State of Knowledge Report

Toxics in Fish Implementation Strategy

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Executive Summary

The Toxics in Fish Vital Sign is focused on the stated objective of reducing exposures to chemicals that have the potential of harm to fish and the people or predators that consume them. The Vital Sign includes four distinct chemical classes (PCBs, PBDEs, PAHs, and EDCs) interacting with three fish species (or species groups: English sole, Pacific herring, salmonids). This range of contaminant and species classes was selected to represent a broad range of chemical fate and transport properties as well as habitats and geography. Collectively, the chemical and biota groups can inform where contaminants occur and how they might affect biota. An Implementation Strategy based on this complex Vital Sign should consider the unique aspects (sources, pathways, uptake, metabolism, species risk exposure, losses, etc.) of each contaminant class and indicator species.

This State of Knowledge report is meant to provide a scientific and technical foundation to the Toxics in Fish (TiF) Implementation Strategy. It is part of an overall package that also includes the TiF Implementation Strategy Narrative, which describes strategies, approaches, and activities for managing contaminants, and the TiF Base Program Analysis, which describes the existing regulatory and programs that address issues related to managing toxic contaminants.

Based on the information reviewed, the following key points should be considered in recovery planning:

- PCBs remain a pervasive problem in the pelagic food web in the Puget Sound basin. PCBs from specific sites (e.g., Superfund sites, Puget Sound Naval shipyard) and bays (e.g., Elliott Bay), likely enter the pelagic food web in Puget Sound's central basin, where they accumulate, biomagnify, and persist. Recovery strategies should address these important sites and bays.

- Benthic fish in urban bays continue to be exposed to PCBs at levels that might cause harm and/or result in consumption advisories. Reduction of PCBs in benthic fish necessitates bay-scale recovery strategies for urban areas such as Elliott Bay/Duwamish River, Commencement Bay, and Sinclair Inlet. Fish in non-urban areas are generally not exposed to harmful levels of PCBs.

- The levels of PBDEs in English sole and Pacific herring are generally below adverse thresholds and declining, suggesting that existing recovery strategies have been effective at protection and restoration. PBDEs in juvenile salmon indicates that there are localized areas with exposures at levels of concern. Targeted actions, for example, aimed at in-river sources, may be more appropriate.

- Liver disease caused by PAHs in English sole (benthic food web) has generally declined throughout the Puget Sound, indicating the overall effectiveness of management programs. However, the contribution of various management actions (e.g., sediment cleanup, source control, removal of creosote-treated pilings, etc.) remains unknown. There are some localized areas (e.g., Duwamish River) with continued high PAH exposures in English sole. Pacific herring (pelagic food web), particularly in south and central Puget Sound, continue to be exposed to high PAH levels.

- Information on the occurrence and impacts of Contaminants of Emerging Concern (CECs) and Endocrine Disrupting Compounds (EDCs) is currently sparse, but improving. Ongoing investigations are finding that these chemical groups are ubiquitous in the marine environment (albeit at very low levels) and some chemicals might be contributing to the reduced survival of important fish species.

These strategy recommendations are based on the wealth of existing environmental monitoring data available in the region. However, there are several key uncertainties which limit our ability to develop precise strategies.

Further, it should also be noted that recovery of the Puget Sound ecosystem regarding toxic contaminants should include a full evaluation of all chemicals across the full ecosystem landscape. The current list of recovery targets is limited. For example, there is not a human health recovery target for Pacific herring and EDC-exposure thresholds are under development.

1 Introduction

This State of Knowledge report is meant to provide a scientific and technical foundation to the Toxics in Fish (TiF) Implementation Strategy. It is part of an overall package that also includes the TiF Implementation Strategy Narrative, which describes strategies, approaches, and activities for managing contaminants, and the TiF Base Program Analysis, which describes the existing regulatory and programs that address issues related to managing toxic contaminants.

The TiF Implementation Strategy was developed based on a conceptual model where human related pressures (e.g., land use development and land use activities) results in chemical inputs to the Puget Sound watershed through a variety of pathways (e.g., stormwater, waste water, air deposition) that lead to contaminant exposures to aquatic species and the humans that consume them. These impacts can be addressed through prevention (through the elimination and control of primary source), mitigation (by managing contaminant loading pathways), or clean ups (by addressing existing contamination in the environment). Strategies to do so are outlined in the TiF Implementation Strategy.

The purpose of this State of Knowledge document is to provide:

- background information on the key indicator species associated with the Toxics in Fish vital sign,
- detail on the chemical properties and fate and transport of the chemicals and chemical classes relevant to the Toxics in Fish Vital Sign,
- an overview of important sources and pathways, and
- considerations, such as effectiveness of approaches and actions, that might be important in the refinement and implementation of the recovery strategies proposed in the Implementation Strategy document.

Information in this document is largely based on a review of the salient literature, including agency reports and publications for regional studies. Despite the wealth of information, there remain uncertainties and gaps in knowledge that impede our ability to effectively manage the impacts of anthropogenic chemicals. The identification and evaluation of research and monitoring priorities to address uncertainties are specifically addressed in the TiF Implementation Strategy Narrative document.

2 Toxics in Fish Vital Sign

The TiF Implementation Strategy was developed to achieve the recovery targets specified under the Toxics in Fish Vital Signs. A complete description of the Vital Sign, including background, the development process, and it indicators and targets is included in the TiF Implementation Strategy narrative. Information is also at the Puget Sound Partnership Toxics in Fish Vital Sign web page.

Only selected details will be repeated here to provide sufficient context for this State of Knowledge report.

2.1 Toxics in Fish Vital Sign Goal

The Puget Sound Partnership Vital Signs reflect a series of recovery goals for the Puget Sound watershed. They describe components related to clean and sufficient water resources, protected and restored habitats, thriving species and food webs, and a healthy and vibrant human population. The Toxics in Fish Vital Sign specifies the following goal:

> Fish populations not harmed by toxic contaminants and fish safe for consumption by predators and humans.

2.2 Toxics in Fish Indicators, Targets, and Thresholds

Toxics in Fish indicators and targets were established based on the stated goal. The Vital Sign indicators are designed to cover: 1) a mix of fish species representing different habitats, 2) a broad range of important chemical classes that are known to cause harm, and 3) recovery targets related to fish health and human health

In each case, targets specify levels of contaminant in a given fish species to be below a given threshold, either based the health of the organism, or the health of predators or humans that consume them.

There are four fish species/life stages that are included as Toxics in Fish Vital Sign indicators.

1. English sole (adults) represent conditions in the benthic food web, primarily related to sediment-bound chemicals;
2. Pacific herring (adults) represent the pelagic food web;
3. Juvenile Chinook salmon represent a salmon species thought to be one of the most susceptible to harm from chemicals (during its outmigration), and;
4. Adult Chinook salmon represent a salmon species widely consumed by humans, and so represent a pathway of toxics from seafood to human.

The Toxics in Fish Vital Sign covers four chemical classes:

1. Polychlorinated biphenyls (PCBs) – persistent and bioaccumulative; chemicals that were designed for various industrial uses, and were banned in the US in 1979. Trace amounts are still being produced in a range of consumer and industrial products.

2. Polybrominated diphenylethers (PBDEs) - persistent and bioaccumulative; primarily flame-retardant compounds that occur in many products such as sofas and television plastics. The primary formulations were banned in 2011 in Washington State.
3. Polycyclic aromatic hydrocarbons (PAHs) - persistent and bioaccumulative (in some animals); hydrocarbons that are produced primarily from burning fossil fuels and from oil spills, and which occur in creosote-treated wood products.
4. Endocrine disrupting compounds (EDCs) – a broad group of chemicals representing a wide range of persistence, potential for bioaccumulation, and modes of action. Their common characteristic is that they interfere with proper functioning of the endocrine system and can affect reproduction.

Recovery goals are based on three types of contamination metrics:

1. Observing a health effect that is directly related to exposure to contaminants. Examples include PAH-related liver disease and EDC-related reproductive impairment in English sole. Monitoring toxicopathic disease (diseases that are directly associated with chemical exposures) can be considered the gold standard for contaminant indicators because it tracks fish health directly.
2. Measuring the levels of contaminants in fish tissues, and comparing these levels against published Critical Tissue Levels (CTL; see Meador et al. (2011)). CLTs are estimates of the level of contamination an organism can tolerate before it exhibits some health effect, or a level that results in unacceptable health risks to humans who consume the fish. This is one of the most common ways to evaluate contaminant indicators.
3. Comparing tissue levels against levels in fish from uncontaminated areas.

These CTLs and other thresholds relating exposure and effects may change as the science behind them improves.

Table 2-1 provides a summary of the Toxics in Fish indicators, targets, and thresholds.

Table 2-1. Toxics in Fish Vital Sign Indicators and associated recovery targets for fish species and contaminants. Also included are an indication of habitat/food web position, and spatial scale of habitat range of each of the indicator species.

Indicator	Recovery Goals				Target habitat / food web	Spatial Scale
	PAHs	PBDEs	PCBs	EDCs		
Pacific Herring	< uncontaminated background (fish health)	95% of samples < 470 ng/g lipid[a] whole body (fish health)	95% of samples < 2,400 ng/g lipid[a] whole body (fish health)	not measured	Pelagic	Basin
English Sole	no PAH related disease (fish health)	95% of samples < 40 ng/g wet wt (human health)	95% of samples < 8 ng/g wet wt (human health)	no contaminant-related reproductive impairment (fish health)	Benthic	Embayment
Chinook Adult	not measured	95% of samples < 40 ng/g wet wt fillet (human health)	95% of samples < 8 ng/g wet wt fillet (human health)	not measured	Pelagic	Puget Sound/ Ocean
Chinook Juveniles	< uncontaminated background (fish health)	95% of samples < 470 ng/g lipid[b] whole body (fish health)	95% of samples < 2,400 ng/g lipid[b] whole body (fish health)	not measured	River mouth/ nearshore marine	River mouth/ estuary/ watershed

Notes:

a. calculated at a tissue lipid concentration of 3%, to normalize lipid levels to a common level all herring may exhibit during periods of low feeding

b. calculated at a tissue lipid concentration of 1%, to normalize lipid levels to a common level all juvenile Chinook may exhibit during a typical out-migration

2.3 Status and Trends: Most Current Toxics in Fish Vital Sign Results

The status and trends of the Toxics in Fish Vital Sign was updated in July 2020 (https://vitalsigns.pugetsoundinfo.wa.gov/VitalSign/Detail/11)

2.4 Updates to the Vital Sign indicator species and chemicals

The suite of Vital Signs that are used to track progress for recovery were re-evaluated in 2020 resulting in a modified suite of Vital Signs and indicators. With specific regard to the Toxics in Fish Vital Sign, the following changes were made:

- The Vital Sign was renamed to Toxics in Aquatic Life to include a larger range of indicator species.
- Caged mussels was added as an indicator species, in addition to those listed in Table 2-1.

- The chemical class of Endocrine Disrupting Compounds has been expanded to include a broader suite of Contaminants of Emerging Concern (CECs). This broadens the definition to include potentially harmful compounds that act by different modes of action rather than strictly endocrine disruption.

The details of these changes will be reflected in future versions of the Vital Sign indicator reports. Relevant background information will be included in this document.

3 Toxics in Fish Background

The purpose of this section is to provide background information that may be useful in the interpretation and comparison of Toxics in Fish-related data, formulation of restoration plans, and evaluation of alternatives. It includes information on indicator species, chemicals of interest including physical and chemical properties, sources, and pathways, and contaminant thresholds.

3.1 Indicator species

This section provides background specific to set of indicator species used in the Vital Sign, and how their lifecycle and behavior might impact the interpretation of monitoring results, and the development of a restoration strategy.

3.1.1 English sole (*Parophrys vetulus*)

Photo: Washington Department of Fish and Wildlife

English sole were selected as an indicator species to represent local benthic condition.

English sole are flatfish that inhabit soft bottom (fine sands and mud) areas of estuaries and bays. Adults can live at depths of over 500 m. Juveniles and adults are carnivorous, eating benthic organisms, primarily marine worms, mollusks, crustaceans, and echinoderms. Primary exposures to toxic contaminants would be via consumption of contaminated benthic organisms and direct exposure to contaminated sediments.

Puget Sound English sole may be territorial and the population stratification could extend down to the level of the individual territory (Day 1976, Myers et al. 1998).

Adults tend to remain localized during the late spring and summer and make seasonal spawning migrations between late August and April. Spawning migrations may range from 30-100 km in Puget Sound (Moser et al. 2013). English sole tend to return to home territory after spawning. English sole probably do not feed during spawning period and so the primary contaminant exposure occurs during summer feeding. Behavioral traits suggest that English sole contaminant profiles are reflective of local, bay scale benthic condition (Moser et al. 2013).

3.1.2 Pacific herring (*Clupea pallasii*)

Photo: USGS Western Fisheries Research Center

Pacific herring are a small-bodied, schooling marine fish species that serve as an indicator for toxic contaminants in Puget Sound's pelagic food web. They not only provide a measure of health for pelagic species but also give an indication of the degree to which predators of herring may be exposed to chemicals as a result of consuming herring. This is especially important because it is difficult or impossible to measure contaminants directly in many predators including seals, dolphins, killer whales, and fish-eating seabirds. It is easier to measure contaminants in their more abundant prey.

Approximately 18 stocks (herring with common spawning ground) spawn within Puget Sound. Genetic studies suggest that only the Cherry Point and Squaxin Pass stocks are genetically discrete while the others are not. The herring spawn in shallow areas along shorelines, between the subtidal and intertidal zones. Eggs are deposited on kelp, eelgrass (*Zostera marina*), and other available structures. Most Washington State herring stocks spawn between mid-January and early April.

The exact feeding patterns of the herring stocks is not well understood. However, contaminant profiles have been used to infer feeding patterns from different stocks (West et al. 2008). The contaminant profiles of herring from Strait of Georgia, Cherry Point, Semiahmoo, and Puget Sound are distinct suggesting unique feeding locations. The contaminant profiles of Puget Sound herring (collected from Squaxin, Quartermaster, and Port Orchard) are all very similar indicating that either they are all feeding at similar locations or the contaminant profiles of their prey is similar across locations throughout central Puget Sound.

Herring feed on planktonic organisms. Calanoid copepods made up the bulk of the diet of juvenile herring in the Strait of Juan de Fuca nearshore zone (Simenstad et al. 1977). Adult herring have been observed to feed heavily on both calanoid copepods and krill (Washington Department of Fish and Wildlife 2014, NOAA Fisheries 2017).

3.1.3 Salmon (*Oncorhynchus* spp.)

Chinook salmon (*Oncorhynchus tshawytscha*). Photo: Washington Department of Fish and Wildlife

Juvenile Chinook salmon. Photo: Columbia River Inter-Tribal Fish Commission

Toxic contaminants are monitored in adult salmon because they are important as food to humans, and to a wide range of predators. Moreover, chemicals can impair salmon health, potentially inhibiting recovery. The highly migratory nature of salmon makes it difficult to determine where they pick up chemicals, however the time they spend in Puget Sound is the main risk factor in their exposure to toxic chemicals (O'Neill and West 2009).

Salmon are an anadromous species and so there is the potential for contaminant exposure in rivers, estuaries, and the marine environment during different phases of their life cycles. Different salmon species have different life cycles (e.g., they spend different amounts of time in rivers and/or the Puget Sound during migration) and the contaminant exposure of an individual fish will vary according to life stage (e.g., juvenile vs adult), species, and stock because each of these factors affects individual contaminant burden. Such factors should be considered when comparing tissue contamination across samples or attempting to utilize tissue contaminant levels to infer environmental condition.

Current monitoring includes both adults and juveniles. Of particular concern is exposures to chemicals in urbanized estuaries or river mouths as juveniles migrate from fresh to salt water. The physiological challenge of moving from freshwater to saltwater makes them particularly susceptible to stressors such as chemical contaminants, and it is in these urban estuaries where many contaminants occur.

Considerations related to salmonid monitoring:

- Adult salmon may undergo contaminant exposure in marine areas outside the Puget Sound. Total contaminant burden in returning spawners likely reflects the condition of the marine environment and not necessarily the condition of the pelagic food web within Puget Sound (O'Neill and West 2009).
    - Adult chinook (O. *tshawytscha*) salmon likely have higher concentrations of PCBs than other Puget Sound species. They feed in Puget Sound for longer durations than other species and feed at a higher trophic level, increasing the likelihood of bioaccumulation.
    - Adult coho salmon (*O. kisutch*) also tend to feed in costal zones increasing the potential exposure to contaminated prey species.
    - Adult sockeye salmon (*O. nerka*), pink salmon (*O. gorbuscha*), and chum salmon (*O. keta*) generally do not reside in Puget Sound but instead are transitory and migrate to the open Pacific Ocean to feed where levels of contamination in prey species is lower.
- Out-migrating juvenile salmon spend time in river mouths and estuaries transitioning from fresh water to marine environments. Juvenile contaminant burdens are likely representative of estuary or nearshore conditions, depending on the location of catch (O'Neill et al. 2015).
- "Blackmouth" Chinook salmon do not migrate out of the Puget Sound. Unlike migrating species, the contaminant exposure is confined to Puget Sound and so Blackmouth contaminant levels are likely to be broadly representative of contaminant levels throughout the pelagic food web in the Puget Sound. O'Neill and West (2009) estimated one-third of Puget Sound Chinook salmon may remain as residents in Puget Sound. A number of salmon hatchery practices have been adopted (e.g., increased rearing times, delayed releases, and rearing in marine net pens) to encourage Chinook and coho salmon to remain as residents in Puget Sound, to support Puget Sound recreational fisheries

3.1.4 Mussels (*Mytilus* spp.)

Bay mussel (Mytilus trossulus). Photo: National Museum Wales.

Caged mussels have been added as a Vital Signs indicator species (reporting beginning in 2021-2022) to represent contamination in the marine nearshore environment. Shellfish are sessile filter feeders that process large volumes of water, and generally lack the metabolic processes for against many common contaminants, so they retain and accumulate chemicals of interest. The uptake of hydrophobic organic contaminants, including PCBs and PAHs mainly occurs as a passive diffusive/equilibrium partitioning process. The key sources for contaminant uptake in mussels are in dissolved-phase in ambient seawater and sorbed to food particles (Beyer et al. 2017). Most PAHs and PCBs are generally not metabolized in mussels and other shellfish, though benzo[a]pyrene specifically has been shown to undergo significant metabolism (Gewurtz et al. 2002). PBDEs have been shown to have a high bioaccumulation potential in mussels (Gustafsson et al. 1999). Tian et al. (2015) compared the ratios of BDE-47/99/153 in clam (*Scapharca subcrenata*) to the ratios in the surrounding waters and found that the proportion of BDE-47 increased, while BDE-99 and -153 decreased suggesting some metabolism.

Shellfish have been used to monitor concentrations of CECs in the marine environment in Puget Sound and elsewhere (Dodder et al. 2014, Maruya et al. 2014a, Maruya et al. 2014b, Granek et al. 2016, James et al. 2020). Due to the wide range of compounds included as CECs, caution should be used when interpreting the measured concentrations in shellfish with environmental occurrences and exposures. For example, the presence of some antibiotics may be the result of natural processes (Hodges et al. 2012), while other contaminants may be susceptible to active export process from the shellfish (Fernandez-Sanjuan et al. 2013).

WDFW have been performing regular monitoring studies with caged mussels, where bay mussels (*Mytilus trossulus*) from a common source are deployed at numerous locations throughout the Puget Sound and analyzed for a range of contaminant (Lanksbury et al. 2010, Lanksbury et al. 2013, Lanksbury et al. 2014, Lanksbury et al. 2017). These programs have been used to report on the status of PCBs, PAHs, PBDEs, and other compounds from throughout the Puget Sound nearshore.

3.2 Chemicals of Concern (PCBs, PAHs, PBDEs, and EDCs)

The Vital Sign was developed based on a suite of chemicals of concern including PCBs, PAHs, PBDEs, and EDCs. This section presents a brief overview of each chemical class and how those properties might affect data interpretation and impact a restoration strategy. Extensive and high quality reviews are available in the literature; e.g., PCBs (Longnecker et al. 1997, ATSDR 2000, Breivik et al. 2002, Borja et al. 2005), PDBEs (de Wit 2002, McDonald 2002, Sjodin et al. 2003, Birnbaum and Staskal 2004, Hites 2004, ATSDR 2017), PAHs (ATSDR 1995, Ravindra et al. 2008), and EDCs (Diamanti-Kandarakis et al. 2009, WHO/UNEP 2012).

3.2.1 Polychlorinated Biphenyls (PCBs)

Polychlorinated Biphenyls are a class of chemical compounds where 2–10 chlorine atoms are attached to a biphenyl molecule (Figure 3-1). There are 209 possible congeners based on the degree of chlorination and the location of the chlorines. PCBs were produced in mixtures, the most common of which were the Aroclors. The Aroclor mixtures were referred to by a 4-digit code: the first two numbers indicated type of mixture, the second two indicated the approximate weight percentage of chlorine in the mixture.

PCBs can also be formed during the production of certain products, including pigments and dyes. Some of the congeners produced as inadvertent by-products of manufacturing are different than the congener groups that were part of the legacy mixes, e.g., PCB-11, which may be a useful marker of the presence of PCBs from non-legacy sources (Hu and Hornbuckle 2010, Rodenburg et al. 2010b, Grossman 2013).

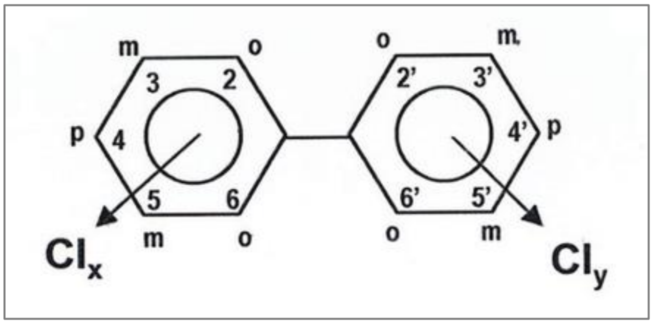

Figure 3-1. General structure of PCB molecule. The number and position of the chlorine molecules (x = 1-5, y = 0-5) results in 209 potential congeners. Location of chlorines is referred to either specifically by number (2-6 or 2'-6') or position (o – ortho, m – meta, p – para).

Relevant properties of PCBs are:

- PCB structure varies according to the degree and position of chlorination. PCBs with no chlorines on 2, 2', 6, and 6' (see Figure 3-1) tend to be planar (the benzene rings align in a single plane).
- PCB toxicity varies according to structure with the planar "dioxin-like" congeners presenting higher toxicity. These include PCB-077, 081, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189. EPA has recommended developing cleanup levels based on the occurrence of these dioxin-like congeners.
- PCBs are hydrophobic, with the more chlorinated molecules being more hydrophobic. All PCBs have reported log $K_{ow} > 5$ (meaning the concentrations are 10^5 higher in oil compared to water in mixed systems) indication that they will sorb strongly to organic particles and accumulate in fatty tissues (Han et al. 2006).
- PCBs are stable and degrade slowly in the environment. The estimated half-lives (the time it takes for environmental concentration to decrease by a factor of two) for PCB congeners to degrade in soils and sediments ranged from ~ 3 years (PCB-028) to ~ 38 years (PCB-180; Sinkkonen and Paasivirta (2000)).
- PCB congeners with more than two chlorines are susceptible to dechlorination by anaerobic bacteria where higher-chlorinated congeners are transformed into lower-chlorinated congeners. Dechlroination of congeners occurs preferentially in the *meta-* or *para-* positions over chlorines in *ortho-* position (Brown Jr. et al. 1987) with most congeners dechlorinated in the *meta-* position (Fagervold et al. 2007). Dechlorination results in an enrichment in low molecular weight *ortho*-substituted congeners, especially PCB-004 (2,2') and PCB-019 (2,2',6). Other congeners that have been reported as markers of dechlorination include PCB-025 (2,3',4), -026 (2,3',5), -032 (2,4',6), and -049 (2,2',4,5') (Rodenburg et al. 2010a).

3.2.1.1 PCBs Analytical

The purpose of this section is to highlight information that may be critical in data interpretation. The comparability of environmental PCB data depends on the laboratory method by which samples were analyzed. Care should be taken when comparing data across different studies or through time. It is often necessary to perform cross validation studies prior to making comparisons (e.g., West et al. (2017)). Additionally, not all methods detect or report on all congeners and so the lack of a particular congener may not necessarily mean that it is not present in the environment, but rather that the compound is not detectable by the given method. Three main methodologies have been used.

- PCB Aroclor analysis (e.g., SW846 Method 8082) estimates the quantity of mass of PCBs present based on presumed Aroclor signatures. This data is not suitable for homolog or congener analysis. For example, many of the PCBs produced as inadvertent by-products were not detected with these methods.
- Low resolution (LR) GC/MS methods (e.g., EPA 8270D) are suitable for identifying homologous groups of PCBs (i.e., groups with the same number of chlorines). The analytical laboratory at NOAA Northwest Fisheries Science Center utilizes a low resolution method to quantify forty-seven PCB congeners (Sloan et al. 2014). Many of the tissue samples analyzed in the Puget Sound were analyzed with this method.
- High resolution (HR) GC/MS methods (e.g., EPA 1668) can be used to determine the concentration of individual congeners in each sample. Some congeners that co-elute and have equal masses may not be resolvable.

3.2.2 PBDEs

Polybrominated diphenyl ethers are a class of brominated hydrocarbons where 2–10 bromine atoms are attached to the diphenyl ether molecule (Figure 3-2**Error! Reference source not found.**). Monobrominated diphenyl ethers are often included. There are 209 possible congeners, though not all congeners exist in commercial mixtures. PBDEs can be categorized by degree of bromination with homolog groups referring to all PBDEs with the same number of bromines. Commercial mixtures included penta- (five bromines per molecule), octa- (eight bromines per molecule), and deca-BDEs (ten bromines per molecule). Sales of penta- and octa-BDEs have been banned in parts of the United States since 2007.

The structure of the molecule varies according to the location and extent of the bromine substitution.

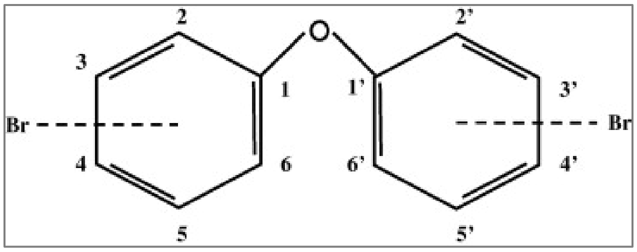

Figure 3-2. General structure of PBDE molecule. Each congener can contain from 2-10 bromine atoms. Bromine location can also be defined relative to the ether bond, with *ortho*- substitutions at 2,2',6,6'; *meta*- substitutions at 3,3',5,5'; and *para*- substitutions at 4,4'.

Relevant properties of PBDEs are:

- PBDEs are stable and degrade slowly in the environment. Estimated half-life (the time it takes for environmental concentration to decrease by a factor of two) is approximately 0.5 years in soils and 1.5 years in sediments (Palm et al. 2002). Debromination results in conversion of higher brominated BDEs to lower-brominated BDEs.
- PBDEs are hydrophobic and have a low solubility in water. They generally are associated with particles and can accumulate in fatty tissues. The log K_{ow} values range from 6.6 for BDE-47 (indicating the concentration of BDE-47 would be $10^{6.6}$ higher in oil compared to water in mixed systems) to 11.1 for BDE-209. The higher value indicates a higher affinity for organics/lipids compared to water.
- The lower-brominated BDEs are slightly volatile and so atmospheric transport may be a consideration (Palm et al 2002). Volatility decreases with extent of bromination.
- There are many considerations that should be taken when evaluating and comparing reported PBDE data, particularly across studies and institutions (Fulara and Czaplicka 2012). Not all analytical methods capture the same range of congeners and so reported total PBDE values may be based on dissimilar information. The analytical laboratory at NOAA Northwest Fisheries Science Center, for example, utilizes a low resolution method to quantify eleven PBDE congeners (Sloan et al. 2014). Many of the tissue samples analyzed in the Puget Sound were analyzed with this method.
- Hydroxylated-BDEs (OH-BDE) and methoxylated-BDEs (MeO-BDE) are structure analogs of PBDEs; neither were artificially synthesized or used in commercial products. OH-BDE are natural products found in the marine environment and may be formed via the metabolism of PBDEs or MeO-PBDEs. MeO-PBDEs appear to be solely natural in origin (Malmvärn et al. 2005).
- PBDE metabolism in fish varies according to congener and fish species. Metabolism generally occurs with congeners containing at least one *meta*-substituted bromine, resulting in the production of debrominated metabolites (Roberts et al. 2011), where the bromine on the *meta*-position is removed.
- The result of PBDE metabolism in fish may be an accumulation of BDE-049 (Dietrich et al. 2015).

3.2.3 PAHs

PAHs are a group of organic compounds containing only carbon and hydrogen that are composed of two or more combined aromatic rings. They are found in heavy hydrocarbons such as tar and creosote and formed during the incomplete combustion of wood and hydrocarbons. Common examples are shown in Figure 3-3. Fate and transport properties vary according to size and structure. The ATSDR has generally grouped them into low molecular weight PAH (2- or 3-ring) and high molecular weight (4- or more rings) for purposes of characterizing fate and transport (ATSDR 1995)

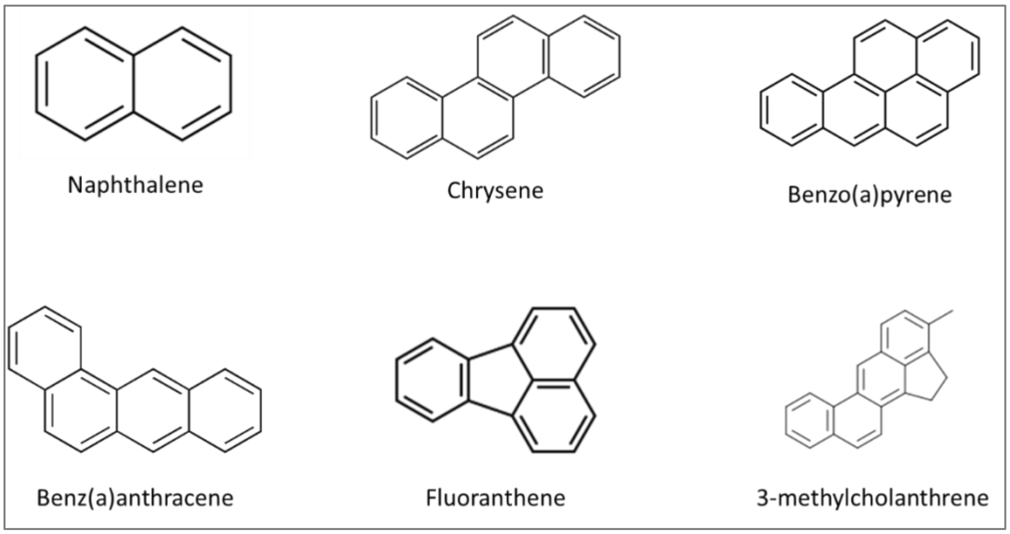

Figure 3-3. Selected PAH compounds.

Relevant properties of PAHs include:

- Overall, PAHs are lipophilic and have very low solubility in water though properties vary by compound. Lipophilicity tends to increase and solubility tends to decrease with number of aromatic rings. A brief summary of properties is shown in Table 3-1.
- PAHs tend to be associated with particles in the aquatic environment.
- In general, low molecular weight PAHs have higher vapor pressure and may volatilize. The higher molecular weight PAHs have low vapor pressures and do not readily volatilize;
- Some PAHs are susceptible to photodegradation and biodegradation in the water column. Persistence in the water column depends on specific compound and environmental conditions;
- PAHs may degrade in the sediments. The half-life of low molecular weight PAHs in sediments is generally on the order of days to weeks. The half-life of high molecular weight PAHs in sediments is on the order of months to years.
- PAHs are rapidly metabolized in fish and so do not bioaccumulate to high levels. Biliary fluorescent aromatic compound (FAC) levels, cytochrome P4501A induction, and DNA adducts are often used as measures of PAH exposure (Johnson et al. 2008a).

Table 3-1. Estimated properties of selected PAHs (ATSDR, 1995)

Compound	Log Kow	Solubility (mg/L at 25C)	Half-life in soils and sediments
Naphthalene	3.3	31	days
Phenanthrene	4.4	1.2	weeks
Chrysene	6.1	0.003	> 0.5 year
Benz(a)anthracene	5.6	0.01	> 0.5 year
Benzo(a)pyrene	6.1	0.002	> 0.5 year

3.2.4 Endocrine Disrupting Compounds and Contaminants of Emerging Concern

Endocrine disrupting compounds are not a single class or group but are classified based on their biological activities. They are defined as, “an exogenous substance or mixture that alters function(s) of the endocrine system and consequently causes adverse health effects in an intact organism, or its progeny, or (sub) populations” (WHO/UNEP 2012). Approximately 800 compounds have been identified as having the potential to affect human endocrine systems, and the majority of chemicals of commerce have not been tested. Since EDCs are such a wide-ranging class of compounds, with diverse physical and chemical properties, it is not possible to make generalizations about environmental behavior.

Contaminants of Emerging Concern applies to a larger group of compounds that are defined by their potential to cause environmental harm. They have been described as compounds that:

- Are known, or suspected to be present in the environment. Their occurrence patterns are being characterized based on advances in analytical instrumentation.
- They are occurring in the environment at levels which may cause harm.
- They are poorly regulated (Diamond et al. 2011).

Endocrine disrupting chemicals are a subset of Contaminants of Emerging Concern.

4 Occurrence of Contaminants in Salish Sea

This section presents a brief summary of regional studies that address the contaminants or species of interest, but are not otherwise included in the Vital Sign reporting. These studies provide additional information on the occurrence and impacts of priority chemicals in the region. The focus is on more recent publications.

4.1 PCB, PBDEs, and PAHs in marine fish

- Lawson et al. (2020) profiled organochlorine contaminant (including PCBs) in 98 resident and transient North Pacific killer whales (*Orcinus orca*) representing ten populations. The populations had distinct chemical "fingerprints" that allowed the inference of foraging habitats. Transients (mammal-eating) had significantly higher OC concentrations than residents (fish-eating), likely because the transient killer whales feed primarily at a higher trophic level than residents. Adult male whales had consistently higher OC levels compared to adult females, regardless of ecotype, suggesting that adult females offload organochlorine contaminants to calves.
  The PCB concentrations were compared to a variety of threshold values, representing different health endpoints. Of the 69 resident killer whales analyzed 62% had blubber ΣPCB levels that exceeded the threshold for immune suppression and vitamin A. All transient and 57% of the resident whales had PCB TEQ concentrations that exceeded the threshold of 209 pg/g lw (defined as the threshold for reproductive dysfunction in ringed seals).
  OC exposure in North Pacific killer whales varied based on ecotype, population and sex/maturity class and is an important consideration when assessing the relative risk of potential health impact from exposures. Overall, the exposures likely cause some health impacts; modeling suggests that such impacts may lead to severe population declines.

- O'Neill et al. (2020) surveyed PCB, PBDE, and DDT concentrations in juvenile Chinook along the Snohomish River. In order to discern the impacts of WWTP effluent, samples were collected in the upper mainstem (no WWTP impacts), lower mainstem (likely WWTP impacts), and distributary channels (some potential WWTP impacts); fish were grouped by natural- or hatchery-origin.
  For PCBs - 0%, 27%, and 29% of natural–origin salmon from the upper mainstem, lower mainstem, and distributary channels, respectively, had (lipid normalized) TPCB concentrations above an adverse critical body residue threshold. 0%, 14% and 0% of the hatchery-origin fish from each sampling group exceeded the threshold. For PDBEs - 73% and 14% of the natural-origin fish from the lower mainstem and the distributary channels had the sum of BDE-047 + BDE-099 ($\sum PBDE_{47,99}$) within the range of concentrations found to alter their immune response and increase disease susceptibility. In contrast, none of the natural-origin Chinook from the upper mainstem, or hatchery-origin had $\sum PBDE_{47,99}$ levels exceeding thresholds.
  Localized sources of PBDEs, in this case likely WWTP effluent discharges, may result in PBDE concentrations high enough to impact out-migrating juvenile Chinook.

- Chen et al. (2018) sampled out-migrating steelhead (*Oncorhynchus mykiss*) from four Puget Sound river systems for the parasite *Nanophyetus salmincola*; a subset were also analyzed for contaminants. The prevalence and parasite load of *N. salmincola* were significantly higher in fish from river systems in central (Green/Duwamish) and southern (Nisqually) Puget Sound than in fish from northern (Skagit) Puget Sound. PBDE concentrations associated with increased disease susceptibility ($\sum PBDE_{47,99}$ > 470 ng/g lw) were observed in 40%, 10%, and 0% of the steelhead sampled from river systems in southern, central, and northern Puget Sound, respectively. PCB levels exceeded threshold of 2400 ng/g lw in 25% of the samples from the central and southern regions of Puget Sound and 17% of samples from northern Puget Sound. Both *N. salmincola* and contaminant levels indicate the potential for reduced marine survival of steelhead from the Nisqually or Green/Duwamish compared to those from the Skagit.

- Conn et al. (2020) measured the concentrations of PCBs, PBDEs, PAHs and other contaminants in Pacific sand lance (*Ammodytes personatus*) from nine locations in Puget Sound. PCBs were detected in fish from all locations in this study ranging in total concentration ($\sum PCB_{209}$) from 2.4 to 42.9 ng/g ww. Lipid normalized concentrations were 1.5 to 25 times below a fish effects health threshold of 2400 ng/g lw, which is the threshold value used in Vital Sign reporting (Table 2-1). PBDEs were detected in sand lance samples from all locations at total PBDE concentration of 0.58 to 4.8 ng/g ww. Tissue concentration for $\sum PBDE_{47,99}$ (max 3.4 ng/g ww) were 3 to 10 times lower than reported effects levels for disease susceptibility and thyroid alteration. PAHs were detected in all samples analyzed with concentrations at one site (Eagle Harbor) being more than five times the levels reported elsewhere. Sand lance from Eagle Harbor were collected adjacent to a former creosote-wood treatment facility with known sediment contamination. Tissues were analyzed for alkylphenol ethoxylates; NP1EO and NP2EO were detected in all samples with the maximum concentration of 5.3 ng/g ww. Although this is lower than potential effects levels for endocrine disruption, the total mass of alkylphenol ethoxylates includes longer chain molecules that were not measured, and so overall exposures may be higher. Chlorinated parrafins were detected in about half of the samples analyzed. However, analytical uncertainties and ambiguous effects levels make it difficult to understand whether their presence might pose an environmental risk.
  Although risk levels to sand lance were generally below threshold values, the PCB concentrations were sufficient that they may biomagnify to levels that can cause adverse impacts on higher trophic level predators including salmon and killer whale.

- Good et al. (2014) analyzed PCB and PBDE concentrations in the six most common fish prey (Pacific sand lance, Pacific herring, northern anchovy, surf smelt, Chinook salmon, and chum salmon) of rhinoceros auklets breeding on Protection Island (eastern end of Strait of Juan de Fuca), Tatoosh Island (off NW tip of Olympic Peninsula), and Destruction Island (WA coast). The results suggested fish from the inland marine waters colony (Protection Island) were more contaminated compared with the fish from the outer coast colonies. Contaminant patterns for the three resident species with spawning populations in Puget Sound (Pacific sand lance, Pacific herring, surf smelt) were quite similar. The patterns were less similar for the three species that range more widely (northern anchovy, Chinook salmon, and chum salmon). For juvenile

Chinook: 30% of fish from protection island exceeded effects threshold for PCBs and 20% exceeded effects thresholds for PBDEs. Elevated PCB and PBDE concentrations in Chinook from the outer coast suggest that they could be from Columbia River stocks. The long-term presence of rhinoceros auklets with areas with prey contaminant profiles similar to those of Protection Island could lead to concentrations known to affect behavior, reproduction, and immune system function seen in other marine birds; it is currently not know, however, if the rhinoceros auklets remain in the area after breeding.

- Cade et al. (2018) reported on the presence of PBDEs and derivatives in seafood obtained from Puget Sound. The shellfish - manilla clams (*Venerupis philippinarum*) and blue mussels (*Mytilus edulis*) – and the English sole were locally sourced from within the Puget Sound. The highest concentration of PBDEs of the fin fish was measured in English sole with average ∑PBDE = 3680 pg/g ww. BDE-47 was the most abundant congener. The ∑
- -BDEs and ∑MeO-BDEs was 117 and 115 pg/g ww, respectively. The concentration of PDBEs in the shellfish ranged from 190 – 520 pg/g ww. The shellfish had a much higher proportion of OH- and MeO-PBDEs compared to the fin fish samples with ∑OH-BDEs = 2005 pg/g ww and ∑MeO-BDEs = 570 pg/g ww.

4.2 Exposure in non-indicator species

Persistent organic pollutants (POP) such as PCBs and PBDEs build up in the food chain, so that higher trophic level predators are the most highly contaminated. Through these trophic interactions, additional species in the food web are vulnerable to contamination and associated lethal and sublethal effects.

- West et al. (2011) - Krill (*Euphausia pacifica* and *Thysanoessa spp*), and particulate organic matter/phytoplankton (POM; material captured by filtering seawater through 20-µm mesh) were sampled and analyzed for toxic contaminants including PCBs, PBDEs, PAHs, and organochlorine pesticides (OCPs). High contaminant levels in both POM and krill were associated with urbanized embayments, suggesting these areas are where POPs enter the pelagic food chain. Overall, PAHs were detected more often and in greater concentration than all other POPs. Aside from Elliott Bay the next greatest PAH concentrations in POM were observed near to shore, and near to obvious PAH sources e.g., marinas and ferry terminals, even in otherwise relatively undeveloped basins.
- Frouin et al. (2013) - PCBs and PBDEs were measured in water (dissolved and particle-bound) and plankton samples collected from the Strait of Georgia. Total PCB concentrations ranged from 52.2 – 364 ng/g lw; total PBDE concentrations ranged from 77.2 – 201 ng/g lw. Unique congener profiles were present in the plankton, dissolved fraction of water, and solids fraction of water. Results provide insight into the biological availability of PCBs and PBDEs to the Strait of Georgia food web.
- In killer whales, total 2,3,7,8-TCDD Toxic Equivalents (TEQ) in most killer whales sampled easily surpassed adverse effects levels established for harbor seals, suggesting that the majority of free-ranging killer whales in the Salish Sea are at risk for toxic effects. The southern resident and transient killer whales of British Columbia can now be considered among the most

contaminated cetaceans in the world (Ross et al. 2000, Ross 2006, Krahn et al. 2007, Krahn et al. 2009, Ross et al. 2009).

- Harbor seal (Phoca vitulina) blubber biopsies were analyzed from locations within Salish Sea across a range of industrialization (e.g., Queen Charlotte Strait, BC, Strait of Georgia, BC, and Puget Sound, WA. Harbor seals in Puget Sound were heavily contaminated with PCBs. Total toxic equivalents to 2,3,7,8-TCDD (TEQ) suggest that Puget Sound seals are at greatest risk for adverse health effects, and that PCBs represent the class of dioxin like contaminants of greatest concern at all sites. Time trends indicate that PBDE concentrations doubled every 3.1 years between 1984 and 2003, but appeared to decline thereafter. Between 1984 and 2003, PCB concentrations declined by 81%. Overall, results suggest that regulations and source controls have reduced inputs of these contaminants to the Salish Sea, consequently reducing the associated health risks to marine wildlife (Ross et al. 2004, Cullon et al. 2005, Ross et al. 2013).
- It is thought that the accumulation of toxics in orcas, derived largely from salmon, a main component of the resident orca diet, is responsible for some calf loss (Ross et al. 2000).

4.3 CECs and EDCs in Puget Sound

Evaluating for exposure and impacts of CECs and EDCs in the environment is based on several lines of evidence. The first is to evaluate for the occurrence of individual chemical in water and sediments, which are in indicator of exposures. Biological samples are also analyzed and these provide a more direct measure of exposures, though metabolic processes, transport within and across different biological compartments, and the analytical challenges of analyzing biological matrices (which is harder than analyzing water or soil) provides additional challenges in data collection and interpretation.

Biomarkers are also used to provide a measure of exposures to chemicals based on known or presumed modes of action. Vitellogenin (Vtg) is amongst the most common biomarker measured in Puget Sound fish. Additional biomarkers are being developed that may provide information on exposures across a wider range of modes of action associated with CEC exposures (Meador et al. 2020).

An additional line of evidence for exposures to EDCs in observations related to altered reproductive timing. As described in Johnson et al. (2008b), the developmental stage of ovaries or testes are examined and classified. Fish are also examined to detect intersex fish. These data for each individual site are compared against the data for fish from the least developed sites, which are taken as an unexposed reference group. Statistically significant differences are used to identify sites with altered reproductive timing.

4.3.1 CECs and EDCs in the Environment

There have been a limited but increasing number of studies evaluating the occurrence and impacts of CECs and EDCs in the marine environment. These are summarized below.

Tian et al. (2020) used high resolution mass spectrometry based method to perform a survey of CECs at 18 sampling locations across the Puget Sound. They identified 87 non-polymeric CECs including pharmaceuticals, herbicides, vehicle-related compounds, plasticizers, and flame retardants. Occurrence concentrations were compared to Predicted No Effects Concentrations (PNECs) and eight CECs were identified with a risk quotient >1, indicating the potential to cause environmental harm. Compounds included perfluorooctanesulfonic acid, bisphenol-S, 2-mercaptobenzothiazole, venlafaxine, fluridone, metsulfuronmethyl, diisononyl phthalate, and hexa(methoxymethyl)melamine.

Meador et al. (2016) sampled juvenile Chinook salmon and Pacific staghorn sculpin (*Leptocottus armatus*) and water from Sinclair Inlet, the Puyallup river estuary, and the Nisqually estuary and analyzed samples for a suite of 150 compounds. Six compounds including sertraline, triclosan, estrone, fluoxetine, metformin, and nonylphenol were detected in water and tissue at concentrations that may cause adverse effects in fish.

A 2012 survey consisting of approximately 50 samples collected throughout Puget Sound indicated the presence of selected pharmaceuticals and personal care products (many of which were EDCs) in nearly every sample (Miler-Schultze et al. 2017). Subsequent samples of surface water runoff entering Puget Sound contained ibuprofen above published risk threshold.

Keil et al. (2011) sampled 22 stations in Barkley Sound, Vancouver Island and 66 stations in Puget Sound and analyzed for a suite of 37 compounds. Fifteen xenobiotics were detected with diethylhexyl phthalate, ethyl vanillin, and benzaldehyde being the most abundant. Concentration were generally 1-2 orders of magnitude higher in Puget Sound compared to Barkley Sound.

Krogh et al. (2017) analyzed wastewater, sea water, sediments, and mussels near two wastewater discharges from Victoria, B.C. for 47 PPCPs; the wastewater from these facilities only undergoes minimal, primary treatment prior to discharge. The concentrations of PPCPs near the outfalls was generally higher than reported elsewhere (Long et al. 2013) but decrease to background within 800 m of outfall. PPCPs in seawater were similar to reported elsewhere in Puget Sound (Meador et al. 2016)

Long et al. (2013) analyzed for 119 pharmaceuticals and personal care products and 13 perfluoroalkyl substances (PFASs) in marine sediments from Puget Sound and Bellingham Bay. Only 14 of the 119 PPCPs and 3 of 13 PFASs were detected. Diphenhydramine was in 87.5% of samples with a maximum concentration of 4.8 ng/g dry weight. Triclocarban was detected in 35.0% of the samples, with a maximum concentration of 16.6 ng/g dry weight.

Persistence and accumulation of EDCs vary according to chemical properties and environmental conditions, e.g., the EDCs bisphenol A, 17β-estradiol, 17α-ethynylestradiol, 4-tert-octyl phenol, and 4-n-nonyl phenol were all found to degrade relatively rapidly in marine water and sediments under aerobic conditions but persisted under anoxic conditions (Ying and Kookana 2003).

4.3.2 Vitellogenin in marine fish

In addition to monitoring for specific chemicals in tissues and water, the xenoestrogenic impacts from EDC exposures has been observed in the region. One of the most common methods for measuring the impacts of EDC exposure is by measuring vitellogenin (Vtg), a protein for the production of egg yolks in reproductive females, in liver or blood (Sumpter and Jobling 1995). It is generally absent from males and so its presence is an established biomarker of (xeno-) estrogen exposures.

Johnson et al. (2008b) surveyed English sole from throughout Puget Sound and found male fish from several urban sites, particularly in Elliott Bay along the Seattle Waterfront, had significantly elevated levels of VTG. At the Elliot Bay sites the reproductive timing appeared altered. In a follow up, da Silva et al. (2013) developed an analytical method for detecting bisphenol-A (BPA), three naturally produced steroidal estrogens; estrone (E1), 17β-estradiol (E2) , 17 β-estriol (E3), and the synthetic estrogen, 17α-ethinylestradiol in fish bile. The method was applied no male English sole collected from urban and non-urban sites from Puget Sound. BPA and E2 were significantly higher at fish from urban sites compared to the non-urban sites.

O'Neill et al. (2016) reported on a survey of EDCs and Vtg gene expression in English sole from ten index site throughout Puget Sound. Samples were analyzed for the suite of EDCs as described in da Silva et al. (2013) in addition to three selective serotonin reuptake inhibitors (SSRIs; fluoxetine, sertraline and citalopram). Vtg was not directly measured in plasma but rather the Vtg messenger Ribonucleic Acid (mRNA) levels were quantified in liver by Reverse Transcriptase quantitative Polymerase Chain Reaction (RT-qPCR). This biomarker focusing on gene expression is reportedly more sensitive, and exposure-response times are more rapid compared to changes in levels of Vtg in plasma (Arukwe et al. 2001). Vtg mRNA may also be more transient compared to Vtg protein, meaning in may not last long after exposures are removed. In a controlled exposure study with fathead minnow, liver Vtg mRNA was detected within 4 h of exposure and returned to normal levels in about 6 d, while plasma Vtg protein was detectable within 16 h of exposure and remained near maximum levels for at least 18 d (Korte et al. 2000). Therefore, Vtg mRNA expression provides a very recent history of potential exposure (i.e., days) whereas plasma Vtg protein levels change more slowly and can provide a broader window of past exposure (i.e., weeks) maximizing the ability to detect elevated Vtg levels and recent exposure to estrogenic chemicals.

Overall, the results indicated: altered reproductive timing in female fish from Seattle Waterfront in Elliott Bay; widespread Vtg induction in male sole, and; little or no recent exposure of English sole to SSRIs. However, Vtg gene expression in male English sole was not significantly correlated with any of the EDCs or total E2-equivalence (EEQ) potentially due to individual variability, differences between exposures and responses, or that the causative chemicals were not quantified.

The complexities of interpreting the relationships between EDC exposures and Vtg mRNA responses in field studies that were highlighted in O'Neill et al. (2016), suggest that Vtg mRNA may not be the most suitable biomarker for evaluating EDC exposure and impacts in the Salish Sea. As such, beginning with the 2017 survey, WDFW has focused on validating and applying measures of Vtg protein in plasma in English sole based on the ELISA method. Results of this work are forthcoming

Note: Add graphic summarizing 2017 sampling results of Vtg in English sole. In process with WDFW.

4.3.3 Other biomarkers in Marine Fish

In addition to the work investigating the presence of Vtg in marine fish, the responses of other biomarkers of chemical exposure have been investigated. Meador et al. (2020) compared the metabolomic profiles of juvenile Chinook salmon collected from an upstream hatchery, two estuaries that receive WWTP effluent, and one reference estuary that does not directly receive effluent. The metabolome patterns for fish from the two WWTP-receiving estuaries were more similar to each other compared to the reference site; the metabolome the reference site and the hatchery upstream were not different. The juvenile Chinook salmon in the impacted estuaries may have been experiencing metabolic disruption without any overt signs of impairment. These results indicate the potential for health impacts of juvenile Chinook salmon and the reduction of likelihood for a successful life cycle when exposed to effluent-related chemicals.

4.4 Mussel Tissue Monitoring

Although the Toxics in Fish Vital Sign indicators currently do not include information or report on mussel tissue monitoring, it is expected that the indicator species will be expanded in 2021 to include mussels. The information reported herein should serve as the basis for initial reporting.

Mussel tissue monitoring in the Puget Sound has evolved over the course of several studies and surveys lead by various organizations. From 1986 to 2012, NOAA's National Status and Trends Mussel Watch Program (MWP) tracked chemical and biological contaminant trends in naturally occurring mussels in Puget Sound.

A 2009/2010 pilot project aimed to adapt NOAA's large-scale program to address nearshore toxics monitoring and needs at the Puget Sound scale. The Washington Department of Fish and Wildlife's (WDFW) Toxics Biological Observation System (TBiOS) team (then called Puget Sound Assessment and Monitoring Program – PSAMP, Toxics in Fish) coordinated with NOAA's Mussel Watch team, Snohomish County Marine Resources Committee, Snohomish Public Works-Surface Water Management, and Washington Sea Grant to sample naturally occurring mussels from throughout the Puget Sound to explore the feasibility of using the program for regional toxics monitoring.

In 2011, NOAA's Mussel Watch Program experienced a funding shortfall, causing a break in their long-term data collection. However, WDFW's TBiOS team continued to partner with NOAA's Mussel Watch Program and conducted wild mussel sampling at historic sites in the winter of 2011/2012.

Further adaptation of the National Mussel Watch program to address Puget Sound goals lead to the 2012/2013 Mussel Watch Pilot Expansion (MWPE) study. This large-scale synoptic survey was the first formal Puget Sound-wide program implemented where caged mussels from a common aquaculture source (Penn Cove Shellfish) were deployed in predator-exclusion cages throughout Puget Sound, expanding monitoring to 108 locations. In tandem with the MWPE, the Tacoma Pierce County Health Department (TPCHD) conducted a complementary gradient study to help define the length of shoreline that represents a "site" for mussel contamination sampling and to measure impacts of land-use on nearshore biota.

Subsequently, WDFW's TBiOS team was asked to manage the nearshore mussel monitoring efforts under the new Stormwater Action Monitoring (SAM) program. In the winter of 2015/2016 the WDFW and regional partners performed the first SAM mussel monitoring survey, focusing on characterizing the spatial extent of tissue contamination in mussels residing inside designated urban growth areas (UGAs) of Puget Sound. Caged mussels from a common aquaculture source were deployed at 73 locations, which included 40 SAM sites and 33 partner sites. In 2016, tissues from a subset of the monitoring sites were analyzed for a suite of 330 trace organic contaminants (i.e., CECs) in order to provide a regional screening in the Puget Sound nearshore. Finally, in 2017/2018, the second SAM mussel monitoring survey was undertaken by WDFW and regional partners.

The results of the most recent investigations are summarized below. A summary of the mussel tissue monitoring programs implemented in Puget Sound is shown in Table 4-1. Links to the complete study reports are included in the notes.

Table 4-1. Summary of mussel tissue monitoring programs in the Puget Sound. Links to the project reports are included in the table notes. Complete data sets for some of studies are available from the Ecology Environmental Information Management systems, using the study IDs that are provided.

Study	Sites[8]	Analysis	EIM Study ID
2009/10 Mussel Watch Pilot Project[1]	20	NA	
Mussel Watch Phase I 2011/2012[2]	23		
Mussel Watch Pilot Expansion (MWPE) 2012/2013[3]	108 (105)	PAHs PCBs PBDEs Organochlorine Pesticides Metals (Pb, Cu, Zn, Hg, As, Cd)	WDFW 11-1916
Mussel Watch Gradient Project Sub-Project of the MWPE Study 2012/13[4]	2	PAHs PCBs PBDEs Organochlorine Pesticides Metals (Pb, Cu, Zn, Hg, As, Cd)	
Stormwater Action Monitoring 2015/16 Mussel Monitoring Survey[5,6]	73 (66)	PAHs PCBs PBDEs Organochlorine Pesticides Metals (Pb, Cu, Zn, Hg, As, Cd)	SAM_MNM RSMP_PC_PMNM2015
Pilot Evaluation of CECs in Mussel Tissues[7]	18	330 organic contaminants (CECs)	
Stormwater Action Monitoring 2017/18 Mussel Monitoring Survey	94	PAHs PCB PBDE Organochlorine Pesticides Metals (Hg, As, Cd, Pb, Zn, Cu, Al)	SAM_MNM SAM_PC_MNM2017

1. https://wdfw.wa.gov/publications/01127
2. https://wdfw.wa.gov/publications/01381
3. https://wdfw.wa.gov/publications/01643
4. https://ecology.wa.gov/DOE/files/a3/a37bd9fe-2d67-4733-bc9a-0547675b02bf.pdf
5. https://wdfw.wa.gov/publications/01925
6. https://ecology.wa.gov/Regulations-Permits/Reporting-requirements/Stormwater-monitoring/Stormwater-Action-Monitoring/SAM-status-and-trends/Puget-Sound-nearshore
7. James et al. (2020)
8. Numbers in parentheses represent the number of sites where mussel cages were successfully recovered

4.4.1 Stormwater Action Monitoring 2015/2016 Mussel Monitoring Survey

The objective of this survey was to characterize contaminants in nearshore biota from inside Puget Sound UGAs. A complete presentation of the data is included in Lanksbury et al. (2017); summary information is provided here. The complete data are available on the Ecology Environmental

Information Management (EIM) website under study ID: SAM_MNM. Data from Pierce County are under study ID: RSMP_PC_PMNM2015.

- PCBs were detected in samples from all sites at concentrations greater than the baseline. There were significant differences in PCB concentrations between baseline, unincorporated – UGA, and city-UGA sites.
- PAHs were detected in samples from all sites at concentrations above the baseline. There were significant differences in PAH concentrations between baseline, unincorporated – UGA, and city-UGA sites.
- PBDEs were detected in 82% of samples at concentrations greater than in the baseline. There were significant differences in PBDE concentrations between baseline, unincorporated – UGA, and city-UGA sites.
- DDTs were detected at 86% of the sites; they were not present above the limit of quantitation at the baseline site. There were significant differences in DDT concentrations between baseline, unincorporated – UGA, and city-UGA sites.
- PCBs, PAHs, PBDEs and DDTs, were correlated with the urbanization and percent impervious surface.
- Zinc, copper, cadmium, arsenic, and mercury were detected in samples from every study site, and lead detected at 86% of the sites. Arsenic, cadmium, copper, lead, mercury, and zinc were present at a few sites at concentrations greater than the baseline sites. There were no significant differences between any of the UGA groups and the baseline sites.
- Zinc was weakly correlated to urbanization; the other metals concentration did not show a clear spatial pattern.

4.4.2 Pilot Evaluation of CECs in Mussel Tissues

As described in James et al. (2020), a subset of tissues from the 2012/2013 Mussel Watch sites were analyzed for a suite of 231 pharmaceuticals and personal care products, 13 perfluorinated compounds, 4 alkylphenols, and 82 pesticides per existing methods. Thirty unique analytes from 11 chemical classes were measured at concentrations above the reporting limits. Three compounds, 4-nonylphenol (4-NP), virginiamycin M1, and sertraline were detected at all 18 locations. The chemotherapy drug melphalan was present at half of the sites at levels of concern. Results of a hydrodynamic simulation was performed to understand the distribution of combined effluent from 99 wastewater treatment facilities; modeled results were consistent with measured concentrations and demonstrated the wide-spread distribution of WWTP effluent throughout the Puget Sound.

4.4.3 Stormwater Action Monitoring 2017/2018 Mussel Monitoring Survey

The primary objective of this survey was to characterize the spatial extent of contamination in nearshore biota within delineated Urban Growth Areas. Thirty eight of the 2015/2016 SAM sites were revisited in 2017/2018, and three new SAM sites were added to replace failed 2015/16 sites (41 SAM sites total).

Additionally, forty four partner sites were included in this survey. A complete presentation of the data is included in Langness et al. (2020); summary information is provided here.

Graphical presentations of PCB, PBDE, and PAH results are shown in Figure 4-1, Figure 4-2, Figure 4-3, and Figure 4-4. It should be noted that the data are presented relative to the constituent concentration measured in previous monitoring campaigns, grouped as the lowest quartile (<25th percentile; green), the middle quartiles (25th percentile to 75th percentile; yellow), and highest quartile (>75th percentile; red).

- A summary of sampling results for PCBs is shown in Figure 4-1.
    - Total PCB[1] concentrations were higher than the baseline mean concentration (i.e., concentration in undeployed mussel tissues) at 88 of 92 sites; some sites in Hood Canal and Willapa Bay were below the baseline value.
    - The median total PCB concentration was higher at the SAM sites in the 2017/2018 study compared to the 2015/2016 study.
    - PCB congeners -118,-138, and -153 were present above the Limit of Quantification (LoQ) at all sites. No other congeners were present at all sites.
    - Sites with the highest concentrations (95th percentile) of PCBs were located in the urbanized south-central Puget Sound basin, while sites with low concentrations were mainly in the remote/least developed Hood Canal basin.
    - There was a significant positive correlation between the concentration of PCBs in mussels and the mean percent of impervious surface in adjacent watersheds.
- A summary of sampling results for PAHs is shown in Figure 4-2.
    - Total PAHs[1] were detected at concentrations above the baseline mean tissue concentration at all sites.
    - The median $\sum_{42} PAH$ concentration was higher at the SAM sites in 2017/2018 compared to the 2015/2016, though the differences were not statistically significant.
    - Benzo[a]pyrene was present above the LoQ at 40 sites sampled in the 2017/2018 study.
    - Mussels from a few SAM sites had PAH concentrations much higher that typically observed in Puget Sound UGAs, suggesting localized point sources.
    - Sites with the highest concentrations (95th percentile) of PAHs were located in the urbanized south-central Puget Sound basin, while sites with low concentrations were mainly in the remote/least developed Hood Canal basin.
    - There was a significant positive correlation between the concentration of PAHs in mussels and the mean percent of impervious surface in adjacent watersheds.

[1] Total PCB concentration were represented by the sum of 17 congeners, then multiplied by a factor of two. Total PBDE concentrations were represented by 11 congeners in the method ($\sum_{11} PBDE$). Total PAH is the sum of 42 PAHs included in the analytical method ($\sum_{42} PAH$). Total DDT is the sum of 6 congeners in the analytical method ($\sum_6$DDT).

- A summary of sampling results for PBDEs is shown in Figure 4-3.
    - Total PBDEs[1] were detected at 86 of 92 sites at concentrations above the baseline mean tissue concentration.
    - PBDEs were not present above the LoQ at six sites.
    - The median $\sum_{11} PBDE$ concentration was lower at the SAM sites from the 2017/2018 study compared to the 2015/2016 study, though the overall differences were not statistically significant.
    - PBDE congeners -28, -66, -85, -155 and -183 were not present above the LoQ at of the sampling locations.
    - Mussels from a few SAM sites had PBDE concentrations much higher that typically observed in Puget Sound UGAs, suggesting localized point sources.
    - Sites with the highest concentrations (95th percentile) of PBDEs were located in the urbanized south-central Puget Sound basin, while sites with low concentrations were mainly in the remote/least developed Hood Canal basin.
    - There was a significant positive correlation between the concentration of PBDEs in mussels and the mean percent of impervious surface in adjacent watersheds.
- A summary of sampling results for DDTs is shown in Figure 4-4.
    - DDTs were detected at 88 of 92 sites at concentrations above the baseline mean tissue concentration.
    - The median $\sum_{6} DDT$ concentration was higher at the SAM sites in 2017/18 study compared to the 2015/16 study.
    - Sites with the highest concentrations (95th percentile) of DDTs were located in the urbanized south-central Puget Sound basin, while sites with low concentrations were mainly in the remote/least developed Hood Canal basin.
    - There was a significant positive correlation between the concentration of DDTs in mussels and the mean percent of impervious surface in adjacent watersheds.

- A suite of metals (Al, As, Cd, Cu, Pb, Hg, and Zn) were evaluated in mussel tissue samples from selected sites in this study.
    - All metals were present above the LoQ at all sites sampled.
    - Sites with the highest concentrations (95^{th} percentile) of metals were located in the urbanized south-central Puget Sound basin, mainly in the Commencement Bay area.
    - There were significant positive correlations between the concentration of lead and zinc in mussels and the mean percent of impervious surface in adjacent watersheds.

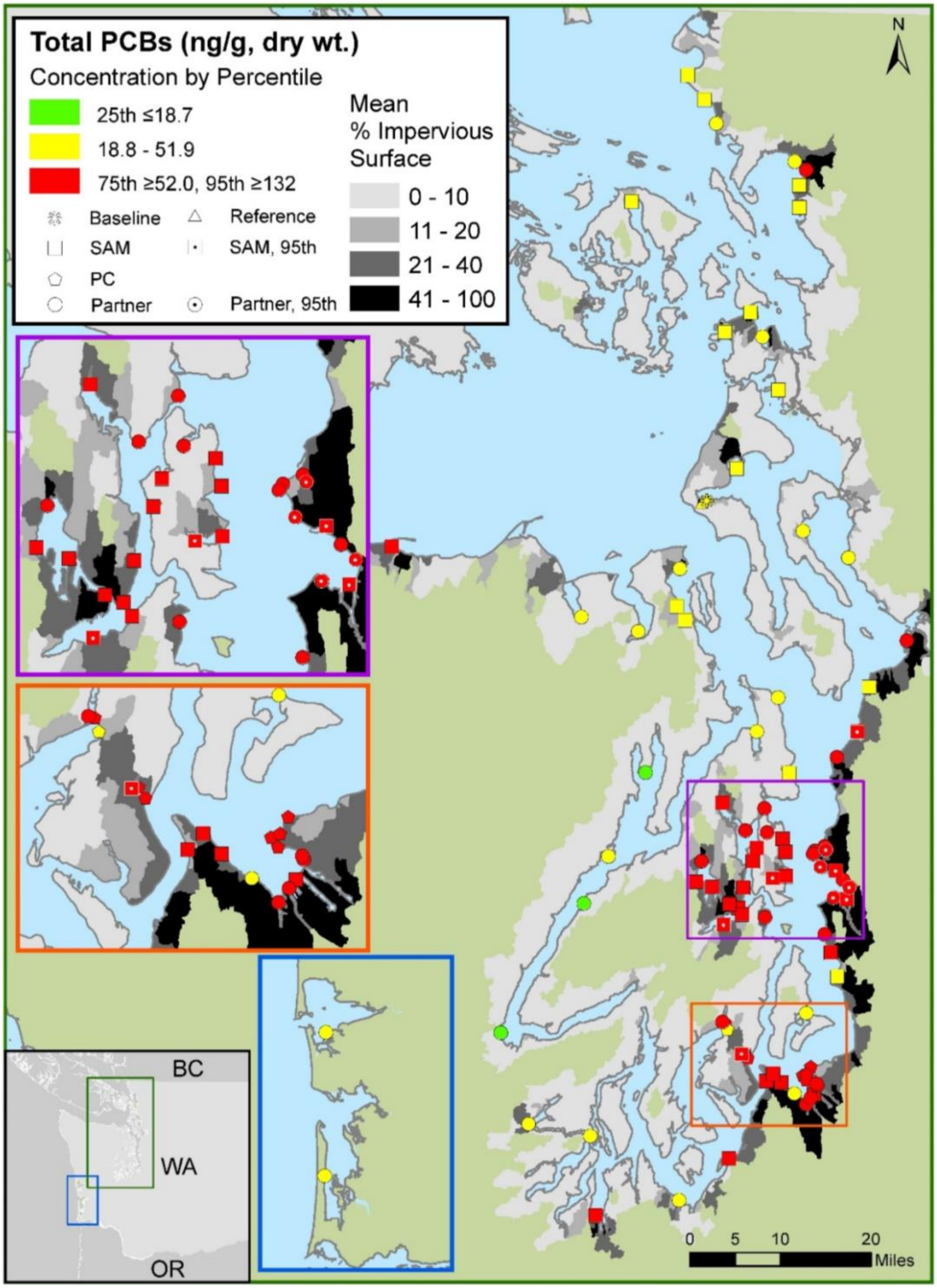

Figure 4-1. Total PCB concentration for all mussel sites in 2017/2018 sampling campaign. The colors are based on the concentration percentiles from mussel monitoring data collected during 2012/13 Mussel Watch Pilot Expansion study and the 2015/16 SAM Mussel Monitoring survey. Green is the lowest quartile (<25th percentile), yellow for the middle quartiles (25th percentile to 75th percentile), and red for the highest quartile (>75th percentile).Figure from Langness et al. (2020).

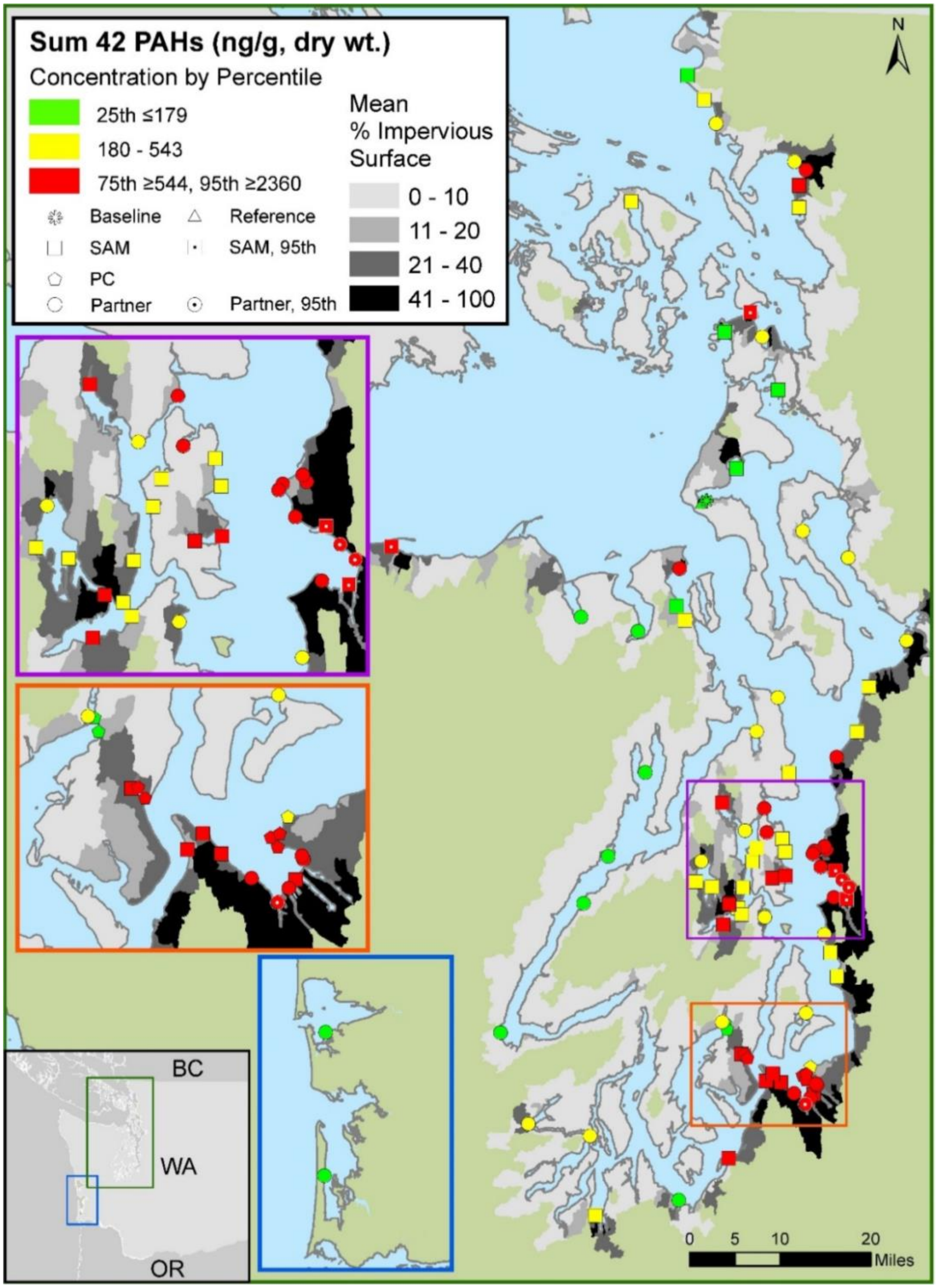

Figure 4-2. PAH concentration for all mussel sites in 2017/2018 sampling campaign. The colors are based on the concentration percentiles from mussel monitoring data collected during 2012/13 Mussel Watch Pilot Expansion study and the 2015/16 SAM Mussel Monitoring survey. Green is the lowest quartile (<25th percentile), yellow for the middle quartiles (25th percentile to 75th percentile), and red for the highest quartile (>75th percentile). Figure from (Langness et al. 2020)

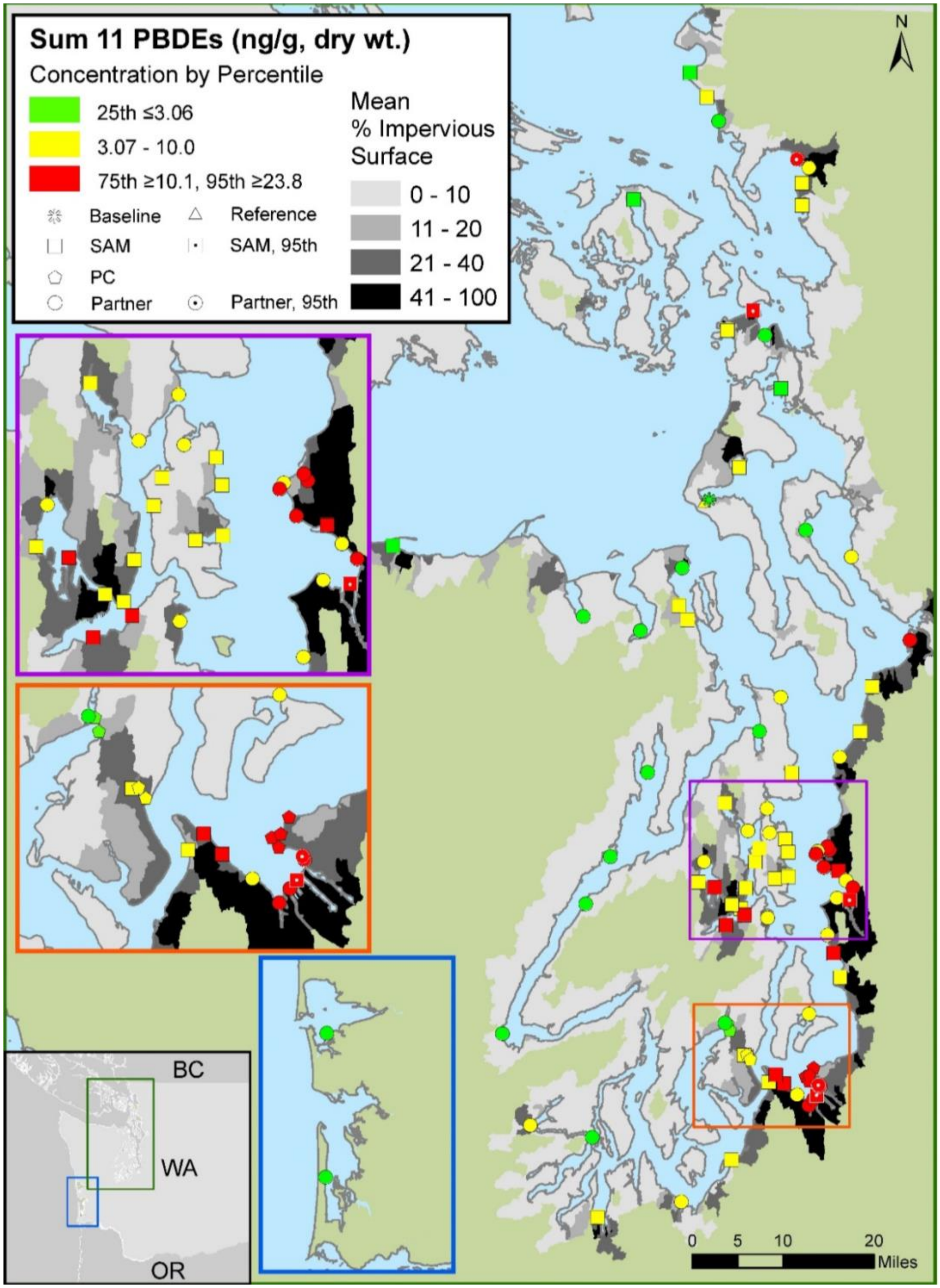

Figure 4-3. PBDE concentration for all mussel sites in 2017/2018 sampling campaign. The colors are based on the concentration percentiles from mussel monitoring data collected during 2012/13 Mussel Watch Pilot Expansion study and the 2015/16 SAM Mussel Monitoring survey. Green is the lowest quartile (<25th percentile), yellow for the middle quartiles (25th percentile to 75th percentile), and red for the highest quartile (>75th percentile). Figure from (Langness et al. 2020)

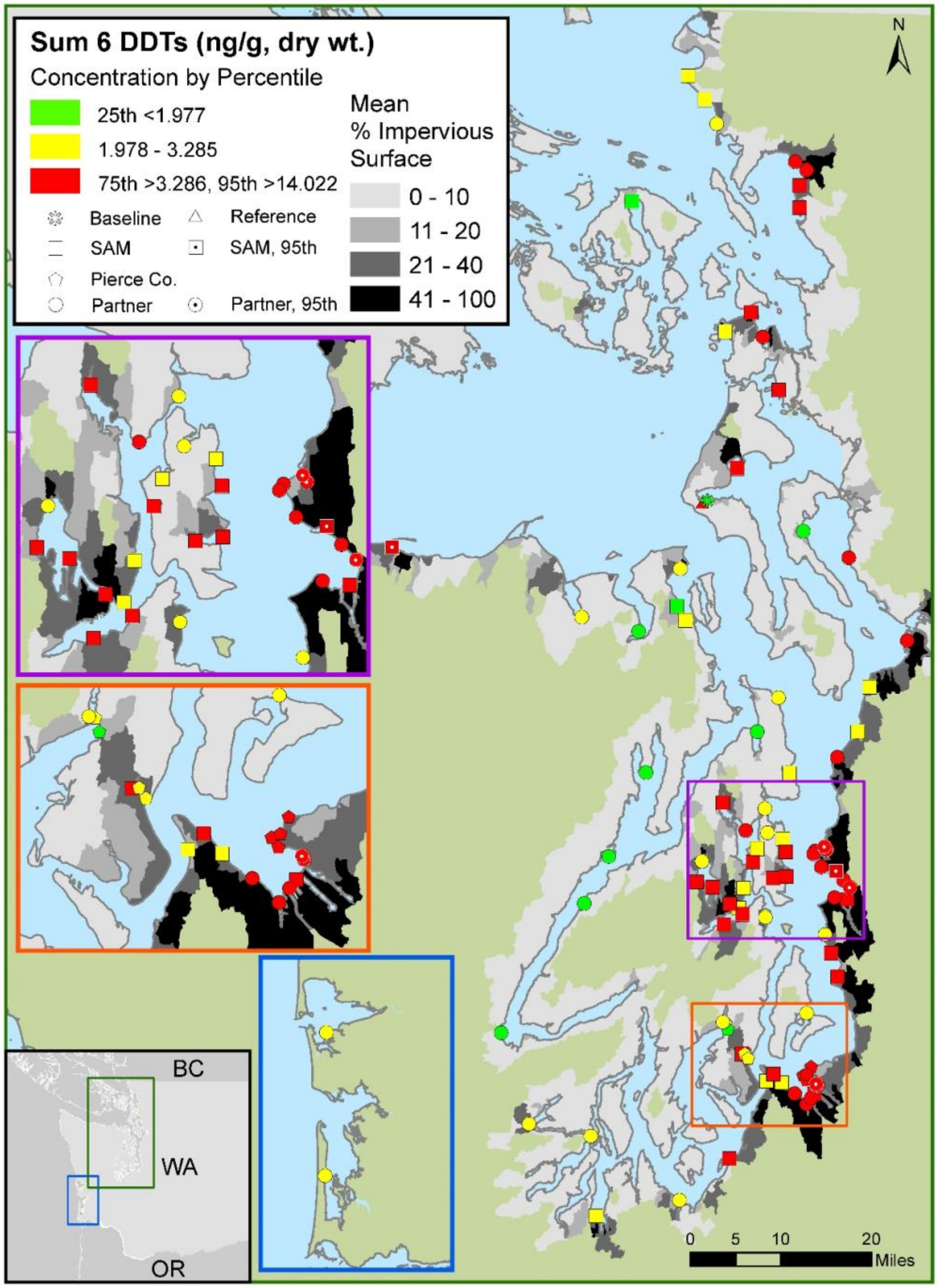

Figure 4-4. DDT concentration for all mussel sites in 2017/2018 sampling campaign. The colors are based on the concentration percentiles from mussel monitoring data collected during 2012/13 Mussel Watch Pilot Expansion study and the 2015/16 SAM Mussel Monitoring survey. Green is the lowest quartile (<25th percentile), yellow for the middle quartiles (25th percentile to 75th percentile), and red for the highest quartile (>75th percentile). Figure from (Langness et al. 2020)

4.4.4 Mussel Tissue Status vs Threshold Values

The Toxics in Fish Vital Signs reporting approach compares the levels of measured contaminants in tissues against threshold values determined to be protective of human health, where there may be a risk from consumption, or to animal health. Existing threshold values are described in Table 2-1. A discussion on thresholds is included in Section 5.1. A discussion on fish consumption rates, which are critical in evaluating risks and, thus, establishing threshold values, is included in Section 6.5.

There are not currently threshold values for contaminants in mussel tissues. However, since mussels are eaten, it is reasonable to utilize the existing human health threshold values for specific contaminants in order to gain a preliminary understanding of potential risk. There are caveats:

- The mussel tissues that are characterized in this study are caged mussels from a common aquaculture source and deployed at a variety of locations throughout the Puget Sound. The measured tissue concentration may not represent the range of concentrations from wild mussels.
- The mussels are deployed during the winter months and this may, or may not represent the tissue concentrations during other times of the calendar year.
- Considerations such as metabolism and depuration may affect chemical concentrations in wild mussels, compared to the values reported here.

The following should be considered a preliminary evaluation of tissue concentrations versus threshold and are presented to inform a more detailed future evaluation.

4.4.4.1 PCBs in mussel tissues vs threshold values

Total PCBs were compared with the fish consumption advisory threshold for total PCBs of 8 ng/g ww for high consumers, which is consistent with the value in current Vital Sign reporting (Table 2-1). Based on the data reported for the 2015/2016 study, 36% of the sites had concentrations that exceeded this value.

For the 2017/2018 study, total PCBs were determined based on the reported concentration of 41 congeners included in the analytical method. For congeners that were reported below the LoQ (i.e., U flag) a random value between 0 and the LoQ was used as the concentration value. Based on this approach, total PCBs exceeded the human health related threshold value of 8 ng/g ww at 37 sites (42%).

4.4.4.2 PAHs in mussel tissues vs threshold values

PAHs have not been reported against tissue levels in current vital sign reporting because fish readily metabolize PAHs and so the expected accumulation of these contaminants is low. Instead, the current Vital Sign reporting compares levels of PAH-associated disease at clean, control sites, with levels at the sites of interest. Mussels do not metabolize PAHs in the same manner (Section 3.1.4) and so there is a potential for accumulation, and exposure to the consumer.

The WADOH fish consumption advisory screening levels for benzo[a]pyrene is 0.2 ng/g ww for general consumers and 0.05 ng/g ww for high consumers. For the 2015/2016 study, as reported in Lanksbury et al. (2017), 21% of sites had mussels that exceeded benzo[a]pyrene concentrations above the DOH screening levels.

For the 2017/2018 study, the LoQ for benzo[a]pyrene ranged from 0.3-0.9 ng/g ww and was present above the LoQ at 40 sites (~ 47%). As such, the concentration of benzo[a]pyrene exceeded the human health threshold level of 0.05 ng/g ww at all sites where it was detected. It is not known whether the threshold level was exceeded at the remaining 44 sites where the concentration was below the LoQ.

4.4.4.3 PBDEs in mussel tissues vs threshold values
For the 2015/2016 study, the levels for total PBDEs were compared to a screening value of 40 ng/g ww (Table 2-1). None of the tissue samples had total PBDE concentrations that exceeded this screening level.

For the 2017/2018 study, the total PBDE concentrations were represented by $\sum_{11} PBDE$. For congeners that were reported below the LoQ (i.e., U flag) a random value between 0 and the LoQ was used as the concentration value. Based on this approach, total PBDEs did not exceed the human health related threshold of 40 ng/g ww at any of the sampling locations.

4.4.4.4 Chlorinated pesticides in mussel tissues vs threshold values
Although chlorinated pesticides are not currently included in the Toxics in Fish Vital Sign reporting, there do exist some human health threshold values, particularly for DDTs. The WADOH fish consumption advisory screening levels for total DDTs are 3 ng/g ww for general consumers and 1.2 ng/g ww for high consumers. For the 2015/2016 study, 11% of sites had total DDT concentrations in mussel tissues that exceeded the high consumer screening level (Lanksbury et al. 2017).

For the 2017/2018 study, total DDTs were calculated as the sum of 6 DDT isomers and metabolites. For compounds that were reported below the LoQ (i.e., U flag) a random value between 0 and the LoQ was used as the concentration value. Based on this approach, total DDTs exceed the human health related threshold of 1.2 ng/g ww at 17 locations (20%).

4.4.4.5 Metals in mussel tissues vs threshold values
Although metals are not currently included in the Toxics in Fish Vital Sign reporting, they are addressed here in order to inform future discussions on effects of metals in biota.

For the 2015/2016 study, none of the metals concentrations exceeded WADOH screening levels or other biological thresholds. However, lead is considered unsafe at any level and it was detected at 88% of all sites with an overall average concentration of 0.4 mg/kg dw (Lanksbury et al. 2017).

Based on the results of the 2017/2018 study:

- Cd was present in tissues above a human health related threshold of 0.4 mg/kg at two sites.
- Cu was present in tissues above a human health related threshold of 4 mg/kg at one site.
- Lead was present in tissues above the LoQ at all sites.
- No other metals were detected above human health related threshold values.

5 Effects of Contaminants in Salish Sea

5.1 Toxics in Fish Thresholds

The Toxics in Fish Vital Sign was developed based on critical tissue levels for fish health, or Washington Department of Health's screening values for human health according to values in the published literature and consumption advisories from the Washington State Department of Health (Meador et al. 2002, Meador et al. 2006, Washington State Department of Health 2006, Johnson et al. 2008b, Sol et al. 2008, Arkoosh et al. 2010). Critical Threshold Levels associated with the Vital Sign are shown in Table 2-1.

These threshold values are generally believed to be protective of human health and the environment. For several reasons, however, thresholds should not be considered to be immutable. First, the science continues to develop and advances in understanding of exposure-response (for example) might justify the alteration of the thresholds. Secondly, the development of threshold includes explicit decisions (e.g., increased risk to human health due to exposure should be < 10^{-6}) and changes in these decisions (e.g., an increased or decreased tolerance of risk) would result in different thresholds. Additionally, there are different methodologies for establishing CTLs and the choice of method could result in changes. For example, the efforts by the European Commission Scientific Committee on Food and the Joint FAO/WHO Expert Committee on Food Additives resulted in a daily health based guidance value of 2.0 pg/kg body weight per day for exposure to dioxin-like PCBs, while the US EPA reported an oral reference dose of 0.7 pg/kg body weight per day for the same compounds (European Food Safety Authority 2015). Both utilized defensible approaches resulting in differing recommendations. It is important to recognize that, while threshold values are necessary for establishing goals and targets, they should be periodically evaluated and updated.

Further, the threshold based on human health criteria have been calculated based on a consumption rate of 175 g/day and were developed for salmon and English sole. The fish consumption rate varies amongst Washingtonians and the current rate may not reflect high consumers (see Section 6.5). Additionally, some consumption surveys indicate that Pacific Herring is commonly eaten (The Suquamish Tribe 2000, Washington State Department of Ecology 2013) and so it may be appropriate to develop a human health-based threshold for that species as well.

For the purposes of establishing targets associated with the Toxics in Fish Vital Sign, exceedances of the existing thresholds are used as a basis to focus recovery activities. Geographical areas and/or compound and species classes that do not exceed threshold may not be considered as priorities.

5.2 Effects of exposure to marine organisms

A brief overview of effects of exposure is summarized below. Additional information is included on: 1) effects evaluations for PBDEs, which may be useful in future evaluation of the effects thresholds that are listed in Table 2-1; 2) the effects evaluations for CECs, and; 3) a review of fish consumption rates that are critical in evaluating human health impacts of contaminants in biota.

5.2.1 PCBs

Effects of PCB exposure have been well studied and reported. These include mortality, impaired growth and reproduction, immune dysfunction, endocrine disruption, neurotoxicity, behavioral alteration, increased susceptibility to disease, and mutagenicity (see Meador et al. (2002) and references therein). Exposure can lead to both individual and population-level impacts.

5.2.2 PAHs

Impacts include cardiac dysfunction and mortality in embryonic and larval herring and salmon (Carls et al. 1999, Barron et al. 2003, Incardona et al. 2004, Meador et al. 2006, Incardona et al. 2009, Hicken et al. 2011, Incardona et al. 2014, Brette et al. 2017). Toxic effects of PAHs on fish embryos may be enhanced by sunlight (Barron et al. 2013).

- Can reduce productivity in subpopulations of English Sole (Sol et al. 2008)
- Chronic embryo mortality has been observed in areas with greatest PAH concentrations (Puget Sound) in English sole embryos (Sol et al. 2008).
- Delayed population declines from early-life PAH exposures are well established (Carls et al. 1999, Yang et al. 2003, Carls and Meador 2009)
- Pacific herring embryos on creosote pilings have decreased hatching success compared to those from distant areas (Vines et al. 2000), accumulate more PAHs and exhibit higher *cyp1a* gene expression compared to background reference (West et al. 2019). Incomplete or improper removal of creosote-treated pilings can result in increased exposures of PAHs for up to a year (West et al. 2019).

5.2.3 PBDEs

- PBDEs may affect thyroid hormone levels and immune function in exposed biota (Hall et al. 2003, Frouin et al. 2010).
- Salmonids exposed to PBDEs may suffer increased disease susceptibility (Arkoosh et al. 2010).
- Exposure to low concentrations of PBDEs in reproductive fish can affect thyroid hormone in offspring and may lead to PBDE-induced toxicity in subsequent nonexposed generations (Yu et al. 2011).
- PBDE exposure may lead to reproduction impairments (Yu et al. 2015).
- Endocrine disrupting effects of PBDEs and OH-BDEs have been reported and that the presence of an OH group greatly increases the endocrine disrupting potential of PBDEs (Wiseman et al. 2011).

5.2.3.1 PBDE effects evaluations

The current threshold values for the vital sign indicator targets for PBDEs are listed as 40 ng/g ww, based on a human consumption threshold, or 470 ng/g lw based on fish health (Table 2-1). Considerations for revising these threshold values include: 1) OH- and MeO-PBDEs, which are not generally quantified with existing analytical methods and that may be associated with the presence of anthropogenic PBDEs, can impart additional toxicity on exposed organisms, and 2) recent studies biological responses, even at low exposure levels.

The presence of OH- and MeO- PBDEs have been reported in marine organisms. MeO-PBDEs are produced by marine bacteria and algae, but not likely from the transformation of anthropogenic PBDEs. OH-PBDEs can be formed through the metabolism of MeO-PBDE in fish (Wan et al. 2009). The transformation of PBDE to OH-PBDE has been reported in mammalian systems (Dong et al. 2010) and through abiotic photochemical processes in aquatic systems (Zhao et al. 2015). OH-PBDEs tend to be more potent than PBDEs. Additionally, BDEs and the OH- and MeO- derivatives have been shown to affect Zebrafish through different sets of receptor-mediated pathways (Liu et al. 2015). As such, assessments of risks of anthropogenic PBDE need to be made against the background of the presence and impacts of the derivatives.

Even in light of those complexities, three recent studies (Arkoosh et al. 2015, 2017, Arkoosh et al. 2018) illustrate the difficulties of exposure assessments even when focusing on simple mixtures of purely anthropogenic PBDEs. These studies focus on exposures to BDE-047 and BDE-099 (two of the most abundant congeners in biological matrices) suggest that biological effects may occur at low concentration, though divergent effects from different congener and nonmonotonic responses complicate the risk assessment. Arkoosh et al. (2015) reported that immune function and disease susceptibility were significantly affected by exposures to BDE-047 or BDE-099, though not consistently across exposure levels; responses were nonmonotonic. For example, salmon had reduced survival in the low BDE-099 exposure compared to control, but not with medium or high BDE-099 exposures. Effects were observed at exposures of ~ 300 ng/g lw (~ 6-9 ng/g ww).

Arkoosh et al. (2017) exposed juvenile Chinook to a range of concentrations of BDE-047 and BDE-099, either as individuals or as 1:1 mixtures. Exposure to BDE-099 alone, or a mixture of BDE-047 and BDE-099 altered thyroid hormone concentration, though exposure to BDE-047 alone did not. Effects were observed at a mixture concentration of 36.8 ng/g ww, though responses appeared nonmonotonic.

Arkoosh et al. (2018) exposed juvenile Chinook to a 1:1 mixture of BDE-047 and BDE-099 at a range of concentrations and, consistent with previous work, described a complex response. For example, disease-induced mortality was increased at exposure concentrations of 10.9, 36.8, and 184 ng/g ww relative to controls, but not at exposure concentrations of 2.8 or 98.6 ng/g ww. Additionally, two measures of immune response were enhanced even at the lowest levels of exposure (2.8 ng/g ww), though, as described, this did not associate with increased disease resistance.

5.2.3.2 PBDEs in Mussels

There has been some work evaluating the effects of PBDE exposures on bivalves, including mussels.

- Blue mussels (*Mytilus edilus*) exposed to 1 µg/L BDE-047 for 3 weeks suffered damaging effects on ovarian follicles and ovocytes in females, and induced spawning in males (Aarab et al. 2006). They concluded that BDE exposures may impair mussel reproductive function and so act as endocrine disruptors.
- Vidal-Liñán et al. (2015) exposed Mediterranean mussels (*Mytilus galloprovincialis*) to BDE-047 at concentrations ranging from 2 – 15 µg/L and measured the response of three biomarkers (glutathione S-transferase (GST), glutathione peroxidase, and acetylcholinesterase (AChE)). They reported that AChE and GST activities were inhibited at all exposure concentrations, suggesting the potential for neurotoxic impacts from BDE-047 exposures.
- Espinosa Ruiz et al. (2019) fed Mediterranean mussels to microalgae containing BDE-047 at concentrations up to 100 ng/L. Results suggested that BDE-047 exposure acted as a stressor against humoral immune parameters, increasing the susceptibility of the exposed organisms to pathogens.

5.2.4 CECs and EDCs

There is growing evidence that some trace organic contaminants can cause impacts to aquatic organisms via endocrine disruption, metabolic and reproductive alterations, and behavior modifications. For example, male fish have been shown to undergo feminization following exposure to contaminants of emerging concern (CEC); such impacts have been observed in Puget Sound and the Columbia River (Lower Columbia River Estuary Partnership 2007, Johnson et al. 2008b, da Silva et al. 2013, O'Neill et al. 2016).

5.2.4.1 CEC effects evaluations

There are ongoing efforts in the region aimed at better understanding the occurrence patterns and potential risk due to the presence of CECs other EDCs in the environment. Not all compounds are bioactive and so they may not lead to any substantial harm or behavior alteration at the generally low concentrations found in Puget Sound. Others, however, may be of concern. Due to the range of potential exposure and impacts of CECs, some effort at prioritization is needed to identify those which may cause harm. A preliminary prioritization effort has begun in order to rationalize and focus investigations (James et al. 2015). This has been followed up by a current project meant to prioritize CECs by comparing occurrence levels in the environment with effects levels. The challenge of this approach is that traditional effects evaluations have only been done for a limited set of chemicals. As such, alternative approaches are needed. Several approaches have been explored in regional risk evaluation related to the presence of CECs. These are summarized below.

<u>Prioritization based on PNEC</u>

An approach for identifying priority CECs is by comparing the occurrence levels in the environment with published Predicted No Effects Levels (PNECs) which is a concentration, below which there will be no impacts on an exposed organism. The determination of PNECs can be performed by experimentation or modelling. For example, Caldwell et al. (2019) determined a PNEC of 1 mg/L for metformin, a widely used antidiabetic that is present in Puget Sound, by applying an assessment factor to the lowest no observed effect concentration from multiple chronic studies with algae, daphnids and fish. In cases where there are not experimental data, quantitative structural activity relationship (QSAR) models are used to predict toxicity and develop PNEC values (Aalizadeh et al. 2017). The limitations of QSAR modeling have been widely noted (e.g., Roy et al. (2016)) and so the use of QSAR-based PNECs should be done with caution.

The prioritization of CECs can then be based on the calculation of a risk factor (RF):

$$RF = \frac{Environmental\ Concentration}{PNEC}$$

Compounds with RF>1 indicate the potential for environmental risk.

This approach was utilized by Tian et al. (2020) who identified a suite of eight CECs in Puget Sound marine waters with RF>1, and that ought to be prioritized for management.

The European Union NORMAN project manages and curates a database system of that provides PNEC and ecotoxicological information (including PNEC values) for ~ 65,000 substances (https://www.norman-network.com/nds/ecotox/; Dulio et al. (2018)). The majority of the PNECs are determined by QSAR modeling, though an increasing number are validated by NORMAN participants.

Prioritization based on In Vitro Assays

There are additional approaches for identifying CECs with the potential to cause environmental harm based on the use of high throughput in vitro assays. The ToxCast and Tox21 programs are generating information on the biological activities based on several hundred high-throughput assays that cover a range of cell responses and approximately 300 signaling pathways (Kavlock et al. 2012, Tice et al. 2013). These assays cover a much wider range of chemicals compared to the traditional, published ecotoxicological evaluations; as such this information should be leveraged to identify priority CECs. Data are available via USEPA Comptox dashboard (https://comptox.epa.gov/dashboard).

Recent examples of this approach include Corsi et al. (2019) and Pinto et al. (2019). In each case, ToxCast bioactivity data were used to develop Activity Concentrations at Cutoff (ACC) values, which are essentially the concentrations which initiate an assay response. ACCs are developed based on bioassays that can be associated with biological responses – some bioassays are based on receptors known to be associated with endocrine disruption. Similar to the risk factor (RF) approach described above, an Exposure Activity Ratio (EAR) is determined by:

$$EAR = \frac{Environmental\ Concentration\ (x\ BCF)}{ACC}$$

A bioconcentration factor (BCF) may be used to translate environmental concentrations to predicted plasma concentrations that may be more representative of exposure concentration that initiates a response (Pinto et al. 2019).

Corsi et al. (2019) indicated that an EAR > 10^{-3} was a reasonable threshold above which a chemical would be of concern.

The Puget Sound Ecosystem Monitoring Program (PSEMP) Toxics workgroup is currently utilizing this approach to identify priority chemicals in the Salish Sea. Results will be used to inform regional monitoring and management of CECs.

Application of Fish Plasma Model

Meador et al. (2017) evaluated the potential impacts of CECs by comparing predicted fish plasma concentration of a suite of CECs with the therapeutic human dose level in plasma. Fish plasma concentration were predicted by a fish plasma model that used measured tissue concentrations. Therapeutic dose levels were available for 70 compounds. 16 out of 24 detected for Chinook and 7 of 14 for sculpin resulted in a Response Ratio > 1 indicating the potential to initiate a biological response.

5.2.4.2 CECs and EDCs in Mussels

There are several studies evaluating the impacts of CECs and EDCs on mussels (see James et al. (2020) and references, therein). Briefly, focusing on those compounds which have been reported in mussels:

- Alkylphenols are toxic to bivalves in the mg/L concentration range, with evidence of endocrine effects including dose-dependent Vtg-like protein induction and alterations in testosterone levels at concentrations as low as 1,000 ng/L (Quinn et al. 2006, Ricciardi et al. 2008, Riva et al. 2010). The reported exposure concentrations are reportedly much lower in Puget Sound suggesting biological impacts to mussels from NP exposures might be minimal.
- Freshwater mussels exposed to SSRIs demonstrated alterations of various biological endpoints including larval metamorphosis rate and algal clearance rate (Di Poi et al. 2014, Gilroy et al. 2017), though at levels that were much higher than reported environmental concentrations.
- Freshwater mussel (*Dreissena polymorpha*) haemocytes showed a clear dose-response effect of DNA damage after in vitro exposure to melphalan (Buschini et al. 2003).
- Marine mussels exposed to metformin was associated with follicle and gamete degeneration and follicle dilation, which could result in disruptions to the reproductive process (Koagouw and Ciocan 2018).

5.2.5 Population-level impacts

- Linking sublethal or lethal effects of contaminants on individuals to population-level effects is difficult, and is presently a major uncertainty for this Vital Sign.
- Linking impairment to population effects requires individual-based modeling, which requires understanding contaminant-life history component links (Spromberg and Meador 2006).
- At present, there are no conclusive data to show that toxic chemicals have been a significant factor in the decrease of Cherry Point or other stocks of herring in Puget Sound (Washington State Department of Natural Resources 2010).
- However, herring populations have suffered age truncation (loss of older age classes), which is consistent with longer time of exposure to POPs (Landis and Bryant 2010).

- Nilsen et al. (2019) identified five key challenges in understanding the potential effects of CECs on populations or food webs. These include: 1) the complexity of mixtures of CECs in the aquatic environment, 2) understanding how sublethal effects of CECs impact aquatic organisms, 3) evaluating the effects of chronic exposures within and across generations, 4) understanding and integrating the outcomes of multiple stressors in addition to CEC exposures, and 5) evaluating the trophic consequences of CEC exposure across aquatic food webs.

6 Impacts on human health

Exposure to toxic contaminants can lead to ill effects for aquatic organisms and the people that consume them. The Vital Sign goal indicates that fish should be safe for human consumption; several of the effects threshold values are based on human health consumption risks. These risk levels were largely developed by DOH (Washington State Department of Health 2006) and are presented.

6.1 Human exposure to PCBs

An extensive review of the research on the health effects of PCBs is presented in the updated Toxicological Profile for PCBs (ATSDR 2000). Briefly, the International Agency for Research on Cancer (IARC) classifies PCBs as human carcinogens. Exposure to PCBs has also been associated with adverse effects to the human immune system, endocrine system and disruption, and reproductive systems.

As described above, PCBs consist of a group of 209 different compounds which vary according the extent and location of chlorination (Figure 3-1). The location of the chlorine affects the overall shape of the PCB molecule. PCBs that lack chlorines in the ortho position (2, 2', 6, or 6') tend to form planar alignments allowing for dioxin-like structures and increasing the probability of interactions with biological receptors. These include PCB-77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189. The relative toxicity of each of these compounds is different and may vary according to species, with PCB-126, -168, and -81 often being the most active. Toxic Equivalency Factors (TEFs) can be used to compare toxicity and determine an overall toxicity of a PCB mixture. The development of cleanup standards based on TEFs has been recommended.

Human exposure effects thresholds have been used by the Washington State Department of Health to establish consumption limits; those thresholds have been adopted into this strategy document (see Table 2-1. Toxics in Fish Vital Sign Indicators and associated recovery targets for fish species and contaminants. Also included are an indication of habitat/food web position, and spatial scale of habitat range of each of the indicator species.

6.2 Human exposure to PBDEs

An extensive review of the research on the health effects of PBDEs is presented in the Toxicological Profile for PBDEs (ATSDR 2017). Briefly, the research classified PBDE as a Group 3 carcinogen (not classifiable as to its carcinogenicity to humans) based on inadequate evidence. The EPA assigns the cancer category Group D (not classifiable as to human carcinogenicity) to mono-, di-, tri-, tetra-, penta-, hexa- octa-, and nona-BDEs; and as "suggestive evidence of carcinogenic potential" for deca-BDE. There is inadequate information on other congeners.

Exposure to BDEs may lead to developmental impacts in children. Results from human studies suggest that PBDE-exposure may be linked to neurodevelopment alterations in children, including impaired cognitive development, impaired motor skills, increased impulsivity, and decreased attention.

Ingestion and dermal absorption of house dust are the major pathways of PBDE exposure in children and adults, accounting for approximately 56-77% of the total PBDE intake. Major PBDE exposure pathway for infants is breast milk (Johnson-Restrepo and Kannan 2009).

6.3 Human exposure to PAHs

An extensive review of the research related to the health effects of exposure to PAHs is presented in the Toxicological Profile for Polycyclic Aromatic Hydrocarbons (PAHs; ATSDR (1995)). Briefly:

There are many different PAHs and they generally occur in mixtures. Seventeen of these have been identified for extensive profiling because: 1) there is more information, 2) they are considered to be more harmful than many of the others, and 3) exposure may be more likely. These are:

- acenaphthene
- acenaphthylene
- anthracene
- benz[a]anthracene
- benzo[a]pyrene
- benzo[e]pyrene
- benzo[b]fluoranthene
- benzo[g,h,i]perylene
- benzo[j]fluoranthene
- benzo[k]fluoranthene
- chrysene
- dibenz[a,h]anthracene
- fluoranthene
- fluorene
- indeno[1,2,3-c,d]pyrene
- phenanthrene
- pyrene

Several have been identified as carcinogens in Table 6-1. The major route of exposure to PAHs is via outdoor or indoor air, smoking or smoke from fireplaces, and through food (Kim et al. 2013a). Drying, smoking, or grilling food can generate PAHs and lead to exposures.

General effects can include:

- Cancer and tumor formation.
- Exposure during pregnancy may lead to reproduction impacts (including offspring of those exposed), increased birth defects and decreased body weight.
- Exposure can negatively affect immune system function.

Table 6-1. Cancer risk for selected PAHs. Compounds not shown were either determined to have no appreciable cancer risk, or were not extensively evaluated. From: https://www.atsdr.cdc.gov/phs/phs.asp?id=120&tid=25

Compound	Department of Health and Human Services	International Agency for Research on Cancer	Environmental Protection Agency
benz[a]anthracene	Known	Probable	Probable
benzo[b]fluoranthene	Known	Possible	Probable
benzo[j]fluoranthene	Known	Possible	
benzo[k]fluoranthene	Known	Possible	Probable
benzo[a]pyrene	Known	Probable	Probable
chrysene		Not classified	Probable
dibenz[ah]anthracene	Known		Probable
indeno[123-cd]pyrene	Known	Possible	Probable

6.4 Human exposure to EDCs

An extensive review of the research on the health effects of exposure to EDCs is presented in the WHO/UNEP report om EDCs (WHO/UNEP 2012)

Briefly:

- Approximately 800 chemicals have been identified with some evidence that they may interfere with hormone receptors, hormone synthesis or hormone conversion.
- Population based observation suggests widespread exposure. These include: low semen quality in young males potentially impairing reproduction; increased global rates of endocrine-related cancers (breast, endometrial, ovarian, prostate, testicular and thyroid) over the past 40–50 years; endocrine-related effects in wildlife populations.
- EDC exposures can impact: female reproductive health (e.g., increased breast cancer), male reproductive health (e.g., increased testicular cancer), altered sex ratio, thyroid-related disorders, neurodevelopmental disorders in children, hormone-related cancer, adrenal disorders, bone disorders, and metabolic disorders (e.g., diabetes).

6.5 Fish Consumption Rates

Eating contaminated fish provides a dose of the various chemicals that are found within the fish tissue and so the more fish eaten by an individual, the higher the doses. As such, determining a “safe” exposure threshold is dependent on the quantity of fish being consumed in addition to other factors. And since fish are exposed to and take up contaminants from the environment, fish consumption rates are often used to establish water quality and sediment quality standards. The water quality criteria in Washington were established based on a fish consumption rate of 175 g/day (WAC 173-201A-240).

According to the results of a set of surveys, there are differences in people's fish consumption rates and practices. Recreational fishers may consume more fish than other people in Washington. Some groups, including Native Americans and Asian and Pacific Islanders, consume much larger amounts of finfish and shellfish (Washington State Department of Ecology 2013). A summary of survey results is presented in Table 6-2.

Table 6-2. Summary of contemporary fish consumption for all shellfish and finfish. All values in g/day. Values are for all fish from all sources. Adapted from Washington State Department of Ecology (2013) and Freimund et al. (2012).

			Percentiles		
Population	**Source of Fish**	**Mean**	**50th**	**90th**	**95th**
National Estimates (consumers only)	NHANES 2003-2006: EPA Statistical Survey Method	56	38	128	168
	NHANES 2003-2006: NCI Statistical Survey Method	19	13	43	57
Columbia River Tribes (1994)		63	41	130	194
Tulalip Tribes (1996)		82	45	193	268
Squaxin Island Tribe (1996)		84	45	206	280
Suquamish Tribe (2000)		214	132	489	797
Lummi Nation (2012)		383	314	800	918
Asian and Pacific Islanders (1999)		117.2	89	242	
Recreational Fishers (compilation of multiple studies)		11–53	1.0–21	13–246	
		6.0–22		42–67	

The fish consumption rates listed in Table 6-2 are generally determined based on surveys of recent (days-to-weeks) behavior. However, these results may be impacted by a "suppression effect" whereby the current degraded and depleted state of a resource diminishes the use of that resource, particularly compared to historical practices (National Environmental Justice Advisory Council 2001). In this case, the presence of contaminants in the fish, fish consumption advisories, degraded habitats, etc. would all result in a lower consumption rate than would otherwise occur (O'Neill 2016). The recognition of suppression has led to the development of heritage fish consumption rates, which describe consumption practices that occurred in the past, and would occur in the present and future in absence of factors that might lead to behavior alteration. A comparison of contemporary and heritage fish consumption rates for the Columbia River basin indicated that heritage rates are one or two orders of magnitude higher than contemporary averages (Harper and Walker 2015a, Harper and Walker 2015b).

7 Toxics in Fish – Contaminant sources, distribution, and loading pathways

This section addresses sources, loadings, pools and distribution of PAHs, PCBs, PBDEs and EDCs, as well as the pathways by which they are delivered to Puget Sound waters and resulting in exposures.

Sources include the processes that result in contaminant production (e.g., wood combustion for PAHs) or products (e.g., transformer oil for PCB) that introduce the contaminants into the Puget Sound watershed. The primary means by which toxicants enter into, or are produced within, the greater Puget Sound watershed are included. Due to differential land use and development the geographic distribution of sources varies throughout the Puget Sound.

Legacy pools - In addition to new sources, there are major pools of legacy contaminants. These legacy contaminants are the result of historic releases and subsequent transport and accumulation to a current environmental compartment (e.g., marine sediments). Many of the compounds of interest in the Toxics in Fish Vital Sign are hydrophobic and degrade slowly. As such, contaminants can enter the Puget Sound and accumulate. The more-labile compounds (e.g., low molecular weight PAHs, many EDCs) will not persist in the Puget Sound environment and so are not likely to form contaminant pools that result in exposure. Legacy contaminant pools may or may not result in exposures.

Pathways describe the means by which contaminants are transported from source or pools to an environment where biological exposures may occur. Commonly discussed pathways include stormwater and wastewater treatment system discharge. Neither of these are the site of original production of contaminants but rather they bring contaminants to an environment where exposures may occur.

The magnitude of the pathways is referred to as the loadings. There are uncertainties in the current loading estimates largely due to limited geographic coverage and low sample replication. In addition, not all potential loadings are accounted for; for example, the loading estimates do not account for an estimated 84,000 kg of PAHs released from vehicle emissions each year (Washington State Department of Ecology and Washington State Department of Health 2012).

The Puget Sound Regional Toxics Loading model was utilized to estimate contaminant fate, transport, and bioaccumulation in Puget Sound (Washington State Department of Ecology 2015a). They reported that loadings estimates from watershed pathways (i.e., surface runoff, POTWs, groundwater) were 5-10 times too low to match observed contaminant masses. In addition, loading estimates from the Straits of Juan de Fuca and Georgia were likely 2-4 times too low based on a comparison of calculated versus observed contaminant masses. Furthermore, the best loading estimates are basin-wide, whereas inputs are local. A summary of Puget Sound-wide loading estimates is presented in Figure 7-1.

Stormwater associated contaminant loadings are seasonal with limited loading occurring during the summer dry period. There may be a spike in loading associated with build-up on land during dry periods, followed by stormwater deliveries during subsequent rain events (Washington State Department of Ecology 2015b).

A key knowledge gap is the relative importance of nearshore sediment sources versus overland flow inputs.

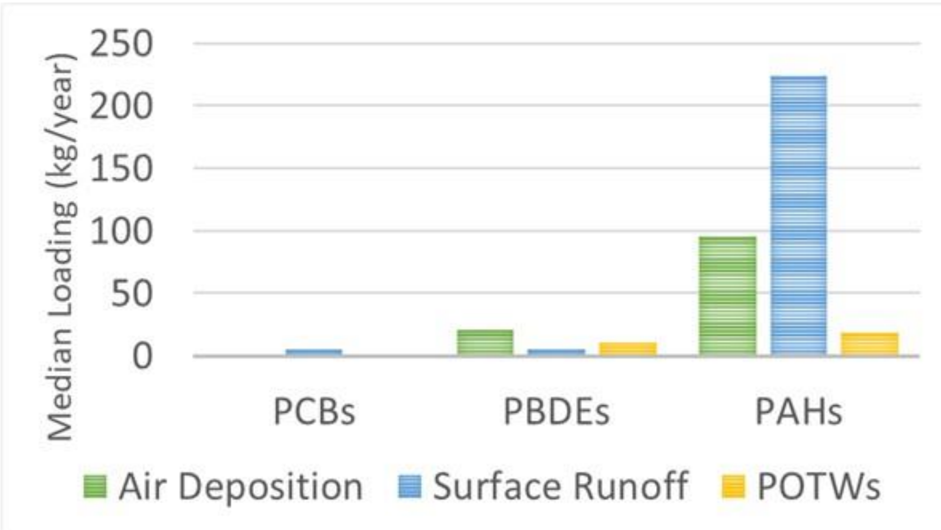

Figure 7-1. Loading estimated by major pathway for PCBs, PBDEs, and PAHs in Puget Sound (Ecology and King County 2011).

7.1 PCBs – Sources, pools, and pathways

Figure 7-2. Overview schematic indicating sources, pools, and pathways for PCBs in Puget Sound.

7.1.1 PCB Sources and Pools

The primary historic sources of PCBs have been curtailed due to regulatory bans. A level of inadvertent production continues to occur, though the congener profiles are generally different compared to the historic production. Notes on sources and pools are presented below.

- PCBs were used in a wide range of industrial and consumer goods for chemical stability and low flammability. Materials containing PCBs include electrical transformers and capacitors, plasticizers, hydraulic fluids, wax and pesticide extenders, lubricants, and adhesives.
- Present sources of PCBs include hazardous waste sites, illegal or improper disposal of industrial wastes and consumer products, leaks from old electrical transformers, municipal and industrial

effluent, runoff from contaminated surfaces, e.g., from clusters of older buildings that have PCB-containing exterior coatings (King County 2013, Washington State Department of Ecology 2015b).

- PCBs were a common additive to sealants used in caulk, around window joints and expansion joints, and some roofing materials. About half of the joint sealant compounds tested in Switzerland contained PCBs. About half of those with PCBs has a chlorine content of 45-55% (Kohler et al. 2005).
- Certain PCB congeners are produced as inadvertent by-products. This inadvertent production of PCBs is allowable at concentrations less than 50 ppb in consumer products.
- PCB regulatory standards are based on measurements from analytical methods (e.g., total Aroclors) that would not necessarily detect inadvertently-produced congeners, such as PCB-11. See Section 3.2.1.1.
- PCB congener profiles differ in new versus legacy products. PCB-11 is common in inadvertent production while it was rarely present in the Aroclor mixtures.
- Washington State Department of Ecology evaluated 68 products for PCBs, focusing on PCB-11, PCB-206, PCB-208, and PCB-209, which are congeners not typically present in the Aroclor mixtures. PCB-11 and PCB-209 were found in 66% and ~10% of the samples tested; PCB-206 and -208 were not present in most of the products tested (Stone 2014).
- Additional sampling was performed on 201 consumer products (Washington State Department of Ecology 2016a). Out of 216 samples, 10 samples contained total PCBs > 0.1 ppm, and 3 samples > 1 ppm. In the 3 samples over 1ppm, 99% of the total PCB consisted of PCB-11. Summary results and regulatory standards are shown below.

Table 7-1. PCB concentrations in products tested by Washington State Department of Ecology (2016a) compared to legacy materials and regulatory standards

Product, Legacy Material, or Regulatory Standard	**Concentration (ppm)**	**Reference**
yellow sidewalk chalk	1.06	Ecology 2016
yellow foam office product	2.31	Ecology 2016
single-serving cereal package	2.32	Ecology 2016
WA marine sediment quality standard	12	WAC 173-204-320
TSCA maximum for inadvertent generation	50	40 CFR Part 761.3
non-PCB electrical transformer	< 50	40 CFR Part 761.3
WA sediment cleanup screening level	65	WAC 173-204-562
PCB electrical transformer requiring registration	≥ 500	40 CFR Part 761.3
legacy joint caulk in Seattle	up to 79,000	King County 2016

- PBC-11 is present in commercial paints, likely due to inadvertent production (Hu and Hornbuckle 2010).
- Davies and Delistraty (2016) estimate the annual releases of PCBs to the environment in Washington State to lamp ballasts (400–1500 kg), inadvertent generation by industrial processes (900 kg), caulk (160 kg), small capacitors (3–150 kg), large capacitors (10–80 kg), pigments and dyes (0.02–31 kg), and transformers (<2 kg).

- Work in San Francisco Bay has shown that PCB-11 is neither persistent nor accumulating in fish (Davis et al. 2014).

7.1.2 PCBs – Pools and Distribution

PCBs are hydrophobic and persistent and so will accumulate in sediments and the food web. Davies and Delistraty (2016) reported that the majority (~97%) of existing PCBs in the Puget Sound ecosystem is contained in the active sediment layer (top 10 cm), <1 % is in the water column, and <3% is in the food web. The pools vary markedly throughout the Puget Sound with urban bays almost always having higher sediment PCB concentrations compared to other areas sampled. Both PCBs and PBDEs are found in higher concentrations in sediments of urban bays and shorelines near Seattle, Tacoma, Bremerton and Everett (Long et al. 2005). Few sites within the urban bays had chemical concentrations which exceeded Washington State Sediment Quality standards (e.g., 17% of Elliot Bay area and 5% of Commencement Bay; (Washington State Department of Ecology 2016c, b) though the lack of exceedances does not necessarily mean lack of PCBs in sediments or lack of transport to the food web.

There is significant uncertainty in the estimates of total mass in Puget Sound sediments.

7.1.3 PCBs – Pathways

An estimate of PCB loads via major pathways is shown in Table 7-2. There is likely a high degree of uncertainty associated with these estimates; Washington State Department of Ecology (2015a) estimated that reported loadings were likely 5-10 times too low. In addition, defined conceptual pathways between sources (e.g., PCBs in lamp ballasts or caulk) and the pathways identified below are not always clear.

Table 7-2. Estimated PCB loads to Puget Sound from selected pathways. Adapted from Washington State Department of Ecology and King County (2011) and Washington State Department of Ecology (2015a)

PCB Pathway	Loading Estimate (2011; kg/year)	Puget Sound Regional Toxic Model (2015; kg/yr)
Groundwater	not analyzed	not analyzed
Air Deposition	1.3	0.4
Surface Runoff	5.3	4.2
POTWs	0.3	0.3
Ocean Exchange	0.8	not calculated
Returning Salmon	0.3	not calculated

7.2 PBDEs – Sources, pools, and pathways

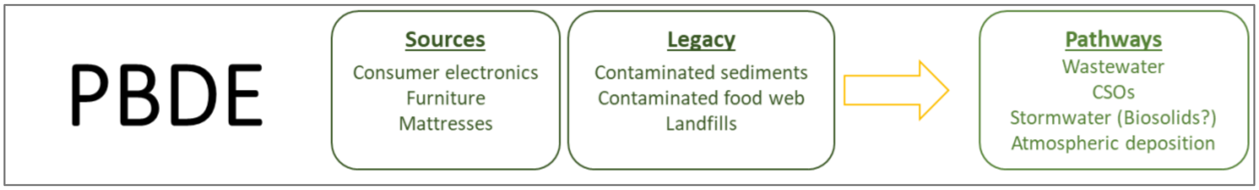

Figure 7-3. Overview schematic indicating sources, pools, and pathways for PBDEs in Puget Sound.

7.2.1 PBDEs – Sources and Pools

The use and production of PBDE homologous groups is currently restricted and so PBDEs entering circulation in new products is limited. PBDEs remain widespread in consumer products that were purchased prior to the phase-out. The primary sources of PBDEs currently are flame retardants in furniture, computer monitors, and other electronics (Washington State Department of Ecology and Washington State Department of Health 2006, Washington State Department of Ecology and King County 2011). The amount of PBDE in products varies depending on the product homolog group used, but typically between 5 and 30% by weight.

PBDE-containing consumer products are typically disposed of in landfills. Some are recycled. Landfills that receive the consumer products can become large pools.

The use of BDEs in new consumer products is limited. They have likely been replaced by alternative halogenated flame retardants (Tao et al. 2016).

7.2.2 PBDEs - pools

- PBDEs are persistent and are transported atmospherically from areas that continue to use them. Environmental cycling has produced large reservoirs of PBDEs in marine sediments (Ross et al. 2009).
- PBDE concentration in sediments is strongly influenced by proximity to sources (Johannessen et al. 2008)
- Transport via trans-Pacific air masses and subsequent surface deposition may provide some continual inputs of PDBEs to Puget Sound, though local urban sources are likely more important (Noel et al. 2009)
- Work from the Strait of Georgia found that urban harbors and municipal outfalls can lead to hot spots for PBDEs in sediments. Congener BDE-209 accounted for 52% of total PBDEs in these areas. Sediments collected from nonurban areas had a higher proportion of BDE-209 compared to shallow samples (66% versus 32%). Both physicochemical properties and environmental processes lead to different PBDE profiles in the sediments of the Strait of Georgia (Grant et al. 2011).
- Plankton monitoring in the Strait of Georgia demonstrated that planktonic uptake may be an important mechanism by which PBDEs enter into the aquatic food web. More highly brominated PBDEs tend to be more preferentially partitioned onto particles. Overall, in the Strait of Georgia

approximately 27 kg of PBDE were in dissolved phase while 1 kg were associated with plankton (Frouin et al. 2013).

7.2.3 PBDEs - Pathways

An estimate of PBDE loads to Puget Sound via major pathways is shown in Table 7-3. Atmospheric deposition and publicly owned treatment works (POTWs) are the largest delivery pathways for PBDE to Puget Sound (Washington State Department of Ecology and King County 2011). However, as with PAHs and PCBs, there remain major uncertainties with the estimations. There are temporal and spatial variations that are not well accounted for in these estimates. For example, King County reported that stormwater is the second largest pathway for PBDEs into local receiving waters (King County 2013). Additional studies have examined other potential (local) sources, e.g., PBDEs that are ubiquitous in landfill leachate and can act as local sources if not well managed (Li et al. 2012).

PBDEs have been used largely in indoor consumer products and so transport pathways likely include volatilization to indoor air and dust followed by subsequent transport to the outdoor environment.

Table 7-3. Estimated PBDE loads to Puget Sound from selected pathways. Adapted from Washington State Department of Ecology and King County (2011) and Washington State Department of Ecology (2015a)

PBDE Pathway	Loading Estimate (2011; kg/year)	Puget Sound Regional Toxic Model (2015; kg/yr)
Groundwater	not calculated	not calculated
Air Deposition	20.3	3.5
Surface Runoff	5.7	4.5
POTWs	10.6	9.9
Ocean Exchange	-11	not calculated

Washington Department of Ecology and Herrera Environmental Consultants (2010) evaluated concentrations of several contaminants in effluent samples from 10 wastewater treatment systems throughout Puget Sound. PBDE levels in effluent varied significantly, but it is unknown if those differences are a result of treatment methods, PBDE influent concentrations, or other factors. Studies performed elsewhere found PBDE removal through wastewater treatment plants to be approximately 90% (Vogelsang et al. 2006, Kim et al. 2013b). Sorption to solids is the primary removal mechanism of PBDEs from the liquid stream (Kim et al. 2013b, c) – the mass of PBDEs is not reduced through wastewater treatment systems, it simply transfers to the solids. As such, the use of sewage sludge as a soil amendment is a potential pathway for the release of PBDEs into the environment (Gockel and Mongillo 2013).

7.3 PAHs – Sources, pools, and pathways

Figure 7-4. Overview schematic indicating sources, pools, and pathways for PAHs in Puget Sound.

7.3.1 PAHs - sources

- Primary sources of PAH compounds in urban airscapes are wood smoke and diesel exhaust (Washington State Department of Ecology and Washington State Department of Health 2012, Brette et al. 2017)
- The Puget Sound Air Deposition Study reported that most atmospheric particles are from petroleum and biomass combustion (Washington State Department of Ecology 2010)
- Creosote treated railroad ties, power poles, and marine pilings were identified as a major source of PAHs to the Puget Sound (Washington State Department of Ecology and Washington State Department of Health 2012)

7.3.2 PAHs - pools

PAHs are hydrophobic, and are therefore associated with suspended particles and/or sediments. PAHs can persist in sediments.

Previous sediment monitoring in the Puget Sound indicated variation with a mean total PAH concentrations ranging from a low of 46 ppb at North Hood Canal to a high of 7,727 ppb at Thea Foss Waterway (Washington State Department of Ecology 2005). The Thea Foss waterway site has since been remediated. PAHs in the majority of stations routinely monitored by the Department of Ecology did not change from 1989-2000; most of the changes that were observed were increases in concentration (Washington State Department of Ecology 2005).

There are localized areas with high PAHs concentration, generally in urbanized bays, near sites of previous industrial activities, or in and around clusters of pilings. Focused remediation efforts have successfully addressed sites throughout the Puget Sound.

- High molecular weight PAHs, the group most often associated with liver disease (Myers et al. 1991), have declined in Elliott Bay sediments (Washington State Department of Ecology 2009), and in English sole bile from Elliott Bay.
- In Commencement Bay there were only selected stations of high PAH concentrations reported in the 2008 and 2014 sampling events, primarily within and near to the Thea Foss waterway. Between 2014 and 1999 the low molecular weight PAH concentrations did not change or increase while concentrations of the high molecular weight PAH remained the same or decreased (Washington State Department of Ecology 2016c). A significant portion of the Thea Foss waterway was dredged or capped from 2002-2006.

- PAH contaminated sediments in Eagle Harbor were addressed by placing a primary cap of clean sediment over the most-contaminated area (1993-1994) with a secondary cap added from 2000-2002. Comparison from before and after the capping indicated a significant decline in legion prevalence and other biomarkers in English sole following the remediation (Myers et al. 2008). Cap maintenance is ongoing.

7.3.3 PAHs - Pathways

Estimates for major pathways to Puget Sound are shown in Table 7-4. There are major caveats and uncertainties associated with these calculations and they should be interpreted with caution. The original reports should be referred to for a complete discussion. The loadings from the Puget Sound Regional Toxics Model (2015) were revised and regenerated from Washington State Department of Ecology and King County (2011)

Table 7-4. Estimated PAH loads to Puget Sound from selected pathways. Adapted from Washington State Department of Ecology and King County (2011) and Washington State Department of Ecology (2015a)

PAH Pathway	Loading Estimate (2011; kg/year)	Puget Sound Regional Toxic Model (2015; kg/year)
Groundwater	284	272
Air Deposition	96	54
Surface Runoff	224	348
POTWs	19	17
Ocean Exchange	not calculated	not calculated
Returning Salmon	not calculated	not calculated

7.4 CECs and EDCs – Sources, pools, and pathways

Figure 7-5. Overview schematic indicating sources, pools, and pathways for EDCs in Puget Sound.

7.4.1 CECs and EDCs - sources

As described above, EDCs include a wide range of compounds that are grouped based on biological effects rather than source, or physical or chemical property. As such, identifying priority sources for EDCs is complicated. A number of pharmaceutical compounds, especially hormones such as steroids or estrogens, have been identified as potential endocrine disruptors though there are numerous compounds in other use categories such as antiseptics (e.g., triclosan), pesticides (e.g., carbofuran, atrazine, lindane), consumer products (e.g., phthalates, bisphenol A) which also have been shown to interact with the endocrine system (Lintelmann et al. 2003, Tijani et al. 2013). Several approaches have been proposed to prioritize monitoring and treatment of trace organic compounds, including EDCs (Kumar and Xagoraraki 2010, Diamond et al. 2011, Vulliet and Cren-Olive 2011). A draft prioritization scheme has been created for the Puget Sound (James et al. 2015).

There are ongoing activities which are focused on further identifying and characterizing EDCs. For example, the EPA Endocrine Disruptor Screening Program (EDSP) was developed to identify potential endocrine disruptors (Tier 1 screening) then determine adverse effects and dose-response relationships (Tier 2 testing). Out of approximately 10,000 candidates for screening—called the Universe of Chemicals—the program has completed Tier 1 tests for only 174 (EPA 2017). Validation of Tier 2 test methods was completed in 2014, and review of the first Tier 2 results is expected to occur 2019-2020 (EPA 2014). Resulting data is expected to provide risk information needed to regulate chemicals under current laws (e.g., Clean Water Act; Toxic Substances Control Act; Safe Drinking Water Act; Federal Food, Drug, and Cosmetics Act).

7.4.2 CECs and EDCs – presence in environment

See Section 4.3.

7.4.3 CECs and EDCs - pathways

There are no regional loading estimates for the suite of endocrine disruption chemicals though there are studies on specific compounds. Bisphenol A, for example, is an endocrine disrupting compound that is used largely in the production of plastics and can be present in finished materials (Careghini et al. 2015). Its presence is diffuse throughout the environment and may be concentrated in landfills and wastewater treatment plant effluent, which can be a significant pathway. It can also persist in biosolids and end up in agricultural or forestry soils amended with biosolids. It has been detected in the bile of English sole (da Silva et al. 2013).

In general, CECs and EDCs may be transported into the environment through stormwater runoff (from urban areas, working farms, etc.) and wastewater (Lubliner et al. 2010, Morace 2012, James et al. 2016a, 2017). Combined Sewer Overflow (CSO) events can often serve as a significant conveyance (Kay et al. 2017). Septic systems have been shown to impact local groundwater and surface water, which can enter into marine systems (Dougherty et al. 2010, James et al. 2016b).

Regionally, there has been significant work characterizing the range of compounds that are present in urban stormwater runoff and determining if those are related to biological impacts (Peter et al. 2018, Peter et al. 2019, Peter et al. 2020). This work clearly demonstrates that there are a suite of compounds that are ubiquitous and unique in stormwater runoff, and these often exceed known effects thresholds in urban streams. Many of these compounds are associated with vehicle use (e.g., tire wear, fluids, etc.) though these aren't the only that had been identified.

Regarding wastewater - there are nearly 100 unique wastewater discharges that act as conveyances of CECs and EDCs into Puget Sound. In order to estimate the cumulative exposures of wastewater-associate compounds to marine biota, James et al. (2020) utilized the Salish Sea Model, a numerical model that simulates the currents and current patterns within the Salish Sea (Khangaonkar et al. 2011, Khangaonkar et al. 2017), to estimate the conservative transport of wastewater throughout the region. Results demonstrate that marine organisms from throughout the region are chronically exposed to a low level of WWTP effluent, with localized areas of higher exposure levels, largely in embayments (Figure 7-6).

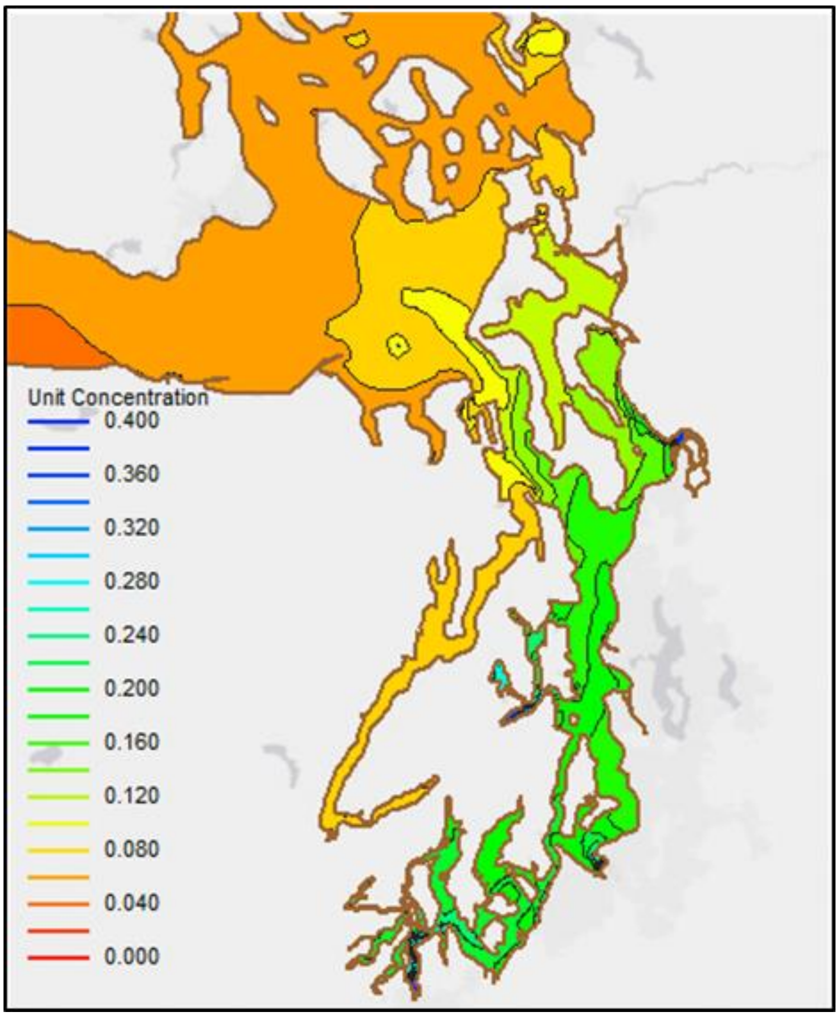

Figure 7-6. Results of a numeric simulation demonstrating the distribution of wastewater effluent from the wastewater treatment plants (n~99) throughout the Salish Sea. The modeled unit concentration in each effluent is ~100. Model results show average relative exposure concentration over ~ 3 month period. Adapted from James et al. (2020).

8 Effectiveness

This section presents a brief discussion on the effectiveness of treatment approaches at reducing toxics loading to Puget Sound. These approaches include: the construction and incorporation of physical infrastructure in the built environment, which range from traditional grey infrastructure such as vaults to low impact development (LID) and green stormwater infrastructure (GSI) approaches including rain gardens, bio infiltration swales, pervious pavement, etc.; the implementation of Best Management Practices (BMPs) such as line cleaning and street sweeping; site remediation and cleanup; and source control and product replacement. Stormwater-related controls receive particular attention since it has been identified as an important pathway for toxics loading to Puget Sound.

This review includes cost, and cost-benefit information where available.

8.1 Stormwater Management

Stormwater runoff can be treated with grey infrastructure and/or LID and GSI installations. The mass of contaminants picked up and transported by stormwater runoff can be reduced through the implementation of BMPs. The source of the contaminants to the environment can be reduced through source control. The first two approaches apply specifically to stormwater, while the latter (source control) can reduce contaminant transport through different pathways such as wastewater or air deposition; it will be addressed below.

An excellent review of the effectiveness of various treatment approaches is included in Mackenzie and McIntyre (2017). Additional important reviews are presented in Ahiablame et al. (2012) and the set of white papers by (AWC and Ecology 2013a, b, c).

8.1.1 Stormwater Management - Low Impact Development

Municipal stormwater is traditionally managed by gray infrastructure, such as curbs, gutters and piping. In contrast, LID, or green infrastructure, attempts to replicate natural flow control and contaminant removal processes through passive processes. Examples of green infrastructure (rain gardens, roof gardens, retention ponds, etc.) use natural drainage features or engineered swales and vegetated contours to infiltrate and treat stormwater runoff. They are generally decentralized installations at the parcel or sub-basin scale. LID can be effective at reducing runoff volumes and contaminant loading (see Mackenzie and McIntyre (2017) and references therein), as well as toxicity (McIntyre et al. 2014, McIntyre et al. 2015, McIntyre et al. 2016). The effectiveness of any given installation, however, is largely dependent on the design and the local site conditions. A brief summary of relevant findings is included below:

- LID installations can reduce runoff quantity through infiltration, evaporation, and transpiration. Reductions generally range from 50-90%, though are dependent on installation size and soil conditions (AWC and Ecology 2013a).

- Peak flow control through LID may be less than achieved through grey-infrastructure due to lower storage capacity (AWC and Ecology 2013a).
- LID installations generally reduce contaminant (TSS, metals, oil and grease) concentrations depending on media utilized. Export of copper and nutrients has been observed (AWC and Ecology 2013a)
- Bioretention treatment improved stormwater quality and reversed nearly all forms of developmental toxicity in zebrafish (McIntyre et al. 2014)
- The standard 60% sand 40% compost (60:40 mix) bioretention soil media specified by Ecology's stormwater management manual cleans urban stormwater runoff sufficiently to protect sensitive life history stages of salmon species (McIntyre et al. 2015).
- Permeable pavements show improvement in most water quality parameters (TSS, phosphorous, nitrogen, metals, PAHs, and herbicides) generally over 50%. Nutrients can be exported (AWC and Ecology 2013a).
- Green roofs can provide some runoff moderation depending on antecedent conditions; prolonged periods of rain can saturate the roof and so additional inputs will overflow without moderation. Contaminant loading to green roofs (and, thus potential for reductions) is low because the inputs are limited to direct rainfall, which is generally clean (Ahiablame et al. 2012)
- Known issues of LID include:
    - There is little reduction of nutrient concentrations from permeable pavement (AWC and Ecology 2013a)
    - There is documented export of copper, nitrogen and phosphorus from bioretention facilities. This is associated with leaching from bioretention media (AWC and Ecology 2013a).

8.1.2 Stormwater Management – Best Management Practices

Stormwater BMPs commonly refer to management approaches that aim to reduce the pollution after it has been released for a site and before it is transported by stormwater runoff into Puget Sound or other sensitive receiving waters. In Puget Sound, several jurisdictions have evaluated BMPs in terms of effectiveness, including the City of Tacoma and the City of Seattle (Poresky et al. 2015, Dorfmeier and Fore 2016). In addition, both jurisdictions performed cost benefit analyses of stormwater control measures, evaluating both the effectiveness of contaminant removal and cost of implementation, operations, and maintenance. Results are summarized in Tables 8-1 through 8-3.

Table 8-1. Average cost effectiveness of sewer line cleaning (line cleaning), regenerative type street sweepers used at high sweeping frequency (street sweeping), stormwater in-pipe filtration at 100% basin coverage (Direct Stormwater Filtration), and pervious pavement. Cost effectiveness was calculated using projected mass contaminant removed over 20 years divided by the total cost (capital, implementation, and operation and maintenance), in thousands of dollars, for each action (Dorfmeier and Fore 2016). DEHP, Bis(2-ethylhexyl)phthalate; TSS, total suspended solids.

Stormwater Treatment	DEHP (lbs/$1000)	TSS (lbs/$1000)	Phenanthrene (lbs/$1000)	Pyrene (lbs/$1000)
Line Cleaning	0.66	4.22	0.02	0.04
Street Sweeping	0.06	1.92	0.00	0.01
Direct Stormwater Filtration	0.02	0.56	0.00	0.00
Pervious Pavement	0.02	0.15	0.00	0.00

Table 8-2. Average cost of stormwater management actions including materials, installation, operation and maintenance (O & M) over 20 years. Street sweeping estimated for vacuum type sweepers at high sweeping frequency; stormwater in-pipe filtration estimates at 100% basin coverage.

Method	Estimated Cost (in thousands of dollars)			
	Capital	Installation	O&M	Total
Sewer Line Cleaning	--	--	$677	$677
Street Sweeping[1]	$45	--	$739	$784
Filtration[2]	$4478	$1791	$3616	$9885
Pervious Pavement	--	$9463	--	$9463

1 Vacuum type street sweepers used at high sweeping frequency

2 Stormwater in-pipe filtration at 100% basin coverage

Table 8-3. Cost effectiveness ratio and estimated cost of stormwater and small combined sewer overflow (CSO) projects included in the City of Seattle's 2014 Integrated Plan (City of Seattle 2014). Cost effectiveness estimates were compared to the sum of the small CSO projects that will be deferred as part of the Integrated Plan. See Poresky et al. (2015) for an effectiveness review of the project.

BMP	**Roadside rain gardens**	**Active treatment of runoff from industrialized areas**	**Street sweeping expansion***	**CSO projects (6 projects)**
Dissolved copper (kg/$1M)	1.7	2	N/A	0.08
Dissolved zinc (kg/$1M)	12	40	N/A	0.36
Fecal coliform bacteria (billion CFU/$1M)	66,000	151,000	8,000	13,000
Oil and grease (kg/$1M)	1,460	2,020	260	42
PBDEs (kg/$1M)	0.011	0.014	0.037	0.00071
PCBs (kg/$1M)	0.007	0.02	0.013	0.0012
Total copper (kg/$1M)	4.8	13	20	0.54
Total zinc (kg/$1M)	40	83	38	1.4
Capital Cost	$188,000 per acre	$96,000 per acre	$350 per acre	$6.5M (average)
Maintenance Cost	$700 per acre	$1300 per acre/yr	$480 per acre/yr	$30,000 per yr (average)

kg = kilograms; CFU = colony forming units; $1M = one million dollars (2014 dollars); N/A = load reduction not quantified. All values presented based on 100 year lifecycle cost estimates with 3 percent discount factor assumed. All values based on 2014 dollars.

* City of Seattle street sweeping enhancement: Planned to increase sweeping to 85% of curbed arterials (for a total 10,600 curb-miles), increasing routes from 24 to 25, enlarging route coverage, extending sweeping season from 40 to 48 weeks per year, and increasing sweeping frequency from biweekly to weekly for some routes

Key points are summarized below:

- Combining different types of stormwater source control methods provides the greatest reduction in pollutants.
- Stormwater line cleaning has been shown to provide large water quality improvements by reducing legacy pollutant loads. The line cleaning program in the City of Tacoma had the highest cost effectiveness ratios of approaches evaluated (Dorfmeier and Fore 2016)
- King County (2016) review of PCB source tracing programs found that stormwater line cleaning and resampling can be effective in determining if PCBs originate from current or legacy sources.

Once historic contamination is removed through cleaning, installing a combination of sediment traps, inline solids, and catch basin solids can isolate areas or parcels with ongoing sources

- Street sweeping expansion is expected to be the most cost effective option for reduction of suspended solids and also provide the greatest load reduction benefit in the City of Seattle. In Tacoma, street sweeping provided higher cost effectiveness ratios across all major contaminants compared to direct stormwater filtration and pervious pavement (Poresky et al. 2015, Dorfmeier and Fore 2016).
- Street sweeping effectiveness varies according to equipment, frequency, road use, and road condition (City of Seattle 2014). It has been recommended that sweeping approximately weekly is required for maximum effectiveness (Kim et al. 2014, Futurewise 2016)
- Pervious materials are an effective way to manage contaminants and reduce volume of stormwater when used in targeted locations to supplement an existing program, or in areas where site conditions are favorable for installation. Use of this application to manage stormwater is limited by installation requirements and cost (Dorfmeier and Fore 2016)
- Seattle found that roadside rain gardens, active treatment of runoff from industrialized areas, and expansion of the street sweeping program removed more pollution per dollar than small CSO projects (Poresky et al. 2015)

8.1.3 Stormwater Management - Barriers to implementing GSI

Implementing policy changes within stormwater regulations remains a challenge. To meet NPDES permit requirements, municipalities have begun to update their codes to incorporate various GSI projects within their jurisdiction (Futurewise 2016). These actions require municipal code changes, pilot projects, and coordinating with proactive developers. Although stormwater management actions are supported by local jurisdictions, many barriers still exist in the implementation of GSI around Puget Sound. Barriers fall into multiple categories including: needs for GSI maintenance, research and cost benefit analyses to decrease uncertainty when implementing BMPs, information on GSI effectiveness, increased communication and outreach, and policy guidance. The major barriers and summary of recommendations from Murphy et al. (2015) and Futurewise (2016) are outlined in Table 8-4.

Table 8-4. Barriers to LID implementation in Puget Sound identified Murphy et al. (2015) and Futurewise (2016). Results are summarized into categories of issues identified, the resulting objective aimed for by addressing the issue, and general recommendations suggested to resolve each.

Identified Issues	Objective	Recommendations
Maintenance	Minimize maintenance and failure risk with GI implementation	Training programs for private property owners
		Provide access to equipment and items for maintaining GI
Cost	Clarify BMP performance for better GI planning	Cost benefit analyses, including maintenance and repair
Research	Decrease uncertainty in BMP effectiveness at various geographic scales	Cost benefit analyses
		LID effectiveness studies at various scales
		Ground water impacts
Education and Outreach	Educate key audiences, disseminate knowledge about what's working in stormwater management, and promote behavior change	Increased communication of research and resources
		Education and training for specific audiences: decision makers, developers, public
		Fact sheets on BMPs
		Stormwater utility rate justification outreach
Policy	Increase standards and compliance to promote green infrastructure implementation and maintenance	Develop implementation guidance for different land types
		Distinguish between different management scales of responsibility (watershed, jurisdiction, region)
		Develop or improve standards and guidelines for common BMPs

8.2 Collaboration and relationship building to support change for environmental management

The effectiveness of projects and programs often pivots on support from communities and interested parties; treatment systems and remedial activities are not done in a vacuum. As such there is value in considering collaboration and relationship building in this context. A few key points are summarized below:

- Public education and behavior change programs work: behaviors can be changed and pollutants can be reduced (Fore 2013)
- Recommended approach: define the desired behavior change, determine who needs to change, identify benefits and barriers to change, remove barriers and test for changes in behavior. Work with experts to create a targeted communication campaign (Fore 2013)
- Collaboration and relationship-building among funders, policy makers, stream managers, urban land managers, and the local community are key to the success of catchment-scale changes in policy and behavior
- Researchers should be prepared to seek funding for both fully implementing catchment-scale projects and maintaining installed infrastructure over time (Bos and Brown 2015, Burns et al. 2015)
- Communities need time to become familiar with new programs and to trust in the process. If behavior change is desired, projects should have realistic expectations about community participation and engagement (Bos and Brown 2015, Burns et al. 2015)

9 Key Uncertainties

The development of coherent recovery strategies is highly dependent on the information related to status and trends, sources and pathways, etc. While much information exists, there remain uncertainties which impair the ability to identify the most valuable actions and/or responses that will lead to the improvement of the Toxics in Fish Vital Sign. Key uncertainties were gathered during several points in the development of this product including the literature review, the experts' panel, and in consultation with groups such as the PSEMP Toxics Workgroup.

Results are included in the Implementation Strategy Narrative.

10 Acronyms and Abbreviations

ATSDR	Agency for Toxic Substances and Disease Registry
BDE	brominated diphenylethers
BMP	Best Management Practice
BPA	bisphenol A
CEC	contaminants of emerging concern
CSO	combined sewer overflow
CTLd	critical tissue level (adverse effect threshold)
DOH	Washington Department of Health
dw	dry weight – concentration reported based on mass of dried sample
Ecology	Washington Department of Ecology
EDC	endocrine disrupting compounds
EDSP	Endocrine Disruptor Screening Program
EPA	U.S. Environmental Protection Agency
FAC	fluorescing aromatic compounds
HR GC/MS	high resolution gas chromatography–mass spectrometry
IARC	International Agency for Research on Cancer
LIO	Local Integrating Organization
LR GC/MS	low resolution gas chromatography–mass spectrometry
lw	lipid weight – concentration normalized based on mass of lipid in sample
MeO	Methoxylated
NEP	National Estuary Program
NGO	non-governmental organization
NOAA	National Oceanic and Atmospheric Administration
NPDES	National Pollutant Discharge Elimination System
NWFSC	Northwest Fisheries Science Center
OH	hydroxylated
PAH	polycyclic aromatic hydrocarbons
PBDE	polybrominated diphenylethers
PCB	polychlorinated biphenyls
POP	persistent organic pollutants
PSEMP	Puget Sound Ecosystem Monitoring Program
PSP	Puget Sound Partnership
TBiOS	Toxics-focused Biological Observing System for Puget Sound
TCSA	Toxic Substances Control Act
TEF	Toxic Equivalency Factor
WDFW	Washington Department of Fish and Wildlife
WDNR	Washington Department of Natural Resources
WHO/UNEP	World Health Organization/United Nations Environment Program

ww	wet weight – concentration reported based on mass of wet sample
WWTP	wastewater treatment plant

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