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Assessing Sources of Toxic Chemicals Impacting Juvenile Chinook

Snohomish River Watershed Source Assessment of PBDE Flame Retardants

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Assessing Sources of Toxic Chemicals Impacting Juvenile Chinook Salmon

Snohomish River Watershed Source Assessment of PBDE Flame Retardants

by

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Abstract

Chinook salmon play a vital role in the Puget Sound ecosystems as prey for endangered southern resident killer whales and are culturally and economically important to tribes and residents of Washington State. Chinook populations are in decline in part due to burdens of toxics accumulated during their seaward migration from Puget Sound rivers. Studies within the Snohomish River watershed have identified polybrominated diphenyl ethers (PBDEs), a group of flame retardant chemicals, in juvenile Chinook at levels which can cause sublethal impacts, reducing their survival. To determine the sources of these chemicals and how they make their way into juvenile Chinook, we performed a contaminant source assessment from 2019 to 2022 in the Snohomish, Skykomish, and Snoqualmie rivers.

Passive samplers, sediments, biofilms, and invertebrate samples were collected from throughout the watershed during low and high flow river conditions. Sources of PBDEs associated with the discharge of treated wastewater were identified in the lower Snohomish mainstem and in the Skykomish River. PBDEs were shown to accumulate in sediments, biofilms, and aquatic insects (invertebrates) throughout the watershed. PBDE concentrations in aquatic insects, and stomach contents and whole-body tissues of juvenile Chinook suggest juveniles are being exposed to PBDEs through their diet in the Snohomish estuary. The results of this study support the need for ongoing efforts to reduce PBDE levels in wastewater paired with long term monitoring of PBDEs in invertebrates and juvenile Chinook in the lower Snohomish mainstem.

Introduction

Background

Chinook salmon are listed as threatened under the endangered species act and play a vital role in the Puget Sound ecosystem as prey for endangered Southern Resident killer whales. Chinook make up to 90% of the resident whales' diets during summer months (Couture et al. 2022), and declining Chinook populations have been identified as a potential cause of the whales' declining numbers (Wasser et al. 2017). Additionally, Chinook are of cultural importance to the Native American tribes of Washington and are of commercial and recreational value.

Chinook salmon rear in freshwater environments during their early life stages then migrate through estuaries into the Puget Sound. This seaward migration causes juveniles to pass through urbanized and developed landscapes, whose restoration and mitigation have been a focal point of Chinook salmon recovery efforts. These developed riverine and estuarine landscapes can impact juvenile Chinook due to the discharge and runoff of toxic pollutants from urban and agricultural sources. Previous surveys of juvenile Chinook suggest they accumulate toxic contaminants during outmigration through urbanized and developed areas. A 2016 detailed study by Washington Department of Fish and Wildlife (WDFW) in the Snohomish River found that 73% of wild origin juvenile Chinook captured contained polybrominated diphenyl ether (PBDE) flame retardants at high enough concentrations to reduce their survival (Carey et al. 2019; O'Neill et al. 2020).

Toxics accumulation in juvenile Chinook occurs across the Puget Sound with over 30% of juveniles sampled in a 2013 study containing body burdens of toxics which could cause sublethal impacts (O'Neill et al. 2015). Similar results have been found in other studies of juvenile Chinook within the Puget Sound, suggesting that toxics accumulation at levels leading to sublethal effects impacting marine survival is common throughout the region (Sloan et al. 2010; Meador et al. 2010). The burden of toxics above certain fish health thresholds in juvenile Chinook has been shown to reduce survival through increased disease susceptibility and predation (Arkoosh et al. 2010; Meador et al. 2010). Due to the widespread impacts of toxics on juvenile Chinook in the Puget Sound region, further investigation is needed to understand where and how toxics are accumulated.

One factor that can impact a juvenile Chinook's exposure to toxics is the amount of time spent in the estuary during outmigration. Sloan et al. (2010) found that wild out-migrating Chinook had significantly higher tissue concentrations of PBDEs than hatchery-raised Chinook in the lower Columbia River and Puget Sound. Similar results were found in WDFW's 2016 survey of juvenile Chinook in the Snohomish River, where only wild origin fish had levels of PBDEs which could potentially cause sublethal effects (O'Neill et al. 2020). These findings suggest that a significant burden of toxic contaminants is acquired during the time juvenile Chinook reside in the estuarine environment.

The life history of Chinook salmon begins in freshwater streams where juveniles rear before either migrating directly to nearshore environments or spend several weeks to months in the estuary (Quinn, 2005). The length of residence time within the estuary is dependent on several

factors including habitat availability, environmental conditions, and life history type (Chamberlin et al. 2021). Chinook of wild origin tend to spend longer periods of time in the estuary, while hatchery raised Chinook are typically held back to increase growth and spend less time in the estuary (Quinn 2005). This may be why previous studies have found lower levels of toxics in hatchery Chinook.

Previous WDFW surveys of juvenile Chinook from the Snohomish estuary have measured whole body tissue concentrations of PBDEs which exceed fish health thresholds (O'Neill et al. 2015). From these surveys focused efforts have formed around determining the potential sources of toxics within the Snohomish watershed. To determine the scale and source of toxic contaminants impacting juvenile Chinook within the watershed, WDFW conducted an intensive survey throughout the out-migration pathway in 2016. The study sampled fish from throughout the estuary and upper tributaries of the Snoqualmie and Skykomish rivers. Wild juvenile Chinook residing in the mainstem of the Snohomish estuary had the highest concentrations of PBDEs in the survey. Hatchery origin fish had significantly lower PBDE tissue concentrations as well as fish collected outside the main stem of the estuary (O'Neill et al. 2020). The study identified the source of these toxics as the discharge of wastewater into the estuary based on unique contaminant patterns and nitrogen stable isotope analysis. From these findings further investigation was needed to determine sources of PBDEs in the Snohomish watershed and their pathway into juvenile Chinook.

Polybrominated Diphenyl Ethers (PBDEs)

Polybrominated diphenyl ethers (PBDEs) are a class of synthetic chemicals developed as additive flame retardants. PBDEs are composed of two phenolic rings linked by an oxygen atom surrounded by 1 to 10 bromine atoms. The chemical group is composed of 209 congeners which vary in the number and placement of bromine atoms on the phenolic rings. The chemicals are grouped into homologs based on the number of bromines and follow a naming convention from mono through deca brominated diphenyl ethers. PBDEs are considered persistent, bioaccumulative, and toxic in the environment.

PBDEs were first introduced in the 1970s as flame retardants used in electronics, plastics, upholstery foams, textiles, and various household products. Three commercial PBDE mixes have been widely used: Pentabromodiphenyl ether (c-pentaBDE), Octabromodiphenyl ether (c-octaBDE), and Decabromodiphenyl ether (c-decaBDE). It should be noted these commercial mixes contain multiple PBDE congeners and are different from the PBDE homologs referenced in this study's results. The global production of PBDEs in 2001 was estimated to be 67,000 metric tons of which about 83% was c-decaBDE (Ecology 2006). In 2004, US manufacturers voluntarily phased out the production of c-pentaBDE and c-octaBDE due to environmental and human health concerns. In 2009, the US Environmental Protection Agency (EPA) and top US manufacturers of c-decaBDE announced a voluntary phase out of production and imports to be completed by 2013. Washington State completed its own phase out of c-decaBDE in 2011 with the restrictions of its use in televisions, furniture, and other consumer and commercial electronics.

Pathways of PBDEs into the environment include volatilization and atmospheric deposition from the manufacturing, use, and recycling of PBDE containing products. Additionally, landfill leachate, stormwater, treated wastewater discharges, and biosolid applications have been identified as pathways of PBDEs entering the environment (Li et al. 2012; Morace 2012; Andrade et al. 2017). PBDEs have been shown to be carcinogenic and toxic in animals, affecting the nervous, immune, and reproductive systems (EPA 2017). Notably PBDEs are known to cause sublethal toxic effects in juvenile salmonids including Chinook. Dietary exposure to PBDEs has been shown to alter thyroid levels and increase disease susceptibility in juvenile Chinook (Arkoosh et al. 2016; Arkoosh et al. 2010).

There are no regulatory levels in Washington State for PBDEs. Previous work by WDFW has compared the tissue burden of PBDEs in juvenile Chinook to an effects threshold derived from laboratory studies at National Oceanic and Atmospheric Administration (NOAA) (Arkoosh et al. 2010;2016). The work of Arkoosh et al. (2010;2016) established a non-monotonic relationship between PBDE dose, tissue burden, and disease susceptibility, where an increased disease susceptibility was found at concentrations ≥ 470 ng PBDE / g lipid; however, this risk declines at concentrations ≥ 2,500 ng PBDE/ g lipid. O'Neill et al. (2015) relied on these tissue burdens in their assessment of juvenile Chinook throughout Puget Sound.

Goals and Objectives

The goal of this study is to identify inputs of PBDEs in the Snohomish River watershed and create a prioritized list of PBDE inputs to mitigate the impacts of toxics on out-migrating juvenile Chinook. In addition to identifying inputs, this study aims to determine the exposure pathway of juvenile Chinook within the Snohomish River.

The objectives of this study were to:

- Monitor concentrations of PBDEs in water, sediments, biofilms, and invertebrates from the Snohomish, Snoqualmie, and Skykomish rivers.
- Determine PBDE water concentration during high flow (Spring) and low flow (late summer) river conditions.
- Identify inputs of PBDEs and areas of concern within the river system
- Determine the exposure pathway of PBDEs from source to juvenile Chinook.

The Department of Ecology's (Ecology's) Toxics Studies Unit (TSU) worked in collaboration with the WDFW Toxics Biological Observation System (TBiOS) team in a coordinated effort to collect and analyze samples for PBDEs. This is part of an ongoing collaborative program between these groups focused upon identifying sources of toxic contaminants impacting juvenile Chinook salmon in Washington's river systems.

This report will outline the findings of a multi-year source assessment of PBDEs in the Snohomish, Snoqualmie, and Skykomish rivers and provide recommendations to reduce the impacts of PBDEs on out migrating juvenile Chinook.

Methods

Snohomish River Watershed (WRIA 7)

The Snohomish River watershed (WRIA 7) is in the Puget Sound drainage and is formed by the confluence of the Snoqualmie and Skykomish rivers. It is the second largest source of freshwater to Puget Sound and drains about 1,856 square miles from its headwaters in the Cascades, through the Puget Sound lowlands, into the Whidbey basin near the city of Everett. Approximately 75% of the drainage is covered in forested land with agricultural and rural land use becoming more prevalent in the lowlands (Figure 1). There are many small towns along the Snoqualmie and Skykomish rivers, the largest of which are Monroe, Sultan, and Duvall. Marysville and Everett are the largest cities within the region with a combined population of 180,469 (2020 census) and surround the Snohomish estuary from the North and South.

The Snohomish estuary consists of a mainstem and three distributary channels: Ebey Slough, Steamboat Slough, and Union Slough. There are many commercial and industrial land uses within the Snohomish estuary including maritime, lumber, sand/gravel, fabrication, and warehouse facilities. There are ten municipal wastewater treatment plants (WWTPs) within the Snohomish watershed which discharge directly to the river system (Table 1). In addition to WWTPs several combined sewer overflows (CSOs) discharge to the Snohomish River.

Table 1. Municipal wastewater treatment plants that discharge to the Snohomish, Snoqualmie, or Skykomish rivers.

Permit number, outfall location and design flow collected from the permit factsheet for each WWTP.

Municipality	Permit Number	Outfall Location	Discharge Waterbody	Design Flow (MGD)
Everett	WA0024490	48.0042, -122.1772	Snohomish River	40.3*
Marysville	WA0022497	48.0356, -122.1731	Snohomish River	12.7*
Lake Stevens	WA0020893	47.9881, -122.1399	Snohomish River	5.01
Snohomish	WA0029548	47.9126, -122.1113	Snohomish River	2.80
Monroe	WA0020486	47.8445, -121.9746	Skykomish River	2.84
Sultan	WA0023302	47.8597, -121.8206	Skykomish River	0.72
Duvall	WA0029513	47.7330, -121.9925	Snoqualmie River	1.30
Carnation	WA0032182	47.6658, -121.9252	Snoqualmie River	0.48
Snoqualmie	WA0022403	47.5392, -121.8322	Snoqualmie River	2.15
North Bend	WA0029351	47.4994, -121.7878	Snoqualmie River	2.58

*WWTP have multiple outfalls, not all of which discharge to the Snohomish River

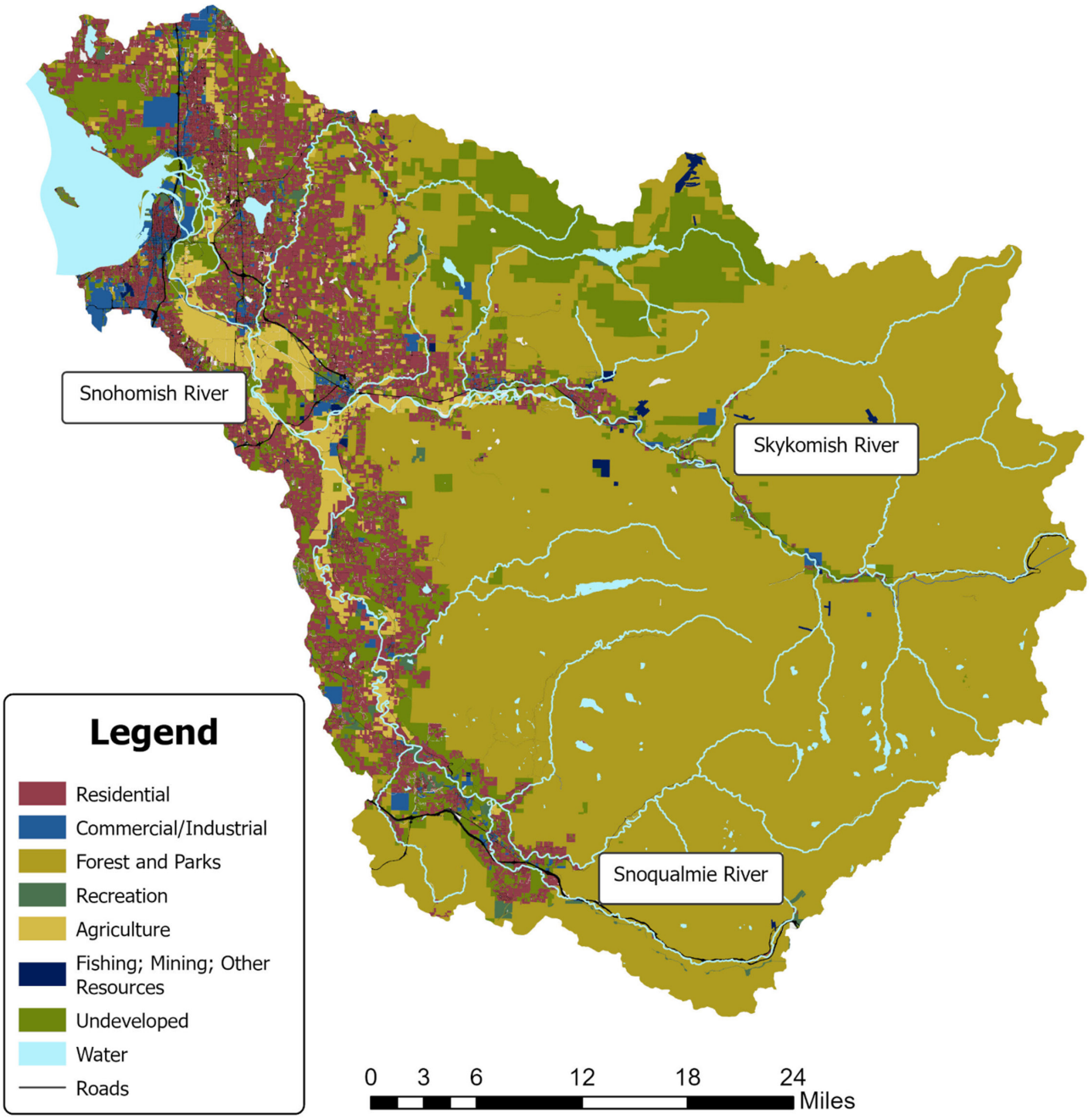

Figure 1. Land use map for the Snohomish River Basin.

Study Locations

Study locations span from the Snohomish estuary up through the Skykomish and Snoqualmie rivers. Sites were chosen based on their proximity to potential PBDE inputs as well as to provide a thorough survey of the river system. Significant evidence has shown WWTP and CSO outfalls are an important pathway of PBDEs into aquatic environments, and therefore areas downstream

of outfalls were prioritized for sampling. Additional sampling locations were selected in the estuary distributary channels (Ebey, Steamboat, and Union Sloughs) to provide a thorough survey of PBDE concentrations within the estuary. After initial sampling events, sites were added in order to bracket (upstream, downstream) locations of interest. Background sites along the Snoqualmie and Skykomish rivers were selected upstream of all WWTP outfalls to provide a reference point for PBDE concentrations. In total 31 sites were sampled over the course of the study period from 2019 to 2022. Site locations are described in Table 2 and displayed in Figure 2. Tables A9 and A11 through A13 describe sampling dates for each sample matrix.

Sampling took place during high (spring) and low flow (late summer/early fall) river conditions. The hydrology of the Snoqualmie and Skykomish rivers is both rainfall- and snowmelt-dominated, and higher flows are usually sustained throughout the winter and spring, but typically highest from April through June (Figure 2). Low flow in the rivers occurs during August and September. There is considerable tidal influence in the lower Snohomish River, up to approximately river mile 20 where the Skykomish and Snoqualmie rivers converge. Saltwater wedges have been observed and modeled in the lower Snohomish during previous water quality studies by Ecology (Cusimano 1997) and others (Hall et al. 2018). The influence of saltwater in the lower Snohomish is dependent on the time of year, tidal cycles, and upstream freshwater flow. During our high flow sampling events saltwater intrusion is minimal and freshwater inputs from the river basin dominate, whereas during low flow periods higher salinities can be measured upriver to our SNOH07.0 sample site (Hall et al. 2018).

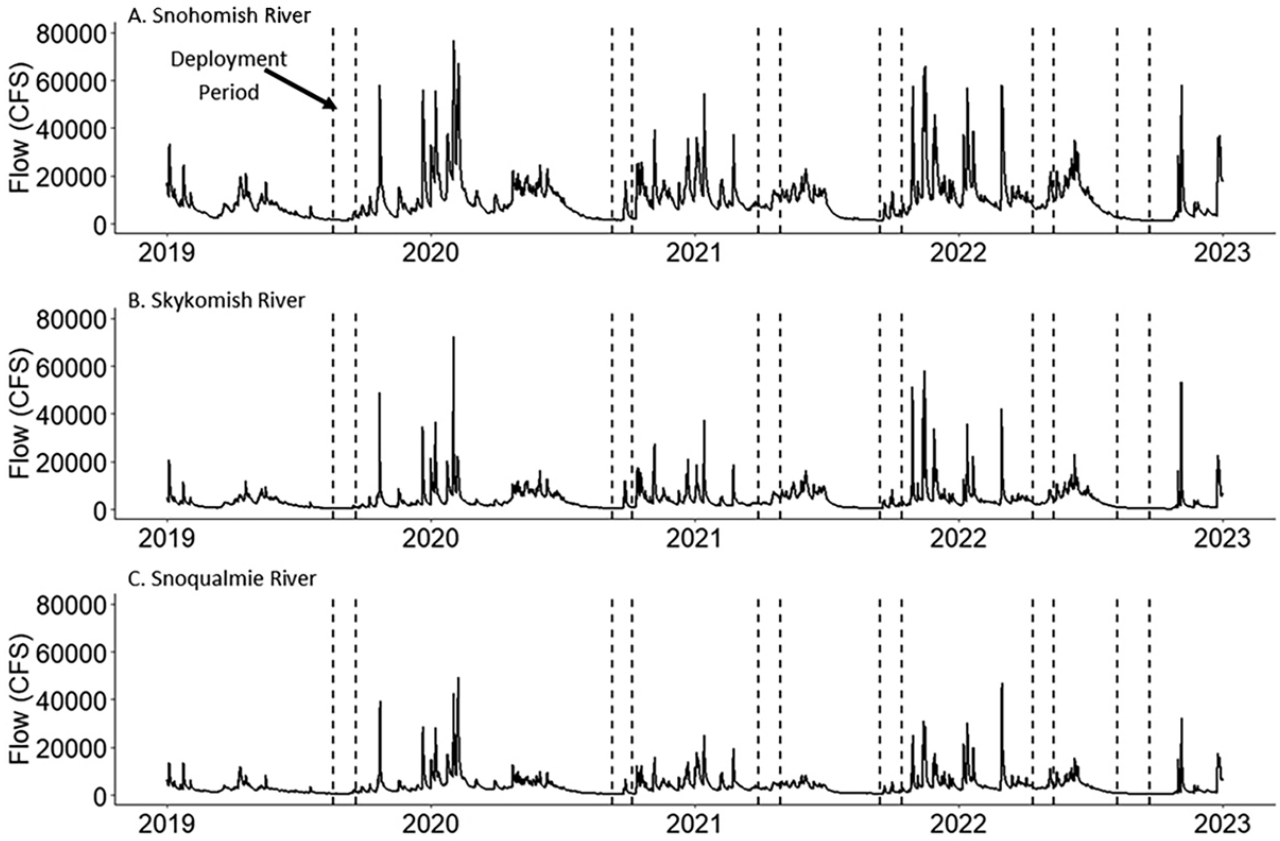

Figure 2. Hydrograph of Snohomish(A), Skykomish(B), and Snoqualmie(C) rivers from 2019 through 2022.

SPMD deployment periods from 2019 through 2022 indicated by vertical dashed lines. River flow data collected from United States Geological Survey (USGS) station 11150800 (Snohomish River) near Monroe, WA; USGS station 12134500 (Skykomish River) near Gold Bar, WA; USGS station 12149000 (Snoqualmie River) near Carnation, WA.

Table 2. Site #, site ID, name, sampling region, geographical coordinates, number of samples per matrix.

Site #	Site ID	Site Name	Sampling Region	Latitude	Longitude	SPMD	Biofilm	Benthic Sediment	Suspended Sediment	Invertebrates
1	SNOH0.0	Snohomish Mouth	Lower Snohomish Mainstem	48.0193	-122.1928	2				
2	SNOH01.8	Dagmar	Lower Snohomish Mainstem	48.0107	-122.1783	6	2	2	5	4
3	SNOH02.5	Snohomish Outfall	Lower Snohomish Mainstem	48.0043	-122.1779	2				
4	SNOH02.9	Langus	Lower Snohomish Mainstem	47.9968	-122.1794	7	1	1	2	11
5	SNOH03.9	Deadwater	Lower Snohomish Mainstem	47.9872	-122.1719	5	1	1	2	
6	SNOH05.0	Snohomish RM 5.0	Upper Snohomish Mainstem	47.9711	-122.1869	3				
7	SNOH07.0	Rotary Park	Upper Snohomish Mainstem	47.9475	-122.1903	5			5	
8	SNOH12.0	City of Snohomish Downstream	Upper Snohomish Mainstem	47.9127	-122.1138	2		1		
9	SNOH13.0	City of Snohomish Upstream	Upper Snohomish Mainstem	47.9080	-122.0923	3		1	1	
10	SNOH20.0	Lord Hill	Upper Snohomish Mainstem	47.8341	-122.0507	1				
11	SS0.0	Steamboat Mouth	Sloughs	48.0363	-122.1837	3		1		
12	SS01.7	Steamboat Upper	Sloughs	48.0228	-122.1545	3		1		
13	ES0.0	Ebey Slough Mouth	Sloughs	48.0471	-122.1802	2		1		2
14	ES1.5	Qwuloolt	Sloughs	48.0348	-122.1591	1				1
15	ES04.3	Ebey Slough RM 4.3	Sloughs	48.0065	-122.1440	1		1		
16	ES05.7	Ebey Slough RM5.7	Sloughs	47.9887	-122.1400	2				

Site #	Site ID	Site Name	Sampling Region	Latitude	Longitude	SPMD	Biofilm	Benthic Sediment	Suspended Sediment	Invertebrates
17	ES06.2	Ebey Slough @ HWY2	Sloughs	47.9789	-122.1436	1				
18	UNION M1	Union Slough 1	Sloughs	47.9984	-122.1632	1		1		
19	UNION M2	Union Slough 2	Sloughs	47.9914	-122.1644	1		1		
20	DITCH 1	Drainage Ditch	Drainage Ditch	48.0066	-122.1763	1		1		
21	SKY23.7	Monroe Downstream 2	Skykomish River	47.8390	-121.9926	1				
22	SKY24.5	Monroe Downstream	Skykomish River	47.8434	-121.9775	5	2			
23	SKY25.0	Monroe Upstream	Skykomish River	47.8463	-121.9691	3				
24	SKY34.5	Sultan Downstream	Skykomish River	47.8572	-121.8222	3	1			
25	SKY35.0	Sultan Upstream	Skykomish River	47.8591	-121.8001	2				
26	SKY40.5	Gold Bar	Skykomish River	47.8540	-121.6976	2	1			
27	SNOQ10.5	Duvall	Snoqualmie River	47.7354	-121.9922	2				
28	SNOQ23.0	Carnation Downstream	Snoqualmie River	47.6660	-121.9254	3	1			
29	SNOQ23.5	Carnation Upstream	Snoqualmie River	47.6612	-121.9277	1				
30	SNOQ40.7	PSE	Snoqualmie River	47.5394	-121.8335	2	1			
31	SNOQ47.8	Little Si	Snoqualmie River	47.4869	-121.7581	3	1			

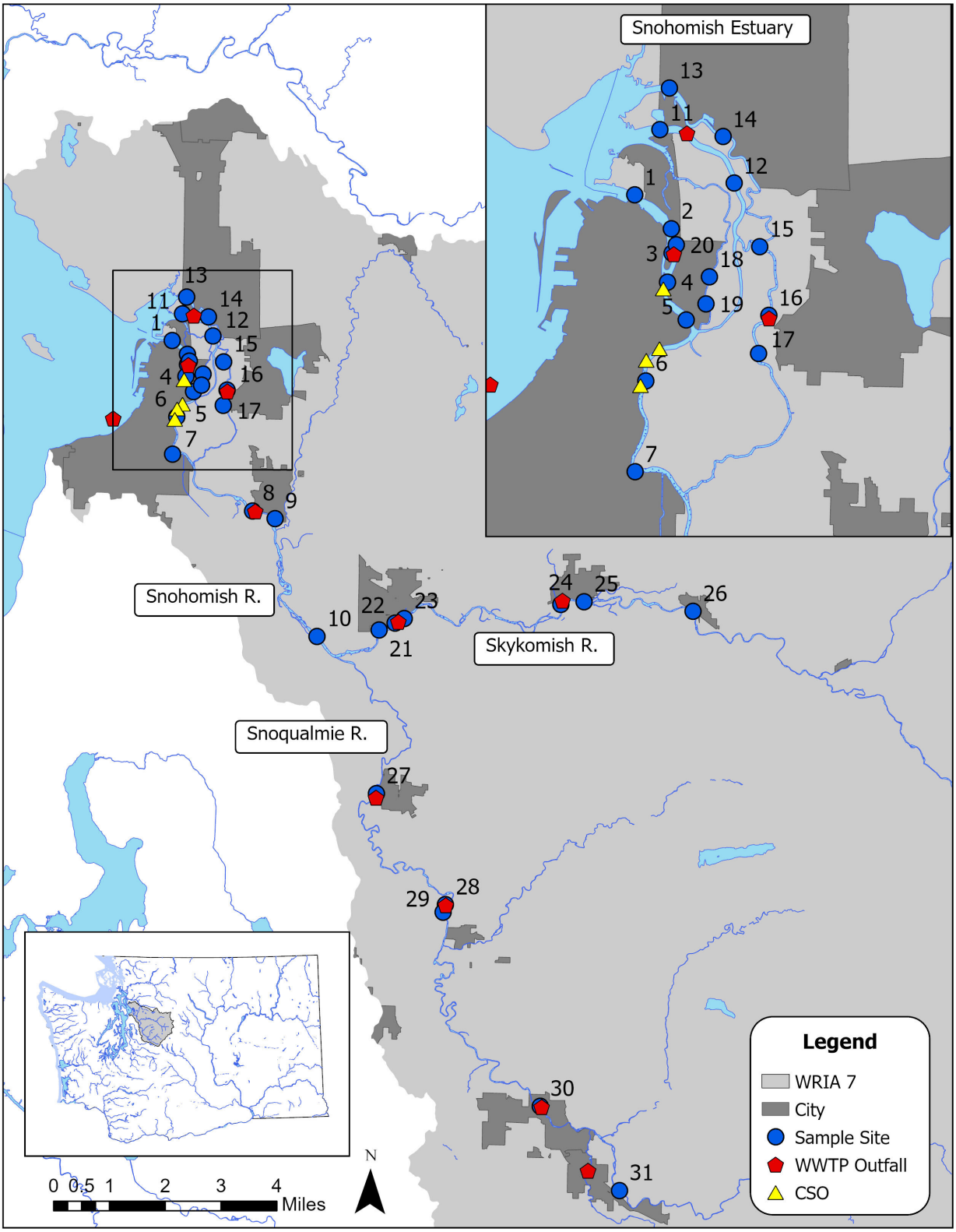

Figure 3. Snohomish, Skykomish, and Snoqualmie rivers sample sites.

Field Methods

Semipermeable membrane devices

Semipermeable membrane devices (SPMDs) were used to determine estimated PBDE water concentrations at each site. SPMDs were deployed for an average of 28 days (+/- 3 days) at each site following standard operating procedures outlined in Ecology (2019). SPMDs are composed of a low-density polyethylene (LPDE) tube (dimensions 91.4 cm x 2.5 cm x 70cm, 95 µm thickness) filled with 1 mL of triolein (high molecular weight lipid) and act as an accumulation device for lipophilic chemicals (i.e., PBDEs). SPMD strips were spiked by the manufacturer with a known mass of performance reference compounds (PRCs) (BDE-10, BDE-38, BDE-126, and BDE-138L), which are used in the modelling of PBDE water concentrations.

SPMDs were shipped from the manufacturer in sealed argon purged 5-gallon steel canisters to limit atmospheric exposure. Care was taken during deployment to limit SPMD atmospheric exposure to less than 30 seconds. Five SPMD strips mounted on stainless-steel carriers were deployed at each site within a specialized stainless-steel canister mounted inside a perforated shade canister to reduce light exposure. Deployment strategy was determined by site, but generally canisters were suspended in the water column from an immovable object (e.g., pilings, tree snag) where available or tethered to a concrete block along the river bottom. At each deployment location a temperature logger was secured to the SPMD canister and to an object above the water level. The temperature data was compared between loggers to determine if the SPMD device remained submerged throughout the sampling period.

Each deployment event included at least two blanks, a field blank and a manufacturing blank. Field blanks consisted of five SPMD strips sealed in a 1-pint steel canister and were exposed to the atmosphere for 30 seconds during the deployment and retrieval events. A manufacturing blank was included with each sampling event and was spiked with PRCs and sent directly to the analytical lab from the manufacturer. All SPMD samples and field blanks were stored at -20°C before and after deployment and shipped on blue ice to the laboratory for PBDE analysis.

Surface Water Grab Samples

Surface water samples were collected at each sampling site three times during the SPMD deployment period. Water samples were collected following standard operating procedures outlined in Ecology (2022a). Grab samples were analyzed for total and dissolved organic carbon (TOC/DOC) and suspended sediment concentration (SSC).

Ancillary water chemistry parameters were collected at each site during grab sample collection. A multiparameter sonde (YSI EXO2) was used to measure pH, specific conductivity, temperature, and dissolved oxygen.

Benthic Sediments

Benthic sediments were collected from a subset of sampling sites during the 2019 low flow sampling event. Site selection was based on the availability of collectable sediments which were limited to the estuary and mainstem of the Snohomish River. Three additional sites were sampled for sediments in 2021. Sediments were collected using a Petite Ponar following standard operating procedure outlined in Ecology (2023). Triplicate Ponar samples were

collected at each site and composited before being subsampled for PBDEs, TOC, total nitrogen (TN), grain size, and stable carbon and nitrogen isotopes.

Suspended Sediments

Suspended sediments were collected from a subset of sites during 2021 and 2022 sampling events. Custom built sediment bottle traps (Figure 4) were used to collect suspended sediments over an approximately 30-day period. Bottle traps were deployed from pilings with the sampling bottles submerged within the water column. Due to tidal fluctuations during the deployment period, sampling depth was not fixed within the water column. Each trap consisted of four pre-cleaned 1L amber glass bottles which passively collected suspended sediments. At the end of the sampling period bottles were capped, transported on ice, and stored at 2°C until being processed. Suspended sediment samples were dewatered by centrifugation at 1000 RPMs for 15 minutes. Dewatered sediments were analyzed for PBDEs, % solids, TOC, and TN.

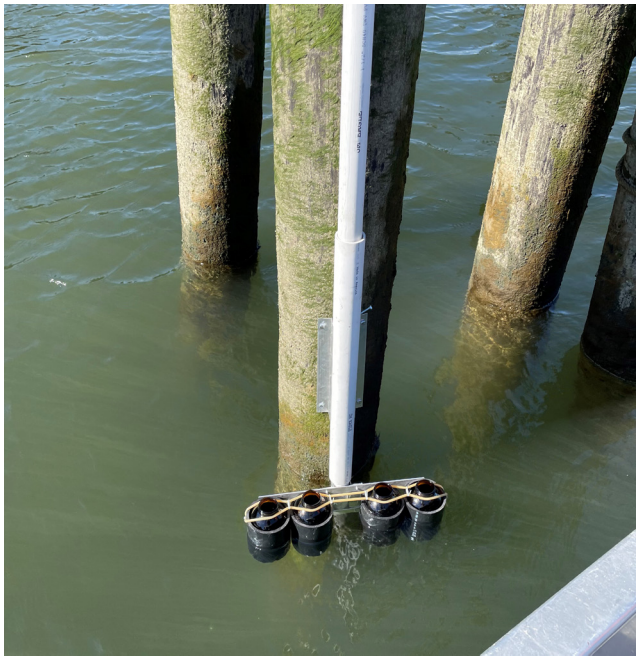

Figure 4. Telescoping suspended sediment bottle trap.

Biofilms

Biofilms were collected from a subset of sampling sites during the 2019 low flow sampling event. Sites were selected due to the presence of collectable biofilms and ranged from the lower Snohomish estuary to the upper Skykomish and Snoqualmie rivers. Biofilms were collected following standard operating procedures for the collection of attached algae (Stevenson and Bahls 1999; Ecology 2022b). At each site biofilms were scraped from river substrates (i.e., rocks and docks) and collected in a stainless-steel bowl. Approximately 10 grams wet weight (ww) of biofilms were collected at each site and transferred into a precleaned glass jar. Biofilm samples were stored on ice while in the field before being frozen at -20°C until submission for PBDE and stable isotope analysis.

Invertebrates

Invertebrate samples were collected from select sites within the Snohomish estuary to gauge the presence of PBDEs in potential juvenile Chinook prey items. Invertebrate collection was limited to sites where invertebrates were present in sufficient amounts to collect. Invertebrates were initially collected during the 2019 low flow sampling event by picking through biofilms. In later collections, during the 2021 and 2022 sampling events, a zooplankton net (60 cm ring net with 200µm mesh) was used to collect invertebrates from the water column. The zooplankton net was deployed at two locations within the Snohomish estuary (SNOH02.9 and ES0.0). Nighttime ebb tides were selected for deployment consistency and due to their higher abundance of invertebrates. The net was deployed at 15 minute intervals, before being retrieved and emptied. Re-deployment took place until sufficient biomass (~10 g ww) was collected. Typically, between 4 and 8 deployment intervals were needed to collect sufficient biomass. The collected biomass was stored in precleaned glass jars on ice during transport and at 2°C until processing within 48hrs of collection. Processing consisted of sieving samples to remove debris and then picking invertebrates from the remaining sample material. A visually representative subsample of individuals was taken for taxonomy and stored in 95% ethyl-alcohol. After processing, samples were homogenized with a mini emulsifier and sub sampled for stable isotopes. Samples were stored at -20°C until submission for PBDE and stable isotope analysis.

Nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) stable isotopes were measured on tissues from several sampling events. The goal of the analysis was to provide further clarity on potential contaminant sources (e.g., treatment plant effluent, Hicks et al. 2017) and use as a possible proxy for the trophic position of study organisms (Post et al. 2002). It is the ratio of stable N isotopes that is often used in ecological studies to investigate questions like ours. Generally, the N isotopic ratio will increase by ~ 3.4 ‰ with each trophic level, due to the excretion of the lighter isotope, ^{14}N, over time (Cabana and Rasmussen 1996; Post 2002). Changes in N source and cycling can also be reflected in the $\delta^{15}N$ of organic N; discrimination of the heavier isotope (^{15}N) leads to fractionation effects during reactions like nitrification, remineralization (ammonification) and denitrification – all of which take place in wastewater treatment plants.

Laboratory Methods

PBDE samples (SPMDs, sediments, biofilms, and invertebrate tissues) were shipped overnight to SGS AXYS in Sidney, B.C., Canada for PBDE analysis. Samples were analyzed for 46 target PBDEs by high resolution gas chromatography/high resolution mass spectrometry (HRGC/HRMS) following laboratory procedure *MLA-033 Analytical Method for the Determination of Brominated Diphenyl Ethers (BDE) and Other Brominated Flame Retardants Revision 06, Version 12* that is based on EPA Method 1614A.

Manchester Environmental Laboratory in Port Orchard, WA performed the following analyses: TOC, DOC, and SSC (water), and TOC/TN and % solids (2021 benthic sediments and all suspended sediments). TOC and DOC water samples were analyzed by method SM5310B. SSC samples were analyzed using ASTMD3977B. TOC/TN in sediments were analyzed using EPA440.0 and % solids by SM2540G. Sediment samples collected in 2019 were analyzed for TOC/TN by UC Santa Cruz Stable Isotope Laboratory in Santa Cruz, CA following method EPA440.0.

Sediment grain size was analyzed using method PSEP 1986 Wet Sieve at Materials Testing and Consulting Inc., Olympia, WA in 2019, and at ALS Environmental, Kelso, WA in 2022.

Carbon and nitrogen stable isotopes in sediments, biofilms, and invertebrate tissue samples collected in 2019 were analyzed by UC Santa Cruz Stable Isotope Laboratory, using a nanoEA-iRMS instrument. Invertebrate tissue samples collected in 2021 and 2022 were analyzed at NOAA's Northwest Fisheries Science Center in Seattle, WA, using a Fisher Scientific Flash 2000 Elemental Analyzer coupled with the Conflo IV interface and analyzed using a Delta V Advantage Isotope Ratio Mass Spectrometer.

Data Reporting and Analysis

Dissolved water concentrations of PBDE congeners were estimated from the mass measured in SPMDs using the USGS SPMD calculator v5.1 (Alvarez 2010). The calculator is based on models developed by Huckins et al. (2006) and utilizes octanol water partitioning coefficients, chemical sampling rates, PRC loss, mass of chemicals sampled, and physical properties of SPMDs to estimate PBDE water concentrations. PRCs BDE-10 and BDE-126 were used in the calculator due to their consistent recovery across all data batches. An average of the estimated water concentration for field blanks and manufacturing blank (day zero) samples was calculated for each batch of SPMDs. The average blank water concentration for each PBDE congener was subtracted from the samples' estimated PBDE water concentration to account for any potential contamination during the manufacturing and deployment of SPMDs. Total PBDE concentrations were calculated for SPMDs as the sum of the di- through octa-BDE congeners excluding results qualified as non-detects (U/UJ) and NJ. Non-detects were treated as zero during the summation of total PBDEs.

Total PBDE concentrations were calculated for sediments, biofilms, and invertebrate tissues as the sum of 43 BDE congeners. Non detects (U/UJ) and results qualified as NJ were not included in the PBDE total calculations.

Spatial and temporal trends in PBDE results were explored using maps and plots to determine locations and timing of elevated PBDE levels. Relative concentrations of PBDE homologs (di-BDE, tri-BDE, tetra-BDE, penta-BDE, hexa-BDE, hepta-BDE, octa-BDE, nona-BDE, deca-BDE — see Acronyms and Abbreviations list for expansions) were calculated and used to explore differences in homolog proportions between sites, flow conditions, and environmental matrixes.

Quality Assurance/Quality Control

All PBDE results were validated by an independent data validator at Manchester Environmental Laboratory using a stage 2B data validation per the technical specification of the following:

- SGS-Axys Method MLA-033 Analytical Method for the Determination of Brominated Diphenyl Ethers (BDE) and Other Brominated Flame Retardants Revision 06, Version 12.
- EPA Method 1614A Brominated Diphenyl Ethers in Water, Soil, Sediment, and Tissue by HRGC/HRMS, May 2010.
- Quality Assurance Project Plan: Assessing Sources of Toxic Chemicals Impacting Juvenile Chinook Salmon; Publication No. 19-03-110, August 2019.
- National Functional Guidelines for High Resolution Superfund Methods Data Review, November 2020.

A description of data quality was provided by the data validator for each batch of results in the form of a written narrative. The validation report included information on laboratory quality control checks, including calibration, ongoing precision and recovery, laboratory blanks, and recovery standards. Quality control checks were assessed against the study's measurement quality objectives outlined in the quality assurance project plan.

Results

QA/QC results

Table A1 provides a summary of the data quality results based on this study's measurement quality objects (MQO). Overall, PBDE results were deemed acceptable with a few exceptions. A summary of qualified results is described below.

PBDE analysis results were reported down to the estimated detection limit for individual congeners. In some cases, reported detection limits exceeded the upper range of sensitivity specified in the MQO. Estimated detection limits are summarized in Table A2.

Several SPMD, tissue, and sediment sample results were qualified due to method blank contamination. Detected sample analyte results <5x the detected method blank result were censored due to method blank contamination and qualified as not detected (UJ qualifier). Method blank censoring rates are summarized in Table A1.

Ongoing Precision and Recovery (OPRs) samples, consisting of a spiked laboratory sample, were included in each extraction batch. All OPR samples met the recovery criterion of 50% – 150% for each media.

Twelve surrogates and one recovery standard were included in each sample analysis. All surrogates in sediments and tissues met the recovery criteria. Low surrogate recoveries for BDE-100L and BDE-209L in SPMDs occurred in <1% and 33% of samples, respectively. BDE-209L recoveries in SPMD samples were consistently at the lower end of the acceptable range (20 – 200%) (Figure A1).

Field and laboratory duplicate samples were analyzed with some batches of SPMDs, tissues (biofilm and invertebrates), and sediment samples. Duplicate samples were not analyzed with some batches due to lost SPMDs and limited tissue volumes. Relative percent difference (RPD) was calculated for results >5x the LOQ in both samples. The duplicate criteria of <50% for RPDs were exceeded for sediment samples from 2019 for BDE-47, BDE-99, BDE-100, BDE-153, and BDE -154, and from 2021 for BDE-154, BDE -206, and BDE -209, and from 2022 for BDE-206, BDE -207, BDE -209. Duplicate tissue samples from 2022 exceeded criteria for BDE-79. 2022 SPMD duplicate samples exceeded the criteria for BDE-105, BDE-181, and BDE-190. Congener results were qualified as estimates (J) for samples and their duplicates which exceeded the criteria. High RPDs in sediment and tissue samples could be due to the natural heterogeneity of these matrixes.

SPMD field blanks and manufacturing blanks (Day Zero SPMDs) were assessed for each sample batch and used to blank subtract calculated PBDE water concentrations. Blank subtraction was performed on individual PBDE congeners based on the average SPMD blank congener concentration for each batch. Total PBDE concentrations in SPMD blanks ranged from 1.47 to 8.76 pg/L (Table A3). The blank subtraction values (i.e., average SPMD blank concentration per batch) were similar between the six SPMD batches, ranging from 2.14 to 5.02 pg/L. Tetra, penta, and octa-BDEs contributed the highest concentrations to blank subtraction values (Table A3).

SPMD Nona and Deca-BDE censoring

Nona and deca-BDEs were censored from SPMD results due to their low theoretical uptake rate, poor surrogate recoveries, and consistent detection in method blanks and SPMD manufacturing blanks.

SPMD chemical uptake rates are modelled in part on water-octanol partitioning coefficients (K_{OW}) for each PBDE congener. Experimentally derived uptake rates have determined SPMDs efficiently sample chemicals with log K_{OW}s between 3 and 8.5 (Huckins et al., 2006). Nona and deca-BDEs' log K_{OW}s range from 10.6 to 11.2 and therefore have a limited theoretical uptake to SPMDs. Additionally, SPMD membrane diffusion, or the transfer of chemicals from water across the membrane barrier into SPMDs is limited by molecular volume and weight (Huckins et al., 2006), therefore nona- and deca-BDEs molecular size and weight limit their diffusion into SPMDs.

Concentrations of nona- and deca-BDEs were detected in SPMD samples but due to low surrogate recoveries and potential degradation during analysis these results could be biased. Surrogate recoveries of BDE-209L were consistently low with 90% of samples having recoveries below 40%, and 33% of samples having recoveries below the MQO threshold of 20%. BDE-209L recoveries are used in the calculation of nona- and deca-BDE concentrations as part of the isotopic dilution analysis method. Due to the low recovery of BDE-209L, nona- and deca-BDE result could be biased high. Additionally, thermal degradation of BDE-209 during analysis can cause the formation of nona-BDEs, causing nona-BDEs to be biased high, and deca-BDE to be biased low.

Nona- and deca-BDE were frequently detected in method blanks and SPMD manufacturing blanks (field blanks and day zero samples) (Table A4), indicating potential sample contamination. These homologs were also frequently detected in field deployed SPMD samples but at similar concentrations to those detected in manufacturing blanks (Table A4). Due to consistent detection and the concentration range of nona- and deca-BDEs in blank samples it is difficult to decern true hits from sample contamination in field deployed SPMDs.

For these reasons nona- and deca-BDE results in SPMDs were excluded from analysis and reporting in this study.

Ancillary Parameters

Results for ancillary parameters (TOC, DOC, SSC, TN, grain size and % solids) for water, sediment, and biofilm samples are provided in Tables A5 thru A8.

TOC and DOC concentrations in water samples ranged from the method reporting limit (0.5 mg/L) to 6.0 and 5.4 mg/L, respectively. TOC and DOC mean concentrations were similar across the watershed and between high and low flow river conditions. SSC ranged from the method reporting limit (0.5 mg/L) to 44 mg/L within the watershed. SSC differed slightly across the waterbodies with highest mean concentration measured in the sloughs and lowest in the Skykomish River. Generally higher SSC concentrations were measured during low flow conditions than high flow.

Benthic sediments, collected mainly in 2019, were composed of fine to very fine grain sands and silts (Table A5). Percent TOC and TN in sediment samples ranged from 0.6 to 5.7% and 0.1 to 0.4%, respectively. Sediments collected from Union Slough had notably higher percentages of TOC than other sloughs and the Snohomish mainstem. Suspended sediment %TOC ranged from

0.9 to 3.3% and was higher at each site when compared to benthic sediments. Percent TN ranges were similar for suspended and benthic sediments. TOC and TN results were similar between high and low flows for suspended sediments except at site SNOH02.9. Insufficient sample volume was collected to assess grain size of the suspended sediments, however visual inspection indicated mostly very fine sands and silts.

Biofilms ranged from 2.1 to 9.4% in TOC and 0.4 to 1.7% for TN. Similar levels of lipids were found in all biofilm samples (0.1 to 0.3%).

Temperature data was collected during each SPMD deployment to assess whether the sampler remained submerged for the entire ~30 days. After SPMD retrieval water and air temperature logs were assessed to determine if SPMDs were exposed to the atmosphere for a significant period of time. Exposed SPMD samples were not analyzed due to potential contamination from atmospheric borne PBDEs. Two SPMD samples from the 2022 low flow sampling events were not analyzed due to atmospheric exposure.

PBDEs in Water

Estimated water concentrations of PBDEs in the Snohomish, Skykomish, and Snoqualmie rivers were measured through the deployment of SPMDs during 6 sampling events (4 low flow & 2 high flow) from 2019 to 2022 (Table A9). Total PBDE concentrations were expressed as the sum of di- through octa-BDEs. The river system was divided into 5 sampling regions for comparison: lower Snohomish mainstem, sloughs (distributary channels), upper Snohomish mainstem, Skykomish River, and Snoqualmie River (Table 2). The lower Snohomish mainstem and sloughs make up the Snohomish estuary.

Total PBDE water concentrations throughout the watershed ranged from 1.2 to 87.5 pg/L with the lowest concentration measured at the Snoqualmie River reference site (SNOQ47.8) and highest at SKY24.5 near the city of Monroe. The lower Snohomish mainstem had the highest mean concentration of PBDEs across the 5 sampling regions (Figure 5A) during high flow conditions (44.5 pg/L), while the Skykomish River had the highest mean concentration during low flow conditions (29.5 pg/L)(Figure 6A). The mean low flow concentration in the Skykomish River was heavily influenced by high levels of PBDEs at site SKY24.5.

The lower Snohomish mainstem consistently had higher PBDE levels during high flow conditions in comparison to sites upstream (upper Snohomish mainstem) and other areas of the estuary (sloughs) (Figure 5). Mean high flow PBDE concentrations in the lower Snohomish mainstem were 2.5 – 3.3 times higher than the mean concentration measured in the sloughs and upper Snohomish. This pattern of elevated concentrations was only observed during high flow conditions. Lower Snohomish mainstem mean low flow PBDE levels were ~2.5 times lower than high flow concentrations and similar to the upper Snohomish and sloughs.

Skykomish River PBDE concentrations were highest during low flow conditions and downstream of the towns of Monroe and Sultan (Figure 6A). PBDE concentrations at SKY24.5, located downstream of Monroe's WWTP outfall, ranged from 36.2 to 87.5 pg/L during low flow conditions. The mean concentration upstream of the outfall (site SKY25.0) was 10.7 pg/L, demonstrating a ~3 – 8 fold increase in concentration between upstream and downstream sites, during low flow conditions. PBDE levels downstream of Sultan's WWTP outfall (site SKY34.5), ranged from 7.9 to 25.1 pg/L during low flow conditions. Downstream concentrations (SKY34.5)

were 3.7 times higher than upstream concentrations (SKY35.0) during low flow conditions in 2021. Concentrations of PBDEs at the Skykomish River reference site (SKY40.5) during low and high flow conditions were 2.3 and 28.2 pg/L, respectively. The reference site was the only location along the Skykomish where PBDE concentrations were higher during high flow conditions.

Among the five sampling regions, mean water concentrations of PBDEs were lowest in the Snoqualmie River. In addition, compared to the other sampling regions, mean PBDE concentrations in the Snoqualmie River were relatively similar across sites and river flow conditions (Figure 6B). Snoqualmie River PBDE concentrations ranged from 2.5 – 15.4 pg/L during low flow conditions, and 2.9 – 7.6 pg/L during high flow. Mean PBDE concentrations during low and high flow were 5.8 pg/L and 6.8 pg/L, respectively. The narrow range of site concentrations and similar mean concentrations between river flow conditions demonstrates PBDE concentrations are similar throughout the Snoqualmie, and near reference site levels.

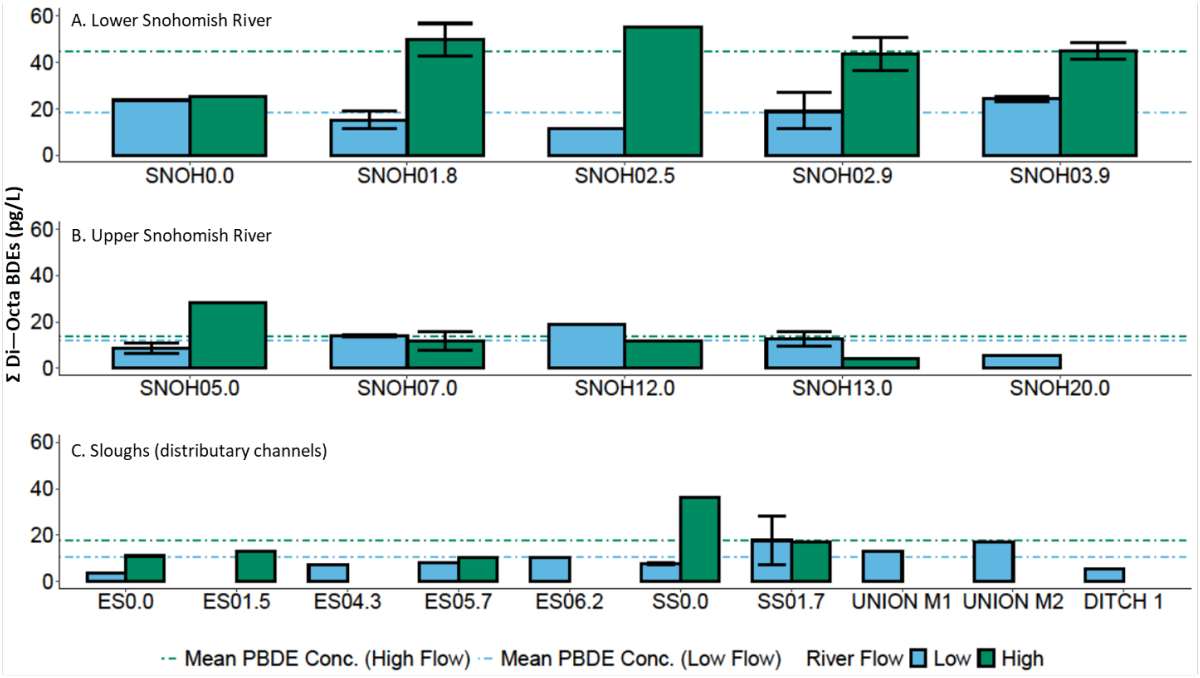

Figure 5. PBDE water concentrations in the Snohomish River and Estuary during low and high flow river conditions.

Total PBDE (sum of di- through octa-BDEs) water concentrations for each station and river flow condition (high/low) are shown. Where more than one sampling event collected data from a site, the concentration represents the mean. Error bars represent the range of PBDE water concentrations at each site. Where no error bars are present only one sample was collected at the site and the point value of that measurement is shown. Where a blank space is present no sample was collected for the flow condition. Mean PBDE concentrations for each sampling region (Lower Snohomish mainstem, Upper Snohomish mainstem, and Sloughs) during high and low flow conditions are indicated by horizontal dashed lines.

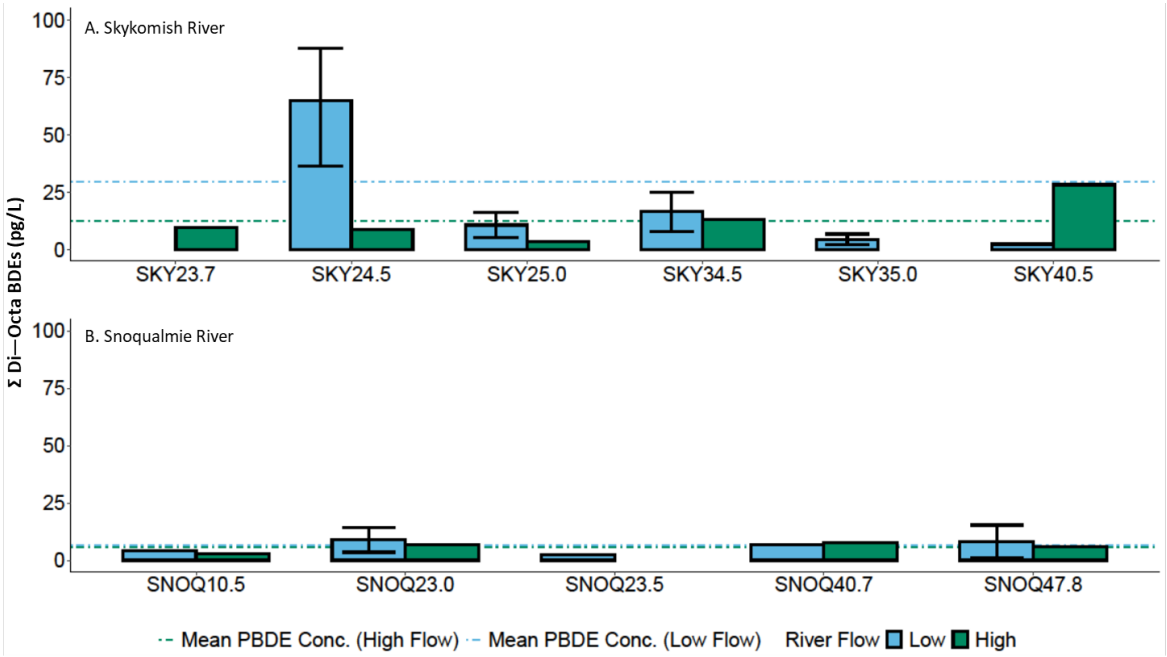

Figure 6. PBDE water concentrations in the Skykomish and Snoqualmie rivers during low and high flow river conditions.

Total PBDE (sum of di- through octa-BDEs) water concentration for each station and river flow condition (high/low) are shown. Where more than one sampling event collected data from a site, the concentration represents the mean. Error bars represent the range of PBDE water concentrations at each site. Where no error bars are present only one sample was collected at the site and the point value of that measurement is shown. Where a blank space is present no sample was collected for the flow condition. Mean PBDE concentrations for each sampling region (Skykomish and Snoqualmie rivers) during high and low flow conditions are indicated by horizontal dashed lines.

PBDE homolog compositions of water were predominantly made up of tetra-, penta-, and hexa-BDEs though relative concentrations of these homologs varied by sampling region and flow (Figure 7). These three homologs were detected at all sites and when summed made up between 45.0 and 99.4% of the total PBDE concentration. While mean relative concentrations for tetra- and penta-BDE were relatively similar across regions, relative concentration ranges were notably broader in the sloughs and Skykomish and Snoqualmie rivers compared to the other sampling regions (Table A10). Additionally, tetra- and penta-BDE relative concentration ranges were larger during low flow conditions.

Di- and tri-BDE homologs were detected at all sites except SNOQ23.0 and made up a minimal proportion of the total PBDEs measured during both flow conditions (Figure 7). Mean relative concentrations ranged from 0.2 – 4.9% for di- and ND – 12.9% for tri-BDE. Sampling region means and ranges were similar within and between flow conditions (Table A10).

Hepta- and octa-BDE homologs were sporadically detected across the river system at a wide range of relative concentrations (Table A10). Octa-BDE homolog was detected in all river zones but only during low flow conditions (Figure 7). Relative concentrations of hepta-BDE homolog group were below 2.0% and were predominately detected during low flow conditions.

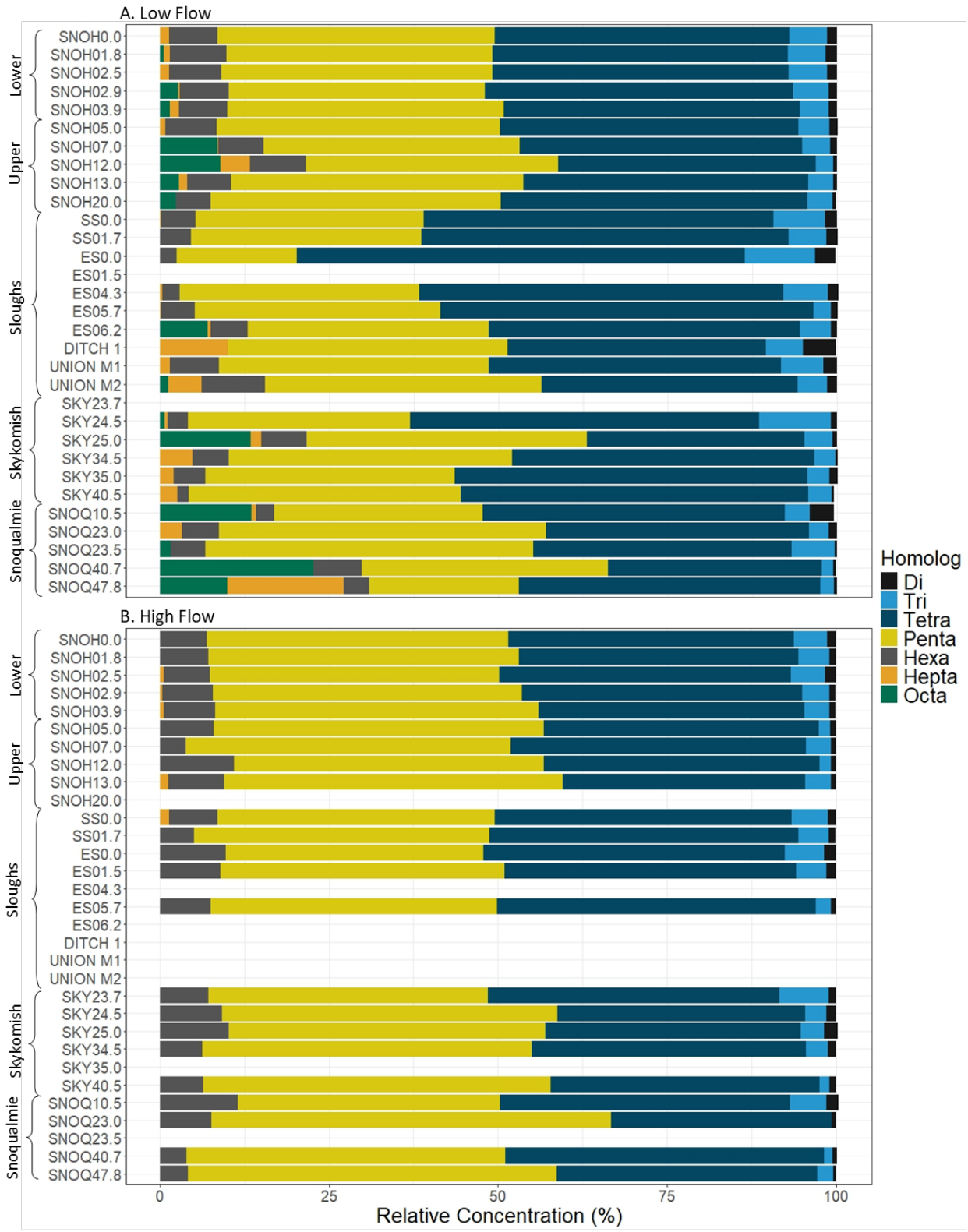

Figure 7. Relative concentrations of PBDE homologs in water from the Snohomish, Skykomish, and Snoqualmie rivers during low flow (A) and high flow (B) river conditions.

Relative concentrations were calculated as [PBDE homolog concentration]/ [Sum di through Octa-BDE concentration].

PBDEs in Sediments

Benthic sediment samples were collected from a subset (n=9) of sites in the Snohomish estuary during the 2019 low flow sampling event. Three additional sites were sampled for benthic sediments in 2021. Results of these surveys are summarized in Table A11. Total PBDE concentrations within the lower mainstem of the Snohomish estuary ranged from 130 to 1395 pg/g. The highest total PBDE concentration was measured at site SNOH13.0 and lowest at SNOH02.9 (Figure 8). The sloughs' (Ebey, Union, and Steamboat) total PBDE concentrations ranged from 346 to 772 pg/g, with the highest concentration measured in Union Slough (UNION M1) and lowest near the mouth of Steamboat Slough (SS0.0).

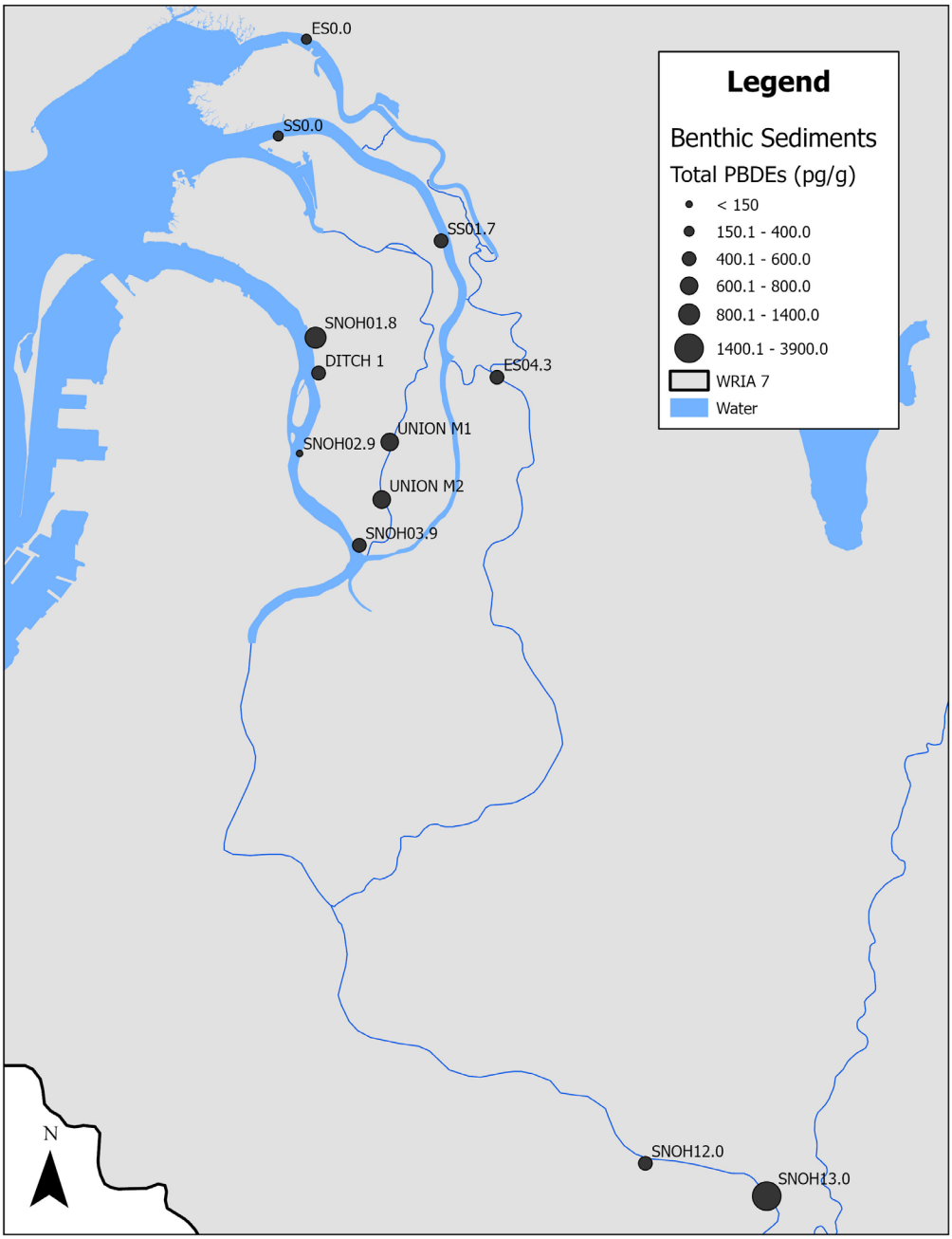

Figure 8. Total PBDE Concentration in benthic sediments.

Suspended sediments were collected along the mainstem of the Snohomish River (sites: SNOH01.8, SNOH02.9. SNOH03.9, SNOH07.0, and SNOH13.0 (2022 only)) during the 2021 low flow and 2022 high flow sampling events. Suspended sediment total PBDE concentrations for the 2021 and 2022 events ranged from 1,323 to 2,504 pg/g and 230 to 1,659 pg/g, respectively (Figure 9). PBDE concentrations were highest at site SNOH01.8 and lowest at SNOH13.0. Total PBDE concentrations were consistently lower at each site during the 2022 high flow event but demonstrated a similar pattern of concentration differences between sites (Figure 9).

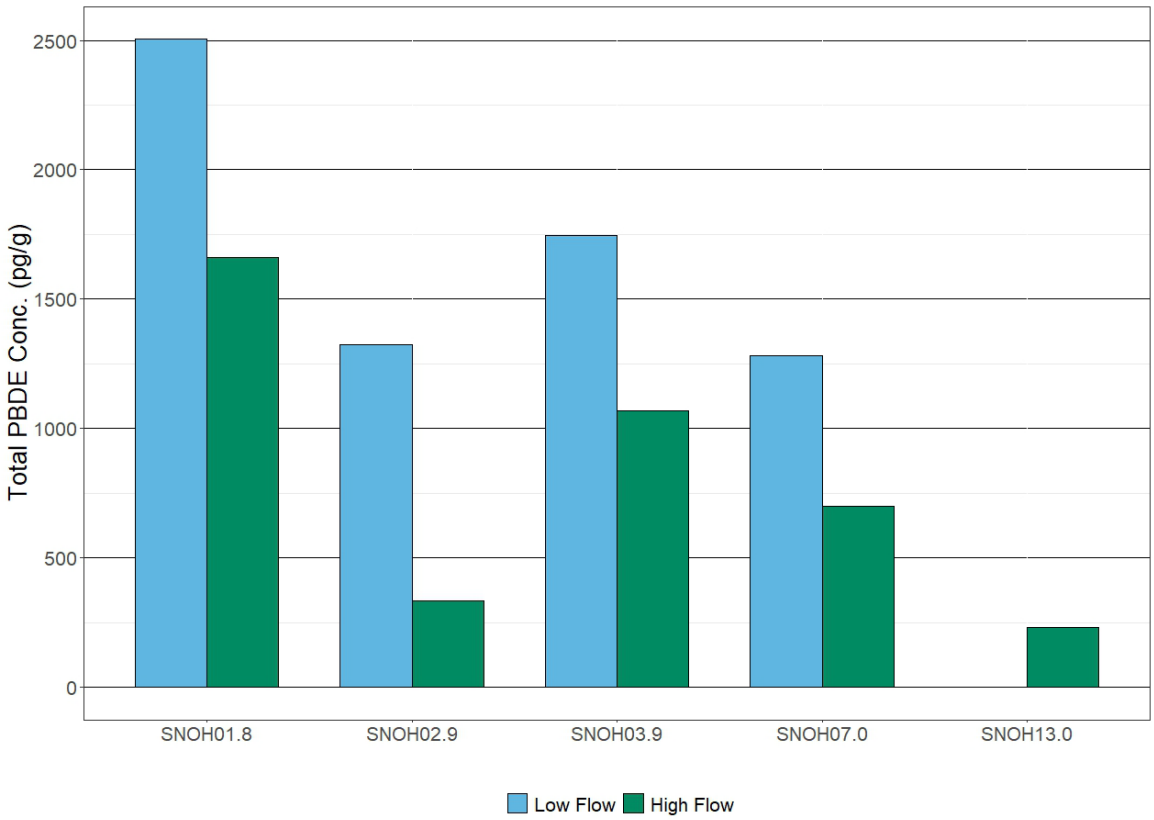

Figure 9. Low vs high flow suspended sediment total PBDE concentrations.

PBDE homolog compositions in benthic sediments were dominated by deca-BDE which accounted for 41.7% – 71.6% of the total PBDE concentration when detected. Deca-BDE was detected in all benthic sediment samples except SNOH02.9 which had a distinctive homolog composition dominated by tetra- and penta-BDEs. Generally, tetra- and penta-BDEs accounted for 4.2% – 18.0% and 7.5% – 23.5% of the total PBDE concentration, respectively. The sum of tetra-, penta-, and deca-BDEs accounted for 72.6% – 91.9% of the total PBDE concentrations in benthic sediment samples.

Homolog compositions in benthic sediments collected from the sloughs were more homogenous (ES0.0, ES04.3, SS0.0, SS01.7, Union M1 & M2) than those collected from the lower Snohomish mainstem (SNOH01.8, SNOH02.9, SNOH03.9) (Figure 10 A). Deca-BDE relative concentrations varied from 57.1% – 64.0% in the sloughs while ranging from 0.0% – 71.6% in the mainstem. The differences in compositions were primarily seen in deca-BDE with tetra- and penta-BDEs being comparable between the mainstem and sloughs except for SNOH02.9.

Suspended sediment PBDE homolog compositions closely resemble those of benthic sediments, with deca-BDE being the dominant homolog measured (Figure 10 A&B). Deca-BDE was detected in all samples and ranged in relative concentration from 40.3% – 70.9%. Relative concentrations of tetra and penta-BDE ranged from 5.8% – 15.5% and 8.1% – 29.9%, respectively. There were slight differences in homolog compositions between low and high flow sampling events. Low flow suspended sediment deca-BDE relative concentrations on average were slightly higher, and tetra and penta-BDEs were slightly lower.

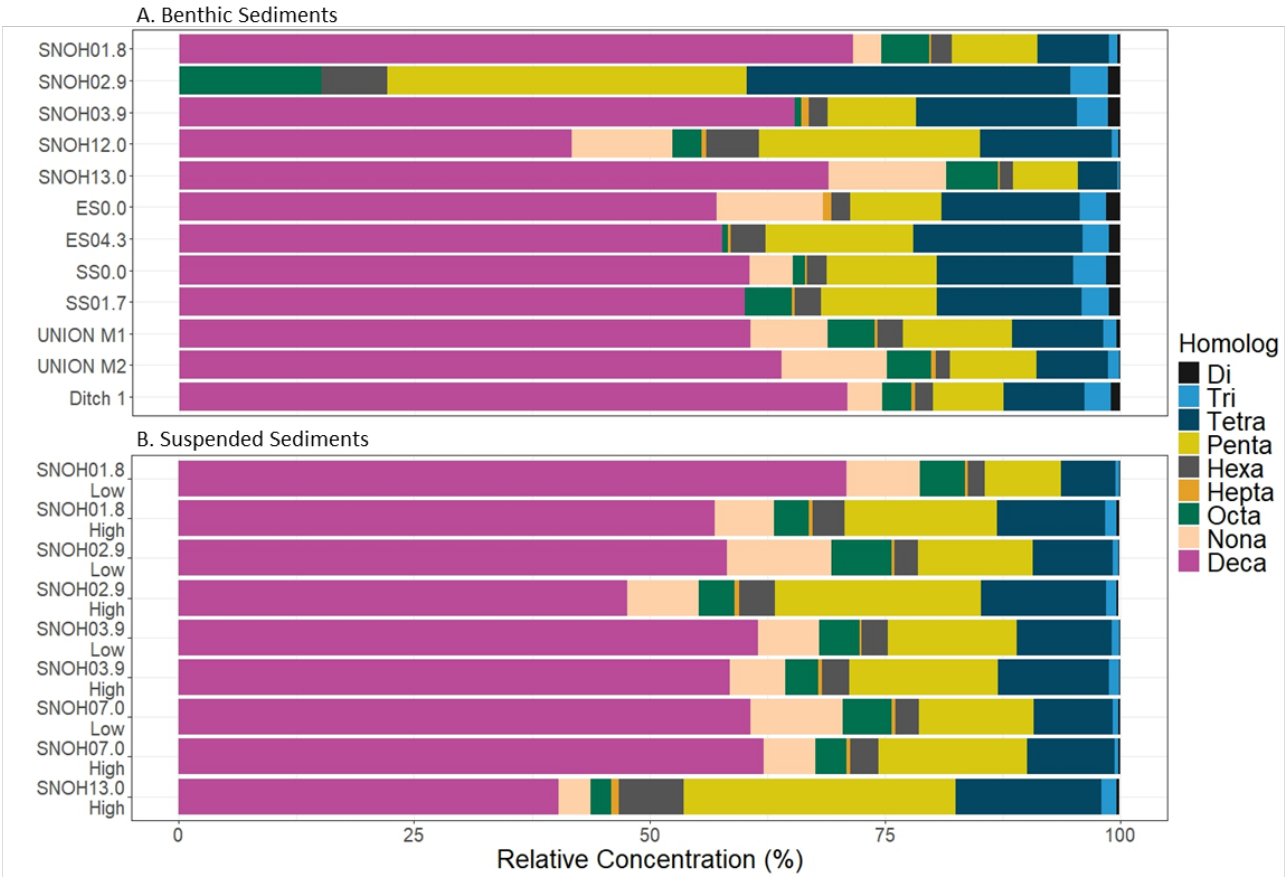

Figure 10. Relative concentrations of PBDE homologs in benthic (A) and suspended (B) sediments.

Relative concentrations were calculated as [PBDE homolog concentration]/ [Sum di through deca-BDE concentration]. Low = Low Flow, High = High Flow.

PBDEs in Biofilms

Biofilms were collected from a subset of sample sites (n=10) during the 2019 low flow sampling event (Table A12). Biofilm total PBDE concentrations were highest in the lower Snohomish main stem, from mile 1.8 to 3.9, where concentrations ranged from 679 – 1,705 pg/g (Figure 11). Skykomish and Snoqualmie river biofilm total PBDE concentrations ranged 119 – 642 pg/g and 11.1 – 338 pg/g, respectively. Elevated concentrations were identified at site SKY34.5 near the town of Sultan on the Skykomish, and at site SNOQ40.7 near the town of Snoqualmie on the Snoqualmie River. Reference site (SNOQ47.8, and SKY40.5) concentrations of total PBDEs were 11.1 pg/g and 125 pg/g demonstrating a considerable difference in background PBDE levels between the Snoqualmie and Skykomish rivers.

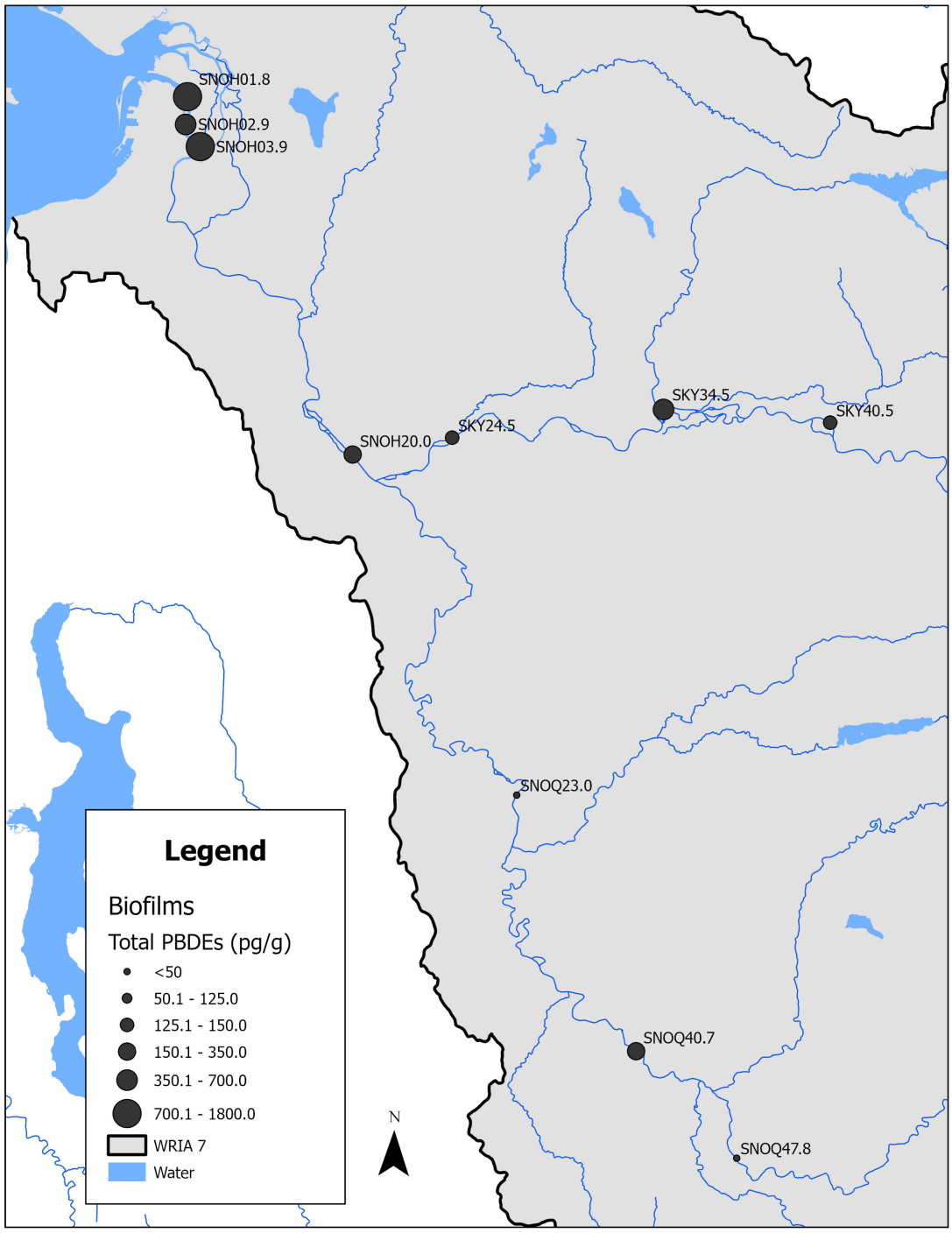

Figure 11. Total PBDE concentrations in biofilms.

The relative concentrations of PBDE homologs in biofilms were dominated by deca-BDE in samples with total PBDE concentration greater than 125 pg/g (Figure 12, indicated by stars). In these samples, deca-BDE comprised 43.6% to 68.8% of the total PBDE concentration and was detected in 100% of the samples. In addition to deca-BDE, tetra and penta-BDEs were also commonly detected. In biofilm samples with less than 125 pg/g of total PBDEs (SNOQ23.0 & SNOQ47.8), deca-BDE was not detected, and penta-BDEs were the dominant congeners measured.

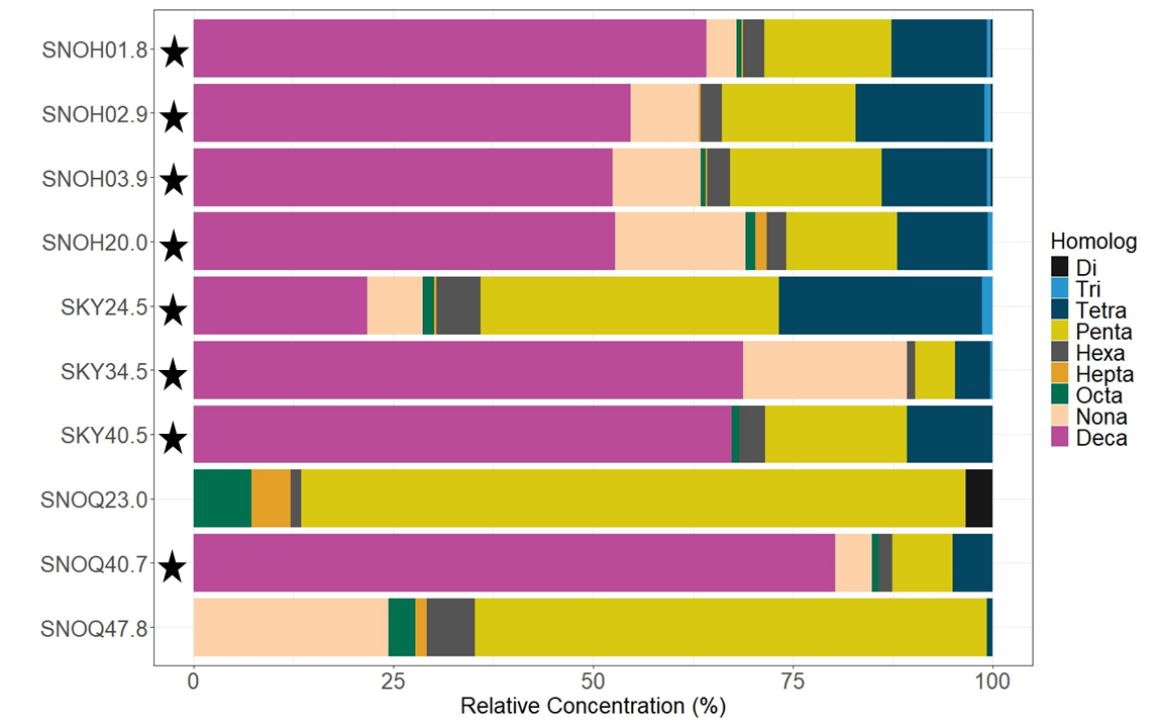

Figure 12. Relative concentrations of PBDE homologs in biofilm samples.

Relative concentrations were calculated as [PBDE homolog concentration]/ [Sum di through deca-BDE concentration]. Stars represent samples with total PBDE concentrations greater than 125 pg/g.

PBDEs in Invertebrate Tissues

Invertebrate collections were composed mainly of amphipods and were generally consistent over time. The main taxon was the benthic or epibenthic species *Americorophium spinicorne*. *A. spinicorne* is a detritivore, that filters out particulate organic matter (Hiebert 2015). Reproduction has been observed in February, March, May, and December; there is no larval stage and individuals inhabit tubes constructed on sediments and hard surfaces. Other consistently present species included *Ampithoe lacertosa, Gnorimosphaeroma insulare, Neomysis mercedis,* and *Eogammarus confervicolus.* All species analyzed for PBDEs are generally in the same feeding guilds (grazers and filterers) and have similar life histories.

Invertebrate tissue samples were collected from two sites (SNOH01.8 & SNOH02.9) in the lower Snohomish mainstem during the 2019 low flow sampling event. These samples indicated that invertebrate tissues contain concentrations of total PBDEs ranging from 1,560 to 1,781 pg/g. Follow up sampling of invertebrate tissues took place monthly from the lower Snohomish mainstem and Ebey Slough in 2021 (April to August) and the mainstem in 2022 (February to September). In both years July samples were not collected due to a lack of biomass in the river. Invertebrate total PBDE results are summarized in Table A13.

Total PBDE concentrations in invertebrate tissues collected from the lower Snohomish mainstem (SNOH01.8 and SNOH02.9) ranged from 1,713 – 9,598 pg/g and 827 – 10,809 pg/g in 2021 and 2022, respectively. Peak concentrations were detected in April of 2021 and March of 2022. A temporal trend in total PBDE concentration was seen in both years with the peak concentrations occurring in spring (March – April) and declining from May through August and September (Figure 13).

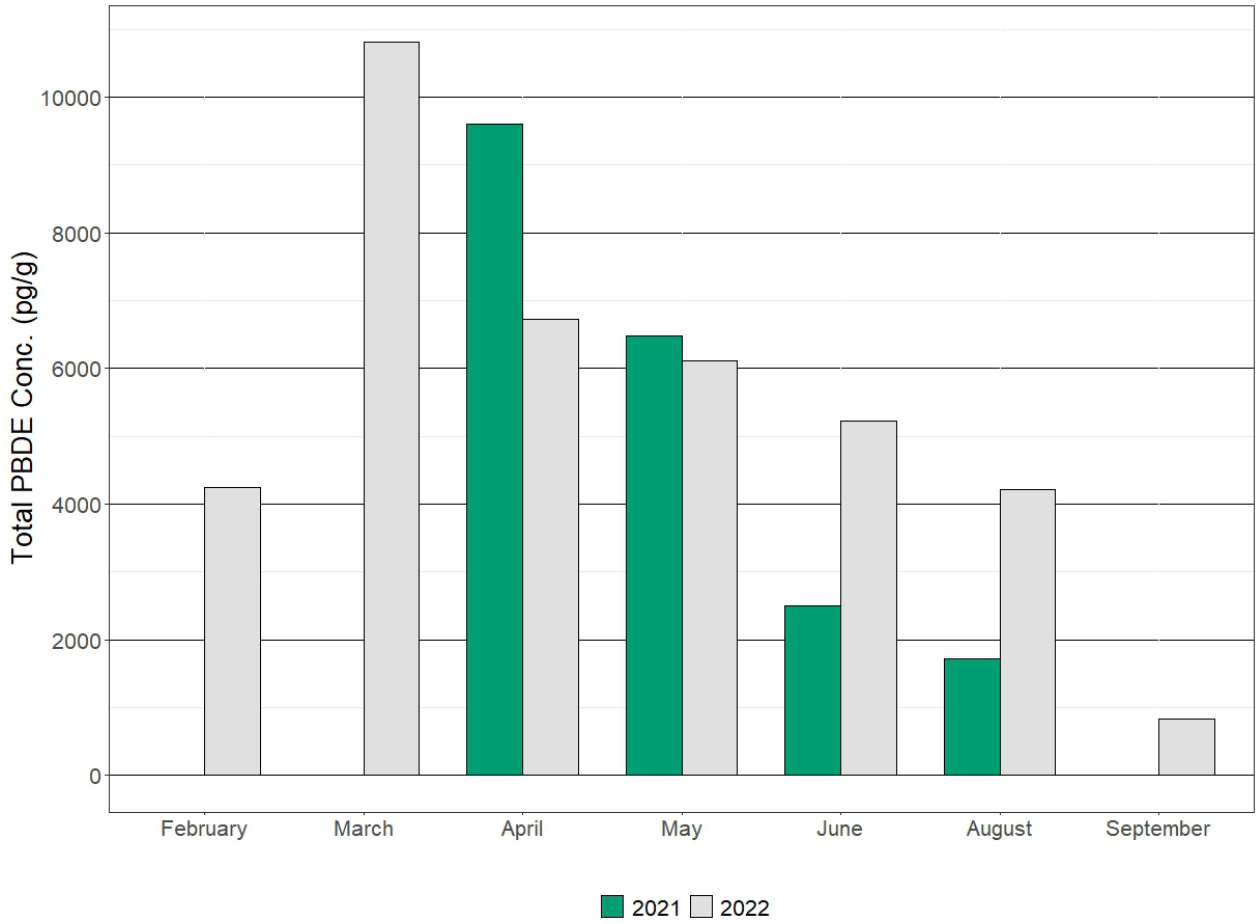

Figure 13. Temporal comparison of Snohomish mainstem invertebrate tissue total PBDE concentrations in 2021 and 2022.

Total PBDE concentrations in invertebrate tissues collected in April, May, and August of 2021 ranged from 1,713 – 9,598 pg/g and 291 – 574 pg/g for the lower Snohomish mainstem (SNOH01.8 and SNOH02.9) and Ebey Slough (ES0.0 and ES1.5), respectively (Figure 14). Total PBDE concentrations were consistently higher in invertebrates collected from the lower Snohomish main stem when compared to Ebey Slough. Lower Snohomish mainstem invertebrate PBDE concentrations were approximately 6 – 18 times greater than Ebey Slough concentrations.

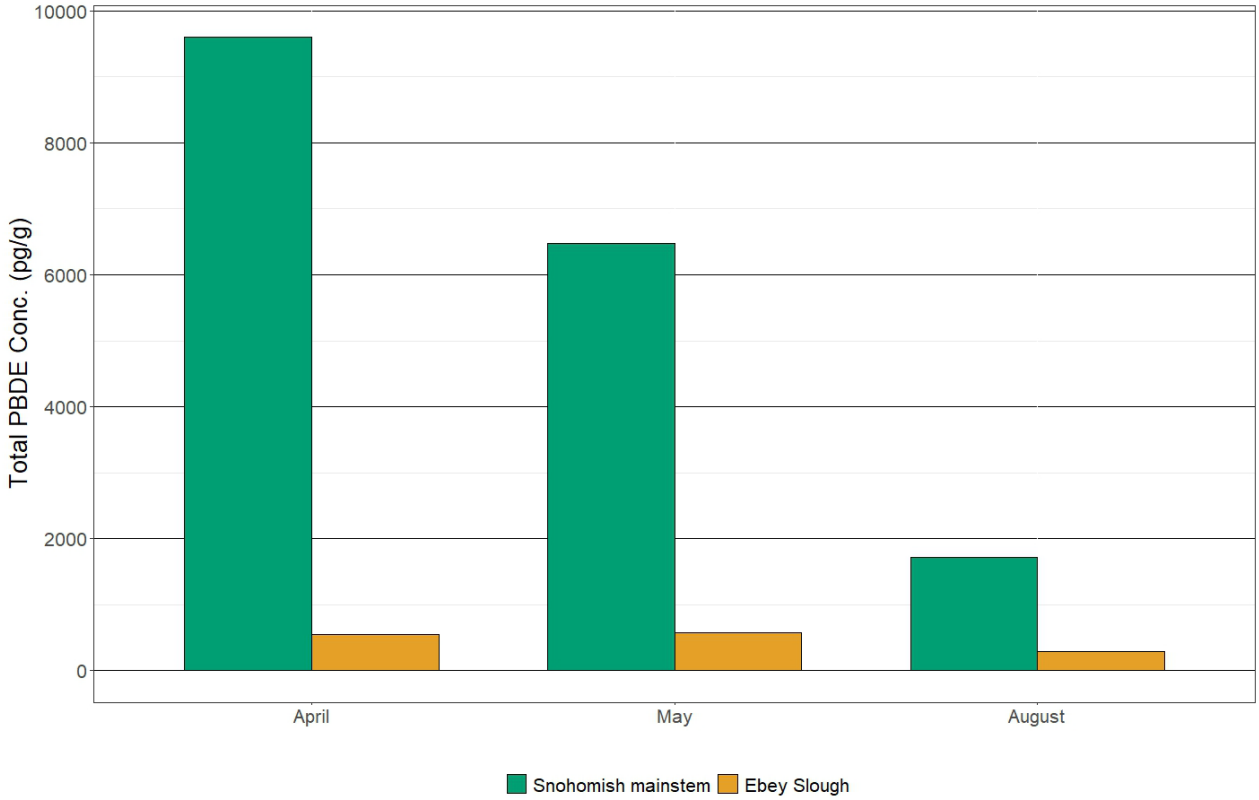

Figure 14. Spatial comparison of total PBDE concentrations in invertebrate tissue collected from the Snohomish mainstem and Ebey Slough.

Invertebrate tissue PBDE concentrations were dominated by tetra- and penta-BDEs accounting for 27.5% – 48.9% and 33.0% –59.9%, respectively, of the total PBDE concentration (Figure 15). The most frequently detected tetra- and penta-BDE congeners were BDE-47 and BDE-99 which were detected in all invertebrate samples. Hexa-BDEs, mainly BDE-153 & 154, were also frequently detected and contributed on average 8% of the total PBDE concentration. Deca-BDE was only detected in 50% of samples and contributed on average 2.9% of the total PBDE concentration.

PBDE homolog relative concentrations were similar between years (2021 & 2022) and sites (Lower Snohomish mainstem & Ebey Slough) with a few exceptions. Deca-BDE was more frequently detected in 2022 (DF = 72%) than 2021 (DF = 29%). In 2021, deca-BDE was only detected in the month of May, where it was measured at both sites, with the highest relative concentration at Ebey Slough (37%). Octa-BDEs were only identified in one sample (Ebey Slough April) but contributed 11% to the total PBDE concentration.

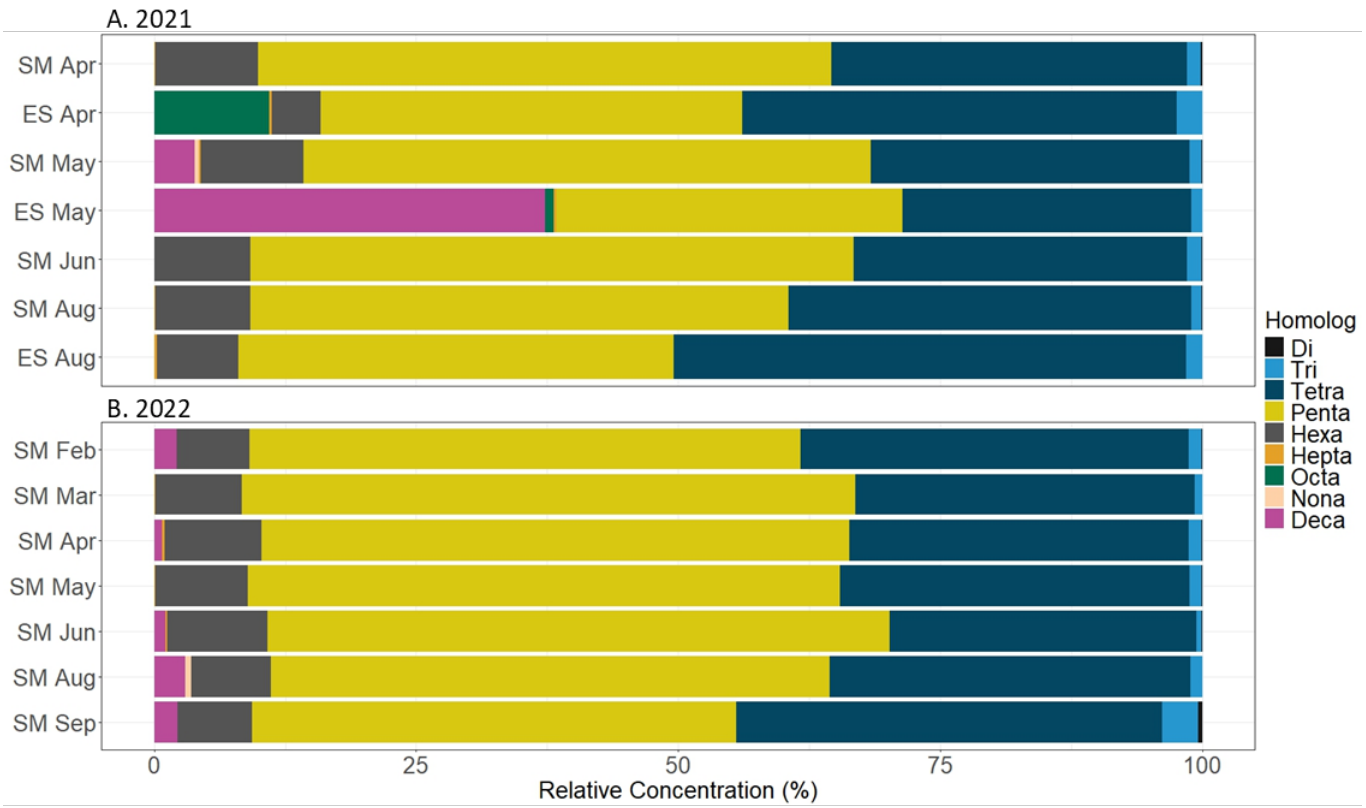

Figure 15. Relative concentration of PBDE homologs in invertebrate samples collected in 2021 (A) and 2022 (B).

SM= Snohomish mainstem, ES = Ebey Slough. Feb = February, Mar = March, Apr = April, Jun = June, Aug = August, Sep = September. Relative concentrations were calculated as [PBDE homolog concentration]/ [Sum di- through deca-BDE concentration].

Stable Isotopes

Biofilm samples collected in 2019 from the Skykomish and Snoqualmie rivers during low flow were analyzed for stable isotopes (Table A12). Results from the Snoqualmie River showed a depleted background ratio (0.3 ‰) and a slight enrichment following the inputs from treatment plants in Snoqualmie and Carnation (Figure 11). Similarly, on the Skykomish River the $\delta^{15}N$ in biofilm from a background sample near Gold Bar was depleted (1.1 ‰) and there was a progressive enrichment in $\delta^{15}N$ downstream following inputs from the Sultan and Monroe treatment plants. The Monroe input was associated with the largest $\delta^{15}N$ enrichment. Effluent from wastewater treatment is often enriched in $\delta^{15}N$, leading to an increase in the isotopic ratio downstream of a discharge location (Hicks et al., 2017).

Three sampling events by WDFW of whole-body juvenile Chinook salmon from the Skykomish River, downstream of the Monroe outfall, has also detailed an enrichment of $\delta^{15}N$ (2016: 14.8‰, 2019: 15.9 ‰, and 2021: 15.2 ‰) in the composite tissue samples. By comparison, two composite whole-body juvenile Chinook salmon samples from the Snoqualmie River in 2021 had an average $\delta^{15}N$ of 7.9 ‰ (A. Carey unpublished data)

The $\delta^{15}N$ of invertebrate tissues from the lower Snohomish River and estuary distributary channels were analyzed from collections in 2019, 2021 and 2022 (Table A13). The 2019 samples were collected from two sites (SNOH01.8 & SNOH02.9) and had a $\delta^{15}N$ of 7.8 ‰ and 8.9 ‰, respectively. In 2021, we collected a more spatially and temporally broad sample from April through August (Figure 16). The samples from Ebey Slough ranged from 6.0 ‰ to 8.1 ‰, while the samples collected from Lower Snohomish mainstem (SNOH01.8 & SNOH02.9) ranged from 4.9 ‰ to 7.7 ‰. Samples from April and May were more depleted. The 2022 samples collected February through August from the Langus Pier (SNOH02.9) documented a relatively consistent $\delta^{15}N$ from February through April, followed by a marked depletion (~ 4 – 5 ‰) in May and June and a subsequent enrichment to winter $\delta^{15}N$ ratios in August and September (Figure 17).

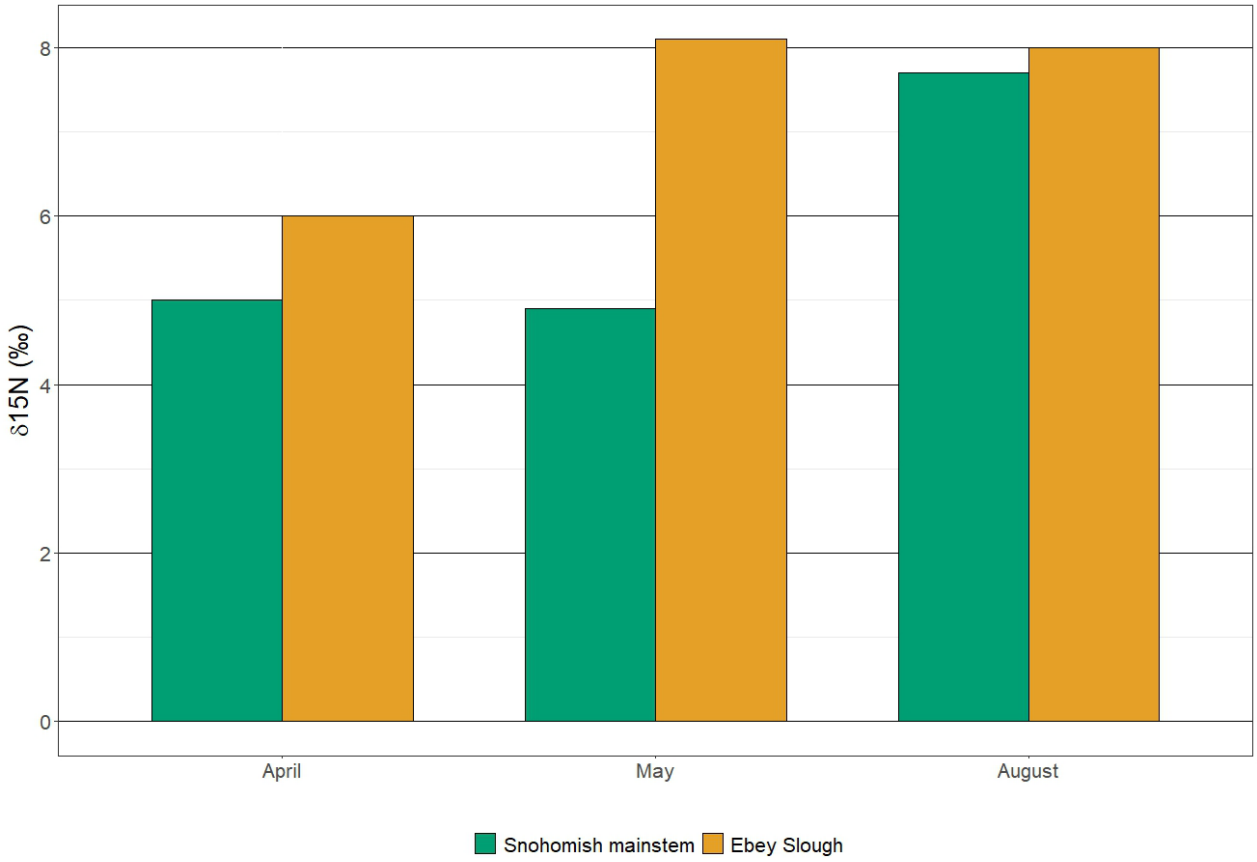

Figure 16 Spatial comparison of $\delta^{15}N$ in invertebrate tissue collected from the Snohomish mainstem and Ebey Slough.

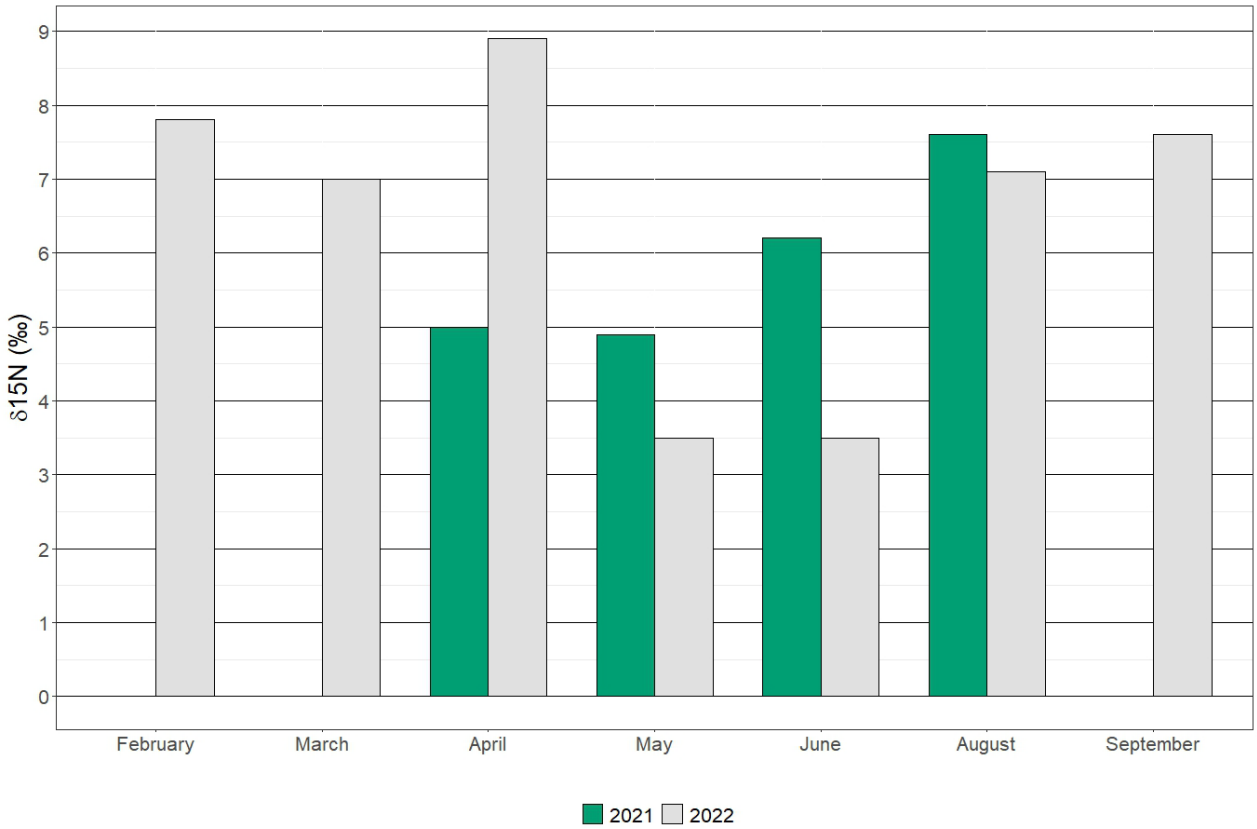

Figure 17. Temporal comparison of Snohomish mainstem invertebrate tissue $\delta^{15}N$ in 2021 and 2022.

Discussion

Areas of PBDE concern

Snohomish River and Estuary

Concentrations of PBDEs in water, sediment, biofilms, and invertebrates varied widely across the Snohomish watershed demonstrating the influence of localized inputs on in-river PBDE concentrations. Two areas within the river system had elevated concentrations of PBDEs in multiple environmental matrices, providing evidence of a localized input. Within the Snohomish River, the main area of notably higher PBDEs is the lower mainstem. This area encompasses a five-mile stretch of the river's estuary from river mile five (SNOH05.0) to the mouth of the Snohomish (SNOH0.0) where the river enters Port Gardner Bay.

Samples of water, biofilms, sediments, and invertebrates collected from the lower mainstem had elevated levels of PBDEs in comparison to other areas of the estuary (i.e., sloughs) and upstream in the upper mainstem. Mean PBDE water concentrations during high flow conditions were 2.5 times higher in the lower mainstem than the surrounding sloughs, and 3.3 times higher than the upper Snohomish mainstem. Biofilm PBDE concentrations were 3 times higher in the lower Snohomish (sites SNOH01.8 & SNOH02.9) than in the upper Snohomish (SNOH20.0). Additionally, invertebrate PBDE tissue concentrations were ~6 – 17 times higher in the lower Snohomish in comparison to Ebey Slough. These elevated PBDE levels indicate an active PBDE input within the lower Snohomish mainstem that is not present in other areas of the estuary or upper Snohomish River.

During the study period from 2019 to 2022, the Everett Water Pollution Control Facility (WPCF) actively discharged treated wastewater into the lower Snohomish mainstem from outfall 015, which is located near site SNOH02.5. Discharge volumes varied throughout the study period, and not all sampling events were conducted during active discharge periods (Table 3).

In 2022 high and low flow sampling events captured water concentrations of PBDEs during periods of active wastewater discharge as well as when no discharge occurred in the lower Snohomish mainstem (Table 3). Active discharge from outfall 015 occurred during the high flow event, while no active discharge occurred during the low flow event. Mean concentrations of PBDEs in the lower mainstem during active discharge were 2.8 times higher than when no discharge occurred. Concentrations of PBDEs were the highest at site SNOH02.5, just downstream of outfall 015 and then decreased moving away (both downstream and upstream) from this site (Figure 18). Due to tidal influx within the lower Snohomish, movement of PBDEs from the outfall is bidirectional and can impact sites upstream of the outfall. These data demonstrate the active source of PBDEs impacting the lower Snohomish mainstem is treated wastewater discharged from outfall 015.

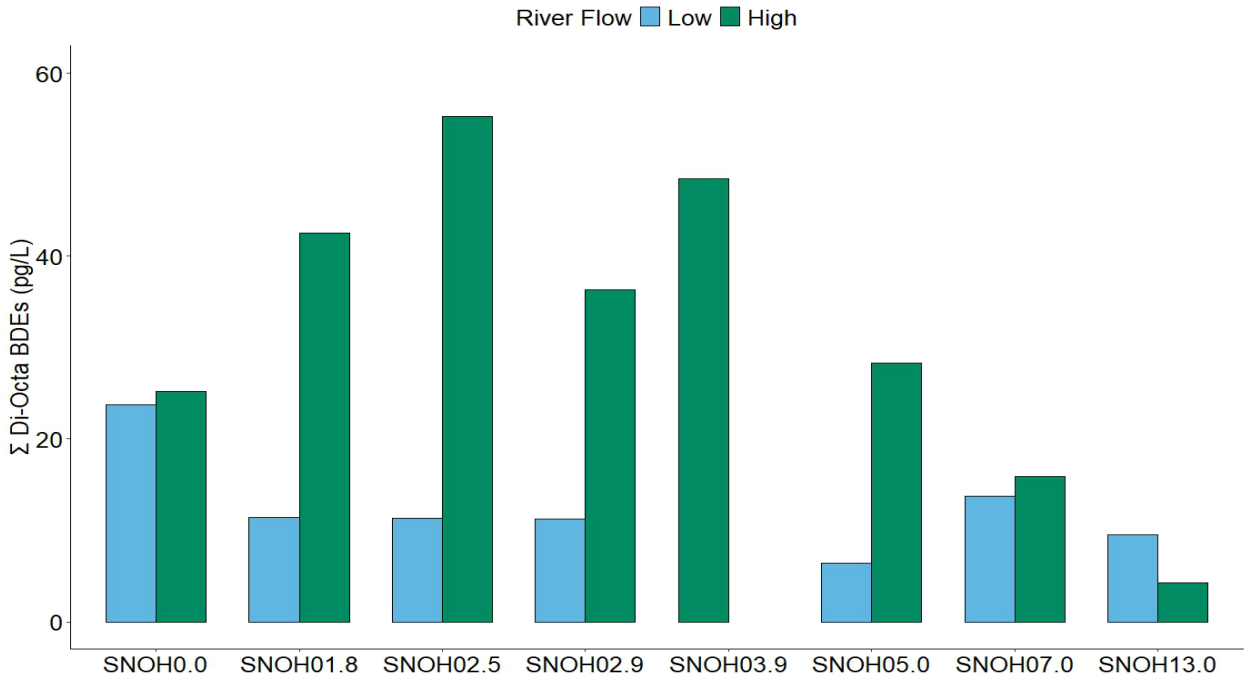

Figure 18. PBDE water concentrations in the Snohomish Mainstem during low and high flow river conditions in 2022.

PBDE water concentrations in the Snohomish mainstem are shown during active treated wastewater discharges from outfall 015 during high flow conditions in the spring of 2022. Water concentrations of PBDE were again measured at each site when no discharge took place during low flow conditions in the late summer of 2022. Where a blank space is present no sample was collected for the flow condition.

Table 3. Discharge records for outfall 015 and Snohomish River CSOs during SPMD sample events and corresponding PBDE water concentrations.

Records of discharge accessed through Ecology's PARIS database[4] and represent an average of daily flows recorded during the specified sampling period. Records of CSO discharge were collected from CSO annual reports for 2019 to 2022 provided by Everett Public Works.

Sampling Dates	River Conditions	Outfall 015 Active	Mean Outfall Discharge (MGD)	CSO Event	CSO Discharge (MG)	t-PBDE in river measured by SPMD (pg/L)$_A$
8/19 – 9/18/19	Low Flow	No		Yes	1.54	26.3
9/8 – 10/5/20	Low Flow	Yes $_B$	4.0	No		13.78
9/13 – 10/14/21	Low Flow	Yes $_C$	5.1	Yes	1.41	22.35
8/8 – 9/22/22	Low Flow	No		No		11.35
3/29 – 4/28/21	High Flow	Yes	11.4	No		51.34
4/13 – 5/12/22	High Flow	Yes $_D$	15.81	Yes	0.24	43.95

A. t-PBDE concentration are the average concentration measured at SNOH01.8, SNOH02.9, and SNOH03.9 during each sampling period. Samples were not collected from all sites during each sampling period.

B. Active discharge from 9/9/20 to 9/25/20.

C. Active discharge from 9/14/21 to 10/14/21.

D. Active discharge except on 5/1/2022

[4] https://apps.ecology.wa.gov/paris/PermitLookup.aspx

Invertebrate tissues sampled in the vicinity of outfall 015 in 2022 had a depleted $\delta^{15}N$ during the months of May and June, which is the end of the juvenile out-migrating residence. The PBDE concentration in the invertebrate tissues was greatest in March and April when the juvenile Chinook are beginning their outmigration residence in the estuary. The results found in this study support those of the 2016 WDFW survey which identified outfall 015 as a probable source of the PBDEs detected in wild juvenile Chinook captured in the lower mainstem due to their distinctive chemical signature, and depleted nitrogen stable isotope ratio (O'Neill et al. 2020). The depleted $\delta^{15}N$ in invertebrate tissues is also consistent with the WDFW observed depleted $\delta^{15}N$ in tissues of wild juvenile Chinook in the lower Snohomish River (O'Neill et al. 2020).

The $\delta^{15}N$ of biota tissues in the receiving waters of WWTP effluent can reflect the N source and processing of N in the plant. Effluent that has not undergone sufficient nitrification or treatment can exhibit a depleted δ15N signal which is taken up by biota in the receiving environment (deBruyn and Rasmussen 2002; Hicks et al. 2017). Conversely, effluent that undergoes treatment to remove excess nitrogen with nitrifying and denitrifying bacteria typically have an enriched δ15N signal compared to background values (Heaton 1986; Savage 2005).

We speculate that the May – June depletion is due to either, (1) a greater input of $\delta^{15}N$ depleted freshwater from the upper Snohomish basin, or (2) a change in the $\delta^{15}N$ of the organic N in the effluent from the Everett treatment plant. Because the depletion of $\delta^{15}N$ in juvenile chinook tissues was not measured in samples throughout the estuary (O'Neill et al. 2020) and appears localized to the lower Snohomish River, the explanation of changes in $\delta^{15}N$ of the organic N in effluent seems more plausible. Over the period of sampling in 2022, the treatment plant was in compliance for discharge rates and concentrations of nitrogen in the effluent (Table 4). Concentrations of N discharged from the plant were fairly consistent; ammonia was the dominant inorganic form being discharged. A shift in $\delta^{15}N$ of the effluent could be a result of a change in processing time and the source of water being treated and flowing through the retention ponds of the treatment plant, which receives residential, commercial, and industrial wastewater and municipal stormwater. A similar progressive depletion of $\delta^{15}N$ in particulate organic matter in treatment plant effluent was noted by deBruyn and Rasmussen (2002) over a summer of sampling in Montreal, Canada.

Table 4. Summary of invertebrate tissue stable isotopes, PBDE concentrations, and discharge rates and nitrogen concentrations from the Everett WPCF (outfall 015).

Records of discharge and nitrogen concentrations were accessed through Ecology's PARIS database[5] and represent monthly averages of weekly composite samples.

Collection Date	Invertebrate d13C (‰)	Invertebrate d15N (‰)	Invertebrate T-PBDE (pg/g)	Average Outfall Discharge (MGD)	Outfall Discharge Ammonia (mg/L)	Outfall Discharge TKN (mg/L)	Outfall Discharge Nitrate + Nitrite (mg/L)
4/27/2021	-26.1	5	9599.0	11.5	25.4	29.1	0.12
5/12/2021	-24.6	4.9	6480.0	11.4	22.7	29.4	0.08
6/25/2021	-22.7	6.2	2496.6	11.9	17.5	25.9	0.34
2/3/2022	-23.7	7.8	4247.9	9.9	21.9	26.5	0.02
3/9/2022	-25.2	7	10809.9	14.6	22.1	26.8	0.13
4/14/2022	-22.4	8.9	6722.7	14.4	21.3	25.1	0.15
5/12/2022	-26.3	3.5	6116.5	12.2	21.0	27.6	0.20
6/16/2022	-26.3 [a]	3.5 [a]	5209.0 [a]	13.9	23.6	31.0	0.25
8/16/2022	-20.2	7.1	4215.25	0	—	—	—
9/13/2022	-24.8	7.6	827.28	0	—	—	—

[a] average of duplicate samples; MGD = millions of gallons per day; TKN = Total Kjeldahl Nitrogen (organic N + ammonia-N)

There are several other WWTPs which discharge to the Snohomish and its estuary (see Table 1); however, the only other discernable increase in PBDE concentrations was measured downstream of the Marysville WWTP 001 outfall at the mouth of Steamboat Slough during high flow sampling in the spring of 2021. Discharge monitoring reports from the Marysville WWTP show active discharge during this sampling period, demonstrating the plant as a potential input of PBDEs into the estuary. No other SPMD deployments coincided with active discharges from this outfall due to the city of Marysville's WWTP pumping and discharging treated wastewater through the city of Everett's deepwater outfall (100) in Port Gardner Bay. Further investigation is needed to determine if outfall 001 is an input of PBDEs to the estuary during active discharge.

Additional potential sources of PBDEs to the Snohomish mainstem are combined sewer overflows (CSOs) located between river miles 3 and 6. These CSOs discharge untreated wastewater during storm events which exceed the capacity of the City of Everett's wastewater infrastructure. These events can occur several times per year and have the potential to impact PBDE concentrations within the lower Snohomish mainstem. While their discharge volume is small in comparison to daily discharges from outfall 015, PBDE concentrations in untreated wastewater are 3 – 4 times higher than in treated wastewater (see section *PBDEs in Wastewater*). Of most concern is CSO outfall SR07 and SR08, located near site SNOH05.0, which over the study period from 2019 to 2022 discharged a combined volume of 15.5 million gallons (Table A14). Due to the infrequent nature of these CSO discharges it is difficult to discern the impact to in-river PBDE concentrations, though it should be noted overlapping sampling and discharge events in 2019 (Table 3) indicated the presence of elevated PBDE concentrations at SNOH05.0. CSO PBDE loading needs to be further investigated to determine what impact these events have on lower Snohomish mainstem PBDE concentrations.

[5] https://apps.ecology.wa.gov/paris/PermitLookup.aspx

Skykomish and Snoqualmie Rivers

Outside of the lower Snohomish River, elevated PBDE concentrations were measured downstream of the city of Monroe's STP outfall in the Skykomish River. Concentrations of PBDEs in water downstream of the outfall (SKY24.5) were 3 – 8 times higher than concentrations upstream (SKY25.0) during low flow conditions. These elevated concentrations were only measured during low flow conditions, while high flow conditions had similar concentrations between sites. This is due to the difference in river flow during these sampling events and the effect of dilution. Wastewater discharges are similar during the high and low flow conditions (Figure 19 B), however less flow in the river (Figure 19 A) results in less dilution of PBDEs entering the river leading to higher concentrations of PBDEs in water.

The enrichment of N stable isotopes in biofilms downstream of the Monroe outfall provides a line of evidence that effluent-derived nutrients are being incorporated into biota in this area. This includes juvenile Chinook from the Skykomish, which have measured δ^{15}N in tissues that are much more enriched than individuals from the Snoqualmie and lower Snohomish rivers (A. Carey unpublished data). However, Skykomish River juvenile Chinook surveyed in 2016 and 2021 did not have elevated concentrations of PBDEs suggesting that the Monroe STP's discharge of PBDEs is not impacting juveniles (O'Neill et al. 2020, A. Carey personal communications, 2024). This may be in part due to the smaller size and limited residence/feeding time of juvenile Chinook in this portion of the river system.

It is noteworthy that samples of resident fish species throughout the Snohomish and Skykomish rivers were found to contain measurable levels of PBDEs (Johnson et al. 2006; Seiders et al. 2007; Mathieu and Wong 2016), suggesting sources of PBDEs within the Skykomish River may be impacting resident fish.

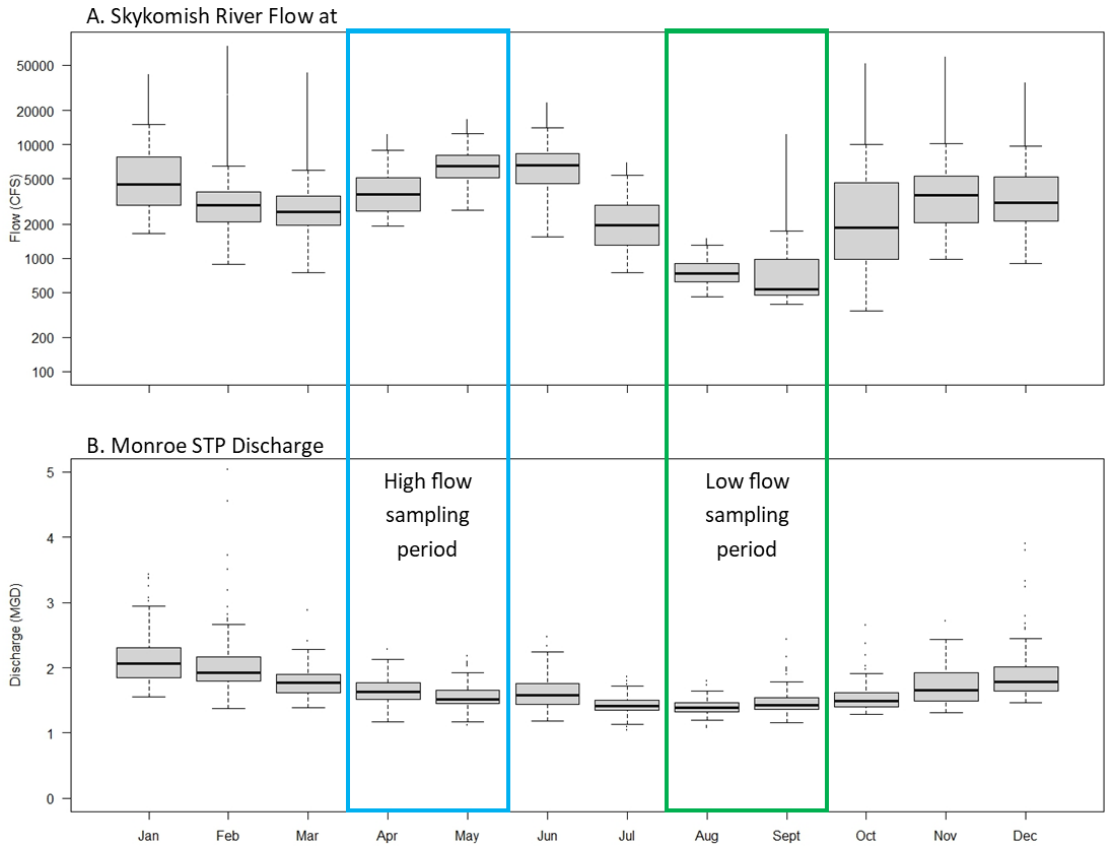

Figure 19. Skykomish River flow and Monroe Sewage Treatment Plant (STP) discharge from 2019 to 2022

Summary of monthly Skykomish River flows and discharges from Monroe STP during the study period from 2019 to 2022. River flows are from USGS station 12134500 near Gold Bar, WA. Records of discharge for Monroe STP were accessed through Ecology's PARIS database[6].

Additional locations in the Skykomish River showed limited influence by potential PBDE inputs. A slightly elevated concentration of PBDEs in water downstream of the Sultan WWTP outfall (site SKY34.5) indicates a potential input of PBDEs from this source. Biofilm PBDE concentrations measured at the site were the highest in the Skykomish River and provide further evidence of a nearby input. Further source confirmation efforts were hindered by lost equipment and additional investigation is needed to confirm the impacts of PBDEs on this section of the river. The PBDE concentrations of the Skykomish River reference site (SKY40.5) were higher and more variable than those measured at the Snoqualmie River reference site (SNOQ47.8), demonstrating the potential for more upstream sources of PBDEs.

The Snoqualmie River had the lowest average concentration of PBDEs of the three rivers and did not show indications of point source PBDE inputs. Water concentrations of PBDEs were similar throughout the river system and river conditions. Similar concentrations between the Snoqualmie reference site and downstream sites indicate river concentrations are near background levels. A slight elevation in biofilm PBDE concentrations at site SNOQ40.7 was not confirmed by water concentrations. The data from the study suggests there are limited sources of PBDE entering the Snoqualmie River.

[6] https://apps.ecology.wa.gov/paris/PermitLookup.aspx

PBDEs in wastewater

This study found that elevated levels of PBDEs within the river basin were associated with the discharge of wastewater from in-river outfalls. While WWTP are not a direct source of PBDEs, they act to concentrate upstream sources of PBDEs through the collection, treatment, and discharge of gray water, sewage, and industrial wastewater. Several studies have characterized the concentration of PBDEs in WWTP influent and effluent within the Snohomish watershed and the greater Western Washington region (Carey et al. 2023; Ecology and Herrera 2010; Meador et al. 2022).

From 2020 – 2022 the Everett WPCF measured influent and effluent concentrations of PBDEs within their facilities during four quarterly sampling events. Total PBDE concentrations ranged from 70,345 to 169,623pg/L in influent samples and from 9,128 to 39,811 pg/L in effluent samples (Table 5). The facility utilizes two outfalls, one located in Port Gardner Bay (outfall 100) and the other in the lower Snohomish mainstem (outfall 015). The highest effluent PBDE concentrations were measured in samples collected from Outfall 015.

Table 5. Everett WPCF influent and effluent PBDE concentrations (pg/L) from 2020 – 2022

Source	n	Min.	Max	Mean	Median
Influent	4	70,345.4	169,623.9	110,220.9	100,457.1
Outfall 100 Effluent	5	9,128.6	20,531.8	16,111.2	16,102.7
Outfall 015 Effluent	7	22,749.7	39,811.0	29,303.9	26,853.0

Total PBDE concentrations are the sum of di- through deca-BDEs. n = number of samples Data provided courtesy Everett Public Works.

Additional surveys conducted in Western Washington WWTPs have found PBDEs in influent and effluent samples at similar concentration ranges as Everett's WPCF. A 2009 survey of 10 Puget Sound POTWs observed total PBDE concentrations in treated wastewater discharges ranging from 8,600 to 135,000 pg/L (Ecology and Herrera 2010). In 2021, King County measured PBDE concentrations in effluent samples from two WWTP facilities. Total PBDE concentrations ranged from 5,039 to 10,743 pg/L (Meador et al. 2022). Ecology measured PBDEs in influent and effluent samples from the Eatonville WWTP in the Nisqually River basin in 2021. Mean influent concentrations were 93,594 pg/L while mean effluent concentrations were 1,855 pg/L (Carey et al. 2023).

In addition to WWTPs, a recent study by Ecology found PBDE concentrations varied widely in discharges of wastewater from a variety of industrial sources; the discharges from these sources were all destined for WWTPs (Wong 2022). Wong (2022) showed that wastewater from an industrial laundry facility had by far the highest PBDE concentration (3,490,000 pg/L). These studies show that wastewater and associated WWTPs are a common pathway of PBDEs throughout Western Washington including the Snohomish watershed.

PBDE Accumulation, Partitioning, and Transport

Accumulation

PBDEs were found throughout the riverine and estuarine environments in water, sediments, biofilms, and invertebrate tissues. Due to their persistence and tendency to accumulate, PBDEs pose a long-term threat to the environment and susceptible species such as juvenile Chinook. While SPMD data shows that dissolved phase concentrations of PBDEs are highest near active sources, we know that PBDEs are more likely to accumulate in organic carbon and lipid rich substrates such as sediments, biofilms, and invertebrate tissues. The accumulation of PBDEs increases their concentration and creates a pathway from source to juvenile Chinook in the Snohomish watershed. Bioconcentration and bioaccumulation of toxics among different environmental media are often described from field measurements and are a useful way to assess the uptake and potential risk of contaminants (Arnot and Gobas 2006).

The accumulation of PBDEs in sediments is well-documented and is due to the chemicals' affinity to adsorb to organic carbon rich substrates (Dinn et al. 2012). Though benthic sediments collected in this study consisted mostly of fine sands and silts with low TOC content, accumulation of PBDEs still occurred at measurable levels. Total PBDE concentrations in bottom sediments collected from the Snohomish estuary were on average ~45,400 times higher than corresponding water concentrations. This accumulation of PBDEs also occurred in suspended sediments where average concentrations were ~58,200 times higher than water concentrations.

In comparison to sediments collected in Port Gardner Bay and Everett Harbor, benthic and suspended sediments from the estuary had lower concentrations of BDE-47, similar concentrations of BDE-99, and higher concentrations of BDE-209 (Figure 20). A potential contributing source of PBDEs to Port Gardner Bay is Everett's wastewater outfall 100. BDE-209 concentrations in treated wastewater discharged from outfall 015 is on average 3.4 times higher than levels discharged to the Port Gardner Bay (Everett Public Works unpublished data), which is a potential cause of higher BDE-209 concentrations within estuary sediments. Estuary sediment BDE-47 and -99 concentrations from this study were similar to samples collected from 2007, 2012, and 2019 in Ebey Slough and the Snohomish River delta (SAIC 2009; Partridge et al., 2014; Marine Sediment Monitoring Team 2022a, b). These sediment results demonstrate there is considerable historical accumulation of PBDEs not only in the Snohomish estuary but in the surrounding marine environments (i.e., Port Gardner Bay, Everett Harbor). Further investigation is needed to understand if there are impacts on biota and uptake into benthic/epibenthic prey organisms from this PBDE accumulation in sediments.

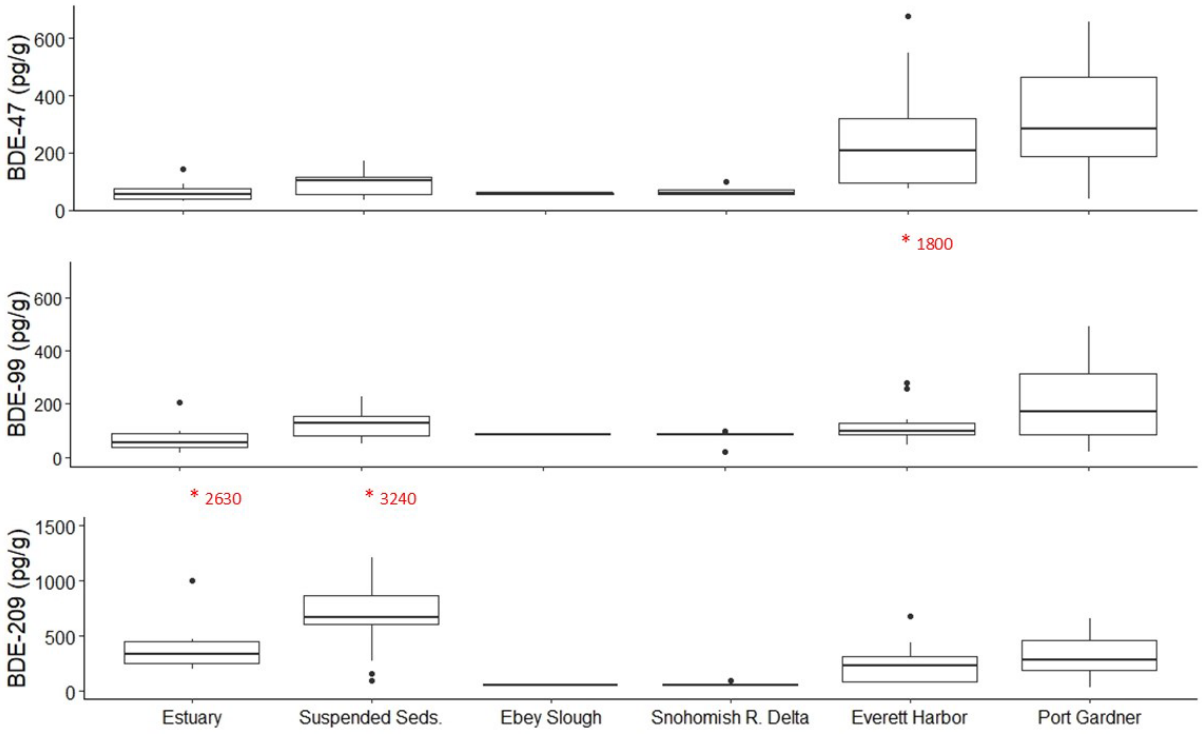

Figure 20. Concentration of BDE-47, -99, and -209 in sediments from the Snohomish estuary and surrounding marine environments

Estuary and suspended sediment (suspended seds.) result are from this study; Ebey Slough, Snohomish River delta (Snohomish R. Delta), Everett Harbor, and Port Gardner results are from 2007, 2012, and 2019 (SAIC 2009, Partridge et al., 2014, Marine Sediment Monitoring Team 2022a, b). Outlier sample concentrations greater than the scale range are shown as (*) with their corresponding concentration. pg/g = picograms/gram.

In addition to accumulating in sediments, biofilms throughout the Snohomish watershed accumulated PBDEs at levels greater than ambient water concentrations. The average total PBDE concentration in biofilms was ~27,800 times higher than ambient water concentrations. The accumulation of PBDEs in biofilms and sediments demonstrates the effects of discharging low levels of PBDEs in high volumes into the watershed.

Due to their persistence, PBDEs accumulate at levels which can begin to impact the food web (e.g., sublethal impacts on juvenile Chinook). Invertebrate tissue samples collected throughout the Snohomish estuary represent prey items for juvenile Chinook residing in the estuary. Average concentrations of PBDEs in invertebrate tissues were 3.5 times higher than biofilms, 9.2 times higher than sediments, and ~275,000 times higher than ambient water concentrations, demonstrating the accumulative effect of PBDEs within the environment and food web. PBDE accumulation in invertebrates provides a pathway from source to juvenile Chinook through the consumption of prey.

Amphipods dominated samples collected from the estuary, of which the predominant species was *Americorophium spinicorne* (Figure 21). Semi-quantitative assessments of representative subsamples of invertebrates showed a few other filter feeders or grazers were present in the samples. *A. spinicorne* is an epibenthic filter feeder that consumes particulate organic matter and has been documented as a prey item for juvenile Chinook residing in the Snohomish estuary during spring months (Cordell et al. 1999).

Figure 21 *A. spinicorne* collected from lower Snohomish mainstem
(photo credit Dany Burgess)

Juvenile Chinook exposure to PBDEs through their diet has been documented in the lower Columbia River and Puget Sound, including the Snohomish estuary (Sloan et al. 2010). Additionally, juvenile Chinook collected by WDFW in 2021 from the lower Snohomish mainstem showed a strong correlation between whole body tissue and gut content concentrations of PBDEs (A. Carey personal communications 2024). PBDE concentrations in juvenile Chinook gut contents were higher than those detected in invertebrates but within an order of magnitude (Table 6). Juvenile Chinook dietary assimilation rates of BDE-47 and BDE-99, the most abundant congeners detected in invertebrate samples, are approximately 50% (Dietrich et al. 2015), demonstrating a significant portion of the PBDEs consumed will accumulate in Chinook tissues.

In addition to availability of prey for juvenile Chinook, timing could also play a role in the transfer of PBDEs through the food web. Results from the 2021 and 2022 temporal sampling of invertebrates found peak tissue concentrations of PBDEs occurred during the spring (March – May) which coincides with the peak juvenile Chinook seaward migration into the Snohomish River as is typical for Puget Sound Chinook (O'Neill et al. 2020; Quinn and Losee 2022). This overlapping occurrence of peak prey PBDE concentrations and juvenile Chinook seaward migration seems to provide the right timing for juvenile Chinook PBDE accumulation in the Snohomish estuary.

Table 6. PBDE concentrations in juvenile Chinook whole body tissues, stomach contents, and invertebrates collected from the Snohomish estuary.

PBDE concentration are the sum of 11 BDEs (BDE-28, 47, 49, 66, 85, 99, 100, 153, 154, 155, 183) reported as ng/g wet weight. Juvenile Chinook whole body tissue and stomach contents sampled in 2021 and unpublished data provided courtesy of WDFW TBiOS. Invertebrates collected from 2019 to 2022. Std Dev = Standard Deviation.

Tissue	Stats	Lower Snohomish Mainstem	Distributary Channels
Juvenile Chinook Whole Body Tissue	n	10	16
	Mean	26	3.8
	Median	22	2.6
	Min	8	0.53
	Max	56	13
	Std Dev	13	3.3
Stomach Contents	n	3	3
	Mean	37	2.9
	Median	38	3
	Min	27	0.71
	Max	46	5
	Std Dev	9.5	2.1
Invertebrates	n	16	3
	Mean	4.4	0.37
	Median	4.3	0.35
	Min	0.8	0.29
	Max	10.4	0.48
	Std Dev	2.8	0.10

The accumulation of PBDEs in juvenile Chinook at levels potentially causing sublethal impacts is well documented in the Snohomish estuary. O'Neill et al. (2020) found that wild origin juvenile Chinook collected in 2016 from the lower mainstem had a mean whole body tissue concentration of 24 ng/g wet weight and had significantly higher levels of PBDEs than fish collected from the upper mainstem and distributary channels. In 2021, a follow up study, which coincided with this study, found similar concentrations of PBDEs in juvenile Chinook collected from the lower Snohomish mainstem (Table 6), which again were elevated in comparison to fish from the distributary channels and the upper watershed. These juvenile Chinook whole body tissue concentrations represent an approximately five-fold increase in PBDE levels over the mean invertebrate concentrations measured during this study. These results, along with the stomach content data, indicate the exposure pathway of PBDEs to juvenile Chinook is through their diet.

Partitioning

Results from this study found that the composition of PBDE homologs were different between environmental media and reflected partitioning of homologs from water to sediments and biofilms. The homolog compositions of treated wastewater discharged to the lower Snohomish mainstem from outfall 015 were composed of similar relative concentrations of tetra-, penta-, and deca-BDE homologs, approximately 30% each (Figure 22). Environmental media collected from the surrounding area had differing proportions of these three homologs. Ambient water homolog compositions were dominated by tetra- and penta-, but also included other lower molecular weight homologs, di and tri-BDEs. As discussed in the *Methods* section, this may be due in part to our use of SPMDs to sample ambient river water PBDE concentrations and the inability of this device to take up the heavier deca-BDE, but it also represents the behavior and possible uptake of deca-BDE in dissolved phase which is limited by its organic carbon-water partition coefficient (K_{OC}).

Sediment (bottom and suspended) and biofilms were dominated by higher molecular weight homologs, including a large proportion of deca-BDE and lower proportions of octa- and nona-BDEs. The differences in homolog make up of ambient water, sediments, and biofilms reflects the differences in partitioning rates between the PBDE homologs and the organic carbon composition of the environmental media.

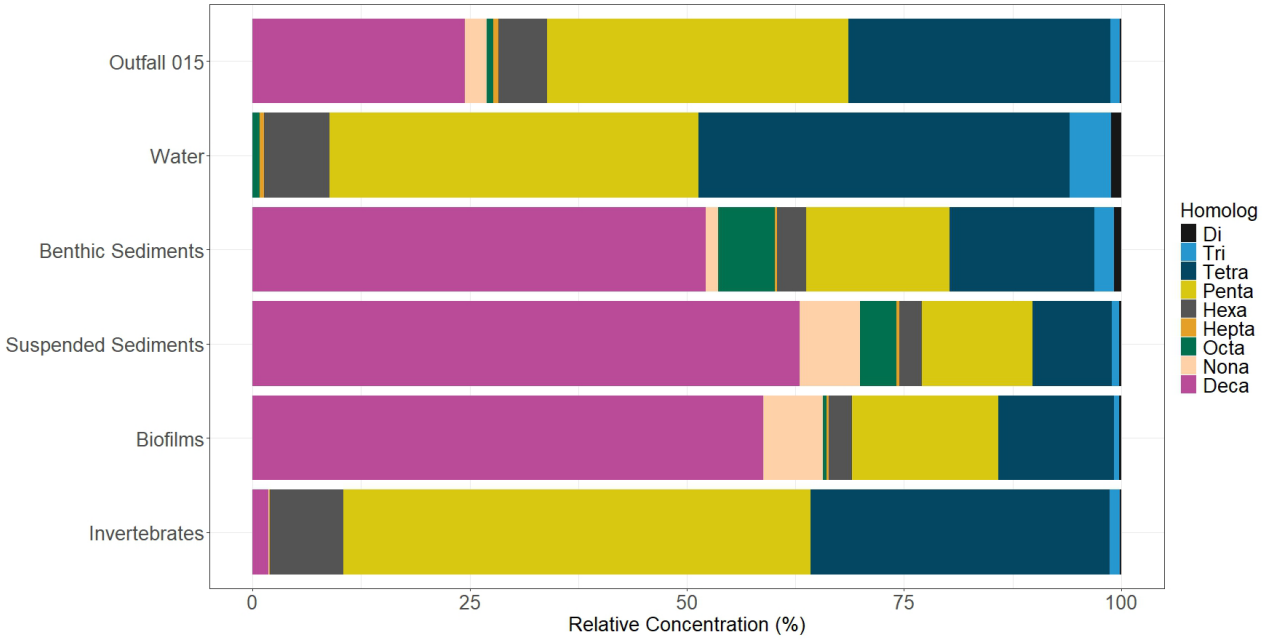

Figure 22. Mean relative concentrations of PBDE homologs in treated wastewater and environmental media collected from the lower Snohomish mainstem

PBDE congeners partition into different environmental media based on their water-particle partitioning coefficient (K_{OC}) and water-octanol partitioning coefficient (K_{OW}). PBDEs in general are hydrophobic and have a strong affinity for partitioning to organic carbon and lipid rich media, but higher molecular weight congeners, such as octa-, nona-, and deca-BDEs have higher K_{OW}s and K_{OC}s limiting their dissolved phase presence in water and causing them to accumulate in sediments and biofilms.

The presence of deca-BDE in treated wastewater is likely due to these whole water samples containing particulate and colloidal matter. As the treated wastewater is discharged to the river system particle bound deca-BDE is incorporated into bottom and suspended sediments as well as biofilm (algae, microbial biomass, and detrital organics). The results of this study are similar to findings of Dinn et al. (2012), who found deca-BDE in significantly higher proportions in sediments near outfalls than in wastewater due to its strong partitioning to particulate matter.

Invertebrates collected from the lower Snohomish mainstem were shown to have higher relative concentrations of tetra- and penta- homologs than their potential food sources: sediments and biofilms (Figure 22). This is potentially due to deca-BDE's strong affinity for particulate matter causing it to pass through the invertebrates' digestive track and limiting tissue assimilation. Booji et al. (2002) described a similar scenario in blue mussels, finding that after depuration deca-BDE tissue concentrations drastically dropped, suggesting deca-BDE was bound and egested on particles. In general, high molecular weight BDEs have been shown to have low bioaccumulation rates in benthic invertebrates and other marine species (Dinn et al. 2012, Mizukawa et al. 2009).

The results of this study suggest that while PBDEs in treated wastewater discharged to the lower Snohomish mainstem are mainly composed of tetra-, penta-, and deca-BDEs, deca-BDE is not being introduced into the pelagic food web at a high rate and is predominantly accumulating in sediments and biofilms. Lower molecular weight PBDEs, including tetra- and penta-BDEs are making their way into the food chain which is of concern due to their potential impact on juvenile Chinook hormone function including growth, immune response, and parr-smolt transformation (Arkoosh et al. 2016).

Transport

While the objectives of this study did not include determining in-river transport mechanisms of PBDEs, observations can be made from collected data to infer potential transport within the river system. Water concentrations of PBDEs were found to be localized to within a few miles of a source. Elevated PBDE water concentrations near outfall 015 and the Monroe STP outfall decreased rapidly within a few miles from the outfalls. This shows that PBDEs attenuate quickly within the water column and either partition out of the dissolve phase or are diluted. Further evidence of this quick attenuation was the lack of additive or cumulative effects from upriver sources in the lower Snohomish mainstem and sloughs. Due to the hydrophobicity of PBDEs their persistence in water is limited as they will quickly bind to particulate matter and eventually settle out of the water column (Akortia et al. 2016).

Suspended sediments provide a potential transport mechanism for particulate-bound PBDEs within the river system. Results from suspended sediments collected in the Snohomish mainstem during the 2021 low flow event showed consistent concentrations of PBDEs from river mile 1.8 to 7.0, whereas PBDE water concentrations during the study were generally only elevated between river miles 1.8 and 5.0 and decreased by river mile 7.0. Additionally, PBDE water concentrations were higher during high flow events while suspended sediment PBDE concentrations were highest during the low flow event. This inconsistency in elevated concentration location and timing between water and suspended sediments may indicate that particulate bound PBDEs are transported further away from a source than dissolved phase PBDEs. Studies in other river systems have found that suspended sediments are important in

transporting PBDEs and other hydrophobic toxics (de Boer et al. 2003; Gregg et al. 2015) and therefore may increase the impacted area of a pollutant source.

Conclusions

Results of this 2019 to 2022 study support the following conclusions:

- Based on water, sediment, biofilm, and invertebrate samples collected during this study, the pathway of PBDEs impacting the lower Snohomish mainstem is the discharge of treated wastewater from the City of Everett's WPCF outfall 015.
- Treated wastewater discharges to the Skykomish River from the City of Monroe's STP cause localized increases in water concentrations of PBDEs during low flow river conditions. To a lesser extent the same is true for the City of Sultan WWTP.
- Despite evidence that effluent-derived nutrients are taken up by Skykomish River biota, migratory juvenile Chinook in this river do not appear to be impacted by PBDEs. Previous studies of resident fish from the Skykomish and Snohomish have documented the accumulation of PBDEs.
- Snoqualmie River PBDE concentrations are near background levels and do not indicate any potential impact from point source discharges.
- PBDE homologs partition differently in surface water, sediments, biofilms, and invertebrates. Surface waters and invertebrates are dominated by tetra- and penta-BDEs. Deca-BDE is the primary homolog in sediments and biofilms.
- PBDEs accumulate in sediments, biofilms, and invertebrates at concentrations greater than those measured in surface waters and wastewater.
- Two years of PBDE concentrations in invertebrate tissues from the lower Snohomish River showed a similar annual trend; the highest PBDE concentrations are in February – March with decreasing concentrations over the summer. The N isotopic ratio of invertebrate tissues suggests a possible change in effluent-derived nutrients in May and June.
- The partitioning and accumulation of tetra- and penta-BDEs into invertebrates exposes juvenile Chinook to PBDEs through their diet. Dietary exposure is potentially compounded by peak invertebrate PBDE concentrations in the spring coinciding with juvenile Chinook seaward migration and residence in the Snohomish estuary.

Recommendations

Results of this 2019 to 2022 study support the following recommendations.

- This study showed that the Everett WPCF 015 outfall is a major contributor to elevated PBDEs in the lower Snohomish mainstem. Current efforts are being undertaken through WPCF's permit to reduce sources of PBDEs from pretreatment dischargers. Continued quarterly monitoring of influent and effluent PBDE concentrations at the facility should be undertaken to determine if these efforts are effective.
- In addition to WPCF monitoring, long term monitoring of the lower Snohomish mainstem should be undertaken to determine if PBDE reduction efforts are reducing concentrations in the environment. Seasonal monitoring of invertebrates within the lower Snohomish could provide an indication of reductions in PBDE levels within the food web and their potential transfer to juvenile Chinook. As part of the continued environmental monitoring effort, PBDE levels within juvenile Chinook should be monitored every 2 to 4 years to determine if PBDE reduction efforts are having an impact on fish tissue concentrations.
- This study showed that PBDEs accumulate in sediments in the lower Snohomish mainstem. These sediments may act as a sink and have the potential to continue to release PBDEs into the food web. Further investigation is needed to determine if sediment accumulated PBDEs are bioavailable and to what degree are they released from and transformed in sediments.
- Suspended sediments collected during this study suggest particulate bound PBDEs are transported further away from sources then dissolved phase PBDE concentrations suggest. Further investigation is needed to determine the transport mechanism of PBDEs and the role particulate matter plays in the movement of these toxics throughout the river system.
- This study did not evaluate CSO contributions to in-river PBDE concentrations. CSOs within the lower Snohomish mainstem should be monitored to determine PBDE discharge concentrations and loads.
- Results from this study showed that elevated levels of PBDEs in the Snohomish and Skykomish rivers were caused by the discharge of treated wastewater from WWTPs. To determine the extent of PBDEs within these facilities, a monitoring study should be performed to determine influent and effluent PBDE concentrations at WWTPs within the Snohomish watershed including the Monroe STP and Sultan WWTP.

References

Personal Communications

Carey, A. WA State Department of Fish and Wildlife [February 15, 2024 & March 12, 2024]

References Cited in Text

Akortia, E., Okonkwo, J., Lupankwa, M., Osae, S., Daso, A., Olukunle, O., & Chaudhary, A. 2016. A review of sources, levels, and toxicity of polybrominated diphenyl ethers (PBDEs) and their transformation and transport in various environmental compartments. Environmental Reviews, 24(3), 253–273. https://doi.org/10.1139/er-2015-0081

Alvarez, D.A., 2010, Guidelines for the use of the semipermeable membrane device (SPMD) and the polar organic chemical integrative sampler (POCIS) in environmental monitoring studies: U.S. Geological Survey, Techniques and Methods 1–D4, 28 p.

Andrade, N., McConnell, L., Anderson, M., Torrents, A., and Ramirez, M. 2017. Polybrominated diphenyl ethers: Residence time in soils receiving biosolids application. *Environmental Pollution (1987)*, *222*, 412–422. https://doi.org/10.1016/j.envpol.2016.12.014

Arkoosh, M., Boylen, D., Dietrich, J., Anulacion, B., Ylitalo, G., Bravo, C., Johnson, L., Loge, F., Collier, T. 2010. Disease susceptibility of salmon exposed to polybrominated diphenyl ethers (PBDEs). Aquatic Toxicology, 98(1), 51–59. https://doi.org/10.1016/j.aquatox.2010.01.013

Arkoosh, M., Van Gaest, A., Strickland, S., Hutchinson, G., Krupkin, A., & Dietrich, J. 2016. Alteration of thyroid hormone concentrations in juvenile Chinook salmon (Oncorhynchus tshawytscha) exposed to polybrominated diphenyl ethers, BDE-47 and BDE-99. *Chemosphere (Oxford)*, *171*, 1–8. https://doi.org/10.1016/j.chemosphere.2016.12.035

Arnot, J. A., and Gobas, F. A. 2006. A review of bioconcentration factor (BCF) and bioaccumulation factor (BAF) assessments for organic chemicals in aquatic organisms. Environmental Reviews, 14(4), 257-297.

Booij, K., Zegers, B., & Boon, J. 2002. Levels of some polybrominated diphenyl ether (PBDE) flame retardants along the Dutch coast as derived from their accumulation in SPMDs and blue mussels (Mytilus edulis). Chemosphere (Oxford), 46(5), 683–688. https://doi.org/10.1016/S0045-6535(01)00232-6

Cabana, G., and Rasmussen, J. B., 1996. Comparison of aquatic food chains using nitrogen isotopes. Proceedings of the National Academy of Sciences, 93: 10844-10847.

Carey, A., Gipe, A., Hobbs, W., O'Neill, S. 2023. Investigating the Source of PBDE Contaminant Exposure in Steelhead Trout within the Major Tributaries of the Nisqually River Basin. Publication FPT23-02. Washington Department of Fish and Wildlife, Olympia.

Carey, A., West, J., Fisk, R., Langness, M., Ylitalo, G., O'Neill, S. 2019. Location Source of PBDE exposure of juvenile Chinook salmon along their out-migrant pathway through the Snohomish River, WA. p. 17 in 2018 Salish Sea Toxics Monitoring Synthesis: A Selection of Research. Edited by C.A. James, R. Jordan, M. Langness, J. Lanksbury, D. Lester, S. O'Neill, K. Song, and C. Sullivan. Puget Sound Ecosystem Monitoring Program. Tacoma, WA. 88 pp: https://www.eopugetsound.org/articles/2018-salish-sea-toxics-monitoring-synthesis

Chamberlin, J., Hall, J., Zackey, W., Leonetti, F., Rustay, M., and Rice, C. 2022. Spatial and Temporal Variability in Chinook Salmon Abundance Reflects Opportunity to Support Life History Diversity in an Estuarine Landscape. Estuaries and Coasts, 45(3), 882–896. https://doi.org/10.1007/s12237-021-00994-3

Cordell, J., Tanner, C., & Aitkin, K. 1999. Fish assemblages and juvenile salmon diets at a breached-dike wetland site, Spencer Island, Washington 1997-98. Fisheries Research Institute, School of Fisheries, University of Washington.

Couture, F., Oldford, G., Christensen, V., Barrett-Lennard, L., & Walters, C. 2022. Requirements and availability of prey for northeastern pacific southern resident killer whales. PloS One, 17(6), e0270523–e0270523. https://doi.org/10.1371/journal.pone.0270523

Cusimano, R.F. 1997. Snohomish River Estuary Dry Season TMDL Study – Phase II. Washington State Department of Ecology, Olympia, WA. Publication No. 97-325. https://fortress.wa.gov/ecy/publications/SummaryPages/97325.html

de Boer, J., Wester, P., van der Horst, A., & Leonards, P. 2003. Polybrominated diphenyl ethers in influents, suspended particulate matter, sediments, sewage treatment plant and effluents and biota from the Netherlands. Environmental Pollution (1987), 122(1), 63–74. https://doi.org/10.1016/S0269-7491(02)00280-4

deBruyn, A.M.H. and Rasmussen, J.B., 2002. Quantifying assimilation of sewage-derived organic matter by riverine benthos. Ecol. Appl. 12, 511–520

Dietrich, J., Strickland, S., Hutchinson, G., Van Gaest, A., Krupkin, A., Ylitalo, G., & Arkoosh, M. 2015. Assimilation Efficiency of PBDE Congeners in Chinook Salmon. Environmental Science & Technology, 49(6), 3878–3886. https://doi.org/10.1021/es5057038

Dinn, P., Johannessen, S., Ross, P., Macdonald, R., Whiticar, M., Lowe, C., & van Roodselaar, A. 2012. PBDE and PCB accumulation in benthos near marine wastewater outfalls: The role of sediment organic carbon. Environmental Pollution (1987), 171, 241–248. https://doi.org/10.1016/j.envpol.2012.07.023

Ecology [Washington State Department of Ecology]. 2019 [recertified/not published]. Standard Operating Procedure EAP001, Version 4.1: Standard Operating Procedures for Conducting Studies using Semi-Permeable Membrane Devices (SPMDs). Olympia, WA. For a copy of this document, please submit a request online or visit ecology.wa.gov/publicrecords.

Ecology [Washington State Department of Ecology]. 2022a [recertified/not published]. Standard Operating Procedure EAP015, Version 1.5: Manually Obtaining Surface Water Samples. Olympia, WA. For a copy of this document, please submit a request online or visit ecology.wa.gov/publicrecords.

Ecology [Washington State Department of Ecology]. 2022b [recertified/not published]. Standard Operating Procedure EAP111, Version 1.14: Periphyton Sampling, Processing, and Identification in Streams and Rivers. Olympia, WA. For a copy of this document, please submit a request online or visit ecology.wa.gov/publicrecords.

Ecology [Washington State Department of Ecology]. 2023 [recertified/not published] Standard Operating Procedure EAP040 Version 1.4: Obtaining Freshwater Sediment Samples. Olympia, WA. For a copy of this document, please submit a request online or visit ecology.wa.gov/publicrecords.

Ecology [Washington Department of Ecology] and Herrera Environmental Consultants, Inc. Phase 3: Loadings of Toxic Chemicals to Puget Sound from POTW Discharge of Treated Wastewater. Ecology Publication Number 10-10-057. December 2010. Olympia, Washington.

EPA (US Environmental Protection Agency). Technical Fact Sheet- Polybrominated Diphenyl Ethers (PBDEs). 2017. https://semspub.epa.gov/work/HQ/190438.pdf

Gregg, T., Prahl, F., & Simoneit, B. 2015. Suspended particulate matter transport of polycyclic aromatic hydrocarbons in the lower Columbia River and its estuary. Limnology and Oceanography, 60(6), 1935–1949. https://doi.org/10.1002/lno.10144

Hall, J.E., Khangaonkar, T.P., Rice, C.A., Chamberlin, J., Zackey, T., Leonetti, F., Rustay, M., Fresh, K., Kagley, A. and Rowse, M., 2018. Characterization of salinity and temperature patterns in a large river delta to support tidal wetland habitat restoration. Northwest Science, 92(1), pp.36-52.

Heaton, T.H.E., 1986. Isotopic studies of nitrogen pollution in the hydrosphere and atmosphere: a review. Chemical Geology: Isotope Geoscience section 59, 87–102

Hicks, K.A., Loomer, H.A., Fuzzen, M.L.M., Kleywegt, S., Tetreault, G.R., McMaster, M.E., and Serovs, M.R. 2017. δ15N tracks changes in the assimilation of sewage-derived nutrients into a riverine food web before and after major process alterations at two municipal wastewater treatment plants. Ecol. Indic. 72, 747–758.

Hiebert, T.C. 2015. *Americorophium spinicorne*. In: Oregon Estuarine Invertebrates: Rudys' Illustrated Guide to Common Species, 3rd ed. T.C. Hiebert, B.A. Butler and A.L. Shanks (eds.). University of Oregon Libraries and Oregon Institute of Marine Biology, Charleston, OR.

Huckins, J.N., Petty, J.D., and Booij, K. 2006. Monitors of organic chemicals in the environment: Semipermeable membrane devices. Springer, New York, USA. 223p.

Johnson, A., Seiders, K., Deligeannis, C., Kinney, K., Sandvik, P., Era-Miller, B., and Alkire, D. 2006. PBDE Flame Retardants in Washington Rivers and Lakes: Concentrations in Fish and QAPP: Assessing Sources of Toxic Chemicals Impacting Juvenile Chinook Salmon 43 Water, 2005-06. Washington State Department of Ecology, Olympia. Publication No. 06-03-027. https://fortress.wa.gov/ecy/publications/SummaryPages/0603027.html

Li, B., Danon-Schaffer, M., Li, L., Ikonomou, M., and Grace, J. 2012. Occurrence of PFCs and PBDEs in Landfill Leachates from Across Canada. Water, Air, and Soil Pollution, 223(6), 3365–3372. https://doi.org/10.1007/s11270-012-1115-7

Marine Sediment Monitoring Team. 2022a. Sediment Quality in Puget Story Map Collection. Washington State Department of Ecology, Olympia. https://storymaps.arcgis.com/collections/aaec1a6656ff43e098d209c75ce00244

Marine Sediment Monitoring Team. 2022b. Focus on: Sediment Quality in East Possession Sound. Washington State Department of Ecology, Olympia. Publication 22-03-025. https://fortress.wa.gov/ecy/publications/SummaryPages/2203025.html

Mathieu, C. and Wong, S. 2016. Brominated Flame Retardants, Alkylphenolic Compounds, and Hexabromocyclododecane in Freshwater Fish of Washington State Rivers and Lakes. Washington State Department of Ecology, Olympia. Publication No. 16-03-012. https://fortress.wa.gov/ecy/publications/SummaryPages/1603012.html

Meador, J., James, A., Ball, S., McIntyre, J. 2022 King County Orca Proviso Wastewater Effluent Discharge Assessment: Impact to Marine Organisms. 2022. King County.

Meador, J., Ylitalo, G., Sommers, F. and Boyd, D. 2010. Bioaccumulation of polychlorinated biphenyls in juvenile Chinook salmon (Oncorhynchus tshawytscha) outmigrating through a contaminated urban estuary: dynamics and application. Ecotoxicology, 19: 141-152. https://doi.org/10.1007/s10646-009-0399-x

Mizukawa, K., Takada, H., Takeuchi, I., Ikemoto, T., Omori, K., & Tsuchiya, K. 2009. Bioconcentration and biomagnification of polybrominated diphenyl ethers (PBDEs) through lower-trophic-level coastal marine food web. *Marine Pollution Bulletin*, *58*(8), 1217–1224. https://doi.org/10.1016/j.marpolbul.2009.03.008

Morace, J. 2012. Reconnaissance of contaminants in selected wastewater-treatment-plant effluent and stormwater runoff entering the Columbia River, Columbia River Basin, Washington and Oregon, 2008-10. *Scientific Investigations Report. United States Geological Survey*, *2012–5068*.

O'Neill, S., Carey, A., Harding, L., West, J., Ylitalo, G., and Chamberlin, J. 2020. Chemical tracers guide identification of the location and source of persistent organic pollutants in juvenile Chinook salmon (*Oncorhynchus tshawytscha*), migrating seaward through an estuary with multiple contaminant inputs. The Science of the Total Environment, 712, 135516–135516. https://doi.org/10.1016/j.scitotenv.2019.135516

O'Neill, S., Carey, A., Lanksbury, J., Niewolny, L., Ylitalo, G., Johnson, L., West, J. 2015. Toxic contaminants in juvenile Chinook salmon (*Oncorhynchus tshawytscha*) migrating through estuary, nearshore and offshore habitats of Puget Sound. Publication FPT16-02. Washington Dept. of Fish and Wildlife, Olympia. https://wdfw.wa.gov/publications/01796

Partridge, V., Weakland, S., Dutch, M., Long, E., Welch, K. 2014. Sediment Quality in Port Gardner Bay, 2012. Washington State Department of Ecology, Olympia. Publication 14-03-035. https://apps.ecology.wa.gov/publications/SummaryPages/1403035.html

Post, D.M., 2002. Using stable isotopes to estimate trophic position: models, methods, and assumptions. Ecology. 83: 703-718.

Quinn, T. 2005. The Behavior and Ecology of Pacific Salmon and Trout. University of Washington Press, Seattle, WA.

Quinn, T., & Losee, J. 2022. Diverse and changing use of the Salish Sea by Pacific salmon, trout, and char. Canadian Journal of Fisheries and Aquatic Sciences, 79(6), 1003–1021. https://doi.org/10.1139/cjfas-2021-0162

Savage, C., 2005. Tracing the influence of sewage nitrogen in a coastal ecosystem using stable nitrogen isotopes. Ambio 34, 145–150.

Science Applications International Corporation (SAIC). 2009. Sediment Characterization Study in Port Gardner and Lower Snohomish Estuary, Port Gardner, WA: Final Data Report. Washington Department of Ecology, Olympia. Publication 09-09-227. https://apps.ecology.wa.gov/publications/SummaryPages/0909227.html

Seiders, K., Deligannis, C., and Sandvik, P. 2007. Washington State Toxics Monitoring Program: Contaminants in Fish Tissue from Freshwater Environments in 2004 and 2005. Washington State Department of Ecology, Olympia. Publication No. 07-03-024. https://fortress.wa.gov/ecy/publications/SummaryPages/0703024.html

Sloan, C., Anulacion, B., Bolton, J., Boyd, D., Olson, O., Sol, S., Ylitalo, G., & Johnson, L. 2010. Polybrominated Diphenyl Ethers in Outmigrant Juvenile Chinook Salmon from the Lower Columbia River and Estuary and Puget Sound, Washington. Archives of Environmental Contamination and Toxicology, 58(2), 403–414. https://doi.org/10.1007/s00244-009-9391-y

Stevenson, R.J. and Bahls, L. Barbour, M.T., J. Gerritsen, B.D. Snyder, and J.B. Stribling, 1999. Rapid Bioassessment Protocols for Use in Streams and Wadeable Rivers: Periphyton, Benthic Macroinvertebrates and Fish, Second Edition. EPA 841-B-99-002. U.S. Environmental Protection Agency; Office of Water; Washington, D.C.

Washington Department of Ecology and Herrera Environmental Consultants, Inc. Phase 3: Loadings of Toxic Chemicals to Puget Sound from POTW Discharge of Treated Wastewater. Ecology Publication Number 10-10-057. December 2010. Olympia, Washington. https://apps.ecology.wa.gov/publications/SummaryPages/1010057.html

Washington State Polybrominated Diphenyl Ether (PBDE) Chemical Action Plan: Final Plan. 2006. Publication 05-07-048. Washington State Department of Ecology, Olympia.

Wasser, S. K., Lundin, J. I., Ayres, K., Seely, E., Giles, D., Balcomb, K., Hempelmann, J., Parsons, K., & Booth, R. 2017. Population growth is limited by nutritional impacts on pregnancy success in endangered Southern Resident killer whales (Orcinus orca). *PloS One*, *12*(6), e0179824–e0179824. https://doi.org/10.1371/journal.pone.0179824

Wong, S. 2022. Chemicals of Emerging Concern in Pretreated Industrial Wastewater in Northwestern Washington State: Screening Study Results, 2021. Publication 22-03-013. Washington State Department of Ecology, Olympia. https://apps.ecology.wa.gov/publications/SummaryPages/2203013.html

Glossary, Acronyms, and Abbreviations

Glossary

Bioaccumulation: The process in which a chemical substance is absorbed in an organism by all routes of exposure as occurs in the natural environment (i.e., dietary and ambient environmental sources) (Arnot and Gobas 2006).

Bioconcentration: The process by which a chemical substance is absorbed by an organism from the ambient environment only through its respiratory and dermal surfaces (i.e., chemical exposure through diet is not included) (Arnot and Gobas 2006).

Bromodiphenyl ether (BDE): A group of organic compounds composed of a diphenyl ether molecule (a benzene ring connected to an oxygen atom, which is connected to another benzene ring) with one or more bromine atoms attached to the aromatic rings. These compounds are part of a larger class of chemicals known as polybrominated diphenyl ethers (PBDEs), which are used primarily as flame retardants in various materials like textiles, electronics, and plastics.

Combined sewer overflow (CSO): A system to discharge combined stormwater and wastewater during high flow weather events to prevent sewer systems from backing up, causing flooding.

Conductivity: A measure of water's ability to conduct an electrical current. Conductivity is related to the concentration and charge of dissolved ions in water.

Decabromodiphenyl ether commercial mix (c-decaBDE): A mixture of various brominated compounds, primarily decabromodiphenyl ether (a flame retardant), used in commercial flame retardant applications.

Decabromodiphenyl ether homolog (Deca-BDE): The polybrominated diphenyl ether congener decabromodiphenyl ether.

Delta 13 C (δ^{13}C): The stable carbon isotope ratio (13C/12C) used in environmental science to trace sources of carbon, such as food sources or environmental changes.

Delta 15 N (δ^{15}N): The stable nitrogen isotope ratio (15N/14N) that is used in ecological and environmental studies to trace nitrogen sources or food webs.

Department of Ecology's Toxics Studies Unit (TSU): A unit within Washington State Department of Ecology responsible for studying toxic substances and their effects on the environment.

Dibromodiphenyl ether homolog (Di-BDE): A group of polybrominated diphenyl ether congeners containing two bromine atoms. Related to flame retardants and used in various industries.

Dissolved organic carbon (DOC): Organic carbon dissolved in water, often from natural or anthropogenic sources, that can affect water quality and ecosystems.

Dissolved oxygen (DO): A measure of the amount of oxygen dissolved in water.

Ebey Slough (ES): A slough located in the Snohomish estuary a water body located in Washington State, part of the Puget Sound watershed.

Effluent: An outflowing of water from a natural body of water or from a man-made structure. For example, the treated outflow from a wastewater treatment plant.

Environmental Information Management database (EIM): A database used by environmental agencies to store and manage environmental data for reporting and decision-making.

Heptabromodiphenyl ether homolog (Hepta-BDE): A group of polybrominated diphenyl ether congeners containing seven bromine atoms. Related to flame retardants used in various industries.

Hexabromodiphenyl ether homolog (Hexa-BDE): A group of polybrominated diphenyl ether congeners containing six bromine atoms. Related to flame retardants used in various industries.

High Resolution Gas Chromatography/High Resolution Mass Spectrometry (HRGC/HRMS): Analytical techniques used for precise identification and quantification of compounds in complex mixtures.

Hydrophobicity: a qualitative term used to describe the extent of interaction between solid and water phases. A hydrophobic substance prefers a nonaqueous environment.

Low-density polyethylene (LDPE): A type of plastic commonly used in packaging and containers, known for its low-density molecular structure.

J Data Qualifier (J): A data qualifier representing an analyte result was positively identified and the associated numerical value is the approximate concentration of the analyte in the sample.

Measurement quality objects (MQO): Tools or metrics used to assess the accuracy and precision of measurement results.

Method Blank (MB): A sample used in laboratory analysis to check for contamination or background levels of a substance in the environment.

NJ Data Qualifier (NJ): The analysis indicates that the presence of the analyte has been tentatively identified, and the associated numerical value represents an approximate concentration. Identification needs further confirmation.

National Oceanic and Atmospheric Administration (NOAA): A U.S. government agency focused on monitoring and researching the oceans, atmosphere, and climate.

Non-detect (ND): Refers to results where a substance was not detected above the limit of detection during analysis.

Nonabromodiphenyl ether homolog (Nona-BDE): A group of polybrominated diphenyl ether congeners containing nine bromine atoms. Related to flame retardants used in various industries.

Not Analyzed (ND): A designation indicating that a sample was not subjected to analysis.

Number of samples (n): The count of individual samples collected for analysis in a study.

Octabromodiphenyl ether homolog (Octa-BDE): A group of polybrominated diphenyl ether congeners containing eight bromine atoms. Related to flame retardants used in various industries.

Organic carbon-water partition coefficient (K_{OC}): A ratio used to describe the distribution of organic compounds between water and organic matter, often used in environmental chemistry.

Parameter: Water quality constituent being measured (analyte). A physical, chemical, or biological property whose values determine environmental characteristics or behavior.

Partition coefficient: the ratio of the concentration of a substance in one matrix or phase to the concentration in a second phase when the two concentrations are at equilibrium. It can describe the hydrophobicity of a substance.

Pentabromodiphenyl ether commercial mix (c-pentaBDE): A group of polybrominated diphenyl ether congeners containing five bromine atoms. Related to flame retardants used in various industries.

Performance reference compound (PRC): A group of compounds spiked into semipermeable membrane devices and used to measure the uptake rate of target analytes.

pH: A measure of the acidity or alkalinity of water. A low pH value (0 to 7) indicates that an acidic condition is present, while a high pH (7 to 14) indicates a basic or alkaline condition. A pH of 7 is considered neutral. Since the pH scale is logarithmic, a water sample with a pH of 8 is ten times more basic than one with a pH of 7.

Point source: Sources of pollution that discharge at a specific location from pipes, outfalls, and conveyance channels to a surface water. Examples of point source discharges include municipal wastewater treatment plants, municipal stormwater systems, industrial waste treatment facilities, and construction sites where more than 5 acres of land have been cleared.

Polybrominated diphenyl ethers (PBDEs): A group of brominated compounds used as flame retardants, known for their persistence in the environment and potential health risks.

Relative percent difference (RPD): A measure of the variation between two values, often used to compare replicates in laboratory experiments.

Reporting Limit (RL): The lowest concentration of a substance that can be reliably measured and reported by a laboratory.

Revolutions per minute (RPM): A unit of rotational speed, used to measure how many full rotations an object makes in one minute.

River mile (RM): A unit of measurement used to describe the location of a point along a river, based on its distance from the river's mouth.

Semipermeable membrane device (SPMD): A device used in environmental sampling to collect pollutants from water or air by allowing certain substances to pass through a membrane.

Sewage Treatment Plant (STP): A facility that treats and processes wastewater to remove contaminants and produce treated water for release or reuse.

Snohomish Mainstem (SM): The main river channel of the Snohomish River in Washington State.

Standard Deviation (SD): A statistical measure of the amount of variation or dispersion in a set of data.

Standard Operating Procedures (SOPs): Established protocols or instructions that outline how specific processes or tasks should be carried out in a consistent manner.

Stormwater: The portion of precipitation that does not naturally percolate into the ground or evaporate but instead runs off roads, pavement, and roofs during rainfall or snow melt. Stormwater can also come from hard or saturated grass surfaces such as lawns, pastures, playfields, and from gravel roads and parking lots.

Suspended sediment concentration (SSC): The concentration of fine particles, such as soil or organic matter, suspended in water.

Tetrabromodiphenyl ether (Tetra-BDE): A group of polybrominated diphenyl ether congeners containing four bromine atoms. Related to flame retardants used in various industries.

Total Kjeldahl Nitrogen (TKN): A measure of organic nitrogen and ammonium nitrogen in water, used to assess water quality.

Total Nitrogen (TN): The sum of all forms of nitrogen in a sample, including organic nitrogen, ammonium, nitrate, and nitrite.

Total Organic Carbon (TOC): A measure of the total amount of organic carbon present in a sample, often used to assess water quality.

Toxics Biological Observation System (TBiOS): A team within the Washington State Department of Fish and Wildlife the monitors the geographic extent and magnitude of toxic contaminants in fish and other organisms in Puget Sound, Washington.

Tribromodiphenyl ether homolog (Tri-BDE): A group of polybrominated diphenyl ether congeners containing three bromine atoms. Related to flame retardants used in various industries.

U Data Qualifier: The analyte was analyzed for but was not detected at the reported level of quantitation.

UJ Data Qualifier (UJ): The analyte was not detected at or above the reported quantitation limit. However, the reported quantitation limit is approximate and may or may not represent the actual limit of quantitation necessary to accurately and precisely measure the analyte in the sample.

United States Geological Survey (USGS): A scientific agency in the U.S. that studies the natural resources and natural hazards of the country.

U.S. Environmental Protection Agency (EPA): A federal agency in the United States that enforces laws and regulations to protect human health and the environment.

Washington State Department of Ecology (Ecology): A state agency in Washington responsible for protecting and improving the environment, including monitoring air, water, and land quality.

Washington Department of Fish & Wildlife (WDFW): A state agency that manages and protects fish, wildlife, and habitats in Washington.

Wastewater Treatment Plant (WWTP): A facility that processes and cleans wastewater before releasing it into the environment or reusing it.

Water-octanol partitioning coefficient (K_{ow}): A value that describes the distribution of a substance between water and octanol (a model for lipid membranes), used to assess the potential for bioaccumulation in organisms.

Water Resource Inventory Area (WRIA): A geographic area used to manage and monitor water resources, particularly in Washington State.

Watershed: A drainage area or basin in which all land and water areas drain or flow toward a central collector, such as a stream, river, or lake at a lower elevation.

Acronyms and Abbreviations

CSO	Combined sewer overflow
BDE	bromodiphenyl ether
c-decaBDE	Decabromodiphenyl ether commercial mix
c-octaBDE	Octabromodiphenyl ether commercial mix
c-pentaBDE	Pentabromodiphenyl ether commercial mix
Deca-BDE	Decabromodiphenyl ether homolog
$\delta^{15}N$	delta 15 N
$\delta^{13}C$	delta 13 C
Di-BDE	Dibromodiphenyl ether homolog
DOC	Dissolved organic carbon
Ecology	Washington State Department of Ecology
EIM	Environmental Information Management database
EPA	U.S. Environmental Protection Agency
ES	Ebey Slough
Hepta-BDE	Heptabromodiphenyl ether homolog
Hexa-BDE	Hexabromodiphenyl ether homolog
HRGC/HRMS	High Resolution Gas Chromatography/High Resolution Mass Spectrometry
J	J Data Qualifier: See Glossary
K_{OC}	Organic carbon-water partition co-efficient
K_{OW}	Water-octanol partitioning coefficient
LPDE	Low-density polyethylene
MB	Method Blank
MQO	Measurement quality objects
n	number of samples
NA	Not Analyzed
ND	Non-detect
NJ	NJ Data Qualifier: See Glossary
NOAA	National Oceanic and Atmospheric Administration
Nona-BDE	Nonabromodiphenyl ether homolog
Octa-BDE	Octabromodiphenyl ether homolog
OPR	Ongoing precision and recovery
PBDE	polybrominated diphenyl ethers
Penta-BDE	Pentabromodiphenyl ether homolog
PRC	Performance reference compound
RL	Reporting Limit
RM	river mile
RPD	relative percent difference
RPM	Revolutions per minute
SD	Standard Deviation
SM	Snohomish Mainstem
SPMD	Semipermeable membrane device
SOP	standard operating procedures
SSC	Suspended sediment concentration
STP	Sewage Treatment Plant

TBiOS	Toxics Biological Observation System
Tetra-BDE	Tetrabromodiphenyl ether
TKN	Total Kjeldahl Nitrogen
TN	Total Nitrogen
TOC	Total organic carbon
Tri-BDE	Tribromodiphenyl ether homolog
TSU	Department of Ecology's Toxics Studies Unit
U	U Data Qualifier: See Glossary
UJ	UJ Data Qualifier: See Glossary
USGS	United States Geological Survey
WDFW	Washington Department of Fish & Wildlife
WRIA	Water Resource Inventory Area
WWTP	Wastewater treatment plant

Units of Measurement

°C	degrees centigrade
CFS	cubic feet per second
cm	centimeter
g	gram, a unit of mass
MGD	million gallons per day
MG	million gallon
mg/L	milligrams per liter
mL	milliliters
pg/g	picograms per gram (parts per trillion)
pg/L	picograms per liter (parts per quadrillion)
µm	micrometer
ww	wet weight

Appendices

Appendix A. Study Results

Appendix A is available only on the internet, linked to this report at:
https://apps.ecology.wa.gov/publications/SummaryPages/2503002.html

Table A1. Summary of the measurement quality objectives and method blank censoring rates for Semipermeable Membrane Device (SPMD), Tissue, and Sediment PBDE analysis

Matrix	Duplicate samples (±50%)	Verification standards (50-150%)	Surrogate Standards (25-150%[a])	Method Detection Limits	Method Blank censoring rate
SPMDs	Pass with exceptions	Pass	72% of samples Pass	Pass with exceptions	5.7%
Tissue	Pass with exceptions	Pass	Pass	Pass with exceptions	0.6%
Sediments	Pass with exceptions	Pass	Pass	Pass with exceptions	7.9%

Table A2. Summary of method detection limits for Semipermeable Membrane Device (SPMD) (pg/sample), Tissue (pg/g), and Sediment (Seds.) (pg/g) PBDE analysis performed for this study. Standard deviation (SD)

PBDE Homolog	SPMD Mean	SPMD Median	SPMD Min	SPMD Max	SPMD SD	Tissue Mean	Tissue Median	Tissue Min	Tissue Max	Tissue SD	Seds. Mean	Seds. Median	Seds. Min	Seds. Max	Seds. SD
Di	12.7	9	2.1	50.6	9.56	0.5	0.2	0.5	3.7	0.69	0.2	0.1	0.1	0.6	0.11
Tri	27	17.8	1.8	167	28.1	0.4	0.3	0.05	2.2	0.51	0.2	0.2	0.1	0.4	0.09
Tetra	8.5	4.9	2	67.1	9.05	0.3	0.1	0.05	2.2	0.51	0.2	0.1	0.1	1.0	0.10
Penta	81.1	54.2	3.2	442	77.1	0.3	0.1	0.05	2.0	0.44	0.3	0.3	0.1	1.7	0.23
Hexa	17.4	8.7	2	98.9	14.8	0.5	0.2	0.05	2.7	0.55	0.3	0.2	0.1	2.7	0.38
Hepta	14.4	9.1	2	97.9	14.8	0.5	0.2	0.05	4.4	0.77	0.4	0.3	0.1	2.1	0.32
Octa	21.8	17.4	2	202	36.1	1.3	0.6	0.05	10.7	2.0	1.0	0.6	0.1	5.5	1.1
Nona	68.4	38.8	3.7	289	64.4	2.3	1	0.05	17.0	3.8	0.9	0.9	0.1	2.6	0.76
Deca	1637	1235	252	8190	1292	18.6	12.4	1.4	86.5	20.3	19.0	17.0	2	74.6	17.5

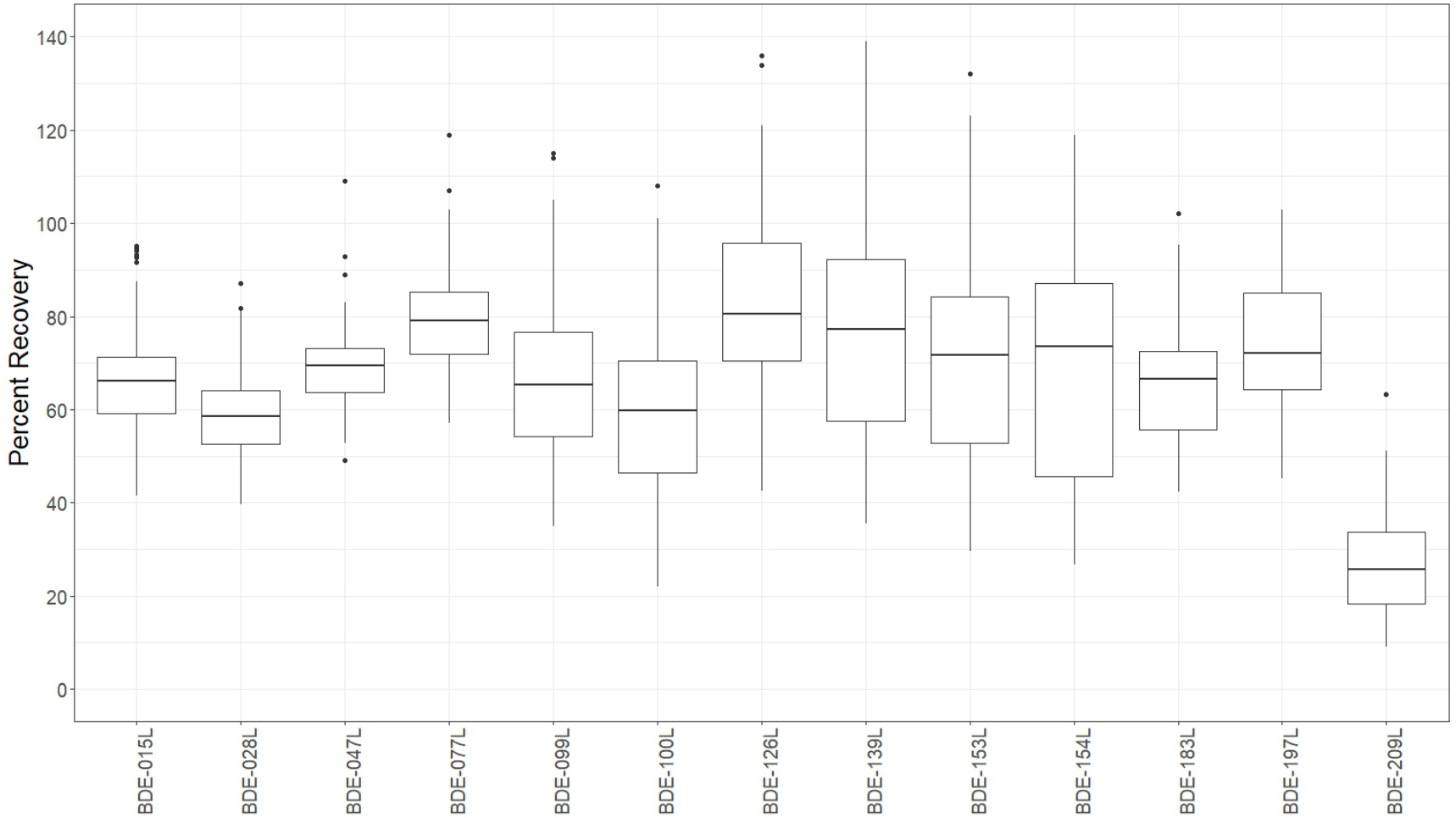

Figure A1 Summary of percent recoveries for PBDE analysis surrogates in SPMD samples.

Table A3. Summary of PBDE concentrations (pg/L) in Semipermeable Membrane Device (SPMD) field blanks and manufacturing blanks (day zero) for each SPMD deployment batch

Sampling Event	Day Zero (Σ Di - Octa-BDE	Field Blank 1 (Σ Di - Octa-BDE	Field Blank 2 (Σ Di - Octa-BDE	Field Blank 3 (Σ Di - Octa-BDE	Di-BDE Event Mean	Tri-BDE Event Mean	Tetra-BDE Event Mean	Penta-BDE Event Mean	Hexa-BDE Event Mean	Hepta-BDE Event Mean	Octa-BDE Event Mean	Σ Di - Octa-BDE Event Mean
2019 Low Flow	4.80	4.88	5.28	5.12	0.03	0.05	1.35	1.73	0.33	0.16	1.38	5.02
2020 Low Flow	1.49	2.66	—	—	0.03	0.03	0.80	0.29	0.08	0.13	0.71	2.08
2021 High Flow	3.63	0.83	1.98	—	0.03	0.04	1.07	1.01	0.21	0.15	1.04	2.14
2021 Low Flow	3.01	4.05	—	—	0.03	0.07	0.67	1.03	0.19	0.15	1.40	3.53
2022 High Flow	2.69	8.76	0.94	—	0.02	0.03	0.24	0.29	0.18	0.14	3.24	4.13
2022 Low Flow	1.47	1.08	4.26	—	0.03	0.04	0.66	0.78	0.19	0.14	1.89	2.27
Study Mean	2.85	3.62*	—	—	0.03	0.04	0.80	0.86	0.20	0.15	1.61	3.20
Study Median	2.85	4.05*	—	—	0.03	0.04	0.73	0.90	0.19	0.15	1.39	2.90
Study Min	1.47	0.82*	—	—	0.02	0.03	0.24	0.29	0.08	0.13	0.71	2.08
Study Max	4.80	8.76*	—	—	0.03	0.07	1.35	1.73	0.33	0.16	3.24	5.02

Table A4. Summary of PBDE homolog concentrations (Conc.) detection rates (detect. rate), and method blank (MB) censoring rates (censor. rate) for Semipermeable Membrane Device (SPMD) samples, SPMD manufacturing blanks (field blanks and day zero blanks), and laboratory method blanks.

Homolog concentration represent the concentrations > detection limits and are reported as pg/sample.

Homolog	SPMD Conc. Mean	SPMD Conc. Median	SPMD Conc. Range	SPMD Detect. Rate	SPMD Censor. Rate	Manu-facturing Blank Conc. Mean	Manu-facturing Blank Conc. Median	Manu-facturing Blank Conc. Range	Manu-facturing Blank Detect. Rate	Manu-facturing Blank MB Censor. Rate	Method Blanks Conc. Mean	Method Blanks Conc. Median	Method Blanks Conc. Range	Method Blanks Detect. Rate
Di	143	74.2	5.9–1800	87.3%	1.1%	34.5	38.2	8.27–71.9	35.3%	12.5%			13.9	4.17%
Tri	673	220	4.21–18900	61.2%	0%	26.9	21.1	2.52–90.0	57.6%	6.12%	19.1	19.1	7.04–31.2	6.67%
Tetra	1289	141	7.38–63500	98.8%	0%	168	41.6	2.14–1770	74.3%	13.9%	48.8	11.8	6.68–265	25.0%
Penta	1441	461	18.6–21400	62.8%	0%	216	195	8.29–1040	61.8%	14.3%	33.3	18.2	3.26–164	36.1%
Hexa	134	63.4	6.11–870	65.0%	0%	29.5	23.0	7.41–91.9	50.0%	11.8%	7.72	5.02	2.76–15.0	19.4%
Hepta	60.6	44.0	3.89–726	43.6%	13.2%	33.5	29.8	12.8–57.5	31.4%	31.2%	7.06	6.28	4.06–12.4	27.8%
Octa	123	81.7	13.1–822	83.3%	30.4%	122	91.0	15.3–648	76.5%	26.9%	22.8	24.4	5.9–36.7	33.3%
Nona	243	194	33.7–1590	80.2%	49.2%	235	177	49.1–1010	85.3%	58.6%	67.4	66.7	38.8–112	66.7%
Deca	2520	2085	767–11400	59.3%	31.2%	3980	1650	1050–22800	76.5%	46.2%	318	318	250–384	41.7%

Table A5. Summary of dissolved organic carbon (DOC), total organic carbon (TOC), and suspended sediment concentration (SSC) for water samples collected during the deployment, midpoint check, and retrieval of Semipermeable Membrane Device (SPMD) samplers.

Results are categorized by waterbody and river flow condition (high and low). % <RL = Percent of results qualified as below the method reporting limit (RL).

Sampling Region	Parameter	Low Flow Mean	Low Flow Median	Low Flow Range	Low Flow % <RL	High Flow Mean	High Flow Median	High Flow Range	High Flow % <RL
Lower Snohomish Mainstem	DOC	1.1	1.0	0.8–1.8	0	1.2	1.1	0.9–1.8	0
	TOC	1.2	1.2	0.9–2.0	0	1.3	1.2	1–1.8	0
	SSC	6.6	4.0	2.0–41	0	4.0	4.0	2.0–9.0	0
Upper Snohomish Mainstem	DOC	1.0	0.8	0.6–2.1	0	1.2	1.1	0.8–1.8	0
	TOC	1.1	0.9	0.7–2.2	0	1.3	1.2	0.8–1.9	0
	SSC	4.5	3.0	1.0–23	0	4.9	3.5	1.0–13	0
Sloughs	DOC	1.8	1.5	0.8–5.4	0	1.4	1.2	0.9–2.1	0
	TOC	2	1.5	0.8–6.0	0	1.5	1.4	1.0–2.3	0
	SSC	9.2	6	3.0–41	0	7.2	6.5	2.0–17	0
Skykomish	DOC	0.8	0.7	0.5–2.1	14.7	0.8	0.7	0.5–1.4	12.5
	TOC	0.9	0.8	0.5–2.2	14.7	0.8	0.8	0.5–1.4	12.5
	SSC	3.4	2	0.5–28	2.56	1.9	1.5	0.5–8.0	6.25
Snoqualmie	DOC	1.2	0.8	0.5–2.4	5.26	0.9	0.8	0.5–1.2	0
	TOC	1.2	0.8	0.6–2.5	0	0.9	0.9	0.6–1.3	0
	SSC	5.0	3.0	0.6–44	0	3.1	3.0	0.7–7.0	0

Table A6. Summary of grain size percent fractions, percent solids, percent total organic carbon (%TOC), and percent total nitrogen (%TN) results for benthic sediments collected in the lower and upper Snohomish mainstem and sloughs.

Site	Sampling Region	% Gravel	%Very Coarse Sand	% Coarse Sand	% Medium Sand	% Fine Sand	% Very Find Sand	% Silt	% Clay	% Solids	% TOC	% TN
SNOH01.8	Lower Snohomish Mainstem	0	0.3	0.7	1.8	27.7	39.6	24.3	5.7	59.2	0.9	0.1
SNOH02.9	Lower Snohomish Mainstem	0.3	0.9	1.9	6.4	66.1	16.4	6.1	1.8	67.7	0.6	0.1
SNOH3.9	Lower Snohomish Mainstem	0.0	0.4	0.8	4.5	35.6	24.6	28.3	5.7	59.6	1.0	0.1
SNOH12.0	Lower Snohomish Mainstem	0.0	1.1	13.8	21.7	11.6	18.4	30.4	3.0	50.3	1.5	0.1
SNOH13.0	Lower Snohomish Mainstem	0.0	0.7	1.3	2.7	12.2	23.9	54.1	5.1	29.2	1.8	0.2
SS0.0	Sloughs	0.0	0.3	1.5	26.6	30.7	15.2	20.6	5.1	62.8	0.8	0.1
SS01.7	Sloughs	0.1	0.4	1.0	5.0	41.4	29.9	17.2	4.9	55.9	0.9	0.1
ES0.0	Sloughs	0.2	0.4	0.6	1.0	28.6	44.3	19.9	4.9	59.1	0.7	0.1
ES04.3	Sloughs	0.0	0.4	0.7	1.1	17.4	37.3	36.1	7.0	55.1	0.9	0.1
Ditch 1	—	1.2	1.4	2.0	2.6	3.0	6.8	69.4	11.7	34.6	3.9*	0.3*
Union Marsh 1	Sloughs	0.2	0.8	2.9	8.6	12.2	14.7	44.7	8.5	49.1	5.7*	0.4*
Union Marsh 2	Sloughs	0.2	0.2	0.5	1.6	34.3	28.6	27.1	4.5	67.4	1.3*	0.1*

*Percent C and N were analyzed at Manchester Environmental Laboratory using method EAP440.1.

Table A7. Summary of percent solids, total organic carbon (%TOC), and total nitrogen (%TN) results for suspended sediments collected during low (2021) and high (2022) flow river conditions.

Site	Sampling Region	Low Flow % solids	Low Flow % TOC	Low Flow % TN	High Flow % solids	High Flow % TOC	High Flow % TN
SNOH01.8	Lower Snohomish Mainstem	45.0	2.8	0.2	48.7	2.8	0.2
SNOH02.9	Lower Snoh. Mainstem	54.8	2.0	0.2	61.6	0.9	0.1
SNOH03.9	Lower Snoh. Mainstem	53.8	3.3	0.2	51.8	3.1	0.2
SNOH07.0	Upper Snoh. Mainstem	49.6	2.4	0.2	52.5	2.5	0.1
SNOH13.0	Upper Snoh. Mainstem	_	_	_	61.4	1.7	0.1

Table A8. Summary of total organic carbon (%TOC), total nitrogen (%TN), lipid (% lipid) results for biofilm samples collected from the Snohomish, Skykomish, and Snoqualmie Rivers.

Site	Sampling Region	%TOC	%TN	%lipid
SNOH01.8	Lower Snoh. Mainstem	6.5	1.1	0.2
SNOH02.9	Lower Snoh Mainstem	9.4	1.7	0.2
SNOH20.0	Upper Snoh. Mainstem	2.1	0.3	0.1
SKY24.5	Skykomish River	4.5	0.7	0.2
SKY34.5	Skykomish River	6.0	0.6	0.1
SKY40.5	Skykomish River	4.0	0.4	0.1
SNOQ23.0	Snoqualmie River	8.2	1.1	0.3
SNOQ40.7	Snoqualmie River	3.2	0.4	0.1
SNOQ47.8	Snoqualmie River	3.7	0.4	0.2

Table A9. Results of PBDEs in SPMDs

SPMD t-PBDE results are the sum of di-octa BDEs

2019 Low Flow

Site ID	Site Name	Sampling Region	Lab ID	Deploy	Retrieve	Deployment Time (Days)	t-PBDE (pg/L)
SNOH02.9	Langus	Lower Snohomish Mainstem	1909042-1	8/22/2019	9/19/2019	28.0	27.15
SNOH03.9	Deadwater	Lower Snohomish Mainstem	1909042-7	8/22/2019	9/19/2019	28.1	25.46
SNOH05.0	Snohomish RM 5.0	Upper Snohomish Mainstem	1909042-8	8/21/2019	9/19/2019	28.9	11.02
SNOH07.0	Rotary Park	Upper Snohomish Mainstem	1909042-9	8/21/2019	9/19/2019	29.0	14.24
SNOH12.0	City of Snohomish Downstream	Upper Snohomish Mainstem	1909042-10	8/20/2019	9/17/2019	28.0	18.86
SNOH13.0	City of Snohomish Upstream	Upper Snohomish Mainstem	1909042-10	8/20/2019	9/17/2019	28.0	15.70
SNOH20.0	Lord Hill	Upper Snohomish Mainstem	1909042-12	8/20/2019	9/17/2019	28.0	5.53
SS0.0	Steamboat Mouth	Sloughs	1909042-2	8/21/2019	9/18/2019	28.0	7.30
SS01.7	Steamboat Upper	Sloughs	1909042-4	8/21/2019	9/18/2019	27.9	7.25
ES0.0	Ebey Slough Mouth	Sloughs	1909042-3	8/21/2019	9/18/2019	28.1	3.66
ES04.3	Ebey Slough RM 4.3	Sloughs	1909042-5	8/21/2019	9/18/2019	28.0	7.30
ES06.2	Ebey @ HW2 bridge	Sloughs	1909042-6	8/21/2019	9/18/2019	27.9	10.42
SKY24.5	Monroe Downstream	Skykomish River	1909042-13	8/19/2019	9/16/2019	28.0	78.74
SKY40.5	Gold Bar	Skykomish River	1909042-15	8/20/2019	9/17/2019	28.0	2.34
SNOQ10.5	Duvall	Snoqualmie River	1909042-16	8/19/2019	9/16/2019	28.0	4.16
SNOQ23.0	Carnation Downstream	Snoqualmie River	1909042-17	8/19/2019	9/16/2019	28.0	14.17
SNOQ40.7	PSE	Snoqualmie River	1909042-18	8/19/2019	9/16/2019	28.0	6.62
SNOQ47.8	Little Si	Snoqualmie River	1909042-19	8/19/2019	9/16/2019	28.0	15.35
—	Day Zero Blank	—	1909042-23	—	—	—	4.80
—	Field Blank (SNOQ)	—	1909042-20	—	—	—	4.88
—	Field Blank (SNOH)	—	1909042-22	—	—	—	5.28
—	Field Blank (SKY)	—	1909042-21	—	—	—	5.12

2020 Low Flow

Site ID	Site Name	Sampling Region	Lab ID	Deploy	Retrieve	Deployment Time (Days)	t-PBDE (pg/L)
SNOH01.8	Dagmar	Lower Snohomish Mainstem	2010022-10	9/8/2020	10/5/2020	26.9	14.42
SNOH02.9	Langus	Lower Snohomish Mainstem	2010022-11	9/8/2020	10/5/2020	26.9	13.15
SKY24.5	Monroe Downstream	Skykomish River	2010022-8	9/8/2020	10/5/2020	26.9	36.16
SKY34.5	Sultan Downstream	Skykomish River	2010022-9	9/8/2020	10/5/2020	26.9	7.87
SNOQ47.8	Little Si	Snoqualmie River	2010022-7	9/8/2020	10/5/2020	27.0	1.16
—	Day Zero Blank	—	2010022-13	—	—	—	1.49
—	Field Blank	—	2010022-12	—	—	—	2.66

2021 Low Flow

Site ID	Site Name	Sampling Region	Lab ID	Deploy	Retrieve	Deployment Time (Days)	t-PBDE (pg/L)
SNOH01.8	Dagmar	Lower Snohomish Mainstem	2110068-1	9/13/2021	10/11/2021	28.1	19.12
SNOH02.9	Langus	Lower Snohomish Mainstem	2110068-3	9/13/2021	10/11/2021	28.1	23.55
SNOH02.9-dup	Langus	Lower Snohomish Mainstem	2110068-4	9/13/2021	10/11/2021	28.1	23.73
SNOH03.9	Deadwater	Lower Snohomish Mainstem	2110068-5	9/14/2021	10/14/2021	30.0	23.01
SNOH07.0	Rotary Park	Upper Snohomish Mainstem	2110068-6	9/14/2021	10/13/2021	29.2	14.24
SS0.0	Steamboat Mouth	Sloughs	2110068-8	9/14/2021	10/13/2021	28.7	7.93
SS1.7	Steamboat Upper	Sloughs	2110068-9	9/14/2021	10/13/2021	28.8	28.12
ES5.7	Ebey Slough RM 5.7	Sloughs	2110068-7	9/16/2021	10/13/2021	27.0	7.99
DITCH 1	Drainage Ditch	Drainage Ditch	2110068-16	9/13/2021	10/11/2021	28.1	5.49
UNION M1	Union Slough 1	Sloughs	2110068-17	9/16/2021	10/14/2021	27.7	12.92
UNION M2	Union Slough 2	Sloughs	2110068-18	9/17/2021	10/14/2021	27.1	16.94
SKY24.5	Monroe Downstream	Skykomish River	2110068-10	9/15/2021	10/12/2021	26.8	56.70
SKY25.0	Monroe Upstream	Skykomish River	2110068-11	9/15/2021	10/12/2021	26.8	16.25

Site ID	Site Name	Sampling Region	Lab ID	Deploy	Retrieve	Deployment Time (Days)	t-PBDE (pg/L)
SKY34.5	Sultan Downstream	Skykomish River	2110068-12	9/16/2021	10/12/2021	26.0	25.13
SKY35.0	Sultan Upstream	Skykomish River	2110068-13	9/16/2021	10/12/2021	26.0	6.75
SNOQ23.0	Carnation Downstream	Snoqualmie River	2110068-14	9/17/2021	10/12/2021	25.2	3.52
SNOQ23.5	Carnation Upstream	Snoqualmie River	2110068-15	9/17/2021	10/12/2021	25.2	2.54
—	Day Zero Blank	—	2110068-20	—	—	—	3.01
—	Field Blank	—	2110068-19	—	—	—	4.05

2022 Low Flow

Site ID	Site Name	Sampling Region	Lab ID	Deploy	Retrieve	Deployment Time (Days)	t-PBDE (pg/L)
SNOH0.0	Snohomish Mouth	Lower Snoh. Mainstem	2209077-13	8/9/2022	9/14/2022	36.0	23.70
SNOH01.8	Dagmar	Lower Snoh. Mainstem	2209077-14	8/9/2022	9/13/2022	35.1	11.44
SNOH02.5	Snohomish Outfall	Lower Snoh. Mainstem	2209077-15	8/9/2022	9/22/2022	43.9	11.34
SNOH02.9	Langus	Lower Snoh. Mainstem	2209077-16	8/9/2022	9/13/2022	35.2	11.27
SNOH05.0	Snohomish RM 5.0	Upper Snoh. Mainstem	2209077-19	8/9/2022	9/14/2022	36.0	6.43
SNOH07.0	Rotary Park	Upper Snoh. Mainstem	2209077-20	8/9/2022	9/14/2022	36.1	13.74
SNOH13.0	City of Snohomish Upstream	Upper Snoh. Mainstem	2209077-21	8/9/2022	9/14/2022	36.0	9.54
SKY24.5	Monroe Downstream	Skykomish River	2209077-22	8/8/2022	9/13/2022	36.0	87.49
SKY25.0	Monroe Upstream	Skykomish River	2209077-23	8/8/2022	9/13/2022	36.0	5.18
SKY35.0	Sultan Upstream	Skykomish River	2209077-24	8/8/2022	9/13/2022	36.0	2.25
—	Day Zero Blank	—	2209077-27	—	—	—	1.47
—	Field Blank	—	2209077-25	—	—	—	1.08
—	Field Blank	—	2209077-26	—	—	—	4.26

2021 High Flow

Site ID	Site Name	Sampling Region	Lab ID	Deploy	Retrieve	Deployment Time (Days)	t-PBDE (pg/L)
SNOH01.8	Dagmar	Lower Snoh. Mainstem	2104049-34	3/29/2021	4/27/2021	28.9	56.75
SNOH01.8-dup	Dagmar	Lower Snoh. Mainstem	2104049-35	3/29/2021	4/27/2021	28.9	56.77
SNOH02.9	Langus	Lower Snoh. Mainstem	2104049-30	3/29/2021	4/27/2021	28.7	50.60
SNOH03.9	Deadwater	Lower Snoh. Mainstem	2104049-31	3/29/2021	4/27/2021	28.9	41.24
SNOH7.0	Rotary Park	Upper Snoh. Mainstem	2104049-32	3/29/2021	4/27/2021	28.9	7.77
SNOH12.0	City of Snohomish Downstream	Upper Snoh. Mainstem	2104049-33	3/29/2021	4/27/2021	28.9	11.64
SS0.0	Steamboat Mouth	Sloughs	2104049-39	3/30/2021	4/28/2021	29.0	36.29
SS1.7	Steamboat Upper	Sloughs	2104049-40	3/30/2021	4/28/2021	29.0	17.19
ES0.0	Ebey Slough Mouth	Sloughs	2104049-36	3/29/2021	4/27/2021	28.9	11.03
ES1.5	Qwuloolt	Sloughs	2104049-37	3/29/2021	4/27/2021	28.9	13.12
ES5.7	Ebey Slough RM 5.7	Sloughs	2104049-38	3/30/2021	4/28/2021	29.0	10.20
SKY23.7	Monroe Downstream 2	Skykomish River	2104049-29	3/30/2021	4/26/2021	27.0	9.78
SKY24.5	Monroe Downstream	Skykomish River	2104049-28	3/30/2021	4/26/2021	27.0	8.80
SKY25.0	Monroe Upstream	Skykomish River	2104049-27	3/30/2021	4/26/2021	27.0	3.44
SKY34.5	Sultan Downstream	Skykomish River	2104049-26	3/31/2021	4/26/2021	26.2	12.99
SKY40.5	Gold Bar	Skykomish River	2104049-25	3/31/2021	4/26/2021	26.3	28.22
SNOQ10.5	Duvall	Snoqualmie River	2104049-24	3/31/2021	4/26/2021	26.0	2.96
SNOQ23.0	Carnation Downstream	Snoqualmie River	2104049-23	3/31/2021	4/26/2021	25.9	6.73
SNOQ40.7	PSE	Snoqualmie River	2104049-22	3/31/2021	4/26/2021	25.8	7.62
SNOQ47.8	Little Si	Snoqualmie River	2104049-21	3/31/2021	4/26/2021	25.8	5.78
—	Day Zero Blank	—	2104049-44	—	—	—	3.63
—	Field Blank	—	2104049-41	—	—	—	0.83
—	Field Blank	—	2104049-42	—	—	—	1.98

2022 High Flow

Site ID	Site Name	Sampling Region	Lab ID	Deploy	Retrieve	Deployment Time (Days)	t-PBDE (pg/L)
SNOH0.0	Snohomish Mouth	Lower Snoh. Mainstem	2205059-11	4/13/2022	5/11/2022	27.9	25.23
SNOH01.8 LB	Dagmar Left Bank	Lower Snoh. Mainstem	2205059-12	4/13/2022	5/11/2022	27.9	28.88
SNOH01.8	Dagmar	Lower Snoh. Mainstem	2205059-13	4/13/2022	5/11/2022	27.9	42.52
SNOH02.5	Snohomish Outfall	Lower Snoh. Mainstem	2205059-14	4/14/2022	5/12/2022	28.0	55.30
SNOH02.9 B	Langus Bottom	Lower Snoh. Mainstem	2205059-16	4/13/2022	5/12/2022	28.8	25.18
SNOH02.9	Langus	Lower Snoh. Mainstem	2205059-15	4/13/2022	5/11/2022	27.9	36.34
SNOH03.9	Deadwater	Lower Snoh. Mainstem	2205059-17	4/14/2022	5/11/2022	27.2	49.83
SNOH03.9-dup	Deadwater	Lower Snoh. Mainstem	2205059-18	4/14/2022	5/11/2022	27.2	47.13
SNOH05.0	Snohomish RM 5.0	Upper Snoh. Mainstem	2205059-19	4/14/2022	5/12/2022	28.0	28.35
SNOH07.0	Rotary Park	Upper Snoh. Mainstem	2205059-20	4/14/2022	5/12/2022	28.0	15.92
SNOH13.0	City of Snohomish Upstream	Upper Snoh. Mainstem	2205059-21	4/14/2022	5/12/2022	28.0	4.23
—	Day Zero Blank	—	2205059-24	—	—	—	2.69
—	field Blank	—	2205059-22	—	—	—	8.76
—	Field Blank	—	2205059-23	—	—	—	0.94

Table A10. Summary of PBDE homolog relative concentrations (%) for Semipermeable Membrane Device (SPMD) samples categorized by waterbody, watershed, and river flow conditions (low and high).

Low Flow Sampling Region	Di Mean	Di Range	Tri Mean	Tri Range	Tetra Mean	Tetra Range	Penta Mean	Penta Range	Hexa Mean	Hexa Range	Hepta Mean	Hepta Range	Octa Mean	Octa Range
Lower Snohomish	1.4	0.9–1.9	5.3	3.8–5.9	44.3	41.7–49.8	39.2	32.4–41.1	7.6	5.5–8.9	0.8	ND–1.4	1.4	ND–10.7
Upper Snohomish	0.9	0.5–1.4	3.9	2.6–5.4	42.3	38.0–45.4	40.4	30.6–43.6	6.9	4.7–8.3	0.9	ND–4.4	4.7	ND–19.4
Sloughs	2.0	1.0–4.9	6.0	2.6–10.4	50.2	37.8–66.1	34.8	17.8–41.3	4.7	ND–9.4	1.6	ND–10.0	0.7	ND–7.0
Skykomish	0.8	0.2–2.2	6.1	2.5–12.9	46.8	27.1–63.6	37.4	28.4–49.6	4.3	1.4–8.4	1.9	ND–7.2	2.7	ND–26.7
Snoqualmie	1.2	0.2–3.6	3.0	0.5–6.3	40.1	14.0–75.0	36.6	20.7–50.9	4.8	ND–8.1	5.9	ND–34.3	8.2	ND–22.7
Watershed	1.2	0.2–4.9	5.1	0.5–12.9	45.2	14.0–75.0	37.6	17.8–50.9	5.7	ND–9.4	2.0	ND–34.3	3.1	ND–26.7

High Flow Sampling Region	Di Mean	Di Range	Tri Mean	Tri Range	Tetra Mean	Tetra Range	Penta Mean	Penta Range	Hexa Mean	Hexa Range	Hepta Mean	Hepta Range	Octa Mean	Octa Range
Lower Snohomish	1.1	0.9–1.6	4.3	3.5–5.0	41.3	38.4–43.2	45.8	42.8–48.6	7.3	6.6–8.3	0.3	ND–0.7	ND	ND
Upper Snohomish	0.8	0.6–1.0	2.9	1.6–4.5	41.0	35.94 6.2	48.2	45.9–50.1	6.9	ND–10.9	0.2	ND–1.2	ND	ND
Sloughs	1.3	0.8–1.8	4.4	2.2–5.8	44.9	43.1–47.2	41.5	38.2–43.7	7.7	5.1–9.7	0.3	ND–1.3	ND	ND
Skykomish	1.3	1.0–2.0	3.8	1.5–7.4	39.6	36.7–43.1	47.6	41.3–51.5	7.8	6.2–10.2	ND	ND	ND	ND
Snoqualmie	0.8	0.3–1.7	2.3	ND–5.4	40.4	32.7–47.2	49.9	38.9–59.1	6.8	3.9–11.5	ND	ND	ND	ND
Watershed	1.1	0.3–2.0	3.7	ND–7.4	41.4	32.7–47.2	46.4	38.2–59.1	7.3	ND–11.5	0.2	ND–1.3	ND	ND

Table A11. Results for PBDEs in benthic and suspended sediments

Sediment t-PBDE are the sum of di- through deca-BDEs. NA = not analyzed

Site ID	Site Name	Sampling Region	Collection Date	Lab ID	Type	t-PBDE (pg/g)	$\delta^{15}N$ (‰)
SNOH01.8	Dagmar	Lower Snohomish Mainstem	9/19/2019	1909042-46	Benthic	1357.48	3.4
SNOH01.8-dup	Dagmar	Lower Snohomish Mainstem	9/19/2019	1909042-66	Benthic	1435.63	3.6
SNOH02.9	Langus	Lower Snohomish Mainstem	9/19/2019	1909042-47	Benthic	130.23	4.0
SNOH03.9	Deadwater	Lower Snohomish Mainstem	9/19/2019	1909042-53	Benthic	504.84	1.8
SNOH12.0	City of Snohomish Downstream	Upper Snohomish Mainstem	9/17/2019	1909042-56	Benthic	513.46	2.7
SNOH13.0	City of Snohomish Downstream	Upper Snohomish Mainstem	9/17/2019	1909042-57	Benthic	3812.85	3.2
SS0.0	Steamboat Mouth	Sloughs	9/18/2019	1909042-48	Benthic	343.86	2.6
SS01.7	Steamboat Upper	Sloughs	9/18/2019	1909042-50	Benthic	553.06	4.0
ES0.0	Ebey Slough Mouth	Sloughs	9/18/2019	1909042-49	Benthic	346.75	4.6
ES04.3	Ebey Slough RM4.3	Sloughs	9/18/2019	1909042-51	Benthic	586.42	3.0
UNION M1	Union Slough 1	Sloughs	10/1/2021	2110058-16	Benthic	772.73	NA
UNION M2	Union Slough 2	Sloughs	10/1/2021	2110058-17	Benthic	688.95	NA
DITCH 1	Drainage Ditch	Drainage Ditch	10/11/2021	2110058-21	Benthic	569.05	NA
SNOH01.8	Dagmar	Lower Snohomish Mainstem	10/15/2021	2110068-31	suspended	1869.48	NA

Site ID	Site Name	Sampling Region	Collection Date	Lab ID	Type	t-PBDE (pg/g)	$\delta^{15}N$ (‰)
SNOH01.8-dup	Dagmar	Lower Snohomish Mainstem	10/15/2021	2110068-30	suspended	4034.74	NA
SNOH02.9	Langus	Lower Snohomish Mainstem	10/15/2021	2110068-32	suspended	1323.67	NA
SNOH03.9	Deadwater	Lower Snohomish Mainstem	10/15/2021	2110068-33	suspended	1932.65	NA
SNOH07.0	Rotary Park	Upper Snohomish Mainstem	10/15/2021	2110068-34	suspended	1280.83	NA
SNOH01.8	Dagmar	Lower Snohomish Mainstem	5/12/2022	2209051-3	suspended	1659.63	NA
SNOH02.9	Langus	Lower Snohomish Mainstem	5/12/2022	2209051-4	suspended	333.70	NA
SNOH03.9	Deadwater	Lower Snohomish Mainstem	5/12/2022	2209051-5	suspended	1069.37	NA
SNOH07.0	Rotary Park	Upper Snohomish Mainstem	5/12/2022	2209051-2	suspended	507.77	NA
SNOH07.0-dup	Rotary Park	Upper Snohomish Mainstem	5/12/2022	2209051-1	suspended	916.69	NA
SNOH13.0	City of Snohomish Upstream	Upper Snoh. Mainstem	5/12/2022	2209051-6	suspended	230.81	NA
SNOH01.8	Dagmar	Lower Snoh. Mainstem	8/16/2022	2209051-7	suspended	870.84	NA
SNOH07.0	Rotary Park	Upper Snoh. Mainstem	8/16/2022	2209051-8	suspended	1179.82	NA
SNOH01.8	Dagmar	Lower Snoh. Mainstem	9/19/2022	2209051-11	suspended	932.40	NA
SNOH07.0	Rotary Park	Upper Snoh. Mainstem	9/19/2022	2209051-9	suspended	1546.77	NA

Table A12. Results for PBDEs, lipids, and nitrogen stable isotopes ($\delta^{15}N$) in biofilms

Biofilm t-PBDE are the sum of di- through deca-BDEs. Tissue samples are a mix of biofilms and invertebrates. NA = not analyzed

Site ID	Site Name	Sampling Region	Lab ID	Collection Date	Type	t-PBDE (pg/g)	% lipid	$\delta^{15}N$ (‰)
SNOH01.8	Dagmar	Lower Snohomish Mainstem	1909042-68	9/19/2019	Biofilm	1195.65	0.16	7.0
SNOH01.8	Dagmar	Lower Snohomish Mainstem	1909042-69	9/19/2019	Tissue	1341.99	0.22	7.1
SNOH02.9	Langus	Lower Snohomish Mainstem	1909042-71	9/19/2019	Biofilm	678.86	0.22	7.7
SNOH03.9	Deadwater	Lower Snohomish Mainstem	1909042-72	9/19/2019	Tissue	1705.33	0.18	7.6
SNOH20.0	Lord Hill	Upper Snohomish Mainstem	1909042-58	9/17/2019	Biofilm	309.04	0.091	5.0
SKY24.5	Monroe Downstream	Skykomish River	1909042-59	9/16/2019	Biofilm	178.89	0.18	5.7
SKY24.5-dup	Monroe Downstream	Skykomish River	1909042-65	9/16/2019	Biofilm	119.87	0.21	NA
SKY34.5	Sultan Downstream	Skykomish River	1909042-60	9/17/2019	Biofilm	642.89	0.08	2.1
SKY40.5	Goldbar	Skykomish River	1909042-61	9/17/2019	Biofilm	125.39	0.098	1.1
SNOQ23.0	Carnation Downstream	Snoqualmie River	1909042-62	9/16/2019	Biofilm	11.49	0.29	2.9
SNOQ40.7	PSE	Snoqualmie River	1909042-63	9/16/2019	Biofilm	338.82	0.12	2.5
SNOQ47.8	Little Si	Snoqualmie River	1909042-64	9/16/2019	Biofilm	14.72	0.21	0.3

Table A13. Results for PBDEs, % lipid and nitrogen stable isotopes in invertebrate tissues

Invertebrate t-PBDE are the sum of di- through deca-BDEs.

Site ID	Site Name	Sampling Region	Lab ID	Collection Date	t-PBDE (pg/g)	% Lipid	$\delta^{15}N$ (‰)
SNOH01.8	Dagmar	Lower Snohomish Mainstem	1909042-67	9/19/2019	1781.31	1.23	7.8
SNOH02.9	Langus	Lower Snohomish Mainstem	1909042-70	9/19/2019	1560.15	1.32	8.9
SNOH02.9	Langus	Lower Snohomish Mainstem	2108068-03	4/27/2021	9598.97	1.66	5.0
ES0.0	Ebey Slough Mouth	Sloughs	2108068-05	4/27/2021	546.58	0.44	6.0
SNOH02.9	Langus	Lower Snohomish Mainstem	2108068-04	5/12/2021	6480.04	1.30	4.9
ES0.0	Ebey Slough Mouth	Sloughs	2108068-06	5/12/2021	574.19	0.53	8.1
SNOH02.9	Langus	Lower Snohomish Mainstem	2108068-08	6/25/2021	2496.58	0.79	6.2
SNOH01.8	Dagmar	Lower Snohomish Mainstem	2108068-01	8/17/2021	1420.73	1.33	7.7
SNOH01.8-dup	Dagmar	Lower Snohomish Mainstem	2108068-09	8/17/2021	2006.06	1.18	7.6
ES1.5	Qwuloolt	Sloughs	2108068-02	8/17/2021	291.43	0.90	8.0
SN0H02.9	Langus	Lower Snohomish Mainstem	2209026-1	2/3/2022	4247.89	1.34	7.8
SN0H02.9	Langus	Lower Snohomish Mainstem	2209026-2	3/9/2022	10809.88	2.61	7.0
SN0H02.9	Langus	Lower Snohomish Mainstem	2209026-3	4/14/2022	6722.72	1.36	8.9
SN0H02.9	Langus	Lower Snohomish Mainstem	2209026-4	5/12/2022	6116.55	2.52	3.5
SN0H02.9	Langus	Lower Snohomish Mainstem	2209026-5	6/16/2022	5190.96	2.75	3.6
SN0H02.9-dup	Langus	Lower Snohomish Mainstem	2209026-6	6/16/2022	5836.58	5.8	3.4
SNOH01.8	Dagmar	Lower Snohomish Mainstem	2209026-7	8/16/2022	4215.25	1.19	7.1
SNOH02.9	Langus	Lower Snohomish Mainstem	2209026-8	9/13/2022	827.28	2.1	7.6

Table A14. Snohomish River Combined Sewer Overflow (CSO) discharge records from 2019 to 2022

Discharge record provided by Everett Public Works as part of the Combined Serwer Overflow Annual Reports submitted to Ecology. MG = million gallons

CSO Outfall	2019 Discharge Volume (MG)	2019 Discharge Duration (hours)	2019 Number of Events	2020 Discharge Volume (MG)	2020 Discharge Duration (hours)	2020 Number of Events	2021 Discharge Volume (MG)	2021 Discharge Duration (hours)	2021 Number of Events	2022 Discharge Volume (MG)	2022 Discharge Duration (hours)	2022 Number of Events
SR01	0.007	0.2	1	0	0	0	0	0	0	0	0	0
SR02	0.2	0.6	1	0	0	0	0	0	0	0	0	0
SR03	0.9	1.5	2	0.43	2	1	0.022	0.3	1	0.35	0.8	1
SR04	0.05	0.5	2	0.77	2.3	2	0.38	1.2	2	0	0	0
SR07	1.1	2.4	7	2	2.8	2	3.1	4.4	6	0.39	0.8	2
SR08	0.8	0.6	1	2.5	2.7	1	4.7	9.4	4	0.9	0.76	1

Table A15. Snohomish River Combined Sewer Overflow (CSO) discharge records summary for study period (2019-2022)

Discharge record provided by Everett Public Works as part of the Combined Serwer Overflow Annual Reports submitted to Ecology. MG = million gallons

CSO Outfall	Total Discharge Volume (MG)	Total Discharge Duration (hours)	Total Number of Events
SR01	0.007	0.2	1
SR02	0.2	0.6	1
SR03	1.702	4.6	5
SR04	1.2	4	6
SR07	6.59	10.4	17
SR08	8.9	13.46	7