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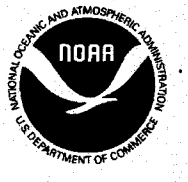

National Status and Trends Program

National Benthic Surveillance Project: Pacific Coast

Organic Chemical Contaminants Cycles I to VII (1984-90)

March 2000

U.S. DEPARTMENT OF COMMERCE
National Oceanic and Atmospheric Administration
National Marine Fisheries Service

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Organic Chemical Contaminants Cycles I to VII (1984-90)

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EXECUTIVE SUMMARY

As a component of National Oceanic and Atmospheric Administration's (NOAA) National Status and Trends (NS&T) Program, the National Benthic Surveillance Project (NBSP) monitored levels of contaminants in sediment and bottomfish and prevalences of pathological conditions in bottomfish at selected sites throughout the U.S. coastline. This memorandum summarizes and interprets the status and trends of the organic chemical contaminants for 50 sites in Alaska, Washington, Oregon, and California for the years 1984-1990 and includes results of 3213 chemical analyses of sediment, fish stomach contents, fish liver, and bile samples. Thirty-one of these sites were in or near urban centers and were selected to be as representative as possible of waste inputs from multiple sources, although the sites were not necessarily representative of entire embayments.

Uniform sampling protocols and state-of-the-art analytical methods have produced an extensive database, which includes detailed information on the distribution of selected polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), and chlorinated hydrocarbon insecticides (CHs) in surficial sediments and stomach contents of selected species of bottom-feeding fish. Coprostanol (COP) was determined in sediment samples as a marker compound of sewage. The PAHs were divided into two classes of compounds: lower molecular weight PAHs (LAHs), having two to three aromatic rings; and higher molecular weight PAHs (HAHs), having four to six aromatic rings. Livers were not analyzed for PAHs because they are of limited value due to the extensive biotransformation of these compounds to more polar metabolic products, most of which are readily excreted into the bile. Therefore, exposure of fish to PAHs was estimated by measuring fluorescent aromatic compounds (FACs) in bile and to PAHs in stomach contents. Concentrations of biliary FACs are reported as benzo[a]pyrene equivalents (FACs-H), representing the HAHs; or naphthalene equivalents (FACs-L), representing LAHs. Liver tissues of fish were analyzed for CHs, PCBs (tri- through decachlorobiphenyls), and several CHs including 2,2-bis(p-chlorophenyl)-1,1,1-trichloroethane and its metabolites (DDTs); chlordanes (a-chlordane and *trans*-nonachlor), dieldrin, and hexachlorobenzene (HCB).

The overall findings from the NBSP for the period 1984-90 indicated that the highest concentrations of most sediment-associated organic contaminants were present in the most highly urbanized areas, and that many of the organic contaminants were bioaccumulated by indigenous marine fish species. The highest levels of organic contaminants were found at sites in San Francisco Bay (Hunters Point, Oakland Estuary), Santa Monica Bay, San Pedro Bay (Los Angeles and Long Beach), San Diego Bay, and Puget Sound (Seattle and Tacoma).

Sampling Site Contamination

In San Francisco Bay, concentrations of PAHs in sediment from sites near the Hunters Point and Oakland Estuary sites were among the highest on the West Coast. These high concentrations were reflected in the levels of FACs in bile of starry flounder (*Platicthys stellatus*) and white

croaker (*Genyonemus lineatus*) from these sites. Concentrations of PCBs, DDTs, chlordanes and dieldrin in sediments from San Francisco Bay tended to be moderate compared to other urbanized bays; however, mean concentrations of these CHs in the livers of starry flounder were significantly higher at sites in San Francisco Bay compared to sites in Oregon and Alaska. White croaker from the Hunters Point and Oakland Estuary sites had concentrations of CHs that were similar to those found in this species from urbanized sites in Southern California, except for concentrations of DDTs which were all significantly higher in croaker from sites in the vicinity of Los Angeles.

Within the Southern California Bight, four of the five sites in Santa Monica Bay had concentrations of DDTs and COP (an indicator of human feces in waste water) in sediment that were among the highest on the West Coast. Sediment concentrations of PCBs and PAHs were moderate compared to other urbanized sites. However, mean concentrations of both DDTs and PCBs in the livers of hornyhead turbot (*Pleuronichthys verticalus*) and white croaker were among the highest found so far on the West Coast, whereas levels of biliary FACs were generally comparable to most urbanized sites.

In nearby San Pedro Bay, all four sites had, by a considerable margin, the highest concentrations of DDTs in sediment and white croaker found among the West Coast sites. With the exception of the Seal Beach site, levels of PCBs in sediment from the sites in this bay were also among the highest on the West Coast, and the levels of PCBs in the livers of white croaker from these sites were also among the highest for this species. Furthermore, the sediment and white croaker from the Long Beach site had high levels of chlordanes compared to most other sites. The levels of PAHs were relatively high in sediments from sites in San Pedro Bay, with levels at the Cerritos Channel site being among the highest along the coast, and these levels were reflected in the levels of FACs found in the bile of white croaker from these sites.

Sediments from two sites in San Diego Bay, the Twenty-eighth Street Pier site, and the north San Diego Bay site, had concentrations of PAHs and PCBs that tended to be higher than most of the other West Coast sites. These high sediment concentrations were also reflected in the high concentrations of FACs in bile and PCBs in livers of white croaker and barred sand bass (*Paralabrax nebulifer*) from these sites.

The site in Seattle's Elliott Bay had sediment concentrations of PAHs and PCBs that were also among the highest on the West Coast. This contamination was also reflected in the elevated concentrations of FACs in bile and PCBs in liver of English sole (*Pleuronichthys vetulus*) from this site. The site in Tacoma's Commencement Bay had concentrations of PCBs in sediment and livers of English sole that were significantly lower than those found at the Elliott Bay site. The levels of most of the other contaminants in sediment and English sole from the Commencement Bay site were not significantly different from those at the Elliott Bay site. The exceptions included sediment and liver tissue concentrations of HCB and concentrations of COP in sediments at the Commencement Bay site which were among the highest on the West Coast.

Sediment and fish [English sole, flathead sole (*Hippoglossoides elassadon*), starry flounder, and fourhorn sculpin (*Myoxocephalus quadricornis*)] samples from sites in Alaska generally had low

concentrations of PAHs, CHs, and their derivatives. The few exceptions were selected sites in the Bering Sea, such as Dutch Harbor, and the Oliktok Point site in the Beaufort Sea. For example, samples of sediment from the Dutch Harbor and Oliktok Point sites had moderate concentrations of HAHs, LAHs, and PCBs, whereas only English sole and flathead sole from Dutch Harbor had relatively high concentrations of FACs-L in bile. Of additional interest was the finding that starry flounder from two other sites in the Bering Sea had concentrations of FACs-H in bile that were significantly higher than for the reference site in Bodega Bay, California. Only flathead sole from the Dutch Harbor site had a mean concentration of CHs that was significantly higher than the reference site.

Statistical Assessments

At many of the sampling sites, concentrations of organic contaminants in sediment appeared to be reflected in the concentrations of these contaminants or their derivatives in fish from these sites. In order to test this hypothesis, Spearman rank correlations were performed for the organic contaminants in three types of samples—sediments, tissues, and stomach contents. The results demonstrated that when data from all fish species were combined, strong associations were found among concentrations of PCBs, chlordanes, DDTs, HCB and dieldrin in sediment and those same chemicals in fish livers, as well as between PAH levels in sediment and their corresponding FACs in fish bile. Concentrations of PCBs, HAHs, DDTs, PAHs, dieldrin, and chlordanes in stomach contents were also highly correlated with those in sediments, liver and/or bile.

Similar, but fewer or less significant, correlations were also found for the individual species. Correlations between levels of PAHs in sediment and/or stomach contents and levels of FACs in bile were found for flathead sole, English sole, white croaker, and barred sand bass. No other correlations were found for flathead sole, perhaps because most flathead sole were captured in relatively uncontaminated sites in Alaska. Among the remaining five species, significant correlations were found for levels of PCBs and chlordanes in sediment and/or stomach contents and levels of these chemicals in livers.

Another important aspect of the NBSP was to evaluate the presence of temporal trends of concentrations of contaminants in sediment and fish. Trends were evaluated by first performing Spearman rank correlations on concentrations of each class of contaminant in sediment and in stomach content and tissue for each fish species at each site. The results of these correlations were tested for consistency using meta-analysis. Only sites for which analyses had been conducted for at least four years over a five year span were used—12 sites met this criterion. Trend analyses were performed for HAHs, LAHs, PCBs, DDTs, dieldrin, and chlordanes. Concentrations of HCB were near or below the limits of detection and were not included in the trends analyses. The trend analyses demonstrated that, of the 72 possible trends, 38 showed no trend, 13 showed decreasing concentrations, and 21 had increasing concentrations. The highest number of increasing trends was among the PAHs, eight for HAHs and five for LAHs, followed by dieldrin (4), chlordanes (2), DDTs (1) and PCBs (2). Among the decreasing trends, chlordanes,

PCBs and DDTs each had three, with dieldrin having two and LAHs one.

Sites with positive trends for concentrations of PAHs were located in both nonurban and urban areas. All three of the nonurban, reference sites in the contiguous U.S. (Dana Point, Bodega Bay and Nisqually Reach) had significant increases in HAHs, LAHs, or both. The urban sites included Hunters Point and Southampton Shoal in San Francisco Bay, Long Beach and Outer Harbor in San Pablo Bay, and Commencement Bay in Puget Sound. The Coos Bay site also had increasing concentrations of chlordanes and DDTs. Positive trends for chlordanes were observed in the Southampton Shoal site, and increases in dieldrin concentrations were found for the Dana Point, South San Diego Bay, Coos Bay and Elliott Bay sites. The only sites with increasing concentrations of PCBs were both in Puget Sound—Commencement Bay and Elliott Bay.

The Dana Point site had the highest number of contaminants with decreasing trends, including chlordanes, DDTs, and PCBs. Decreasing trends for dieldrin and DDTs were also found at the Hunters Point site, and for PCBs and LAHs at the South San Diego Bay site. The only other decreasing trends were found for the following individual contaminant classes at single sites: dieldrin (Bodega Bay), DDTs (Hunters Point), chlordanes (Nisqually Reach), PCBs (San Pedro Bay Outer Harbor), and chlordanes (West Santa Monica Bay).

Because the NBSP was a national program in which samples of sediment and fish were collected and analyzed in very similar ways, it is possible to compare the results found on the West Coast with other regions of the U.S. Such a comparison shows that, with the exception of a few sites in Boston Harbor, levels of PAHs in sediments from urban sites on the north Atlantic Coast were similar to those from urban sites on the West Coast. Levels of PAHs in sediments from sites in the inner portions of Boston Harbor were highest in the U.S. Similarly, aside from a site in New Bedford Harbor, levels of PCBs in sediments from urban sites on the north Atlantic and West Coasts were similar. The New Bedford Harbor site had the highest sediment levels of PCBs so far found in the U.S. Again, with the exception of sites in the above-mentioned harbors, levels of PAHs and their derivatives and PCBs in winter flounder (*Pleuronectes americanus*) from these urbanized sites on the north Atlantic Coast were also comparable to those in fish from similar sites on the Pacific Coast. In contrast, levels of DDTs in sediment and fish from sites in the Los Angeles area were the highest found in the U.S., whereas levels of DDTs in sediment and fish from other sites in the north Atlantic and West Coasts were similar.

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PREFACE

The National Benthic Surveillance Project (NBSP) was initiated in 1984 by NOAA as a component of the National Status and Trends (NS&T) Program, which is designed to document the status of and to assess long-term changes in the environmental quality of the Nation's coastal and estuarine waters. The NBSP is a cooperative effort between the National Marine Fisheries Service and NOAA's National Ocean Services (NOS). The specific objectives of the NBSP were:

- Measurement of concentrations of chemical contaminants in sediment and in species of bottom-dwelling fish at selected sites in urban and nonurban embayments.
- Determination of the prevalences of diseases in these same fish species.
- Evaluation of temporal trends in these parameters.

This technical memorandum summarizes the results of the organic chemical contaminant analyses for the first seven years of the West Coast portion of NBSP. Although it is an overview of findings as well as a detailed presentation and treatment of these data, it is not meant to comprehensively review the related marine pollution literature. However, pertinent references are included in discussions of the most significant aspects.

INTRODUCTION

The National Benthic Surveillance Project (NBSP) compiled information on levels of chemical contaminants in the surface (top 2 cm) of bottom sediment in selected coastal and estuarine areas of the U.S., and on the levels of these contaminants (or their derivatives) in selected species of bottomfish (Brown et al. 1998, Varanasi et al. 1989b). The measurement of tissue concentrations of contaminants in marine fish bridges the gap between those contaminants that are associated with sediment, and those found in fish. Moreover, because fish are mobile, concentrations of contaminants in their tissues give an indication of contamination over a wider area than measuring contaminants in sediment alone. Because the intent of NBSP was to provide broad geographical coverage, the number of sites that could be sampled in an individual bay or estuary was necessarily limited. The urban sampling sites were specifically located in areas that integrate waste inputs from a number of sources, and were not directly adjacent to any known point sources of contaminants or established dredge disposal areas. Thus, the data from an individual sampling site are not intended to fully describe the environmental status of an entire embayment, but they do provide information on the status of representative sites.

The use of uniform sampling protocols and rigorous analytical techniques in the NBSP permitted the assembly of an extensive and robust database that documented levels of contaminants in sediments and in selected marine bottom-feeding fish on a nationwide basis. The establishment of such a Nationwide database is a fundamental requirement in documenting present conditions (status) and in establishing a scientifically meaningful baseline from which future changes in environmental quality (trends) can be measured.

This technical memorandum presents the results of the first seven years (1984-1990) of the West Coast component of the NBSP. Although a large body of data is available regarding contaminants in freshwater habitats of the U.S. [for example through the Environmental Protection Agency's (EPA) online computer database system, Staples et al. 1985], little information of this nature has been collected for marine and estuarine habitats (Zarba 1989). Therefore, these data serve to assess the contamination at a given site within a bay, which can indicate whether an embayment should be investigated more extensively in the future, either by the NBSP or by other suitable projects.

Project Rationale

Sediments are a known repository for a large number of organic contaminants, particularly those that are relatively insoluble in marine or estuarine water (Means et al. 1980, O'Connor and Huggett 1988). Fish have been shown to accumulate a variety of organic contaminants from sediments (Eadie et al. 1982, Gossett et al. 1983, Varanasi et al. 1985). Hence, it is important to analyze samples of sediments for organic contaminants, including polycyclic aromatic hydrocarbons (PAHs), chlorinated hydrocarbons (CHs) and coprostanol (COP). Total organic carbon and particle grain size was also determined in sediment samples. Selected organic contaminants, known to be or suspected of being toxic to marine organisms, are listed in Table 1 and are discussed in the next section. The urban sampling sites were specifically located in areas that integrated waste inputs from a number of sources,

and were not directly adjacent to any known point sources of contaminants or established dredge disposal areas.

Eight species of bottom-feeding fish listed in Table 2 were collected, with each species being captured at one or more reference sites. Fish were analyzed for the organic contaminants (or metabolites thereof) listed in Table 1, which are discussed in the next section. Stomach contents were analyzed for the same analytes as sediment. Liver tissues were also analyzed for the same CHs as were sediments with the exception of PAHs. Liver tissues were not analyzed for PAHs due to the extensive biotransformation of these compounds to more polar metabolic products, most of which are readily excreted into the bile (Varanasi et al. 1989a). Therefore, exposure of fish to PAHs was determined by measuring fluorescent aromatic compounds (FACs) in bile using high-pressure liquid chromatography with fluorescence detection (Krahn et al. 1984, 1986a). Concentrations of biliary FACs are reported as benzo[a]pyrene (FACs-H) or naphthalene (FACs-L) equivalents on a wet weight basis.

The same species of fish was used when evaluating and comparing exposures to contaminants at different locations, to avoid the confounding issue of species-specific differences. Interspecies differences in the uptake and fate of toxic chemicals by fish have been reported (Varanasi et al. 1986, 1987, 1989a) and could pose difficulties in extending interpretations of specific results to general situations.

Chemical analysis of stomach contents from the target bottom-feeding fish species was an additional means of assessing contaminant exposure. Although the manner and degree to which bottomfish are exposed to the various classes of sediment-associated contaminants are not well understood, it is generally held that diet can be an important route. Measurement of concentrations of contaminants in the fish tissues and in the benthic organisms comprising their diet can provide both a short term (diet) and a longer term (tissues) indication of exposure to contaminants. Due to the unevenness in distribution of sediment-associated contaminants in a given area, the contaminant levels found in benthic food species (fish or invertebrate) tend to be more representative of the overall degree of contamination for a given area than are the levels determined from a small number of sediment samples. The importance of determining the taxonomic distribution of stomach contents is discussed below in the section dealing with metabolites of PAHs.

Thus, a minimum of three environmental compartments were sampled at each site (sediment, stomach contents and liver/bile of at least one target species of bottom-dwelling fish). Liver and bile of a fish will be considered as a single environmental compartment because FACs in bile indicate exposure to PAHs, and CHs in liver reflect exposure to CHs.

Rationale for Selection of Organic Contaminants

The findings presented here are organized around the major classes of organic contaminants listed in Table 1, most of which are or are closely related to "Priority Pollutants" (Sittig 1980). Selection of these chemicals was based in part on recommendations of NOAA experts over the years (Lauenstein et al. 1993), and on the periodic revalidations of NOAA's environmental specialists, including those of the

Table 1. Organic compounds determined during NBSP Cycles I-VII (1984-90). Polycyclic aromatic hydrocarbons (PAHs) and chlorinated hydrocarbons (CHs) were determined in sediment and fish stomach contents, and CHs were also determined in fish livers. Fluorescent aromatic compounds (FACs) were determined only in bile; coprostanol was determined only in sediment.

PAHs	CHs
Low Molecular Weight (LAHs)	*Polychlorinated Biphenyls (PCBs)*
naphthalene§	dichlorobiphenyls†
2-methylnaphthalene	trichlorobiphenyls
1-methylnaphthalene	tetrachlorobiphenyls
biphenyl	pentachlorobiphenyls
2,6-dimethylnaphthalene	hexachlorobiphenyls
acenaphthylene£	heptachlorobiphenyls
acenaphthene	octachlorobiphenyls
2,3,5-trimethylnaphthalene£	nonachlorobiphenyls
fluorene	decachlorobiphenyl£
phenanthrene	
anthracene	*DDTs*
1-methylphenanthrene	o,p'-DDE
	p,p'-DDE
High Molecular Weight (HAHs)	o,p'-DDD
fluoranthene	p,p'-DDD + o,p'-DDT‡
pyrene	p,p'-DDT
benz[a]anthracene	
chrysene	*Chlordanes*
benzo[b+k]fluoranthenes£•	a-chlordane*
benzo[e]pyrene	trans-nonachlor*
benzo[a]pyrene	
perylene	
indeno[1,2,3-cd]pyrene£	
dibenz[a,h]anthracene	*Dieldrins*
benzo[g,h,i]perylene£	dieldrin
	aldrin
Fluorescent Aromatic Compounds (Biliary FACs)	
low molecular weight (FACs-L)	*Miscellaneous CHs*
high molecular weight (FACs-H)	lindane (g-BHC)
	mirex
Steroidal Sewage Tracer (Sediment)	hexachlorobenzene
coprostanol	heptachlor
	heptachlor epoxide

§ could not be determined accurately in stomach contents for Cycle V
£ determined beginning in Cycle III; not available for Cycles I and II
• reported separately beginning in Cycle V
† not determined after Cycle II
‡ reported separately beginning in Cycle IV
* included in "chlordanes" totals throughout this report

Table 2. Total numbers of chemical analyses of sediment, fish stomach contents, fish liver, and fish bile samples performed during NBSP Cycles I-VII (1984-90; 3213 analyses total).

Sediment analyses[1,2,3]: 417

Fish analyses

Species	Stomach contents[1,2]	Liver[1]	Bile	Total
Fourhorn sculpin	4	11	10	25
Flathead sole	10	31	185	226
English sole	25	69	365	459
Starry flounder	34	92	445	571
White croaker	53	156	702	911
Hornyhead turbot	21	56	228	305
Barred sandbass	16	38	145	199
Black croaker	6	16	78	100
Total	169	469	2158	2796

[1]Includes duplicate analyses.
[2]Numbers of analyses for individual PAHs and CHs may differ slightly from total number of analyses for technical or analytical reasons.
[3]Data for clay/silt ratio and total organic carbon (TOC) were not available for all samples.

Environmental Conservation Division. Also included is a natural steroid, coprostanol (COP), which is a sewage tracer because of its origin in human intestines. The principal classes of the organic contaminants listed in Table 1 are further described below.

PAHs

The PAHs are widely distributed throughout the marine environment and commonly occur in sediments in urban coastal and estuarine areas. They are composed of fused benzene rings, with or without alkyl substituents (e.g., methyl groups). In this study the general class of PAHs is divided into two subclasses:

- lower molecular weight PAHs having two to three aromatic rings (LAHs)
- higher molecular weight PAHs having four to six aromatic rings (HAHs)

LAHs

Relative to the HAHs, the LAHs are more volatile and water-soluble, and moreover, tend to be more readily taken up and metabolized by fish, and the metabolites excreted. The LAHs also are generally known for their acute toxicity. Sources of LAHs include all fossil fuels and discharges of incomplete combustion of them, as well as crude oil (Clark and Brown 1977).

HAHs

In contrast to the LAHs, the HAHs are less water soluble and generally more tightly sorbed to sediment. HAHs are present in crude oil, fossil fuels and in combustion residue (e.g., soot) from all incomplete combustion processes, including natural processes such as forest fires. Because of efficient metabolic transformations, the LAHs and HAHs are generally not bioaccumulated *per se* to any great extent in fish (Meador et al. 1995). The HAHs are also known more for their chronic toxicity than acute toxicity. One of the more thoroughly studied HAHs, benzo[a]pyrene (BaP), has been shown to be carcinogenic in mammals and fish (Osborne and Crosby 1987, Hendricks et al. 1985).

Metabolites of PAHs

Fish possess a significant capability, primarily in the liver, to readily metabolize PAHs and related aromatic compounds to more polar products (metabolites) that are not detectable in customary PAH analytical procedures (Varanasi et al. 1982, 1989a). Instead, the PAH metabolites are determined by their fluorescence which often is increased relative to parent compound fluorescence.

The metabolites pass into the bile for excretion. Because the target fish species possess gall bladders, the bile can be readily collected for analytical separation of components by high performance liquid chromatography (HPLC) with fluorescence quantitation (Krahn, 1986a). This fluorescence is relatively diagnostic of the PAH metabolites, which are also termed “fluorescent aromatic compounds” (FACs).

CHs

Unlike the PAHs, levels of CHs in fish can be directly measured in tissues, because the CHs are more resistant to metabolic conversion than AHs (Bickel and Muehlebach 1980, Stein et al. 1984, Stein et al. 1987). The CHs listed in Table 1 were measured in both the stomach contents and the livers of the target fish as well as in sediments.

PCBs

Among the CHs, the PCBs are persistent contaminants of estuarine sediments and biota that are very widespread. Commercial formulations of PCBs are mixtures of individual chlorinated biphenyls (congeners) varying according to the numbers of chlorines and their ring positions on the biphenyl. Prior to the 1975 congressional ban on PCB manufacture, various mixtures of some 209 individual PCBs were used extensively in electrical transformers, capacitors, paints, waxes, inks, dust control agents, paper and pesticides (Sittig 1981).

The toxicity of PCBs in terrestrial and aquatic organisms has been the object of considerable research (e.g., Safe 1984, 1994). Adverse effects, including reduced growth, diminished reproductive potential, immunosuppression, and carcinogenesis, have been reported in a variety of species (Casillas et al. 1994, Johnson et al. 1998, Gilbertson 1989, Myers et al. 1994). The U.S. EPA has recommended a maximum 24-hour average exposure of 0.03 µg/L PCBs in seawater as the water quality standard. However, few studies with marine species have focused on effects in animals exposed to sediment-associated PCBs or to PCBs contaminating their diet. Thus, the point above which environmental quality is significantly compromised by sediment concentrations of PCBs is not well understood.

DDTs

The toxicity of waterborne DDT to diverse aquatic species has been well documented (Holden 1972). More recent findings indicate that DDTs are endocrine disrupters that cause reproductive impairment and sexual abnormalities (Colborn et al. 1993). However, little is known concerning (a) the toxicity and bioavailability of sediment-associated DDTs, (b) the toxic effects on fish of dietary intake of DDTs, or (c) the toxicological significance of the DDT degradation products, DDE and DDD, in sediments or tissues of aquatic or terrestrial organisms. Of note is Sittig's (1980) finding that DDE is much less toxic than DDT when administered orally to mice.

Hexachlorobenzene, Chlordanes, and Dieldrin

Of the remaining CHs in Table 1, HCB (which is chemically unique in that it possesses a benzene ring as well as chlorine atoms), the chlordanes (which were grouped for statistical purposes) and dieldrin were selected for presentation on the basis of their use as pesticides.

Changes in Analytes Determined During First Seven Years of Program

One problem encountered in monitoring programs is the lack of standard methods which can complicate interpretation of the results (Edmondson 1991), especially with regard to assessment of long-term trends; therefore, all concentrations of chemicals used in this report are based solely on trace chemicals that were analyzed for all seven cycles using the same methods. The list of organic chemicals analyzed in bottom sediment was expanded in 1986 (Cycle III) to add two additional LAHs (acenaphthene and 2,3,5-trimethylnaphthalene), three additional HAHs (benzo[b+k]fluoranthenes, indeno[1,2,3-cd]pyrene, and benzo[g,h,i]perylene), decachlorobiphenyl, and a number of individual PCB congeners. In addition, analysis of dichlorobiphenyls was dropped at this time. Naphthalene was not reported for stomach contents samples for 1988 (Cycle V) due to analytical problems. These changes in analytes determined as well as a few changes in analytes reported are noted in Table 1.

METHODS

Field Sampling Protocols

Samples of sediment and target fish tissues (livers, bile, and stomach contents) were collected along the Pacific Coast during the first seven years of the NBSP at the 50 sites shown in Figures 1 and 2 and listed in Table 3 (all sites were not sampled each year). Thirty-seven of these sites were located in or near urbanized embayments. The remaining 13 sites were located in nonurban areas, four of which were used as reference or comparison sites (shown in italics in Table 3). Alaska had 13 sites; Washington, 3 sites; Oregon, 3 sites; and there were 31 sites in California. The names of the West Coast NBSP sampling sites and the target fish are also listed in Table 3. Included in this table are site codes which are used throughout the text, tables, figures and appendices.

Sediments were collected at three stations per site, and three sediment grabs taken at each sampling station were composited for analysis. Up to 60 bottom-feeding fish per species were obtained by otter trawl at each site. Fish were collected in the spring and summer months using an otter trawl; individual tows lasted ca. 5 min and covered about 0.2 nautical miles (300 m). Fish were selected for analysis according to length and the tissues were excised aboard ship, and frozen at - 20°C. A portion of the stomach contents samples were composited according to species, cruise, and site, and preserved in 10% buffered formalin for taxonomy.

Analytical Procedures

Sediments were analyzed for selected polycyclic PAHs and CHs according to Krahn et al. (1988) and Sloan et al. (1993). Coprostanol was determined in sediment by the method of Krahn et al. (1989). All analyte concentrations for sediment and tissue samples are reported on a dry weight basis. The PAHs are reported as either LAHs or as HAHs (Table 1). Chlorinated hydrocarbons are divided into three groups of compounds: PCBs; 2,2-bis(p-chlorophenyl)-1,1,1-trichloroethane and its metabolites (DDTs); and chlordanes (a-chlordane and *trans*-nonachlor).

Total organic carbon in sediments was determined instrumentally with a carbon-hydrogen-nitrogen analyzer on samples treated by the procedure of Hedges and Stern (1984). Particle size determination on sediment samples differentiated the clay-silt component of the sediment (i.e., particles <63 micron in diameter) from the larger particles. Sediments were separated into the two fractions using standard wet seiving techniques derived from EPA methods (Plumb 1981). Total organic carbon and particle size analyses were done at the NMFS Southeast Fisheries Science Center laboratory.

Liver tissues were analyzed for the same CHs as were sediments, but not for PAHs because PAHs are extensively biotransformed to more polar metabolic products in the liver. Therefore, exposure of fish to PAHs was estimated by measuring FACs in bile using high-pressure liquid chromatography with fluorescence detection (Krahn et al. 1984, 1986a). Concentrations of biliary FACs are reported as

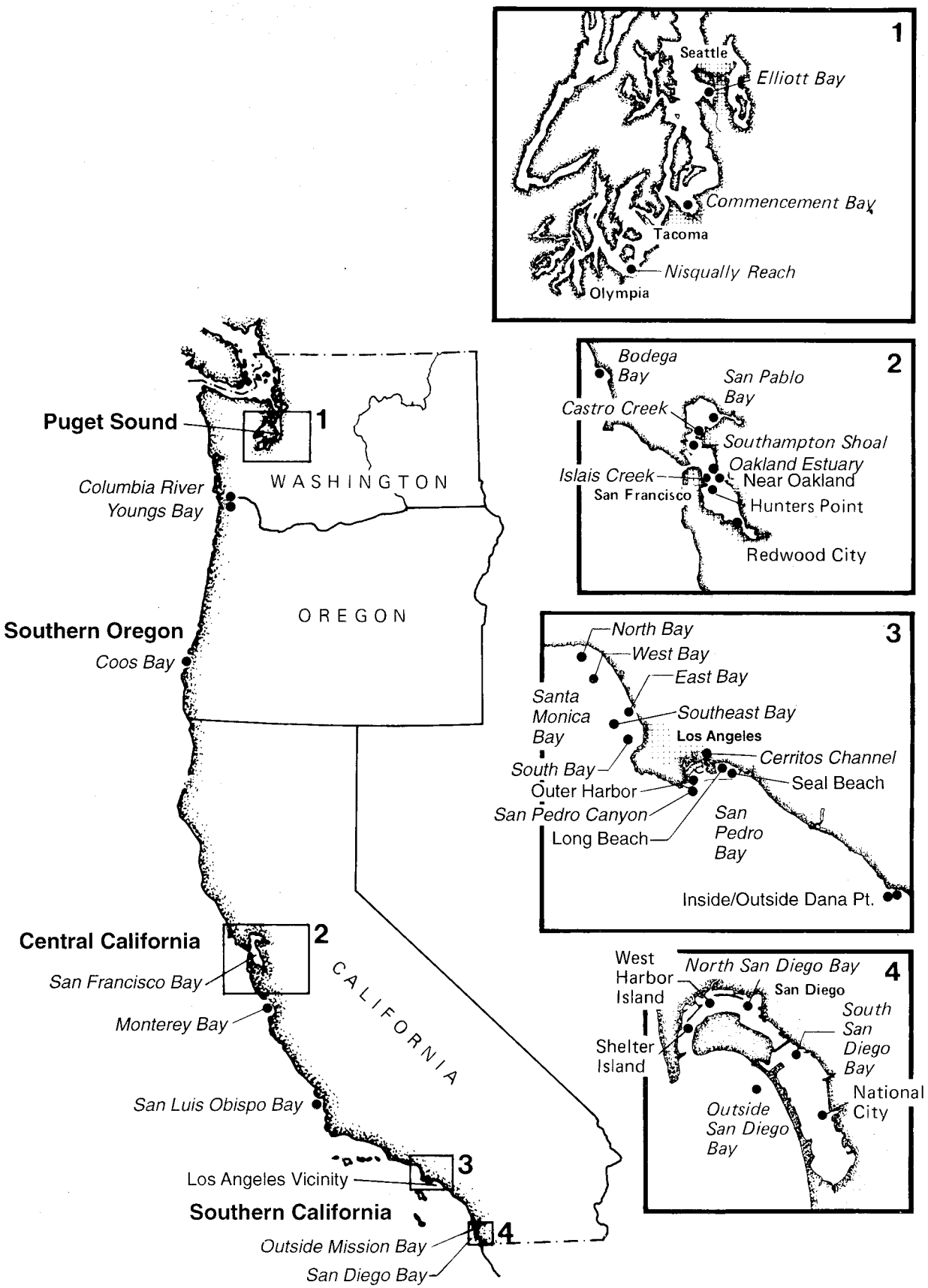


Figure 1. Map of the Pacific Coast of the contiguous United States showing the locations of sampling sites.

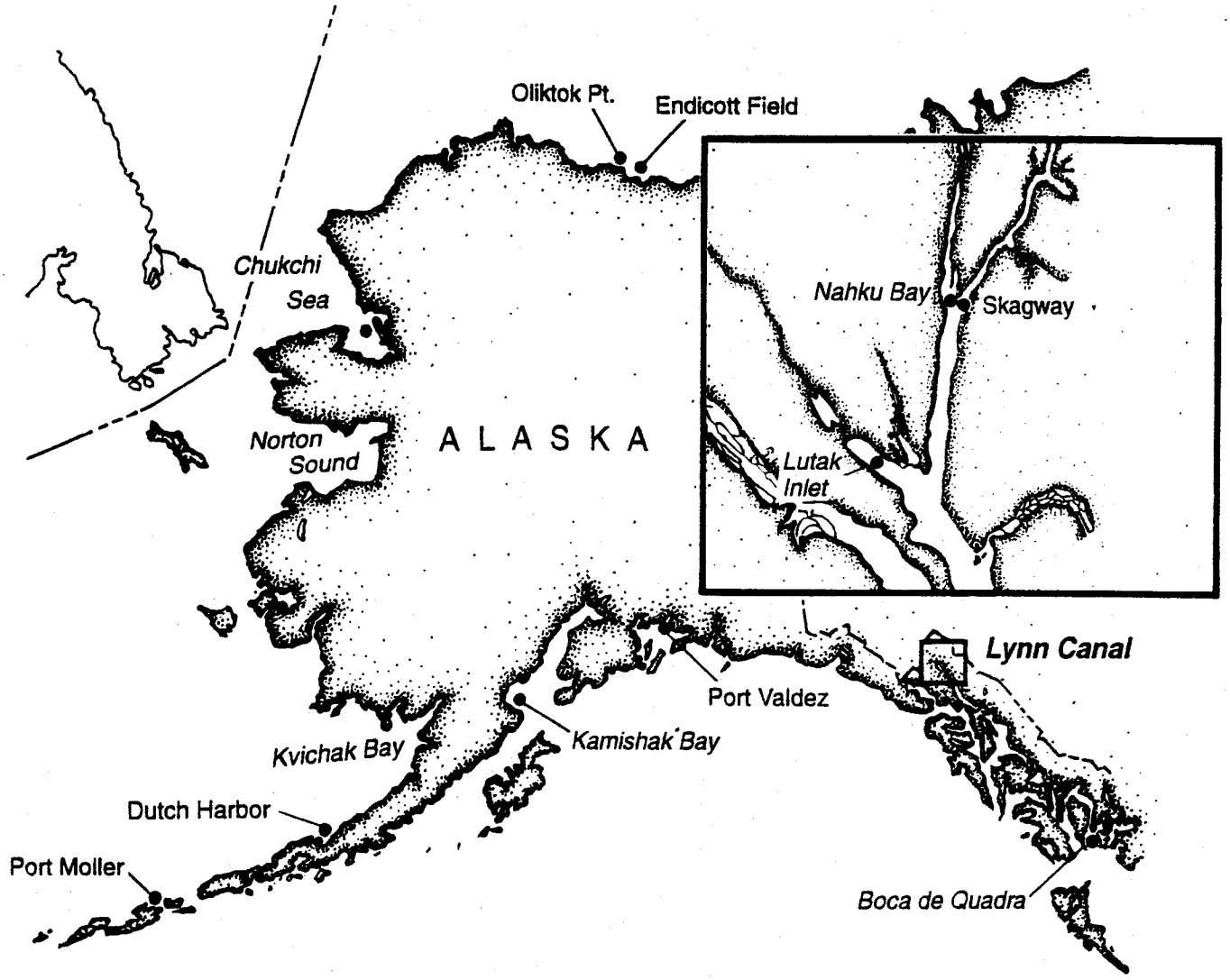


Figure 2. Map of Alaska showing locations of sampling sites.

Table 3A. California sites sampled 1984-90 for the National Benthic Surveillance Project (NBSP). Reference sites in *italics*.

Site name	Site location abbrev.	Lat.	Long.	Years sampled 1984	1985	1986	1987	1988	1989	1990	Target fish species*
south San Diego Bay	SDBTE	32°41.0'	117°08.2'	•	•	•	•	•	•		BSb
National City	SDBNC	32°40.1'	117°07.6'				•	•			BSb
north San Diego Bay	SDBNO	32°43.2'	117°11.3'			•	•	•			WhC, BSb, BlC
W Harbor Island	SDBHI	32°43.4'	117°12.7'			•	•	•	•		WhC
Shelter Island	SDBSI	32°42.5'	117°13.7'				•	•	•		BlC
outside San Diego Bay	SDBOU	32°39.2'	117°11.5'	•	•						HhT
outside Mission Bay**	MIBOU	32°47.3'	117°14.8'					•	•	•	BlC
*Dana Point, Outside***	DANOU	33°27.0'	117°41.0'	•	•	•	•	•	•	•	WhC, BSb, HhT
Dana Point, Inside	DANIH	33°27.5'	117°42.1'				•	•			WhC, BSb, HhT
San Pedro outer harbor	SPBOH	33°42.6'	118°15.4'		•	•	•	•	•	•	WhC
Long Beach	SPBLB	33°44.6'	118°10.6'		•	•			•	•	WhC
Seal Beach	SPBSB	33°44.1'	118°08.8'		•	•	•			•	WhC, HhT
San Pedro Canyon	SPBBR	33°42.0'	118°15.7'	•							HhT
Cerritos Channel	SPBCC	33°45.7'	118°15.3'				•	•	•	•	WhC
east Santa Monica Bay	SMBEA	33°53.2'	118°25.8'	•	•			•			HhT
SE Santa Monica Bay	SMBSE	33°52.5'	118°27.0'					•	•	•	WhC
N Santa Monica Bay	SMBNO	33°59.3'	118°35.9'						•		WhC, HhT
S Santa Monica Bay	SMBSO	33°47.5'	118°29.0'						•	•	HhT
west Santa Monica Bay	SMBWE	33°56.5'	118°33.3'		•		•	•	•	•	WhC, HhT
San Luis Obispo Bay	SLUOB	35°06.5'	120°45.0'					•			WhC
Monterey Bay	MONIH	36°37.6'	121°52.3'		•	•				•	EnS
Moss Landing**	MONML	36°48.0'	121°48.0'			•					EnS
San Pablo Bay	SFBSP	38°02.9'	122°17.6'	•	•	•	•	•		•	WhC, StF
Castro Creek	SFBCC	37°58.8'	122°24.8'				•	•			StF
Southampton Shoal	SFBSS	37°53.0'	122°24.0'	•	•	•	•	•	•	•	StF
Islais Creek Channel	SFBIC	37°44.8'	122°23.0'			•					WhC
near Oakland	SFBOA	37°47.5'	122°20.3'	•							WhC
Hunters Point	SFBHP	37°42.0'	122°21.5'	•	•	•	•	•	•	•	WhC, StF
Oakland Estuary	SFBOE	37°47.0'	122°21.0'				•	•	•	•	WhC, EnS
Redwood City	SFBRC	37°33.4'	122"11.2'			•	•	•			WhC
*Bodega Bay***	BODNO	38°18.3'	123°02.5'	•	•	•	•	•	•	•	EnS, WhC, StF

* BSb = barred sandbass, WhC = white croaker, BlC = black croaker, HhT = hornyhead turbot, EnS = English sole, and StF = starry flounder.

** These sites are considered nonurban.

Table 3B. Sites in Oregon, Washington, and Alaska sampled 1984-90 for the National Benthic Surveillance Project (NBSP). Reference sites in *italics*..

State	Site name	abbrev.	Site location Lat.	Long.	Years sampled 1984	1985	1986	1987	1988	1989	1990	Target fish species*
OR	Coos Bay	COONB	43°22.6'	124°12.5'	•	•	•		•	•	•	StF
OR	Columbia River estuary**	COLDS	46°13.2'	123°55.6'	•	•	•			•		StF
OR	Columbia R.Youngs Bay	COLYB	46°10.0'	123°50.0'								StF
WA	*Nisqually Reach***	PUGNR	47°06.7'	122°41.9'	•	•	•				•	EnS
WA	Commencement Bay	PUGCB	47°16.7'	122°25.0'	•	•	•			•	•	EnS
WA	Elliott Bay	PUGEB	47°35.5'	122°21.4'	•	•	•			•	•	EnS, FlS
AK	*Lutak Inlet***	LUTCR	59°18.7'	135°31.5'	•		•					FlS
AK	Nakhu Bay**	NAHES	59°28.1'	135°20.3'	•							FlS
AK	Skagway	SKASR	59°26.6'	135°19.7'			•					FlS
AK	Kamishak Bay**	GOAKB	59°11.7'	153°39.6'			•			•		FlS
AK	Port Valdez	PWSPV	61°06.5'	146°15.5'			•			•		FlS
AK	Boca de Quadra**	BOCBP	55°16.5'	130°32.5'			•					FlS
AK	Dutch Harbor	BERDH	53°54.0'	166°29.9'			•	•				FlS
AK	Port Moller	BERPM	56°06.4'	160°41.0'			•	•				FlS
AK	Kvichak Bay**	BERKB	58°41.0'	157°36.0'				•				FlS
AK	Norton Sound**	NORNO	64°20.0'	165° 33.0'					•			StF
AK	Chukchi Sea**	CHKRD	67°29.5'	164°02.8'				•	•			StF
AK	Prudhoe Bay	BEAPB	70°20.8'	147°58.0'		•	•					FhS
AK	Oliktok Point	BEAOP	70°30.4'	149°53.4'		•	•					FhS

* EnS = English sole, StF = starry flounder, FhS = fourhorn sculpin, and FlS = flathead sole.

** These sites are considered nonurban. Prudhoe Bay and Oliktok Point are nonurban but are associated with petroleum production.

benzo[a]pyrene equivalents (FACs-H), representing the HAHs; or naphthalene equivalents (FACs-L), representing LAHs as ng of FACs per g of bile. The excitation/emission wavelengths used were 290/335 nm (appropriate for naphthalene and structurally similar compounds) and the other set to 380/430 nm (appropriate for benzo[a]pyrene and structurally similar compounds). The concentrations of FACs in bile were determined using naphthalene and benzo[a]pyrene as external standards and converting the fluorescence response of bile to naphthalene or benzo[a]pyrene equivalents. Stomach contents samples were analyzed for the same suites of analytes as sediment samples except for COP.

For stomach content taxonomy, various prey items were sorted and identified by phylum. Each category of prey was damp-dried and weighed with a precision of 0.0001 g.

Statistical Analyses

Because the number of observations per site were different, the relative concentrations of contaminants in sediment and in fish tissue were compared statistically using the GT2 multiple comparison method (Gabriel 1978; Sokal and Rohlf 1981). The computation and plotting of a GT2 multiple comparison analysis is equivalent to performing a one-way analysis of variance (ANOVA) followed by a multiple-range test (Kleinbaum et al. 1988). In graphical displays of GT2 comparison intervals, those that do not overlap are significantly different ($p \leq 0.05$). Because environmental chemical concentrations are often log-normally distributed (Travis and Land 1990), the comparison intervals were calculated for contaminant concentrations (x) transformed using $\ln(x+1)$, resulting in comparison intervals that are asymmetric about the arithmetic mean.

The Spearman rank correlation method (Zar 1984), a bivariate non-parametric procedure, was used to examine relationships among chemical concentrations in sediments and fish. This method is preferred for situations in which an underlying normal distribution or linearity cannot be reasonably assumed. Spearman rank correlation was also used to analyze temporal trends in certain classes of contaminants over the period 1984-90. Only sites for which four years of data covering at least a five year span were selected. Temporal trends were evaluated for three environmental compartments: sediment and stomach contents and liver/bile of at least one target species of bottom-dwelling fish. Each tissue and sediment concentration was assumed to be a representative sample of the site for the year it was obtained and each concentration was used as an individual value in the trends analyses rather than using a mean value (per analyte per substrate) for the year (Gilbert 1987).

Meta-analysis techniques (Mullen and Rosenthal 1985 and Mullen 1989) were used to test for consistent trends in levels of contaminants in sediment and fish at each site. These techniques have been applied extensively to studies in the social sciences and medicine to test for consistency in experimental results across independent studies of relationships between risk factors and measurable effects. Meta analysis thereby integrates comparable studies into cumulative indications of association. The temporal trends computed in this study do not conform in the strictest sense to meta-analysis assumptions of independence as they are not derived from separate studies. However, it was assumed that the compartments analyzed were distinct enough to allow for synthesis into a single test of trend for each contaminant analyzed at a site. The significance levels for the Spearman rank correlations for pertinent

compartments (sediment, stomach contents and bile concentrations for PAHs and sediment, stomach contents and liver concentrations for all others contaminants) were transformed to Z-values, combined and then transformed back to a single p-value per Mullen and Rosenthal (1985) and Mullen (1989). The resultant significance level gives an indication of the consistency across compartments and statistical certainty with which a prevailing trend in concentration exists for a particular contaminant at a site.

RESULTS AND DISCUSSION

This report is based on 3213 chemical analyses of sediment, fish stomach contents, fish liver, and fish bile samples performed during NBSP west coast Cycles I-VII (1984-1990, Table 2). Mean concentrations of organic contaminants in sediment from each of the sampling sites is available for a one to seven year period, and these mean values will be compared over a coast-wide basis. However, due to potential species-specific differences among the target fish species, comparisons of mean concentrations of organic contaminants (or their derivatives) will be made only within the same species.

Particle Size Characteristics and Percent Total Organic Carbon

The percent silt-clay in sediment (i.e., the fraction of a sediment sample that contains the particles < 63 µm in diameter) for the West Coast NBSP sites ranged from 4.3% for Nisqually Reach in south Puget Sound to 99.0% for Port Valdez, Alaska (Fig 3; particle size and organic carbon were not determined for all samples). Within the NOAA Status and Trends Program, sediment samples with silt-clay values ≤ 20% are considered to be sandy and contaminant data from sandy sediments are not generally used because of complications encountered when normalizing contaminant data to percent silt-clay (NOAA 1988). Sediments from 37 of the 42 sites had percentage silt-clay values ≥ 20% and among these sites the percentage silt-clay values were generally not significantly different. The sites with values more than 40% silt-clay were primarily located in protected embayments sheltered from the open ocean. Nonurban sites had values ranging from 5% (Nisqually Reach in Puget Sound, Washington) to 40% (Dana Point, California). Sites with sediments having low percentage silt-clay values were usually located in open coastal bays, such as Bodega Bay or Monterey Bay, California or they were in areas of high riverine flows, such as Nisqually Reach (Washington) or the Columbia River estuary (Washington/ Oregon).

Concentrations of total organic carbon (TOC) tended to be highest in sediments from urban sites, even though some of the highest concentrations were detected in sediments from nonurban sites in Alaska—Boca de Quadra and Oliktok Point (Fig. 3). Fifteen urban sites had concentrations of TOC that were significantly higher than those from the reference site with the highest comparison interval (Dana Point): National City, south Bay, and north Bay in San Diego Bay; Long Beach and Outer Harbor in San Pedro Bay; Hunters Point, Oakland Estuary, Oakland, and Redwood City in San Francisco Bay; Coos Bay in southern Oregon; Youngs Bay in the Columbia River estuary; Elliott Bay and Commencement Bay in Puget Sound; and Dutch Harbor in Alaska. The high TOC values for sediments from Boca de Quadra and Oliktok Point were likely due to the presence of large amounts of glacial silt.

A correlation ($r_s = 0.68$, $p \leq 0.0001$) was found between the TOC values and the percent clay-silt of sediment samples. For example, six of the seven sediments with the lowest TOC values also had the lowest percent clay-silt values. Nevertheless, a few examples were found where TOC and clay-silt values for sediments were substantially different. This lack of agreement is presumably due to the differing geological and biological components in sediments among the sampling sites. For example, sediment from the Port Valdez, Alaska site was largely of glacial origin, thus it had a high percentage of

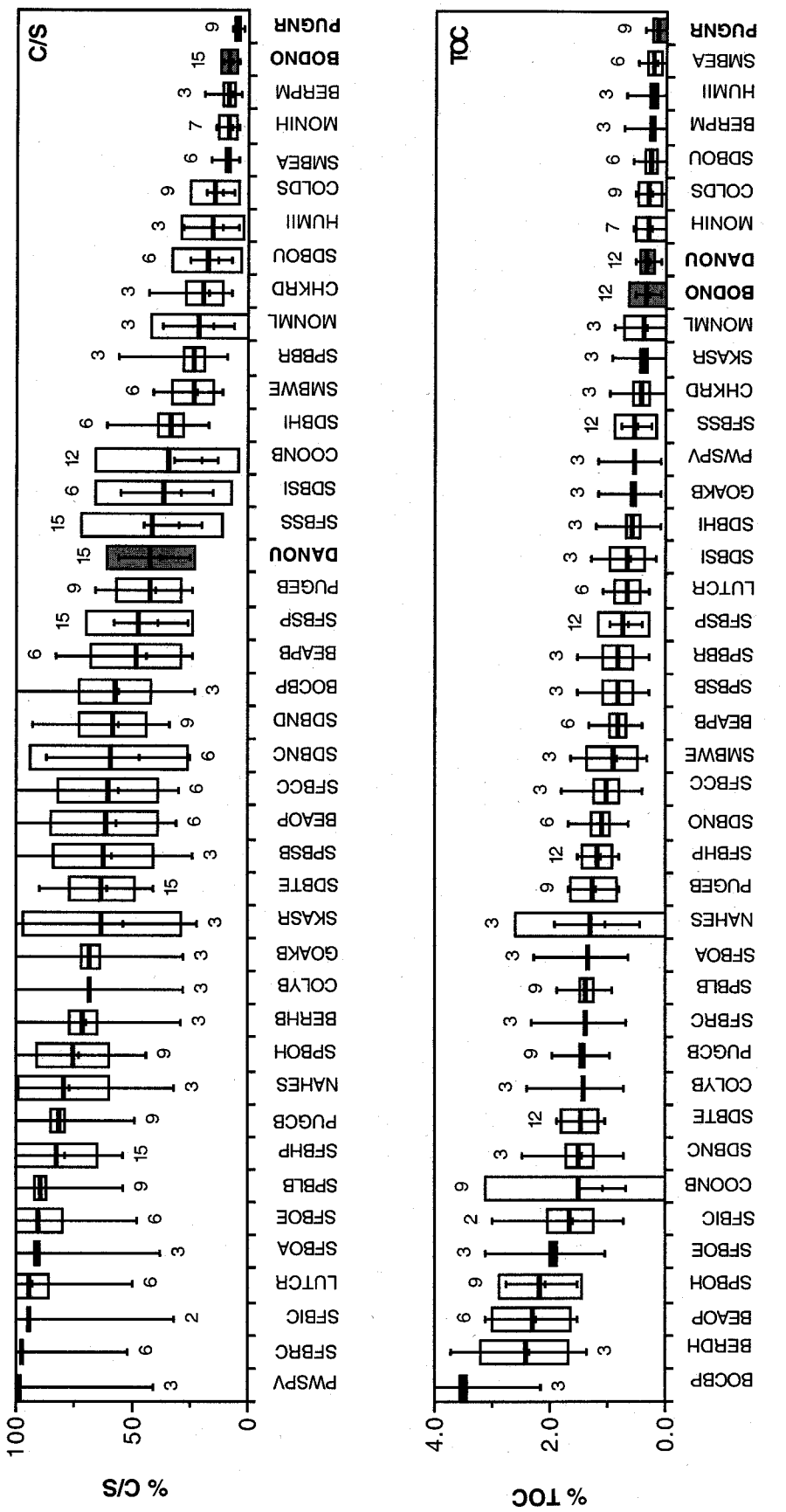


Figure 3: Percent clay/silt (C/S) and percent total organic carbon (TOC) in sediments (dry weight basis) for years 1984-1990.

Arithmetic mean [▬] ± 1 standard deviation box [ ], geometric mean [-] ± comparison interval [I], and reference sites [▬]. Sites are ranked in order of arithmetic mean concentrations along the x-axis (see Table 3 for site abbreviations; numbers for each site indicate sample size).

clay-silt and moderate TOC values; whereas sediment from the Boca de Quadra site, which had large amounts of forest debris, had a moderate percentage of clay-silt and high levels of TOC.

Organic Contaminants in Sediments

Figures 4-6 display the mean concentrations (arithmetic and geometric) and their standard deviations for organic contaminants in sediments from each site for years 1984-1990. Coprostanol was analyzed in sediments collected in years 1987-1990. The classes of organic contaminants included PAHs, PCBs, DDTs, chlordanes, dieldrin, HCB and coprostanol. For comparative purposes the concentrations of contaminants in sediments from each of the sampling sites were related to those at the Outside Dana Point reference site in Southern California. This site was chosen as the primary reference site for sediments because the mean percentage silt-clay value (42%) was more representative of urban sites than were the values for sediments from the other reference sites.

The results of the sediment chemistry analyses from this study will be primarily compared with results produced by NOAA's Mussel Watch Project (NOAA 1989), a component of the NS&T Program. Although a number of previous studies of organic contaminants in sediments from Pacific coastal areas have been published, these studies used analytical methods that are not strictly comparable to the methods used in the present study. The Mussel Watch Project collects and analyzes sediment associated with molluscs using analytical procedures comparable to those of NBSP, and collections were made over a somewhat similar time period (1986 to the present, NOAA 1989). Moreover, many of the Mussel Watch sites were located near NBSP sites. In addition, both projects participate in the same rigorous NS&T quality assurance (QA) program. Under the supervision of the National Institute of Standards and Technology (formerly the National Bureau of Standards), the performances of NS&T laboratories were carefully tested through annual intercalibration exercises with certified reference standards.

PAHs

Concentrations of PAHs (Table 1) measured in sediment are presented here primarily for the purpose of comparing these data with other studies which measure summed PAHs. However, for the purposes of this report, the emphasis will be placed on presenting the concentrations of LAHs and HAHs

LAHs and HAHs

The concentrations of LAHs and HAHs were near or below detection limits at most nonurban sites. Twenty-six urban sites had concentrations of LAHs and HAHs that were significantly higher than those from the Dana Point reference site (Fig 4). Twenty of these sites were in California: all five sites in San Diego Bay; all five sites in San Pedro Bay; three sites in Santa Monica Bay; all five sites in San Francisco Bay; and the two sites in San Pablo Bay. The other six sites were the Coos Bay site in OR; the Elliott Bay and Commencement Bay sites in Puget Sound, WA; the Endicott Field and Oliktok Point sites near Prudhoe Bay, Alaska; and the Dutch Harbor site in Alaska.

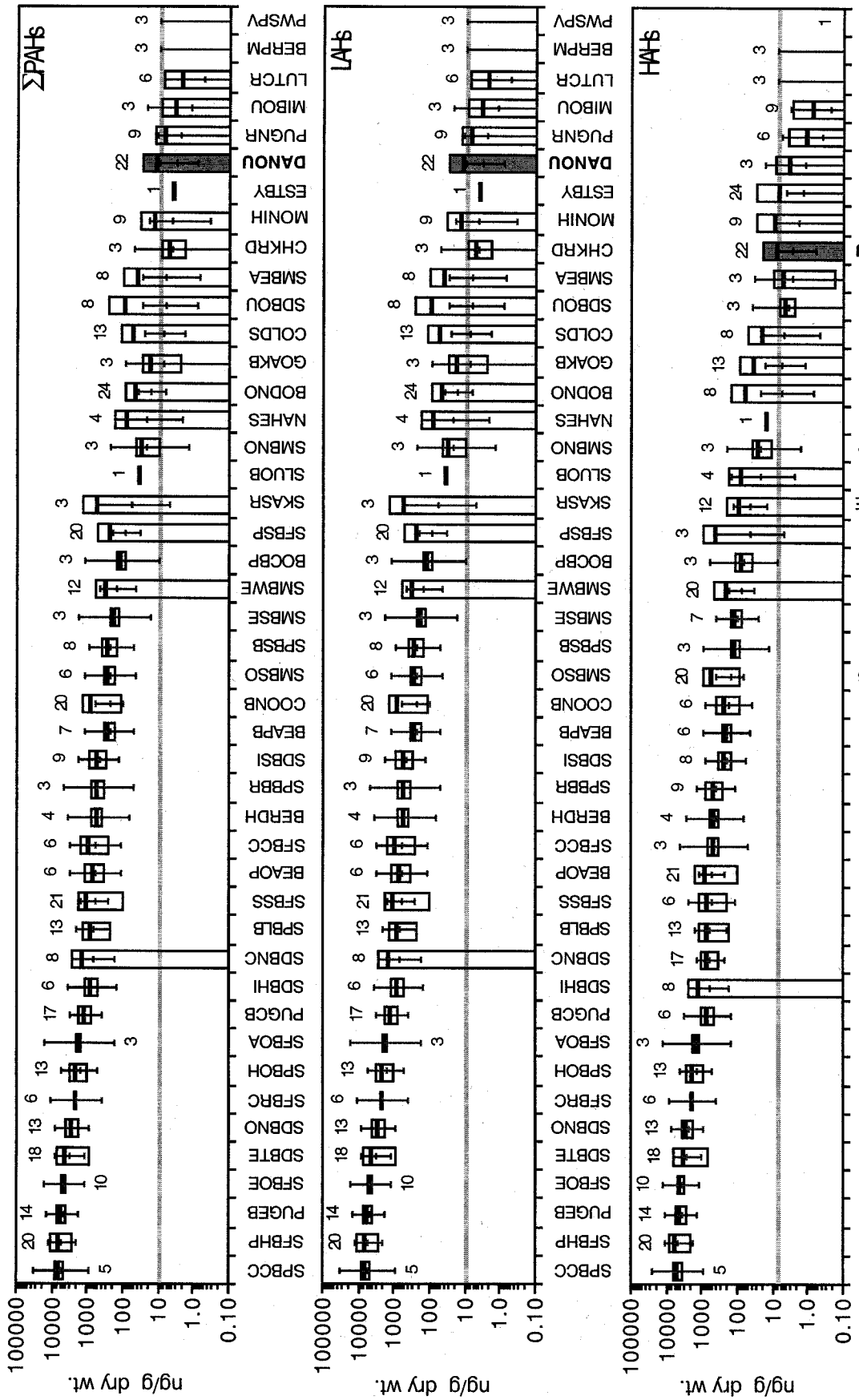


Figure 4. Total PAHs, LAHs, and HAHs in sediment (ESTBY is identity for Estero Bay which is north of San Luis Obispo).

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▒ …] for the comparison sites [■]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

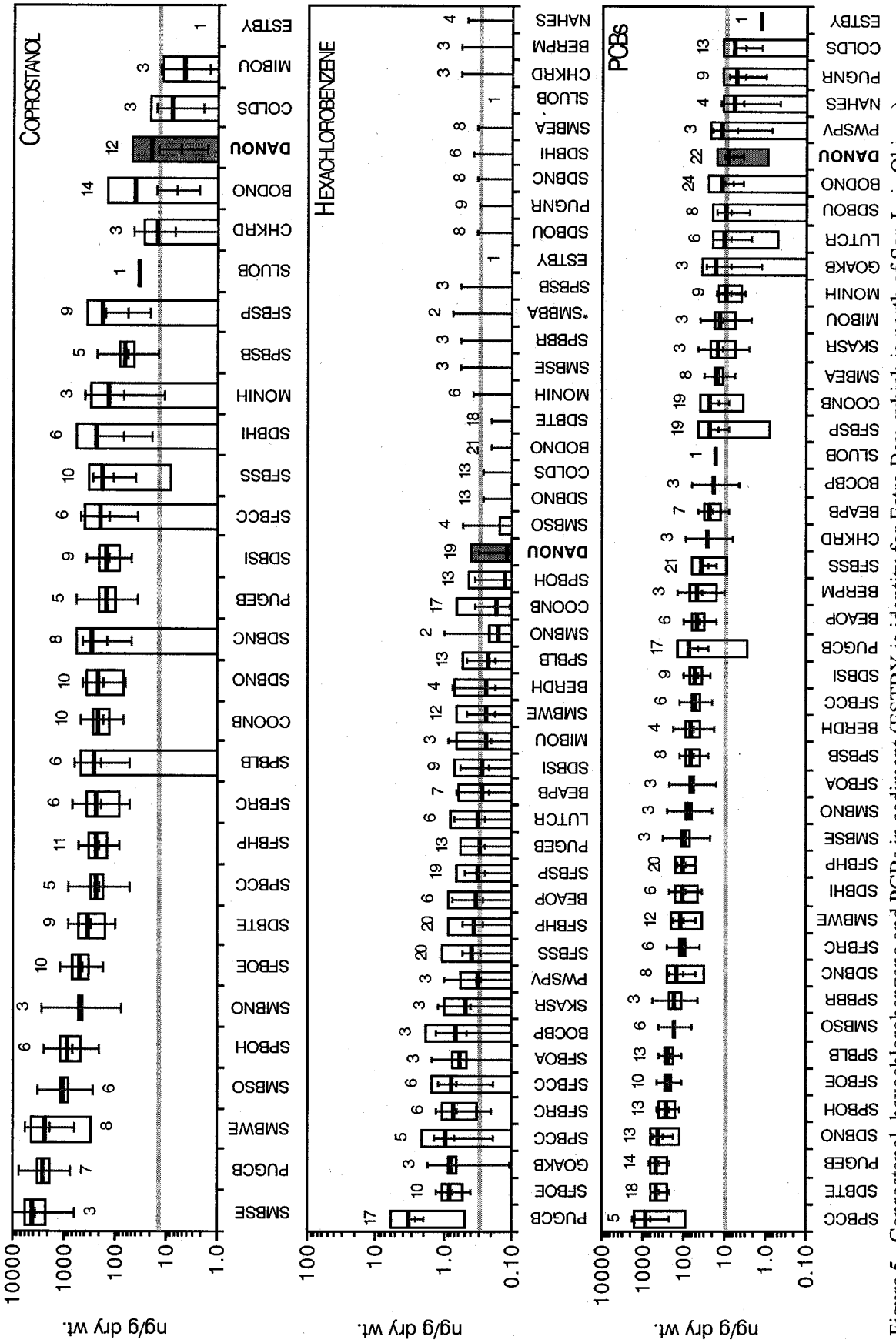


Figure 5. Coprostanol, hexachlorobenzene and PCBs in sediment (ESTBY is identity for Estro Bay which is north of San Luis Obispo).

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [▭], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [….▒….] for the comparison sites [▓]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

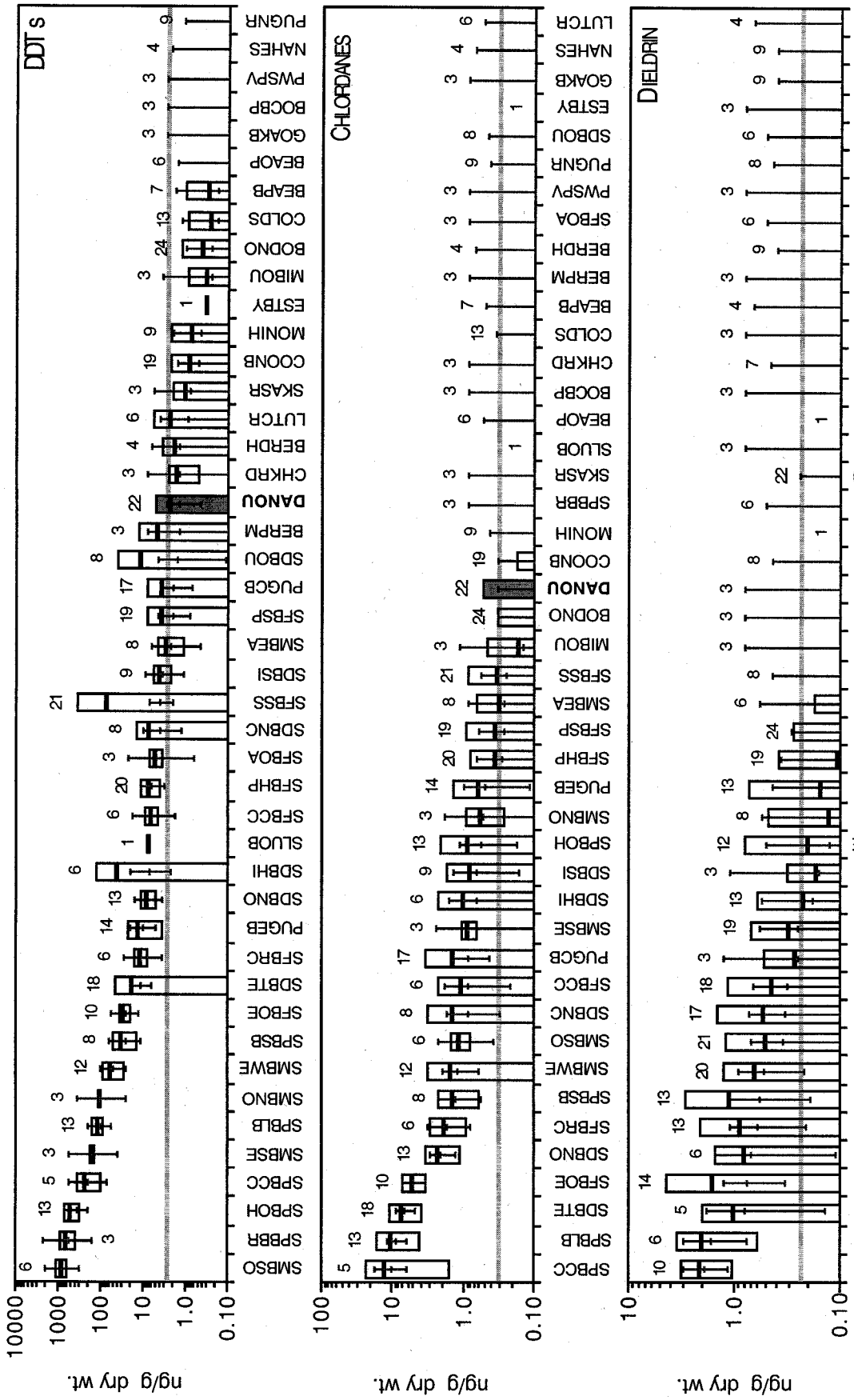


Figure 6. DDTs, chlordanes, and dieldrin in sediment (ESTBY is identity for Estero Bay which is north of San Luis Obispo).

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▒ …] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

Coprostanol

Coprostanol analyses were conducted on samples from 30 sites sampled during cycles 4-7 (1987-90). Twenty-one sites had concentrations of coprostanol that were significantly higher than those from the Dana Point reference site: all sites in San Diego Bay; the Outer Harbor, Long Beach and Cerritos Channel sites in San Pedro Bay; all the sites in Santa Monica Bay; all the sites in San Francisco Bay; the Coos Bay site, and both Commencement Bay and Elliott Bay sites in Puget Sound (Fig 5).

Hexachlorobenzene

Relatively low concentrations of HCB was detected in sediment samples (mean concentrations were less than 10 ng/g). Mean concentrations of HCB were significantly higher in sediments from only two sites (Commencement Bay and Oakland Estuary) compared to Dana Point sediment (Fig 5).

PCBs

PCBs were found in sediments from all of the West Coast sites, including the sites in Alaska. This contrasts with the PAHs (which, as noted above, were either not detected at nonurban sites or were at very low concentrations). Twenty-four sites had concentrations of total PCBs that were significantly higher than those from the Dana Point reference site (Fig 5). Significantly higher concentrations were found in sediment from all five sites San Diego Bay; all five sites in or near San Pedro Bay; four sites in west and south Santa Monica Bay; all five sites in San Francisco Bay; the Castro Creek site in San Pablo Bay; the sites in Commencement and Elliott Bays in Puget Sound, and the Oliktok Point and Dutch Harbor sites in Alaska.

DDTs

Fifteen sites had sediment with mean concentrations of DDTs that were significantly higher than that of the Dana Point reference site (Fig 6). With the exception of the site in Elliott Bay, all of these sites were in California. Within San Diego Bay, these sites included the North and South Bay sites. Significantly higher concentrations were also found in sediment from all sites in San Pedro Outer Harbor, four sites in Santa Monica Bay, and three sites in south San Francisco Bay. Sediment from all of the reference sites and sites in Alaska had concentrations of DDTs at or below the limits of detection (ca. 0.1 ng/g, dry weight).

Chlordanes

Of the 10 sites that had sediment concentrations of chlordanes that were significantly higher than those of the Dana Point reference site, nine were in California and one was in Commencement Bay. The sites in California included the North and South San Diego Bay sites; the Long Beach, Cerritos Channel, and Seal Beach sites in San Pedro Bay; the West and South Santa Monica Bay sites, and the Oakland Estuary and Redwood City sites in San Francisco Bay (Fig 6).

Dieldrin

Dieldrin was detected in sediment samples from 21 sites at concentrations < 10 ng/g dry weight (Fig 6). The only sites with mean sediment concentrations of dieldrin significantly higher than for Dana Point were Oakland Estuary and Redwood City in San Francisco Bay, and Elliott Bay in Puget Sound. Concentrations were below limits of detection at nonurban sites and all sites in Alaska.

Discussion of Sediment Chemistry Data

Because differences in analytical methods and quality control procedures can interfere with comparisons between different studies, comparisons of these findings with other similar types of monitoring/ reconnaissance studies that used analytical methods similar to ours, and were included in the NOAA Status and Trends Quality Assurance Program.

The concentrations of LAHs, HAHs, PCBs and DDTs in sediments collected in 1986 and 1987 from five Mussel Watch sites (NOAA 1991) located within 2 km of corresponding NBSP sites were very similar to those found in the present study. In San Diego Bay, sediment from the West Harbor Island NBSP site had mean concentrations of HAHs, PCBs and DDTs 720±320, 100±63, and 38±84 ng/g dry weight, respectively, compared to mean concentrations (n = 6) of 725±384, 96±80, and 9±8 ng/g dry weight, respectively for sediments from the Mussel Watch site located near Harbor Island. (Concentrations of contaminants for the Mussel Watch sites were reported as normalized to sediment particle size, therefore, for the purpose of this report, the values were converted to a dry weight basis.) Further north, in San Pedro Harbor, the concentrations of HAHs, PCBs and DDTs in sediment from the Outer Harbor NBSP site (1700±900, 260±130, and 500±210 ng/g, respectively) were similar, or slightly lower, than those found in sediment from a Mussel Watch site adjacent to a nearby fishing pier (1750±1300, 200±60, and 750±830 ng/g, respectively, n = 6). The concentrations of HAHs, PCBs and DDTs in sediments from two NBSP sites in San Francisco Bay—near Redwood City (1800±300, 100±28 and 11±4 ng/g, respectively) and Oakland (1400±320, 61±12, and 5±2 ng/g) were approximately the same as those for Mussel Watch sites located adjacent to the Dumbarton bridge (2000±540, 70±34, and 14±6, n = 6) and Emeryville (1600±910, 74±38, and 28±11 ng/g, n = 6), respectively. In Puget Sound, only one set of sites were close enough to justify a comparison of concentrations of HAHs, PCBs and DDTs: the Commencement Bay NBSP site (680±370, 70±67, and 3±4 ng/g, respectively) and the Mussel Watch site at Browns Point (860±360, 46±11, and 3±2 ng/g, n = 6). Again these concentrations were very similar. Overall, these results demonstrate that studies conducted in the same geographical area, using closely similar techniques, can indeed furnish highly synoptic results.

Sediment concentrations of DDTs reported by Brown et al. (1986) and by our laboratory (Malins et al. 1987) near our present Southeast Santa Monica Bay site were 100 ng/g and 120 ng/g, respectively. These values compare well with the 150±26 ng/g of this study, attesting to the relatively steady state of DDTs in Santa Monica Bay sediments.

Organic Contaminants in Fish

Because no single species of fish inhabits all of the West Coast sampling sites, nine bottom-feeding species were needed to characterize the exposure of marine fish to contaminants along the entire coast (Table 2). The results of chemical analyses of tissues for each fish species will be presented on a regional basis, and will include data for both urban and nonurban sites. The mean concentrations of FACs in bile, CHs in liver and CHs and PAHs in stomach contents are presented for each species, beginning with the species collected in the southernmost sites and proceeding in a northern direction.

The results of analyses of chemical contaminants in the stomach contents (i.e., food organisms) of fish will be presented to provide information on the types and concentrations of chemicals these fish are exposed to through their diet; they will also provide one of several measures of exposures to environmental contaminants that facilitate better interpretation of the overall findings. Although little scientific information is available on the relative proportions of chemical contaminants in stomach contents that can be absorbed and transported to tissues of the host fish, most of the chemicals measured in this study readily pass through gastrointestinal membranes (Gobas et al. 1988). However, a difficulty in interpreting chemical analyses for stomach contents is the fact that many of the target species are opportunistic feeders. Thus, the taxonomic composition of food organisms for a certain species will vary from site to site. Because some prey organisms, such as crustaceans, may be more effective than others (e.g., molluscs) at biotransformation of organic compounds (Varanasi et al. 1985), it is important that the differences in the taxonomic composition of food organisms for the same species at different sites also be determined. For example, if the host fish are from an area high in PAH contamination, this may not be reflected in the stomach contents if the latter consist mostly of crustaceans.

Barred Sand Bass

Specimens were taken from barred sand bass collected from three sites in San Diego Bay, one site outside of Mission Bay, and two sites from Dana Point. These latter two sites included one in the harbor and a site on the outer portion of the point which is a reference site. Samples of bile were taken from fish collected from all six sites, whereas bile, liver and stomach content samples were collected from two sites each in San Diego Bay and Dana Point.

The mean concentrations of FACs-H and FACs-L in the bile of barred sand bass from the National City site, the south San Diego Bay site, and the north San Diego Bay site were all significantly higher than that for bass from the Dana Point reference site (Fig 7). Although mean concentration of FACs-L in bass from the outside Mission Bay site were similar to those in bass from Dana Point reference site, however, concentrations of FACs-H from these Mission Bay bass were similar to those from San Diego Bay sites.

Mean concentrations of PCBs (Fig 8) and dieldrin (Fig 9) in the livers of barred sand bass from south San Diego Bay and National City were significantly higher than those for the Dana Point sites. However, the reverse was true for DDTs (Fig 8). Mean concentrations of DDTs in barred sand bass livers were approximately the same at both sites in San Diego Bay, but were significantly lower than

those for bass from the Dana Point sites. For HCB, the mean concentration in bass from the south San Diego Bay site was significantly lower than that for Dana Point bass, and the mean value for bass from the National City site was not significantly different from either of the other sites. All of these HCB values were quite low (≤ 10 ng/g), however. For chlordanes, only bass from the south San Diego Bay site had significantly higher liver concentrations compared to bass from the Dana Point sites (Fig 8).

Mean concentrations of LAHs in the stomach contents of barred sand bass from the two sites in San Diego Bay were not significantly different from those found in bass from Dana Point (Fig 10). However, the mean concentration of HAHs in bass from south San Diego Bay was significantly higher than for bass from Dana Point. Similarly, the mean concentration of PCBs in the stomach contents of barred sand bass from the south San Diego Bay site was significantly higher than that in bass from Dana Point, but the mean concentration in bass stomachs from the National City site was not significantly different from either of the other two sites (Fig 11). Conversely, no significant differences were found among the levels of DDTs, chlordanes, dieldrin, or HCB in stomach contents of bass from any of the three sites (Figs 11 and 12)—the mean concentrations of dieldrin and HCB were ≤ 10 ng/g.

Barred sand bass from Dana Point appear to have been exposed to higher levels of DDTs than would be expected for such a nonurban area with low concentrations of DDTs in sediments. However, the proximity of Dana Point to San Pedro Bay, where sediment levels of DDTs were among the highest measured (Fig 6, also Bascom 1982), may account for these unexpected levels in the stomach contents (Fig 11). For example, food organisms and/or fish containing DDTs may move from the San Pedro Bay area to Dana Point. Chemical and taxonomic analyses of the stomach contents from barred sand bass support this explanation (McCain et al. 1992). The stomach contents of barred sand bass from the Dana Point site had a mean concentration of DDTs at least twice that of sand bass from the San Diego Bay sites. The food organisms found in these stomach content samples from sand bass from the Dana Point site consisted of approximately equal proportions of fish (44%) and arthropods (52%). Both groups include several readily mobile species. Thus, it can be speculated that the elevated levels of DDTs in the livers of barred sand bass from the Dana Point site were due in part to consumption of some food organisms that were exposed to DDTs in areas away from this site (e.g., the vicinity of San Pedro Bay).

Although comparisons of chemical contaminants in fish in this report have focused on inter-site differences, it is also of interest to note the differences in bioaccumulation of contaminants by fish species captured during different seasons of the year. Most of the chemistry data in this report are from analyses of samples collected during June and July. However, in 1987, barred sand bass were collected from the South San Diego Bay site during February as well. No significant seasonal differences were found in the concentrations of PCBs and DDTs in liver and FACs in bile from barred sand bass (McCain et al. 1992).

Black Croaker

Mean concentrations of FACs-H in the bile of black croaker from the Shelter Island site and the North and South San Diego Bay sites were significantly higher than those for black croaker from the outside Mission Bay reference site (Fig 13). For FACs-L, a significant difference was also found in mean concentrations in croaker bile from the North and South San Diego sites compared to black croaker

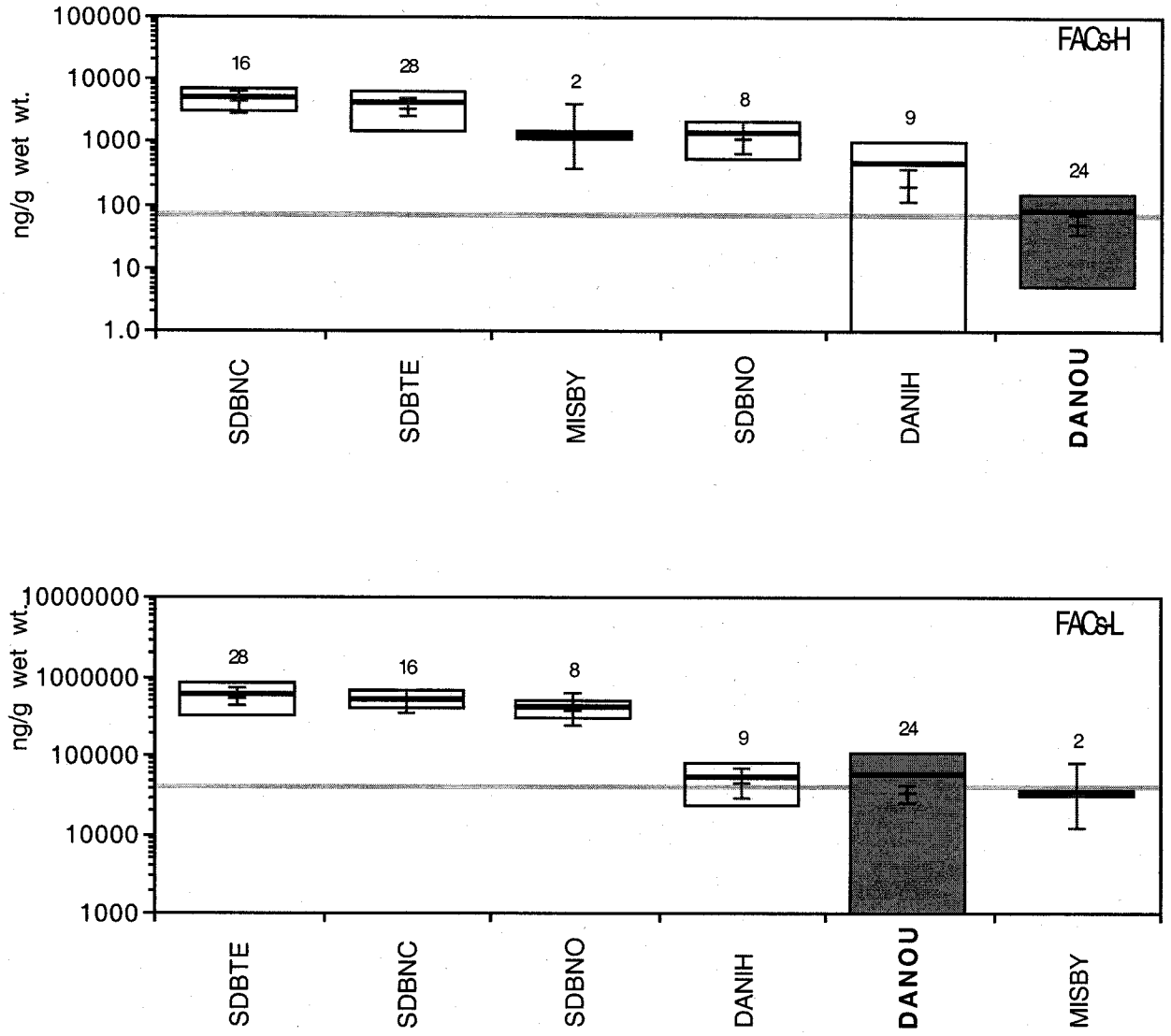


Figure 7. FACs-H and FACs-L in bile of barred sand bass. Only fish were collected from a site in Mission Bay (MISBY) and only bile was analyzed. MISBY is not listed in Table 3.

Arithmetic mean [▬] concentrations, (ng/g, wet weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▒▒ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

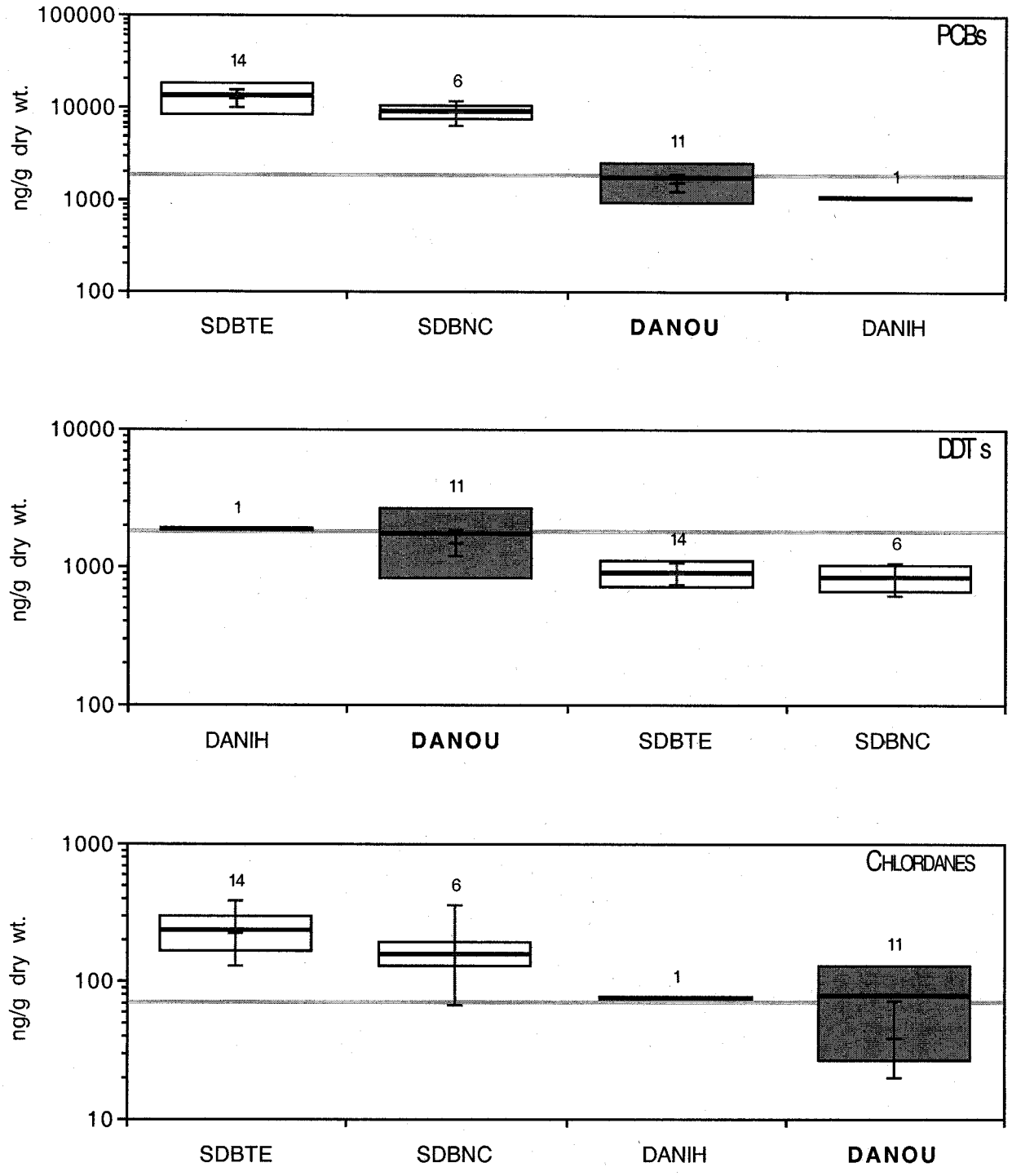


Figure 8. PCBs, DDTs, and chlordanes in livers of barred sand bass.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▬ …] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

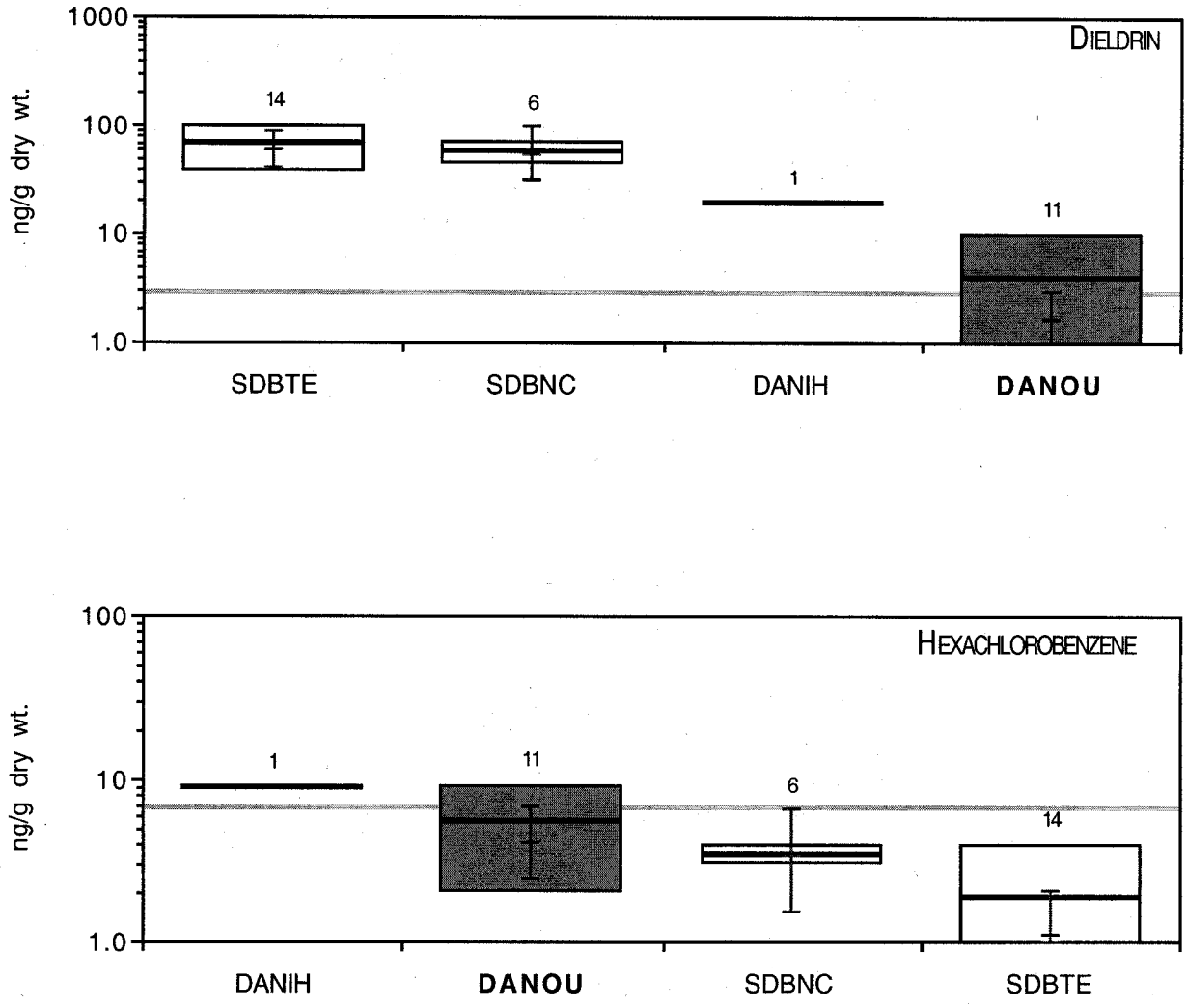


Figure 9. Dieldrin and hexachlorobenzene in livers of barred sand bass.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… … …] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

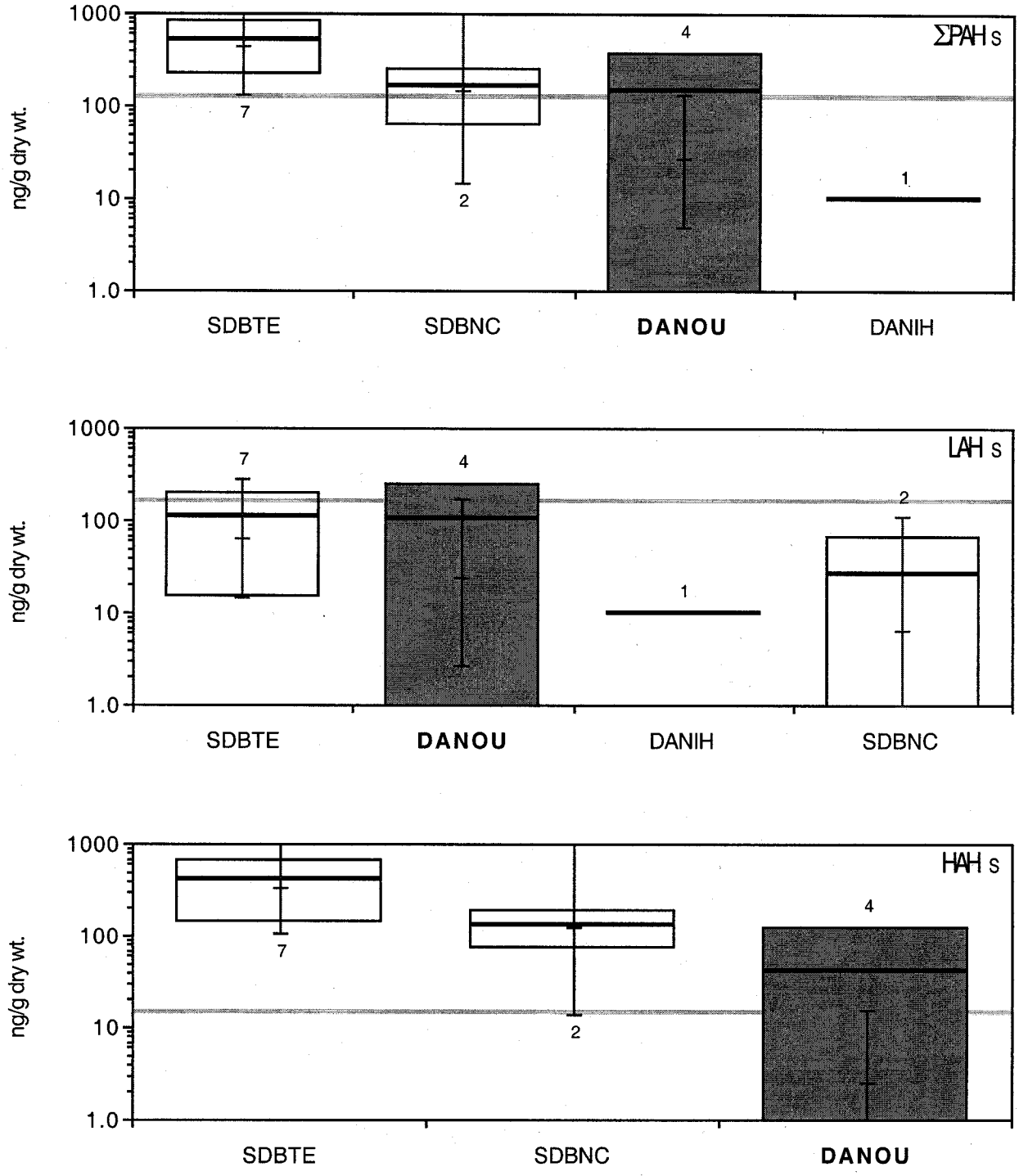


Figure 10. Total PAHs, LAHs, and HAHs in stomachs of barred sand bass.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▒▒ …] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

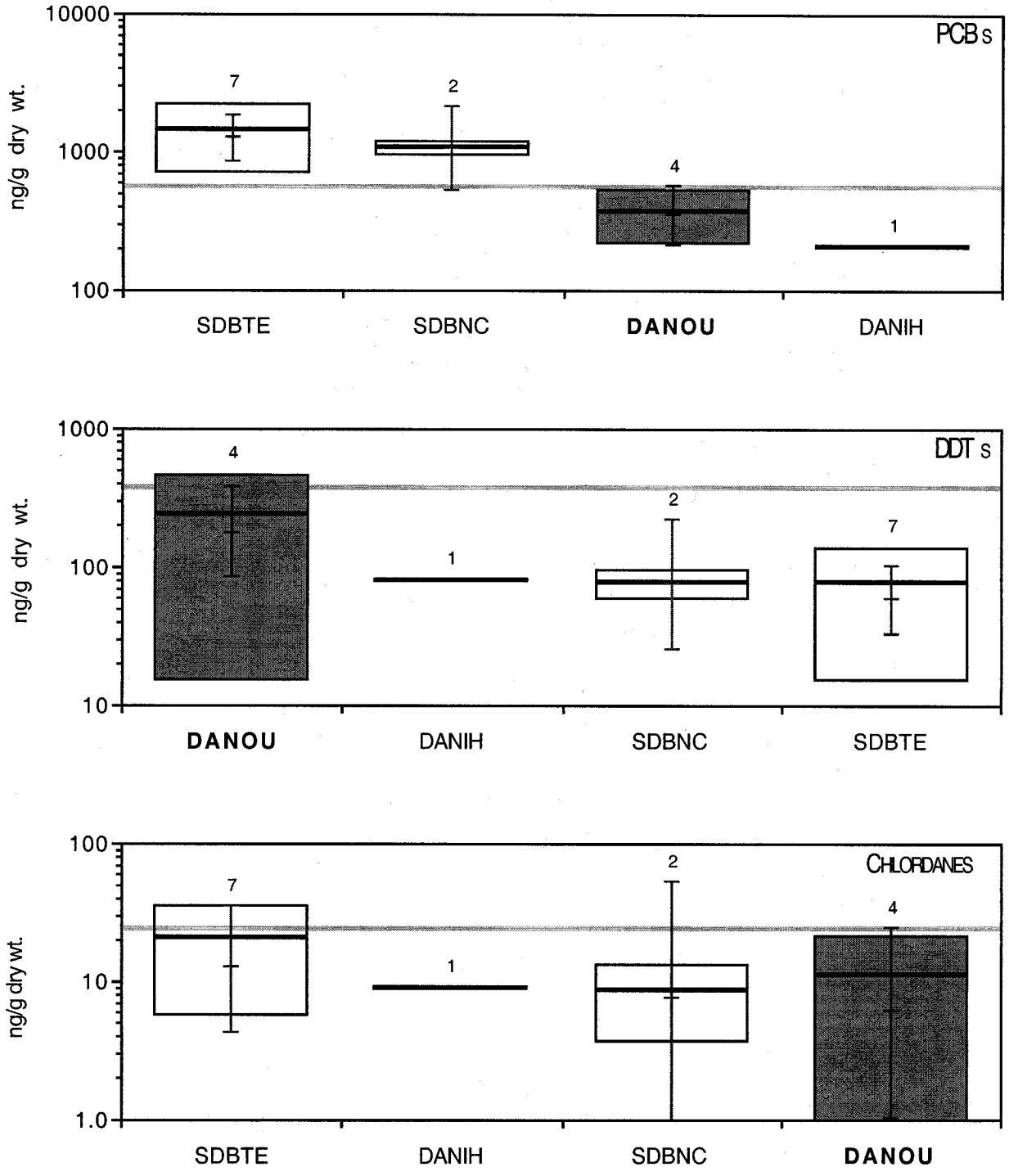


Figure 11. PCBs, DDTs, and chlordanes in stomachs of barred sand bass.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▒▒ …] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

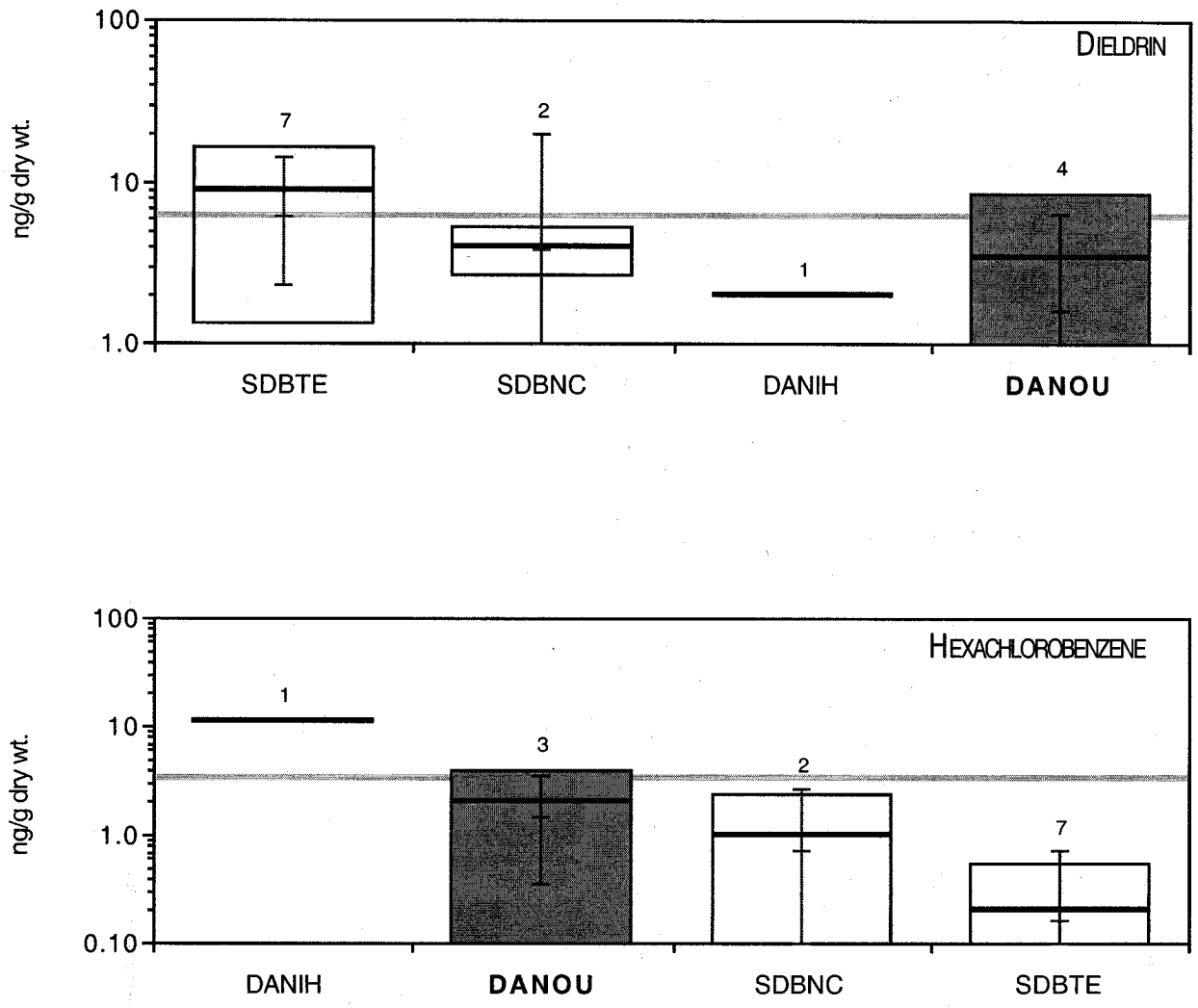


Figure 12. Dieldrin and hexachlorobenzene in stomachs of barred sand bass.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [▭], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ... ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

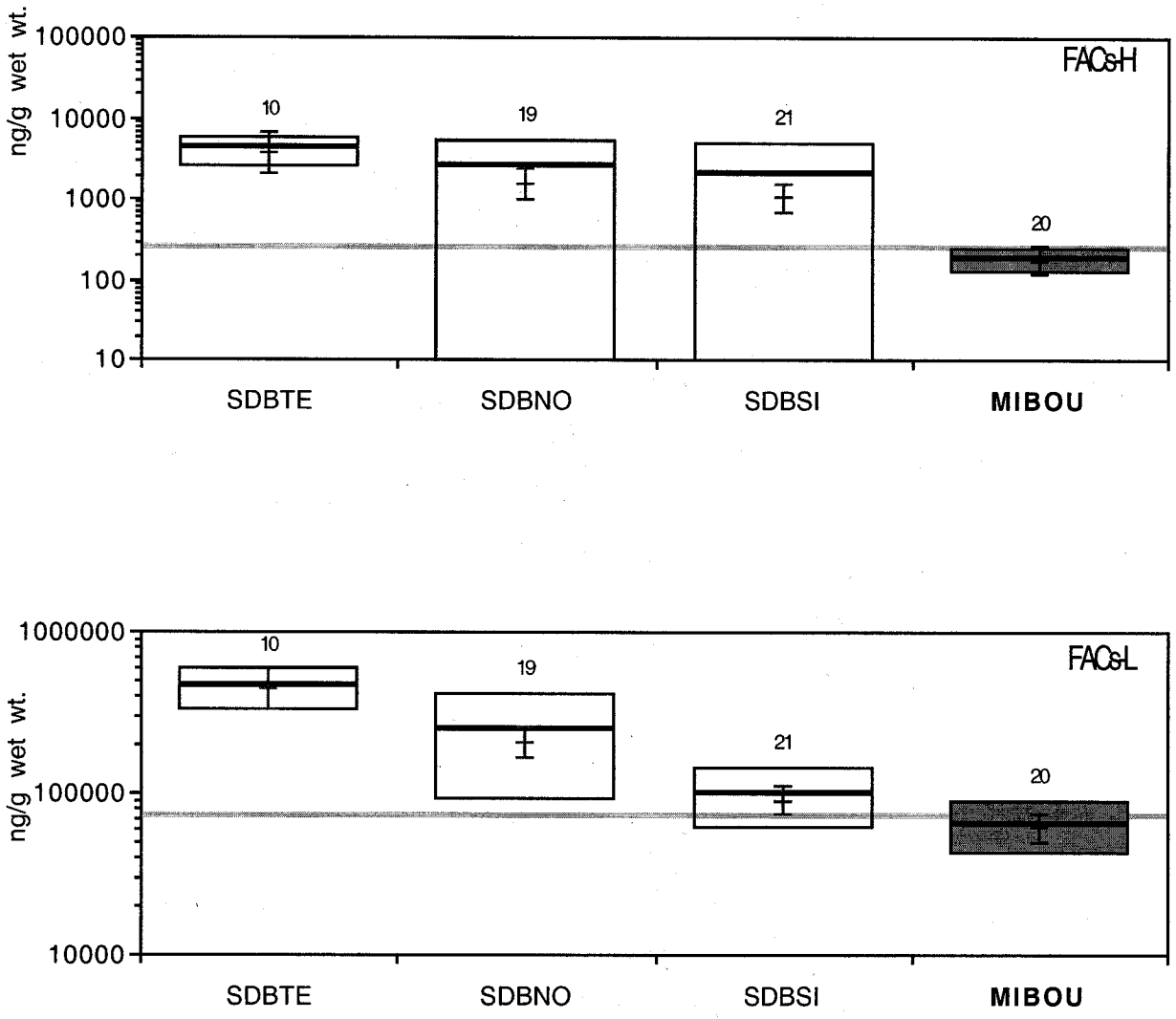


Figure 13. FACs-H and FACs-L in bile of black croaker.

Arithmetic mean [▬] concentrations, (ng/g, wet weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▬ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

from the outside Mission Bay site. The relatively high concentration of FACs-L in black croaker from the reference area may have been due to surface runoff entering Mission Bay (McCain et al. 1992).

Mean concentrations of PCBs, chlordanes, and dieldrin in black croaker livers from the San Diego Bay sites were comparable, and were significantly higher than those from the outside Mission Bay site (Figs. 14 and 15). Concentrations of DDTs were also significantly higher in black croaker from the North and South Bay sites, but not from the Shelter Island site, whereas concentrations of HCB were near or below the limits of detection at all sites.

Because black croaker from the North San Diego Bay site did not have sufficient stomach contents for chemical analysis and only one composite from the outside Mission Bay site was analyzed, statistical tests were not performed, and the data are not presented. Nevertheless, the data for the outside Mission Bay site provided useful information. Mean concentrations of PAHs were 300 ± 120 ng/g in the stomach contents of black croaker from the Shelter Island site, whereas no PAHs were detected in the stomach contents of black croaker from the outside Mission Bay reference site. These findings at the outside Mission Bay site are in contrast to the moderate concentrations of FACs-L measured in the bile of croaker from this site, suggesting that diet may not have been an important route of uptake of LAHs. Uptake through the water column may have been a more important mode of exposure.

Concentrations of PCBs in the stomach contents of black croaker from the Shelter Island site were 990 ± 380 ng/g, about twice that found from the outside Mission Bay reference site (410 ng/g), with a similar ratio for the DDTs at the two sites (99 ± 71 ng/g for Shelter Is vs. 54 ng/g for outside Mission Bay). Chlordanes were <15 ng/g, dieldrin was < 5 ng/g, and HCB was < 4 ng/g in the stomach contents of black croaker from all sites.

White Croaker

At all sites, except the north and south San Diego Bay sites, white croaker were collected during the summer; however, at these two San Diego Bay sites during 1987 and 1988, this species was also collected during the winter**.** Unless stated otherwise, the following results are for the summer sampling only.

Mean concentrations of FACs-H in the bile of white croaker were highest at the Cerritos Channel site in San Pedro Bay, the north San Diego Bay site, and the Oakland Estuary site in San Francisco Bay (Fig 16). Values of FACs-H from the other urban sites were also significantly higher than those for the Dana Point reference site, with the exception of the near Oakland and Redwood City sites in San Francisco Bay, San Luis Obispo site, and the west Santa Monica Bay site. A similar pattern of concentrations was observed for FACs-L in the bile of white croaker, except the two sites in San Pablo Bay also had concentrations not significantly different from the reference site.

As was mentioned above, samples of white croaker were also collected at the North San Diego Bay site, during the winter (February) of 1987 and 1988. A significant seasonal difference was found between the mean concentration of FACs-L in the bile of white croaker collected from this site in the summer (96.0 ± 43.0 µg/g, wet weight, n = 15) and in the winter (220.0 ± 25.0 µg/g, wet weight, n = 5). The importance of the season of capture as a determining factor of the levels of FACs in white

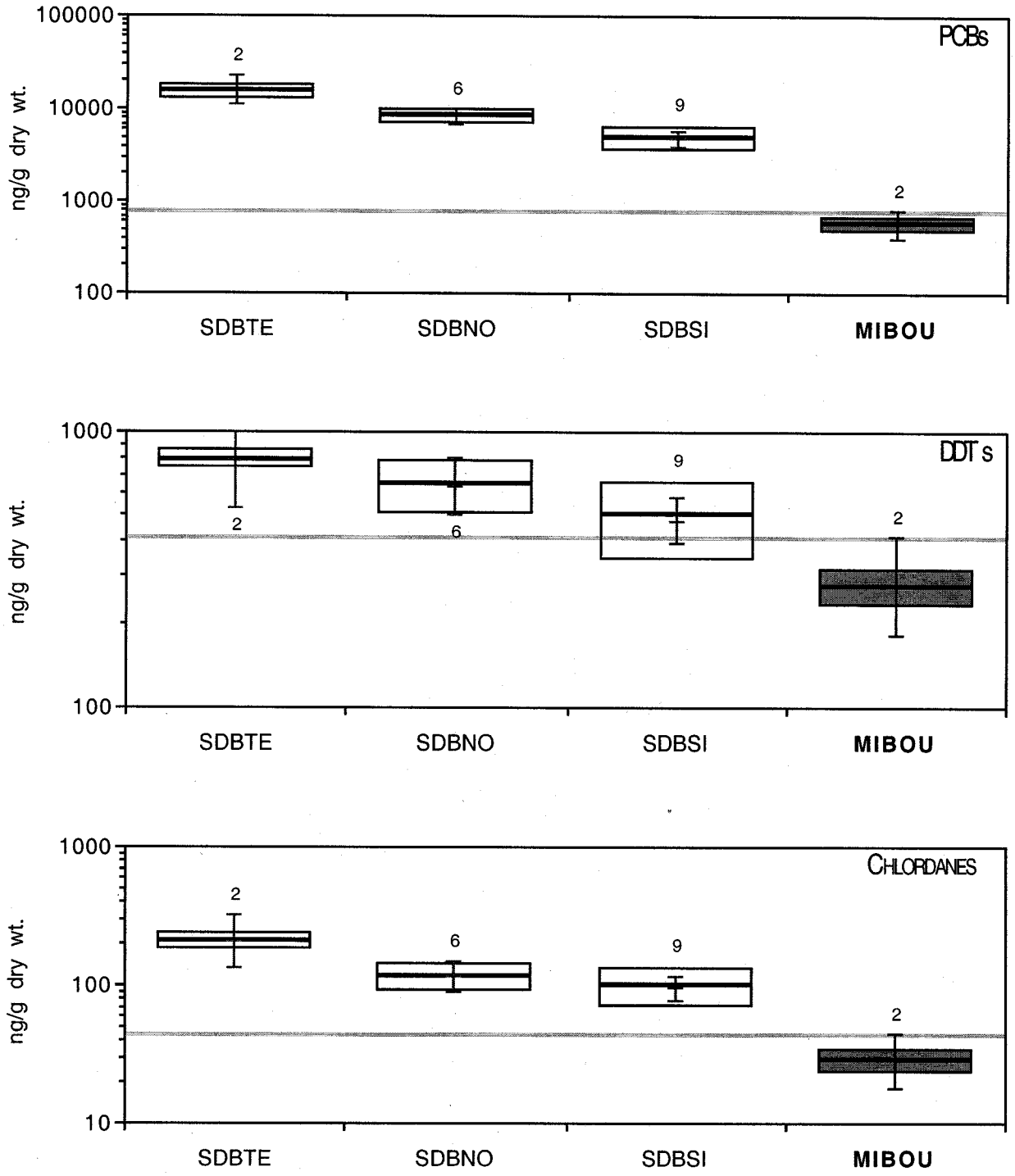


Figure 14. PCBs, DDTs, and chlordanes in livers of black croaker.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▒▒ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

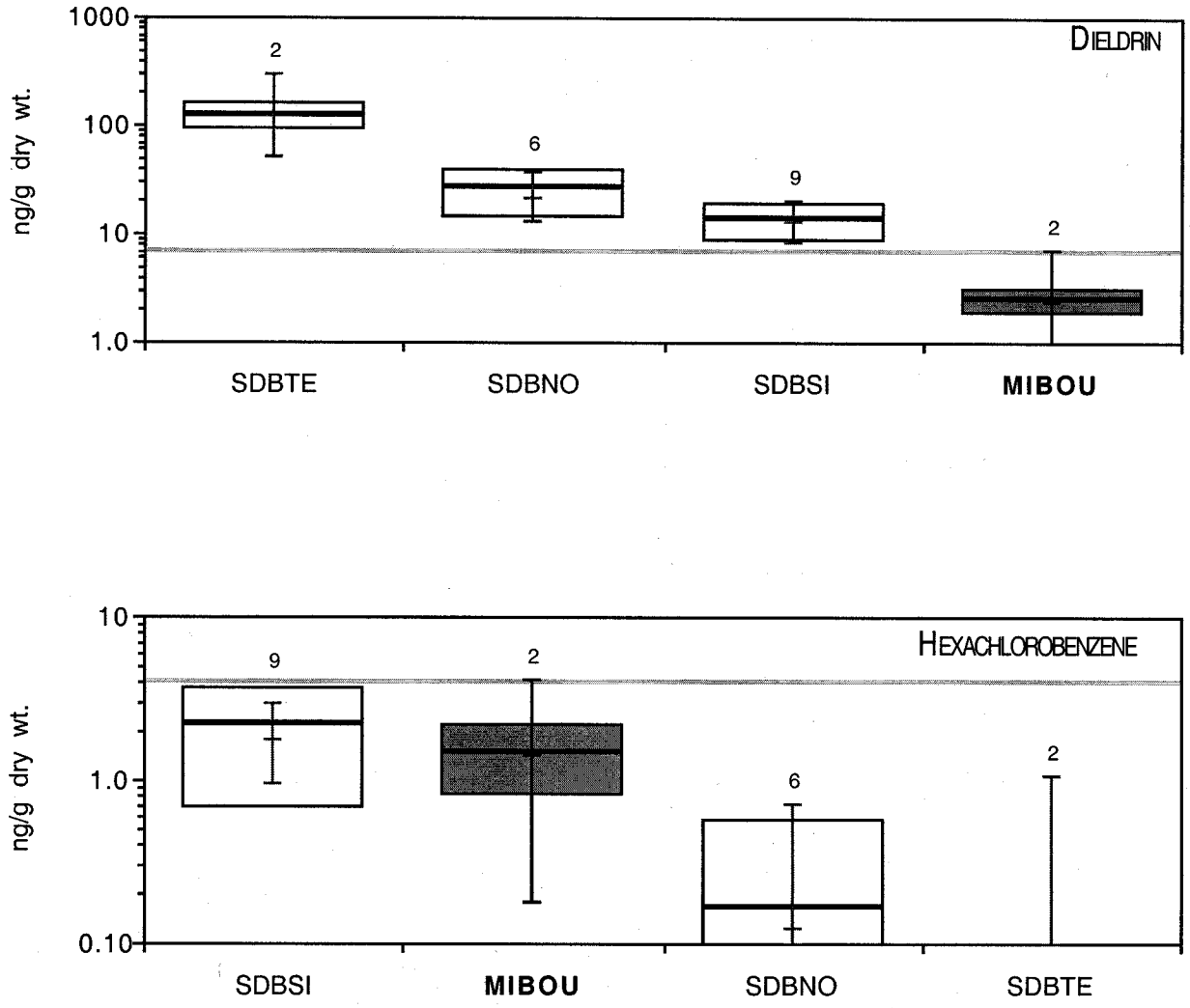


Figure 15. Dieldrin and hexachlorobenzene in livers of black croaker

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▬ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

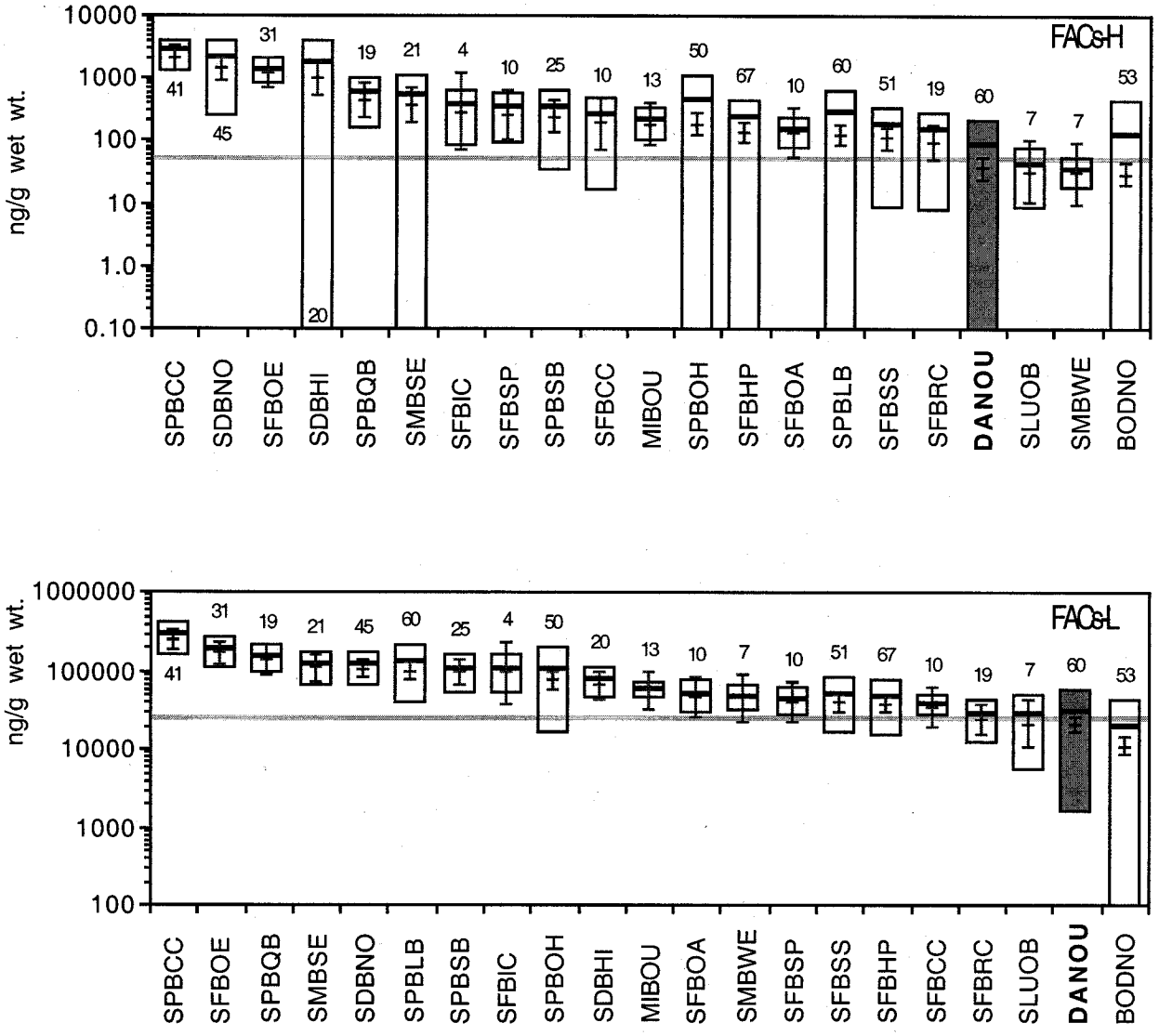


Figure 16. FACs-H and FACs-L in bile of white croaker.

Arithmetic mean [▬] concentrations, (ng/g, wet weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▒▒ ...] for the comparison sites [■]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

croaker was also supported by previous studies. Malins et al. (1987) reported bile levels of FACs-H for white croaker (3,700 ± 3,100 ng/g, n = 7) collected in the winter from a site in the Long Beach/Los Angeles vicinity were substantially higher than those found in the present study (97 ± 98 ng/g, n = 42). These two sites were within 1.3 km of each other. Two factors may have influenced these seasonal differences. First, previous studies of English sole in Puget Sound suggest that as they approach their spawning season the levels of bile metabolites can increase. This may be due, in part, to a reduction in feeding activity and the consequent retention of bile in the gall bladder (Collier and Varanasi 1991, Collier et al. 1992). In white croaker, the peak of the spawning season is in January and February (Love et al. 1984), so that the higher bile levels of FACs for croaker collected in the winter (Malins et al. 1987) may reflect such a seasonal effect. Second, southern California receives more rainfall in winter compared to summer, and urban runoff is known to result in high levels of PAHs entering the marine environment (Hoffman et al. 1984). This runoff may have resulted in higher levels of FACs in bile.

Mean concentrations of PCBs in the liver of white croaker from most urban-associated sites were significantly higher than those for white croaker from the nonurban sites at Dana Point and in Bodega Bay (Fig 17); the exception was the Southampton Shoals site in San Francisco Bay. However, only the mean concentrations of DDTs in the livers of white croaker from all four sites in San Pedro Bay, and east and southeast Santa Monica Bay sites were significantly higher than for the Dana Point reference site (Fig 17). The mean concentrations of chlordanes were also significantly higher in croaker from three sites in San Pedro Bay (Long Beach, Cerritos Channel and Seal Beach) as well as the north San Diego Bay site (Fig 17) than in white croaker from the reference site. Levels of HCB in white croaker livers were all ≤ 8 ng/g (Fig 18).

Of interest was the finding that the mean concentration of DDTs in liver of white croaker from the Dana Point site was significantly higher than those in fish from the Redwood City site in San Francisco Bay and the Bodega Bay site. As mentioned above, barred sand bass and white croaker from Dana Point appear to have been exposed to higher levels of DDTs than would be expected for such a nonurban area with low concentrations of DDTs in sediments. Because white croaker tend to form schools and are relatively mobile, it is likely that the elevated levels of DDTs in white croaker from the Dana Point were at least partially the result of previous exposures of these individuals to DDTs in other areas prior to migration to Dana Point.

The mean concentrations of dieldrin in the livers of white croaker were significantly higher at five of six sites in San Francisco Bay (Southampton Shoal being the exception), the North Bay and Harbor Island sites in San Diego Bay, and the Long Beach site in San Pedro Bay compared to white croaker from the Bodega Bay and Dana Point sites (Fig 18). Moreover, the concentrations in white croaker from the San Francisco Bay and Long Beach sites were significantly higher than those for many other urban sites, including sites in San Pedro Bay and Santa Monica Bay.

A significant, but minor, seasonal difference was found between the mean concentration of PCBs in livers of white croaker collected from the North San Diego Bay site during the summer (6.7 ± 0.2 µg/g, dry weight, n = 3) compared to that found in croaker collected during the winter (4.4 ± 0.9 µg/g, dry weight, n = 3). High levels of CHs in the livers of white croaker from San Pedro Bay were reported previously. For example, Malins et al. (1987) reported PCB concentrations of 13,000 and 5,300 ng/g, respectively, in two composites of five livers of fish from sites near the Long Beach and San Pedro

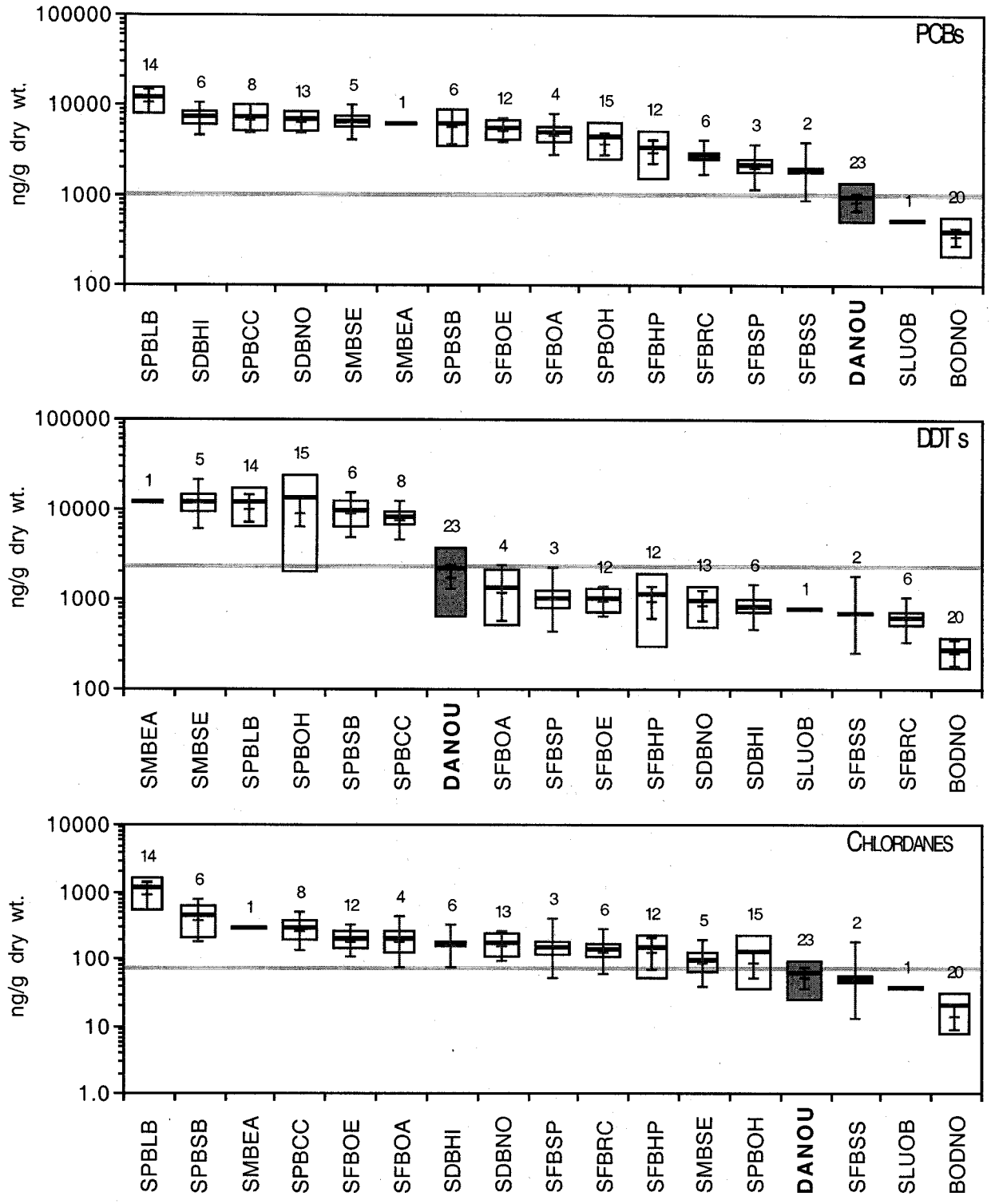


Figure 17. PCBs, DDTs, and chlordanes in livers of white croaker.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▒▒ …] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

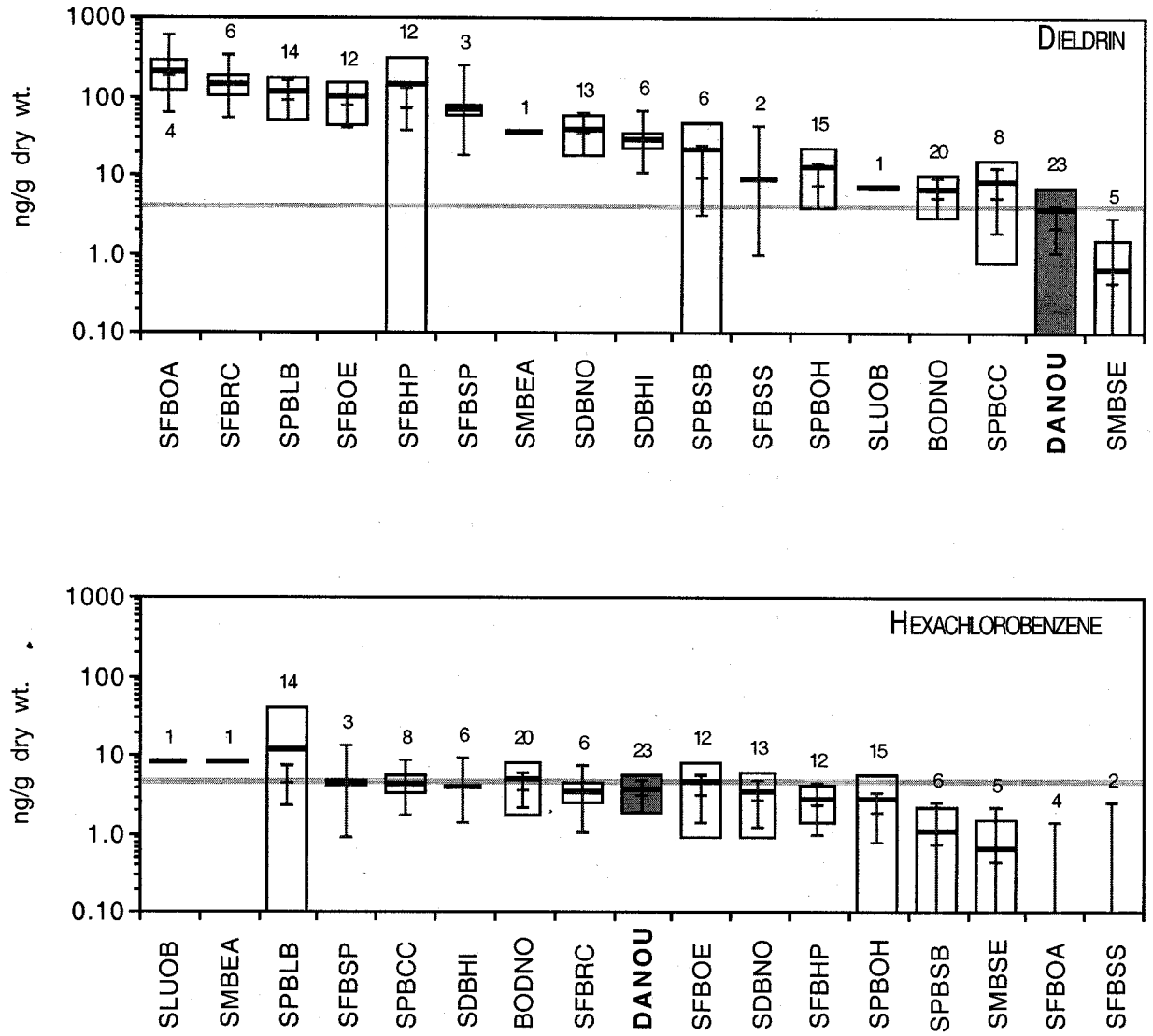


Figure 18. Dieldrin and hexachlorobenzene in livers of white croaker.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▬ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

Outer Harbor sites, compared to 12,000 ± 4,200 ng/g (n = 11) and 4,700 ± 2,200 ng/g (n = 12), respectively, in the present study. At the Outer Harbor site, the levels of DDTs in white croaker livers found in the two studies were somewhat similar: 22,000 ng/g (Malins et al. 1987) vs. 15,000 ± 12,000 ng/g found in the present study. However, at the sites near Long Beach, the previous study reported levels of DDTs (41,000 ng/g) which were over three times higher than those we report here (13,000 ± 5,200 ng/g). The significance of this difference is not presently known.

Concentrations of HAHs in stomach contents of croaker from most urban sites were significantly higher than for Dana Point; the exceptions were the Seal Beach and Redwood City sites (Fig 19). For PCBs, concentrations were also significantly higher in the stomach contents of croaker from most of the urban sites, with the exception of two sites in San Francisco Bay (Hunters Point and Redwood City) and the Seal Beach site in San Pedro Bay compared to those in croaker from the two reference sites, Bodega Bay and Dana Point (Fig 20). On the other hand, concentrations of DDTs in the stomach contents of croaker were significantly higher at only three sites in San Pedro Bay—Outer Harbor, Long Beach, and Cerritos Channel (Fig 20). Only the Oakland Estuary site had croaker with stomach contents having significantly higher concentrations of dieldrin compared to those in croaker from the two reference sites, although all concentrations were low (e.g., ≤ 10 ng/g). With two exceptions, no significant differences were found among the mean concentrations of LAHs, chlordanes and HCB in the stomach contents of white croaker from any of the West Coast sites (Fig 19 -21); the exceptions were the concentrations of LAHs in croaker from the Oakland Estuary site which was higher than the Dana Point site and the mean concentrations of chlordanes in the stomach contents of white croaker from Bodega Bay were lower than those from Dana Point. The concentrations of HCB in these stomach contents samples were quite low (<10 ng/g) and thus more difficult to detect significant differences.

Hornyhead Turbot

Concentrations of FACs-H, FACs-L, DDTs and PCBs in hornyhead turbot were significantly higher for all of the urban-associated sites, with the exception of the San Diego Bay Outer Harbor site, compared to levels in turbot from the Dana Point nonurban site (Figs. 22 and 23). The mean concentrations of FACs-H and DDTs in turbot from the San Diego Bay Outer Harbor site were not significantly different from those for Dana Point turbot. For chlordanes, only mean concentrations in the livers of hornyhead turbot from four of the Santa Monica Bay sites were significantly higher than those from the Dana Point site (Fig 23). Concentrations of dieldrin in turbot livers were significantly higher only at the San Pedro Canyon site, whereas no significant differences in mean concentrations (≤10 ng/g) of HCB in turbot liver were found among any of the sites (Fig 24).

Stomach contents of turbot from the west, southeast and south Santa Monica Bay sites had concentrations of PCBs and DDTs which were significantly higher than those from Dana Point (Fig 26). However, no significant differences were found in the concentrations of PAHs, HAHs, LAHs, chlordanes, dieldrin or HCB in the stomach contents of hornyhead turbot among fish from any of the sites (Fig 25 - 27).

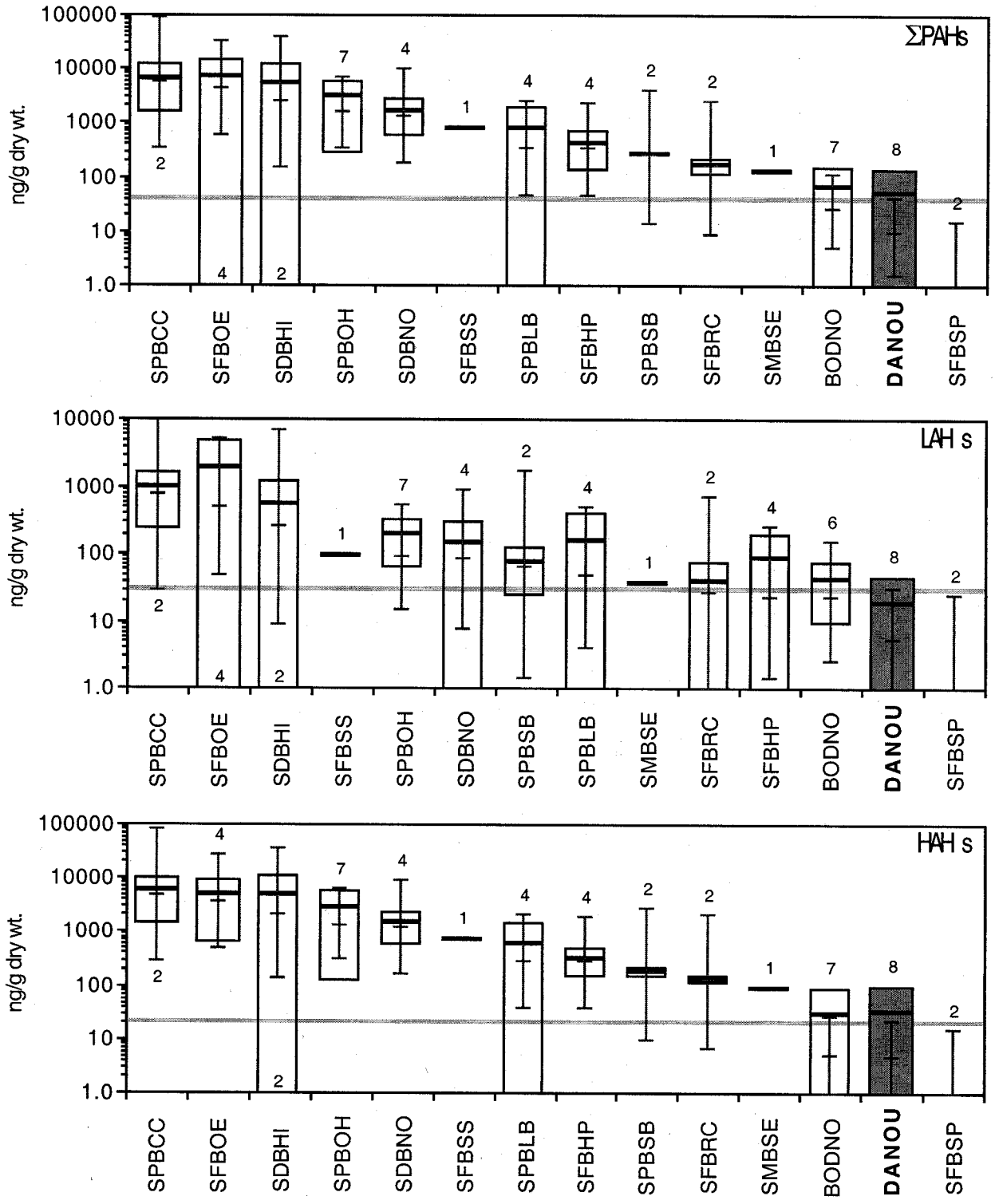


Figure 19. Total PAHs, LAHs, and HAHs in stomachs of white croaker.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▭ …] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

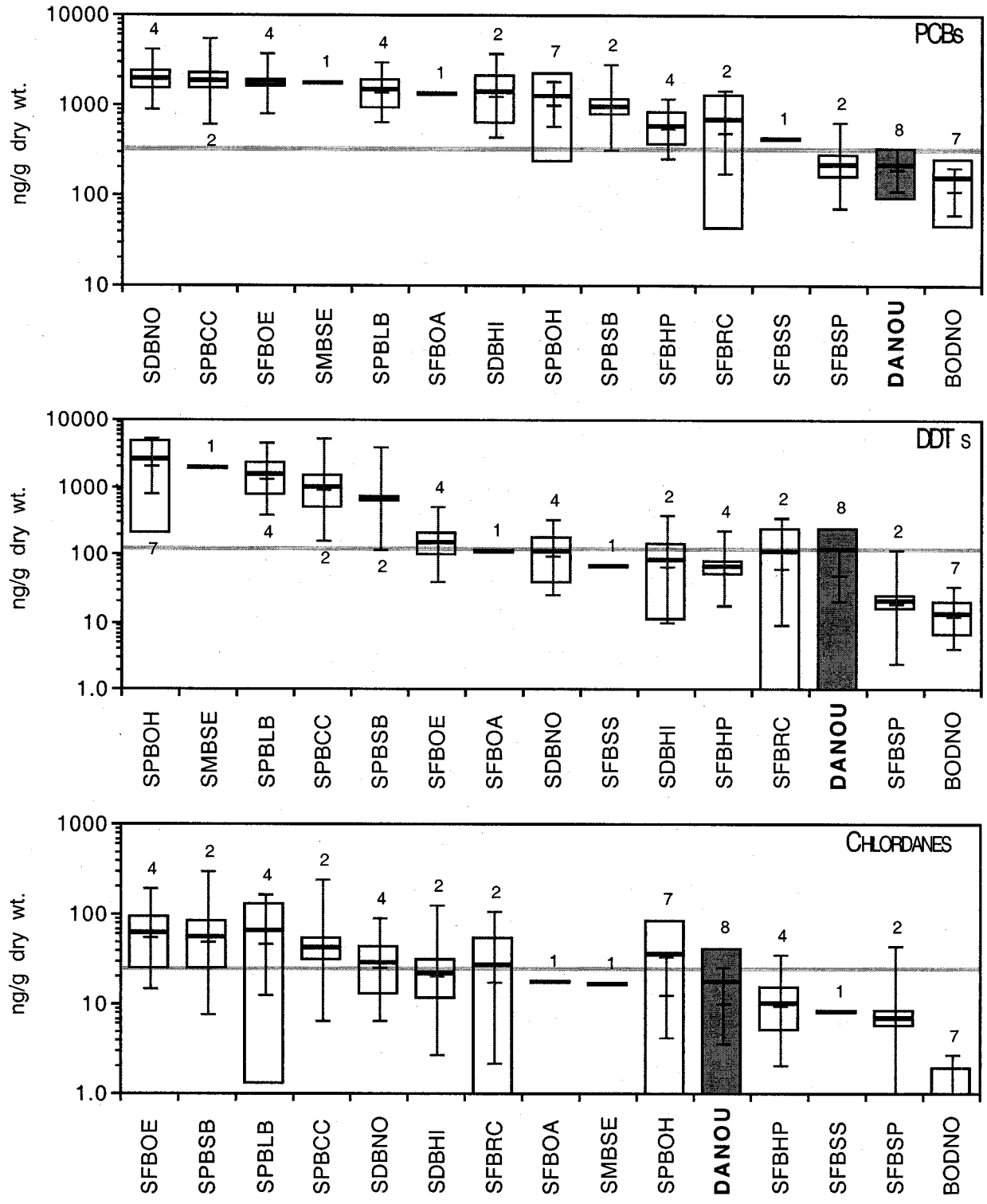


Figure 20. PCBs, DDTs and chlordanes in stomachs of white croaker.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▒▒ …] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

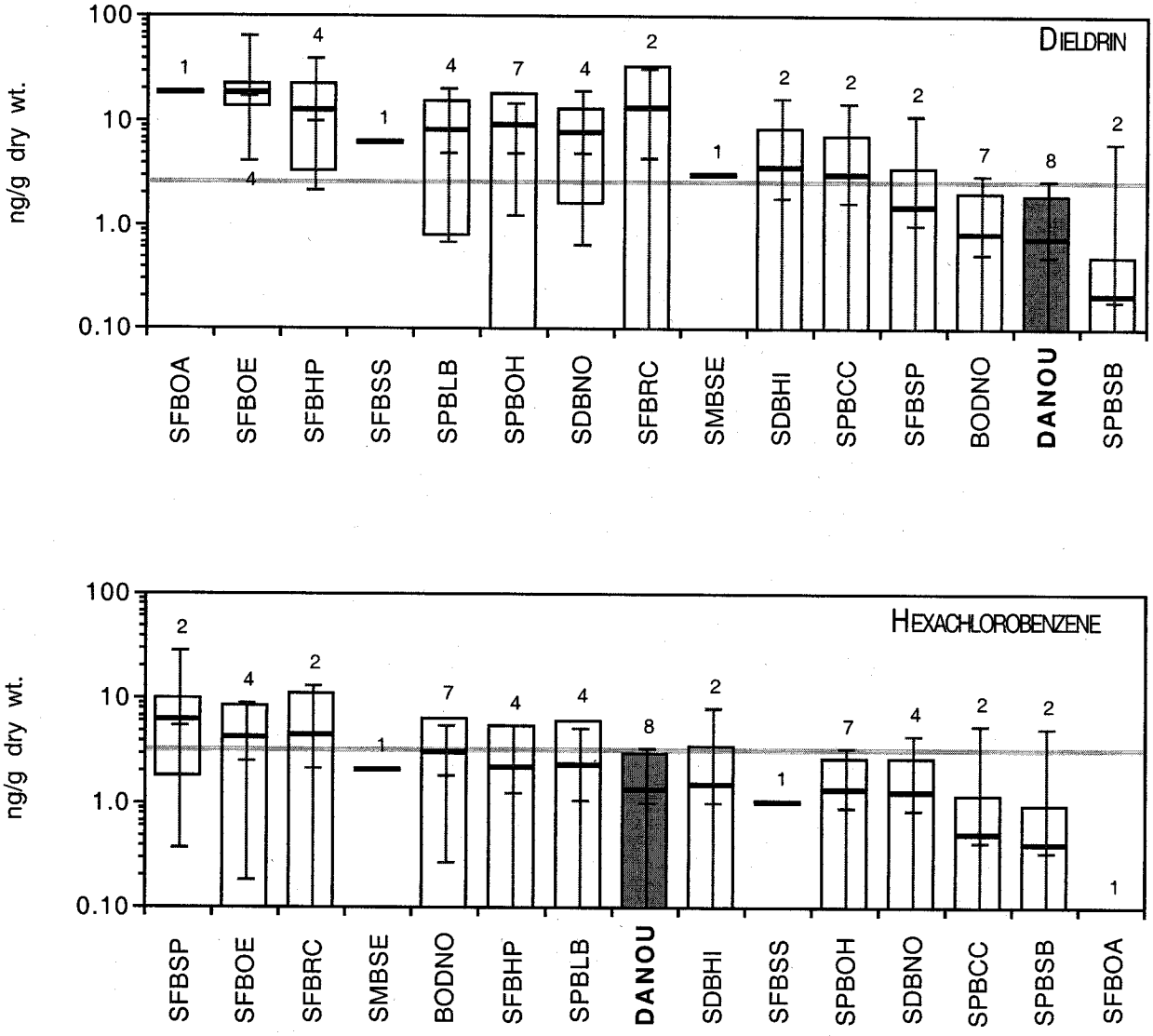


Figure 21. Dieldrin and hexachlorobenzene in stomachs of white croaker.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▒▒▒ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

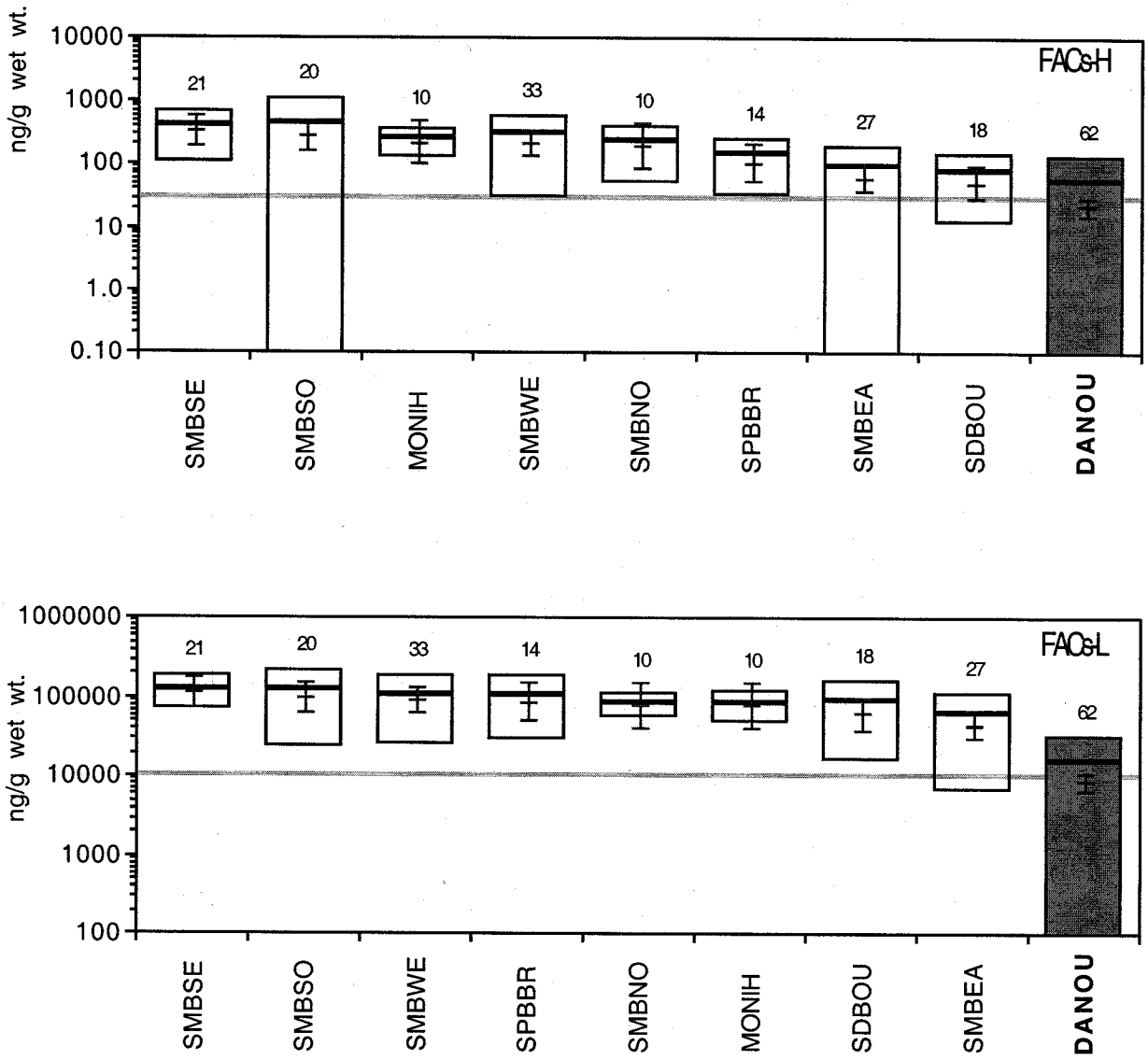


Figure 22. FACs-H and FACs-L in bile of hornyhead turbot.

Arithmetic mean [▬] concentrations, (ng/g, wet weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▬ ...] for the comparison sites [■]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

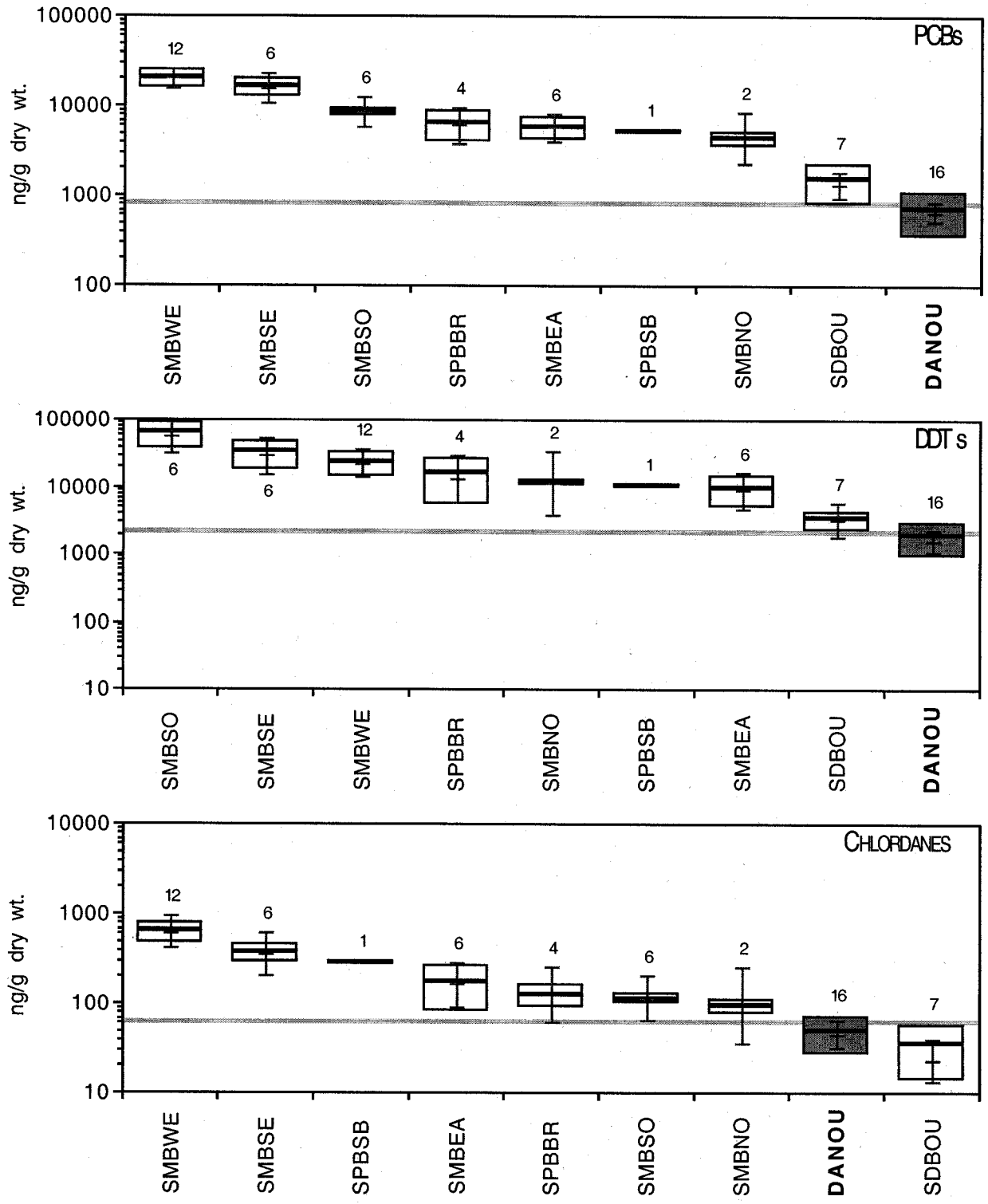


Figure 23. PCBs, DDTs, and chlordanes in livers of hornyhead turbot.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▒ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

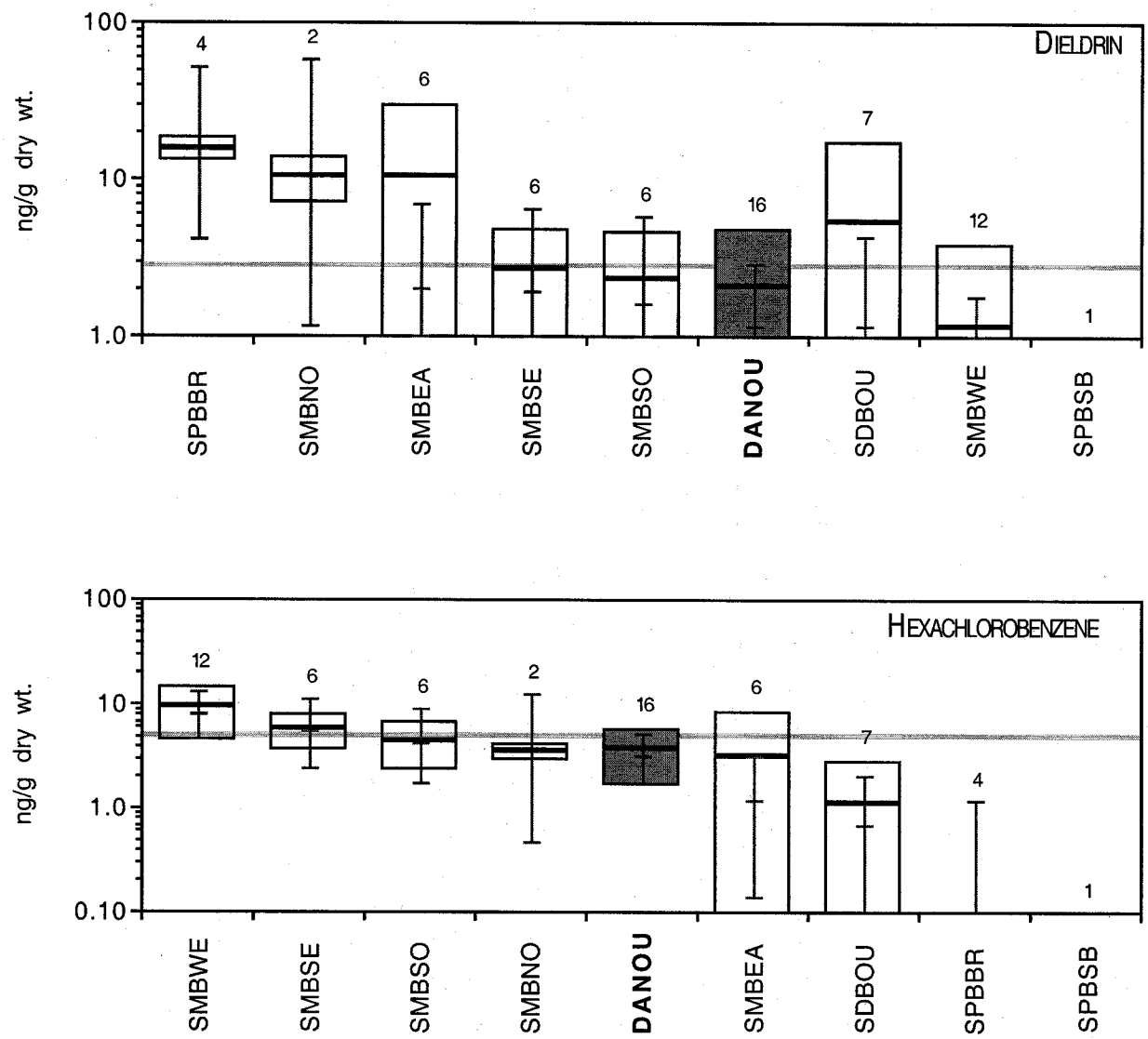


Figure 24. Dieldrin and hexachlorobenzene in livers of hornyhead turbot.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ... ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

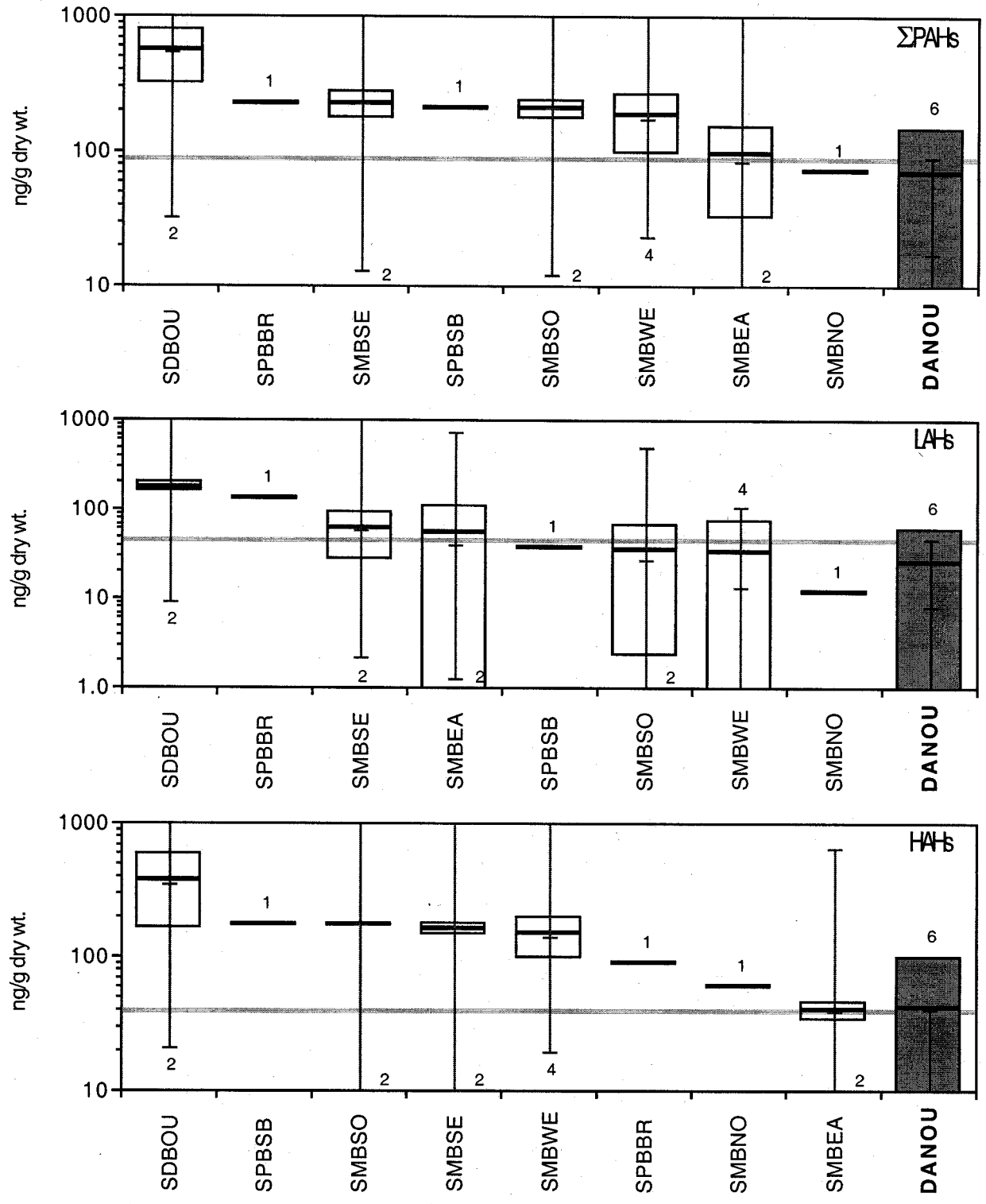


Figure 25. Total PAHs, LAHs, and HAHs in stomachs of hornyhead turbot.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▬ …] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

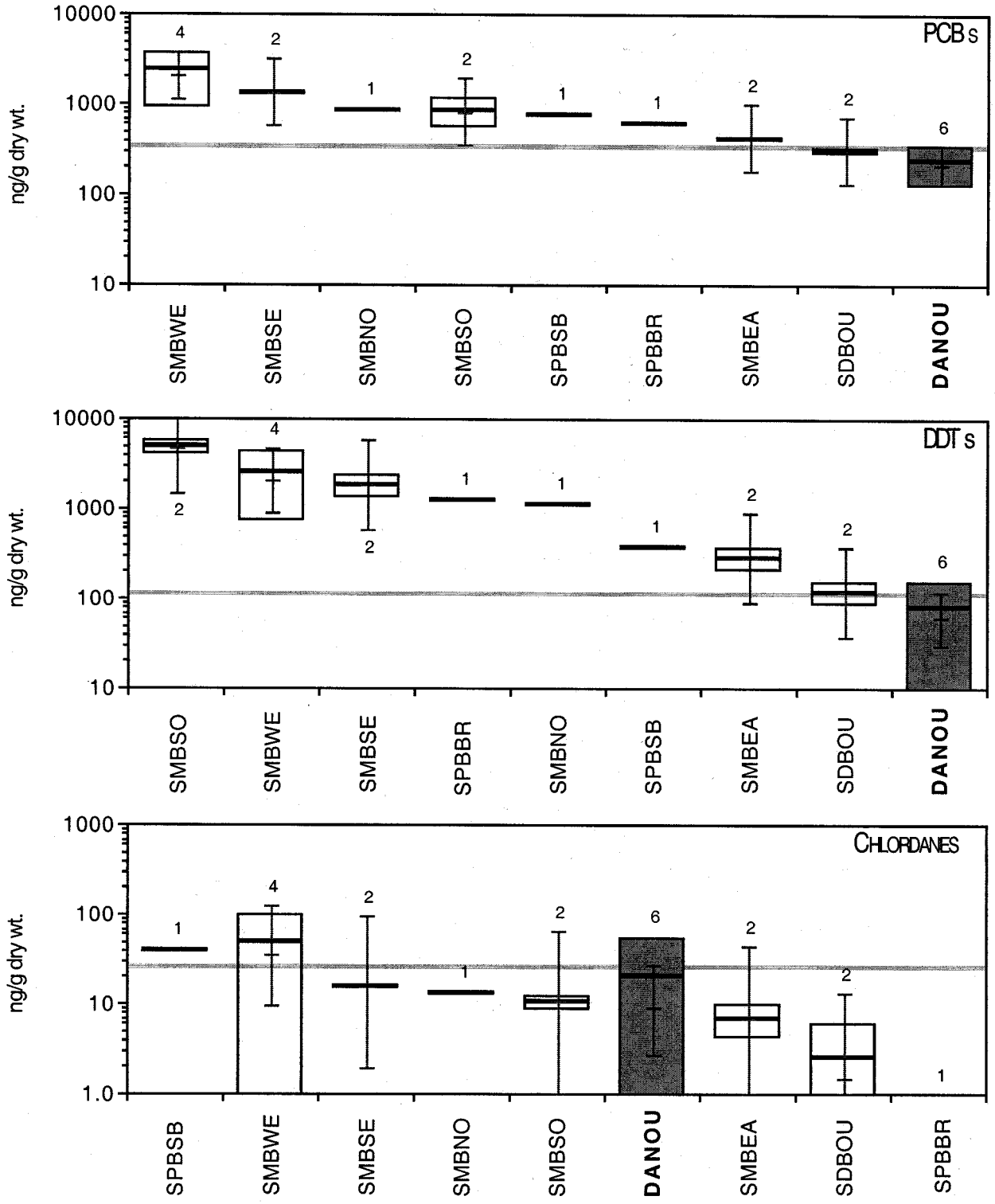


Figure 26. PCBs, DDTs, and chlordanes in stomachs of hornyhead turbot.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▬ …] for the comparison sites [■]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

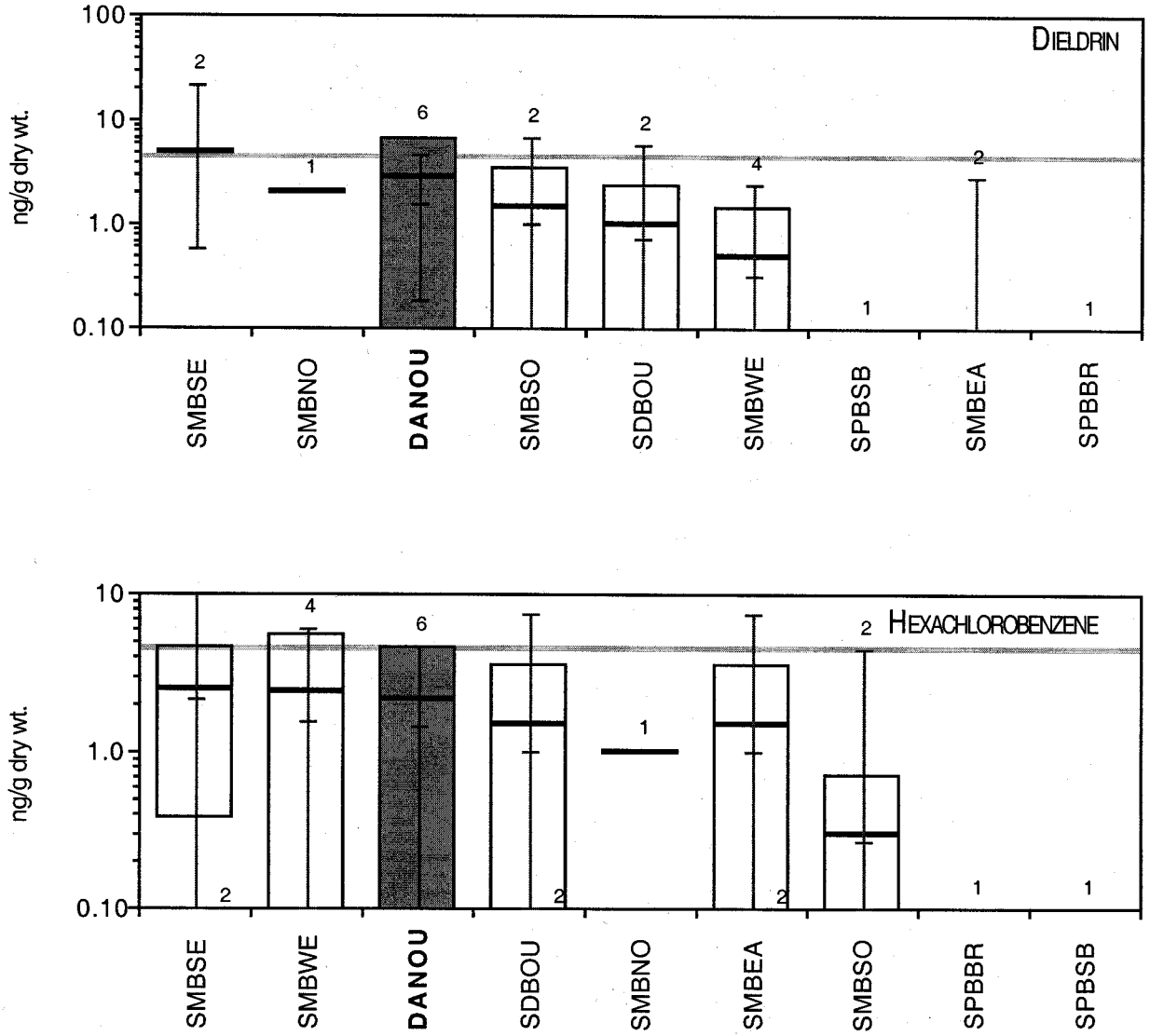


Figure 27. Dieldrin and hexachlorobenzene in stomachs of hornyhead turbot.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▬ …] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

Starry Flounder

In general, most of the sites in San Francisco Bay and Coos Bay had starry flounder with relatively high concentrations of both FACs-H and FACs-L. Of the 15 sampling sites for which mean concentrations of FACs-H in the bile of starry flounder were determined, only three sites had values that were not significantly different from Bodega Bay (the reference site for starry flounder); these sites were the Youngs Bay site near the mouth of the Columbia River, the site in the Chukchi Sea, and the site in the inner portions of Castro Creek near San Pablo Bay (Fig 28). Similarly, only three of the fifteen sites had mean concentrations of FACs-L that were not significantly different from that for the Bodega Bay site (Chukchi Sea, Kvichak Bay in the Bering Sea and Norton Sound, Fig 28).

Overall, mean concentrations of PCBs, DDTs, chlordanes and dieldrin in the livers of starry flounder were significantly higher at sites in San Francisco Bay compared to sites in Oregon and Alaska (Figs. 29 and 30). The mean concentrations of PCBs and dieldrin in the livers of starry flounder from the sites in San Francisco Bay were also significantly higher than for flounder from the nearby reference site in Bodega Bay. In contrast, among the sampling sites in San Francisco Bay, only the mean concentration of DDTs in the liver of starry flounder from the Southampton Shoals site was significantly different from that for Bodega Bay. The mean concentration of chlordanes in flounder from the Hunters Point and San Pablo Bay sites were also significantly higher than for the Bodega Bay site. Mean concentrations of HCB in flounder livers were $\leq$ 10 ng/g at all sites.

Almost no statistically significant differences among the sampling sites were found for mean concentrations of PAHs, PCBs, chlordanes, dieldrin, or HCB in stomach contents of starry flounder (Figs. 31-33). The few exceptions included the mean concentrations of DDTs in the stomach contents of flounder from the Southampton Shoal and Castro Creek sites that were significantly higher than those for flounder from Coos Bay site (Fig 32), and the mean concentrations of HAHs in flounder from the Hunters Point and Southampton Shoals sites in San Francisco Bay were significantly higher than for the Bodega Bay site (Fig 31). Concentrations of chlordanes, dieldrin, or HCB were all $\leq$ 10 ng/g.

English sole

English sole had the widest geographical distribution of any of the target fish species. They were most abundant at sites in Puget Sound and Monterey Bay, but were collected as far north as Dutch Harbor, Alaska, and as far south as outside of San Diego Bay. However, English sole was not the primary target fish species in Alaska and certain sites in Southern California, so for sole from these sites, only analyses for FACs in bile were performed because the analyses are relatively inexpensive. The results of analyses for FACs-H demonstrated that concentrations in sole from sites in the contiguous U.S. were generally significantly higher than those at the Bodega Bay reference site, whereas the concentrations in sole from the Alaska sites were not significantly different from the Bodega Bay sole (Fig 34). The exceptions were the low concentrations at the site near Monterey, CA, and at the Nisqually Reach reference site in Puget Sound. A similar pattern of bile concentrations was observed for mean concentrations of FACs-L in English sole, with some pronounced exceptions: (1) the mean concentration of FACs-L at Dutch Harbor, Alaska, site was among the highest measured in the program, and (2) mean concentrations at the Monterey Bay sites and the Boca de Quadra site were

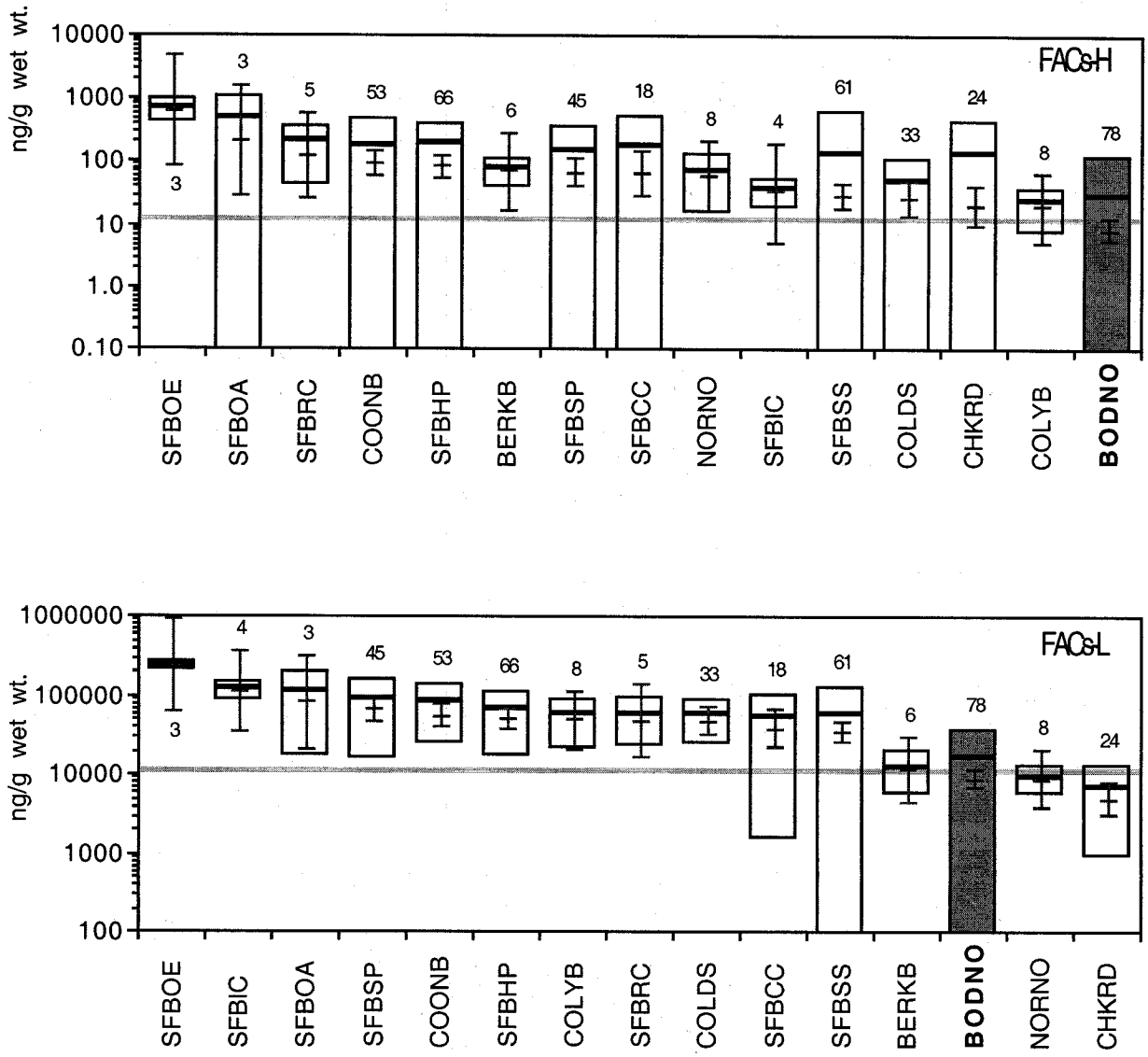


Figure 28. FACs-H and FACs-L in bile of starry flounder.

Arithmetic mean [▬] concentrations, (ng/g, wet weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▒▒▒ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

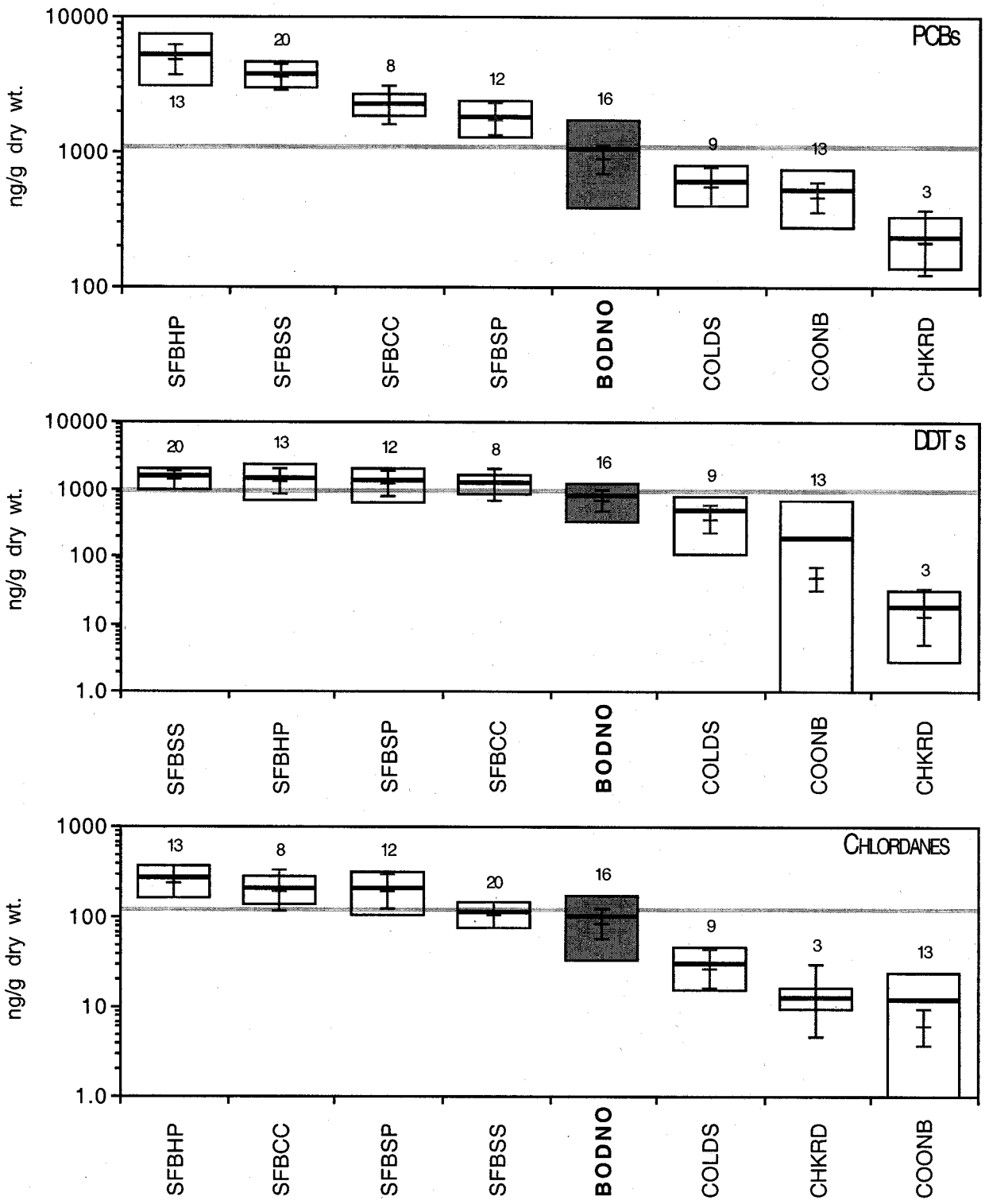


Figure 29. PCBs, DDTs, and chlordanes in livers of starry flounder.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▒ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

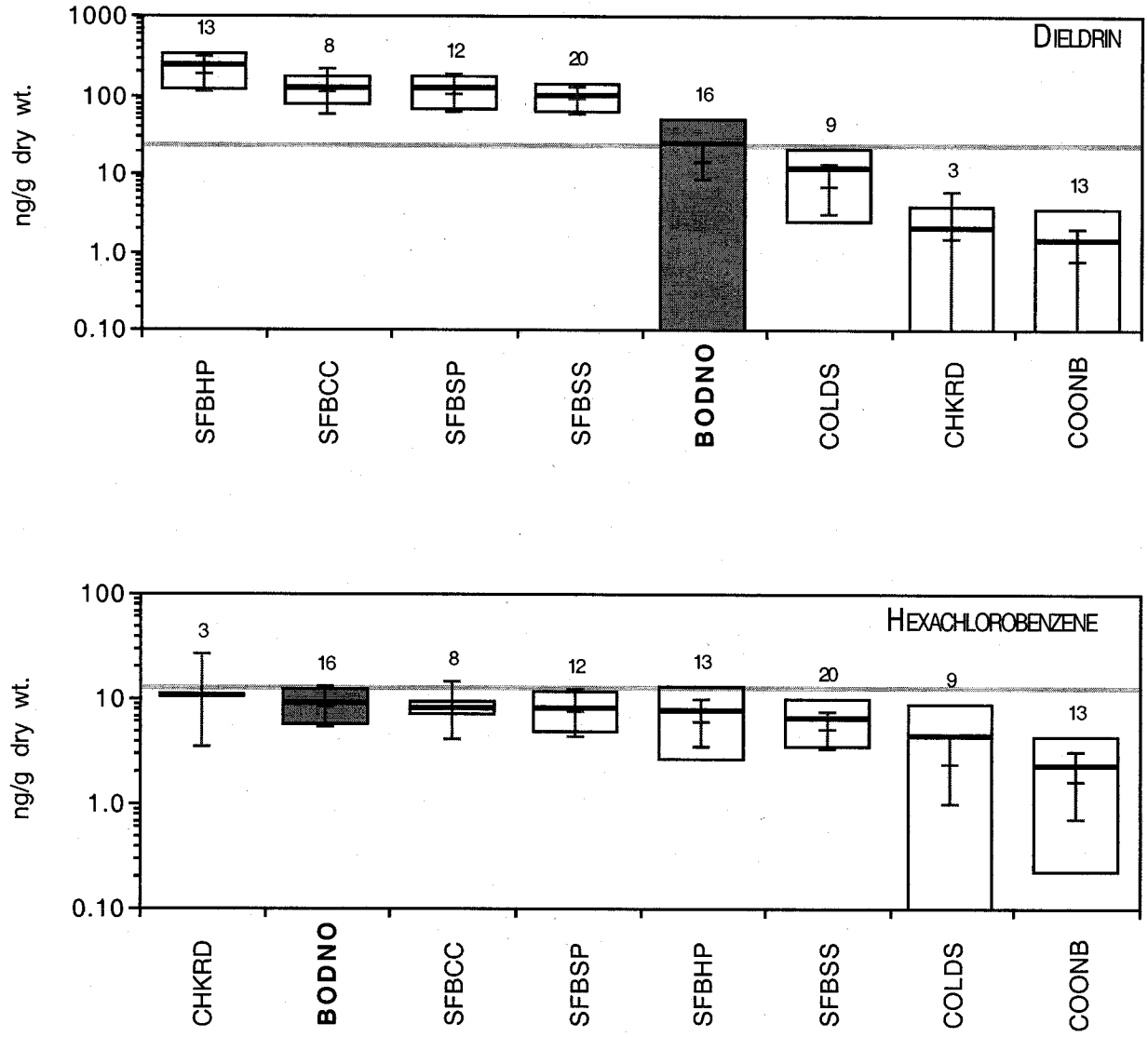


Figure 30. Dieldrin and hexachlorobenzene in livers of starry flounder.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ... ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

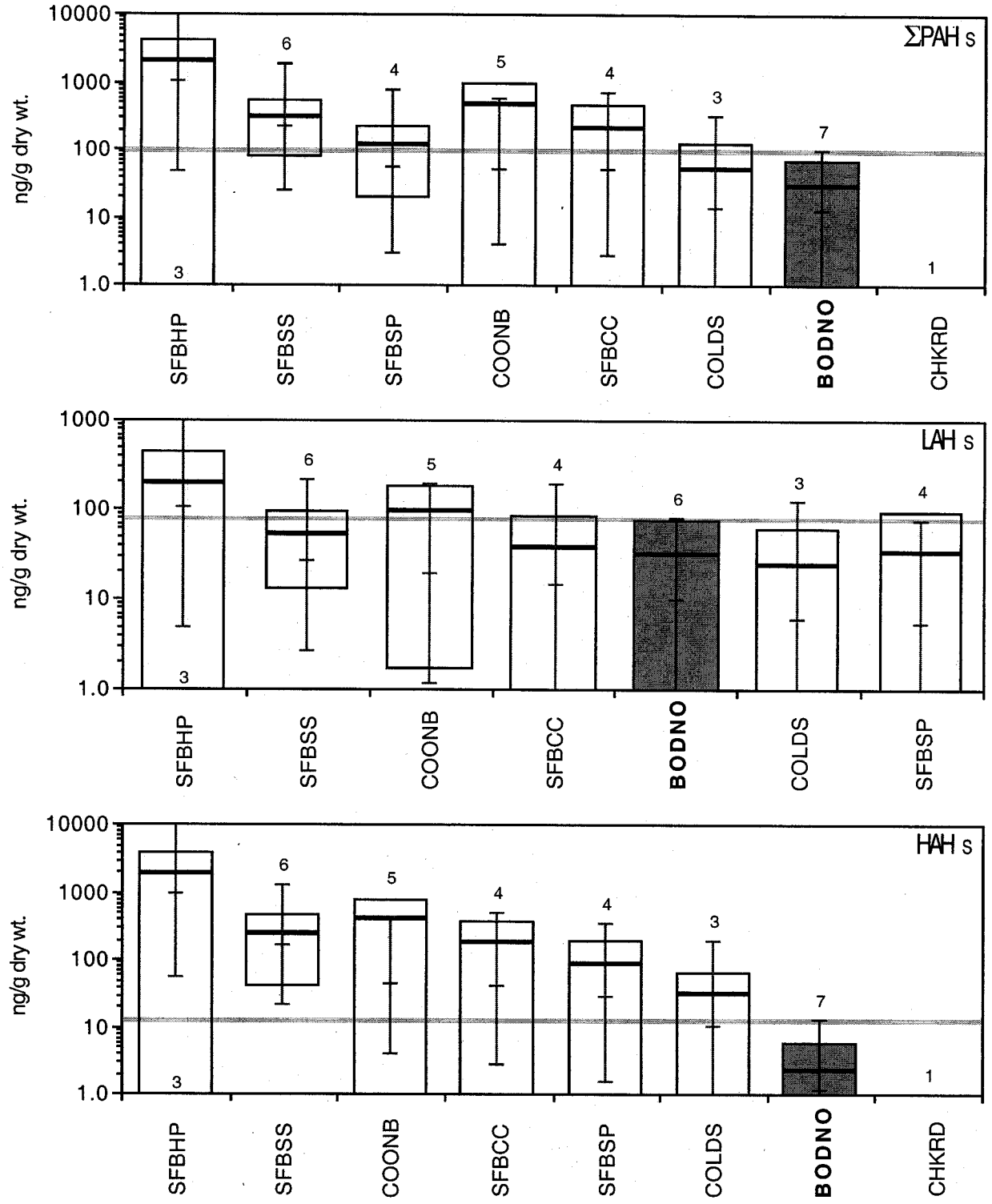


Figure 31. Total PAHs, LAHs, and HAHs in stomachs of starry flounder.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▒▒ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

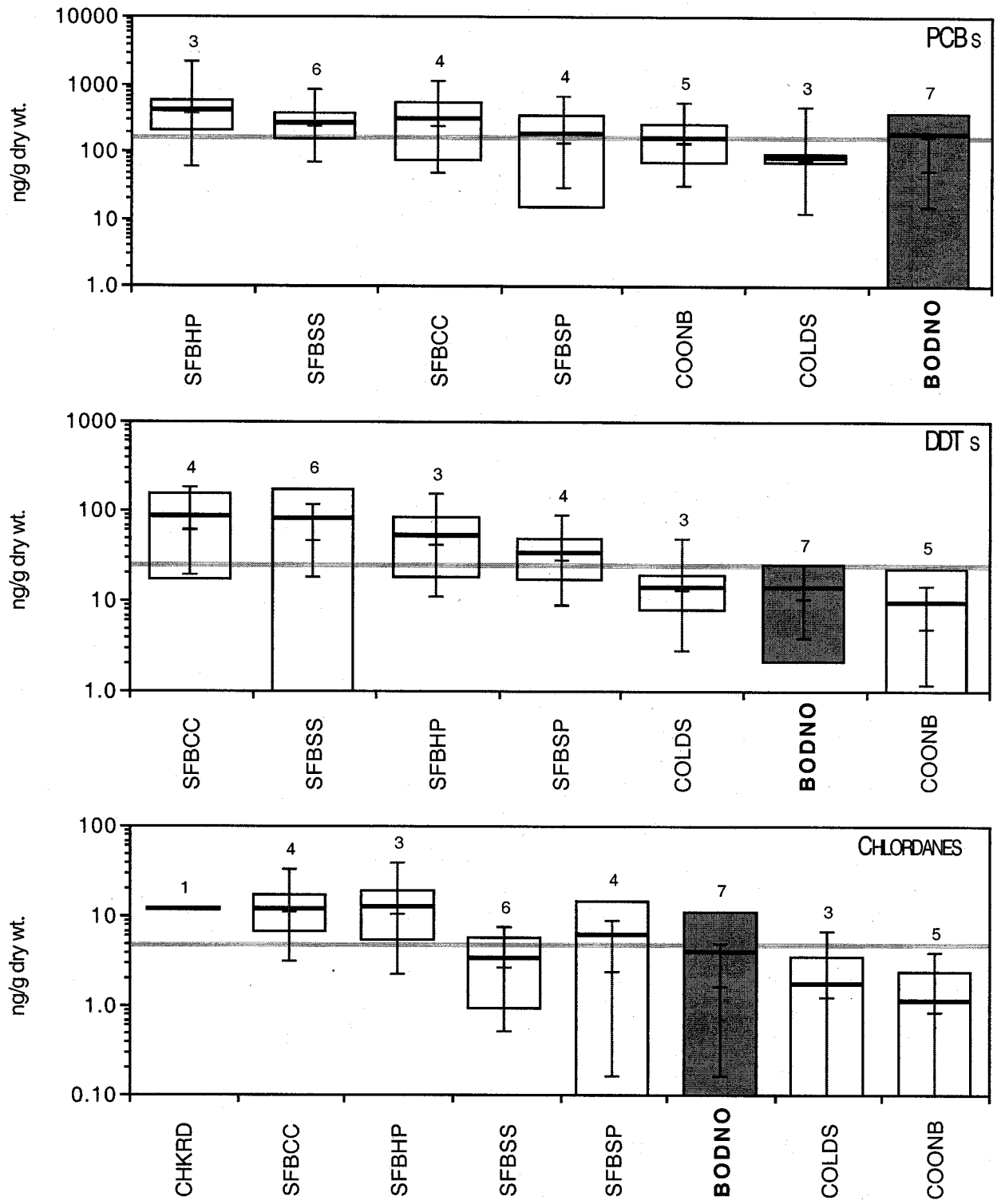


Figure 32. PCBs, DDTs, and chlordanes in stomachs of starry flounder.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▒▒▒ …] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

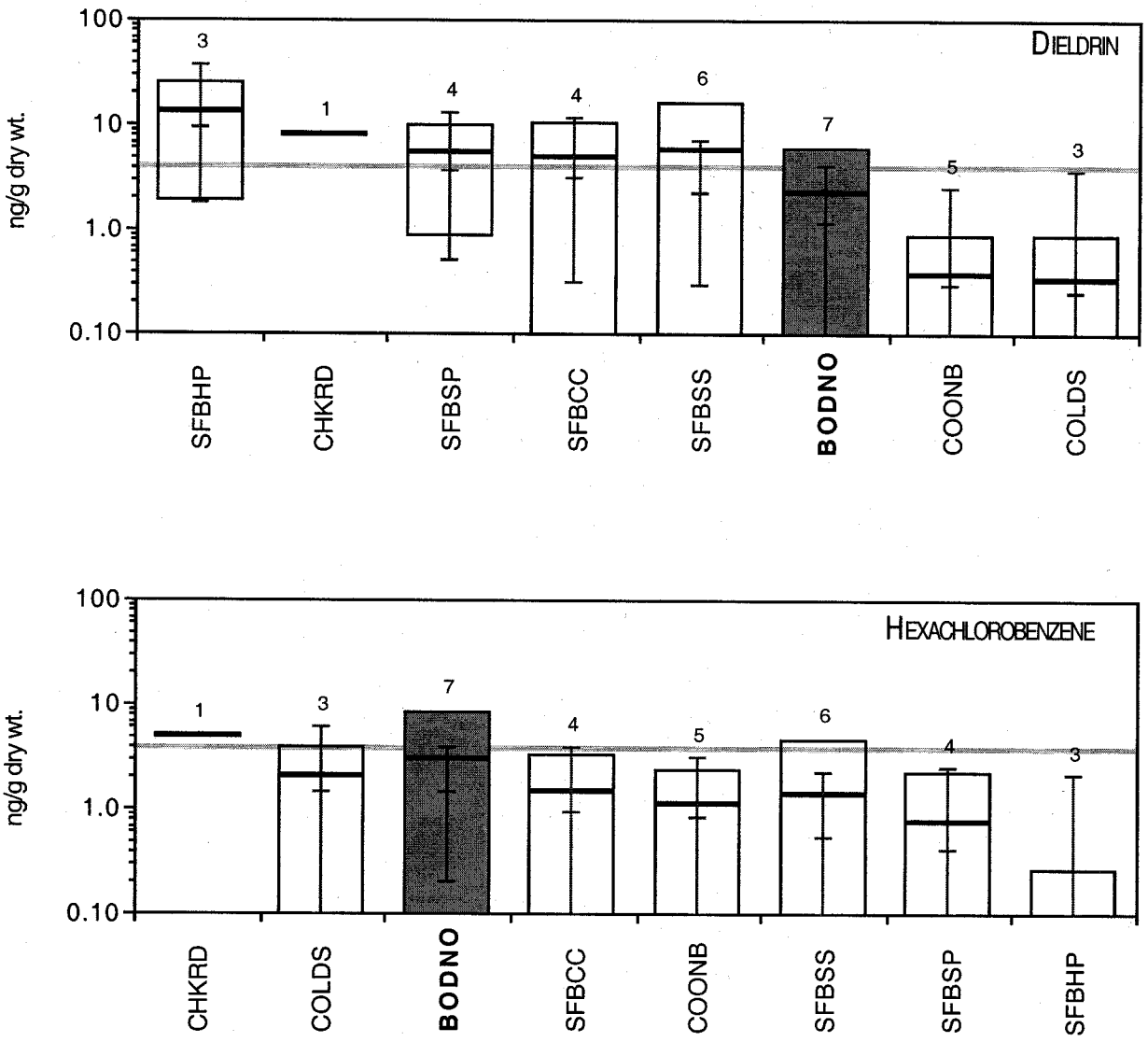


Figure 33. Dieldrin and hexachlorobenzene in stomachs of starry flounder.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▒▒ …] for the comparison sites [■]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

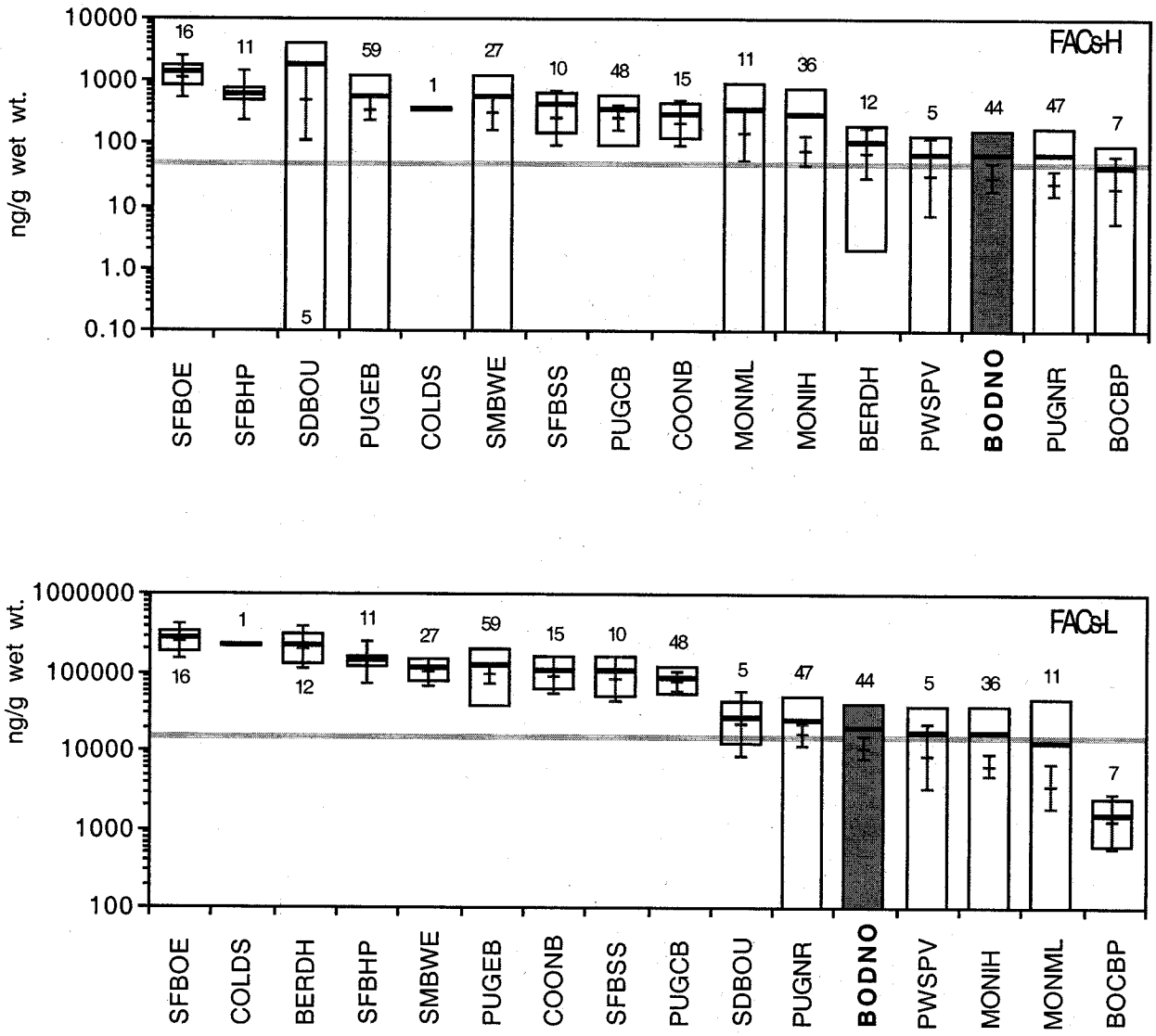


Figure 34. FACs-H and FACs-L in bile of English sole.

Arithmetic mean [▬] concentrations, (ng/g, wet weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▬ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

significantly lower than for the Bodega Bay site. Values for sole from sites away from major urban areas were not significantly different from Bodega Bay sole (Fig 34).

The mean concentration of FACs-H reported here for English sole from the Elliott Bay site is typical of the values reported for English sole from selected urban sites in Puget Sound (Malins et al. 1985, Krahn et al. 1986b), such as sites near Everett and Mukilteo. However, in the most contaminated waterways of Puget Sound (i.e., the Duwamish Waterway and Eagle Harbor), levels of FACs-H in English sole bile were three to eight times higher (Krahn et al. 1986b) than those reported here. On the other hand, bile levels at nonurban sites (e.g., Useless Bay and President Point) in Puget Sound were similar to those found at the nonurban sites in this present study.

The high mean concentration of FACs-L in the bile of English sole from the Dutch Harbor site is surprising considering the relatively low concentrations of LAHs measured in the sediments from this site. A major source of the LAHs to which the sole may have been exposed was likely waterborne petroleum products associated with fishing vessel activity in this harbor which is a major staging area for commercial fishing in the Bering Sea (Otto 1981).

English sole from the Elliott Bay and Commencement Bay sites had significantly higher liver concentrations of PCBs, DDTs, HCB and chlordanes than did sole from the two reference sites (Nisqually Reach and Bodega Bay, Figs. 35 and 36). Although only a single liver composite was analyzed for sole from the Oakland Estuary site, concentrations of PCBs, DDTs, and chlordanes were among the highest found. There was no statistical differences for the concentrations of dieldrin in sole livers among the sites (Fig 36).

The concentrations of PCBs and DDTs found in the livers of English sole from Puget Sound in the present study are similar to those reported previously in studies conducted during the late 1970s and early 1980s. For example, the concentrations of PCBs and DDTs in a single composite of five livers from sole captured in 1979 near the present sampling site along the Seattle Waterfront in Elliott Bay, were 9,200 and 820 ng/g respectively (Malins and al. 1980). These values are comparable to those reported here for PCBs and DDTs in multiple analyses of sole from the Elliott Bay site (10,000 ± 4,200 ng/g and 760 ± 490 ng/g, respectively, n = 9). In addition, concentrations of PCBs and DDTs in composites of five livers of English sole from sites near the present site in Commencement Bay were 6,400 and 350 ng/g, respectively (Malins and al. 1980), which are similar to the present measurements of 4,000 ± 850 and 660 ± 270 ng/g, respectively, n = 10. These comparisons suggest that the levels of PCBs and DDTs in Elliott and Commencement Bays did not change appreciably during the years from 1979 to 1990.

With the exception of LAHs and HAHs no significant differences were found among mean concentrations of all of the chemical contaminants in the stomach contents of English sole from any of the sampling sites (Figs. 37 - 39). Whereas mean concentrations of HAHs were significantly higher in sole from the Elliott Bay and Commencement Bay sites compared to the Bodega Bay site, the stomach content concentrations of LAHs at these two sites were not significantly different from the Bodega Bay site, but they were significantly higher than those for the site in Nisqually Reach. These findings are consistent with data from analyses of other samples from Elliott Bay: high concentrations of HAHs in sediment and high FACs-H concentrations in bile of English sole (Johnson et al. 1994).

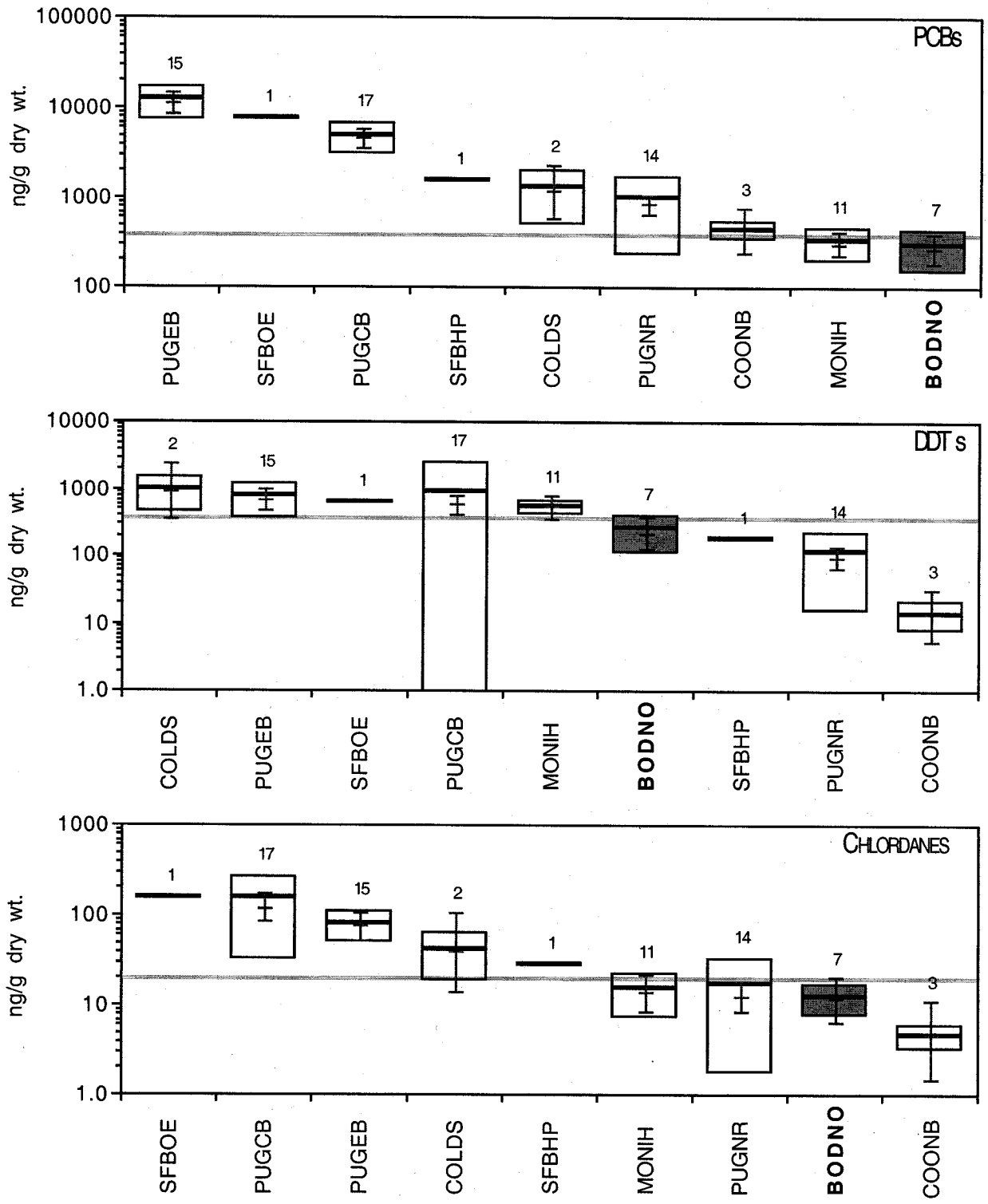


Figure 35. PCBs, DDTs, and chlordanes in livers of English sole.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▒▒ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

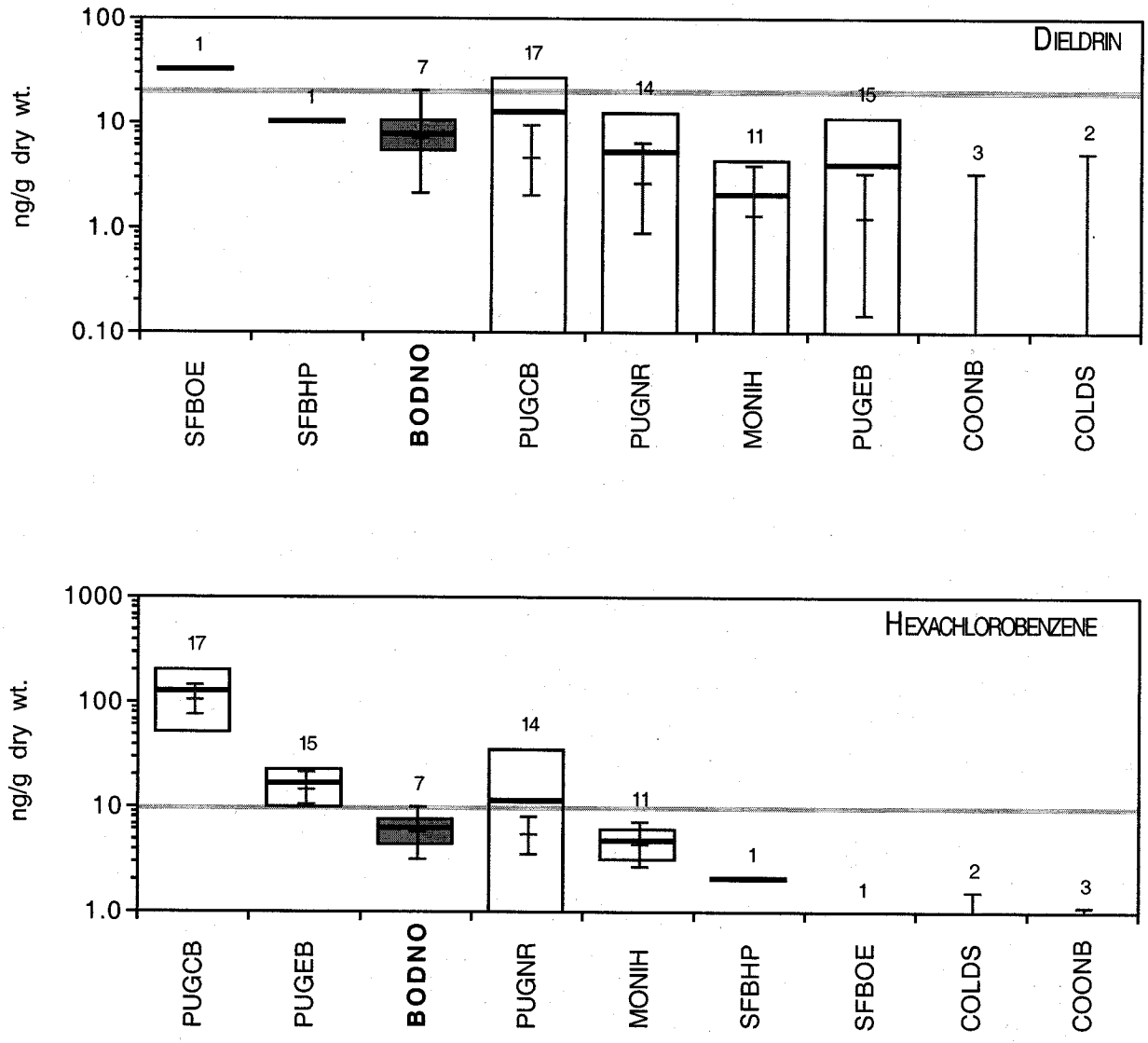


Figure 36. Dieldrin and hexachlorobenzene in livers of English sole.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▬ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

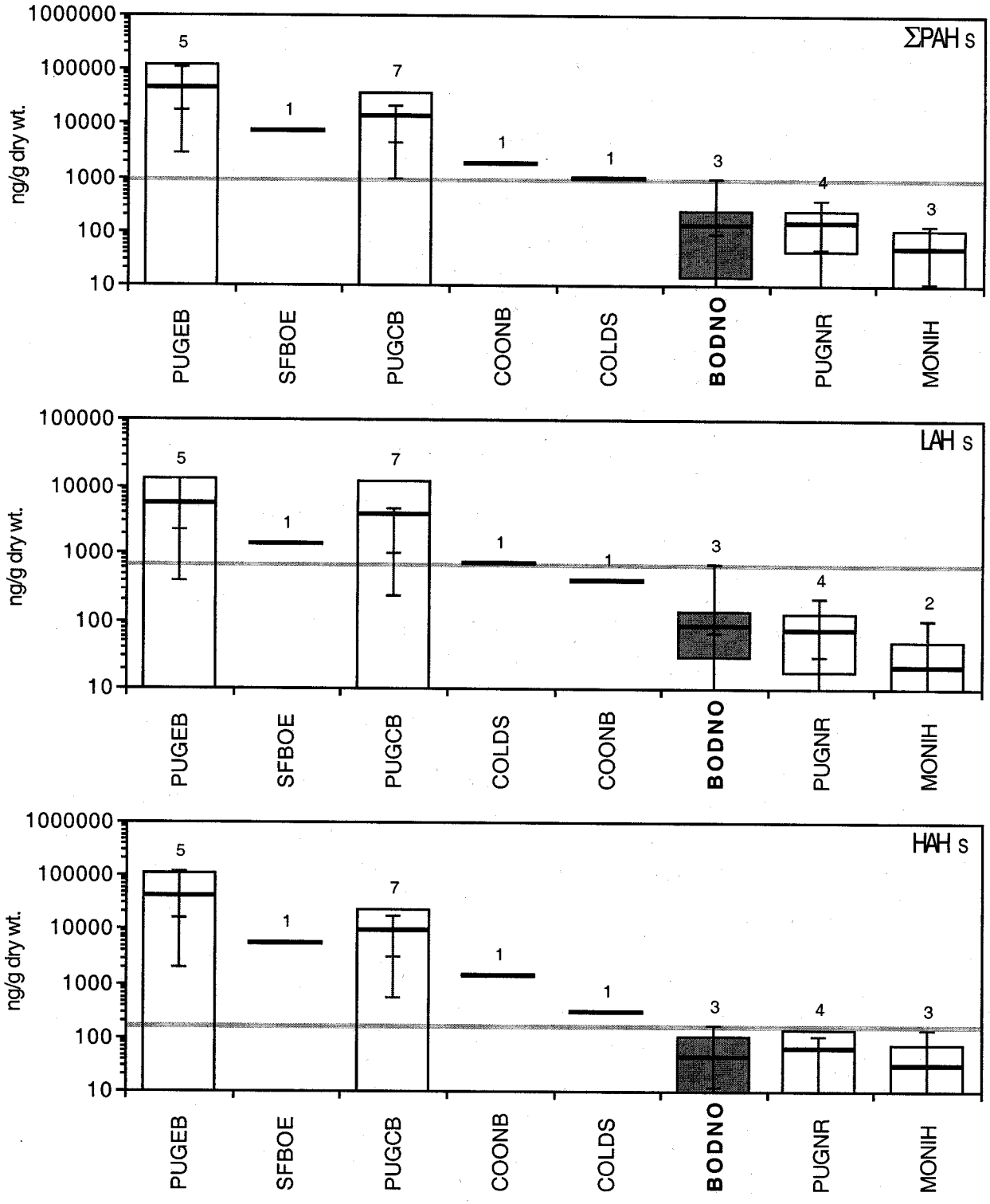


Figure 37. Total PAHs, LAHs, and HAHs in stomachs of English sole.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▬ …] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

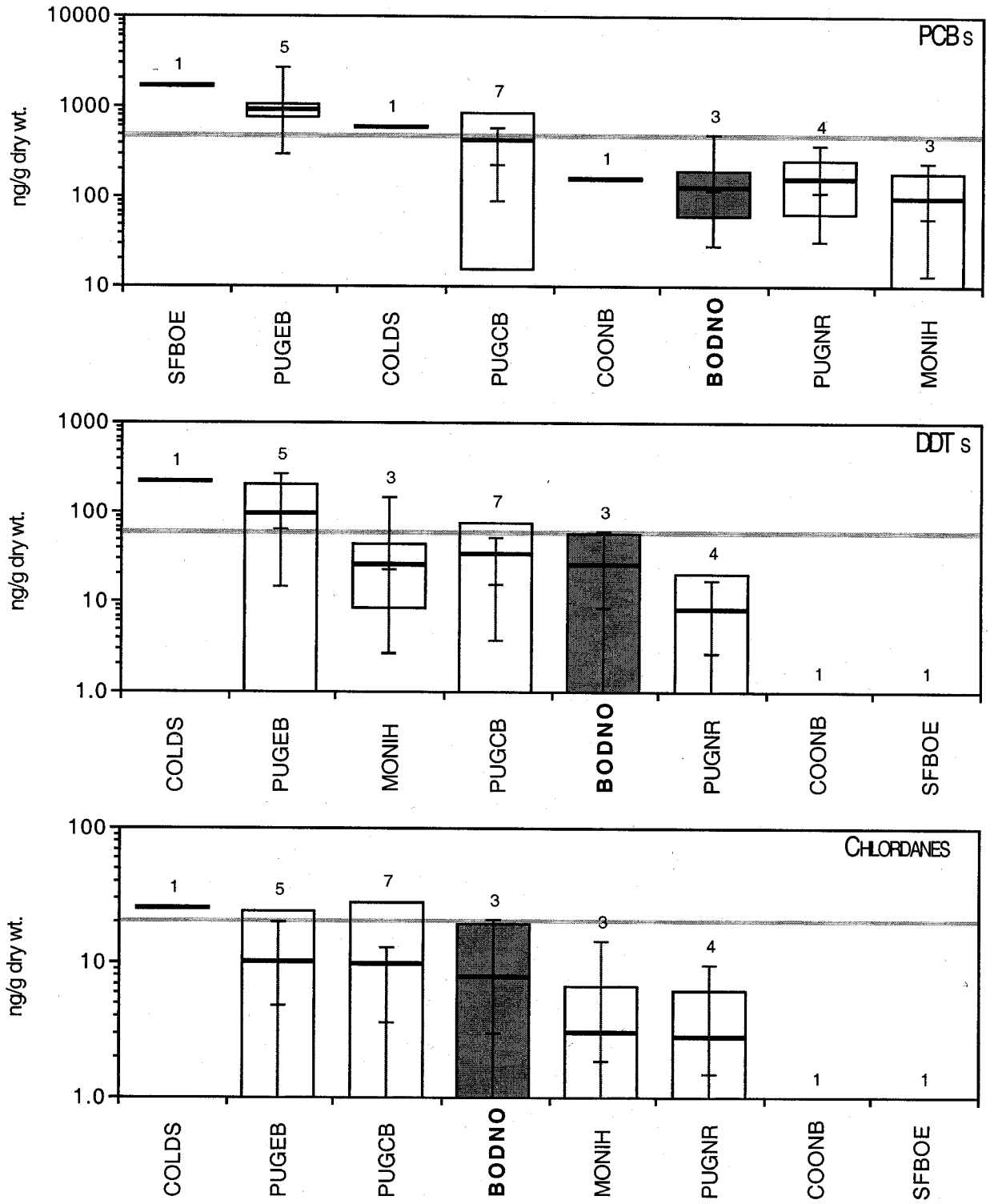


Figure 38. PCBs, DDTs, and chlordanes in stomachs of English sole.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [… ▒▒ …] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

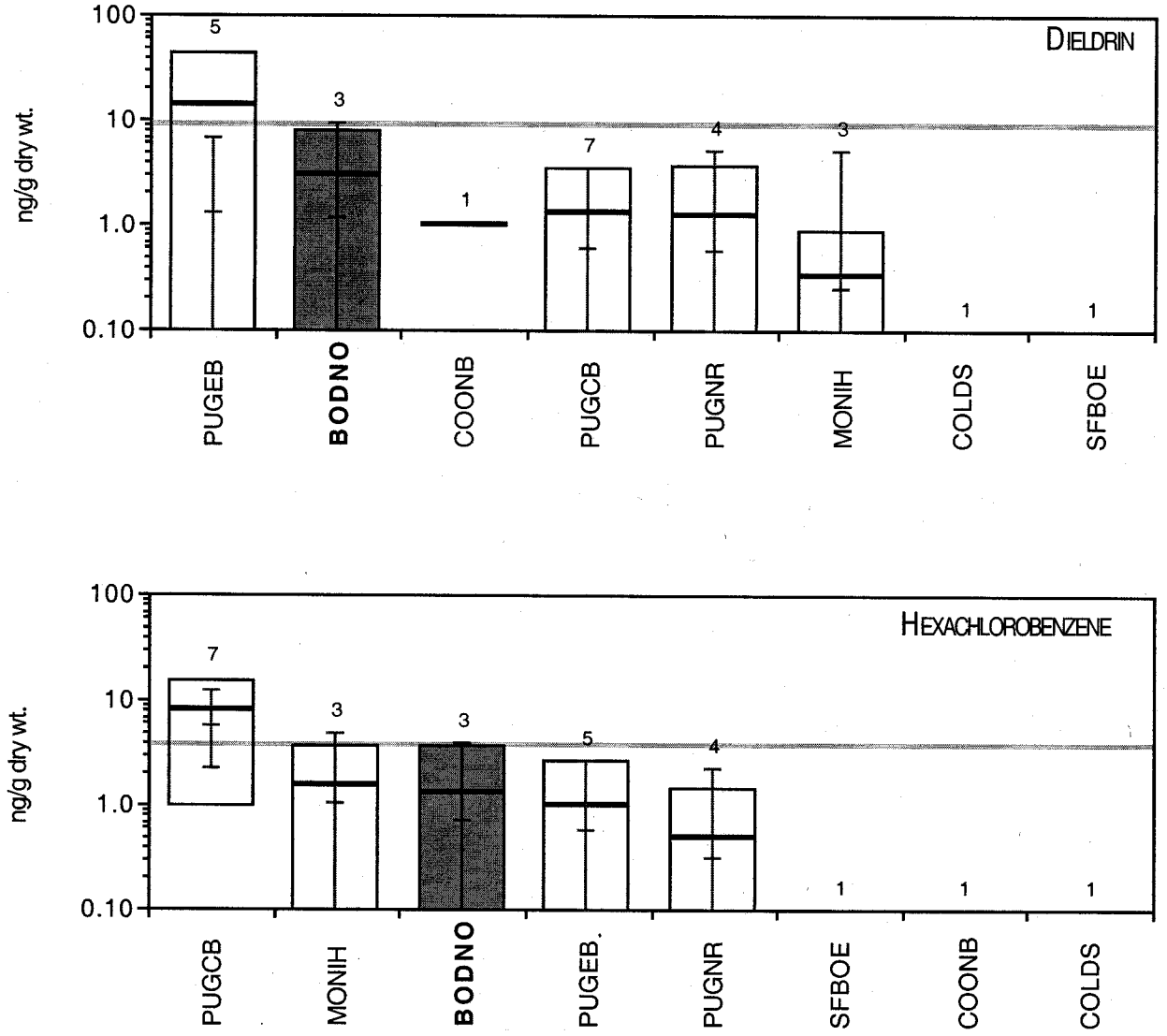


Figure 39. Dieldrin and hexachlorobenzene in stomachs of English sole.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▬ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

Flathead Sole

Flathead sole were collected from two sites in Puget Sound (Elliott and Commencement Bays) and in eight Alaskan sites. This species was not a primary target species for Puget Sound, therefore a limited number chemical analyses were performed, including biliary FACs and CHs in single composites of liver from fish from the Elliott Bay site to provide an urban comparison to the Alaskan data. The Lutak Inlet site was the Alaskan site designated as a reference site; however, two other sites were located in relatively nonurban areas—Boca de Quadra and Kamishak Bay.

The mean concentrations of FACs-H in the bile of flathead sole from the Elliott Bay, Commencement Bay, and Dutch Harbor sites were significantly higher than for the Lutak Inlet reference site, whereas the mean concentration in sole from the Kamishak Bay site was significantly lower than that for the reference site (Fig 40). The mean concentrations of FACs-L in the bile of flathead sole from these three sites, as well as from the Skagway site, were significantly higher than for any of the other sites. Within the Alaskan sites, the highest mean concentration of PCBs in the liver of flathead sole was found at the Dutch Harbor site, which was significantly higher than those in sole from the nonurban sites (Fig 41). Nevertheless, the concentration of PCBs in flathead sole from the Elliott Bay site in Seattle was approximately 7 times as high as that for the Dutch Harbor site. Mean concentrations of DDTs, chlordanes, dieldrin and HCB in flathead sole livers from all of the Alaskan sites were either not significantly different from, or were lower compared to that for the reference site (Fig 41). As described for liver concentrations of PCBs, concentrations of DDTs, chlordanes and dieldrin in sole from Elliott Bay were several times as high as those for sole from Alaskan sites (Fig 42). Too few composites of stomach contents from flathead sole were collected from Alaskan sites to permit statistical tests on the chemistry data.

Fourhorn Sculpin

Fourhorn sculpin were collected from the three sites on the Beaufort Sea. Sufficient amounts of bile for more than a single analysis were obtained only in sculpin from the Endicott Field site, hence statistical comparisons were not possible. Generally, however, the concentrations of FACs in the bile of fourhorn sculpin were low (Fig 43) compared to other Alaskan fish species, such as English sole and flathead sole from Dutch Harbor or Elliott Bay (Figs. 34 and 40, respectively).

Concentrations of PCBs in the livers of fourhorn sculpin from the Oliktok Point site were several fold higher than those in fish from the Endicott Field site (Fig 44). No appreciable differences were found between concentrations of DDTs, HCB, dieldrin or chlordanes in the livers of sculpin from these two sites (Figs. 44 and 45). The mean concentrations of DDTs, chlordane and dieldrin were low, typical of nonurban sites; however, the mean concentrations of HCB in the livers of sculpin from both sites were somewhat higher than expected—at least double that found in flathead sole livers from the other Alaskan sites.

Only one composite of stomach contents was obtained from sculpin collected at the Oliktok Point site, precluding statistical treatment of the data. Nevertheless, a few qualitative observations can be made about the data. The stomach contents in fourhorn sculpin from the Endicott Field and Oliktok Point

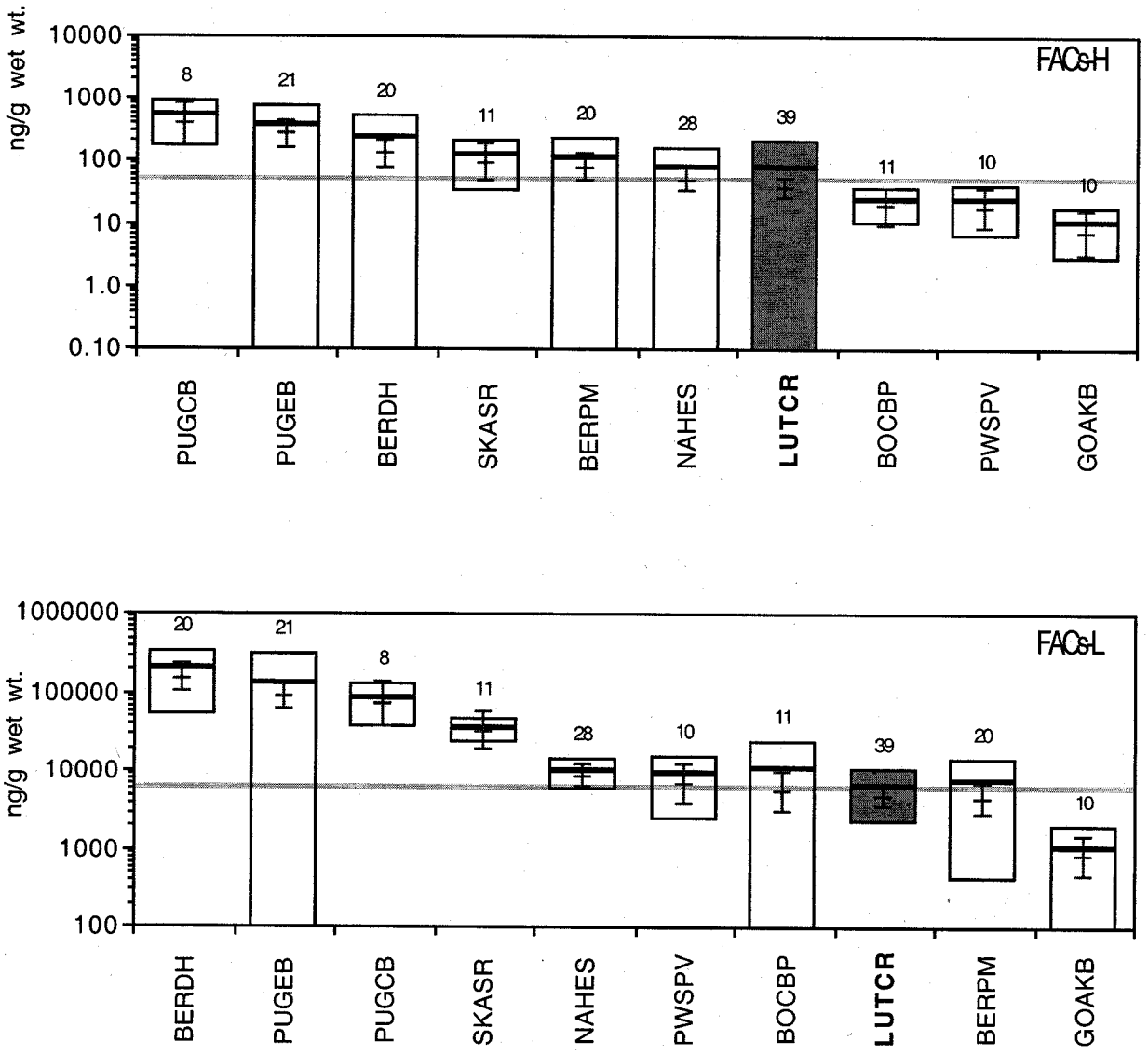


Figure 40. FACs-H and FACs-L in bile of flathead sole.

Arithmetic mean [▬] concentrations, (ng/g, wet weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ... ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

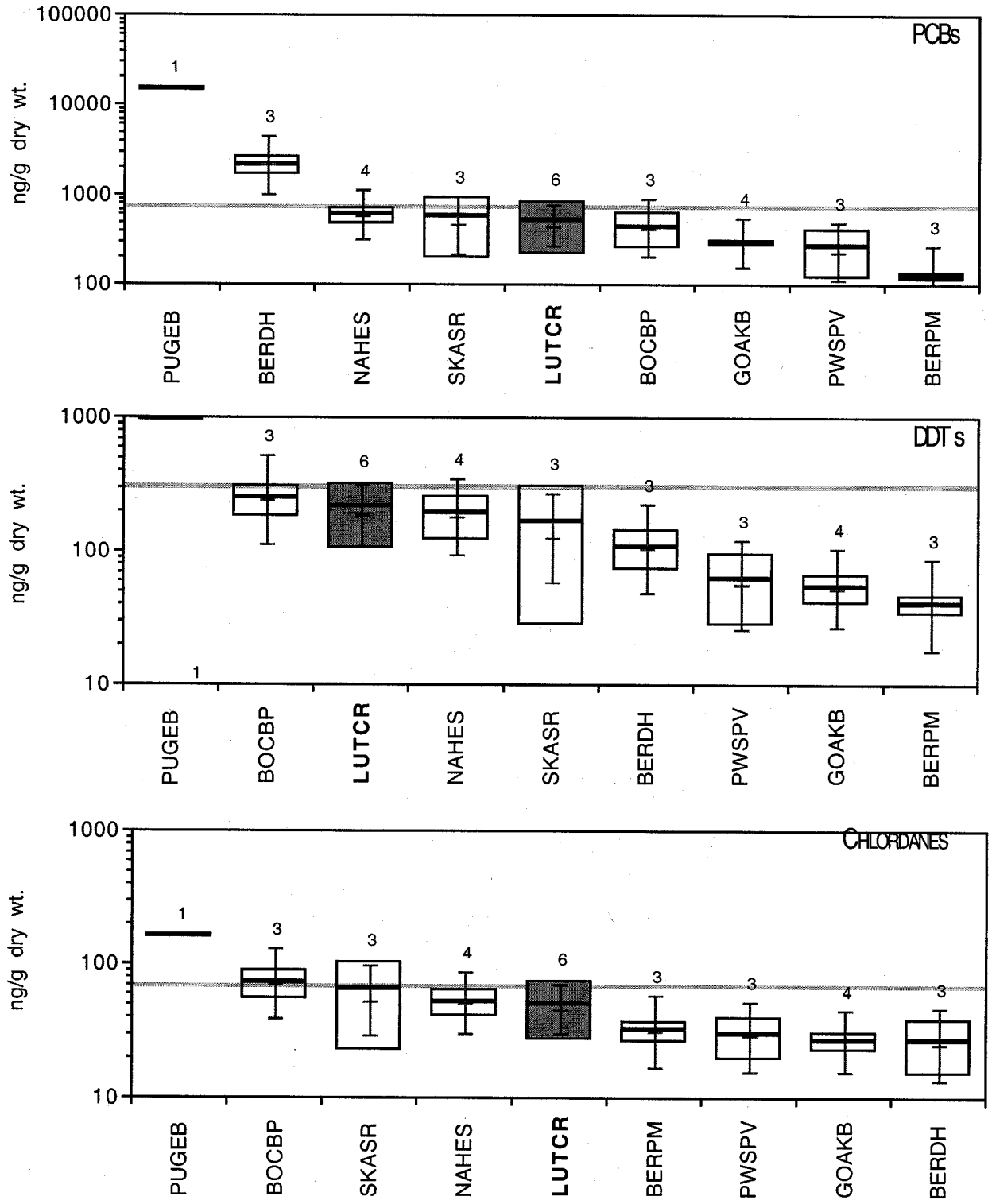


Figure 41 . PCBs, DDTs, and chlordanes in livers of flathead sole.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ▒ ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

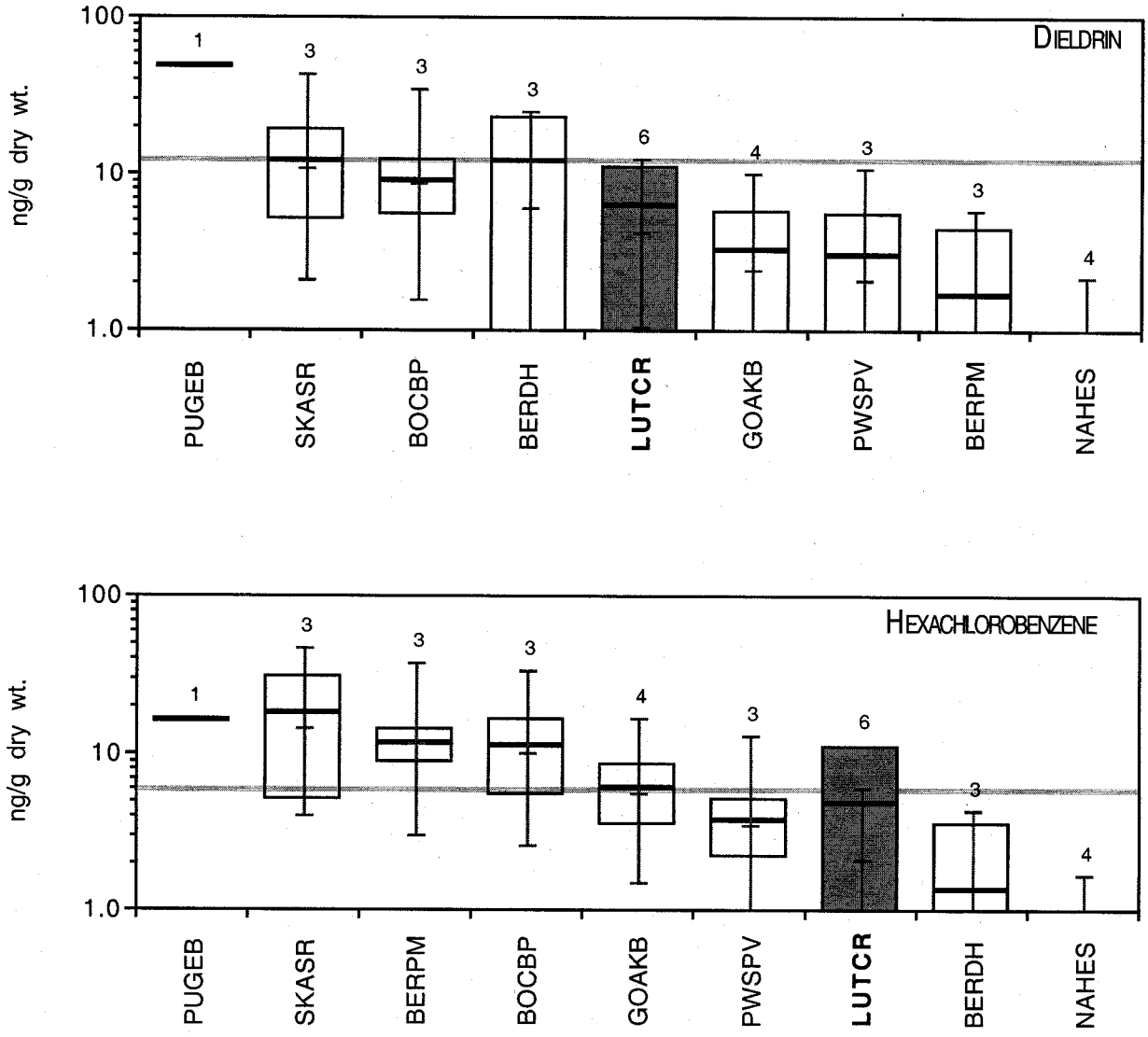


Figure 42. Dieldrin and hexachlorobenzene in livers of flathead sole.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□], geometric mean [-] ± comparison interval [I], and maximum comparison interval value [... ... ...] for the comparison sites [▩]. Sites are ranked in order of geometric mean concentrations along the x-axis (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

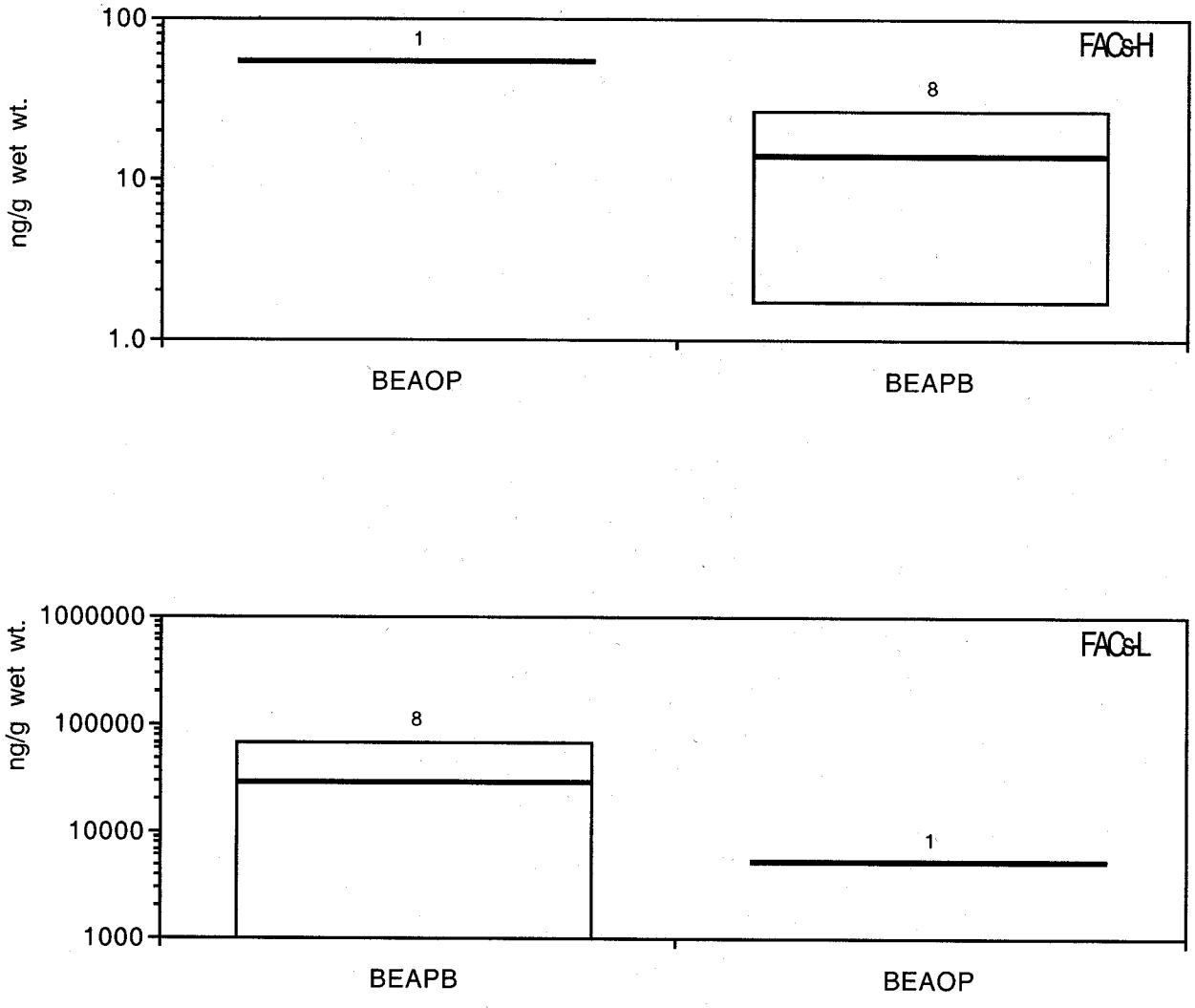


Figure 43. FACs-H and FACs-L in bile of fourhorn sculpin.

Arithmetic mean [▬] concentrations, (ng/g, wet weight) ± 1 standard deviation box [□] (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

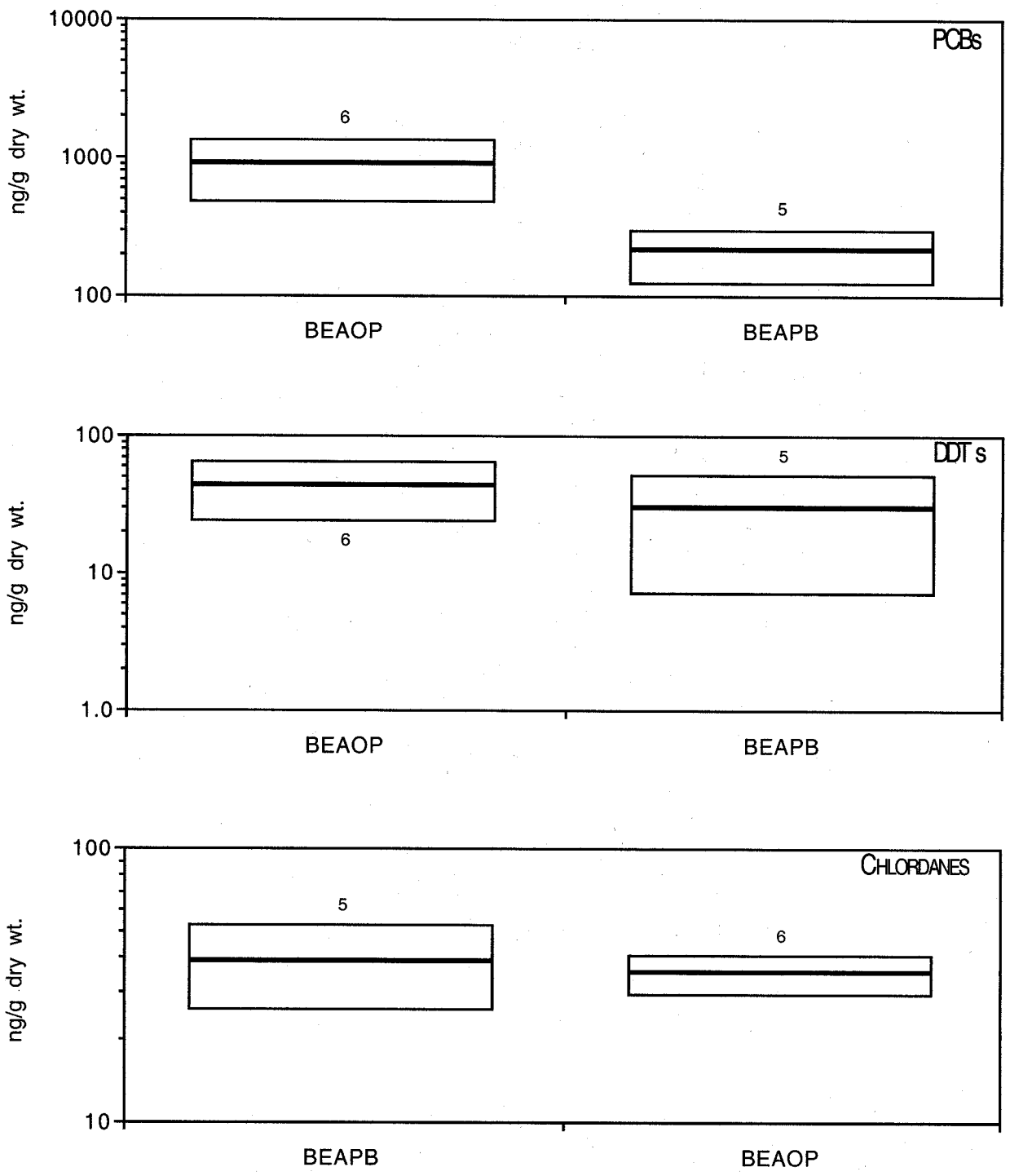


Figure 44. PCBs, DDTs, and chlordanes in livers of fourhorn sculpin.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□] (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

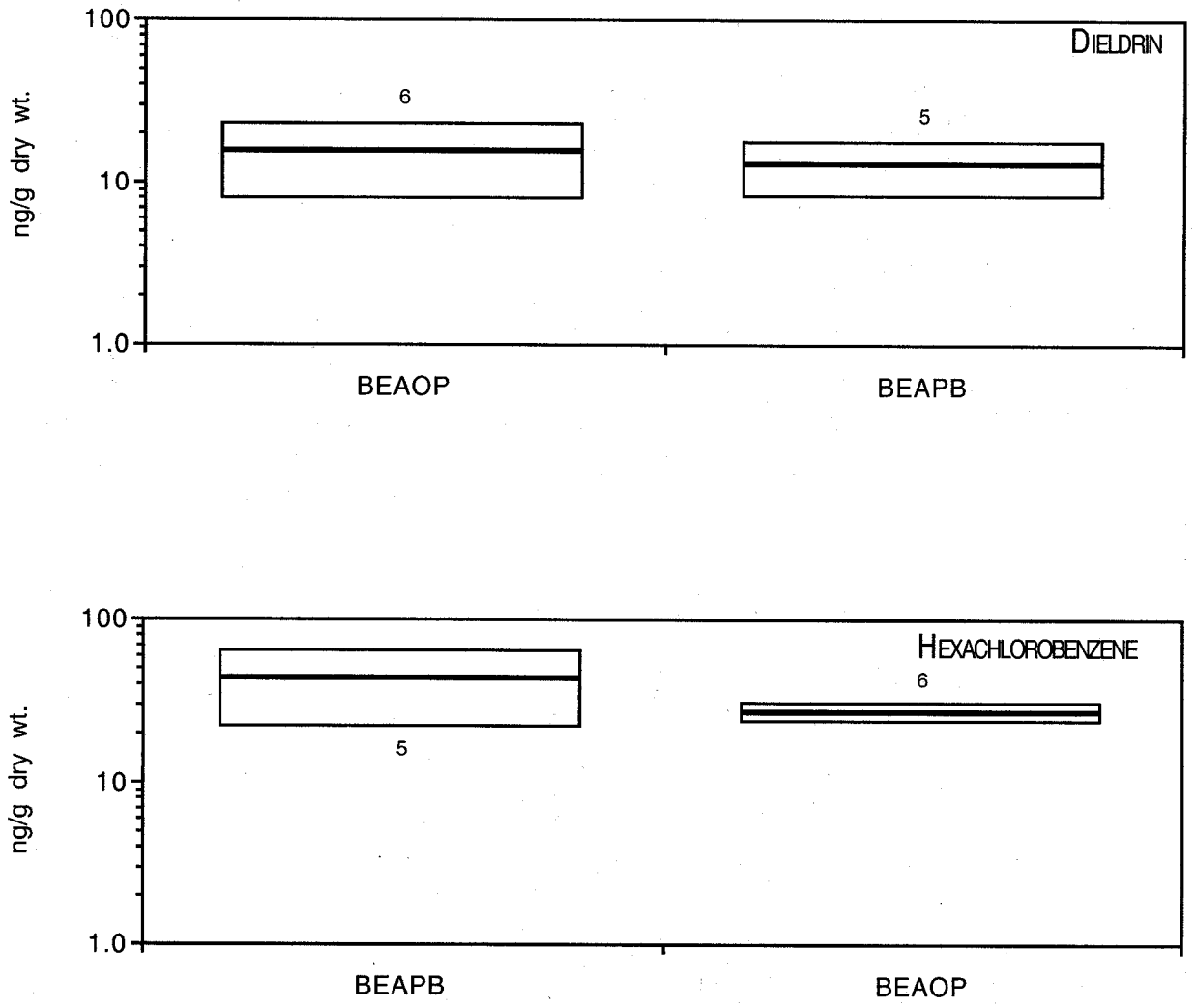


Figure 45. Dieldrin and hexachlorobenzene in livers of fourhorn sculpin.

Arithmetic mean [▬] concentrations, (ng/g, dry weight) ± 1 standard deviation box [□] (see Table 1 for chemical abbreviations and Table 3 for site abbreviations; numbers for each site indicate sample size).

sites contained almost exclusively LAHs (110 ng/g and 99 ng/g, respectively). Concentrations of PCBs were 100 ± 10 ng/g and 230 ng/g, respectively, for these sites; all other organic contaminants were ≤ 16 ng/g.

Fish Diet Composition and Contaminant Uptake

The uptake and fate of chemical contaminants by benthic organisms vary widely among phyla (and in some cases, among taxonomic groups within phyla). For example, rates of cytochrome P-450-dependent monooxidation are slower in molluscs than in arthropods, both *in vivo* and *in vitro* (Varanasi et al. 1985). As a result, metabolism of PAHs is much slower in molluscs than in arthropods. Likewise, metabolism of PAHs is much slower in annelids than in vertebrates (Meador et al. 1995). For example, when fish (English sole), shrimp, molluscs, and amphipods were exposed to the same contaminated sediment for four weeks, the shrimp and fish had no detectable parent PAHs, whereas, during the same period, the molluscs and amphipods accumulated parent PAHs, with the amphipod species accumulating the highest concentration of PAHs (Varanasi et al. 1985). The authors concluded that factors other than metabolism, such as feeding mode and physiological processes, play a role in determining bioaccumulation of chemical contaminants by benthic organisms. Therefore, it is important to determine the taxonomic composition of the food organisms found in stomach contents of fish to better understand the process of food chain transfer of chemical contaminants.

Historically, studies of the food habits of fishes have been conducted to provide information about the interactions between fish populations and prey populations, with the goal of improving management of fish stocks. The present study, however, was undertaken within the context of a national pollution monitoring program, and pertains to the process of food chain transfer of chemical contaminants rather than to fisheries management.

The taxonomic analyses of stomach contents revealed that, with few exceptions, the eight fish species considered had fed primarily on benthic invertebrates (Fig 46). Statistical analyses performed on stomach contents of fish included mean percentages of the prey phyla of the food types of greatest interest, namely arthropods, molluscs, vertebrates (i.e., small fish), and annelid worms, because of their relevance to pollution studies. This section will deal primarily with these four prey phyla.

To assess the uptake and fate of contaminants from prey organisms, the percentages of the two prey phyla which metabolize PAHs (vertebrates and arthropods) were combined, as were the two which do not (molluscs and annelids). When the fish species were plotted according to the average percentages of these two combined categories in their diets (Fig 47), it was evident that English sole and hornyhead turbot consumed primarily non-metabolizers, whereas fourhorn sculpin, barred sand bass, black croaker, and flathead sole consumed primarily metabolizers. White croaker and starry flounder each consumed both categories of prey.

The dietary composition of flathead sole from Dutch Harbor may provide clues which could better define the exposure of this species to LAHs. Flathead sole from this site had the highest bile concentrations of FACs-L found in this species, yet the stomach contents from these fish had no

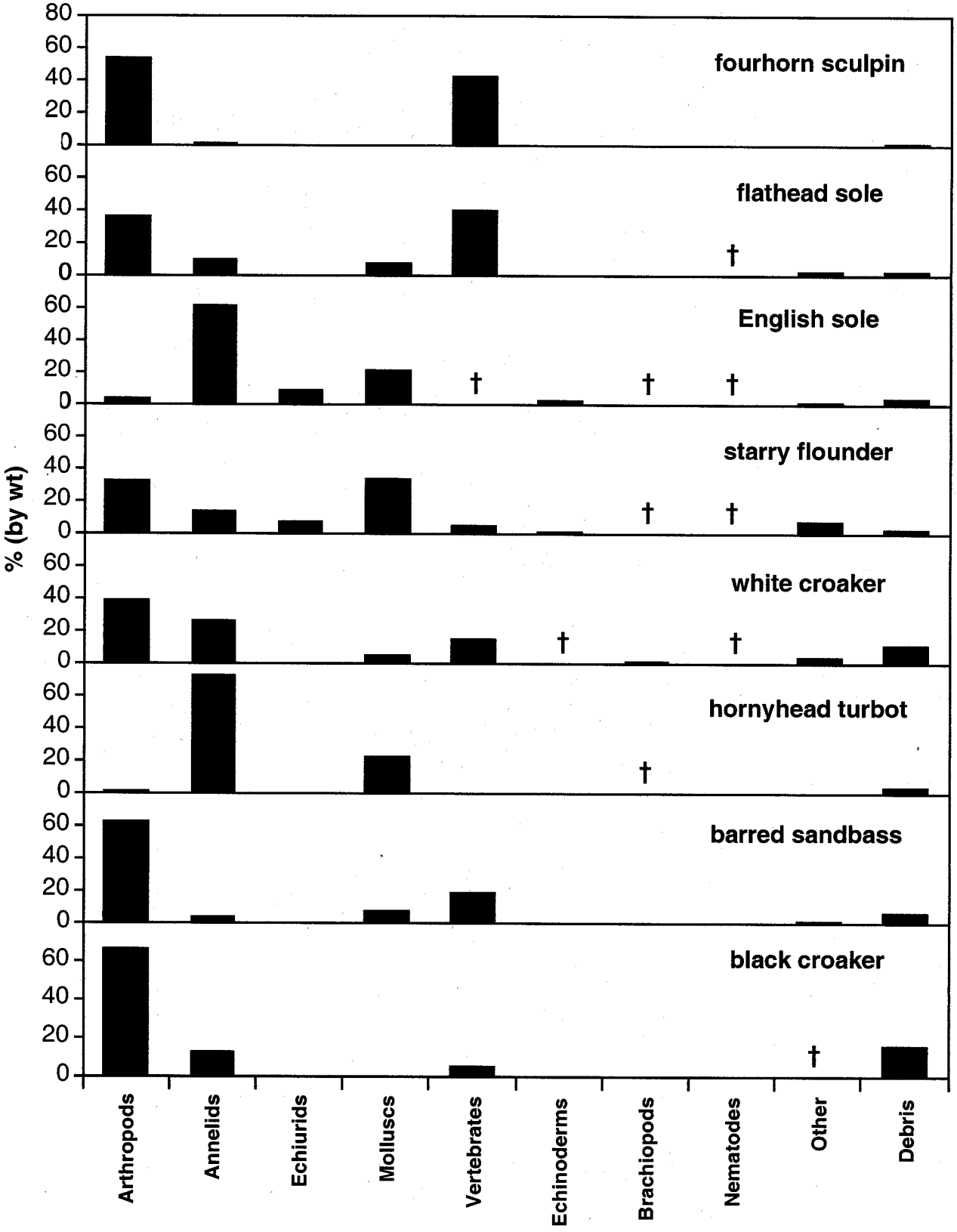


Figure 46. Frequency of occurrence (% by weight) for food types found in stomach contents of eight bottom-feeding fish species collected along the West Coast of North America during the years 1984-88, based on 2 to 33 samples per species. Prey phyla are listed in descending order of overall importance in the diets of the eight species combined; † indicates trace percentage (<0.5%).

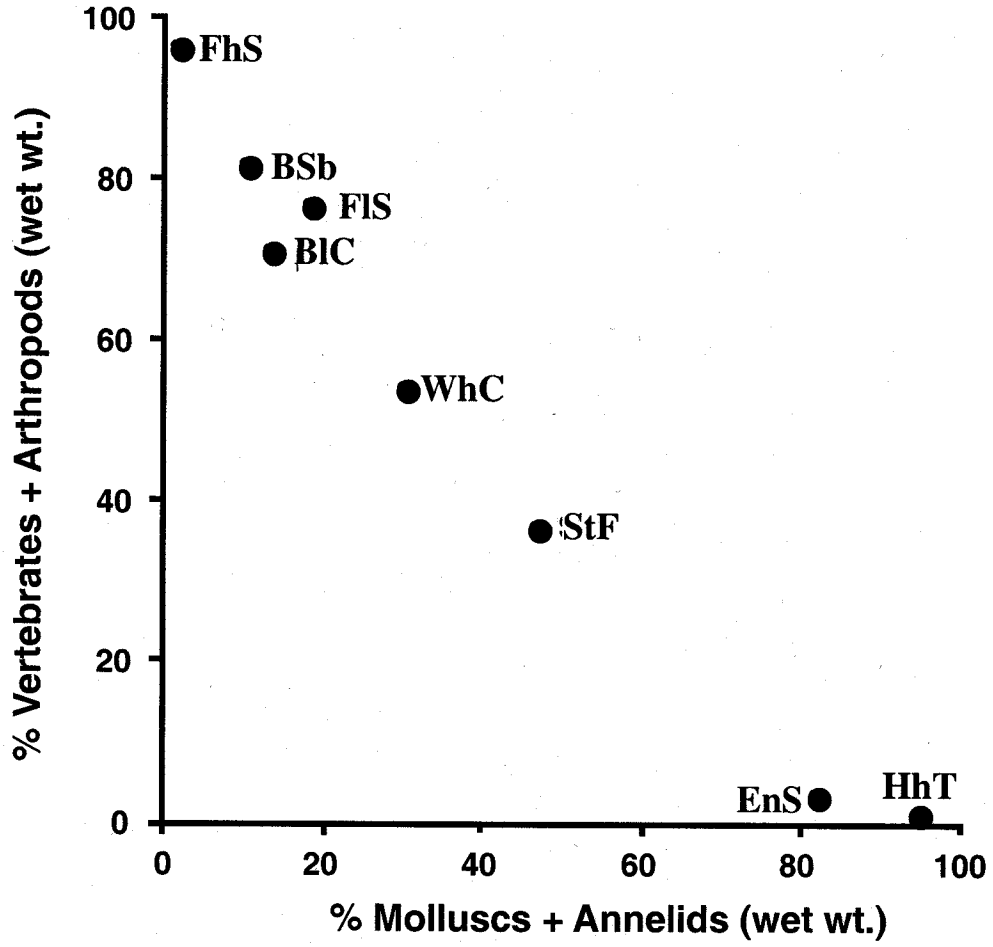


Figure 47. Frequency of occurrence (% by weight) for two major prey categories relating to food-chain transfer of contaminants found in stomach contents of eight species of bottom-feeding fish during the years 1984-88. Vertebrates and arthropods readily metabolize PAHs, whereas mollucs and annelids metabolize PAHs less efficiently. (See Table 3 for fish abreviations).

detectable levels of LAHs. The absence of LAHs in the stomach contents of these fish may have been related in part to the finding that the stomach contents of fish from Dutch Harbor consisted primarily of other fish and arthropods, both of which readily metabolize LAHs to polar compounds that are not detectable by routine GC/MS procedures. As was discussed previously, an explanation for this observation could be that another important route of uptake of LAHs for flathead sole in Dutch Harbor was via the water column.

Statistical Correlations

Spearman's rank correlations (Zar 1984) were carried out for the concentrations of the major categories of organic contaminants in the following comparisons for all species combined: sediments vs. tissues (liver and bile), sediments vs. stomach contents, and stomach contents vs. liver or bile. In addition, similar correlations have been investigated for the six target species of fish for which the number of sites sampled was large enough: flathead sole, English sole, starry flounder, white croaker, hornyhead turbot, and barred sand bass. Data used for these correlations were taken from analyses of samples collected during Cycles I-V.

All species

Highly significant correlations ($p \leq 0.0001$) were found between concentrations of PCBs, chlordanes, DDTs, and dieldrin, respectively, in sediment and levels of these compounds in fish livers of all target species combined (Table 4). Similar strong associations ($p \leq 0.0001$) were found between both HAHs in sediment vs. FACs-H in bile and LAHs in sediment vs. FACs-L in bile, respectively. Concentrations of PCBs, HAHs, DDTs, PAHs, dieldrin, and chlordanes in stomach contents were also highly correlated ($p<0.001$ to $p<0.0001$) with those in sediments, liver and/or bile (Table 4).

In addition, strong positive correlations were also found between concentrations of chemicals in sediment and those in fish tissues when selected species were tested individually (Tables 5-10). These correlations were less significant than those for all species combined, due in part to smaller sample sizes.

Barred sand bass

For this species, a positive correlation ($p \leq 0.01$) was found between sediment levels vs. liver levels of PCBs (Table 5). At a lower level of significance, positive correlations ($p \leq 0.05$) were also found between sediment vs. bile or liver levels of LAHs and chlordanes, respectively.

White croaker

For white croaker, strong positive correlations ($p \leq 0.0001$) were found between sediment and stomach contents levels of PCBs, DDTs, and HAHs; stomach contents and bile or liver levels of PAHs, PCBs, DDTs, and HAHs; and sediment and bile or liver levels of PCBs and FACs-H, respectively (Table 6). Positive correlations of lower significance ($p \leq 0.001$) were found between stomach

Table 4. Spearman rank correlations among concentrations of chemicals in sediment, stomach contents, and liver or bile. Correlations for fish tissue are for all eight major species[a] combined.

Chemical class	Sediment vs. stomach contents[b]	Sediment vs. liver or bile[c]	Stomach contents vs. liver or bile[d]
LAHs	0.39****	0.52****	0.36***
HAHs	0.68****	0.60****	0.54****
PAHs	0.61****	0.49****	0.56****
PCBs	0.72****	0.77****	0.76****
DDTs	0.63****	0.57****	0.69****
Chlordanes	0.50****	0.68****	0.52****
Dieldrin	0.60****	0.56****	0.53****
Hexachlorobenzene	0.21*	0.47****	0.28**

[a] Species are: four-horn sculpin, flathead sole, English sole, starry flounder, white croaker, hornyhead turbot, barred sand bass, and black croaker.

[b] Stomach contents values were for individual composite samples; sediment values were site means for a given year based on three samples; n = 100 for PAHs and 102 for chlorinated compounds.

[c] Sediment values were site means; liver values were species means for specific sites for a given year based on three composite samples; n = 118 for PAHs and 106 for chlorinated compounds.

[d] Stomach contents values were for individual composite samples; liver values were species means for specific sites for a given year based on three composite samples; n = 94 for PAHs and 102 for chlorinated compounds.

* = $p < 0.05$
** = $p < 0.01$
*** = $p < 0.001$
**** = $p < 0.0001$

Table 5. Spearman rank correlations among concentrations of chemicals in sediment, stomach contents, and liver or bile of barred sand bass.

Chemical class	Sediment vs. stomach contents[a]	Sediment vs. liver or bile[b]	Stomach contents vs. liver or bile[c]
LAHs	0.32	0.69*	-0.11
HAHs	0.55	0.66	0.11
PAHs	0.53	0.32	0.42
PCBs	0.65	0.83**	0.61
DDTs	-0.35	-0.58	0.32
Chlordanes	0.31	0.75*	0.01
Dieldrin	0.46	0.34	0.30
Hexachlorobenzene	0.50	0.23	0.57

[a] Stomach contents values were for individual composite samples; sediment values were site means for a given year based on three samples; n = 7 for PAHs and 10 for chlorinated compounds.

[b] Sediment values were site means; liver values were species means for specific sites for a given year based on three composite samples; n = 10 for PAHs and 10 for chlorinated compounds.

[c] Stomach contents values were for individual composite samples; liver values were species means for specific sites for a given year based on three composite samples; n = 7 for PAHs and 10 for chlorinated compounds.

* = $p < 0.05$
** = $p < 0.01$
*** = $p < 0.001$
**** = $p < 0.0001$

Table 6. Spearman rank correlations among concentrations of chemicals in sediment, stomach contents, and liver or bile of white croaker.

Chemical class	Sediment vs. stomach contents[a]	Sediment vs. liver or bile[b]	Stomach contents vs. liver or bile[c]
LAHs	0.22	0.46**	0.53**
HAHs	0.70****	0.62****	0.67****
PAHs	0.66***	0.48**	0.77****
PCBs	0.84****	0.76****	0.74****
DDTs	0.82****	0.59***	0.70****
Chlordanes	0.36	0.61***	0.59***
Dieldrin	0.57**	0.59***	0.64***
Hexachlorobenzene	0.11	0.33	0.18

[a] Stomach contents values were for individual composite samples; sediment values were site means for a given year based on three samples; n = 28 for PAHs and 29 for chlorinated compounds.

[b] Sediment values were site means; liver values were species means for specific sites for a given year based on three composite samples; n = 36 for PAHs and 30 for chlorinated compounds.

[c] Stomach contents values were for individual composite samples; liver values were species means for specific sites for a given year based on three composite samples; n = 28 for PAHs and 29 for chlorinated compounds.

* = $p < 0.05$
** = $p < 0.01$
*** = $p < 0.001$
**** = $p < 0.0001$

contents and liver levels of dieldrin and chlordanes, sediment and liver levels of these same compounds and of DDTs as well, and sediment vs. stomach contents levels of PAHs. Correlations of yet lower significance ($p \leq 0.01$) were found for sediment vs. stomach contents levels of dieldrin, stomach contents vs. bile levels of LAHs, and sediment vs. bile levels of PAHs and LAHs, respectively.

Hornyhead turbot

Positive correlations ($p \leq 0.01$) were found between stomach contents and liver levels of DDTs and PCBs of hornyhead turbot, between sediment vs. liver levels of PCBs, and between sediment vs. stomach contents levels of DDTs, respectively (Table 7). At $p \leq 0.05$, positive correlations were found between sediment vs. liver levels of chlordanes and HCB, and between sediment vs. stomach contents levels of PCBs. Sediment dieldrin concentrations were always below the detection limit at sites where hornyhead turbot were collected, so correlations between sediment and liver or stomach contents could not be performed.

Starry flounder

For this species, a strong positive correlation ($p \leq 0.0001$) was found between sediment and stomach contents levels of dieldrin (Table 8). At a slightly lower level of significance ($p \leq 0.001$), positive correlations were found between sediment and stomach contents levels of DDTs, sediment and liver levels of PCBs, and stomach contents and liver levels of dieldrin. At the $p \leq 0.01$ level of significance, positive correlations were found for sediment vs. stomach contents levels of chlordanes and PCBs, sediment vs. liver levels of DDTs and chlordanes, and stomach contents vs. liver levels of DDTs. Positive correlations of lowest significance ($p \leq 0.05$) were found for sediment vs. stomach contents levels of HAHs and PAHs, sediment vs. liver levels of HCB and dieldrin, and stomach contents vs. liver levels of chlordanes and PCBs.

English sole

For English sole, strong positive correlations ($p \leq 0.0001$) were found between sediment vs. biliary FACs-L, and between sediment and stomach contents levels of HAHs, respectively (Table 9). At somewhat lower levels of significance, positive correlations ($p \leq 0.001$) were also found between sediment and liver levels of the chlordanes, PCBs, and HCB, between sediment and biliary FACs PAH, and between stomach contents vs. bile levels of PAHs and FACs-L, respectively. At the $p \leq 0.01$ level of significance, positive correlations were found for LAHs and PAHs in sediment vs. stomach contents, and DDTs and HAHs in sediment vs. liver or bile. Positive correlations of lowest significance ($p \leq 0.05$) were found for DDTs and HAHs in stomach contents vs. liver or bile.

Flathead sole

For flathead sole, positive correlations ($p \leq 0.05$ to $p \leq 0.01$) were found between sediment and bile levels of HAHs and PAHs, respectively (Table 10). Sample size was relatively small for flathead sole ($n = 9$ to 11). This species was collected in Alaska, where sediment contaminant levels were generally low, so strong correlations between sediment, liver or bile, and stomach contents would not be

Table 7. Spearman rank correlations among concentrations of chemicals in sediment, stomach contents, and liver or bile of hornyhead turbot.

Chemical class	Sediment vs. stomach contents[a]	Sediment vs. liver or bile[b]	Stomach contents vs. liver or bile[c]
LAHs	0.04	0.39	0.36
HAHs	0.36	0.13	0.25
PAHs	0.38	0.14	0.47
PCBs	0.71*	0.81**	0.81**
DDTs	0.79**	0.51	0.82**
Chlordanes	0.42	0.72*	0.43
Dieldrin	--[d]	--[d]	0.39
Hexachlorobenzene	0.17	0.61*	0.56

[a] Stomach contents values were for individual composite samples; sediment values were site means for a given year based on three samples; n = 10 for PAHs and 11 for chlorinated compounds.

[b] Sediment values were site means; liver values were species means for specific sites for a given year based on three composite samples; n = 11 for PAHs and 11 for chlorinated compounds.

[c] Stomach contents values were for individual composite samples; liver values were species means for specific sites for a given year based on three composite samples; n = 10 for PAHs and 11 for chlorinated compounds.

[d] All sediment values for dieldrin below detection limit.

* = $p < 0.05$
** = $p < 0.01$
*** = $p < 0.001$
**** = $p < 0.0001$

Table 8. Spearman rank correlations among concentrations of chemicals in sediment, stomach contents, and liver or bile of starry flounder.

Chemical class	Sediment vs. stomach contents[a]	Sediment vs. liver or bile[b]	Stomach contents vs. liver or bile[c]
LAHs	0.29	0.06	0.10
HAHs	0.50*	0.25	0.31
PAHs	0.48*	0.18	0.21
PCBs	0.53**	0.65***	0.49*
DDTs	0.68***	0.59**	0.62**
Chlordanes	0.62**	0.54**	0.50*
Dieldrin	0.73****	0.42*	0.63***
Hexachlorobenzene	-0.08	0.43*	0.33

[a] Stomach contents values were for individual composite samples; sediment values were site means for a given year based on three samples; n = 25 for PAHs and 25 for chlorinated compounds.

[b] Sediment values were site means; liver values were species means for specific sites for a given year based on three composite samples; n = 26 for PAHs and 26 for chlorinated compounds.

[c] Stomach contents values were for individual composite samples; liver values were species means for specific sites for a given year based on three composite samples; n = 25 for PAHs and 25 for chlorinated compounds.

* = $p < 0.05$
** = $p < 0.01$
*** = $p < 0.001$
**** = $p < 0.0001$

Table 9. Spearman rank correlations among concentrations of chemicals in sediment, stomach contents, and liver or bile of English sole.

Chemical class	Sediment vs. stomach contents[a]	Sediment vs. liver or bile[b]	Stomach contents vs. liver or bile[c]
LAHs	0.76**	0.75****	0.82***
HAHs	0.91****	0.56**	0.58*
PAHs	0.71**	0.68***	0.83***
PCBs	0.48	0.85***	0.57
DDTs	0.48	0.74**	0.59*
Chlordanes	0.50	0.86***	0.33
Dieldrin	0.48	-0.09	-0.18
Hexachlorobenzene	0.36	0.81***	0.45

[a] Stomach contents values were for individual composite samples; sediment values were site means for a given year based on three samples; n = 12 for PAHs and 12 for chlorinated compounds.

[b] Sediment values were site means; liver values were species means for specific sites for a given year based on three composite samples; n = 21 for PAHs and 13 for chlorinated compounds.

[c] Stomach contents values were for individual composite samples; liver values were species means for specific sites for a given year based on three composite samples; n = 12 for PAHs and 12 for chlorinated compounds.

* = $p < 0.05$
** = $p < 0.01$
*** = $p < 0.001$
**** = $p < 0.0001$

Table 10. Spearman rank correlations among concentrations of chemicals in sediment, stomach contents, and liver or bile of flathead sole.

Chemical class	Sediment vs. stomach contents[a]	Sediment vs. liver or bile[b]	Stomach contents vs. liver or bile[c]
LAHs	0.04	0.55	0.10
HAHs	0.56	0.69*	0.55
PAHs	-0.04	0.78**	0.10
PCBs	0.09	0.22	0.42
DDTs	-0.20	-0.01	0.47
Chlordanes	-0.47	0.52	0.02
Dieldrin	--[d]	--[d]	-0.17
Hexachlorobenzene	0.56	0.31	0.16

[a] Stomach contents values were for individual composite samples; sediment values were site means for a given year based on three samples; n = 9 for PAHs and 10 for chlorinated compounds.

[b] Sediment values were site means; liver values were species means for specific sites for a given year based on three composite samples; n = 11 for PAHs and 10 for chlorinated compounds.

[c] Stomach contents values were for individual composite samples; liver values were species means for specific sites for a given year based on three composite samples; n = 9 for PAHs and 10 for chlorinated compounds.

[d] All sediment values for dieldrin below detection limit.

* = $p < 0.05$
** = $p < 0.01$
*** = $p < 0.001$
**** = $p < 0.0001$

expected. In fact, sediment dieldrin concentrations were so low (always below the detection limit) that correlations between sediment and liver or stomach contents could not be performed.

Summary for fish species

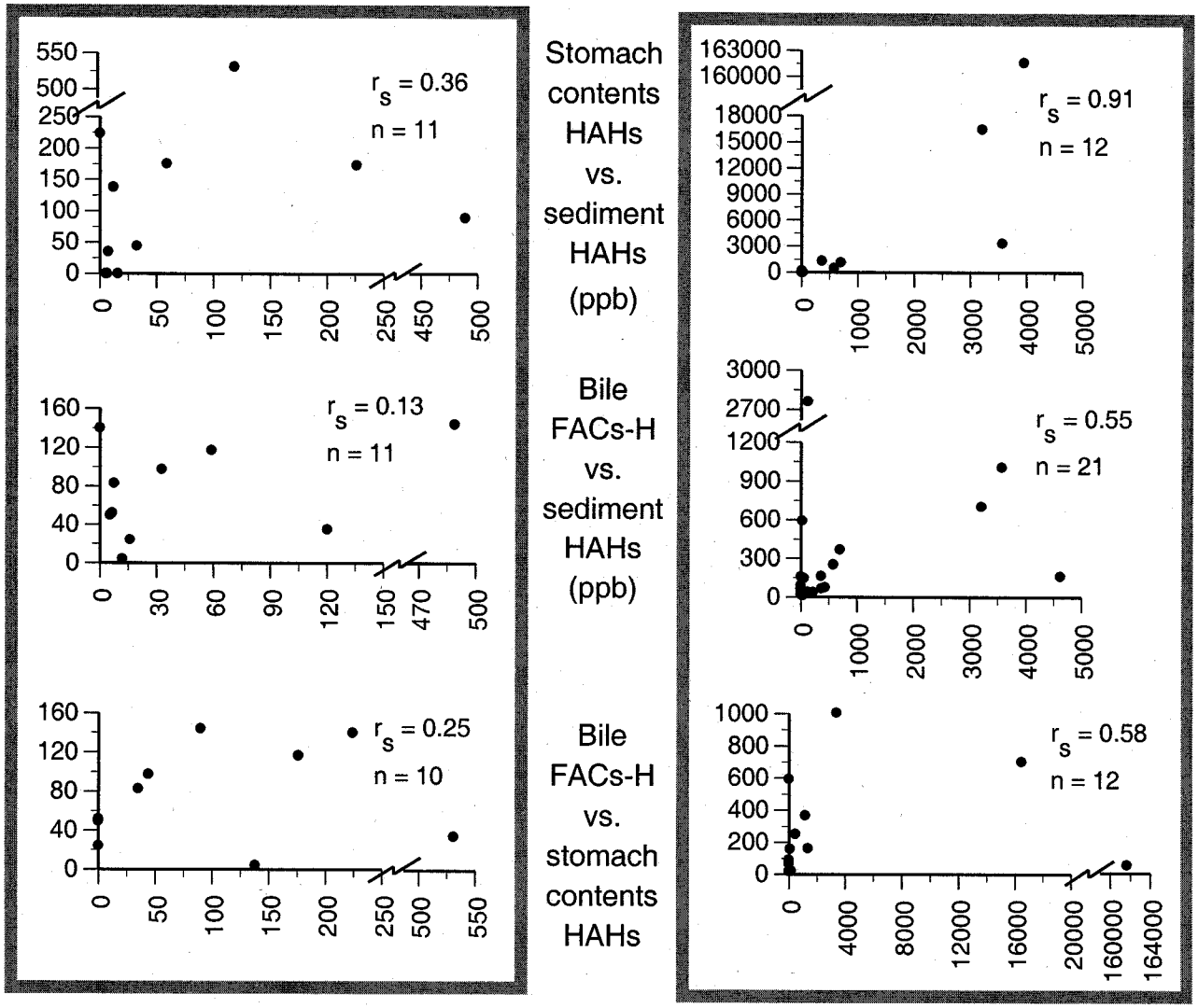
All statistically significant correlations ($p \leq 0.05$) for individual species were positive. White croaker had the largest number of statistically significant correlations ($p \leq 0.01$, 19 out of 24 categories), followed by starry flounder with 15, English sole with 14, hornyhead turbot with 7, barred sand bass with 3, and flathead sole with 2. The number of significant correlations per species is only partially a reflection of the number of samples collected. For example, more than twice as many samples of starry flounder were collected as were obtained of English sole, but English sole had almost as many significant correlations as did starry flounder.

Of the significant correlations ($p \leq 0.05$) for individual species, 27 were for sediment vs. liver or bile, 17 were for stomach contents vs. liver or bile, and 16 were for sediment vs. stomach contents. PCBs had the largest number of correlations in the individual species (11 out of 18 correlations), followed by DDTs (10), chlordanes (8), PAHs (8), HAHs (8), and LAHs (6). Thus persistent compounds tended to show more significant correlations than those more readily metabolized.

Although interspecies comparisons of tissue levels of chemicals are complicated by potential species-specific differences, these very strong correlations demonstrate the important relationships between concentrations of toxic chemicals in sediments and the uptake of these chemicals by sediment-associated organisms. This relationship is clearly apparent upon examination of the data. For example, the sites with the highest sediment concentrations of PCBs and PAHs (south San Diego Bay and Elliott Bay) were also the sites that had fish species with the highest concentrations of PCBs in liver tissues and FACs-H in bile (barred sand bass and English sole, respectively).

Summary Analysis of Statistical Correlations

Contaminant concentrations in environmental compartments (sediment, stomach contents and liver or bile) were in many cases highly intercorrelated for each bottom-dwelling fish species. To the extent that sediment, stomach contents, and liver or bile levels are intercorrelated for a given species, measurement of any one compartment provides a good index of levels of environmental contaminants. Because a large number of relationships among species, chemical classes, and environmental compartments could be examined, it is useful to summarize the results given in Tables 4-10 in another way. Correlations among the chemical concentrations measured in the three environmental compartments are summarized graphically in Fig 48 for two species, hornyhead turbot and English sole. The top part of the figure shows pairwise scattergrams of the concentrations of FACs-H (bile) and HAHs (sediment, stomach contents) for each of the two species. To provide a graphical comparison between species, the 3 correlation coefficients computed for each species are plotted on a 3-dimensional graph at the bottom of the figure. Each of the compartment pairs is associated with one axis of the graph (i.e., each species is represented by a separate point). The correlation coefficients for bile FACs-H vs. stomach contents



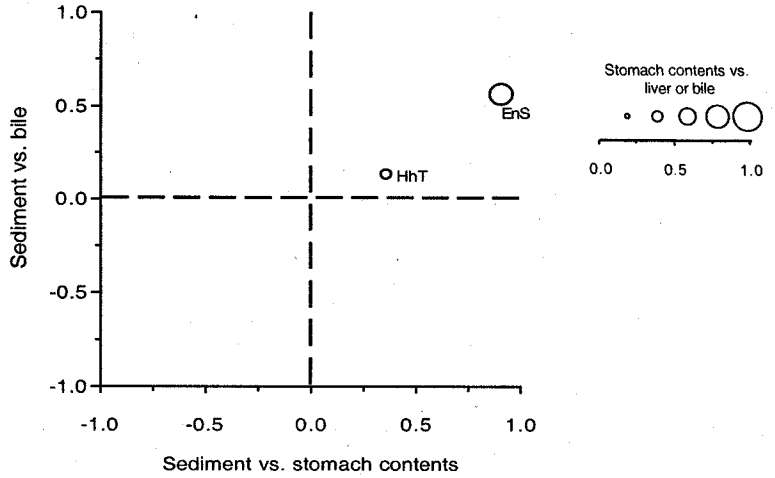


Figure 48. Summary analysis illustrating example of a species exhibiting high intercorrelations among types of environmental compartments for HAHs (English sole) and one showing low intercorrelations for the same class of contaminants (hornyhead turbot). Correlations are assessed using the Spearman rank correlation coefficient (r_s) based on annual site means for all samples of a given type. See Table 3 for species abbreviations.

HAHs) are plotted on the axis perpendicular to the page, and the values are represented by the size of the plotted circle. The graph shows that sediment and stomach contents levels of HAHs and those of bile FACs-H are highly correlated ($rs > 0.50$ in all pairs) for English sole. In contrast, concentrations in the three compartments are not well correlated for hornyhead turbot ($rs < 0.40$ in all cases).

Summary analyses for the chemical classes are given in Figures 49-51. For LAHs, HAHs, and PAHs, considered separately, English sole exhibited consistently high correlations among sediment, stomach contents, and bile (Fig 49). In comparison, all other species showed lower intercorrelations for LAHs, but only hornyhead turbot and starry flounder showed consistently low correlations for HAHs. For total PAHs, starry flounder was unusual in having a high correlation between sediment and stomach contents levels, but low correlations between sediment or stomach contents levels and bile levels.

For PCBs, all species except flathead sole had relatively high correlations among sediment, stomach contents, and liver levels. In contrast, HCB levels in one compartment did not provide a good prediction of levels in another (Fig 50). For the pesticides, levels of DDTs among the three compartments were generally intercorrelated except in the cases of flathead sole and barred sand bass. Levels of chlordanes were not highly intercorrelated for most species, and for dieldrin intercorrelations were particularly poor for English sole and flathead sole (Fig 51).

Seven of the sites sampled contained sediments with < 20% silt-clay, indicating predominance of sand and gravel. These types of sediments are less likely to accumulate as high a level of chemical contaminants than are sediments with more silt-clay (Means et al. 1980). Whereas some of these sites with < 20% silt-clay had low concentrations of many contaminants, these sites are primarily nonurban sites that receive smaller amounts of chemicals from anthropogenic sources. Such nonurban sites were needed as comparison sites, as it was difficult to find nonurban sites located in muddy areas. Moreover, when the sites with < 20% silt-clay were eliminated from the correlations between levels of chemicals in sediments, stomach contents and tissues, the strength of the correlations was not adversely affected. This suggests that even though a few of the sampling sites had both < 20% clay-silt and low levels of sediment-associated contaminants, the sediment contaminants concentrations generally reflected the target fish species exposure.

Statistical Analysis of Trends

Trend analysis was conducted for sites which had at least four years of analytical data covering a span of five years or more. The trend analysis was a two-step process: the first step of the process involved Spearman rank correlations of contaminant concentrations for each of three types of “compartments” (sediment, liver/bile and stomach contents of each fish species) for each of 12 sites that met the above mentioned criteria. (See Appendix A for a listing of the results of these correlations.). The second step consisted of meta-analysis to consider changes in contaminant concentrations over time in all compartments measured at a site. The results of the meta-analyses will be presented in this section. There are several advantages to considering all three compartments simultaneously. Sediment data are available for each site for all years it was sampled because sediment can always be collected, whereas tissue samples were either not always collected or were collected from different species during some

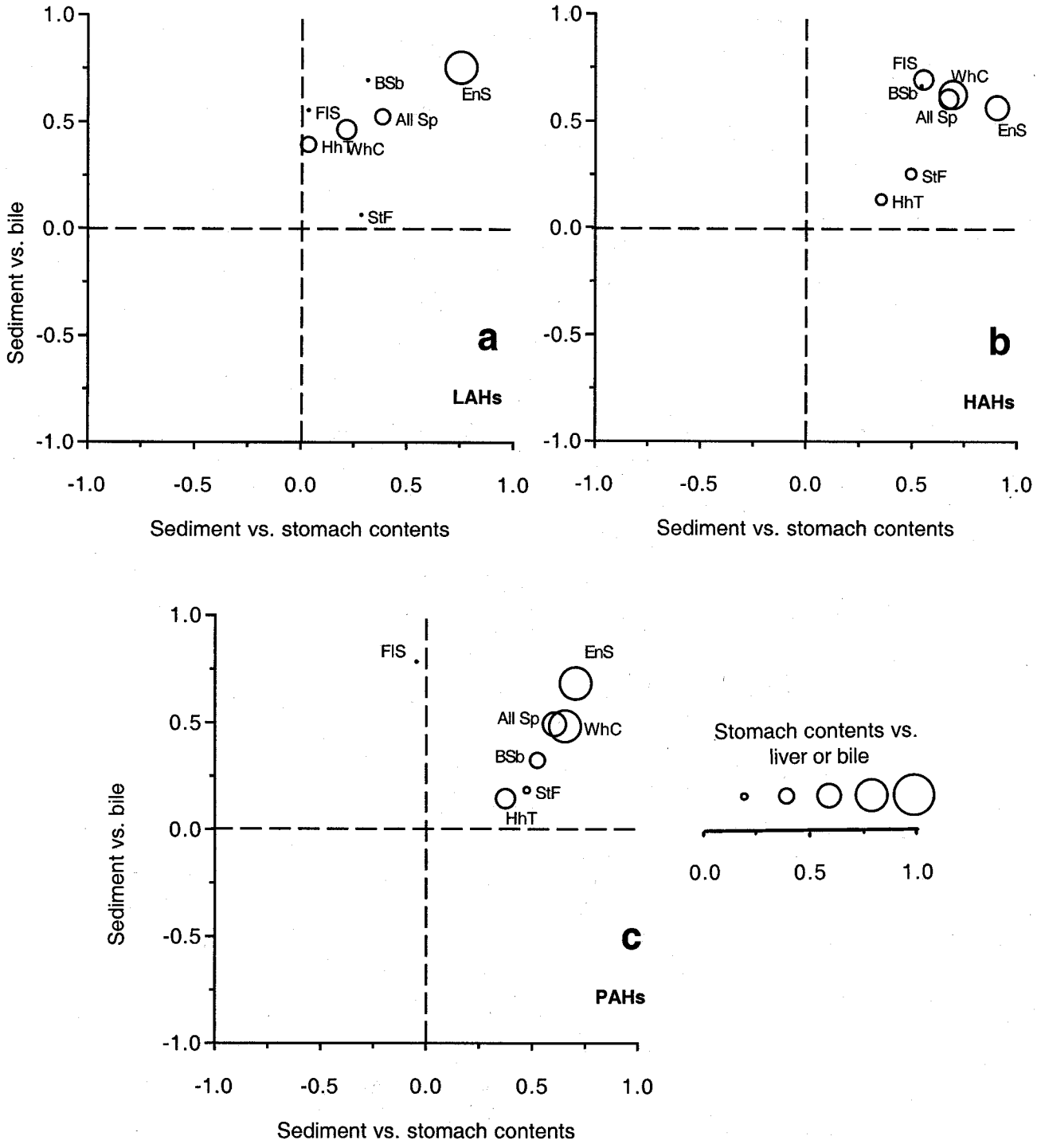


Figure 49. Summary analysis showing intercorrelations among Spearman rank correlation coefficients for chemical concentrations in sediment, bile, and stomach contents for (a) LAHs, (b) HAHs, and (c) PAHs for six of the eight species of bottom-dwelling fish separately and for all eight species combined (fourhorn sculpin and black croaker are omitted due to small sample size; see Table 3 for species abbreviations).

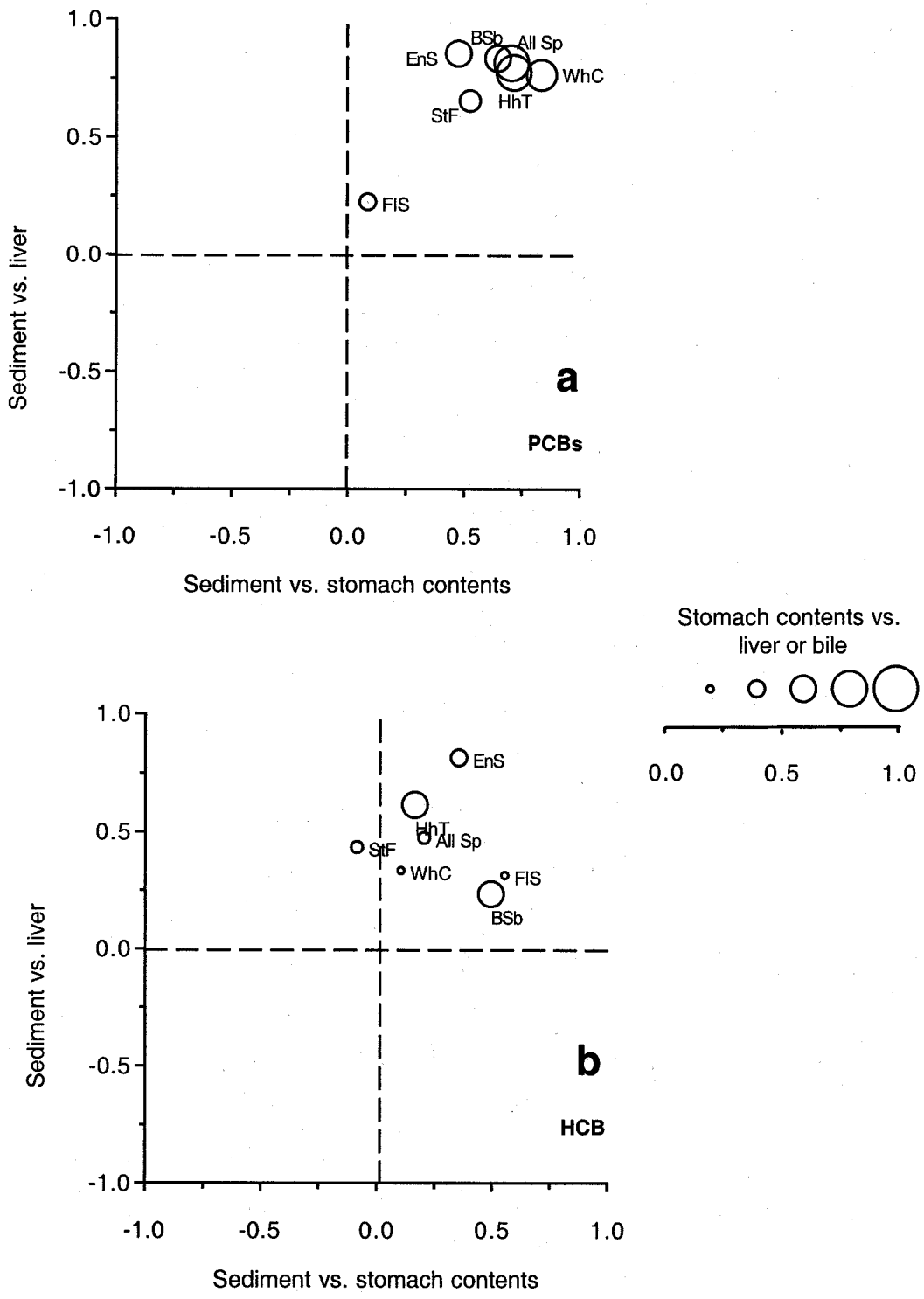


Figure 50. Summary analysis showing intercorrelations among Spearman rank correlation coefficients for chemical concentrations in sediment, liver, and stomach contents for (a) PCBs and (b) hexachlorobenzene for six of the eight species of bottom-dwelling fish separately and for all eight species combined (fourhorn sculpin and black croaker are omitted due to small sample size; see Table 3 for species abbreviations).

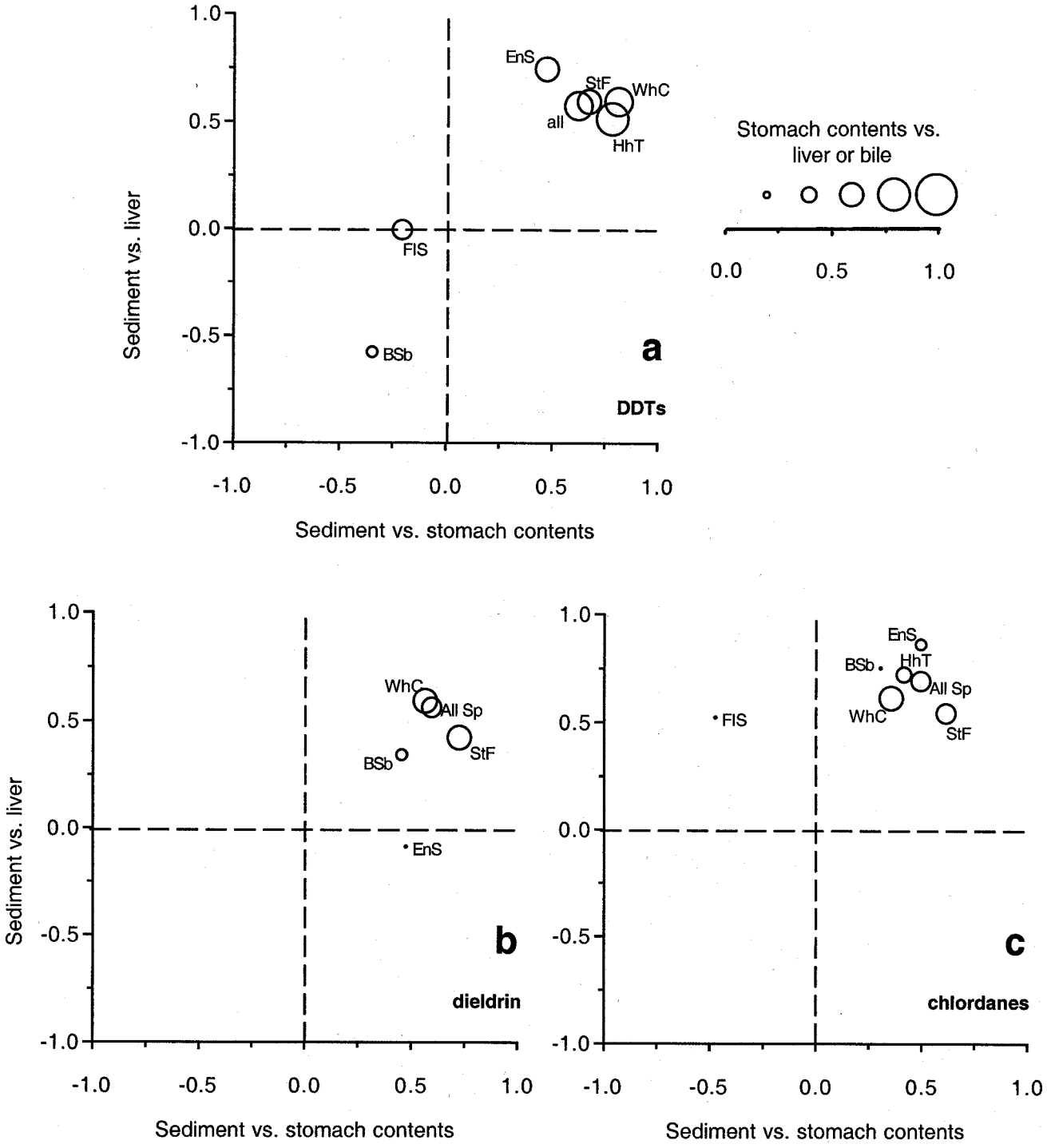


Figure 51. Summary analysis showing intercorrelations among Spearman rank correlation coefficients for chemical concentrations in sediment, liver, and stomach contents for (a) DDTs, (b) dieldrin, and (c) chlordanes for six of the eight species of bottom-dwelling fish separately and for all eight species combined (fourhorn sculpin and black croaker are omitted due to small sample size; see Table 3 for species abbreviations).

years at certain sites due to population fluctuations. Tissue, on the other hand, reflects bioavailability and is not subject to bioturbation. Furthermore, a trend detected in one compartment at a site using Spearman rank correlation is more convincing if a similar trend is also evident in other compartments at that site.

Trends were assessed for chlordanes, dieldrin, DDTs, HAHs, LAHs, and PCBs. Because concentrations of HCB in sediment and tissues were near or below the limits of detection at most sites, trends for HCB will not be reported. Of the 72 possible trends (12 sites and 6 chemical classes), 29% (21 of 72) showed an increasing trend, 53% (38 of 72) showed no trend in concentrations, and 18% (13 of 72) showed decreasing concentrations (Table 11). The highest number of increasing trends was among the PAHs, eight for HAHs and five for LAHs, followed by dieldrin (4), chlordanes (3), DDTs (1) and PCBs (2). Among the decreasing trends, chlordanes, PCBs and DDTs each had three, with dieldrin having two and LAHs one.

Sites with positive trends for concentrations of PAHs were located in both nonurban and urban areas. All three of the nonurban reference sites in the contiguous U.S.—Dana Point, Bodega Bay and Nisqually Reach —had significant increases in HAHs, LAHs, or both. The urban sites with increasing trends for these PAHs included Hunters Point and Southampton Shoal in San Francisco Bay, Long Beach and Outer Harbor in San Pablo Bay, and Commencement Bay in Puget Sound. The Coos Bay site also had increasing concentrations of chlordanes and DDTs. Positive trends for chlordanes were observed at Southampton Shoal, and increases in dieldrin concentrations were found for the Dana Point, South San Diego Bay, Coos Bay and Elliott Bay sites. The only sites with increasing concentrations of PCBs were both in Puget Sound—Commencement Bay and Elliott Bay.

The Dana Point reference site had the highest number of contaminants with decreasing trends, including chlordanes, DDTs, and PCBs. Decreasing trends for dieldrin and DDTs were also found at the Hunters Point site, and for PCBs and LAHs at the South San Diego Bay site. The only other decreasing trends were found for the following individual contaminant classes at single sites: dieldrin (Bodega Bay), DDTs (Hunters Point), chlordanes (Nisqually Reach), PCBs (San Pedro Bay Outer Harbor), and chlordanes (West Santa Monica Bay).

In those 38 cases for which temporal trends in contaminant concentration were not detected, high natural variability and sample size constraints may have resulted in insufficient statistical power to detect underlying temporal gradients. The importance of resolving this question points to the need for continued collection of chemical data to increase the number of observations per site and thereby reduce the statistical uncertainty potentially masking temporal trends. Moreover, the persistence of CHs and the consistent increases in concentrations of PAHs, which are persistent in certain abiotic compartments (e.g., sediments), should be of concern to resource managers, because research is showing linkages between exposures of marine organisms to these chemicals and a variety of pathological conditions (Myers et al. 1994, Myers et al. 1998) and adverse effects, including impaired reproduction (Johnson et al. 1998), growth (Casillas et al. 1995) and immunocompetence (Arkoosh et al. 1998).

The temporal trends found in this project differ in some respects from those recently reported for concentrations of the same classes of chemicals in mussel (*Mytilus edulis*) tissues collected and

Table 11. Summary of trend analyses: meta-analysis results for organic contaminants by site for the 12 long-term sites during the period 1984-90. Chemical and site abbreviations are given in Table 3. All Spearman rank correlations are included in the Appendix.

Site	Chlordanes			Dieldrin			∑DDTs			∑HAHs			∑LAHs			∑PCBs		
BODNO	(+)	0.1145		(-)	0.0433	*	(-)	0.2355		(+)	0.0001	****	(+)	0.0001	****	(+)	0.0571	
PUGCB	(+)	0.1961		(+)	0.4152		(+)	0.2235		(+)	0.0001	****	(+)	0.0640		(+)	0.0001	****
COONB	(+)	0.0001	****	(+)	0.0001	****	(+)	0.0445	*	(-)	0.2449		(+)	0.3529		(+)	0.1720	
DANOU	(-)	0.0001	****	(+)	0.0001	****	(-)	0.0191	*	(+)	0.0001	****	(+)	0.0001	****	(-)	0.0001	****
PUGEB	(+)	0.2673		(+)	0.0058	**	(+)	0.1997		(-)	0.0808		(-)	0.3278		(+)	0.0016	**
SFBHP	(-)	0.0768		(-)	0.0001	****	(-)	0.0061	**	(+)	0.0001	****	(+)	0.0001	****	(+)	0.1173	
SPBLB	(-)	0.0546		(-)	0.0554		(-)	0.0287	*	(+)	0.0012	**	(+)	0.0178	*	(+)	0.1389	
SFBSS	(+)	0.0199	*	(-)	0.4797		(+)	0.1135		(+)	0.0001	****	(-)	0.1905		(+)	0.0753	
PUGNR	(-)	0.0309	*	(+)	0.3524		(-)	0.2922		(+)	0.0460	*	(+)	0.0840		(-)	0.2104	
SPBOH	(+)	0.3398		(+)	0.0957		(-)	0.1293		(+)	0.0001	****	(+)	0.0001	****	(-)	0.0004	***
SDBTE◇	(-)	0.1269		(+)	0.0057	**	(+)	0.4297		(-)	0.2990		(-)	0.0002	***	(-)	0.0001	****
SMBWE◇	(-)	0.0004	***	(+)	0.0755		(+)	0.1313		(+)	0.1042		(+)	0.2663		(-)	0.2637	

Total sites with trends (p<0.05)							Totals
positive	2	4	1	7	5	2	21
negative	3	2	3	1	1	3	13
Total sites with no trend (p>0.05)							
	7	6	8	4	6	7	38

**** = $p < 0.0001$
*** = $p < 0.001$
** = $p < 0.01$
* = $p < 0.05$

◇ Spearman rank correlation coefficients did not meet our criteria for inclusion in meta analysis for FACs at SDBTE or for sediment at SMBWE or PUGNR.

analyzed between approximately 1986 and 1993 (O'Connor 1996). Spearman rank correlations were conducted for 44 sites on the West Coast of the U.S. and significant ($p \leq 0.05$) negative temporal trends in mussel concentrations of DDTs and PCBs were found at three and four sites, respectively; no positive trends were found. No significant trends for PAHs were reported. These differences from the findings of the present study may be due to a number of factors, including the fact that O'Connor (1996) examined trends in mussel tissues only, whereas the present study examined trends in sediment and fish. Mussels are suspension feeders and are generally located around the periphery of embayments attached to rocks and other structures, whereas the target fish used in this project are bottom feeders associated with areas of embayments where contaminated sediments are generally deposited.

SUMMARY

The results presented in this report parallel in many ways those reported in previous reports (Varanasi et al. 1988, 1989b) on the first three years of the West Coast component of the NBSP. These reports included data for 31 West Coast sites sampled between 1984 and 1986, whereas data for four additional years for 50 sites (the original 31 sites plus 19 additional sites) are reported here. These additional years of data and sites dramatically strengthened the statistical power of the current data analyses. For example, in Varanasi et al. (1989b) concentrations of HAHs in sediments were significantly higher at only two sites compared to the Dana Point reference site. However, in the present report, 27 sites had significantly higher concentrations of HAHs than did the Dana Point site. Similarly, whereas they reported sediment concentrations of PCBs significantly higher at five sites relative to Dana Point, in the present report 24 sites had significantly higher concentrations. Two factors were likely major contributors to the differences between these reports. The effects of intrasite variability in chemical concentrations, so critical to intersite statistical significance in the first three years of this study (Varanasi et al. 1989b), diminished as more analyses accumulated in the succeeding four years. The increased numbers of samples tended to reduce the intrasite variability and provided more accurate appraisals of between-site differences. Also, the log transformed chemistry data presented in GT-2 plots in this report visually facilitated the drawing of statistical distinctions between the highly contaminated and minimally contaminated sites.

In general, results of the first seven years of the Pacific Coast portion of the NBSP demonstrated that the highest levels of organic contaminants were found in sites in San Diego Bay, San Pedro Bay (Los Angeles and Long Beach), Santa Monica Bay, San Francisco Bay, and Elliott Bay (Seattle). Intermediate levels of pollution were found in Monterey Bay, San Pablo Bay, and Commencement Bay (Tacoma), whereas sites in Oregon and Alaska were among the least polluted.

In addition to defining the geographical distribution and levels of contaminants at selected sites on the West Coast, a major goal of this program was to evaluate possible temporal trends in these contaminants. This program is unique among long-term monitoring programs because it addresses trends in surficial sediments and associated fish. There might be some objections to the use of data from surface sediment analyses rather than using sediment cores because surface sediments are more subject to bioturbation. Surface sediment may not reflect contaminant inputs in a precise way due to bioturbation; however, every stratum in a core sample was once a surface layer and likely also underwent bioturbation. Furthermore, many coastal marine sites are unsuitable for coring due to natural and human-induced mixing, and to the presence of rocks, wood, or other debris. This is particularly true in industrialized areas.

Several chemical classes were demonstrated to have increasing or decreasing temporal trends at one or more of the 12 sites for which trend analyses were performed. A variety of factors can explain these trends. Increasing trends in HAHs and/or LAHs were observed at five urban sites located near growing population centers, including Los Angeles, San Francisco, and Tacoma. There are several potential sources of PAHs in urban areas (e. g., industry, ship activity and automobile traffic) which result in release of PAHs onto roadways and into the atmosphere and waterways. Subsequently, PAHs are

discharged into marine waters through precipitation and surface-water runoff (Hoffman et al. 1984). The finding that another urban site, Elliott Bay, monitored in this project showed no temporal trend in concentrations of PAHs may have been due in part to regulatory measures instituted in this bay over the past several years [e.g., relocation of a major sewage outfall and reductions in the number of combined sewer overflow events (Crecelius et al. 1995)]. To achieve decreases in concentrations of these pollutants in the marine environment near urban areas, additional control and remediation measures may be required.

The increasing temporal trends in PAHs also observed at the nonurban sites (Dana Point, Bodega Bay, and Nisqually Reach) need to be interpreted with caution. All three sites are located near the urban areas that also demonstrated increasing trends in PAHs and, thus, could be influenced by the redistribution of contaminants emanating from these areas. Also, the trends at the Dana Point and Bodega Bay sites were highly significant. Nevertheless, the concentrations of PAHs in the sediment and fish stomach contents from these three sites were quite low, generally near or below the limits of detection. Concentrations of biliary FACs were more variable, but were also generally low compared to all other sites.

The only increasing temporal trends in PCBs were found at the two urban sites in Puget Sound, located in Commencement and Elliott Bays. Both of these sites were located near waterways and nearshore areas which were previously shown to have relatively high concentrations of PCBs in sediment (Malins et al. 1984). Thus, because PCBs are no longer being manufactured, it is likely that the redistribution of sediment particles and other materials, as well as movement of fish, from nearby areas with accumulations of PCBs into these sampling sites, which were located in depositional areas in deeper areas of the bays, may have accounted for this observed trend in concentrations of PCBs.

A similar phenomenon may have influenced the finding of increasing temporal trends in DDTs at the Coos Bay site. At the Coos Bay site, the concentrations of DDTs in sediment and in starry flounder were among the lowest measured in this study. Moreover, the trend for Coos Bay was only weakly significant; thus this trend must be viewed with caution. These pesticides are persistent and were widely used in agriculture and in urban areas, as well. They are transported into coastal waters by geochemical processes. Even though production and use of DDT was halted in the 1970s, agricultural urban areas may still serve as a major DDT reservoir.

Of the 72 temporal trends investigated, only 13 (18%) were decreasing and most of these were found among the CHs. With the exception of the Nisqually Reach site in Puget Sound and the Hunters Point site in San Francisco Bay, all of these decreasing trends among CHs occurred at sites in Southern California. Decreasing trends of PCBs, DDTs, and chlordanes were found at the Dana Point reference site. Of interest were the presence of similar trends for sites in nearby San Pedro Bay, including PCBs at the Outer Harbor site and DDTs at the Long Beach site. It is possible that the same management actions aimed at source control of CHs may have affected all three of these sites. The decreasing temporal trend observed for PCBs at the South San Diego Bay site in the present study was also reported by McCain et al. (1992); however, they reported the results of Spearman rank correlations and meta-analysis for only 5 years of data. Thus, the present findings strengthen their observations. Possible explanations for this apparent decrease may be related to source control and clean-up efforts instituted by the California Regional Water Quality Control Board in the industrial areas of San Diego

Bay (Peter Michael, California Regional Water Quality Control Board, personal communication). The only other Southern California site with a decreasing trend was West Santa Monica Bay for levels of chlordanes.

Decreasing trends outside of southern California were found at the Hunters Point site in San Francisco Bay (DDTs), and Nisqually Reach in Puget Sound (chlordanes). Due to the low concentrations of chlordanes in sediment (below detection limits) and fish tissues ($\leq$ 10 ng/g) from the Nisqually Reach site, the observed trend must be viewed with caution. Decreasing trends for PAHs were found at only one site: a highly significant trend ($p \leq 0.001$) for LAHs in South San Diego Bay. The reasons for this trend is not currently understood.

In summary, the NBSP was successful in generating an extensive overview of the recent status of environmental quality in coastal waters. Such knowledge is important for effective management of the nation's highly productive coastal habitats and the resources they support. The seven-year (1984-1990) results of the West Coast portion of the NBSP demonstrate that concentrations of PAHs and CHs in sediments generally correlate well with levels of these compounds or their derivatives in bottom-dwelling fish. Such findings emphasize the importance of measuring contaminant levels in both the physical (sediment) and biological compartments. However, the variability in many of the measured parameters–due to natural variation or patchiness of contaminant distributions in many urban areas–points to the need for continued collection of chemical data to provide a sound statistical basis for the analyses of trends in pollutant concentrations.

The results also suggest that, since the mid-1980s, the concentrations of the persistent CHs, such as PCBs and DDTs in sediment and fish, show no consistent temporal trends, whereas levels of PAHs, which are non-point source contaminants, showed consistent increases at both nonurban and urban near-coastal sites. These findings indicate that, to achieve further decreases in concentrations of anthropogenic pollutants, additional control and remediation measures would appear to be required.

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APPENDIX A

Appendix 1. Results of Spearman rank analysis of data used for trends analysis (year sampled vs. Chemical concentration). Abbreviations and footnotes are defined on page 121.

Site	Chemical	Sample type	Species	n value	rho	Trend	signif (2t)	stars	# years	Min Conc	Max Conc
BODNO	Chlordanes	liver	starry flounder	16	-0.271	-	0.309		6	22	280
BODNO	Chlordanes	liver	white croaker	20	-0.126	-	0.598		7	5	50
BODNO	Chlordanes	sediment		24	0.292	+	0.166		7	0.1	1
BODNO	Chlordanes	stomach	starry flounder	7	0.2	+	0.666		6	0.6	20
BODNO	Chlordanes	stomach	white croaker	7	0.401	+	0.352		7	2	3
BODNO	Dieldrin	liver	starry flounder	16	-0.218	-	0.418		6	3	100
BODNO	Dieldrin	liver	white croaker	20	-0.211	-	0.371		7	3	15
BODNO	Dieldrin	sediment		24	-0.07	-	0.747		7	0.2	1
BODNO	Dieldrin	stomach	starry flounder	7	0.036	+	0.969		6	0.4	10
BODNO	Dieldrin	stomach	white croaker	7	0.118	+	0.784		7	0.6	3
BODNO	FACs H	bile	white croaker	53	0.567	+	< 0.0001	****	7	1	1843
BODNO	FACs H	bile	starry flounder	77	0.332	+	0.003	**	6	1	820
BODNO	FACs L	bile	starry flounder	77	0.337	+	0.003	**	6	640	140000
BODNO	FACs L	bile	white croaker	53	0.68	+	< 0.0001	****	7	1000	171303
BODNO	Hexachlorobenzene	liver	white croaker	20	0.296	+	0.205		7	2	11
BODNO	Hexachlorobenzene	liver	starry flounder	16	-0.064	-	0.814		6	3	16
BODNO	Hexachlorobenzene	sediment		21	0.056	+	0.809		6	0.2	0.2
BODNO	Hexachlorobenzene	stomach	starry flounder	7	0.578	+	0.161		6	0.8	15
BODNO	Hexachlorobenzene	stomach	white croaker	7	-0.072	-	0.907		7	1	10
BODNO	Sum DDTs	liver	starry flounder	16	-0.531	-	0.034	*	6	249	1891
BODNO	Sum DDTs	liver	white croaker	20	-0.142	-	0.551		7	119	484
BODNO	Sum DDTs	sediment		24	0.066	+	0.759		7	0.1	3
BODNO	Sum DDTs	stomach	white croaker	7	0.342	+	0.448		7	7	29
BODNO	Sum DDTs	stomach	starry flounder	7	0.414	+	0.352		6	3.7	35
BODNO	Sum HAHs	sediment		24	0.224	+	0.293		7	2	98
BODNO	Sum HAHs	stomach	white croaker	7	0.556	+	0.193		7	4	159

**** = p<0.0001
*** = p<0.001
** = p<0.01
* = p<0.05

Site	Chemical	Sample type	Species	n value	rho	Trend	signif (2t)	stars	# years	Min Conc	Max Conc
BODNO	Sum HAHs	stomach	starry flounder	7	0.676	+	0.106		6	3	9
BODNO	Sum LAHs	sediment		24	0.251	+	0.238		7	9	93
BODNO	Sum LAHs	stomach	white croaker	6	0.371	+	0.496		6	21	98
BODNO	Sum LAHs	stomach	starry flounder	6	-0.309	-	0.574		5	12	117
BODNO	Sum PCBs	liver	white croaker	20	-0.203	-	0.391		7	190	972
BODNO	Sum PCBs	liver	starry flounder	16	-0.024	-	0.929		6	242	2959
BODNO	Sum PCBs	sediment		24	0.263	+	0.214		7	0.9	68
BODNO	Sum PCBs	stomach	white croaker	7	0.179	+	0.724		7	17	329
BODNO	Sum PCBs	stomach	starry flounder	7	0.414	+	0.352		6	2	534
COLDS	Dieldrin	sediment		13	0.119	+	0.698		4	2	2
COLDS	Hexachlorobenzene	sediment		13	-0.159	-	0.603		4	0.3	0.3
COLDS	Sum DDTs	sediment		13	0.077	+	0.803		4	0.3	2
COLDS	Sum HAHs	sediment		13	-0.493	-	0.087		4	5	128
COLDS	Sum LAHs	sediment		13	-0.462	-	0.112		4	3	54
COLDS	Sum PCBs	sediment		13	-0.245	-	0.42		4	0.7	16
PUGCB	Chlordanes	liver	English sole	17	-0.338	-	0.185		5	44	530
PUGCB	Chlordanes	sediment		18	0.594	+	0.009	**	5	0.5	7
PUGCB	Chlordanes	stomach	English sole	7	-0.21	-	0.666		5	4	50
PUGCB	Dieldrin	liver	English sole	17	-0.51	-	0.036	*	5	9	41
PUGCB	Dieldrin	sediment		17	0.181	+	0.487		5	0.3	3
PUGCB	Dieldrin	stomach	English sole	7	0.647	+	0.106		5	4	5
PUGCB	FACs H	bile	English sole	48	0.358	+	0.013	*	5	28	1200
PUGCB	FACs L	bile	English sole	48	0.093	+	0.528		5	28000	178958
PUGCB	Hexachlorobenzene	liver	English sole	17	-0.294	-	0.252		5	48	340
PUGCB	Hexachlorobenzene	sediment		17	0.095	+	0.717		5	0.6	11
PUGCB	Hexachlorobenzene	stomach	English sole	7	0.148	+	0.724		5	4	23
PUGCB	Sum DDTs	liver	English sole	17	-0.314	-	0.22		5	224	6797
PUGCB	Sum DDTs	sediment		17	0.373	+	0.14		5	0.7	13
PUGCB	Sum DDTs	stomach	English sole	7	0.222	+	0.609		5	7	127

**** = p<0.0001
*** = p<0.001
** = p<0.01
* = p<0.05

Site	Chemical	Sample type	Species	n value	rho	Trend	signif (2t)	stars	# years	Min Conc	Max Conc
PUGCB	Sum HAHs	sediment		17	0.329	+	0.198		5	210	1722
PUGCB	Sum HAHs	stomach	English sole	7	0.63	+	0.132		5	468	42510
PUGCB	Sum LAHs	sediment		17	-0.108	-	0.681		5	199	929
PUGCB	Sum LAHs	stomach	English sole	7	0.556	+	0.193		5	169	22650
PUGCB	Sum PCBs	liver	English sole	17	0.341	+	0.18		5	3001	8587
PUGCB	Sum PCBs	sediment		17	0.763	+	0.0004	***	5	4.5	252
PUGCB	Sum PCBs	stomach	English sole	7	0.148	+	0.724		5	26	1032
COONB	Chlordanes	liver	starry flounder	13	0.501	+	0.081		5	4	38
COONB	Chlordanes	sediment		19	0.37	+	0.119		6	0.3	0.5
COONB	Chlordanes	stomach	starry flounder	5	0.564	+	0.33		5	1	2.7
COONB	Dieldrin	liver	starry flounder	13	0.66	+	0.014	*	5	1	7
COONB	Dieldrin	sediment		19	0.319	+	0.183		6	0.4	1
COONB	Dieldrin	stomach	starry flounder	5	0.783	+	0.146		5	0.9	1
COONB	FACs H	bile	starry flounder	53	-0.223	-	0.108		5	12	1700
COONB	FACs L	bile	starry flounder	53	-0.186	-	0.182		5	4500	290000
COONB	Hexachlorobenzene	liver	starry flounder	13	0.465	+	0.109		5	2	6
COONB	Hexachlorobenzene	sediment		17	0.625	+	0.007	**	6	0.3	2
COONB	Hexachlorobenzene	stomach	starry flounder	5	0.667	+	0.232		5	0.7	3
COONB	Sum DDTs	liver	starry flounder	13	0.021	+	0.946		5	10	1742
COONB	Sum DDTs	sediment		19	0.209	+	0.39		6	0.4	6
COONB	Sum DDTs	stomach	starry flounder	5	0.5	+	0.439		5	4	32
COONB	Sum HAHs	sediment		20	0.388	+	0.091		6	11	1343
COONB	Sum HAHs	stomach	starry flounder	5	-0.41	-	0.439		5	338	931
COONB	Sum LAHs	sediment		20	0.303	+	0.194		6	29	879
COONB	Sum LAHs	stomach	starry flounder	5	0.205	+	0.679		5	127	210
COONB	Sum PCBs	liver	starry flounder	13	-0.347	-	0.245		5	175	1026
COONB	Sum PCBs	sediment		19	0.597	+	0.007	**	6	1.2	71
COONB	Sum PCBs	stomach	starry flounder	5	-0.4	-	0.556		5	36	262
DANOU	Chlordanes	liver	white croaker	23	-0.544	-	0.007	**	7	25	164

**** = p<0.0001
*** = p<0.001
** = p<0.01
* = p<0.05

Site	Chemical	Sample type	Species	n value	rho	Trend	signif (2t)	stars	# years	Min Conc	Max Conc
DANOU	Chlordanes	liver	hornyhead turbot	16	-0.079	-	0.77		6	17	93
DANOU	Chlordanes	liver	barred sandbass	11	0.204	+	0.548		4	41	163
DANOU	Chlordanes	sediment		22	0.017	+	0.939		7	2	2
DANOU	Chlordanes	stomach	barred sandbass	4	-0.2	-	0.916		4	8	25
DANOU	Chlordanes	stomach	white croaker	8	-0.108	-	0.796		7	2	79
DANOU	Chlordanes	stomach	hornyhead turbot	6	-0.771	-	0.092		6	2	89
DANOU	Dieldrin	liver	barred sandbass	11	0.901	+	0.0002	***	4	4	19
DANOU	Dieldrin	liver	white croaker	23	0.457	+	0.028	*	7	2	12
DANOU	Dieldrin	liver	hornyhead turbot	16	0.69	+	0.003	**	6	2	10
DANOU	Dieldrin	stomach	white croaker	8	0.796	+	0.022	*	7	0.9	3
DANOU	Dieldrin	stomach	barred sandbass	4	0.949	+	0.334		4	3	11
DANOU	Dieldrin	stomach	hornyhead turbot	6	0.145	+	0.739		6	1	11
DANOU	FACs H	bile	hornyhead turbot	62	0.307	+	0.015	*	6	1	437
DANOU	FACs H	bile	white croaker	60	0.437	+	0.0005	***	7	2	750
DANOU	FACs L	bile	hornyhead turbot	62	0.298	+	0.019	*	6	160	69211
DANOU	FACs L	bile	white croaker	60	0.386	+	0.002	**	7	4100	166342
DANOU	Hexachlorobenzene	liver	hornyhead turbot	16	0.285	+	0.285		6	3	8
DANOU	Hexachlorobenzene	liver	white croaker	23	0.041	+	0.852		7	2	7
DANOU	Hexachlorobenzene	liver	barred sandbass	11	0.787	+	0.004	**	4	4	12
DANOU	Hexachlorobenzene	sediment		19	-0.008	-	0.973		6	0.5	1
DANOU	Hexachlorobenzene	stomach	barred sandbass	3	1	+	0.334		3	2	4
DANOU	Hexachlorobenzene	stomach	white croaker	8	0.055	+	0.886		7	0.9	5
DANOU	Hexachlorobenzene	stomach	hornyhead turbot	6	0.029	+	0.999		6	1	6
DANOU	Sum DDTs	liver	barred sandbass	11	0.288	+	0.391		4	632	3820
DANOU	Sum DDTs	liver	white croaker	23	-0.362	-	0.089		7	720	5997
DANOU	Sum DDTs	liver	hornyhead turbot	16	-0.304	-	0.253		6	62	4012
DANOU	Sum DDTs	sediment		22	-0.052	-	0.819		7	0.4	12
DANOU	Sum DDTs	stomach	hornyhead turbot	6	-0.543	-	0.289		6	29	227
DANOU	Sum DDTs	stomach	barred sandbass	4	-0.2	-	0.916		4	96	572

**** = p<0.0001
*** = p<0.001
** = p<0.01
* = p<0.05

Site	Chemical	Sample type	Species	n value	rho	Trend	signif (2t)	stars	# years	Min Conc	Max Conc
DANOU	Sum DDTs	stomach	white croaker	8	0.503	+	0.191		7	32	401
DANOU	Sum HAHs	sediment		22	-0.177	-	0.431		7	1	48
DANOU	Sum HAHs	stomach	white croaker	8	0.185	+	0.666		7	5	182
DANOU	Sum HAHs	stomach	hornyhead turbot	6	0.455	+	0.353		6	23	138
DANOU	Sum HAHs	stomach	barred sandbass	4	-0.258	-	0.916		4	162	162
DANOU	Sum LAHs	sediment		22	-0.093	-	0.68		7	1	25
DANOU	Sum LAHs	stomach	barred sandbass	4	0.4	+	0.75		4	13	322
DANOU	Sum LAHs	stomach	white croaker	8	0.086	+	0.841		7	4	72
DANOU	Sum LAHs	stomach	hornyhead turbot	6	0.324	+	0.496		6	10	92
DANOU	Sum PCBs	liver	hornyhead turbot	16	-0.438	-	0.089		6	310	1924
DANOU	Sum PCBs	liver	barred sandbass	11	-0.17	-	0.617		4	507	3505
DANOU	Sum PCBs	liver	white croaker	23	-0.496	-	0.016	*	7	450	2164
DANOU	Sum PCBs	sediment		22	-0.235	-	0.291		7	0.5	33
DANOU	Sum PCBs	stomach	white croaker	8	0.036	+	0.932		7	75	476
DANOU	Sum PCBs	stomach	hornyhead turbot	6	-0.943	-	0.017	*	6	96	398
DANOU	Sum PCBs	stomach	barred sandbass	4	0.6	+	0.416		4	240	603
PUGEB	Chlordanes	liver	English sole	15	-0.022	-	0.938		5	35	143
PUGEB	Chlordanes	sediment		14	0.181	+	0.536		5	0.2	2.9
PUGEB	Chlordanes	stomach	English sole	5	0.1	+	0.936		5	2	34
PUGEB	Dieldrin	liver	English sole	15	0.039	+	0.89		5	4	21
PUGEB	Dieldrin	sediment		14	0.627	+	0.017	*	5	1	9
PUGEB	Dieldrin	stomach	English sole	5	0	-	1		5	69	69
PUGEB	FACs H	bile	English sole	59	-0.43	-	0.0007	***	5	40	3600
PUGEB	FACs L	bile	English sole	59	-0.076	-	0.565		5	26000	447918
PUGEB	Hexachlorobenzene	liver	English sole	15	0.033	+	0.907		5	6	31
PUGEB	Hexachlorobenzene	sediment		13	0.321	+	0.285		5	0.2	0.8
PUGEB	Hexachlorobenzene	stomach	English sole	5	-0.112	-	0.806		5	1	4
PUGEB	Sum DDTs	liver	English sole	15	0.076	+	0.787		5	203	1814
PUGEB	Sum DDTs	sediment		14	0.284	+	0.324		5	2.1	33

**** = p<0.0001
*** = p<0.001
** = p<0.01
* = p<0.05

Site	Chemical	Sample type	Species	n value	rho	Trend	signif (2t)	stars	# years	Min Conc	Max Conc
PUGEB	Sum DDTs	stomach	English sole	5	-0.3	-	0.679		5	29	294
PUGEB	Sum HAHs	sediment		14	0.347	+	0.224		5	2037	7146
PUGEB	Sum HAHs	stomach	English sole	5	0.5	+	0.439		5	3369	161640
PUGEB	Sum LAHs	sediment		14	-0.188	-	0.519		5	590	1847
PUGEB	Sum LAHs	stomach	English sole	5	0.5	+	0.439		5	361	17870
PUGEB	Sum PCBs	liver	English sole	15	0.556	+	0.031	*	5	5296	21607
PUGEB	Sum PCBs	sediment		14	-0.056	-	0.849		5	192	903
PUGEB	Sum PCBs	stomach	English sole	5	0.6	+	0.33		5	712	1109
SFBHP	Chlordanes	liver	starry flounder	13	-0.376	-	0.206		4	103	410
SFBHP	Chlordanes	liver	white croaker	12	-0.564	-	0.056		4	65	400
SFBHP	Chlordanes	sediment		20	0.309	+	0.185		6	0.2	1.3
SFBHP	Chlordanes	stomach	white croaker	4	0.4	+	0.75		4	6	18
SFBHP	Chlordanes	stomach	starry flounder	4	0.2	+	0.916		4	4	17
SFBHP	Dieldrin	liver	white croaker	12	-0.904	-	< 0.0001	****	4	14	550
SFBHP	Dieldrin	liver	starry flounder	13	-0.695	-	0.008	**	4	33	460
SFBHP	Dieldrin	sediment		20	0.634	+	0.003	**	6	0.4	2
SFBHP	Dieldrin	stomach	starry flounder	3	-1	-	0.334		3	2	25
SFBHP	Dieldrin	stomach	white croaker	4	-0.8	-	0.334		4	3	26
SFBHP	FACs H	bile	starry flounder	65	0.094	+	0.458		6	1	1152
SFBHP	FACs H	bile	white croaker	67	0.755	+	< 0.0001	****	6	14	1019
SFBHP	FACs L	bile	white croaker	67	0.702	+	< 0.0001	****	6	6600	143360
SFBHP	FACs L	bile	starry flounder	65	0.053	+	0.678		6	1900	224831
SFBHP	Hexachlorobenzene	liver	starry flounder	13	-0.001	-	0.996		4	2	19
SFBHP	Hexachlorobenzene	liver	white croaker	12	-0.228	-	0.476		4	2	4
SFBHP	Hexachlorobenzene	sediment		20	0.415	+	0.069		6	0.1	2
SFBHP	Hexachlorobenzene	stomach	white croaker	4	0.2	+	0.916		4	0.6	7
SFBHP	Hexachlorobenzene	stomach	starry flounder	3	0.866	+	1		3	0.3	0.3
SFBHP	Sum DDTs	liver	starry flounder	13	-0.564	-	0.045	*	4	685	3749
SFBHP	Sum DDTs	liver	white croaker	12	-0.485	-	0.11		4	490	3419

**** = p<0.0001
*** = p<0.001
** = p<0.01
* = p<0.05

Site	Chemical	Sample type	Species	n value	rho	Trend	signif (2t)	stars	# years	Min Conc	Max Conc
SFBHP	Sum DDTs	sediment		20	0.618	+	0.004	**	6	1.9	13
SFBHP	Sum DDTs	stomach	starry flounder	3	-1	-	0.334		3	18	84
SFBHP	Sum DDTs	stomach	white croaker	4	-0.6	-	0.416		4	52	86
SFBHP	Sum HAHs	sediment		20	0.635	+	0.003	**	6	1672	15370
SFBHP	Sum HAHs	stomach	white croaker	4	0.6	+	0.416		4	115	529
SFBHP	Sum HAHs	stomach	starry flounder	3	1	+	0.334		3	192	4094
SFBHP	Sum LAHs	sediment		20	0.578	+	0.008	**	6	260	2567
SFBHP	Sum LAHs	stomach	white croaker	4	0.8	+	0.334		4	20	250
SFBHP	Sum LAHs	stomach	starry flounder	3	1	+	0.334		3	23	460
SFBHP	Sum PCBs	liver	white croaker	12	-0.152	-	0.637		4	1848	8941
SFBHP	Sum PCBs	liver	starry flounder	13	-0.333	-	0.266		4	2159	9781
SFBHP	Sum PCBs	sediment		20	0.661	+	0.002	**	6	25	261
SFBHP	Sum PCBs	stomach	starry flounder	3	-0.5	-	1		3	233	607
SFBHP	Sum PCBs	stomach	white croaker	4	-0.4	-	0.75		4	436	949
SPBLB	Chlordanes	liver	white croaker	14	-0.034	-	0.909		5	142	2130
SPBLB	Chlordanes	sediment		13	-0.517	-	0.071		4	4	26
SPBLB	Chlordanes	stomach	white croaker	4	0.4	+	0.75		4	14	158
SPBLB	Dieldrin	liver	white croaker	14	-0.512	-	0.061		5	12	250
SPBLB	Dieldrin	sediment		13	-0.012	-	0.969		4	0.7	4
SPBLB	Dieldrin	stomach	white croaker	4	0.4	+	0.75		4	6	18
SPBLB	FACs H	bile	white croaker	60	0.443	+	0.0004	***	5	8.8	1570
SPBLB	FACs L	bile	white croaker	60	0.33	+	0.01	**	5	16000	456732
SPBLB	Hexachlorobenzene	liver	white croaker	14	0.644	+	0.013	*	5	2	110
SPBLB	Hexachlorobenzene	sediment		13	0.059	+	0.848		4	0.5	0.8
SPBLB	Hexachlorobenzene	stomach	white croaker	4	-0.738	-	0.416		4	1	8
SPBLB	Sum DDTs	liver	white croaker	14	-0.297	-	0.302		5	1766	22152
SPBLB	Sum DDTs	sediment		13	-0.017	-	0.956		4	11	171
SPBLB	Sum DDTs	stomach	white croaker	4	-0.6	-	0.416		4	475	2356
SPBLB	Sum HAHs	sediment		13	-0.061	-	0.843		4	365	2374

**** = p<0.0001
*** = p<0.001
** = p<0.01
* = p<0.05

Site	Chemical	Sample type	Species	n value	rho	Trend	signif (2t)	stars	# years	Min Conc	Max Conc
SPBLB	Sum HAHs	stomach	white croaker	4	-0.4	-	0.75		4	72	1920
SPBLB	Sum LAHs	sediment		13	-0.048	-	0.876		4	38	291
SPBLB	Sum LAHs	stomach	white croaker	4	-0.4	-	0.75		4	4	510
SPBLB	Sum PCBs	liver	white croaker	14	0.435	+	0.12		5	6005	18036
SPBLB	Sum PCBs	sediment		13	-0.112	-	0.715		4	141	327
SPBLB	Sum PCBs	stomach	white croaker	4	0	-	0.916		4	1061	2179
PUGNR	Chlordanes	liver	English sole	14	-0.485	-	0.078		4	7	67
PUGNR	Chlordanes	stomach	English sole	4	-0.105	-	0.916		4	4	7
PUGNR	Dieldrin	liver	English sole	14	0.081	+	0.784		4	2	28
PUGNR	Dieldrin	stomach	English sole	4	0.258	+	0.916		4	5	5
PUGNR	FACs H	bile	English sole	47	0.13	+	0.383		4	1	807
PUGNR	FACs L	bile	English sole	47	0.22	+	0.138		4	2500	147393
PUGNR	Hexachlorobenzene	liver	English sole	14	-0.768	-	0.001	**	4	2	96
PUGNR	Hexachlorobenzene	stomach	English sole	4	-0.258	-	0.916		4	2	2
PUGNR	Sum DDTs	liver	English sole	14	-0.191	-	0.514		4	31	450
PUGNR	Sum DDTs	stomach	English sole	4	0.105	+	0.916		4	6	26
PUGNR	Sum HAHs	stomach	English sole	4	0.738	+	0.416		4	93	166
PUGNR	Sum LAHs	stomach	English sole	4	-0.2	-	0.916		4	69	140
PUGNR	Sum PCBs	liver	English sole	14	-0.143	-	0.626		4	356	3378
PUGNR	Sum PCBs	stomach	English sole	4	-0.4	-	0.75		4	16	230
SFBSS	Chlordanes	liver	starry flounder	20	-0.09	-	0.706		6	31	189
SFBSS	Chlordanes	sediment		21	0.438	+	0.047	*	6	0.1	1.5
SFBSS	Chlordanes	stomach	starry flounder	6	0.203	+	0.655		6	2	7
SFBSS	Dieldrin	liver	starry flounder	20	-0.553	-	0.011	*	6	11	190
SFBSS	Dieldrin	sediment		21	0.352	+	0.118		6	0.5	2
SFBSS	Dieldrin	stomach	starry flounder	6	0.441	+	0.353		6	2	27
SFBSS	FACs H	bile	white croaker	51	0.339	+	0.015	*	4	14	664
SFBSS	FACs H	bile	starry flounder	61	0.007	+	0.959		6	1	4000
SFBSS	FACs L	bile	starry flounder	61	-0.099	-	0.45		6	4200	500000

**** = p<0.0001
*** = p<0.001
** = p<0.01
* = p<0.05

Site	Chemical	Sample type	Species	n value	rho	Trend	signif (2t)	stars	# years	Min Conc	Max Conc
SFBSS	FACs L	bile	white croaker	51	0.012	+	0.932		4	2900	172987
SFBSS	Hexachlorobenzene	liver	starry flounder	20	0.559	+	0.01	*	6	3	11
SFBSS	Hexachlorobenzene	sediment		20	0.649	+	0.002	**	6	0.2	3
SFBSS	Hexachlorobenzene	stomach	starry flounder	6	0.778	+	0.06		6	0.5	8
SFBSS	Sum DDTs	liver	starry flounder	20	-0.321	-	0.168		6	292	3035
SFBSS	Sum DDTs	sediment		21	0.679	+	0.0007	***	6	0.2	1393
SFBSS	Sum DDTs	stomach	starry flounder	6	-0.429	-	0.422		6	10	257
SFBSS	Sum HAHs	sediment		21	0.35	+	0.12		6	32	2201
SFBSS	Sum HAHs	stomach	starry flounder	6	0.371	+	0.496		6	35	614
SFBSS	Sum LAHs	sediment		21	0.228	+	0.32		6	10	878
SFBSS	Sum LAHs	stomach	starry flounder	6	-0.6	-	0.23		6	21	93
SFBSS	Sum PCBs	liver	starry flounder	20	-0.198	-	0.402		6	1150	5031
SFBSS	Sum PCBs	sediment		21	0.516	+	0.017	*	6	3.8	86
SFBSS	Sum PCBs	stomach	starry flounder	6	-0.086	-	0.911		6	152	455
SFBSP	Chlordanes	sediment		19	0.41	+	0.081		5	0.3	1.5
SFBSP	Hexachlorobenzene	sediment		19	0.357	+	0.133		5	0.1	1
SFBSP	Sum DDTs	sediment		19	0.439	+	0.06		5	0.3	13
SFBSP	Sum HAHs	sediment		20	0.009	+	0.969		5	1.6	754
SFBSP	Sum LAHs	sediment		20	0.15	+	0.528		5	4.5	134
SFBSP	Sum PCBs	sediment		19	0.315	+	0.189		5	2	73
SPBOH	Chlordanes	liver	white croaker	15	-0.155	-	0.582		6	51	410
SPBOH	Chlordanes	sediment		13	0.843	+	0.0003	***	4	0.5	4
SPBOH	Chlordanes	stomach	white croaker	7	-0.927	-	0.008	**	5	4	114
SPBOH	Dieldrin	liver	white croaker	15	-0.002	-	0.995		6	2	27
SPBOH	Dieldrin	sediment		13	0.803	+	0.001	***	4	2	4
SPBOH	Dieldrin	stomach	white croaker	7	-0.778	-	0.048	*	5	0.9	25
SPBOH	FACs H	bile	white croaker	50	0.384	+	0.006	**	5	7	2868
SPBOH	FACs L	bile	white croaker	50	0.526	+	< 0.0001	****	5	7200	396466
SPBOH	Hexachlorobenzene	liver	white croaker	15	0.354	+	0.195		6	1	12

**** = p<0.0001
*** = p<0.001
** = p<0.01
* = p<0.05

Site	Chemical	Sample type	Species	n value	rho	Trend	signif (2t)	stars	# years	Min Conc	Max Conc
SPBOH	Hexachlorobenzene	sediment		13	0.234	+	0.441		4	0.6	1
SPBOH	Hexachlorobenzene	stomach	white croaker	7	0.155	+	0.724		5	1	3
SPBOH	Sum DDTs	liver	white croaker	15	-0.089	-	0.752		6	788	37249
SPBOH	Sum DDTs	sediment		13	-0.142	-	0.644		4	5	786
SPBOH	Sum DDTs	stomach	white croaker	7	-0.148	-	0.724		5	981	7555
SPBOH	Sum HAHs	sediment		13	0.414	+	0.16		4	426	3584
SPBOH	Sum HAHs	stomach	white croaker	7	0.889	+	0.014	*	5	94	6330
SPBOH	Sum LAHs	sediment		13	0.332	+	0.268		4	82	783
SPBOH	Sum LAHs	stomach	white croaker	7	0.148	+	0.724		5	51	386
SPBOH	Sum PCBs	liver	white croaker	15	-0.107	-	0.704		6	451	7944
SPBOH	Sum PCBs	sediment		13	-0.207	-	0.498		4	101	532
SPBOH	Sum PCBs	stomach	white croaker	7	-0.852	-	0.023	*	5	516	3465
SDBTE	Chlordanes	liver	barred sandbass	20	-0.564	-	0.01	**	5	97	401
SDBTE	Chlordanes	sediment		21	-0.083	-	0.72		6	2	15
SDBTE	Chlordanes	stomach	barred sandbass	9	0.618	+	0.073		5	9	41
SDBTE	Dieldrin	liver	barred sandbass	20	-0.548	-	0.012	*	5	27	130
SDBTE	Dieldrin	sediment		21	0.78	+	< 0.0001	****	6	0.8	2
SDBTE	Dieldrin	stomach	barred sandbass	9	0.422	+	0.25		5	4	24
SDBTE	Hexachlorobenzene	liver	barred sandbass	20	0.366	+	0.112		5	2	7
SDBTE	Hexachlorobenzene	sediment		21	-0.226	-	0.325		6	0.1	0.1
SDBTE	Hexachlorobenzene	stomach	barred sandbass	9	0.533	+	0.145		5	0.6	1
SDBTE	Sum DDTs	liver	barred sandbass	20	-0.471	-	0.036	*	5	441	1243
SDBTE	Sum DDTs	sediment		21	0.112	+	0.63		6	5	117
SDBTE	Sum DDTs	stomach	barred sandbass	9	0.633	+	0.073		5	25	193
SDBTE	Sum HAHs	sediment		21	-0.087	-	0.707		6	459	13270
SDBTE	Sum HAHs	stomach	barred sandbass	9	-0.051	-	0.879		5	95	967
SDBTE	Sum LAHs	sediment		21	-0.169	-	0.465		6	58	6414
SDBTE	Sum LAHs	stomach	barred sandbass	9	-0.864	-	0.005	**	5	6	272
SDBTE	Sum PCBs	liver	barred sandbass	20	-0.623	-	0.003	**	5	6051	24908

**** = p<0.0001
*** = p<0.001
** = p<0.01
* = p<0.05

Site	Chemical	Sample type	Species	n value	rho	Trend	signif (2t)	stars	# years	Min Conc	Max Conc
SDBTE	Sum PCBs	sediment		21	-0.438	-	0.047	*	6	125	872
SDBTE	Sum PCBs	stomach	barred sandbass	9	0.385	+	0.292		5	505	2695
SMBWE	Chlordanes	liver	hornyhead turbot	12	-0.497	-	0.101		4	380	1020
SMBWE	Chlordanes	stomach	hornyhead turbot	4	-1	-	0.084		4	12	125
SMBWE	Dieldrin	liver	hornyhead turbot	12	0.2	+	0.533		4	7	7
SMBWE	Dieldrin	stomach	hornyhead turbot	4	0.775	+	0.416		4	2	2
SMBWE	FACs H	bile	hornyhead turbot	33	0.28	+	0.115		4	37	1100
SMBWE	FACs L	bile	hornyhead turbot	33	-0.062	-	0.732		4	32000	470000
SMBWE	Hexachlorobenzene	liver	hornyhead turbot	12	-0.466	-	0.127		4	6	21
SMBWE	Hexachlorobenzene	stomach	hornyhead turbot	4	0.2	+	0.916		4	0.7	7
SMBWE	Sum DDTs	liver	hornyhead turbot	12	0.605	+	0.037	*	4	13758	44616
SMBWE	Sum DDTs	stomach	hornyhead turbot	4	-0.8	-	0.334		4	840	5014
SMBWE	Sum HAHs	stomach	hornyhead turbot	4	-0.4	-	0.75		4	91	210
SMBWE	Sum LAHs	stomach	hornyhead turbot	4	0.8	+	0.334		4	13	96
SMBWE	Sum PCBs	liver	hornyhead turbot	12	0.108	+	0.738		4	13640	28925
SMBWE	Sum PCBs	stomach	hornyhead turbot	4	-0.8	-	0.334		4	909	4302

**** = p<0.0001
*** = p<0.001
** = p<0.01
* = p<0.05

FOOTNOTES FOR APPENDIX TABLE

Spearman Rank Analysis

Site abbreviations are listed in Table 3.

Abbreviations of chemical names are in Table 1.

The Spearman rank correlation coefficient and its two-tailed significance level are listed as *rho* and *signif(2t)*, respectively.

Samples were not collected every year at all of the sites, therefore *# years* is used to show the number of annual cycles within the 7 year series for which samples were collected.

Min- and Max Conc shows the minimum and maximum analyte concentrations for each site (FACs concentrations are ng/g bile; all other concentrations are ng/g dry weight).