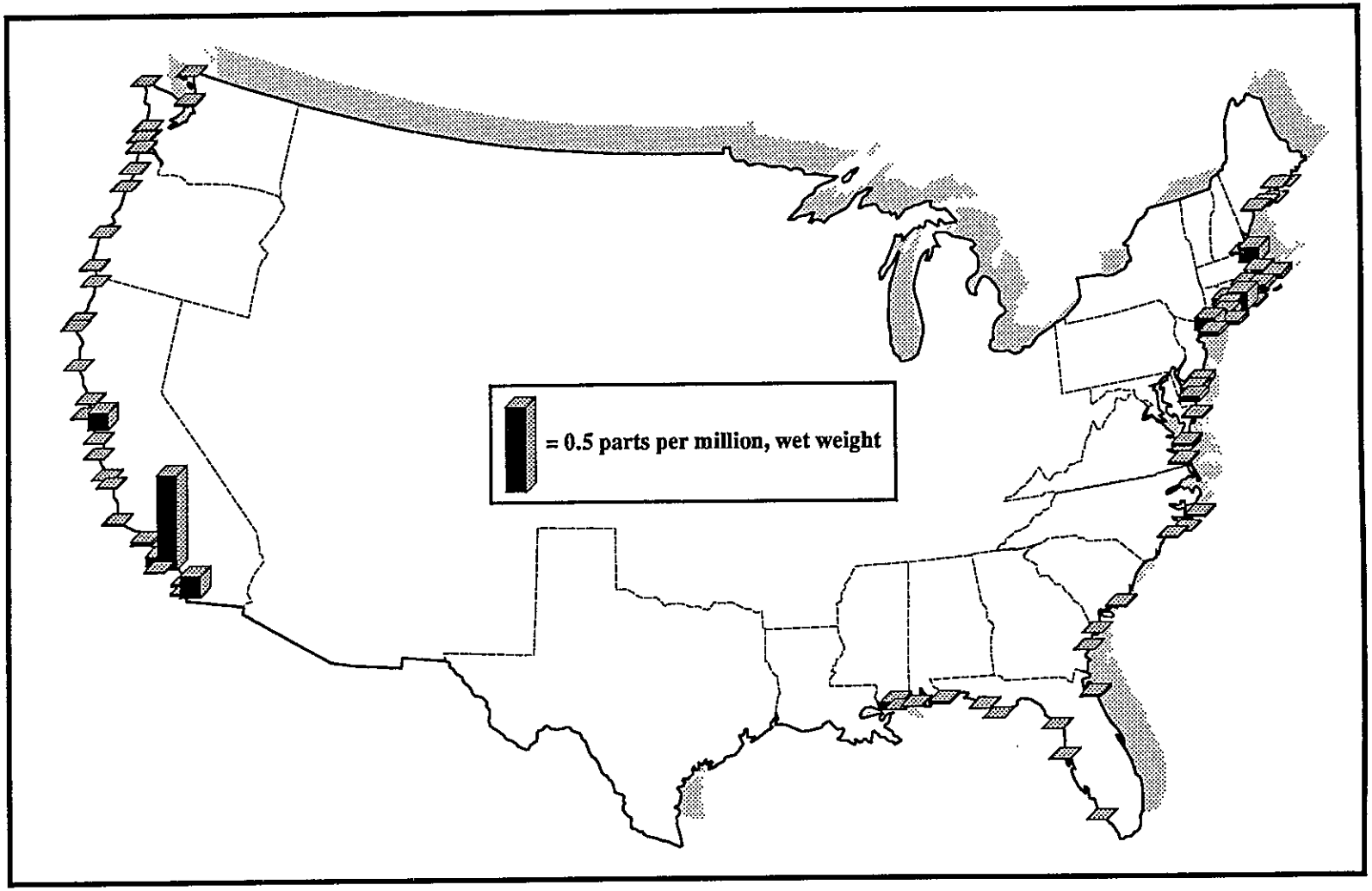


Figure 6.1. PCB (Aroclor 1254) concentrations in whole bivalves sampled at 71 U.S. sites in 1976. U.S. EPA Mussel Watch data from Farrington et al. (1982). Moisture content used to convert from given dry weight values from Goldberg et al. (1978) and in original data supporting Farrington et al. (1982).

6.1.2 Temporal Trends

Because of analytical and other sampling inconsistencies, it is difficult to judge from national bivalve monitoring programs if PCB concentrations have been increasing or decreasing on a national basis. The 1976 EPA Mussel Watch focused on determination of Aroclor 1254 and (noted previously in Section 4.1.1) resulted in a national grand median of 0.009 ppm ww for the 71 sites surveyed. The 1986 NOAA NS&T Mussel Watch Project determined PCB concentrations by chlorination number for bivalves from 144 sites. Preliminary reduction of these data yields a national grand median tPCB concentration of 0.015 ppm ww (range 0.009 to 0.680). While these medians agree within a factor of 2, they cannot be compared without making some correction or adjustment to one or both data sets.

While it is difficult to discern from national programs whether PCBs have been increasing or decreasing over the past decade on a national basis, it is clear concentrations have decreased in bivalves from specific localities subjected to local long-term monitoring, and that seasonal variation can be significant.

On the Pacific Coast, the longest continuous bivalve monitoring for PCBs is at Royal Palms State Park at Whites Point on the Palos Verdes Peninsula. The site is at the base of the Whites Point outfalls through which the County Sanitation Districts has discharged over 350 million gallons per day of treated sewage for several decades. Coastal (California) mussels at this site were monitored two to four times per year for chlorinated pesticides and PCBs between summer of 1971 and fall of 1982 by the SCCWRP. During this period, annual average tPCBs declined at least 20-fold from about 0.4 ppm ww in 1971 to less than 0.02 ppm ww in 1982 (Figure 6.2) commensurate with declines of sewage-born PCBs from source control (as in SCCWRP, 1986). Declines also were recorded for Oceanside during the SCCWRP 1971 and 1974 surveys and California State Mussel Watch (CSMW) surveys, 1976 through 1986 (Hayes and Phillips, 1987; Figure 6.2). Although beginning at very low concentrations, the CSMW also recorded declining but fluctuating concentrations at two other long-term monitoring sites in California, Trinidad Head and Bodega Head (Figure 6.2).

In Florida, Wilson and Forester (1968) conducted monthly PCB monitoring for nearly 8 years (1969-1976) at a reference site in East Bay, and at a nearby site in Escambia Bay experiencing contamination from a known PCB discharge. Average annual Aroclor 1254 concentrations in oysters dropped about 6-fold from about 12 ppm ww in 1969 to about 2 ppm ww in the first half of 1976 (Figure 6.2).

On the Atlantic Coast, several data sets can be merged to document long-term trends of PCB contamination in bivalves. As previously noted (Section 5.1), PCBs were measured in Chesapeake Bay oysters in 1971 and 1972 by the NPMP and 1976 to 1986 in various bay segments by the states of Maryland and Virginia. Concentrations declined several-fold to over 10-fold during this period, depending on the segment. In the mainstem of the upper Chesapeake Bay, values were simply low and variable (Figure 6.2). In addition, the EPA Mussel Watch program team returned to several Atlantic sites--Boston Harbor, Narragansett Bay, Rhode Island, and Beaufort, North Carolina--more than three

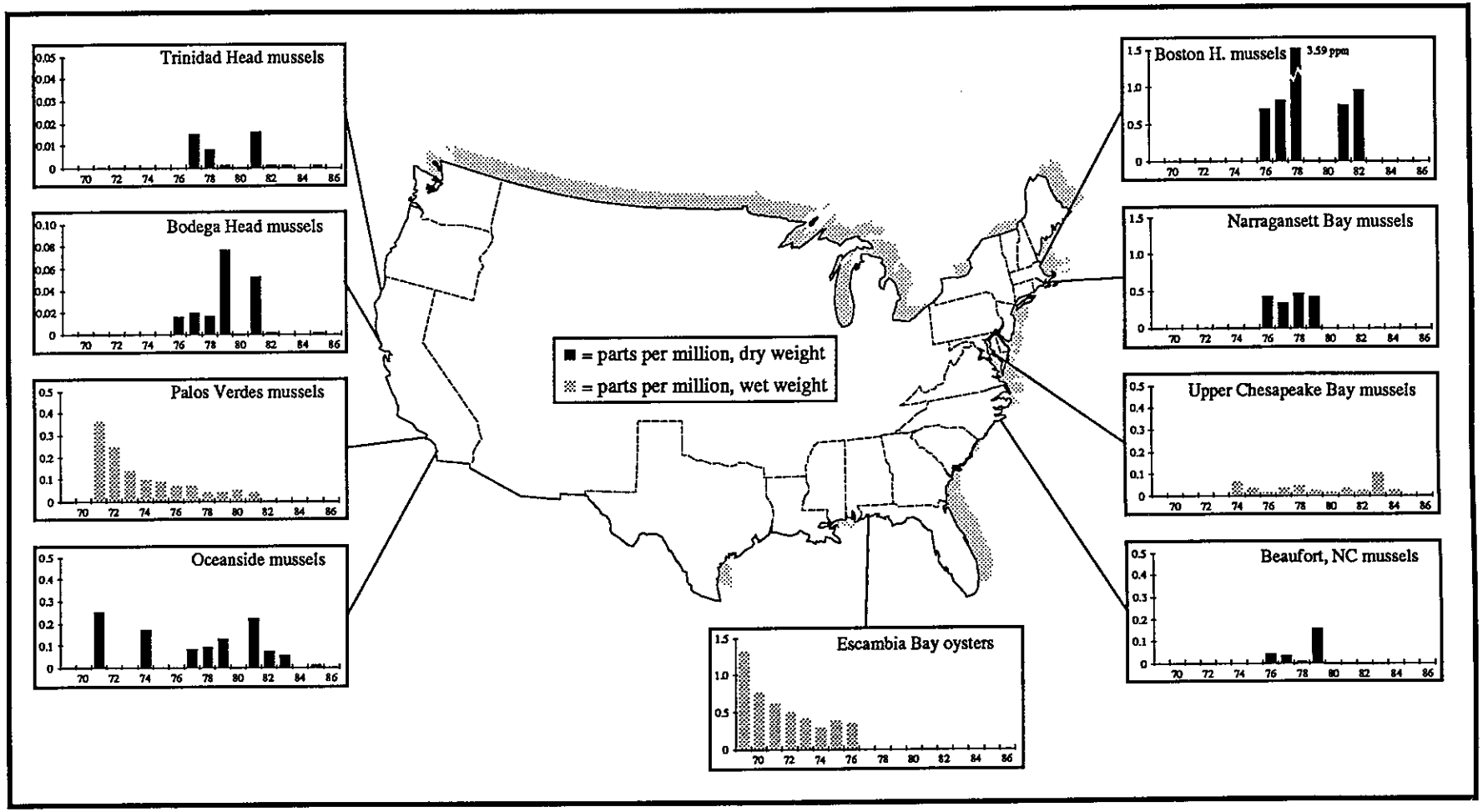


Figure 6.2. Annual mean PCB concentrations in mussels and oysters at nine sites sampled annually or occasionally since 1969. Data from sources cited in text. Note differences in scale and reporting units.

times between 1976 and 1982 (Farrington et al., 1982) In Boston Harbor, concentrations of Aroclor 1242 in Bay mussels appeared to peak in 1978 and return to the range of 1976-77 concentrations in 1981-82 (shown as 0.7 to .9 ppm dw, or about 0.07 to 0.09 ppm ww, in Figure 6.2, upper right). Concentrations remained in the range of 0.5 to 1.3 ppm dw in the 1986 NOAA Mussel Watch survey (OAD, 1987). In Narragansett Bay, Aroclor 1254 concentrations were fluctuating in the range of 0.4 to 0.5 ppm dw between 1976 and 1979 (Farraington et al., 1982; Figure 6.2). In the 1986 NOAA Mussel Watch, concentrations of tPCBs were about 0.2 ppm dw, suggesting the possibility of a decline since the late 1970s. The 1981 concentrations in Beaufort, North Carolina, mussels was higher in 1981 (about 0.15 ppm dw) than in 1976-77 (0.05 ppm dw; Figure 6.2). The recent (1986) NOAA Mussel Watch recorded concentrations in the range of 0.02 to 0.103 ppm dw at two other North Carolina sites (NOAA, 1987).

In addition to these long-term trends, several of the data sets provide special information about short-term, high-frequency variations of PCB content in bivalves. For example, the monthly sampling in East Bay and Escambia Bay, Florida, by Wilson and Forrester (1978) clearly showed a 3- to 5-fold seasonal variation superimposed over the long-term trend. Other monthly and quarterly local bivalve monitoring data sets exist that also reveal seasonal variations in this range suggesting a natural source of variation that may need to be included to statistically evaluate the significance of apparent long-term trends.

In summary, the use of bivalves to monitor PCBs began at scattered localities between 1969 and 1972, but full nationwide sampling was not implemented until 1976 (EPA Mussel Watch). During this period, it appears that a few sites were identified with very high concentrations (Elizabeth River, Virginia; Pensacola, Florida; and Palos Verdes, California) all of which had substantially decreased PCB concentrations by the onset of the EPA Mussel Watch in 1976. That survey, nevertheless, documented relatively high concentrations in various additional urban embayments of the Northeast and Southwest. Local surveys continued to document declining or at least generally, but seasonally variable, unchanging concentrations through the early 1980s. Preliminary comparisons of 1976 EPA data with the 1986 NOAA NS&T Mussel Watch program, suggest no dramatic changes in PCB concentrations have occurred on a national basis despite obvious decreases at former "hot spots".

6.2 Trends in flatfish

Flatfish, (flounder, sole, and dabs) live and feed on the bottom, allowing contact with the sediment reservoir of contaminants, thus making them popular choices for the monitoring of local contaminant levels. Flatfish are presumed not to migrate far, which would make them good indicators of local contamination.

6.2.1 Nationwide Trends

Prior to the NOAA 1984 Benthic Surveillance Project, there was no single national survey that synoptically surveyed PCBs in flatfish from many U.S. coastal areas. However, our data search revealed that during 1980, flatfish were independently sampled at many Atlantic, Gulf, and Pacific estuarine, coastal, and offshore sites. We pooled these data to get a better idea of large-scale coastal and offshore trends in this one time period.

PCB concentrations in muscle or flesh were compared using data for several species of flatfish sampled during the one year, 1980, by several investigators. Levels of total PCBs in coastal flatfish from onshore sites of the Gulf of Mexico waters (less than 0.1 ppm ww) were lower than those from Atlantic or Pacific coastal areas sampled (greater than 0.1). The highest concentrations of PCBs (greater than 2.0 ppm ww) occurred in fish from New Bedford Harbor, Massachusetts (Figure 6.3). Flatfish muscle from the New York Bight contained a mean level of 0.18 ppm ww. This is one-twentieth of the PCB levels found in striped bass from the same area (see Chapter 7). Of five Pacific Coast sites sampled in 1980, Elliott Bay and the Palos Verdes Peninsula had the highest levels of total PCBs in flatfish muscle (Figure 6.3). Offshore fishes in both the northeast and in the Gulf of Mexico were lower yet, below 0.05, and many below 0.005, ppm ww, (Figure 6.3; ERCO 1981). It is thus apparent that flatfish across the Northeast coastal shelf experienced as much as a 500-fold gradient in PCBs from offshore to some estuarine sites, such as New Bedford Harbor.

In summary, the national estuarine and nearshore patterns of PCB contamination in flatfish identify contaminated areas similar to those from previous or current bivalve and other estuarine fish surveys. In addition, however, due largely to the East and Gulf coastal shelf surveys by ERCO (1980 and 1981), it was clear that PCB contamination in 1980 of flatfish decreased dramatically with distance from shore.

6.2.2 Temporal Trends

As reviewed in Chapter 4, lack of sampling and analytical continuity, and an apparent lack of great change, makes it impossible to discern the direction of long-term trends in PCB contamination in fish on a national basis, at least from national programs. This clearly applies to flatfish. However, local monitoring programs have been conducted for many years in several locations and these do provide some data for establishing trends in flatfish contamination at these specific sites.

On the Pacific Coast, long-term trends can be developed for PCBs in muscle of flatfish from Elliott Bay, in Puget Sound and for at least five sites in the Southern California Bight. In Elliott Bay, concentrations of tPCBs in flatfish muscle sampled between 1977 and 1983 varied about 0.1 and 0.6 ppm ww, with the highest mean concentration occurring in 1980 and the lowest in 1983 (Figure 6.4). In Southern California, concentrations in flatfish muscle were lower in the early 1980s than in the mid-1970s in Santa Monica Bay, San Pedro Bay, at Catalina Island, the Palos Verdes Peninsula, and off Orange County (Figure 6.4).

Table 6.2. Flatfish species and data sources used for assessment of trends in PCBs.

Common Name	Data Source
American plaice	7
Bigmouth sole	3
Broad flounder	7
C-O turbot	4
California halibut	2, 3
Dover sole	1, 3, 4, 9
English sole	3, 4, 10
Fourspot flounder	5, 7
Gulf flounder	7
Pacific halibut	4
Pacific sanddab	3, 4, 8
Rex sole	4
Shoal flounder	7
Slender sole	4
Southern flounder	8
Speckled sanddab	3
Starry flounder	9
Summer flounder	5, 7, 8
Windowpane flounder	4, 6, 7, 8
Winter flounder	5, 6, 7, 8, 9
Yellowtail flounder	7

1. Young et al., in press
2. SCCWRP, unpublished data in support of MBC and SCCWRP, 1980.
3. County Sanitation Districts of Orange County (CSDOC), California, unpublished data.
4. County Sanitation Districts of Los Angeles County (CSDLA), California, Unpublished data.
5. Kolek and Cuervals, 1982.
6. Northeast Monitoring Program, NOAA, 1982.
7. ERCO 1980 and 1981.
8. Gadbois and Maney, 1982
9. Sherwood, 1982 (supporting data).
10. Malins et al., 1980b.

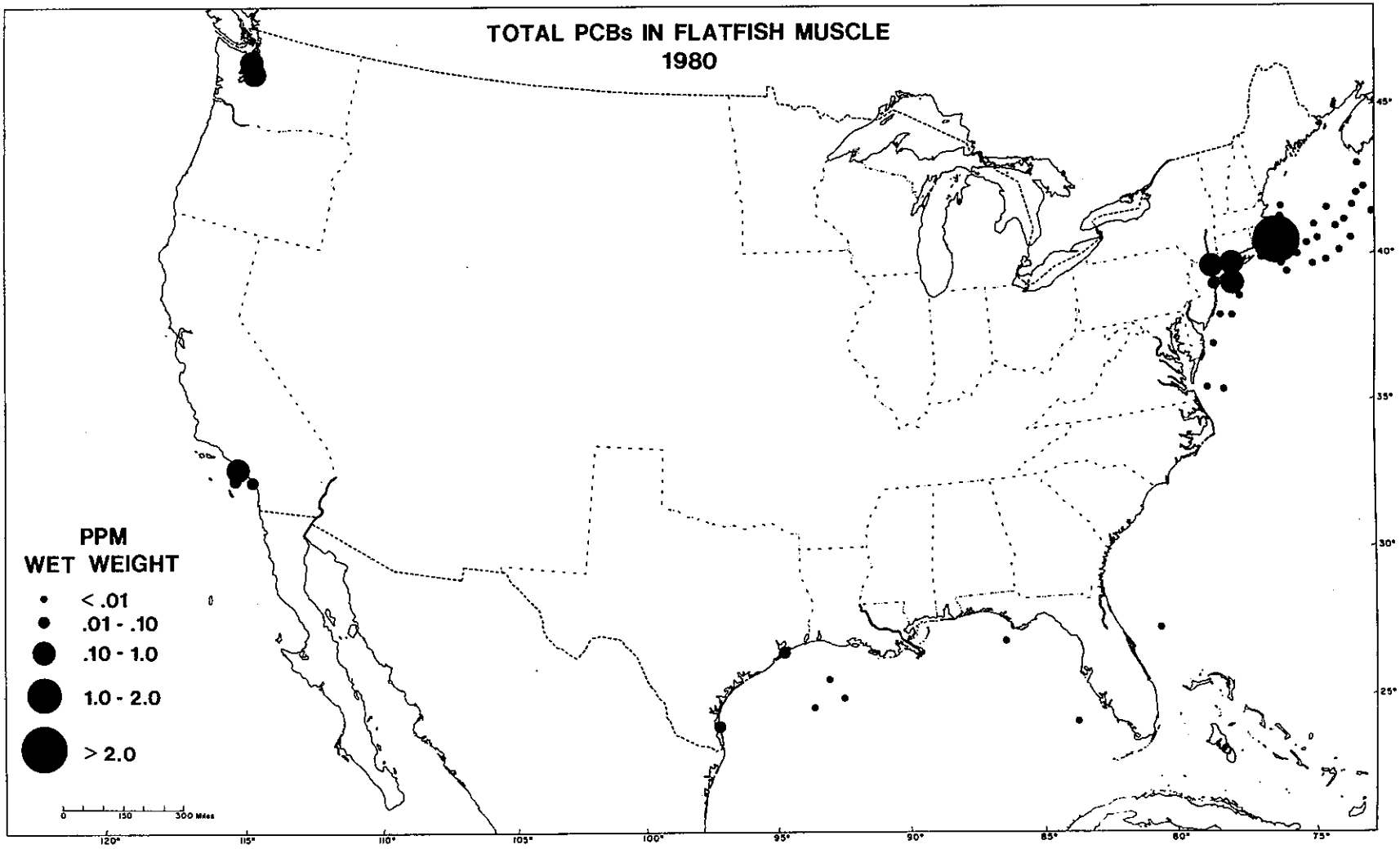


Figure 6.3. Concentration of tPCBs in muscle of flatfish from 43 sites or regions sampled during 1980.

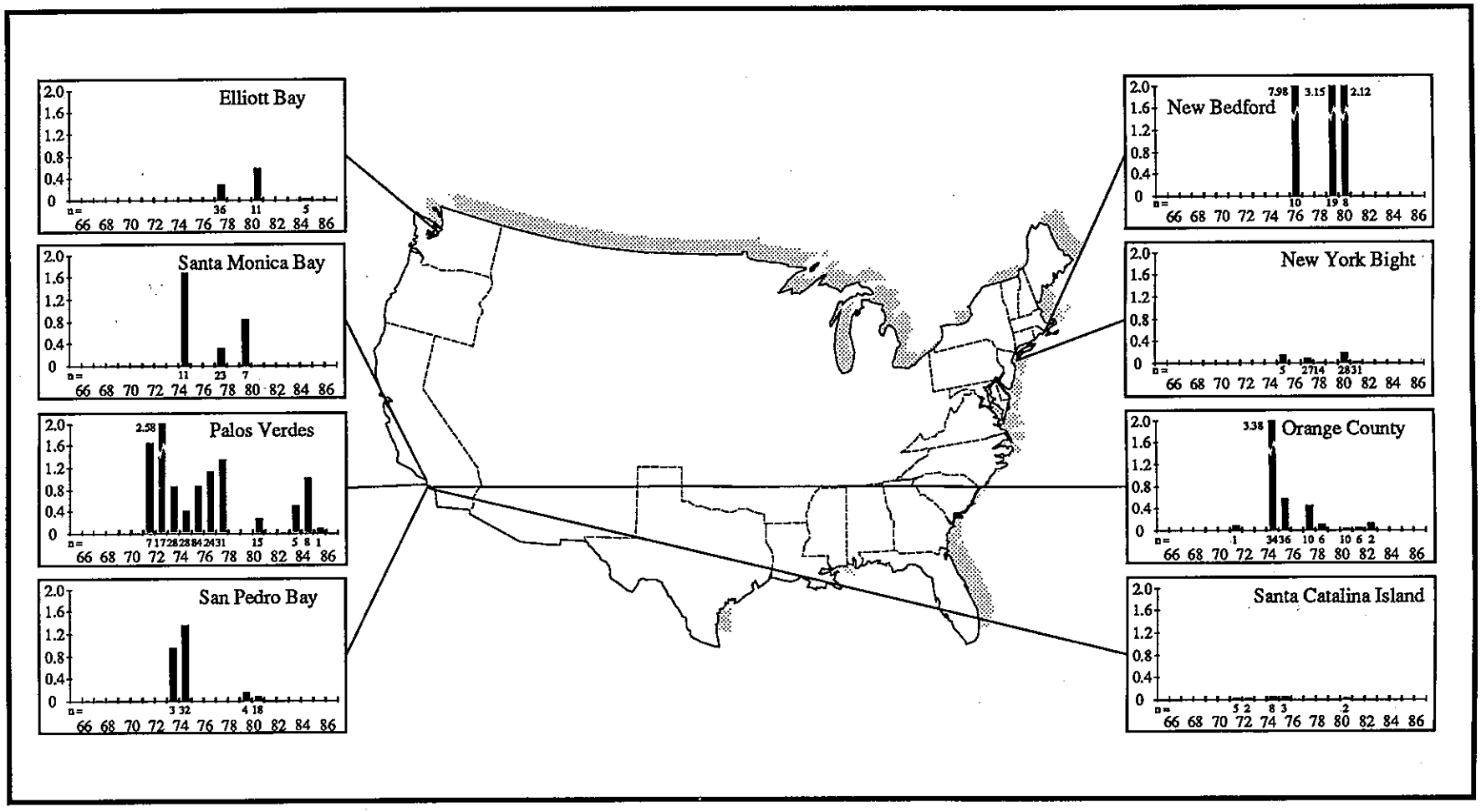


Figure 6.4. Annual mean PCB concentrations in flatfish flesh or muscle at eight sites sampled annually or occasionally since 1971. Data from sources cited in text.

At Palos Verdes, one known epicenter of now-controlled PCB inputs, there was considerable interannual variation in the means, with an apparent increase from about 1.0 to several ppm ww 1973 to 1977, a sharp decline to 0.2 ppm ww in 1980, a rise to 1.0 in 1984, and again a decline to below 0.1. Some of this apparent interannual variability may be due to year-to-year differences in the mix of species used in the analysis. Nevertheless, the overall trend in the last decade is downward by at least an order of magnitude; for one species, Dover sole, interannual variation is less, and the long-term decline clearer (D.R. Young, U.S. EPA, Newport, Oregon, personal communication).

Less frequent sampling in Santa Monica Bay suggested a decrease in flatfish contamination (Figure 6.4, from about 2.0 in 1974 to 0.9 ppm ww in 1979, but the data base is clearly inadequate. Off the Orange County outfall area, the PCB decline was dramatic, from 1.4 ppm ww in 1974 to about 0.1 ppm ww in 1982. The general decline follows decreasing inputs from local sewage due to source control initiated during the mid 1980s (see SCCWRP, 1986). Concentrations in San Pedro Bay flatfish, between Palos Verdes and Orange County also decreased dramatically from 3.8 ppm ww in 1974 to 0.2 ppm ww in 1980 (Figure 6.4). Finally, while concentrations in muscle of flatfish from Catalina Island were apparently always low (below 0.05 ppm ww, Figure 6.4) they too declined between 1974 and 1980.

There appear to be no long-term data for flatfish along the Gulf of Mexico Coast comparable to that for Southern California. Indeed, there appear to be no comparable long-term data for other fish species or other fish tissues at Gulf of Mexico sites with the notable exception of Gulf menhaden monitored by Stout (1981); these data show low levels relative to the rest of the United States and declining PCB concentrations during the 1970s as reviewed in Chapter 7.

Two Northeast Coast areas yield data for inferring trends of PCBs in flatfish muscle. Repeated sampling of flatfish in New Bedford Harbor between 1976 and 1980 suggest substantial (4-fold) declines in flatfish PCB concentrations from about 8.0 ppm ww in 1976 to 2.1 ppm ww in 1980; additional recent data from the Massachusetts Division of Marine Fisheries are being evaluated to confirm this trend and extend it to more recent years. New Bedford Harbor is the site of major PCB contamination and the concentrations in flatfish muscle, at least through 1980, clearly exceeded the current U.S. FDA action limit of 2.0 ppm ww in edible tissue

New York Bight flat fish were resampled several times between 1975 and 1981 for PCBs in flatfish. Relatively low but fluctuating concentrations (less than 0.05 to greater than 0.3 ppm ww) were apparent with no obvious increasing or decreasing trend.

This close-up review of PCB trends in flatfish suggests that at six sites in three urbanized areas, levels of PCBs in muscle of flatfish have declined since the mid-1970s by factors of 2 to 10 times.

The trends are apparent despite the inclusion of various species at different time periods. The major decreases in concentrations appeared during the mid-to-late-1970s. These data for flatfish, largely from local monitoring programs, show generally more obvious declines in PCB contamination than are evident on a national basis from aperiodic national programs of other species except menhaden (Stout, 1981; Chapter 7). The local site data selected for this analysis are inadequate to extrapolate to the national level. The possibility exists that local monitoring efforts have been more consistent than national efforts and that additional careful assessments of these and other local monitoring programs would resolve nationwide trends for PCBs not otherwise evident from past nationwide programs. For example, in the case of Southern California, it seems clear there have not only been declining PCB levels at former point source sites (e.g., Santa Monica Bay, Palos Verdes, and Orange County) but also regionwide, by factors of 2 to 10 times over the past decade.

6.3 Summary and Conclusion

Based on available evidence, highest PCB levels in the muscle of flatfish and in whole bivalves have occurred in urban embayments of the Pacific and Atlantic coasts. This is also true for fish liver data as described in Chapter 4. Flatfish muscle sampled in offshore waters had lower PCB levels than those captured in bays by a factor of 10 to 100. The Gulf Coast appears to have been undersampled, but evidence suggests that PCBs are not a major contaminant for bivalves from Gulf areas except in the Pensacola Escambia Bay area. In the Pacific, highest levels of PCBs were documented in flatfish and bivalves from San Diego Harbor, San Pedro Harbor, Palos Verdes Peninsula, Santa Monica Bay, and Elliott Bay along the Pacific Coast. Atlantic Coast sites with highly contaminated flatfish and shellfish include Boston Harbor, New Bedford Harbor, the Hudson River, Long Island Sound, and the New York Bight.

PCB levels appear to be variable in bivalve mollusks from Boston Harbor and declining in flatfish, as well as other species (Chapters 4, 5, and 7), from the New York Bight, Palos Verdes Peninsula, San Pedro Harbor and Orange County, and Puget Sound. The body of evidence examined to date indicates that PCBs in some coastal and estuarine flatfish and bivalves of the United States were declining by the early 1980s. However, because of inter-project analytical differences the decreases are not evident at the national level from national programs, are also not great at sites away from former historical point sources and require statistical confirmation. Local monitoring programs may be yielding clearer pictures of trends than have past national programs that lack interannual continuity. A challenge is to determine trends in less urbanized estuarine regions and also in biota of the shelf, slope, and EEZ. A considerable body of existing data from local programs is available for such an analysis.

In conclusion, there appears to be a sufficient body of historical data on PCBs in Atlantic and Gulf coast bivalves and Atlantic Coast and Gulf of Mexico offshore flatfish, to make some comparative judgments about recent (circa 1980) geographic patterns and some longer term trends. The data base needs to be further examined for information on Gulf of Mexico coastal and estuarine flatfish. Further, we narrowly focused here on bivalves and flatfish muscle tissue to be consistent with the recent (Matta et al.) assessment of trends for the West Coast. A large amount of data (more than 5,000 samples) exist for other species, but may require innovative trophic or taxonomic compositing for making additional inferences about spatial and temporal trends in PCBs.

7.0 TRENDS IN SELECTED RESOURCE SPECIES

While all species are valuable from an ecological and aesthetic point of view, public attention is frequently focused on only a few species of recreational or commercial value. What can be said about trends in chlorinated pesticide and PCB contamination in these species? This chapter uses national and local data to review trends for two groups of fishes, menhaden and striped bass.

7.1 Menhaden

Menhaden are small- to medium-sized, herring-like fishes of great commercial and ecological importance in the Atlantic and Gulf of Mexico. Wherever they occur they are prominent prey of larger fishes, sea birds, and marine mammals. They are a principal marine forage fish of North American waters. And finally, they are the focus of significant roe fisheries and reduction fisheries yielding meal used as poultry food and oil used in a variety of edible and inedible products. Indeed, they are a naturally oily fish, making them and their products prime candidates for accumulating and monitoring PCBs and chlorinated pesticides.

Data were encountered for over 800 samples of menhaden. PCB and pesticide contamination in menhaden was investigated annually from 1969-1977 during an NMFS monitoring program (Stout et al., 1981). PCB and pesticide residues were measured in whole fish, oil, and fish meal from 314 samples that were collected from 11 menhaden processing plants along the Atlantic and Gulf coasts. An NPMP monitoring survey analyzed 89 whole juvenile menhaden from the east coast and Gulf estuaries during 1972-1976 (Butler and Schutzmann, 1978).

The New York Department of Environmental Conservation (NYDEC) and the New Jersey Department of Environmental Protection (NJDEP) produced 60 PCB residue values of whole and filleted menhaden from the New York Bight and Long Island Sound during 1975-1978. In addition, unpublished data of whole menhaden PCB levels from this region were provided by Virginia Stout (NMFS, Seattle, Washington).

7.1.1 PCBs in Menhaden

Overall, PCB residue levels were highest in menhaden from the Northeast Coast but generally declined in all four substrates analyzed (whole fish, oil, fish meal, and muscle) during the sampling period 1969-1979 (Stout et al., 1981). This trend was apparent for whole fish in several areas but not in whole fish from Long Island Sound or in fillets from New York Harbor Menhaden in which PCB levels increased during the late 1970s (Figure 7.1). The increase is mainly due to a relatively high mean PCB concentration for 1978 and 1981. Unfortunately, sampling of whole menhaden after 1976 was not performed in any study; therefore, a comparison of this upward trend to that at any other site is not possible.

The highest PCB concentrations measured by Stout occurred in oil, meal, and whole fish from the New York Bight area. This is not surprising since many other species of fish from this area also contained relatively high concentrations of PCBs. Juvenile estuarine menhaden from North Carolina contained the lowest concentration of PCBs during the sampling period.

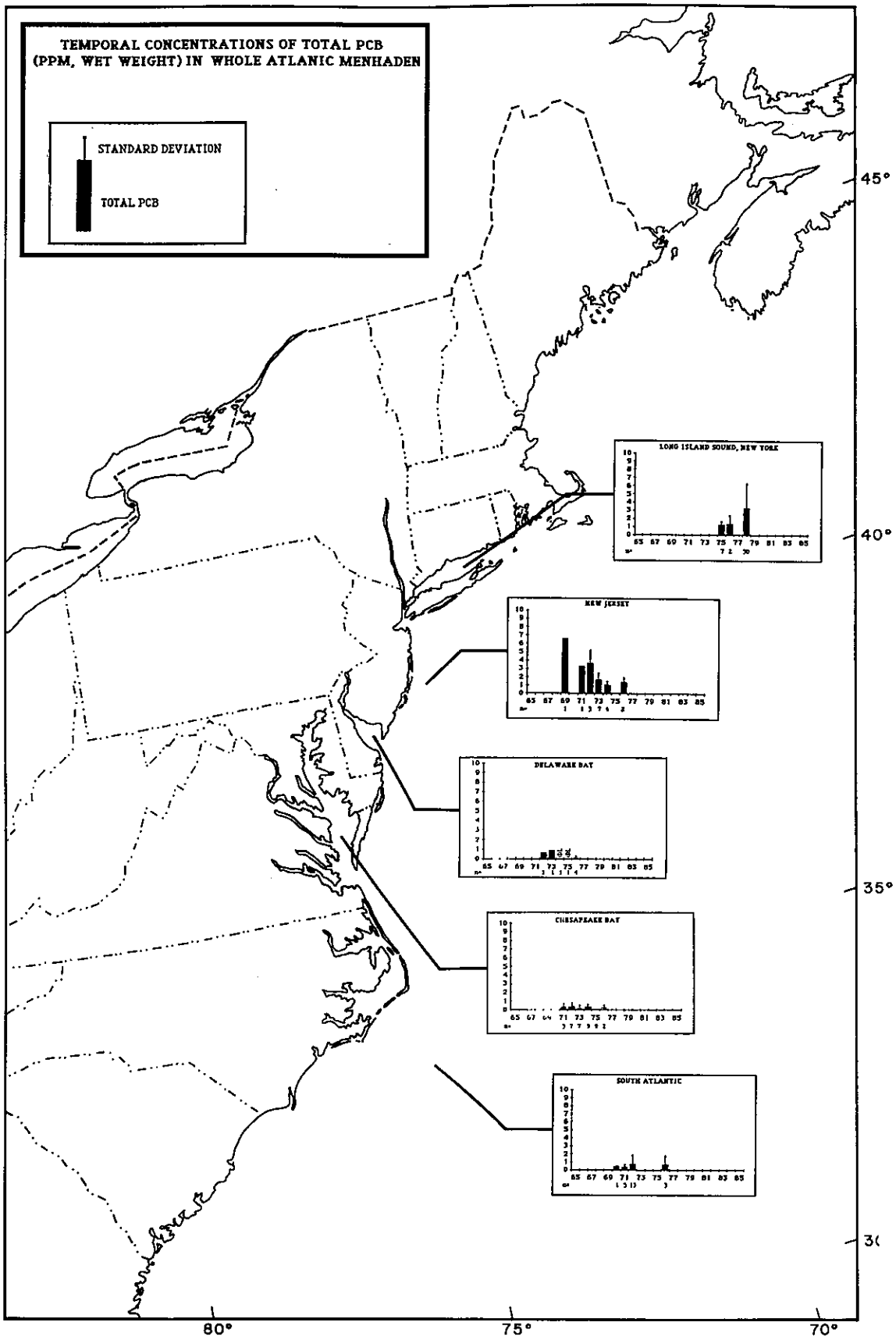


Figure 7.1. Annual variations of average tPCBs (Aroclor) in whole Atlantic menhaden from five Atlantic Coast areas 1969-78. Based on data from sources cited in text.

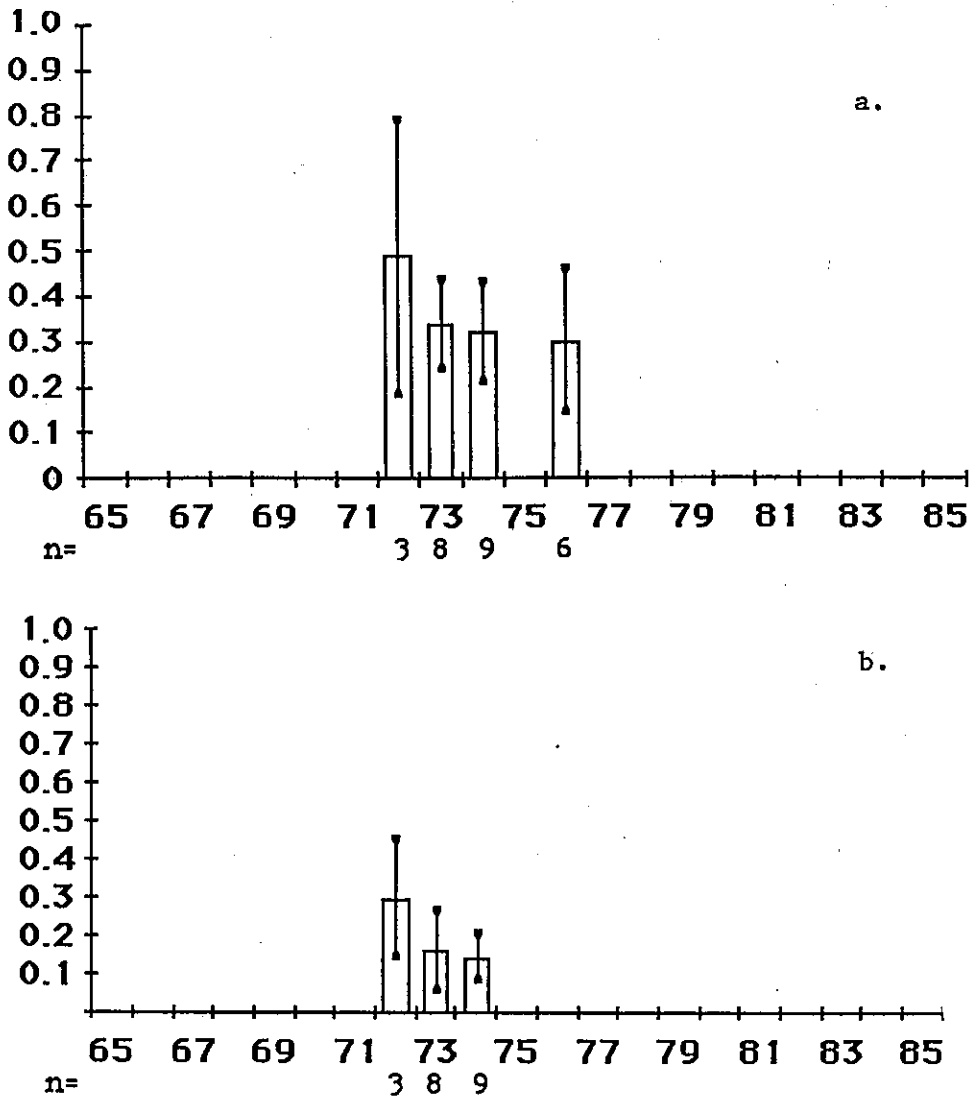


Figure 7.2. Annual variations of average tPCBs (Aroclor, ppm ww) in menhaden delivered to reduction plants in Louisiana and Mississippi, 1972-76: (a) whole fish, (b) meal. Based on original data supporting Stout et al., 1981.

Since data are sparse after 1979, it is difficult to accurately determine the recent status of PCB contamination in Atlantic and Gulf menhaden, but the results of sampling in the 1970s suggest that levels in all substrates had been declining.

7.1.2 DDT in Menhaden

Data on DDT in whole Gulf and Atlantic menhaden are mostly available from the efforts of Stout et al. (1981) and the NPMP monitoring survey on juvenile estuarine fish (Butler and Schutzmann, 1978) (Figures 7.3 and 7.4).

Whole Atlantic menhaden from lower Chesapeake Bay had higher mean DDT levels than fish caught from the upper regions of the Bay. The year with highest concentration was 1970 (0.795 ppm ww; Figure 7.3). Since 1971, DDT levels in both upper and lower Chesapeake Bay dropped below 0.1 ppm ww. DDT levels in Atlantic menhaden from Rehoboth Beach, Delaware and Delaware Bay were at or below 0.1 ppm ww from 1972 to 1976 (Figure 7.3). A decline from very high DDT levels was also evident for Atlantic menhaden found in Raritan Bay, New Jersey. Mean DDT concentration values dropped tenfold from 1969 to 1976 (Figure 7.3).

Three of the four sites sampled for juvenile whole Atlantic menhaden between 1970 to 1976 on the North Carolina coast, as well as one site from Georgia, all reported DDT levels less than 0.1 ppm ww (Figures 7.3 and 7.4). Adult whole Atlantic menhaden from Onslow Bay, North Carolina had the highest overall mean DDT level in the southeast region with highest mean concentration occurring in 1972 (0.5 ppm ww).

The highest total DDT level documented in any menhaden were from Port Harlingen, Texas in 1972 when the mean value of two samples of whole juvenile Gulf menhaden was 23 ppm ww (Figure 7.4). In the 4 years that followed, the yearly mean DDT values declined and leveled off to one tenth of the original value. In the surrounding Laguna Madre area juvenile Gulf menhaden DDT levels were highest in 1974 and 1975. DDT levels in adult whole Gulf menhaden from Mississippi Sound declined slightly from 1970 to 1974, but in 1976 an increased mean value was observed (Figure 7.4).

Three surveys analyzed menhaden samples for pesticides other than DDT. NPMP, Stout et al. (1981), and Foehrenbach et al.(1971) measured up to 12 pesticides in 386 samples of Gulf or Atlantic menhaden. The pesticides most commonly searched for were dieldrin and endrin.

7.1.3 Other Pesticides

Most of 356 samples contained measurable levels of dieldrin. The maximum value, 3.17 ppm ww, was detected in menhaden oil from New Jersey in 1969 (Stout et al., 1981). The highest level detected in whole menhaden was 0.45 ppm ww in a 1970 sample from Hempstead Harbor, New York Dieldrin was also detected in menhaden from Flanders Bay, New York; the Chesapeake Bay; Rehobeth, Delaware; North Carolina; Louisiana; Moss Landing, Mississippi; Oso, Texas; Port Harlingen, Texas; and Arroyo City, Texas.

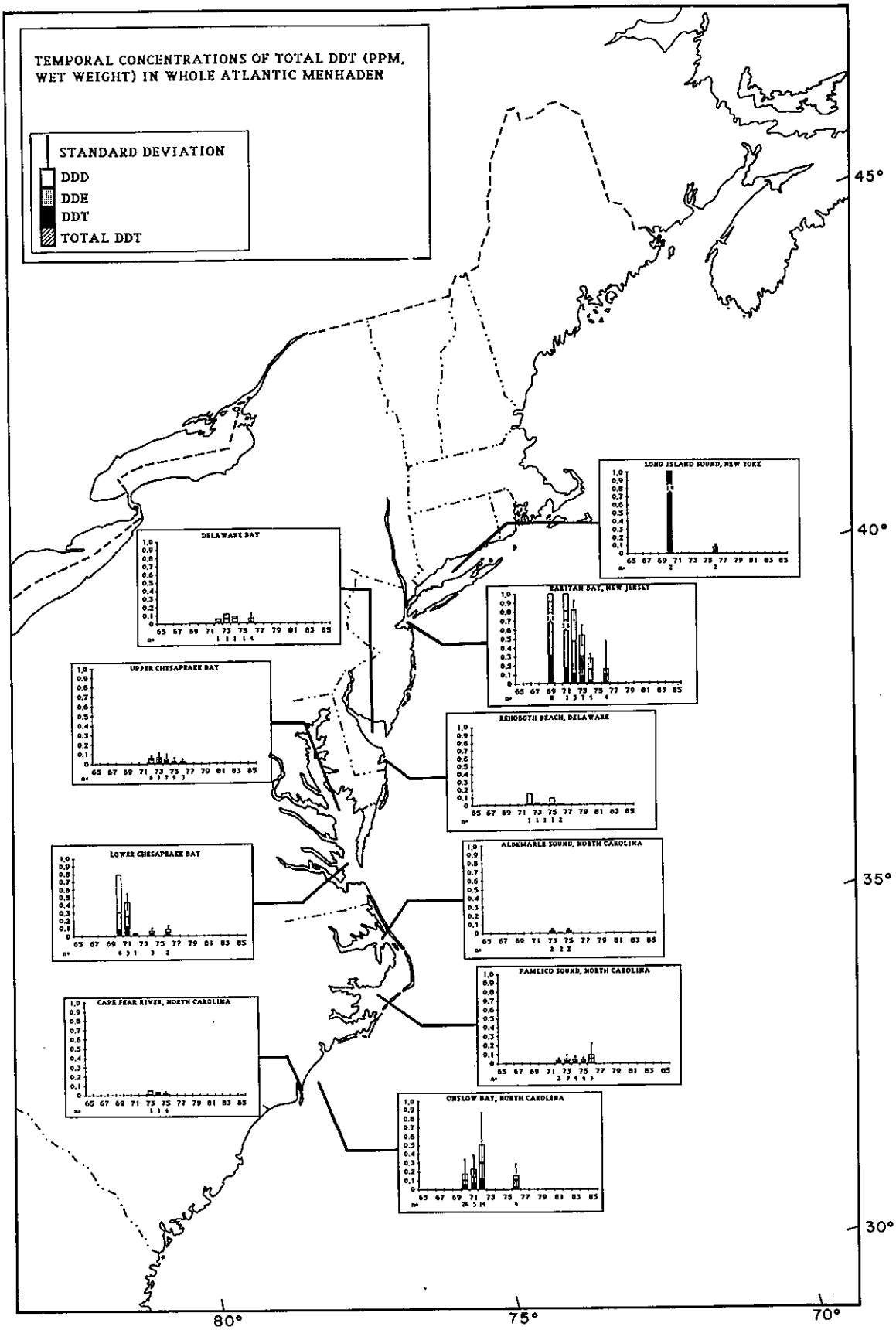


Figure 7.3. Annual variations of average tDDT in whole Atlantic menhaden from 10 collecting or reduction plant sites along the Atlantic Coast, 1969-76. Based principally on original data supporting Stout et al., 1981 and Butler and Schutzmann, 1978. See text for more details.

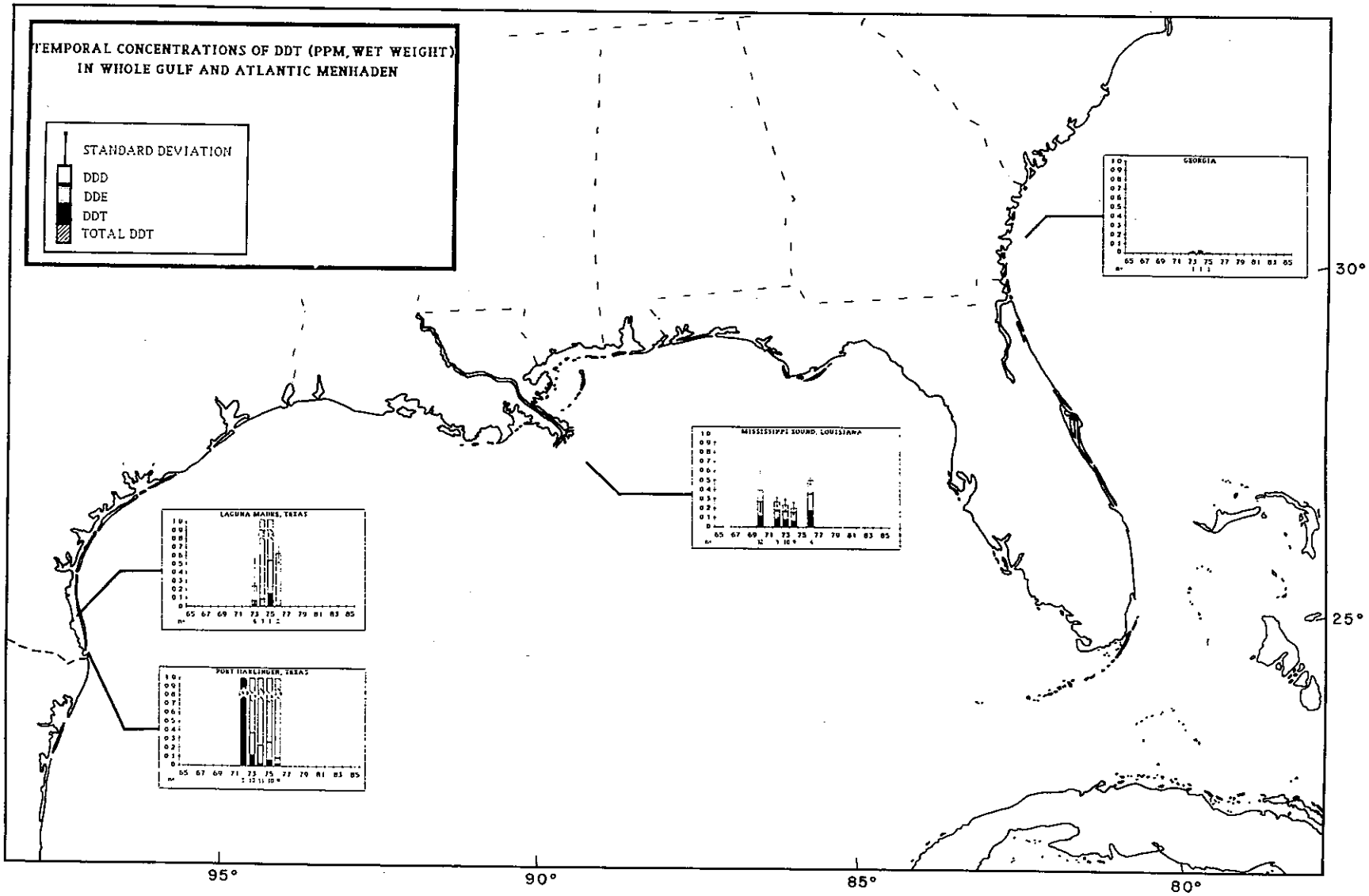


Figure 7.4. Annual variations of average tDDT in whole Gulf and Atlantic menhaden from four Southeast and Gulf of Mexico collecting or reduction plant sites, 1970-76. Based principally on original data supporting Stout et al., 1981 and Butler and Schutzmann, 1978. See text for more details.

Endrin was less frequently detected than dieldrin. The highest level (0.259 ppm ww) was detected in fish oil from Louisiana in 1974 (Stout et al., 1981). The highest level in whole fish was 0.197 ppm ww from a sample from Port Harlingen, Texas in 1974 (data supporting Butler and Schutzmann, 1978). Endrin was also detected in menhaden from New Jersey, the Chesapeake Bay, North Carolina, and Arroyo City, Texas.

Chlordane was detected in nine samples of menhaden from Port Harlingen and Arroyo City, Texas; Oyster Bay, New York; and the Chesapeake Bay. The maximum level was found in a sample of whole menhaden from Port Harlingen, Texas in 1975 (0.20ppm ww).

Toxaphene was detected in 43 samples of menhaden from Port Harlingen and Arroyo City, Texas and Moss Landing, Mississippi. The maximum level, 8.8 ppm ww, was detected in whole fish from Port Harlingen, Texas in 1974. Three samples from Port Harlingen also contained the herbicide dachthal (DCPA) in 1974 (up to 0.164 ppm ww; see Chapter 5).

Pesticides not detected in menhaden by NPMP (Butler and Schutzmann, 1978), Stout et al. (1981) or Foehrenbach et al. (1971) were aldrin, heptachlor, heptachlor epoxide, lindane, endosulfan, methoxychlor, mirex, and trifluralin.

7.2 Striped Bass

No other estuarine sport fish has been subject to as great of a concern in the United States as the striped bass. Ranging from the maritime provinces of Canada south to Georgia, and with significant introduced populations in California and Oregon, the striped bass now captures the imagination of fishermen on both coasts. The striped bass is an anadromous fish, migrating from the ocean to spawn in lower reaches of rivers.

Data were encountered on PCB levels in over 5,000 samples of striped bass. Most (3,500 samples) were generated by the NYDEC in response to the PCB-contamination of the Hudson River as reported in Sloan et al. (1984) and described above. Most of the balance of the data comes from San Francisco Bay (based on original data supporting Whipple et al., 1983) but there are additional comparative samples from southern California, Coos Bay, the Chesapeake Bay, Delaware Bay, New Jersey bays, Rhode Island, Massachusetts, and North Carolina.

Striped bass were sampled most heavily between 1977 and 1980. Grouping all samples analyzed during these years allows a comparison of PCB levels in at least 10 areas of the Pacific and Atlantic coasts (Figure 7.5). In this comparison striped bass in the Hudson River were the most contaminated with PCBs, containing a mean level of almost 9 ppm ww during these years. Striped bass from other areas of New York and from New Bedford Harbor were also highly contaminated (containing between 1.5 and 4 ppm ww total PCBs).

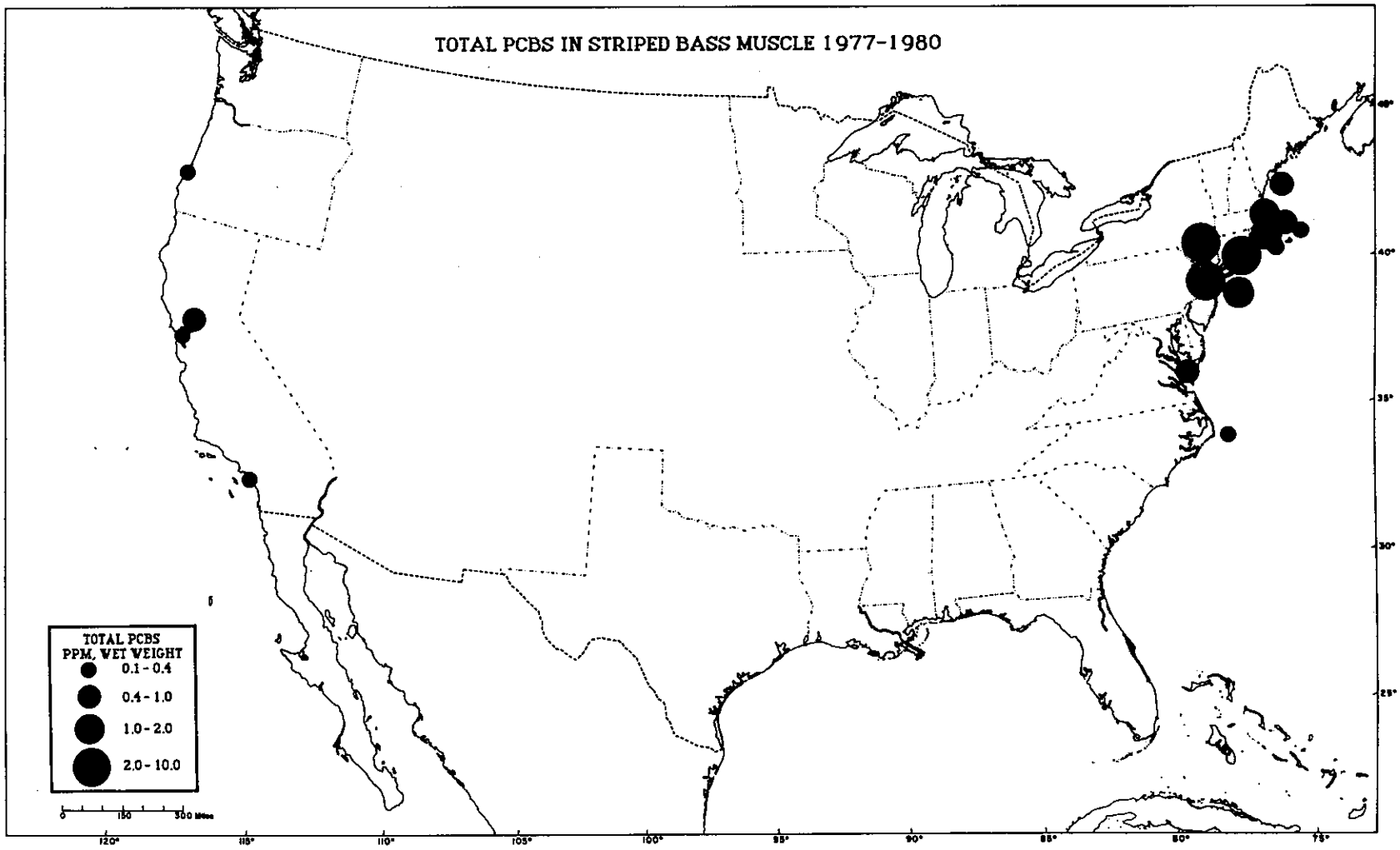


Figure 7.5. Average tPCBs in muscle of striped bass from 16 sites or regions during 1977-80. Based on data from various sources cited in text.

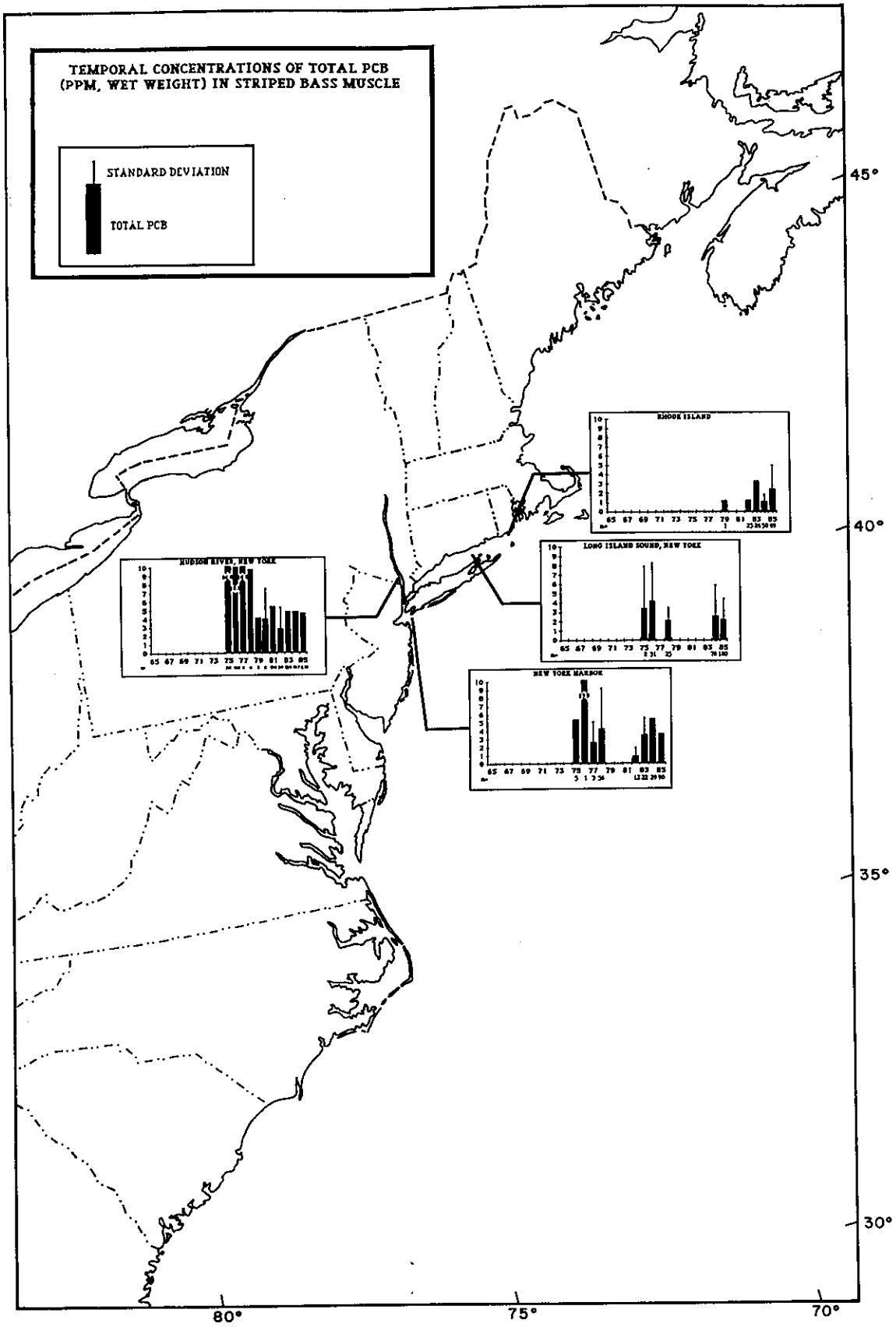


Figure 7.6. Annual variations of average tPCB (Aroclors) in muscle of striped bass from four Atlantic Coast sites or regions, 1976-85. Based on data from various sources cited in text.

Temporal trends of total PCBs in striped bass have not followed the same course in all Atlantic areas. In three areas, levels have declined since 1974, but not dramatically (Figure 7.6). PCBs in fish from the Hudson River have declined from 24 ppm ww in 1975 to 4.6 ppm ww in 1985. Similar, though less dramatic, declines occurred in fish from Long Island Sound and New York harbor. Meanwhile, PCB levels in Rhode Island striped bass have increased since 1979. In all four regions, concentrations were converging at a level of about 3.0 ppm ww, or just above the FDA action limit of 2.0 ppm ww.

Surprisingly, DDT has been analyzed in only approximately 250 samples of striped bass since 1970. This is only 5 percent of the samples subjected to PCB analysis. Most samples were generated by national surveys (Whipple et al. (1978), Butler et al. (1976), Stout, unpublished data, and CEMP (Butler, 1978). These surveys collected fish from New York, Chesapeake Bay, San Francisco Bay, Umpqua River, Coos Bay, and Newport Bay.

The widest distribution of sampling occurred between 1973 and 1976. Combining data on fish collected during these years shows that flesh of striped bass from the Sacramento River had the highest total DDT level--1.04 ppm ww (Figure 7.7). Temporal trends in total DDT levels in striped bass can be developed for the San Francisco Bay and Chesapeake Bay. Through 1981, striped bass total DDT levels appeared to decline in San Francisco Bay but was variable and increasing in Chesapeake Bay (Figure 7.8).

Several surveys analyzed striped bass for pesticides other than DDT. Up to 116 samples have been analyzed for 14 pesticides. The pesticide most commonly searched for and detected was dieldrin. Although most samples contained levels less than detection limits, highest levels of dieldrin were detected in striped bass gonads from the Hudson River in 1982 (0.18 ppm ww). These fish also contained the highest levels found in flesh (0.042 ppm ww). Other areas where dieldrin was detected in striped bass include the Chesapeake Bay, the San Joaquin and Sacramento Rivers, and Montauk Point, New York.

Another commonly sampled pesticide in striped bass was aldrin, but only four samples contained measurable levels, all below 0.01 ppm ww. These samples were taken between 1978 and 1982 in San Francisco Bay, Long Island Sound, and the New York Bight. Alpha-chlordane has also been commonly measured in striped bass. Gonads of fish from the San Joaquin and Sacramento Rivers contained up to 0.17 ppm ww in 1978. Fish from the Hudson River contained highest levels of alpha-chlordane in flesh and gonads (0.9 ppm dw and 0.53 ppm ww, respectively).

Toxaphene has rarely been searched for in striped bass samples, but was detected in gonads of striped bass from San Francisco tributaries: four samples contained more than 1 ppm ww toxaphene in 1978 and 1980, with the maximum level (1.9 ppm ww) detected in 1978.

Trans-nonachlor was found in striped bass at concentrations up to 0.82 ppm ww in the San Joaquin River in 1980. The maximum level of heptachlor epoxide in striped bass was found in a sample from the Potomac River in 1981.

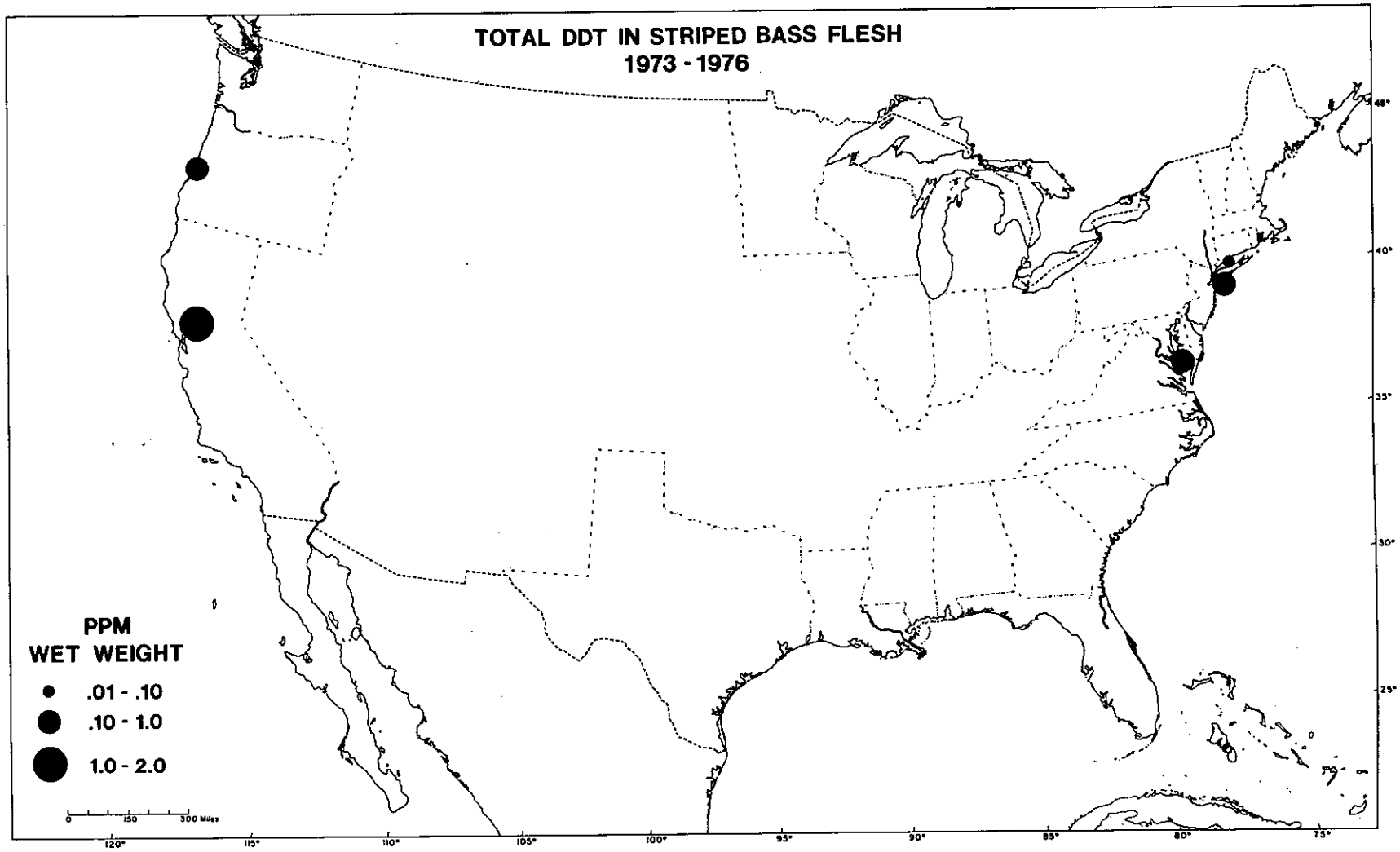
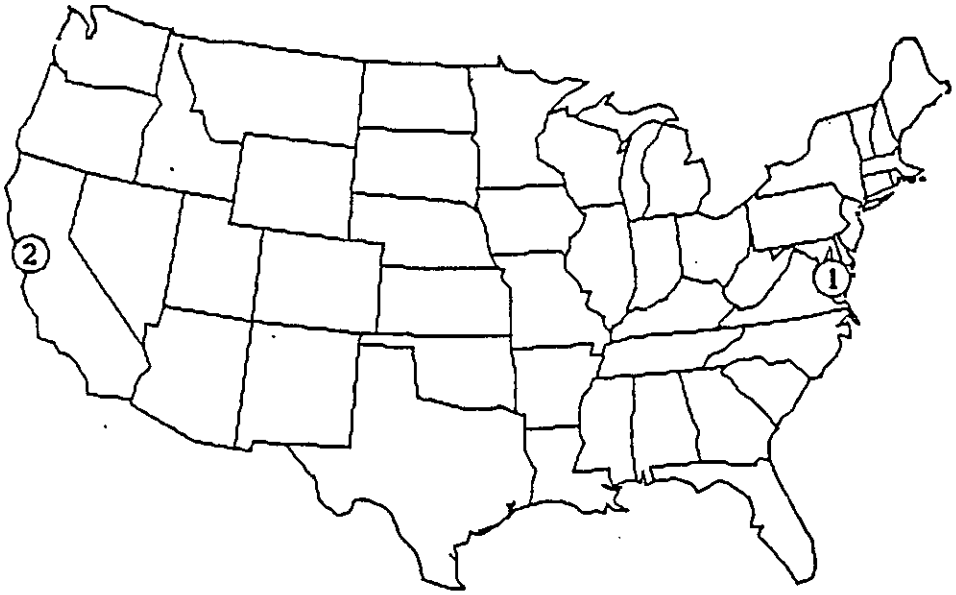
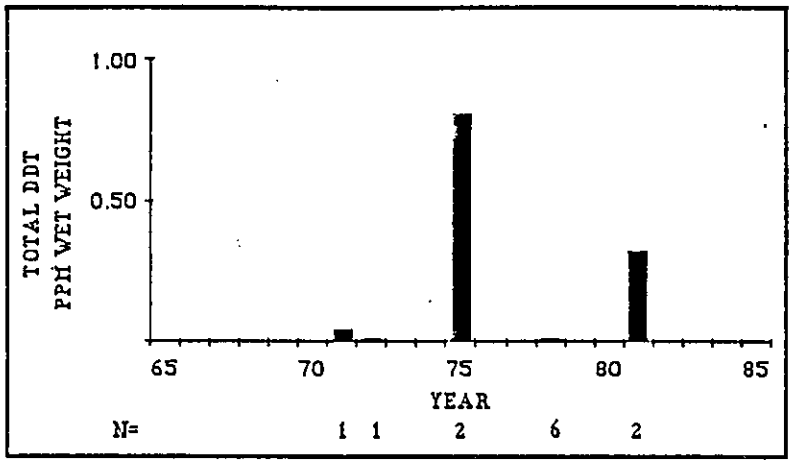


Figure 7.7. Average tDDT in flesh of striped bass from five sites, 1973-76. Based on data from several sources cited in text.



1. CHESAPEAKE BAY



2. SAN FRANCISCO BAY

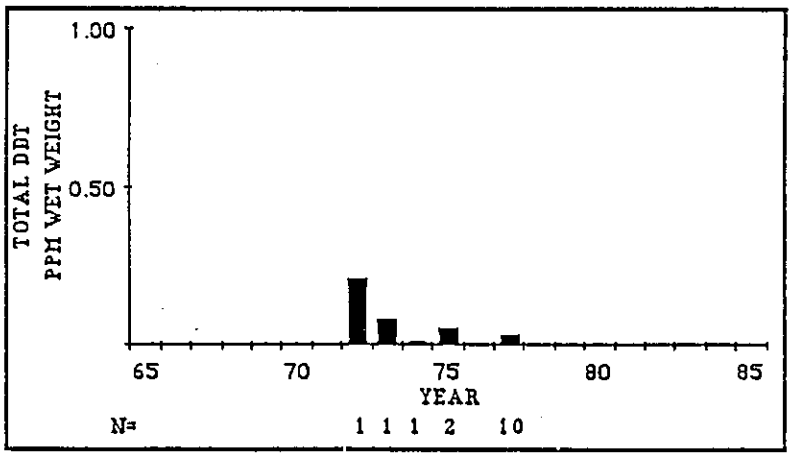


Figure 7.8. Annual variations of average total DDT in whole young striped bass from the Chesapeake Bay and from San Francisco Bay, 1971-81. Based largely on data supporting Butler and Schutzmann, 1978.

Mirex, heptachlor, and lindane have not been reported in striped bass above a detection limit of 0.02 ppm ww. Levels of endrin have not exceeded 0.04 ppm ww detection limit. However, these pesticides have been rarely analyzed in striped bass.

7.3 Other Resource Species

Many other resource species--anadromous, estuarine, and offshore--have been subject to analyses of PCBs and chlorinated pesticides. As noted elsewhere in this report, the bivalves--oysters, clams and mussels--have been particularly well monitored in part due their status as "indicator" or "sentinel" species. The NS&T Program is continuing to focus on these.

In addition, there have been other resource-specific surveys not yet reviewed in detail by this project. One of the most important of these is the recently completed 1984-85 survey of PCBs in Atlantic bluefish explicitly directed to the entire population from Florida to New England (NOAA, 1986). A particularly important aspect of this study was the explicit nature of the sampling and survey design which was clearly focused on determining the extent to which various size groups approached or exceeded the FDA action limit of 2.0 ppm ww. Only the largest fish exceeded this value, with several concentrations exceeding 40.0 ppm ww. In addition, that study also included analyses of 10 chlorinated pesticides including DDTs, four chlordane-related chemicals, HCB, lindane, and dieldrin. When coupled with older data from other local and regional surveys, this survey should represent a hallmark effort in the history of monitoring chlorinated chemicals in coastal fish resources.

There also remain older resource-focused surveys on other species that remain largely unknown and largely unreviewed. Principal among these on the Atlantic Coast, is a 1969 interstate survey of chlorinated pesticides in blue crab populations along the southeast coast (North Carolina to Florida, Mahood et al., 1970). In this survey, blue crab composites were collected monthly from 20 sites (5 per state) and analyzed for DDE, DDD, DDT, dieldrin, and mirex. When re-examined and coupled with more recent data, it may be possible to document regional and long-term trends in this species and also to determine the extent to which seasonal variability affects contaminant burdens in crabs.

7.4 Summary and Conclusion

Menhaden, a fish of both the open coast and estuary, was clearly contaminated by PCBs and chlorinated pesticides along much of the mid-Atlantic coast and in specific areas of the southeast and Gulf of Mexico. The most contaminated menhaden, considering all pesticides, were fish from Arroyo Colorado, Texas. Where concentrations of PCBs and DDT were high in the early 1970s, levels clearly declined through the late 1970s; however, few fish or sites were sampled in the 1980s. New sampling along the entire Atlantic and Gulf coasts is needed to confirm that trends of the previous decade have continued to the present. In any case, there was evidence for large-scale, regionwide decreases in PCBs, which was not evident in other national monitoring programs.

The highest concentrations of PCBs in striped bass have and continue to come from fish in the Hudson River. The most recent data and analyses from the NYDEC (Sloan et al., 1987) suggest a continuing slow decline and that these fish will not be below the FDA action limit of 2.0 ppm ww until after the year 2019. A more regional view, as reported here, also suggests that concentrations may continue to increase in fish from peripheral areas. To confirm these projections and hypotheses, a long-term region-wide (northeast) monitoring commitment is needed.

Hudson River striped bass also contained highest levels of PCBs, dieldrin, and chlordane while fish from the Sacramento and San Joaquin Rivers contain highest levels of DDT, toxaphene, and trans-nonachlor. Highest levels of these compounds have been detected in gonads. Trends with time are variable for DDT in San Francisco Bay.

Considering that over 5,000 samples of striped bass have been analyzed for PCBs, it was disappointing to find so few (less than 250) analyzed for other chemicals.

Additional review of the data is needed to identify other resource species--both coastal and estuarine that may hold promise for developing long-term trends or large-scale geographic patterns.

8.0 DISCUSSION AND IMPLICATIONS

This report describes how much data already exists for documenting the geographic distribution and long-term trends of PCB and chlorinated pesticide contamination in U.S. coastal fish and shellfish. By splicing together data from different surveys it then reconstructed the history of contamination on a national basis for 17 specific chemicals or chemical groups. Finally, by way of examples, this report described the extent to which existing data can be used to document spatial, temporal taxonomic trends in specific bays and species groups.

This chapter summarizes the salient features of this review by focusing on information that could be productively filled with addition analysis or monitoring. Specifically, it examines previously-presented information in terms of its continuity and adequacy in identifying geographic patterns, species representativeness, and additional measurements needed to determine present trends in PCBs and chlorinated pesticide contamination.

8.1 Chemicals of Concern

Which chemicals are of concern and which are not? To address this question, each chemical was classified according to several criteria concerning direction of trends and adequacy of knowledge. This classification appears in Table 8.1 and is reviewed below.

8.1.1 Contaminants of Continuing Concern

PCBs, DDTs, dieldrin, and chlordane are candidates for continued assessment and monitoring on a national basis. Each of these chemicals have been geographically widespread in coastal and estuarine organisms; that is, they have been readily measured contaminants at more than just a few isolated "hot spot" sites. Concentrations have declined, at least at former "hot spots", but at least three remain of current concern as evidenced by fishery advisories and closures. These conclusions can be reached despite a considerable amount of discontinuity in sampling locations, species, and duration of programs on a national basis.

Declining PCB contamination of coastal fish and shellfish was demonstrated in several regions and resource species; however, additional analysis of existing data is needed to determine whether or not there has been a decrease in contamination on a national basis. Stout (1987), in a review of trends in PCB contamination of both fish and wildlife, concludes that "...PCB levels have fallen in some areas where they were previously highest but are increasing in previously clean areas and remaining more or less constant where low levels were already noted." Further, she predicts that "...until the 680,000 metric ton reservoir of PCBs produced worldwide since 1929 is destroyed or immobilized, PCBs will continue to accumulate in aquatic and terrestrial organisms." The NYDEC estimates that at the present rate of change, it may not be possible to reopen the Hudson striped bass fishery until the year 2019 when flesh concentrations should be below the FDA action limit of 2.0 ppm ww (Sloan et al., 1987). As concentrations decline in heavily or moderately contaminated areas, such as the Hudson, Boston Harbor, New Bedford Harbor, several bays of Puget Sound, and San Diego Bay, will they

indeed increase to concentrations of significance in adjacent or remote areas? Clearly, continued monitoring is needed to confirm the geographic distribution and direction and magnitude of changes of PCB contamination. And, not only is further monitoring required, it is probably required at sites not currently experiencing significant PCB contamination. In short, nationwide monitoring should continue into the foreseeable future.

Table 8.1 PCBs and chlorinated pesticides listed in terms of status of knowledge and recommendations for surveillance in fish and shellfish on a nationwide basis.

1. CONTAMINANTS OF CONTINUING CONCERN; MAINTAIN SURVEILLANCE

   PCBs
   DDT (mainly DDE)
   Dieldrin
   alpha (cis-)-chlordane
   trans-nonachlor

2. CONTAMINANTS OF UNCERTAIN NATIONAL CONCERN

   Endosulfan
   Toxaphene
   PCP (Pentachlorophenol)
   DCPA (dacthal)
   Endrin
   Kelthane
   Kepone

3. CONTAMINANTS APPARENTLY NOT OF NATIONAL CONCERN; REDUCE SURVEILLANCE FREQUENCY

   Aldrin
   Methoxychlor
   Heptachlor and Heptachlor epoxide
   Mirex
   HCB (hexachlorobenzene)
   Lindane

DDT concentrations in coastal fish and shellfish decreased dramatically on a nationwide basis after 1972 (Chapter 4). The decline in DDT contamination was clearly evident in shellfish and fish of specific bays and estuaries, along the open coast and offshore in the open seas. Data was sufficient to indicate that, compared to today, coastal marine life of the United States contained 100 times more contamination from DDT 20 years ago. By the late 1970s, high concentrations, at or near the FDA action limit of 5.0 ppm ww, were only found among fish in the immediate coastline of the Los Angeles area. Nonetheless, DDT metabolites remained common nationally at

detectable levels in the mid-1980s with geographical concentration patterns similar to those observed in the late 1960s and early 1970s (i.e., higher in Southern California and Delaware Bay than elsewhere). Furthermore, fish with levels approaching the FDA action limit are occasionally discovered along the California coastal shelf beyond the Los Angeles area.

Continued monitoring of tDDT in fish and shellfish may or may not be needed on a national basis, but it clearly is needed locally in areas of high past use or disposal including the Southern California Bight, Delaware Bay, and several agriculture areas such as the Arroyo Colorado, Monterey Bay, and San Francisco Bay. Further, since the metabolite DDE is a contaminant of the acaracide kelthane and has recently appeared in wildlife as a result of kelthane use (Hunt et al., 1986 and Risebrough et al., 1986), it may be important to continue monitoring DDE on a national basis.

Prior to 1972, dieldrin was the third most frequently occurring organochlorine pesticide in U.S. shellfish. Concentrations were generally moderate (less than the FDA action limit of 0.3 ppm ww) in shellfish or fish in larger embayments of the northeast, southeast, and Gulf of Mexico and at apparently isolated localities along the Pacific Coast and in the Hawaiian Islands. Declining dieldrin concentrations occurred nationally although the magnitude of the decline has not been estimated. Dieldrin continues to be the third or fourth most common and concentrated organochlorine on a nationwide bases.

Dieldrin remains a common contaminant but apparently not at concentrations of concern, at least in terms of public health. Unless research suggests otherwise, it need only be monitored on a national basis periodically until questions of public health significance are resolved.

Contamination of coastal and estuarine fish and shellfish by chlordane compounds (chlordanes and nonachlors) occurred at apparently isolated localities on all three coasts and Hawaii. Recent data (since the mid-1970s) was insufficient to determine if chlordane has been a more widespread contaminant beyond the immediate vicinities of urban and agriculture areas. In any case, in the mid-1980s, concentrations in some urban areas were sufficiently high (at or near the FDA action limit of 0.3 ppm ww) to warrant seafood consumption warnings (Baltimore Harbor and a Connecticut estuary). Furthermore, chlordane compounds collectively compete with dieldrin as the third or fourth most abundant organochlorine on a nationwide basis in the mid-1980s.

Continued nationwide monitoring of chlordane compounds is justified on the basis that it remained in use through 1986, is common in fish from some urbanized areas, occurred in oceanic animals in the early 1980s, and is the cause of at least one continuing advisory. The data base examined in preparing this report was inadequate to establish trends and additional data is required to further clarify the geography and direction and magnitude of trends.

8.1.2 Contaminants of Uncertain National Concern

Sufficient uncertainty exists about the national status of seven chemicals or chemical groups to warrant additional monitoring or assessment. Six of these--endosulfan, toxaphene, PCP, kelthane, kepone, and DCPA (dacthal) have not been subject to a national coastal survey, but there is evidence from local monitoring programs that these still might be important contaminants of estuarine organisms near agriculture areas or chemical production facilities. The current status of toxaphene in the vicinity of a production plant near Brunswick, Georgia and in the Arroyo Colorado, is unknown. We do not know if contamination has spread beyond these areas or if organisms in other estuaries have experienced increasing toxaphene contamination in concert with that experienced by inland fishes of the midwest and California. Presumably kepone has only been a contaminant of the James River, Virginia. We do not know if concentrations of endosulfan, kepone, PCP, or DCPA are important beyond sites of recent or past local monitoring. We do not know if kelthane is a new contaminant of national significance. These uncertainties are a direct result of a lack of recent nationwide sampling, that is, a lack of continuity since the mid-1970s. A review of recent use information and of state monitoring efforts might help resolve these uncertainties, but cannot replace an actual national survey.

A fifth pesticide, endrin, was surveyed in the 1960s and 1970s, occurred in both coastal and estuarine organisms, apparently declined in concentration but has been excluded from nationwide surveys since then. Presumably endrin is no longer a contaminant of coastal organisms, but to be certain, it too should be resurveyed.

8.1.3 Contaminants Apparently Not of National Concern

For several reasons, six additional chemicals or chemical groups do not appear to be of immediate concern and do not have great justification for frequent monitoring on a national basis. Aldrin, heptachlor (and its epoxide), mirex (dechlorane), HCB, and lindane were included in national coastal surveys from the late 1960s and continuing through 1986. They were rarely detected above the 0.01 ppm ww historical detection limits in fish or shellfish and in 1984 did not occur above that concentration in fish liver samples from 48 sites sampled on a nationwide basis. Aldrin, which is no longer in use, is rapidly metabolized to dieldrin so there is no reason to expect it now. Heptachlor is mainly a minor constituent of technical chlordane and might be worth investigating in areas of significant chlordane and nonochlor contamination. Mirex is no longer used, and even near areas of past application was rarely detected in estuarine biota (once it was correctly identified). HCB and lindane (gamma HCH) probably continue to be locally important contaminants in Puget Sound and Hawaii, respectively, but are not so nationally.

The last chemical, methoxyclor, was used to replace DDT, but it has rarely been reported in measurable concentrations in past national programs or in recent state and local programs.

Ongoing national monitoring programs could benefit greatly reduce the frequency of measuring these chemicals and increasing the effort on those of uncertain national status.

8.1.4 Other Encountered Chemicals

During the 1960s and 1970s, the organochlorine pesticides were replaced by carbamate and organophosphorous pesticides. Data were encountered for some of these in estuarine organisms and it may be important to consider them in future nationwide monitoring efforts. The pesticides and maximum concentrations in ppm ww included ethyl parathion (0.07), methyl parathion (0.12), ethion (0.15), trithion (0.21), and DEF (0.14) measured in tissues of spotted sea trout from the Arroyo Colorado during the 1972-76 NPMP estuarine fish surveys (P. Butler, unpublished data); methyl- and ethyl parathion and ethion were also measured in fish from Oso Bay, Texas. In California, chlorpyrifos occurred in bay mussels from Point Mugu Lagoon and Anaheim Bay and in freshwater mussels near the head of Newport Bay, in Southern California, and upstream of Elkhorn Slough in Monterey County. Diazinon also occurred in freshwater mussels upstream of Elkhorn Slough (Stephenson et al., 1986). No national coastal surveillance programs have been undertaken since 1976 that explicitly include measures of these second and third generation pesticides. Further analysis of local survey reports could reveal additional data.

8.2 National Geographic Completeness

Has past sampling been geographically complete? Are there important parts of the coast or embayments that have been inadequately sampled or monitored?

From a national viewpoint, past and recent sampling, has been surprisingly complete. For example, by 1976, the NPMP had sampled sites from all coastal states except New Hampshire. Today, all except New Hampshire have or are being sampled as part of the NS&T Program.

Nevertheless, there remain several major stretches of coast that have been poorly sampled.

The least known areas of all in terms of recent PCB and chlorinated pesticide contamination include most of Alaska, the entire Atlantic coast of Florida, the coast of Oregon, Delaware Bay, Hawaii, the Virgin Islands, Puerto Rico, and perhaps the Pamlico Sound complex of North Carolina. The opportunity should be taken soon to intensify sampling in these areas or locate and analyze locally collected data.

Caution should be exercised in assuming that contamination always increases from the sea toward the most urbanized or industrialized segments of estuaries or that pelagic fish of the coastal shelf are less prone to contamination than estuarine bottomfish or bivalves. For example, the case was presented that DDT concentrations may have been higher in fish from offshore sites than in bays during the late 1960s and early 1970s along the West Coast. PCB concentrations are nearly as high in bluefish from open coastal areas (NOAA, 1986) as in striped bass from areas such as the Hudson River estuary.

We know of no current programs dedicated to the monitoring of coastal shelf or offshore fish or shellfish populations, but it is clear from the 1986 Cooperative Bluefish Sampling Program (NOAA, 1986) that fishes of the open coast are not contaminant free. Perhaps an infrequent survey of fishes of the EEZ is needed to confirm that contamination is not spreading seaward as a result of, or in spite of, coastal and estuarine clean up efforts. A periodic resurvey of bluefish along the Atlantic coast would build upon that recent data base. Likewise, resurveys of ocean species sampled during other previous synoptic surveys in the Pacific (e.g. Stout and Beezhold, 1961), Atlantic, or Gulf of Mexico (e.g. Baird et al., 1974; Giam et al., 1973; ERCO, 1980 and 1981) would build upon existing data in those regions.

In contrast to the coastal and estuarine situation, there has, and continues to be, detailed and extensive monitoring data for PCBs and chlorinated pesticides in inland and freshwater fishes (e.g., Schmitt et al., 1985). The possibility seems to exist that freshwater fish are more contaminated than marine or estuarine fish on a national basis. Many of the NPMP sites described by Schmitt et al. (1981) are located near or upstream of the estuarine sites reviewed here. Moreover, that program has identified trends and patterns that may be relevant in interpreting these data, especially with respect to locations past or recent "hot-spots" caused by past pesticide applications or pesticide production waste discharge. In addition to coordinating coastal with inland monitoring, it would be worthwhile to analyze both sets of historical data together.

8.3. National Sampling vs. Local Conditions

To what extent can national sampling activities characterize local conditions and trends? That is, to what extent do a few sites in a bay or estuary adequately represent local conditions or warn of local aromatics?

These questions were partly addressed in Chapter 5 by focusing on spatial and temporal trends in three widely separated estuaries.

In the case of Chesapeake Bay and San Francisco Bay, it is clear that one or a few sites inadequately represent conditions and trends for the estuary as a whole. Each estuarine segment has its own peculiar suite of contaminants and history. In the case of the long and narrow Arroyo Colorado, two or three sites were sufficient to establish that there was a substantial down-channel gradient of contamination by several pesticides. The lesson is that a few sites might be adequate for long-term characterization and monitoring, but the data can be correctly interpreted only after the bay has been systematically characterized. It can be correctly inferred that a high level of contamination at a single characterization site in a bay indicates a significant contamination problem somewhere in the bay, but it cannot be inferred that a low level of contamination at a single site indicates there is not significant contamination in the bay. Therefore, it is just as important to monitor and characterize apparently clean embayments (as judged from a few sites) as it is to do so in obviously contaminated areas, particularly if there is other evidence of possible contamination such as recent or historical pesticide application in the drainage area.

On the other hand, national surveys are not intended to characterize conditions in specific embayments, estuaries, or localities, but they do provide valuable points of reference so that conditions in specific embayments can be placed in a larger, national perspective.

8.4 Species

To what extent have important species been adequately monitored and which species are most useful?

Although nearly 600 species of U.S. fish and shellfish have been sampled, most of the existing data come from only a dozen species or larger taxonomic groups, namely bivalve molluscs, flatfish, and several groups of pelagic and coastal fishes.

The bulk of existing historical data is for oysters of several species and, secondarily, for mussels and other bivalves. Several hundred historical sites nationwide were established one to two decades ago as part of the NPMP and/or as part of individual state programs.

A problem, not investigated in this review, is that the same species have not been resampled at the same sites in subsequent national or synoptic surveys. For example, at Coyote Point in San Francisco Bay, the NPMP monitored oysters (1965-77) whereas the California Mussel Watch Program monitored mussels. This inconsistency adds uncertainty in establishing trends using absolute concentrations and requires subsequent reviewers to attempt normalizing data in some fashion. Site- and species-specific continuity is needed to insure that bivalve monitoring data are really comparable over time.

Indeed, perhaps the best use of historical data can be made by occasionally returning to these species at the sites where they were formerly sampled. If such an effort was undertaken even once, as many as 200 site-specific time series could be extended by one decade for a cumulative total of two to three decades.

Many important resource species are represented in the data. It was possible to document trends in contamination of two selected species: striped bass and menhaden. Results indicated that Gulf menhaden from Port Harlingen, Texas, like other fish from this area, were highly contaminated with a variety of chlorinated pesticides including DDT, endrin, chlordane, and toxaphene whereas elsewhere (New Jersey and Delaware Bay) Atlantic menhaden experienced high contamination by only one or two chemicals (DDT, PCBs) or experienced low-level contamination (i.e., southeast United States and northern Gulf of Mexico). Past monitoring of menhaden and menhaden products was very useful in demonstrating how pesticides and PCBs declined through the 1970s.

Striped bass from the Hudson River, New York, contained the highest levels of PCBs, dieldrin and chlordane whereas those from San Francisco Bay contained the highest levels of DDT, toxaphene and trans-nonachlor (a-chlordane). Highest levels were in gonads. Declining concentrations were apparent

for DDT In San Francisco Bay and for PCBs in Hudson River and Long Island Sound fish; nonetheless, as recently as 1986, concentrations remained above the FDA action limit of 2.0 ppm ww PCB in striped bass from New York State and adjacent waters.

Flatfish, croakers (drums), striped bass, and small pelagic fish (such as menhaden) have been intensely sampled on all coasts and should be targeted for future muscle tissue monitoring nationwide. Nationwide monitoring, if extended to muscle tissue for these groups, could, at many tens of sites, markedly enhance the interpretation of trends established nearly a decade ago. Intriguing monitoring possibilities exist for several species groups. For example, processing and reduction plants on all three coasts could be revisited to obtain fish and oil from menhaden (Atlantic and Gulf), anchovy (California) and herring (Pacific Northwest, Northeast, and Alaska) for confirming the direction of previously established contamination trends reflecting major coastal and estuarine regions.

Several additional points, not covered in the text of this report, emerge from examination of the data. Additional research could be productively focused on comparing residue trends among various species so that more species could be authoritatively combined into comparable "target taxa." For example, it may be possible to lump rockfishes and cod-like fishes into a group so that existing and future data from nearly 150 species might be made directly comparable.

Crustaceans, namely crab, shrimp, and lobster, have been somewhat overlooked in recent national and regional monitoring programs. Yet there are some regionally important historical and existing data bases such as the 1969 Cooperative Blue Crab surveys along the Atlantic Coast (Mahood et al., 1970). It would be instructive to repeat this survey. The cancer crabs of the Pacific and Atlantic northeast and the blue crabs and their relatives of the mid-Atlantic and Gulf of Mexico and pandalid and panaeid shrimp remain important fishery targets everywhere and could be historically useful targets of a new nationwide reconnaissance survey.

In selected open coastal areas, time series or at least synoptic surveys have been established in the past for lesser known offshore species such as the mesopelagic fishes off California (MacGregor, 1974) and in the Gulf of Mexico (Baird et al., 1974). Continued or renewed sampling of these taxa at historical sites could provide a new benchmark for the extent to which PCBs and chlorinated pesticides are increasing or decreasing in EEZ pelagic ecosystem.

8.5 Tissues

There has been a considerable inconsistency among programs in the selection of tissues for monitoring trends in PCBs and chlorinated pesticides in fish. The NPMP used whole juvenile fish whereas the NS&T Program uses only liver tissues, and others only muscle. This inter-organ disparity has made it impossible to compare some data sets without invoking some kind of normalization procedure (which has not been done). Normalizing procedures should be tested to see if there are ways of relating data among samples of whole fish, muscle, and liver tissue. Alternatively, concerned agencies should be consulted in the future so that the most useful tissue or substrates are analyzed at some sites on a continuing basis.

8.6 Summary of Needs

In summary, several actions are needed to more adequately assess the geographic distribution, temporal trends, and significance of PCB and pesticide contamination in U.S. coastal fish and shellfish. On a national basis, monitoring should be continued for PCBs, DDE, the chlordanes and possibly dieldrin. Toxaphene, endosulfan, PCP, DCPA, endrin, kelthane, and kepone should be surveyed nationally at least once to establish if there is any current cause for concern. Other pesticides such as aldrin, methoxychlor, heptachlor, heptachlor epoxide, mirex, HCB, and lindane do not need to be monitored frequently on a national scale. Additional monitoring focus should be placed on edible tissues of larger coastal predatory fish, such as striped bass and blue fish. Organisms from the Atlantic coast of Florida, Hawaiian waters, several areas in Alaska, and commercial organisms from U.S. high seas fisheries should be checked for PCB and pesticide trends. A nationally centralized and easily accessible PCB and pesticide data base should be completed and used to receive and process new data from local, state, and federal programs. A comparison of contamination trends among inland, estuarine, coastal, and oceanic fish and shellfish would help provide additional perspective on identifying resources and chemicals of concern and help set priorities for additional cleanup and mitigation.

Finally, additional tools need to be developed to help interpret monitoring data. It is possible correlations between fish and bivalve data would allow prediction of contaminant levels in fish from results of bivalve monitoring programs. Determination of pesticide ratios among organs of several fish species would allow for more realistic seafood risk assessments. Also, research is clearly needed to identify tissue concentrations that are in fact hazardous to fish and shellfish.

The great magnitude of past sampling is evident. With judicious selection of sites and species, it is possible for future programs to extend trends already established. It is already possible to extend trends from a subset of these chemicals into the immediate future on a national and large-scale regional basis. The upsurge of local and regional monitoring since the late 1970s deserves additional careful review since these programs may serve to enhance the interpretation of spatial and temporal trends at the national level. Continued review of both existing and new data will help determine the best sampling strategies for extending trends at specific sites.

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APPENDIX A

COMMON AND SCIENTIFIC NAMES OF ORGANISMS CITED

Common Name	Scientific Name
alewife	*Alosa pseudoharengus*
anchovy, bay	*Anchoa mitchilli*
anchovy, silverside	unknown
bass, barred sand	*Paralabrax nebulifer*
bass, striped	*Morone saxatilis*
bluefish	*Pomatomus saltatrix*
bonito, Pacific	*Sarda chiliensis*
bowfin	*Amia calva*
clam, Asiatic	*Corbicula fluminea*
clam, hard	*Mercenaria mercenaria*
clam, soft	*Mya arenaria*
crab, blue	*Callinectes sapidus*
croaker, Atlantic	*Micropogonius undulatus*
croaker, white	*Genyonemus lineatus*
cunner	*Tautogolabrus adspersus*
eel, American	*Anguilla rostrata*
flounder, arrowtooth	*Atheresthes stomias*
flounder, broad	*Paralichthys squamilentus*
flounder, dusky	*Syacium papillosum*
flounder, eyed	*Bothus ocellatus*
flounder, fourspot	*Paralichthys oblongus*
flounder, fringed	*Etropus crossotus*
flounder, gulf	*Paralichthys albigutta*
flounder, shoal	*Syacium gunteri*
flounder, southern	*Paralichthys dentatus*
flounder, starry	*Platichthys stellatus*
flounder, summer	*Paralichthys dentatus*
flounder, yellowtail	*Limanda ferruginea*
flounder, window pane	*Scophthalmus aquosus*
flounder, winter	*Pseudopleuronectes americanus*
flounder, witch	*Glyptocephalus cynoglossus*
gar, alligator	*Lepisosteus spatula*
goatfish (not specified)	Mullidae
halibut, California	*Paralichthys californicus*
halibut, Pacific	*Hippoglossus stenolepsis*
herring, Atlantic	*Clupea harengus harengus*
iao, (silverside)	*Pranesus insularium*

mackerel, king	Scomberomorus cavalla
menhaden, Atlantic	Brevoortia tyrannus
menhaden, gulf	Brevoortia patronus
milkfish	Elops affinis
mullet (not specified)	Mugilidae
mussel, bay	Mytilus edulis
mussel, coastal (California)	Mytilus californianus
mussel, northern horse	Modiolus modiolus
mussel, ribbed	Modiolus demissus
oyster, American (eastern)	Crassostrea virginica
oyster, Olympia oyster	Ostrea lurida
oyster, Pacific	Crassostrea gigas
perch, white	Morone americana
plaice, American	Hippoglossoides platessoides
rockfish (not named by species)	Sebastes spp.
sablefish	Anoplopoma fimbria
salmon	Oncorhynchus spp.
sanddab, longfin	Citharichthys xanthostigma
sanddab, Pacific	Citharichthys sordidus
sanddab, speckled	Citharichthys stigmaeus
sculpin, fringehead	unknown
shad, gizzard	Dorosoma cepedianum
shrimp, brown	Penaeus aztecus
smelt, delta	Hypomesus transpacificus
sole, bigmouth	Hippoglossina stomata
sole, Dover	Microstomus pacificus
sole, English	Parophrys vetulus
sole, flathead	Hippoglossoides elassodon sole,
gray (witch flounder)	Glyptocephalus cynoglossus
sole, petrale	Eopsetta jordani
sole, Rex	Glyptocephalus zachirus
sole, rock	Lepidopsetta bilineata
sole, sand	Psettichthys melanostictus
sole, slender	Lyopsetta exilis
sole, yellowfin	Limanda aspera
spot	Leiostomus xanthurus
sucker, spotted	Minytrema melanops
tarpon	Megalups atlanticus
trout, spotted sea	Cynoscion nebulosus
tuna, yellowfin	Thunnus albacares
turbot, C-P	Pleuronichthys coenosus
turbot, hornyhead	Pleuronichthys verticalis
whitefish (not specified)	unknown

APPENDIX B

ACRONYMS

BHC	benzene hexachloride
CA	California
CEMP	Cooperative Estuarine Monitoring Program
CSDOC	County Sanitation Districts of Orange County
CSDLA	County Sanitation Districts of Los Angeles County
CSMW	California State Mussel Watch
CT	Connecticut
DDE	dichloraphenyl dichloroethylene
DDT	dichlorophenyltrichloroethane
DEF	s,s,s-tributyl phosphorotrithioate (a cotton defoliant)
dw	dry weight
EEZ	Exclusive Economic Zone
EPA	Environmental Protection Agency
FDA	Federal Food and Drug Administration
FL	Florida
FW	freshwater
GA	Georgia
HCB	hexachlorobenzene
HCH	hexachlorocyclohexane
IL	Illinois
km	kilometer
LACSD	Los Angeles County Sanitation District
MA	Massachusetts
MD	Maryland
N	number of samples
NAS	National Academy of Sciences
NC	North Carolina
NC DEHC	North Carolina Department of Health and Conservation
NESDIS	National Environmental Satellite, Data & Information Service
NMFS	National Marine Fisheries Service
NOAA	National Oceanic and Atmospheric Administration
NPMP	National Pesticide Monitoring Program
NS&T	National Status and Trends
NSSP	National Shellfish Sanitation Program
NY	New York
NYDEC	New York Department of Environmental Conservation
OAD	Ocean Assessments Division

PCA	pentachloro-anisole
PCB	polychlorinated biphenyls
PCDD	polychlorinated dibenzodiorin
PCP	pentachlorophenol
ppb	parts per billion, ug/kg, or ng/g
ppm	parts per million, mg/kg, or ug/g
SC	South Carolina
SCCWRP	Southern California Coastal Water Resource Project
tDDT	total DDT
tPCB	total PCB
TX	Texas
USDA	United States Department of Agriculture
VIMS	Virginia Institute of Marine Sciences
VSHD	Virginia State Health Department
VA	Virginia
VIMS	Virginia Institute of Marine Sciences
VSHD	Virginia State Health Department
WA	Washington
ww	wet weight