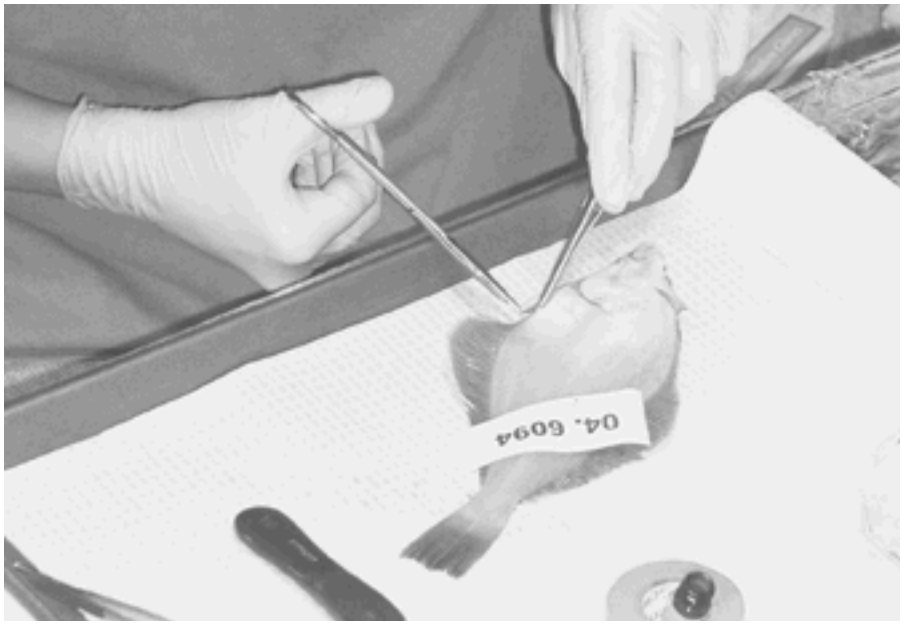
4 Toxic Contamination



A scientist studies an English sole from Elliott Bay. | *Sarah Brace*

1. Overview

In the past 150 years, people have released a wide variety of chemicals into Puget Sound and watersheds, many of which are toxic to humans, animals, and plants. While contamination by a number of toxics, such as lead, polychlorinated biphenyls (PCBs), and dioxins, has been reduced by use restrictions, other chemicals continue to be used and many enter into Puget Sound through stormwater runoff, wastewater discharges, and nonpoint sources, adding to a legacy of contamination.

Puget Sound is unique among North American estuaries, because of its geologically young, deep, narrow, fjord-like structure. Several shallow sills restrict the entry of deep oceanic water into Puget Sound, which reduces flushing of these inland marine and estuarine waters compared to the other urbanized estuaries of North America. Thus, toxic chemicals that enter Puget Sound remain longer within the system, and the trapping of toxics means that biota are subject to increased exposure. This hydrologic isolation also puts Puget Sound at higher risk from nutrients and pathogens that may enter the system.

The combination of hydrologic isolation with the persistent (resisting degradation) and bioaccumulative (increasing within in organisms over time) nature of many chemical contaminants creates additional risk for the Puget Sound ecosystem. For example, chinook salmon that remain as residents in Puget Sound (both as a result of natural tendencies and hatchery practices), rather than migrate to the ocean, are several times more contaminated than other chinook populations along the West Coast. Another disturbing indication of this is found in Pacific herring, one of Puget Sound's keystone forage fish species. These fish live almost all of their lives in pelagic waters, so one might suspect they would be among the least contaminated of fish species. However, PSAMP scientists have shown high body burdens of PCBs in this species from the central and Southern basins of Puget Sound—comparable to herring from northern Europe's severely contaminated Baltic Sea.

Table 4-1. Chemicals of concern in Puget Sound.
(Source: PSAT)

Metals (and organometals)	Organic compounds
Arsenic	Polychlorinated biphenyls (PCBs)
Cadmium	Polycyclic aromatic hydrocarbons (PAHs)
Copper	Pesticides
Lead	Dioxins and furans
Mercury	Phthalate esters
Tributyl tin	Polybrominated diphenyl ethers (PBDEs)
	Hormone-disrupting chemicals – including bisphenol A, nonylphenol, 17b-estradiol, and ethynylestradiol

The toxic contaminants that harm or threaten the health of the Puget Sound ecosystem include chemicals designed and synthesized to meet industrial needs, agricultural products such as pesticides, byproducts of manufacturing or the combustion of fuel, fossil fuels, and naturally occurring toxic elements that may become unusually highly concentrated in the environment because of human uses or other activities. Table 4-1 lists chemicals currently of highest concern in Puget Sound. Release of these chemicals to the environment can occur through designed and controlled human actions (e.g., application of pesticides or the discharge of wastes through outfall pipes, smokestacks, and exhaust pipes) or as unintended consequences of human activities (e.g., oil and chemical spills, leaching from landfills, and runoff of chemicals from the deterioration or wear of roofs, pavement, and tires).

Key findings reported in this chapter include:

- Approximately one percent of Puget Sound sediments are highly degraded, 31 percent are of intermediate quality, and 68 percent are of high quality. The **degraded sediments** (as measured by toxicity, chemistry, and benthic infauna) are mainly associated with urban embayments that are often located near river deltas and other highly productive nearshore habitat of importance to Puget Sound species.
- Chinook salmon from Puget Sound have nearly three to five times the **PCB** levels of chinook from Alaska, British Columbia, and Oregon.
- Flame retardants, or **polybrominated diphenyl ethers** (PBDEs) occurred in 17 percent of sediment sites sampled in Hood Canal in 2004 and were detected in 16 percent of samples from 10 Puget Soundwide sediment sampling sites in 2005.
- **PBDEs** are now second to PCBs in order of importance in the Puget Sound food web. PBDEs in English sole from urban areas are almost 10 times higher than those levels measured in sole from the Georgia Basin. Herring from Puget Sound have nearly three times the levels of PBDEs in Georgia Basin herring. Harbor seals from Puget Sound have over twice the PBDEs found in seals near Vancouver, BC. Scientists estimate that PBDE levels are doubling every four years in marine mammals, including harbor seals and orcas, and will surpass PCB levels in these species by 2020.
- In Puget Sound sediments, polycyclic aromatic hydrocarbons (**PAHs)** have not changed significantly over the past decade, except in Bellingham Bay, Port Gardner, and Anderson Island, where levels have increased. Point Pully (in central Puget Sound) had a significant decrease in PAHs during this same period.

- In Dungeness crab, **PAH** exposure was six times higher in urban areas than in non-urban areas. English sole had three to four times the PAH exposure in urban areas, compared to non-urban areas.
- English sole from Elliott Bay and the Thea Foss Waterway had four to six times the risk of developing liver lesions, (typically associated with **PAH** exposure), compared to sole from Hood Canal or the Strait of Georgia.
- Six **endocrine-distrupting compounds** (bisphenol A, estradiol, ethynylestradiol, and three phthalates) were detected in more than 20 percent of surface-water samples collected in King County's lakes, rivers, streams, and stormwater discharges.
- Male English sole from several Puget Sound locations (including 30 percent of males from Elliott Bay) are producing an egg-protein (vitellogenin) normally found only in female fish. This finding suggests that these fish have been exposed to **endocrine-disrupting compounds**.
- In Longfellow Creek, an urban stream in Puget Sound, **pre-spawn mortality** occurred in 25 to 90 percent of female salmon returning to the stream between 2002 and 2005, suggesting that contaminants from stormwater are posing a threat to the spawning success of salmon in urban streams.

Pathways of Toxics into Puget Sound

Toxic chemicals can be introduced into Puget Sound through:

- Discharges of wastewater and stormwater through outfalls.
- Nonpoint runoff and groundwater discharges to surface waters.
- Spills.
- Atmospheric deposition.
- Release of by-products from other contaminants that break down or change in the environment over time.
- Import of contaminants through biological migrations.
- Re-suspension, re-circulation, and bioaccumulation of contaminants into other organisms or other parts of the ecosystem.

2. Sediment Quality in Puget Sound

Chemical contaminants from industrial and municipal point sources, stormwater runoff, and atmospheric deposition in the Puget Sound watershed are generally discharged, flow, or fall into the nearest water body. Ultimately, most make their way to Puget Sound. Those that are not water-soluble typically bind to silt and clay particles in the water and settle to the bottom. Bottom sediments in Puget Sound are final repositories for many chemical contaminants and serve as records of what is being (or has been) released into the environment.

a. Sediment Monitoring

Some contaminants have physical and chemical properties that bind them tightly to the sediments, and they become biologically unavailable to organisms that contact them. Many remain bioavailable and, whenever organisms live in or ingest contaminated sediments, they can be directly harmed or, indirectly, can accumulate these chemicals in their tissues and transfer them to other animals in the food web.

Collection and analysis of Puget Sound sediments have been conducted over many years to reveal the identities and quantities of contaminants that have been released into the environment and have accumulated in various locations. Sediment analyses also measure the harm chemical contaminants may cause to the estuarine organisms that live in or on them.

Assessing the condition of Puget Sound's sediments

The Sediment Quality Triad Index was developed as a weight-of-evidence approach that combines the results of the sediment chemistry, toxicity, and benthic invertebrate analyses generated in this study to classify the overall quality of the sediment samples. Four categories of sediment quality were generated to define each station and, ultimately, each sediment monitoring region and strata of the study. They are:

High Quality: No degradation detected in any of three test parameters.

Intermediate/High Quality: Degradation detected in one of three test parameters.

Intermediate/Degraded Quality: Degradation detected in two of three test parameters.

Degraded Quality: Degradation detected in all three test parameters.

Status and Trends

Ecology developed estimates of degraded sediments in Puget Sound from 1997 to 1999[1] (Long, et al. 2003, 2005). These estimates indicated that approximately one percent of Puget Sound sediments are degraded, 31 percent are of intermediate quality, and 68 percent are of high quality. Recent sediment quality data was collected in 2002 and 2003 from 81 new stations in three additional Puget Sound regions, including the San Juan Archipelago, the eastern Strait of Juan de Fuca, and Admiralty Inlet (Long, et al. in prep.). Data on chemistry, toxicity, and benthic infaunal community structure from all 381 stations were used in a Sediment Quality Triad Index to identify spatial (geographic) patterns and spatial extent of degraded sediment quality in eight monitoring regions, five strata (such as harbors and bays and for the entire Puget Sound study area. These data complete the sediment quality baseline for Puget Sound, spanning from 1997 through 2003.

Sediment monitoring regions were defined by their hydrologic, bathymetric, and geological features, as well as by the distribution of biota and differences in the degree of sediment quality in these regions (Figure 4-1). The majority of stations and the highest percent of the area from the Strait of Georgia, Whidbey Basin, Admiralty Inlet, Hood Canal, and South Sound regions were of high quality. Samples classified as degraded were collected in the Whidbey Basin, central Sound, Hood Canal, and south Sound regions, representing 0.2 percent, 2.3 percent, 0.9 percent, and 0.1 percent, respectively, of the area within each region. Degraded sediments in these four regions were identified primarily from Everett Harbor, Elliott and Commencement bays, Port Gamble, Port Ludlow, and Budd Inlet. The largest percentages of stations and areas with intermediate sediment quality were found in the San Juan Archipelago, Strait of Juan de Fuca, central Puget Sound, and south Puget Sound regions.

Sediment monitoring strata—harbor, urban, basin, passage, and rural—are defined by their major geographic features and degree of anthropogenic activity (Long et al. 2003). The strata also differed dramatically in their degrees of sediment quality (Figures 4-2: Basins, Harbors, Passages, Rural and Urban). The largest percentages of stations (47 percent) and areas (14 percent) with degraded sediment quality were found in the harbor stratum, while five percent of the stations and four percent of the area were degraded in the urban strata. Intermediate sediment quality was also most pervasive in harbors, followed by urban strata. Highest sediment quality was measured in passage, basin, and rural strata.

When calculated for the whole 1997-2003 Puget Sound baseline study area, the Sediment Quality Triad Index indicated that sediments from approximately 7 miles^2 (19 km^2), or 0.8 percent of the study area, were degraded (Table 4-2, page 138). Sediments with intermediate quality were distributed over 318 miles^2 (826 km^2), or about 35 percent of the area. High-quality sediments were found in 596 miles^2 (1543 km^2) representing 65 percent of the study area.

Impacts to the Ecosystem

The health of Puget Sound may be negatively affected by even a small proportion (0.8 percent) of degraded sediments. The areas classified as degraded are located in and around the urban/industrial embayments of the Sound, primarily in river deltas known to be critical nearshore habitat for many species. Most of the Puget Sound species identified as endangered or threatened rely on the nearshore habitat and their declines may be, in part, related to degraded sediments.

[1]For PSAMP and NOAA's National Status and Trends Program.

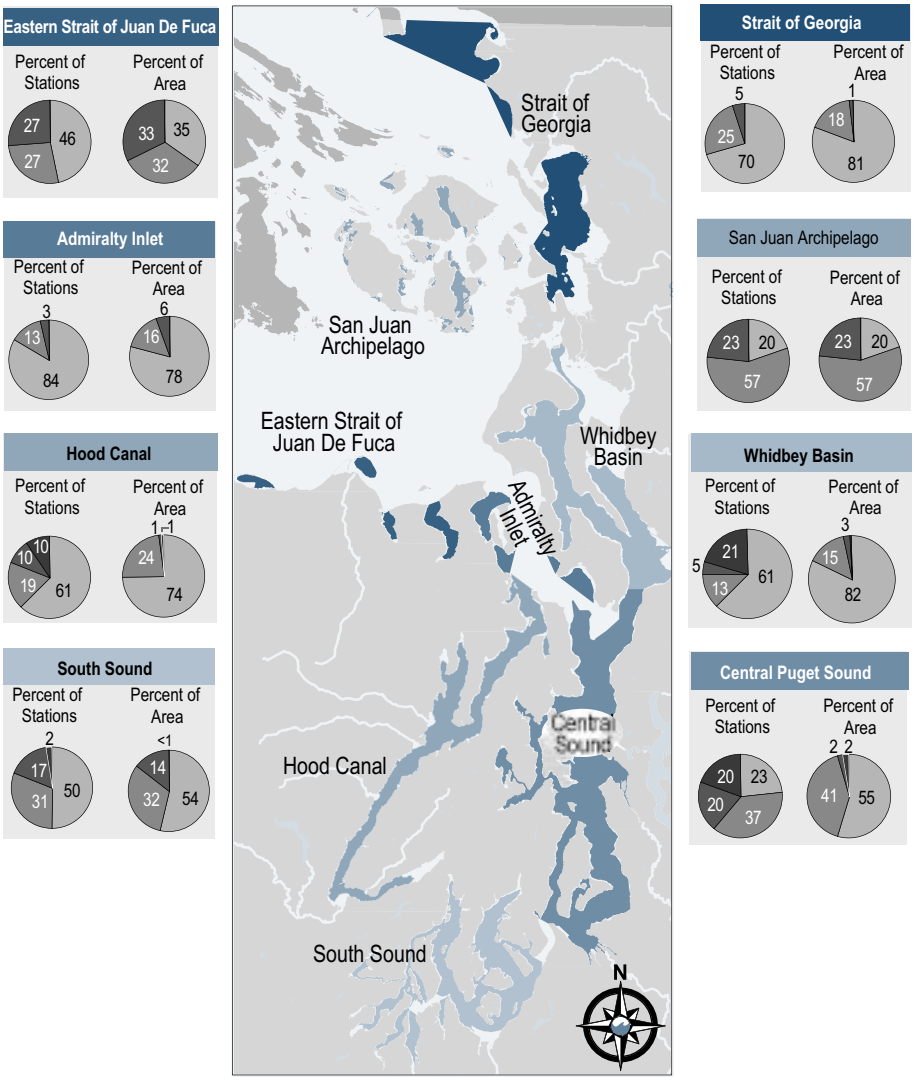


Figure 4-1. Sediment Quality Triad Index in eight Puget Sound monitoring regions, 1997-2003. Percent of stations and percent of area, representing each index category, are depicted for each region.
(Source: Ecology)

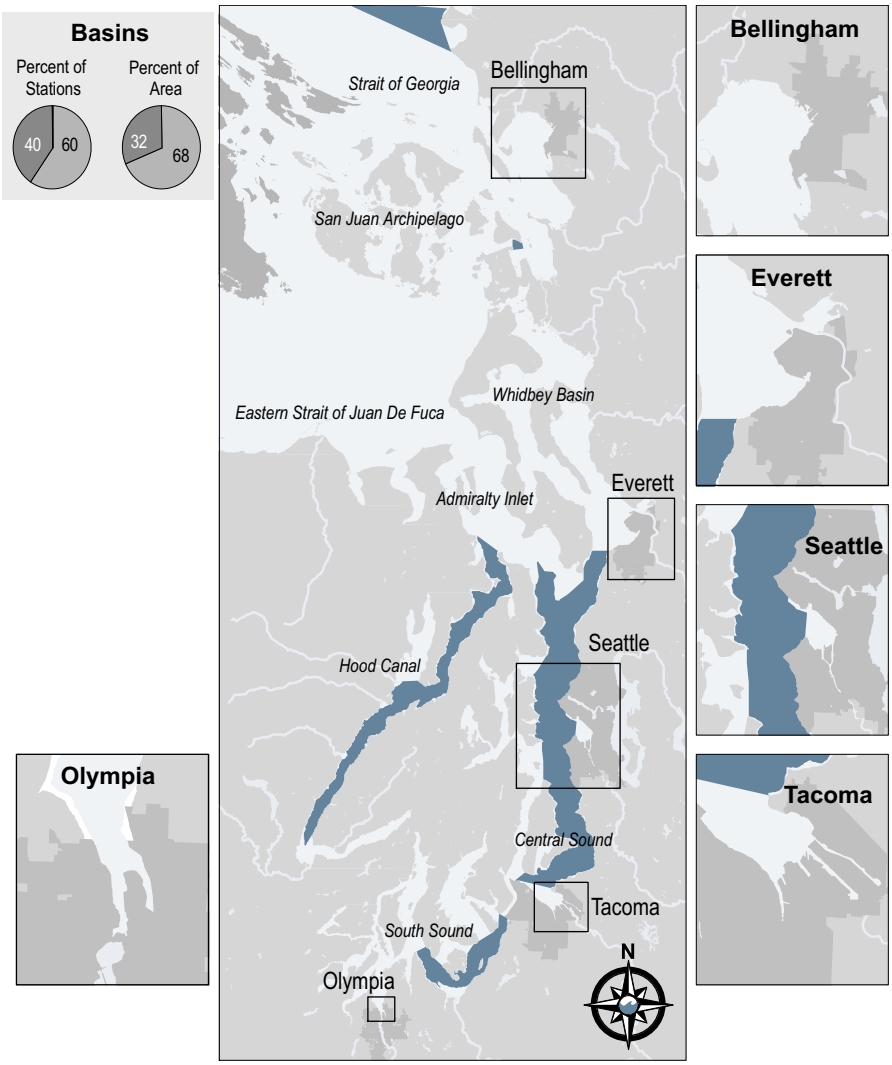
Sediment Quality Triad Index
- High
- Intermediate/High
- Intermediate/Degraded
- Degraded

High levels of toxic chemicals are present or are linked to health impairment in organisms that reside in, or whose food resources are tied to, the more urban and industrialized embayments of central and southern Puget Sound (PSAT 2002, 2004). Examples include high levels of PAHs and/or PCBs measured in the bodies of shellfish (Dungeness crab), fish (English sole, demersal rockfish, coho salmon, and Pacific herring), birds (bald eagle eggs collected from Hood Canal), and marine mammals (southern Puget Sound harbor seals and southern resident orca whales), all associated with the more highly contaminated Central and Southern Puget Sound basins. Different populations of many of these species residing in northern Puget Sound and the Strait of Georgia or feeding on prey from these cleaner locales had lower contaminant levels in their tissues.

In addition to focused attention on the 0.8 percent of the study area with degraded sediments, there is a sizeable area (34.6 percent) classified with intermediate quality sediments. Intermediate quality sediments may work together with other environmental stressors such as low dissolved oxygen levels and climate change to further negatively impact Puget Sound. Future attention and monitoring must occur in these areas to determine whether sediment conditions improve, remain stable, or deteriorate further.

Figure 4-2. Basins. Sediment Quality Triad Index in five Puget Sound monitoring strata 1997-2003. Percent of stations (left pie charts) and percent of area (right pie charts) representing each index category are depicted for each stratum.
(Source: Ecology)

Sediment Quality Triad Index
- High
- Intermediate/High
- Intermediate/Degraded
- Degraded



Human Health Consequences

As a natural resource, Puget Sound depends on high-quality sediments to maintain a viable ecosystem and economy. Toxic chemicals in sediments are a major concern, because the trophic web of Puget Sound is detritus-based (Kennish 1997). Animals that live in or on contaminated sediments may absorb the toxic chemicals and distribute them throughout the food web.

Human health risks occur primarily through consumption of fish and shellfish that have bioaccumulated contaminants. DOH and local health districts around the Sound have issued fish and shellfish consumptions advisories that warn people not to eat contaminated seafood (PSAT 2004). These advisories have been made primarily in the Whidbey Basin and Central Puget Sound, two regions identified by PSAMP with some of the largest percentages of degraded sediments. Possible health concerns from exposure to contaminants may include effects on neurological, reproductive, and immune systems, as well as cancer.

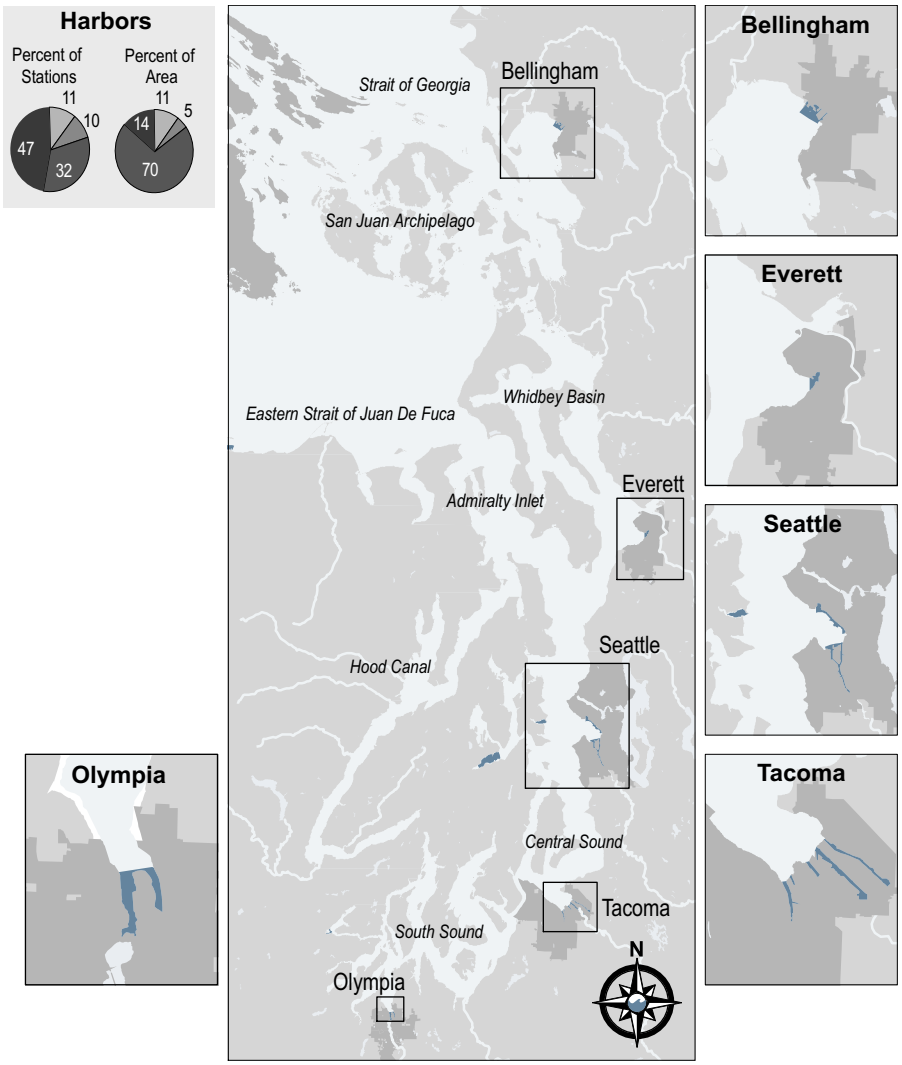
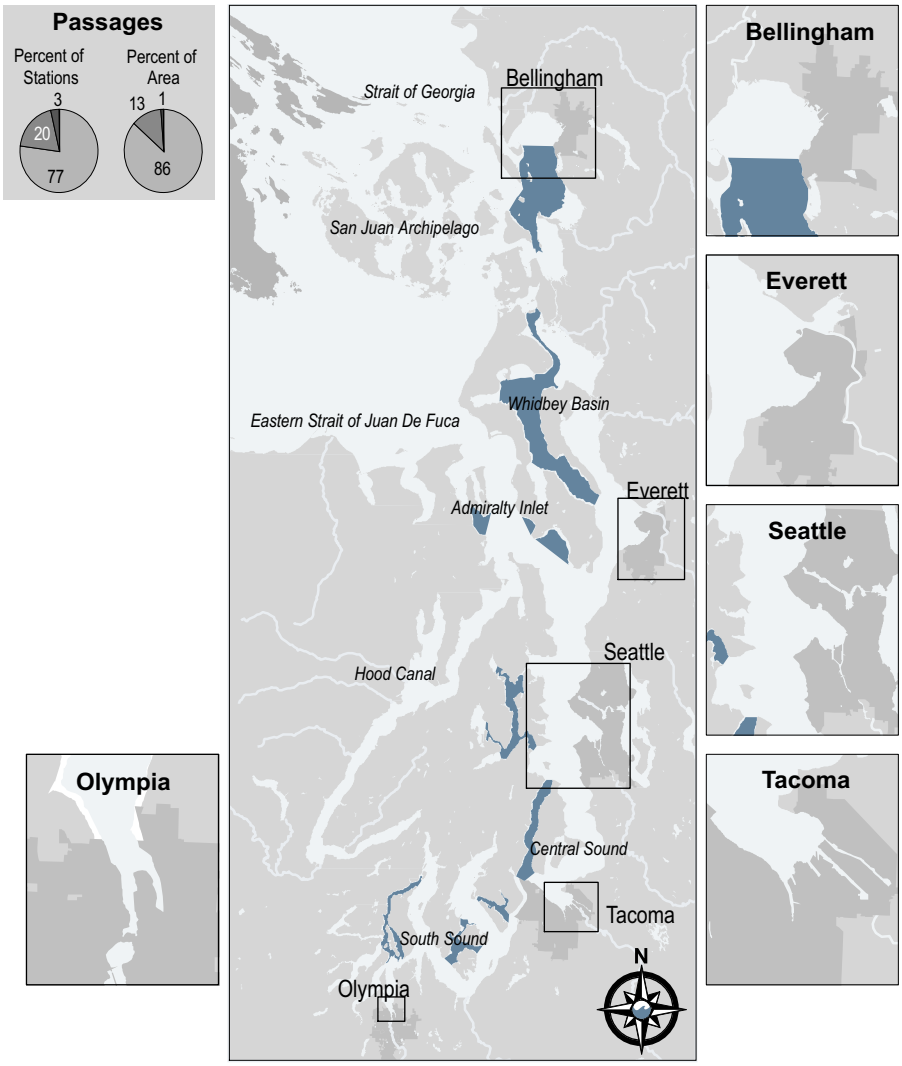


Figure 4-2. Harbors.

Sediment Quality Triad Index

- High
- Intermediate/High
- Intermediate/Degraded
- Degraded

Figure 4-2. Passages.



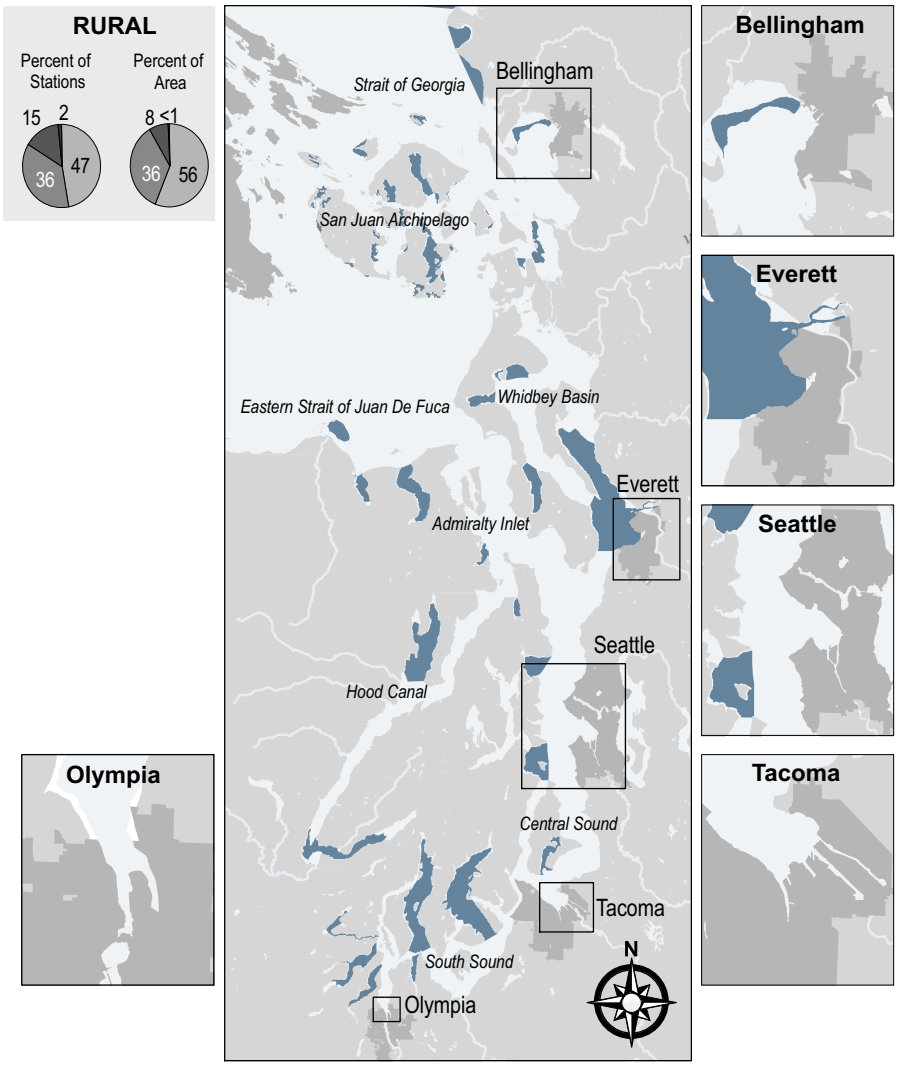


Figure 4-2. Rural.

Sediment Quality Triad Index

- High
- Intermediate/High
- Intermediate/Degraded
- Degraded

Figure 4-2. Urban.

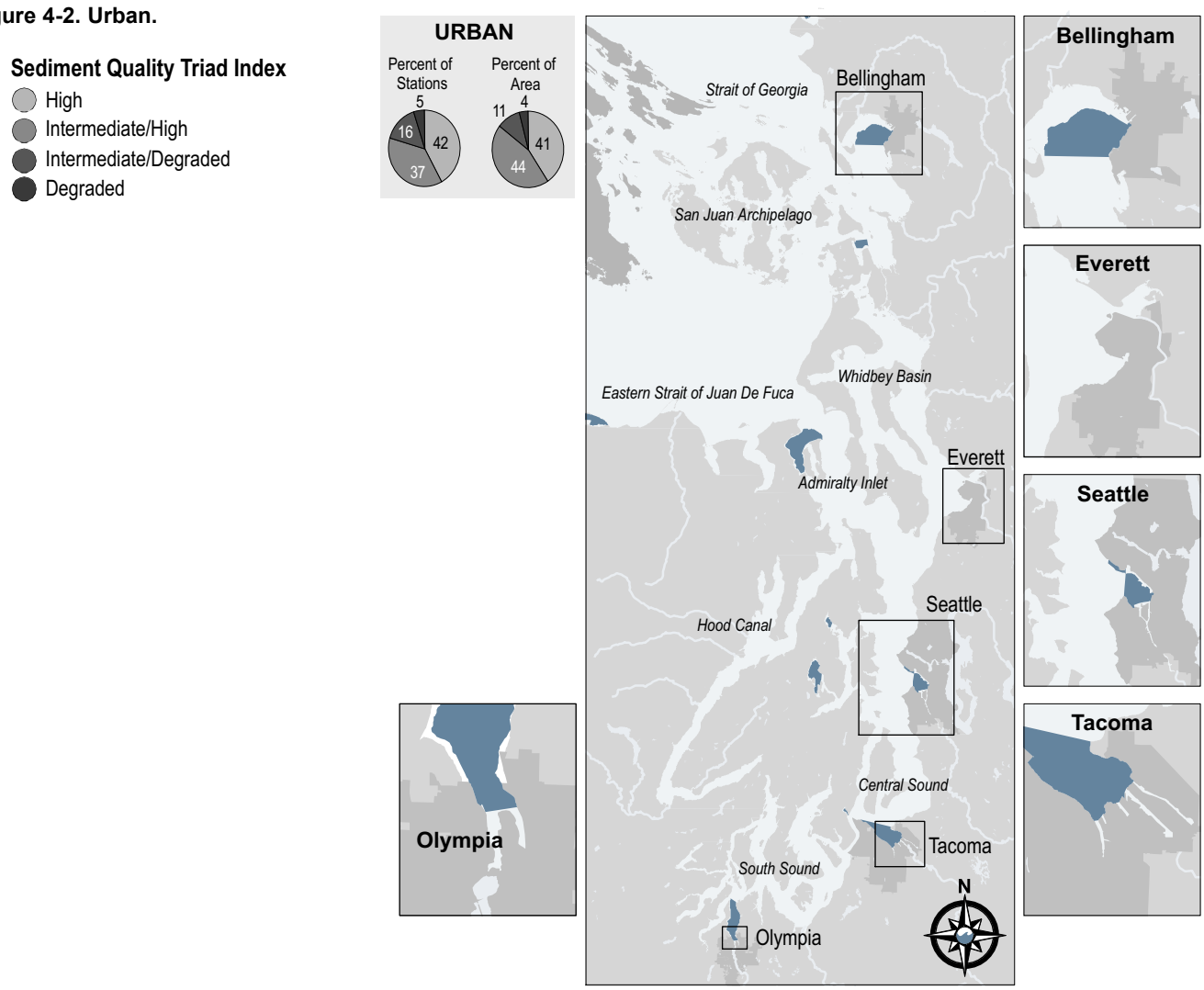


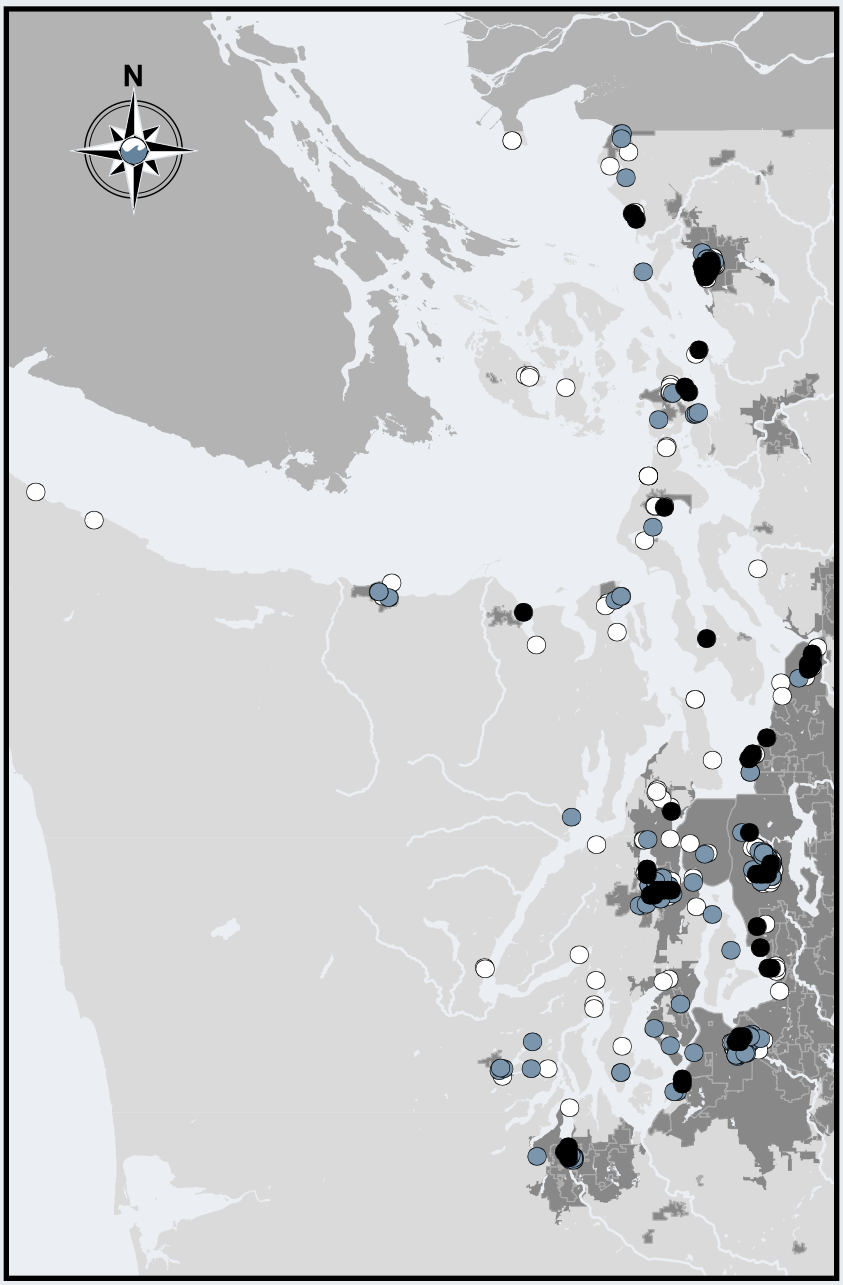
Table 4-2. Sediment Quality Triad Index in the entire Puget Sound study area, 1997-2003. The index rates sites based on chemistry, toxicity, and benthic infauna community structure. (Source: Ecology)

Sediment Quality Index Category	Stations		Area	
	No.	percent	mile²/km²	percent
Total Study area	381	100.0	1481/2389	100.0
High	176	46.2	959/1543	64.6
Intermediate/High	114	29.9	429/692	29.0
Intermediate/Degraded	55	14.4	84/134	5.6
Degraded	36	9.4	12/19	0.8

Superfund and Other Contaminated Sediment Site Cleanup Efforts in Puget Sound

EPA's Superfund program has been investigating and cleaning up contaminated Puget Sound sediments since 1980. A total of 3.85 million cubic yards of contaminated sediments have been dredged and 206 acres capped at six Puget Sound Superfund sites, with more sites under investigation (Figure 4-3). Contaminants addressed in these cleanups include PCBs, PAHs, other organic contaminants, and metals.

Additionally, Ecology's Toxic Cleanup Program is currently commencing work to clean up sediment contamination at smaller sites outside major urban areas. Target sites will be chosen, based on each site's relative ecological sensitivity and importance (for instance, whether the site provides habitat for juvenile fish and invertebrates). The first site chosen for cleanup is an area of wood-waste contamination in Port Gamble (C. Asher, Ecology's Toxic Cleanup Program, pers. comm.). Cleanup and monitoring are scheduled to occur through 2007.



Impaired marine sediment sites

Impaired marine sediment sites are based on Ecology's 303(d) impaired sediment listings (categories 4a and 4b)

Puget Sound state and federal upland cleanup sites

- State cleanup sites waiting remedial action. (115 sites)
- State and federal sites withremedial action in progress. (553 state and federal sites)

Ecology defines a Puget Sound contaminated upland sediment site as any site within 1/2 mile of the Puget Sound shoreline.

Puget Sound Urban Areas

Figure 4-3. Current contaminated sites in Puget Sound region (through mid-2006). Impaired marine sediment sites are locations where in-water sediments have been tested and found to have toxic pollutants that exceed state standards. Locations marked in blue are awaiting clean up. Those marked in white have clean up in progress.
(Source: Ecology)

Benthic, demersal, pelagic —what's the difference?

Benthic organisms, such as tube worms, sculpins, and sole, spend most of their lives living on or in the sediments of the seafloor.

Demersal organisms, such as rockfish and Pacific cod, are associated with the seafloor.

Pelagic organisms, such as herring and salmon, dwell in open waters, rarely making contact with the seafloor.

3. Persistent Bioaccumulative Toxics in the Food Web

Persistent bioaccumulative toxics (PBTs) are chemicals that persist in the environment because they resist degradation from environmental or metabolic processes. They also accumulate in tissues or fat of animals and humans and are only slowly metabolized, so that, over time, their concentrations increase in individuals. Many of these contaminants also increase (bio-magnify) as they move throughout the food web.

Most of Washington's urban and industrial centers, and the majority of its population, are located along Puget Sound's shoreline. These centers represent ongoing and historical sources of toxic contaminants, including PBTs, that are deposited into the Puget Sound estuary by natural transport and sedimentation processes. Most of the contaminants that enter the Sound are thought to attach to particles and settle out of the water column, to accumulate in bottom sediments. PSAMP has documented that the bottom sediments in urban bays of Puget Sound (and the bottomfish species associated with them) are contaminated with PBTs, and significant efforts are underway to remediate contamination in these bays. However, recent PSAMP studies have shown that Puget Sound's important pelagic species (Pacific herring and salmon) are more contaminated than herring or salmon from the Georgia Basin and other coastal estuaries of the west coast of North America. These new studies have shown that PBTs permeate the Puget Sound food web, not only in its bottom-dwelling species, but in the pelagic component of the food web as well.

Ecology recently adopted a rule (WAC Chapter 333) that identifies PBTs of greatest concern for Washington, establishes criteria for their selection, and outlines preparations for PBT chemical action plans. PCBs and PBDEs are prominent PBT groups in Ecology's rule, and they are also toxics of high prominence in PSAMP's monitoring, along with others such as aldrin/dieldrin, benzo(a)pyrene, chlordane, DDT, dioxins and furans, hexachlorobenzene, mercury, and toxaphene.

Toxic contaminants enter the Puget Sound ecosystem via water (e.g., river or stream inputs, point sources like industrial or wastewater discharges or nonpoint sources like stormwater and other runoff) the atmosphere (transport from both local and distant sources) and in the bodies of migrating organisms (biotransport). Puget Sound's semi-enclosed nature, combined with the depth and current patterns of its basins, entrain water, nutrients, toxic contaminants, and organisms, resulting in a physically and biologically isolated ecosystem that tends to retain introduced chemical contaminants. Fat-bonding, or lipophilic contaminants, such as PCBs and PBDEs, can be taken up and retained by plankton, or attach to particles and settle into the bottom sediments. PBTs retained by plankton are rapidly assimilated into the food web and accumulated by pelagic consumers such as zooplankton, and forage fish and then amplified and recycled throughout the food web to high-level predators like salmon, orcas, birds, and humans.

a. Benthic Food Web

The benthic food web comprises the complex, interrelated predator-prey relationships that exist among the plants, animals, and microbes inhabiting the seafloor. In the soft sediments of Puget Sound, benthic organisms include bacteria, foraminifera, micro- and macroalgae, sedentary species including small, tube-dwelling and burrowing invertebrates, bivalve mollusks (such as clams)

that live within the sediments, and larger, mobile surface-dwelling or burrowing invertebrates (e.g., crabs and shrimps), and fishes. Chemicals absorbed by these sediment-dwelling species can accumulate in their bodies and become magnified in the predators that consume them.

Although PBTs may be concentrated in soft sediments, the area, or zone of biological impact of those concentrated PBTs may be much larger, because of the movement patterns and feeding relationships of its occupants. Fish and invertebrate species that live on harder or more consolidated substrates such as cobble or rocky reefs (substrates where PBTs are unlikely to accumulate) often forage, or search for prey, in surrounding soft sediments, where PBTs may accumulate. Conversely, organisms that are exposed to PBTs in the soft sediments can move around and be consumed by organisms in less contaminated areas. This biotransport of PBTs can occur from a very small scale to a hemispheric scales, when migratory fish and bird species pick up PBTs in one part of the world and transport them to another.

i. PBTs in the Sediments

Sediment monitoring by Ecology and NOAA indicates that PCB-contaminated sediments are mainly concentrated in urban and industrialized bays, and the remainder of Puget Sound sediments are relatively uncontaminated (Long et al. 2005). Recent data also indicate that PBDEs are detectable in our urban bays and estuaries, although at lower levels than in other urban estuaries on the West Coast.

Status and Trends

In 2004, Ecology scientists added PBDEs to the list of chemicals measured in sediments collected annually for PSAMP. PBDE levels were measured for five congeners in June 2004 and 12 congeners in April 2005 from 10 long-term sediment monitoring stations in Hood Canal. PDBE levels will also be assessed in future years. Data from these samples will establish a baseline for PBDE concentrations in sediments throughout Puget Sound, from which changes over time can later be determined, and allow comparisons with levels measured nationally and worldwide. Also, comparisons of levels in marine waters and biota can be made to assess the role of these contaminants in both benthic and pelagic food webs.

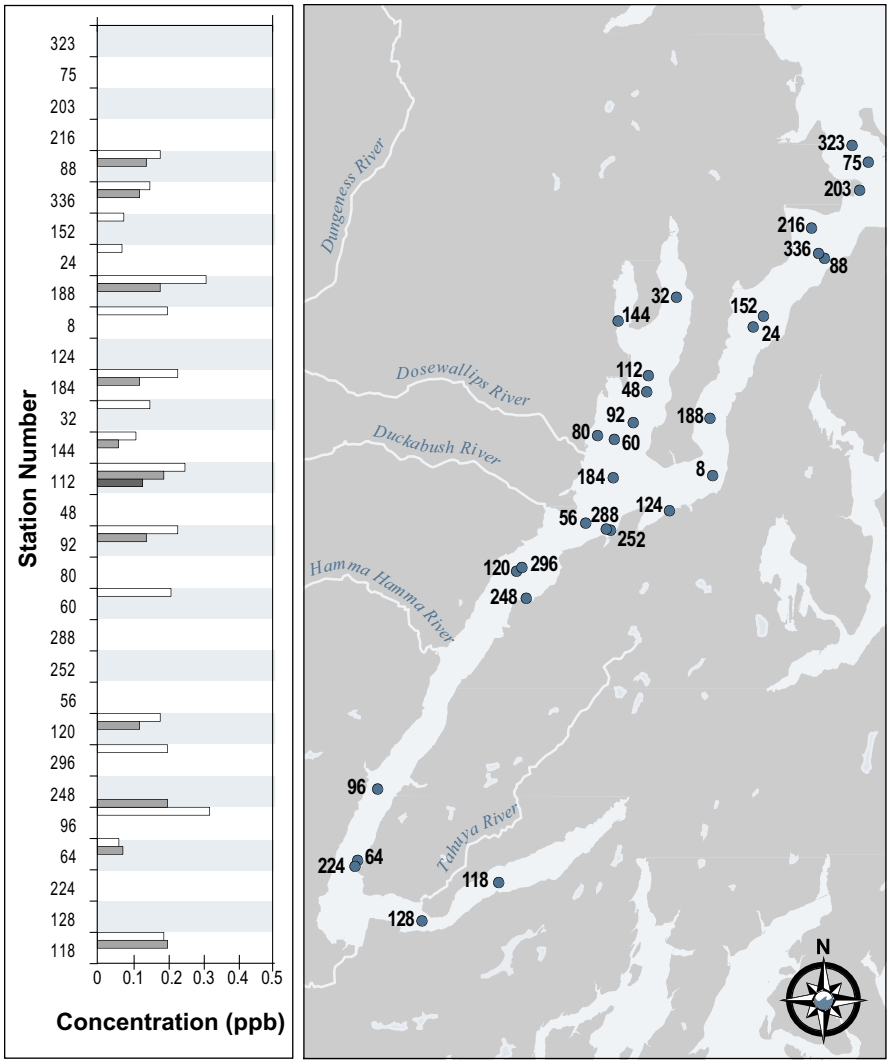
For the 2004 PSAMP sediment quality survey in Hood Canal, five PBDE congeners (BDE-47, -99, -100, -153, and -154) were measured in sediments from 30 randomly selected stations. These congeners are predominant in commercially available PBDE mixtures, and their levels in the environment have been measured worldwide. Of 180 samples, 17 percent had detectable levels of PBDE congeners, while 83 percent were below the level of detection. Congener BDE-47 was detected in 17 of 30 samples, congener BDE-99 was detected in 10 of 30 samples, and congeners BDE-100 and -153 were detected in one of 30 samples. Congener BDE-154 was not detected (Figure 4-4). All detection values that fell below the laboratories reporting limit were estimates.

Concentrations of a broader suite of PBDEs were measured in sediments collected in April 2005 at 10 PSAMP temporal sediment monitoring stations located throughout Puget Sound. Congeners measured included the five penta-BDE congeners measured in 2004, plus BDE-209 (the primary congener in the commercial deca-BDE mixtures), and BDE-49, -66, -71, -138, -183, and -184. Of 422 samples, 16 percent of the samples had detectable levels of PBDE congeners, while 83 percent were below the level of detection. As in Hood Canal, congeners BDE-47 and -99 were detected most frequently. Congener BDE-47 was detected

PCBs

Polychlorinated biphenyls (PCBs) are synthetic organic molecules characterized by a double phenyl ring surrounded by one to 10 chlorine atoms. There are 209 different individual PCB chemical compounds, called congeners, that differ only in the number and placement of chlorine atoms. PCBs were invented for use in many industries; as electrical and thermal insulators for hydraulic equipment, plasticizers in paints, plastics, and rubber products; in pigments, dyes, and carbonless copy paper; and many other applications. These compounds were designed to resist degradation, which makes them long-lasting for their intended use. Unfortunately, this characteristic makes them persistent in the environment, and the chemical structure of these molecules can be highly toxic. The unintended consequence of releasing PCBs is persistent toxic contamination of aquatic food webs. Prior to cessation of their production in the U.S. and Canada in the 1970s, more than 1.5 billion pounds of PCBs were manufactured in the U.S. alone.

Figure 4-4. Flame retardants (PBDEs) in Hood Canal. In June 2004, sediments from 30 randomly selected stations throughout Hood Canal were sampled for five congeners of PBDEs (BDE-47, -99, -100, -153, and –154). These congeners are predominant in commercially available penta-BDE mixtures, and their levels in the environment have been measured worldwide. Congeners BDE-47 and -99 were detected in 17 and 10 of 30 samples, respectively, and BDE-100 and -153 were each detected in one of 30 samples. Congener BDE-154 was not detected in any of the samples.
(Source: Ecology)



PBDEs

Polybrominated diphenyl ethers (PBDEs) are chemicals that were designed as flame-retardants for common household items, including textiles and electronics. Like PCBs, they are persistent, bioaccumulative, and toxic. PBDEs are similar in structure to PCBs except that they contain bromine instead of chlorine atoms. There are also 209 congeners of PBDEs. In January 2006, Ecology and DOH published a chemical action plan for PBDEs; their recommendations for reducing PBDEs in the environment include educating the public on minimizing exposure to PBDEs, prohibiting the manufacture, distribution, or sale of new products containing penta- and octa-PBDEs, encouraging the legislature to ban deca-PBDEs, working with stakeholders to encourage manufacturers to develop safer, effective alternatives, and ensuring that workers in certain industries are not exposed to unacceptable levels of PBDEs.

at all 10 stations, while BDE-99 was detected at five of the stations, including the deep, depositional Shilshole and Point Pully stations, as well as at the stations near urban and industrial areas, such as Port Gardner, Sinclair Inlet, and the Thea Foss Waterway. Congener BDE-209 was also detected at these five stations and at the station in Budd Inlet. Congener BDE-49 was detected at three of the stations, congener BDE-71 was detected at two of the stations, and BDE-66 and -100 were detected at only one station. Congeners BDE-138, -153, -154, -183, and -184 were not detected in any samples (Figure 4-5).

Comparisons of the Puget Sound sediment PBDE data with data from the San Francisco estuary and various European and Asian marine surveys indicated that congeners BDE-47, -99, and -209 were found in the highest concentrations worldwide. Levels of BDE-209 in Puget Sound were the lowest of all measured, while BDE-47 and -49 were lower only in three of the six other surveys (Figure 4-6).

ii. PBTs in English Sole

English sole are bottom-dwelling flatfish, widely distributed throughout Puget Sound and coastal regions of the northeastern Pacific Ocean. They are closely associated with the bottom sediments, have relatively high site fidelity, and consume benthic invertebrates. English sole is an ideal indicator species because it

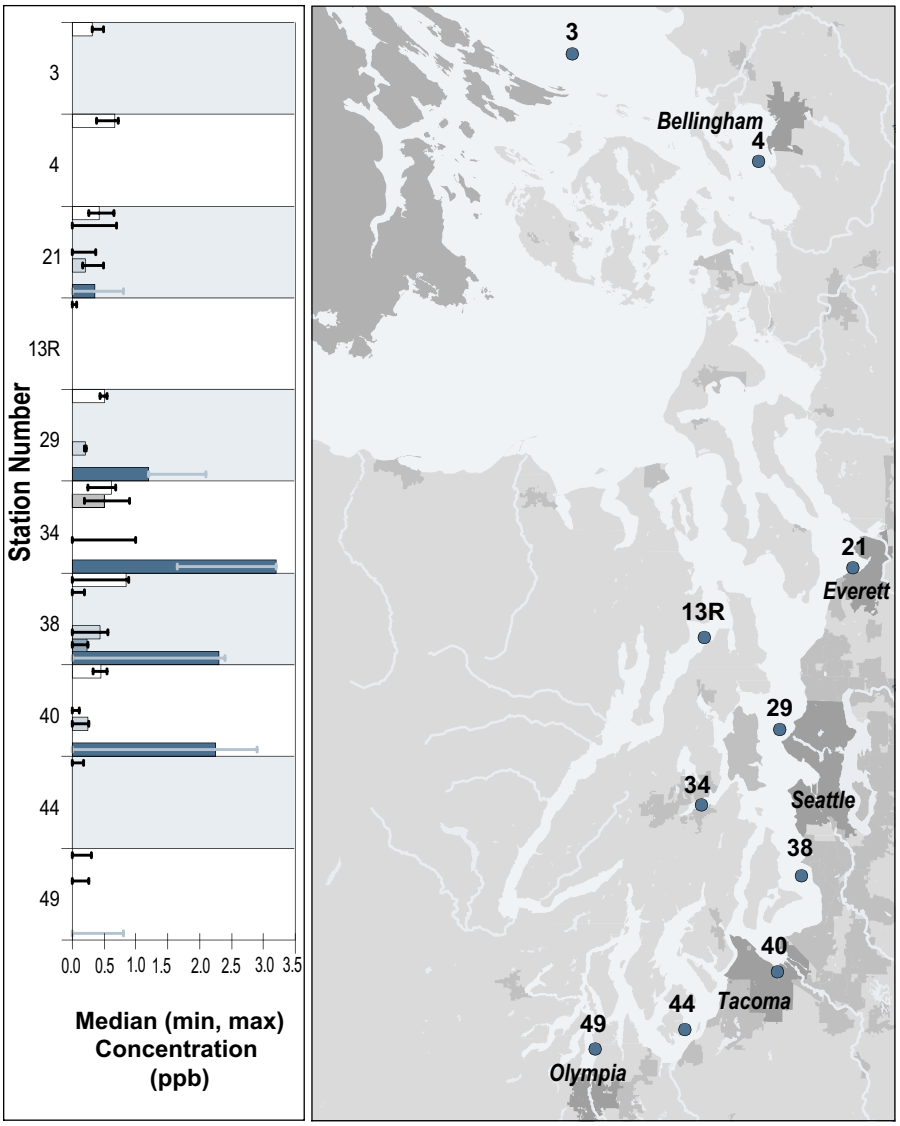


Figure 4-5. Flame retardants (PBDEs) in Puget Sound. Concentrations of a broader suite of PBDEs were measured in sediments collected in April 2005 at 10 PSAMP temporal sediment monitoring stations located throughout Puget Sound. Congeners measured included the five penta-BDE congeners measured in 2004, plus BDE-209 (the primary congener in commercial deca-BDE mixtures), and BDE-49, -66, -71, -138, -183, and -184. As in Hood Canal, BDE-47 and -99 were detected most frequently. Congener BDE-47 was detected at all 10 stations and BDE-99 was detected at five of the stations, including the deep, depositional Shilshole and Point Pully stations, and the stations near urban and industrial areas, including Port Gardner, Sinclair Inlet, and the Thea Foss Waterway. Congener BDE-209 was also detected at these five stations, and at the station in Budd Inlet. Congeners BDE-49, -66, -71, and -100, were detected at three, one, two, and one station(s), respectively. Congeners BDE-138, -153, -154, -183, and -184 were not detected in any samples. (Source: Ecology)

PBDE Congener Monitoring

Monitoring Station

BDE Congeners

BDE-47
BDE-49
BDE-66
BDE-71
BDE-99
BDE-100
BDE-209

is abundant, found in many habitats, and the levels of certain contaminants (such as PCBs) in its tissues reflect the levels of contaminants in the sediments at sites where it lives.

Status and Trends

WDFW scientists have sampled English sole consistently from the early 1990s to present at eight index sites for describing temporal trends in PBTs, and periodically at over 40 other randomly selected sites in north, central, and southern Puget Sound to describe the spatial distribution of PBTs in this species. In addition, they conducted more intensive focus studies in the three most contaminated urban bays of Puget Sound (Elliott Bay in 1997, Sinclair Inlet in 1998, and Commencement Bay in 1999) to gain a better understanding of contaminant patterns near to their sources.

PBT accumulation in fish is mostly closely related to the magnitude of PBTs in their environment. However, other factors, such as a fish's fat content, its position in the food web (trophic status), age, and gender can also determine the extent to which PBTs in the environment are accumulated. WDFW scientists have already documented elevated levels of PCB in the muscle tissue of English sole from

Figure 4-6. Concentration of 12 BDE congeners measured in marine sediments worldwide. Congeners BDE-47, -99 and -209 are detected in Puget Sound sediments but at relatively low levels compared to the UK or San Francisco measurements. (Source: Ecology)

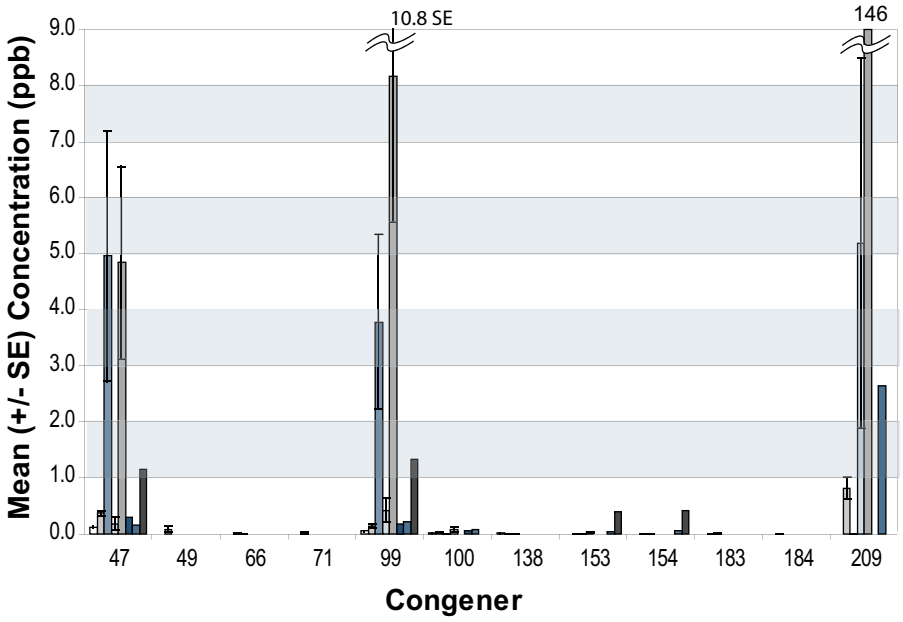
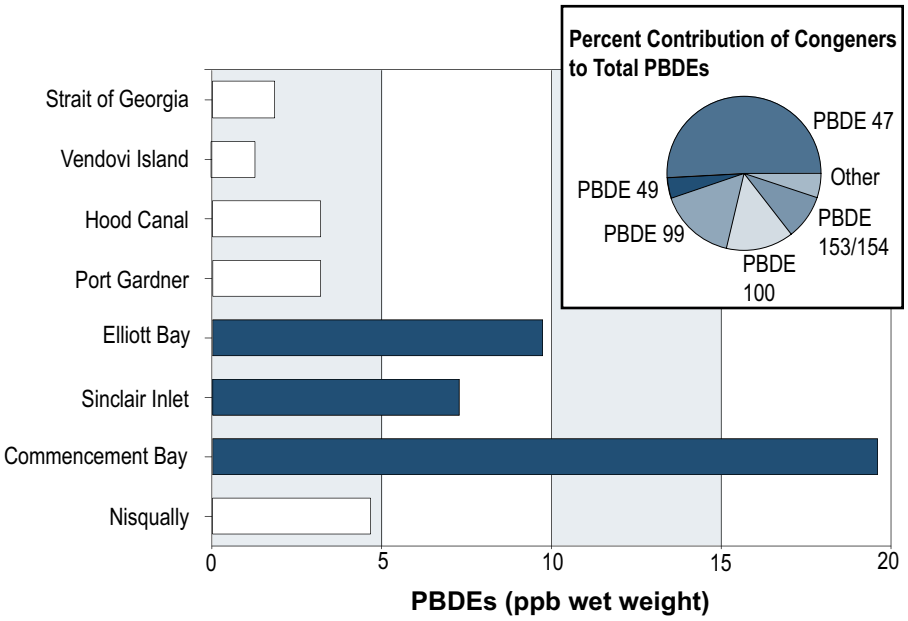


Figure 4-7. PBDE concentrations in English sole. Fish were sampled from three urban (shaded bar) and five non- or near-urban (open bar) Puget Sound locations in 2003. Each bar represents a composite value from 20 skinless muscle fillets. Highest concentrations of PBDEs were measured in fish from urban bays in central and southern Puget Sound. (Source: WDFW)



urban harbors and bays of Puget Sound, and have shown that PCB accumulation in English sole can be predicted primarily by the concentration of PCBs in sediments where the fish live, with additional accumulation as fish age (O'Neill and West, in prep.). Sediment PCB concentration was the stronger predictor, accounting for 70 percent of variation in English sole PCBs, while age contributed only four percent of the variation observed. In general, the more contaminated the sediments, the more contaminated the fish, and older fish from contaminated areas had slightly higher PCB exposures than did younger fish. In contrast, mercury accumulation in English sole can be predicted mostly by fish age (63 percent of the observed variation) and only slightly by sampling location (four percent of the observed variation).

Recent WDFW data indicate that PBDEs behave in a fashion similar to PCBs – PBDE levels are elevated in English sole from the urbanized bays of Puget Sound (Figure 4-7), at concentrations considerably higher than those reported

by Ikonomou et al. (2006) in English sole in the Georgia Basin. The pattern, or relative proportion of congeners that make up the total PBDE concentration was similar among all English sole samples, regardless of where the samples were collected, yet it was different from the PBDE congener pattern observed in sediments. This suggests that individual PBDE congeners move through the food web at different rates.

iii. PBTs in Rockfish

Quillback, copper, and brown rockfish are long-lived demersal predators, usually associated with rocky substrate. Because of their longevity and high position in the food web, their probability of exposure to PBTs is great. These species have small home ranges, suggesting that they will reflect local patterns of contamination. Because they are popular targets of recreational fisheries in Puget Sound, rockfish also represent a food-web pathway through which contaminants can move from the environment to humans.

Populations of some rockfish species are in decline, making their utility as indicators problematic. Rockfish have become rare in many areas of Puget Sound, including the South Puget Sound and Whidbey basins, so they are unsuitable as broad-scale indicators. WDFW scientists began sampling rockfish in 1989 but ended targeted sampling in 1997, after which specimens were taken only as by-catch from other efforts. Although sample sizes are small in some cases, adequate numbers have been collected to characterize three of the four highly urbanized embayments of central Puget Sound, non-urban areas of central Puget Sound, Admiralty Inlet, and the San Juan Islands.

Status and Trends

WDFW scientists have sampled rockfish from six of WDFW's nine Sport Fisheries Management Areas (Figure 4-8). They observed the greatest PCB exposure in fish from Elliott Bay (Management Area 10) and Sinclair Inlet (Management Area 13)—two of the most contaminated embayments in central Puget Sound (Figure 4-9). PCBs were also present at lower levels in rockfish from Commencement Bay (Management Area 11) and Port Gardner (Management Area 8-1), and in non-urban locations in Management Area 11. PCBs were rarely detected in rockfish from all other management areas, which reflects either a lack of exposure to PCBs (i.e., their habitats were uncontaminated), or that samplers failed to represent the full range of individuals in a population. For example, Commencement Bay habitats are contaminated with PCBs; however, rockfish are rare in the areas sampled by WDFW, and only very young fish (from two to eight years of age) were obtained from Commencement Bay.

WDFW scientists have observed that male rockfish can accumulate measurably greater concentrations of PCBs than females can. This probably results when females lose PCBs during reproduction, as they transfer PCB-laden fats to their eggs and larvae. This may happen in other species as well; however, rockfish present a particularly good opportunity to observe the phenomena, because they live long enough to accumulate high PCB levels (males) and lose PCBs through many reproductive cycles (females) (Figure 4-10). Although PCBs were highly variable in both sexes, the greatest concentrations (exceeding 200 ng/g wet wt.) were measured in the oldest males (15 to 30 years of age) from Elliott Bay and Sinclair Inlet. PCBs were almost always relatively low in females; their greatest concentration was 156 ng/g in a 9-year-old female from Sinclair Inlet (not shown) and 90 ng/g in a 10-year-old female from Elliott Bay. PCBs in females from both locations declined after this age, probably related to the start of their reproductive life.

What are congeners and Aroclors?

PCB and PBDEs are categories, or families, of chemical compounds. At their core, PCBs and PBDEs are both dual-benzene-ring structures, linked by a single carbon-to-carbon bond. With PCBs, the number of chlorine atoms and their position on the dual-ring core defines the resulting chemical, called a **congener**. PBDEs have identical dual-ring structures at their cores; however, chlorine atoms are replaced with bromine. For both PCBs and PBDEs, there are 209 congeners.

PCBs can be directly measured by quantifying and combining the 209 congeners (or a subset) that exist in a medium such as sediments or tissue, or they can be estimated by comparing the signal observed in a medium with well-known, commercial mixtures of congeners. In the Pacific Northwest, the most commonly used of these mixtures was **Aroclors**. Aroclor is a trade name applied to a number of specific PCB products, each of which contains varying degrees of chlorination, depending on the intended use of the product. For example, Aroclor 1260 contains 60 percent of its weight as chlorine, whereby Aroclor 1254 contains 54 percent, based on the specific mixture of congeners included in the product.

Figure 4-8. WDFW's Recreational Fisheries Management Units. Samples were collected from all nine Management Areas, but PCBs were rarely detected in Management Areas 6 and 7. Highest PCB levels measured in rockfish were from Management Areas 10 and 11. (Source: WDFW)

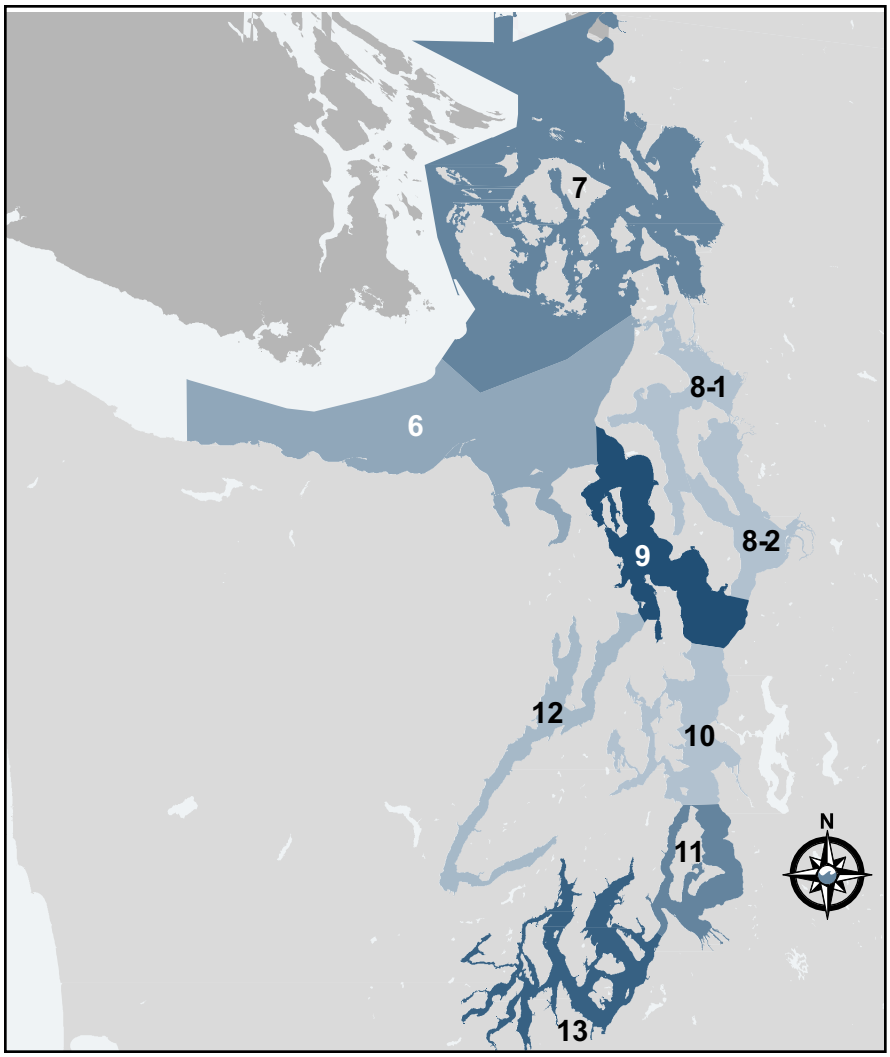
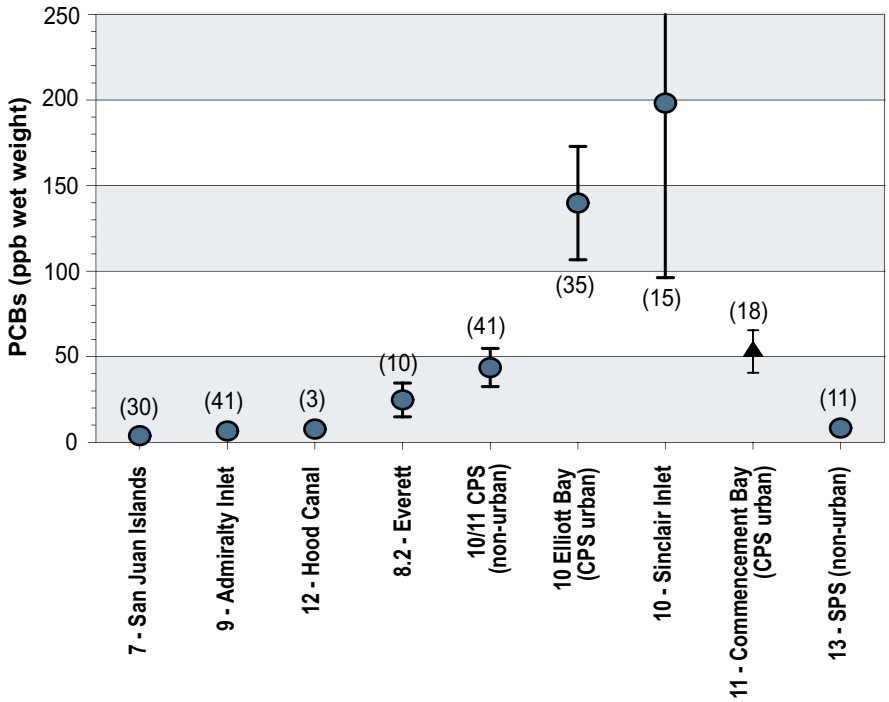


Figure 4-9. PCBs in Rockfish. Average PCB concentration (wet weight, ± 95 percent confidence interval) in three rockfish species from six WDFW Fishery Management Areas in Puget Sound. Sample sizes are indicated in parentheses. Rockfish from three urban bays, Elliott Bay, Sinclair Inlet, and Commencement Bay, had the highest levels. All samples were measured as sum of Aroclors, except Commencement Bay, which was calculated to an Aroclor-equivalent from a congener-based method. (Source: WDFW)



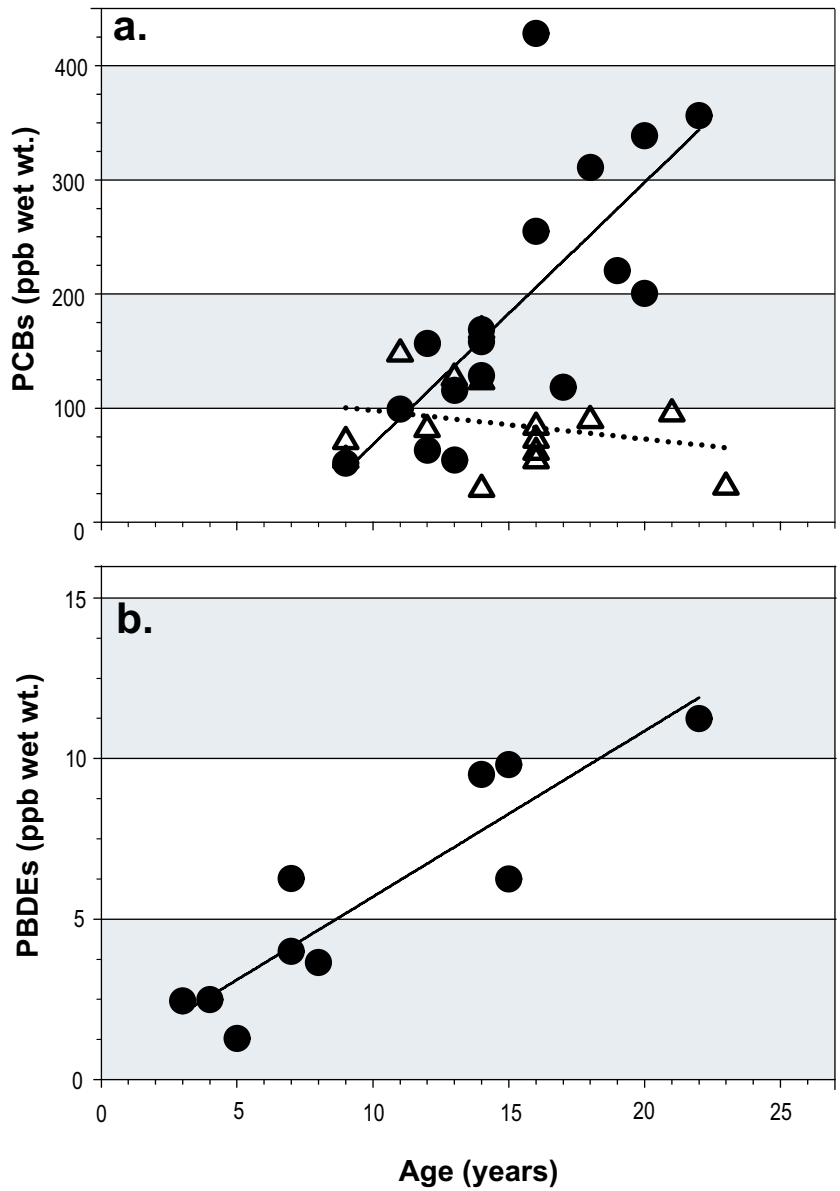


Figure 4-10. **Accumulation of (a) PCBs and (b) PBDEs with age in male rockfish sampled from the Seattle waterfront**. Unlike male rockfish, females do not accumulate PCBs with age, probably because they maternally transfer their body burdens of PCBs to their young. Female rockfish were not sampled for PBDEs, but it is expected that, like with PCBs, PBDE accumulation will remain low as fish age.
(Source: WDFW)

(a) PCB accumulation
Males ● r^2=0.54 **Females** △ r^2=0.06

(b) PBDE accumulation
Males ● r^2=0.83 **Females** △ Not sampled

As in English sole, PBDEs appear to behave similarly to PCBs in rockfish. Based on a subset of male quillback rockfish sampled along the highly urbanized Seattle waterfront[2], male rockfish also appeared to accumulate PBDEs as they aged (Figure 4-10); however, PBDE concentrations were lower than the PCB levels observed for these same fish, possibly indicating that environmental levels of PBDEs have not yet reached those of PCBs, a conclusion that is supported by PBDE results reported in the Sediments chapter of this update.

iv. PBTs in Lingcod

Lingcod are voracious benthic carnivores that are typically associated with rocky or vegetated habitats, and they consume a wide range of fish and invertebrate prey. They are sedentary, exhibiting a relatively small home range. Because of their large size and piscivorous (fish-consuming) natures, they are one of the few predators of adult rockfish. These characteristics indicate that the lingcod's risk of exposure to PBTs is relatively high.

[2] PCBs were measured as the sum of Aroclors.

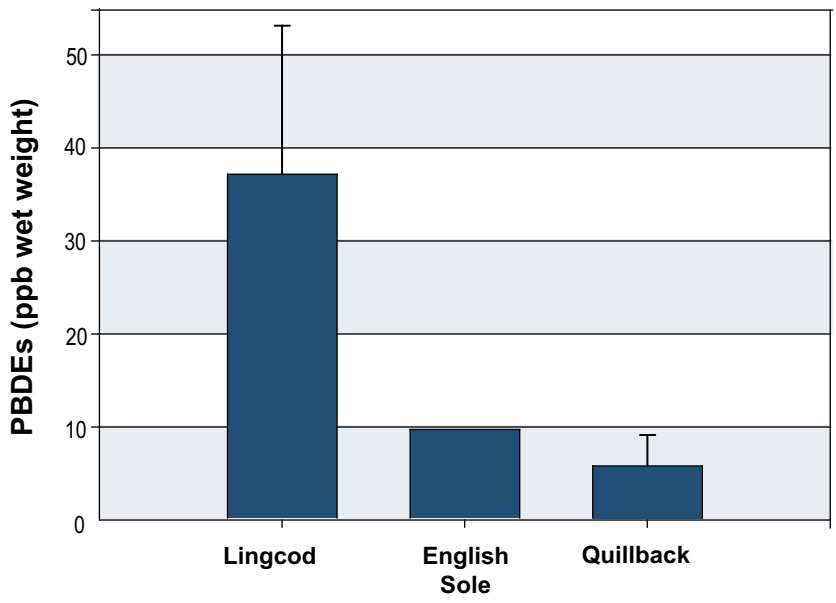


Figure 4-11. Average total PBDE concentrations detected in muscle tissue of benthic and demersal fish species sampled along the Seattle waterfront in 2003. Lingcod values are based on an average of five females (95 percent confidence interval). English sole sample is a composite sample of 20 fish, and quillback rockfish represents the average of 10 males.
(Source: WDFW)

Status and Trends

In 2003, WDFW collected lingcod, quillback rockfish, and English sole along the Seattle waterfront for PBDE analysis. PBDE concentrations were highest in lingcod, followed by English sole and quillback rockfish (Figure 4-11). Additional data (not shown) indicate that PBDEs did not accumulate with age in lingcod, although only females were sampled; as with rockfish, it is expected that PBDEs would accumulate to higher concentrations in male lingcod.

b. Pelagic Food Web

The pelagic food web comprises the complex, interrelated predator-prey relationships that exist among the plants, animals, and microbes that inhabit the water column of Puget Sound. The foundation of the pelagic food web consists of phytoplankton (single-celled algae) that fix sun energy via photosynthesis. Accordingly, they grow only in the sunlit surface layer (photic zone) of Puget Sound, which is typically less than 60 feet in depth on average. Phytoplankton is consumed by protozoa and zooplankton, which, in turn are prey of higher levels of marine food web species, including fish and whales.

Even though pelagic and benthic food webs are presented separately in this update, these two webs are also strongly linked. Pelagic species often consume benthic species, and vice versa. In addition, many marine organisms, including most benthic fishes and invertebrates produce planktonic eggs or larvae which mix with zooplankton species that spend their entire lives in the water column.

PBTs that enter the Puget Sound must pass through the water column before they reach the sediments, and it is this phase where pelagic organisms can intercept PBTs. PBTs can be absorbed by or attached to phytoplankton, microbes, and others, thereby directly introducing PBTs in the pelagic food web. In addition, sediment-PBTs may become re-suspended into the water column by storms, dredging, or animals' activities (e.g., digging and burrows), making them available to the pelagic food web. Once in the biota, PBTs can accumulate and biomagnify.

PSAMP scientists and their collaborators evaluate water-column contamination by measuring PBTs directly in the water, as well as in various species selected as

indicators. Recent results from King County's water-column sampling efforts in Elliott Bay and the Duwamish River, planktivorous blue mussel sampling from NOAA's Mussel Watch program, and WDFW's Pacific herring and Pacific salmon monitoring efforts are presented in the following sections of this update.

i. King County Water PCB Levels

King County completed a Water Quality Assessment of the Duwamish River and Elliott Bay in 1999, to evaluate the effects of combined sewer overflows (CSOs) in these water bodies. The Environmental Fluids Dynamic Computer Code (EFDC) model—a hydrodynamic and fate and transport numerical model—was used as part of this assessment, to estimate chemical concentrations under conditions with and without CSO discharges. King County is currently updating the EFDC model, in part to refine the predictions of total PCB concentrations in the water column of the Lower Duwamish Waterway.

Status and Trends

King County collected a limited number of water samples in the Duwamish River, Green River, and Elliott Bay. Data from the latter two locations will provide information on boundary conditions and for measuring PCBs in the water column, as inputs from freshwater flows and marine tidal flows to the Duwamish River. Water samples were collected in August, September, November, and December 2005 from four locations, identified in Figure 4-12, and date are listed in Table 4-3.

The August and September samples represent dry-weather, low-flow conditions and the November and December samples represent wet-weather, higher-flow conditions. All samples were analyzed for 209 PCB congeners. These data constitute a relatively small sample set, and King County is currently in discussion with other parties involved in the Lower Duwamish Waterway cleanup efforts, to evaluate the possibility of further study of PCBs in the Duwamish River.

ii. PCBs in Mussels

Since 1986, NOAA's National Status and Trends (NS&T) Program has been monitoring contaminants in mussels (*Mytilus edulis* and *M. californicus*) from Puget Sound, the Strait of Juan de Fuca, and the Pacific Ocean coast of Washington as part of their Mussel Watch program. The NS&T Program analyzes nearly 150 separate chemicals in whole soft tissue from composites (consisting of tissue from multiple individuals mixed into one sample) of mussels collected at each of each of 17 sites in Washington and one at the Columbia River south Jetty. Samples were collected annually to 1994 and every other year since then.

Mussel tissue is considered an indicator of the water-column contamination, because this species consumes (by filtering) primarily phytoplankton. The Mussel Watch data can help track changes in contaminants, including PBTs, PAHs, and metals that accumulate in mussel tissue, providing a gauge of local water quality near monitoring sites.

Status and Trends

Data from five Washington sites in 2003 indicate that total PCB concentrations in mussels have averaged slightly higher than the 2003/2004 national median of 50 ppb dry weight (Figure 4-13). Fourmile Rock, located near Elliott Bay in Seattle, had especially high PCB levels in the early 1980s, (more than 1,500 ppb) followed by general decline to 262 ppb dry weight in 2002. This level though lower, is still more than five times the national median for PCBs in mussels.

Figure 4-12. King County's 2005 water column PCB congener sampling locations in Puget Sound.
(Source: KC DNRP)

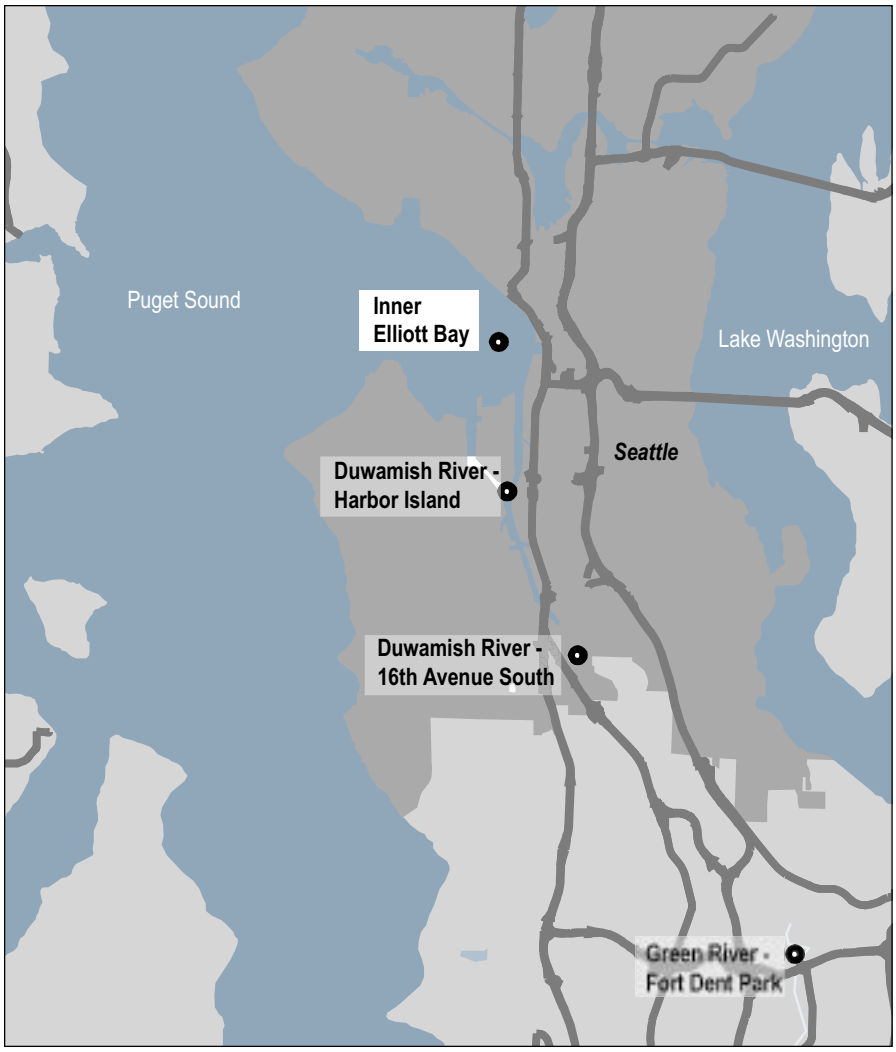
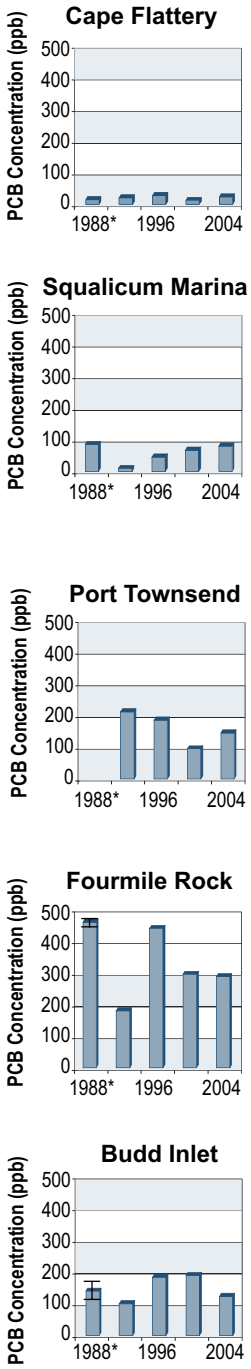


Table 4-3. The total PCB concentrations detected during the four sampling periods in the Duwamish River, Elliott Bay, and Green River, 2005. The Duwamish River and Green River sites generally had higher PCB levels in the late summer (August and September), whereas the Elliott Bay site had lower levels during this period.
(Source: KC DNRP)

		Total PCBs	(pg/L)	
Station/Depth	**Aug-05**	**Sep-05**	**Nov-05**	**Dec-05**
Inner Elliott Bay (15 meters)	65.6	152	151	131
DR – Harbor Island (1 meter)	1,800	1,100	616	2,050
DR – Harbor Island (salt wedge)	1,810	No Data	261	679/524*
DR – 16th Ave. S. (1 meter)	1,430/1,620*	1,160	474	1,130
DR – 16th Ave. S. (salt wedge)	3,120	1,720	185	1,340
Green River (surface)	248	814	933/113*	82.9

*Field replicate samples

Figure 4-13. PCBs in Puget Sound mussels. Average PCB levels in Puget Sound mussel tissue—approximately 80 ppb dw—are 60 percent higher than the national median of 50 ppb dry weight. Fourmile Rock, located near Seattle, had times the national median PCB levels in 2002, 262 ppb dry weight. However, this level is considerably lower than concentrations in the mid-1980s. (Source: NOAA)



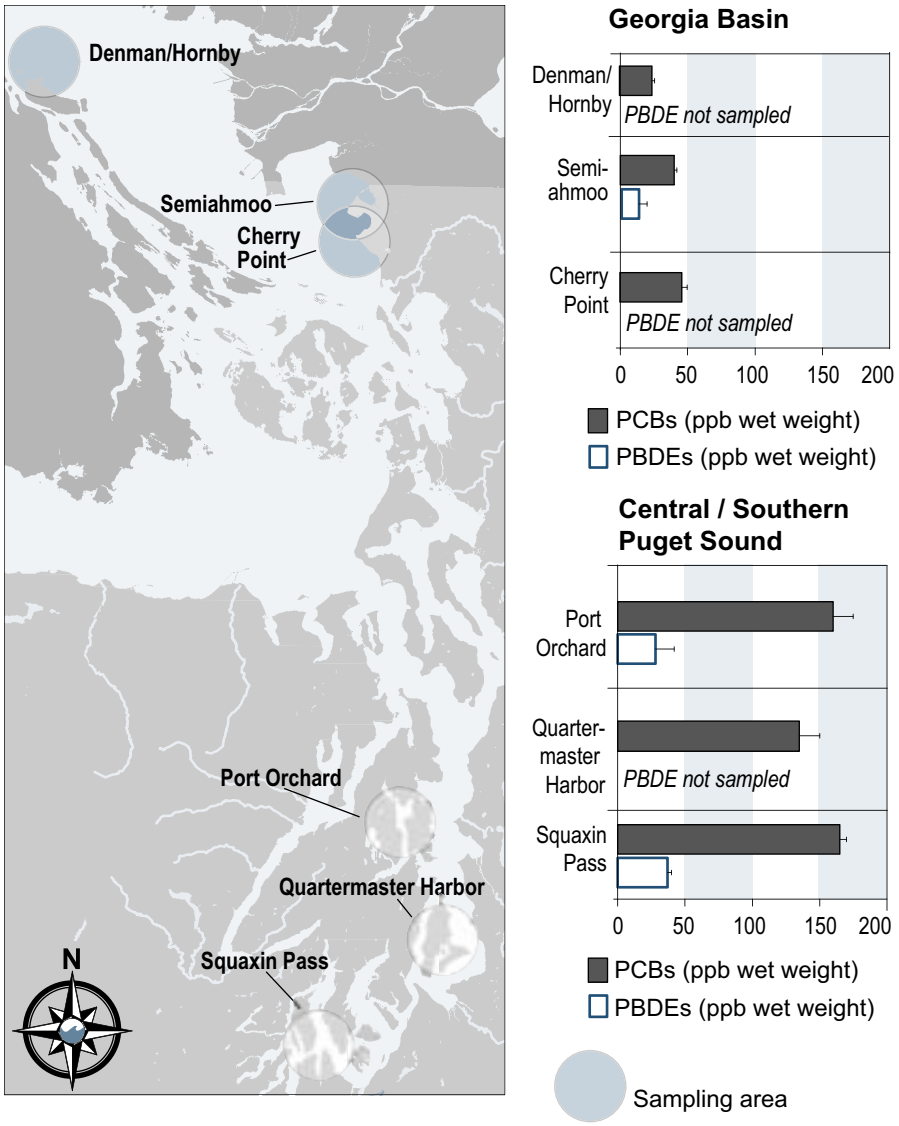
Elsewhere throughout Puget Sound and the Straits of Juan de Fuca, PCB concentrations were about 100 to 200 ppb in the mid-1980s and have since declined to below 100 ppb by 2002, with the exception of a slight increased in the mid-1990s. This peak was highest at the Fourmile Rock and Duwamish Head (near Seattle) sites but also occurred elsewhere in Puget Sound, such as at Sinclair Inlet. Following this peak, PCB concentrations have been declining again.

Generally, over the past two decades, sites with the lowest PCB concentrations were along Washington's outer coast, in northern Puget Sound and the Straits of Juan de Fuca.

iii. PCBs in Herring

Pacific herring are important prey to many fish species, as well as seabirds and marine mammals. Consequently, the health of these higher trophic level organisms is linked to the health of herring in Puget Sound and the Georgia Basin ecosystem. PCBs and other lipophilic compounds that may be present in the environment are accumulated in fatty fish such as herring. Also, because of their pelagic schooling behavior, average contaminant exposures in adult spawning stocks of herring will likely reflect environmental contamination from the geographic areas in which they reside. WDFW collects whole body samples for PSAMP, as general indicators of contamination of forage fishes in the pelagic food web.

Figure 4-14. Concentrations of PCBs and PBDEs in whole-body samples of Pacific herring. Fish were sampled from six spawning populations in the Georgia Basin and Puget Sound. Concentrations of PBDEs in central and southern Puget Sound herring samples are almost three times higher than concentrations in herring from the southern Georgia Basin. (Source: WDFW)



Status and Trends

The levels of PCBs in Pacific herring sampled from central and southern Puget Sound from 1999 to 2004 are four to nine times higher than those from the Georgia Basin sites (Figure 4-14), and showed no trend from 1999 through 2004 (Figure 4-15). The levels in Puget Sound herring are similar to those measured recently in herring from the heavily industrialized Baltic Sea—long considered one of Europe's most contaminated inland seas (Figure 4-16).

Pacific herring sampled in central and southern Puget Sound in 2004 also exhibited PBDE levels almost three times greater than those in herring from the southern Georgia Basin (Figure 4-14). Although trend data are not yet available for PBDEs in Pacific herring, data from other studies show that, unlike PCBs, PBDEs are rapidly increasing in the marine food web (Ikonomou et al. 2006).

Elevated PBT levels in Pacific herring from Puget Sound are another indication that the Puget Sound water column is contaminated with PBTs. Pacific herring consume zooplankton, which have no obvious, direct trophic connections to benthic biota. Phytoplankton may directly intercept PBTs as they enter marine waters from outside sources (e.g., atmospheric transport, point sources, and

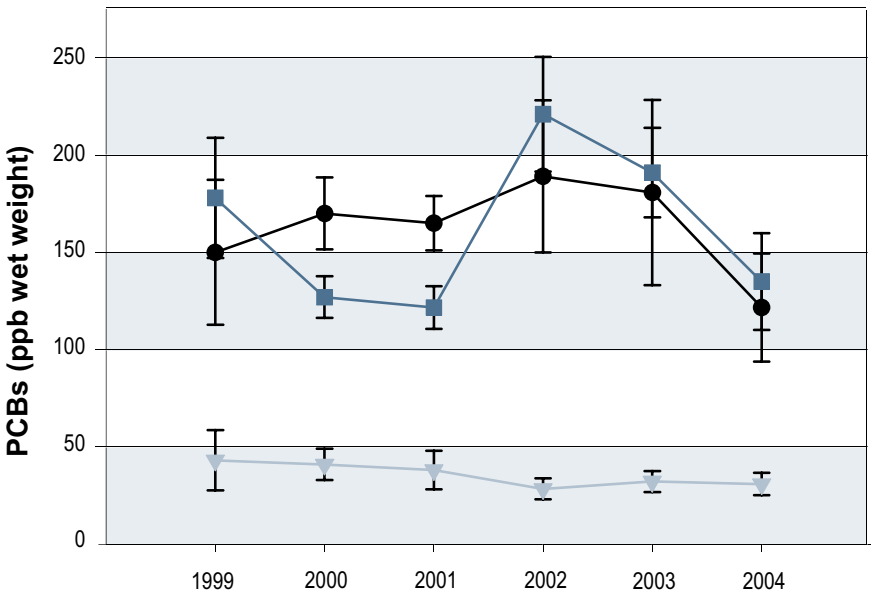


Figure 4-15. Trends of PCBs in Pacific herring. Samples were from two Puget Sound (Squaxin and Port Orchard) and one Georgia Basin (Semiahmoo) population from 1999 through 2004. PCB levels in Puget Sound herring were four to nine times higher than the Georgia Basin population. (Source: WDFW)

- Port Orchard
- Semiahmoo
- Squaxin

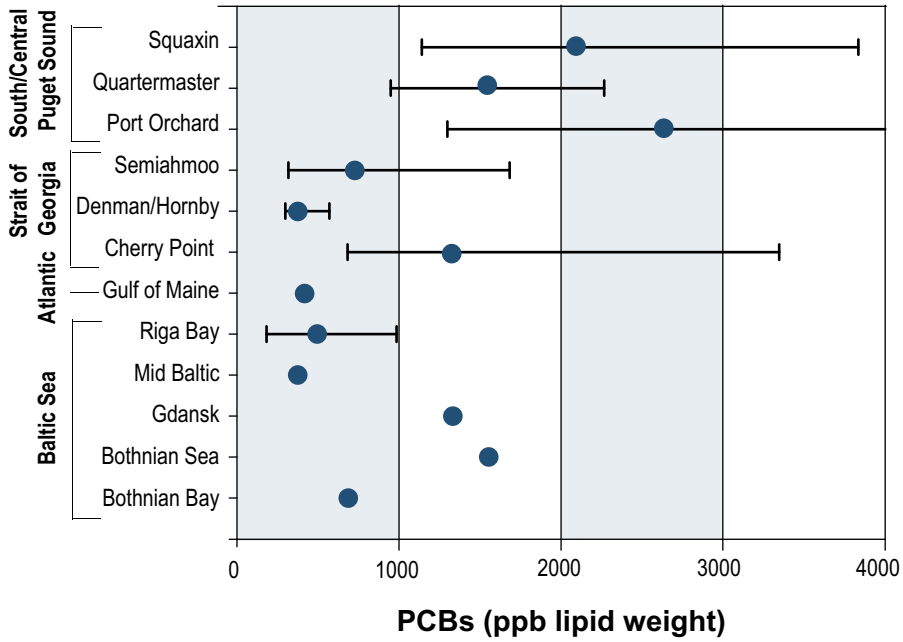


Figure 4-16. A comparison of current PCB levels in herring from the Puget Sound and Georgia Basin with other locations worldwide. Data points (circles) show reported measures of central tendency (mean or median) of lipid-weight concentrations, and short vertical lines indicate minimum and maximum reported values, when available. PCB levels in herring from three Puget Sound populations exceeded levels measured from one Atlantic Ocean and five Baltic Sea locations. (Source: WDFW)

nonpoint source fresh waters) before they settle to sediments or, perhaps, from internal processes, such as when PBTs are re-suspended from sediments via storms, waves, and other physical processes. Contaminants in phytoplankton can be transferred to zooplankton, which are then transferred to herring. This is because many zooplankton are larvae of benthic fish and invertebrates, which may be contaminated by maternal transfer of PBTs from female benthic fishes and invertebrates that feed in contaminated urban harbors and bays.

iv. PBTs in Pacific Salmon

The level of exposure to PBTs in Pacific salmon and other fishes depends for the most part on where the salmon live and what they eat. Highly migratory species such as Pacific salmon can encounter a wide range of contaminant conditions in

their lives, from highly polluted estuaries like the lower Duwamish River to the comparatively cleaner waters of the Pacific Ocean. However, the great majority of PBT accumulation by salmon occurs not in their freshwater or estuarine phase, but in the marine phase of their lives (O'Neill et al. 1998). Habitats used by salmon in their marine phase include the inland marine waters of Puget Sound and the Georgia Basin, as well as the Pacific Ocean, and it is in this phase where most of the salmon's growth occurs.

In their first year at sea, pink, chum, and sockeye salmon rapidly migrate northward and westward through marine waters of the West Coast to the open waters of the North Pacific Ocean, Gulf of Alaska, and Bering Sea (Quinn 2005). Hence, the majority of their growth occurs at a great distance from coastal pollution sources. In contrast, chinook and coho salmon have a more coastal marine distribution along the continental shelf (Quinn 2005) and so may be more exposed to PBTs from coastal pollution sources.

The PSAMP herring contaminant studies (described in Section 3b.iii of this chapter) have indicated that the pelagic food web of the central and southern Puget Sound is more contaminated with PBTs than is the Strait of Georgia. Hence, salmon populations that originate from the central or southern Puget Sound are more likely to be exposed to PBTs because they feed on contaminated Puget Sound prey during their migration to and from the ocean. Moreover, salmon that remain as residents in central or southern Puget Sound (rather than migrating to the ocean) have a greater likelihood of PBT exposure because they feed on Puget Sound prey for a greater proportion of their lives.

In addition to proximity of salmon to contaminant sources, the position of each species in the food web (i.e., what they eat) can strongly affect its exposure to PBTs. The five species of salmon that occur in Puget Sound and Georgia Basin utilizes a wide range of feeding strategies. Chinook salmon occupy the highest position in the food web and, therefore, have the greatest likelihood of exposure to and biomagnification of PBTs such as PCBs. Chinook salmon consume fish and invertebrates, but more of their food comes from herring and other forage fish. In contrast, pink and chum salmon eat mostly invertebrates that accumulate lower levels of contaminants.

Status and Trends

The following sections present recent PSAMP analyses that support the hypothesis that residency (or time spent) in Puget Sound increases exposure of coho and chinook salmon to persistent bioaccumulative toxics like PCBs and PBDEs. Some wild chinook salmon, and, to a lesser extent, coho salmon, may naturally reside in Puget Sound—particularly populations from southern Puget Sound. In addition, some artificial rearing practices are designed to encourage residency in Puget Sound (such as delaying the release of yearling chinook salmon from hatcheries and extending rearing of coho salmon in marine net pens) which may inadvertently increase the fish's exposure to PCBs. Results include both artificially reared salmon and wild salmon.

Coho Salmon

PCB accumulation in adult coho salmon returning to spawn appears to be primarily related to the fish's migration distance through, or their residency in, Puget Sound (Figure 4-17). Agate Pass net-pen-reared coho, whose release had been delayed to encourage Puget Sound residency, exhibited the greatest PCB concentrations of any coho group sampled by PSAMP in the period from 1998 through 2002. All other coho, which consisted of a mixture of wild fish and

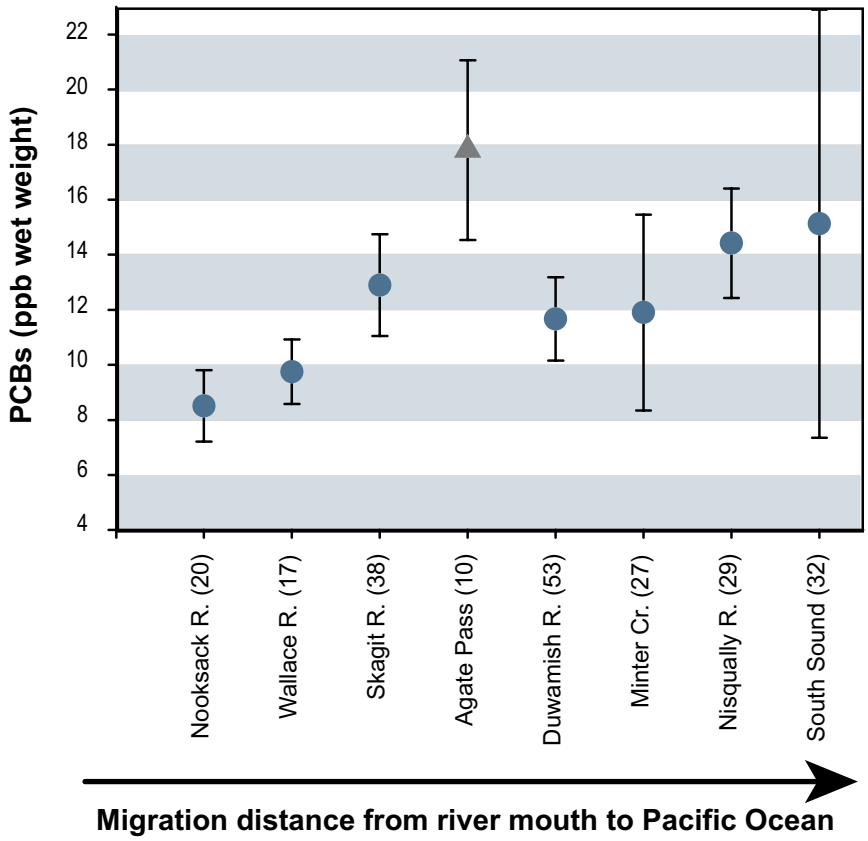


Figure 4-17. Average PCB concentration in fillets from adult coho salmon sampled from 1998 through 2002. The highest PCB levels (congeners) were measured in net-pen-reared fish whose release was delayed to prolong their residence time in Puget Sound. In all other groups, a strong trend of increasing PCBs is evident in coho salmon populations as their migration distance through Central and Southern Puget Sound increased.
(Source: WDFW)

● Hatchery (normal release) and Wild

▲ Net Pen - delayed release

hatchery-reared fish that had been released with normal timing, exhibited an increase in PCB concentrations, with increasing migration distance through central and southern Puget Sound. This corroborates previous PSAMP work by O'Neill et al. (1998), who documented higher PCB concentrations in wild coho salmon returning to spawn in central and southern Puget Sound, relative to coho returning to the Nooksack and Skagit rivers in the North Puget Sound and southern Georgia Basin. These results support the migration-distance hypothesis, or, alternatively, suggest that coho salmon populations from southern areas of Puget Sound may have a greater proportion of naturally-occurring Puget Sound-resident individuals. PSAMP scientists are continuing these studies, and have collected coho salmon from a number of locations throughout Puget Sound in 2006.

Chinook Salmon

A comparison of PCBs in chinook salmon from Puget Sound and other West Coast populations indicated that overall, Puget Sound chinook salmon fillets are nearly three times more contaminated than fillets of chinook salmon from other West Coast populations (Figure 4-18). Data shown are from Puget Sound samples that were collected 1992-1996 and data from other areas were adapted from other reports (Krahn et al. 2002; Hites et al. 2005; Missildine et al. 2005; EPA 2002). As mentioned previously, chinook salmon accumulate most of their PCBs in the marine environment, so the elevated PCB levels in Puget Sound chinook salmon suggest that these fish distribute themselves differently in marine waters from other populations along the West Coast. Moreover, the PCB concentrations in Puget Sound chinook salmon varied greatly among individuals, further suggesting that not all Puget Sound chinook salmon have the same marine distribution.

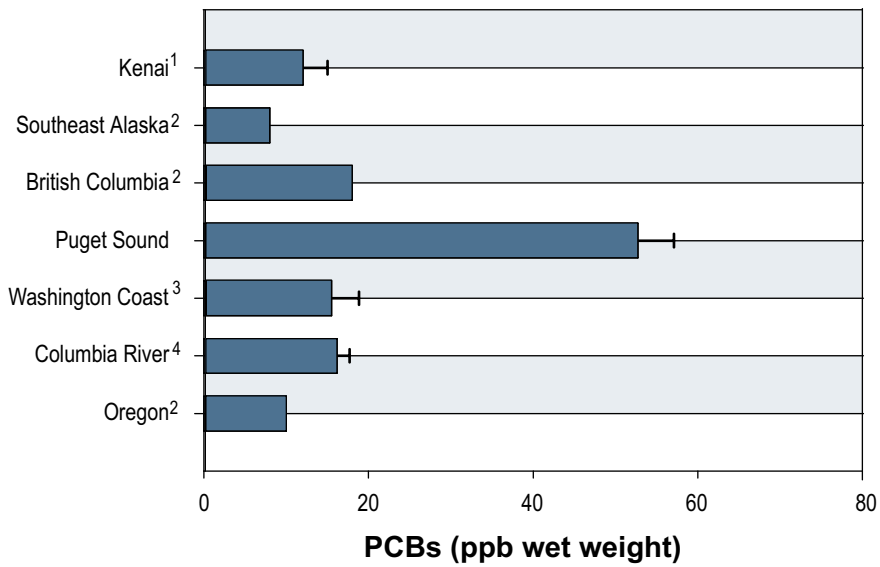


Figure 4-18. PCBs in chinook fillets. Average PCB concentration in chinook salmon fillets (wet weight, with 95 percent confidence interval) sampled from Alaska, British Columbia, Puget Sound, the Columbia River, and the coast of Washington and Oregon. Data for Puget Sound were based on fillet samples collected by PSAMP from 1992–1996 and the results for the other six locations were adapted for use here from other reports. Chinook salmon fillets from Puget Sound had nearly three times the concentrations of PCBs than chinook salmon from other areas. This may be due to longer residency times in Puget Sound. (Source: WDFW)

Adult Puget Sound chinook salmon typically migrate to coastal waters rather than offshore (Myers et al. 1998), however the total time a chinook salmon spends feeding in coastal waters as opposed to residing within Puget Sound waters is unknown, and probably highly variable. Based on the range of contaminants observed in PSAMP studies, a significant proportion of Puget Sound chinook salmon are probably at least partially resident in Puget Sound, and some Puget Sound chinook may reside in these waters year-round. Feeding within Puget Sound on herring and other forage fish exposes the chinook salmon to higher contaminant levels than along the Pacific Ocean coast or in the open ocean.

A recent independent PSAMP/NOAA Fisheries study of PBT accumulation in whole body samples of individual chinook salmon along the West Coast confirmed that summer/fall chinook from Puget Sound were considerably more contaminated than other West Coast populations (Figure 4-19). Furthermore, Puget Sound resident chinook salmon (i.e., chinook salmon captured in central Puget Sound outside the normal migration time for adult fall chinook salmon) had the highest concentrations of PBTs.

PBDEs were measured above the limit of detection (1 ppb) in five of these seven chinook salmon groups, and concentrations ranged from roughly one-fifth to one half that of PCBs. In addition, the pattern of PBDEs matched that of PCBs, suggesting that PBDEs and PCBs behave similarly in the ecosystem.

c. Extended Food Web

i. PBTs in Osprey

Ospreys are long-lived, fish-eating birds of prey with high nest fidelity and that, typically, catch fish within short distances of their nest sites. Nesting osprey in the Pacific Northwest are migratory, spending winters in southern Mexico and northern Latin America, where industrial contamination is low (Henny et al. 2002). The osprey is a good avian indicator species of toxic, persistent, and bioaccumulative contaminants, such as PCBs, as well as selected emerging toxics (such as PBDEs and herbicides). Because osprey fish close to their nesting sites, contaminants in osprey eggs reflect local exposures to chemicals in breeding areas. Figure 4-20 shows an osprey with a flatfish (known to contain high concentrations of PCBs and other contaminants).

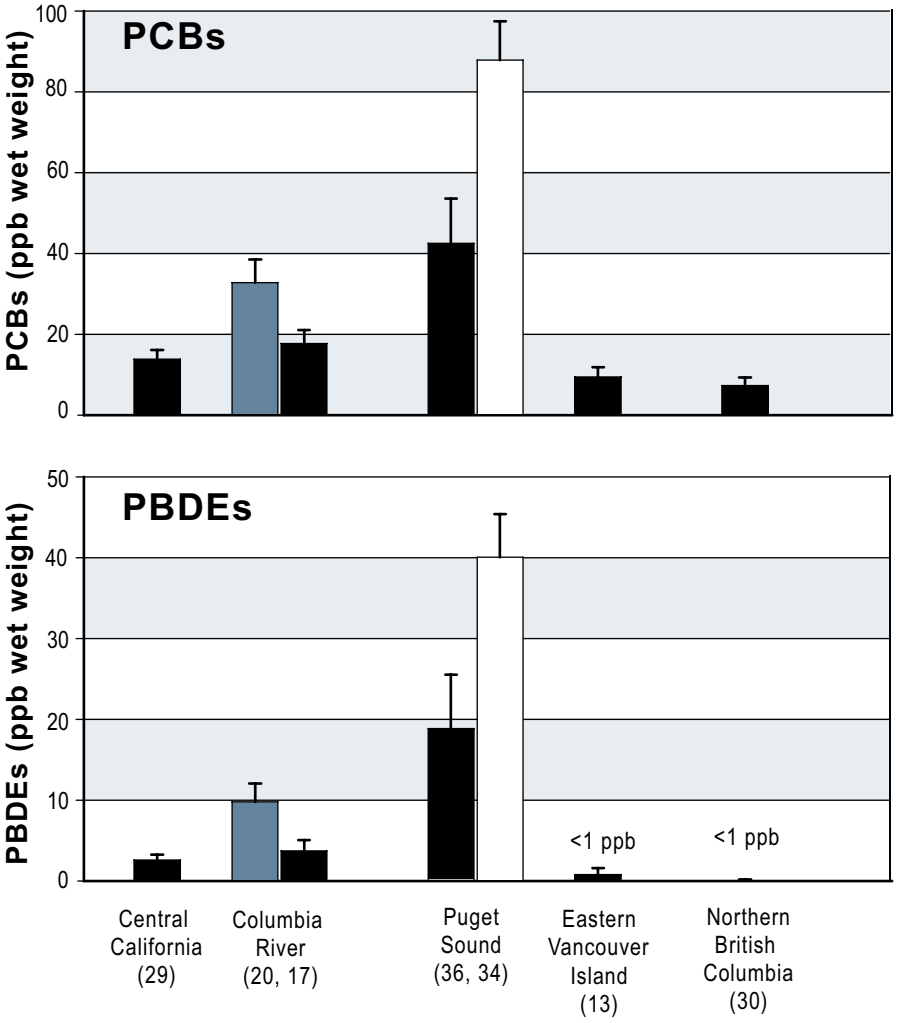


Figure 4-19. PCBs and PBDEs in whole body chinook. Average concentrations of PCBs and PBDEs (with 95 percent confidence interval) in whole body samples of individual chinook salmon caught in terminal fishing areas. River populations represented include fish returning to the Sacramento/San Joaquin (central California); Columbia (spring and fall run); Nooksack, Duwamish, and Nisqually Rivers (Puget Sound); the Fraser and Nimpkish Rivers (eastern coast of Vancouver Island, Georgia Basin); and the Skeena River (northern B.C.). Additional data are shown for sub-adult chinook salmon that were resident in Puget Sound in the winter months.
(Source: WDFW)

Spring Run
Fall Run
Sub-adult Winter Resident

Figure 4-20. Osprey along the Duwamish River. Left: Osprey delivers a starry flounder to its young in a nest on a piling near Everett Harbor. Right: Osprey nest on Duwamish River with two eggs. (Photos: Chuck Henny, USGS)

Status and Trends

Since 1993, scientists with the Contaminant Biology Program of the USGS Forest and Rangeland Ecosystem Science Center in Corvallis, Oregon, have conducted studies of nesting osprey populations in Washington and Oregon to assess contaminant exposure and impacts. Initial findings, based on studies conducted along the Columbia River (Henny et al. 2004) and Willamette River in Oregon (Henny et al. 2003), show that some contaminants biomagnify from fish to an osprey egg by a factor of up to 174-fold.

USGS scientists studied ospreys nesting on the Snohomish River delta near Everett Harbor in 2002 and the Duwamish River and Lake Washington area in 2003. The researchers evaluated contaminant concentrations, reproductive performance, foraging locations, and prey species preferences. During the 2002 investigation, scientists collected four osprey eggs from nests near Everett Harbor and in 2003 collected 11 eggs from the Duwamish Waterway/Duwamish River and vicinity (see Figure 4-20 for photograph of osprey eggs). These eggs were analyzed for legacy contaminants (e.g., PCBs. DDT, dioxins, and furans) as well as PBDEs and herbicides. The herbicide analysis resulted in the first reported detection of herbicides in osprey eggs (Chu et al. 2006). Additional analysis of 2002 osprey and cormorant egg samples from near Everett Harbor is planned in order to compare residue concentrations of PBDEs and herbicides from this site with that of osprey eggs from Everett Harbor and the Duwamish River.

ii. Harbor Seals

Harbor seals provide an integrated signal of food web contamination in Puget Sound. Feeding on a wide variety of fish and invertebrate species, they are particularly vulnerable to contamination by PBTs. While many PBTs were banned in North America following widespread environmental contamination and associated impacts on wildlife in the 1970s (e.g., PCBs and DDT), PBDEs represent an emerging PBT concern. One of the three commercial PBDE products, deca (a name that refers to its molecular structure) remains on the market, ensuring continued inputs into Puget Sound by way of wastewater, landfill leachate, incinerator ash, and nonpoint source pollution.

Studies of contaminants in harbor seal pups in the Puget Sound and Georgia Basin region were carried out by Department of Fisheries and Oceans Canada.

Status and Trends

Although regulation led to a dramatic reduction in PCBs in harbor seals from Puget Sound's Gertrude Island between the early 1970s and the present (Calambokidis et al. 2001), recent research has found an exponential increase in concentrations of PBDEs in harbor seals during the period from 1984 to 2003 (Figure 4-21) (Ross et al. 2005). While PBDE concentrations in 2003 were present at relatively low levels, (1/15th the concentration of PCBs in seals), the trend observed suggests that PBDE concentrations are doubling every four years and, by 2020, will surpass PCBs as the number-one PBT concern in Puget Sound seals.

Impacts to the Ecosystem

Harbor seal food-basket studies indicate that PBDEs are now second, after PCBs, in order of importance in Puget Sound food webs, having surpassed DDT (Cullon et al. 2005). As local sentinels of food-web contamination, harbor seals offer strong evidence that the Puget Sound ecosystem is rapidly becoming contaminated with a chemical that can be harmful to other animals, including seabirds and orcas (Ross 2006).

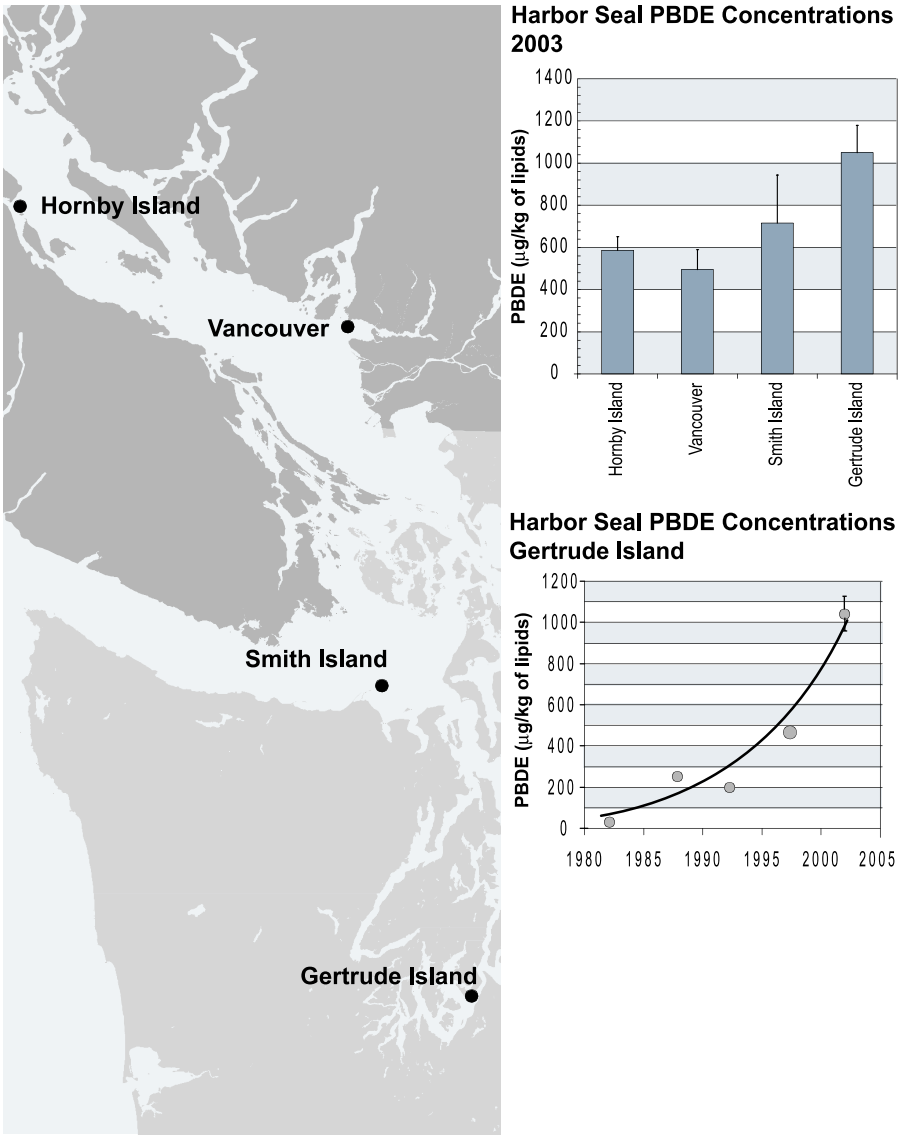


Figure 4-21. PBDEs in harbor seals from the Georgia Basin and Puget Sound. The map illustrates the locations of collection sites in 2003 near Hornby Island, Vancouver, B.C., Smith Island and Gertrude Island, and Puget Sound. Seals pups near Gertrude Island in Southern Puget Sound had nearly twice the PBDE levels of Vancouver seals (upper graph). Analysis of archived tissue (lower graph) shows the exponential increase in PBDEs since the early 1980s.
(Source: Fisheries and Oceans Canada)

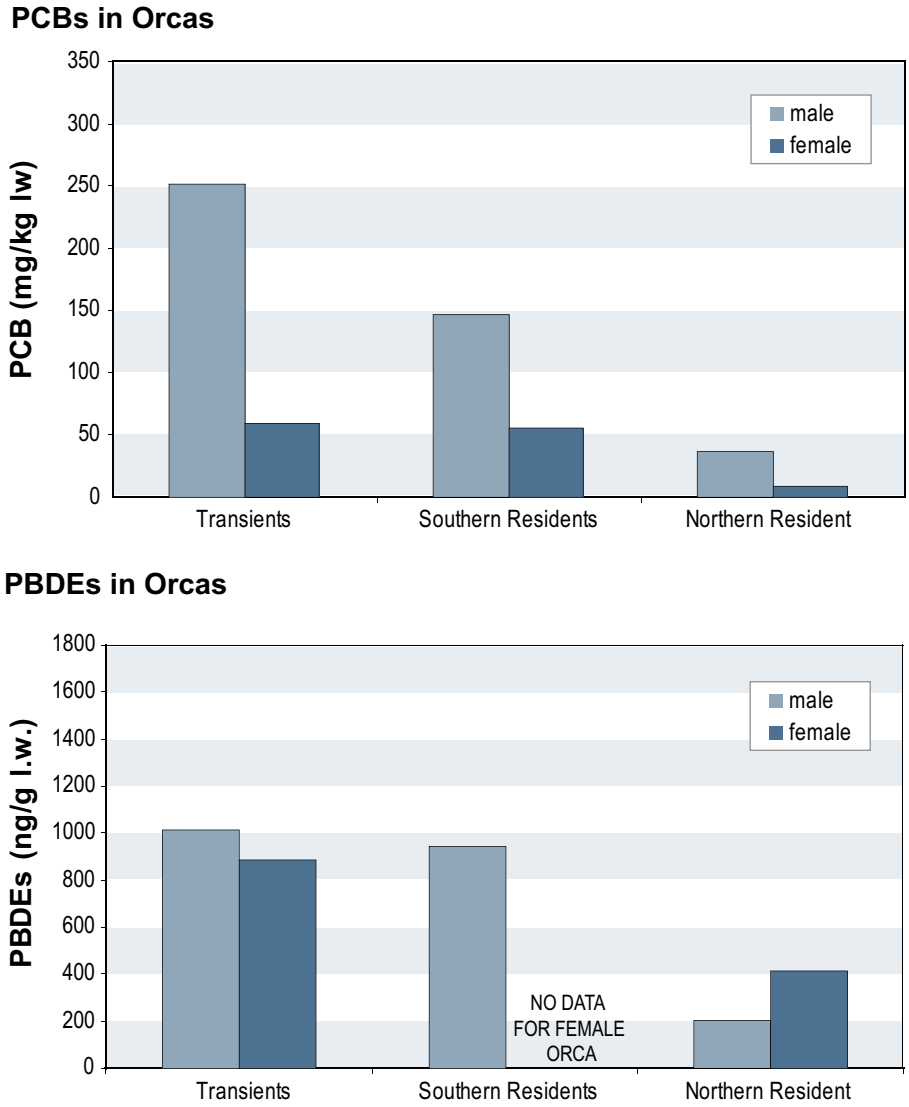
iii. PBTs in Orcas

The southern resident orcas spend a portion of the year in Puget Sound, from March to September. Their exact whereabouts the remainder of the year is not known although members of the southern residents have been sighted in Monterey, California. Throughout their range, the southern resident orcas consume primarily fish, and preferably salmon. Their position at the top of the food chain is reflected in the high level of contaminants in their tissue. Females tend to have lower PBT levels as they typically pass a considerable amount of contaminants to their offspring through fetal development and nursing.

Status and Trends

Southern resident orcas feed on salmon returning to Puget Sound and the Georgia Basin in the summer and fall and have three times the levels of PCBs than northern resident orcas that feed on salmon returning further north of the Georgia Basin region (Ross et al. 2000, Rayne et al. 2004, Ross 2006). Transient whales which occasionally visit Puget Sound have the highest PBT levels of the three populations sampled (Figure 4-22). This is consistent with the transient's

Figure 4-22. PBDEs and PCBs in transient, southern resident, and northern resident orcas from samples collected between 1993-1996. Transients occasionally visit Puget Sound and prey primarily on marine mammals, and the levels of PCBs and PBDEs are highest in the transient population. Southern residents have 3-4 times the levels of these compounds compared to the northern residents, which feed further north of the Puget Sound Georgia Basin region. Both southern and northern resident orcas feed mainly on salmon. (Source: Department of Fisheries and Oceans Canada)



preference for marine mammals which are higher trophic species. Even still, both northern and southern resident orcas are more contaminated with PBTs than other north Pacific resident orca populations (Ylitalo et al. 2001; Herman et al. 2005). PBDE levels are increasing exponentially in these animals and are expected to surpass PCB levels by 2020.

iv. PBTs and Humans; analysis of breast milk from several countries

The highest levels of PBDEs in human tissues have been found in the U.S. and Canada, nations which produce and purchase the greatest amount of products with PBDE flame retardants. Levels of PBDEs in human tissues (fat, blood, and breast milk) in the U.S. are 10 to 40 times higher than reported for Europe and Japan and appear to be increasing (Figure 4-23). Studies conducted on animals show that prenatal exposure to PBDEs can impact the brain, affecting behavior and learning after birth and into adulthood. Animal studies have also shown that PBDEs can affect the thyroid and liver. Currently, the levels of PBDEs that cause these effects in animals are higher than the levels of PBDEs that most people encounter. However, if trends continue, similar effects could be seen in humans.

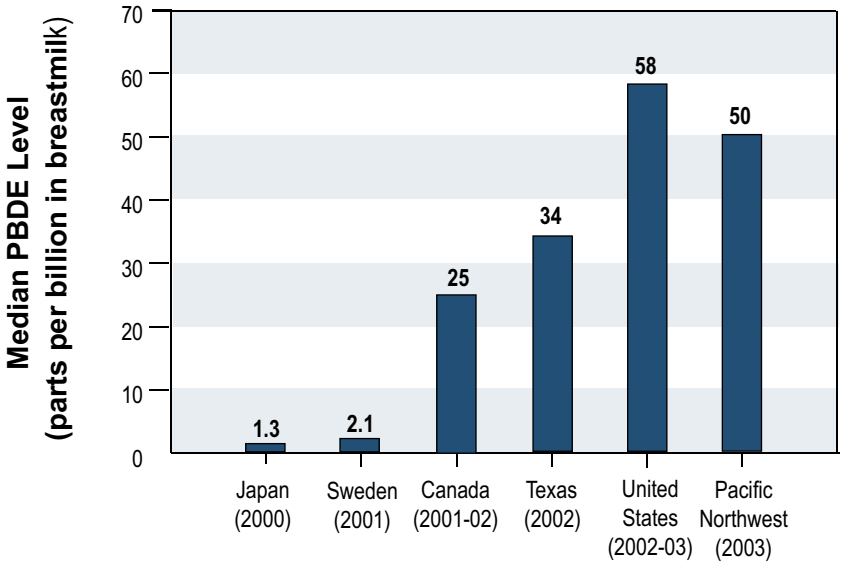


Figure 4-23. PBDEs in breast milk from Japan, Sweden, Canada and the U.S. in 2003. Numbers in the figure are the median ppb measurements from regions sampled. Overall, the highest levels of PBDEs were found in the U.S. The Pacific Northwest had the highest median levels in the country at 50 ppb. Sweden, with a median level of 2.1, banned PBDEs in the early 1990s out of health concerns. (Source: Sightline Institute)

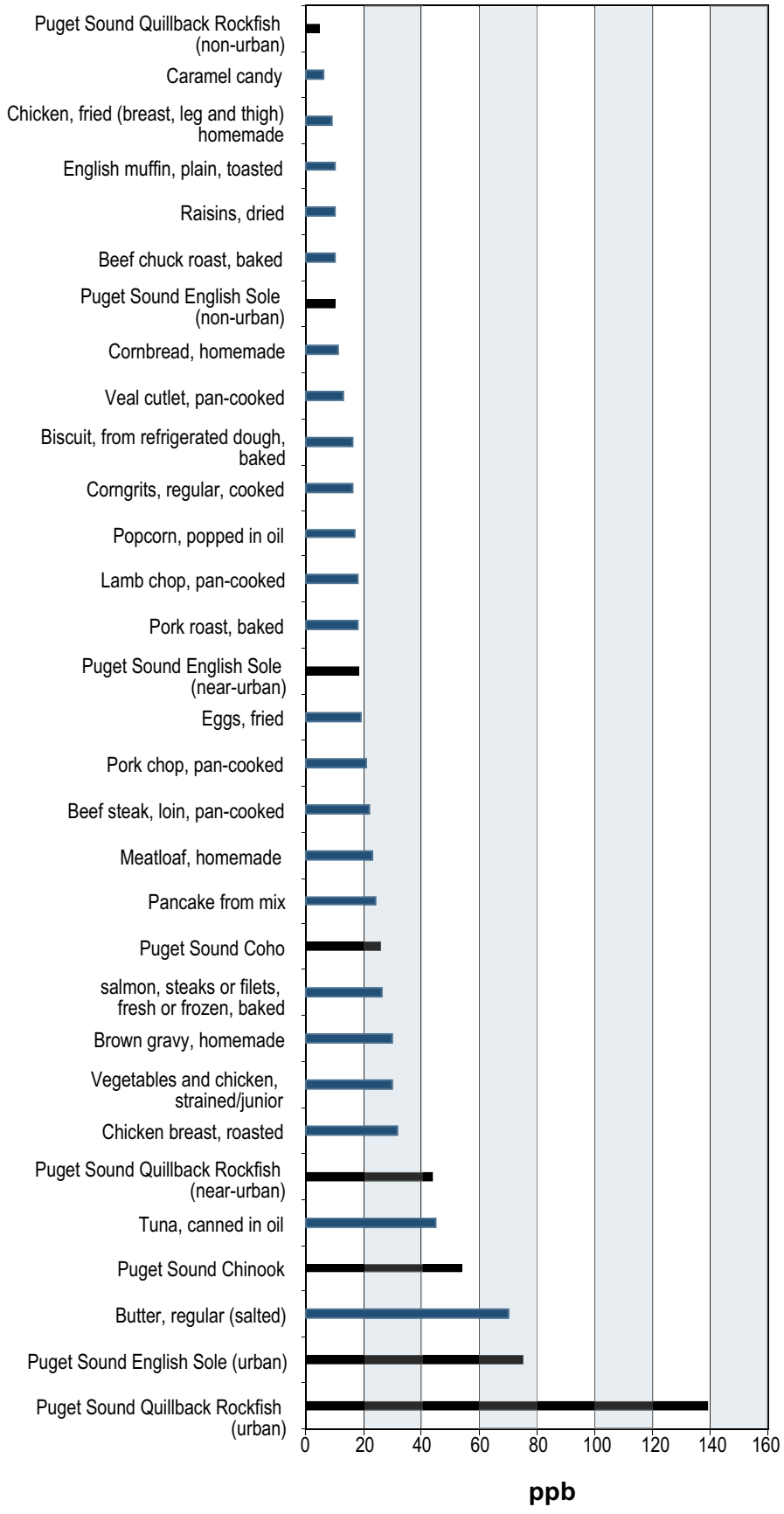
Human Health Consequences

Recent studies have attempted to quantify and compare the risks of eating contaminated fish with the benefits associated with their ingestion. Further work is expected on this subject, as more reports on fish contaminant levels and human health become available. Limited data show a link between fish consumption and a decrease in development of some cancers. Eating fish has also been associated with impacts on brain function, including protection against cognitive decline.

Studies have analyzed items from a wide range of food groups for PCB content, to determine the relative risk from consuming shellfish and fish from Puget Sound (Figure 4-24). These results are limited because, for many foods listed, only one or a few samples were analyzed.

DOH provides information to the public on fish and shellfish consumption advisories issued for specific water bodies in Washington, warning of chemical contamination. Most Puget Sound advisories address consumption of fish from urban areas (Dyes Inlet, Eagle Harbor, Sinclair Inlet, the Duwamish River, Commencement Bay, and Budd Inlet). WDFW found that PCBs and mercury in bottom fish are higher in urban bays, lower near urban sites, and approach background levels in most non-urban areas.

Figure 4-24. PCBs in common foods. Samples include fish from Puget Sound and results are reported in micrograms per kilogram sampled. Commercial foods were sampled as part of the U.S. Food and Drug Administration's total diet study and market-basket survey. In most cases, data are limited by small sample sizes.
(Source: DOH)



4. PAHs in Puget Sound

PAHs are toxic and carcinogenic chemicals formed by the incomplete burning of organic matter, including petroleum, oil, coal, and wood (Newman and Unger 2003). Vehicles release PAHs into the atmosphere in exhaust emissions and deposit them on the ground through oil and gasoline leaks. Such leaks can be transported to streams, rivers, and estuaries in stormwater runoff. Long-term aquatic sediment core studies in Puget Sound and nationwide found that PAH levels peaked in the mid-1940s through 1960s. Decreases were seen in the 1970s and 1980s, followed by more recent increases. It is believed that the early declines in PAH concentrations can be attributed to the switch from coal to oil and natural gas for home heating, improvements in industrial emissions controls, and increases in the efficiency of power plants. More recent increases have been linked to increasing urban sprawl and vehicle traffic in urban and suburban areas (Lefkovitz et al. 1997; Van Metre et al. 2000, in press). Recent studies by USGS have also measured high PAH concentrations in stormwater runoff from parking lots sealed with coal-tar-based asphalt sealants (Mahler et al. 2005).

a. PAHs in sediments

Ecology sampled sediments at 10 fixed stations each spring from 1989 through 2000 (Figure 4-25). Stations were chosen from a variety of habitats and geographic locations in Puget Sound. Sediments from each station were analyzed for particle size, organic carbon content, and the presence of more than 120 chemical contaminants, as well as the types and abundances of sediment-dwelling organisms.

Chemical contaminants in the sediments were measured yearly from 1989 through 1996 and again in 2000. The contaminants examined included priority pollutant and ancillary metals, as well as organic compounds, such as PAHs, chlorinated pesticides, and PCBs. Changes in sediment condition over this time included increases in levels of PAHs at some stations.

Status and Trends

Concentrations of most PAH compounds did not change significantly over the study period; however, most of those that *did* change increased in concentration. There was a significant overall increase in benzofluoranthenes and increases in PAHs at stations in Bellingham Bay, Port Gardner, and Anderson Island. In contrast, there was a significant decrease in PAHs at the Point Pully station (Table 4-4).

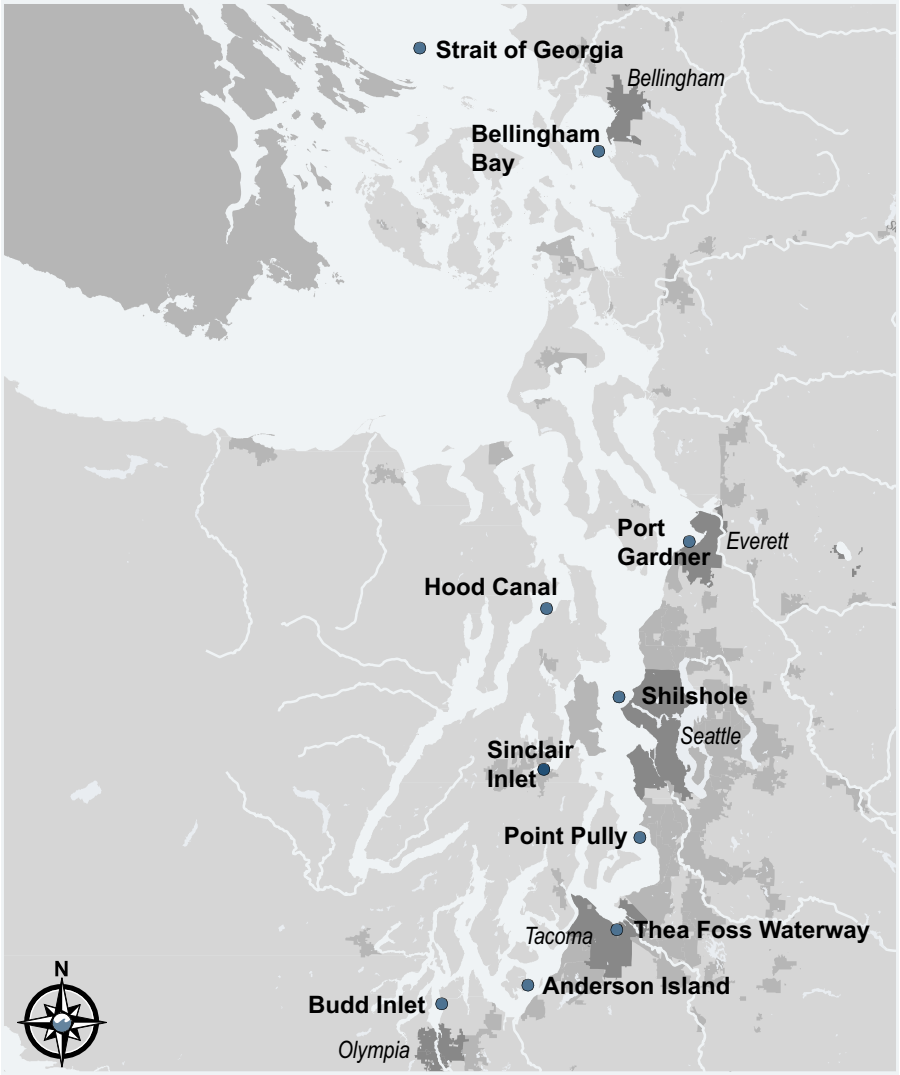
Impacts to the Ecosystem

The PSAMP sediment monitoring program provides a vital record of sediment conditions in Puget Sound and gives insight into the effects of both natural and human-driven stressors on the estuary. The fixed sentinel stations monitored in this program can raise red flags, highlighting important environmental changes affecting Puget Sound.

b. Biota Exposure to PAHs

Exposure studies allow researchers to determine if an organism has been exposed to a compound or contaminant in the environment. The concentration of a known compound is typically measured from muscle tissue, lipids, or the whole organism. Exposure studies do not indicate whether the organism's health has been affected.

Figure 4-25. Location of PSAMP's 10 long-term monitoring sites in Puget Sound.
(Source: Ecology)



i. PAHs in Mussels

Since 1986, NOAA's National Status and Trends Mussel Watch Program has been monitoring contaminants in mussels from Puget Sound, the Strait of Juan de Fuca, and the Pacific Ocean coast of Washington. The Mussel Watch Program analyzes nearly 150 separate chemicals in whole soft tissue from composites of two mussel species collected at each of 12 sites in Puget Sound and four sites on the Pacific Ocean coast. Samples were collected annually until 1994 and every other year since then. The Mussel Watch program data can help track changes in contaminants, including PBTs, PAHs, and metals that accumulate in mussel tissue, providing a gauge of water quality near monitoring sites.

Status and Trends

Median concentrations of PAHs[3] in mussel tissue from Puget Sound sites range from 200 to 4,000 parts ppb dry weight (dw). These concentrations range from one to more than 10 times the national median value of 220 ppb dw.

The pattern of PAH concentrations in mussel tissue in Puget Sound has varied over the last two decades. PAH levels declined in the mid-1980s, then increased

[3]TPAHs in ppb dry weight is the sum of 18 parent compounds of dry tissue weight.

PAH Compounds	Station Change 1989-1996 vs. 2000											Station Trend 1989-2000										
	Strait of Georgia	Bellingham Bay	Hood Canal	Port Gardner	Port Gardner[oc]	Shilshole	Sinclair Inlet	Point Pully	Thea Foss WW	Anderson Island	Budd Inlet	Strait of Georgia	Bellingham Bay	Hood Canal	Port Gardner	Port Gardner[oc]	Shilshole	Sinclair Inlet	Point Pully	Thea Foss WW	Anderson Island	Budd Inlet
LPAHs																						
2-Methylnaphthalene	↑	↑		--	--	--	--	--	↑			↑	--	--	--	--	--	--	--	--	--	--
Acenaphthene				--	--		--		--						--	--	--	--	--	--		--
Acenaphthylene				--	--		↑	--	--					--	--	--	--	--	--	--	--	--
Anthracene	--	--		--	--	--	--	↓	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Fluorene	--	--		--	↑	--	--	--	--		--	--	--		--	↑	--	↑	--	--	--	--
Naphthalene	--	↑		--	--	--	--	--	↑	--	--	↑	--	--	--	↑	--	--	--	--	--	--
Phenanthrene	--	--	--	--	--	--	--	↓	--	↑	--	--	--	--	--	--	--	--	--	--	--	--
Retene	--	--		--	--	--	--	--	↑	--	--	--	--	--	--	--	--	--	--	↑	--	--
Total LPAH	↑	↑	--	--	--	--	--	--	--	↑	↑	↑↑	--	--	--	↑	--	--	--	--	--	--
HPAHs																						
Benzo(a)anthracene	--	↑		--	↑	--	--	--	--	↑	--	--	--	--	--	--	--	--	--	--	--	--
Benzo(a)pyrene	--	--		--	--	--	--	--	--	↑	↑	--	--	--	--	--	--	--	--	--	--	--
Total Benzofluoranthenes	↑	↑	--	--	↑	↑	--	--	--	↑	↑	--	--	--	--	--	--	--	--	--	--	--
Benzo(g,h,i)perylene	--			--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Chrysene	--	--		--	--	--	--	↓	--	↑	--	--	--	--	--	--	--	--	--	--	--	--
Dibenzo(a,h) anthracene						--	--	--	--						--	--	--	--	--	↓		
Fluoranthene	--	--	--	--	--	--	--	↓	--	↑	--	--	--	--	--	--	--	--	--	--	--	--
Indeno(1,2,3-c,d)pyrene	--	--		↑	↑	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Perylene	↑	↑		--	--	↑	--	--	--	--	--	--	--		--	--	--	--	--	--	--	--
Pyrene	--	↑	--	--	↑	--	--	↓	--	↑	↑	--	--	--	--	--	--	--	--	--	--	--
Total HPAH	--	↑	--	--	--	--	--	--	--	↑	--	↑	--	--	--	--	--	--	--	--	--	--
Total PAH	--	↑	--	--	--	--	--	--	--	↑	↑	↑	--	--	--	--	--	--	--	--	--	--

Table 4-4. Changes and trends in both low- and high-molecular-weight PAH compound concentrations in sediments collected from 1989 through 2000. While most PAH compounds did not change significantly during the study period, those that did change generally increased in concentration. Blank boxes indicate no change. Double arrows at the Strait of Georgia indicate greater change, compared to the other sites.
(Source: Ecology)

↑,↓..... increase, decrease (α=0.05)

↑↑, ↓↓ ... increase, decrease (α = 0.01)

-- ..... no change

blank..... insufficient data

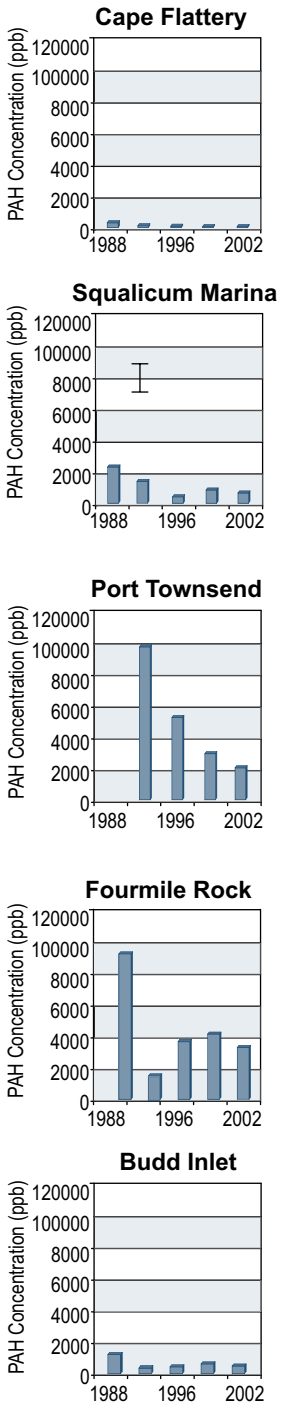
Shaded results indicate changes for all stations combined for a single compound or for all compounds combined for a single station, at α=0.05 (dark) or α=0.10 light.

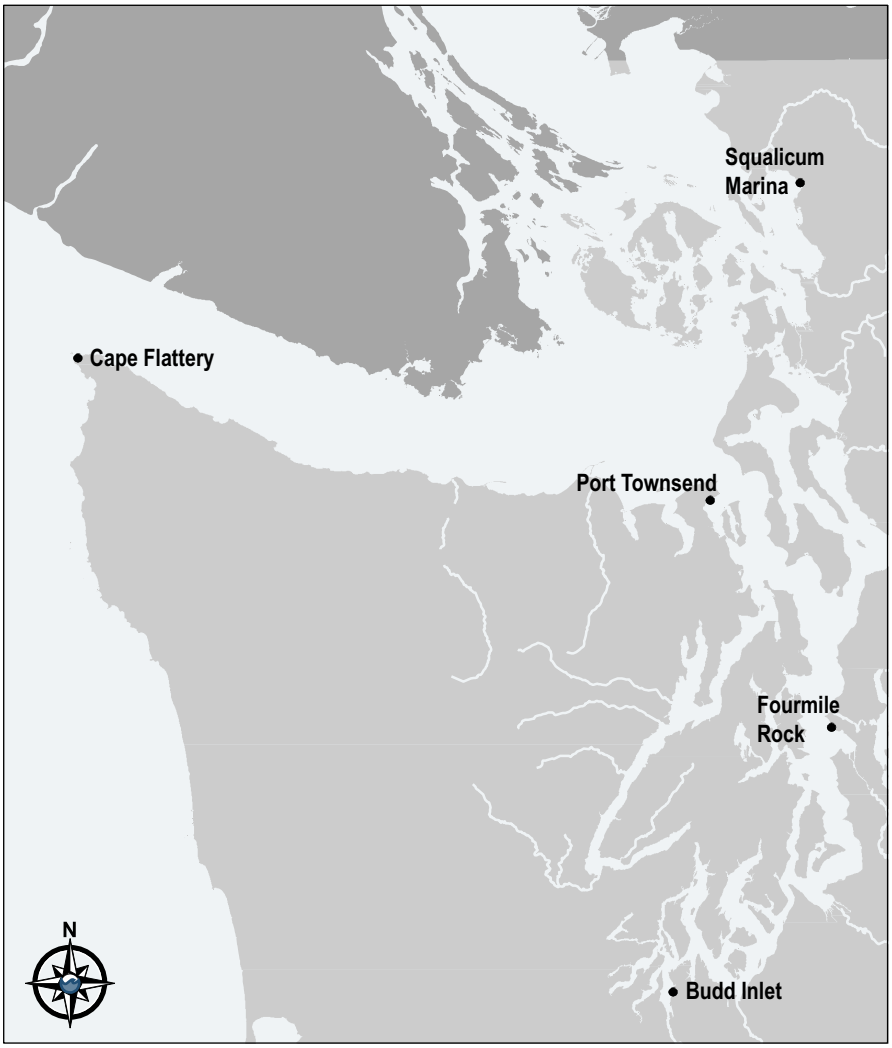
[oc] = data normalized to organic carbon.

slightly in the mid-1990s. Overall, PAHs in mussel tissue appears to be in decline. Everett Harbor had the highest concentration in 2000, at 31,750 ppb (off the scale in Figure 4-26); however, concentrations at that site were not consistently that high over the years. A second site with high mussel PAH concentrations was Port Townsend. The Port Townsend Mussel Watch site is located at the south side of the marina and sampling did not begin here until 1990. Nevertheless, this site had the highest concentrations in 1990—8,000 to 12,000 ppb dw. Those concentrations fell to about 2,500 ppb dry weight by 2002.

Elsewhere throughout Puget Sound and the Strait of Juan de Fuca, PAH concentrations were about 1,000 to 2,000 ppb dry weight in the mid-1980s. Concentrations had declined to below 1,000 ppb dry weight by 2002; however, as with PCBs, there was an increase in PAH concentration in the mid-1990s, most notably at Fourmile Rock, Everett Harbor and Duwamish Head. Generally, during

Figure 4-26. Total PAHs in Puget Sound mussel tissue. PAH levels have generally declined, with highest concentrations in Everett and Fourmile Rock near Seattle. Median tPAH levels from all national sites sampled 2002/2003 was 220 ppb dry weight). (Source: NOAA)





the past two decades, sites with the lowest TPAH concentrations (<200 ppb dry weight but as low as 10 to 20 ppb dry weight) were along the Pacific Ocean coast and in northern Puget Sound.

ii. Crabs and Fish

PSAMP scientists have previously described PAH-exposure of English sole, Pacific herring, rockfish, and Dungeness crab in the *2000* and *2002 Puget Sound Updates*. Combining results for all species for multiple years between 1998 and 2005[4] indicates that the greatest exposure to PAHs generally occurs in urbanized embayments or shorelines (Figure 4-27). Concentration of PAHs was six times greater in Dungeness crab from one urban location (Thea Foss Waterway) than two non-urban locations (Vendovi Island and the Cherry Point shoreline). The bottom-dwelling fish species (English sole and rockfish) also exhibited three to four times greater exposure (as measured by the PAH metabolic biliary phenanthrene) to PAHs in the Thea Foss Waterway, Elliott Bay, Sinclair Inlet, and

[4]Sampling occurred in the following years for the following species:
Dungeness crabs - 2001
Pacific herring - 1999, 2000, 2001, 2002, and 2004
English sole - 1998, 1999, 2000, 2001, 2003, 2005
Rockfish - 1998, 1999, 2002 and 2001 however, not all rockfish species were sampled in all years.

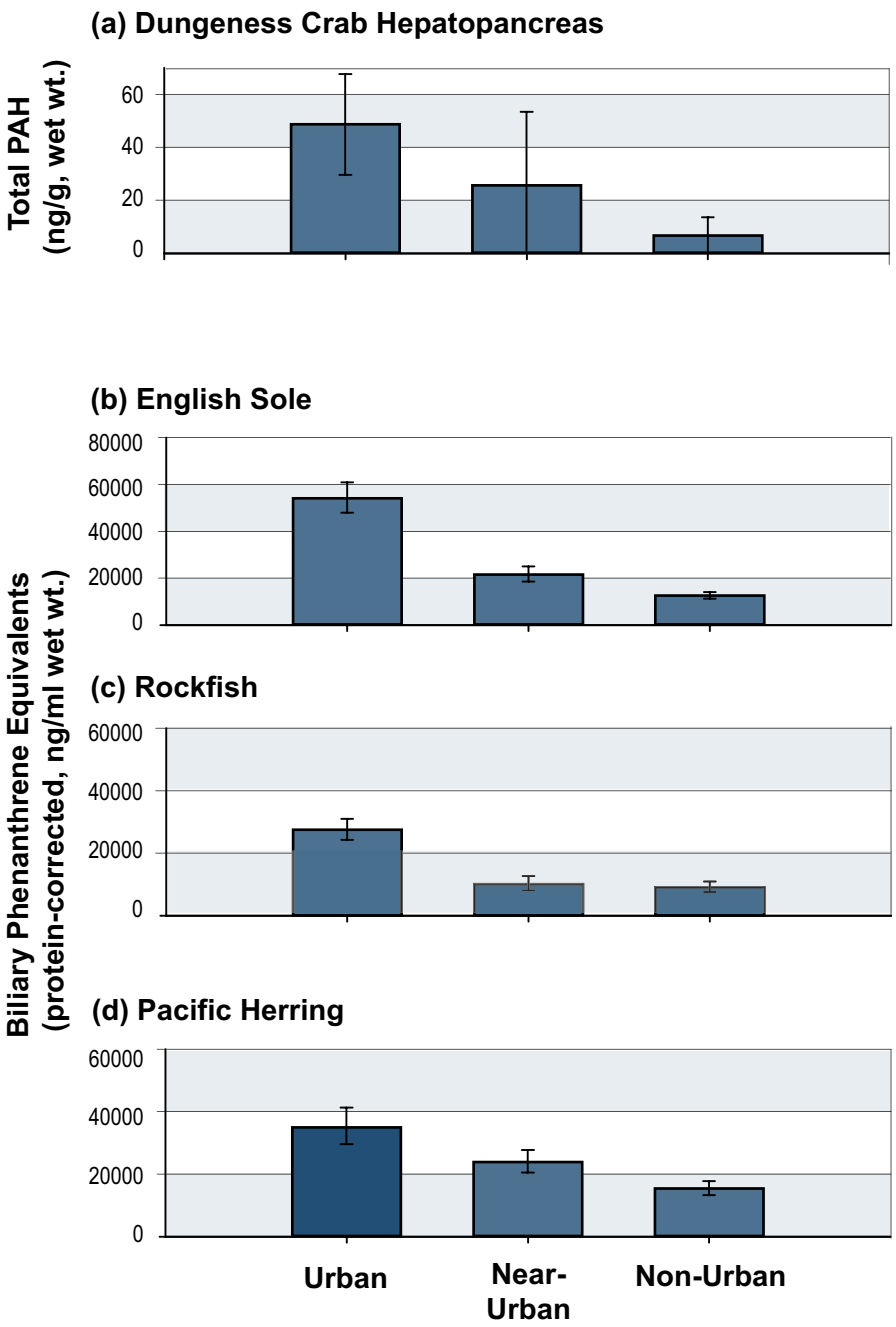


Figure 4-27. Exposure to PAHs in urban embayments. Exposure to PAHs (as indicated by the presence of the biomarker biliary phenanthrene in the bile) is greatest in urbanized embayments or shorelines for bottom-dwelling species (Dungeness crab, English sole, and rockfish), or in urbanized basins (Central Puget Sound) for a pelagic species such as Pacific herring. PAHs were measured directly in crab tissue and as PAH-metabolites in fish bile. (Source: WDFW)

Port Gardner, than at non-urban sites. For the pelagic species (Pacific herring), PAH exposure of an urban population (Port Orchard/Madison stock in central Puget Sound) was more than twice that of a non-urban herring (Semiahmoo stock in the Strait of Georgia).

c. Effects of PAHs on Biota

i. English Sole

In previous editions of the *Puget Sound Update*, PSAMP, WDFW and NOAA scientists have described the connection between PAH exposure and its effects, primarily liver disease and reproductive impairment in English sole. PSAMP scientists have been monitoring both exposure and effects in English sole as an indicator of the health of bottom-dwelling fishes in Puget Sound.

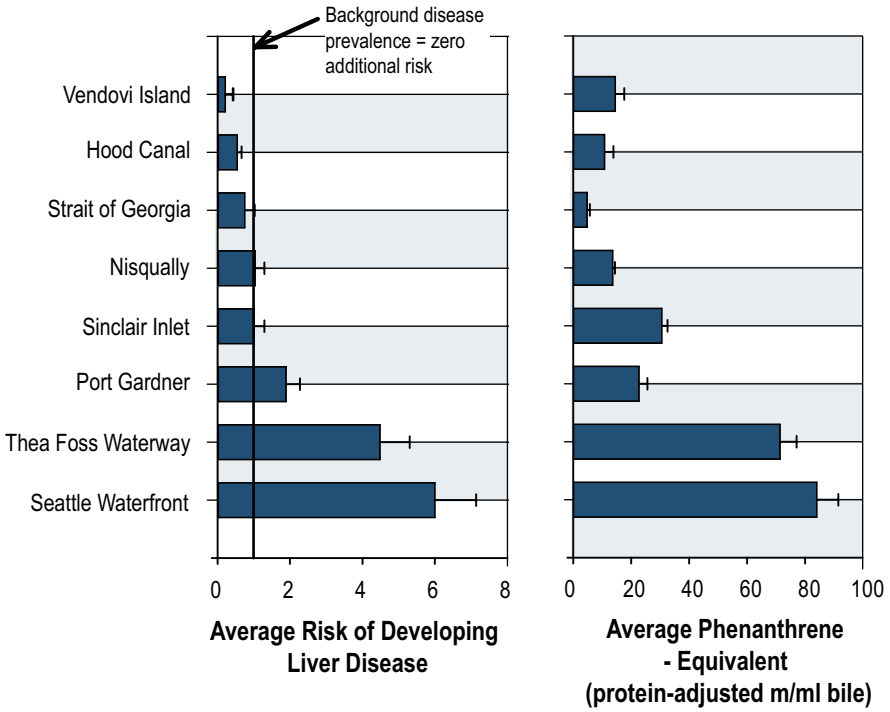


Figure 4-28. Risk of developing liver disease and exposure to PAHs in English sole. Study was conducted at eight long-term monitoring stations. English sole from two urban locations exhibited a high risk of developing liver disease. The Seattle waterfront had six times the risk and the Thea Foss Waterway had four times the risk relative to uncontaminated baseline stations. Both urban locations had relatively high exposures to PAHs as reflected in the measured phenathrene, a byproduct of PAH metabolism (>60 ug/ml). Port Gardner exhibited a relatively small risk (two times the baseline) and relatively low PAH-exposure, while all other locations exhibited risk equivalent to or less than baseline and low PAH exposure. Risks were averaged over a 17-year period, and PAH exposure was averaged over an eight-year period.
(Source: WDFW)

Status and Trends

Observations for PAH-related liver disease in English sole for a period spanning 17 years, from 1989 through 2005, from six locations, and for slightly shorter periods from two additional locations are summarized in this update. PAH exposures have been measured since 1998 in sole from these eight locations, which represent a range of conditions from highly urbanized or industrialized to more remote uncontaminated locations.

Of the eight long-term English sole stations that PSAMP scientists monitor, both PAH exposure and the risk[5] of developing liver disease have been consistently greatest in individuals taken from two urban locations—the Seattle waterfront and Thea Foss Waterway (Figure 4-28).

English sole from another urban location, Sinclair Inlet, have consistently exhibited low prevalences of liver disease and PAH exposure, even though these fish exhibit high concentrations of PCBs and other toxics characteristic of Puget Sound's urban habitats (Figure 4-27). Sole from Port Gardner exhibited disease risk of two times the baseline, while sole from five other sites exhibited disease risk equivalent to or less than baseline. PAH exposure was low in all six locations, relative to that of the Seattle waterfront and Thea Foss Waterway.

High prevalence of liver disease in English sole has also been reported in Eagle Harbor, where PAH source control and cleanup have occurred, and where NOAA scientists have documented the recovery of English sole health over the past decade. (See Focus Study: Eagle Harbor Capping on page 172.)

[5] Risk is calculated using logistic regression to compare the prevalence of liver disease (i.e., the number of fish exhibiting any disease, as a percentage of the total) at a location, to prevalence of disease in English sole from 19 uncontaminated background, or baseline locations. The risk at baseline locations is 1.0, and the risk at other locations is expressed as the increased or decreased predicted likelihood of developing disease relative to that baseline (e.g., 10x). The analysis accounts for the natural increased likelihood that English sole will develop liver disease as they age, even in the absence of PAHs and other toxicopathic compounds.

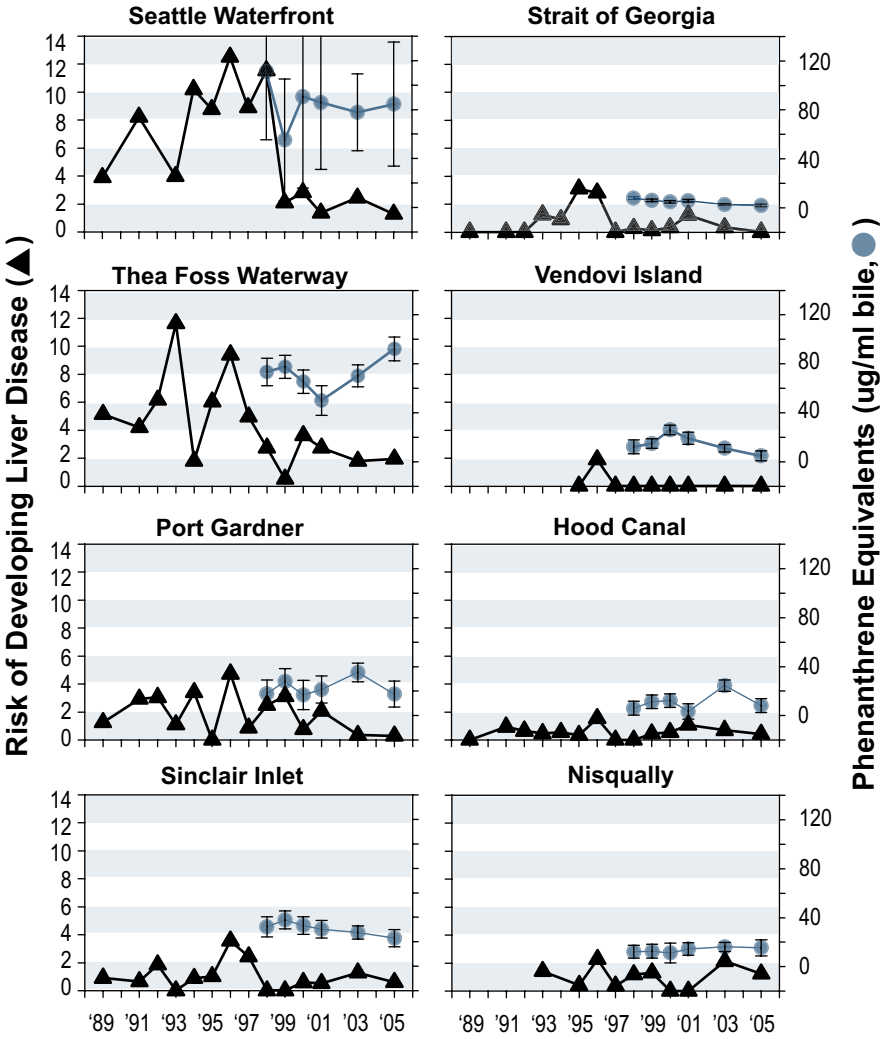
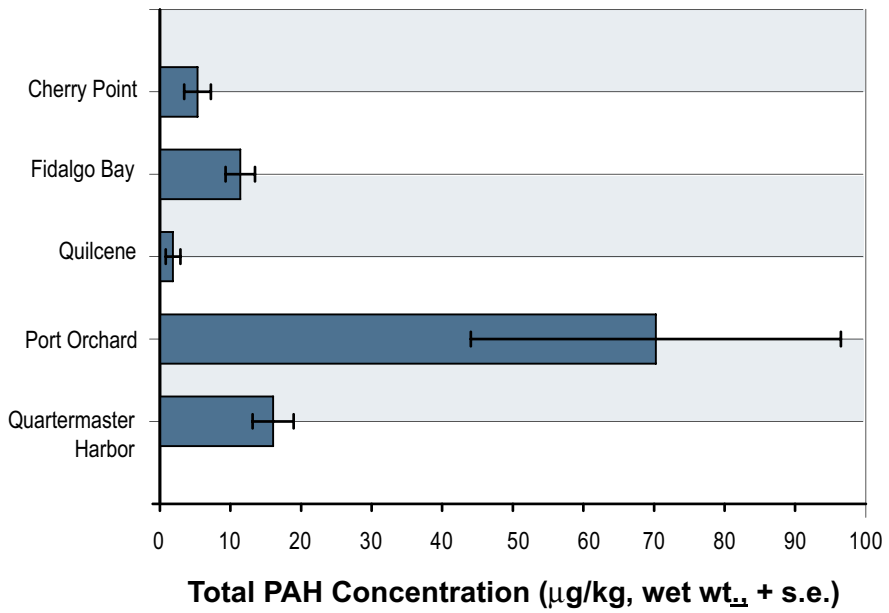


Figure 4-29. Trends in liver disease risk and exposure to PAHs in English sole. Long-term trends in risk of developing liver disease (triangles), 1989–2005, and in exposure to PAHs (circles), as measured by the biomarker biliary phenanthrene equivalents in the bile, 1997-2005. Both the Seattle waterfront and Thea Foss Waterway had higher risk of developing liver disease, compared to other locations in Puget Sound. Analysis is based on six composite samples per year. (Source: WDFW)

In the 17 years that liver disease has been monitored in Puget Sound's English sole, decreasing trends have been observed at the Seattle waterfront and Thea Foss Waterway—the two urban locations that have historically exhibited the greatest overall disease prevalences (Figure 4-29). Risk at the Seattle waterfront increased over the first 10 years of monitoring (1989-1998), with an average of 8.5 times the baseline in that period and a peak in 1996 of 12.5 times the baseline. Risk at the Seattle waterfront declined sharply from 1999 through 2005, with an average of two times the baseline risk during this period. A similar decline occurred at the Thea Foss Waterway, with average and peak risks of six times the baseline and 12 times the baseline prior to 1998, and an average of two times the baseline from 1998 through 2005.

It is difficult to correlate time trends in PAH exposure with liver disease, because PSAMP's monitoring of PAH exposure in English sole bile began in 1998, missing the period of high liver disease prevalence in the early to mid-1990s. It is possible that slowing declines in liver disease at the Seattle waterfront and Thea Foss Waterway result from localized sediment cleanup measures and source controls; however, this is not clear from PAH exposure measures. This could result from the time lag that occurs between source removal (exposure) and recovery from liver disease. In addition, English sole probably forage over a larger range than is encompassed by individual recovery (e.g., sediment capping) efforts. Hence,

Figure 4-30. PAHs in herring eggs. Total PAHs were greatest in spawned herring eggs sampled from the Port Orchard/Madison spawning grounds. (Source: WDFW)



during sediment years when sediment cleanup is occurring, variability in PAH exposure may increase, while overall exposure decreases. This appears to be the situation at the Seattle waterfront.

ii. Herring Eggs

PSAMP scientists have assessed PAH exposure in developing embryos of Pacific herring. This otherwise pelagic species spawns adhesive eggs on intertidal and shallow subtidal structures, especially on algae and seagrasses. Shoreline habitats are particularly susceptible to large and small oil spills and to PAH inputs from shoreline sources, such as runoff and river inputs. PSAMP scientists sampled developing herring embryos from five locations throughout Puget Sound in 1999, 2001, and 2002, representing five of the Sound's major spawning stocks.

Status and Trends

Embryos of Port Orchard/Madison herring in their typical spawning habitats (along the northern and western shore of Bainbridge Island and northern Port Orchard) exhibited five to 15 times the concentrations of Total PAH (TPAH) than those from four other locations throughout the Sound, including two near oil refineries or transfer stations (Fidalgo Bay and the Cherry Point shoreline) (Figure 4-30). Port Orchard/Madison embryos also exhibited a high variability in TPAH concentrations, which was related to differences in specific collection locations and developmental stages of the embryos (Figure 4-31). One Port Orchard/Madison spawning location, Point Bolin, exhibited low TPAH, while another, Hidden Cove, exhibited relatively high TPAH. In addition, TPAH increased in Hidden Cove embryos as they aged: samples of 10-day-old embryos had up to four times the TPAH concentrations of three-day-old embryo from the same location, collected a week prior. TPAH concentrations observed in embryos from Hidden Cove were high enough to suspect toxicological impacts. In one experiment (Carls et al. 1999), developing herring embryos were exposed to aqueous PAHs at concentrations of TPAH between 22 and 108 ng/g (embryo tissue, wet weight) and became malformed or died. Total PAH concentrations in four of five Hidden Cove samples were well above the 22 ppb lower threshold, and samples from Point Bolin, as well as all others from the four other spawning stocks (Figure 4-30), were well below this threshold.

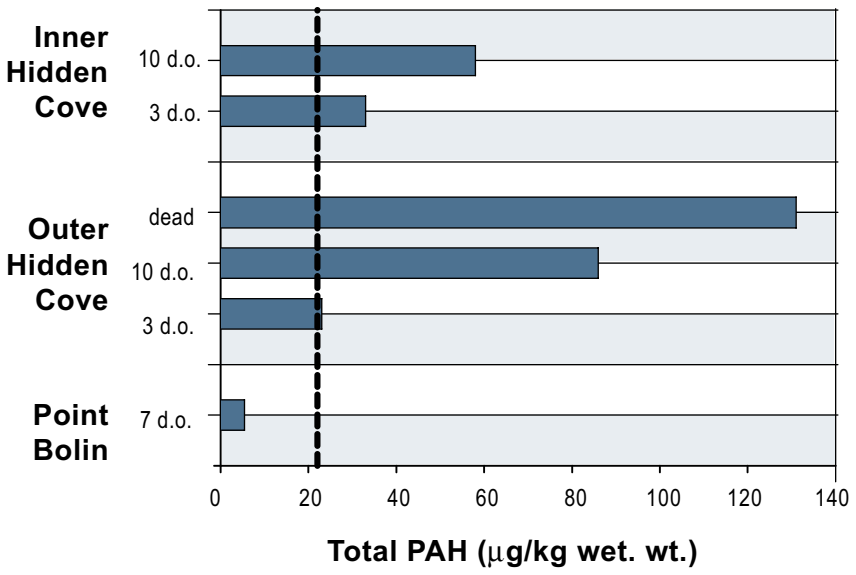


Figure 4-31. Variability in herring eggs from Port Orchard. High variability in Total PAH (TPAH) from Port Orchard/Madison spawned eggs is related to specific spawning location and age of embryos. High TPAH concentrations were observed in embryos taken from two locations within Hidden Cove, but not from Point Bolin, 5.5 miles (9 km) away. In addition, 10-day-old embryos had greater TPAHs than did those recently spawned (three-days-old), indicating that PAHs accumulated in embryos as they developed. Greatest TPAH levels were observed in dead embryos (probably older than 10 days). The vertical dashed line indicates a total PAH concentration, or threshold, beyond which exposed embryos begin to die.
(Source: WDFW)

d. PAHs and Creosote

Creosote is an effective tar-based wood preservative that has been in use for over 100 years, to preserve railroad ties, telephone and power poles, wharf and pier pilings, beach access stairways, railings, and other landscaping features. There are a variety of commercial creosote formulas containing as many as 300 different chemicals, a large portion of which are PAHs, known to be toxic. PAHs are associated with disease and other health problems in English sole and other marine fish. Researchers at NOAA's Northwest Fisheries Science Center state that failure to achieve acceptable minimum levels of PAHs in sediments will result in impaired productivity of fish stocks (Johnson, Collier, and Stein 2002).

Creosote wood debris has been observed on beaches throughout Puget Sound. Whether freshly washed up or buried in the intertidal zone for decades, this debris can leach creosote continually. Researchers with the Skagit Marine Resources Committee found that even 60-year-old pilings are leaching creosote daily into the marine environment, and a scratch with a finger nail can bring fresh chemicals to these pilings' surfaces (Dinnel 2005). One cubic foot of creosote-treated wood contains at least 20 pounds (9 kg) of creosote.

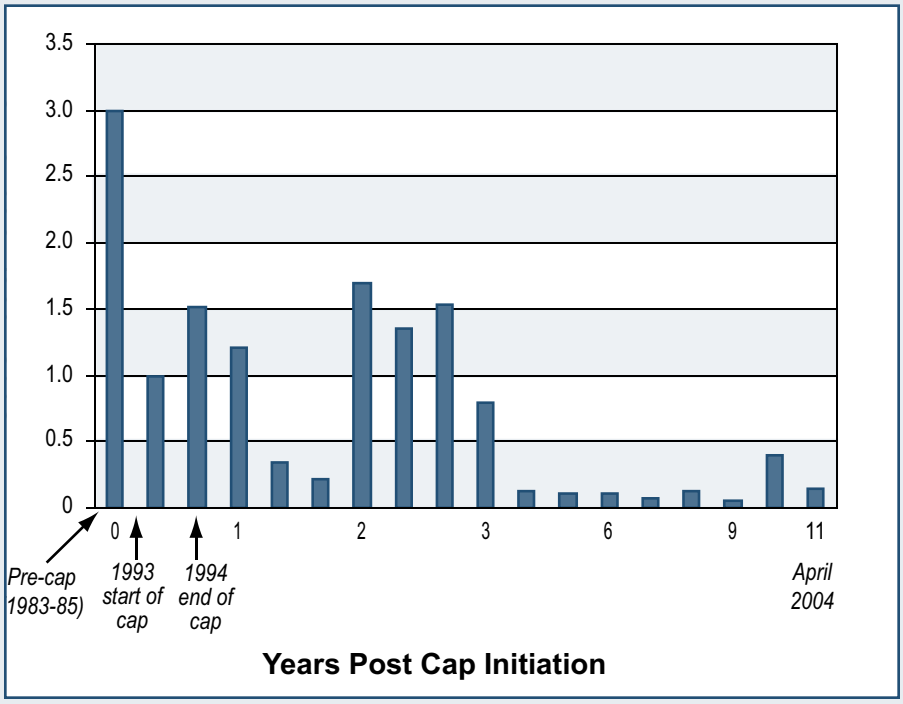
Status and Trends

NWSC and DNR have identified creosote removal as a high priority and are actively surveying and removing creosote debris from beaches. Marine Resources Committees in Whatcom and Skagit counties and the Padilla Bay National Estuarine Research Reserve organized a creosote removal program between 2002 and 2005. The goals of the program are to inventory and remove creosote debris on Northern Puget Sound and the Strait of Juan de Fuca beaches, better understand the rate of deposition of creosote debris on the beaches, and identify sources of creosote contamination. To date, the program has resulted in the removal of 275 tons (124 kg) of debris from 112 miles (180 km) of shoreline in Whatcom, Skagit, and Island counties and in Padilla Bay (Table 4-5, Page 174). Large removal projects at Dungeness Spit National Wildlife Refuge, Lake Hancock, Jetty Island, and San Juan Historical Park are scheduled to occur in late 2006.

FOCUS STUDY

Restoration of English Sole Health Following Capping of Contaminated Sediments in Eagle Harbor

Figure 4-32. Risk of liver lesions in English sole in Eagle Harbor, 1983-2004. Risk of lesion occurrence defined as 1.0 at start of capping. Relative risks at other sampling timepoints determined by stepwise logistic regression, after accounting for fish age. (Source: NOAA)



Eagle Harbor on Bainbridge Island was designated as a Superfund site by the EPA in 1987 because of high sediment concentrations of PAHs released chronically from a nearby creosoting facility. These high levels of PAHs were associated with adverse biological effects in resident fish species. Earlier studies (1983-1986) by NOAA scientists on English sole from this site showed high incidence (up to 80 percent) of liver lesions, including tumors in resident fish.

Scientists have demonstrated in multiple field studies that these lesions are strongly and consistently associated with PAH exposure, and have also shown that liver lesions can be induced in sole by injections of a PAH-rich fraction extracted from Eagle Harbor sediment. Further studies from 1986 through 1988 incorporated biochemical biomarkers of PAH exposure and effects. Prior to site remediation, liver lesion occurrence and biomarker values in English sole from Eagle Harbor were among the highest in Puget Sound.

As part of a combined EPA/Army Corps of Engineers effort, a cap of clean sediment was placed over the most contaminated portions of Eagle Harbor in 1993 and 1994 in an attempt to immobilize PAH-contaminated sediments. NOAA Fisheries scientists found that liver lesion prevalences and biomarker values just before capping were somewhat reduced, compared to historical data, consistent with the closure of a creosoting facility and implementation of shore-based source controls.

Fish were collected immediately after capping and at regular intervals up to about two years after capping. Scientists found that toxicopathic liver lesion risk (a

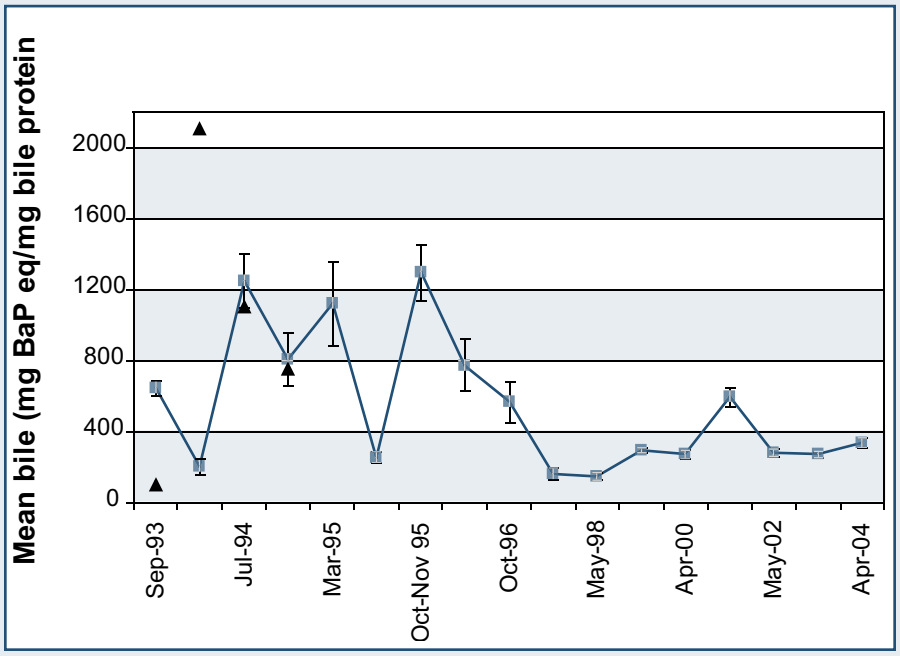


Figure 4-33. Biliary FACs (a biomarker for PAH exposure) in English sole from Eagle Harbor. Biliary FACs have declined since the late 1990s following capping.
(Source: NOAA)

Mean Bile FACs, BaP, PC
Reference sites

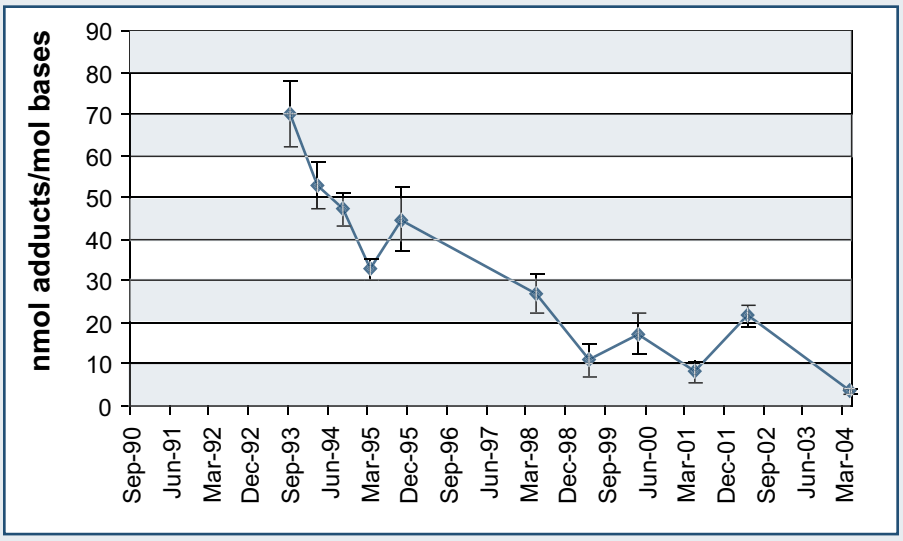


Figure 4-34. DNA adducts (a biomarker for PAH exposure) in English sole from Eagle Harbor. The prevalence of this biomarker has declined in English sole since capping took place in 1993.
(Source: NOAA)

calculated parameter that is based on lesion prevalence and fish age), and levels of two biomarkers—a metabolite of PAH that binds to the DNA found in liver cells (hepatic DNA adducts) and a metabolite of PAHs found in bile (biliary FACs)—were highly variable, relative to values prior to cap placement. Over the entire monitoring period there was an overall decrease in risk for hepatic lesions in English sole (Figure 4-32). There was also a decline in the two biomarkers; biliary FACs (Figure 4-33); and hepatic DNA adducts (Figure 4-34). In particular, the risk of hepatic lesion occurrence in English sole has been consistently low (> 0.20), compared to lesion risk at cap initiation (1.0), from approximately four years after sediment cap placement through April 2004.

These results show that the sediment capping process has been relatively effective in reducing PAH exposure and associated biological effects in resident flatfish species.

Table 4-5. Creosote wood debris removal in northern Puget Sound, between 2002 and 2005. In total, 275 tons of creosote were removed from 112 miles of beach. (Source: Northwest Straits Commission).

Site	Creosote debris removed	Shoreline in project area
Whatcom County shoreline	100 tons	
Padilla Bay	30 tons	26 miles
Skagit County	35 tons	80 miles
Island County	210 tons	6 miles, plus Double Bluff Beach

Impacts to the Ecosystem

Studies with herring embryos demonstrated that water-diffusible compounds from creosote-treated pilings disrupted normal development, and proximity to the pilings was directly correlated to survival (Vines et al. 2000). Northwest Straits beaches are frequently spawning sites for surf smelt and sand lance—two of the local forage fish that are important prey for salmon, marine birds, and other wildlife.

5. Oil Spills

Each year, commercial ships transport about 15.8 billion gallons of crude oil and refined petroleum products through Puget Sound. The total number of vessels and the amount of oil that each vessel can carry have both increased, therefore increasing the risk of oil spills in Puget Sound. For example, newer container ships can now carry up to 3.8 million gallons of fuel, while oil tankers carry upwards of 40 million gallons. Additional sources of potential oil spills are large marine oil terminals, refineries, oil pipelines, land transportation and smaller commercial or recreational boats. A major oil spill in Puget Sound could be catastrophic for Puget Sound marine life and shorelines.

The following section summarizes the overall oil spill statistics for Puget Sound.

Status and trends

Since 2005, there have been no **major** spills (10,000 gallons or more). There have been 19 **serious** spills (25 gallons or more) with about 4,000 gallons reaching Puget Sound. Of this amount, commercial vessels spilled at least 3,160 gallons. Figure 4-35 shows locations of oil spills in Puget Sound since 1998.

Since 1998, Puget Sound and its tributaries experienced one major spill (in 1999), and 165 serious spills, totaling at least 350,000 gallons (Figure 4-36). During each of the last nine years, the total number of oil spills reported to Ecology has stayed about the same, while the number of serious spills has decreased from 23 to about 13 spills per year (Figure 4-37). Spills larger than 25 gallons are included in this tally.

While it is difficult to characterize trends in these low-probability high-impact events, it appears that the volume of oil released from large spills has steadily declined in the last 15 years.

a. Dalco Oil Spill, 2004

An estimated 1,000 gallons of unknown oil product was released to the waters of southern Puget Sound on October 14, 2004, in the vicinity of Commencement Bay and Dalco Passage near Vashon and Maury islands. Oiled beaches were reported on the southern ends of both Vashon and Maury islands, and oil was reported on Puget Sound waters in Colvos Passage and central Puget Sound.

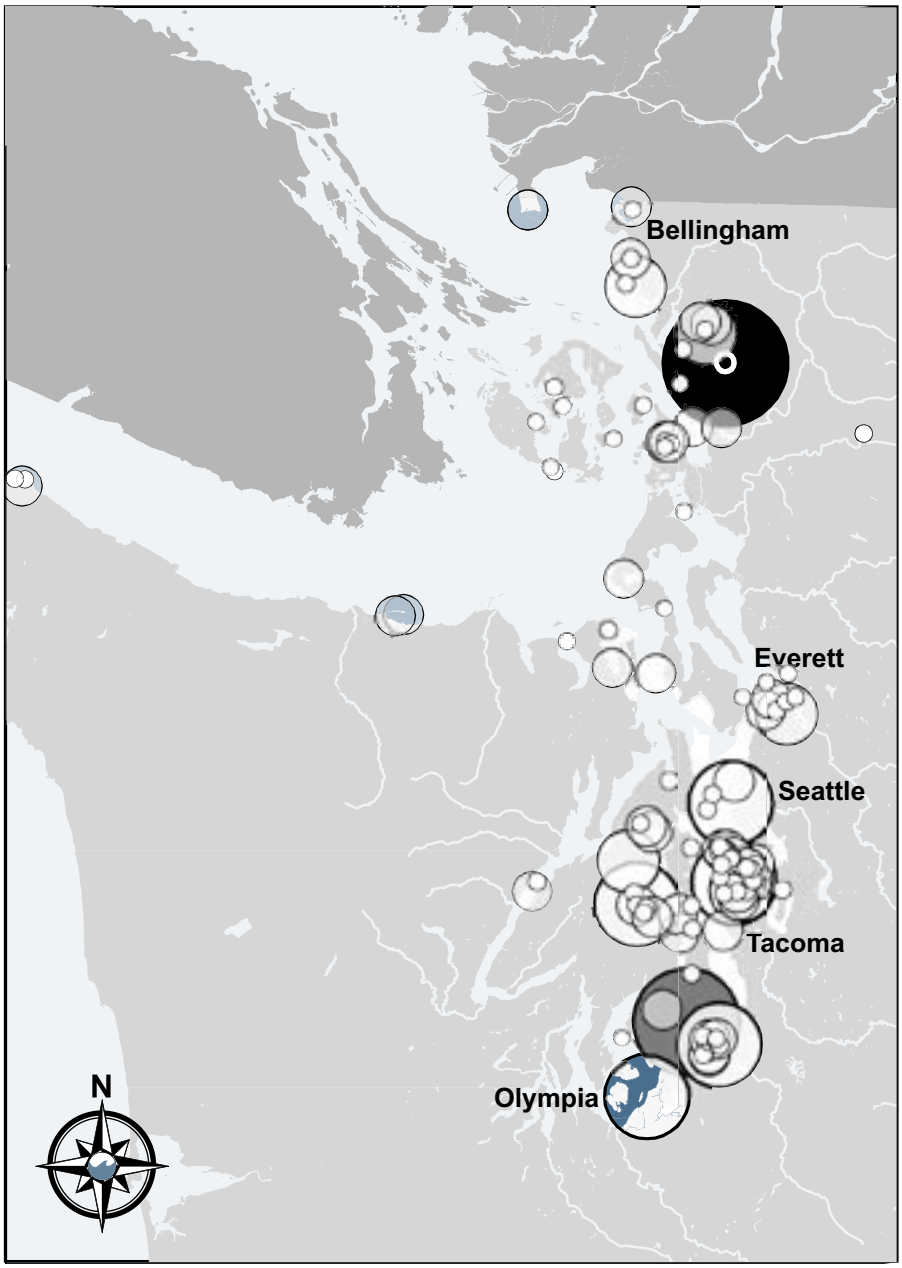


Figure 4-35: **Map of location and volume of oil spills in Puget Sound 1998-2006**. The cumulative volume of oil spilled into Puget Sound during this period is approximately 350,000 gallons, from 165 serious and one major spill.
(Source: Ecology)

Location and volume of oil spills, 1998-present (in gallons)

Serious spills
(25 - 10,000 gallons)

- 25 - 100 gals (110 spills)
- 101 - 1,000 gals (55 spills)
- 1,001 - 2,500 gals (5 spills
- 2,501 - 5,000 gals (5 spills
- 5,001 - 7,200 gals (1 spill - 7,200 gals: Dalco Passage 2004)

Major spills
(> 10,000 gals)

- > 10,000 gals (1 spill - 277,200 gals: Whatcom Creek 1999)

Ecology and WDFW staff collected intertidal sediment samples, along with some tissue and water samples, from the area of immediate impact, as well as from other areas in King, Pierce, and Kitsap counties.

Status and Trends

King County conducted a reconnaissance survey of intertidal sediments to aid in evaluating impacts to both King County properties and other properties located within its borders. The survey involved the collection of 30 intertidal sediment samples from 18 stations located on Vashon and Maury islands and along the mainland shoreline of south King County (Figure 4-38). The samples were analyzed for a variety of chemical and physical parameters to aid in evaluating impacts to Puget Sound shorelines from the oil spill.

Figure 4-36. Total volume of oil spilled per year from 1998-2006. (Source: Ecology)

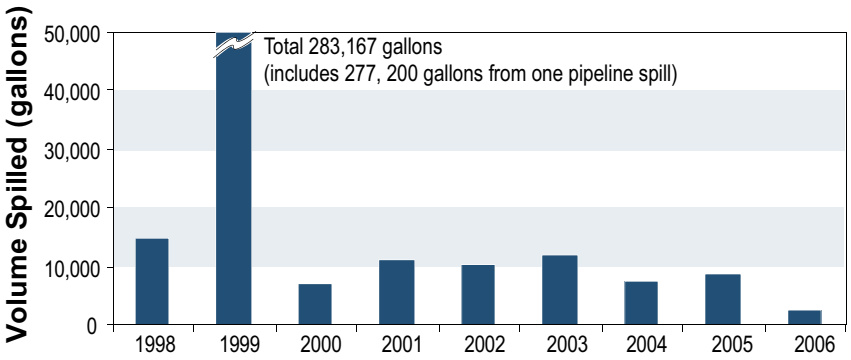


Figure 4-37. **Total number of oil spilled in Puget Sound from 1998-2006.** Spills of 25 gallons or more are included in this total. (Source: Ecology)

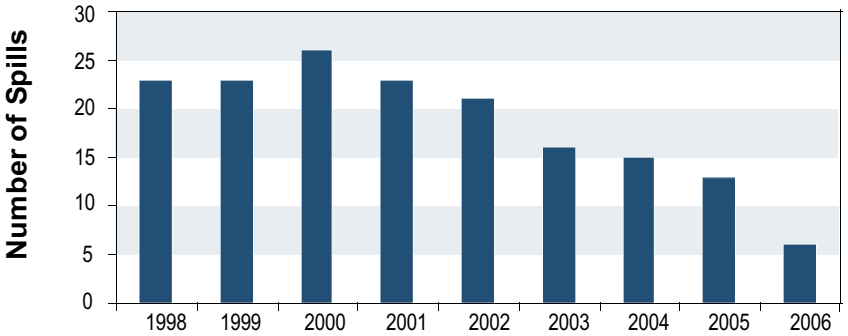


Figure 4-38. Sites of sampling for Dalco oil spill residue. (Source: KC DNRP)

- Single Sample
- Three-sample Transect

Single samples were collected from the plus 7-foot-tide height (referenced to mean lower-low water) at 14 of the sampling stations. A three-sample transect was performed at each of the other four stations, with samples collected from the previous high water mark (indicated by the wrack line), the plus 7-foot-tide height, and the water line at the time of sampling.

All 30 samples were analyzed for petroleum hydrocarbons and PAHs, and those samples collected from the four southernmost stations were also analyzed for PCBs.

Petroleum hydrocarbons were not detected at any of the 18 stations sampled. Trace levels of several PAH compounds were detected at many of the stations sampled. Elevated PAH concentrations were detected at the Tahlequah ferry dock station; however, these concentrations were still well below the Puget Sound Apparent Effects Thresholds[6]. It appears that elevated PAH concentrations detected at the Tahlequah station are the result of legacy contamination from the dock and normal ferry operations. PCBs were not detected at any of the four stations sampled, and PCB analysis was not performed on samples collected from the other 14 stations, based on petroleum hydrocarbon and PAH analytical results.

Based on the results of this intertidal sediment reconnaissance survey, it does not appear that the Dalco Passage oil spill had a lasting impact on intertidal sediment quality at the 18 locations from which King County collected samples.

b. Oil Spill at Point Wells, 2003

On December 30, 2003, nearly 5,000 gallons of diesel oil spilled into Puget Sound at Point Wells near Richmond Beach. Over the next two days, the oil came ashore along 1.5 miles of the northern shore of Port Madison, between Point Jefferson and Indianola. The pristine beach and an adjacent marsh were heavily oiled, and shellfish and other intertidal organisms were contaminated. Cockles, native littleneck, butter, manila, and eastern softshell clams had elevated levels of PAHs, particularly near the marsh. PAH levels ranged from 173 to 17,000 parts per billion.

Status and Trends

During the week following the oil spill, federal and state agencies, the Shoreline Cleanup Assessment Team (SCAT) and the Responsible Party (RP) carried out wildlife surveys from boats and helicopters and on foot along 30 miles of shoreline. Survey teams attempted to rescue 23 animals, including six birds that were oiled. Only two of the birds survived. One oiled seal pup was captured but subsequently died of pneumonia.

DOH's Office of Food Safety and Shellfish immediately closed the shoreline to shellfish harvest, due to concern for public health risks. By one month later, about 700 gallons of oil were recovered and 180,000 pounds of solid waste were removed, including 14 tons of oil-coated pea gravel from the upper intertidal zone.

There are no accepted international or federal limits for oil-related contaminants in shellfish. Seafood contaminated by oil is considered adulterated, meaning the contaminant may impart a taste or smell. Consumption of adulterated seafood is not regarded as an acute health risk.

[6] The Apparent Effects Thresholds (AFT) is the concentration of a contaminant above which statistically significant likelihood of adverse affects is expected (EPA 1988).

6. Endocrine Disrupting Compounds

Endocrine disruptors are compounds that interfere with the normal actions of hormones in humans, fish, and wildlife. Environmental estrogens are an especially troublesome class of endocrine disruptors. These compounds include naturally occurring estrogens in plants and in animal waste, as well as synthetic estrogens and estrogen-like compounds (xenoestrogens) that are structurally similar to natural hormones. Xenoestrogens, which include some chlorinated pesticides, birth control pills, plastics and plasticizers, and surfactants in soaps and other personal care products, act as hormone mimics and may block natural hormone functions.

a. Monitoring for Endocrine Disrupting Compounds in Puget Sound

King County initiated a pilot monitoring study in 2003 to determine if Endocrine Disrupting Chemicals (EDCs) are present in surface waters of King County and, if so, at what concentrations. The County also wanted to better understand the potential for EDCs to effect aquatic life, and King County's data were compared to data from laboratory exposure studies. Samples were collected from marine waters, large lakes, rivers, and smaller streams, from stormwater discharges on Seattle's State Route 520 Bridge, and from stormwater discharges in the Sammamish River valley. These initial surveys were not intended to provide a comprehensive assessment of EDCs in King County waters. These data will be used to determine if future monitoring is warranted, and if so, to guide development of such a monitoring program.

Data for 16 EDCs were collected (Table 4-6). Five chemicals were never detected: estrone, methyltestosterone, progesterone, testosterone, and vinclozolin. Six chemicals were detected in greater than 20 percent of the samples: bisphenol A (BPA—a plasticizer), 17-β estradiol (a natural hormone known as E2), and 17α ethynylestradiol (a synthetic hormone, EE2), and three phthalates (bis (2-ethylhexyl) phthalate, diethyl phthalate, and di-N-octyl phthalate). The remaining five chemicals—nonylphenol (NP) (surfactant breakdown product), the phthalates benzyl butyl phthalate, bis(2-ethylhexyl) adipate, dimethyl phthalate, di-N-butyl phthalate—were detected less frequently. Many of the detected phthalates were also commonly detected in laboratory and field blanks, suggesting that their results be interpreted with caution.

Overall, the maximum detected concentrations for several chemicals were in undiluted stormwater runoff; other chemicals were detected at relatively similar concentrations across all water types. For the purposes of this preliminary study, and based on the limited data for some water types, only coarse differences between marine, lake, stream, and limited point-sources were distinguishable.

b. Effects of Endocrine Disrupting Compounds on English sole

Recent studies reveal that environmental estrogens are present in Puget Sound, based on monitoring vitellogenin induction in male English sole. Vitellogenin is the egg yolk protein produced by female fish during the reproductive season. Male fish, whose natural estrogen levels are low, don't normally produce vitellogenin. However, male English sole from several sites in Puget Sound have detectable levels of vitellogenin in their blood, signaling that these residential bottom-feeding fish are experiencing significant exposure to estrogenic compounds in their habitat.

Table 4-6 Maximum detected endocrine disrupting compound concentrations by water type. (Source: KC DNRP)

Chemical (µg/L unless noted)	N	FOD (%)	Marine	Lakes	Stream/ River-Dry weather	Stream/ River-Wet weather	100% Road/ Bridge Runoff	100% Storm-water
Benzyl Butyl Phthalate	72	12.5	0.01	ND	0.011	ND	0.96	2.06
Bis(2-ethylhexyl)adipate	127	9.4	ND	ND	1.02	ND	0.65	0.036
Bis(2-Ethylhexyl)Phthalate	30	100	40.5	13.1	15.8	4.61	20.3	--
Diethyl Phthalate	67	23.9	--	ND	ND	0.55	2.55	ND
Dimethyl Phthalate	164	15.2	--	0.014	0.02	0.022	0.193	1.71
Di-N-Butyl Phthalate	55	9.1	--	--	0.31	ND	0.9	ND
Di-N-Octyl Phthalate	163	29.4	--	0.0396	0.06	0.68	3.36	0.5
Bisphenol A	181	24.9	ND	0.046	0.44	0.934	9.14	1.57
Total 4-Nonylphenol	272	15.8	0.254	0.149	0.46	0.836	44.2	8.9
17-β Estradiol (ng/L)[1]	362	0.3	ND	ND	13.0	ND	ND	ND
17-β Estradiol (ng/L)[2]	344	27.9	0.5	0.6	1.1	0.5	N/A	1.2
Estrone	362	0	ND	ND	ND	ND	ND	ND
17α Ethynylestradiol (ng/L)[1]	362	0	ND	ND	ND	ND	ND	ND
17α Ethynylestradiol (ng/L)[2]	343	23.9	ND	0.9	0.63	2.0	ND	5.9
Methyltestosterone	362	0	ND	ND	ND	ND	ND	ND
Progesterone	362	0	ND	ND	ND	ND	ND	ND
Testosterone	362	0	ND	ND	ND	ND	ND	ND
Vinclozolin	362	0	ND	ND	ND	ND	ND	ND

— No usable samples for matrix

Bold Maximum of all detections

ND All values non-detect in matrix

N Number valid samples, by chemical, all matrices

FOD Frequency of detection based on usable samples.

1 Analysis by GCMS

2 Analysis by ELISA

NA Not Analyzed

PSAMP monitoring studies have shown that, of all the sites sampled, those in Elliott Bay have the highest number of males (over 30 percent samples) with detectable levels of vitellogenin in their blood.

Female sole are also affected by exposure to the estrogenic compounds at the Elliott Bay sites. Researchers have found that some females begin spawning at a younger age and demonstrate altered reproductive timing. Over time, these physiological changes induced by increase exposure to estrogen compounds in the environment, coupled with other stressors, may challenge the future reproductive success of English sole.

As yet, we are unsure about the specific compounds that are responsible for vitellogenin induction and other reproductive abnormalities in English sole. Monitoring studies are beginning to characterize various pharmaceutical and wastewater compounds in the Puget Sound and its associated watersheds that could have estrogenic activity.

Laboratory studies are underway with salmon, sole, and other species to determine the sensitivity of fish species to xenoestrogens. In one study, male salmon were exposed to ethinyl estradiol, the synthetic estrogen found in birth controls pills, at concentrations within the range that has been measured at Puget Sound sites. These fish showed changes in reproductive hormone levels that could disrupt their reproductive cycles. Effects were found in English sole exposed to the natural estrogen, 17-beta estradiol. In another study, zebrafish were exposed to similar concentrations of ethinyl estradiol, and their sexual behavior was observed. Exposed males became less aggressive, and their mating success was reduced. Laboratory exposure studies with English sole are also helping to identify industrial chemicals that might be causing vitellogenin production in male sole. Male sole exposed to the plasticizer bisphenol A or the surfactant nonylphenol produced vitellogenin; those exposed to phthalates did not.

Table 4-7. Summary of trace metal results at Station LTDF01, 1988-2004
(Source: KC DNRP)

Metal	Frequency of Detection	Detection Limit Range[1]	Concentration Range[1]	SQS[1]
Arsenic	10/12	3.1 – 12	9.8 – 15.2	57
Cadmium	7/12	0.18 – 0.70	0.23 – 0.49	5.1
Chromium	12/12	0.31 – 1.2	27.9 – 47.8	260
Copper	12/12	0.23 – 0.93	37.4 – 65.2	390
Lead	12/12	1.8 – 7.0	48.0 – 69.7	450
Mercury	12/12	0.033 – 0.047	0.425 – 0.911	0.41
Silver	10/12	0.23 – 0.93	0.63 – 1.68	6.1
Zinc	12/12	0.31 – 1.2	74.4 – 100	410

[1]All values reported in mg/kg DW.

7. Metals in Puget Sound

a. Metals in Elliott Bay Subtidal Sediments

King County has monitored sediment quality at one station along the Seattle waterfront as part of its ambient sediment monitoring program. Subtidal sediment samples have been collected from Station LTDF01 (Table 4-7) and analyzed for metals, among other chemicals since 1988.

Samples were collected annually from 1988 through 1993, in 1995, and biennially from 1996 through 2004. Included in the suite of metal analytes have been the eight trace metals regulated under the Washington State Sediment Management Standards: arsenic, cadmium, chromium, copper, lead, mercury, silver, and zinc. Table 4-7 summarizes analytical results for these eight trace metals. It includes the frequency of detection, the range of detection limits, the range of detected metal concentrations, and the Sediment Quality Standard (SQS) chemical criterion for each metal. Data in the table are presented in units of milligram per kilogram, normalized to dry weight.

Arsenic, cadmium, and silver were not detected in every sample, and detected concentrations of these three metals have generally been just above the analytical limit of detection. Concentrations of arsenic, cadmium, chromium, copper, lead, silver, and zinc have been consistent over the 16-year monitoring period, and all detected sediment concentrations of these metals have been well below their respective SQS chemical criteria.

Concentrations of mercury, however, have consistently been above the SQS chemical criterion of 0.41 mg/kg. Figure 4-39 presents sediment mercury concentrations at Station LTDF01 between 1988 and 2004.

Mercury concentrations exceeded the Cleanup Screening Level (CSL) in sediment samples collected during the first eight monitoring events. The CSL is the higher of the two Sediment Management Standards chemical criteria. Since 1998, however, sediment mercury concentrations have been below the CSL but still exceeding the SQS. The data indicate a possible downward trend in mercury concentrations at Station LTDF01 (Figure 4-39).

b. Metals in Puget Sound Sediments

As part of PSAMP, Ecology sampled sediments at 10 fixed stations each spring, from 1989 through 2000 (Figure 4-40). Stations were chosen from a variety of habitats and geographic locations in Puget Sound. Sediments from each station

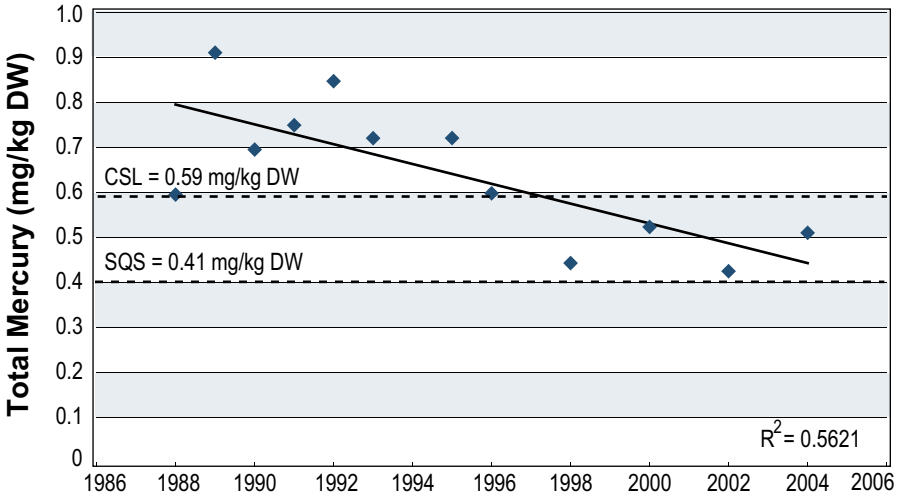


Figure 4-39. Mercury concentrations at King County's Elliott Bay station, 1988-2004. Dry-weight mercury concentrations along with Sediment Quality Standard (SQS) and Cleanup Screening Level (CSL) chemical criteria, and line of regression showing a downward trend in concentration.
(Source: KC DRNP)

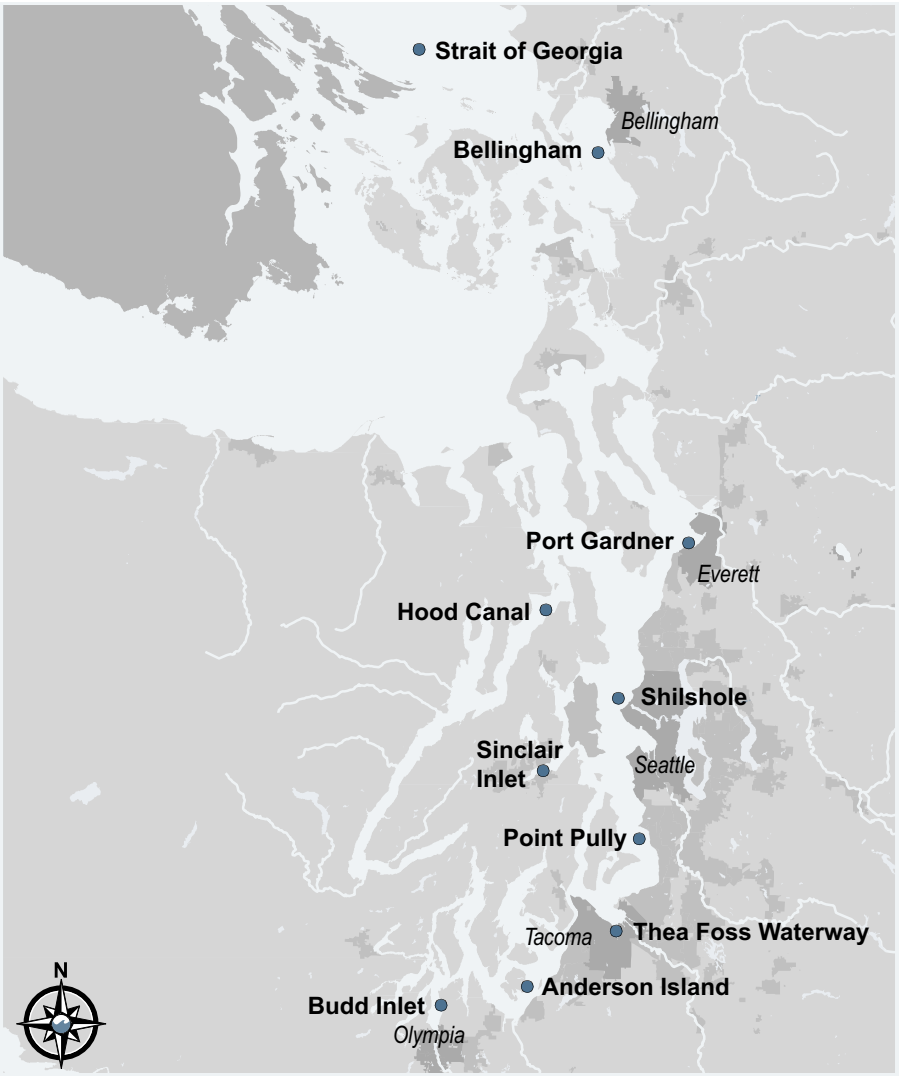


Figure 4-40. Location of 10 long-term PSAMP sediment monitoring stations in Puget Sound.
(Source: Ecology)

were analyzed for particle size, organic carbon content, and the presence of more than 120 chemical contaminants, as well as the types and abundances of sediment-dwelling organisms.

Chemical contaminants in the sediments were measured yearly from 1989 through 1996 and again in 2000. The contaminants examined included priority pollutant and ancillary metals, as well as organic compounds, such as PAHs, chlorinated pesticides, and PCBs. Changes in sediment condition over this time period included decreases in levels of metals at some stations.

Status and Trends

The concentrations of most metals did not change significantly over the study period. Those that did change generally decreased. There was a significant decrease in copper across all stations and significant decreases in metals in general at stations in Port Gardner and Budd Inlet (Table 4-6).

Impacts to the Ecosystem

Toxic metals enter the environment as waste from industrial manufacturing and mining, municipal wastewater, combustion products, and agricultural pesticides (Newman and Unger 2003). Nationwide, metal concentrations in fresh water and estuarine sediments have exhibited declines, similar to those observed in this study, since the mid-1970s. These trends may reflect decreases in emissions to air and water from municipal and industrial sources, following the implementation of federal clean water and air regulations. However, despite these improvements, metal concentrations remain above sediment quality guidelines in many urban bays of Puget Sound, emphasizing the need for continued monitoring and cleanup (Lefkovitz et al. 1997, Mahler et al. 2004).

The PSAMP long-term monitoring program provides a vital record of sediment conditions in Puget Sound and gives insight into the effects of both natural and human-driven stressors on the estuary.

c. Metals in Biota

Since 1986, NOAA's National Status and Trends (NS&T) Program has been monitoring contaminants in mussels from Puget Sound, the Strait of Juan de Fuca and the Washington coast. The NS&T Program analyzes nearly 150 separate chemicals in whole soft tissue from composites of mussels (*Mytilus edulis* and *M. californicus*) collected at each of 17 sites in Washington and one at the Columbia River south jetty. Samples were collected annually to 1994 and every other year since then. In this report, five of the 17 sites in Washington are summarized.

Status and Trends

Trends for five metals from five locations (Budd Inlet, Port Townsend, Squalicum Marina in Bellingham, Fourmile Rock near Seattle, and Cape Flattery near Neah Bay on the outer coast) are shown in figures 4-41 a through e. From 1988-1990, three samples per site were collected annually. After 1990, the sample size dropped to one sample per site every two years. Data shown is the median concentration of metals.

Average mercury levels in Puget Sound mussels are 20 percent higher than the national median of 0.1 ppm dw. Mercury concentrations are highest in Bellingham Bay, where the median concentration rages between 1.5 and 2.6 ppb dry weight. Cape Flattery, the closest to the coast, had levels ranging 0.9 and .2 ppb. The remaining three sites have lower mercury levels, with some variability across the 17 year monitoring period.

Metals	Station Change 1989-1996 vs. 2000										Station Trend 1989-2000									
	Strait of Georgia	Bellingham Bay	Hood Canal	Port Gardner	Shilshole	Sinclair Inlet	Point Pully	Thea Foss WW	Anderson Island	Budd Inlet	Strait of Georgia	Bellingham Bay	Hood Canal	Port Gardner	Shilshole	Sinclair Inlet	Point Pully	Thea Foss WW	Anderson Island	Budd Inlet
Priority Pollutant																				
Antimony						--	--		--							--	--	--	--	
Arsenic	--	--	--	--	↓	↓	--	↓	↓	--	--	--	--	--	--	--	--	--	--	--
Cadmium						--				↓	--	--	--	--	--	--	↑	--	↑	--
Chromium	--	--	--	--	↑	--	--	↑	↑	↓	--	--	--	--	--	↓	--	--	--	--
Copper	--	↓	--	↓	↓	--	↓	--	↓	↓	--	--	--	--	--	↓	↓	--	--	--
Lead	--	--	--	--	--	--	--	↓	--	↓	--	--	--	--	--	--	--	--	--	↓
Mercury	--	↓		--	--	--	--	↓	--	↓	--	--	--	--	--	--	↓	--	↓	--
Nickel	--	--	--	↓	--	↓	--	--	--	↓	--	--	--	--	--	--	--	--	--	--
Silver	--	--	--	↓	--	--	↓	--		--	--	--		--	--	--	--	--	--	--
Zinc	--	↓	--	↓	--	--	--	↓	--	↓	--	--	--	--	↓	↓↓	↓	--	--	--
Ancillary																				
Aluminum	--	--	--	↓	--	--	--	↓	--	↓	--	--	--	--	--	--	--	--	--	--
Iron	--	↓	--	↓	--	--	↓	--	↑	↓	--	--	--	--	--	--	--	--	--	--
Manganese	--	--	--	--	--	↓	↓	--	--	--	--	--	--	--	--	--	--	--	--	--

Table 4-6. Changes and trends in metals concentrations in sediments. Samples were collected from 1989 through 2000 for the PSAMP sediment monitoring program. Most metals declined in concentrations during this period, except for chromium and cadmium, which showed increases in some locations.
(Source: Ecology)

↑,↓..... increase, decrease (α=0.05)

↑↑, ↓↓ ..... increase, decrease (α = 0.01)

-- ..... no change

blank..... insufficient data

Shaded results indicate changes for all stations combined for a single compound or for all compounds combined for a single station, at α=0.05.

Puget Sound mussel samples are close to the national median of 2.1 ppb with the exception of Budd Inlet, where levels reached approximately 12 ppb in 2002-2003. The high variability in cadmium levels at this site may be due in part to the small sample size (one sample every two years) may account for the large inter-year variability seen in lead at this site. But local sources of cadmium may be causing this site to have overall higher levels than other Mussel Watch sites in Puget Sound.

Lead concentrations in mussels from the five sites is close to the national median of .77 ppb, with the exception of Budd Inlet and Cape Flattery which both had peaks in the mid-1990s (3.7 and 1.7 ppb respectively). Since this time, the levels at these two sites have returned levels below 1.0 ppb.

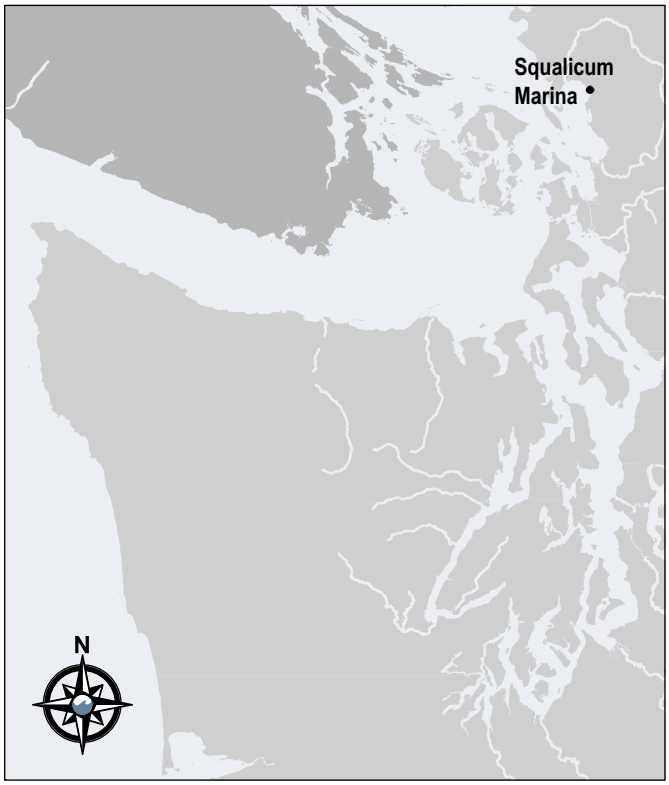
Since the Mussel Watch program began, zinc levels in Washington mussels from the five sites have exceeded the national median of 110 ppb dry weight for all sites. Budd Inlet has only slightly exceeded the national median (approximately 140 ppb in the late 1980s) but in recent decades has hovered near 100. Bellingham and Fourmile Rock have had the highest zinc levels, ranging in the 150-250 ppb concentration over the past two decades.

Copper levels in mussels have been fairly steady at Budd Inlet, Port Townsend, Fourmile Rock and Cape Flattery, showing little change since 1985. In Bellingham Bay, however, copper levels have increased steadily in the past 6 years, with currently levels about twice the national median for copper (8.0 ppb dry weight). Levels in Bellingham have been higher than other sites since monitoring began in the early 1980s.

Copper disrupts sense of smell in fish

The sense of smell in salmon is controlled by the olfactory sensory organ, a delicate structure appropriately called a "rosette" for its radial arrangement of folded tissue. The organ is designed to detect odors as they are carried by water through the olfactory cavity. Studies have shown that exposure to dissolved copper at levels close to those measured in stormwater can impair a fish's sense of smell. This can cause critical behavior changes in fish, affecting predator avoidance, natal stream imprinting, homing, and mating synchronization.

Figure 4-41a. Trends of metals in mussel tissue in Puget Sound. Mussel Watch data for five metals (mercury, lead, copper, zinc and cadmium) from four sites throughout Puget Sound and one at Cape Flattery near the outer coast is shown. The Squalicum Marina in Bellingham had the highest median mercury, zinc and copper levels.
(Source: NOAA National Mussel Watch Program).



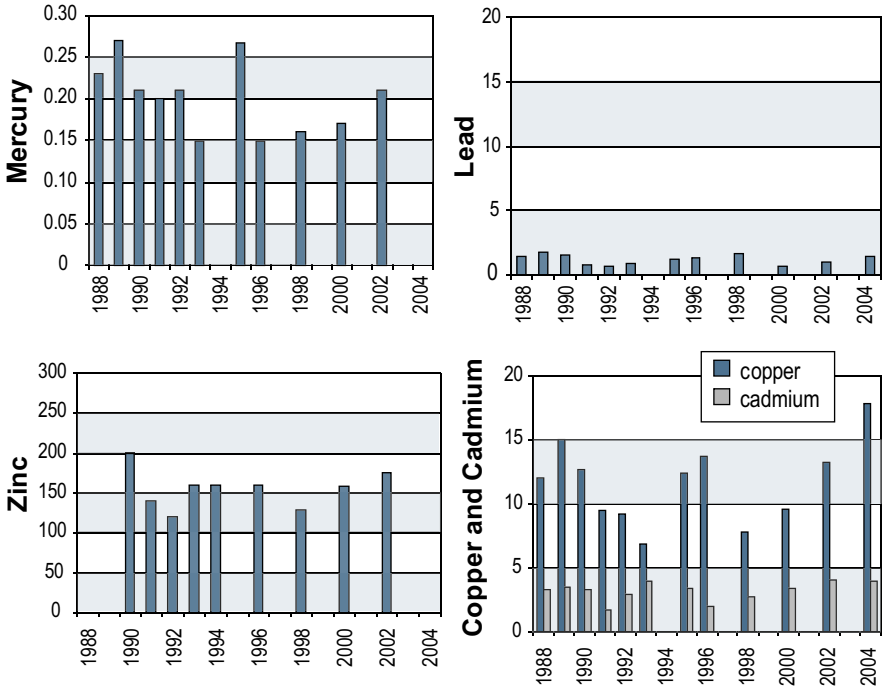


Figure 4-41b. Trends of metals in mussel tissue at Cape Flattery.

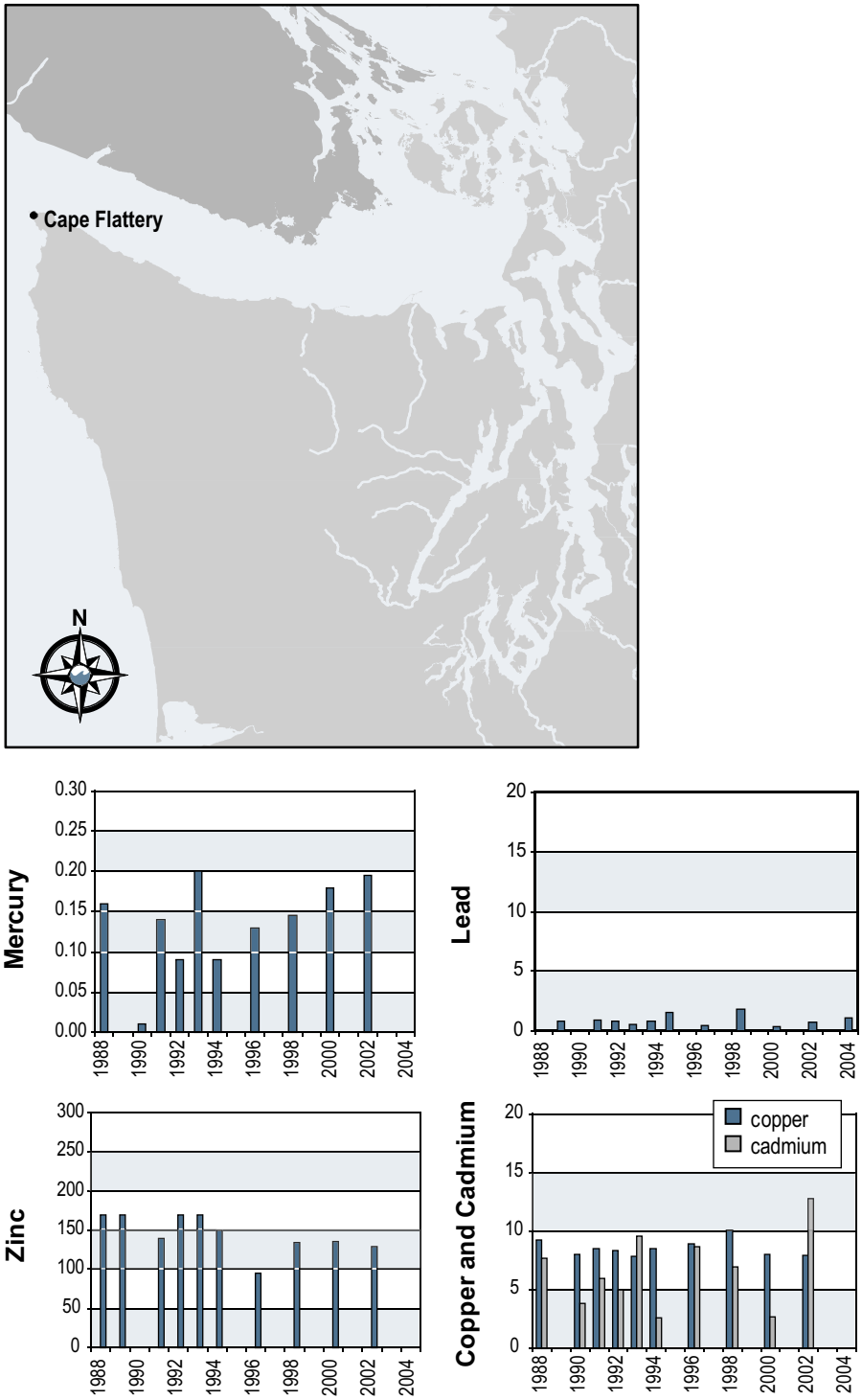
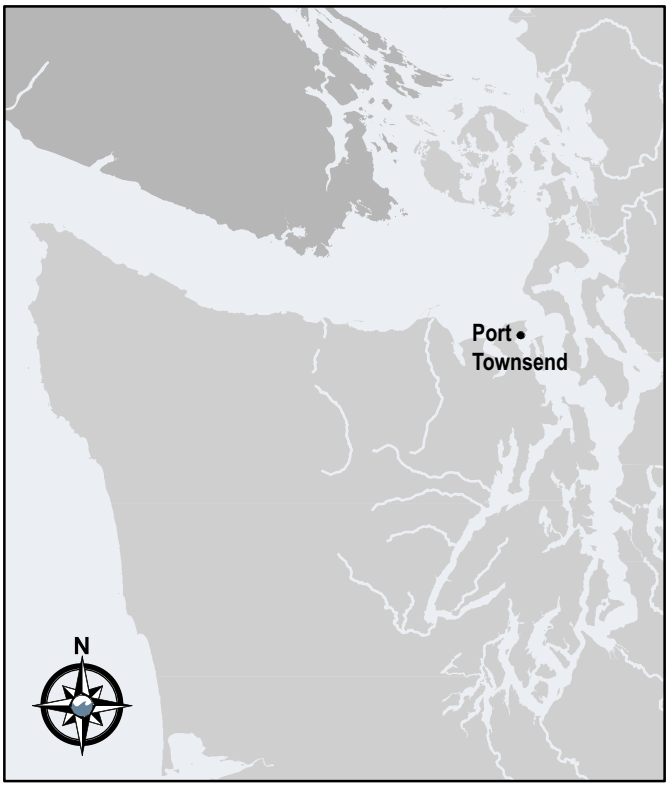
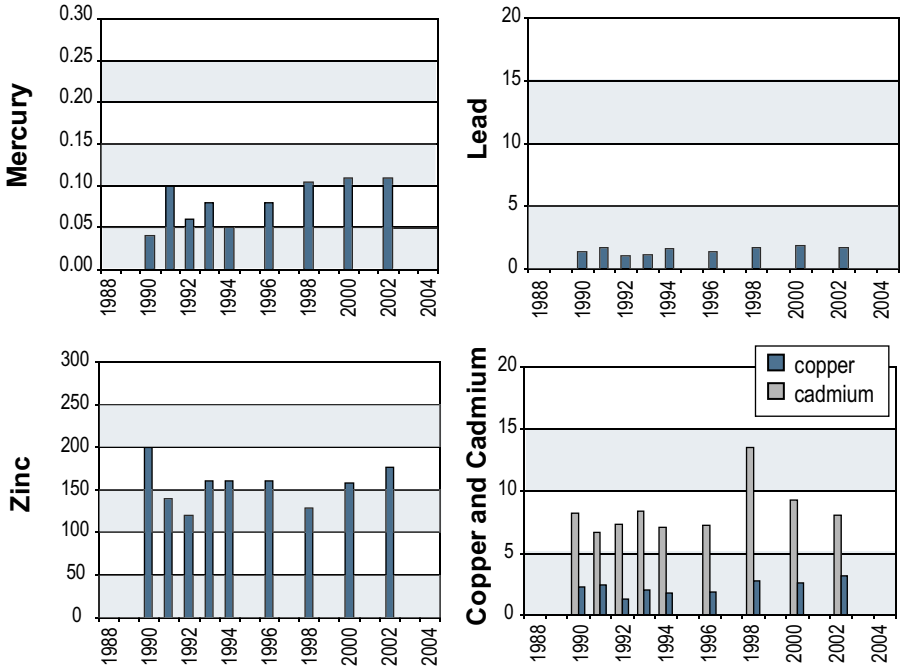
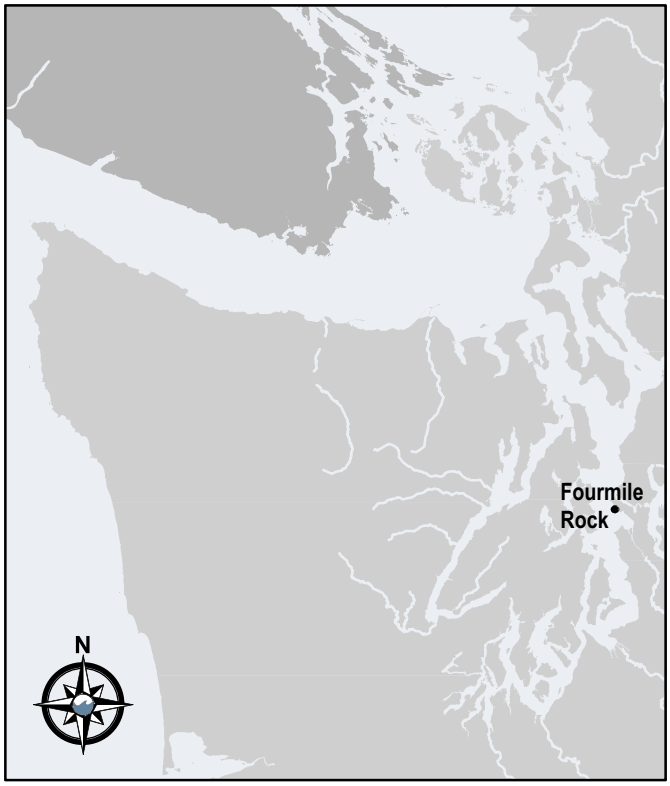


Figure 4-41c. Trends of metals in mussel tissue at Port Townsend in Puget Sound.







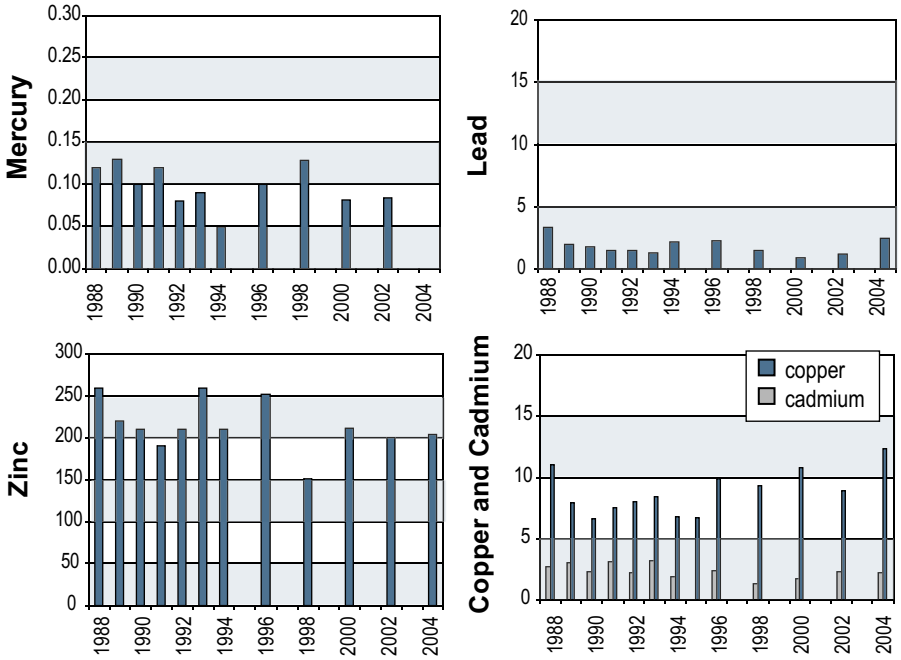


Figure 4-41d. Trends of metals in mussel tissue at Fourmile Rock in Puget Sound.

Figure 4-41e. Trends of metals in mussel tissue at Budd Inlet in Puget Sound.

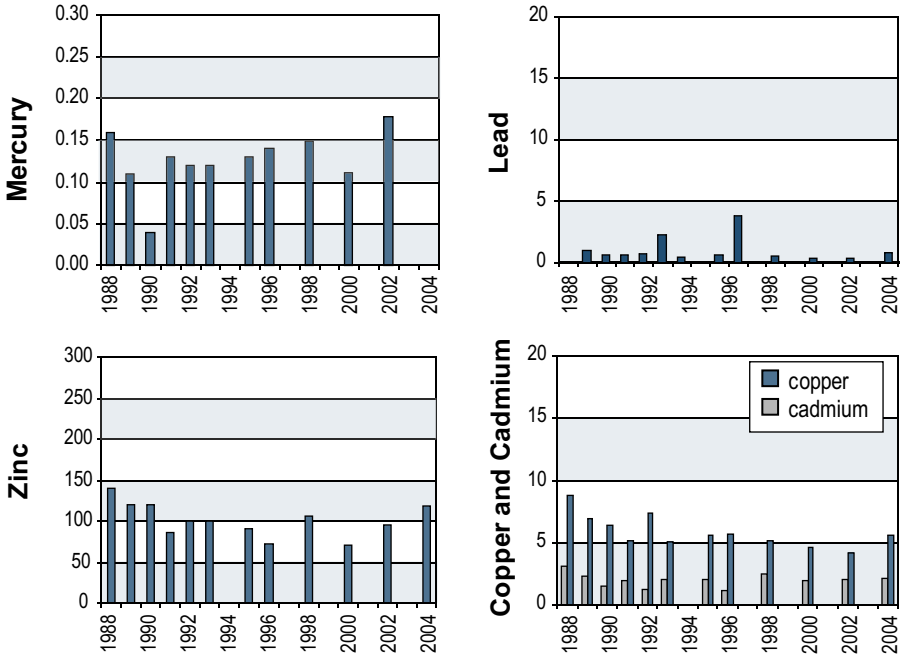


Figure 4-42. Dead coho salmon with 100 percent egg retention. (Courtesy of Sarah McCarthy, NOAA Fisheries Science Center).

8. Effects of Urban Runoff on Biota in Freshwater Systems

Many restoration projects in Puget Sound focus on streams running through urban areas that have barriers to fish passage or unsuitable physical habitat for spawning salmonids. Beginning in the late 1990s, agencies in the greater Seattle area began conducting salmonid spawner surveys to evaluate the effectiveness of local stream restoration efforts. These surveys detected a surprisingly high rate of mortality among migratory coho salmon females that were in good physical condition but had not yet spawned (Figure 4-42). In addition, adult coho from several urban streams showed a similar progression of symptoms (disorientation, lethargy, loss of equilibrium, gaping, and fin splaying) that rapidly led to death of the affected animals. This phenomenon was termed pre-spawn mortality (PSM). Researchers from NOAA Fisheries' Northwest Fisheries Science Center, in partnership with City of Seattle - Seattle Public Utilities and USFWS's Western Washington Fish and Wildlife Office, have been studying adult coho spawners in Puget Sound streams to determine the causes and geographical extent of these acute fish die-offs.

Status and Trends

PSM has been consistently observed over the past several years in many lowland urban streams in the Puget Sound region, with overall rates ranging from approximately 25 to 90 percent of females returning to spawn. Coho die-offs in these streams are generally associated with large rain events. Continuous daily surveys over the past four years in a representative urban stream in West Seattle have revealed female coho PSM rates ranging from 66 to 89 percent, compared with less than one percent in a forested reference stream (Table 4-9). Although the precise cause of PSM in urban streams remains unknown, conventional water quality parameters (e.g., temperature and DO) and disease do not appear to be causal. The weight of evidence suggests that adult coho, which enter small urban streams following fall storm events, are acutely sensitive to pollutants in nonpoint source stormwater runoff. Research is underway to investigate potential linkages between land-use patterns and PSM, as well as the effects of degraded stormwater on other life history stages of coho. Additional studies are being conducted to predict the population-level impacts of PSM on coho stocks that return to spawn throughout the Puget Sound region.

Table 4-9. Pre-spawn mortility in urban and rural streams from 2002-2005. Neither disease nor conventional water quality parameters (temperature and dissolved oxygen) appear to be the cause of the mortality, suggesting that pollution sources in stormwater may be the culprit.
(Source: NOAA Fisheries Science Center)

	Year	N	PSM
Longfellow Creek (urban)	2002	57	86%
	2003	18	66%
	2004	9	89%
	2005	75	89%
Fortson Creek (non-urban)	2002	114	<1%

Impacts to the Ecosystem

Collective results to date suggest that stormwater runoff has important negative impacts on both the survival and reproductive success of coho salmon in urban and urbanizing watersheds. The current aim is to determine the consequences of spawner and embryo mortality on healthy populations of wild coho throughout the Puget Sound Basin.

9. Recommendations

In the *2002 Puget Sound Update*, recommendations were provided based on the results from the studies summarized in the report. The recommendations for toxic contaminants work and progress made through 2006 on those recommendations are summarized below:

Recommendation from the *2002 Update* for Toxic Contaminants	Progress made through 2006 on recommendations in the *2002 Update*
As much as possible, studies should be interdisciplinary in nature such that the contaminant data can be integrated with population data and life history patterns. Understanding of the currently unexplained variability in some contaminant data will require such an approach.	Progress is difficult to document. However, WDFW and NOAA continue to collaborate on studies of PAH exposure in English sole and PTB concentrations in fish species in Puget Sound.
Further pilot studies are needed to assess toxic contaminant impacts in previously understudied species, to fully evaluate ecosystem effects of these contaminants.	No progress to report.
Continued monitoring is needed for biota affected by contaminants, even when contamination levels and productivity are improving, as long as a contaminant impact is observed. This will ensure that recovery proceeds as expected and important causal factors have not been overlooked.	• Continued studies of the affects of PAHs on English sole (WDFW). • Continued studies on vitelloginin levels in male English sole (NOAA).
Further studies are needed to better understand sources of the recent increases in benzoic acid in sediments and shellfish as well as the ecological implications of these increases.	No progress to report.
For contaminants that are increasing in Puget Sound sediments, scientists need to quantify sources, and policy-makers need to determine if current controls are inadequate to control these pollutants.	No efforts to quantify sources have been initiated as of August 2006 but interagency discussions have produced proposals for near- and long-term work to assess loads of toxic chemicals to Puget Sound.
WDFW needs to further investigate the sources and pathways of contaminants in Pacific herrings. Emphasis should be on assessing whether dredged material management, contaminated sediment cleanup, or wastewater discharge control could reduce herring contaminant exposure.	No progress to report.

Recommendation from the *2002 Update* for Toxic Contaminants	Progress made through 2006 on recommendations in the *2002 Update*
Scientists need to use developing food web models for the Strait of Georgia and South Puget Sound Basin and information on contaminant burdens in various organisms, to see if they can accurately describe the major pathways of accumulation to rockfish, salmon, harbor seals, orcas, and Hood Canal bald eagles. These studies can identify gaps and encourage additional studies to fill them. This work can also identify the leading opportunities to reduce the accumulation of toxics in the food web.	No efforts to develop and apply food web models have been initiated as of August 2006 but discussions have begun about projects that if funded, could begin in early 2007 or the 2007-2009 biennium.
Scientists need to continue efforts to relate fish contamination and disease to sediment contamination, especially at areas such as the Seattle waterfront and Thea Foss Waterway, to try and learn more about how fish respond to cleanup efforts and sediment disturbances.	Studies of the risk of developing liver lesions in English sole continued at the Seattle waterfront, Thea Foss Waterway, and other sites in Puget Sound. Linkage to clean up or capping efforts is implied. (NFSC/WDFW)
More focused monitoring is needed to measure the effectiveness of alternative contaminant control measures.	No progress to report.

Moving Forward on Puget Sound Science

In looking ahead to what recommendations to report in future editions of the *Puget Sound Update,* it makes sense to focus on the goals and strategies that have been recommended in *2006 The Puget Sound Partnership Final Report*, the PSAT *2007-2009 Conservation and Recovery Plan for Puget Sound,* and the 2006 PSAMP Review. Collectively, these three sources provide targets and goals developed and supported by a large scientific community and reflect both short-term (two year) and long-term considerations for protecting and restoring Puget Sound's health.

The following bullets summarize the goals and strategies put forth by the Puget Sound Partnership, PSAT, and PSAMP that are related to toxic contaminants (Chapter 4 of this report). Progress towards these goals and strategies will be reviewed in the next edition of the *Puget Sound Update*.

Puget Sound Partnership Final Report (from Appendix A)

Goal: Puget Sound species and the web of life thrive.

- Terrestrial, aquatic and marine species exist at variable levels into the future and biodiversity of the overall ecosystem is naturally maintained.
- Invasive species do not significantly reduce the viability of native species and the functioning of the food web.
- The harvest of fish, wildlife, shellfish and plants is balanced, viable, and ecosystem based.

2007-2009 Conservation and Recovery Plan for Puget Sound

Priority 1. Clean up contaminated sites and sediments

- Continue to identify and clean up contaminated sites.
- Manage navigation dredging operations to clean up contaminated areas whenever possible and prevent contamination of unconfined disposal sites.

Priority 2. Prevent Toxic Contamination

- Reduce the use and generation of toxic chemicals.
- Reduce the release of toxic chemicals to the environment.
- Improve spill prevention and response.
- Educate residents to change behaviors to reduce toxic contamination.
- Study toxics in Puget Sound.

The Role of Science

Strategies:

- Continue ongoing monitoring of the status and trends of key components of the Puget Sound ecosystem.
- Provide scientific information to stakeholders, decision-makers and the public.
- Direct new monitoring activities to focus on the effectiveness of management activities and policy initiatives.
- Develop a roadmap to prioritize, finance and conduct focused research on emerging topics or research questions that are brought forth through PSAMP and science programs.

Detailed recommendations for further research and monitoring

Many of the following recommendations are an outcome of the 2005-2006 PSAMP review and have been included as actions in the *2007-2009 Puget Sound Conservation and Recovery Plan*. Progress towards these and previous recommendations will be reported in the next *Puget Sound Update*.

Toxics assessment

- Threats to human and marine wildlife health from exposure to major contaminants (PCBs, PBDEs, mercury, PAHs, metals, and pesticides) and new, emerging contaminants (such as pharmaceuticals and personal care products) are identified and measured in key indicators in the food web, including mussels, Pacific herring, salmon, and seals.
- The sources and contribution of key toxic contaminants from terrestrial, atmospheric, and marine discharge sources are determined. This information is used to determine toxic loading in sediment and key fish, mammals, and water bodies in Puget Sound.
- PSAMP status and trends monitoring of sediments is continued to determine spatial extents of contamination, toxicity, and benthos impairment within regions of Puget Sound.
- Develop a new urban embayment layer to PSAMP regional and strata layers for spatial extent calculations, as well as an assessment of sediment quality to measure success of contaminated site cleanup. Monitor sediment quality at multiple scales (Soundwide, regional, strata, and bay) for conventional contaminants and newly emerging contaminants. Also monitor sediment quality on intertidal lands.

Toxics Management

- Develop comprehensive and extensive integrated contaminant monitoring plans to track pathways and burdens and to link to human consumption advisories.

Modeling

- Develop a quantitative model to determine baseline levels of inputs and the fate of toxics in Puget Sound with explicit consideration of forage fish (by age, class, and location), birds, fish, and mammals.
- Develop a conceptual model of Puget Sound, using data from PSAMP, the Puget Sound Nearshore Partnership, and other science programs, to communicate and organize scientific information, relationships, and results.

Processes and Connections

- Develop biological indicators (invertebrates, fish, birds and mammals) of toxic exposure and effects at multiple taxonomic levels.
- Monitor effects of mixtures and interactions of nutrients, organics, and metals, not isolated contaminants.