October 2003

FINAL

Status, Trends and Effects of Toxic Contaminants in the Puget Sound Environment

PREPARED FOR:

Puget Sound Action Team

Olympia, WA

PREPARED BY:

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STATUS, TRENDS AND EFFECTS OF TOXIC CONTAMINANTS IN THE PUGET SOUND ENVIRONMENT

FINAL

Prepared for

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AET	Apparent effects threshold
µg·g^{-1}	Micrograms per gram (= parts per million)
µg·kg^{-1}	Micrograms per kilogram (= parts per billion)
µg·L^{1}	Micrograms per liter (= parts per billion)
AIRS	Aerometric Information Retrieval System
As	Arsenic
ATSDR	Agency for Toxic Substances and Disease Registry
AVS	Acid volatile sulfide
BAT	Best available technology
BCF	Bioconcentration factor
BCT	Best control technology
BCWMSP	British Columbia/Washington Marine Science Panel
BMP	Best management practices
BOD	Biochemical oxygen demand
CCMP	Comprehensive Conservation and Management Plan
Cd	Cadmium
cm·s^{-1}	Cubic meters per second
COPC	Contaminant of potential concern
CSL	Cleanup Screening Level
CSO	Combined sewer overflow
Cu	Copper
CWA	Clean Water Act
DBT	Dibutyl tin
DO	Dissolved oxygen
DoD	Department of Defense
dw	Dry weight
EDC	Endocrine disrupting compound
ECC	Environmental Cooperative Council
Eh	Reduction-oxidation potential
EIS	Environmental impact statement
EOF	Emergency overflows
EPA	Environmental Protection Agency (also, USEPA)
ERA	Ecological Risk Assessment

ERL	Effects range low
ERM	Effects range medium
EVS	EVS Environment Consultants
FAC	Fluorescing Aromatic Compounds
FAO	Food and Agriculture Organization
FCA	Foci of cellular alteration
FDA	Food and Drug Administration (federal)
fw	Fresh weight
GBCWWG	Georgia Basin Clean Water Work Group
GSI	Gonadosomatic index
Hg	Mercury
HPAH	High molecular weight polycyclic aromatic hydrocarbons
HSPF	Hydrologic Simulation Program Fortran
IEMPOP	International Experts Meeting on Persistent Organic Pollutants
LPAH	Low molecular weight polycyclic aromatic hydrocarbons
lw	Lipid weight
MBT	Monobutyltin tin
MELP	British Columbia Ministry of Environment, Lands and Parks
MWLAP	British Columbia Ministry of Water, Land and Air Protection
$mg \cdot g^{-1}$	Milligrams per gram (= parts per thousand)
$mg \cdot kg^{-1}$	Milligrams per kilogram (= parts per million)
$mg \cdot L^{1}$	Milligrams per liter (= parts per million)
ML	Maximum level
MOSS	Marine Outfall Siting Study
MSMP	Marine Sediment Monitoring Program
MSMS	Marine Safety Management System
$mt \cdot y^{-1}$	Metric tonnes per year
MTCA	Model Toxics Control Act
MWLAP	BC Ministry of Water Lands and Parks
$\mu g \cdot kg^{-1}$	Micrograms per kilogram (= parts per billion)
$\mu g \cdot L^{-1}$	Micrograms per liter (= parts per billion)
NATA	National Air Toxics Assessment
NAWQA	National Water Quality Assessment
NBSP	National Benthic Surveillance Program
$ng \cdot g^{-1}$	Nanograms per gram (= parts per billion)

NOAA	National Oceanic and Atmospheric Administration
NPDES	National Pollutant Discharge Elimination System
NPS	Non-point source pollution
NS&T	National Status and Trends (Program)
OP	Organophosphorous (pesticides)
PAH	Polycyclic aromatic hydrocarbon
Pb	Lead
PBT	Persistent, bioaccumulative toxin
PCB	Polychlorinated biphenyl
PCDD	Polychlorinated dibenzo-*p*-dioxin
PCDD	polychlorinated dibenzo-*p*-dioxins
PCDF	Polychlorinated dibenzofuran
PCDF	polychlorinated dibenzofurans
PEL	Probable effects level
pg·g^{-1}	Picograms per gram (= parts per trillion)
PHOBEA	Photochemical Ozone Budget of Eastern North Pacific Atmosphere
PMEL	Pacific Marine Environmental Laboratory
POM	Particulate organic matter
ppb	Parts per billion
ppm	Parts per million
PSAMP	Puget Sound Ambient Monitoring Program
PSDDA	Puget Sound Dredge Disposal Analysis
PSEP	Puget Sound Estuary Program
PSTWG	Puget Sound Toxics Work Group
PSAT	Puget Sound Action Team
PSGBTWG	Puget Sound/Georgia Basin Toxics Work Group
PSWQAT	Puget Sound Water Quality Action Team
PTI	PTI Environmental Services
SDN	Specific degenerative/necrotic lesions
SEM	Simultaneously extractable metals
SL	Screening level
SMIC	Sea-surface microlayer
SMS	Sediment Management Standards
Sn	Tin
SQS	Sediment Quality Standard

SQV	Sediment quality values
TBT	Tributyltin
TEL	Threshold effects level
TEQ	Toxic equivalency quotient
TMDL	Total maximum daily load
TOC	Total organic carbon
TPH	Total petroleum hydrocarbon
TSS	Total suspended solids
USEPA	US Environmental Protection Agency
USFWS	US Fish and Wildlife Service
USGS	United States Geological Service
WDFW	Washington State Department of Fish and Wildlife
WDNR	Washington State Department of Natural Resources
WDOE	Washington State Department of Ecology
WDOH	Washington State Department of Health
WHO	World Health Organization
WRIA	Water Resource Inventory Area
ww	Wet weight
WWTP	Wastewater treatment plant

GLOSSARY

advection	The horizontal movement of a substance, such as sediment in water.
biliary FAC	A measure of the metabolites of polycyclic aromatic hydrocarbons (PAH) found in the bile of vertebrates, used as a surrogate for exposure to PAHs
bioaccumulation	A general term, meaning that an organism stores within its body a higher concentration of a substance than is found in its environment. Includes uptake of substances from water (=bioconcentration) and from food.
bioavailable	The fraction of the total chemical in the surrounding environment which can be taken up by organisms. The environment may include water, sediment, suspended particles, and food items.
bioconcentration	Accumulation of a chemical by an aquatic organism directly from the water, to a higher concentration than is found in the water. The bioconcentration results from simultaneous processes of uptake and elimination.
bioconcentration factors	A unitless value describing the degree to which a chemical can be concentrated in the tissues of an organism in the aquatic environment. It is calculated by dividing the concentration of a specific chemical in an aquatic organism by the concentration of the chemical in the water in which the organism was living. BCF is usually determined experimentally. Dangerous bioaccumulative toxicants would usually have BCF's with values in the thousands or tens of thousands.
biomagnification	The result of processes of bioconcentration and bioaccumulation by which tissue concentrations of bioaccumulated chemicals increase as the chemical passes up through the food chain. The term implies an efficient transfer of chemical from food to consumer, so that residue concentrations increase systematically from one trophic level to the next.
biomethylation	The process by which microbes add a methyl group to a compound, often a metal.
carcinogen	A substance or chemical which induces cancer in living organisms.

DNA adduct	Damaged segments of DNA.
embryotoxic	Causing injury to the embryo.
fecundity	Refers to the number of offspring produced by a female organism.
fetotoxic	Causing injury to the fetus.
histopathologic	Having an adverse affect on tissue.
immunotoxic	Causing injury to the immune system.
imposex	A condition in which a female organisms develops male sexual organs.
mutagen	A chemical that causes an alteration of the inherited genetic material, i.e., the DNA of the genes. In the narrowest sense, the chemical alters the genetic material of paternal or maternal sex cells.
teratogen	A substance that causes alteration in the developing cells, tissues, or organs at the embryonic stage of development. An agent that increases the incidence of congenital malformations.
vitellogenesis	The production of vitellogenin, a protein synthesized by the livers of female fish in preparation for reproduction.

ACKNOWLEDGEMENTS

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EXECUTIVE SUMMARY

The Puget Sound Toxics Work Group (PSTWG) was established to determine the status, trends, and gaps regarding toxic chemicals in the waters, sediments, and biota of Puget Sound. To help achieve this goal, the PSTWG contracted EVS Environment Consultants Ltd. (EVS) to conduct a literature review and to interview experts working on toxics issues in the basin, focusing on selected metals and organic contaminants including: arsenic, cadmium, copper, lead, mercury, tributyltin, polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), pesticides, dioxins and furans, and phthalate esters. The product of this review is a summary of the sources of the contaminants and the pathways by which they enter Puget Sound, an assessment of water, sediment and tissue concentrations vis-à-vis regulatory criteria, and identification of data gaps.

The review of the available data highlighted concerns at both a broader scale such as identifying/quantifying sources and trophic transfer of persistent, bioaccumulative toxics (PBTs) in Puget Sound, to more specific questions such as what are the potential ecological effects of the elevated lead concentrations observed in Sinclair Inlet fish. A companion document summarizes recommendations for further research and monitoring of toxics in Puget Sound to answer these and other questions.

CONTAMINANT SOURCES AND PATHWAYS

Puget Sound has a long history of intensive industrial activities, which have left a legacy of contaminated sites along the shore, particularly in the urban centers of Seattle and Tacoma. Toxic metals and organic chemicals continue to enter Puget Sound through a variety of direct and indirect sources, including: municipal and industrial wastewater discharges (e.g., smelters, oil refineries, pulp and paper mills); contaminated sites; surface runoff; groundwater seepage; oil and other chemical spills; disturbance of contaminated sediments through dredging; and atmospheric deposition. Regulatory controls, improved technology, and clean-up efforts appear to have resulted in decreasing contaminant discharges to the Sound. However, there is a lack of information regarding overall contaminant loading from the various sources.

WATER QUALITY

Water quality in Washington State is regularly monitored through the Puget Sound Ambient Monitoring Program (PSAMP), which focuses on conventional parameters that characterize the physical properties, nutrient conditions and sanitary quality of Puget Sound's marine waters. The program does not, however, address toxic contaminants, and only limited data were available from other studies for an assessment of toxic metals and organics in the water column of Puget Sound. These data suggested that contaminant concentrations in the water column were highest in urban/industrial embayments such as

Commencement Bay, however, they have decreased during the last two decades as a result of contaminated-site cleanup and source control efforts. Still of concern, however, are contaminant concentrations at the sea-surface microlayer, which can be several orders of magnitude higher than those detected in the subsurface water column.

Because of the complex behavior of metals and organic contaminants in seawater, and a propensity for these chemicals to partition into sediments and biota, water quality monitoring needs to be focused at locations where the effects may occur, such as in the vicinity of point source discharges (e.g., industrial effluent discharges, combined sewer overflows [CSOs], stormwater outfalls) and at the sea-surface microlayer.

SEDIMENT QUALITY

Many contaminants have an affinity for the organic or inorganic components of sediment particles. As a result, sediments often act as a contaminant sink with substantially higher concentrations than in the overlying water. In Puget Sound "hot spots" of toxic contaminants have been shown to alter and reduce benthic invertebrate communities, and cause disease in aquatic organisms closely associated with the sediments (e.g., liver lesions in English sole). Historically contaminated sediments also continue to contribute metals and organic compounds to higher trophic level organisms through food chain transfer.

A comprehensive basin-wide study was recently conducted by the National Oceanic and Atmospheric Administration (NOAA) in partnership with PSAMP (PSAMP-NOAA 2002). The purpose of the study was to characterize sediment quality throughout Puget Sound, rather than focusing on known or suspected areas of contamination. Over the course of three years (1997-1999), three hundred samples were collected from randomly selected stations. Metals such as arsenic, copper, lead, and mercury were detected throughout the Sound, whereas cadmium was detected at 20-50% of the stations and TBT at 0-80% of the stations. The highest concentrations were usually found in the urban embayments of Bellingham Bay, Everett Harbor, Sinclair Inlet, Elliott Bay and Commencement Bay. Organic contaminants such as polycyclic aromatic hydrocarbons (PAHs) were also ubiquitous, while phthalate esters, polychlorinated biphenyls (PCBs), dibenzofurans and DDT were detected at fewer stations.

Chemical analysis of sediment cores taken from six locations in Central Puget Sound (Lefkovitz et al. 1997) indicated that metals and organic contaminant concentrations generally peaked around the 1960s, coinciding with peak use, production and/or discharge in the U.S. Since that time, contaminant concentrations have decreased, but still remain higher than pre-1890 concentrations.

Current monitoring and data gaps include:

- Insufficient characterization of background concentrations of some contaminants such as PCBs;

- Insufficient data regarding the presence/composition of individual PCB or dioxin congeners in sediments to support human health risk assessments; and
- Laboratory methods in use for some compounds may have higher detection limits than environmental concentrations of concern.

BIOTA

Once contaminants are in the water and sediments of the Sound, portions of the contaminant load can become available to aquatic biota. Both chronic and acute effects can occur when aquatic organisms take up and assimilate various contaminants into their tissues. Sediments are often the initial source of contaminants; invertebrates living in the sediments can assimilate contaminants through dermal contact or ingestion. Benthic fish feed on the invertebrates, taking in the contaminant body burdens of their prey, as well as any incidental ingestion from the sediments. These benthic fish then become food items for larger pelagic fish, which are in turn fed upon by killer whales and otters. Humans may consume both lower and higher trophic level marine organisms.

The highest contaminant body residues have been found in organisms in closer proximity to contaminant sources and/or in organisms occupying positions high in the food web, indicating continued exposure of aquatic organisms off Puget sound to toxic contaminants.

Invertebrates

The National Oceanic and Atmospheric Administration's (NOAA's) Mussel Watch program is the only comprehensive and long-term data set of metal and organic contaminant concentrations in Puget Sound invertebrates, and little is known about the ecological effects at the individual, population or ecosystem level of measured contaminant tissue burdens in benthic infauna and shellfish.

The available data indicate that arsenic tissue residues regularly exceed human health guidelines, however, there is uncertainty in whether or not the metal is in a bioavailable form. PCB and DDT are also detected in almost all the samples analyzed, with the highest concentrations measured in Commencement Bay and Elliott Bay biota. PCB in mussel and crab edible tissue generally exceeded human health cancer risk benchmarks, particularly in Central Puget Sound.

Current monitoring and data gaps include:

- There is a lack of contaminant levels and effects in benthic infauna, other than large mollusks that are harvested;
- Contaminant levels and effects in crabs not well studied;

- There is insufficient information regarding presence/composition of individual PCB congeners to support human health risk assessments.

Fish

Fish tissue contaminant burdens and the toxic effects of metals and organic contaminants have been studied at numerous locations throughout Puget Sound. Findings of research conducted to date indicate that fish living in or migrating through the urban/industrial embayments of Central and South Puget Sound accumulate metals and organic contaminants to a greater degree than fish from non-urban sites, and are at higher risk for developing diseases, lowered reproductive success and impaired immune system function.

The most studied fish in Puget Sound is the English sole. Because it lives in close contact with bottom sediments and consumes benthic invertebrates, it is a useful sentinel species for exposure to contaminated sediments. English sole accumulate arsenic, lead and organochlorines such as PCBs, however, the contaminant of primary concern appears to be polycyclic aromatic hydrocarbons (PAHs). While the overall mortality rate of adult English sole from contaminated sites in Puget Sound was similar to fish from relatively uncontaminated sites (Landahl et al. 1997), chronic effects such as toxicopathic liver lesions, reproductive impairment, and immunosuppression have been documented (Collier et al. 1998; Arkoosh et al. 1996).

Salmon exhibit a unique life history in that they reproduce in fresh water and mature to adulthood in the ocean. Juveniles migrating from their natal creeks to the marine environment pass through estuaries which are sometimes co-located with major urban-industrial areas. There, they are exposed to contaminants through their diet of benthic organisms and have demonstrated impaired immune system function in response (Arkoosh et al. 1998; Varanasi et al. 1993). The contribution of contaminants accumulated by juveniles migrating through contaminated estuaries to the contaminant burden of adults is negligible (O'Neill et al. 1998). However, there is a lack of information regarding the relative importance of other local (Puget Sound) versus global (Pacific Ocean) sources of contaminants such as PCBs to adult salmon. Moreover, the ecological significance of measured adult contaminant burdens is unknown.

Current monitoring and data gaps include:

- The reproductive effects of many contaminants of concern are unknown;
- The cause of endocrine disruption in male rockfish form Elliott Bay is unknown;
- The relative importance of global versus local sources of PCB to Puget Sound salmon is unknown.

Mammals

Limited data indicate that top carnivorous marine mammals such as harbor seals and killer whales accumulate organic contaminants. PCB concentrations in harbor seals have decreased significantly between 1972 and 1997 but have now stabilized, mirroring the trend in decreasing concentrations observed in the Mussel Watch program. This indicates that while overall PCB concentrations in Puget Sound have decreased, the compound continues to be persistent in the food web.

While relatively little research has been conducted in Puget Sound specifically, PCBs have been linked to immune system impairment in marine mammals. For example, captive feeding studies in which harbor seals were fed polyhalogenated aromatic carbon-contaminated (i.e., PCB, PCDD, PCDF) herring showed that total PCB blubber concentrations of 17 $mg \cdot kg^{-1}$ were associated with immunosuppression (Ross et al. 1996). On this basis, both harbor seals and killer whales in Puget Sound are thought to be at risk.

Current monitoring and data gaps include:

- There are insufficient data to determine temporal trends of organochlorine compounds in killer whales;
- Further investigation is required to determine the relative importance of local (i.e., contaminated sites in Puget Sound) versus international (i.e., global atmospheric transport) sources of persistent toxics that bioaccumulate to higher trophic consumers such as carnivorous marine mammals;
- The mechanisms by which persistent toxics that bioaccumulate affect marine mammals and the risks associated with current contaminant burdens are not fully understood.

Birds

Relatively few studies have been conducted on the birds of Puget Sound, in part because it is difficult to find an appropriate sentinel species. Consequently, there are insufficient data to assess the potential effects of measured tissue concentrations or to determine temporal and spatial trends of contaminant tissue burdens. Limited studies of surf scoters overwintering in Commencement Bay indicate that the migratory birds do accumulate contaminants, however, histopathological analyses indicated the birds were in good health (Mahaffy et al. 1997). Further sample collection from other areas in Puget Sound is necessary to increase the overall sample size and to provide a comparison between birds overwintering in different locations.

While surf scoters have apparently experienced no effects related to contaminant exposure and accumulation, the eggshells of blue herons and glaucous-winged gulls in the early 1990s were significantly thinner than those measured prior to the advent of organochlorine compounds (Speich et al. 1992). The eggshells of eagles from Hood

Canal are also thinner than they were at the beginning of the 20th century, and the population of eagles in Hood Canal has not recovered from the effects of wide-spread DDT use to the same degree as populations in other areas of Washington State (Mahaffy et al. 2001). PCB concentrations in Hood Canal eagle eggs were greater than reported thresholds above which effects may be observed. The source of PCBs, however, is unknown as sediment and fish tissue contaminants were not high enough in Hood Canal to account for the elevated PCB levels measured.

Other monitoring and data gaps include:

- There are not enough data to assess the spatial variability in contaminant uptake by birds utilizing urban/industrial estuaries versus more isolated estuaries;
- The sublethal effects of contaminants measured in Puget Sound birds is unknown;
- There are not enough data to assess the relationship between contaminant burdens and eggshell thinning in Puget sound waterfowl.

EMERGING ISSUES

During the last several years, some issues in the study of environmental contaminants have been moving to the forefront and may have implications for the direction of research in Puget Sound's marine environment. These issues include: the appropriateness of extrapolating laboratory toxicity studies to field conditions; the ecological relevance of the phototoxicity of polycyclic aromatic hydrocarbons (PAHs); the level of "recently" introduced organic compounds (e.g., polybrominated diphenyl ethers; pharmaceuticals and personal care products); and the speciation and potential bioavailability of cadmium in marine organisms consumed by mammals.

1. INTRODUCTION

1.1 BACKGROUND AND HISTORY

In 1992, the British Columbia Premier Mike Harcourt and Governor Booth Gardner of Washington State signed an Environmental Cooperation Agreement that committed the province and state to promote and coordinate mutual efforts to ensure the protection, preservation and enhancement of our shared environment for the benefit of current and future generations. Shortly after the agreement was signed, the British Columbia/Washington Environmental Cooperative Council (ECC) was created to address the numerous issues transcending the boundary between British Columbia and Washington. The ECC formed the Puget Sound/Georgia Basin International Task Force to focus on environmental issues in the Strait of Georgia and Puget Sound. The Puget Sound/Georgia Basin International Task Force membership includes representatives from federal, provincial and state agencies, tribes, First Nations, and regional organizations from both sides of the international boundary (PSGBITF 2001).

In 1993, the British Columbia/Washington Environmental Cooperative Council appointed a Marine Science Panel (the Panel) comprised of six university and government marine scientists from British Columbia and Washington. The Panel was charged with making recommendations for management of the shared marine waters. After a year of analysis, the Panel presented twelve recommendations to the ECC in August 1994 which it believed the governments and citizens of the region should undertake as remedies for the highest priority environmental concerns. The EEC directed the Puget Sound/Georgia Basin International Task Force to develop actions to implement the Marine Science Panel's recommendations (PSGBITF 2001).

The International Task Force divided the Marine Science Panel's recommendations into high, medium and low priorities. Controlling toxic waste discharges was assigned a medium priority rating. The Panel noted that while some sources of toxic chemicals are being reduced, non-point sources of toxic chemicals remained a major obstacle to controlling sediment and microbial contamination in the shared waters. The Panel recommended that efforts to bring all point sources of toxic chemicals continue and additional emphasis be placed on controlling non-point sources of pollution including surface water runoff from urban and industrial areas (BCWMSP 1994).

The Task Force established Toxic Chemical Work Groups in British Columbia and Washington in 1998. The goals of these groups are to identify the toxic chemicals in shared waters, evaluate their risk, and propose to the British Columbia/Washington Environmental Cooperative Council and regulatory agencies a strategy to minimize this risk to the environment and economic well-being of our communities (PSGBITF 2001).

1.2 GEORGIA BASIN CLEAN WATER WORK GROUP

The Georgia Basin Clean Water Work Group (GBCWWG), co-chaired by Narender Nagpal of BC Ministry of Air, Land and Water and Ed Wituchek of Environment Canada (Pacific/Yukon Region), developed a Terms of Reference in 1998 charging the group to identify and prioritize potential risks to the environment and human health from anthropogenic toxic substances, including input and loadings of toxic chemicals, areas of significant contamination, impacts of population growth, and the transport of toxic substances across the international border. The GBCWWG is to notify the Puget Sound/Georgia Basin International Task Force of any urgent environmental or health issues, pertaining to toxic substances, requiring immediate action and the actions required. The Terms of Reference also include the identification of gaps in existing knowledge including research and monitoring, and a requirement to prioritize the filling of these gaps. A report addressing these issues will be provided to the Puget Sound/Georgia Basin International Task Force. The development of this report will include public consultation and will be coordinated with the United States counterparts. The report will develop options for mitigating the risks to the environment and human health from anthropogenic toxic substances, including strategies for implementation of these mitigation measures. These measures will be developed for toxic substances originating in Canada affecting only Canadian waters and, in cooperation with the United States work group, for toxic substances affecting the shared waters. The implementation plan will identify lead agencies and their potential roles (PSGBTWG 1998).

1.3 PUGET SOUND TOXICS WORK GROUP

The Puget Sound Toxics Work Group (PSTWG) is co-chaired by Mike Gallagher of the Washington Department of Ecology, and Mike Letourneau of the Region 10 office of the United States Environmental Protection Agency. Representatives on the PSTWG consist of state, federal, local governments, and representatives from tribes, businesses and non-government organizations residing in the Puget Sound region. A complete listing of members and their affiliations can be found in Table 1-1. The PSTWG met four times between 1999 and 2002:

- October 27, 1999 – Tacoma;
- May 18, 2000 – Olympia;
- January 31, 2001 – Seattle; and
- October 26, 2001 – Seattle.

In addition, a joint meeting of the Puget Sound Toxics Work Group and the Georgia Basin Clean Water Work Group was held on June 21, 2000 in Mount Vernon.

Table 1-1: Members of the Puget Sound Toxics Work Group

NAME	AFFILIATION
Co-Chairs	
Mike Gallagher	Department of Ecology
Mike Letourneau	US EPA Region 10
Environmental Interest Organizations	
Laurie Valeriano	Washington Toxics Coalition
Soundkeeper	Puget SoundKeepers Alliance
Kathy Fletcher	People for Puget Sound
Business/Industry Organizations	
Grant Nelson	Association of Washington Business
Bill Dewey	Taylor Shellfish Farms
Lincoln Loehr	Heller-Ehrman-White-McAuliffe
Tribal Nations	
Grant Kirby	Northwest Indian Fisheries Commission
Denise Dailey	Makah Tribe
Don Llopfer	Stillaguamish Tribe
Michael Cochrane	Northwest Indian College
Washington State Agencies	
Ann Wick	Washington Department of Agriculture
Gary Bailey	Washington Department of Ecology
Cheryl Niemi	Washington Department of Ecology
Sandi O'Neill	Washington Department of Fish and Wildlife
James E. West	Washington Department of Fish and Wildlife
Steve Jeffries	Washington Department of Fish and Wildlife
Joan Hardy	Washington Department of Health
Allison Ray	Washington Department of Transportation
Tim Goodman	Washington Department of Natural Resources
John Dohrmann	Puget Sound Action Team
Scott Redman	Puget Sound Action Team
Federal Agencies	
Dave Kendall	US Army Corps of Engineers
Mary Mahaffy	US Fish and Wildlife Service
Tracy Collier	NOAA
Christine Psyk	USEPA
Roseanne Lorenzana	USEPA
Dana Davoli	USEPA
Bruce Duncan	USEPA
Lon Kissinger	USEPA
Public Ports	
Eric Johnson	Washington Public Ports Association
Other	
Andrea Copping	Washington Sea Grant Program
Dan Riley	Western State Petroleum Association
Peter Hildebrandt	Environmental Consultant
John Calambokidis	Cascadia Research

Note: This list comprises members on the PSTWG mailing list; an average of 12 people attend workgroup meetings. The statements made in this report do not necessarily reflect the views of the members or their affiliations.

1.4 OVERVIEW OF REPORT

The Puget Sound Toxics Work Group was established to determine the status, trends, and gaps regarding toxic chemicals in the waters, sediments, and biota of Puget Sound. The Work Group works cooperatively with the Georgia Basin Clean Water Work Group to identify areas of mutual concern within shared waters of Puget Sound and Georgia Basin. It is the intent of the work group to report to the British Columbia/Washington Environmental Cooperation Council on their findings and make recommendations on furthering the control of toxic discharges into Puget Sound (PSTWG 2000).

At their May 18, 2000 meeting, the Puget Sound Toxics Work Group agreed, for discussion and reporting purposes, to adopt the physical and political boundaries established by the Marine Science Panel for Puget Sound. Figure 1-1 depicts the boundaries of Puget Sound and Georgia Basin as recognized by the Marine Science Panel. These boundaries extend to the northwestern point (Neah Bay) of Washington State, the southern marine water boundaries near Olympia, Washington, the Cascade mountains to the east and the international border to the north (BCWMSP 1994).

For the purpose of discussions and the generation of this report, the Puget Sound Toxics Work Group also agreed to adopt the Marine Science Panel's definition of a toxic which is any chemical that results in acute or chronic impairment of a biological organism. The Marine Science Panel defines impair as:

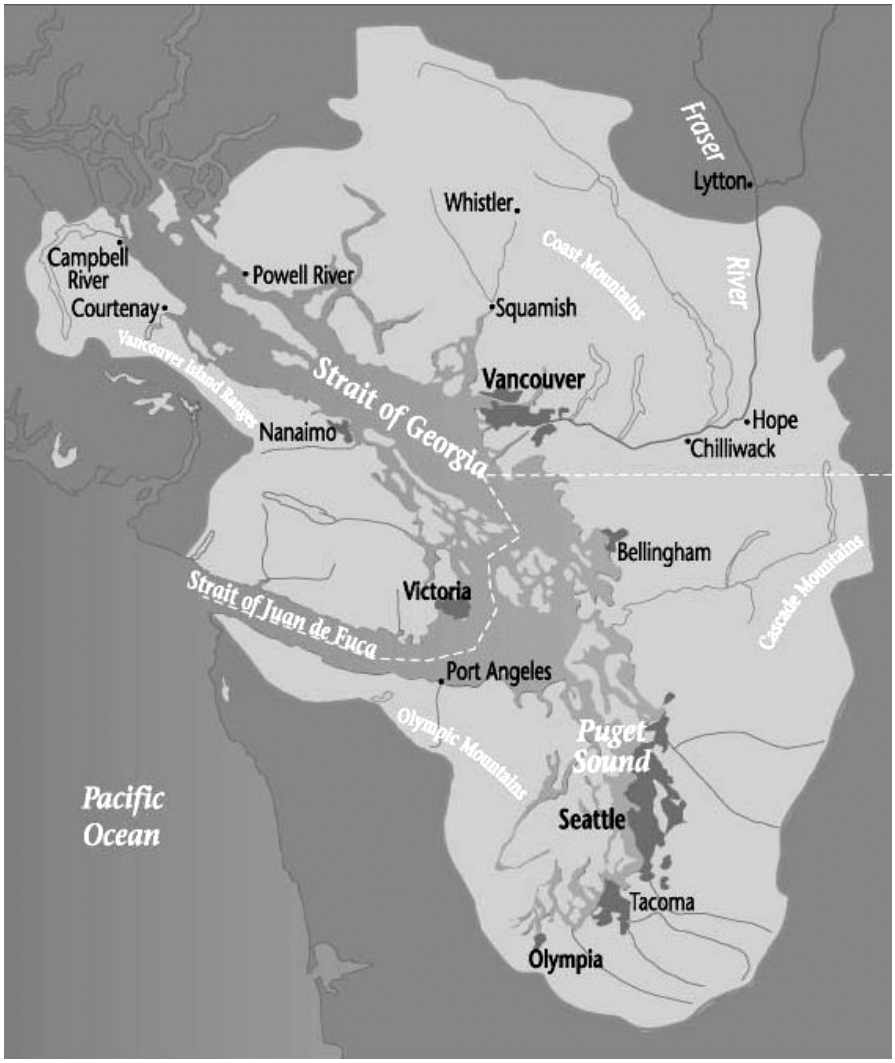
> to diminish strength, value, quantity or quality synonym: injure, biological as: of, pertaining to, caused by, or affection life or living organisms and organism as: any living individual; any plant or animal (BCWMSP 1994).

The Puget Sound Toxics Work Group, was provided funding by the Puget Sound Action Team, (pass-through funds from Region 10 EPA) to hire a contractor for research and generation of parts [the body] of this report. In addition, the PSTWG requested that the contractor propose recommendations for additional research and monitoring of toxics in Puget Sound. This report was developed utilizing published and unpublished reports, and interviews with PSTWG members and other experts working on toxics issues in Puget Sound. This report has been reviewed and approved by the PSTWG and the recommendations provided in this report include recommendations made by the contractor (EVS Environment Consultants) and the PSTWG. While this report is intended to meet the charge made to the PSTWG by the Puget Sound/Georgia Basin International Task Force, it is not intended to be a definitive report on all the toxics issues in Puget Sound.

This report is organized in a manner to give the reader a general overview of the toxic chemicals of concern in the Puget Sound Ecosystem, the pathways by which they enter and move through the various media and biota, and the effects of these chemicals on the biota. Where sufficient data exists, this report provides information on concentration trends of the various chemicals and compares concentrations to existing regulatory

criteria. In addition, this report identifies those agencies responsible for regulating and monitoring toxics in Puget Sound, and describes research efforts conducted by non-regulatory organizations.

Figure 1-1: Boundaries of Puget Sound and Georgia Basin



1.5 PUGET SOUND TOXICS WORK GROUP CO-CHAIRS

Questions and comments regarding this report can be made directly to the co-chairs of the Puget Sound Toxics Work Group.

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2. ENVIRONMENTAL CHEMISTRY AND TOXICITY OF SELECTED CHEMICALS OF CONCERN IN THE PUGET SOUND ECOSYSTEM:

The purpose of this section is to provide general background information on the sources, environmental chemistry and toxicity of the selected metals and organic contaminants that are the subject of this document.

2.1 METALS

All metals have a known natural abundance in the environment, as geochemical components of sediments, soils and rocks (Table 2-1). Natural weathering processes mobilize these compounds and transport them into streams, rivers and eventually, the ocean. However, greater quantities of metals generally enter the aquatic environment through anthropogenic sources such as fossil fuel combustion, industrial emissions, the discharge of municipal waste waters and via surface runoff (Table 2-2). For example, anthropogenic input of arsenic is estimated to be three times that from natural sources and the highest concentrations of arsenic in the natural environment occur near active and abandoned gold- and base-metals mines and ore processing facilities (Environment Canada 1993a).

Table 2-1: Natural abundance of metals in the earth's crust and the normal range found in soils.

ELEMENT	EARTH'S CRUST ($mg \cdot kg^{-1}$ dw)	NORMAL RANGE IN SOILS ($mg \cdot kg^{-1}$ dw)	ELEMENT	EARTH'S CRUST ($mg \cdot kg^{-1}$ dw)	NORMAL RANGE IN SOILS ($mg \cdot kg^{-1}$ dw)
Arsenic	1.5	0.1 - 40	Molybdenum	1.5	0.1 – 40
Cadmium	0.1	0.01 - 2.0	Nickel	80	2 – 750
Chromium	100	5 - 1500	Selenium	0.05	0.1 – 5
Cobalt	20	0.5 - 65	Silver	0.07	0.01 – 8
Copper	50	2 - 250	Thallium	0.6	0.1 - 0.8
Lead	14	2 - 300	Tin	2.2	1 – 200
Manganese	950	20 - 10000	Vanadium	160	3 – 500
Mercury	0.05	0.01 - 0.5	Zinc	75	1 – 900

Source: Alloway (1995)
dw – dry weight

Table 2-2: Summary of anthropogenic sources of metals to the aquatic environment.

Metal	Sources	
Arsenic	• Industrial emissions, • Insecticides, • Wood preservatives, • Fossil fuel combustion, • Industrial wastes,	• Past applications of lead-arsenate; • Active and abandoned gold- and base-metals mines, ore processing facilities
Cadmium	• Base-metal smelting and refining; • Metal plating; • Manufacture of pigments, batteries and plastics;	• Fossil fuel combustion; • Solid waste disposal and • Sewage sludge applications
Copper	• Industrial effluents, particularly from smelting, refining, and metal plating industries; and	• Municipal waste waters
Lead	• Manufacture of gasoline additives, pigments, alloys, and ammunition; • Vehicle maintenance and equipment repair; • Antifouling paint on ship hulls and bridges; • Tanks and piping;	• Petroleum refining; • Batteries; • Heavy metal soaps; • Automobile exhaust; • Past applications of lead-arsenate; • Municipal waste water discharges
Mercury	• Fossil fuel combustion; • Metal mining and reprocessing; • Removed and improperly disposed dental amalgam material	• Chloralkali plants; • Disposal of fluorescent lamps, thermometers, automobile light switches, and thermostats;
TBT	• Antifouling paint additive on ship and boat hulls, docks, fishnets, and buoys; • Fungicidal wood preservatives;	• Textile disinfectants; • Stabilizers in PVC resin.

Sources: Atkinson 1992; Eisler 1987a; Eisler 1988a; Environment Canada 1993a; Environment Canada 1994a; EXTOXNET 1996a; Garrett and Shrimpton 1997; USEPA 1985

2.1.1 Environmental Chemistry

The chemistry and behavior of metals in water can be complex and are dependent on a number of factors. Metals in the aquatic environment can exist in dissolved form, adhered to particulates, as part of organic and/or inorganic complexes, and in various oxidation states. In the marine environment, most metals will partition directly into sediments. Key factors influencing the chemistry, partitioning and bioavailability of metals include Eh (i.e., reduction-oxidation, or redox, conditions), pH, hardness, and the presence of organic carbon and other compounds with which metals will combine.

2.1.1.1 Arsenic

Arsenic is constantly being cycled in the aquatic environment and because of the natural presence of arsenic (largely as arsenate or As^{5+}) in seawater, is ubiquitous in the living tissue of marine organisms (Eisler 1988a). Arsenic in water is primarily in a dissolved ionic state with very little associated with particulates. Background concentrations of total arsenic in the ocean are typically in the order of $< 10\ \mu g \cdot L^{-1}$. The metal will precipitate out of the water column in association with iron and other compounds. Once in the sediments, arsenic can remain stable for 100 years or more. Arsenic in ocean sediments ranges from non-detectable to 455 $mg \cdot kg^{-1}$ on a dry weight (dw) basis, while sediments from areas contaminated by smelters can have concentrations ranging from 200 to 3,500 $mg \cdot kg^{-1}$ dw. Arsenic forms both inorganic and organic compounds, depending on pH, redox potential and other physical parameters (Eisler 1988a).

2.1.1.2 Cadmium

Cadmium is a natural constituent of surface waters and occurs in higher concentrations in seawater than in freshwater. Typical background concentrations of cadmium in coastal seawaters are around 0.05 $\mu g \cdot L^{-1}$ (Eisler 1985a). Cadmium reaches sediments from the water column primarily through uptake by plankton or binding of the metal to organic matter and the subsequent settling of detritus (Environment Canada 1994a). Background cadmium concentrations range from 30 to 1,000 $\mu g \cdot kg^{-1}$ in marine sediments.

Cadmium is consistently recycled in a similar pattern exhibited by nutrients (Burton and Stathan 1990, cited in Environment Canada 1994a). For example, elevated cadmium levels may be found in coastal areas without identifiable anthropogenic sources as a result of the oceanographic process of upwelling. Plants also play an important role in the cycling of cadmium as the metal can be taken up from the sediments by aquatic macrophytes, and then released to the water column when the plant decays (Eisler 1985a).

2.1.1.3 Copper

Total copper concentrations in ocean water range from 0.06 to 6.7 $\mu g \cdot L^{-1}$, while contaminated estuaries have concentrations ranging from 3 to 176 $\mu g \cdot L^{-1}$ (Eisler 1998). Copper in the water column is primarily associated with particulate matter and therefore is generally not bioavailable. The distribution of copper is controlled mainly by physical processes such as advection (i.e., horizontal movement in the water column) and settling of particulates. In Puget Sound, for example, advection removes about 60% of the total copper added to the main basin while the remaining 40% is deposited into the sediments (Paulson et al. 1988). Contaminated estuarine sediments can have copper concentrations in excess of 2,000 $mg \cdot kg^{-1}$ while uncontaminated estuaries have concentrations in the order of 10 $mg \cdot kg^{-1}$.

2.1.1.4 Lead

The solubility of lead in water is low and the metal tends to concentrate in the surface microlayer of the water column (particularly if organic matter is present), bind to suspended solids or precipitate into the sediments (Eisler 1988b). Ambient lead concentrations in seawater are ~0.005 $\mu g \cdot L^{-1}$, and in coastal sediments range from 1 to 912 $mg \cdot kg^{-1}$ (ATSDR 1999). Lead in the environment has decreased significantly since it was banned as an additive in gasoline, however, sediments are considered a significant global reservoir of lead (Eisler 1988b). As a result, lead-contaminated sediments continue to be a source of the metal to aquatic organisms. Plants and crustaceans can take lead up from the sediments via absorption, and when they die, or molt in the case of crustaceans, lead is released (Eisler 1988b).

2.1.1.5 Mercury

Mercury concentrations in the open ocean are < 0.01 $\mu g \cdot L^{-1}$, while in coastal seawaters they are < 0.02 $\mu g \cdot L^{-1}$ (Eisler 1987a). Background sediment concentrations of mercury are in the order of 0.02-0.1 $mg \cdot kg^{-1}$, while sediments near significant human sources such as chloralkali plants can have concentrations > 10 $mg \cdot kg^{-1}$. Mercury in water and sediment is mainly inorganic; however, inorganic mercury can be liberated from a sediment sink via methylation, even after the original source ceases (Grey et al. 1995). Biomethylation is enhanced by large amounts of available mercury, large numbers of bacteria, the absence of strong complexing agents, near-neutral pH, high temperatures, and moderately aerobic environments.

Background concentrations of mercury in biota are usually < 1.0 mg $Hg \cdot kg^{-1}$ fw, while organisms from contaminated areas can have tissue concentrations significantly > 1.0 mg $Hg \cdot kg^{-1}$ fw, primarily in the form of methylmercury (Eisler 1987a).

2.1.1.6 Tributyltin

Other than some methyltin compounds, organotins are entirely synthetic, therefore any measurable concentration of tributyltin is related to a human source (Eisler 1989). The highest environmental concentrations of TBT have generally been found in the vicinity of marinas and shipyards. For example, at UK marinas, reported TBT concentrations are in the order of 2.0 $\mu g \cdot L^{-1}$, while in coastal waters ~0.5 $\mu g \cdot L^{-1}$ concentrations have been reported. TBT can be as much as 10,000 times higher in the sea-surface microlayer than in the water column, likely due to partitioning into petroleum hydrocarbon sheens at surface (Eisler 1989). Observed sediment concentrations range from non-detectable to ~1.5 $mg \cdot kg^{-1}$.

TBT on its own is quite unstable and will break down in the environment unless it is combined with an element such as oxygen. Degradation rates of TBT can be quite rapid in the water column, however, it readily adsorbs to sediments and suspended solids,

where upon the chemical becomes highly persistent (i.e., a half-life ranging from months to years; Garrett and Shrimpton 1997). Micro-organisms, aquatic biota and photolysis are the main pathways of degradation or removal of TBT from the aquatic environment. Once degraded, TBT forms less toxic, as well as less hydrophobic products such as dibutyltin (DBT), monobutyltin (MBT) and ultimately inorganic tin.

2.1.2 Toxicity

The toxicity of metals to aquatic organisms ranges widely from slight reduction in growth rates to mortality and may be manifested acutely (i.e., after a short term exposure) or over a longer term (i.e., chronically). The expression of toxic effects is dependent on several factors including:

- Exposure route, duration and concentration;
- The form of the metal at the time of exposure (e.g., inorganic arsenic is more toxic than the organic form, while methylmercury is more toxic than inorganic mercury), which can be affected by site-specific physical and chemical conditions (e.g., pH, redox);
- External and internal synergistic, additive or antagonistic interactions of co-occurring contaminants (e.g., cadmium has been observed to reduce the teratogenic effects of methylmercury on fish);
- Sensitivity of a given organism (e.g., mollusks are generally less sensitive to metals than other aquatic phyla);
- Life stage (e.g., embryonic and larval stages of benthic organisms are generally more sensitive);
- Physiological ability to detoxify and/or excrete the metal (e.g., some vertebrates synthesize a protein, metallothionein, which can sequester metals); and,
- The condition of the exposed organism (e.g., a fish that is stressed by elevated water temperatures is more sensitive to toxicant exposure).

2.1.2.1 Arsenic

Arsenic forms both inorganic and organic compounds, depending on pH, redox potential and other physical parameters (Eisler 1988a). In the recent past, it was understood that the inorganic forms of arsenic were responsible for the toxic effects in organisms exposed to the metal. However, there is increasing concern regarding the toxicity of methylated arsenic species, as well (Thomas et al. 2001). Arsenic is bioaccumulated by marine organisms, concentrating in the fatty tissues, but does not biomagnify through the food chain.

Marine organisms can contain up to 100 mg $As \cdot kg^{-1}$ dw, compared to typical background concentrations of < 1 $mg \cdot kg^{-1}$ in freshwater organisms. However, the tissue burden in

marine animals is usually in an organic form, arsenobetaine, that is not toxic to either the organism or the consumer (Eisler 1988a; Francesconi and Edmonds 1993).

The preponderance of research regarding the effects of arsenic have pertained to exposure to inorganic arsenic, such as may occur via poisoning from pesticides (ATSDR 2000a).

Invertebrates - Water concentrations greater than 100 $\mu g \cdot L^{-1}$ inorganic arsenic have been observed to cause reduced survival and embryonic malformations in marine invertebrates (Environment Canada 1993a). Sediment concentrations in the range of 100 – 700 $mg \cdot kg^{-1}$ dw have had similar effects, ultimately resulting in a reduction in the abundance of higher level benthic invertebrates.

Fish - Fish exposed to inorganic arsenic via diet were observed to have impaired growth, avoided food and had impaired food metabolism (Eisler 1988a). Gill hemorrhaging and necrosis of the liver, heart and ovaries have been observed in fish exposed to arsenic salts via water.

Birds - Symptoms of acute inorganic arsenic poisoning in birds include muscular incoordination, slowness, jerkiness, unkempt appearance, and seizures within a few hours of exposure, with death following a day or two later (Eisler 1988a). Early life stages are the most sensitive to arsenic exposure (Environment Canada 1993a).

Mammals - Chronic effects in mammals exposed to arsenic is not common, likely due to rapid detoxification and excretion of low doses (Eisler 1988a). Rather, arsenic toxicity is generally acute and related to consumption of contaminated vegetation and water. Signs of acute exposure include gastric disturbance, extreme weakness, collapse and death. In cases where chronic exposure to organic arsenic forms have resulted in effects, depressed growth and immune impairment have been observed.

Humans - Acute poisoning in adult humans usually results from the ingestion of large doses of inorganic arsenic (e.g., poison) or from inhalation (ATSDR 2000a). Skin lesions (cancerous and non-cancerous), and lung, bladder and urinary tract cancers have been documented in people exposed to inorganic arsenic from contaminated drinking water (NRC 2001). Arsenic has also been implicated in reproductive effects, however, the studies available to date are inconclusive. The organic form of arsenic, to which humans would be exposed through consumption of fish/shellfish, has not been demonstrated to have effects at the doses typically consumed (ATSDR 2000a).

2.1.2.2 Cadmium

The free ion (Cd^{2+}) is the most toxic form of cadmium. Cadmium at the background water and sediment concentrations described in Section 2.1.1.2 can be toxic when conditions such as pH shift to favor the free ion. For example, cadmium bound to sediment particles may become bioavailable once ingested due to internally reduced pH levels (NOAA 1994).

The toxicity of cadmium is lessened at lower temperatures and higher salinities, therefore marine organisms tend to be less susceptible to cadmium-related effects than are freshwater organisms (Eisler 1985a). As well, marine biota generally contain significantly higher cadmium residues in their tissue than do freshwater or terrestrial organisms, probably due to higher total cadmium levels in seawater (NOAA 1994).

While cadmium is naturally occurring in the aquatic environment, it is not a biologically essential element. Cadmium is bioaccumulated by all trophic levels, but does not biomagnify (ATSDR 1999a). The metal has been identified as an endocrine disrupting compound (EDC), and teratogenic effects are higher from cadmium exposure than from other metals.

Invertebrates - Lethal effects resulting from cadmium exposure have been observed in marine invertebrates at water concentrations ranging from 5-5,000 $\mu g \cdot L^{-1}$, and sediment concentrations ranging from 50-200 $mg \cdot kg^{-1}$ (Environment Canada 1994a). Sublethal effects (including inhibition of reproduction, decreased growth and population alterations) may occur at water concentrations of 0.2-2 $\mu g \cdot L^{-1}$, and at sediment concentrations of ~6-10 $mg \cdot kg^{-1}$. Crustaceans are the most sensitive marine invertebrates to cadmium exposure.

Fish - High water-borne cadmium concentrations appear to affect fish via gill damage and accumulation, while lower-concentration, longer-term exposures affect the intestine and kidney (Eisler 1985a). Sublethal effects include reduction in growth, survival and fecundity, respiratory disruption, abnormal muscle contractions, and altered enzyme levels. Teratogenic effects include edema, microcephalia and developmental malformations.

Birds - Birds are relatively resistant to cadmium. When exposure concentrations and durations are high enough though, sublethal effects including growth inhibition, anemia, and testicular damage (Eisler 1985a).

Mammals - Mammals also appear to be relatively resistant to cadmium, however, when exposure concentrations and durations are high enough, teratogenic effects have been observed (Eisler 1985a).

Humans - The effects of environmental exposure of humans to cadmium have not been studied extensively and have focused on people living in the vicinity of point sources such as smelters or who were chronically exposed via contaminated rice. The subjects in such studies have manifested decreased bone density and kidney dysfunction (Environment Canada 1994a). Carcinogenicity in humans has been observed as a result of inhalation of cadmium (i.e., smoking, occupational exposure to aerosols/dust).

2.1.2.3 Copper

Copper is an essential element and is readily taken up, and therefore generally found in most animal tissues. It is important in several enzyme systems in vertebrates, and marine

gastropods use copper in a blood pigment (Eisler 1998). When copper is ingested in excess of nutritional requirements, the metal may be stored in the liver or bound to the protein metallothionein. The ability of the liver to store copper varies among species (Leland and Kuwabara 1985). The concentration of copper in biota appears to decrease with increasing trophic level. The free ion Cu^{2+} is considered the most toxic form of copper (Eisler 1998).

Invertebrates - Copper can affect transport across epithelial surfaces in marine gastropods, and can interfere with oxygen binding to the oxygen-carrying molecule in crabs (Eisler 1998).

Fish - Acute water-borne copper toxicity to fish results through gill damage and subsequent osmoregulatory failure (Leland and Kuwabara 1984). The metal also affects the transport of oxygen across the gill membrane (Eisler 1998). Fish synthesize metallothionein, which can sequester copper, thereby rendering it biologically unavailable.

Birds - Copper can effect growth rates, egg production and development in birds (USEPA 2003).

Mammals - Mammals are relatively tolerant to copper. However, chronic exposure of piscivorous wildlife to the metal can lead to liver damage, impaired reproduction, reduced resistance to diseases, jaundice and death. Copper consumed in excess of metabolic requirements is sequestered by metallothionein in gastrointestinal cells, stored in the liver or incorporated into bile, and eventually excreted (Eisler 1998).

Humans - In humans, toxic exposure to copper is more likely to occur as a result of inhalation (e.g., occupational exposure to dust) or dermal contact than ingestion of contaminated fish/shellfish. Gastrointestinal effects (i.e., anorexia, nausea, diarrhea) have been observed in people exposed to copper in drinking water (ATSDR 1990).

2.1.2.4 Lead

Lead will bioconcentrate in aquatic organisms and older organisms tend to have higher concentrations (particularly in bony tissue), but lead has not been observed to biomagnify through the food chain (Eisler 1988b). Organic forms such as trialkyl lead and tetraalkyl lead are more toxic than inorganic lead. Lead has been identified as an EDC.

Invertebrates - Many species of aquatic organisms, including mollusks, can have tissue concentrations in the order of 1,000 mg lead$\cdot kg^{-1}$ fw (soft tissue) without suffering any apparent effects in the individual or their progeny (Eisler 1988b). Lead appears to be selectively accumulated in the digestive gland of mollusks rather than in the edible tissue. Waterborne lead concentrations of ~ 1.0 µg $Pb^{2+}\cdot L^{-1}$ have been observed to affect copepod reproduction.

Fish - Lead is primarily accumulated in the epidermis (skin and scales), bone and intestine of fish, with very little accumulating in edible tissues (Pascoe and Blanchet 1993). Uptake is greatest during periods of rapid growth. Acutely lethal concentrations of lead cause excess mucous production in the gills, which can result in asphyxiation of the fish (Eisler 1988b). Symptoms of lead poisoning in fish exposed to sublethal water-borne lead concentrations (i.e., > 10 $\mu g \cdot L^{-1}$) include spinal curvature, anemia, renal dysfunction, neuropathological effects, growth inhibition and reproductive effects (Eisler 1988b).

Birds - Birds are most seriously affected (i.e., reduced survival; impaired reproduction) by lead in the form of shot, either through direct ingestion which typically happens to waterfowl like ducks and geese, or through the ingestion of waterfowl that have eaten lead shot, which may happen to birds of prey like eagles (Eisler 1988b). The form of lead ingested is important; carnivorous birds ingesting prey contaminated by other forms of lead have not exhibited the same toxic effects.

Mammals - There are insufficient data to determine the effects of lead in feral mammals.

Humans - In humans, lead causes gastrointestinal distress, and can interfere with the synthesis of heme, a component of the oxygen-carrying molecule in blood, resulting in anemia. Impaired brain function and neurobehavioral effects are also significant effects (ATSDR 1999b).

2.1.2.5 Mercury

Methylmercury is the most toxic and most readily accumulated and retained form of mercury in aquatic biota, and once it enters a biological system it is very difficult to eliminate (Clement Associates 1985). Mercury is the only metal known to biomagnify up food chains, making it of particular risk to humans and animals in the upper trophic levels.

Toxic effects generally occur because mercury binds to proteins and alters protein synthesis (Beckvar et al. 1996). Both inorganic and methylmercury are taken up easily by aquatic organisms, however, methylmercury is accumulated much more efficiently and is depurated much more slowly. Mercury is considered an EDC.

Invertebrates - Low concentrations of mercury in water can delay larval development, prolonging the length of the pelagic stage and thereby increasing the potential for predation (Beckvar et al. 1996). Mercury in sediments has been observed to inhibit burrowing response and larval settlement, therefore also increasing the risk of predation. Invertebrates usually contain lower proportions of methylmercury than fish or mammals, although there are significant interspecies differences (e.g., 1% in a polychaete compared to 100% in Dungeness crab).

Fish - Acute exposure of fish to mercury can cause increased respiration, sluggishness, and a loss of equilibrium (Eisler 1987a), while chronic exposure can result in effects such as brain lesions, emaciation, cataracts, changes in behavior (i.e., inability to capture food, abnormal motor coordination). Maternally-transferred mercury can exert a greater toxic effect on developing embryos than mercury in water or sediments (Beckvar et al. 1996).

Birds - Signs of mercury poisoning in birds include impaired muscular control including lack of coordination, falling and slowness (Eisler 1987a). Mercury also affects the reproduction, growth and development, and behavior of birds. The highest concentrations of mercury in birds exposed to mercury have been found in the brain, followed by the liver, kidney and muscles.

Mammals - At relatively low concentrations in mammals, mercury can adversely affect reproduction, growth and development, behavior, blood and serum chemistry, motor coordination, vision, hearing, histology, and metabolism (Eisler 1987a). Larger mammals like seals appear to be more resistant to mercury poisoning than smaller ones like river otters. Marine mammals can bind mercury with metallothionein and selenium, rendering it non-toxic (Beckvar et al. 1996).

Humans - In adult humans the target organs for mercury toxicity are the brain/central nervous system (especially the sensory, visual and auditory sections associated with coordination) and the kidneys (Eisler 1987a). Mercury can also be maternally transferred to developing fetuses, causing brain damage, incoordination, blindness and seizures.

2.1.2.6 Tributyltin

Bioconcentration of TBT by aquatic organisms is high, but degradation (i.e., debutylation) tends to be sufficiently rapid that the compound is not biomagnified (Eisler 1989).

Invertebrates - TBT effects are greatest during larval and juvenile life stages, and crustaceans tend to be particularly sensitive (USEPA 1997). Bivalve mollusks are also sensitive and when exposed to low concentrations of TBT in water, adverse effects included mortality of larvae, abnormal development of the shell and gonad, and reduced larval settlement and growth (Garrett and Shrimpton 1997). Imposex, the development of male sex organs in females, has been observed in neogastropods near industrial/marine shipping areas (Garrett and Shrimpton 1997). Imposex has been observed world-wide and has resulted in the extinction of some populations of the more sensitive gastropod species in areas of heavy TBT-pollution due to reduced reproductive capacity (Matthiessen and Gibbs 1998).

Fish - The acute toxicity of TBT to fish is variable, ranging from water concentrations of 1.5-200 $\mu g \cdot L^{-1}$. Sublethal effects have been observed at the lower end of this range and include enzyme disruption, impaired growth, behavioral abnormalities, histopathological

changes, reduced egg hatchability, and gross internal abnormalities (Garrett and Shrimpton 1997).

Birds - Research regarding the effects of TBT on birds is scarce. However, TBT has been observed to affect egg production, eggshell thickness, fertility and hatchability at high doses (EXTOXNET 1996a).

Mammals - High doses in mammals can affect endocrine glands (i.e., upset pituitary, gonad and thyroid hormones), reproductive, immune and central nervous systems, and bone structure (EXTOXNET 1996a).

Humans - Research regarding the effects of TBT on humans, particularly through oral exposure, is limited (ATSDR 1992). In one case, neurotoxic effects (e.g., vertigo, headache, altered consciousness, visual impairment, convulsions) and death were observed in people administered a drug containing organotins. Organotins have not been identified as mutagenic, teratogenic or carcinogenic (Eisler 1989).

2.2 ORGANIC COMPOUNDS

Some organic compounds, such as polycyclic aromatic hydrocarbons (PAHs) and dioxins and furans can be naturally occurring (e.g., produced during forest fires) or are byproducts of industrial processes, and have no known uses. PAHs and dioxins and furans continue to be discharged into the environment from both natural and anthropogenic sources. Other organics such as polychlorinated biphenyls (PCBs) are entirely synthetic and were not found in the environment prior to initial production in the mid-20th century. Even though the manufacture and use of PCBs and organochlorine pesticides ceased in the 1970's, they can still be found in the aquatic environment today and continue to affect aquatic organisms. The anthropogenic sources of selected organic contaminants are summarized in Table 2-3.

2.2.1 Environmental Chemistry

Organic compounds range from the simple methane molecule to long-chained, multi-ringed, halogenated structures that vary in persistence in the environment and effects on aquatic organisms. The fate and transport of organic compounds in environmental systems is controlled by the partitioning of the compound between sediment, suspended particulates, pore water, surface water and biota. The observed partitioning of non-ionic organic chemicals like PCB is due to sorption to organic phases, including dissolved organic matter in pore water and sedimentary organic matter. The extent to which a chemical is associated with organic matter relative to their dissolved aqueous concentrations is related to a number of factors including molecular weight, and number and position of chlorine atoms in the case of chlorinated compounds.

Table 2-3: Summary of anthropogenic sources of selected organic compounds.

Organic Compound	Sources	
PCBs	• Cooling and insulating fluids in industrial transformers and capacitors; • Hydraulic fluids; • Heat transfer fluids; • Plasticizers;	• Paints; • Inks; • Dust control agents; • Carbonless paper; and • Pesticides.
PAHs	• Aluminum smelters; • Metallurgical and coke production; • Creosote-treated products (e.g., marine pilings); • Petroleum spills;	• Municipal effluents (including stormwater runoff); and • Deposition of atmospheric PAHs.
Pesticides	• Runoff from agricultural areas; • Runoff from residential areas;	• Industrial emissions.
Dioxins and furans	• Manufacture of chlorophenols and their derivatives (used in wood treatment); • From bleaching processes in pulp and paper mills; • Incomplete incineration of other chlorinated organic compounds such as PCBs; • Ash residues and emissions of municipal waste incinerators; • Fossil fuel power plants;	• Internal combustion engines; • Beach bonfires; • Residential burning of garbage; • Home fireplaces; • Cigarette smoke; and • Trace impurities in some commercial herbicides such as 2,4,5-t, 2,4-d and 2,4,6-trichlorophenol.
Phthalate esters	• Plasticizer in polyvinyl emulsions; • Antifoaming agents; • Suspension agent for solids in aerosols; • Lubricant for aerosol valves;	• Perfume solvent and fixative; • Skin emollient; • Plasticizer in nail polish and hair spray; and, • Municipal waste sludges.

Sources: Alford-Stevens 1986; Atkinson 1992; Environment Canada 1994b; IEMPOP 1995; Kociba and Schwetz 1982a,b; NOAA 1994; PTI 1991.

2.2.1.1 Polychlorinated biphenyls (PCBs)

Polychlorinated biphenyls are complex mixtures of individual compounds composed of two connected benzene rings with 1 to 10 chlorine atoms attached in one of 209 combinations (Clement Associates 1985). The individual compounds, known as congeners, have different chemical and physical properties based mostly on the degree of

chlorine substitution and, to a lesser extent, on the position of the chlorine substitution (Phillips 1986; Safe 1990). Commercial mixtures of PCBs have a variety of trade names, but the most familiar is Aroclor (produced by the Monsanto Corporation in the United States). Different Aroclors were made based on their chlorine content by weight (i.e., Aroclor 1254 is 54% chlorine by weight, while Aroclor 1260 is 60% chlorine) and are complex mixtures of different congeners with very different properties and potentially different toxicological effects. For example, di-ortho coplanar PCB is about 5,000 times less toxic than 3,3',4',5-pentachlorobiphenyl (Safe 1990). Environmental PCB concentrations are often reported as Aroclor equivalents or total PCBs, or as individual congeners depending on the analytical method used.

Adsorption to organic material in sediments and soils is probably the major fate of at least the more highly-chlorinated PCBs. Once bound, PCBs may persist for years with slow dispersion providing continuous, low-level exposure to the surrounding environment. Some soil micro-organisms can biodegrade low-chlorinated PCB. Heavier PCB (i.e., those with five or more chlorines) can slowly be photolyzed by ultraviolet light, via reductive dechlorination, in aquatic environments (Clement Associates 1985; Sawhney 1986). Anaerobic bacteria can also degrade higher chlorinated congeners, however, degradation is often incomplete (ATSDR 2000b).

Although they are no longer produced and used in the United States, PCBs in sediments can be a significant and continual source of contamination for aquatic organisms, with uptake occurring directly via respiratory intake of interstitial or overlying water, dermal sorption, and ingestion of sediment, or indirectly through trophic transfer. Sediment PCB availability to aquatic organisms depends on the concentrations of the specific PCB compound present, physical properties of the sediment, environmental factors, and characteristics of the organisms (Clement Associates 1985).

2.2.1.2 Polycyclic aromatic hydrocarbons (PAHs)

Polycyclic aromatic hydrocarbons (PAHs) are organic compounds made up of carbon and hydrogen atoms grouped into rings containing five or six carbon atoms. PAHs fall into two categories: low molecular weight PAH compounds (LPAHs) composed of fewer than four rings and high molecular weight PAH compounds (HPAHs) of four or more rings (Environment Canada 1994b; Eisler 1987b). PAHs commonly measured in environmental media are listed in Table 2-4.

Table 2-4: List of commonly measured low and high molecular weight polycyclic aromatic hydrocarbons (PAHs).

LOW MOLECULAR WEIGHT PAHs (LPAHs)	HIGH MOLECULAR WEIGHT PAHs (HPAHs)	
Acenaphthene	Benzo[a]anthracene	Chrysene
Acenaphthylene	Benzo[b]fluoranthene	Dibenzo[a,h]anthracene
Anthracene	Benzo[k]fluoranthene	Fluoranthene
Fluorene	Benzo[g,h,i]perylene	Indeno[1,2,3-cd]pyrene
Naphthalene	Benzo[a]pyrene	Pyrene
Phenanthrene		

Because of their low water solubility, PAHs tend to adsorb to particulate matter. Sediments are the major environmental sink for PAHs in the aquatic environment; as a result, most PAHs remain relatively near their point sources (Neff 1979). HPAHs such as benzo[a]pyrene predominate in sediments as LPAHs are more likely to be lost through physical processes.

PAHs in sediment are persistent; for example, the biodegradation half-life of benzo[a]pyrene in sediments can be up to 58 years (Environment Canada 1994b). However, some degradation does occur and is controlled by many factors. The most rapid biodegradation of PAH compounds occurs at the water/sediment interface, and is attributed to the oxidation of PAHs by prokaryote organisms as the first step in metabolism (Brooks 1993). HPAHs are more resistant to microbial degradation than lighter weight PAHs. Furthermore, degradation of PAHs decreases with increasing anaerobic conditions in sediments, and PAHs in oxygen-poor basins or in anoxic sediments can persist indefinitely (Eisler 1987b).

PAHs are highly hydrophobic and lipophilic chemicals, therefore bioaccumulation is a key process in their fate (Nagpal 1993). Uptake of PAHs is dependent on the specific compound (e.g., anthracene versus chrysene), but generally, where sediment concentrations are elevated, benthic organisms obtain a majority of their PAHs from sediment through their ability to mobilize PAHs from the sediment/pore water interface (Eisler 1987b).

2.2.1.3 Pesticides

Pesticides are a broad group of chemicals (including organic and inorganic compounds) used to kill or otherwise control pest organisms (i.e., weeds, insects, fungus). They may target particular organisms, or biological pathways (e.g., cholinesterase activity) or they may affect a broad spectrum of organisms.

Organochlorine (OC) pesticides are synthetic chlorinated hydrocarbons that have low volatility, are chemically stable, highly soluble in lipids, and have a slow rate of

biotransformation and degradation. DDT, aldrin/dieldrin, and endosulfan are examples of OC pesticides that have been used in the Puget Sound area. Degradation of organochlorines is frequently partial, resulting in a product with reduced chlorination but remaining highly persistent (e.g., DDT degrades to DDD and DDE, both of which are persistent). Some chemicals have comparatively short half lives but the resulting metabolites are either equally toxic (e.g., heptachlor and its active metabolite, heptachlor epoxide) or much more toxic and persistent that the parent compound (e.g., oxychlordane is much more toxic to mammals than its parent compound chlordane).

The persistence of these compounds in the environment and their tendency towards bioconcentration and biomagnification within various food chains led to the decline in use and eventual ban of organochlorine pesticides in North America and Europe. Many developing nations continue to use them due to the low cost of manufacture, effectiveness, and limited available alternatives. As a result, they remain contaminants of concern internationally (Brown 1978; Ecobichon 1991).

The environmental fate and effects of organochlorine pesticides is complex due to the differences in behavior of even closely related compounds. In general, however, the major sink for OC pesticides in the marine environment is sediments (IEMPOP 1995). Because they are lipophilic and resistant to metabolism, organochlorines tend to bioaccumulate in the fatty tissues of marine organisms and biomagnify up the food chain.

Organophosphorus (OP) and carbamate pesticides are much less persistent than organochlorine pesticides, with degradation occurring in weeks rather than years. Diazinon, malathion and parathion are examples of OP pesticides and carbofuran is an example of a carbamate pesticide that have been used in the Puget Sound area. Diazinon is considered relatively persistent for an OP pesticide (e.g., 75% lost after 12 weeks) while parathion and malathion are lost from soil after 1 week. In water, most OP and carbamate insecticides are chemically hydrolyzed and biodegraded by microorganisms under aerobic conditions (Eisler 1985b; 1986b). Bioaccumulation does not appear to be an important fate process for either OP or carbamate pesticides as they tend to readily convert to non-toxic breakdown products.

Synthetic pyrethroids are the most recent group of organic insecticides to be manufactured and were introduced less than 20 years ago. These compounds are synthetic analogues of natural pyrethrins (which are found in a particular species of chrysanthemum). Pyrethroid pesticides appear to partition rapidly to sediment and organic particles once they enter the aquatic environment. The persistence of pyrethroid pesticides in the aquatic environment appears to be limited (e.g., they have been detected for, at most, several months in sediments following introduction into the aquatic environment). The relatively limited persistence following field application suggests that exposure to aquatic species is temporary, intermittent, and restricted. The persistence of pyrethroids in aquatic species is largely unknown, although this class of compounds is expected to be rapidly metabolized by both invertebrate and fish species. As with

persistence in sediments, tissue residues may seasonally fluctuate in a pattern consistent with agricultural uses (EVS 1993).

Inorganic pesticides include metal oxides such as chromated copper arsenate (CCA) which is used as a wood preservative (Hutton and Samis 2000). See Section 2.1.1 for a discussion regarding the environmental chemistry of copper and arsenic.

2.2.1.4 Dioxins and Furans

Polychlorinated dibenzo-*p*-dioxins (PCDDs) are part of a class of parameters more commonly referred to as chlorinated dioxins. Similarly, polychlorinated dibenzofurans (PCDFs) are part of a class of parameters more commonly referred to as chlorinated furans. There are 75 congeners of dioxins and 135 congeners of furans, varying in the number and placement of chlorine atoms (Safe 1990). Dioxins include 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-TCDD), which is considered the most toxic form and is widely distributed in the environment (PTI 1991; Environment Canada 1993c).

Dioxins and furans are persistent and, because of their hydrophobic nature and general resistance to metabolism, biomagnify in organisms. Dioxins and furans move from sediments into organisms via direct accumulation from sediment and from pore water into the dermis and gills of benthic invertebrates and forage fish. They are then transferred to fish and birds via ingestion. Some exposure will also occur through the incidental ingestion of contaminated sediments (Eisler 1986c).

2.2.1.5 Phthalate Esters

The most widely used and commonly detected phthalate ester in the environment is bis(2-ethylhexyl)phthalate (DEHP). DEHP concentrations in surface waters near industrial facilities that use or produce it have ranged from 0.6 to 2,400 $\mu g \cdot L^{-1}$, whereas average seawater concentrations are in the order of 0.005 to 0.7 $\mu g \cdot L^{-1}$ (ATSDR 2002b). Marine and freshwater sediment concentrations near industrial outfalls generally range from 6.0 to 1,500 $\mu g \cdot kg^{-1}$, while in the vicinity of combined sewer overflows, reported concentrations ranged from 900 to 27,000 $\mu g \cdot kg^{-1}$. Fish/shellfish tissue concentrations ranging from 2 to 32,000 $\mu g \cdot kg^{-1}$ have been reported.

Environmental transport of phthalates most frequently occurs by adsorption onto suspended solids and complexation with organic compounds. They readily interact with organic substances in water and soil, forming a very soluble compound in water (Clement Associates 1985). Bioaccumulation is considered an important fate process as a variety of unicellular and multicellular organisms take up and accumulate phthalate esters. However, because biodegradation is common, and phthalate esters are metabolized by multicellular organisms, it is unlikely that long-term bioaccumulation or biomagnification will occur (Environment Canada 1994c).

2.2.2 Toxicity

Organic compounds have a wide range of effects on aquatic organisms, from reproductive impairment such as reduced fecundity and viability of offspring, developmental impairment such as brain and skeletal deformations and reduced growth, to acute mortality of both adults and juveniles. Of particular concern are the persistent, bioaccumulative and toxic compounds (or PBTs). Because many organics are lipophilic, they tend to accumulate in fatty tissues unless the organism has a mechanism for metabolizing and excreting the compound. The contaminants can then be biomagnified up the food chain, resulting in exponentially higher concentrations in higher trophic level organisms such as carnivorous marine mammals.

As with metals, the manifestation of toxic effects related to organic contaminants is dependent on several factors including:

- Exposure route, duration and concentration;
- The composition of the chemical (e.g., LPAH are more toxic but less carcinogenic than HPAH);
- Sensitivity of a given organism (e.g., birds are less sensitive to synthetic pyrethroids than invertebrates and fish are);
- Life stage (e.g., embryonic and larval stages of benthic organisms are generally more sensitive);
- Physiological ability to detoxify and/or excrete the compound (e.g., fish are able to rapidly excrete the OP pesticide chlorpyrifos); and
- The condition of the exposed organism (e.g., a fish that is stressed by elevated water temperatures is more sensitive to toxicant exposure).

2.2.2.1 PCBs

PCB compounds, particularly those with a single or no chlorine at the carbon-carbon bond linking the rings of the PCB molecule, are thought to induce aryl hydrocarbon (Ah) hydroxylase activity which is believed to be responsible for the majority of toxic effects associated with PCBs (Safe 1990). Each PCB congener has a different affinity for the Ah receptor molecule and therefore different degrees of effects. Different congeners can also interact with one another (e.g., synergistically), consequently, the mixture of congeners is important in determining the ultimate effects in the environment. PCBs with larger numbers of orthosubstituted chlorine atoms result in non-Ah receptor mediated effects and some PCB metabolites have anti-estrogenic effects.

The more toxic PCB congeners (i.e., coplanar PCBs) are structurally related to dibenzo-*p*-dioxins, and are believed to have similar modes of toxic action as dioxins. The concentration of the dioxin-like PCBs can be converted into toxic equivalents (TEQs),

and compared to dibenzo-*p*-dioxin concentrations on a relative basis. Section 2.2.2.4 (Dioxins and Furans) includes a more detailed description of TEQs.

PCBs are bioaccumulated by organisms and biomagnified through the food web, with higher concentrations found in higher trophic level organisms. Different species may preferentially take up different congeners, however, in general, greater fractions of congeners with 4 to 6 chlorine atoms are found in organisms (ATSDR 2000b).

Invertebrates - PCBs have a wide variety of effects on aquatic organisms and there are significant interspecies differences in sensitivities to PCBs, even among species that are closely related taxonomically (Eisler 1986a). Most studies of the effects of PCBs on benthic invertebrates have shown adverse effects on survival, growth, and reproduction.

Fish - Effects of PCBs on fish include mortality, growth-related effects, behavior responses, biochemical alterations, and adverse reproductive effects. Of particular concern are the effects of dioxin-like PCB congeners, which have the same toxic mechanism as 2,3,7,8-tetrachlorinated dibenzo-*p*-dioxin, or 2,3,7,8-TCDD (Walker and Peterson 1991; Zabel et al. 1995). 2,3,7,8-TCDD and these dioxin-like PCB congeners cause early life stage mortality associated with blue-sac disease, which involves subcutaneous yolk sac edema (Wisk and Cooper 1990; Walker et al. 1991).

In addition, numerous field studies have reported increased mortality, pathologic anomalies, and biochemical changes in feral fish collected from ecosystems where PCBs have been detected and correlated with PCB tissue burdens (Niimi 1996). These observations include reduced hatchability and poor survival of larvae taken from feral organisms and reared in the laboratory (Mac and Schwartz 1992; Ankley et al. 1991).

Birds - A substantial amount of research demonstrates that adverse reproductive effects occur in piscivorous bird populations exposed to PCBs and dioxins in the Great Lakes Region (Jones et al. 1993, 1994; Tillitt et al. 1992; Giesy et al. 1994a,b). Piscivorous birds display a number of symptoms similar to those observed in other avian species exposed to planar halogenated hydrocarbons in the laboratory including altered biochemical homeostasis, physical deformities, fetotoxicity, and teratogenesis. In addition to embryo mortality, PCBs cause edema and beak malformations, such as crossed beaks in double-crested cormorants (Firestone 1973; Schrankel et al. 1982; Brunström and Darnerud 1983).

Mammals - Wildlife, especially mink, are particularly susceptible to adverse effects from exposure to specific PCB congeners, including the non-ortho and mono-ortho substituted PCBs (Leonards et al. 1995). Residues from PCBs can cause mortality or serious reproductive complications in mammals. Other clinical signs of PCB toxicity include anorexia, liver and kidney degeneration, and gastric ulcers, which have been observed in mink fed PCB-contaminated coho salmon (Wren 1991).

Immunotoxicological effects have been demonstrated in captive feeding studies of harbor seals (Ross et al. 1996) and while immune response has not been specifically studied in

killer whales, PCB concentrations in the blubber of killer whales found in Washington State waters exceed those shown to affect harbor seals (Ross et al. 2000).

In addition, the 12 dioxin-like PCB congeners have been shown to cause endocrine disruption in various species of aquatic biota.

Humans - In humans, PCBs can disrupt the production and disposition of thyroid hormones and impair the immune system (ATSDR 2000b). Infants and small children exposed to PCBs through breast milk have exhibited often subtle neurological impairment. However, human epidemiological studies are often confounded by the fact that individuals may also be exposed to co-occurring chlorinated dioxins and furans. Aroclor mixtures have been shown to be carcinogenic in rats and are potentially carcinogenic in humans.

2.2.2.2 PAHs

PAH compounds share common properties but are highly variable in terms of their toxicity (Eisler 1987b). For example, LPAH compounds exhibit acute toxicity to some organisms, with toxicity increasing as alkyl substitution increases (Van Luik 1984). However, they are considered non-carcinogenic. Conversely, HPAH compounds are less toxic but can be carcinogenic, mutagenic or teratogenic to a wide variety of organisms including fish, amphibians, birds, and mammals (Moore and Ramamoorthy 1984; Eisler 1987b; Environment Canada 1994b).

Although PAHs are rapidly bioaccumulated, they can also be quickly metabolized and eliminated from most organisms (e.g., forage fish and birds; Eisler 1987b). In most fish, PAHs are metabolized and excreted, and concentrations in fish tissue are generally low. Bioaccumulation is usually short term, and biodegradation and biotransformation become the more common fate processes (Clement Associates 1985), with biotransformation and excretion rates varying widely among fish species (Meador et al. 1995). Biomagnification of PAHs up the food chain is not expected because higher level organisms such as fish can eliminate PAHs and their metabolites (Environment Canada 1994b; Eisler 1987b), which are occasionally more carcinogenic than the parent PAH compound (O'Connor and Huggett 1988).

Invertebrates - Mollusks and other invertebrates do not metabolize PAHs as efficiently as fish and higher vertebrates, therefore, concentrations can accumulate in tissue, with higher lipid contents in aquatic organisms often resulting in increasing bioconcentration factors (Eisler 1987b). Effects of PAHs observed in benthic invertebrates include inhibited reproduction, delayed emergence, sediment avoidance, and mortality (Eisler 1987a; Landrum et al. 1991). In a study of PAH toxicity to the amphipod *Diporeia*, the mechanism identified as most likely responsible for observed acute toxic responses to PAHs was narcosis (Landrum et al. 1991).

Fish - PAH-contaminated sediment, such as may be found in the Puget Sound area, has been linked to adverse effects in fish, including reproductive impairment, immune dysfunction, increased incidence of liver lesions, and other histopathological endpoints (Malins et al. 1987; Johnson et al. 1988; Varanasi et al. 1992). Fin erosion and liver abnormalities have also been observed in fish exposed to extracts from PAH-contaminated sediments (Fabacher et al. 1991). Other studies report sublethal effects on the cellular immune system (reduced macrophage activities) in fish exposed to PAH-contaminated sediments that could result in increased susceptibility to disease (Weeks and Warinner 1984, 1986; Weeks et al. 1986).

Birds – Birds experimentally exposed to PAHs experienced increased liver weight and increased blood flow to the liver (Eisler 1987b). It has been suggested by some researchers that embryos are able to metabolize PAHs into more toxic intermediate products than adults do, and that avian embryos have a greater capacity to do so than the embryos and fetuses of mammals.

Mammals – The primary toxic effect of PAHs in mammals is carcinogenicity. PAHs, particularly the higher molecular weight compounds, cause genetic injury to cells through a breakdown product of the parent compound (Eisler 1987b). The target organs are dependent on the individual PAH compound, but generally PAHs tend to selectively attack proliferating cells.

Humans - There is little information available regarding human health effects from exposure to PAH. A majority of the research that is available has been related to inhalation exposure (e.g., smoke, coal tar emissions), although it is expected that consumption of PAH-contaminated fish/shellfish also poses a risk to humans, based on laboratory studies with mice (ATSDR 1995). Specific PAH compounds (i.e., benz[a]anthracene, benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, chrysene, dibenz[a,h]anthracene, and indeno[1,2,3-c,d]pyrene) have been listed as probable human carcinogens by the USEPA.

2.2.2.3 Pesticides

Organochlorine pesticides

Organochlorine pesticides include a wide variety of compounds, with varying degrees of toxicity. Because of their different chemical properties, organochlorine pesticides may also be bioaccumulated and metabolized to varying degrees (Baird 1999). The effects of DDT and chlordane on various aquatic organisms and humans are presented as examples of the effects of organochlorine pesticides in the environment.

Invertebrates - DDT is highly toxic to invertebrates and early developmental stages appear to be more susceptible to DDT that adults (EXTOXNET 1996c). Invertebrates exposed to organochlorines may suffer from mortality, immobilization and reproductive effects (Eisler 1990).

Fish – Organochlorines such as DDT are highly toxic to fish and early developmental stages appear to be more susceptible than adults (EXTOXNET 1996c). Symptoms of organochlorine exposure in fish include hyperexcitability, increased respiration rate, erratic swimming, loss of equilibrium, convulsions and rapid death (Eisler 1990).

Birds - DDE, a metabolite of DDT, interferes with the enzyme that regulates the distribution of calcium, leading to egg-shell thinning in some bird species (Baird 1999). Symptoms of exposure to other organochlorines include: sluggishness, ruffled feathers, reduced food intake and weight loss (Eisler 1990).

Mammals - In mammals, organochlorines cause abnormalities in the central nervous system, are teratogenic and reproductive toxicants, and most are potent carcinogens (Eisler 1990; Clement Associates 1985).

Humans - Human exposure can result from occupational exposure to pesticides or through the consumption of fish and aquatic invertebrates exposed to organochlorine pesticides. Organochlorine pesticides target the central nervous system, however, liver, kidney, and immune system may also be affected (ATSDR 2002a).

OP and Carbamate Pesticides

Organophosphorus and carbamate pesticides impede nervous system activity in a wide range of organisms (i.e., insects to humans) by disrupting cholinesterase activity (Baird 1999). OPs and carbamates are much more acutely toxic to humans and other mammals than OC pesticides are. The effects of chlorpyrifos on various aquatic organisms and humans are presented as an example of the effects of organophosphorus pesticides in the environment. Chlorpyrifos is a broad spectrum insecticide and has been detected in the tissues of some Puget Sound organisms.

Invertebrates - Arthropods are the most sensitive invertebrates, and mollusks the least sensitive to chlorpyrifos (Eisler 1988c). Symptoms of the inhibition of cholinesterase activity levels in the brain include sluggishness, incoordination, delayed maturation and growth and reproductive impairment. Chlorpyrifos is excreted from invertebrates.

Fish - As a result of cholinesterase inhibition, chlorpyrifos can cause decreased survival and growth in fish (EXTOXNET 1996d), however, fish tend to be less sensitive to OP-pesticides than invertebrates are (Eisler 1986b). Chlorpyrifos is rapidly excreted by fish, with equilibration with the surrounding environment occurring within 24 to 48 hours (Eisler 1988c).

Birds – Birds are relatively sensitive to chlorpyrifos with lethal doses (i.e., LD_{50}) in the range of 8-100 $mg \cdot kg^{-1}$ depending on the species (EXTOXNET 1996d). Mallards exposed to 125 $mg \cdot kg^{-1}$ chlorpyrifos laid significantly fewer eggs. Symptoms of exposure include muscular incoordination, rapid breathing, muscular weakness, fluffed feathers, prostration, spasms, coma and convulsions (Eisler 1988c).

Mammals - Symptoms of exposure to OP-pesticides in mammals are similar to those in birds and include hyperexcitability, muscular incoordination, rapid breathing, muscular weakness, tremors, prostration, spasms, coma, and convulsions (Eisler 1988c). Mammals are generally more resistant to OP-pesticides than birds are (Eisler 1986b).

Humans - Exposure of humans to chlorpyrifos is most likely to occur through inhalation or ingestion of pesticide residues, during or after application to crops (ATSDR 1997). Humans exposed to short-term, low levels of chlorpyrifos may experience dizziness, fatigue, nausea and intestinal distress, while high exposure concentrations may cause symptoms such as paralysis, seizures and loss of consciousness.

Synthetic pyrethroids

Synthetic pyrethroids have high insecticidal properties but low mammalian toxicity. They are highly fat soluble, but easily degraded and therefore do not tend to bioaccumulate (EXTOXNET 1994).

Invertebrates - Pyrethroids affect the nervous system of invertebrates, causing paralysis (EXTOXNET 1996b).

Fish - Pyrethroids are also highly toxic to fish, affecting the nervous system in the same way as in invertebrates (EXTOXNET 1996b). Lethal concentrations of permethrin are in the order of 0.002 – 0.008 $mg \cdot L^{-1}$ for several freshwater fish.

Birds - Birds are relatively insensitive to the effects of pyrethroids (EXTOXNET 1996b).

Mammals - Mammals tend to be less sensitive to pyrethroids than do fish, and are able to metabolize the compounds in this group of chemicals (ATSDR 2001). Aquatic mammals are not likely to be affected by ingesting lower trophic organisms that have been exposed to pyrethroids.

Humans - Effects of pyrethroids on humans are usually related to inhalation exposure or consumption of large quantities of the pesticide itself (e.g., residues on food), rather than consumption of contaminated fish and shellfish (ATSDR 2001). Oral exposure may result in gastrointestinal distress and nervous system impairment. Because pyrethroids are quickly metabolized, sublethal effects are often reversible.

Inorganic pesticides

See Section 2.1.2 for a discussion of the toxic effects of copper and arsenic.

2.2.2.4 Dioxins and furans

The toxic effects of dioxins and furans to fish and birds occur at low doses (Eisler 1986c). Although there is debate throughout the scientific literature, dioxins and furans are also considered to have long-term carcinogenic effects (USEPA 1994a).

The mode of action of all dioxins and furans is very similar to 2,3,7,8-TCDD, the most toxic and most extensively studied congener. For this reason, the toxicity of all dioxin and furan congeners is expressed in terms of toxicity equivalency factors (TEFs) relative to 2,3,7,8-TCDD. For example, octachlorodibenzo-*p*-dioxin has a TEF of 0.001, while 3,3',4,4',5-pentachlorobiphenyl has a TEF of 0.1 (Baird 1999). TEFs can be specific to a particular group of organisms such as mammals or fish. Multiplying the TEFs by the measured concentration of the compound then results in a toxicity equivalent concentration (or TEQ), which is used to assess the potential toxic effects of dioxins and furans, and dioxin-like PCBs, as a group, or to determine relative contribution of specific dioxin or furan or dioxin-like PCB compounds to toxicity.

Invertebrate - Invertebrates are relatively resistant to the effects of dioxin; growth and reproductive effects have not been observed at concentrations that have been reported to be lethal to fish (Eisler 1986c).

Fish - The maternal transfer of 2,3,7,8-TCDD is an important exposure route for fish and results in both reproductive and development effects in the offspring. Hemorrhages, craniofacial malformations, edema and mortality have been observed prior to and during hatching as well as the sac-fry stage of development of salmonids (Spitsbergen et al. 1991; Walker et al. 1994; Johnson et al. 1998).

Mature fish appear to be less sensitive to dioxins than young fish, however, adverse effects including fin erosion, hemorrhaging, cranial deformation, edema (swelling in connective tissue, or cavity), tumors, and histopathological damage have been observed (Cook et al. 1991). Substantial mortality to older fish is expected to occur when wet weight accumulations reach in the range of 1000-15,000 $ng \cdot kg^{-1}$ (USEPA 1993b). In general, older and larger fish die last, while smaller or younger specimens succumb first (Norris and Miller 1974).

Birds - Like some PCB congeners, dioxins and furans can cause disruptions to the endocrine system, resulting in reproductive effects such as reduced fecundity and hatchability in birds (Takatsuki 2001), as well as beak deformities and edema, deformities in developing brains of young birds, and decrease in weight gain and skeletal growth.

Mammals - Mammals exposed to dioxin may exhibit decreased body weight, liver damage, changes in blood and bone marrow, shrinking of the thymus, and death. Lethal oral doses of 2,3,7,8-TCDD for mammals varies greatly among different species (Environment Canada 1993b). Atrophy of the thymus is a consistent finding in mammals poisoned by 2,3,7,8-TCDD, as well as suppression of thymus-dependent cellular immunity, especially among young animals, may contribute to their death (Rozman et al. 1984). Developing mammalian fetuses are especially sensitive to 2,3,7,8-TCDD; the frequencies of stillbirths increase with relation to maternal exposure to 2,3,7,8-TCDD. This same relationship has been noted with regards to teratogenic effects such as cystic kidney, cleft palate, and spinal column deformities among live births (Ramel 1978).

Studies also indicate that 2,3,7,8-TCDD is an extremely potent porphyrogenic agent. Porphyria is a condition characterized by fragility of the skin, photosensitivity, and accumulation of porphyrins in the liver (Cantoni et al. 1981).

Humans - A majority of what is known or suspected regarding the effects of dioxins and furans in humans is based on laboratory studies on animals as human epidemiological studies are often confounded by the fact that individuals may also be exposed to other, co-occurring chlorinated contaminants (ATSDR 1998). Effects that have been associated with human exposure to materials contaminated with 2,3,7,8-TCDD in some studies include cancer, dermal (e.g., chloracne), hepatic and thyroid dysfunction, diabetes, and cardiovascular, respiratory, immunological, neurological, and reproductive effects.

2.2.2.5 Phthalate esters

The toxic effects of phthalate esters on aquatic organisms and fish/shellfish consumers have not been well studied. Phthalates are readily metabolized in higher organisms and not likely to bioaccumulate.

Invertebrates - Lower molecular weight phthalates in water are more acutely toxic to invertebrates than heavier phthalates (Adams et al. 1995).

Fish - Lower molecular weight phthalates in water are more acutely toxic to fish than heavier phthalates (Adams et al. 1995). Chronic exposure of fish to dibutyl phthalate (a lower molecular weight compound) has been observed to affect growth (Environment Canada 1994d).

Birds - There was insufficient information to describe the effects of phthalate esters on birds, however, there are indications that toxicity to birds is low.

Mammals - Effects in feral mammals exposed to phthalate esters via consumption of contaminated fish/shellfish have not been studied (ATSDR 2002b).

Humans - Effects in humans exposed to phthalate esters via consumption of contaminated fish/shellfish have not been studied (ATSDR 2002b).

3. SOURCES AND PATHWAYS OF TOXIC CONTAMINANTS IN THE PUGET SOUND ENVIRONMENT

Puget Sound is affected by toxics in surface water, and also from contaminants that originate from other source areas, including sediment, ground water, and atmospheric deposition. This section begins with a brief overview of how water quality in Puget Sound is regulated and monitored, and places the discussion of contaminant sources in a larger context (Section 3.1). A discussion of sources of toxic contaminants to Puget Sound and different transport pathways is provided in Section 3.2. Section 3.3 includes a summary of historical contaminant sources by region. Finally, Section 3.4 outlines current monitoring initiatives and potential data gaps.

3.1 REGULATORY OVERVIEW

3.1.1 Background – The Clean Water Act

Water quality in Washington is managed by the state, under the auspices of the federal Clean Water Act (CWA), Public Law 95-217. The CWA was enacted in 1972, and is the most comprehensive piece of legislation dealing with the environmental quality of the Nation's waters, covering both marine and freshwater systems (USEPA 1994b).

The Washington Department of Ecology (WDOE) is the principal state environmental agency that manages water quality in accordance with the federal CWA as well as other state and local laws. WDOE has implemented a watershed approach to managing water quality, for both point source and nonpoint-source water quality activities. Puget Sound is divided into several Water Resource Inventory Areas (WRIAs). There are also many individual water bodies within Puget Sound that are regulated because of toxic contaminants.

In the state of Washington, sediment quality is regulated through Sediment Management Standards under the Clean Water Act because it is recognized that contaminated sediments can have a significant effect on the overall health of water quality and, therefore, the aquatic ecosystem. The Model Toxics Control Act, promulgated in 1988, is an additional tool used by WDOE to manage sediment quality. The purposes of the Act were to clean-up contaminated sediments, improve the management of hazardous wastes, and prevent future sediment contamination by pollution prevention (WDOE 2001a). The state Sediment Management Standards were also developed under the authority of the Puget Sound Water Quality Authority Act, the Pollution Disclosure Act of 1971, the Water Resources Act of 1971, and the state Environmental Policy Act.

3.1.2 Regulation of Point Sources of Toxic Contaminants in Puget Sound

A point source of pollution is defined by regulations as any discernible, confined conveyance, but not including return flow from irrigated fields. In other words, it is a static, identifiable contaminant source, such as a waste water outfall. Section 402 of the CWA established the National Pollutant Discharge Elimination System (NPDES) and requires that any direct discharger of pollutants to the surface waters of the United States obtain an NPDES permit before the discharge can take place. The U.S. Environmental Protection Agency (USEPA) and WDOE further categorize permitted point sources as major and minor discharges. This distinction is determined during the evaluation of several aspects of the effluent (e.g., volume, contaminants present) that is described in the permit application process.

Each NPDES permit contains effluent limitations (maximum concentrations of contaminants), effluent monitoring requirements, and facility specific requirements (spill plan, pollution prevention plan, etc.). The effluent limit established for each contaminant is either technology-based or water quality-based. Technology-based limitations (known as Best Control Technology [BCT] or Best Available Technology [BAT] limitations) are established by an engineering analysis and an economic test. If a technology-based limitation results in effluent concentrations which exceed the water quality standards a mixing zone may be authorized. If the effluent concentration then has a reasonable potential to violate the water standards past the boundary of a mixing zone, a more stringent water quality-based effluent limit is placed in the permit such that the water quality standards will be met at the boundary of the mixing zone.

Each permit issued under the NPDES system contains site-specific requirements (e.g., monitoring programs) and maximum concentrations for contaminants in the effluent. The specific allowable concentrations may be established based on best available treatment technologies (i.e., all known, available and reasonable methods of prevention, control and treatment [AKART]). Alternatively, water quality-based limits may be set, in which case, effluent quality must be such that it protects the designated beneficial uses of the given receiving waterbody, either at the point of discharge or at the edge of an initial dilution zone (i.e. the area of initial mixing of the effluent into the receiving environment) around the outfall. Essentially, the standards regulating point sources are quite different, giving environmental managers multiple options. The AKART and effluent-based standards apply to the quality of the effluent prior to discharge, while the receiving water standards apply to a total allowable contaminant concentration for a given water body.

Section 403 of the Clean Water Act sets criteria to prevent unreasonable degradation of the marine environment. These requirements include and authorize imposition of any additional effluent limitations, including zero discharge, necessary to protect the receiving waters to attain the objectives of the CWA. Therefore, direct discharge to Puget Sound is regulated under Sections 403 (Ocean discharge criteria) and 404 (permits for dredged or fill material).

Dredging and dredged material disposal is monitored under the auspices of the Puget Sound Dredged Disposal Analysis (PSDDA) program. The PSDDA partners include: the U.S. Army Corps of Engineers, the U.S. Environmental Protection Agency, and Washington State Departments of Ecology and Natural Resources (WDNR). Furthermore, although three of the four agencies (i.e., the Corps, US EPA, WDOE) have independent regulatory authority under the CWA to manage water and sediment quality, and WDNR has final say on the use of the state aquatic lands, all four agencies share responsibility for expenses, studies, review of projects, management of the program and communication with the public.

In 1991, Washington State adopted the majority of PSDDA's procedures in regulatory form as the Sediment Management Standards (SMS), under the State's water quality and waste clean-up regulations. EPA Region 10 approved the SMS in accordance with the Federal Clean Water Act.

3.1.3 Regulation of Nonpoint Sources of Toxic Contaminants in Puget Sound

Non-point source (NPS) pollution occurs when contaminants are discharged over a wide area, or from a number of small inputs, rather than from a single identifiable source (e.g., large scale acid rain; watershed erosion in areas of logging). NPS is now the leading cause of water pollution in Washington. Because of the diffuse nature of NPS, control is usually exerted through implementation of best management practices (BMPs). Current state activities are focused on reducing fecal coliform counts and limiting pesticides in discharges to aquatic receiving environments.

In order to approach the problem of quantifying nonpoint source water quality issues, the EPA has developed a Total Maximum Daily Load (TMDL) rule, which was published in 2000 under the authority of Section 303 of the Clean Water Act (USEPA 2000). A TMDL is a calculation of the maximum amount of a pollutant that a waterbody can receive (point and nonpoint), and still meet water quality standards. The EPA generally does not oversee implementation or enforcement of TMDLs; rather, water quality standards are set by the state. The TMDL calculation must include a margin of safety to ensure that the waterbody can be used for the purposes the State has designated. The calculation must also account for seasonal variation in water quality.

In compliance with Section 303(d) of the CWA, WDOE identifies waterbodies that fail to meet water quality standards (i.e., for water, sediment, or tissue) designed to protect beneficial uses including fishing, swimming, boating, to industrial and agricultural purposes, and fish habitat. The *Final 1998 Section 303(d) List* of waterbodies impaired because of toxic contaminants in sediment or tissues included many bays in Puget Sound, such as Bellingham Bay, Everett Harbor, Duwamish Waterway, Elliott Bay, Commencement Bay, Budd Inlet, Dyes Inlet, Hood Canal (North), and other localized

areas (Table 3-1). Further region-specific information is presented in Section 3.3 of this report. WDOE is currently drafting the 2002 303(d) list (WDOE 2002).

Following mention on the 303(d) list, WDOE prepares water cleanup plans, including TMDLs, to improve the health of the waterbody in consultation with local interests. WDOE will then propose and establish conditions in discharge permits and nonpoint-source management plans to reduce pollution, and a monitoring plan to evaluate the effectiveness of the cleanup plan. Currently the only TMDL that has been done for toxics in marine sediments is the Bellingham Bay Pilot Study (WDOE 2001b).

Three laws at the state level work in concert with federal legislation to manage nonpoint source pollution:

- The State Environmental Policy Act offers the most powerful legal protection of receiving environments through its EIS (environmental impact statement) and impact mitigation requirements.
- The Shoreline Management Act is used to regulate the development of the state's wetlands, riparian, and estuary/embayment habitats.
- The Growth Management Act was promulgated to curb the environmental effects of urban sprawl, and requires the protection of critical habitat areas and resource lands.

3.1.4 Washington Department of Ecology PBT Strategy

In December 2000, the Washington Department of Ecology released the report *Proposed Strategy to Continually Reduce Persistent, Bioaccumulative Toxins (PBTs) in Washington State* (WDOE 2000), which was drafted at the direction of the Washington State Legislature. During the 2000 legislative session, lawmakers adopted legislation directing Ecology to develop for review by the legislature "…a proposed long-term strategy to address persistent, bioaccumulative, and toxic chemicals in the environment." In the fall of 2000, WDOE conducted an extensive public consultation on a draft PBT strategy. Public meetings were held in Bellingham, Spokane, Kennewick, Vancouver and Seattle, and received extensive comments from public interest organizations, business and agricultural interests, other government agencies, and from individual citizens. After the consultation process, WDOE submitted a proposed PBT strategy (which can be obtained at http://www.ecy.wa.gov/pubs/0003054.pdf) to the House Agriculture & Ecology Committee and the Senate Environment, Energy, and Water Resources Committee for their consideration during the 2001 legislative session.

Table 3-1: Summary of Puget Sound waterbodies included on the *1998 303(d) List of Impaired and Threatened Waterbodies* and the parameters for which they were listed.

WATER BODY	PARAMETER[a]	
	SEDIMENT	TISSUE
North Puget Sound		
Bellingham Bay	Arsenic, copper, lead, mercury, PCB[b], LPAH[c], HPAH[d], phthalate[e], dibenzofuran	
Strait of Georgia	Cadmium, PCB, LPAH, HPAH	
Padilla Bay, Fidalgo Bay and Guemes Channel		PCB
Port Gardner and Everett Harbor	Mercury, PCB, LPAH, HPAH	
Possession Sound	Arsenic, cadmium, copper, lead, mercury, LPAH, HPAH, phthalate, dibenzofuran	
Central Puget Sound		
Central Puget Sound	Mercury, PCB, LPAH, HPAH, dibenzofuran	
Duwamish Waterway	Arsenic, cadmium, copper, lead, mercury, PCB, LPAH, HPAH, phthalate	PCB
Elliott Bay	Arsenic, cadmium, copper, lead, mercury, PCB, LPAH, HPAH, phthalate, dibenzofurans	
Dyes Inlet and Port Washington Narrows	Cadmium, mercury, phthalate	Arsenic, mercury, HPAH, phthalate
Eagle Harbor	Mercury, HPAH	Arsenic, HPAH, PCB
Port Gamble Bay		Dieldrin
Port Orchard, Agate Passage and Rich Passage		Arsenic
Sinclair Inlet	Arsenic, cadmium, copper, lead, mercury, LPAH, HPAH, phthalate	Arsenic, PCB, aldrin, dieldrin,
South Puget Sound		
Commencement Bay	Arsenic, cadmium, copper, lead, mercury, PCB, LPAH, phthalate, dibenzofurans	Dieldrin
Thea Foss Waterway		PCB
Budd Inlet	Copper, mercury, PCB, LPAH, HPAH, phthalate, dibenzofurans	PCB, HPAH
Hood Canal		
Hood Canal North	Copper, lead, mercury, LPAH, HPAH, phthalate, dibenzofurans	

Source: WDOE (2002)

[a] only the parameters described in Section 2 are included here; other parameters may also have contributed to the 303(d) listing

[b] PCB includes total or individual Aroclors

[c] LPAH includes one or more compound

[d] HPAH includes one or more compound

[e] phthalate includes one or more compound

The PBT Strategy envisions the continual reduction of risks to human health and Washington's environment from exposures to PBTs, and focuses on the following goals:

- Reduce and phase-out existing sources of PBT chemicals;
- Clean up PBT chemicals from historical sources;
- Prevent new sources of PBT chemicals;
- Build partnerships to promote efforts to reduce and eliminate PBT chemicals;
- Improve regulatory and non-regulatory approaches;
- Identify and prioritize additional PBT chemicals;
- Improve public awareness and understanding of PBT problems and solutions; and
- Promote the development of information needed to make informed decisions on measures to reduce PBT chemicals.

The PBT Strategy initially identified nine PBT chemicals (Table 3-2) that are known to be present in Washington's environment. In addition, the strategy included a process for screening these and some 56 additional chemicals and prioritizing them for future reduction actions. Chemical-specific action plans, to be developed by WDOE in collaboration with other agencies for specific "high-priority chemicals," will be the primary means by which specific reduction actions and activities will be developed and implemented.

Table 3-2: Summary of PBT chemicals initially identified by the PBT Strategy

STARTER LIST OF PBTS
Aldrin/dieldrin
Benzo[a]pyrene
Chlordane
DDT (and DDD/DDE)
Dioxins and furans
Hexachlorobenzene
Mercury
PCBs
Toxaphene

Source: WDOE (2000)

WDOE completed the following actions during the1999-2001 Biennium:

- Used the Environmental Protection Agency's (EPA) Waste Minimization Prioritization Tool (WMPT) defining characteristics to add PBT chemicals. A list of 65 chemicals that Ecology plans to screen was identified in Appendix E of the PBT Strategy.
- Coordinated among WDOE programs to ensure increased collaboration on cross-media effects.

- Worked with EPA, other state and local agencies, and interested parties on continued development and implementation of both this state-specific strategy and national PBT- related initiatives.
- Continued to pursue grant opportunities from EPA.
- Coordinated existing indicators to track PBT-reduction successes and failures.

During the 2001-2003 Biennium, WDOE completed the following actions:

- Jointly, with the Washington Department of Health, Ecology developed a chemical action plan for mercury. The Ecology/Health Mercury Chemical Action plan was released in February 2003 (http://www.ecy.wa.gov/pubs/0303001.pdf).
- Developed and implemented a *PBT Baseline Monitoring Program* for long-term monitoring.
- Developed and released for public review a draft PBT "Working List", which expanded the original PBT list from 9 PBTS to 22 PBTs (Table 3-3).

Table 3-3: Draft PBT working list (list of 22 PBTs by category)

CATEGORY	SUBSTANCE
Metals [1]	Cadmium, Lead, Mercury
Pesticides	Dicofol, Endosulfan, Lindane, Methoxychlor, Pendimethalin, Pentachlorobenzene/pentachloronitrobenzene, Trifluralin, 1,2,4,5-tetrachlorobenzene
Banned Pesticides	Aldrin/dieldrin, Chlordane, DDT/DDD/DDE, Heptachlor epoxide, Toxaphene
Organic Chemicals	Pentabromodiphenyl ether, Hexachlorobenzene, Hexachlorobutadiene
Combustion By-products	Dioxins and furans, PAHs
Banned Organic Chemicals	PCBs

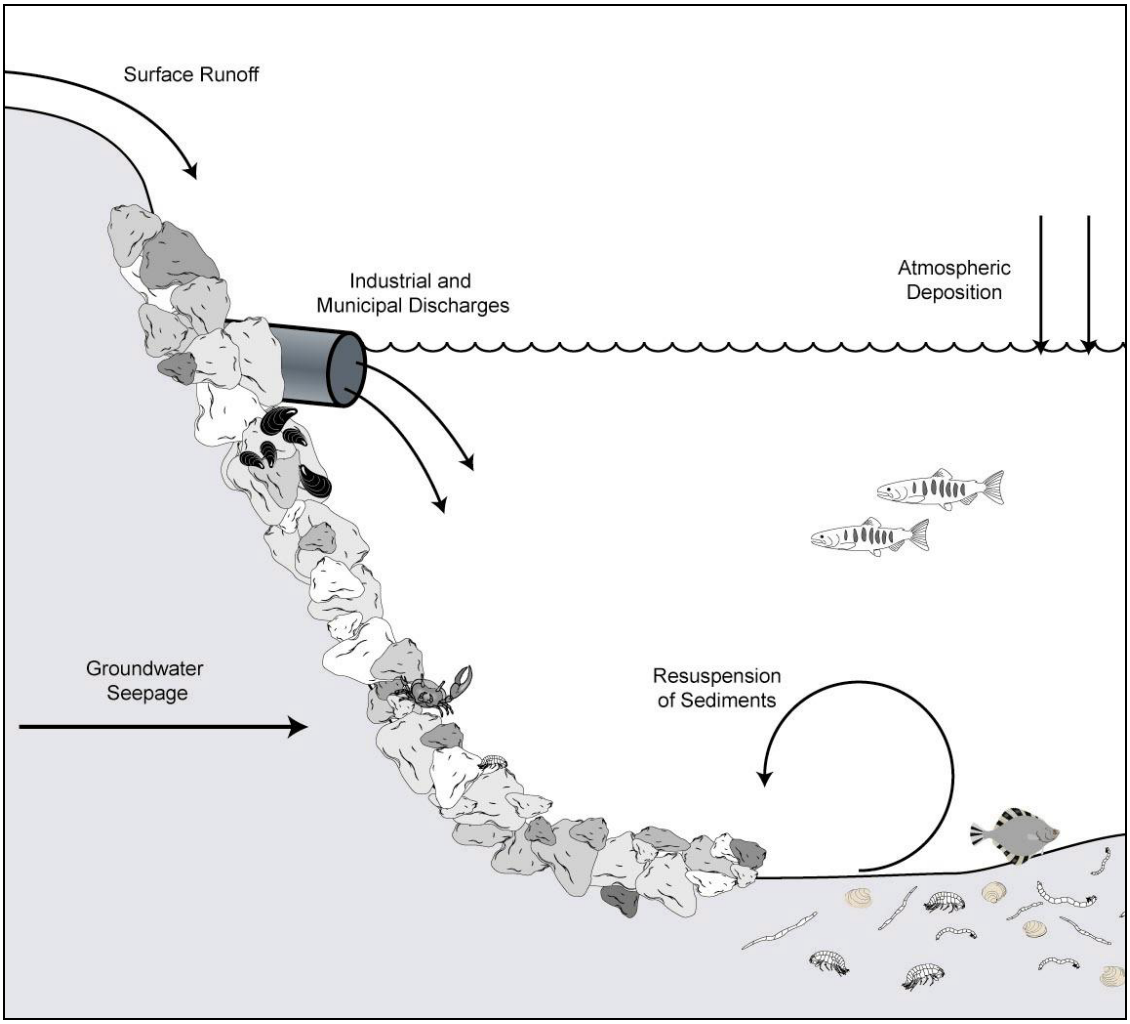
[1] EPA is currently working with its Science Advisory Board to develop comprehensive cross-agency guidance for assessing the hazards and risks of metals. Until this issue posed to EPA's Science Advisory Board is addressed, Ecology is including this footnote on the Draft PBT Working List identifying that any metals on the working list are currently undergoing this review and that Ecology will revise any PBT Working List so as to be consistent with EPA's waste minimization treatment of metals.

Currently, during the 2003-2005 Biennium, Ecology is implementing the Mercury Chemical Action Plan.

3.2 CONTAMINANT SOURCES AND PATHWAYS TO THE ENVIRONMENT

The contaminants discussed in this document reach the aquatic environment through a variety of sources and human activities (Tables 2-2 and 2-3). In the Puget Sound area, industrial and municipal discharges, groundwater seepage, atmospheric deposition, and resuspension of sediments can result in the exposure of marine organisms in the receiving environment to both metals and organic contaminants (Figure 3-1). The contaminants from some sources (i.e., operationally-contaminated industrial sites) may reach the aquatic environment by one or more of these pathways, depending on the nature of the contaminant.

Figure 3-1: Simplified conceptual model of contaminant sources and pathways to the Puget Sound aquatic environment



3.2.1 Municipal and Industrial Wastewater Discharges

The largest wastewater contributions to Puget Sound come from various industrial facilities and municipal sewer systems (TetraTech 1988b). Typical industries located near Puget Sound include: bleach plants, copper smelters, coal handling sites, chrome and silver plating industries, docks, dry cleaners, metal industries, wood treatment facilities, organic chemical manufacturing plants, oil refineries, pulp mills, petroleum distributors, paint and ink industries, photography industries, plastics, roofing, rubber manufacturers, ship building and repair, and scrap yards (PSEP 1991 and references therein).

Industrial and municipal wastewater outfalls are considered point sources and effluent discharges from them are regulated by a NPDES permit, as discussed in Section 3.1. Figure 3-2 shows the locations of active permitted discharges (as of 2002) from municipal waste water systems; petroleum refineries and bulk transfer stations; wood preserving facilities; ship building and repair facilities; foundries and smelting operations; and pulp and paper mills.

Municipal wastes may be conveyed in separate pipes than stormwater (i.e., double sewer systems), or they may be conveyed with stormwater in a single, or combined system. Hydraulic overloads and other wastewater treatment plant (WWTP) malfunctions can lead to the discharge of untreated wastewater into Puget Sound. The combined sewer systems occasionally experience overloading during storm events. To prevent backups in the system, planned overflow points called combined sewer overflows (CSOs) were added. These CSOs are mostly considered non-permitted point sources of pollution to Puget Sound (Jones & Stokes Associates and Tetra Tech 1983).

Both metal and organic contaminants are commonly measured in WWTP effluent. Frequently detected contaminants in Puget Sound outfalls include several metals (Cu, Cd, Zn, As, Cr, Ni, Pb) and PAHs. Hydrocarbons are widespread in urban settings and are frequently detected in both stormwater and CSO samples, typically reported as oil and grease and/or total petroleum hydrocarbons (TPH). The most toxic components of TPH are thought to be PAHs, which were measured more frequently than oil and grease or TPH. PAHs, mostly phenanthrene and fluoranthene, are organic pollutants of concern in both stormwater and CSOs. Phthalates are a concern in both stormwater and CSOs, and are of particular concern in Elliott Bay.

The types of contaminants that may be expected in discharges from an industrial site depend on the type of operation it is; for example, wood preserving facilities using creosote may be sources of PAHs, while shipyards may contribute metals from antifouling paints such as TBT (Table 3-4).

Figure 3-2: Locations of active permitted municipal and industrial discharges to Puget Sound (as of 2002)

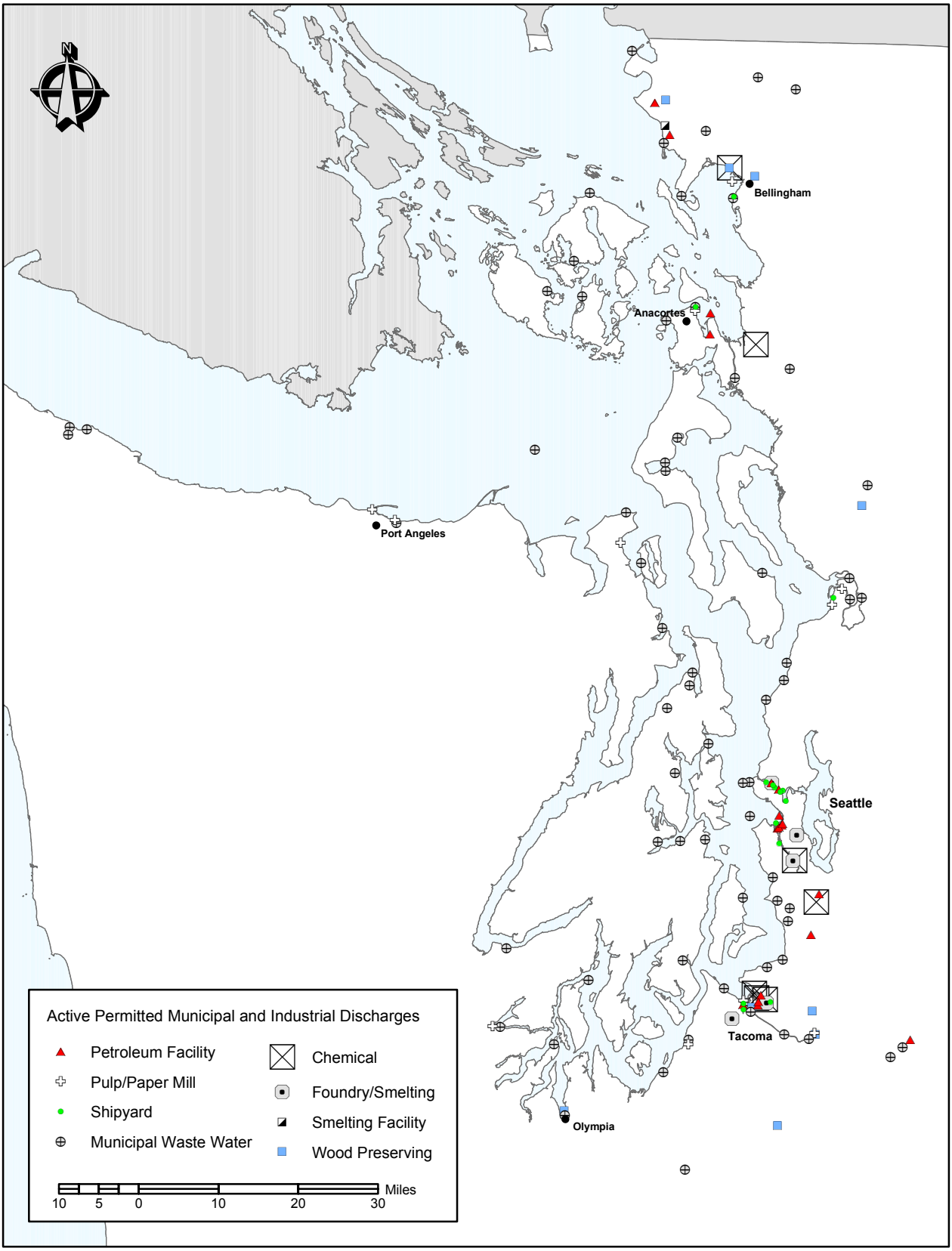


Table 3-4: Summary of municipal and industrial sources of contaminants in Puget Sound.

FACILITY TYPE	POTENTIAL CONTAMINANTS
Wood preserving	PAH, arsenic, copper, chromium
Ship building and repair	TBT, cadmium, copper
Petroleum refinery/transfer station	PAH
Foundry/smelting	metals
Pulp/paper mill (processes involving chlorine)	dioxins and furans
Wastewater treatment plants	metals, PAHs, phthalates

3.2.2 Industrial Waste (Contaminated) Sites

Puget Sound has a long history of intensive industrial activities, which have left a legacy of contaminated sites along the shore, particularly in the urban centers of Seattle and Tacoma. The groundwater underneath and/or sediments adjacent to such sites may still be a source of contaminants to marine biota today.

The primary industries that caused historic contamination in the Sound include aluminum smelters, oil refineries, and pulp and paper mills. Superfund sites along the shores of Puget Sound include Commencement Bay (Tacoma), several naval facilities (e.g., Puget Sound Naval Shipyard and others), the Duwamish Waterway and Harbor Island in Elliott Bay and Eagle Harbor in Central Puget Sound, many of which have been cleaned up or are in the process of being cleaned up. The locations of Puget Sound Superfund sites are shown in Figure 3-3. Washington State's Toxic Cleanup program has also identified numerous sites in and around Puget Sound with contaminated groundwater, soil, surface water and sediments (Figure 3-4). Figure 3-5 illustrates the abundance of various contaminant classes at these suspected and confirmed contaminated sites. Where there is a functional pathway, these sites have the potential to contribute contaminants to Puget Sound's marine environment. For region-specific information on contaminated sites in Puget Sound, see Section 3.3.

Figure 3-3: Locations of Superfund sites in Puget Sound (as of 2002).

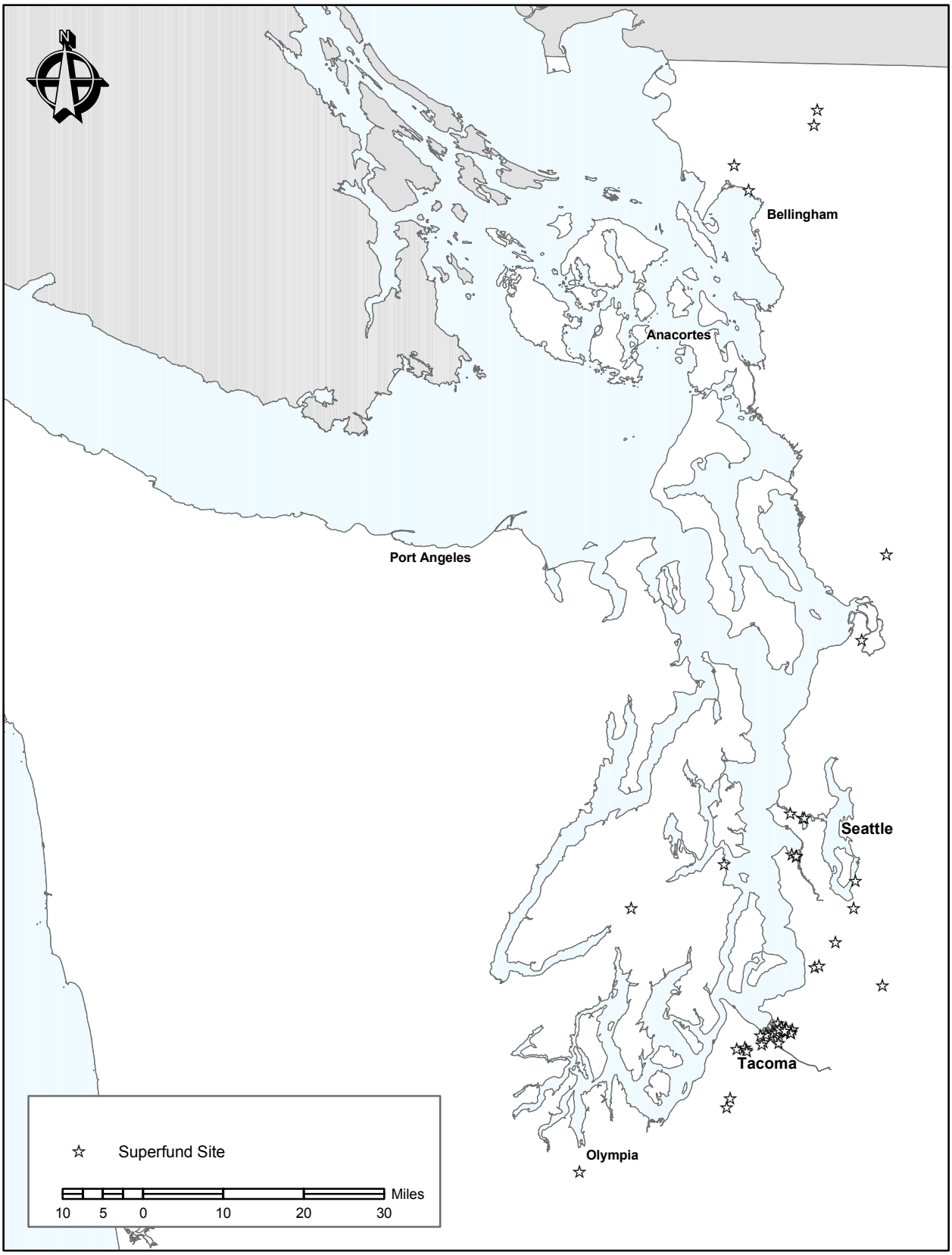


Figure 3-4: Locations of Toxics Cleanup (MTCA) sites in Puget Sound (as of 2002).

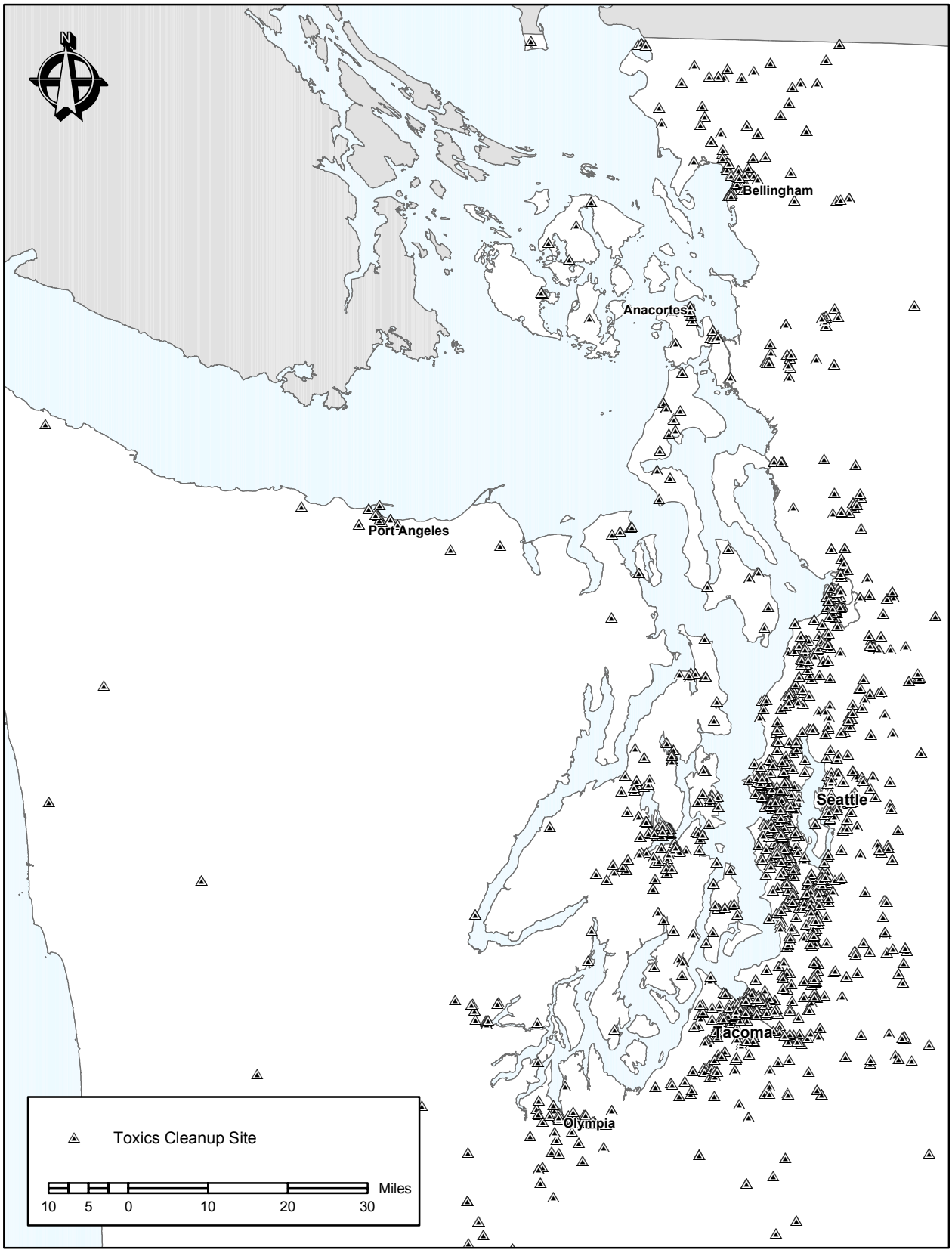
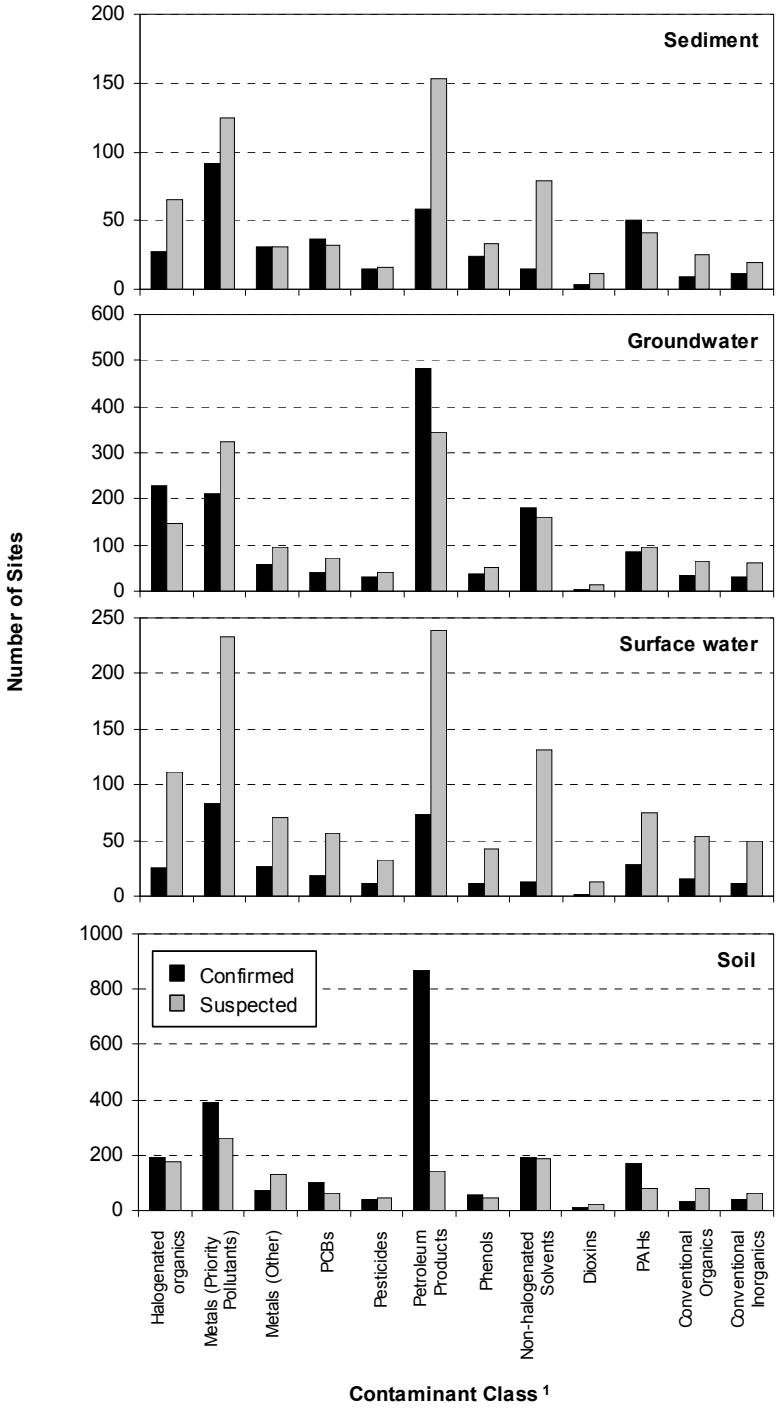


Figure 3-5: Selected contaminant classes present at both confirmed (black columns) and suspected (gray columns) MTCA contaminated sites in counties adjacent to Puget Sound (as of 2002).



Notes:
[1] Sites may be contaminated with more than one chemical class.
Source: WDOE 2002

3.2.3 Surface Runoff

In the Puget Sound area, stormwater runoff may be discharged into local streams or directly into Puget Sound via separate stormwater collection systems, or it may be conveyed to WWTPs via a combined sewer system. Storm sewer outfalls are considered to be point source discharges to the aquatic environment. Toxic contaminants typically found in urban stormwater include metals, PAHs and other hydrocarbons, phthalate esters, PCBs, and pesticides (Table 3-5).

Table 3-5: Potential sources of toxic contaminants typically found in urban stormwater.

CONTAMINANT	POTENTIAL SOURCES	
Metals (dissolved, particulate)	Motor vehicle operation Copper water pipes Galvanized culverts Landfill leachate Pigments in paints	Road salt Industrial discharges Atmospheric deposition Illicit dumping Poor waste disposal practices
PAH and other hydrocarbons	Motor vehicle operation Burning of fossil fuels, fuel spills Leaking underground fuel tanks	Creosoted structures Asphalt particles Natural sources, combustion
Phthalate esters	Leaching of plastic products	
PCBs	Past applications of PCB-laden adhesives and caulking compounds	Stockpiled waste PCB Transformer leakage
Pesticides	Pest control (golf courses, cemeteries, lawns, gardens) Illicit dumping	

Source: BC Research Corporation (1992)

Surface drainage, including agricultural runoff, is a NPS pollution discharge to Puget Sound, and is difficult to quantify. Although pollutant contributions of direct surface runoff are probably negligible relative to that carried by storm drain systems, there may be cases (e.g., hazardous materials landfills, old manufacturing plants) that have significant localized effects.

The relative loading volume of contaminants between surface runoff and point discharges such as WWTP outfalls is highly dependent on the land use of the drainage area. In urban areas, modification of the land surface (particularly the area of impervious surfaces) results in pervasive change to runoff water quantity and quality (Hill et al. 2000). Accurate land use/land cover data are critical for calculating contaminant loadings in surface water runoff and land cover is a primary input parameter for numerical hydrologic models such as the Hydrologic Simulation Program Fortran (HSPF), widely used by the surface-water management agencies of King County, Snohomish County, the cities of Seattle and Bellevue. There is little consistency, however, in spatial modeling techniques, or collection and processing methods of land-cover data, resulting in a wide

range of nonpoint source loading calculations. In addition, most Puget Sound modeling to date has focused on conventional biological water quality parameters such as nutrients or bacteria, rather than organic contaminants or heavy metals. Nener and Wernick (1997) compared point source loading (calculated from waste discharge permit information) to estimated loadings from urban runoff in the Fraser River delta (i.e., Greater Vancouver) area of British Columbia. They found that in the worst case scenarios, estimated surface runoff loadings of contaminants such as lead and copper were higher than from permitted discharges.

There are numerous freshwater tributaries to Puget Sound. Although point and non-point sources may contribute to the contaminants carried by these rivers and streams, they are themselves considered point sources. WDOE has collected water quality data on some of these rivers since 1977, including hydrological data, conventional water quality parameters, metals and pesticides (Jones & Stokes Associates and Tetra Tech 1983).

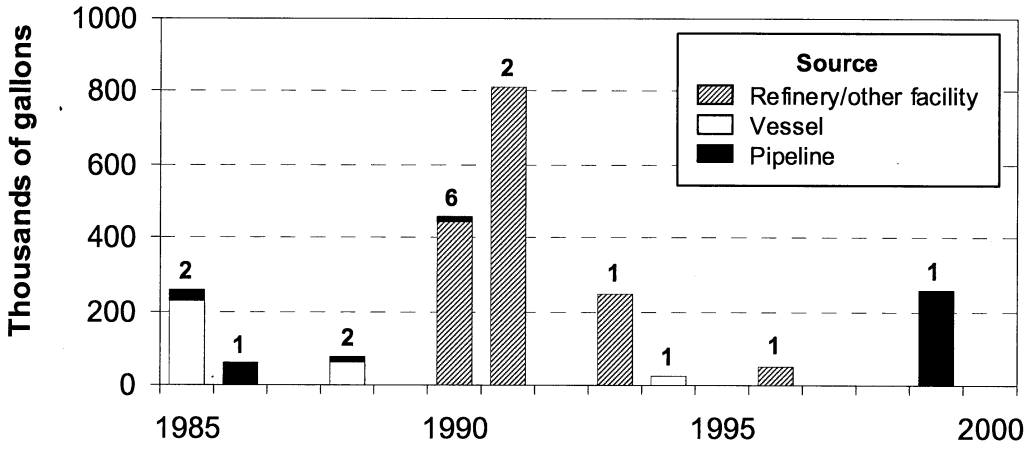
3.2.4 Ground Water Discharges

Transport of contaminants into Puget Sound via groundwater is considered a non-point source of pollutants. Leaching of contaminants into groundwater may occur as a result of improper waste disposal practices and accidental spills. Data on the quantity and type of contaminants discharged into the Puget Sound via groundwater is limited. Some potential sources of groundwater contamination include pesticides in agricultural and urban areas, and synthetic organic chemicals in urban and industrial areas (Brown and Caldwell, Inc. 1985). Figure 3-5 illustrates the number of confirmed and suspected contaminated groundwater sites in counties adjacent to Puget Sound and the types of chemical of concern.

3.2.5 Oil Spills

The risk of oil spills to the Sound is related to significant refining activities, tanker ship loading and unloading, transportation of oil via land-based pipelines. About 15 billion gallons of crude oil and refined petroleum products are moved through the Sound on an annual basis (PSWQAT 2002a). Furthermore, significant commercial cargo traffic travels through Puget Sound to and from port facilities. Figure 3-6 shows the total volume and source of “major” (i.e., > 10,000 gal.) oil spills in the Puget Sound Basin between 1985 and 2000. While some of the spilled material was contained, a large volume reached both the terrestrial and marine environments of the Basin (PSWQAT 2002a). Between 1993 and 2001, 191 oil spills of 25 to 10,000 gallons in volume (classified as “serious” spills) were recorded, from which a total volume of 73,400 gallons reached the marine waters of the Sound.

Figure 3-6: Total volume and source of major (i.e., >10,000 gal.) oil spills in the Puget Sound Basin between 1985 and 2000. The number of spills is indicated by the number at the top of the column.



Source: PSWQAT (2002a)

3.2.6 Dredging

Dredging can result in the resuspension and redistribution of contaminants that had been buried by sediment deposition (Jones & Stokes Associates and Tetra Tech 1983). A study along Seattle's waterfront has shown that sediments in the water column and depositing onto remediated sites as a result of vessel traffic and construction activities had contaminant concentrations in excess of the Washington Sediment Management Standards (Michelsen et al. 1998). Dredging and filling are considered to be a non-point source of contaminants to Puget Sound.

The PSDDA program conducts testing on samples representative of over 10 million cubic yards of sediment and manages the disposition of an average of 8 million cubic yards of dredged sediments per year. Under the PSDDA program, if bioassay tests indicate that the dredged material is more toxic than the surrounding sediments, the dredged material must be removed from the site. This ensures that the dredge site will have the same, if not better, water and sediment quality conditions found prior to dredging.

Every year extensive physical and chemical monitoring is conducted by the Army Corps of Engineers (the Corps) and the Department of Natural Resources (DNR), respectively, and the data entered by WDOE into a multi-task relational database called SEDQUAL. Comparison of historical data and new data is conducted to determine trends. Monitoring results at the end of each dredging year cycle have shown continued improvement in the quality of the sediments and waters in the disposal areas.

PSDDA's tiered testing approach allows results from bioassays, bioaccumulation and benthic studies to be used to make the determination whether the sediments are suitable

for dredging and disposal in Puget Sound. Sediments that fail the tests can still be dredged, but must be placed in confined disposal sites, usually upland, although near-shore aquatic sites can be proposed if appropriately managed. This approach allows the applicant to decide whether they wish to spend minimum funds and effort by exclusively testing for bulk sediment chemistry (the most inexpensive of options), and abiding by its established trigger levels, or simultaneously conducting biological toxicity testing to determine whether the sediments will or will not affect marine populations.

3.2.7 Atmospheric Deposition

Atmospheric deposition of contaminants may occur directly into Puget Sound or indirectly through surface water runoff from land into Puget Sound. Mechanisms of transport via the atmosphere can occur in three different ways; the exchange of gases at the air-water interface, deposition of particulates onto the water surface and through precipitation (Jones & Stokes Associates and Tetra Tech 1983). Atmospheric deposition has been observed to result in contaminant enrichment of the sea-surface microlayer compared to the underlying water column (Hardy et al. 1985).

Coordinated research on atmospheric source of contaminant deposition is limited and different methodologies are used to monitor the same contaminant in different environmental compartments, sometimes resulting in data that are not comparable. For example, aerosol data are measured for POM (polycyclic organic matter) while PAHs are measured in water and sediments. A standardized design for monitoring atmospheric deposition of contaminants would be necessary in order to better integrate atmospheric data into watershed models. In order to determine the significance of atmospheric deposition of toxics to aquatic systems, a focus on local conditions and compounds of interest is also needed (Wesely and Hicks 2000).

Atmospheric deposition is diffuse, covering a large geographic area, a range of ecosystems, and a large number of pollutants. While there are well-defined environmental effects and health risks related to atmospheric deposition of toxics in the Eastern U.S., few contamination problems in the West have been directly traced to atmospheric deposition. The pollutants of concern on the Pacific coast are primarily toxics (mercury, copper, zinc, and other heavy metals, dioxins, PCBs, PAHs, both current-use and banned pesticides, and several volatile or semi-volatile organics), but nitrogen may also be a problem in some areas (PSEP 1991).

There are few data showing how much atmospheric deposition, either directly to the water body or indirectly to the watershed, arrives at the coast or what affect it has on coastal ecosystems. Deposition can come from one or more of the following sources: local, regional, supraregional, or long-range transport (e.g., from Asia). Long-range transport of pollutants from Asia to North America has become the subject of significant research. There has been and likely will continue to be increases in coal and other industrial emissions from China and other developing Asian economies. Air from Asia

can be transported to the west coast of the U.S. in as little as five days (Jaffe et al. 1999). Pollutant storms are easily identifiable because the large dust particles are transported to the U.S. only during discrete meteorological events. The large particle pollutants must be entrained in high air currents in order for them to reach the coast of North America. Recently, the Photochemical Ozone Budget of Eastern North Pacific Atmosphere (PHOBEA) project (Jaffe, pers. comm. 2002) is using instantaneous measurement techniques at Checka Peak Observatory in the NW corner of the Olympic Peninsula.

The Puget Sound Water Quality Action Team (PSWQAT) conducted an EPA-funded study in 1990 to determine if atmospheric deposition was a significant source of contaminants in Puget Sound (PSEP 1991). Both metals and organic contaminants were measured in areas with a high concentration of industrial activities. Aerosol and deposition data were modeled to apportion aerosol data to sources and to estimate the spatial distribution of contaminants. The study found that the load from atmospheric deposition to the water column is low compared to direct discharges into the water, except potentially in the sea-surface microlayer. Exposure to contaminants during development may harm organisms that spend this critical time in the microlayer (Section 4.1).

Deposition of lead and zinc, though small, was important to the total mass loading of these two metals (PSEP 1991). Atmospheric deposition also has strong seasonal variation. Deposition in the summer is dominated by larger particles originating from local sources, whereas in the winter, deposition is in the form of finer particles, primarily from wood smoke.

3.3 SOURCES OF CONTAMINANTS TO PUGET SOUND BY REGION

Region-specific contaminant sources are discussed below: Northern Puget Sound (from the US border south to Everett Harbor); Central Puget Sound (including Elliott Bay); Southern Puget Sound (including areas around Tacoma and Olympia); and Hood Canal.

3.3.1 Northern Puget Sound

3.3.1.1 Everett Harbor

The Everett Harbor area is served by both combined and double sewer systems. The double sewer system was installed in the newer sections of Everett in order to limit overloading the sewers or the treatment plant by allowing stormwater runoff to be routed to surrounding streams and lakes while the sanitary sewage goes directly to the wastewater treatment plant (City of Everett 2002). The combined portions of the sewer system have CSOs that are limited to one overflow event a year. Three municipal WWTPs are located in the Everett area: Mukilteo, Marysville and Everett (Figure 3-7). The Mukilteo plant discharges into the Port Gardner area, the Marysville plant discharges

into Ebey Slough, and the Everett plant, which has a history of hydraulic overloads, discharges into the Snohomish River.

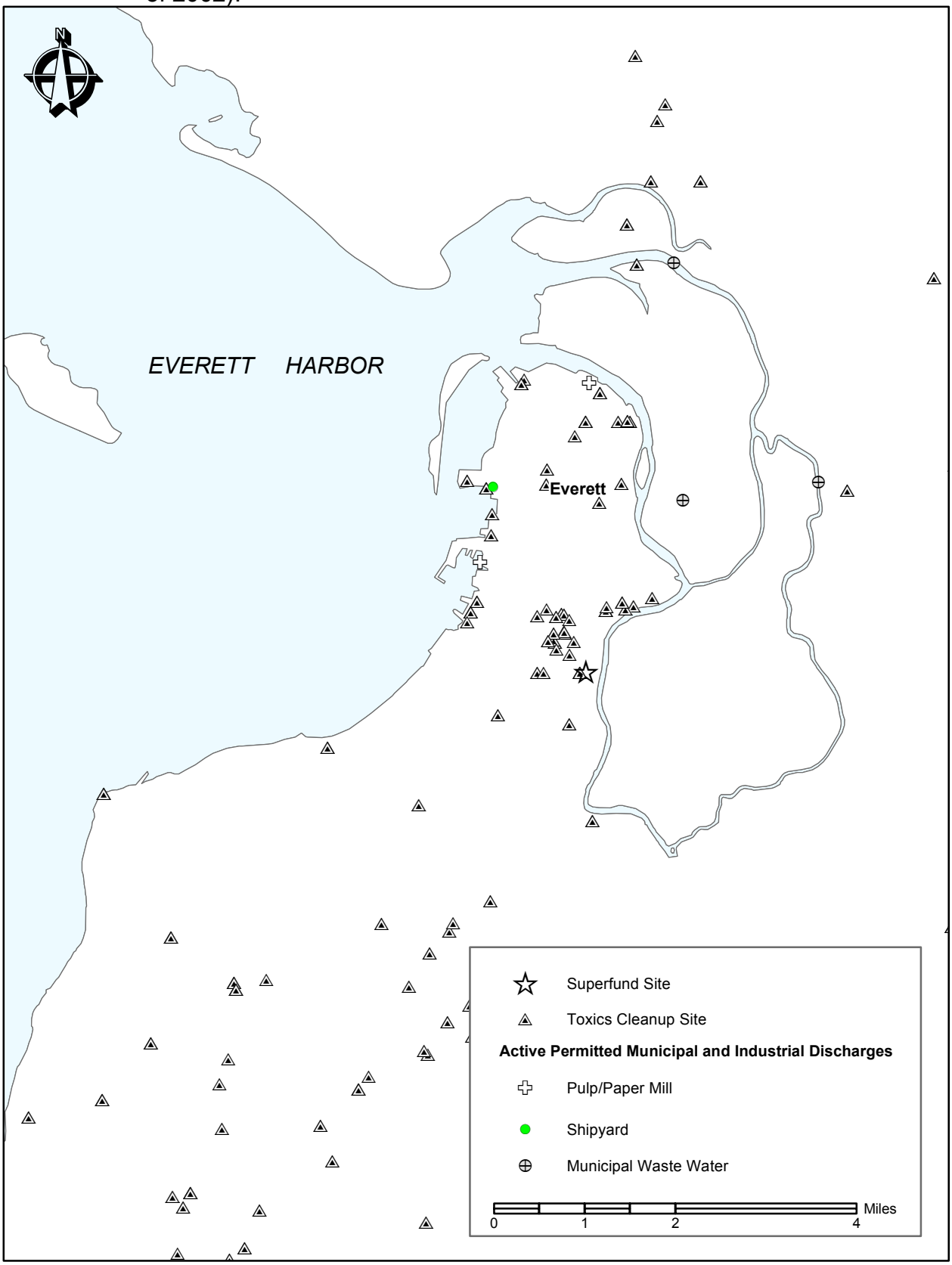
The majority of surface water runoff into Everett Harbor is via natural drainage by the Snohomish River and its sloughs. Land use in the area is mostly rural agricultural so there are few developed storm sewer networks. Some storm drains collect runoff from the more urban areas and discharge it into the Snohomish River and Everett Harbor. Runoff monitoring data is not available but the pollutant loading is thought to be insignificant (TetraTech 1988b)

Industrial discharge to Everett Harbor consists of both permitted and non-permitted direct discharges, surface runoff, and CSO overflow events. The types of industries that are permitted under NPDES to directly or indirectly discharge into Everett Harbor include pulp and paper mills, a fuel supply company, an airport, marinas, boat building and repair companies, and lumber companies. Permitted industrial facilities discharge treated process wastewater, untreated non-contact cooling water, and stormwater directly into the waterways. Facilities that are permitted to indirectly discharge are connected to the municipal treatment plants via the sewer system. Other sources of industrial contaminant migration into Everett Harbor include leachates and spills (TetraTech 1988b).

Facilities that have the potential to contribute to groundwater contamination in the Everett Harbor area include two landfills, a fuel supply depot and a Boeing test facility. The Everett Landfill is currently only operated as a solid waste transfer station. The landfill is unlined enabling the possible leaching of contaminants into surrounding groundwater. The Tulalip landfill was closed in 1979 because of concern over wetland destruction, water contamination, and complaints about odor problems (TetraTech 1988b). In September 2000 the construction of a capping system was completed for the Tulalip landfill.

Nine sediment cleanup sites under the WDOE Toxics Cleanup Program are located in Everett Harbor (Figure 3-7). The main landuse activity associated with contamination is industrial, however, military operations have also contributed to contamination. Most of the sites at which cleanup has been completed are associated with the former Weyerhaeuser Everett facility (WDOE 2001c).

Figure 3-7: Locations of Superfund and Toxics Cleanup sites and active permitted municipal and industrial discharges in the vicinity of Everett Harbor (as of 2002).



3.3.1.2 Bellingham Bay

Active permitted discharges to Bellingham Bay include municipal WWTPs, a shipyard, a pulp and paper mill, and wood preserving and chemical facilities (Figure 3-8).

Nine sites in Bellingham Bay are listed for sediment cleanup (WDOE 2001c). Much of the cleanup in Bellingham Bay is in the early stages, as indicated by the large number of sites in the initial investigation phase. Bellingham Bay is the subject of a pilot project involving local, state, and Federal agencies, as well as native bands and businesses to address bay-wide cleanup of sediment sites. A draft Environmental Impact Statement (EIS) for the Bellingham Bay Comprehensive Strategy was issued in July 1999. Most of the causes of contamination are industrial, except for a historic municipal landfill (WDOE 2001c).

As discussed earlier, the only TMDL that has been conducted for toxics in marine sediments is the Bellingham Bay Pilot Study (WDOE 2001b). Concentrations of mercury in the range of 2-5 $mg \cdot kg^{-1}$ have been measured in Bellingham Bay and are related to waste discharge from a mercury-cell chloralkali plant that operated there from the 1960's to the 1990's (Crecelius et al. 1975).

3.3.1.3 Other

Additional current or historical contaminant sources in the North Puget Sound region include contaminated sites in Padilla Bay and Port Angeles which are under investigation, and a naval site on Whidbey Island, which has been cleaned up (WDOE 2001c).

3.3.2 Central Puget Sound

3.3.2.1 Elliott Bay and King County

Contamination in Elliott Bay has resulted from a long history of maritime and industrial activity. A total of 27 contaminated sites have been identified in Seattle: 19 in Elliott Bay and 8 in the Duwamish River and Waterway (WDOE 2001c). Clusters of contaminated sites surround Harbor Island, with a few isolated sites along the western shore of Elliott Bay on the Seattle downtown waterfront. Cleanup of several sites in Elliott Bay is underway and with more in the remedial investigation and feasibility study phase. Toxic Cleanup and Superfund sites in the Elliott Bay and King County area are shown in Figure 3-9.

Figure 3-8: Locations of Superfund and Toxics Cleanup sites and active permitted municipal and industrial discharges in the vicinity of Bellingham Bay (as of 2002).

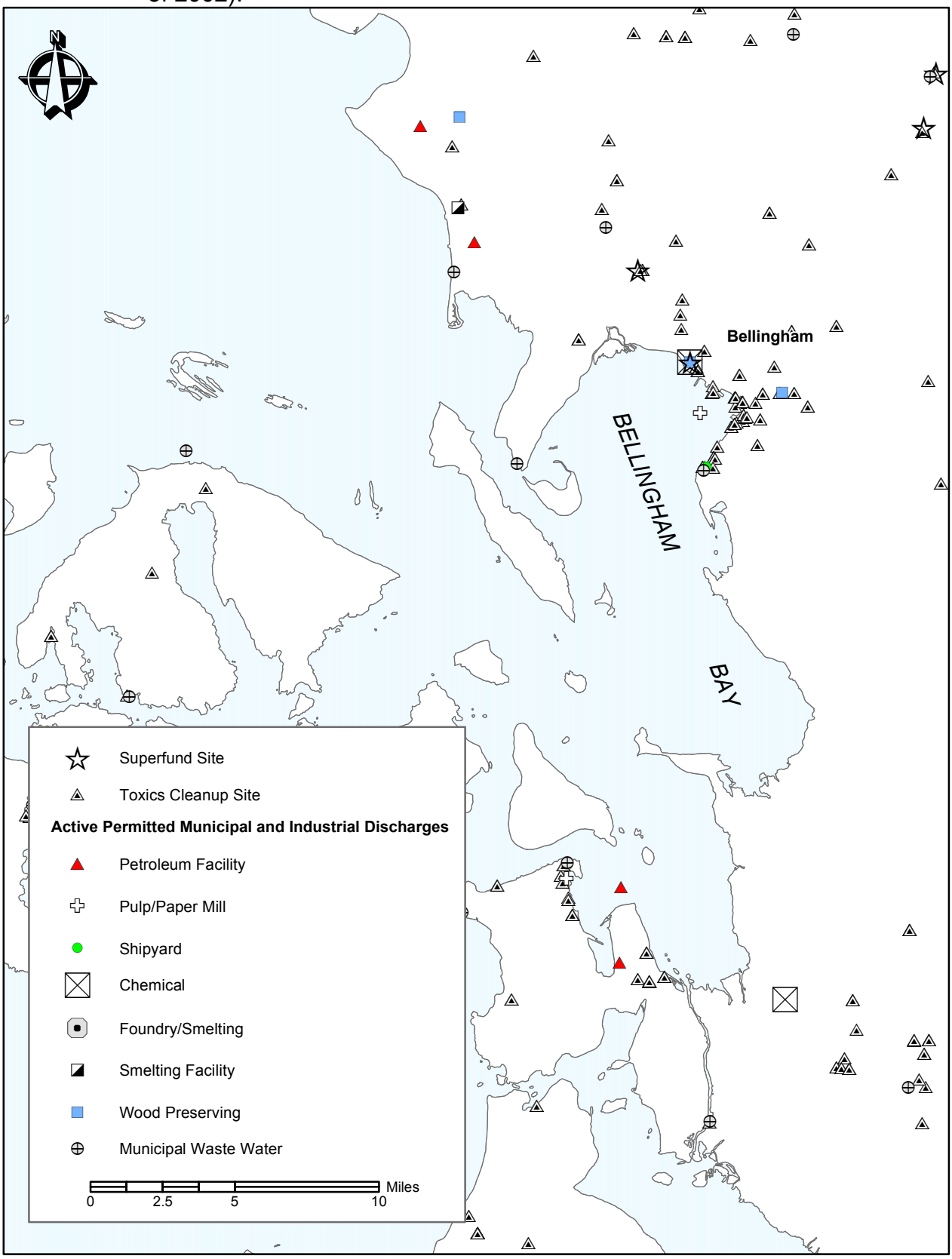
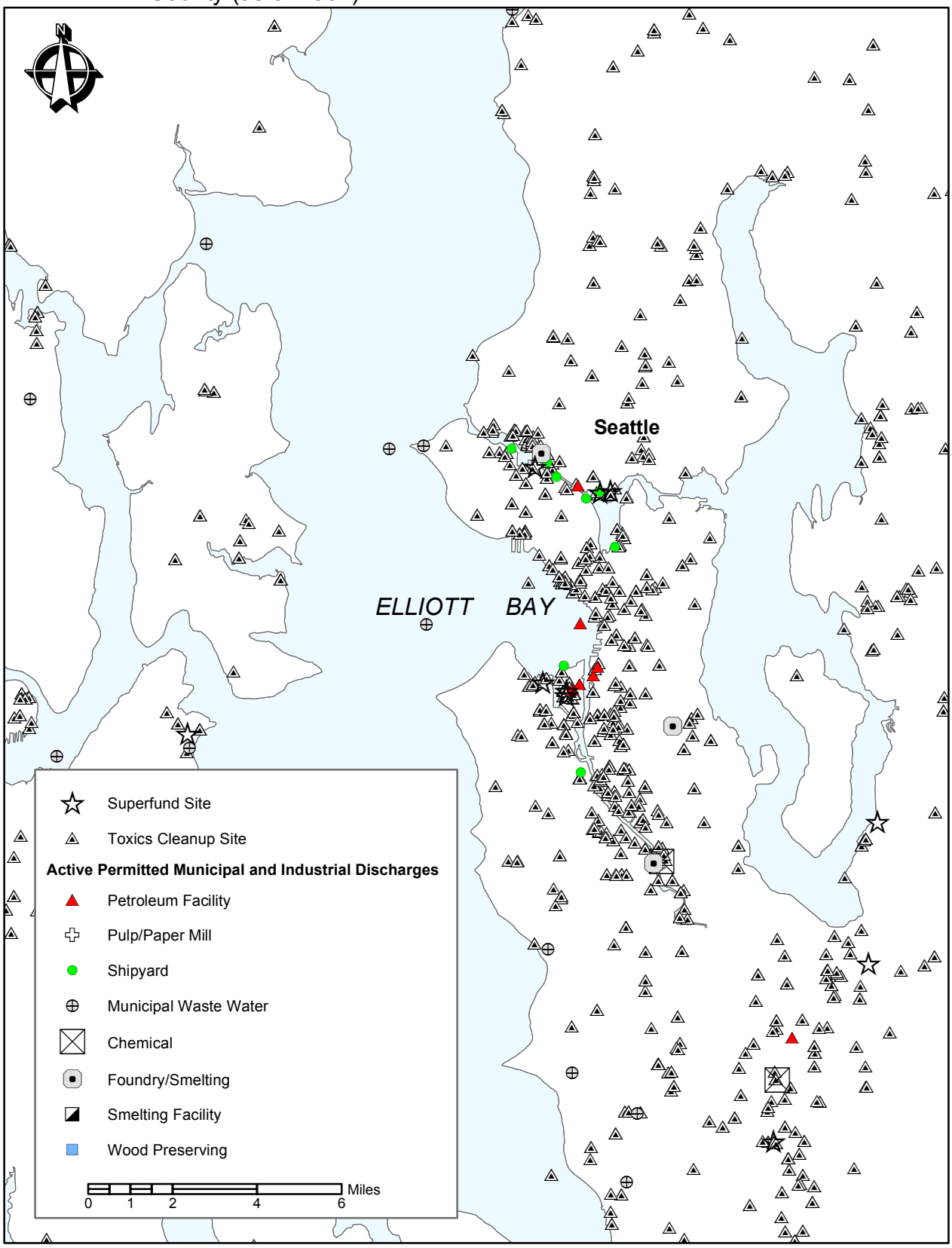


Figure 3-9: Locations of Superfund and Toxics Cleanup sites and active permitted municipal and industrial discharges in the vicinity of Elliott Bay and King County (as of 2002).



Three WWTPs (West Point, South Plant [previously Renton] and Vashon Island) and two CSO treatment plants (Alki and Carkeek) discharge treated effluent into Elliott Bay and Central Puget Sound (King County 2001). The outfalls for these treatment plants discharge at least 1,000 ft offshore (TetraTech 1988b). The City of Seattle maintains a smaller sewer collector and trunk lines and METRO (which was absorbed by the King County Department of Metropolitan Services in 1995) operates a large interceptor system that transports wastewater from the city collector system to the area treatment plants.

King County has been collecting background data in anticipation of a new regional wastewater treatment plant in southern Snohomish County that will discharge treated effluent through a new outfall. The Marine Outfall Siting Study (MOSS) was initiated in late 1998 in support of siting a suitable outfall location (King County 2001).

The most recent results from the MOSS program showed that metal concentrations at all offshore and intertidal stations were below the Washington State acute and chronic marine water quality criteria. This is consistent with results from part of a Puget Sound Interim Study from Central Puget Sound that showed effluent from the West Point sewage outfall did not greatly increase the existing concentrations of the heavy metals measured in the water column and in the biota (Schell et al. 1977). The study indicated, however, an increase in Pb and Hg concentrations measured in the intertidal organisms near West Point. Historical dumping of dredged material or sludge may be responsible for some of the increase in metals concentrations. Schell et al. (1977) estimated that less than 1.5% of the total concentration of Cu, Pb, and Zn was derived from the West Point plant at that time.

Twenty different organic compounds (i.e., PAHs, phthalates) were detected in MOSS stations. Previous work has shown evidence of surface water contamination by PAHs from municipal wastewater discharges. For example, Barrick (1982) reported an annual mass emission of 30.6 $mt \cdot yr^{-1}$ in 1978-79 for resolved AHs in the effluent from the West Point plant, which derives from sources such as street-dust accumulation in the city and undegraded fuels oils. Street runoff is the most probe source for the bulk of the PAHs. Barrick (1982) calculated a total flux of aliphatic and aromatic hydrocarbons to central Puget Sound that exceeds 1100 $mt \cdot yr^{-1}$. The discharge is sufficient to account for the build of fossil aliphatic hydrocarbons of anthropogenic origin currently accumulating in central Puget Sound sediments.

CSOs discharging into Elliott Bay include overflow from the City of Seattle combined sewer system and the King County interceptor system. The interceptor system transports wastewater to the treatment plant. King County CSOs account for 94% of total CSO discharge to the Bay (TetraTech 1988b). A CSO water quality assessment in the Duwamish River and Elliott Bay indicated that there are potential risks to benthic invertebrates, birds and people related to sediment contaminants in the vicinity of CSO outfalls (King County 1999). However, due to historic contamination and current stormwater and industrial inputs, the removal of CSO outfalls is not expected to reduce the risk of exposure to aquatic organisms, wildlife and human users of the system.

Both permitted and non-permitted industrial effluents are discharged to Elliott Bay (Figure 3-9). Industrial process wastewater is not permitted to be discharged. Both private and city storm drains empty into Elliott Bay. Private storm drains generally serve areas immediately adjacent to the waterways and serve fairly small drainage basins. The drainage basin areas served by City of Seattle storm drains range in size from 4 to 1,400 acres (TetraTech 1988a).

Contaminated groundwater is also a possible source of toxic pollutants to Elliott Bay. Specific types of sites in the area that may contribute to contaminated groundwater include abandoned landfills, active bulk petroleum facilities, inactive bulk petroleum storage facilities, and industrial sites. These sites contribute to the contamination through improper waste disposal practices and accidental contaminant spills (TetraTech 1988a).

Other causes of contamination include ship traffic such as at Harbor Island West Waterway, refueling spills, and ship discharges such as at Todd/Lockheed. CSO contamination occurs in several areas such as Harbor Island, Denny Way CSO, East waterway and Piers 46-48 and 58.

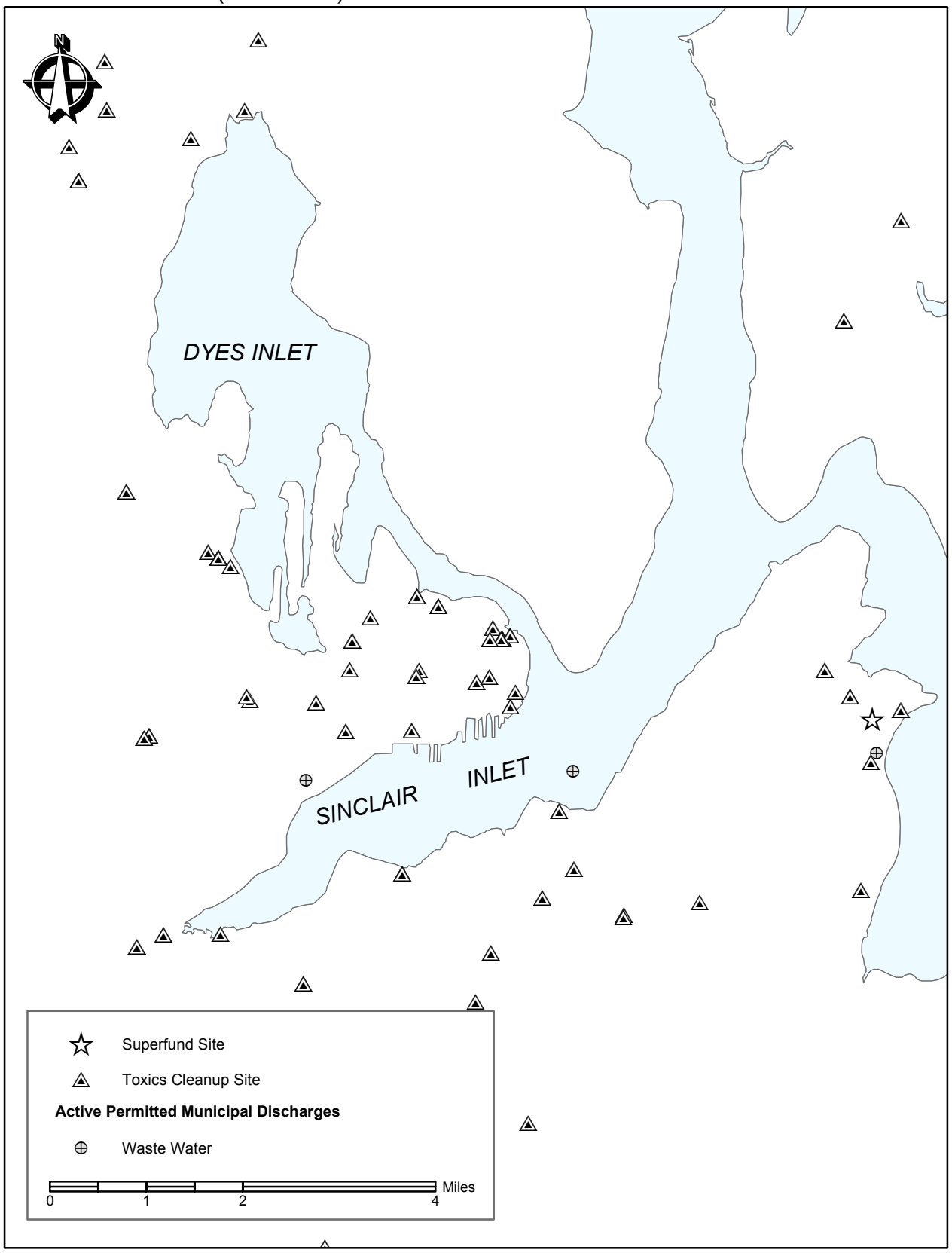
3.3.2.2 Sinclair and Dyes Inlets

Sinclair Inlet, Dyes Inlet, and Eagle Harbor and Liberty Bay (on the eastern Kitsap Peninsula near Bremerton), are considered here together. Eleven listed contaminated sites are located in this area (WDOE 2001c). Much of the contamination in Sinclair Inlet, Dyes Inlet and Liberty Bay sediments resulted from naval operations while in Eagle Harbor the contamination is related to a former wood treatment facility. Most of the Bremerton/Kitsap sites are in the latter phases of cleanup. Two sites are completed and five sites are in the design, cleanup, or monitoring phase (WDOE 2001c). Toxic Cleanup and Superfund sites in the Sinclair and Dyes Inlets area are shown in Figure 3-10.

The two current discharges of secondary treated effluent into Sinclair Inlet are from the City of Bremerton WWTP, and the Port Orchard/Kitsap County Sewer District. There are no active WWTPs discharging into Dyes Inlet or the Port Washington Narrows. The City of Bremerton sanitary sewer system includes both CSOs and emergency overflows (EOF). The EOFs discharge raw sewage into receiving waters during periods of pump station malfunction. Two CSOs and four EOFs discharge into Sinclair Inlet, nine CSOs and five EOFs discharge into Port Washington Narrows, and one CSO discharges into Port Orchard Narrows (TetraTech 1988c).

Surface water runoff is discharged to Sinclair and Dyes Inlets from storm drain outfalls and natural drainage. Runoff from the Puget Sound Naval Shipyard and the cities of Bremerton, Port Orchard and Silverdale is collected by established storm drain systems. In the outlying areas natural drainage channels and culverts discharge surface water runoff into the Puget Sound (TetraTech 1988c).

Figure 3-10: Locations of Superfund and Toxics Cleanup sites and active permitted municipal and industrial discharges in the vicinity of Sinclair and Dyes Inlets (as of 2002).



Sinclair and Dyes Inlets contain some of the most contaminated sediments in Puget Sound. Contaminants of concern include potential carcinogens such as PCBs and PAHs and toxic metals such as chromium, lead and mercury. The highest concentration of PAHs was found at the east end of the shipyard near the Bremerton Ferry Terminal, while the second highest concentration was found in the middle of the passage between Manette and Annapolis. High levels of PCBs have been reported near Bremerton's sewage treatment plant, near the Gorst tide flats, near Annapolis, and even in the center of the harbor, as well as near the shipyard. Heavy metals, including mercury and cadmium, were found in central Sinclair Inlet, near Bremerton's sewage treatment plant, southwest of Port Orchard Marina and several other areas. In many samples, mercury was 10 to 100 times higher than background.

The concentration and flux of metals from the sediment to overlying Puget Sound water was measured in Sinclair Inlet as part of naval base monitoring (Johnson 1993; Chadwick et al. 1994). Despite high concentrations of metals in the sediments, results suggested low availability and mobility of these metals because of high AVS relative to SEM concentrations. Higher flux rates were measured for metals with lower sulfide solubility (i.e., Ni and Zn).

Like Sinclair Inlet but perhaps not as severe, Dyes Inlet has a number of pollution hot spots identified by past studies. Significant levels of hydrocarbons, mercury, chromium and nickel were found offshore a mile south of Silverdale, near Chico Bay, in Ostrich Bay and in Phinney Bay. English sole from Dyes Inlet have significantly elevated concentrations of PCBs and chromium in their tissues, according to the Urban Bay Action Plan. Ostrich Bay is of particular concern because of its proximity to Jackson Park, a federal Superfund site where tons of Navy ordnance and chemicals were dumped into a landfill along the shore. A fish consumption advisory has been issued for all bottom fish and shellfish in the area due to presence of toxic contaminants in the marine environment (WDOH 2002).

3.3.3 Southern Puget Sound

3.3.3.1 Commencement Bay

Commencement Bay near Tacoma was one of the first locations in the state where sediment cleanup was initiated and the work there contributed to the foundation of sediment management in Washington State. Twelve separate locations comprise the Commencement Bay Superfund Site and are associated with the industrial history of Tacoma, including the former ASARCO smelter. All 12 sites are in the latter stages of cleanup (WDOE 2001c).

A number of industries including a pesticide manufacturing facility, a rock-wool plant, steel slags, powdered metal plant, shipbuilding facilities, marinas using arsenic boat

paints, and the Tacoma Smelter have released arsenic-containing materials since 1931, resulting in As sediment concentrations ranging from 0.38 to 1260 $mg \cdot kg^{-1}$ (Davis et al. 1997). Other substances of concern in Commencement Bay and its waterways have included copper, lead, mercury, zinc, PAHs, PCBs and other chlorinated hydrocarbons (EVS 1995). These substances have been found to cause damage (e.g., mortality, behavioral dysfunction, susceptibility to disease) to the benthic invertebrates, salmonids, flatfish and birds inhabiting Commencement Bay.

Recently, WDOE announced significant reduction in the amount of toxic metals discharged to the bay. A combination of industrial activities, shipyard operations, and storm water have contributed to pollution in Commencement Bay, however, several of these facilities have implemented better source control measures during the past decade. Current Toxic Cleanup and Superfund sites in the Commencement Bay area are shown in (Figure 3-11).

Permitted municipal and industrial discharges to Commencement Bay and its waterways include: WWTPs; petroleum facilities; a pulp and paper mill; shipyards; and chemical plants (Figure 3-11).

3.3.3.2 Budd Inlet

Four contaminated sediment sites related to industrial activities, municipal sewage discharges and shipyard operations, have been identified for clean up in Budd Inlet, two of which have been completed (WDOE 2001c). Toxic Cleanup sites in the Budd Inlet area are shown in Figure 3-12.

There are two permitted waste discharges to Budd Inlet, one for a WWTP and the other for a wood preserving facility (Figure 3-12).

3.3.4 Hood Canal

Much of the shoreline surrounding Hood Canal is rural in nature and the vacation homes are served by septic tanks. The only NPDES permitted discharge is located in Lynch Cove and serves a resort inn (Paulson et al. 1993). Industrial activities and naval operations at four sites in Hood Canal have required sediment cleanup (WDOE 2001c). The two naval sites have been completed while the industrial sites are under initial investigation. Toxic Cleanup sites in the Hood Canal area are shown in Figure 3-13.

Figure 3-11: Locations of Superfund and Toxics Cleanup sites and active permitted municipal and industrial discharges in the vicinity of Commencement Bay (as of 2002).

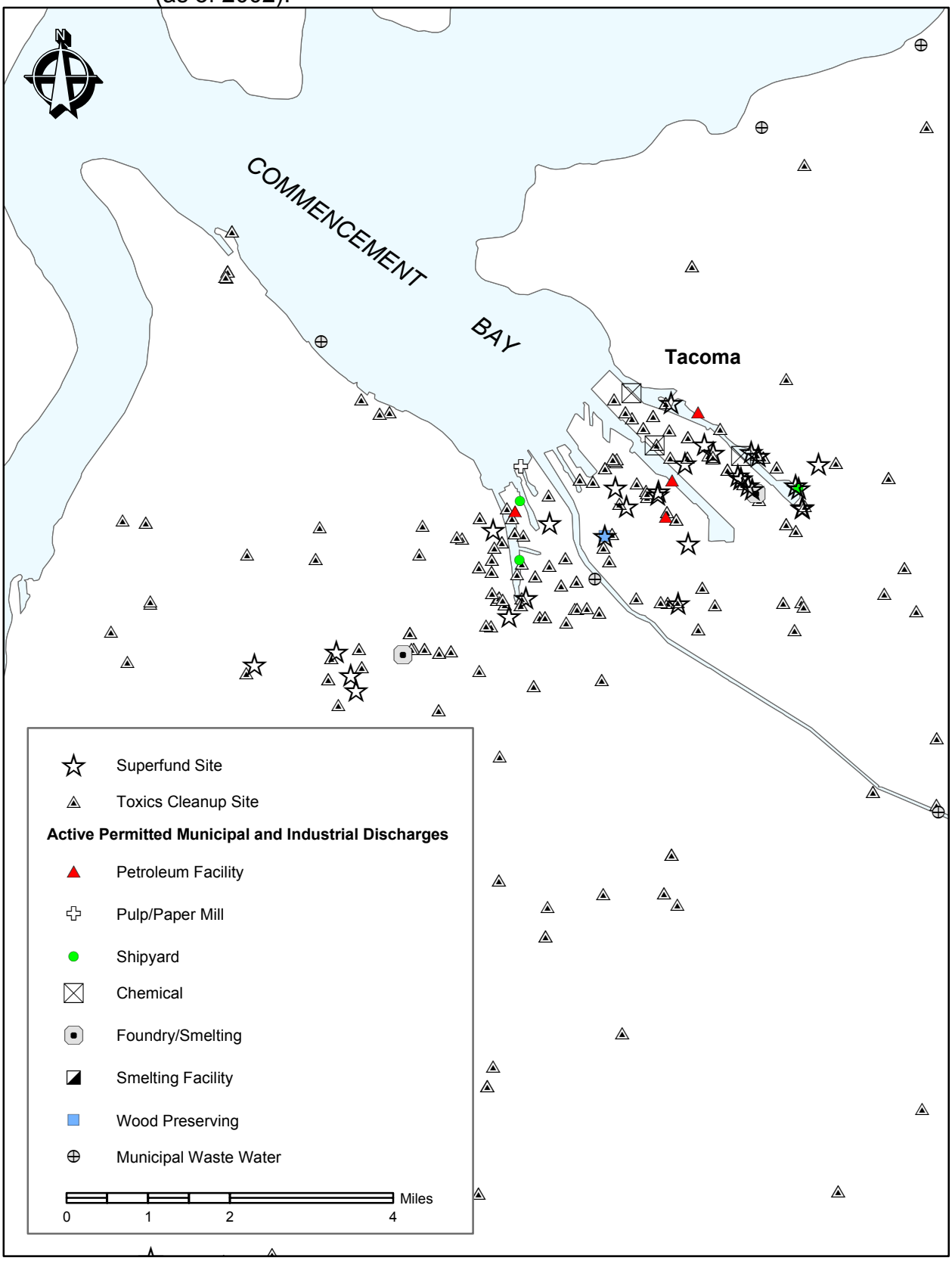


Figure 3-12: Locations of Superfund and active permitted municipal and industrial discharges in the vicinity of Budd Inlet (as of 2002).

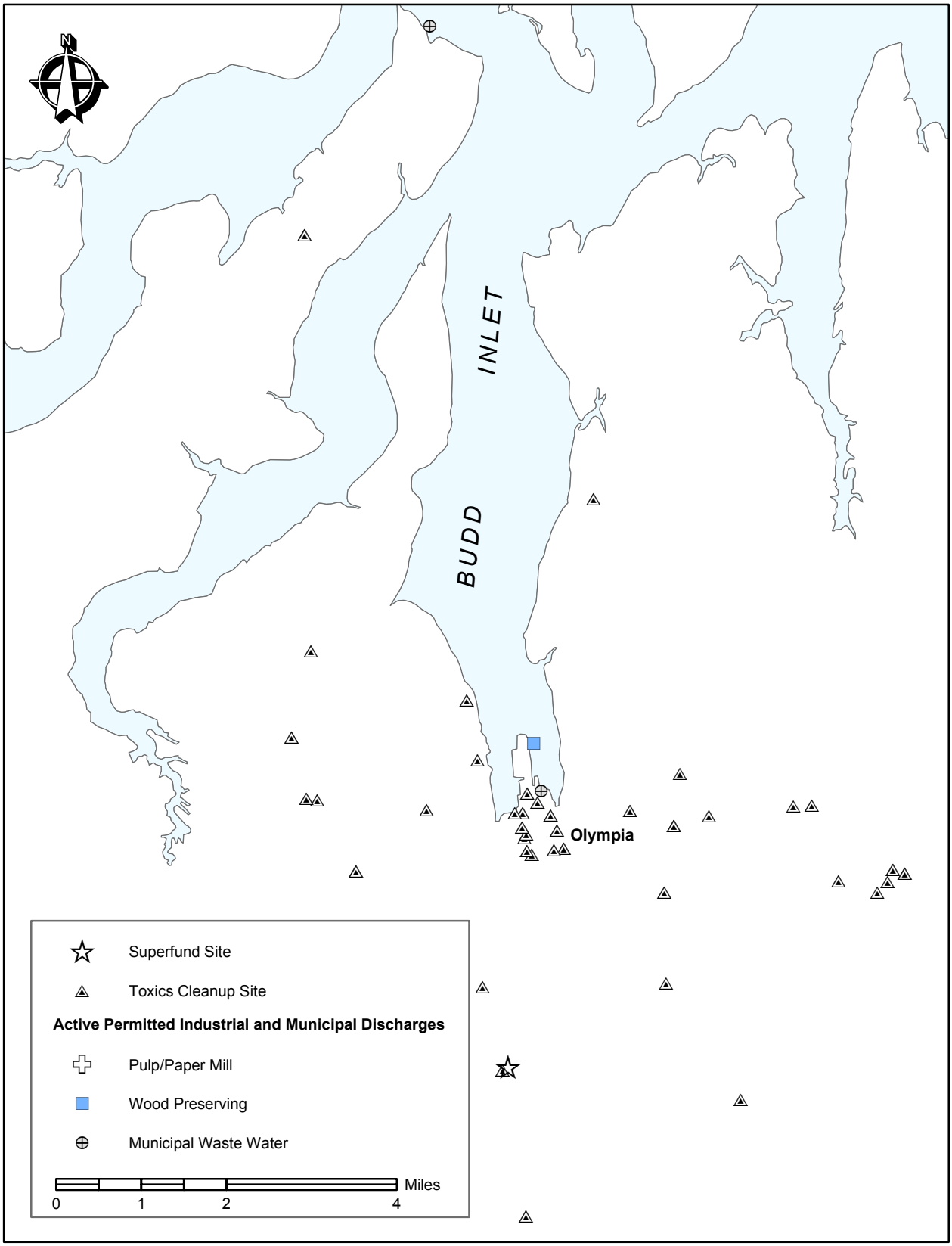
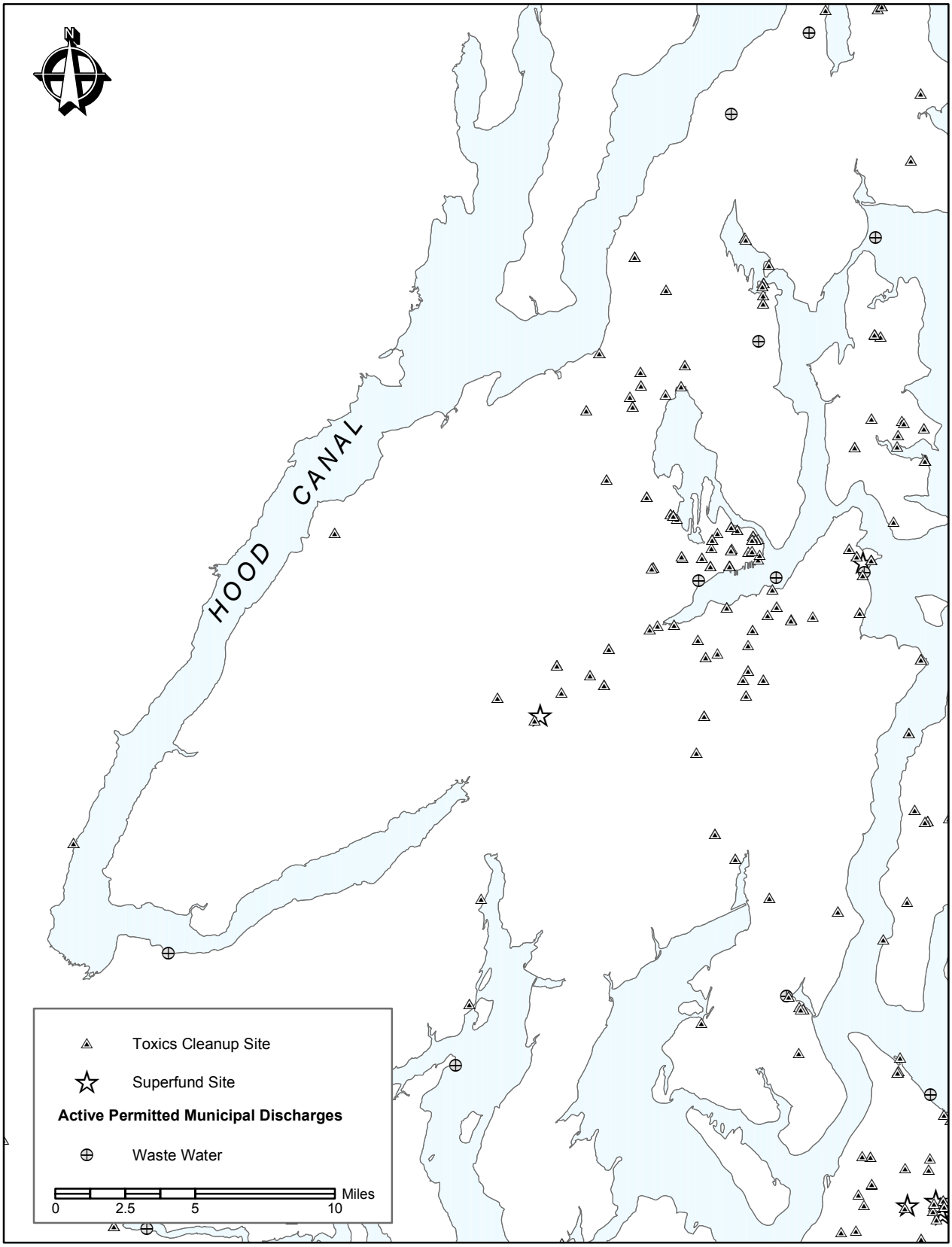


Figure 3-13: Locations of Superfund and Toxics Cleanup sites and active permitted municipal and industrial discharges in the vicinity of Hood Canal (as of 2002).



3.4 MONITORING PROGRAMS

3.4.1 Marine Environment Monitoring Programs

3.4.1.1 Puget Sound Ambient Monitoring Program

The Puget Sound Ambient Monitoring Program (PSAMP) was established in the late 1980s to assess the environmental quality of Puget Sound and to evaluate the effectiveness of the *Puget Sound Water Quality Action Plan* (PSWQAT 2000a). The overall design and direction of PSAMP is defined by the Puget Sound Water Quality Action Team, while its member agencies direct and implement the various studies. Through PSAMP, eight major ecosystem components are monitored: the quality of sediments, freshwater and marine waters; the health of fish, shellfish, birds and mammals; and the condition of physical habitats. The programs addressing three of these components are highlighted below:

Marine Sediment Monitoring Program (MSMP) – The overall goal of the MSMP is to document both natural and human-induced changes in sediment quality throughout Puget Sound (Llansó et al. 1998; data have been reported in two volumes - Llansó et al. 1998a, b). A total of 76 stations were established and have been sampled by WDOE staff on a regular or rotational basis since 1989. The MSMP uses the triad approach (i.e., sediment chemistry, benthic community structure, and laboratory toxicity testing) to assess sediment quality. Metals and organic contaminant concentrations at stations monitored by the MSMP were generally low near detection limits, except at specific stations closely associated with urban or industrial centers, such as Sinclair Inlet, Dyes Inlet, Eagle Harbor, Elliott Bay and Commencement Bay.

The results of PSAMP's recent sediment assessment initiative in partnership with NOAA are discussed in Section 4.2.

Marine Waters Monitoring Program – WDOE's Marine Waters Monitoring Program is focused on assessing basic water quality conditions (i.e., temperature, salinity, dissolved oxygen, nutrients) throughout Puget Sound and along Washington's outer coast (Newton et al. 1998). Toxic metals and organic contaminants are not measured as part of this program.

Fish Contaminants and Fish Health – WDFW staff implement the Fish Component of PSAMP, the overall objective of which is to assess tissue burdens and health effects of a variety of metals and organic contaminants in fish and macroinvertebrates (West et al. 2001a). Ten marine fish species representing different life history patterns and feeding strategies have been monitored annually or intermittently at over 50 stations throughout Puget Sound since 1989. The results of this program are discussed in Section 4.3.2.

3.4.1.2 National Oceanic and Atmospheric Administration – National Status and Trends Program

NOAA initiated the National Status and Trends (NS&T) Program in 1984 to determine the current status of U.S. estuarine and coastal waters (NOAA 2002). The NS&T Program involves long-term monitoring of contaminants and other environmental conditions at more than 350 sites along U.S. coasts through a number of projects, including the following:

National Benthic Surveillance Program (NBSP) – Through the NBSP, chemical contaminants in bottom-dwelling fish and the sediments on which they lived were monitored at 120 sites nation-wide between 1984 and 1993. The biological effects of the measured tissue burdens (i.e. toxicopathic liver disease) were also assessed.

Sediment Coring Project – Ten estuaries were selected in 1989 as locations from which sediment core samples would be collected to determine temporal trends in sediment contaminant concentrations. The results for Puget Sound are discussed in Section 4.2.4.

Mussel Watch Project – NOAA has used bivalve mollusks (e.g., mussels and oysters) since 1986 to monitor the quality of coastal and estuarine environments. Samples are collected from representative areas rather than near known or suspected contaminant sources. Data for mussels collected from Puget Sound are discussed in Section 4.3.1.

3.4.1.3 Pacific Marine Environmental Laboratory

In 1979, scientists at the Pacific Marine Environmental Laboratory (PMEL) began an investigation of the sources, transformation, transport and fate of pollutants in Puget Sound and its watershed. The process studies were undertaken to understand the role of flocculation in trace metal transport. Subsequently, the research centered on the role of suspended sediments in transporting and redistributing trace metals and organics in the main basin of the Sound. Twenty-eight cruises were undertaken between 1979 and 1986 in two urban embayments (Elliott and Commencement Bays) and the open waters of Puget Sound. In addition to the water column trace metal and ancillary data, sediment trap, sediment column solid phase and sediment column interstitial phase (pore water) data were collected (Paulson et al. 1991).

3.4.2 Freshwater Environment Monitoring Programs in the Puget Sound Basin

3.4.2.1 United States Geological Survey

The Puget Sound Basin National Water Quality Assessment (NAWQA) Surface Water Studies assesses the quality of the more than 100 streams and rivers that discharge to

Puget Sound and incorporates this information into the U.S. Geological Survey (USGS) national assessment of water quality. These studies included:

Retrospective Studies – provide a first-cut analysis of general water-quality conditions within the study unit using existing water-quality data. Information on the occurrence and distribution of nutrients and pesticides in Puget Sound Basin streams was used to calculate the nutrient loads from major rivers to Puget Sound. A large existing nutrient data set collected primarily by the Washington State Department of Ecology, the USGS, and King County METRO allowed the calculation of average annual nutrient loads (inorganic nitrogen and phosphorus) for the major rivers discharging to Puget Sound. These loading estimates reveal water-quality differences between major watersheds in the Basin and allow estimates of total nutrient input to Puget Sound from surface-water sources (Embrey and Inkpen 1998; Inkpen and Embrey 1998).

Pesticides have been evaluated in small streams in the Puget Sound Lowland in recent years by WDOE, the U.S. Environmental Protection Agency, and researchers from Western Washington University. Analysis of water samples for pesticides is used to determine the occurrence and distribution of these chemicals in small streams draining predominantly agricultural and urban land in the Puget Sound Lowland (Bortleson and Davis 1997).

Monitoring fish and benthic invertebrate communities, assessing aquatic habitat, and analyzing for contaminants in fish tissue have been done by WDOE, researchers from the University of Washington and Western Washington University, U.S. Forest Service, U.S. Army Corps, the Salmon and Steelhead Habitat and Assessment Project, and U.S. Environmental Protection Agency. Puget Sound NAWQA biologists compiled basin-wide information and data and evaluated the status and trends of aquatic biota, summarized factors affecting aquatic biota, and provided a topical bibliography in the report *Water Quality Assessment of the Puget Sound Basin, Summary of Stream Biological Data Through 1995* (Black and Silkey 1998)

Fixed Station Water-Quality Sampling (1996-1998) – has been conducted at 11 sites: Thornton Creek in the Lake Washington basin, and 10 stations in the Nooksack, Green, and Skokomish River basins. These sites were sampled at least once a month to evaluate the integrated effects of various land uses on water quality, using data from sites located near the mouths of watersheds. Data were used to address the specific effects of individual land uses, using sites located in smaller upstream watersheds comprised of predominantly forested, agricultural, suburban, or urban land uses (Ebbert et al. 2000).

Synoptic Sampling (1998) – evaluates the distribution of an individual water-quality constituent or group of constituents over a geographically dispersed area. In 1998, pesticide transport during storm runoff by streams draining urban-suburban areas was evaluated (Voss et al. 1999). The streams sampled included: Sunset Creek, Valley Creek, Juanita Creek, Lewis Creek, Thornton Creek, South Fork Thornton Creek, Thornton

Creek below Jackson Park Golf Course, Lyon Creek, Longfellow Creek, Miller Creek, Des Moines Creek, and Little Soos Creek.

3.5 DATA GAPS

- Lack of recent information on contaminants in stormwater runoff to determine trends and need for source control measures;
- Lack of information on contaminant loading from CSOs and EOFs.

4. FATE AND EFFECT OF CONTAMINANTS IN PUGET SOUND

As shown in Section 3, contaminants can enter the Puget Sound aquatic environment via a number of pathways. Once these contaminants are in the water and sediments of the Sound, portions of the contaminant load can become available to aquatic biota. Both chronic (i.e., sublethal effects such as reduced fecundity or growth, or behavioral modifications that occur after prolonged exposure) or acute (i.e., mortality that occurs after a short exposure to high concentrations) effects can occur when aquatic organisms take up and assimilate various contaminants into their body tissues. Numerous properties of the contaminants (as summarized in Section 2), as well as of the sediments and of individual marine species, interact in the final expression of toxicity.

Figure 4-1, a conceptual model of PCB transfer pathways and aquatic receptor species in the lower food web of Puget Sound, illustrates the complexity of interactions between environmental compartments, and identifies potential exposure pathways that may be significant in the Sound. Sediments are often the initial source of contaminants; invertebrates living in the sediments can assimilate contaminants through dermal contact or ingestion. Benthic fish feed on the invertebrates, taking in the contaminant body burdens of their prey, as well as any incidental ingestion from the sediments. These benthic fish then become food items for larger pelagic fish, which are in turn fed upon by killer whales and otters. The primary exposure pathway for humans is the consumption of contaminated fish and shellfish. This conceptual model is intended to demonstrate the variety of pathways and receptors that might be applicable to Puget Sound as a whole; site-specific conditions, especially changes in land use patterns, will alter the presence (and significance) of the applicable pathways and receptors.

This section is broken down into discussions of the toxic contaminants identified in Section 2, and includes:

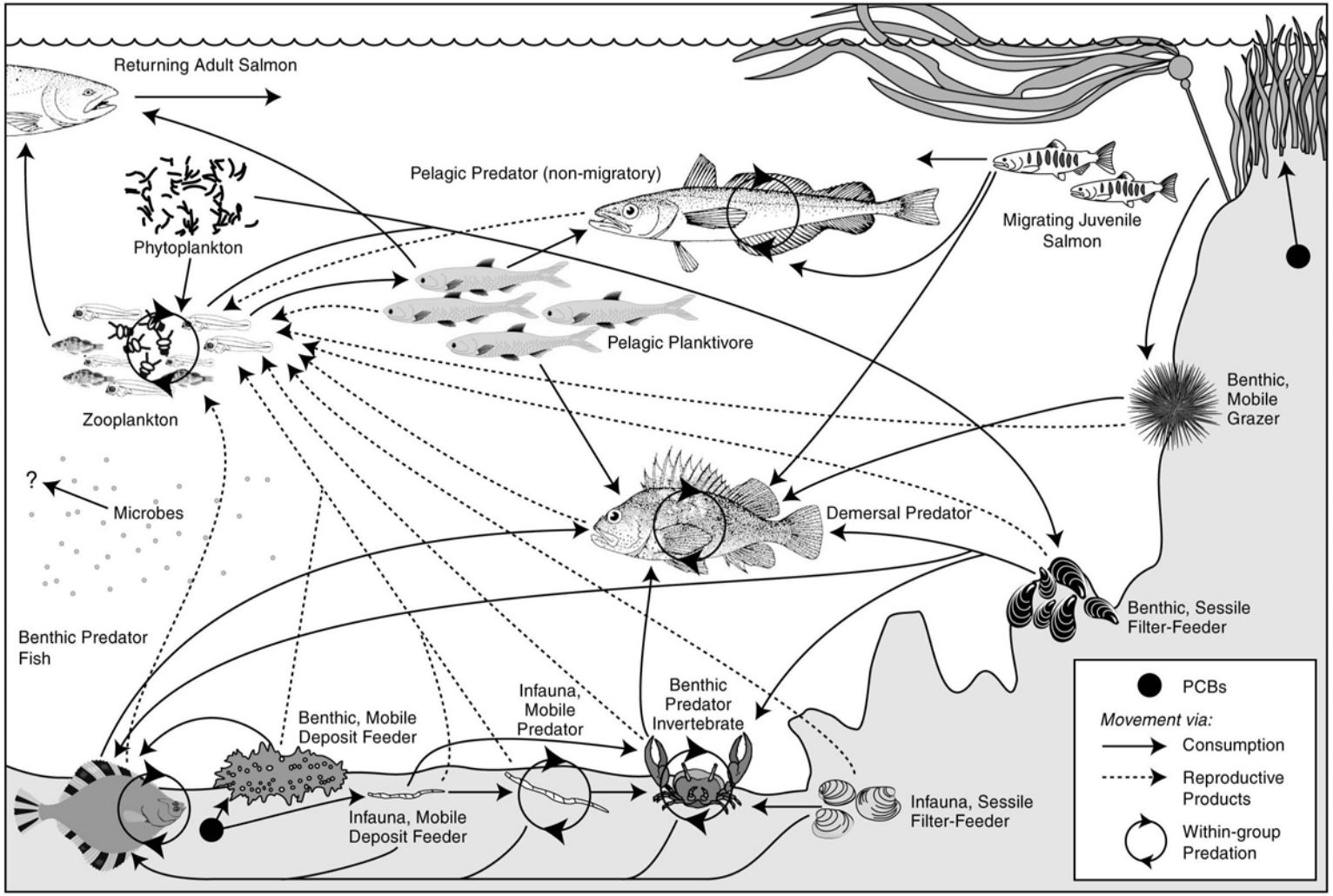
- metals such as arsenic, copper, cadmium, lead, mercury and tributyltin; and,
- organics such as PCBs, PAHs, pesticides, dioxins and furans, and phthalate esters.

The environmental fate of the contaminants in the Sound are further detailed according to their respective exposure media (i.e., sediment, water).

4.1 WATER

Puget Sound is a complex collection of embayments, deep water channels and industrialized waterways. Tidal exchange from the Straits of Georgia and Juan de Fuca results in considerable dilution of the water column, while at the same time shallow sills on the ocean floor restrict the movement of deeper waters (Harrison et al. 1994). Due to the diverse physical oceanography of Puget Sound, water quality conditions are varied.

Figure 4-1: Conceptual model for PCB cycling in the lower food web of the Puget Sound aquatic environment (courtesy of J. West and S. O'Neill of WDFW).



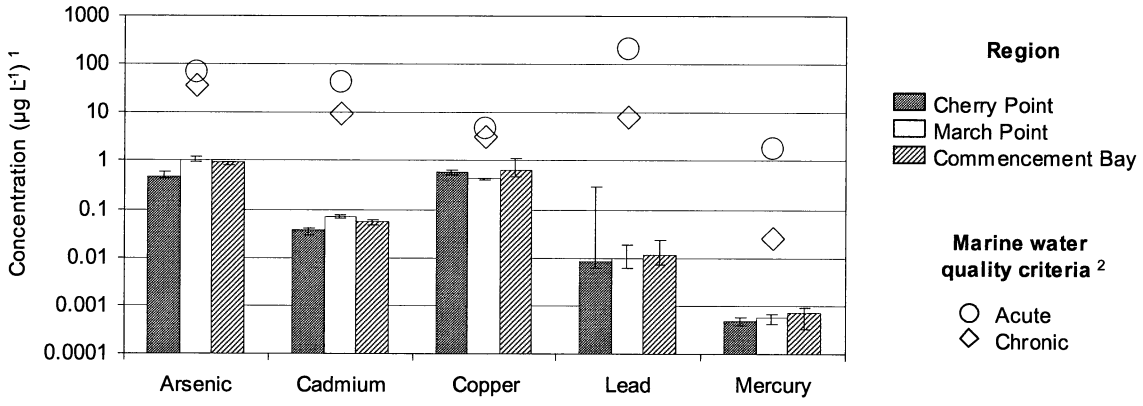
Water quality monitoring in Puget Sound, such as that conducted by PSAMP, assesses ambient physical properties such as salinity, dissolved oxygen (DO_2), and temperature, as well as nutrient concentrations and bacterial counts. The quality of the water column tends to be good in terms of these conventional parameters (i.e., they meet applicable water quality criteria), however, some areas are sensitive to eutrophication (Newton et al. 1998). Toxic contaminants are not included in most ambient monitoring programs for a number of reasons. Many contaminants of concern tend to be particle reactive and settle out of the water column and into the benthic sediments. The dilution of the water column via tidal flushing will also mask water-borne contaminants except in localized areas such as adjacent to point source discharges, non-point source surface runoff, or marine spills. Water quality data that have been collected regarding toxic contaminants indicate that source control programs implemented in the 1970's helped to reduced quality problems in the water column related to the forest industry and wastewater treatment plants (PSWQA 1991). However, there may be on-going water quality issues near hazardous waste facilities, industrial effluent discharges, stormwater outfalls, and combined sewer overflows.

A less well-understood part of the water column is that of the sea-surface microlayer (SMIC). The interface between the sea and the atmosphere is highly productive and can support dense populations of crustacean larvae that are preyed upon by juvenile salmonids. As well, the pelagic eggs of several commercially important fish species such as sole, flounder and rockfish float at the sea surface (Hardy and Word 1986). Studies in the 1980s determined that the concentrations of some contaminants at the sea surface, both metals and organics, exceeded ambient water quality criteria and were toxic in both laboratory and *in situ* toxicity tests (Hardy et al. 1987a,b).

4.1.1 Review Of Existing Water Quality Data

Studies of water-borne contaminants in Puget Sound have been conducted with various objectives and geographical foci. For example, Crecelius and Cullinan (1998) conducted a study of the presence of 13 U.S. EPA priority pollutant metals in areas representative of three different regions of Puget Sound. In each region (i.e., Cherry Point northwest of Bellingham; March Point near Anacortes; Commencement Bay at Tacoma) samples were collected from ten stations laid out in a grid fashion and covering an area of about 18 km^2. Analysis for metals was conducted using new U.S. EPA methods capable of detecting low concentrations in seawater. The results for dissolved arsenic, cadmium, copper, lead and mercury are summarized in Figure 4-2. Total metals concentrations were similar indicating that the dissolved fraction predominated. The authors indicated that the results presented are the only known data regarding regional concentrations of a number of metals in Puget Sound.

Figure 4-2: Water column metals concentrations in three regions of Puget Sound. Columns are mean concentrations, with range indicated by the error bars.



Notes:
[1] All concentrations are reported as dissolved, except for mercury, which is total.
[2] Water quality criteria are from Chapter 173-201A WAC (Ecology, November 1997).
Source: Crecelius and Cullinan (1998)

King County conducts regular sampling of offshore and intertidal waters in Puget Sound in fulfillment of requirements related to an NPDES permit to determine whether or not wastewater discharges are affecting the receiving environment (King County 2001). The geographical area covered by this sampling corresponds to the Central Puget Sound region. The King County monitoring program for 1999 and 2000 assessed 108 organic compounds, including chlorinated pesticides, PCBs, chlorinated herbicides, organophosphorus pesticides, PAHs, phthalates and metals. Of these, chlorinated pesticides and herbicides, PCBs and organophosphorus pesticides were not detected in any offshore samples. Table 4-1 summarizes the results for organic compounds that were detected. The four stations (underwater park at Brackett's Landing; Edwards Point; Golden Gardens Park; Shilshole Bay) with the most frequent detections of PAHs were located near piers, docks and marinas that may be sources of creosote (King County 2001).

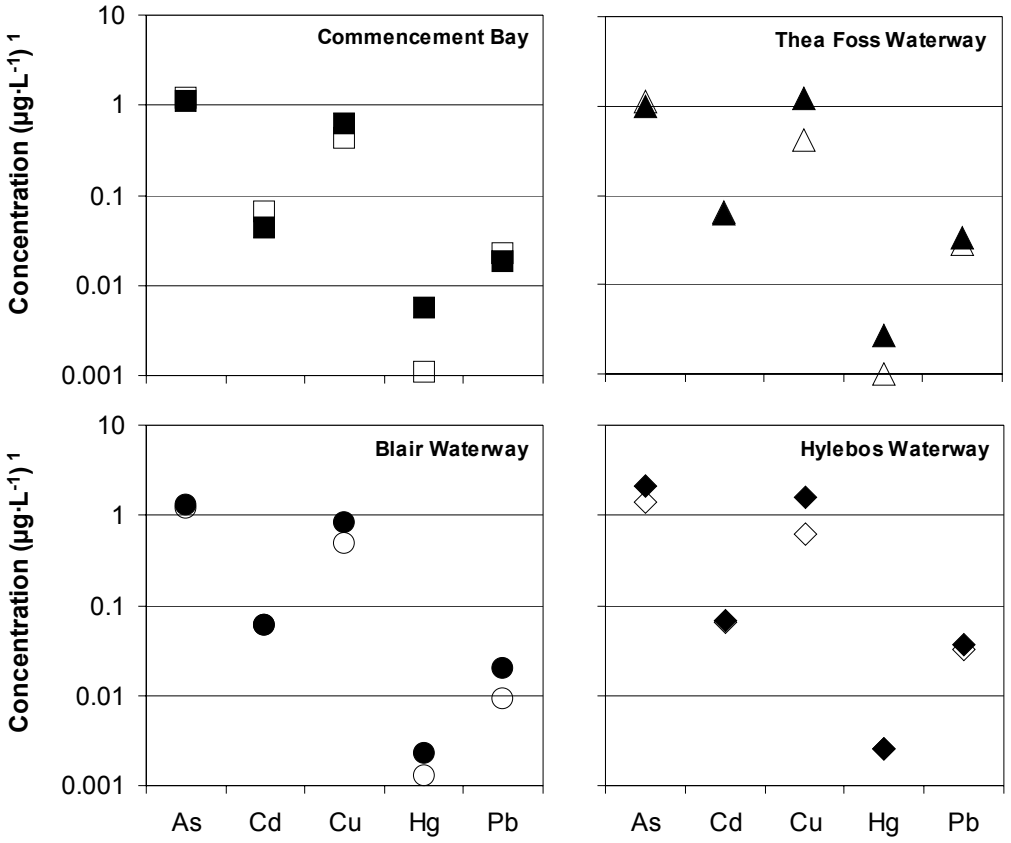
Table 4-1: Organic contaminant concentrations and frequency of detection (FOD) in the Puget Sound waters of King County (1999-2000)

Parameter[a]	Offshore		Intertidal	
	Concentration[b]	FOD[c]	Concentration	FOD
Phthalates				
Benzyl butyl phthalate	0.048 (0.0081-0.28)	2.6	NR	
Bis(2-ethylhexyl) phthalate	1.7 (0.14-37)	21	2 (0.71-15.4)	15
Di-N-butyl phthalate	0.17 (0.070-0.26)	0.6	0.74 (0.53-0.83)	3
Di-N-octyl phthalate	0.011 (0.0054-0.063)	3.5	0.0092 (0.0064-0.83)	14
Diethyl phthalate	0.024 (0.0020-0.070)	1	0.1 (0.082-0.25)	12
Dimethyl phthalate	0.0068 (0.0047-0.14)	17	0.009 (0.0054-0.019)	11
PAHs				
Acenaphthene	NR		0.0075 (0.0058-0.084)	4.5
Anthracene	NR		0.023 (0.019-0.026)	3
Benzo[a]anthracene	0.014 (0.014-0.014)	0.3	0.015 (0.015-0.015)	1.5
Benzo[a]pyrene	0.017 (0.017-0.017)	0.3	0.014 (0.0094-0.018)	3
Benzo[b]fluoranthene	0.011 (0.011-0.011)	0.3	0.014 (0.0089-0.019)	3
Benzo[k]fluoranthene	0.0090 (0.0090-0.0090)	0.3	0.014 (0.0078-0.02)	3
Chrysene	0.015 (0.015-0.015)	0.3	0.025 (0.025-0.025)	1.5
Fluoranthene	0.0052 (0.0048-0.030)	1.3	0.008 (0.0051-0.051)	32
Fluorene	NR	0	0.0067 (0.006-0.0095)	11
Naphthalene	0.019 (0.012-0.021)	1	0.017 (0.013-0.036)	18
Phenanthrene	0.005 (0.0049-0.0086)	5.1	0.0088 (0.0052-0.048)	36
Pyrene	0.0053 (0.0048-0.037)	2.3	0.011 (0.0052-0.032)	15

Source: King County (2001)
[a] Only parameters detected are reported
[b] Concentration is $\mu g \cdot L^{-1}$, reported as median with range in brackets
[c] FOD – frequency of detection, in %
NR – not reported

Localized areas have also been monitored to determine background concentrations and to assess source control efforts. Washington Department of Ecology conducted an assessment of metals concentrations in Commencement Bay and three of it's waterways (i.e., Thea Foss, Blair, Hylebos) between November 1997 and August 1998 to determine changes in the receiving water quality following the clean-up of several Superfund sites (Johnson and Summers 1999). Samples collected from the surface (0.5 m) and deeper in the water column (8 m) were measured for a number of metals. The results for arsenic, cadmium, copper, lead and mercury are shown in Figure 4-3. Surface water concentrations of copper and lead were significantly higher than deep water concentrations.

Figure 4-3: Summary of selected metals concentrations at the surface (0.5 m; solid symbols) and at depth (8 m; white symbols) in Commencement Bay and its waterways in 1997 and 1998.



Notes:
[1] All results are reported as median concentrations (in $\mu g \cdot L^{-1}$) for dissolved metals except mercury, which is reported as total recoverable.
Source: Johnson and Summers (1999)

In the mid-1980s chemical analyses and toxicity tests were conducted on the sea-surface microlayer in Puget Sound in order to determine the sources, fate and effects of toxic contaminants at the atmosphere-hydrosphere interface. Mean concentrations of total "SMIC metals" (i.e., metals with an affinity for the sea-surface microlayer such as cadmium, lead, zinc, silver and copper) ranged from <0.05 $mg \cdot L^{-1}$ in Central Sound and Sequim Bay to ~1.4 $mg \cdot L^{-1}$ in Elliott Bay (Hardy et al. 1987b). Mean concentrations of total PAHs were also lowest in Central Sound and Sequim Bay (<20 $\mu g \cdot L^{-1}$), while the highest mean was for Port Angeles Harbor (~650 $\mu g \cdot L^{-1}$). A principal components analysis determined the relative importance of contaminant class to decreased larval survival in sand sole embryo toxicity tests to be: pesticides and metals > aromatic compounds > saturated compounds > PCBs. However, further analysis showed that all contaminants classes tended to co-occur at high concentrations so the precise cause of toxicity was not identifiable.

Sources of the SMIC contaminants were suggested to be direct or indirect atmospheric deposition of metals and fossil fuel combustion byproducts, and the upwelling of domestic sewage from subsurface outfalls (Hardy et al. 1987b). An additional potential source for contaminants in the SMIC is the suspension of benthic sediment during dredging (Word et al. 1987).

TBT concentrations in the Puget Sound water column have been measured for a number of years, however, the data are proprietary in nature (Cardwell, pers. comm. 2002), and were not available for inclusion in this document.

4.1.2 Comparison to Water Quality Standards

Water chemistry was screened against the *Water Quality Standards for Surface Waters of the State of Washington* (Chapter 173-201A WAC) and the USEPA *National Recommended Water Quality Criteria: 2002*, which are summarized in Appendix A1. Both sets of benchmarks have values to protect against acute (i.e., short-term exposure resulting in injury or death) and chronic (i.e., long-term or repeated exposure resulting in injury or death) effects to aquatic organisms. The USEPA has also developed water quality criteria for the protection of human health. The criteria used in this assessment are based on an increased lifetime cancer risk level of 10^{-6} due to the consumption of aquatic organisms.[1]

The Washington State and USEPA numerical water quality criteria for the protection of aquatic organisms apply to the dissolved fraction of the metal as this generally approximates the bioavailable form of the metal. Metals in the water column may exist in a dissolved phase or adhered to particulate material. Dissolved metals are defined as those that will pass through a 0.45 μm filter.

In comparison, water quality studies may report concentrations as total metals, and permit criteria for waste-water discharges may require the analysis of total recoverable metals. Total metals and total recoverable metals both refer to the sum of dissolved metals and those adhered to particulates that will not pass through a 0.45 μm filter. The difference between the two is the digestion method (e.g., the use of an acid to release adsorbed metals) used.

4.1.2.1 Metals

The concentrations of arsenic, cadmium, copper, lead and mercury in all the samples measured by Crecelius and Cullinan (1998) were below the acute and chronic marine water quality criteria set by the State (Figure 4-2). In King County's Puget Sound waters,

[1] The manifestation of effects related to the consumption of marine organisms are dependent on a number of factors including exposure route, form of the compound, and diet (Section 2). Subsistence and recreational consumers may be at greater risk of exposure to contaminants in fish and shellfish.

arsenic, cadmium, copper, lead and mercury were detected at almost 100% of the offshore and intertidal water samples measured in 1999 and 2000, however, they were all below the marine water quality acute and chronic criteria, as well (King County 2001). Metals concentrations in Commencement Bay and its waterways were, for the most part, similar to those measured in the Puget Sound waters of King County (Johnson and Summers 1999). All of the reported arsenic concentrations exceeded the level for the protection of human consumers of fish, however, USEPA is currently reassessing the criterion (USEPA 2002).

In comparison, metals concentrations detected in the sea-surface microlayer were several orders of magnitude higher than those detected in the subsurface water column and approached or exceeded both the acute and chronic criteria for marine waters. For example, sea-surface concentrations of copper in Elliott Bay ranged from 9.8 $\mu g \cdot L^{-1}$ to 3,215 $\mu g \cdot L^{-1}$. However, comparison of SMIC contaminants to existing water quality criteria may not be meaningful because metal speciation and therefore toxicity of contaminants may be markedly different than in the subsurface water column (Hardy 1982).

4.1.2.2 Organics

Washington State does not have water quality criteria for the organic parameters detected in King County's Puget Sound waters (Table 4-1), whereas the nine organochlorine pesticides, one organophosphate pesticide, and total PCBs for which there are criteria were not detected at all. The British Columbia Ministry of Water, Land and Air Protection (MWLAP; formerly the Ministry of Environment, Lands and Parks – MELP) has established marine criteria for five of the PAHs detected by King County. Naphthalene, acenaphthene, fluorene, and chrysene were all lower than the MWLAP criteria, while benzo[a]pyrene slightly exceeded the marine criterion of 0.01 $\mu g \cdot L^{-1}$ (Nagpal 1993).

4.1.3 Spatial Trends in Water Quality

Crecelius and Cullinan (1998) determined that the water quality of the Cherry Point and March Point regions were not affected by local sources of contamination. Conversely, there appear to be local sources of contamination in Commencement Bay as significantly higher concentrations of mercury, copper and lead were measured, compared to Cherry and March Points. As well, Commencement Bay had greater within-region variability than the other two regions. The data collected by Johnson and Summers (1999) further describes the spatial variability of surface water quality in Commencement Bay and its waterways (Figure 4-3). Generally, Thea Foss and Hylebos Waterways had greater metals concentrations than did Blair Waterway and outer Commencement Bay.

Spatial variation in metals concentrations of the sea-surface microlayer were even more striking (Table 4-2). Urban embayments such as Elliott Bay and Commencement Bay had

copper and lead concentrations that were orders of magnitudes greater than less-industrialized areas like Sequim Bay and the Central Sound channel (Hardy et al. 1987b). PCB concentrations were non-detectable in Sequim Bay, low in one sample from Central Sound and as high as 3,894 $ng \cdot L^{-1}$ in one sample from Elliott Bay. The effect of elevated contaminant levels in urban embayments was illustrated during *in situ* embryo toxicity testing (Hardy et al. 1987a). The normal live hatch of sand sole eggs from Port Angeles Harbor was 4% of that in Sequim Bay, while Commencement Bay had 42% normal live hatch and Elliott Bay had 75%. All of the larvae tested at the Commencement Bay station exhibited morphological abnormalities such as bent spines. Sea-surface microlayer contaminants may also be deposited on intertidal substrates during ebb tide and affect epibenthic and infaunal species (Gardiner 1992).

Table 4-2: Concentrations of selected metals in the sea-surface microlayer of Puget Sound

	CONCENTRATION[a]		
LOCATION	CADMIUM	COPPER	LEAD
North Puget Sound			
Port Angeles	X	16	310
Sequim Bay	0.26 (0.1-0.6)	10 (1.0-34)	4.9 (0.7-17)
Central Puget Sound			
Elliott Bay	0.84 (0.2-2.7)	800 (9.8-3,200)	64 (3.9-160)
Central Sound	0.72 (0.1-1.7)	12 (1.3-37)	17 (1.4-56)
South Puget Sound			
Commencement Bay	0.53 (0.2-1.6)	110 (2.7-630)	91 (0.6-650)

Source: Hardy et al. (1987b)
[a] all concentrations in $\mu g \cdot L^{-1}$, mean with range in brackets
X = not measured

4.1.4 Temporal Trends in Water Quality

4.1.4.1 Metals

Johnson and Summers (1999) compared their data set with studies conducted in the mid-1980s and found that similar concentrations of arsenic, cadmium and lead were reported then (with the exception of mercury, which was not measured in the earlier studies). Historical data cited in Johnson and Summers (1999) indicated that nearshore arsenic concentrations in the Blair Waterway were as high as 120 $\mu g \cdot L^{-1}$ in 1984, and 59 $\mu g \cdot L^{-1}$ in the center of the channel, compared to 2.0 $\mu g \cdot L^{-1}$ measured in 1997. The change is attributed to cleanup and source control of known metals sources such as ASARCO slag (used to stabilize roadways and in the manufacture of rock wool), scrap yards, marinas and shipyards.

4.1.4.2 Organics

Temporal trends in organic contaminants in the water column could not be assessed due to a lack of data.

4.1.5 Summary of Water Quality

4.1.5.1 State of Knowledge

Water quality in Washington State is regularly monitored through PSAMP. The program focuses on conventional parameters that characterize the physical properties, nutrient conditions and sanitary quality (i.e., bacterial counts) of Puget Sound's marine waters. The data provide information regarding the health of the Sound in terms of potential for eutrophication and depressed dissolved oxygen concentrations, which have implications for aquatic biota and human users of Puget Sound's waterways. The program does not, however, address toxic contaminants such as metals and organic compounds in the water column and there are limited current data regarding metals and toxic organics in the water column.

Limited data were available for an assessment of toxic metals and organics in the water column of the Puget Sound ecosystem. The data that were available suggest that water column contaminants are highest in urban/industrial embayments, but that metals concentrations in places such as Commencement Bay and its waterways have decreased as a result of contaminated site cleanup efforts. Furthermore, contemporary data showed that metals and organic contaminant concentrations were below both chronic and acute water quality criteria for the Puget Sound waters of King County and for Commencement Bay. Initial studies of the sea-surface microlayer, however, show that this biologically important ecotone may be at risk for adverse effects from metal and organic contaminant enrichment.

Because of the complex behavior of metals and organic contaminants in seawater, and a propensity for these chemicals to partition into sediments and biota, water quality monitoring needs to be focused at locations where the effects may occur, such as in the vicinity of point source discharges (i.e., industrial effluent discharges, CSOs, stormwater outfalls) and at the sea-surface microlayer. Such monitoring is important for identifying the need for and success of source control programs.

4.1.5.2 Contaminant Effects

- Toxic contaminants in the sea-surface microlayer (i.e., metals, petroleum hydrocarbons, chlorinated organics) in urban/industrial embayments have been reported to reduce hatchability of sand sole eggs during *in situ* toxicity tests (Hardy et al. 1987b).

4.1.5.3 Monitoring and Data Gaps

Monitoring and data gaps and other issues related to metals and organic contaminants in the water column of Puget Sound include the following:

- Chemical and toxicological analysis of contaminants in the sea-surface microlayer were conducted in the 1980s; there are no data to provide an assessment of current conditions in the SMIC or to determine temporal trends;
- Toxic contaminants other than metals are infrequently measured in the water column, for example, organophosphate pesticides which are less persistent than other pesticides but can have significant effects on biota;
- The comparison of historical (where available) and current data is difficult due to changes in laboratory analytical techniques and in water quality criteria (i.e., dissolved versus total recoverable);
- There is a lack of monitoring to document changes in environmental health related to source control efforts (Mearns, pers. comm. 2002);
- TBT data for the water column have been collected in Puget Sound but the results are proprietary in nature (Cardwell, pers. comm. 2002) and therefore not publicly available

4.2 SEDIMENT

Many contaminants have an affinity for the organic or inorganic components of sediment particles (Section 2). As a result, sediments often act as a contaminant sink with substantially higher concentrations than in the overlying water. During the last decade, chemical contamination of aquatic sediments has been recognized as a serious problem in some U.S. coastal waters. In Puget Sound, for example, hot spots of toxic contaminants have been shown to alter and reduce the benthic community (both abundance and species diversity), to interfere with cellular and physiological processes, and to cause disease in fish. Contaminants may also affect higher trophic organisms such as mammals through bioaccumulation or biomagnification in the food web. Potential sources of contaminants include historical activities on adjacent upland areas as well as present discharges (Section 3). Most hot spots are in areas of high vessel traffic, industrial activities, or poor flushing and are often located near urban centers (NOAA 1994).

4.2.1 Review of Existing Data

A considerable amount of research has been conducted on sediment quality in Puget Sound. The Sediment Quality Information System (SEDQUAL), for example, contains the data from over 400 sediment surveys, for a total of 5,000 stations sampled between 1950-1997 (Long et al. 1999). Studies represented in this database include environmental

assessments for Superfund sites, requests to have waterbodies de-listed (i.e., removed from the 303(d) listing of impaired/threatened waterbodies), and legal settlements regarding spills. Sampling locations for studies represented in this database are focused on suspected or known areas of sediment contamination, and the data were collected during several decades. The SEDQUAL data do not necessarily represent current sediment quality conditions due to sediment cleanup efforts.

Two monitoring programs have addressed sediment from a basin-wide perspective, rather than evaluating areas with the highest contaminant concentrations. First, as part of PSAMP, WDOE's Marine Sediment Monitoring Team has sampled 76 core (i.e., fixed) and rotational monitoring stations located away from contaminant sources for over 180 toxic chemicals (including metals, PAHs, and organochlorines) to determine ambient sediment conditions in Puget Sound (PSWQAT 2002b). Currently, 10 of the original 76 stations continue to be monitored at five-year intervals.

Second, through the National Status and Trends Program, the National Oceanic and Atmospheric Administration (NOAA) and WDOE partnered together to characterize sediment quality throughout Puget Sound. Over the course of three years, samples were collected from three hundred randomly selected stations in North Sound (the U.S.-Canada border to Everett), Central Sound (Everett Harbor through Commencement Bay), South Sound (south of Tacoma Narrows through to Shelton), and Hood Canal/Port Townsend (PSWQAT 2002a). The sampling program followed the Sediment Quality Triad approach, in which bulk chemistry, toxicity in laboratory tests, and benthic-community structure of the site is assessed (Long and Chapman 1985). The chemical analyses included metals, chlorinated organics, PAHs, organotins, and phthalate esters. Sediment toxicity was determined in the laboratory using amphipod survival, sea urchin fertilization, Microtox®, and cytochrome P450 RGS. Long et al. (1999; 2000; 2002) then used the data to extrapolate from each sampling point to the larger surrounding area in order to estimate the spatial extent and magnitude of sediment degradation in Puget Sound.

The following overview of contaminants concentrations in Puget Sound sediments is based on the data collected through the joint PSAMP-NOAA sampling effort between 1997 and 2000 (PSAMP-NOAA 2002).

Arsenic, copper, lead and mercury were detected in all of the samples analyzed from North, Central and South Puget Sound and Hood Canal sediment stations (Table 4-3). In comparison, cadmium was detected in as few as 19 samples from North Puget Sound to as many as 94 samples in Central Puget Sound, while TBT was not detected in Hood Canal samples to as many as 86 samples in Central Puget Sound. Metals concentrations generally varied from one to two orders of magnitude in each of the three regions. TBT concentrations, however, varied as much as 10,000-fold.

PAH compounds were detected in almost all of the samples collected from North, Central and Sound Puget Sound and Hood Canal (Tables 4-4, 4-5, 4-6 and 4-7). Generally, the

lowest concentrations were measured in North Puget Sound sediments, whereas Central and South Puget Sound sediments had the highest concentrations of both low and high molecular weight PAHs. Phthalate ester compounds were detected in fewer than 25% of the samples and also varied considerably from the median concentrations. While the highest median and maximum PAH concentrations were consistently found in the South Puget Sound region, maximum concentrations of different phthalate esters were not found in one region only. For example, the highest concentration of bis(2-ethylhexyl) phthalate was in a sample from North Puget Sound, while the highest concentration of di-n-butyl phthalate was in a sample from Central Puget Sound. Phthalate esters were not detected in any Hood Canal samples. Total PCBs were detected in as few as 21 samples in North Puget Sound to as many as 89 samples in South Puget Sound, however, the highest concentration was measured in a sample from North Puget Sound. Total DDT was detected in a greater number of Central Puget Sound samples than either the North or South. Central Puget Sound also had the highest median concentration of DDT, however, the highest maximum occurred in South Puget Sound. Dibenzofuran concentrations were detected in similar numbers of samples and at similar ranges of concentrations in all three regions.

Table 4-3: Summary of selected metal concentrations in North, Central and South Puget Sound sediments (1997-2000).

	CONCENTRATION[a, b]				
PARAMETER	MEAN	MEDIAN	MIN	MAX	FOD[c]
North Puget Sound (n=105)					
Arsenic	9.5	7.6	2.9	200	105
Cadmium	1.2	0.91	0.54	2.9	19
Copper	38	33	4.4	460	105
Lead	12	6.7	3.0	190	105
Mercury	0.12	0.080	0.010	0.81	105
TBT	25	5.3	0.0033	420	47
Central Puget Sound (n=105)					
Arsenic	12	6.5	1.6	500	105
Cadmium	0.43	0.30	0.10	1.7	94
Copper	42	30	4.0	330	105
Lead	35	22	2.6	500	105
Mercury	0.24	0.13	0.010	1.5	105
TBT	90	9.2	0.49	3,100	86
South Puget Sound (n=84)					
Arsenic	8.6	7.6	2	57	84
Cadmium	0.43	0.25	0.1	2.3	58
Copper	42	34	1.6	420	84
Lead	24	15	4.5	260	84
Mercury	0.14	0.09	0.01	1.6	84
TBT	31	7	3	430	26
Hood Canal (n=21)					
Arsenic	6.4	5.9	2	17	21
Cadmium	0.37	0.14	0.1	1.8	14
Copper	38	29	0.75	110	21
Lead	8.5	5.9	2.2	19	21
Mercury	0.054	0.054	0.017	0.12	21
TBT	nd	nd	nd	nd	0

Source: Long et al. (1999, 2000, 2002) and PSAMP-NOAA (2002)

[a] all concentrations reported as $mg \cdot kg^{-1}$ dw, except for TBT which is $\mu g \cdot kg^{-1}$ dw

[b] concentrations reported for strong acid digestion method

[c] FOD = frequency of detection, or the number of samples in which parameter was detected

dw = dry weight

nd = not detected

n = total sample size

Table 4-4: Summary of selected organic contaminant concentrations in North Puget Sound sediments.

	CONCENTRATION[a]				
PARAMETER	MEAN	MEDIAN	MIN	MAX	FOD[b]
LPAH					
Acenaphthene	44	3.9	0.32	670	89
Acenaphthylene	18	4.1	0.13	110	104
Anthracene	72	12	0.46	1,200	105
Fluorene	62	12	0.98	990	101
Naphthalene	118	15	1.1	1,400	103
Phenanthrene	210	63	4.8	2,300	103
HPAH					
Benzo[a]anthracene	88	29	0.85	1,200	105
Benzo[a]pyrene	59	29	0.27	600	105
Benzo[g,h,i]perylene	42	27	0.59	260	105
Chrysene	113	40	1.5	1,600	105
Fluoranthene	326	78	3.0	4,500	105
Indeno[1,2,3-c,d]pyrene	41	27	0.39	280	102
Pyrene	284	71	2.2	3,800	105
Phthalate Esters					
Bis (2-ethylhexyl) phthalate	3,600	400	113	38,000	14
Butylbenzylphthalate	29	28	12	42	14
Di-n-butylphthalate	1,000	410	130	5,600	26
Di-n-octyl phthalate	23	23			1
Diethyl phthalate	49	49	45	53	2
Dimethyl phthalate	100	100	31	170	2
Chlorinated organics					
Total PCB	190	9.2	3.3	3,400	21
Dibenzofuran	63	9.8	0.88	1,300	103
Total DDT	2.0	1.6	1.1	3.6	12

Source: PSAMP-NOAA (2002)
[a] concentration reported as $\mu g \cdot kg^{-1}$ dw
[b] FOD = frequency of detection, or the number of samples in which parameter detected (out of 105)
dw = dry weight

Table 4-5: Summary of selected organic contaminant concentrations in Central Puget Sound sediments.

	Concentration[a]				
Parameter	**Mean**	**Median**	**Min**	**Max**	**FOD[b]**
LPAH					
Acenaphthene	52	3.9	0.48	1,700	89
Acenaphthylene	31	4.0	0.05	19	104
Anthracene	130	12	0.97	1,100	105
Fluorene	55	12	0.76	830	101
Naphthalene	200	15	1.9	8,400	103
Phenanthrene	280	63	3.3	3,800	103
HPAH					
Benzo[a]anthracene	200	29	1.5	1,800	105
Benzo[a]pyrene	300	29	1.3	2,900	105
Benzo[g,h,i]perylene	160	27	1.4	1,000	105
Chrysene	260	40	2.6	1,700	105
Fluoranthene	870	78	4.9	43,000	105
Indeno[1,2,3-c,d]pyrene	160	27	1.2	1,200	102
Pyrene	620	71	4.5	14,000	105
Phthalate Esters					
Bis (2-ethylhexyl) phthalate	510	400	140	1,000	14
Butylbenzylphthalate	50	47	7.7	92	20
Di-n-butylphthalate	560	410	70	2,900	26
Di-n-octyl phthalate	16	16	-	-	1
Diethyl phthalate	21	17	5.1	92	21
Dimethyl phthalate	19	11	3.3	65	12
Chlorinated organics					
Total PCB	150	69	2.5	2,000	67
Dibenzofuran	59	14	1.1	2,000	99
Total DDT	6.4	4.9	0.21	21	46

Source: PSAMP-NOAA (2002)
[a] concentration reported as $\mu g \cdot kg^{-1}$ dw
[b] FOD = frequency of detection, or the number of samples in which parameter detected (out of 105)
dw = dry weight

Table 4-6: Summary of selected organic contaminant concentrations in South Puget Sound sediments.

	Concentration[a]				
Parameter	**Mean**	**Median**	**Min**	**Max**	**FOD[b]**
LPAH					
Acenaphthene	70	10	0.41	2,400	75
Acenaphthylene	64	13	0.18	1,200	84
Anthracene	150	28	0.37	4,200	83
Fluorene	79	15	0.059	2,300	82
Naphthalene	260	54	1.4	7,300	79
Phenanthrene	420	90	2	11,000	84
HPAH					
Benzo[a]anthracene	220	53	0.94	5,400	84
Benzo[a]pyrene	260	63	1.5	5,900	84
Benzo[g,h,i]perylene	170	59	0.92	3,300	84
Chrysene	340	85	1.5	8,200	84
Fluoranthene	540	130	2.2	15,000	84
Indeno[1,2,3-c,d]pyrene	160	46	0.15	3,300	84
Pyrene	660	160	1.8	19,000	84
Phthalate Esters					
Bis (2-ethylhexyl) phthalate	1,100	850	120	4,700	7
Butylbenzylphthalate	54	45	0.35	130	6
Di-n-butylphthalate	270	272	69	470	2
Di-n-octyl phthalate	21	24	6.8	33	3
Diethyl phthalate	200	67	23	840	7
Dimethyl phthalate	23	18	8.2	94	12
Chlorinated organics					
Total PCB	53	12	3.4	830	68
Dibenzofuran	63	18	0.55	1,400	84
Total DDT	5.9	1.9	0.54	30	19

Source: PSAMP-NOAA (2002)
[a] concentration reported as $\mu g \cdot kg^{-1}$ dw
[b] FOD = frequency of detection, or the number of samples in which parameter detected (out of 84)
dw = dry weight

Table 4-7: Summary of selected organic contaminant concentrations in Hood Canal sediments.

	Concentration[a]				
Parameter	**Mean**	**Median**	**Min**	**Max**	**FOD[b]**
LPAH					
Acenaphthene	20	3.7	0.49	264	21
Acenaphthylene	19	7.4	1.0	88	21
Anthracene	51	11	2.9	540	21
Fluorene	31	14	2.4	230	21
Naphthalene	260	34	3.4	3,500	21
Phenanthrene	110	48	11	670	21
HPAH					
Benzo[a]anthracene	36	16	2.1	330	21
Benzo[a]pyrene	48	22	2.1	470	20
Benzo[g,h,i]perylene	23	19	2.2	62	17
Chrysene	51	22	3.5	440	21
Fluoranthene	190	49	5.4	2,200	21
Indeno[1,2,3-c,d]pyrene	38	22	1.4	360	17
Pyrene	200	46	4.8	2,400	21
Phthalate Esters					
Bis (2-ethylhexyl) phthalate	nd	nd	nd	nd	0
Butylbenzylphthalate	nd	nd	nd	nd	0
Di-n-butylphthalate	nd	nd	nd	nd	0
Di-n-octyl phthalate	nd	nd	nd	nd	0
Diethyl phthalate	nd	nd	nd	nd	0
Dimethyl phthalate	nd	nd	nd	nd	0
Chlorinated organics					
Total PCB	5.6	5.1	2.8	12	21
Dibenzofuran	26	7.1	1	290	21
Total DDT	0.61	0.58	0.54	0.72	3

Source: PSAMP-NOAA (2002)
[a] concentration reported as $\mu g \cdot kg^{-1}$ dw
[b] FOD = frequency of detection, or the number of samples in which parameter detected (out of 21)
dw = dry weight
nd = not detected

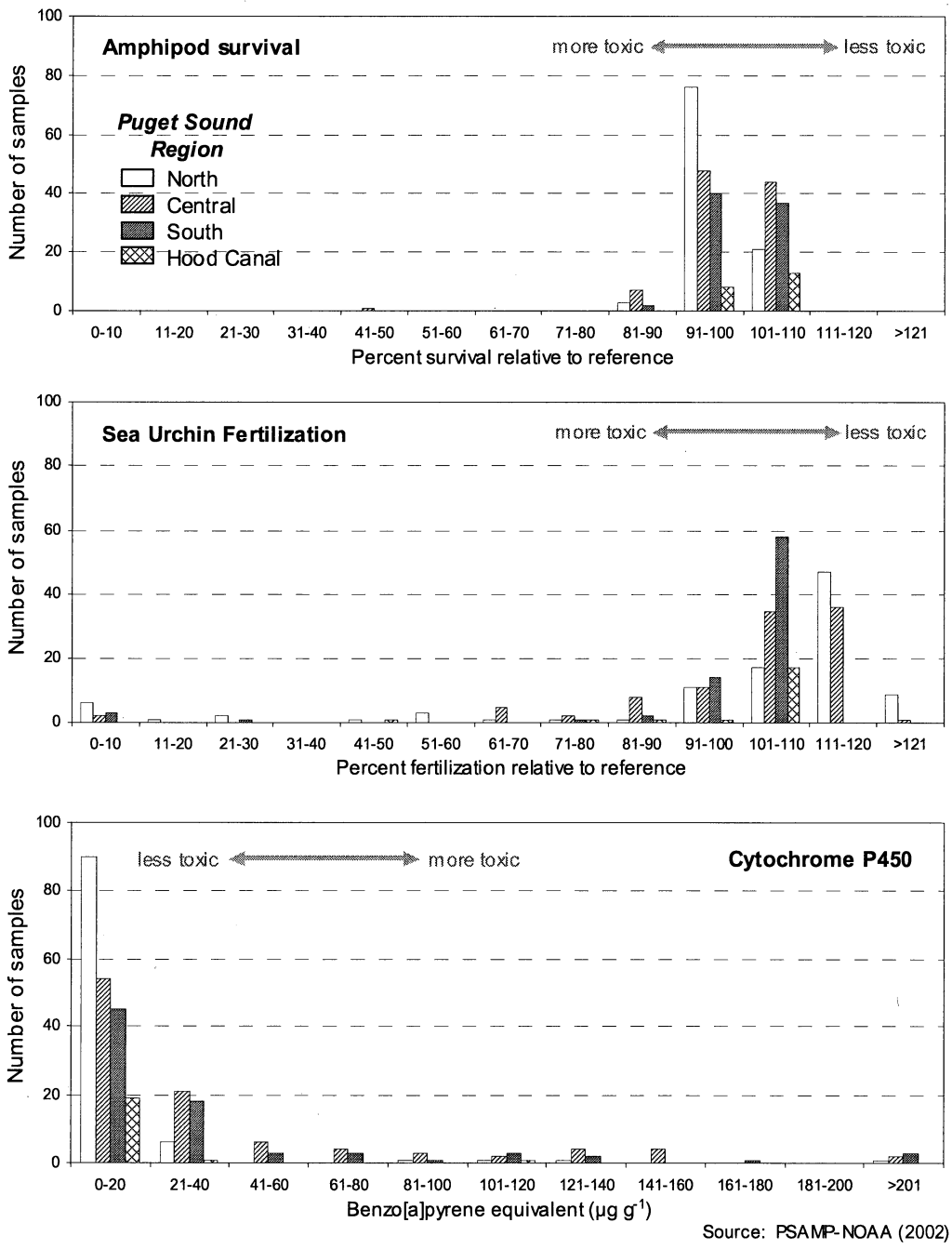
4.2.1.1 Sediment Toxicity Testing

The toxicity of sediment samples collected between 1997 and 1999 were tested using several methods to support a weight-of-evidence approach for assessing the overall potential for adverse environmental effects due to sediment contaminants (Long et al. 1999). The tests, conducted on different phases of the sediments (i.e. solid phase; pore water; solvent extract) and using several toxicological endpoints (i.e. acute mortality, physiological impairment), included the following:

- 10-d amphipod (*Ampelisca abdita*) survival through exposure directly to the solid phase of the bulk sediment sample. The *A. abdita* acute mortality test is commonly used to assess the toxicity of sediments, and is generally not confounded by grain size and ammonia content of the sample;
- Sea urchin (*Strongylocentrotus purpuratus*) fertilization through exposure to pore water extracted from the sediment sample. This method tests the pore water, in which toxic contaminants may be highly bioavailable, and the effects of contaminants on early life stages;
- Microbial luminescence (Microtox®) through exposure to an organic solvent extract of the sediment sample. This test is indicative of the potential toxicity associated with contaminants bound to the sediment particles, and is not affected by grain size or ammonia content of the sample; and,
- Human Reporter Gene System (Cytochrome P450) Response Assay in which transgenic cells derived from the human hepatoma line and linked to the firefly luciferase gene are exposed to an organic solvent of the sediment sample and the response is reported as benzo[a]pyrene equivalents. This assay provides an estimate of sediment-bound contaminants (such as dioxins, furans, HPAH, co-planar PCBs) that may cause chronic and/or carcinogenic effects in benthic organisms.

Cytochrome P450 induction occurred more frequently than adverse responses in the other three toxicity tests described above (Figure 4-4), particularly in the urban-industrial embayments of Central Puget Sound (PSWQAT 2002b). Sea urchin fertilization was also adversely affected to a greater degree by samples from the Central Puget Sound area. Overall, the estimated spatial extent of toxicity as determined by the Cytochrome P450 test was 586 km^2, compared to 94 km^2 for the sea urchin fertilization test, and 11 km^2 for the Microtox® test (PSWQAT 2002b). Amphipod survival was affected in <0.1 km^2 of Puget Sound.

Figure 4-4: Frequency distribution of results for amphipod survival, sea urchin fertilization, and Cytochrome P450 tests on Puget Sound sediments.



4.2.1.2 *Benthic Invertebrate Community Data*

The basic premise behind benthic invertebrate community indices is evaluation of the structure of the benthic community at a disturbed site relative to an undisturbed reference

site. A number of benthic invertebrate community indices are currently available in the primary literature and in use by various state and federal agencies. These indices evaluate a number of different community metrics, such as species diversity, abundance, biomass and pollution sensitivity, to determine an overall assessment of the health of a waterbody. Expected responses of the benthic invertebrate community to pollution include:

- Reduced species diversity;
- Community dominance by one or two key taxa;
- Reduced total biomass (fewer invertebrates), or reduced relative biomass (per any taxa of interest)

An inherent weakness in the application of many invertebrate indices to sites with an abundance of chemical or heavy metals contamination is that most indices were developed as a measure of organic pollution (primarily nutrient enrichment, such as BOD or excess nitrogen), so they are not necessarily sensitive to chemical pollution, such as might result from high concentrations of heavy metals or chlorinated organic compounds like PCBs. In the case of toxic xenobiotics such as these compounds, no existing benthic invertebrate pollution sensitivity classification system is sufficiently comprehensive to allow these organisms to be used for as indicators of habitat contamination (Wogram and Liess 2001).

Results of sediment toxicity tests are frequently compared to benthic invertebrate community data from the same location as the test sediments. The state of Washington, under the Sediment Management Standards, typically considers a ≥50% reduction in invertebrate abundance at the test sites relative to the reference sediments as a biologically significant adverse impact.

Twenty percent of the stations sampled during the PSAMP-NOAA program were found to have impaired benthic communities based on metrics such as abundance, richness, eveness and dominant taxa (PSWQAT 2002b). In North Puget Sound, the benthic communities of some stations in Bellingham Bay, Oak Harbor, Everett Harbor, and Port Gardiner were found to be impaired, while stations in Sinclair and Dyes Inlets, Elliott Bay and the Duwamish Waterway in Central Puget Sound had altered benthic communities. Sampling stations in South Puget Sound's Commencement Bay waterways, Gig Harbor, Budd Inlet and Oakland Bay at Shelton, as well as Port Ludlow and Port Gamble in North Hood Canal also had impaired benthic invertebrate communities.

4.2.1.3 Weight of Evidence

Based on a weight of evidence assessment of the data collected (i.e., integration of bulk chemistry, benthic community and toxicity data), only 1% of the total Puget Sound area was found to have significantly altered benthic communities and laboratory toxicity in

association with high sediment contaminant concentrations, while 57% of Puget Sound maintained an abundant and diverse benthic community despite the occurrence of laboratory toxicity and/or elevated contaminants (Table 4-8; Long et al. 2001). The most seriously degraded sediments occurred in Everett Harbor, the lower Duwamish River, Sinclair Inlet, Commencement Bay and its waterways, Olympia Harbor and along Seattle's waterfront. The remaining 42% of Puget Sound had healthy benthic communities and was free of toxicity and elevated contaminants. These areas tended to be deep basins or shallow bays adjacent to non-urban areas.

Table 4-8: Summary of the spatial extent of sediment degradation in Puget Sound based on a sediment quality triad approach (i.e., bulk chemistry, laboratory toxicity, benthic community).

	NORTHERN PUGET SOUND	CENTRAL PUGET SOUND	SOUTH PUGET SOUND	TOTAL PUGET SOUND
Total area (km^2)	773.9	731.7	857.7	2363
Significant toxicity, high contaminant concentrations, altered benthic community				
n	10	18	11	39
Area (km^2)	10.3	8.1	4.4	22.8
% of area	1.3	1.1	0.5	1.0
Significant toxicity, high contaminant concentrations, abundant/diverse benthic community				
n	16	18	5	39
Area (km^2)	81.7	91.6	39.0	212.3
% of area	10.6	12.5	4.5	9.0
Toxic or contaminated (not both), abundant/diverse benthic community				
n	53	39	49	141
Area (km^2)	530.2	272.6	332.8	1135.6
% of area	68.5	37.3	38.8	48.1
Not toxic or contaminated, abundant/diverse benthic community				
n	21	25	35	81
Area (km^2)	151.7	359.3	481.4	992.4
% of area	19.6	49.1	56.1	42.0

Source: Long et al. (2001)
n = number of samples

4.2.2 Comparison to Sediment Quality Standards

For the purposes of evaluating the quality of sediments in Puget Sound, chemistry data were compared to the Washington State Sediment Management Standards and NOAA sediment quality guidelines (Appendix A2).

1. Washington State Sediment Management Standards (Chapter 173-204, updated in 1995):

    a. Sediment Quality Standards (SQS) – these standards are intended to correspond to a sediment quality that will result in no adverse acute or chronic effects on biological resources. These values essentially represent the state's lower-bound for sediment screening.

    b. Cleanup Screening Level criteria (CSL) – these standards are intended to establish minor adverse effects levels. Sites above this level are identified as cleanup sites of potential concern, while sites below this level are identified as cleanup sites of low concern. These criteria represent the state's upper-bound for sediment screening, as they are less conservative (less protective) than the SQS.

2. NOAA Sediment Quality Guidelines (Long et al. 1995)

    a. Effects range low (ERL): The ERL is the lower 10^{th} percentile of the effects studies for a chemical. Concentrations below the ERL represent a minimal effects range where effects would be rarely observed.

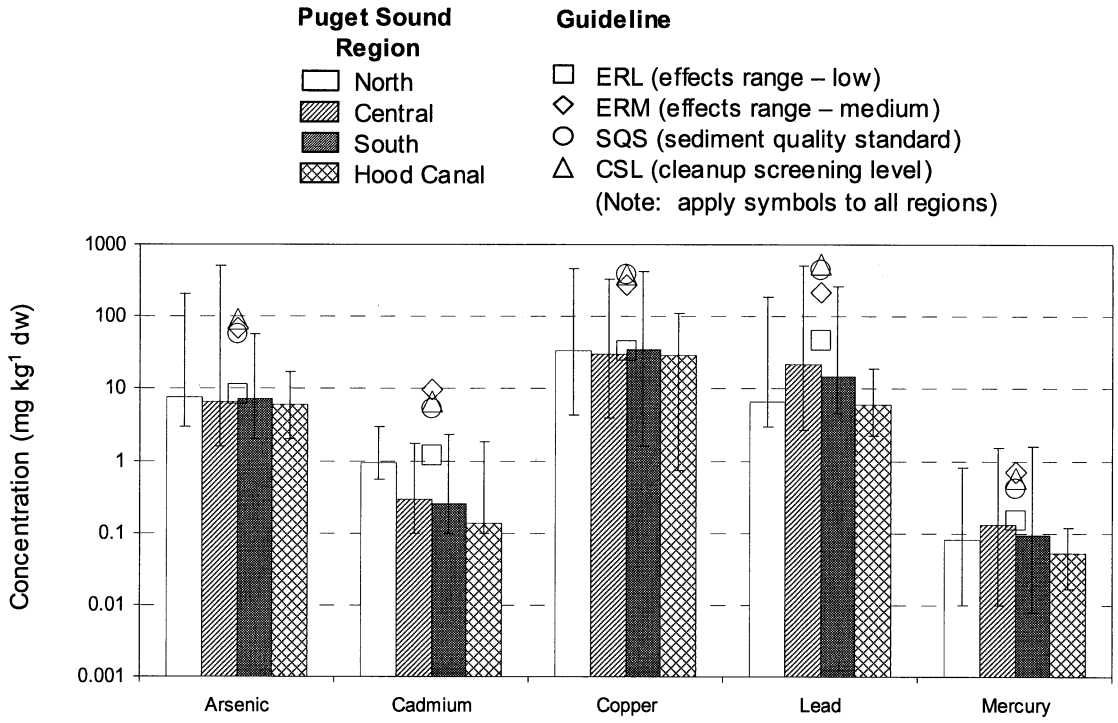
    b. Effects range median (ERM): The ERM is the median, or 50^{th} percentile, or the effects data for a chemical. Concentrations above the ERM represent a probable effects range, where effects would frequently occur.

Because several chemical and physical factors affect the bioavailability of metals, total sediment metals concentrations are generally not predictive of the toxic effects of these trace elements (Adams et al. 1992; Burton and Scott 1992). As discussed in Section 2, the manifestation of biological effects related to exposure to these contaminants is dependent on several other factors such as bioavailability and interactions between different contaminants (i.e., additive and synergistic behavior). Therefore, the purpose of comparing the available sediment data with sediment quality standards is to highlight areas of potential concern rather than providing an assessment of risk.

4.2.2.1 Metals

The greatest exceedances of either the Sediment Quality Standards (SQS) or the Cleanup Screening Levels (CSL) were for arsenic in North and Central Puget Sound samples, and for mercury in Central Puget Sound samples (Figure 4-5). Concentrations of cadmium, copper and lead were either at or below the guidelines for all four regions. The median concentration was usually one order of magnitude lower than the sediment standards, indicating that a substantial number of samples had low metals concentrations. When compared to the NOAA guidelines, particularly the lower of the two (i.e., ERL) there were exceedances for all metals in all four areas.

Figure 4-5: Summary of metal concentrations in North, Central and South Puget Sound and Hood Canal sediments compared to sediment quality guidelines. Columns are median concentrations in mg kg^{-1} dw and error bars show range.



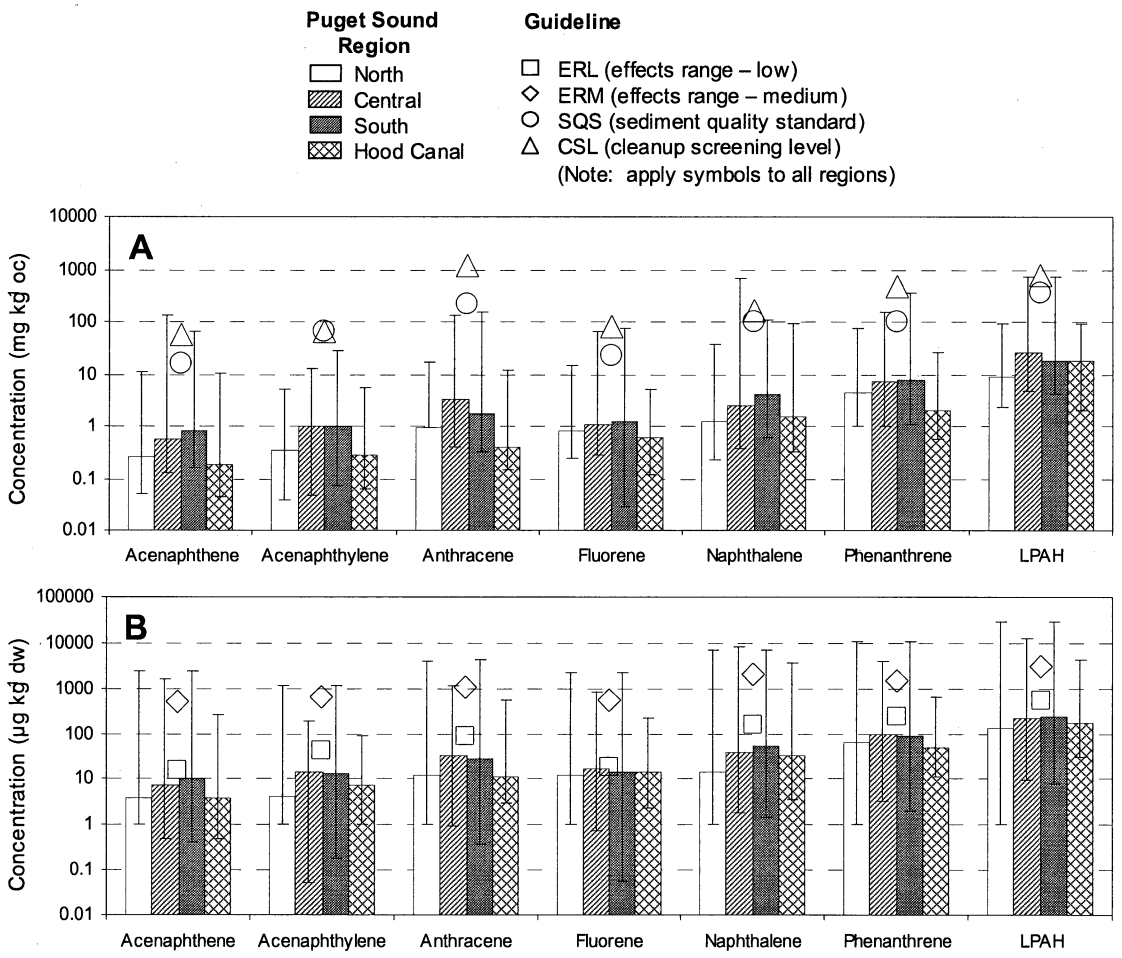
Source: PSAMP-NOAA (2002)

Currently, Washington State does not have a sediment quality guideline for TBT. Meador et al. (2002), however, have recommended a sediment TBT concentration of 6,000 ng g^{-1} oc (i.e., organic carbon normalized) for the protection of salmonid prey species, primarily benthic invertebrates. This recommended criterion was not exceeded in any of the Puget Sound samples.

4.2.2.2 Organics

Both low and high molecular weight polycyclic aromatic hydrocarbons were detected in almost all of the samples from North, Central and South Puget Sound and Hood Canal. However, only about 20% of the samples had concentrations exceeding either the SQS or CSL (Figures 4-6A and 4-7A). A greater number of samples from all four areas exceeded the NOAA sediment guidelines, which are based on dry weight concentrations rather than organic-carbon normalized concentrations (Figures 4-6B and 4-7B). The median concentration for most of the parameters was at or just below the ERL, while the maximum concentrations of most parameters exceeded the ERM.

Figure 4-6: Summary of LPAH concentrations in Puget Sound sediments compared to sediment quality guidelines (**A**: organic-carbon normalized data compared to SQS and CSL; **B**: dry weight data compared to ERL and ERM). Columns are median concentrations, with range indicated by error bars.

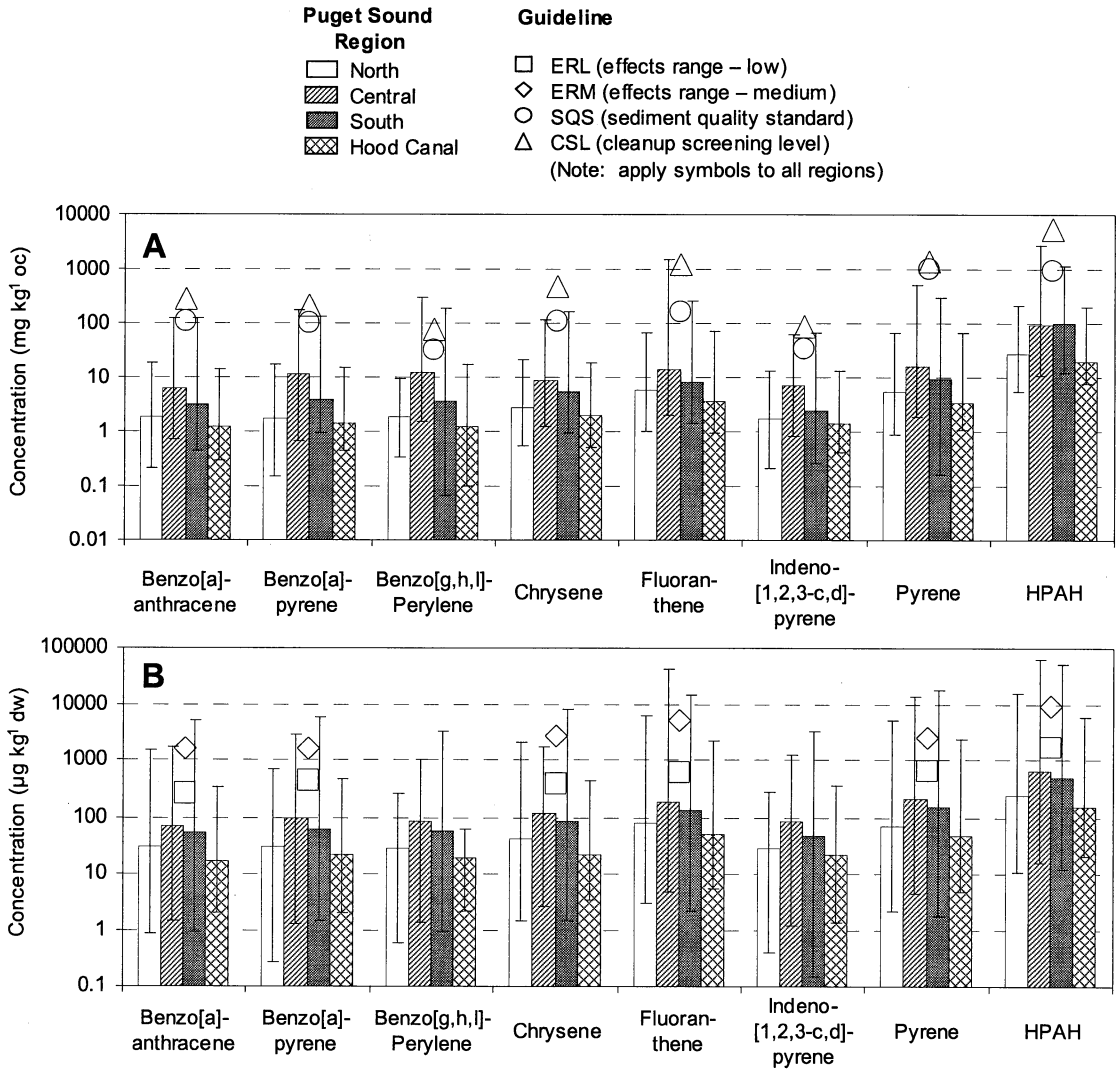


Source: PSAMP-NOAA (2002)

The median organic carbon-normalized concentrations of all PAH compounds were one to two orders of magnitude lower than any of the sediment quality values (SQVs). The regional pattern of SQV exceedances is generally consistent with the pattern of bulk chemistry concentrations shown in Tables 4-4, 4-5, 4-6 and 4-7. For example, the greatest concentration of naphthalene was measured in a sample from Central Puget Sound as was the greatest exceedance of SQVs. In the case of other PAHs (i.e., fluorene), however, the highest concentrations were in South and Central Puget Sound sediments, while Central Puget Sound sediments exceeded the SQV to the same degree. This may be due to the relative total organic carbon (TOC) content in the sediments of the three regions. Total organic carbon (TOC) concentrations in Central Puget Sound (mean ~ 1.4%; max ~ 4.2%) were lower than in either North (mean ~ 1.9%; max ~9.9%) or Sound Puget Sound

(mean ~1.8%; max ~7.9%). Washington State's SQVs for organic contaminants are set on a carbon-normalized basis, as the presence of organic carbon affects the bioavailability of compounds such as PAHs.

Figure 4-7: Summary of HPAH concentrations in Puget Sound sediments compared to sediment quality guidelines. (**A**: organic-carbon normalized data compared to SQS and CSL; **B**: dry weight data compared to ERL and ERM). Columns are median concentrations, with range indicated by error bars.

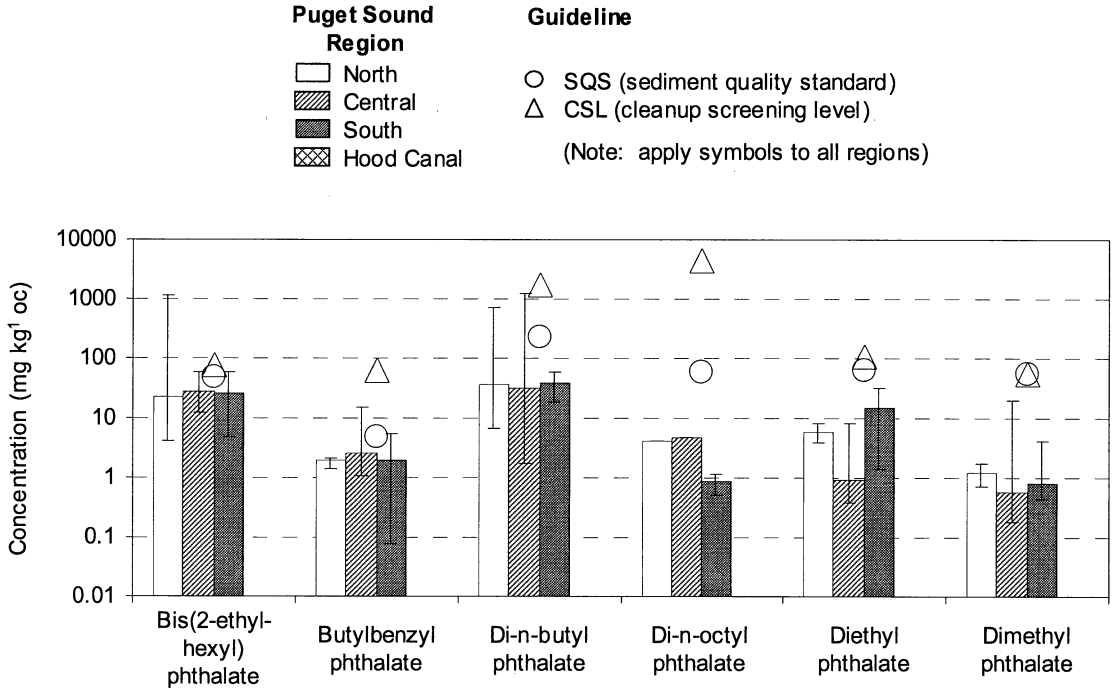


Source: PSAMP-NOAA (2002)

Phthalate ester compounds were detected in 1 to 30% of the samples, while fewer than 5% occurred at concentrations exceeding the SQS. The SQS were exceeded for bis(2-ethylhexyl) phthalate, butylbenzyl phthalate and di-n-butyl phthalate only (Figure 4-8). TOC content of the sediments appeared to have the same effect on the regional pattern of

bulk chemistry versus actual SQV exceedances, as was observed for PAHs. For example, while overall concentrations of butylbenzyl phthalate were highest in South Puget Sound sediments, Central Puget Sound sediment concentrations exceeded the SQS in a greater number of samples and to a greater degree (on a carbon-normalized basis).

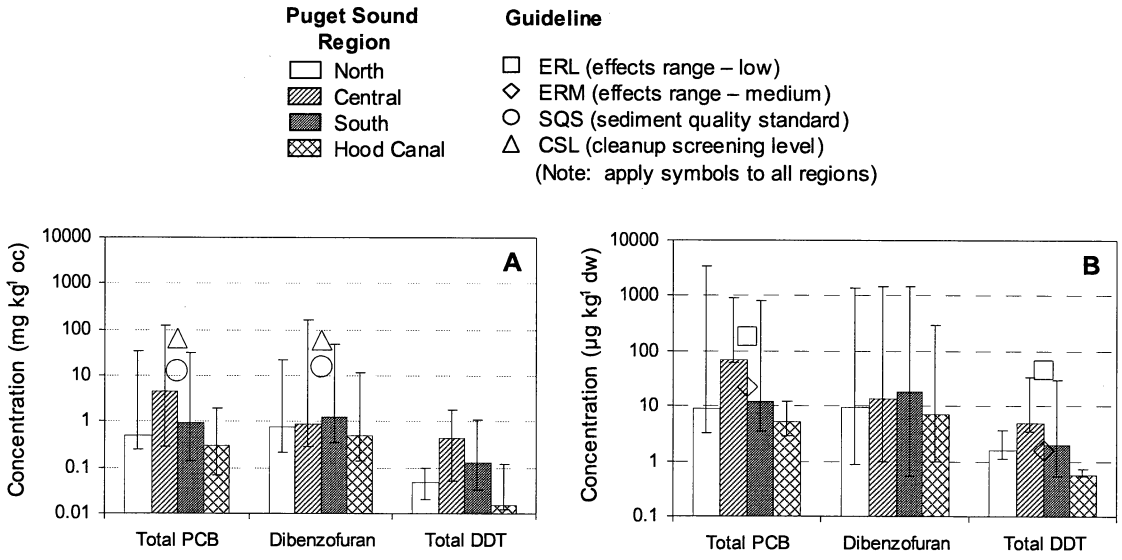
Figure 4-8: Summary of phthalate ester concentrations in Puget Sound sediments compared to sediment quality guidelines. Columns are median carbon-normalized concentrations in mg kg^{-1} oc and error bars show range. (Note: all Hood Canal samples below MDL).



Source: PSAMP-NOAA (2002)

The highest PCB, dibenzofuran and DDT concentrations were generally found in North and South Puget Sound. However, there were relatively few exceedances of the SQVs in these two regions (Figure 4-9A). In comparison, while Central Puget Sound had similar or lower concentrations of the three compounds than the other regions, there were considerably more exceedances of the SQVs; as many as half of the samples with detected PCB concentrations and almost all of the samples with detected DDT/DDT metabolites concentrations exceeded the SQV. The PCB concentrations in some Central Puget Sound samples also exceed the CSL, which is the less-protective standard. These results again illustrate the importance of organic carbon content in affecting the bioavailability of sediment contaminants. Figure 4-9B shows the same data as raw bulk sediment concentrations (i.e., not OC-normalized) compared to the NOAA sediment benchmarks. The general pattern of PCB, dibenzofuran and DDT concentrations on a regional basis was the same for the dry weight data, however, there were a greater number of benchmark exceedances for total PCB.

Figure 4-9: Summary of total PCB, dibenzofurans and total DDT concentrations in Puget Sound sediments compared to sediment quality guidelines (**A**: organic-carbon normalized data compared to SQS and CSL; **B**: dry weight data compared to ERL and ERM). Columns are median concentrations, with range indicated by error bars.



Source: PSAMP-NOAA (2002)

As of 2001, WDOE had identified 112 contaminated sediment cleanup sites in Puget Sound, covering an area of 3,400 acres (WDOE 2001c). Twenty-two of these sites have been cleaned up or require no remedial action, eleven sites are currently undergoing remediation, and an additional 79 require clean-up (65 of which are in the investigation and design phases of cleanup).

4.2.3 Spatial Trends in Sediment Quality

4.2.3.1 Metals

Figures 4-10 through 4-15 illustrate the spatial distribution of metals concentrations in Puget Sound (based on the recent PSAMP-NOAA sampling program). Metals concentrations were compared to the Washington State SQS in order to provide some context to the measured values, with the exception for TBT for which there is no standard.

The highest concentration of all metals were found in the major urban/industrial bays such as Elliott Bay in Seattle, Commencement Bay and its waterways in Tacoma, Everett Harbor, Bellingham Bay and Sinclair Inlet. An unusually high mercury concentration was reported for a sample from Totten Inlet.

Lead concentrations measured in the recent PSAMP-NOAA program are shown together with data from SEDQUAL in Figure 4-13 to provide an example of the effects of source control on environmental contaminants. Historic data show that lead in samples from the major urban/industrial embayments often exceeded the SQS, whereas no recent samples exceeded the sediment standard.

4.2.3.2 Organics

Figures 4-16 through 4-20 illustrate the spatial distribution of organic contaminant concentrations in Puget Sound (based on the recent PSAMP-NOAA sampling program). Where possible concentrations were compared to the Washington State SQS or the NOAA ERL in order to provide some context to the measured values.

As with metals, the highest concentrations of organic contaminants were generally found in the urban/industrial areas of Elliott Bay, Commencement Bay and its waterways, Everett Harbor and Sinclair Inlet. Outside of these areas, organic contaminants were largely non-detectable.

Figure 4-10: Spatial distribution of arsenic in Puget Sound based on PSAMP-NOAA 1997-1999 data.

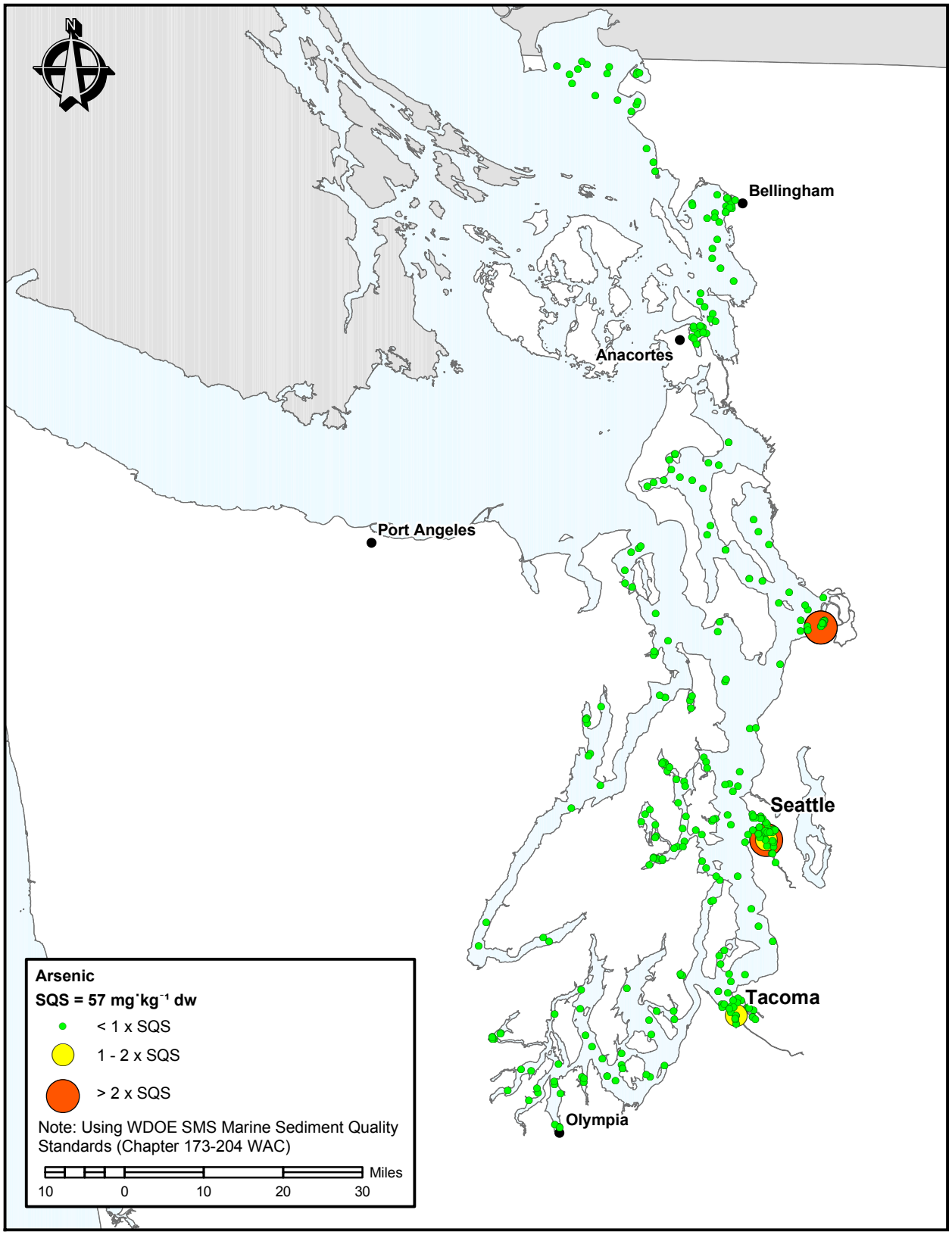


Figure 4-11: Spatial distribution of cadmium in Puget Sound based on PSAMP-NOAA 1997-1999 data.

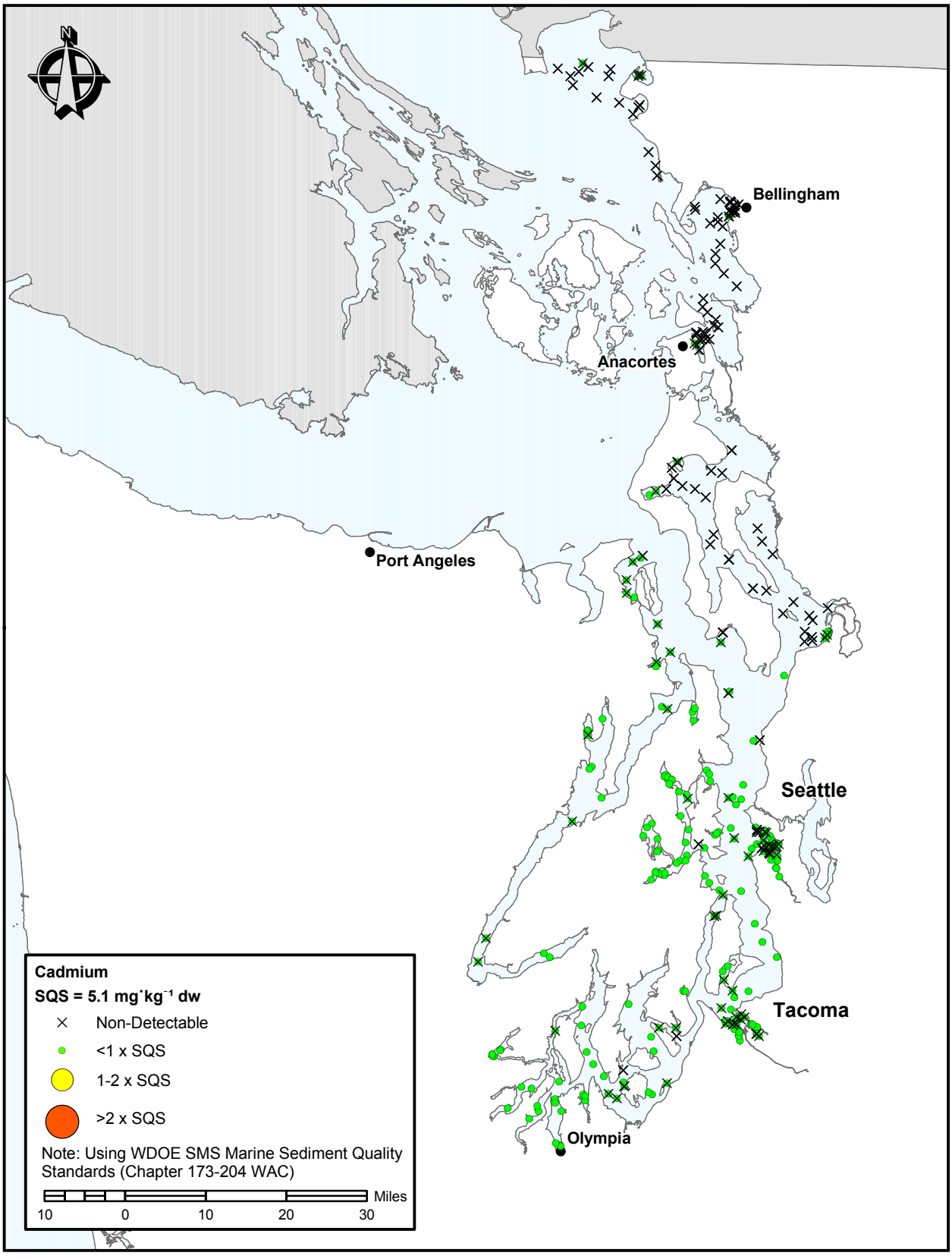


Figure 4-12: Spatial distribution of copper in Puget Sound based on PSAMP-NOAA 1997-1999 data.

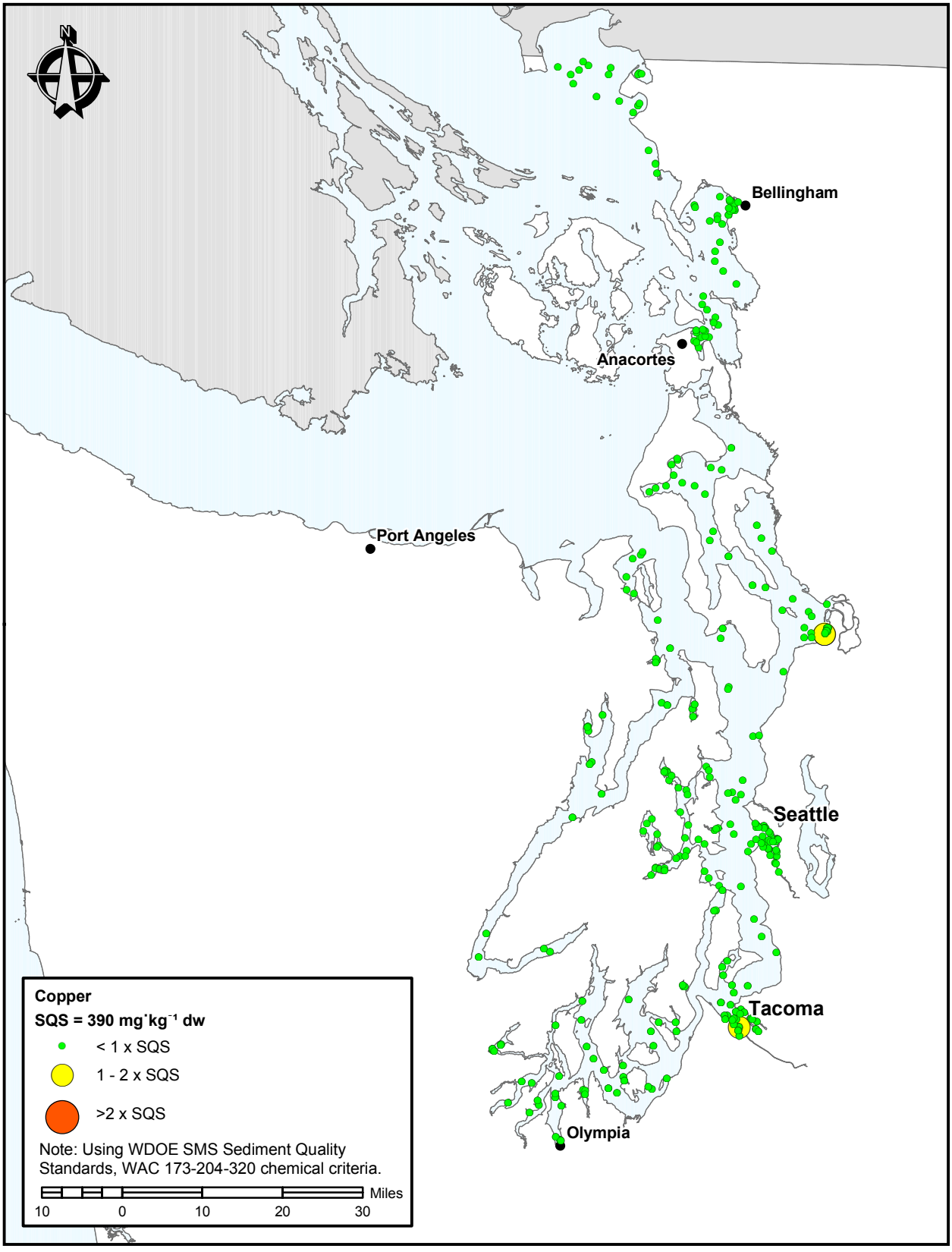


Figure 4-13: Spatial distribution of lead in Puget Sound based on PSAMP-NOAA 1997-1999 data and historical data from SEDQUAL.

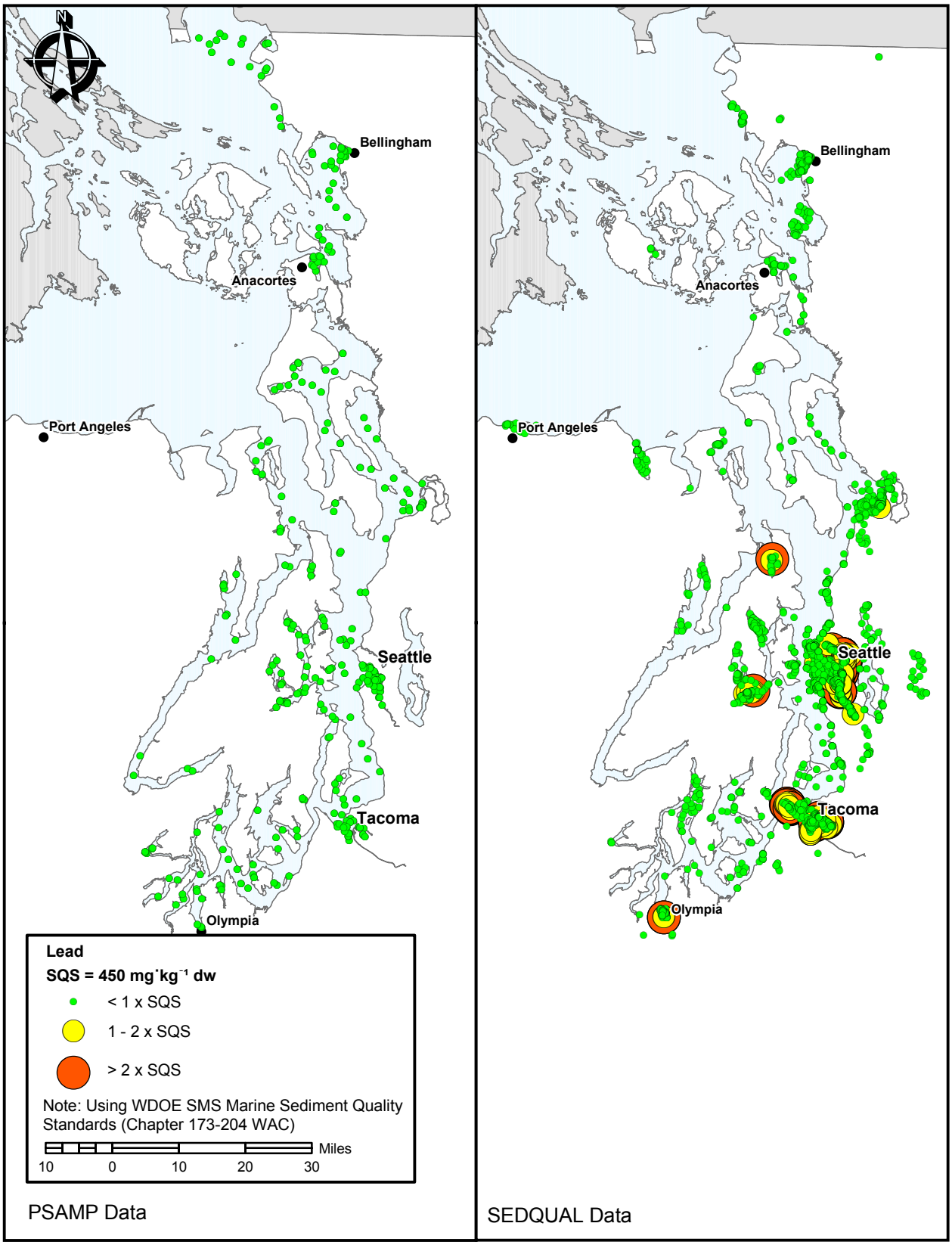


Figure 4-14: Spatial distribution of mercury in Puget Sound based on PSAMP-NOAA 1997-1999 data.

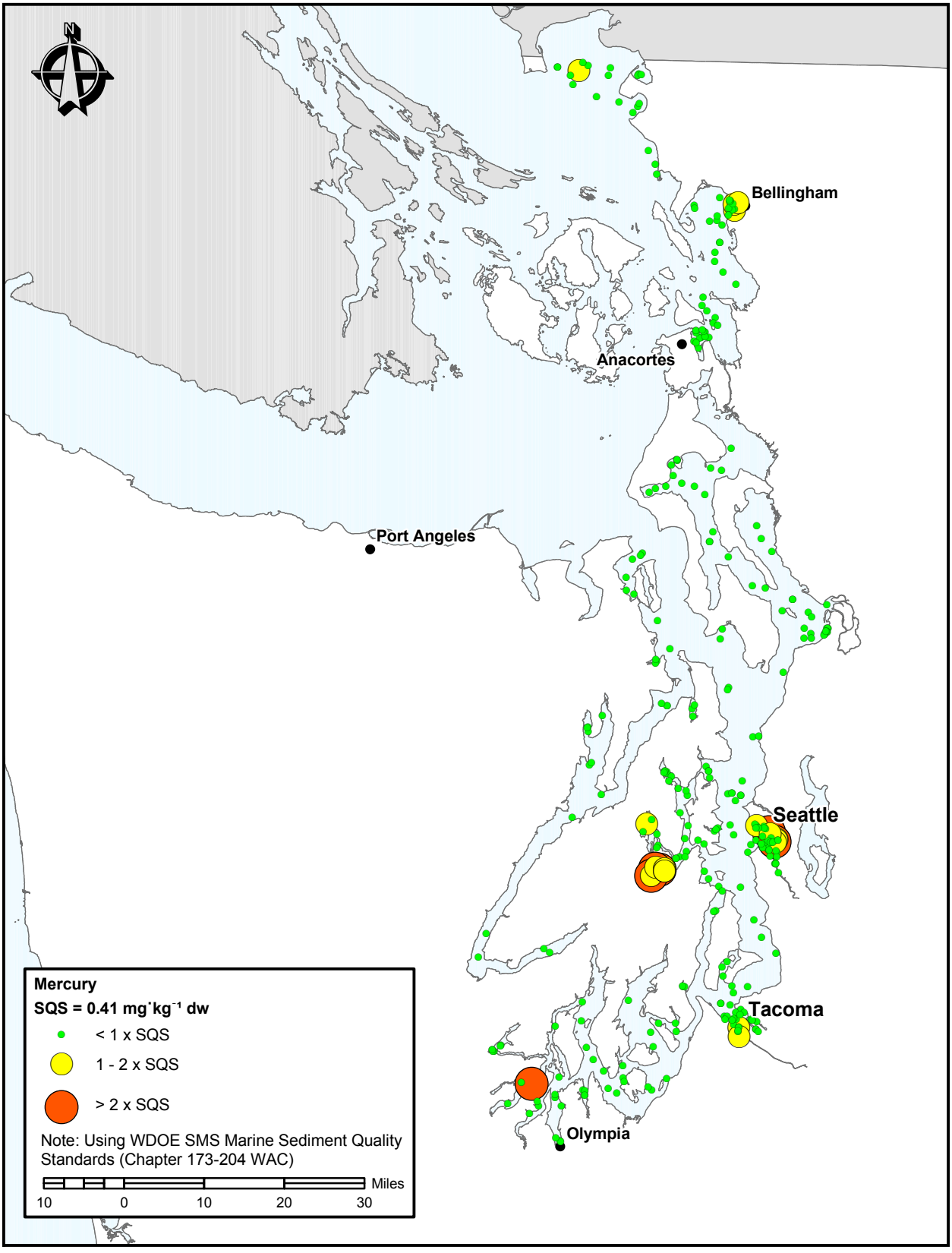


Figure 4-15: Spatial distribution of tributylin in Puget Sound based on PSAMP-NOAA 1997-1999 data.

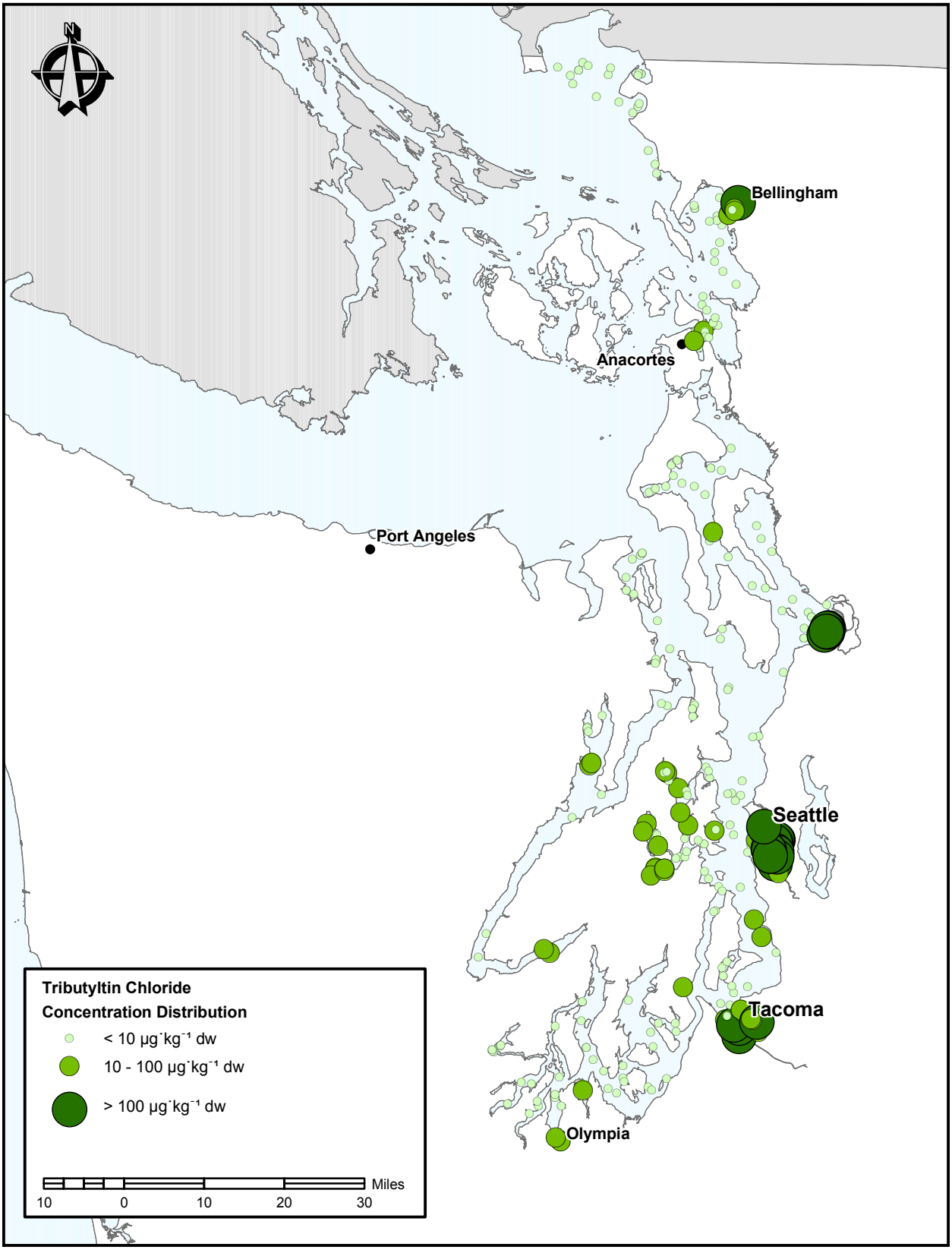


Figure 4-16: Spatial distribution of Total PCB in Puget Sound based on PSAMP-NOAA 1997-1999 data.

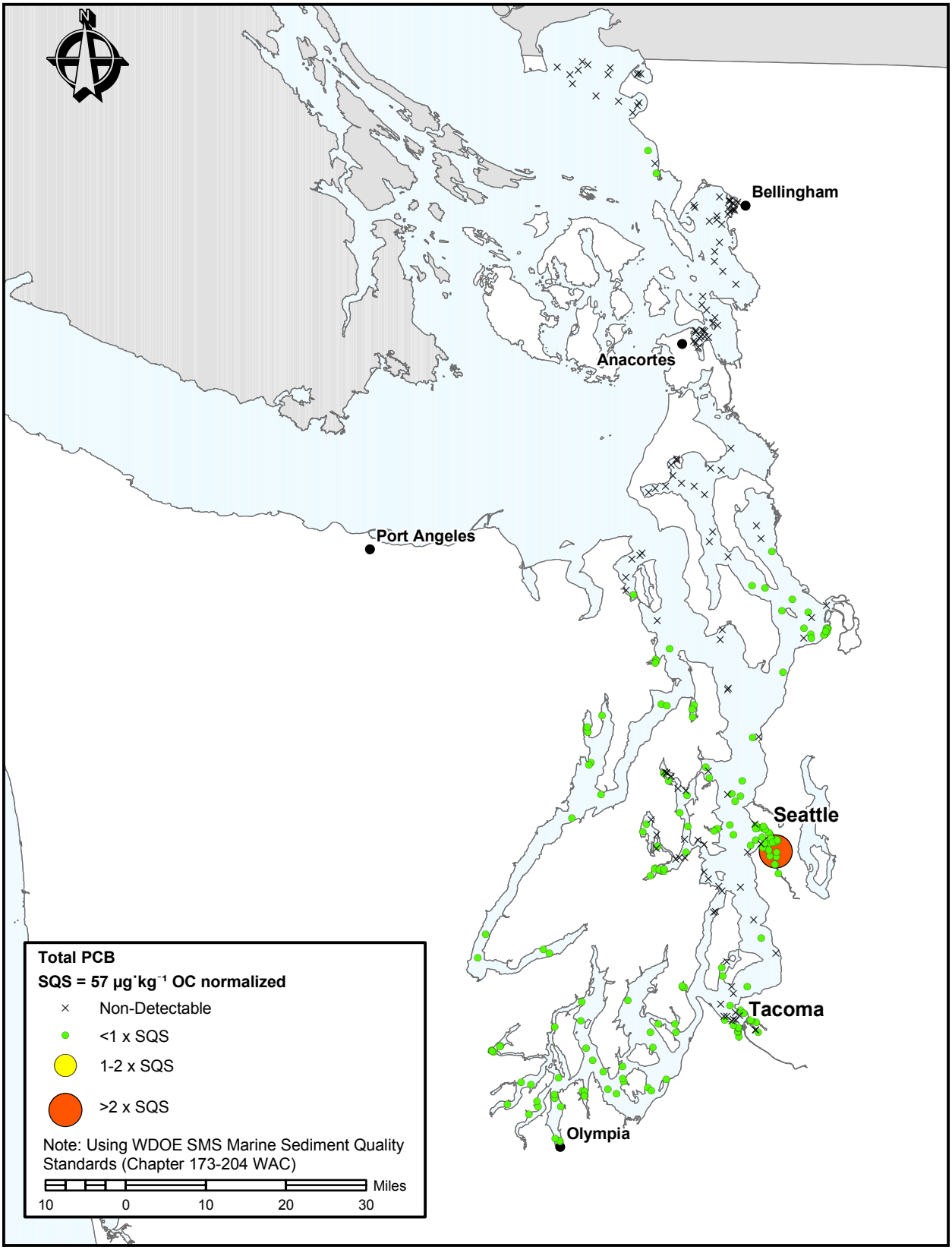


Figure 4-17: Spatial distribution of DDT in Puget Sound based on PSAMP-NOAA 1997-1999 data.

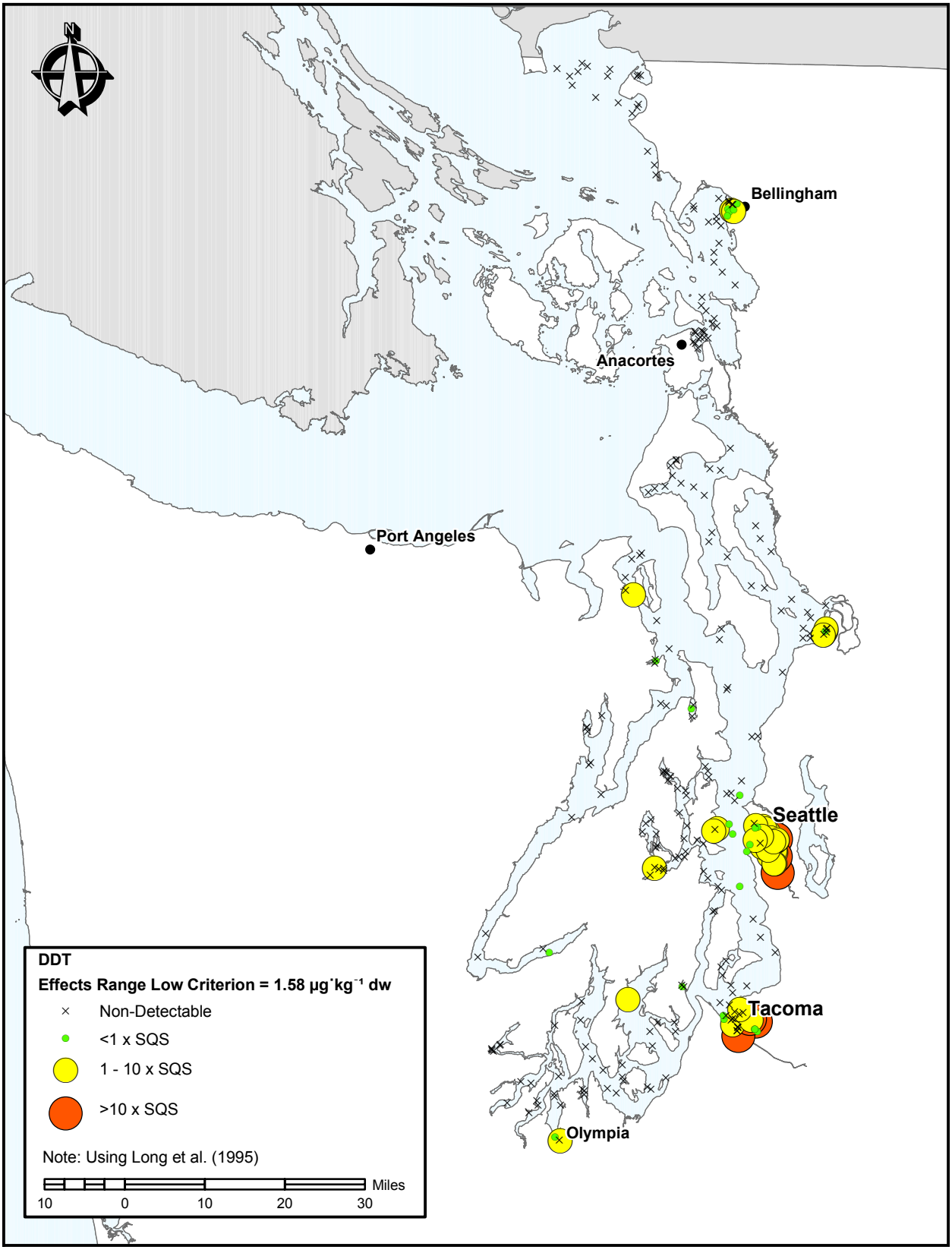


Figure 4-18: Spatial distribution of dibenzofuran in Puget Sound based on PSAMP-NOAA 1997-1999 data.

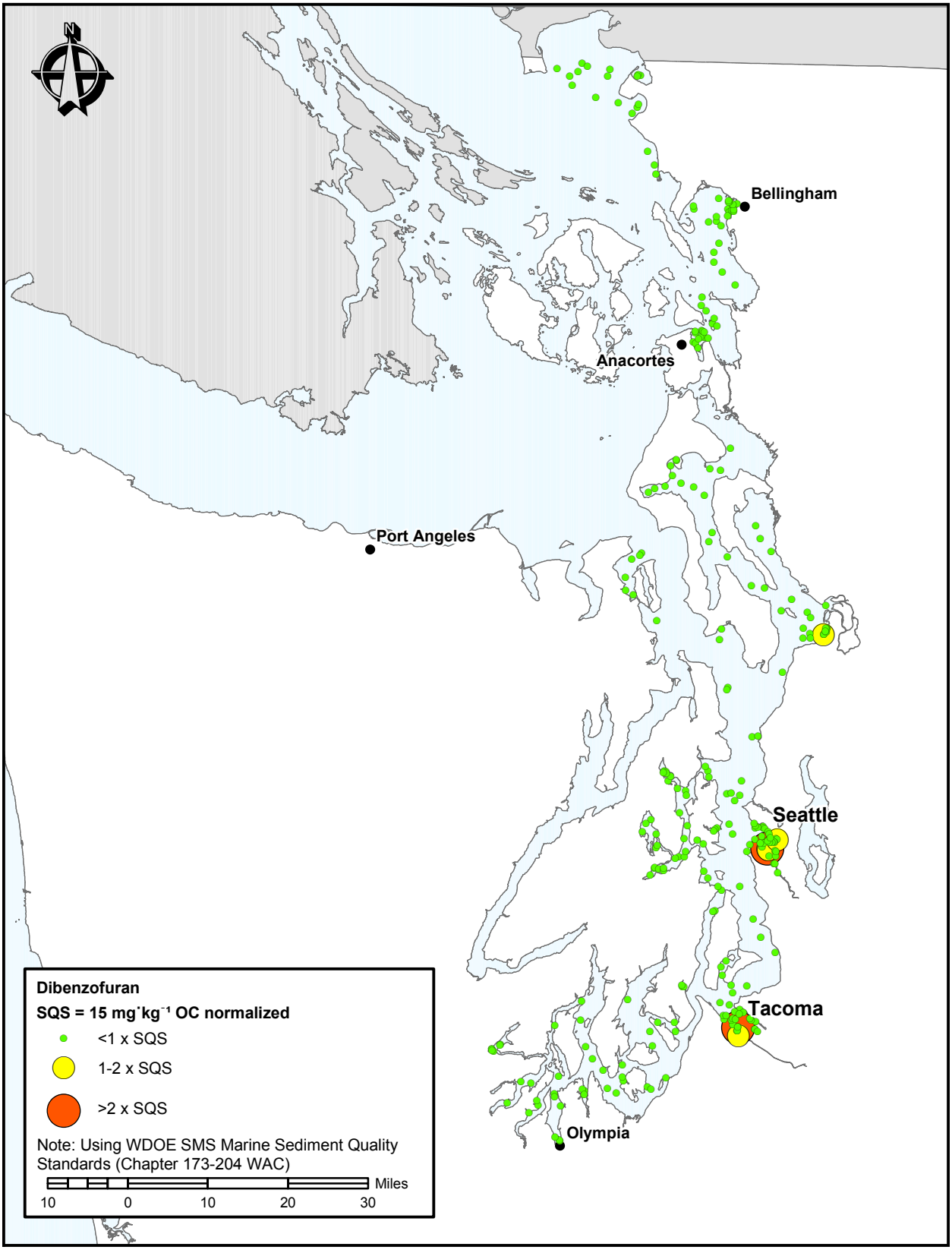


Figure 4-19: Spatial distribution of total PAHs in Puget Sound based on PSAMP-NOAA 1997-1999 data.

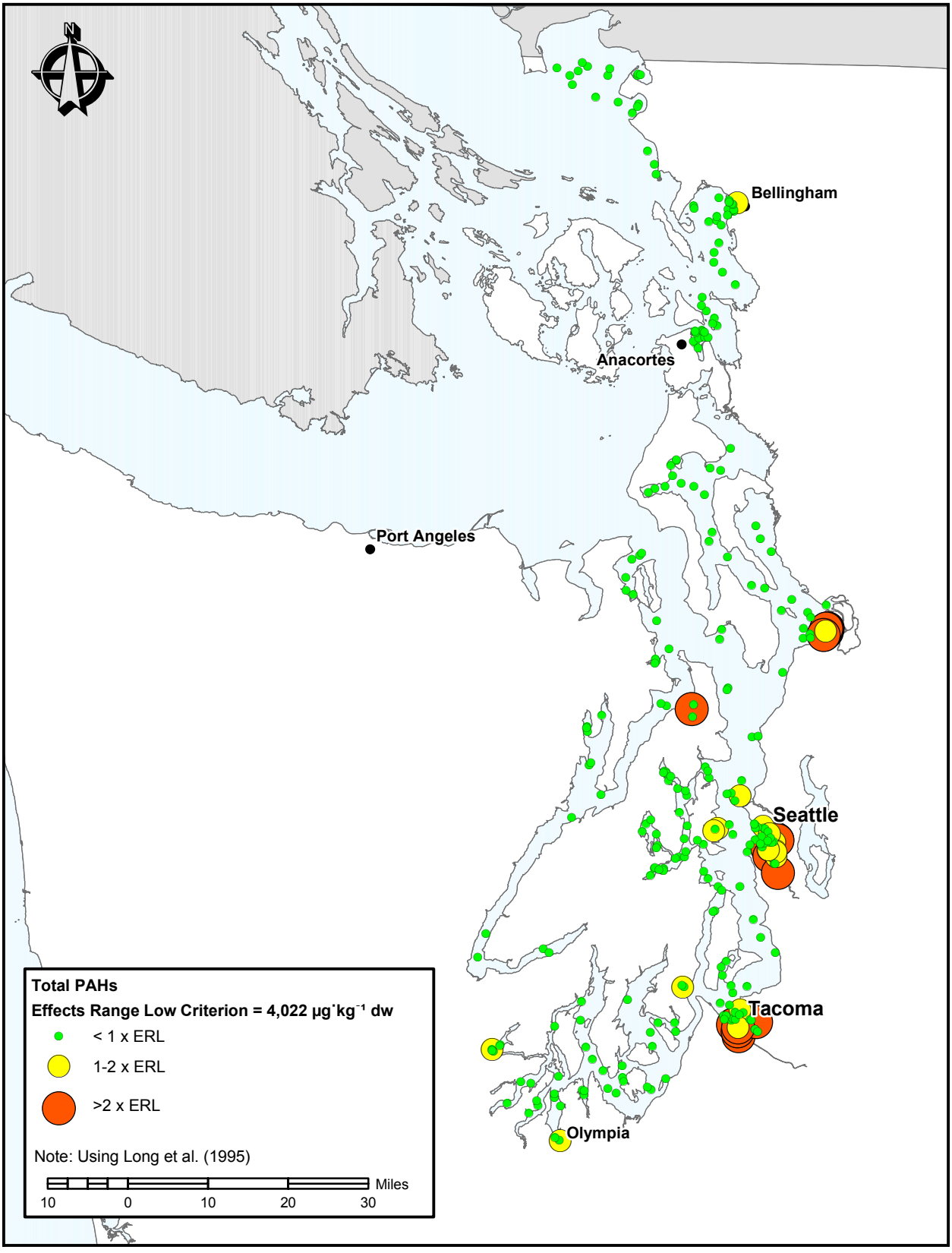
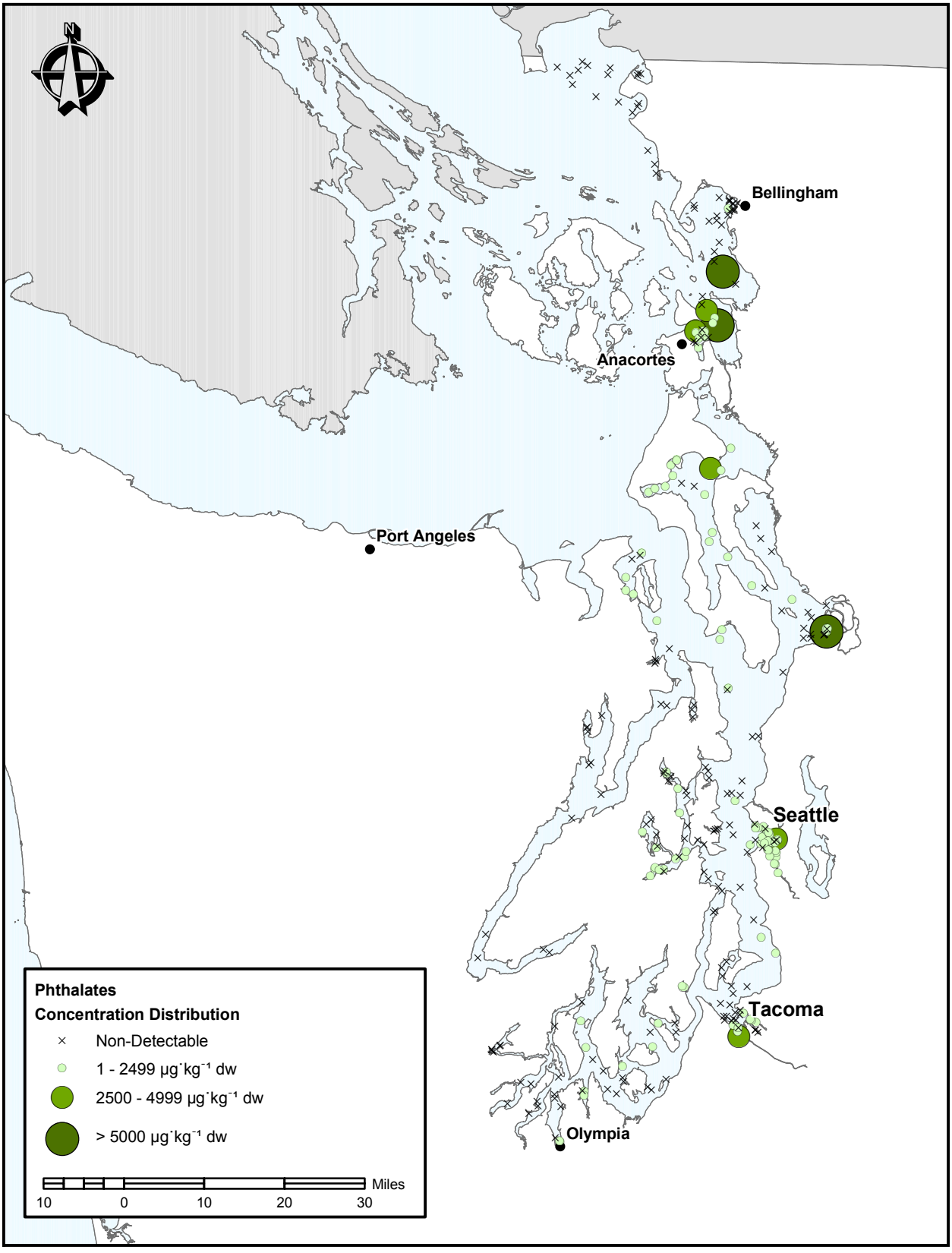


Figure 4-20: Spatial distribution of total phthalates in Puget Sound based on PSAMP-NOAA 1997-1999 data.



Overall, sediments from urban-industrial areas exhibited the highest sediment contaminant concentrations, the greatest toxicity in laboratory tests, and the most highly altered benthic communities (Long et al. 1999, 2000, 2002). This is consistent with the locations of sediment clean up sites in Puget Sound (Table 4-9; Figure 3-4).

Table 4-9: Sediment cleanup sites in Puget Sound identified by the WDOE Toxics Cleanup Program as of 2001.

WATERBODY[a]	NUMBER OF SITES
North Puget Sound	
Bellingham Bay	9
Everett/Port Gardner	9
Fidalgo Bay	6
Central Puget Sound	
Bremerton/Kitsap Inlets	11
Elliott Bay	19
Duwamish River/Waterway	8
South Puget Sound	
Commencement Bay and its waterways	12

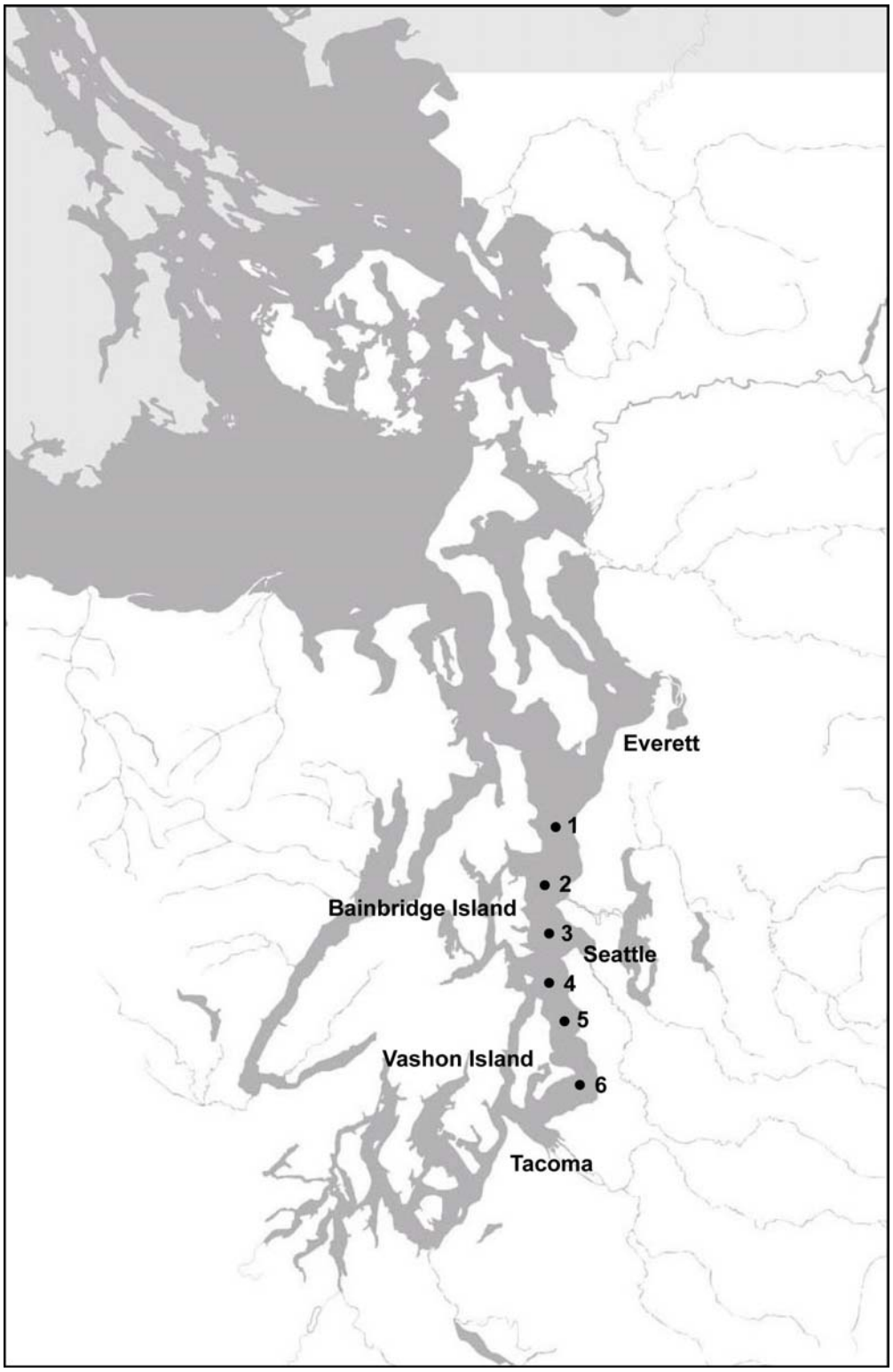
Source: WDOE (2001c)
[a] only waterbodies with more than 3 cleanup sites are listed

4.2.4 Temporal Trends in Sediment Quality

Temporal trends in sediment quality were assessed using two data sets. The first data set was for sediment core samples were collected from six locations in Central Puget Sound (Figure 4-21) as part of NOAA's National Status and Trends Program. Approximately 25 2-cm sections were cut from each core and analyzed for metals, PAH, PCB and chlorinated pesticides, nutrients, butyltins and grain size (Lefkovitz et al. 1997). The sections were also age-dated to assist in determining historical trends in chemical composition of the sediments. The sediment core data is useful for determining historical trends in the deep sediments of Puget Sound.

The second data set was for PSAMP's Marine Sediment Monitoring Program, which has sampled 76 fixed and rotational stations throughout Puget Sound between 1989 and the present. While the sampling stations are located away from known contaminant sources, the data provide a modern and broader geographical perspective regarding sediment contaminant trends. The data represent surface conditions in each year.

Figure 4-21: Location of sediment core sampling stations in Puget Sound (Lefkovitz et al. 1997)



4.2.4.1 Metals

Arsenic concentrations peaked around 1965, decreased until 1970 and have remained stable since (Figure 4-22). Arsenic concentrations were highest in Core 5 located between Tacoma and Seattle, compared to a core off Elliott Bay, and the period of arsenic enrichment coincides with the operation of the ASARCO smelter in Tacoma between 1890 and 1986 (Yake 2001). The elevated arsenic concentrations measured in Core 6 in East Passage indicate that sediment contaminants were likely being distributed northward via advection (i.e., horizontal movement of water). Mercury concentrations in the sediment cores peaked around 1960 and have fluctuated widely since then rather than stabilizing. The temporal and spatial pattern of mercury concentrations is similar to that of arsenic and may also be related to the operation of the ASARCO smelter (Yake 2001). Mercury is the only chemical parameter measured by Lefkovitz et al. (1997) to exceed sediment quality guidelines. Lead also peaked around 1965 and decreased substantially starting at the time lead was banned as a gasoline additive (Yake 2001). Despite the apparent decreases in these three metals, concentrations remain higher than pre-1890. Cadmium and copper did not appear to become as enriched in the area of the sediment core, but have been consistently higher than pre-1890 concentrations.

Four cores were sampled for butyltins, however, all results were lower than the detection limit and therefore not profiled (Lefkovitz et al. 1997).

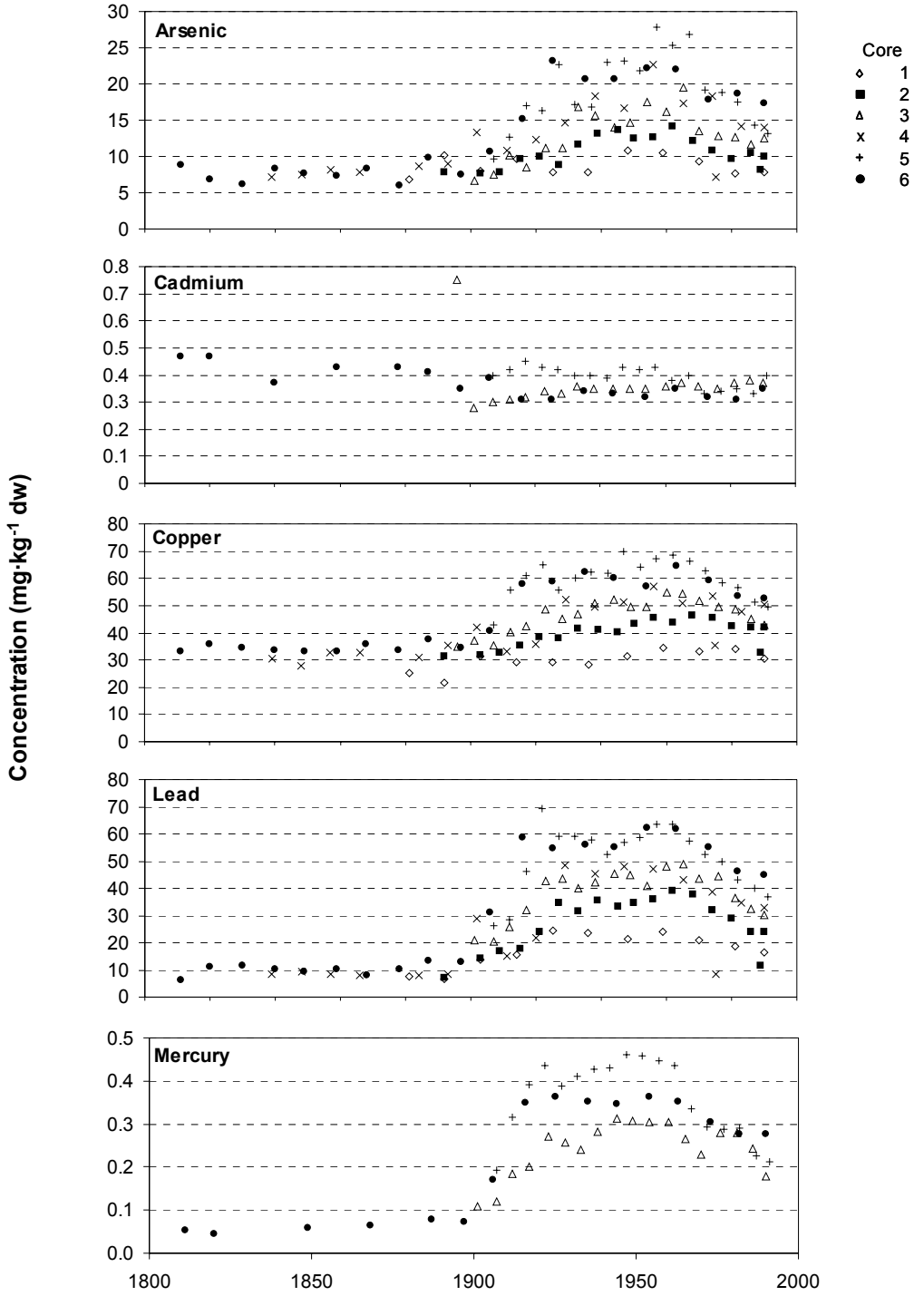
Sediment chemistry has been variable between 1989 and 2001 throughout Puget Sound although there appears to be a general downward trend for all metals at stations like Sinclair Inlet (Figure 4-23).

4.2.4.2 Organics

The highest total PCB concentrations measured in the core sediments was ~35 $\mu g \cdot kg^{-1}$ dw for the Elliott Bay site around 1960. Unlike the metal parameters, organic contaminants in the sediment cores tended to show a south to north increase in concentrations (Figure 4-24). Since the EPA restriction on production and use of PCBs in 1977, sediment concentrations have steadily decreased. Total DDT concentrations also peaked around 1960, coinciding with peak production in the U.S., then decreased until 1980 when there was a temporary upward spike. At the coring station off Seahurst Park between Tacoma and Seattle, the sudden increase in DDT concentration was much larger and exceeded the 1960s peak by 2 $\mu g \cdot kg^{-1}$ dw. Total PAH concentrations were greatest in the 1940s, and were likely related to coal-burning at the time (Yake 2001).

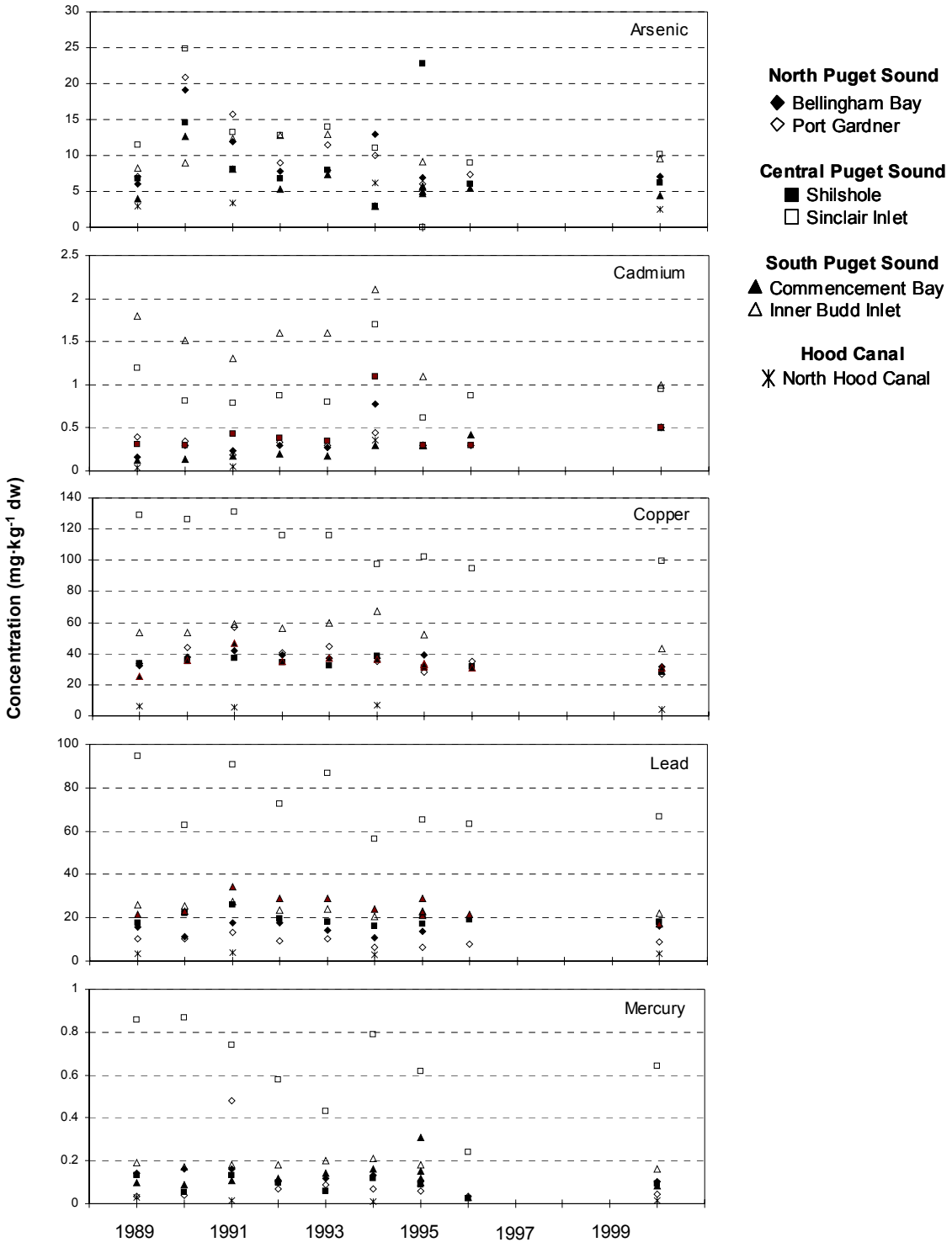
Figure 4-25 shows the recent trends in total PCBs and total PAHs. Both organic parameters appear to be stable at most stations. Exceptions include a peak in PCB concentrations in Sinclair Inlet sediments, and high year to year variability in PAH concentrations in Commencement Bay. DDTs and dieldrin were not detected in any samples from any stations between 1989 and 2001.

Figure 4-22: Trends in the concentrations of selected metals in Puget Sound sediment core samples from 1880 to 1990. Sediment core sampling locations are shown in Figure 4-21.



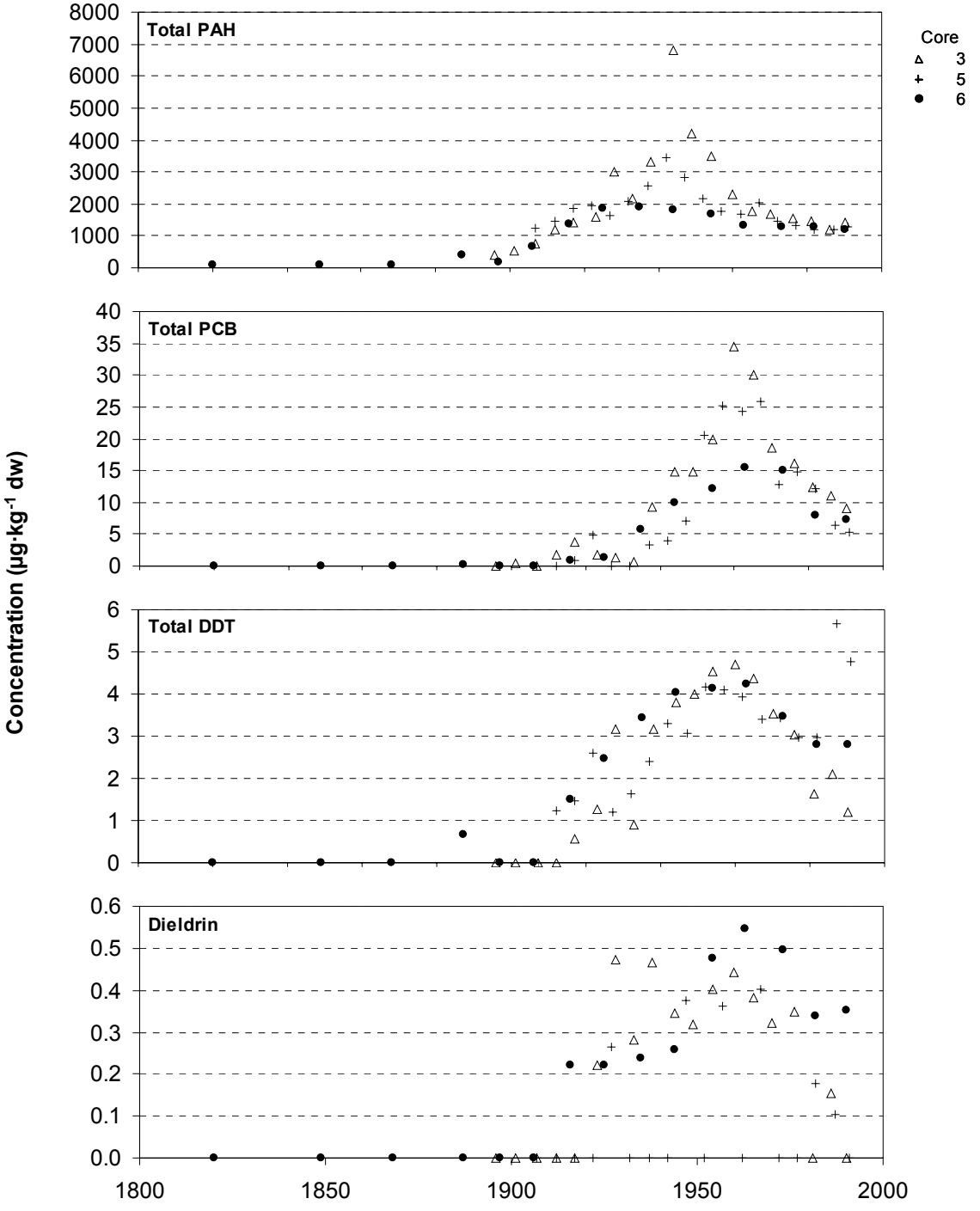
Source: Lefkovitz et al. (1997)

Figure 4-23: Trends in the concentrations of selected metals in surficial sediments at selected MSMP stations in Puget Sound from 1989 to 2001.



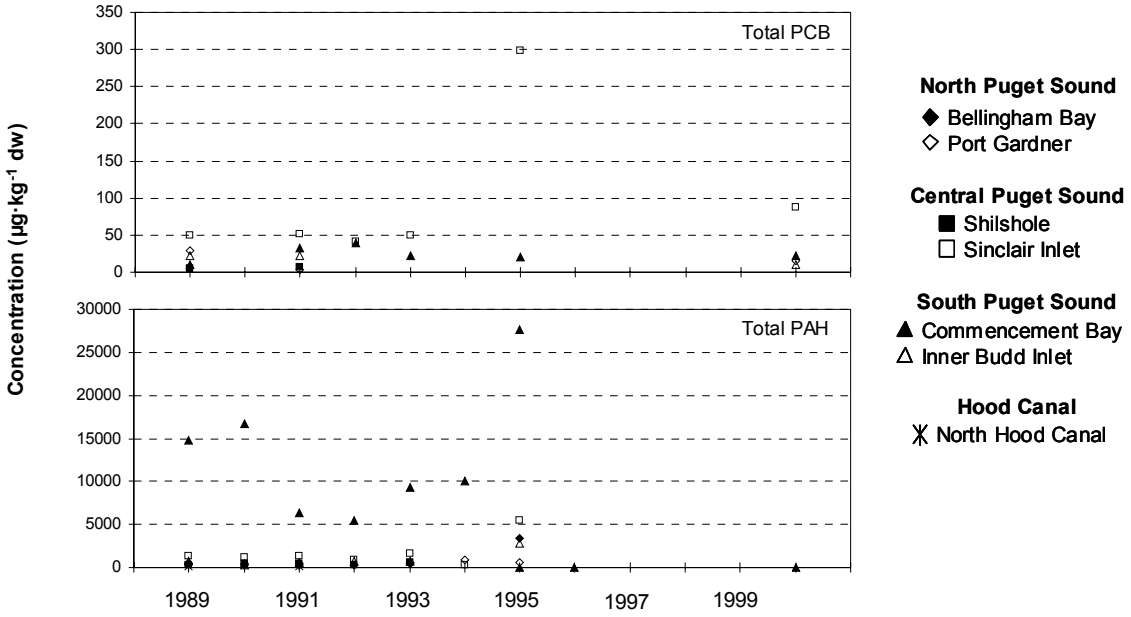
Source: PSAMP (2002)

Figure 4-24: Trends in concentrations of selected organic contaminants in Puget Sound sediment core samples from 1880 to 1990. Sediment core sampling locations are shown in Figure 4-21.



Source: Lefkovitz et al. (1997)

Figure 4-25: Trends in the concentrations of selected organic contaminants in surficial sediments at selected MSMP stations in Puget Sound from 1989 to 2001.



Source: PSAMP (2002)

4.2.5 Sediment Summary

4.2.5.1 State of Knowledge

A recent (1997-1999) Puget Sound-wide sediment sampling program measuring bulk sediment chemistry, laboratory toxicity and benthic community structure indicated that a majority of the Sound has healthy surficial sediments. The discharge of certain chemicals into the Puget Sound environment have decreased as a result of source control efforts (e.g., removal of lead from gasoline), and significant cleanup efforts have been conducted at contaminated sediment sites in many of the Sound's urban/industrial embayments. However, sediment contaminants continue to be a problem in some areas.

Sediment data collected in the last decade indicate that:

- The highest contaminant levels are located in hot spots such as urban/industrial harbors and near municipal and industrial effluent discharges, with a general trend of more contaminants in the central basin;
- Historically contaminated sediments continue to be a source of persistent bioaccumulative toxics, such as PCB, to the marine food chain;

- Laboratory toxicity tests are not always predictive of effects on benthic invertebrate communities related to sediment contaminants;
- The presence/absence of benthic organisms can be an indicator of how contaminated sediments are, although sediments where there is a healthy benthic community can still contribute to effects in higher trophic level organisms through food chain transfer of the contaminants;
- There is evidence that source control efforts and *in situ* capping are resulting in decreased PAH sediment concentrations at some cleanup sites (i.e., Eagle Harbor);
- Lead sediment concentrations have decreased significantly as a result of source control efforts.

4.2.5.2 Contaminant Effects

- Sediment PAHs have been implicated in the elevated incidence of liver disease in the English sole of contaminated areas such as the urban/industrial waterways of Elliott Bay and Commencement Bay.

4.2.5.3 Monitoring and Data Gaps

Monitoring and data gaps and other issues related to metals and organic contaminants in the sediments of Puget Sound include the following:

- It is likely that not all historically contaminated sediments have been identified;
- There are no recent sediment core data (Yake 2001; Mearns 2001). As well, cores have only been collected from the central basin of Puget Sound;
- Sediment data are not consistently entered into a single database, rather, they may be entered into SEDQUAL or a national database maintained by NOAA, or may not be entered at all;
- Background concentrations of some contaminants such as PCB have not been characterized sufficiently;
- Current laboratory methods in use for some compounds may have higher method detection limits than environmental concentrations of concern;
- There are insufficient data regarding the presence/composition of individual PCB or dioxin congeners in sediments to support human health risk assessments.

4.3 BIOTA

Puget Sound is a physically complex ecosystem with a wide range of habitat types supporting diverse and abundant aquatic life. Contaminants observed in the water column and sediments may exert acute and/or chronic effects on the organisms that come into direct contact with them, either via dermal exposure or dietary intake. The mechanisms of bioaccumulation and biomagnification result in the further trophic transfer of contaminants through the food web. As a result, localized contamination has been observed to have a far-reaching effect on some species.

In order to monitor the effects of contaminants on the biota of Puget Sound, PSAMP and other studies have sampled a variety of organisms for tissue contaminant burdens and evidence of effects (Table 4-10). The results of these monitoring programs and research projects are summarized in the following sections.

Table 4-10: Marine species that have been monitored for tissue burden and effects of toxic contaminants in the Puget Sound environment.

Invertebrates	**Mammals**
Mussels (*Mytilus edulis*)	Harbor seal (*Phoca vitulina*)
Japanese littleneck clam (*Tapes japonica*)	Killer whale (*Orcinus orca*)
Littleneck clam (*Prototheca staminea*)	Gray whale (*Eschrichtius robustus*)
Butter clam (*Saxidomus giganteus*)	River otter (*Lutra canadensis*)
Japanese oyster (*Crassostrea gigas*)	
Dungeness crab (*Cancer magister*)	**Fish**
Graceful crab (*Cancer gracilis*)	English sole (*Pleuronectes vetulus*)
	Starry flounder (*Platichthys stellatus*)
Birds	Rock sole (*Pleuronectes bilineatus*)
Western grebe (*Aechmophorus occidentalis*)	Chinook salmon (*Oncorhynchus tshawytscha*)
Bald eagle (*Haliaeetus leucocephalus*)	Coho salmon (*O. kisutch*)
Double-crested cormorant (*Phalacrocorax auritus*)	Chum salmon (*O. keta*)
Pelagic cormorant (*P. pelagicus*)	Pacific herring (*Clupea pallasi*)
Pigeon guillemot (*Cepphus columba*)	Copper rockfish (*Sebastes caurinus*)
Great Blue Heron (*Ardea herodias*)	Quillback rockfish (*S. maliger*)
Glaucous-winged gull (*Larus glaucescens*)	Brown rockfish (*S. auriculatus*)
Surf Scoter (*Melanitta perspicillata*)	Yelloweye rockfish (*S. ruberrimus*)
	Pacific cod (*Gadus macrocephalus*)

4.3.1 Invertebrates

Invertebrate species in Puget Sound occupy a wide range of habitats and employ a variety of feeding strategies. They may be exposed to contaminants by dermal contact and diet, while other species such as mussels inhabit areas less proximal to contaminated

sediments but may bioaccumulate contaminants from particulate matter in the water column. In the following section, invertebrates will be discussed in two groups:

- benthic infauna (i.e., burrowing bivalve mollusks, polychaete worms and crustaceans as well as other phyla); and
- shellfish (i.e., crustaceans and mollusks that live on the substrate), which are often consumed by humans.

4.3.1.1 Review of Existing Data

Benthic Infauna

Because burrowing species of invertebrates live in direct contact with benthic substrates and are prey items for carnivorous organisms, they play a critical role in the trophic transfer of contaminants up through the food web. However, there do not appear to be any comprehensive studies on toxic contaminant levels in the benthic infauna of Puget Sound. In the absence of such studies, some information can be inferred from the analyses of stomach contents from fish that consume benthic and epibenthic organisms. For example, English sole on the West Coast of North America have been documented to consume arthropods, annelids, echiurids, and mollusks (McCain et al. 2000) while juvenile salmonids inhabiting estuaries consume cumaceans and amphipods in addition to small fish (McCain et al. 1990).

LPAH, HPAH, PCB and DDT concentrations in the stomach contents of juvenile chinook were significantly higher for fish from the Duwamish Waterway and Commencement Bay than from the Nisqually Estuary in 1989, and all but DDT were again significantly higher in the urban waterways in 1990 (Stein et al. 1995; Table 4-11). As discussed in Section 4.3.2, the elevated concentrations in stomach contents were reflected in the fish tissue concentrations of the same contaminants. The data illustrate two main points: (1) the high interannual variability sometimes observed even at highly contaminated sites; and (2) the trophic transfer of toxic contaminants from the sediments to the benthic infauna and then to higher trophic status organisms.

Table 4-11: Concentrations of selected organic contaminants in the stomach contents of juvenile chinook from three different estuaries in Puget Sound (1989-1990).

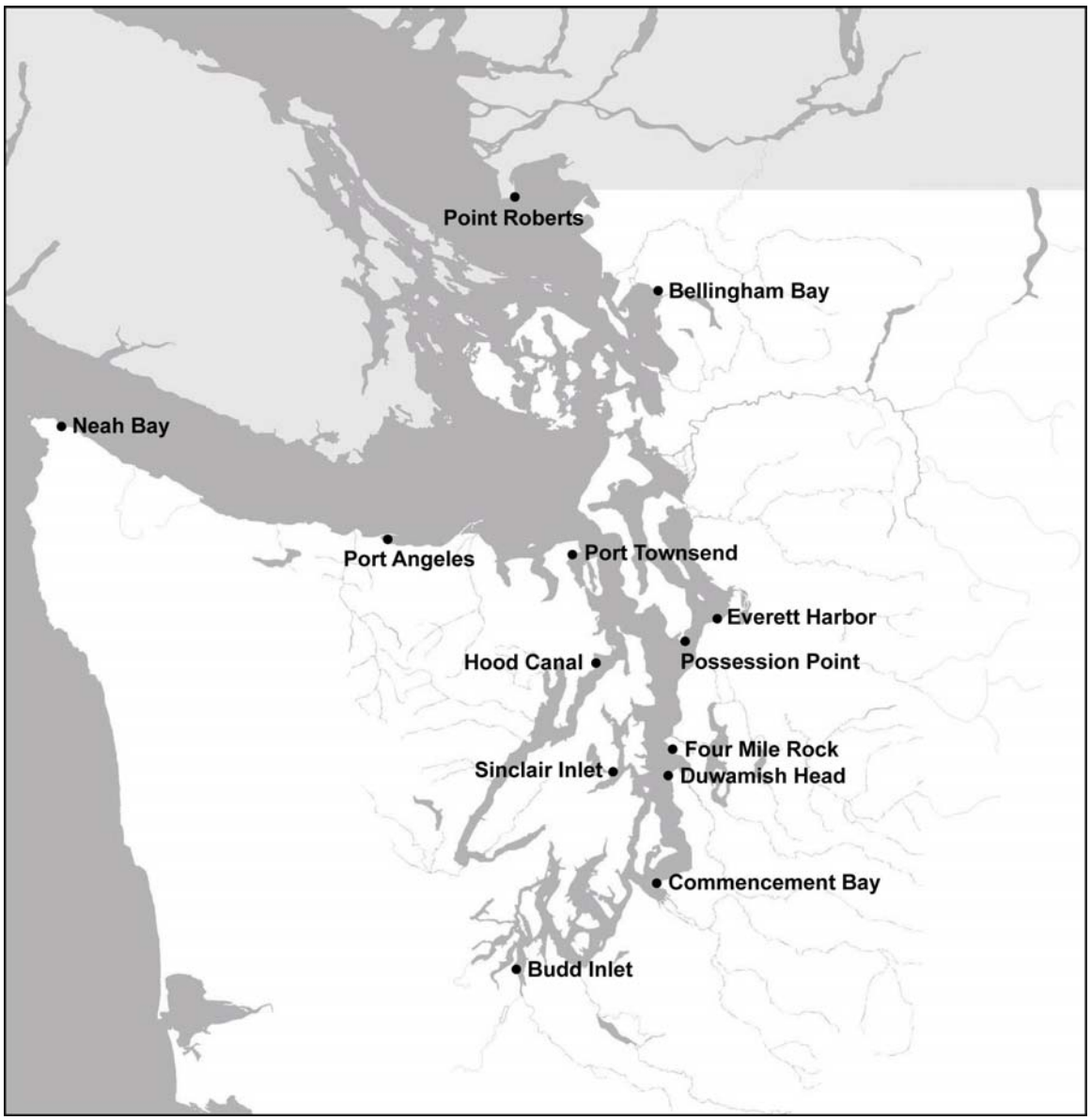
Year/Site	N	Contaminant[a]			
		LPAH[b]	HPAH[c]	PCB[d]	DDT[e]
1989					
Duwamish Waterway	3	120,000 ± 120,000	21,000 ± 15,000	300 ± 40	29 ± 11
Commencement Bay	3	3,200 ± 460	2,800 ± 870	190 ± 50	27 ± 9.0
Nisqually River Estuary	3	30 ± 2.0	4.0 ± 2.0	22 ± 10	2.0 ± 0.6
1990					
Duwamish Waterway	3	34,000 ± 18,000	17,000 ± 11,000	260 ± 90	13 ± 7.0
Commencement Bay	2	39,000 ± 33,000	37,000 ± 33,000	260 ± 70	33 ± 13
Nisqually River Estuary	2	18 ± 3.0	6.5 ± 2.2	33 ± 8.0	9.1 ± 3.0

Source: Stein et al. (1995)
n = the number of samples
[a] concentration expressed is mean ± standard error of the mean; $ng \cdot g^{-1}$ on a wet weight (ww) basis
[b] LPAH – the sum of 8 compounds
[c] HPAH – the sum of 12 compounds
[d] PCB – the sum of homologue classes trichlorobiphenyls to nonachlorobiphenyls plus decachlorobiphenyl
[e] DDT – the sum of *p,p'*-DDT, *p,p'*-DDE, and *p,p'*-DDD.

Shellfish

The only comprehensive survey of contaminants in shellfish of Puget Sound is through the national Mussel Watch program, which uses mussels as a sentinel species on the Pacific Coast (Lauenstein et al. 1997). The focus of the program, which began in 1986, is to monitor overall trends in marine environmental quality rather than the health of the organisms. Mussels filter feed in the water column and are good bio-integrators of water-borne contaminants, although Ylitalo et al. (1999) found that mussels tended to have lower concentrations of contaminants than did species more closely associated with the sediments at similar sampling locations. Ten sites are regularly monitored in Puget Sound (Lauenstein et al. 1997), and the data for 17 elements and 52 organic compounds or groups of compounds are available from NOAA electronically (NOAA 2002). Sampling locations are shown in Figure 4-26.

Figure 4-26: Mussel Watch sampling locations in Puget Sound (NOAA 2002).



In 1998, arsenic concentrations did not vary considerably from site to site, whereas there was an almost ten-fold difference in copper, lead and mercury concentrations (Table 4-12). TBT concentrations in mussels were the most variable, ranging from 5 ng (Sn)·g^{-1} at the monitoring site in Hood Canal to 187 ng (Sn)·g^{-1} near Elliott Bay at Four Mile Rock. Metals concentrations in Puget Sound mussels were generally similar to the national average, with the exception of TBT at Four Mile Rock, which was one order of magnitude higher.

Table 4-12: Summary of selected metals concentrations in Puget Sound mussels (*Mytilus edulis*) as determined in the 1998 Mussel Watch Program. Also shown are the national averages for *M. edulis* and all mollusks analyzed.

	METAL[a]					
LOCATION	ARSENIC	CADMIUM	COPPER	LEAD	MERCURY	TBT
North Puget Sound						
Point Roberts	7.07	1.95	8.36	1.39	0.05	18
Bellingham Bay	7.21	2.73	7.78	1.61	0.16	60
Neah Bay[b]	-	-	-	-	-	-
Port Angeles	6.34	5.44	8.64	0.38	0.09	11
Port Townsend	8.29	2.75	13.50	1.73	0.10	12
Everett Harbor	7.82	2.44	9.38	3.48	0.25	44
Central Puget Sound						
Possession Point	5.51	1.64	8.34	0.58	0.06	9
Four Mile Rock	6.16	1.33	9.32	1.51	0.13	187
Duwamish Head	8.18	2.69	7.08	1.65	0.08	54
Sinclair Inlet	6.99	1.89	7.13	1.08	0.09	9
South Puget Sound						
Commencement Bay	7.51	2.29	9.16	2.59	0.08	17
Budd Inlet	7.41	2.45	5.19	0.57	0.15	9
Hood Canal	6.95	2.97	10.20	0.58	0.09	5
National Average[c]						
Mytilus edulis	8.37 (7.76-14.2)	2.03 (0.53-5.98)	11 (5.19-19.8)	2.34 (0.38-6.72)	0.19 (0.054-0.55)	32 (0-390)
All mollusks[d]	9.96 (3.86-68.7)	3.93 (0.38-17.3)	163 (5.19-3,460)	1.83 (0.23-28.9)	0.15 (0-0.55)	40 (0-650)

Source: NOAA (2002)

[a] Concentrations reported as $\mu g \cdot g^{-1}$ dw, except TBT which was reported as $ng(Sn) \cdot g^{-1}$ dw.

[b] Neah Bay not sampled in 1998

[c] Range shown in brackets

[d] Seven different mollusk species are used depending on regional availability

Organic contaminants in mussels were also variable from site to site, with the highest concentrations occurring in urban embayments, particularly in Central Puget Sound (Table 4-13). For most sites, the concentrations of organic compounds were generally well below the national average. PCB, DDT and dieldrin concentrations at Four Mile Rock near Elliott Bay were at the national average, while PAH concentrations were at the national maximum observed in 1998. The highest chlorpyrifos concentration measured in Puget Sound mussels was in Everett Harbor; the level of the pesticide there was similar to the national maximum.

Table 4-13: Summary of selected organic contaminant concentrations and lipid content in Puget Sound mussels (*Mytilus edulis*) as determined in the 1998 Mussel Watch Program. Also shown are the national averages for *M. edulis* and all mollusks analyzed.

Location	Parameter[a]					
	PCB[b]	DDT[c]	PAH[d]	Dieldrin	Chlor-pyrifos	Lipid
North Puget Sound						
Point Roberts	21	5.3	560	0.8	0	3.7
Bellingham Bay	42	16	1,900	3.3	4.6	4.3
Neah Bay[e]	-	-	-	-	-	-
Port Angeles	19	7.1	420	0.9	0	5.3
Port Townsend	74	7.4	6,200	0.4	0	4.2
Everett Harbor	85	9.3	9,000	2.8	20	4.8
Central Puget Sound						
Possession Point	62	14	400	2.1	0	-
Four Mile Rock	270	350	20,000	25	1.3	6.8
Duwamish Head	180	19	5,000	0	0	6.1
Sinclair Inlet	120	16	300	3.1	0	5.5
South Puget Sound						
Commencement Bay	85	15	1,700	3.4	0	5.9
Budd Inlet	89	10	670	2.8	2.2	7.8
Hood Canal	14	3.9	1,200	1.2	1.6	4.6
National Average[f]						
Mytilus edulis	240 (13.1-2,500)	140 (3.9-2,100)	1,800 (66-20,000)	26 (0-860)	2.46 (0-21)	5.6 (2.8-8.6)
All mollusks[g]	120 (4.7-2,500)	87 (3.9-2,100)	730 (15-20,000)	12 (0-860)	2.48 (0-22)	5.3 (1.3-12)

Source: NOAA (2002)

[a] Concentrations reported as $ng \cdot g^{-1}$ dw, except lipid which is %.

[b] Sum of 18 PCB congeners

[c] Sum of *o,p'*-DDT, *p,p'*-DDT, *o,p'*-DDE, *p,p'*-DDE, *o,p'*-DDD, and *p,p'*-DDD

[d] Sum of 16 PAH compounds

[e] Neah Bay not sampled in 1998

[f] Range shown in brackets

[g] Seven different mollusk species are used depending on regional availability

The Washington State Pesticide Monitoring Program (WSPMP) was initiated in 1992 by the Department of Ecology to assess pesticide residues in surface and groundwater throughout the state. As part of the monitoring program, mussel tissues were collected from five Puget Sound sites (i.e., Padilla Bay, Duwamish Waterway, Hylebos Waterway, Chambers Creek and Budd Inlet) in 1995 (Johnson and Davis 1996). Forty-three pesticides, pesticide breakdown products and PCBs were measured in composite samples consisting of >30 mussels from each site. DDT and its derivatives DDE and DDD, as well as PCBs were detected at all of the sites. Dieldrin and endosulfan were detected at

~70% of the sites while organophosphate pesticides were not detected in any of the samples. The largest number of analytes detected and the highest concentrations were in mussel tissues from the Hylebos Waterway in Commencement Bay. In contrast, the highest concentrations of organic contaminants measured in the Mussel Watch program in 1998 were found in the vicinity of Elliott Bay (i.e., Duwamish Head and Four Mile Rock; Table 4-13).

Krishnakumar et al. (1994, 1999) analyzed mussel tissue from several sites in Puget Sound (i.e., Coupeville and Double Bluff on Whidbey Island, Four Mile Rock and Secrest near Seattle, Commencement Bay, Sinclair Inlet, Eagle Harbor and Oak Bay) for various metal and organic contaminants. The purpose of these studies was to determine whether or not there was a relationship between contaminant levels and biological effects such as hemic neoplasia (i.e., leukemia), a commonly occurring condition in Puget Sound mussels. While the researchers observed elevated PAHs, chlorinated hydrocarbons (PCBs, DDT, other pesticides) and metals in some urban mussels relative to reference sites, an increased incidence of hemic neoplasia was not found (Krishnakumar et al. 1999). However, pathological changes in lysosomes from digestive cells of mussels collected from urban/higher contaminant level areas were observed (Krishnakumar et al. 1994). As well, mussels from urban embayments were found to have reduced growth compared to those from reference areas.

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are byproducts of industrial processes and may be found in municipal sewage and discharges from pulp mills and wood treatment facilities (Section 2). In 1991, a reconnaissance survey was conducted at several sites in Puget Sound to determine if crabs were accumulating dioxins and furans and to determine potential health risks to consumers of the crabs (PTI 1991). Four different crab species (i.e., Dungeness, red, rock and graceful crabs) were collected from twelve sites based on potential contaminant sources such as pulp mills (i.e., Everett Harbor, Bellingham Bay, Port Angeles Harbor), oil refineries (i.e., March Point, Cherry Point), wastewater treatment plants (i.e., West Point near Seattle), wood treatment facilities (i.e., Eagle Harbor), marinas (i.e., Eagle Harbor, Sinclair Inlet near Bremerton, South Elliott Bay), and agricultural areas (i.e., Skagit River estuary). Hepatopancreas and edible muscle tissues were analyzed for PCDD and PCDF congeners and lipid content. The results of chemical analyses were converted to toxic equivalent concentrations (TEQs; see Section 2.2.1.4 for definition). Hepatopancreas TEQ concentrations ranged from 2.0 $ng \cdot kg^{-1}$ in Dungeness crab from Dungeness Bay, the reference area, to 62 $ng \cdot kg^{-1}$ in Dungeness crab from Everett Harbor. TEQs in edible muscle ranged from 0.28 $ng \cdot kg^{-1}$ in Dungeness crab from Bellingham Bay to 2 $ng \cdot kg^{-1}$ in graceful crab from Eagle Harbor. In comparison, TEQs in the hepatopancreas of Howe Sound, British Columbia crabs in 1991 were between 100 and 200 $ng \cdot kg^{-1}$, and have since decreased to ~10 $ng \cdot kg^{-1}$ in 1998 (Environment Canada 2001).

Dioxin-like and total chlorinated biphenyls (CBs) were also evaluated in hepatopancreas and muscle tissue of Dungeness crab from four locations in Puget Sound as part of a country-wide assessment of eight commercially and recreationally important marine species (Table 4-14). As with mussels monitored as part of the Mussel Watch program, the highest concentrations of CBs were observed in crabs from urban embayments of Central and South Puget Sound (Ylitalo et al. 1999).

Table 4-14: Chlorinated biphenyl (CB) concentrations in hepatopancreas and muscle tissue of Dungeness crabs from Puget Sound.

Location	Hepatopancreas			Muscle		
	Total CBs[a]	CB TEQs[b]	% Lipid	Total CBs	CB TEQs	% Lipid
Central Puget Sound						
Elliott Bay	8,800 ± 7,300	140 ± 160	4.6	120 ± 70	1.3 ± 0.81	1.6
Useless Bay	340 ± 220	7.4 ± 4.6	9.6	5.7 ± 1.8	0.07 ± 0.04	0.50
South Puget Sound						
Commencement Bay	1,900 ± 1,700	30 ± 24	5.9	43 ± 34	0.59 ± 0.51	0.87
Nisqually Reach	400 ± 200	6.6 ± 3.4	7.7	8.6 ± 3.6	0.09 ± 0.06	0.37

Source: Ylitalo et al. (1999)
[a] concentration is mean ± standard deviation; $ng \cdot g^{-1}$ ww
[b] concentration is mean ± standard deviation; pg 2,3,7,8-TCDD $eq \cdot g^{-1}$ ww

PAH concentrations in Dungeness crab hepatopancreas were recently measured as part of a pilot study by PSAMP and WDFW's Oil Spill Team (OST) to determine if crabs would be a useful sentinel species for assessing ecological damage related to oil spills (PSWQAT 2002b). Unlike many vertebrate species, crabs are not able to metabolize and excrete PAHs to any great degree. The pilot study indicated that Dungeness crabs accumulated PAHs correlated to the level of sediment contamination at the four sampling locations in the study. The PAH concentration in composite samples of hepatopancreas from Commencement Bay crabs (~50 $\mu g \cdot kg^{-1}$) was significantly higher than that found in Port Gardner (~25 $\mu g \cdot kg^{-1}$), Vendovi Island (~10 $\mu g \cdot kg^{-1}$), or Cherry Point (~5 $\mu g \cdot kg^{-1}$) crabs. Pending funding, the researchers plan to measure PAH concentrations in the edible tissue of Dungeness crabs to determine potential human health risks associated with consuming the crabs. Smaller species such as the graceful crab are also being assessed for use as a sentinel species.

Other studies on contaminants in shellfish have been conducted for the purpose of verifying CWA Section 303(d) listings or to address health concerns expressed by human consumers of shellfish. For example, Dyes Inlet and Port Washington Narrows were listed as impaired waterbodies in 1998 because of elevated antimony, phthalate esters and individual PAHs in the edible tissues of crabs and clams; samples were collected by WDOE in 2001 to determine whether or not the tissue burdens of various contaminants was still of concern (Roose and Johnson 2002a). WDOE found that the contaminants of

concern were not detectable in crab muscle from Ostrich Bay, while clams from Port Washington Narrows had measurable concentrations of PAHs, unlike in previous studies.

WDOE collected clams and crabs from several 303(d) listed waterbodies (Sinclair Inlet, Dyes Inlet, and Eagle Harbor) and reference sites (Southern Hood Canal and Sequim Bay) in 2001 and 2002 to assess arsenic speciation (Roose and Johnson 2002b). Total arsenic concentrations in native and Japanese littleneck clams from the listed waterbodies ranged from 1.9-4.2 $\mu g \cdot g^{-1}$ on a wet weight (ww) basis, while at the reference stations, total arsenic was observed at 2.3-3.4 $\mu g \cdot g^{-1}$ ww. Of the total arsenic present in the clams from the 303(d) listed sites, 0.4-1.2% was in the form of inorganic arsenic. Clams from the reference stations contained 0.7-1.0% inorganic arsenic. The inorganic arsenic concentrations in samples from both the exposed and reference sites exceeded the 303(d) listing criterion of 0.006 $\mu g \cdot g^{-1}$ ww. The researchers concluded that the observed concentrations were naturally occurring and recommended that the 303(d) waterbodies be de-listed for arsenic.

Fourteen composite edible tissue samples from Dungeness crabs, littleneck and butter clams, Japanese oysters and mussels were sampled in Padilla Bay in 1999 for over 130 potentially toxic metals and organic compounds that are of concern for human health (Johnson 2000). The highest metal concentration for crabs was arsenic (5,230-8,390 $\mu g \cdot kg^{-1}$ ww), followed by mercury (41-75 $\mu g \cdot kg^{-1}$ ww) and lead (11-33 $\mu g \cdot kg^{-1}$ ww). Cadmium was not detected at all. Conversely, arsenic concentrations were lower in clams and oysters (1,360-2,600 $\mu g \cdot kg^{-1}$ ww) while cadmium levels were higher (211-1,440 $\mu g \cdot kg^{-1}$ ww). These concentrations were not substantially different than those found at the reference station. Few organic contaminants were detected in crab tissue (two or three samples had DDE, PCB, and TCDF only) compared to oysters, in particular (TBT, pesticides, TCDF and PCBs). These results illustrate interspecies differences in contaminant uptake. Furthermore, only two of the contaminants measured, arsenic and TCDF, exceeded screening levels for the protection of human health, however, arsenic speciation was not conducted to determine the proportion of inorganic arsenic present. The Washington Department of Health (WDOH) recommended further monitoring of arsenic to determine speciation in edible tissues and the associated health risks.

4.3.1.2 Comparison to Tissue Screening Benchmarks

Washington State has not developed tissue residue benchmarks for the protection of piscivorous wildlife. Therefore, where available, tissue residues were compared to 1 in 100 increased lifetime cancer risk levels and non-carcinogenic effects levels developed by New York State for aquatic biota in the Niagara River (Newell et al. 1987). In that document, it was proposed that the 1 in 100 cancer risk level is adequate to prevent decreases in population related to cancer induced by a given toxic compound. The benchmarks used in this study are summarized in Appendix A3.

Two USEPA documents were drawn on for tissue residue benchmarks for the protection of human health (USEPA 1989; 1997). As for wildlife, two levels of protection have been derived: a 1 in 100,000 increased lifetime cancer risk concentration and a noncarcinogenic effects concentration. In humans, no cancer risk level is considered acceptable, however, regulatory agencies generally regard a risk range of 10^{-4} to 10^{-6} as being tolerable.

These tissue benchmarks have been used in this report as a screening tool only, and have not been used to determine risk to human consumers. The manifestation of effects related to the consumption of marine organisms with tissue residues of metals and organic contaminants are dependent on a number of factors including exposure route, form of the compound, and diet[2] (Section 2). Tissue concentrations exceeding these benchmarks should be flagged for further study.

Metals

The only available benchmarks for screening tissue metal concentrations were for the protection of human health. Arsenic concentrations in mussels[3] as well as other marine invertebrates (Mussel Watch; Johnson and Davis 1996; Krishnakumar et al. 1994; Johnson 2000) exceeded the 1 in 100,000 cancer risk criterion for the protection of human health at both urban and non-urban sites, whereas the non-carcinogenic hazards criterion was not exceeded at any site. All other metals concentrations were below both the 1 in 100,000 cancer risk and non-carcinogenic hazard benchmarks for the protection of human health. The exceedances of the arsenic benchmark should not necessarily cause any concern to fish and shellfish eaters because arsenic is ubiquitous in the tissue of marine organisms, and is usually in an organic form that is not toxic to either the organism itself or consumers of the organism (Eisler 1988a; Francesconi and Edmonds 1993).

Organics

PCB concentrations in mussel and crab edible tissues (Mussel Watch; Johnson and Davis 1996; Krishnakumar et al. 1994; Ylitalo et al. 1999) generally exceeded the 1 in 100,000 cancer risk benchmark particularly for samples from urban embayments in Central Puget Sound. Hepatopancreas PCB concentrations exceeded the 1 in 100,000 cancer risk benchmark in samples from both urban and non-urban embayments. The non-carcinogenic hazard benchmarks for the protection of human health and both wildlife benchmarks were not exceeded for PCBs.

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[2] Subsistence and recreational consumers may be at greater risk of exposure to contaminants in fish and shellfish.

[3] Contaminant data for mussels is often reported as dry weight concentrations because lipid content is often highly variable from individual to individual. However, tissue quality values are usually reported as fresh or wet weight. Therefore, in order to compare the two, dry weight tissue concentrations for mussels were converted to wet weight assuming ~ 85% moisture content (data not shown).

Dioxin concentrations in crab hepatopancreas from Everett Harbor (PTI 1991) exceeded the 1 in 100,000 cancer risk level but not the non-carcinogenic hazard level. The benchmarks for chlorinated pesticides were not exceeded in any species.

About twenty mussel samples from Elliott Bay (Four Mile Rock) between 1986 and 1998 (Mussel Watch) exceeded the 1 in 100,000 cancer risk level for benzo[a]pyrene, as did seven samples from Port Townsend, three samples from Everett Harbor, and one from Commencement Bay. The 1 in 100,000 cancer risk level for chrysene was also exceeded for ten samples from Elliott Bay, and one for each of Bellingham Bay, Port Townsend and Budd Inlet.

4.3.1.3 Spatial Trends

Infaunal and benthic invertebrates from urban/industrial estuaries generally contained the highest contaminant levels (Johnson and Davis 1995; Krishnakumar et al. 1994; PTI 1991; Stein et al. 1995; Ylitalo et al. 1991). On a regional level (i.e., North, Central, and South Puget Sound, Hood Canal), however, there did not appear to be a clear geographical trend in metals concentrations (Table 4-12), or a clear relationship between known contaminant sources and concentrations of metals in mussels. For example, the highest concentrations of metals like arsenic, cadmium and lead measured at Washington State Mussel Watch stations were at Cape Flattery on the outer Olympic Peninsula (Mearns 2001). The elevated metals concentrations in mussels at the Pacific coast sites in Washington may be related to ocean upwelling of particular trace elements, following patterns in nutrient cycling. In comparison, elevated organic compound concentrations were measured predominantly in Central Puget Sound. This is consistent with the numbers and magnitude by which sediment concentrations of these same contaminants exceed SQV in the Central Puget Sound area (Section 4.2.2.2).

Another measure of the geographical distribution of contaminants in shellfish is the location at which fish consumption advisories have been posted (Figure 4-27). A majority of the sites are located in Central Puget Sound and are related to organic contaminants, although mercury and other metals were also of concern at some sites (Table 4-15).

Figure 4-27: Location of fish consumption advisories in Puget Sound (WDOH 2002)

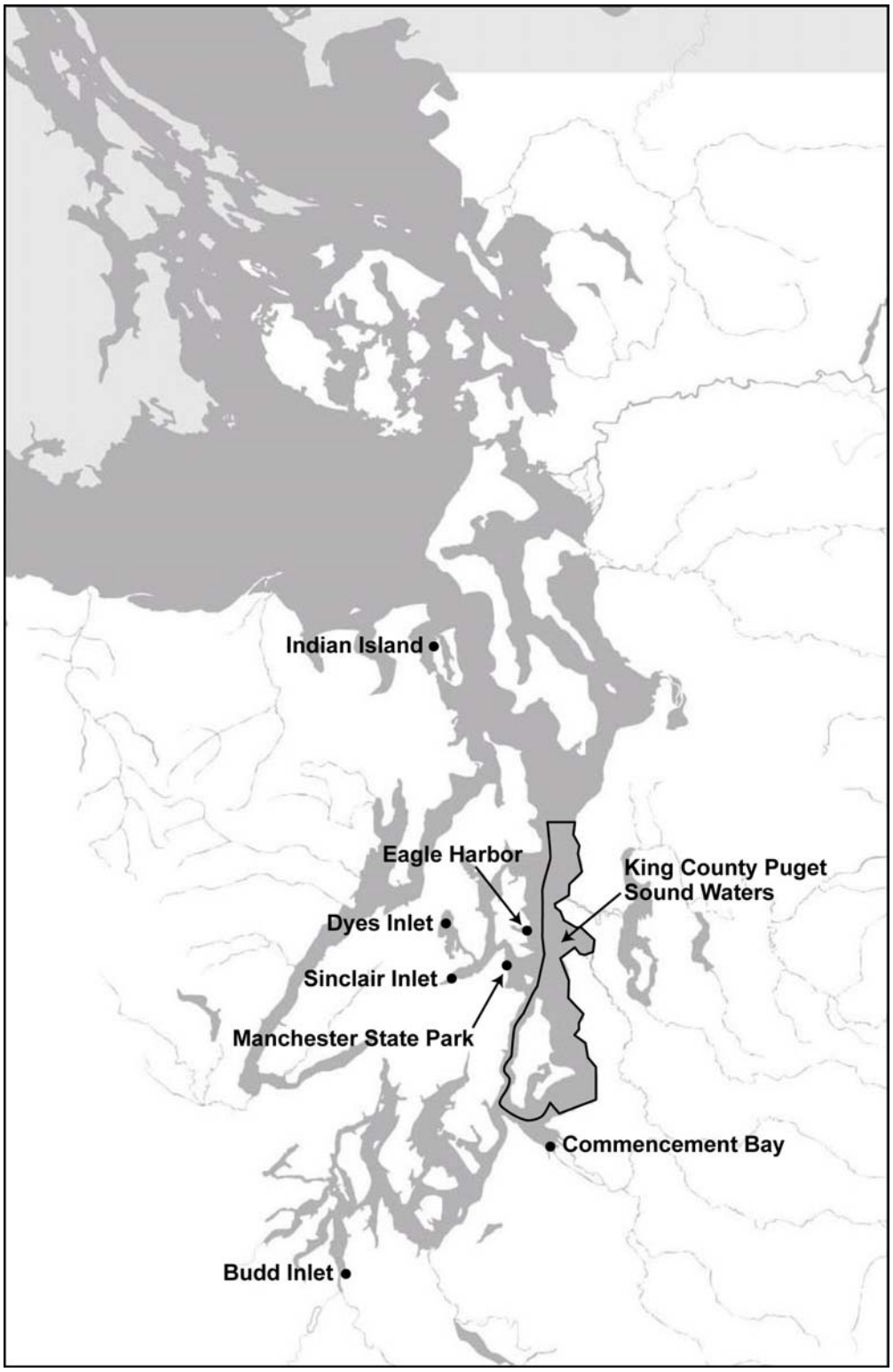


Table 4-15: Fish consumption advisories issued for locations in Puget Sound (as of 2002).

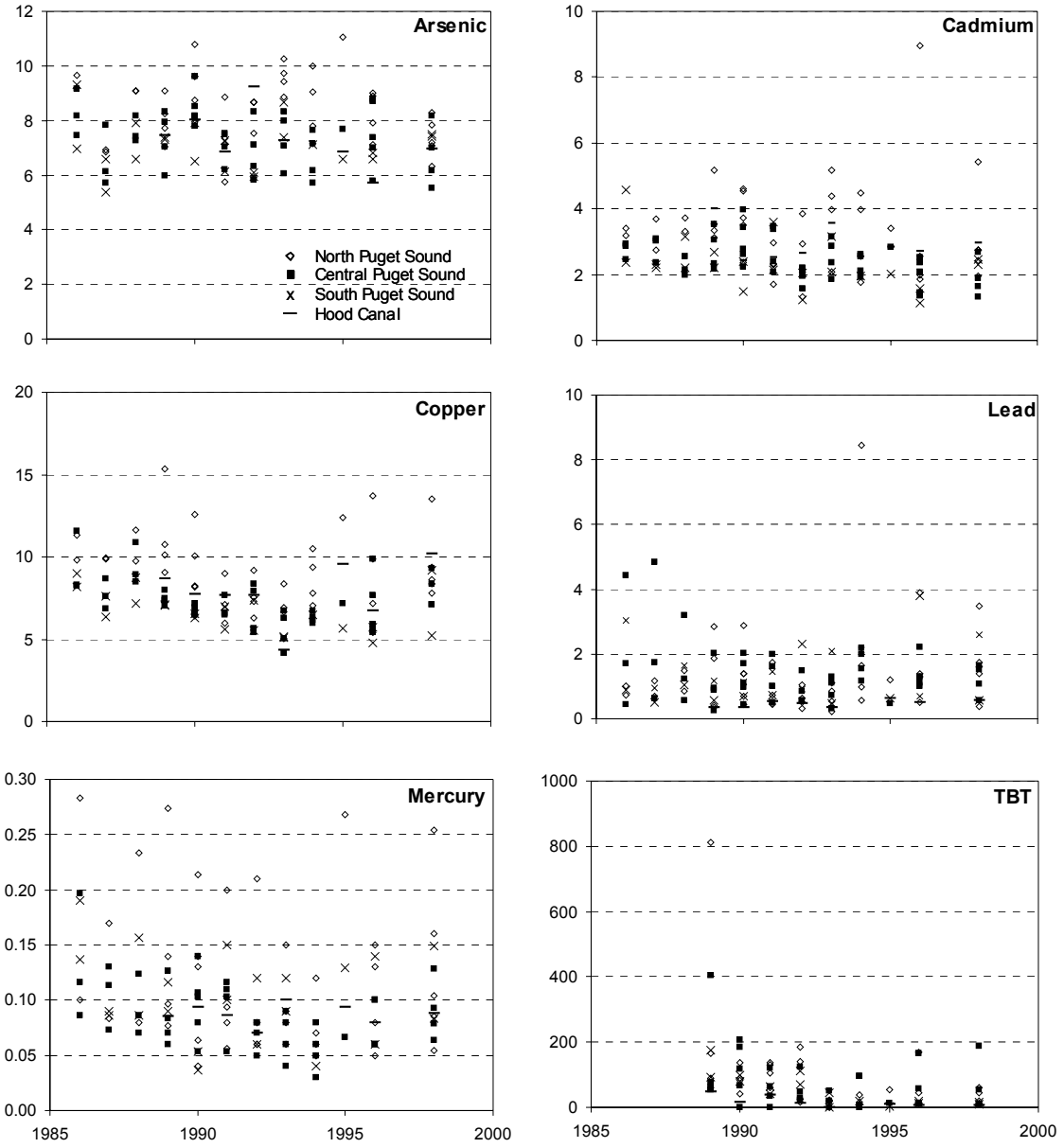
LOCATION	FISH TYPE	CONTAMINANT OF CONCERN
North Puget Sound		
Indian Island in and around Boggy Spit	Shellfish	Pesticides and metals
Central Puget Sound		
Dyes Inlet – west side of Ostrich Bay in the vicinity of the Jackson Park Naval housing development	All bottom fish and all shellfish, including crab	Naval ordnance
Eagle Harbor between Wing Point and Bainbridge Island	All bottom fish and all shellfish, including crab	PAHs, mercury
Sinclair Inlet – south end	All shellfish including crab, and all bottom fish including rockfish	Mercury and PAHs
King County – all Puget Sound waters	Bottom fish, shellfish, seaweed	General – historical industrial discharges
Manchester State Park outside of Clam Bay	All shellfish	PCBs and dioxins
South Puget Sound		
Commencement Bay	Bottom fish, shellfish and crab	PCB, diethylphthalate, metals
Budd Inlet near Eastbay Marina (related to hazardous waste site Cascade Pole)	All shellfish	Creosote, volatile organic compounds, pentachloro-phenol, and dioxins

Source: WDOH (2002)

4.3.1.4 Temporal Trends

The Mussel Watch program has been in operation since 1986 and therefore provides a good data set from which to determine temporal trends of contaminants in invertebrate organisms. Generally, metals concentrations in mussels from the ten Mussel Watch sites in Puget Sound were variable from year to year and without any discernable trends between 1986 and 1998 (Figure 4-28). However, unusual peaks in cadmium at Port Angeles in 1996 (~9 $\mu g \cdot g^{-1}$), lead at Everett Harbor in 1994 (~8 $\mu g \cdot g^{-1}$), and mercury at Everett Harbor in 1998 (~0.25 $\mu g \cdot g^{-1}$) occurred. Copper and mercury concentrations varied most in mussels from Bellingham Bay, where there was an apparent decrease in both metals from the late 1980's to the mid 1990's, after which there was a trend toward increasing concentrations. Mercury subsequently decreased after 1995, while copper continued to increase. TBT concentrations in the mussels from most stations decreased dramatically from 1989 to 1990 and appear to have remained stable since that time.

Figure 4-28: Trends in selected metals concentrations in mussels from North, Central and South Puget Sound and Hood Canal. Concentrations are reported in $\mu g \cdot g^{-1}$ dw, except TBT which is ng $(Sn) \cdot g^{-1}$ dw.

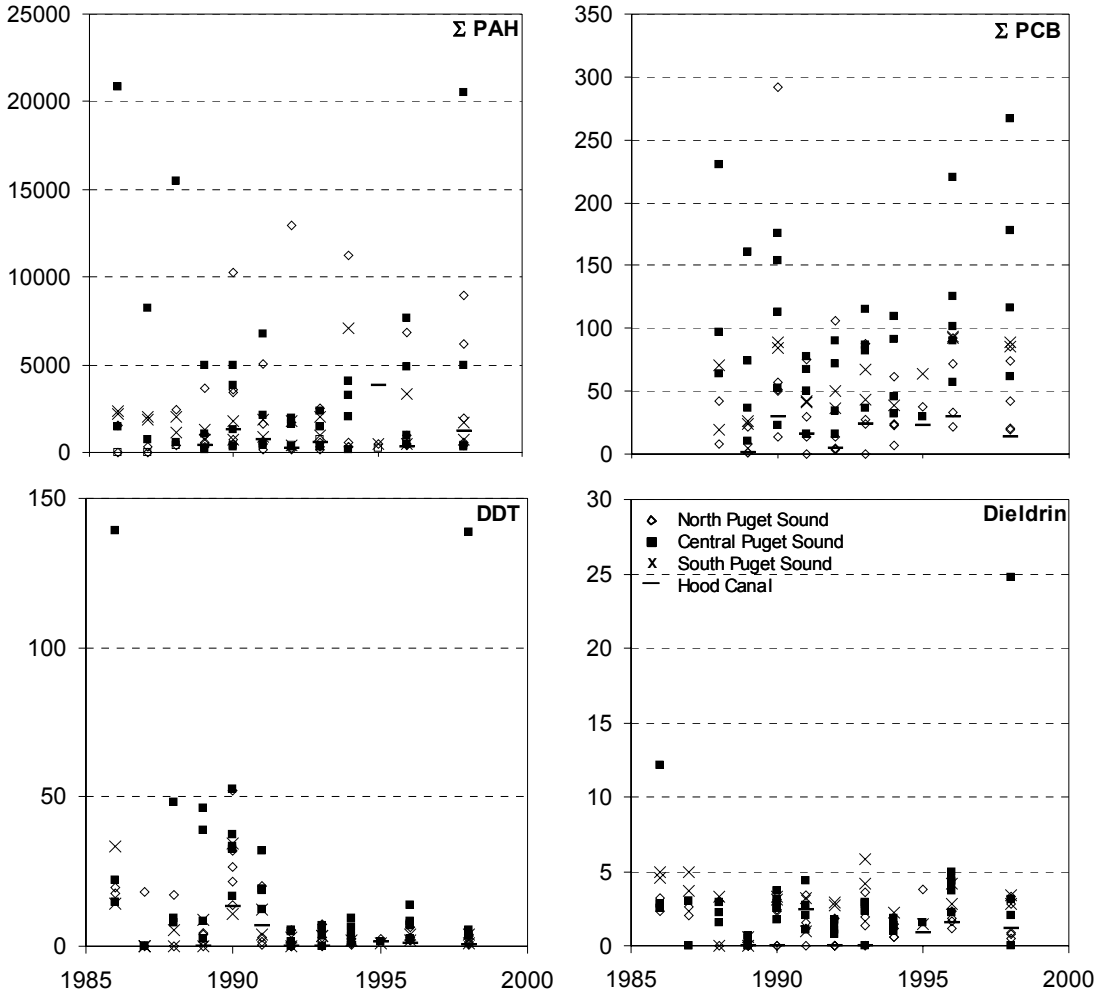


Source: NOAA (2002)

The concentrations of organic compounds in mussels fluctuated more widely than did the metals, although there appeared to be a general downward trend between 1986 and 1998 (Figure 4-29). The highest concentrations of PAHs, PCBs, DDT and dieldrin were measured in mussel samples from Four Mile Rock near Elliott Bay in the mid 1980s. All four chemicals decreased through to the mid 1990s, at which time there was a sudden and large increase from 1996 to 1998. 1999 and 2000 data (not shown) indicate that PCB

concentrations at the Four Mile Rock station decreased again (PSWQAT 2002b). A similar increase and subsequent decrease was also observed in remote areas such as the outer Washington coast and Alaska, suggesting a large-scale event such as a temporary increase in global transport and atmospheric deposition of PCBs (PSWQAT 2002b). With the exception of PAHs in Everett Harbor mussels, the concentrations of PAHs, PCBs, DDT and dieldrin at most sites remained stable after the initial decline observed up to the early 1990s. The trend in PAH concentration from Port Townsend mussels in North Puget Sound was inconsistent with the other sites in that it increased in the mid 1990s while PAH concentrations were relatively lower and stable at the remaining sites.

Figure 4-29: Trends in selected organic compound concentrations in mussels from North, Central and South Puget Sound and Hood Canal. Concentrations are reported in $ng \cdot g^{-1}$ dw. (ΣPAH is the sum of 18 PAH compounds; ΣPCB is the sum of 18 congeners)



Source: NOAA (2002)

Not enough data were available to determine temporal trends in the contaminant burdens of other invertebrate species.

4.3.1.5 Summary of Toxic Contaminants in Invertebrates

State of Knowledge

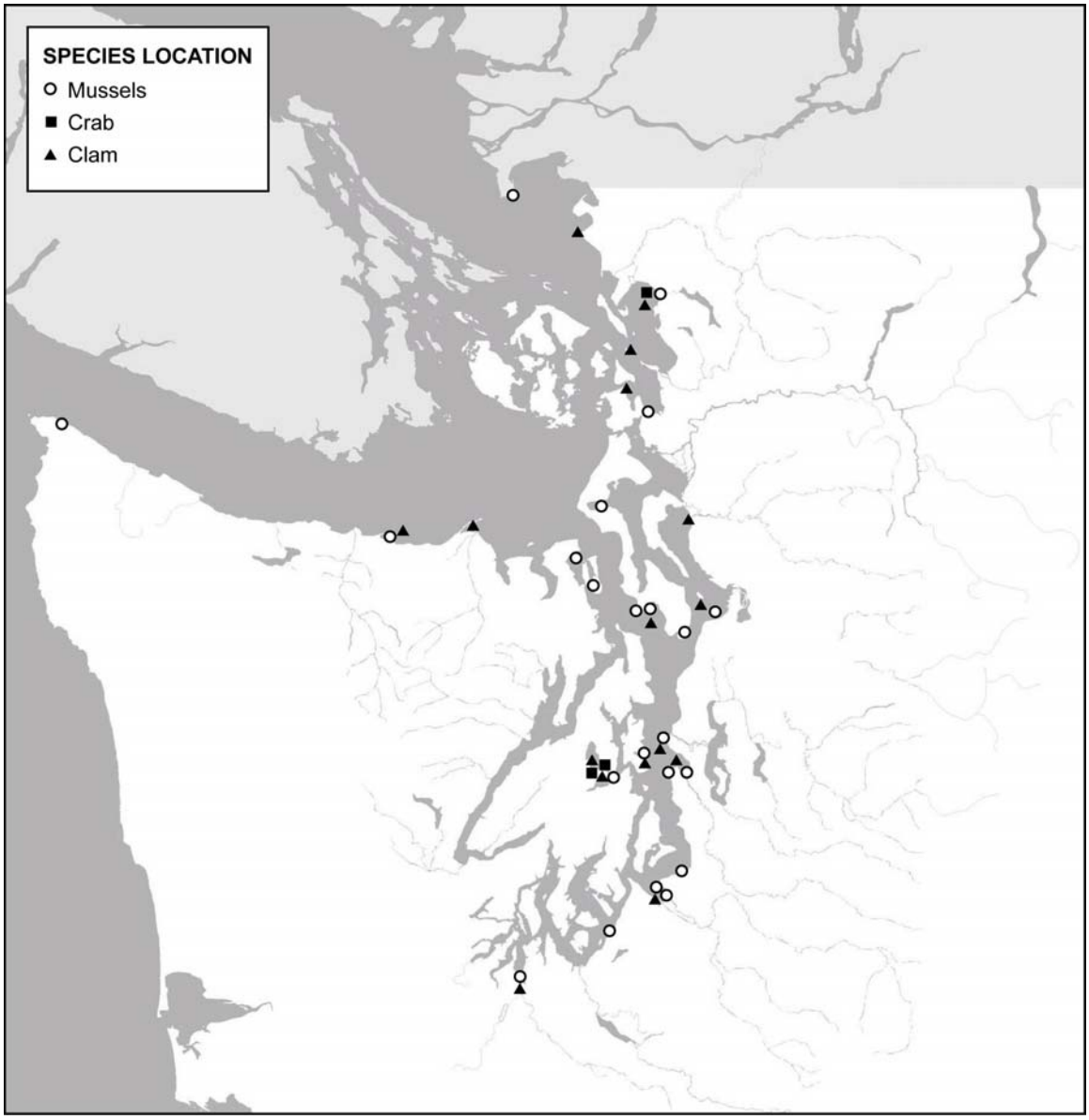
In addition to the Mussel Watch locations shown in Figure 4-26, other shellfish have also been sampled throughout Puget Sound (Figure 4-30). NOAA's Mussel Watch program is the only comprehensive and long-term database of metal and organic contaminant concentrations in Puget Sound mollusks.

These studies have shown that:

- Because mussels are filter feeders, they tend to have lower tissue contaminant burdens than invertebrates more closely associated with sediments (Ylitalo et al. 1999). Any assessment regarding contaminant trends will tend to be an underestimate of what is happening with benthic organisms such as clams and crabs;
- Arsenic concentrations in marine invertebrates regularly exceeded prescribed screening benchmarks for the protection of piscivorous wildlife and human health. However, the arsenic is predominantly in a form that is not toxic to the organism or consumers;
- Shellfish from both 303(d) listed and reference locations exceeded the inorganic arsenic listing criterion (0.006 $\mu g \cdot g^{-1}$), however, the observed concentrations were likely due to naturally occurring conditions in Puget Sound (Roose and Johnson 2002b);
- There is no clear geographic trend in tissue metals residues that can be related to human activities. For example, cadmium concentrations were found to be highest in mussels from the outer Washington coast;
- PCBs and DDT have been detected in almost all of the tissue samples analyzed, while dieldrin has been detected in approximately two thirds of the samples and other organochlorine pesticides have not been detected at all;
- The highest organochlorine residues have been found in organisms from Commencement Bay and Elliott Bay;
- PCB concentrations in mussel and crab edible tissues generally exceeded the 1 in 100,000 cancer risk benchmark, particularly for samples from urban embayments in Central Puget Sound;

- PCB concentrations in mussel and crab edible tissue did not exceed the noncarcinogenic effects concentrations for either human health or piscivorous wildlife;
- Benthic organisms consumed by juvenile salmonids in the Duwamish Waterway and Commencement Bay have higher concentrations of organic contaminants (i.e., PAH, PCB, DDT) than do food organisms from uncontaminated areas (Stein et al. 1995).

Figure 4-30: Locations at which shellfish have been sampled for metals and organic contaminants



Contaminant Effects

- Mussels in urban embayments have been observed to have lower growth and higher rates of pathological changes in digestive cells than mussels from remote areas (Krishnakumar et al. 1994);
- Little is known about the ecological effects at the individual, population or ecosystem level of measured contaminant tissue burdens in benthic infauna and shellfish.

Monitoring and Data Gaps

Monitoring and data gaps and issues related to metals and organic contaminants in the invertebrates of Puget Sound include the following:

- Contaminant levels and effects in benthic infauna, other than large mollusks that are harvested, have not been studied;
- Contaminant levels and effects in crabs and other shellfish are not well studied;
- Studies of contaminants in crabs are often measured in the hepatopancreas tissue for ecological studies, while assessment programs with the objective of determining human health effects related to shellfish consumption usually use muscle tissue, or the "edible" portion of the animal. Therefore, data collected for one purpose may not be useful for the other. Research into the use of extrapolation factors would be valuable;
- The speciation of some metals, such as arsenic, in shellfish and therefore the bioavailability to mammalian (including human) consumers, is not well understood;
- Other than the Mussel Watch program, there are no comprehensive, long-term data sets for marine invertebrates, therefore, it is difficult to assess temporal contaminant trends;
- There are insufficient data regarding the presence/composition of individual PCB or dioxin congeners in invertebrate tissue to support human health risk assessments;
- The relationship between sediment and tissue concentrations is poorly understood, including the preferential uptake of different contaminant forms (e.g., PCB or dioxin congeners);
- There are no Washington State or Puget Sound-specific tissue residue screening benchmarks for marine invertebrates.

4.3.2 Fish

4.3.2.1 Review Of Existing Data

To date, the most comprehensive survey of contaminants in fish tissue has been conducted by the Puget Sound Ambient Monitoring Program (West et al. 2001a). Ten different fish species representing different life histories and feeding strategies were collected between 1989 and 1999 from stations throughout Puget Sound representing urban, near-urban and non-urban conditions. Fish tissues were analyzed for contaminants including chlorinated pesticides, PCBs, aromatic hydrocarbons, chlorinated aromatic hydrocarbons, phthalate esters and selected toxic metals. Additional interpretive analysis of the data presented in West et al. (2001a) is provided in a number of papers, which will be discussed below, as will further studies conducted on individual species and in site-specific areas.

Flat Fish

The most studied fish in Puget Sound is the English sole. This species is widely distributed throughout Puget Sound, lives in close contact with bottom sediments and consumes benthic invertebrates. Therefore, it is a useful sentinel species for exposure of aquatic organisms to contaminated sediments. Fish were collected regularly from eight locations, and intermittently from 54 locations throughout Puget Sound between 1989 and 1999 (West et al. 2001a). Muscle and liver tissues were analyzed for a suite of metals and organic contaminants, and bile was analyzed for Fluorescing Aromatic Compounds in bile (biliary FAC). Because fish rapidly metabolize and excrete PAHs, fish tissue residue concentrations of parent PAH compounds do not provide a useful measure of exposure to fish (Varanasi et al. 1989). Therefore, the breakdown products of PAH metabolism found in bile (i.e., biliary FACs) are measured instead.

Mercury and copper concentrations in the muscle tissues of English sole were low compared to other fish sampled by PSAMP (West et al. 2001a). Arsenic, however, was considerably higher, ranging from 1.8 to 20.0 $mg \cdot kg^{-1}$ (compared to chinook salmon in which arsenic ranged from 0.09 to 1.80 $mg \cdot kg^{-1}$). While the mean lead concentration was similar to that of other species, the maximum value (0.110 $mg \cdot kg^{-1}$ in Sinclair Inlet) was an order of magnitude higher than any other species. Furthermore, age-specific lead concentrations in Sinclair Inlet English sole were significantly higher than those in English sole from all other Puget Sound stations (PSWQAT 2000b). Total PCBs were higher in English sole than all other fish species sampled except Pacific herring, while total DDTs were lower in English sole.

Researchers from NOAA conducted a series of studies on fish from the Hylebos Waterway in the mid 1990s to determine the relationship between contaminant exposure and fish injury (Collier et al. 1998). As part of the *Flatfish Toxicopathic Injury Study*,

adult English sole were collected from several sites in the Hylebos Waterway and a reference site near Colvos Passage. Liver, bile and stomach contents were analyzed for contaminant concentrations, while Cytochrome P4501A (CYP1A) activity and DNA damage were measured in liver tissues. CYP1A induction is a biochemical response to organic contaminants and is used as a biomarker for contaminant exposure, as is DNA damage (also referred to as DNA adducts). The research team observed that biliary FAC concentrations generally paralleled aromatic hydrocarbon levels in the stomach contents (Collier et al. 1998). Elevated concentrations of chlorinated hydrocarbons such as PCB, hexachlorobenzene, and DDT in the stomach contents were also reflected in elevated liver concentrations of the same contaminants. Hepatic CYP1A activity and levels of DNA adducts were significantly higher in fish from the Hylebos Waterway than in fish from Colvos Passage.

While the overall mortality rate of adult English sole from contaminated sites in Puget Sound was similar to the fish from relatively uncontaminated sites (Landahl et al. 1997), chronic effects such as toxicopathic liver lesions, reproductive impairment, and immunosuppression have been documented (Collier et al. 1998; Arkoosh et al. 1996). In the NOAA study described above, exposure to PAHs (as estimated by biliary FAC), PCB and DDT and its derivatives were each shown statistically to be a significant risk factor in the prevalence of several lesion types such as neoplasms, foci of cellular alteration (FCA), proliferative lesions, and specific degenerative/necrotic lesions (SDN). The co-occurrence of contaminants, as is the case in the Hylebos Waterway, can make it difficult to determine relative ecological importance of the different classes of chemicals. However, English sole from locations with high concentrations of PCB in the absence of elevated PAH had the same prevalence of liver lesions as fish from less contaminated reference sites (PSWQAT 1998). Furthermore, the mode of action of PAH hepatic toxicity has been experimentally shown to be consistent with what occurs in the field (discussed in Collier et al. 1998). Therefore, PAH exposure appears to be the main ecologically significant risk factor.

An additional component of the NOAA research on fish injury in the Hylebos Waterway was the *Flatfish Reproductive Injury Study* (Collier et al. 1998). Adult English sole were collected from several sites in the Hylebos Waterway and a reference area between September 1994 and April 1995, during the known period of vitellogenesis (i.e., the production of the protein vitellogenin by the liver of female fish in preparation for reproduction) in this species. As in the *Flatfish Toxicopathic Injury Study*, chemical analyses of liver tissue, bile and stomach contents were conducted. Additional analyses included histopathological examination of ovaries and measurement of reproductive status (i.e., estradiol in plasma; gonadosomatic index [GSI] – a measure of gonad development). Gravid fish showed similar exposure to contaminants as the fish examined in the liver lesion study. Furthermore, juvenile sole exhibited precocious (i.e., early) sexual maturation and adults showed inhibited gonadal development (Collier et al. 1998). Both PAH and chlorinated hydrocarbons were determined to be statistically significant

risk factors for the precocious maturation, while exposure to PAHs alone was a significant risk factor for the inhibited ovarian development. The influence of nutritional status and chemical exposure on fecundity (number of eggs produced per female) and egg weight were studied for English sole from three contaminated urban waterways and one reference station. While contaminant levels did not appear to affect egg development, PCB concentrations were statistically correlated with reduced egg weight and plasma vitellogenin levels, and biliary FACs were correlated with higher rates of ovarian lesions and reduced fecundity (Johnson et al. 1997). Spawning success rates for English sole in Hylebos Waterway have not been studied, however, Collier et al. (1998) estimated that the fish from this area would only produce about 25% of the viable offspring produced by English sole from the reference site in Colvos Passage.

Rockfish

Rockfish are long-lived (60-90 years) carnivorous fish that tend to occupy relatively small areas on rocky reefs. Because of their age and trophic status, rockfish may bioaccumulate higher concentrations of contaminants than other bottom fish such as English sole (West et al. 2001a). Quillback, copper and brown rockfish were collected from 24 stations in Puget Sound between 1989 and 1999 and analyzed for PCBs, DDTs and metals (West et al. 2001a). Mercury concentrations ranged from 0.004 $mg \cdot kg^{-1}$ in quillback rockfish to 1.2 $mg \cdot kg^{-1}$ in yelloweye rockfish. West and O'Neill (1998) assessed the mercury concentrations in rockfish from six of the 24 sampling locations and found that mercury increased with age in the rockfish from all sampling locations. Fish from Sinclair Inlet (urban), Elliott Bay (urban), and Foulweather Bluff (non-urban) had higher age-specific mercury concentrations than did fish from the other sites. These results illustrate the potential importance of trophic transfer of contaminants from contaminated areas to less-contaminated or "pristine" sites. There did not appear to be any difference in accumulation based on gender or lipid content.

Total PCB concentrations ranged from 1.7 $\mu g \cdot kg^{-1}$ in quillback rockfish to 123 $\mu g \cdot kg^{-1}$ in brown rockfish (West et al. 2001a). West and O'Neill (1998) observed a significant correlation between age and PCBs in male rockfish from the two urban sampling locations (Elliott Bay; Sinclair Inlet) but not in female rockfish (Table 4-16). Unlike mercury, PCBs may be transferred from female rockfish of reproductive age to their eggs. West et al. (2001b) observed vitellogenesis in two of eleven male rockfish from Elliott Bay indicating exposure to exogenous estrogens or estrogen mimics, however, exposure to PCB, a known pseudo-estrogen, may or may not be the cause. The reproductive effects of vitellogenesis in male rockfish and the transfer of organic contaminants to offspring are unknown at this time.

Table 4-16: Mean concentrations of PCB (as sum of Aroclors 1254 and 1260) in rockfish from Puget Sound.

	MALE		FEMALE	
LOCATION	CONCENTRATION[a]	AGE[b]	CONCENTRATION	AGE
Urban	211.84 (±149.60)	18.8	93.98 (±52.03)	15.7
Near-urban	52.61 (31.67)	11.7	39.33 (39.37)	15.8
Non-urban	4.9 (2.69)	14.5	4.0 (0.00)	13.9

Source: West et al. (2001b)
[a] Data are reported as mean with standard deviation in brackets, and concentrations are in $\mu g \cdot kg^{-1}$ ww.
[b] Ages are in years.

Mean biliary FAC-BaP concentrations ranged from 380 $ng \cdot mL^{-1}$ bile (n=94) in quillback rockfish to 1,164 $ng \cdot mL^{-1}$ bile in brown rockfish (n=10). The ecological significance of these biliary FAC levels was not discussed.

Salmon

Stein et al. (1995) collected outmigrating juvenile chinook salmon from the Duwamish Waterway and Commencement Bay in order to determine contaminant exposure and uptake during the estuarine residence time of this species. PCB, DDT and PAHs were reported for stomach contents, while PCB, DDT and biliary FAC concentrations were reported for liver tissue. Mean liver concentrations of total PCB were up to ~450 $ng \cdot g^{-1}$ ww for fish from the Duwamish Waterway in 1989. In comparison, PCBs in fish from the Nisqually Estuary were significantly lower at ~100-150 $ng \cdot g^{-1}$ ww. These higher PCB levels were attributed to dietary exposure; the stomach contents of juvenile chinook from the Duwamish were 300 $ng \cdot g^{-1}$ ww, compared to 190 $ng \cdot g^{-1}$ ww in Commencement Bay and 22 $ng \cdot g^{-1}$ ww in the Nisqually Estuary. This work confirmed a pilot study conducted in the Duwamish Waterway by McCain et al. (1990).

While PAHs in the stomach contents of fish from the Duwamish were significantly higher than in Commencement Bay, biliary FAC concentrations were not significantly different between the two sites, suggesting that the PAHs were not bioavailable (Stein et al. 1995). In a similar study conducted in the Hylebos Waterway of Commencement Bay, Stehr et al. (1998) found that chum salmon juveniles showed higher indicators of exposure (i.e., cytochrome P4501A activity; DNA adducts) than did chinook salmon, suggesting that chum salmon may be more susceptible to biological injury from contaminant exposure than chinook salmon.

Estuaries provide important rearing habitat for juvenile salmonids. During their estuarine residence time, which may last from days to months depending on the species and/or the stock, the young fish undergo physiological changes that allow them to migrate from freshwaters to the marine environment. At this time, juvenile salmonids may be susceptible to physical stressors and disease. Varanasi et al. (1993) injected hatchery-

reared chinook salmon with an organic solvent extract of contaminated sediment from the Duwamish Waterway and observed immunosuppression (i.e., decreased ability of B cells to produce antibodies to specific antigens), demonstrating that contaminant exposure of juvenile salmonids during estuarine residence time may result in adverse biological effects. In order to further assess the effect of contaminant exposure on immune function in juvenile salmonids, Arkoosh et al. (1998) challenged young chinook from the Duwamish Waterway with a marine pathogen, *Vibrio anguillarum*. The liver concentrations of total PCBs, DDT and biliary FACs for fish from the Duwamish Estuary were slightly lower than those reported in Stein et al. (1995), however, they were still significantly higher than in fish from the reference location. Higher cumulative mortality as a result of infection by *V. anguillarum* occurred in the fish from the Duwamish as compared to those from a hatchery and a non-urban estuary. Furthermore, decreased disease resistance persisted in the Duwamish chinook even two months after being removed from the contaminant source.

Adult salmon returning to spawn in the freshwater tributaries of Puget Sound were captured annually between 1990 and 1998, but not 1991 (West et al. 2001a). The muscle tissue from coho and chinook salmon were analyzed for a number of metal and organic contaminants (Table 4-17); PCBs were one of the few contaminants that appeared to accumulate in Puget Sound salmon. O'Neill et al. (1998) interpreted the data from the PSAMP fish monitoring program in terms of percent lipid, location caught, origin of the fish (i.e., hatchery or wild), gender and age (chinook only). Chinook had significantly higher PCB concentrations (reported as total Aroclors) and total DDT than did coho, potentially explained by the fact that chinook adults tend to be older when they return to spawn, they have a higher lipid content, and they tend to consume a greater percentage of fish than do coho. O'Neill et al. (1998) suggested that the contribution of contaminants accumulated by juveniles migrating through contaminated estuaries to the PCB burden of adult salmon was negligible, and that other sources in Puget Sound (i.e., related to contaminated sites) and/or the North Pacific Ocean (i.e., related to atmospheric deposition) were more important. The ecological significance of the PCB concentrations measured in adult chinook and coho is unknown to date. Furthermore, the maternal transfer of PCBs to eggs and the resulting reproductive effects in Puget Sound salmon have not been examined.

Table 4-17: Summary of contaminants in muscle tissue of adult coho and chinook salmon captured in Puget Sound.

	COHO SALMON		CHINOOK SALMON	
	RESULT	N	RESULT	N
Contaminant				
Σ Aroclors (µg·kg^{-1} ww)[a]	33 (6.0-130)	233	54 (12-220)	217
Σ DDTs (µg·kg^{-1} ww)[b]	12 (3.0-39)	266	21 (4.3-59)	228
Mercury (mg·kg^{-1} ww)	0.049 (0.025-0.110)	108	0.093 (0.051-0.16)	106
Copper (mg·kg^{-1} ww)	0.57 (0.23-1.01)	103	0.57 (0.26-1.20)	101
Arsenic (mg·kg^{-1} ww)	0.64 (0.09-1.60)	103	0.69 (0.09-1.80)	101
Lead (mg·kg^{-1} ww)	0.03 (0.02-0.04)	103	0.03 (0.02-0.04)	101
Physical data				
Lipids (%)	3.3 (0.2-15)	265	3.4 (0.2-22)	227
Composite age (yrs)	3	288	4 (2-5)	225
Composite length (mm)	560 (370-720)	288	753 (391-1010)	232

Source: West et al. (2001a)
Data reported as means with range shown in brackets.
N = number of samples
[a] Σ Aroclors = sum of Aroclors 1248, 1254 and 1260
[b] Σ DDT = sum of p,p'DDT, p,p'DDE and p,p'DDD
NA = not analyzed

Herring

A pilot study of toxic contaminants in Pacific herring stocks of Puget Sound was initiated in 1995 and routine monitoring began in 1999 (West et al. 2001a). Mean whole body total PCBs for all herring analyzed was 130 µg·kg^{-1} ww. The highest concentrations, 200 µg·kg^{-1}, were measured in fish from Port Orchard in central Puget Sound, compared to 17 µg·kg^{-1} ww in fish from the Strait of Georgia (O'Neill and West 2001). O'Neill and West (2001) suggested that the higher concentrations measured in the Port Orchard herring stock are due to elevated levels of organic contaminants in the food web of Central Puget Sound, and may put them at risk for adverse biological effects. Total DDT concentrations ranged from 13 µg·kg^{-1} ww in fish from Cherry Point to 47 µg·kg^{-1} in fish from Port Orchard. The maternal transfer of contaminants to spawned eggs is currently being assessed in a pilot study (O'Neill, pers. comm. 2002).

4.3.2.2 Comparison to Tissue Screening Benchmarks

Tissue screening benchmarks are described in more detail in Section 4.3.1.2. and summarized in Appendices A3 and A4.

Metals

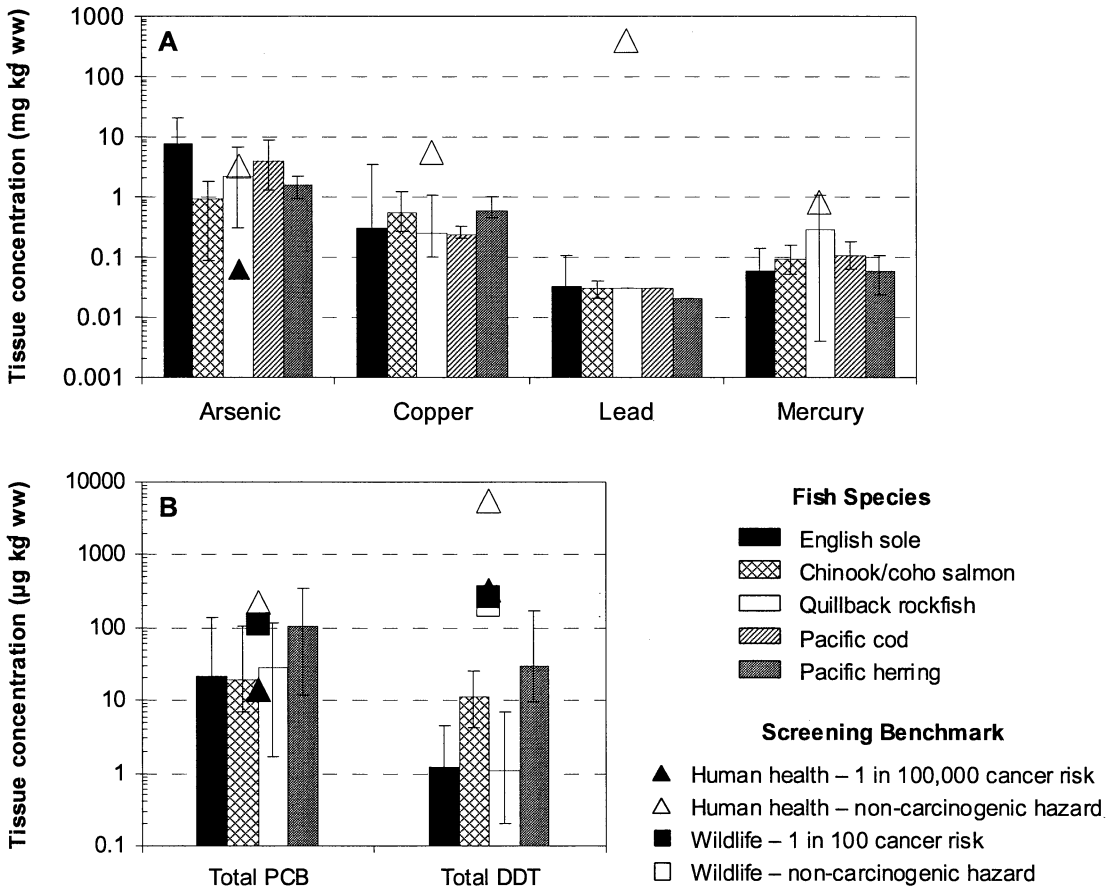
The only available benchmarks for screening tissue metal concentrations were for the protection of human health. The arsenic noncarcinogenic effects benchmark was exceeded by some samples for all fish species, while all fish samples exceeded the 1 in 100,000 cancer risk level. There is some evidence that arsenic tends to be higher in lower trophic organisms such as flatfish. As with invertebrates, the exceedances of the arsenic benchmark should not necessarily cause any concern to fish consumers because arsenic is ubiquitous in marine organisms and is usually in a form that is not toxic (Eisler 1988a; Francesconi and Edmonds 1993). The mercury benchmark for noncarcinogenic effects was exceeded only by a few samples of quillback rockfish. Rockfish are long-lived fish occupying a high trophic level in the food chain and tend to accumulate mercury to a greater degree than the other fish species analyzed. Health officials recommend that consumers of fish limit their intake of species such as rockfish due to the risk of mercury exposure (WDOH 2003). None of the fish species had tissue concentrations exceeding the copper or lead benchmarks.

Organics

PCB and DDT screening benchmarks were available for both the protection of human health and piscivorous wildlife. Some samples of all the fish species tested had PCB tissue concentrations exceeding the benchmark for both the noncarcinogenic effects and the 1 in 100 cancer risk level for the protection of wildlife, while the DDT benchmarks were not exceeded in any tissue sample.

Almost all of the fish tissue samples exceeded the 1 in 100,000 cancer risk level for PCBs while only Pacific herring samples exceeded the noncarcinogenic effects level for the protection of human health. The DDT benchmarks were not exceeded in any tissue samples.

Figure 4-31: Summary of concentrations of selected (**A**) metals and (**B**) organic compounds in the tissue of Puget Sound fish compared to screening benchmarks for the protection of human health and piscivorous wildlife. Columns indicate mean concentrations in muscle tissue (except for Pacific herring which was measured as whole body) with error bars showing range.



Source: West et al. (2001a)

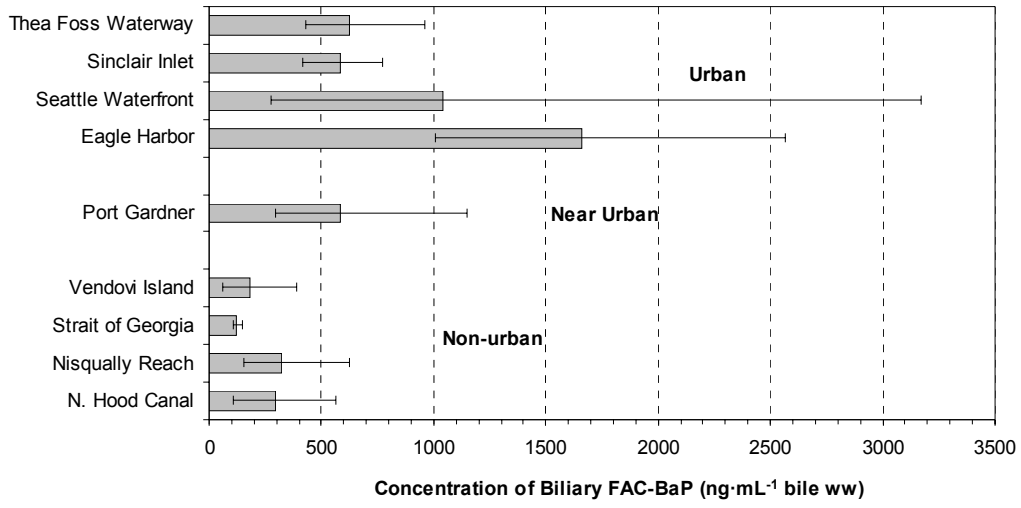
4.3.2.3 Spatial Trends

The spatial variation of contaminants in fish and resulting adverse biological effects is clearest for fish species or life stages with a lower trophic status and/or a high affinity for localized habitats, and therefore more closely associated with contaminant sources. Both juvenile salmonids and flatfish like the English sole tend to consume prey species associated with the benthic environment. For example, the food organisms comprising the stomach contents of juvenile chinook from the Duwamish Waterway consisted of cumaceans, amphipods and small fish (McCain et al. 1990). Cumaceans and amphipods

are crustaceans that feed on microorganisms and organic matter in benthic sediments. English sole collected on the West Coast of the U.S. were found to consume arthropods, annelids, echiurids, mollusks and debris (McCain et al. 2000). Several studies have shown that contaminant levels in stomach contents are reflected in elevated tissue contaminant burdens, contaminant metabolites, or biomarker levels (West et al. 2001a; Collier et al. 1998; Stein et al. 1995; Varanasi et al. 1993; McCain et al. 1990).

Figure 4-32 shows the spatial variation of biliary FAC-BaP (benzo[a]pyrene equivalents) in English sole from a number of locations in Puget Sound. An assessment of data collected from these sites and others by WDFW staff showed that the risk of English sole developing liver disease was highest (10x) at the Seattle waterfront compared to the baseline reference site in Hood Canal (PSWQAT 2002b). The other sites determined to have a greater risk compared to the reference site included Commencement Bay (8x), Sinclair Inlet (2x), and Port Gardner (3x). English sole from the Strait of Georgia sampling location were also determined to have a higher risk (2x) of liver disease than from the Hood Canal site. A number of reports confirm that liver lesions in English sole from urban waterways, particularly where PAH contamination is significant (such as Elliott Bay and Commencement Bay) are more prevalent than in sole from near-urban or non-urban waterways such as Hood Canal or Port Susan, (Collier et al. 1998; O'Neill et al. 1998).

Figure 4-32: Biliary benzo[a]pyrene FAC concentrations in English sole from non-urban, near-urban and urban areas of Puget Sound. Columns are mean concentrations for data from 1989-1999, with error bars indicating the range.



Source: West et al. (2001a)

Johnson et al. (1994) compiled data regarding survival rates and indicators of reproductive success for four sites in Puget Sound and found that English sole from

moderately (Sinclair Inlet) to highly contaminated (Duwamish Waterway, Eagle Harbor) sites had a much lower overall reproductive success than fish from an uncontaminated area (Port Susan).

West et al. (2001b) detected PCBs in 100% of rockfish sampled from urban areas, 97.4% of fish from near-urban areas and 6.7% from non-urban areas. Furthermore, rockfish from urban areas (Sinclair Inlet and Elliott Bay) had significantly higher concentrations of PCB Aroclors 1254 and 1260 than those from near-urban areas (Central Puget Sound) and non-urban areas (San Juan Islands and Admiralty Inlet). Mercury concentrations in urban rockfish, particularly those from Sinclair Inlet and Elliott Bay, were also higher than in fish from near- or non-urban sites.

The spatial relationship between contaminant sources and tissue burdens for pelagic fish like adult salmon and herring are less clear. As discussed above, tissue concentration of organic contaminants in juvenile salmonids was related to proximity to contaminated estuaries such as in the Duwamish Waterway and Commencement Bay. While adverse biological effects were noted from this exposure, the contribution of contaminants from this life stage to the adult stage migrating back to freshwater was found to be negligible. Instead, adult tissue burdens of PCB were suggested to be from Puget Sound proper and/or the North Pacific Ocean. Overall, salmon returning to natal streams in southern Puget Sound were found to have higher PCB concentration than fish returning to freshwater tributaries in the north (O'Neill et al. 1998). This difference is possibly due to a longer feeding period in south Puget Sound and therefore greater duration of exposure to PCB sources in central and southern Puget Sound.

The contaminants in Pacific herring are also difficult to associate with specific urban waterways/contaminated sites. However, O'Neill and West (2001) found that Pacific herring from south and central Puget Sound had significantly higher concentrations of PCBs than did northern Puget Sound/Georgia Basin stocks. As well, they observed higher PAH values in herring from central Puget Sound area compared to south and north Sound and Strait of Georgia. These results indicate a general "signature" of organic contaminants in the food web of central Puget Sound, which corresponds with what is known about spatial trends in sediment contaminants (Section 4.2.3).

4.3.2.4 Temporal Trends

Assessing temporal trends in contaminant accumulation by fish is difficult for a number of reasons, including:

- Lack of a sufficient number of data points through time;
- High within-year variability in data sets that have been collected over a sufficient period of time; and
- A strong correlation between age and/or lipid content and contaminant accumulation by some fish.

Collier et al. (1998), however, were able to compare their findings with studies done by other researchers on liver injury of English sole from the Hylebos Waterway, and found no appreciable change in the incidence of liver lesions since the 1970s, implying that exposure to PAH-contaminated sediments has not decreased. Based on results from PSAMP, contaminants in fish tissues have not changed, although there may be an increase in effects from PAHs (O'Neill, pers. comm. 2002).

A recent study of the influence of reducing exposure to PAHs on the incidence of liver lesions in English sole was conducted in Eagle Harbor where a historic contaminated site has been undergoing cleanup (PSWQAT 2000b). Remedial actions included upland source control (i.e., the installation of subsurface sheetpile wall and a groundwater pump and treat system) and capping of intertidal and subtidal sediments. Initial studies in the mid-1980s indicated that 75% of the English sole population exhibited toxicopathologic liver lesions and elevated biomarkers. Since source control and capping, the fish in Eagle Harbor have shown a significantly reduced risk of liver lesions, biliary FAC concentrations and hepatic DNA adducts.

4.3.2.5 Summary of Toxic Contaminants in Fish

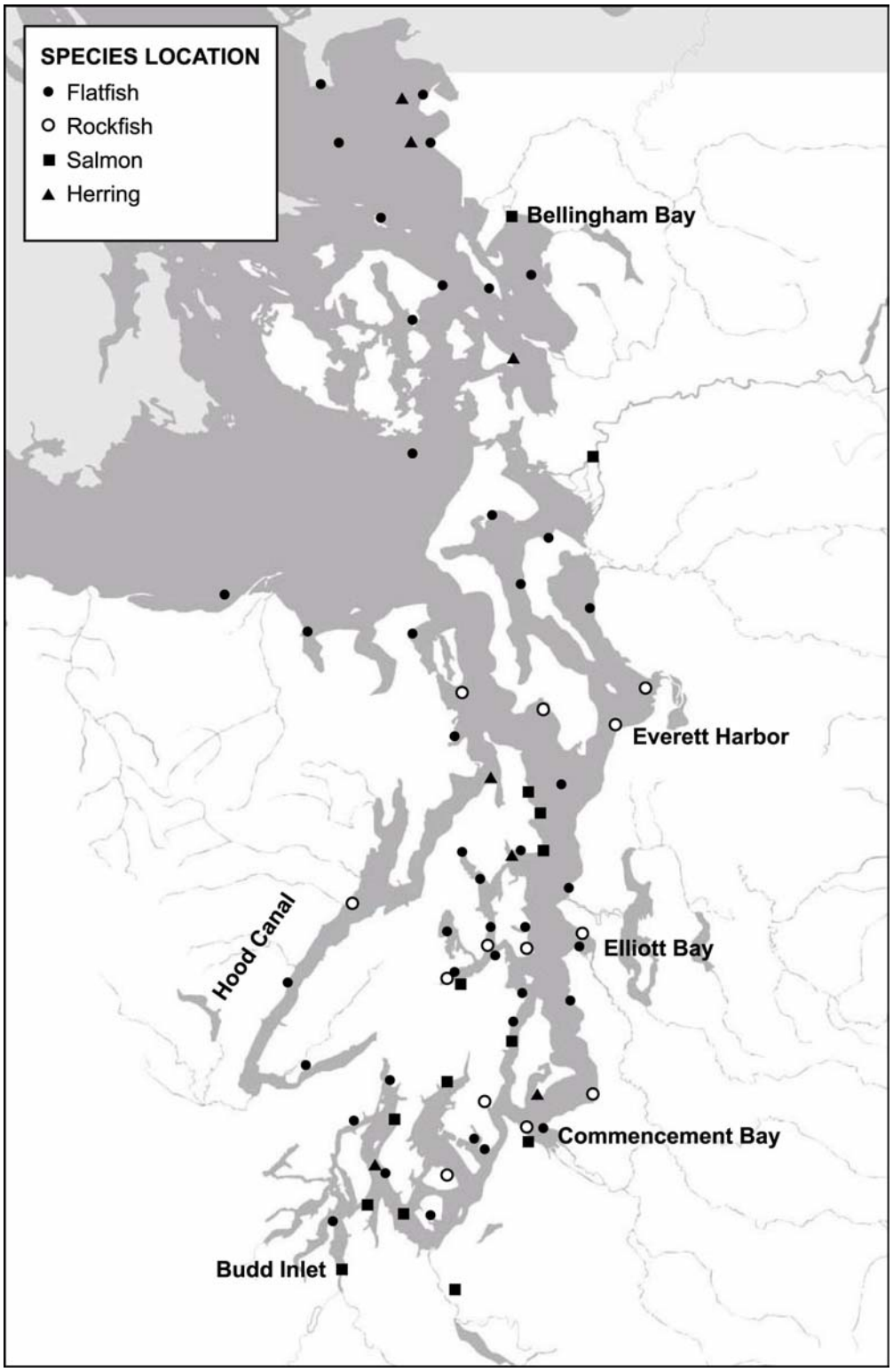
State of Knowledge

Fish tissue contaminant burdens and the toxic effects of metals and organic contaminants have been studied for numerous locations throughout Puget Sound (Figure 4-33). The PSAMP program is responsible for a large part of what is known about contaminants in fish. Other studies have addressed fish tissue contaminants on a nation-wide scale or limited to one or two embayments, Commencement and Elliott Bays in particular.

These studies have shown that:

- Contaminant burdens are species-specific and may be affected by trophic level, gender, age, and lipid content of the fish;
- Spatial trends in the tissue burden of fish is more apparent for species that are "sedentary" or which occupy a lower trophic status;
- Fish from urban and near-urban areas had the highest tissue burdens of metals and organic contaminants. One exception was for rockfish from Foulweather Bluff, a non-urban station near the outlet of Hood Canal where samples had age-specific mercury concentrations comparable to the urban embayments Elliott Bay and Sinclair Inlet (West and O'Neill 1998);
- The tissue benchmark for the protection of human health from mercury was exceeded in a few samples of quillback rockfish;

Figure 4-33: Locations at which fish have been sampled for tissue concentrations of metals and organic contaminants.



- The tissue benchmark for the protection of human health from PCB cancer risks was exceeded in almost all of the fish tissue sampled by West et al. (2001a), while only Pacific herring samples exceeded the noncarcinogenic effects level for PCB;
- Tissue benchmarks for the protection of wildlife from both carcinogenic and non-carcinogenic effects of PCB were exceeded in some fish of all the species for which data were available (West et al. 2001a);
- The highest overall PCB concentrations have been measured in Pacific herring (however, it should be noted that herring tissue burdens were assessed on a whole-body basis rather than muscle tissue only; West et al. 2001a);
- There is a signature of organic contaminants in Central Puget Sound herring (O'Neill and West 2001);
- English sole from Sinclair Inlet have higher age-specific lead concentrations than do sole from other Puget Sound stations (PSWQAT 2000);
- PCB has been detected in 100% of rockfish from urban embayments, 97% from near urban areas, and 7% from non-urban sites (West et al. 2001b);
- Female rockfish may transfer PBC to their offspring, whereas they do not transfer mercury (West and O'Neill 1998);
- Outmigrating juvenile salmon have been observed to accumulate PCBs and DDT in contaminated estuaries such as the Duwamish Waterway and Commencement Bay (Stein et al. 1995);
- PCBs are one of the few contaminants observed to accumulate in adult salmon, and adult chinook salmon have higher concentrations than do coho salmon (O'Neill et al. 1998);
- Adult salmon returning to natal streams in South Puget Sound were found to have higher PCB concentrations than fish returning to Central Puget Sound streams (O'Neill and West 1998);
- Organochlorine contaminants measured in adult salmon returning to natal streams in the Puget Sound area may be the result of exposure of the fish to the toxic compounds in the North Pacific Ocean or from Puget Sound sources. The relative contribution of contaminants accumulated by salmon as they outmigrate through contaminated estuaries during their juvenile life stage is considered negligible (O'Neill et al. 1998);
- PCB tissue benchmarks for the protection of piscivorous wildlife from cancer were exceeded in some fish samples.

Contaminant Effects

- English sole are at higher risk for developing liver lesions in areas with PAH contamination (Collier et al. 1998), while other closely related species such as starry flounder do not appear to be similarly affected (Collier, pers. comm., 2002);
- Exposure to PAH and chlorinated hydrocarbons are risk factors for several adverse reproductive effects in English sole including precocious sexual maturation, inhibition of gonadal development and decreased egg weight (Collier et al. 1998);
- The reproductive success of English sole is lower in moderately to highly contaminated sites (Johnson et al. 1994);
- Vitellogenesis has been observed in male rockfish from Elliott Bay, indicating exposure to an as of yet unidentified endocrine disrupting compound (West et al. 2001b);
- Exposure to toxic contaminants during their estuarine residence time can cause impaired immune function in juvenile salmonids (Varanasi et al. 1993);
- Chum salmon juveniles showed higher indicators of exposure to contaminants (e.g., damaged DNA; higher CYP1A activity) than chinook salmon, suggesting that chum may be more susceptible to biological injury from the contaminants (Stehr et al. 1998).

Monitoring and Data Gaps

Monitoring and data gaps related to metals and organic contaminants in the fish of Puget Sound include the following:

- The reproductive effects of many contaminants of concern are not known;
- PCBs appear to be transferred from female fish to their eggs, however, the effect of contaminant exposure via maternal transfer is unknown at this time. PSAMP is conducting a pilot study of contaminants in herring eggs (O'Neill and West 2001);
- There are not enough data to assess temporal trends in the contaminant burdens and effects in many fish species;
- The ecological significance of measured biliary-FAC concentrations in rockfish is unknown;
- The potential population effects from sublethal exposures to contaminants are not known. This may be difficult to quantify because many other factors (i.e., harvest rates, natural changes in the physical environment) contribute to population dynamics;

- There is evidence of endocrine disruption (i.e., vitellogenesis) in the male rockfish collected from Elliott Bay, however, the cause was not clear;
- The speciation of arsenic in fish, and therefore the bioavailability to mammalian consumers, is not well understood;
- The relative importance of global versus local sources of PCB to Puget Sound salmon is unknown;
- There are no regulatory tissue residue screening benchmarks for Puget Sound, either for the protection of the fish themselves or for the protection of piscivores;
- There are insufficient data regarding the presence/composition of individual PCB or dioxin congeners in fish tissue to support human health risk assessments;
- The relationship between sediment and tissue concentrations is poorly understood, including the preferential uptake of different contaminant forms (e.g., PCB and dioxin congeners).

4.3.3 Marine Mammals

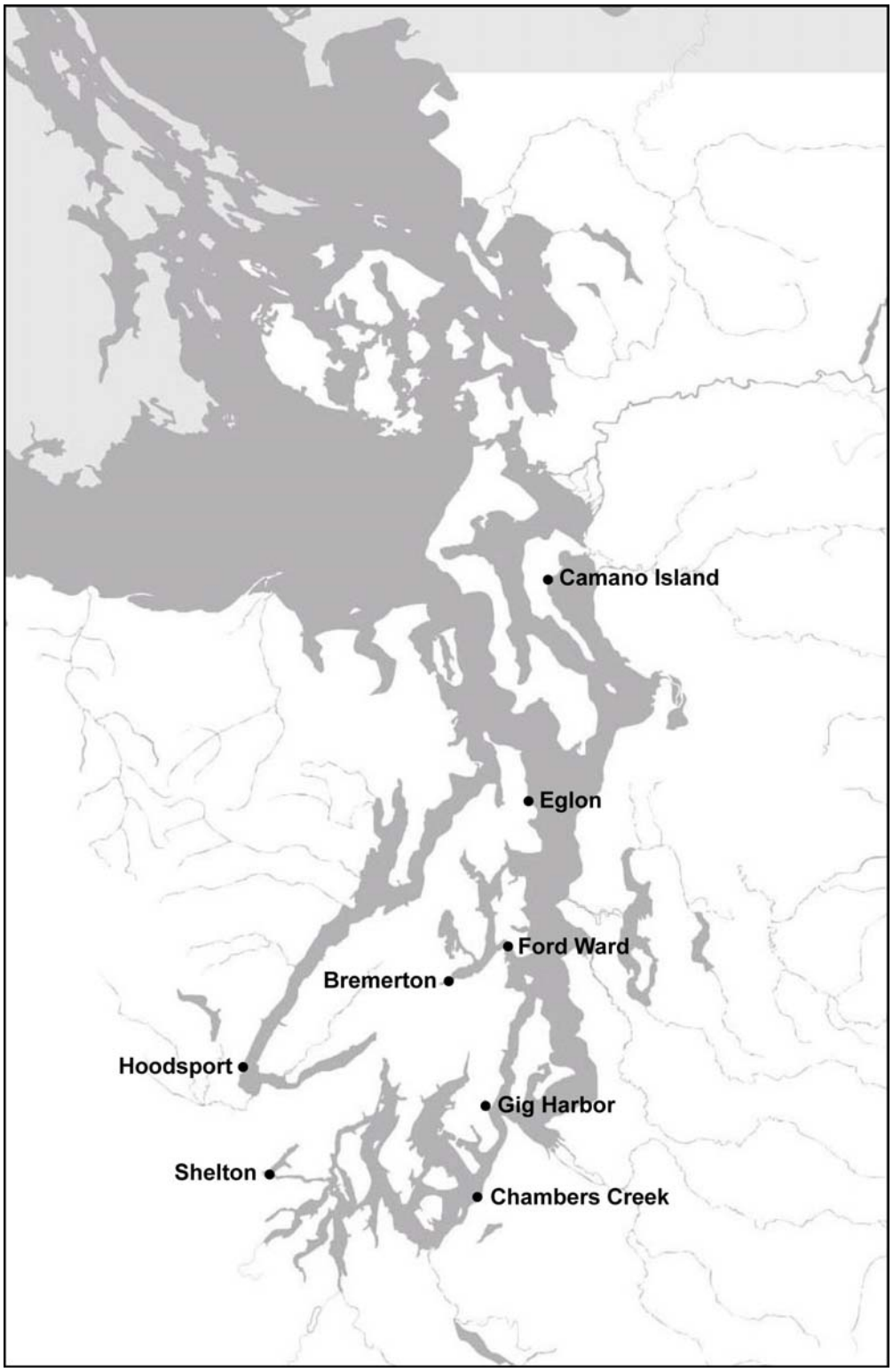
4.3.3.1 Review of Existing Data

Carnivorous mammals such as harbor seals and killer whales occupy the top of the marine food chain and are therefore good indicators of the continued presence of persistent, bioaccumulative toxics such as PCBs and organochlorine pesticides throughout the marine environment, even decades after the chemicals were banned from use in the United States.

River Otters

The livers from 22 river otters trapped at eight locations in Puget Sound (Figure 4-34) were analyzed for 17 metals, 20 organochlorines, 47 PCB congeners, 10 dioxins and 12 furans as part of an overall study investigating the potential for endocrine disruption in young male otters (Grove et al. 2001). River otters are top predators and eat a wide variety of fish and invertebrate species; furthermore, they are non-migratory. Therefore, they are considered to be a useful sentinel species for assessing the exposure of aquatic wildlife to and the effects of harmful contaminants. The results for mercury, cadmium, butyltins, DDE, PCBs, and dioxins were reported, however, there was no interpretive analysis of the data so the ecological significance of the findings is not yet known.

Figure 4-34: Sampling locations for river otters collected in Puget Sound (Grove et al. 2001).



Mercury was detected in the livers of all individuals and concentrations ranged from ~2 ppm dw in an age class 0 otter to ~31 ppm dw in an age class 2+ otter, both from Fort Ward. Cadmium was only detected in adult (age class 2+) otters with the exception of one age class 1 individual from Hoodsport, in which the highest concentration (~1.8 ppm dw) was found. Total butyltin concentrations ranged from ~50 $ng \cdot g^{-1}$ ww at the reference location to ~2,600 $ng \cdot g^{-1}$ ww near Fort Ward. Of the three butyltin species measured in Puget Sound river otters, very low concentrations of TBT were detected, compared to di- and monobutyltin. In comparison, the total butyltin concentrations measured in the livers of sea otters found dead on the California coast between San Francisco and Santa Barbara ranged from 40 to 9,200 $ng \cdot g^{-1}$ ww and tributyltin was the predominant form of the compound measured, suggesting recent exposures to butyltin sources (Kannan et al. 1998). Butyltin concentrations were highest in sea otters that died of infectious diseases.

DDE liver concentrations were less than 50 ppb ww for all age classes at most sites (Grove et al. 2001). In comparison, the concentrations in age class 2+ river otters at the Bremerton sampling location ranged from ~25 ppb ww to ~270 ppb ww, while an age class 0 river otter from Camano Island had a concentration of ~100 ppb ww. PCB concentrations were also highest in an age class 2+ otter from Bremerton (Table 4-18) regardless of the congener reported (i.e., total; Aroclor 1254:1260; individual congeners), with the exception of PCB 126, which was highest in an otter from Chambers Creek. Dioxin concentrations varied geographically, depending on the congener reported. For example, 2,3,7,8-TCDD liver concentration (detected in only five animals) was highest in an age class 2+ river otter from Eglon (Central Puget Sound). OCDD was detected in the largest number of animals and the highest concentrations were in age class 2+ otters from Fort Ward (Central Puget Sound) and Chambers Creek (South Puget Sound).

Harbor Seals

Organochlorine contaminants have been monitored in harbor seals in a number of studies. In 1990, Hong et al. (1996) collected blubber and liver samples from eight pups found dead on Smith Island, where Puget Sound and the Strait of Juan de Fuca meet, and Gertrude Island in southern Puget Sound. The tissue samples were analyzed for PCBs, *p,p'*-DDE, hexachlorobenzene and mirex. Total PCB concentrations in blubber ranged from 1.3 ppm ww at Smith Island to 16 ppm ww at Gertrude Island (Table 4-18). Ross et al. (1998) conducted biopsies on healthy, young harbor seals at Gertrude Island in 1996 to collect blubber for PCB, PCDD and PCDF analysis and found an average PCB concentration of 16.9 ppm lw. The authors also converted the concentration of the dioxin-like PCBs, PCDDs and PCDF detected to total Toxic Equivalents (TEQs); PCBs were found to contribute about 95% of the TEQ in harbor seal blubber. The ecological relevance of these findings are described in Ross et al. (1996), who summarize captive feeding studies in which the immune system was found to be compromised in harbor seals fed polyhalogenated aromatic hydrocarbon-contaminated (i.e., PCB, PCDD, PCDF) herring. The total PCB concentration in the blubber of the test animals in which

immunosuppression was observed was found to be 17 $mg \cdot kg^{-1}$ on a lipid weight (lw) basis. Based on known PCB concentrations in blubber, Ross et al. (1996) identified the harbor seal population of Puget Sound to be at risk to impaired immune function.

Metals concentrations in livers and kidneys of dead harbor seals collected in Puget Sound were analyzed in the 1980s and found to be within the range measured in pinnipeds from other areas of the world (Calambokidis et al. 1984, cited in Calambokidis et al. 1991).

Table 4-18: Summary of PCB concentrations in marine mammals in Puget Sound.

SPECIES/LOCATION	GENDER	AGE	TISSUE	CONC.[a]	UNITS	SOURCE
River Otters						
Bremerton	Male	Young	Liver	3.8 (0.4-10.1)	$\mu g \cdot kg^{-1}$ ww	Grove et al. 2001
Fort Ward	Male	Young	Liver	0.5 (0.2-0.9)	$\mu g \cdot kg^{-1}$ ww	Grove et al. 2001
Eglon	Male	Young	Liver	0.2	$\mu g \cdot kg^{-1}$ ww	Grove et al. 2001
Gig Harbor	Male	Young	Liver	0.4 (0.3-0.5)	$\mu g \cdot kg^{-1}$ ww	Grove et al. 2001
Hoodsport	Male	Young	Liver	0.1	$\mu g \cdot kg^{-1}$ ww	Grove et al. 2001
Harbor Seals						
Smith Island	NA	Pup	Blubber	1.7 (1.3-2.1)	$mg \cdot kg^{-1}$ ww	Hong et al. 1996
Smith Island	NA	Pup	Liver	9 (6-14)	$\mu g \cdot kg^{-1}$ ww	Hong et al. 1996
Gertrude Island	NA	Pup	Blubber	13.1 (9.2-16)	$mg \cdot kg^{-1}$ ww	Hong et al. 1996
Gertrude Island	NA	Pup	Liver	490 (290-970)	$\mu g \cdot kg^{-1}$ ww	Hong et al. 1996
Gertrude Island	NA	Pup	Blubber	16.9 (±2.90)	$mg \cdot kg^{-1}$ lw	Ross et al. 1998
Killer Whale						
Southern residents	Male	Adult	Blubber	146.3 (± 32.7)	$mg \cdot kg^{-1}$ lw	Ross et al. 2000
Southern residents	Female	Adult	Blubber	55.4 (± 19.3)	$mg \cdot kg^{-1}$ lw	Ross et al. 2000
Transients	Male	Adult	Blubber	251.2 (± 54.7)	$mg \cdot kg^{-1}$ lw	Ross et al. 2000
Transients	Female	Adult	Blubber	58.8 (± 20.6)	$mg \cdot kg^{-1}$ lw	Ross et al. 2000

[a] Concentrations reported as means with range or standard error of the mean in brackets.
ww - wet weight
lw - lipid weight
NA - not available

Gray Whales

Tissue samples and stomach contents from dead gray whales beached in California, Washington and Alaska were analyzed for a number of contaminants including chlorinated hydrocarbons, polycyclic aromatic hydrocarbons and metals (Varanasi et al. 1994). Mercury concentrations in liver tissue were up to 100 $ng \cdot g^{-1}$ ww, reported for a whale stranded on Hartstene Island in southern Puget Sound, and 120 $ng \cdot g^{-1}$ ww in a whale stranded near Lyre River in the Strait of Juan de Fuca. Varanasi et al. (1994) determined that these results were low compared to mercury concentrations in the livers

of other marine mammals. Gray whales filter feed bottom sediments for benthic invertebrates. The relative exposure of gray whales to metals contaminants compared to whales that are top predators would depend in part on the bioavailability of a contaminant such as mercury in sediments. The concentrations of copper, cadmium and lead in liver and kidney tissue were similar for whales from all sampling locations. All of the gray whales sampled by Varanasi et al. (1994) had detectable levels of chlorinated hydrocarbons. However, there were no regional differences in organics levels in tissues, and the authors concluded that the gray whale was not useful as a sentinel species due to considerable migratory distances they travel and the short period of time actually spent in Puget Sound.

Killer Whales

Three discrete killer whale communities inhabit the coastal waters of Washington, Alaska and British Columbia: southern and northern residents which eat primarily salmon, and transient whales which eat primarily marine mammals. The southern resident community ranges from Puget Sound to midway up both the west and east coasts of Vancouver Island. In order to assess differences in contaminant levels of the different killer whale communities, Ross et al. (2000) collected blubber biopsy samples and analyzed them for PCBs, dioxins and furans. Total PCB concentrations (Table 4-18) were higher in all samples than those detected in marine mammals from other parts of the world, including the industrialized areas of eastern North America and northern Europe. The total PCB concentrations in blubber were also higher than the threshold concentration in harbor seals above which immune system effects were observed (Ross et al. 1996). Conversely, total dioxin and furan concentrations in both the southern resident and transient killer whale populations were considered to be low (Ross et al. 2000). The authors concluded that the elevated PCB levels observed in these whale communities may make them more vulnerable to disease, compounding the potential effects of other stressors such as increased human activities in the marine waters of Washington and British Columbia and decreased food supply.

Age and gender appear to have an effect on the total PCB concentrations measured in killer whales (Ross et al. 2000). PCBs were initially variable and increasing in immature male and female whales. PCB levels in male whales continued to be variable and increased with age. When the females reached reproductive age (around 15 years old), however, PCB concentrations decreased and remained low until the whales reached reproductive senescence, after which time PCBs began accumulating again. A majority of the PCB burden in female whales of reproductive age was transferred to their offspring.

The ultimate pathway by which PCBs accumulate in killer whales is not clear. Chinook salmon comprise a significant portion of the diet of southern residents (Ford et al. 1998), and as discussed in Section 4.3.2.1, chinook salmon captured in southern Puget Sound had elevated PCB concentrations. However, chinook salmon spend a majority of their

adult life in the Pacific Ocean and Ross et al. (2000) suggested they are exposed via atmospheric deposition and trophic transfer of PCBs in the North Pacific.

Information regarding the concentrations and effects of metals contaminants in killer whales was not available.

4.3.3.2 Comparison to Tissue Screening Benchmarks

Tissue quality screening benchmarks do not exist for marine mammals. However, studies have indicated that immune dysfunction occurs in harbor seals at PCB concentrations of 17 $mg \cdot kg^{-1}$ lw in blubber. Based on this value, Puget Sound harbor seals and killer whales are at risk of immunosuppression (Ross et al. 1996).

4.3.3.3 Spatial Trends

The spatial variation of contaminant burdens in Puget Sound River otters depended on the compound, and in some cases the specific congener (Grove et al. 2001). Of the metals contaminants detected, butyltin concentrations were consistent throughout Puget Sound, although higher than at the reference station. Mercury concentrations were highest in otters from Fort Ward in Central Puget Sound, while cadmium was highest in an otter from Hood Canal. Chlorinated organic contaminants were generally highest in otters from near Bremerton in Sinclair Inlet, although 2,3,7,8-TCDD was highest in an otter from Eglon. Total H7CDD and OCDD were highest in otters from Fort Ward in Central Puget Sound and Chambers Creek in South Puget Sound.

The blubber in seals from Gertrude Island had significantly higher concentrations of total PCBs, coplanar PCBs, *p,p'*-DDE and mirex than seals from Smith Island (Hong et al. 1996; Table 4-18). The study did not include an assessment of diet, however, the authors suggested two possible reasons for the different contaminant burdens observed: (1) seals from each island may be predisposed to different levels of PCB exposure through availability of different prey species that are either more or less contaminated; (2) alternatively, the seals could be consuming similar prey items, and proximity to contaminant sources is controlling exposure. In either case, the data suggest greater trophic transfer of organochlorine contaminants to harbor seals in Puget Sound proper than further north. Further evidence of this is provided by Ross et al. (2001) who found a geographical decrease in PCB concentrations in the blubber of seals from south in Washington to north in British Columbia (Puget Sound – 18 $mg \cdot kg^{-1}$ lw; Strait of Georgia – 2.5 $mg \cdot kg^{-1}$ lw; Queen Charlotte Sound 1.1 $mg \cdot kg^{-1}$ lw).

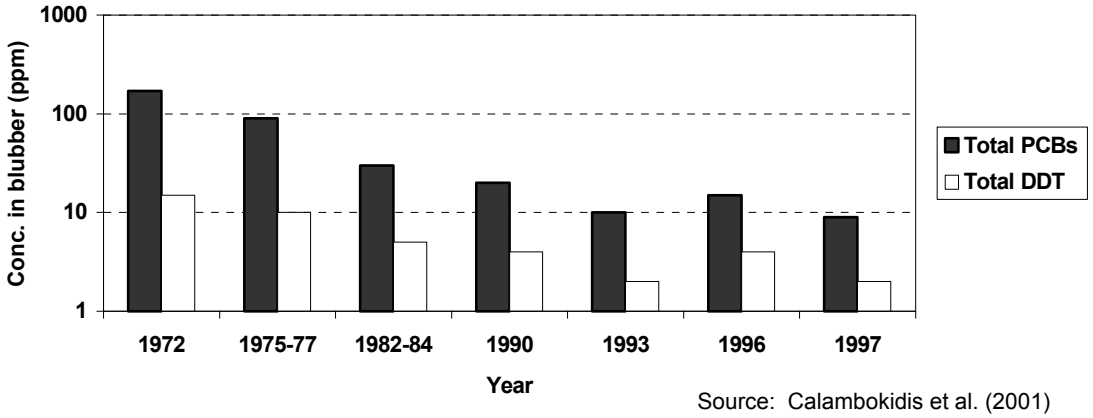
Of the three killer whale populations inhabiting the west coast of North America, transients (males in particular) had the highest blubber concentrations of PCBs, followed by southern residents, which spend part of their time in Puget Sound, and northern residents, which are not found in Puget Sound waters (Ross et al. 2000). Dioxin and furan

concentrations were low in all three populations and not significantly different from each other.

4.3.3.4 Temporal Trends

Calambokidis et al. (2001) compared PCB and dioxin concentrations in harbor seals collected at four to five year intervals starting in 1972. The study involved a comparison of analytical techniques, re-analysis of archived samples, and inter-laboratory variability to ensure that pooling current and earlier data was appropriate. Historical analyses of samples were found to be consistent with results using modern methods, therefore the researchers decided that all the data could be pooled for a trend analysis. The results indicated that PCBs and DDT concentrations declined in the 1970s but have now stabilized (Figure 4-35). As discussed earlier, however, organochlorine residues in harbor seals remain at levels known to cause immune dysfunction. The temporal trend of PCBs in the mammals mirrors that observed in the Mussel Watch program (Section 4.3.1.3), indicating that while PCB concentrations have decreased, they continue to be persistent in the food web.

Figure 4-35: Trends in PCB and DDT concentrations in Puget Sound harbor seal blubber between 1972 and 1997.



Source: Calambokidis et al. (2001)

Not enough data were available to determine temporal trends of contaminants in killer whales. However, anecdotal evidence indicates that the highest PCB concentrations measured in a killer whale to date were in an animal that was found beached on the Olympic Peninsula in January of 2002 (CBC 2002). The whale reportedly had a PCB concentration 25% greater than any observed before. Furthermore, the results were unusual because the whale was a female of reproductive age, which usually transfer organic contaminants to their offspring.

4.3.3.5 Summary of Toxic Contaminants in Marine Mammals

State of Knowledge

Metals and organic contaminant concentrations have been measured in marine mammal tissue (i.e., liver, blubber) at a number of locations throughout Puget Sound (Figure 4-36). Southern resident killer whales have been sampled from the Straits of Juan de Fuca and Georgia. The number of studies on marine mammals is small compared to other biota.

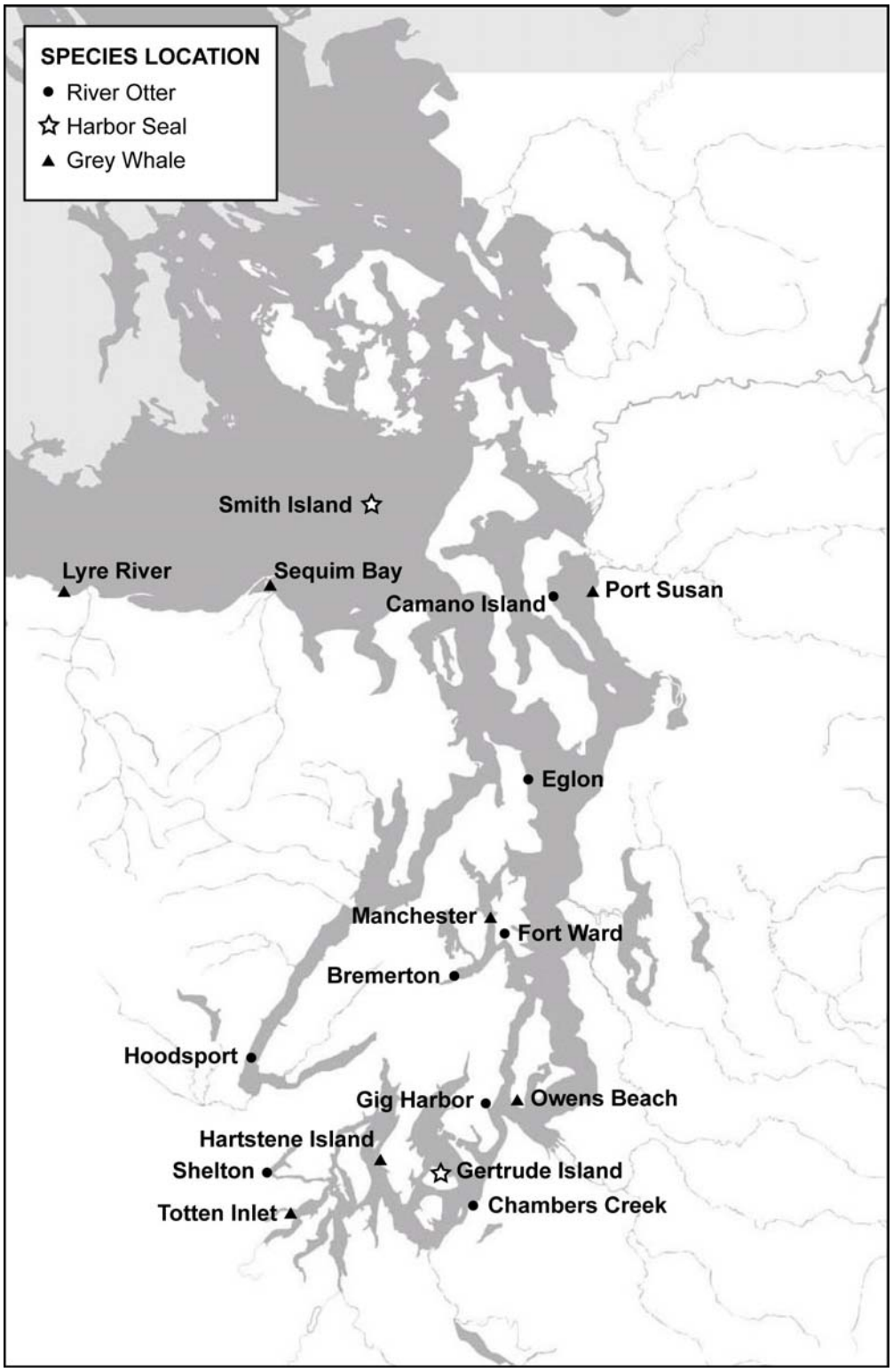
These studies have indicated that:

- Metals concentrations in harbor seals were found to be comparable to pinnipeds in other parts of the world (Calambokidis et al. 1984, cited in Calambokidis et al. 1991);
- The concentration of PCBs in killer whales inhabiting Puget Sound and the Washington coast is higher than in the whales from any other industrialized area in North America and northern Europe (Ross et al. 2000);
- Dioxin and furan concentrations are low in killer whales (Ross et al. 2000);
- PCB concentrations in harbor seals have decreased since the 1970s but now appear to have stabilized (Calambokidis et al. 2001);
- Gray whales, which filter sediments for benthic invertebrates, were found to have lower contaminant concentrations relative to carnivorous marine mammals;
- Gray whales have detectable levels of chlorinated hydrocarbons, however, no regional differences were found due to the migratory patters of the species (Varanasi et al. 1994).

Contaminant Effects

- Harbor seals in Puget Sound are at risk for impaired immune function due to PCBs (Ross et al. 1996);
- The concentration of PCBs in the blubber of killer whales frequenting Puget Sound waters exceeds the threshold above which immunosuppression may be expected (Ross et al. 1996).

Figure 4-36: Locations at which marine mammals have been sampled for tissue concentrations of metals and organic contaminants



Monitoring and Data Gaps

Monitoring and data gaps related to metals and organic contaminants in the marine mammals of Puget Sound include the following:

- There are insufficient data to determine temporal trends of persistent, bioaccumulative toxics (PBTs) like PCBs and other organochlorine compounds in killer whales;
- Further investigation is required to determine the relative importance of local (i.e., contaminated sites in Puget Sound) versus international (i.e., global atmospheric transport) sources of persistent bioaccumulative toxics like PCBs to higher trophic consumers such as killer whales;
- The mechanisms by which PBTs affect marine mammals and the risks associated with current contaminant burdens are not fully understood (Ross, pers. comm. 2002);
- Congener-specific data for sediment, water, air and biota, which would behave as a "fingerprint" and help delineate sources and transportation of PBTs in Puget Sound are not available (Ross, pers. comm. 2002);
- There is a lack of information regarding the sources, transport and fate of unregulated PBTs, particularly related to higher trophic level species such as seals, whales and human consumer groups (Ross, pers. comm. 2002).

4.3.4 Birds

4.3.4.1 Review of Existing Data

Few studies have been conducted on contaminants in the marine birds of Puget Sound, in part because finding an appropriate sentinel species (i.e., non-migratory; spends a substantial portion of its life cycle in Puget Sound; forages on marine prey species; is abundant throughout Puget Sound) is difficult (Mahaffy, pers. comm. 2002). As part of PSAMP, the U.S. Fish and Wildlife Service (USFWS) and Washington Department of Fish and Wildlife (WDFW) evaluated pigeon guillemots, great blue heron and surf scoters against these criteria and found the scoters to be the only suitable sentinel species (Mahaffy et al. 1997). Surf scoters are migratory birds that breed in the northern boreal forests of North America and overwinter along both Pacific and Atlantic coasts. The Puget Sound population of this species feeds exclusively in the marine environment, primarily on benthic organisms.

Surf scoters were collected from Commencement Bay in October at the start of their overwintering period, and again in February to determine if they accumulated metals or organochlorine compounds (Mahaffy et al. 1997). Biliary FACs were measured to determine exposure to PAHs. The late winter samples of surf scoters had significantly

higher liver concentrations of mercury than the fall samples, while copper, cadmium, and zinc decreased significantly during the overwintering period (Table 4-19).

Table 4-19: Summary of selected metals and organic contaminant concentrations in the livers of surf scoters sampled from Commencement Bay in fall and late winter (1995-1996).

		SAMPLING PERIOD			
PARAMETER	UNITS	FALL (N=20)[a]	FOD[b]	LATE WINTER (N=20)	FOD
Metals					
Arsenic	ppm dw	2.6 (0.84-4.3)	NR [c]	2.9 (0.73-6.6)	NR
Cadmium*	ppm dw	8.3 (2.6-18)	NR	6.0 (2.3-9.9)	NR
Copper*	ppm dw	81 (45-160)	NR	45 (31-71)	NR
Lead		ND [d]	NR	ND	NR
Mercury*	ppm dw	2.1 (0.82-14)	NR	2.6 (1.2-6.9)	NR
Organochlorines					
p,p'-DDE	ppb ww	NC [e] (ND – 54)	55	25 (ND – 54)	95
Total PCBs	ppb ww	63 (ND – 96)	65	70 (NC – 120)	85
Biliary FACs					
Naphthalene	ppt [f]	22,000 (7,200-62,000)	NR	24,000 (6,000-68,000)	NR
Phenanthrene	ppt	9,300 (2,900-28,000)	NR	11,000 (4,700-29,000)	NR
Benzo[a]pyrene	ppt	160 (41 – 350)	NR	190 (87 – 410)	NR

Source: Mahaffy et al. (1997)
[a] Concentrations reported as mean with range in brackets
[b] FOD = frequency of detection in %
[c] NR = not reported
[d] ND = not detected
[e] NC = not calculated. Mean was not calculated when frequency of detection was < 60%
[f] equivalents per gram of bile
* Fall sampling period significantly different than late winter sampling period (at $p < 0.05$)

Mercury accumulation also occurred in surf scoters sampled from Commencement Bay in the mid-1980s (Mahaffy et al. 1997). PCBs and *p,p*'-DDE were the only organochlorine compounds detected in the birds, and both were detected in a greater number of birds during the late winter sampling period than in the fall. While concentrations of these contaminants were higher in the late winter than the fall, the difference was not significant. Biliary FACs were also slightly higher in the late winter samples, but were not significantly different than the fall samples. Organic contaminant levels were found to be lower than threshold concentrations above which effects are documented and histopathological analysis indicated that the surf scoters were in good health. The authors concluded that contaminants did not appear to be adversely affecting the birds, however, they recommended further sample collection from other areas in

Puget Sound to increase the overall sample size and to provide a comparison between birds overwintering in different locations.

With the advent of persistent and bioaccumulative chemicals such as DDT, eagle populations in the United States declined dramatically. Since the use of DDT was banned in the 1970s, eagle numbers have slowly increased and recovery goals for successfully occupied territories and number of young per occupied territory have been met for most of Washington State, with the exception of Hood Canal and a few other locations (Mahaffy et al. 2001). In order to determine if environmental contaminants were still influencing eagle productivity in Hood Canal, USFWS and WDFW initiated a study to measure PCB, DDT and dioxin/furan concentrations in bald eagle eggs, and to evaluate the source of contaminants (Mahaffy et al. 2001). Addled eggs were collected from nests in Hood Canal territories with a history of low productivity, and when available from nests outside of Hood Canal (i.e., the outer coast of Washington or the Skagit River) for comparison. Almost all of the eggs collected from Hood Canal nests had thinner shells (6.1% thinner) than the mean eggshell thickness of bald eagle eggs collected prior to the use of DDT, and all of the eggs collected either in or outside of Hood Canal had detectable levels of PCBs and *p,p'*-DDE. The mean total PCB concentrations (as determined by congener-specific analysis) in Hood Canal bald eagle eggs was 6,490 $ng \cdot g^{-1}$ fw (range = 3,660-13,100 $ng \cdot g^{-1}$), compared to a mean of 2,680 $ng \cdot g^{-1}$ fw (range = 1,130-5,400 $ng \cdot g^{-1}$) for eggs from sites outside of Hood Canal. The concentrations measured in all of the Hood Canal eggs exceeded reported threshold concentrations (i.e., 4.0 $\mu g \cdot g^{-1}$ fw) above which effects may be observed. Conversely, DDE and dioxin/furan concentrations were lower than those reported to affect productivity. The researchers determined that sediment contaminant and fish tissue concentrations were not high enough in Hood Canal to account for the elevated PCBs measured in the eggs and recommended that in future studies, other prey items such as marine mammals and fish-eating birds be assessed as potential sources.

Other bird species that have been studied for metals and organic contaminants in the past include western grebes, great blue herons, Pigeon guillemots, glaucous-winged gulls, double-crested and pelagic cormorants. Western grebes, piscivorous birds that overwinter in Puget Sound, were collected from Commencement Bay between October 1985 and February 1986 and analyzed for metals, DDE, PCB and chlordanes (Henny et al. 1990). The birds were found to accumulate mercury, arsenic, potentially cadmium, DDE, PCBs, and chlordanes while wintering in Commencement Bay. The toxicological significance of the increased body burdens of these contaminants was unknown as the birds appeared to be in good health. Little is known about critical body residues or sublethal effects of the metal and organic contaminants on this bird species. Grebes were common prey items for Bald eagles nesting in Puget Sound (Knight et al. 1990), therefore, there is potential for further trophic transfer of contaminants.

Speich et al. (1992) collected eggs from double-crested cormorants, pelagic cormorants, great blue herons, glaucous-winged gulls and pigeon guillemots from throughout Puget

Sound to determine the effects of organochlorine residues on eggshell thinning. The eggshells of the cormorants and pigeon guillemots were not significantly different than those measured prior to 1947 (i.e., pre-DDT production). In comparison, the eggs of great blue herons were significantly thinner than prior to 1947, in both industrial and agricultural areas. Table 4-20 summarizes the PCB and DDT concentrations in eggs from six Puget Sound heronries. Eggshell thicknesses were not correlated with PCB concentrations, while there was a significant, though weak, negative correlation with DDT concentrations. Organochlorine residues at the concentrations measured did not appear to have an adverse effect on reproductive success (Speich et al. 1992; Fitzner et al. 1988). While eggshell thickness in glaucous-winged gulls was also significantly lower than pre-1947 values, the concentrations of PCB and DDT were not as high as in herons from similar locations (i.e., Smith Island, Seattle, Tacoma). Neither PCB nor DDT concentrations were correlated with eggshell thickness, and Speich et al. (1992) concluded that other factors were contributing the eggshell thinning.

Table 4-20: Summary of PCB and total DDT concentrations in great blue heron eggs from Puget Sound.

	PCBs		Total DDT	
Location	N[a]	Concentration[b]	N	Concentration
Northern Puget Sound				
Samish Island	5	2.90 (0.98-7.1)	5	0.72 (0.24-1.4)
March Point	5	1.8 (1.1-2.7)	5	1.6 (0.09-6.3)
Central Puget Sound				
Seattle	6	15 (6.3-37)	6	2.2 (0.95-4.3)
Southern Puget Sound				
Tacoma	5	5.5 (3.3-8.6)	5	1.6 (1.3-1.9)
Nisqually	5	1.4 (0.90-2.1)	5	0.42 (0.13-0.80)
Totten Inlet	4	1.4 (0.81-2.2)	4	0.35 (0.05-0.72)

Source: Speich et al. 1992

[a] N = number of samples

[b] geometric mean with 95% confidence intervals in brackets; reported as $\mu g \cdot g^{-1}$ ww

Metals concentrations have also been measured in great blue herons. Eggs from nests, hatchlings, and eggs with developed embryos on the ground were collected from a colony near Fort Lewis in southern Puget Sound, as well as colonies in southeastern Washington and northern Idaho (Blus et al. 1985). The observed tissue concentrations are summarized in Table 4-21. Despite the proximity of the Fort Lewis colony to a smelter operating in Tacoma at the time of the study, the birds did not appear to accumulate metals in concentrations high enough to affect reproductive success.

Table 4-21: Summary of metals concentrations in great blue heron eggs, advanced embryos/hatchlings and near fledglings from the Fort Lewis colony in 1981 and 1982.

		METAL CONCENTRATIONS[a]				
AGE	N	ARSENIC	CADMIUM	COPPER	LEAD	MERCURY
Egg	3	ND[b]	NA[c]	0.46 (0.29-0.5)	NA	0.06 (0.04-0.12)
Advanced embryos and hatchlings [d]	1	1.1	0.22	1.1	NA	0.06
Near fledgling [e]	3	26.1 (9.9-90)	NA	65.0 (57-83)	ND	1.14 (0.82-1.5)

Source: Blus et al. (1985)
[a] concentrations reported as geometric means with range in brackets; $\mu g \cdot g^{-1}$ ww
[b] ND = not detected
[c] NA = not analyzed
[d] Whole body analyzed
[e] Liver only analyzed

4.3.4.2 Comparison to Tissue Screening Benchmarks

There are no tissue screening benchmarks for assessing potential contaminant effects in birds. However, researchers have estimated a total PCB threshold of > 4.0 - 6.0 $\mu g \cdot g^{-1}$ fw in eagle eggs, above which reproduction and population effects may be expected. Eggs collected from Hood Canal between 1992 and 1994 exceeded this threshold (Mahaffy et al. 2001).

For DDT, the threshold above which reproductive and population effects may occur is ~3.6 $\mu g \cdot g^{-1}$ fw. Almost all of the eagle eggs collected from Hood Canal by Mahaffy et al. (2001) were below this threshold.

4.3.4.3 Spatial Trends

A majority of the studies on contaminants in birds have focused on specific locations with known contamination such as Commencement Bay in Tacoma. The only study in which contaminants were measured throughout Puget Sound was the one done by Speich et al. (1992). As Table 4-20 shows, PCB and DDT concentrations were highest in eggs from great blue heron colonies located in Seattle and Tacoma. The greatest adverse effects, however, were detected in northern Puget Sound where the land use was predominantly agricultural.

4.3.4.4 Temporal Trends

Migratory birds in Commencement Bay were found to accumulate metals and organic contaminants during their overwintering period in the bay (Mahaffy et al. 1997; Henny et

al. 1990). Not enough data were available to assess long term temporal trends in contaminant burdens in Puget Sound birds.

4.3.4.5 Summary of Toxic Contaminants in Birds

State of Knowledge

Tissue concentrations of metals and organic contaminants have been measured in birds from several locations in Hood Canal and South Puget Sound, with several of the studies focusing on contaminant accumulation during overwintering in Commencement Bay (Figure 4-37).

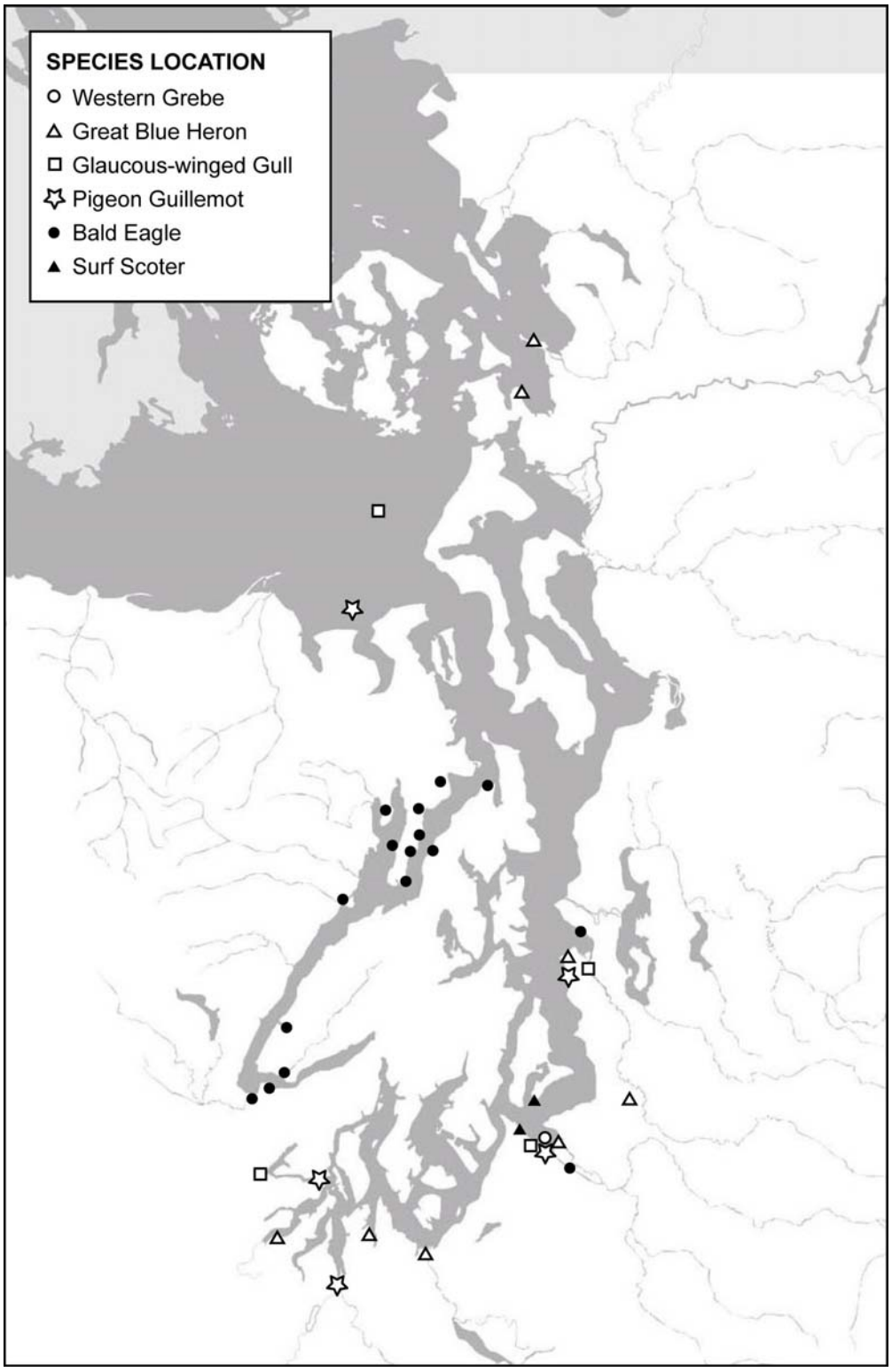
These studies have shown that:

- Surf scoters have been determined to be a useful sentinel species because they are non-migratory; they spend a substantial portion of their life cycle in Puget Sound; they forage on marine prey species; and they are abundant throughout Puget Sound (Mahaffy, pers. comm. 2002);
- Surf scoters overwintering in Commencement Bay accumulated mercury, but without apparent effects (Mahaffy et al. 1997);
- Eagle populations have been increasing throughout Washington State except in the Hood Canal area (Mahaffy et al. 2001);
- Birds from a heronry near a Tacoma smelter did not appear to accumulate metals (Blus et al. 1985);
- PCB concentrations in heron eggs are highest at a heronry near Seattle (Speich et al. 1992);
- The greatest adverse effects observed in herons from agricultural areas in North Puget Sound (Speich et al. 1992).

Contaminant Effects

- Despite the fact that birds overwintering in Commencement Bay (i.e., surf scoters, western grebes) accumulated metals and organic contaminants, they appear to be in good health;
- PCB concentrations in eagle eggs collected from Hood Canal exceeded the threshold level above which adverse reproductive effects have been observed to occur;
- Great blue herons and glaucous-winged gulls have experienced egg-shell thinning, however, PCB and DDT concentrations were not correlated with egg-shell thickness.

Figure 4-37: Locations from which birds have been sampled for tissue concentrations of metals and organic contaminants



Monitoring and Data Gaps

Gaps in data regarding metals and organic contaminants in the birds of Puget sound include the following:

- The source of PCBs in the eagles of Hood Canal is not known as sediment and fish tissue concentration were found to not be high enough to account for the elevated egg concentrations;
- The potential ecological significance of contaminant accumulation in migratory birds overwintering in urban estuaries is unknown as the birds appear to be in good health;
- There are not enough data to assess the spatial variability in contaminant uptake by birds utilizing urban/industrial estuaries versus more isolated estuaries;
- The sublethal effects of contaminants measured in birds is unknown;
- There are not enough data to assess the relationship between contaminant burdens and eggshell thinning in Puget Sound waterfowl;
- There are no Washington State or Puget Sound-specific tissue residue screening benchmarks for birds.

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5. STATUS OF TOXIC CONTAMINANTS IN PUGET SOUND

As shown in Section 4, numerous studies regarding the concentrations in and effects of toxic metals and organic contaminants on Puget Sound's marine ecosystem have been conducted. None of these studies, however, have provided a comprehensive assessment for the entire Sound. One of the difficulties in making such an assessment of the overall behavior and effects of contaminants in the Puget Sound environment using the available studies is that data for different environmental compartments (i.e., sediment, water, invertebrates, fish) are infrequently collected synoptically (i.e., from each compartment at the same time and from the same location). Furthermore, there is high variability in the tolerance of different species as well as the conditions under which toxicity is manifested.

In the absence of a comprehensive study, four data sets representing different environmental compartments, including sediment, mussels, flatfish and rockfish (Table 5-1), were compared to illustrate the broader geographical distribution of the selected contaminants as well as the potential for transfer of contaminants between trophic levels. These four components were chosen because:

- They collectively represent different exposure and trophic levels;
- Large numbers data for them have been collected from throughout Puget Sound;
- The datasets were available electronically.

Table 5-1: Summary of data used to assess overall status of toxic metals and organic contaminants in the Puget Sound environment

GRAPH	EXAMPLE OF	DESCRIPTION OF DATASET	SOURCE
A Sediment	Primary contaminant source to aquatic biota	Data collected between 1997 and 1999	PSAMP-NOAA 2002
B Mussels	Filter feeding organisms, good integrators of water-borne contaminants	All data collected between 1986 and 1998	Mussel Watch 1998
C Flatfish	Associated with benthic environment, low trophic level	All data collected between 1989 and 1999, English sole and starry flounder data pooled	West et al. 2001a
D Rockfish	Associated with benthic environment, high trophic level, long-lived	All data collected between 1989 and 1999, quillback, brown, copper, and yelloweye rockfish data pooled	West et al. 2001a

A series of figures were prepared using these datasets (Figures 5-1 through 5-10). The data on each graph have been grouped by Puget Sound region (e.g., North, Central, South, Hood Canal), arranged by rank (i.e., ascending order), and compared to available

numerical screening benchmarks. To provide clarity, the graphs are annotated with location names.

This exercise was not intended to be a risk assessment for either aquatic organisms inhabiting Puget Sound or human consumers of fish and shellfish from the Sound. Rather, it was conducted to provide some context for the information presented in Section 4 as well as a general overview of the relative concern related to each of the contaminants discussed.

5.1 ARSENIC

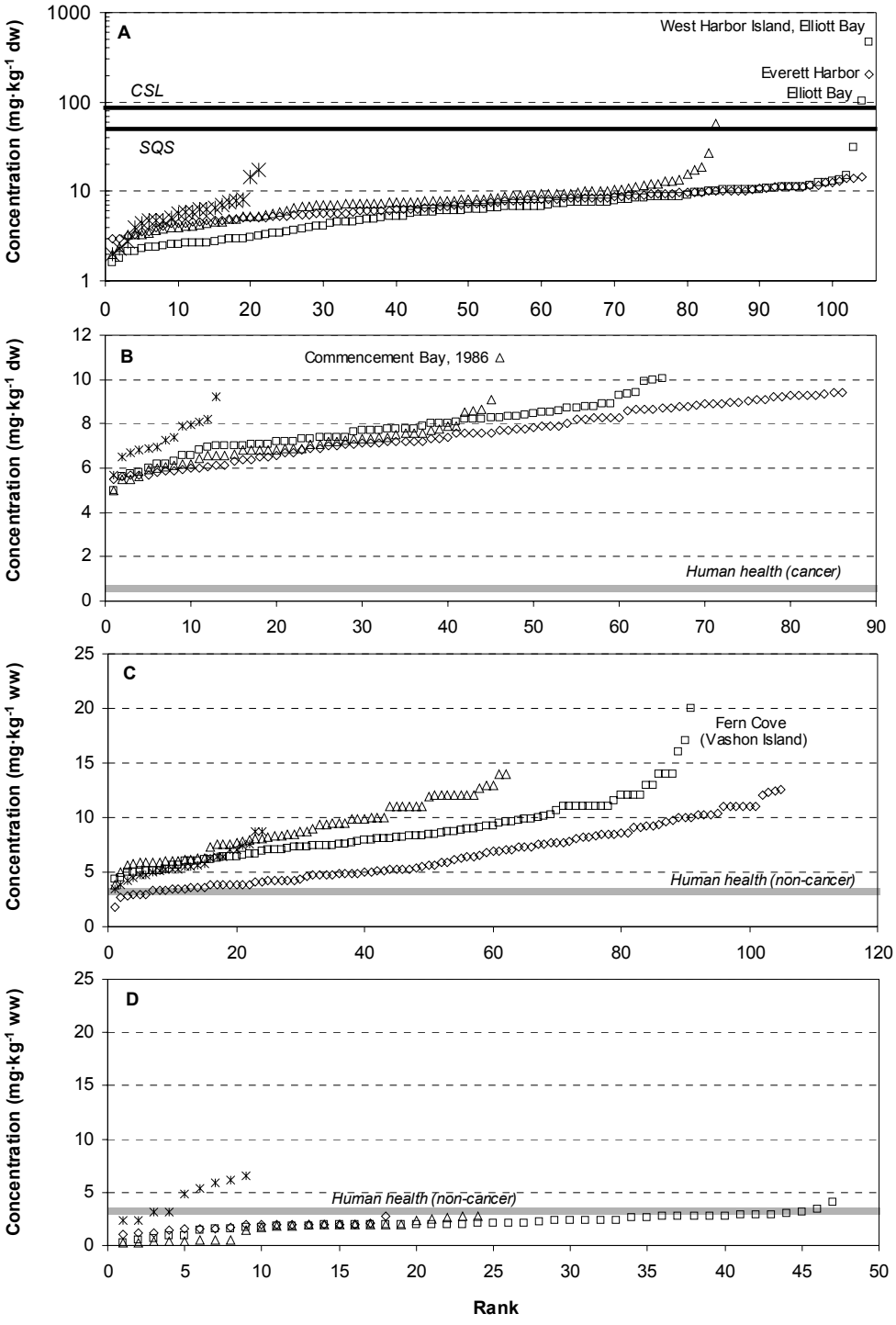
Very few sediment samples collected by PSAMP-NOAA exceeded either of the Washington State sediment standards (Figure 5-1), however, the *1998 303(d) List of Impaired and Threatened Waterbodies* (1998 303[d] list) for arsenic in sediment indicated that Bellingham Bay, Possession Sound, Duwamish Waterway, Sinclair Inlet and Commencement Bay had exceedances of the Sediment Management Standards (WDOE 2002). Current arsenic concentrations in sediments are significantly lower than those reported in SEDQUAL.

Arsenic concentrations in mussels exceeded the 1 in 100,000 cancer-risk criterion for the protection of human health at both urban and non-urban sites, whereas the non-carcinogenic hazards criterion was not exceeded at any site. All flatfish tissue concentrations also exceeded the cancer-risk criterion while only some rockfish (in Hood Canal and Central Puget Sound) exceeded the human health non-cancer level. The highest concentrations in English sole were observed at Fern Cove on Vashon Island.

The 1998 303(d) listing included Dyes Inlet and Port Washington Narrows, Eagle Harbor, Port Orchard/Agate Passage/Rich Passage, Sinclair Inlet due to arsenic tissue concentrations. However, there is a significant amount of uncertainty regarding the potential for human health effects from the levels of arsenic observed, which WDOE acknowledges in the 1998 303(d) decision matrix. Johnson and Roose (2002b) recently recommended that the above waterbodies be removed from the 303(d) list for exceedances of inorganic arsenic concentrations in tissues as they concluded that the elevated concentrations observed in the clam tissues (i.e., >0.006 $\mu g \cdot g^{-1}$) were naturally occurring in Puget Sound.

Except in localized areas where there may be significant historical sediment (such as in Central Puget Sound related to the operation of a smelter in Commencement Bay), or effluent discharges with elevated concentrations, arsenic is most likely not an overall contaminant of concern for Puget Sound.

Figure 5-1: Comparison of arsenic concentrations in Puget Sound (**A**) sediments; (**B**) mussels; (**C**) English sole and (**D**) rockfish. (SQS – Sediment Quality Standard; CSL – Cleaunup Screening Level; ◇ - North Puget Sound; □ - Central Puget Sound; △ - South Puget Sound; ✳ - Hood Canal).



5.2 CADMIUM

The highest cadmium concentrations were measured in samples from Everett Harbor, Olympia (East Bay), Thea Foss Waterway in Commencement Bay, and Lynch Cove in Hood Canal, however, all sediment samples were below the Washington State's sediment management standards (Figure 5-2). The 1998 303(d) listing indicates that sediments from the Strait of Georgia, Possession Sound, Duwamish Waterway, Elliott Bay, Dyes Inlet and Port Washington Narrows, Sinclair Inlet, and Commencement Bay had cadmium levels exceeding the Sediment Management Standards (WDOE 2002). Historically, sediment cadmium concentrations exceeded the SQS > 10x at locations such as Elliott Bay in Central Puget Sound, and the former ASARCO site near Tacoma, a Commencement Bay waterway, and Budd Inlet in South Puget Sound.

There were no obvious geographical trends for cadmium concentrations in mussels in relation to sediments. Cadmium cycling in the marine environment strongly resembles that of nutrients (Section 2.1.1.2), therefore, elevated concentrations are often found on Washington's outer coast despite the absence of the anthropogenic sources that are common in Puget Sound.

Cadmium was not measured in fish tissues by PSAMP.

Except in localized areas where there may be significant sediment contamination or effluent discharges with elevated concentrations, cadmium is most likely not an overall contaminant of concern for Puget Sound.

5.3 COPPER

The highest copper concentrations were measured in samples from Everett Harbor, Middle Waterway in Commencement Bay, and Harbor Island in Elliott Bay, however, only two samples exceeded the Washington State Sediment Management Standards (Figure 5-3). The 1998 303(d) listings indicate additional exceedances of the copper standards for sediments in Bellingham Bay, Possession Sound, Duwamish Waterway, Elliott Bay, Sinclair Inlet, Commencement Bay, Budd Inlet, North Hood Canal. Historically, sediment concentrations of copper exceeded the SQS > 10x at the former ASARCO site and in Elliott Bay (SEDQUAL).

All tissue concentrations were below the screening benchmarks for the protection of human health, and there were no obvious geographical trends for copper concentrations in mussels or fish in relation to sediments.

Except in localized areas where there may be significant sediment contamination or effluent discharges with elevated concentrations, copper is most likely not an overall contaminant of concern for Puget Sound.

Figure 5-2: Comparison of cadmium concentrations in Puget Sound (**A**) sediments; and (**B**) mussels. (SQS – Sediment Quality Standard; CSL – Cleaunup Screening Level; ◇ - North Puget Sound; ☐ - Central Puget Sound; △ - South Puget Sound; ✳ - Hood Canal).

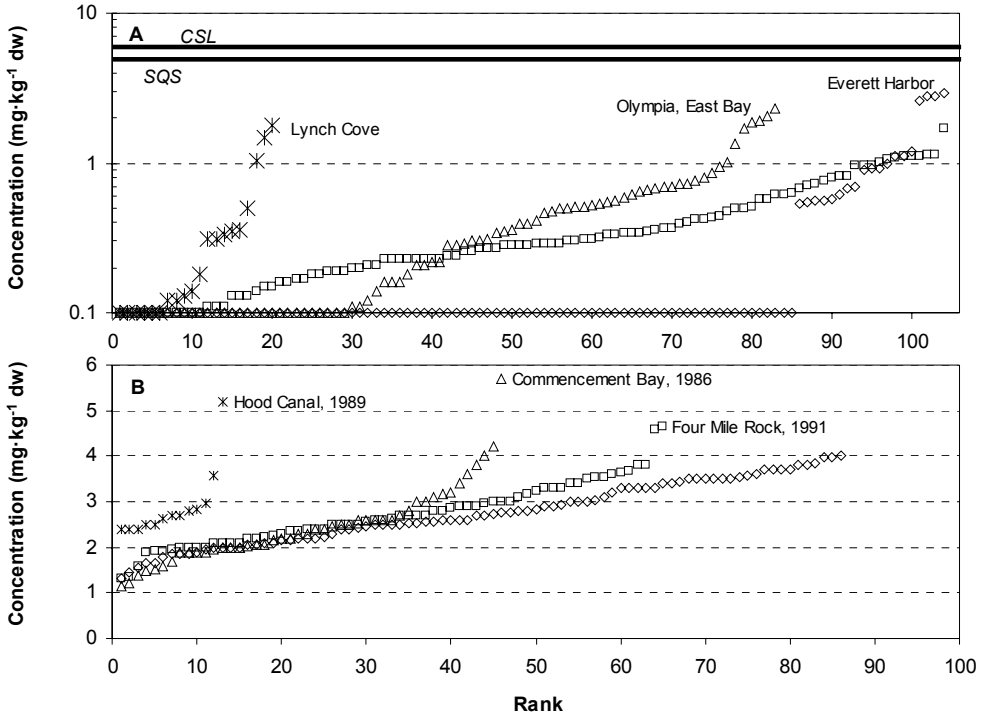
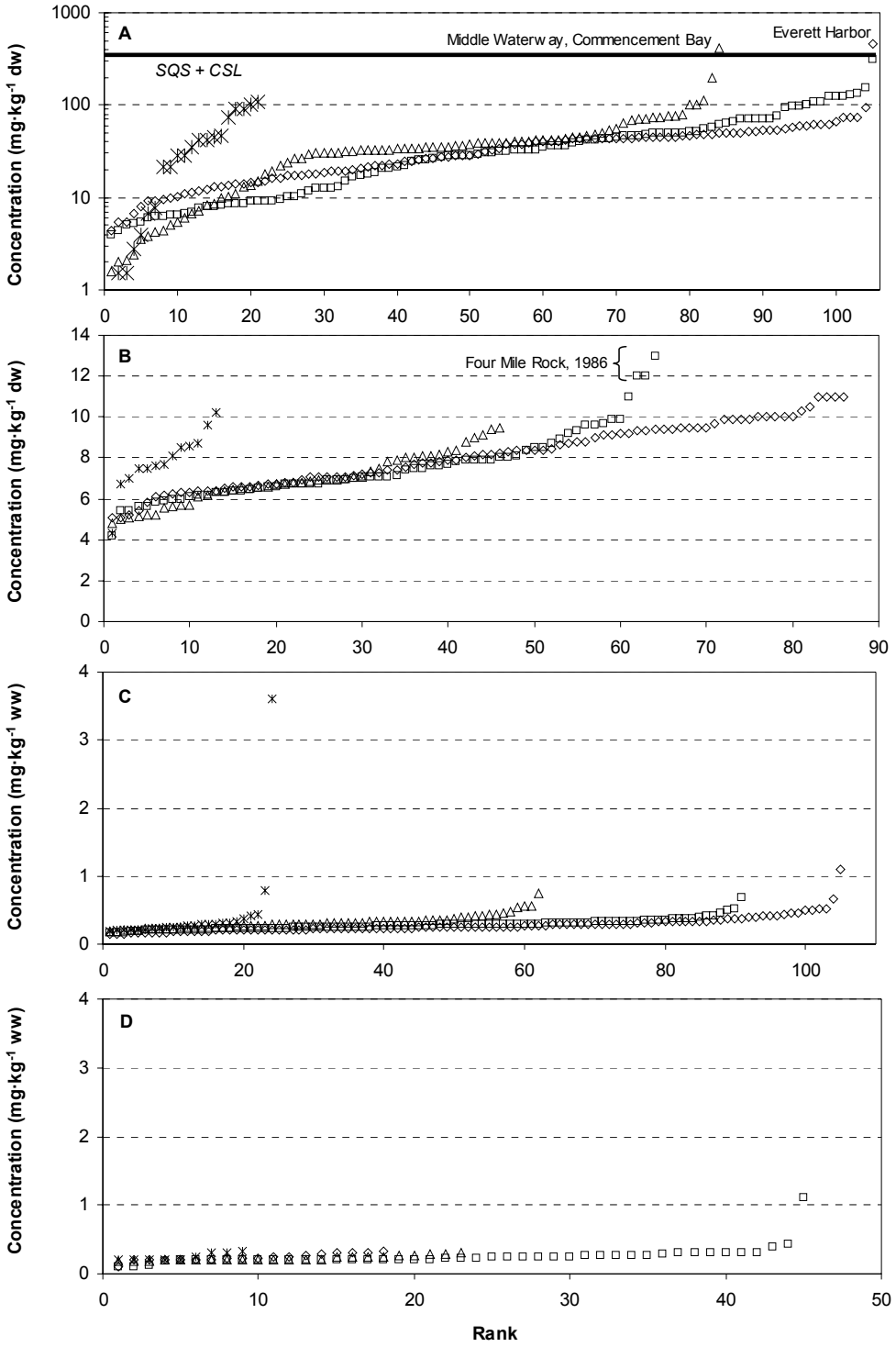


Figure 5-3: Comparison of copper concentrations in Puget Sound (**A**) sediments; (**B**) mussels; (**C**) English sole and (**D**) rockfish. (SQS – Sediment Quality Standard; CSL – Cleaunup Screening Level; ◇ - North Puget Sound; □ - Central Puget Sound; △ - South Puget Sound; ✳ - Hood Canal).



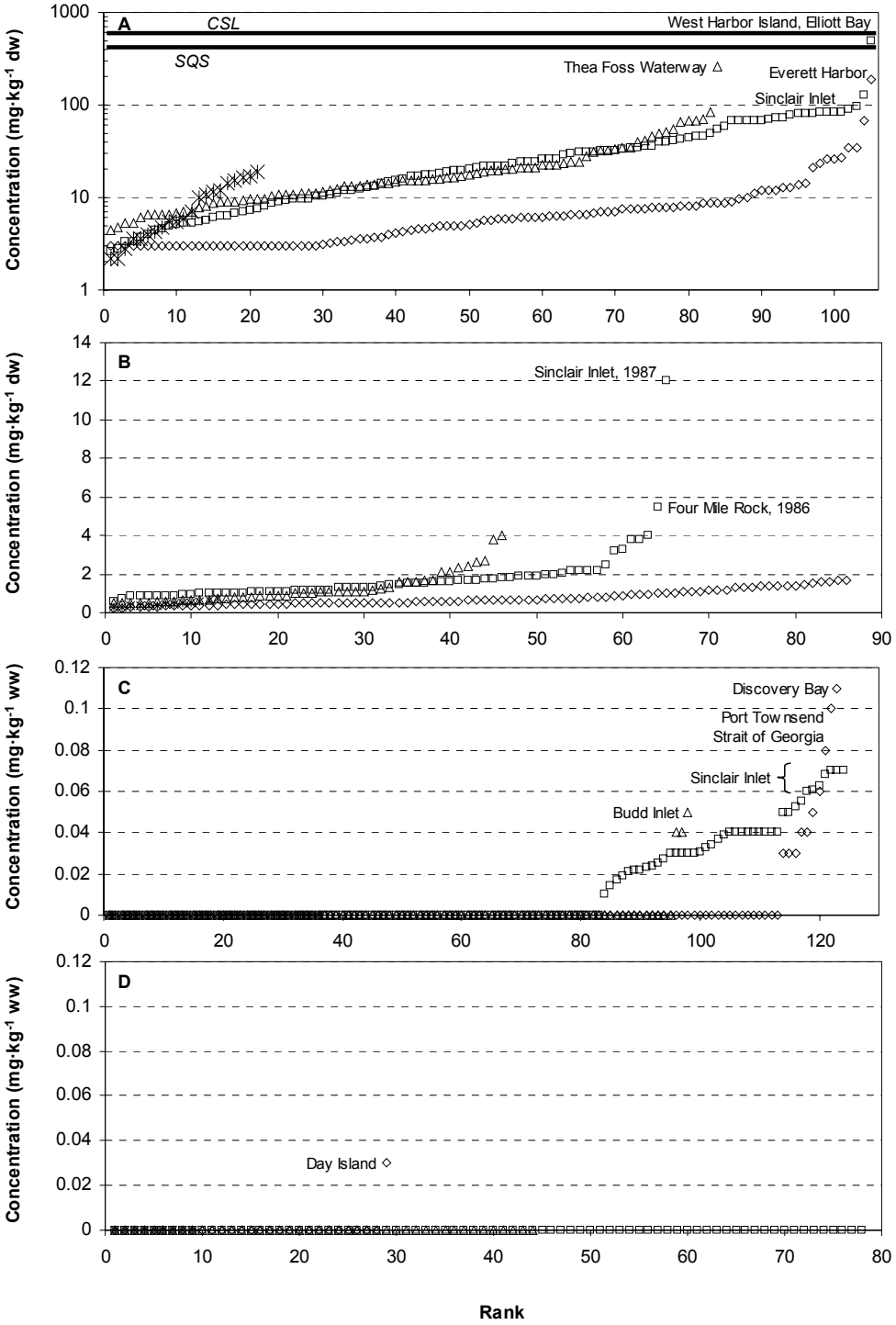
5.4 LEAD

The only exceedance of Sediment Management Standards for lead was in a sample from Harbor Island in Elliott Bay (Figure 5-4). The highest concentration for South Puget Sound was in Thea Foss Waterway, while Everett Harbor had the highest concentration in North Puget Sound. The 1998 303(d) listing indicates further exceedances in Bellingham Bay, Possession Sound, Duwamish Waterway, Elliott Bay, Sinclair Inlet, Commencement Bay, North Hood Canal (WDOE 2002). Historical sediment studies (SEDQUAL) indicated that the highest lead concentrations (i.e., > 10x SQS) occurred in the vicinity of Commencement Bay (i.e., near the former ASARCO site) and Elliott Bay. Only a few samples from Sinclair Inlet exceeded the SQS, and only by a small amount (i.e., < 5x).

None of the tissue samples for either mussels or fish exceeded the screening benchmarks for the protection of human health. However, English sole appeared to accumulate lead to a greater degree than rockfish, and there is a spatial trend in lead concentrations that points to a potential problem for English sole in Sinclair Inlet in particular. The highest single sample concentrations of lead in English sole were from Discovery Bay (lead detected in 2 of 6 samples), Port Townsend (lead detected in 2 of 15 samples, and the Strait of Georgia (lead detected in 2 of 21 samples). However, lead was more consistently measured in English sole from Sinclair Inlet (detected in 23 of 26 samples) and age-specific concentrations in these fish were higher than those from other locations. These data reflect the fact that lead concentrations in Central Puget Sound sediments were consistently higher in Sinclair Inlet (Figure 4-23). The significance of these lead concentrations are presently unknown, however, lead at sublethal concentrations is reported to cause developmental, reproductive and neurological problems in fish (Section 2.1.2.4).

Except in localized areas where there may be significant sediment contamination or effluent discharges with elevated concentrations, lead is most likely not an overall contaminant of concern for Puget Sound.

Figure 5-4: Comparison of lead concentrations in Puget Sound (**A**) sediments; (**B**) mussels; (**C**) English sole and (**D**) rockfish. (SQS – Sediment Quality Standard; CSL – Cleaunup Screening Level; ◇ - North Puget Sound; □ - Central Puget Sound; △ - South Puget Sound; ✳ - Hood Canal).



5.5 MERCURY

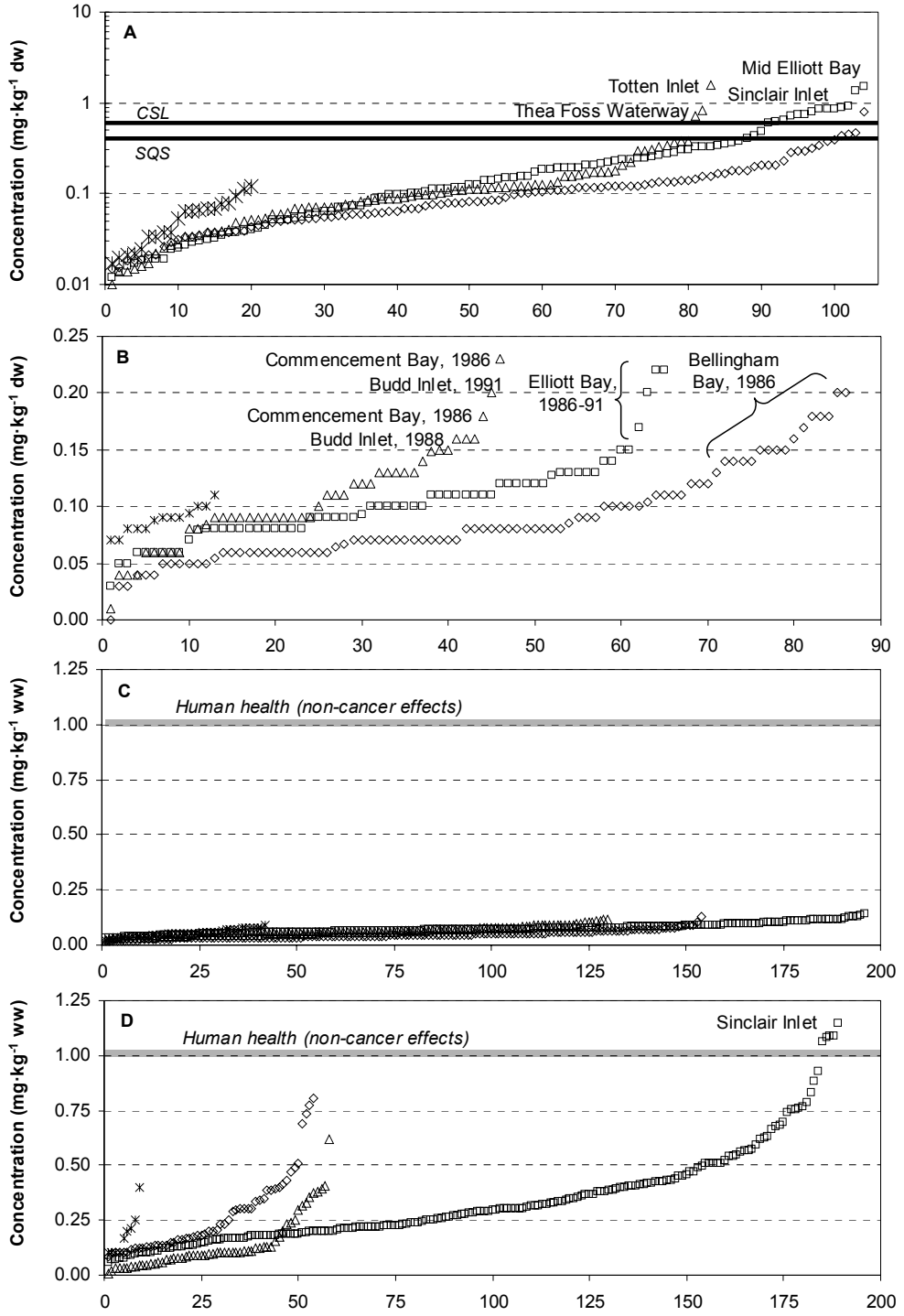
Several sediment samples exceeded both the SQS and CSL standards for mercury, particularly in Totten Inlet and Thea Foss Waterway in South Puget Sound, Elliott Bay and Sinclair Inlet in Central Puget Sound and Boundary Bay in North Puget Sound (Figure 5-5). Long-term sediment monitoring by PSAMP indicates that Sinclair Inlet sediments have consistently exceeded the SQS, while other stations in Puget Sound have been below the sediment standard (Figure 4-23). Historically, the highest (i.e., > 10x SQS) concentrations were found in the Commencement Bay waterways, near the former ASARCO site, Elliott Bay and its waterways, Sinclair Inlet and Bellingham Bay. Waterbodies on the 1998 303(d) list for mercury in sediments included:

- Bellingham Bay,
- Port Gardner and Everett Harbor,
- Central Puget Sound,
- Duwamish Waterway,
- Elliott Bay,
- Eagle Harbor,
- Sinclair Inlet,
- Dyes Inlet and Port Washington Narrows,
- Commencement Bay,
- Budd Inlet, and
- North Hood Canal (WDOE 2002).

While mercury concentrations in mussels did not exceed the screening benchmarks for the protection of human consumers, the spatial pattern of the metal in mussel tissue reflects historical sediment contamination patterns. Mercury concentrations in English sole were also lower than the human health non-cancer effects level. In comparison, it is evident that rockfish in Puget Sound accumulate mercury; tissue concentrations for rockfish from Sinclair Inlet exceeded the human health non-cancer level, while many samples from Sinclair Inlet, Elliott Bay, and the San Juan Islands had concentrations approaching the non-cancer effects level. Furthermore, rockfish from Sinclair Inlet (urban), Elliott Bay (urban), and Foulweather Bluff near the outlet of Hood Canal (non-urban) had higher age-specific mercury concentrations than did fish from other areas, indicating a greater degree of exposure. Fish consumption advisories have been issued for fish and shellfish from Eagle Harbor and Sinclair Inlet due to elevated tissue mercury concentrations.

Because of its persistence and wide-spread distribution, mercury continues to be a contaminant of concern in Puget Sound.

Figure 5-5: Comparison of mercury concentrations in Puget Sound (**A**) sediments; (**B**) mussels; (**C**) English sole and (**D**) rockfish. (SQS – Sediment Quality Standard; CSL – Cleaunup Screening Level; ◇ - North Puget Sound; □ - Central Puget Sound; △ - South Puget Sound; ✳ - Hood Canal).

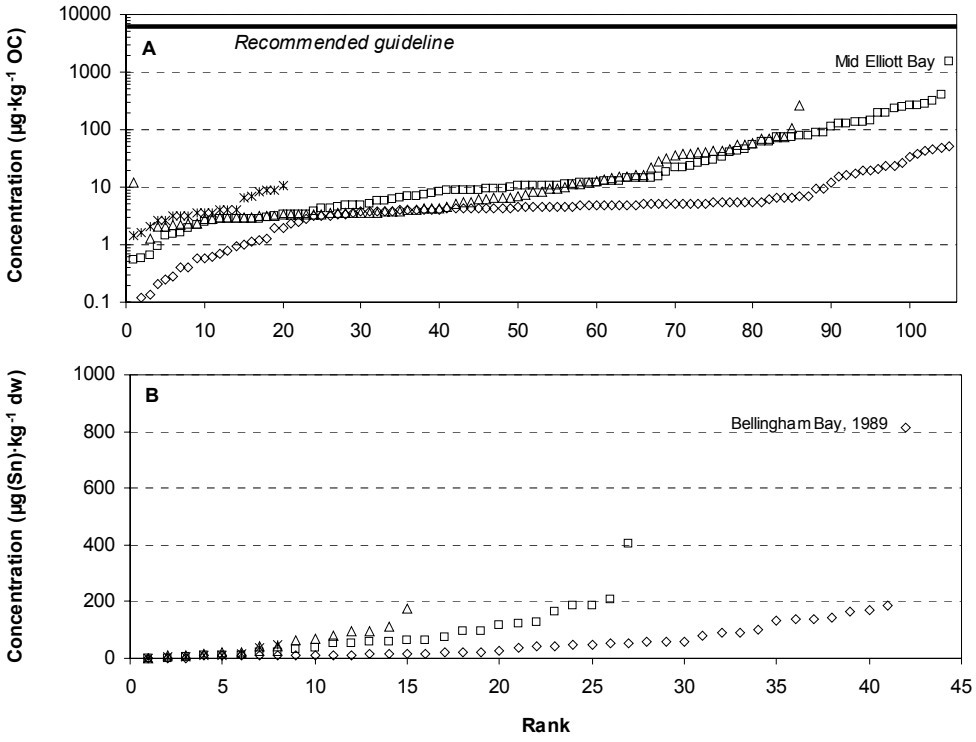


5.6 TRIBUTYLTIN

The data available for an overview of tributyltin contamination in Puget Sound is limited to sediment and mussel tissue concentrations. TBT was measured in only a small number of liver samples from English sole (Figure 5-6). The highest TBT sediment concentrations were measured in Elliott Bay, which is not unexpected given that it is a major port in Puget Sound. Cardwell et al. (1999) evaluated the risks for chronic effects from TBT in water to the marine organisms of Puget Sound. They determined that marinas presented the greatest risks (3%), followed by shipyards (1%), and harbors (2%). Fish and shellfish habitats adjacent to these kinds of sites were determined to have a 0% risk from TBT.

Except in localized areas where there may be significant sediment contamination or effluent discharges with elevated concentrations, TBT is likely not an overall contaminant of concern for Puget Sound.

Figure 5-6: Comparison of TBT concentrations in Puget Sound (**A**) sediments; and (**B**) mussels. (◇ - North Puget Sound; □ - Central Puget Sound; △ - South Puget Sound; ∗ - Hood Canal).



5.7 PCBs

Exceedances of the Washington State Sediment Management Standards were most common in sediments from Elliott Bay in Central Puget Sound, while only one sample from North Puget Sound (i.e., Everett Harbor) had elevated PCB concentrations and three samples from South Puget Sound (i.e., Vashon Island; Hylebos Waterway) had elevated PCBs (Figure 5-7). The 1998 303(d) list indicates further SMS exceedances in Bellingham Bay, Strait of Georgia, Port Gardner/Everett Harbor, Central Puget Sound, Duwamish Waterway, Elliott Bay, Commencement Bay, and Budd Inlet.

PCB concentrations in mussels were generally highest in the early 1990's, in samples from Port Townsend and Elliott Bay. However, in 1998, there was a spike in mussel tissue concentrations at Elliott Bay. This temporary increase in PCBs may have been as a result of a sudden global increase in atmospheric PCBs as similar mussel tissue increases were noted in remote sampling locations on Washington's outer coast and in Alaska (PSWQAT 2002b). An alternate explanation is that there was a local disturbance in Elliott Bay (e.g., resuspension of contaminated sediments during a site cleanup or construction) as concurrent increases in DDT and PAHs were also observed.

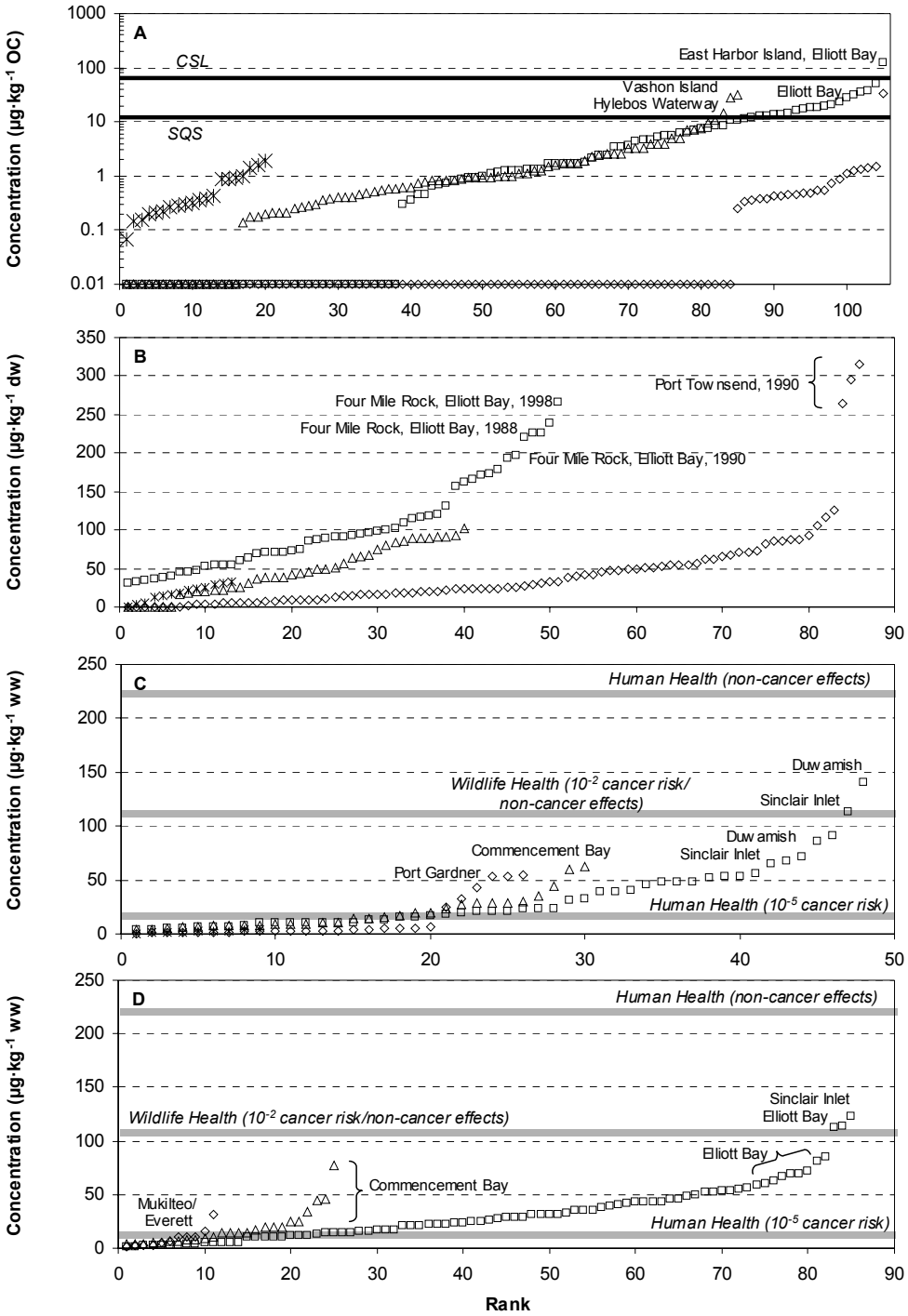
A significant number of fish tissue samples also contained elevated PCB concentrations. Indeed, most of the samples from Central Puget Sound and approximately half of the samples from North and South Puget Sound exceeded the 1 in 100,000 cancer risk level for the protection of human consumers. Fish from Hood Canal, in comparison were below this tissue benchmark. 1998 303(d) list included the following waterbodies due to PCB tissue residues: Padilla Bay/Fidalgo Bay/Guemes Channel, Duwamish Waterway, Eagle Harbor, Sinclair Inlet, Thea Foss Waterway, Budd Inlet (WDOE 2002). A fish consumption advisory has been issued for Manchester State Park and Commencement Bay, also due to elevated tissue PCB concentrations (WDOH 2002).

English sole and rockfish from Sinclair Inlet and Elliott Bay may pose both a 1 in 100 cancer risk and non-cancer effects risk to wildlife based on PCB tissue residues. Elevated PCB concentrations have also been measured in Pacific salmon and herring and the fish themselves may be at risk for adverse biological effects (O'Neill and West 2001).

PCBs have been observed at potentially harmful levels in higher trophic level organisms as well. While overall PCB concentrations have decreased in harbor seals (Figure 4-35) they appear to have stabilized at a level that still puts them at risk of immune system dysfunction (Ross et al. 1996). Killer whales also have potentially harmful PCB concentrations.

Collectively, the different lines of evidence point to a PCB signature in Central Puget Sound. Despite the fact that PCB was banned from use and production three decades ago, it is still circulating in the food web and continues to pose a risk to Puget Sound biota and human consumers of fish and shellfish from the Sound.

Figure 5-7: Comparison of total PCB concentrations in Puget Sound (**A**) sediments; (**B**) mussels; (**C**) English sole and (**D**) rockfish. (SQS – Sediment Quality Standard; CSL – Cleaunup Screening Level; ◇ - North Puget Sound; □ - Central Puget Sound; △ - South Puget Sound; ✳ - Hood Canal).



5.8 PAHs

Polycyclic aromatic hydrocarbons have been assessed here in two different ways:

1. Using total dry weight PAH concentrations in sediments compared to ERM/ERL benchmarks, and total PAH concentrations in mussels for an overall view of PAHs (Figure 5-8); and
2. Using organic carbon-normalized benzo[a]pyrene concentrations in sediments, dry weight benzo[a]pyrene concentrations in mussels, and benzo[a]pyrene fluorescing aromatic hydrocarbons (B[a]P-FAC) in flatfish and rockfish bile for an overview of potential contaminant transfer between environmental compartments (Figure 5-9).

Several sediment sampling locations had total PAH concentrations in excess of the ERL benchmark but only two (i.e., Thea Foss Waterway and Elliott Bay) exceeded the ERM. Elevated PAH concentrations (one or more LPAH or HPAH) in sediments have contributed to the 1998 303(d) listing of the following waterbodies: Bellingham Bay, Strait of Georgia, Port Gardner/Everett Harbor, Possession Sound, Duwamish Waterway, Elliott Bay, Eagle Harbor, Sinclair Inlet, Commencement Bay, Budd Inlet, and North Hood Canal.

In comparison, total PAH concentration only appeared to be elevated in mussels from the Elliott Bay area, where relatively high concentrations were reported for the mid-1980s and for a sample in 1998. A spike in tissue concentrations of PCB and DDT were also noted for the same 1998 Elliott Bay sample, perhaps indicating some localized disturbance of contaminated sediments in Elliott Bay or temporary wastewater discharge to the Bay.

Benzo[a]pyrene concentrations in sediments and mussels exhibited a similar regional distribution pattern as total PAH concentrations. However, there were a greater number of exceedances of the sediment benchmarks. The difference in the number of exceedances could be due to larger concentrations of the HPAH relative to total PAH concentration or because the sediment benchmarks used were derived in different ways, with one applying to dry weight concentrations and the other to OC-normalized concentrations. Benzo[a]pyrene concentrations in mussels in the mid 1980s exceeded the 1 in 100,000 lifetime increased cancer risk for human consumers.

B[a]P-FAC concentrations were highest in Elliott Bay rockfish, while Sinclair Inlet rockfish and Eagle Harbor flatfish also had relatively high levels. PAHs in fish and/or shellfish tissue from Dyes Inlet/Port Washington Narrows, Eagle Harbor, and Budd Inlet have also contributed to the 303(d) listing for these waterbodies, while fish consumption advisories have been issued for Eagle Harbor, Sinclair Inlet and Budd Inlet.

While B[a]P-FAC concentrations were highest in rockfish, English sole appear to be more sensitive to the harmful effects of PAHs. The overall mortality rate of adult English

sole from contaminated sites in Puget Sound has not been found to be any higher than the fish at relatively clean sites (Landahl et al. 1997). However, chronic effects such as liver lesions, reproductive impairment and immunosuppression have been documented and PAH implicated as a contributing if not primary factor (Collier et al. 1998; Arkoosh et al. 1996).

The clean up of contaminated sediments has reduced the risk of harmful effects to aquatic organisms at some sites (e.g., Eagle Harbor). However, PAHs continue to be released into the aquatic environment from additional historically-contaminated sediments, through atmospheric deposition and surface runoff of combustion products or leaching from creosote-treated wood. Overall, the concentrations of PAH in fish tissue have not changed (O'Neill, pers. comm. 2002) and the incidence of liver lesions in English sole has not decreased from the 1970s (Collier et al. 1998). Furthermore, human consumers of fish and shellfish from Puget Sound may be at continued risk from the effects of PAH.

Figure 5-8: Comparison of total PAH concentrations in Puget Sound (**A**) sediments; and (**B**) mussels. (ERM – Effects Range Medium; ERL – Effects Range Low; ◇ - North Puget Sound; □ - Central Puget Sound; △ - South Puget Sound; ∗ - Hood Canal).

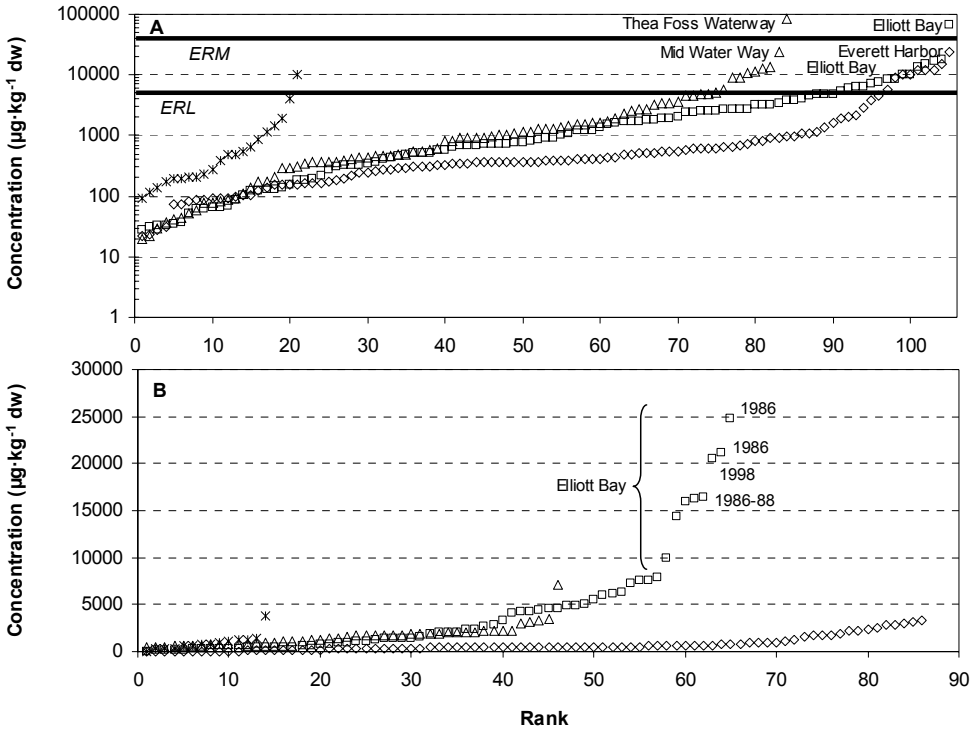
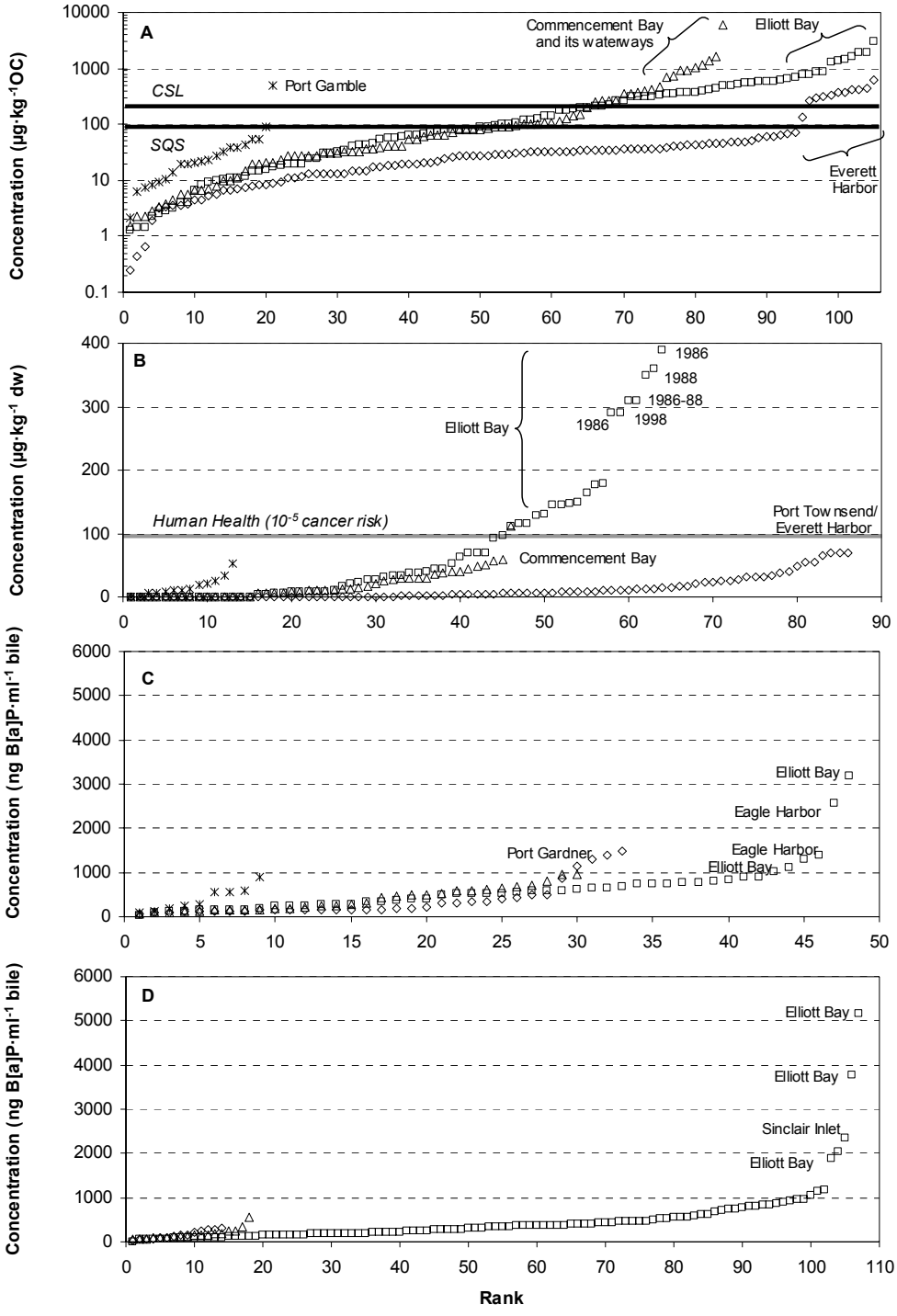


Figure 5-9: Comparison of benzo[a]pyrene concentrations in Puget Sound (**A**) sediments; and (**B**) mussels; and B[a]P-FAC concentrations in (**C**) English sole and (**D**) rockfish bile. (SQS – Sediment Quality Standard; CSL – Cleaunup Screening Level; ◇ - North Puget Sound; ☐ - Central Puget Sound; △ - South Puget Sound; ✳ - Hood Canal).

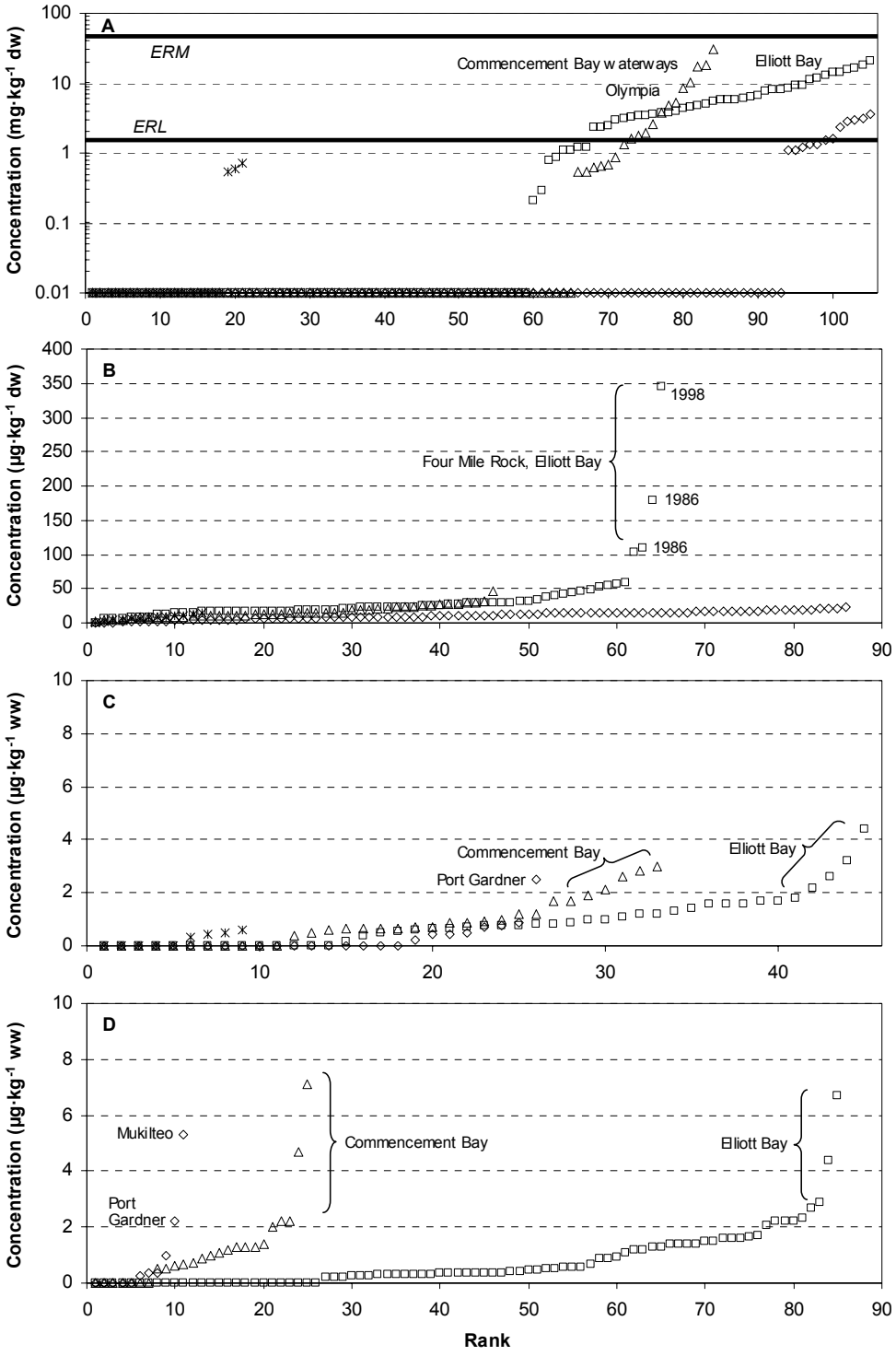


5.9 DDT

DDT (and its metabolites) is an example of an organochlorine pesticide that can be found in the marine environment of Puget Sound. For the assessment, DDT concentrations were reported as dry weight concentrations and compared to the NOAA ERL and ERM benchmarks as Washington Sediment Management Standards do not exist for the compound. None of the samples had DDT concentrations in excess of the ERM, while several samples from Commencement Bay and it waterways, Olympia in Budd Inlet and Elliott Bay exceeded the ERL (Figure 5-10). Only mussels from Elliott Bay exhibited elevated DDT concentrations, while fish tissue concentrations followed a similar regional pattern as the sediments. However, none of the tissue samples exceeded benchmarks for the protection of either human consumers or piscivorous wildlife.

These data demonstrate that while DDT was banned from production and use in the United States some time ago, a reservoir of the organochlorine pesticide remains in the environment (i.e., agricultural soils that may transported to the Sound via surface runoff; historically-contaminated sediments).

Figure 5-10: Comparison of total DDT concentrations in Puget Sound (**A**) sediments; (**B**) mussels; (**C**) English sole and (**D**) rockfish. (ERM – Effects Range Medium; ERL – Effects Range Low; ◇ - North Puget Sound; □ - Central Puget Sound; △ - South Puget Sound; ✳ - Hood Canal).



5.10 SUMMARY

The overall effect of toxic metals and organic chemicals in the marine environment of Puget Sound is difficult to quantify at the population or ecosystem level as there are many factors causing stress to aquatic biota and influencing their numbers (e.g., habitat loss; overharvesting; climate change). However, specific contaminant-induced effects, such as liver lesions and reproductive impairment in English sole due to PAH exposure, have been identified in several urban and near-urban embayments.

A significant volume of sediment contaminants has been assessed and cleaned up in areas such as Elliott Bay and Commencement Bay and its waterways, and sediment assessment and cleanup continue today. However, it is difficult to quantify the current and future potential for exposure of aquatic organisms to toxic metals and organic chemicals because they are exposed to a combination of historical (sometimes unidentified) and current deposits.

Of the contaminants discussed in this report, the most troubling are those that are persistent, that bioaccumulate and biomagnify in the food chain and that are toxic (PBTs). An example of such a contaminant are PCBs. Despite the fact that PCBs were banned from production and use in North America decades ago, the contaminant is still cycling through the food web and top-carnivore trophic levels such as killer whales are showing increasingly higher concentrations. The data shown in Figure 5-7 indicate that PCBs persist in the sediments of Puget Sound, particularly in Central Puget Sound, and that some fish have high enough concentrations of PCBs in their tissue to present a health risk to both human and wildlife consumers.

General statements that can be made about the status of toxics in Puget Sound include:

- The highest contaminant body residues have been found in organisms in closer proximity to contaminant sources and/or in organisms occupying positions high in the food web, indicating continued exposure of aquatic organisms of Puget Sound to toxic contaminants.
- There is evidence that with source control, there is decreasing exposure to toxics and subsequent decreases in adverse biological effects (e.g., Eagle Harbor).
- Not enough is known about the reproductive effects (induced by endocrine disrupting compounds, for example) or the temporal trends of contaminants.
- There are some human health risks associated with consuming Puget Sound biota. This is demonstrated by the number of fish consumption advisories as well as some of the tissue data which indicate levels that increase risks of cancer and non-carcinogenic effects to consumers.

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6. EMERGING ISSUES

During the last several years, some issues in the study of environmental contaminants have been moving to the forefront and may have implications for the direction of research in Puget Sound's marine environment:

Laboratory Toxicity Test Versus Field Studies – The sediment quality triad approach (Long and Chapman 1985) to assessing the overall health of sediments, options for dredgate disposal and remediation requirements has become a commonly used tool during the last decade. There are questions, however, about the relationship between laboratory toxicity tests and the abundance and diversity of benthic fauna in the natural environment, and the ability of laboratory tests to predict effects in the field.

Phototoxicity of PAHs – Several researchers have determined via laboratory studies that exposure to UV light can affect the toxicity of PAHs and in some jurisdictions, criteria to protect aquatic organisms from photo-induced toxicity have been established based on laboratory toxicity data. However, the ecological (*in situ*) relevance of this phenomenon is not well understood (McDonald and Chapman 2002).

Expression of PCB Toxicity in Terms of Congeners Versus Aroclors – PCB concentrations in sediments and tissues have frequently been expressed in terms of total PCB and as Aroclors. There is increasing interest in assessing toxicity in terms of individual congeners as an alternative because it is recognized that different organisms can preferentially take up different congeners.

Endocrine Disrupting Compounds – New laboratory analytical techniques and increasing understanding of the endocrine systems in both vertebrate and invertebrate species have led to increasing concern about the endocrine disrupting properties of various chemicals. Endocrine disrupting compounds may affect thyroid function, decrease fertility and hatching success, feminize male organisms, masculinize female organisms, or alter immune function (Colborn et al. 1993 and references therein).

Persistent, Bioaccumulative Toxins (PBTs) – PBTs are a certain group of chemicals that have the ability to persist in the environment for a long time, build up in human or animal tissue, and have toxic effects on humans or wildlife. Even when released in very small amounts, they accumulate and can cause environmental problems. WDOE has identified 9 PBTs in need of further long-term reductions as part of the agency's *Proposed strategy to continually reduce persistent, bioaccumulative toxins (PBTs) in Washington State – December 2000*.

Trophic Transfer Of PBTs – The mechanisms by which persistent and toxic compounds move through the food web are not well understood. For example, there is growing concern about PCB contamination in marine mammals, however, the sources of the contaminant are not known.

Polybrominated Diphenyl Ethers – PBDEs are constituents in fire retardants and have come under scrutiny in Europe and on the eastern coast of the U.S. PBDEs are structurally similar to PCBs and are being found in increasingly higher concentrations in the marine environment.

Cadmium in Oysters – Fisheries and Oceans Canada scientists have been studying cadmium in oysters not proximal to anthropogenic sources in response to concerns that farmed oysters would not meet Food and Agriculture Organization (FAO) existing or proposed lower guidelines for the protection of human consumers (Kruzynski, pers. comm. 2002). The scientists concluded that the elevated tissue burdens of cadmium were naturally occurring, however, the bioavailability of the cadmium has not been studied yet.

Arsenic Speciation In Shellfish – Arsenic is another metal that accumulates in shellfish as a result of the naturally high concentrations of arsenate (As^{5+}) in seawater. There is apparently no evidence of ecosystem effects related to the accumulation of arsenic in plankton and filter-feeding shellfish, and arsenic is generally not bioavailable to mammalian consumers (Crecelius, pers. comm. 2002). However, there is still a human health a concern regarding how much As may be bioavailable. EPA is currently developing new laboratory analytical techniques to help answer this question.

Objectives of Data Collection – PSAMP is starting to look at dioxins and PCB in crab, however, the data collected for this program (i.e., hepatopancreas tissue) is not necessarily suitable for determining the risk to mammalian consumers. Conversely, data collected for assessing risk to mammalian consumers is not necessarily appropriate for determining effects in the prey species. Examination of potential extrapolation factors would be valuable.

Pharmaceuticals and Personal Care Products (PPCPs) – PPCPs comprise a broad, diverse collection of thousands of chemical substances including prescription and over-the-counter therapeutic drugs, fragrances, cosmetics, sun-screen agents, diagnostic agents, nutraceuticals, biopharmaceuticals, and many other substances. These chemicals, whether applied externally, or ingested, have the potential to be excreted or washed into sewage systems and from there discharged to the aquatic and terrestrial environments.

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APPENDICES

APPENDIX A

Regulatory Standards and Screening Benchmarks

APPENDIX A1: Washington State water quality standards for selected chemicals and USEPA National Recommended Water Quality Criteria

Chemical	Washington State Water Quality Standards ($\mu g \cdot L^{-1}$)		USEPA National Recommended Water Quality Criteria: 2002 ($\mu g \cdot L^{-1}$)		
			Sea Water		Human Health[b]
	Acute	Chronic	Acute	Chronic	
Metals[a]					
Arsenic	36.0	69.0	36	69	0.14[c]
Cadmium	42.0	9.3	40	8.8	-
Copper	4.8	3.1	4.8	3.1	-
Lead	210.0	8.1	210	8.1	-
Mercury (inorganic)	1.8	0.025	1.8	0.94	-
Polycyclic Aromatic Hydrocarbons					
LPAH					
Acenaphthene	-	-	-	-	990
Anthracene	-	-	-	-	40,000
HPAH					
Benzo[a]anthracene	-	-	-	-	0.018
Benzo[a]pyrene	-	-	-	-	0.018
Benzo[g,h,i]perylene					0.018
Chrysene	-	-	-	-	0.018
Indeno[1,2,3-c,d]pyrene	-	-	-	-	0.018
Pyrene	-	-	-	-	4,000
Phthalate Esters					
Bis(2-ethylhexyl)phthalate	-	-	-	-	2.2
Butylbenzyl phthalate	-	-	-	-	1,900
Chlorinated pesticide and PCBs					
4,4'-DDT	-	-	0.13	0.001	0.00022
DDT	0.13	0.001			
Dieldrin	-	-	0.71	0.0019	0.00054
PCB	10.0	0.030	-	0.03	0.00064

Sources: Water Quality Standards for Surface Waters of the State of Washington (Chapter 173-201A WAC); USEPA (2002)

[a] Criteria refer to dissolved metals

[b] Human consumption of aquatic organisms; increased lifetime cancer risk level 10^{-6}

[c] USEPA is currently reassessing this value

APPENDIX A2: Washington State sediment quality criteria and NOAA sediment quality guidelines for selected chemicals

Chemical	Washington State Criteria		NOAA Guidelines	
	SQS	CSL	ERL	ERM
Metals	*mg·kg^{-1} dry weight*		*mg·kg^{-1} dry weight*	
Arsenic	57	93	8.2	70
Cadmium	5.1	6.7	1.2	9.6
Copper	390	390	34	270
Lead	450	530	46.7	218
Mercury	0.41	0.59	0.15	0.71
TBT	NA	NA	NA	NA
Polycyclic Aromatic Hydrocarbons	*mg·kg^{-1} organic carbon*		*µg·kg^{-1} dry weight*	
LPAH				
Acenaphthene	16	57	16	500
Acenaphthylene	66	66	44	640
Anthracene	220	1,200	85.3	1,100
Fluorene	23	79	19	540
Naphthalene	99	170	160	2,100
Phenanthrene	100	480	240	1,500
Sum of LPAH[1]	370	780	552	3,160
HPAH				
Benzo[a]anthracene	110	270	261	1,600
Benzo[a]pyrene	99	210	430	1,600
Benzo[g,h,i]perylene	31	78	NA	NA
Chrysene	110	460	384	2,800
Fluoranthene	160	1,200	600	5,100
Indeno[1,2,3-c,d]pyrene	34	88	NA	NA
Pyrene	1,000	1,400	665	2,600
Sum of HPAH[2]	960	5,300	1,700	9,600
Total PAH[3]	NA	NA	4,022	44,792
Phthalate Esters	*mg·kg^{-1} organic carbon*			
Bis(2-Ethylhexyl)phthalate	47	78	-	-
Butylbenzylphthalate	4.9	64	-	-
Diethylphthalate	61	110	-	-
Dimethylphthalate	53	53	-	-
Di-N-butyl phthalate	220	1,700	-	-
Di-N-octyl phthalate	58	4,500	-	-
Chlorinated organics	*mg·kg^{-1} organic carbon*		*µg·kg^{-1} dry weight*	
Dibenzofuran	15	58	-	-
4,4'-DDE	-	-	2.2	27
Total DDT	-	-	1.58	46.1
Total PCB[4]	12	65	22.7	180

Sources: Chapter 173-204 WAC Sediment Management Standards (SMS), updated in 1995; Long, et al. (1995).
SQS = Sediment Quality Standard; CSL = Cleanup Screening Level; ERL = Effects Range Low; ERM = Effects Range Medium.
[1] Sum of 6 LPAHs for Washington State SMS and sum of 7 LPAH for NOAA guidelines.
[2] Sum of 9 HPAHs for Washington State SMS and sum of 6 HPAHs for NOAA.
[3] Sum of 13 PAHs for NOAA guidelines.
[4] As total Aroclors for Washington State and as total congeners for NOAA.

APPENDIX A3 – Tissue benchmarks for the protection of piscivorous wildlife for selected contaminants

Chemical	New York[a]		Canada[b]
	1 in 100 cancer risk	Non-carcino-genic effects	Maximum
Chlorinated Organic			
Aldrin/dieldrin	0.022	0.12	-
2,3,7,8-TCDD	$2.5x10^{-6}$	$3x10^{-6}$	-
Total DDT	0.27	0.2	1[c]
Total PCB	0.11	0.11	0.11[d]

[a] Concentrations in $mg \cdot kg^{-1}$ ww; Newell et al (1987)
[b] Concentrations in $mg \cdot kg^{-1}$ ww
[c] For the protection of fish consuming birds (Environment Ontario 1984)
[d] Maximum concentration; BCMOELP (1998)

APPENDIX A4 – Tissue benchmarks for the protection of human health for selected metals and organics

Chemical	USEPA[a]			Canada[b]
	1 in 100,000 Cancer Risk[c]	Non-carcinogenic Effects[d]	Legal limit[e]	Maximum
Metals				
Arsenic	0.062	3.2	-	3.5[f]
Cadmium	-	5.4	-	-
Copper	-	400	-	-
Lead	-	-	-	0.8[f]
Mercury (inorganic)	-	1.1	1.0	0.5[f]
Methyl mercury	-	1.1	-	-
Polycyclic Aromatic Hydrocarbons				
LPAH				
Acenaphthene	-	650	-	-
Anthracene	-	3,200	-	-
Fluorene	-	430	-	-
Naphthalene	-	430	-	-
HPAH				
Benzo(a)anthracene	0.15	-	-	-
Benzo(a)pyrene	0.015	-	-	0.004[g]
Chrysene	15	-	-	-
Fluoranthene	-	430	-	-
Indeno(1,2,3-c,d)pyrene	0.15	-	-	-
Pyrene	-	320	-	-
Phthalate Esters				
Bis(2-ethylhexyl)phthalate	7.7	220	-	-
Butylbenzylphthalate	-	2,200	-	-
Diethylphthalate	-	8,600	-	-
Dimethylphthalate	-	110,000	-	-
Di-N-butyl phthalate	-	1,100	-	-
Di-N-octyl phthalate	-	220	-	-
Chlorinated Organics				
Total DDT	0.32	5.4	-	5[h]
Dieldrin	0.0067	0.54	-	-
Chlorpyrifos	-	32	-	-
Dibenzofuran	-	43	-	-
2,3,7,8-TCDD	6.9×10^{-7}	2.0×10^{-5}	-	-
PCB	0.014	0.22	2.0	2.0[h]

[a] Concentrations in $mg \cdot kg^{-1}$ ww
[b] Concentrations in $mg \cdot kg^{-1}$ ww
[c] cancer risk level 10^{-5} (USEPA 1997)
[d] non-cancer hazard (USEPA 1997)
[e] legal limit for fish and fisheries products (USEPA 1989)
[f] BCMOELP (1998)
[g] BCMOELP (1998); for low consumers
[h] H&WC (1990)

APPENDIX B

List of Persons Contacted

Appendix B: List of Persons Contacted

NAME	AGENCY
Peter Chapman	EVS Environment Consultants
Tracy Collier	National Oceanic and Atmospheric Administration
Kathleen Collins	Puget Sound Water Quality Action Team
Eric Crecelius	Batelle Laboratories
Dana Davoli	U.S. Environmental Protection Agency
Bruce Duncan	U.S. Environmental Protection Agency
Chris Field	Puget Sound Water Quality Action Team
Tim Goodman	Washington Department of Natural Resources
Ellie Hale	Puget Sound Water Quality Action Team
Joan Hardy	Washington Department of Health
John Hardy	Western Washington University
Grant Kirby	Northwest Indian Fisheries Commission
Lon Kissinger	U.S. Environmental Protection Agency
Robert Kotchenruther	Puget Sound Water Quality Action Team
Roseanne Lorenz	U.S. Environmental Protection Agency
Mary Mahaffy	U.S. Fish and Wildlife Service
Russ McMillan	Washington Department of Ecology
Alan Mearns	National Oceanic and Atmospheric Administration
Jan Newton	Washington Department of Ecology
Sandra O'Neill	Washington Department of Fish and Wildlife
Christine Psyk	U.S. Environmental Protection Agency
Peter Ross	Fisheries and Oceans Canada
Nat Scholz	National Oceanic and Atmospheric Administration
Jim Simmons	King County
Kimberley Stark	King County Department of Natural Resources
James West	Washington Department of Fish and Wildlife
Jacques White	People for Puget Sound