Brominated Flame Retardants, Alkylphenolic Compounds, and Hexabromocyclododecane in Freshwater Fish of Washington State Rivers and Lakes

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Brominated Flame Retardants, Alkylphenolic Compounds, and Hexabromocyclododecane in Freshwater Fish of Washington State Rivers and Lakes

by

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Water Resource Inventory Area (WRIA) and 8-digit Hydrologic Unit Code (HUC) numbers for the study area:

Study Location	WRIA	HUC
Banks Lake	42	17020014
Clear Lake	43	17020013
Kitsap Lake	15	17110019
Lake Whatcom	1	17110004
Mayfield Lake	26	17080005
Pierre Lake	60	17020002
Sawyer Lake	9	17110013
Lake Stevens	7	17110011
Mid-Columbia River	31	17070101
Snake River	35	17060107
Snohomish River	7	17110011

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Abstract

The Washington State Department of Ecology (Ecology) conducted a study in 2014 to evaluate current levels of emerging contaminants and persistent, bioaccumulative, toxic chemicals (PBTs) in freshwater fish tissue in Washington State. Ecology collected a total of 44 fish tissue samples from 11 waterbodies located throughout the state, across a range of land use types and contamination potential. The fish tissue samples were analyzed for chemicals that are either on the state's current PBT List or are emerging contaminants that require more information: polybrominated diphenyl ethers (PBDEs), alternative brominated flame retardants, alkylphenolic compounds, and hexabromocyclododecane (HBCD).

In general, detected concentrations of analytes were in the order of PBDEs > alkylphenolic compounds > HBCD > alternative brominated flame retardants. PBDEs were detected at the highest frequency (100% of samples) and at the highest concentrations. Total PBDEs (T-PBDEs) ranged from 332 – 46,000 ng/Kg ww. The alkylphenolic compounds 4n-octylphenol and mono- and di- nonylphenol ethoxylates were present in fewer samples (48%) than PBDEs, but at concentrations within the range of T-PBDE concentrations (445 – 4,080 ng/Kg ww). Alpha-HBCD was detected in 27% of samples with concentrations ranging from 115 – 362 ng/Kg ww.

Eighty-nine percent of samples contained one or more of the alternative flame retardants analyzed, at low levels. Decabromodiphenylethane (DBDPE) was present in the highest concentrations of the four non-BDE flame retardant analytes, ranging from 14.1 – 304 ng/Kg ww, followed by 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE) which ranged from 0.682 – 44.5 ng/Kg ww. These results suggest that replacement chemicals for PBDEs may persist in the environment of Washington State. Hexabromobenzene and pentabromoethylbenzene were detected at concentrations ranging from 0.100 – 2.20 ng/Kg ww.

The results of this study should be used to support prioritization of chemicals to be addressed by Ecology through chemical actions plans and other efforts to reduce toxics in the state.

Acknowledgements

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Introduction

Background

The Washington State Department of Ecology (Ecology), in collaboration with other state agencies, develops chemical action plans (CAPs) to identify, characterize, and evaluate uses and releases of persistent, bioaccumulative, and toxic chemicals (PBTs) in the state. The agencies use CAPs to compile information and recommend actions to protect human health and the environment. CAPs are developed for one chemical or chemical group at a time.

The PBT Rule laid out a process to select which PBTs are given priority for CAP development (WAC 173-333-410). In 2007, Ecology published a *Multiyear PBT Chemical Action Plan Schedule* that outlined priority PBTs and set forth a schedule in which Ecology will address the chemicals (Gallagher, 2007). Ecology periodically reviews and, as appropriate, updates the multiyear schedule. The PBT List, which the multiyear schedule draws from, will be re-prioritized in the future. It may be expanded to include chemicals that exhibit one or more of the PBT characteristics (i.e., very persistent or very bioaccumulative) or are released into the environment on a regular basis, rendering them *pseudo-persistent*.

To support reprioritization of PBTs and to know whether new chemicals should be added to the list, data are needed on the occurrence and levels of these chemicals present in Washington's environment. This study provides data on select emerging contaminants and PBTs in freshwater fish of Washington State. Ecology collected freshwater fish tissue samples from 11 waterbodies throughout the state for analysis of brominated flame retardants (BFRs), alkylphenolic compounds, and hexabromocyclododecane (HBCD).

Brominated Flame Retardants

Brominated flame retardants are a broad class of chemicals used in consumer products, such as furniture and electronics, to prevent or slow the spread of fire. Polybrominated diphenyl ethers (PBDEs) are a group of flame retardants belonging to this class that are identified as PBTs and are widespread in the environment. Ecology developed a CAP for PBDEs in 2006, after growing concern that the chemicals were dramatically increasing in people and in the environment (Ecology et al., 2006). In 2008, a Washington State law prohibited the use of two commercial formulations of PBDEs – Penta-BDE and Octa-BDE – in consumer products, and restricted the use of Deca-BDE in mattresses, residential upholstered furniture, computers, and televisions (RCW 70.76). The state law required Ecology to issue a finding that safer alternatives to Deca-BDE were available before the restrictions took effect.

Chemical manufacturers stopped production of penta-BDE and octa-BDE in the mid-2000s, and phased out most uses of deca-BDE by the end of 2012. As commercial uses of PBDEs were phased out, manufacturers started using alternative flame retardants as replacements to meet flammability standards. Some of the replacement chemicals for PBDEs are also brominated, and little is known about their toxicity and fate in the environment. Modeling studies suggest that some of the alternative BFRs have similar hazard profiles to PBDEs and may persist in the environment (EPA, 2014a; Kuramochi et al., 2014). The U.S. Environmental Protection Agency (EPA) has completed alternatives assessments for flame retardants in several different product applications and safer alternatives have been identified (https://www.epa.gov/saferchoice/design-environment-alternatives-assessments).

Additive flame retardants are not chemically bound to material and leach out of products over time, accumulating in indoor dust. These chemicals are released to the environment when textiles with indoor dust on them are washed and traces are delivered to wastewater treatment plants (WWTPs), ultimately discharging to river systems through WWTP effluent (Schreder and La Guardia, 2014). Atmospheric deposition and surface runoff have also been identified as important pathways for PBDE contamination in Washington State aquatic systems (Norton et al., 2011; PNNL, 2010; Herrera, 2011).

BFRs analyzed in this study include PBDEs and the following alternative flame retardants: 1,2-Bis(2,4,6-tribromophenoxy)ethane (BTBPE), decabromodiphenylethane (DBDPE), hexabromobenzene (HBBz), and pentabromoethylbenzene (PBEB).

Alkylphenolic Compounds

Alkylphenolic compounds are a large class of chemicals that consist of a carbon chain and attached phenol ring. They include nonylphenol (NP) and octylphenol (OP), which are used to produce alkylphenol ethoxylates. They are commonly used in the production and manufacture of detergents and other cleaning products, emulsifiers, personal care products, and are widely applied in various industrial processes. NP and nonylphenol ethoxylates (NPEs) make up 80 –85% of the volume of alkylphenolic compounds on the market (EPA, 2010).

Alkylphenolic compounds are not currently on the agency PBT List, but are contaminants of emerging concern, and NP and OP are considered a chemical of high concern to children by Ecology (WAC 173-334-110). The Washington State Children’s Safe Products Act required manufacturers to report to Ecology if NP or OP are used in children’s products. Manufacturer reporting shows that NP is frequently used in the making of children’s products, particularly as a contaminant in footwear or as a solvent or stabilizer in toys. The EPA developed a chemical action plan for NP and NPEs (EPA, 2010a) and launched the Safer Detergents Stewardship Initiative in 2006 to identify safer alternative to NPEs and encourage voluntary phase-out of the chemicals.

Alkylphenolic compounds enter into the environment primarily through wastewater treatment plant discharges, but stormwater, septic systems, atmospheric deposition, and pesticide applications may be important pathways as well (reviewed by Klosterhaus et al., 2012a). In the environment, higher chain NPEs break down into degradation products that include NP, nonylphenol monoethoxylates (NP1EO), and nonylphenol diethoxylates (NP2EO), which are persistent, low-to-moderately bioaccumulative, and highly toxic to aquatic organisms (EPA, 2014b). This study included the following alkylphenolic compound analytes: 4n-NP, NP1EO, NP2EO, and 4n-OP.

Hexabromocyclododecane

Hexabromocyclododecane (HBCD) refers to a technical mixture composed primarily of alpha, beta, and gamma diastereoisomers. It is used as a flame retardant in extruded (XPS) and expanded (EPS) polystyrene for building insulation, as well as in furniture textiles, automotive upholstery, and other consumer products such as electronics. HBCD exhibits high aquatic toxicity and is a human health concern for reproductive, developmental, and neurological effects, based on animal studies (EPA, 2010b).

Ecology included HBCD on the agency PBT List but has not scheduled it for development of a CAP. EPA released an action plan summary for HBCD in 2010 (EPA, 2010b) and has recently issued an alternatives assessment for its use in XPS and EPS insulation (EPA, 2014c).

HBCD can be transported long distances and has been found in many different environmental media throughout the world (Covaci et al., 2006). Sources to the environment generally include diffuse particulate releases to soil during construction and demolition of XPS- or EPS-insulated buildings and through the use or disposal of products containing HBCD (EPA, 2010b). Particulates containing HBCD are transferred to air or stormwater runoff and through wastewater treatment plant effluent and landfill emissions (EPA, 2010b). Alpha-, beta-, and gamma-HBCD isomers were analyzed in this study.

Study Design

Ecology collected fish samples from 11 waterbodies throughout the state in the fall of 2014. Samples were analyzed for BFRs, alkylphenolic compounds, and HBCD. The study locations are displayed in Figure 1 and described in Table 1. Waterbodies were selected for this study to be spatially distributed throughout the state and to represent a variety of land use types with varying degrees of contamination potential. Study locations also covered a range of surface elevations and waterbody/watershed sizes.

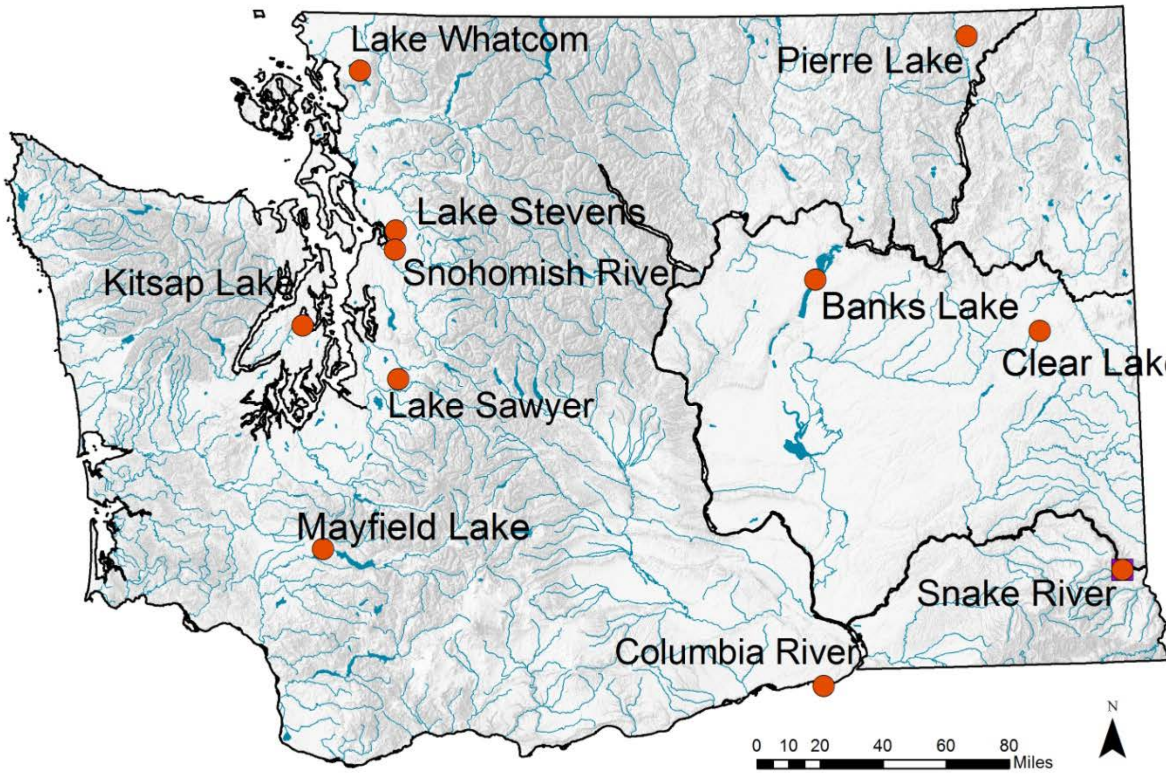

Figure 1. 2014 Fish Collection Locations.

Three urban waterbodies – two lakes and one river – were chosen to represent waterbodies with more significant contaminant sources such as urban stormwater and wastewater treatment plant (WWTP) effluent. The urban waterbodies included Lake Stevens and Kitsap Lake (urban stormwater inputs) and the Snohomish River (stormwater and WWTP discharge). The Snake River and Columbia River sites have moderate contamination potential due to wastewater treatment plant effluent; however, these sites drain large areas and may have a more diluted signal than Snohomish River. Other sites chosen to represent moderate contamination potential include Clear Lake, Lake Whatcom, and Lake Sawyer. Land use surrounding these sites include a mix of undeveloped (brush steppe or forested) land and residential development.

Two waterbodies were chosen to reflect primarily forested watersheds – Mayfield Lake in western Washington and Pierre Lake in eastern Washington. The land surrounding these two lakes is relatively undisturbed forestland. However, Mayfield Lake does receive municipal wastewater treatment plant effluent from the city of Mossyrock.

One to three different species of fish were collected from each study location. Field staff aimed to collect sufficient numbers of fish at each waterbody for analysis of two composite samples of a predator and two composite samples of a bottom feeder species. Composites of skin-on fillet tissue from the predator species were analyzed to provide data applicable to human health concerns and for comparability among studies (e.g., Johnson et al., 2006). Bottom feeder species were analyzed as composites of whole body tissue to obtain data on ecological exposure, and also for comparability to previous studies (e.g., Johnson and Friese, 2012). Species collected from each lake for analysis of fillet and whole body composites are listed in Table 1.

Collection goals were met with the following exceptions. No bottom feeder species were encountered at Pierre Lake or Snohomish River. Efforts to collect a predatory species were also unsuccessful at Snohomish River, and therefore mountain whitefish and peamouth were collected from Snohomish River instead.

Table 1. Study Locations and Fish Species Analyzed.

Study Location	Predatory Species Analyzed (fillet)	Bottom Feeder Species Analyzed (whole body)	Elevation (ft)	Lake Area (acres)	Watershed Area (sq mi)	Predominant Land Type
Lakes						
Banks Lake	SMB	BBH	1,570	27,000	---	Agricultural
Clear Lake	LMB	TCH	2,344	316	10	Brush steppe
Kitsap Lake	LMB	BBH	156	250	3	Urban
Mayfield Lake	NPM	LSS	450	2,200	1,400	Forested
Pierre Lake	SMB	---	2,000	110	27	Forested
Sawyer Lake	LMB	BBH	512	300	13	Resid./forested
Lake Stevens	LMB	BBH	210	1,000	7	Urban
Lake Whatcom	SMB	BBH	312	5,000	56	Resid./forested
Rivers						
Mid-Columbia River	SMB	LSS	343	---	2,214,000	Agricultural
Snake River	LMB, NPM	LSS	760	---	107,500	Agricultural
Snohomish River	MWF, PEA	---	40	---	1720	Urban

SMB: smallmouth bass; BBH: brown bullhead; LMB: largemouth bass; TCH: tench;
NPM: northern pikeminnow; LSS: largescale sucker; MWF: mountain whitefish; PEA: peamouth.

Methods

Sample Collection and Preparation

Ecology field crews carried out fish collections using boat electro-shocking and gill-netting techniques. All field collections and sample preparations followed Ecology Standard Operating Procedures for *Collection, Processing, and Preservation of Finfish Samples* (Sandvik, 2014a) and *Resecting Finfish Whole Body, Body Parts, or Tissue Samples* (Sandvik, 2014b). Fish composite samples consisted of 3-5 fish skin-on fillets or whole bodies, depending on species, per sample.

Ancillary data were measured either in the field or laboratory and recorded. Fish lengths and weights were recorded in the field, and fish sex was determined in the laboratory during fish processing. Aging structures were sent to Washington Department of Fish and Wildlife biologists for determination of ages.

Laboratory Analysis

The compounds analyzed in this study deviated slightly from the target analyte list in the Quality Assurance (QA) Project Plan: alkylphenolic compounds were analyzed instead of chlorinated paraffins. The contract laboratory changed their reporting procedure for chlorinated paraffins during the course of this study prior to sample shipment to the laboratory. The procedure for chlorinated paraffins utilized a 3:1 signal to noise ratio for reporting, with estimated sample specific detection limits (SDL) of 5 ng/g or less. After receiving additional data from other programs, the laboratory identified a minimum concentration for positive identification for the confident identification of chlorinated paraffins. This value was above that expected of ambient fish tissue concentrations and therefore chlorinated paraffins were dropped from the analyte list. Alkylphenolic compounds were added to the analyte list because they exhibit PBT characteristics, yet data are lacking on these compounds in freshwater fish of Washington State.

BFRs, alkylphenolic compounds, HBCD, and lipids were analyzed by AXYS Analytical Services, LTD. Table 2 displays the target analyte suites for this study and Table 3 displays the methods followed for laboratory analysis. Table 4 displays the median estimated detection limits and median estimated quantitation limits achieved for all analyses.

Table 2. Analyte Suites and Individual Chemicals Analyzed.

Analyte Suite	Analyte	Acronym
Brominated Flame Retardants (BFRs)	Polybrominated diphenyl ethers [1]	PBDEs
	Pentabromoethylbenzene	PBEB
	Hexabromobenzene	HBBz
	1,2-Bis(2,4,6-tribromophenoxy)ethane	BTBPE
	Decabromodiphenylethane	DBDPE
Alkylphenolic compounds	4-Nonylphenol (branched and linear forms)	4-NP
	4-n-Octylphenol (linear isomer)	n-OP
	4-Nonylphenol monoethoxylates (branched and linear isomers)	NP1EO
	4-Nonylphenol diethoxylates (branched and linear isomers)	NP2EO
Hexabromocyclododecane (HBCD)	alpha-HBCD	a-HBCD
	beta-HBCD	b-HBCD
	gamma-HBCD	g-HBCD

[1] congeners:
'-7, -8/11, -10, -12/13, -15, -17/25, -28/33, -30, -32, -35, -37, -47, -49, -51, -66, -71, -75, -77, -79, -85, -99, -100, -105, -116, -119/120, -126, -128, -138/166, -140, -153, -154, -155, -181, -183, -190, -203, -206, -207, -208, -209

Table 3. Analytical Methods.

Analyte	Method	Reference
BFRs	HR-GC/MS	AXYS MLA-033
Alkylphenolic compounds	LC-MS/MS	AXYS MLA-080
HBCD	LC-MS/MS	AXYS MLA-070
Lipids	Gravimetric	EPA 1614/AXYS MLA-033

HR-GC/MS: high resolution gas chromatography/mass spectrometry
LC-MS/MS: liquid chromatography – mass spectrometry/mass spectrometry

Table 4. Median Estimated Detection Limits and Estimated Quantitation Limits.

Analyte Suite	Analyte	EDL	EQL
BFRs	PBDEs*	0.2 ng/Kg	5.0 ng/Kg
	BDE-209	9.0 ng/Kg	50 ng/Kg
	BTBPE	1.5 ng/Kg	---
	DBDPE	20 ng/Kg	---
	HBBz	0.1 ng/Kg	---
	PBEB	0.1 ng/Kg	---
Alkylphenolic compounds	4-NP	500 ng/Kg	---
	n-OP	500 ng/Kg	---
	NP1EO	500 ng/Kg	---
	NP2EO	500 ng/Kg	---
HBCD	a-HBCD	---	100 ng/Kg
	b-HBCD	---	100 ng/Kg
	g-HBCD	---	100 ng/Kg

*Except for BDE-209

Data Quality

The QA coordinator at Ecology's Manchester Environmental Laboratory (MEL) reviewed and verified all analytical data for this study following EPA's *National Function Guidelines for Superfund Organics Methods Data Review* (EPA, 2014d). MEL provided written case narratives to the project manager with a description of the quality of the data, including method of analysis, instrument calibration, and results of quality control (QC) tests. All QC tests outlined in the QA Project Plan were performed for the analyses. MEL also provided electronic data deliverables with final data values and qualifiers.

All samples were properly preserved and analyzed within method holding times. All analytes were within acceptance limits for the initial calibration and continuing calibration verification checks. QC tests generally met acceptance criteria outlined in the methods and QA Project Plan. The following sections describe QC test results and qualifications made to the data. Case narratives are available upon request from the project manager.

Brominated Flame Retardants

All PBDE congener concentrations below the lowest calibration standard were qualified "J" as estimates, because results were derived from responses outside the calibration range. All non-detect results were reported down to the estimated detection limit and flagged "UJ". PBDE congeners 206, 207, and 208 may be formed from degradation of BDE-209 during the analytical procedure; therefore, results for these congeners should be considered "maximum" concentrations. Results for congeners BDE- 206, 207, and 208 were qualified "J" as estimates.

Results for the alternative BFRs (BTBPE, DBDPE, HBBz, and PBEB) were calculated from a single point calibration standard concentration; therefore, data were qualified "J" as estimates. For results that did not meet isotope abundance ratio acceptance limits, but all other identification criteria were met, values were qualified "NJ", to indicate that the analyte has been tentatively identified and the associated result is an estimate. Several method blanks contained target analyte concentrations between the detection limit and quantitation limit. Results less than ten times the concentration found in associated method blanks were qualified "U" as non-detects and raised to either the quantitation limit or amount detected in the sample if higher than the quantitation limit.

Relative percent differences (RPDs) were calculated for laboratory duplicates and the associated source sample where both results were greater than five times the quantitation limit. RPDs met the measurement quality objective (MQO) of <40%, with average RPDs of 3%, 9%, and 8% in the three samples analyzed in duplicate. Laboratory control samples (LCS) had an average recovery of 98%, and all were within MQOs.

Alkylphenolic Compounds

The analytical method for alkylphenolic compounds used a single point calibration standard for calculation of results. All data for the alkylphenol analytes should be considered estimates and have been qualified "J".

Method blank results for 4-NP ranged from 9.7 – 15.6 ng/g. All 4-NP samples had concentrations less than ten times the blank contamination and were qualified "U" as a non-detect at either the quantitation limit or the amount detected in the sample if higher than the quantitation limit. Small amounts of NP1EO and NP2EO were detected in several method blanks below the reporting cut-off of 0.5 ng/g. Because concentrations below the 0.5 ng/g level are much less certain, sample results were qualified only when they were less than three times the blank concentration. Results for these samples were qualified with a "UJ" at the level of detection.

One surrogate sample recovery was slightly below the acceptance limit of 40% (13C6-NP1EO = 35%). Affected results that use the labeled compound for quantification were qualified as estimates. Duplicate sample RPDs were not calculated, as no results were greater than five times the quantitation limit. LCS recoveries were within MQOs, with an average recovery of 94%.

Hexabromocyclododecane

All HBCD data met MQOs with the following exception. The surrogate ^{13}C12-beta-HBCD was not recovered in sample 1412023-22. The associated sample result was rejected.

No other problems were encountered with the HBCD analysis. Three laboratory duplicates were analyzed by the laboratory. No HBCD analytes were detected in two of the samples. One sample contained alpha-HBCD above the quantitation limit in both the source sample and duplicate. The relative percent difference was 8%. All method blanks were free of the target analytes at or above the quantitation limit. LCS recoveries were within MQOs, with an average recovery of 94%.

Results and Discussion

A total of 44 fish tissue samples collected from 11 waterbodies throughout the state were analyzed for BFRs, alkylphenolic compounds, and HBCD during this study. In general, concentrations of analytes were in the order of total (T-) PBDEs > alkylphenolic compounds > HBCD > non-BDE flame retardants. An overview of detected concentrations is displayed in Figure 2, and the full data set is available in Appendices A and B. Data can also be obtained through Ecology's Environmental Information Management database at https://fortress.wa.gov/ecy/eimreporting/.

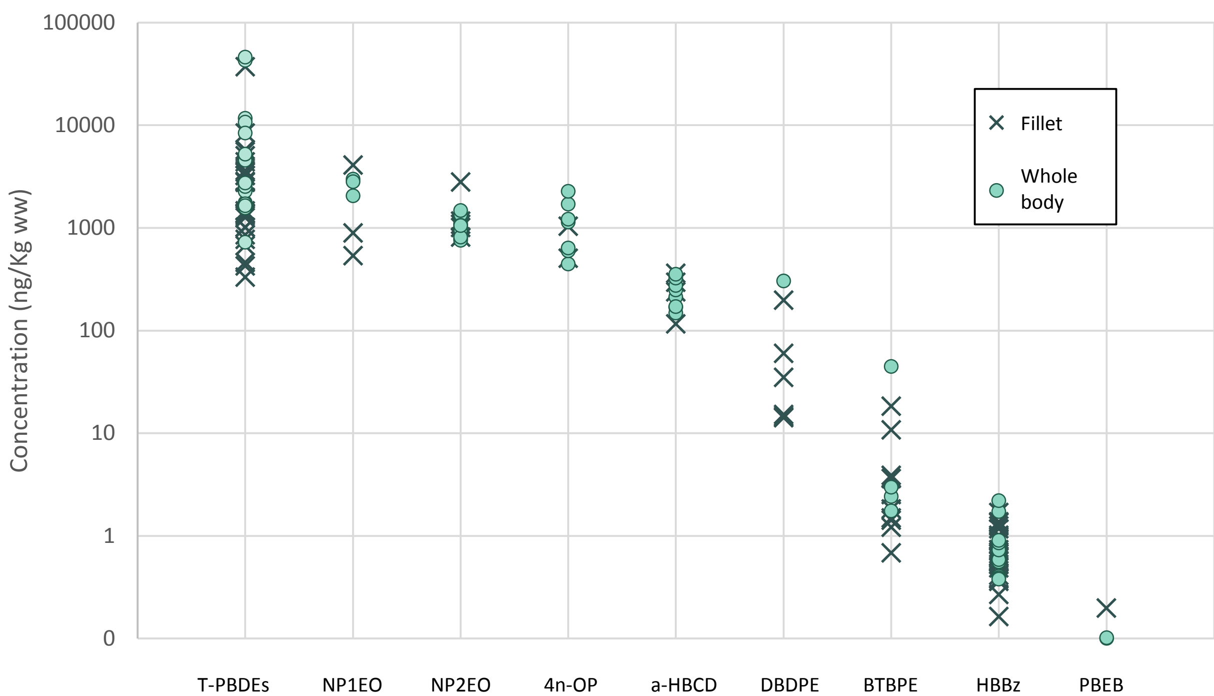

Figure 2. Concentrations of Individual Compounds Analyzed in Fish Tissue Samples Collected from 11 Washington State Rivers and Lakes in 2014.

Figure includes detected concentrations only.
Note the logarithmic axis.

Results of the individual analytes, as well as comparisons with other findings, are presented in the following sections. Summed ("T-") values and summary statistics include detected compounds only. Results qualified as estimates ("J") and tentatively identified ("NJ") were included in summed values and statistics. All results are reported on a wet weight (ww) basis, unless otherwise noted. In comparisons to other studies, concentrations were lipid-normalized and are indicated as lipid weight (lw).

Brominated Flame Retardants

PBDEs

Table 5 summarizes T-PBDE concentrations measured in fillet and whole body fish tissues sampled for this study. PBDEs were detected in all samples analyzed. Concentrations in fillet samples ranged from 332 – 37,000 ng/Kg, with a median of 1,730 ng/Kg. Whole body composites were slightly higher in T-PBDEs, ranging from 725 – 46,000 ng/Kg (median = 3,650 ng/Kg).

Table 5. Statistical Summary of PBDE Flame Retardant Results in Fish Tissue.

Analyte	Tissue Type	No. of Detections	Detection Frequency	Min (ng/Kg ww)	Max (ng/Kg ww)	Mean (ng/Kg ww)	Median (ng/Kg ww)
T-PBDEs*	Fillet	26	100%	332	37,000	3,830	1,730
T-PBDEs*	Whole body	18	100%	725	46,000	8,700	3,650

*Sum of detected PBDE congeners listed in Table 2.

Figure 3 displays the study locations ranked by mean T-PBDE concentrations. T-PBDE concentrations were highest in two Columbia River largescale sucker samples and a single Snohomish River mountain whitefish sample. T-PBDE concentrations in these three samples were in the range of 36,000 - 46,000 ng/Kg, whereas the majority of the samples fell between 1,000 - 10,000 ng/Kg. Fillet samples of predator species in Sawyer, Clear, and Pierre Lakes were lowest in T-PBDEs, all containing concentrations <1,000 ng/Kg.

The two Columbia River largescale sucker samples also had the greatest percent lipids (11.6% and 14.8%). Lipid-normalized T-PBDEs values for these two samples were at the higher end, but within the range of the rest of the data set. The Snohomish River mountain whitefish, however, was relatively low in lipids (2.49%), and the lipid-normalized T-PBDEs concentration was greater than four times the next highest value. The Snohomish River estuary has also been identified as a hotspot for T-PBDEs in juvenile salmon in comparison to other areas of Puget Sound (O'Neill et al., 2015).

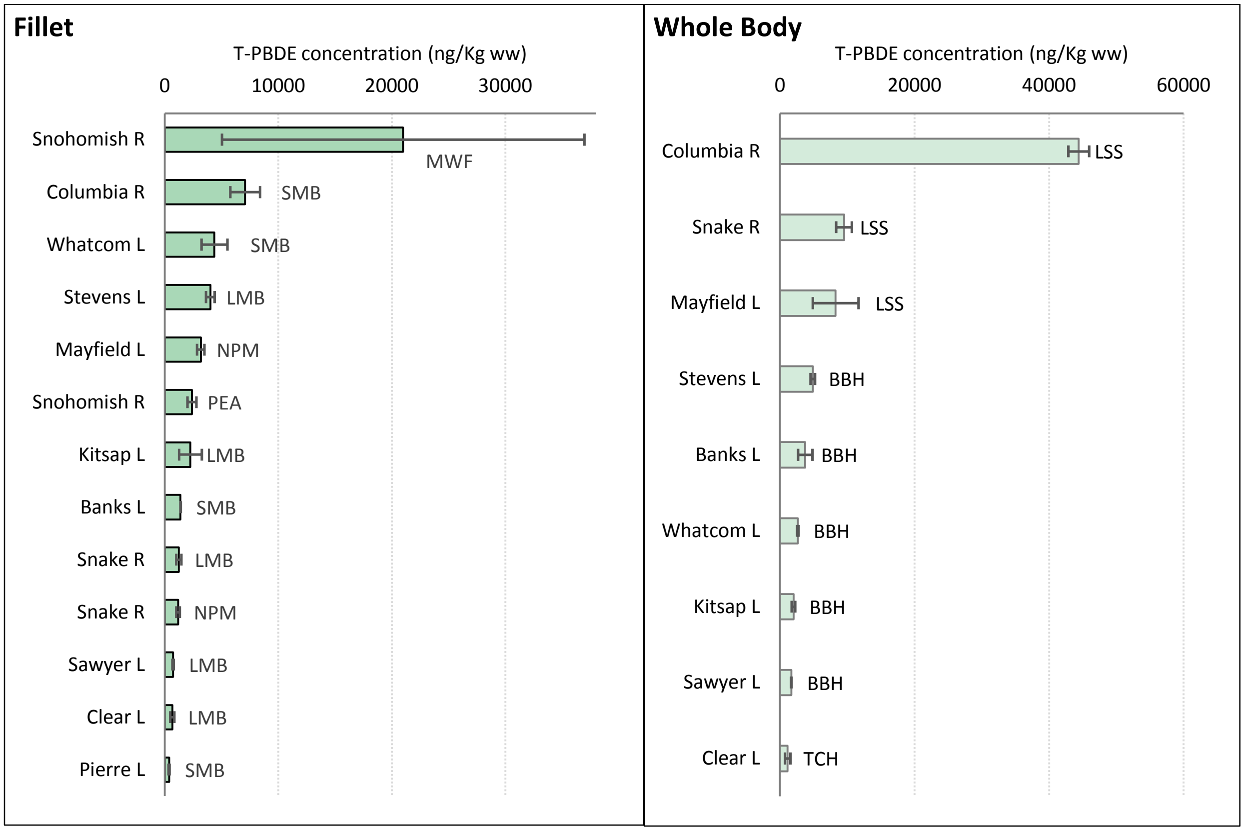

Figure 3. Mean Concentrations (bars) and Ranges (error bars) of T-PBDEs in Fillet and Whole Body Fish Tissue.

n: 2 for all samples.
Species code of each sample is given to the right of blue bars: MWF: mountain whitefish, SMB: smallmouth bass, LMB: largemouth bass, NPM: northern pikeminnow, PEA: peamouth, LSS: largescale sucker, BBH: brown bullhead, TCH: tench.

Forty congeners were analyzed as part of the PBDE suite. Of the 40 congeners, 11 were detected in every sample. The dominant congeners of the Penta-BDE commercial flame retardant – BDE-47, -99, and -100 – contributed the majority of the PBDE burden in samples, with an average contribution of 53%, 15%, and 11% to the total, respectively. The average BDE-47 concentration was 3,470 ng/Kg (median = 1,150 ng/Kg), and mean concentrations of BDE-99 and -100 were 807 ng/Kg and 772 ng/Kg (median = 334 and 289 ng/Kg), respectively. BDE-209 – the primary congener in Deca-BDE – was detected in all but four of the samples, at an average of 3% of the total PBDE burden. However, in two samples the percent contribution was much higher: 30% in a smallmouth bass from the Columbia River and 28% in a northern pikeminnow from Mayfield Lake. BDE-49, -154, and -153 also had average contributions of 3% to the total.

The T-PBDE concentrations measured in this study were within the range of fish tissue samples analyzed by other statewide Ecology fish tissue studies (Figure 4). The three highest samples – from the Columbia River (largescale sucker whole body and smallmouth bass fillet) and Snohomish River (mountain whitefish fillet) – were around the 80th percentile of the statewide data (data set of 2001-2014 freshwater fish tissue surveys accessed from Ecology's EIM database). All other samples fell between 6 - 60% of the statewide data.

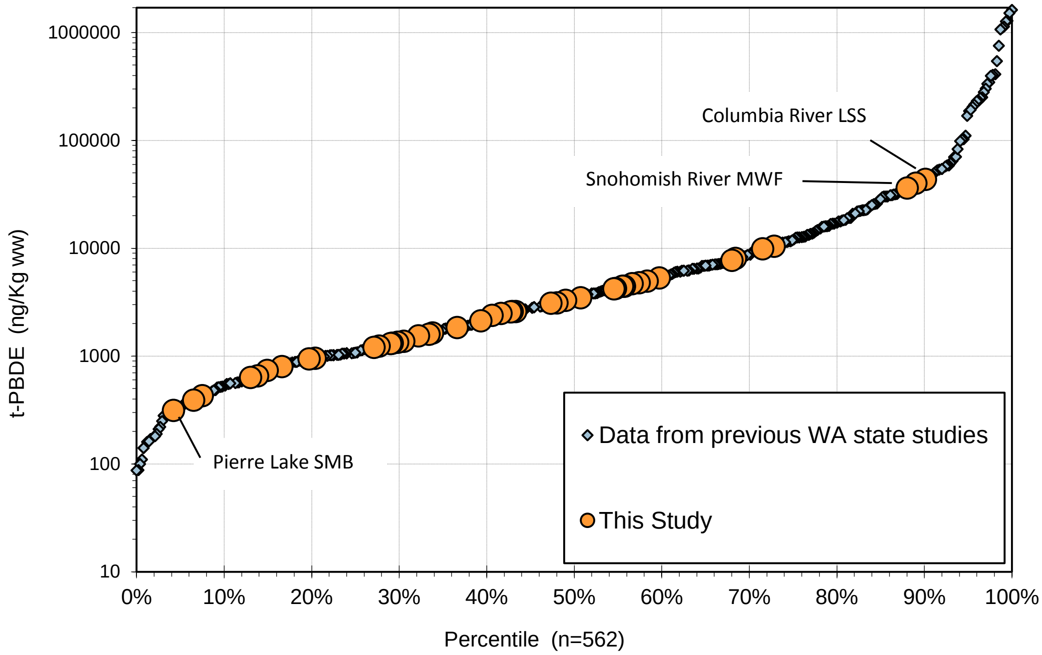

Figure 4. Cumulative Frequency Distribution for T-PBDEs in Fillet and Whole Body Fish Tissue Collected in Washington State, 2001 - 2014.

Data from previous studies include freshwater fish tissue from general environmental monitoring studies entered in Ecology's EIM database.
All data points represent the sum of 13 PBDE congeners; fillet and whole body tissue samples are combined.
LSS: largescale sucker; MWF: mountain whitefish; SMB: smallmouth bass.

BTBPE, DBDPE, HBBz, and PBEB

All four of the alternative brominated flame retardants analyzed (BTBPE, DBDPE, HBBz, and PBEB) were detected in this study, with at least one of the analytes detected in 89% of samples. Results are summarized in Table 6 and graphically displayed in Figure 5. HBBz was detected most frequently out of the four analytes, with 77% of the samples containing HBBz at or above the estimated detection limit of 0.1 ng/Kg. BTBPE and DBDPE analyses had higher estimated detection limits (1.5 and 20 ng/Kg, respectively) and were detected less frequently – in 32% and 16% of samples. PBEB was detected infrequently (7% of samples), with an estimated detection limit of 0.1 ng/Kg.

Table 6. Statistical Summary of Non-BDE Flame Retardant Results in Fish Tissue.

Analyte	No. of Detections	Detection Frequency	Min* (ng/Kg ww)	Max* (ng/Kg ww)	Mean* (ng/Kg ww)	Median* (ng/Kg ww)
BTBPE	14	32%	0.68	44.5	6.96	2.47
DBDPE	7	16%	14.1	304	91.4	35.0
HBBz	34	77%	0.163	2.20	0.792	0.666
PBEB	3	7%	0.100	0.198	0.133	0.102

*Statistic includes detected values only.

DBDPE was present in the highest concentrations of the four analytes, ranging from 14.1 – 304 ng/Kg. Two samples from Mayfield Lake (a largescale sucker and northern pikeminnow) were particularly high in DBDPE (304 and 197 ng/Kg), while samples from Pierre Lake, Snohomish River, Sawyer Lake, and Lake Whatcom ranged from 14.1 – 60.1 ng/Kg. Concentrations of BTBPE were the next highest of the four analytes. BTBPE ranged from 0.682 – 44.5 ng/Kg, with the maximum concentration found in a largescale sucker sample from Mayfield Lake. Other waterbodies with detections of BTBPE included Pierre Lake, Snohomish River, Lake Stevens, and Lake Whatcom. HBBz was detected in all waterbodies at low concentrations (range = 0.163 – 2.20 ng/Kg). PBEB was present in the lowest concentrations (< 0.2 ng/Kg), with detections only in Snohomish River and Lake Stevens samples.

Median concentrations of non-BDE flame retardants were two to three orders of magnitude lower than T-PBDE concentrations. BDE-209 (the main component of the largely phased out commercial flame retardant, Deca-BDE) was detected more often, but in similar concentrations to DBDPE, a major replacement for Deca-BDE. DBDPE is structurally similar to deca-BDE and behaves similarly in the environment, exhibiting environmental persistence and long-range atmospheric transport characteristics (Ricklund et al., 2010). BDE-209 is known to break down into lower brominated congeners metabolically in some species of fish (Roberts et al., 2011; Stapleton et al., 2004), whereas debromination of DBDPE in fish tissue has not been established (He et al., 2012; Wang et al., 2012). BDE-209 was found at a similar percent contribution among the species and sample types in this study.

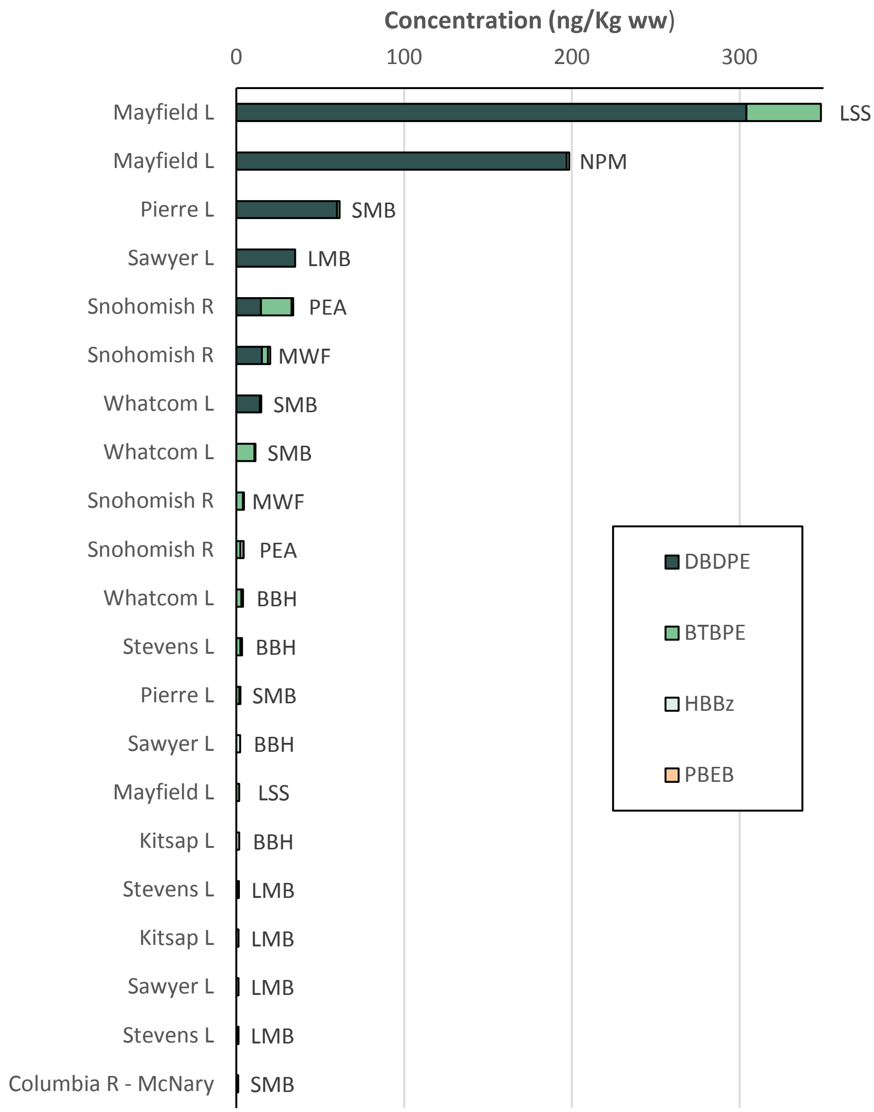

Figure 5. Non-BDE Concentrations in Fish Tissue Samples Ranked by Sum of DBDPE, BTBPE, HBBz, and PBEB.

Only samples with a summed concentration greater than 1.0 ng/Kg are shown.
LSS: largescale sucker; NPM: northern pikeminnow; SMB: smallmouth bass; LMB: largemouth bass; PEA: peamouth; MWF: mountain whitefish; BBH: brown bullhead.

Comparison to Other Studies

The only other regional data on alternative brominated flame retardants in Washington freshwater fish tissue comes from USGS studies on the Columbia River. BTBPE was analyzed for but not detected in samples of largescale sucker and larval lamprey tissue from the Lower Columbia River system (Nilsen et al., 2014; Nilsen et al., 2015). Similar to the current study's results, non-BDE brominated flame retardants in fish tissue have been detected infrequently and in low concentrations in other parts of North America. Non-BDE flame retardants in fish from similar trophic levels in the Great Lakes and St. Lawrence River system were found in similar frequency and lipid-normalized concentrations as the current study (Ismail et al., 2009; Law et al., 2006; Houde et al., 2014). Detected concentrations of BTBPE and DBDPE in the Washington fish samples ranged from 0.03 – 1.06 ng/g lw and 0.49 – 9.0 ng/g lw, respectively. Law et al. (2006) reported BTBPE in Lake Ontario fish samples between <MDL and 1.48 ng/g lw, and DBDPE concentrations ranging from <MDL – 3.30 ng/g lw. BTBPE, HBBz, and PBEB were not detected in San Francisco Bay fish samples (Klosterhaus et al., 2012b).

Alkylphenols and Alkylphenol Ethoxylates

Results of alkylphenolic compounds analyzed in fish tissue are summarized in Table 7 and shown in Figure 6. Forty-eight percent of fish tissue samples contained one or more of the following alkylphenolic compounds analyzed: 4n-OP, NP1EO, and NP2EO. 4-NP was not detected at levels greater than 10 times the lab blank contamination in any of the samples. All four analytes had mean estimated detection limits of 500 ng/Kg. NP2EO was detected in the highest frequency – 34% of samples – at concentrations ranging from 757 – 2,790 ng/Kg, followed by 4n-OP with a 20% detection frequency and range of 445 – 2,270 ng/Kg. NP1EO was detected in 14% of samples (range = 537 – 4,080 ng/Kg).

Table 7. Statistical Summary of Alkylphenolic Compounds Results in Fish Tissue.

Analyte	No. of Detections	Detection Frequency	Min* (ng/Kg ww)	Max* (ng/Kg ww)	Mean* (ng/Kg ww)	Median* (ng/Kg ww)
4n-OP	9	20%	445	2,270	1,060	1,040
4-NP	0	0%	---	---	---	---
NP1EO	6	14%	537	4,080	2,230	2,440
NP2EO	15	34%	757	2,790	1,170	1,060

*Statistic includes detected values only.

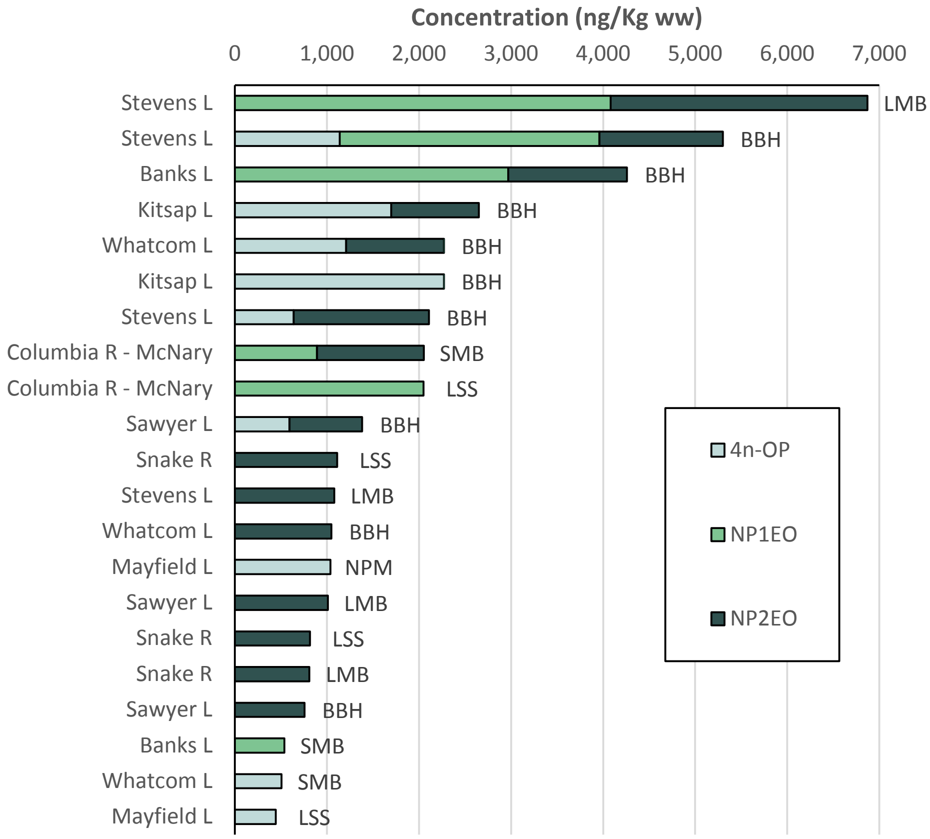

Figure 6. Alkylphenol and Alkylphenol Ethoxylates Concentrations in Fish Tissue Samples Ranked by Sum of 4n-OP, NP1EO, and NP2EO.

LMB: largemouth bass; BBH: brown bullhead; SMB: smallmouth bass; LSS: largescale sucker; NPM: northern pikeminnow. Only detected results are included in graph.

Alkylphenolic compounds in the environment are primarily associated with WWTP effluent, the dominant pathway to aquatic systems (Soares et al., 2008). However, this study found 4n-OP and nonylphenol ethoxylates in fish collected from waterbodies with no direct WWTP effluent discharge, suggesting that stormwater and septic systems may be important pathways for these waterbodies. Lake Stevens samples contained the highest concentrations and the greatest number of detections. All sites with the exception of Clear Lake, Pierre Lake, and Snohomish River had multiple samples with detections of alkylphenolic compounds.

Comparison to Other Studies

Despite the lipophilic characteristics of alkylphenolic compounds, few studies have reported their alkylphenolic compound concentrations on a lipid-weight basis. The wet weight concentrations of alkylphenolic compounds found in the current study were low compared to those reported in the literature. Total NPEs (sum of NP1EO and NP2EO) in the Washington State fish samples ranged from <0.5 – 6.87 ng/g ww (mean = 1.71 ng/g ww), three orders of magnitude lower than largemouth bass collected near wastewater treatment plant (WWTP) outfalls in the Chicago River, IL (T-NPEs ranged from 530 – 8,280 ng/g ww; Lozano et al., 2012) and two orders of magnitude lower than carp collected from the Cuyahoga River (range = 32 – 920 ng/g ww; Rice et al., 2003). The Washington State fish were somewhat closer to, but still lower than, concentrations of T-NPEs in largemouth bass collected from "reference" condition waterbodies in Illinois (T-NPE range = 30 – 110 ng/g ww; Lozano et al., 2012) and carp collected from a site upstream of urban and WWTP influence in Ohio (ng/g ww; Rice et al., 2003). Barber et al. (2015) reported concentrations of OP, NP1EO, and NP2EO in similar species of freshwater fish from the Great Lakes and Upper Mississippi River system that were 1-2 orders of magnitude higher than the current study's samples.

Hexabromocyclododecane

HBCD was analyzed as three diastereoisomers in the fish tissue samples: alpha-, beta-, and gamma- HBCD. Table 8 provides a statistical summary of the HBCD results. Beta- and gamma- HBCD were not detected above the quantitation limit of 100 ng/Kg in any of the samples analyzed; only alpha-HBCD was detected. Studies of HBCD in fish tissue have found that alpha-HBCD is the dominant isomer that accumulates in biota (Law and Herzke, 2011).

A total of 12 fish tissue samples (27%) contained alpha-HBCD above quantitation limits (shown in Figure 7). Detected concentrations ranged from 116 - 362 ng/Kg, with a mean of 242 ng/Kg and a median of 243 ng/Kg. The highest concentration was found in a Lake Whatcom smallmouth bass sample, followed by largescale sucker samples from Mayfield Lake and Columbia River. HBCD was also detected in samples from Banks Lake, Clear Lake, and the Snohomish River.

Table 8. Statistical Summary of HBCD Results in Fish Tissue.

Analyte	No. of Detections	Detection Frequency	Min* (ng/Kg)	Max* (ng/Kg)	Mean* (ng/Kg)	Median* (ng/Kg)
alpha- HBCD	12	27%	116	362	242	243
beta- HBCD	0	0%	---	---	---	---
gamma- HBCD	0	0%	---	---	---	---

*Statistic includes detected values only.

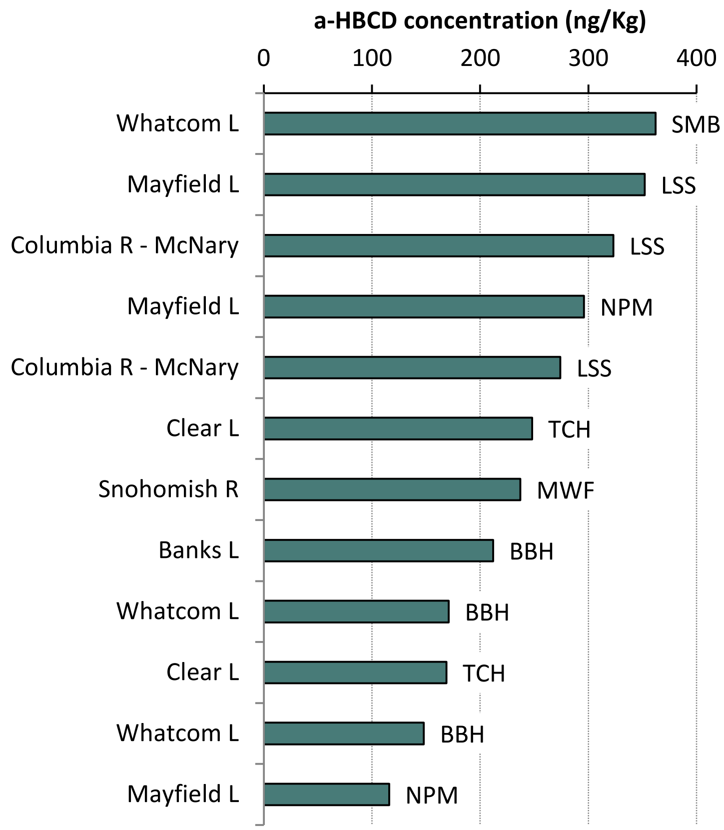

Figure 7. Detected alpha-HBCD Concentrations in Fish Tissue Samples.

SMB: smallmouth bass; LSS: largescale sucker; NPM: northern pikeminnow; TCH: tench; MWF: mountain whitefish; BBH: brown bullhead. Only detected results are included in graph.

Comparison to Other Studies

These results are quite similar to HBCD concentrations found by Johnson and Friese (2012) in fish tissue from four urban/industrial Washington State waterbodies. Detected concentrations of alpha-HBCD (the only isomer found) in fillets of common carp and whole body largescale sucker samples from their study ranged from 2.3 – 11.3 ng/g lw (median = 6.1 ng/g lw), while the current study range was 2.2 – 13.5 ng/g lw (median = 7.3 ng/g lw).

Lipid-normalized HBCD concentrations found in the current study are much lower than those reported for North American bottom-feeder fish samples collected near point sources and are at the lower end of mixed trophic-level fish from diffuse-source waterbodies compiled by Chen et al. (2011) (Law et al., 2006; Tomy et al., 2004). T-HBCD concentrations in freshwater fish samples compiled by Chen et al. (2011) ranged from 290 – 5,010 ng/g lw (median = 2,540 ng/g lw) near point sources and from 2.8 – 370 ng/g lw (median = 20 ng/g lw) in diffuse source waterbodies. However, the current study's results are very similar to T-HBCD lipid weight concentrations reported in fish tissue collected from San Francisco Bay, with T-HBCD in samples of shiner surfperch ranging from 2.5 – 24.7 ng/g lw (median = 6.5 ng/g lw) (Klosterhaus et al., 2012b).

Spatial Trends

The lipid-normalized concentration data showed some general spatial patterns with implications for sources and transport mechanisms. Figure 8 shows lipid-weight contaminant concentrations as a percent of the total across sites. PBDEs tended to be highest in rivers receiving WWTP effluent (Snohomish River and Columbia River). Whereas, non-BDE flame retardant concentrations were highest in lakes receiving a mix of WWTP effluent and atmospheric deposition (Mayfield Lake) and atmospheric deposition alone (Pierre Lake). In particular, DBDPE and BTBPE were detected in Pierre Lake, where atmospheric deposition is likely the only source of these current-use flame retardants. However, the compounds also appeared in lakes with other potential pathways, such as stormwater (Sawyer and Whatcom Lakes). Nonylphenol ethoxylates and OP concentrations were not primarily associated with WWTP inputs, as would be expected based on their dominant exposure via WWTP effluent (Klosterhaus et al., 2012a). Lipid-normalized concentrations of nonylphenol ethoxylates and octylphenol were highest in urban lakes on the west side of the state where stormwater is the likely transport mechanism.

No pattern was evident for HBCD concentrations in fish tissue. Lipid-normalized HBCD concentrations were very similar among fish samples from the Snohomish River and Mayfield, Whatcom, Clear, and Banks Lakes; all of which have varied land uses and potential sources of transport. Atmospheric deposition, stormwater, and WWTP effluent all likely play important roles in HBCD contamination of these waterbodies.

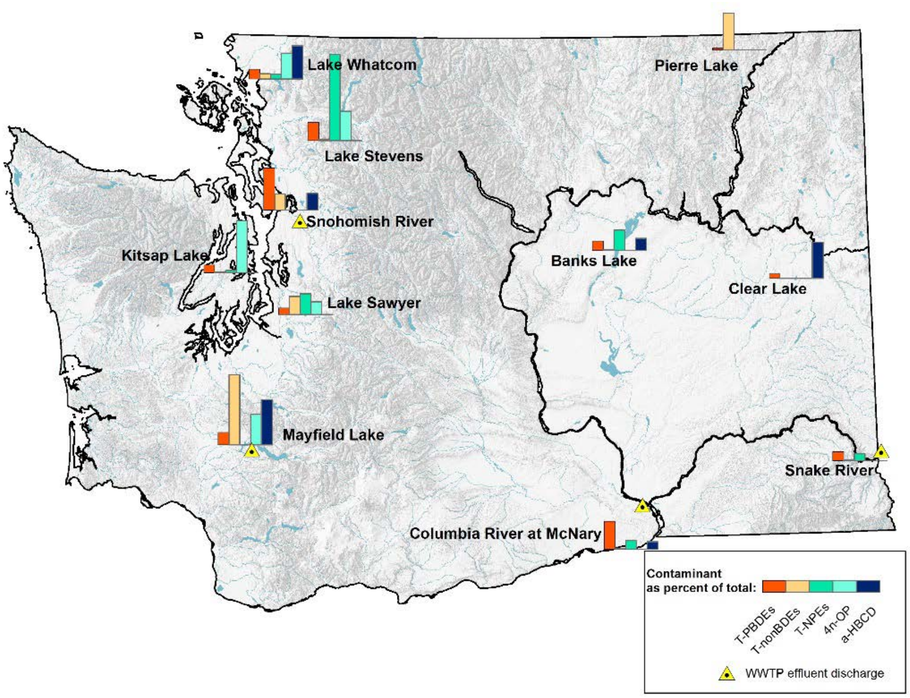

Figure 8. Contaminant Concentrations (ng/g lipid weight) Normalized by Percent of Total for the 2014 Study Locations.

Conclusions

A total of 44 fish tissue samples collected from 11 waterbodies throughout the state were analyzed for brominated flame retardants (BFRs), alkylphenolic compounds, and HBCD during this study. Results of this study support the following conclusions:

- All compounds except for 4-NP, beta-HBCD, and gamma-HBCD were detected in this study. In general, detected concentrations of analytes were in the order of T-PBDEs > alkylphenolic compounds > HBCD > non-BDE flame retardants.
- PBDEs were detected at the highest frequency (100% of samples) and at the highest concentrations of all analytes. T-PBDE concentrations ranged from 332 – 46,000 ng/Kg ww, with a median of 2,700 ng/Kg ww. The three highest concentrations – in fish collected from the Columbia and Snohomish Rivers – were around the 80^{th} percentile of previously reported PBDE data for Washington State freshwater fish, while the rest of the samples fell between 6 – 60% of the statewide data.
- The alkylphenolic compounds NP1EO, NP2EO, and 4n-OP were present in 48% of samples. Concentrations of the detected individual compounds ranged from 445 – 4,080 ng/Kg ww, which are low compared to data from other freshwater systems in North America.
- alpha-HBCD was detected in 27% of samples, with concentrations ranging from 115 – 362 ng/Kg ww. Lipid-normalized HBCD concentrations in this study are very similar to those previously recorded from four urban/industrial Washington State waterbodies and were comparable to HBCD fish tissue levels reported in other parts of North America with diffuse sources.
- Eighty-nine percent of samples contained one or more of the non-BDE flame retardants analyzed. DBDPE was present in the highest concentrations of the four non-BDE flame retardant analytes, ranging from 14.1 – 304 ng/Kg ww, followed by BTBPE which ranged from 0.682 – 44.5 ng/Kg ww. HBBz and PBEB were detected at concentrations ranging from 0.100 – 2.20 ng/Kg ww. Lipid-normalized non-BDE concentrations were similar to those found in other parts of North America.
- DBDPE was found less often, but in similar concentrations to BDE-209, the main congener used in the largely phased out flame retardant Deca-BDE. DBDPE is currently a replacement flame retardant for Deca-BDE.
- T-PBDEs were highest in waterbodies receiving WWTP effluent, whereas non-BDE flame retardant concentrations were highest in waterbodies influenced by WWTP effluent and atmospheric deposition. Non-BDE flame retardants also appeared in lakes with other potential pathways, such as stormwater. Alkylphenolic compounds were found in waterbodies not directly receiving WWTP effluent and were highest in urban lakes. No pattern was apparent for HBCD concentrations in fish tissue, as lipid-normalized concentrations were similar across waterbodies receiving atmospheric deposition, stormwater, and WWTP effluent.

Recommendations

The results of this study should be used to support prioritization of chemicals to be addressed by Ecology through chemical action plans (CAPs) and other efforts to reduce toxics in the state. Specific recommendations from this study include the following:

- Nonylphenol ethoxylates and octylphenol should be considered for inclusion in Ecology's PBT List, if new chemicals are added to the list in the future. Additional monitoring of NP levels in fish tissue is warranted, given that the lab blank contamination in this study prohibited detections. Future monitoring of alkylphenolic compounds in waterbodies of Washington State receiving wastewater treatment plant effluent should be considered.
- The brominated flame retardants DBDPE and BTBPE should also be considered for inclusion in Ecology's PBT List, if new chemicals are added. While a CAP has been developed for PBDEs (Ecology et al., 2006) and PBDEs have been largely phased out of commercial use, the results from this study suggest that replacement chemicals for PBDEs may persist in the environment in Washington State.
- Future monitoring of these compounds should include additional media, such as surface water, sediments, and wastewater treatment plant effluent.
- The finding of two current-use brominated alternative flame retardants in fish tissue in this study indicates that exploratory monitoring of additional halogenated flame retardants is warranted.

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Appendices

Appendix A. Biological Data on Fish Samples

Table A-1. Biological Data on Fish Samples Analyzed for this Study.

Waterbody	Species	Collect Date	Sample ID	Number of Fish in Composite	Sample Type	Mean Length (mm)	Mean Weight (g)	Mean Age (yr)
Banks L	Brown bullhead	10/15/2014	1412023-01	3	Whole	345	631	7.3
	Brown bullhead	10/15/2014	1412023-02	3	Whole	290	332	6.0
	Smallmouth bass	10/15/2014	1412023-03	5	Fillet	422	959	8.4
	Smallmouth bass	10/15/2014	1412023-04	4	Fillet	360	610	4.5
Clear L	Largemouth bass	10/14/2014	1412023-05	5	Fillet	313	562	3.0
	Largemouth bass	10/14/2014	1412023-06	5	Fillet	462	2102	9.4
	Tench	10/14/2014	1412023-07	5	Whole	335	555	3.4
	Tench	10/14/2014	1412023-08	4	Whole	429	1033	11.5
Columbia R - McNary	Largescale sucker	10/16/2014	1412023-09	5	Whole	599	2273	17.6
	Largescale sucker	10/16/2014	1412023-10	5	Whole	498	1424	12.2
	Smallmouth bass	10/16/2014	1412023-11	3	Fillet	357	676	3.7
	Smallmouth bass	10/16/2014	1412023-12	2	Fillet	420	1157	5.5
Kitsap L	Brown bullhead	9/16/2014	1412023-13	5	Whole	201	105	1.0
	Brown bullhead	9/16/2014	1412023-14	4	Whole	253	229	1.3
	Largemouth bass	9/16/2014	1412023-15	5	Fillet	285	382	1.4
	Largemouth bass	9/16/2014	1412023-16	4	Fillet	457	1985	5.5
Mayfield L	Largescale sucker	10/27/2014	1412023-17	5	Whole	486	1181	12.2
	Largescale sucker	10/27/2014	1412023-18	5	Whole	446	883	11.6
	Northern pikeminnow	10/27/2014	1412023-19	5	Fillet	441	806	14.4
	Northern pikeminnow	10/27/2014	1412023-20	5	Fillet	346	348	7.2
Pierre L	Smallmouth bass	10/13/2014	1412023-21	5	Fillet	350	620	7.6
	Smallmouth bass	10/13/2014	1412023-22	5	Fillet	309	389	6.8
Sawyer L	Brown bullhead	10/21/2014	1412023-23	4	Whole	207	107	1.0
	Brown bullhead	10/21/2014	1412023-24	4	Whole	246	174	1.0
	Largemouth bass	10/21/2014	1412023-25	5	Fillet	334	558	2.8
	Largemouth bass	10/21/2014	1412023-26	5	Fillet	285	325	2.0
Snake R	Largemouth bass	10/15/2014	1412023-27	5	Fillet	423	1281	5.2
	Largemouth bass	10/15/2014	1412023-28	5	Fillet	303	446	1.8
	Largescale sucker	10/15/2014	1412023-29	5	Whole	513	1386	14.2
	Largescale sucker	10/15/2014	1412023-30	5	Whole	461	1008	10.6
	Northern pikeminnow	10/15/2014	1412023-31	3	Fillet	257	141	3.0
	Northern pikeminnow	10/15/2014	1412023-32	4	Fillet	219	85	3.0
Snohomish R	Mountain whitefish	11/19/2014	1412023-33	5	Fillet	239	105	1.2
	Mountain whitefish	11/19/2014	1412023-34	5	Fillet	310	262	4.2
	Peamouth	10/29/2014	1412023-35	5	Fillet	253	146	5.0
	Peamouth	10/29/2014	1412023-36	5	Fillet	217	94	4.0
Stevens L	Brown bullhead	10/28/2014	1412023-37	3	Whole	300	366	2.0
	Brown bullhead	10/28/2014	1412023-38	5	Whole	215	122	1.0
	Largemouth bass	10/28/2014	1412023-39	5	Fillet	252	256	1.0
	Largemouth bass	10/28/2014	1412023-40	4	Fillet	281	363	1.3
Whatcom L	Brown bullhead	9/30/2014	1412023-41	5	Whole	241	189	1.0
	Brown bullhead	9/30/2014	1412023-42	5	Whole	224	162	1.0
	Smallmouth bass	9/30/2014	1412023-43	4	Fillet	382	880	5.0
	Smallmouth bass	9/30/2014	1412023-44	5	Fillet	423	1312	7.8

Appendix B. Laboratory Data on Fish Samples

Table B-1. Laboratory Data on Fish Samples Analyzed for this Study.

Waterbody	Sample ID	Species Code	Sample Type	T-PBDEs (ng/Kg)	BTBPE (ng/Kg)	DBDPE (ng/Kg)	HBB (ng/Kg)	PBEB (ng/Kg)	alpha-HBCD (ng/g)	beta-HBCD (ng/g)	gamma-HBCD (ng/g)	4n-OP (ng/g)	4-NP (ng/g)	NP1EO (ng/g)	NP2EO (ng/g)	Lipids (%)
Banks Lake	1412023-01	BBH	W	4834	1.78 UJ	33.5 UJ	0.403 NJ	0.113 UJ	0.0988 U	0.0988 U	0.0988 U	1.11 UJ	17.4 UJ	1.02 UJ	0.574 UJ	3.78
	1412023-02	BBH	W	2672	1.7 UJ	108 UJ	0.391 NJ	0.114 UJ	0.212	0.0975 U	0.0975 U	0.45 UJ	17.1 UJ	2.97 J	1.29 J	3
	1412023-03	SMB	F	1393	2.32 UJ	62 UJ	0.412 NJ	0.111 UJ	0.096 U	0.096 U	0.096 U	4.71 UJ	55.6 UJ	0.481 UJ	1.11 J	1.16
	1412023-04	SMB	F	1363	7.32 UJ	111 UJ	0.488 J	0.112 UJ	0.0991 U	0.0991 U	0.0991 U	0.673 UJ	16.2 UJ	0.467 UJ	0.575 UJ	1.41
Clear Lake	1412023-05	LMB	F	459	7.59 UJ	107 UJ	0.574 J	0.115 UJ	0.0969 U	0.0969 U	0.0969 U	0.463 UJ	13.5 UJ	0.463 UJ	0.463 UJ	1.24
	1412023-06	LMB	F	854	4.76 UJ	54.2 UJ	0.269 J	0.113 UJ	0.0966 U	0.0966 U	0.0966 U	0.485 UJ	13.5 UJ	0.485 UJ	0.551 UJ	1.57
	1412023-07	TCH	W	1561	4.35 UJ	48 UJ	0.665 NJ	0.112 UJ	0.248	0.1 U	0.1 U	0.498 UJ	9.12 UJ	0.498 UJ	0.498 UJ	2.67
	1412023-08	TCH	W	725 NJ	1.16 UJ	37.8 UJ	0.16 UJ	0.115 UJ	0.169	0.0982 U	0.0982 U	0.485 UJ	13.5 UJ	0.485 UJ	0.588 UJ	1.44
Columbia River at McNary	1412023-09	LSS	W	42867	5.75 UJ	40.6 UJ	0.558 NJ	0.115 UJ	0.274	0.0998 U	0.0998 U	10.3 UJ	23.3 UJ	2.05 J	0.576 UJ	11.6
	1412023-10	LSS	W	45956	8.11 UJ	24.5 UJ	0.689 NJ	0.116 UJ	0.323	0.0995 U	0.0995 U	6.88 UJ	13.6 UJ	0.79 UJ	0.582 UJ	14.8
	1412023-11	SMB	F	5779 J	3.98 UJ	40.5 UJ	1.17 J	0.113 UJ	0.0987 U	0.0987 U	0.0987 U	0.5 UJ	13.5 UJ	1.33 UJ	0.591 UJ	1.73
	1412023-12	SMB	F	8385	3.58 UJ	20.3 UJ	0.885 NJ	0.115 UJ	0.0979 U	0.0979 U	0.0979 U	1.05 UJ	13 UJ	0.892 J	1.16 J	3.19
Kitsap Lake	1412023-13	BBH	W	1762	5.5 UJ	42.1 UJ	1.73 J	0.115 UJ	0.0998 U	0.0998 U	0.0998 U	1.7 J	21 UJ	0.742 UJ	0.952 J	4.3
	1412023-14	BBH	W	2269	4.59 UJ	11.5 UJ	0.588 NJ	0.116 UJ	0.0968 U	0.0968 U	0.0968 U	2.27 J	20.3 UJ	1.23 UJ	0.566 UJ	4.4
	1412023-15	LMB	F	1259	1.32 UJ	43.7 UJ	1.37 NJ	0.114 UJ	0.0984 U	0.0984 U	0.0984 U	0.49 UJ	13.6 UJ	0.49 UJ	0.689 UJ	1.31
	1412023-16	LMB	F	3252	1.27 UJ	50.4 UJ	0.747 UJ	0.18 UJ	0.0966 U	0.0966 U	0.0966 U	0.495 UJ	11.2 UJ	0.865 UJ	0.495 UJ	2.03
Mayfield Lake	1412023-17	LSS	W	11659	44.5 J	304 J	0.418 UJ	0.102 UJ	0.0989 U	0.0989 U	0.0989 U	0.445 J	21.7 UJ	0.444 UJ	0.509 UJ	7.24
	1412023-18	LSS	W	4877	1.75 NJ	75.6 UJ	0.617 UJ	0.133 UJ	0.352	0.0989 U	0.0989 U	1.83 UJ	31.7 UJ	1.46 UJ	0.498 UJ	6.71
	1412023-19	NPM	F	3498	1.42 J	197 NJ	0.821 UJ	0.154 UJ	0.296	0.1 U	0.1 U	1.04 J	12.5 UJ	0.495 UJ	0.51 UJ	2.19
	1412023-20	NPM	F	2836	2.35 UJ	86.6 UJ	0.974 NJ	0.276 UJ	0.116	0.0987 U	0.0987 U	0.463 UJ	9.73 UJ	0.834 UJ	0.463 UJ	1.56
Pierre Lake	1412023-21	SMB	F	332	1.49 NJ	60.1 NJ	0.501 UJ	0.109 UJ	0.0971 U	0.0971 U	0.0971 U	0.58 UJ	11.4 UJ	0.493 UJ	0.617 UJ	1.76
	1412023-22	SMB	F	424	1.82 NJ	17.5 UJ	0.561 NJ	0.0986 UJ	0.0972 U	REJ	0.0972 U	0.493 UJ	10.2 UJ	0.493 UJ	0.493 UJ	1.46
Sawyer Lake	1412023-23	BBH	W	1711	2.5 UJ	30.3 UJ	2.2 J	0.115 UJ	0.099 U	0.099 U	0.099 U	0.595 J	12.4 UJ	0.75 UJ	0.789 J	2.67
	1412023-24	BBH	W	1640	2.69 UJ	28.7 UJ	0.767 UJ	0.187 UJ	0.0978 U	0.0978 U	0.0978 U	1.34 UJ	16.5 UJ	0.448 UJ	0.757 J	2.23
	1412023-25	LMB	F	671	2.41 UJ	35 J	0.547 UJ	0.139 UJ	0.097 U	0.097 U	0.097 U	0.49 UJ	14.3 UJ	1.03 UJ	0.49 UJ	1.04
	1412023-26	LMB	F	775	2.54 UJ	52.6 UJ	1.25 NJ	0.129 UJ	0.0949 U	0.0949 U	0.0949 U	0.5 UJ	15.4 UJ	1.67 UJ	1.01 J	0.79

Table B-1 Continued.

Waterbody	Sample ID	Species Code	Sample Type	T-PBDEs (ng/Kg)	BTBPE (ng/Kg)	DBDPE (ng/Kg)	HBB (ng/Kg)	PBEB (ng/Kg)	alpha-HBCD (ng/g)	beta-HBCD (ng/g)	gamma-HBCD (ng/g)	4n-OP (ng/g)	4-NP (ng/g)	NP1EO (ng/g)	NP2EO (ng/g)	Lipids (%)
Snake River	1412023-27	LMB	F	1468	3.37 UJ	40.4 UJ	0.723 UJ	0.178 UJ	0.0985 U	0.0985 U	0.0985 U	0.49 UJ	15.4 UJ	0.49 UJ	0.5 UJ	1.37
	1412023-28	LMB	F	1014	1.52 UJ	96.5 UJ	0.667 NJ	0.132 UJ	0.0984 U	0.0984 U	0.0984 U	0.498 UJ	13.1 UJ	0.498 UJ	0.81 J	1.24
	1412023-29	LSS	W	10703	0.896 UJ	84.3 UJ	0.429 UJ	0.108 UJ	0.0968 U	0.0968 U	0.0968 U	4.71 UJ	55.6 UJ	0.481 UJ	1.11 J	5.77
	1412023-30	LSS	W	8368	1.66 UJ	9.4 UJ	0.379 NJ	0.0955 UJ	0.0971 U	0.0971 U	0.0971 U	3.07 UJ	43 UJ	0.483 UJ	0.815 J	7.62
	1412023-31	NPM	F	1318	1.88 UJ	8.9 UJ	0.163 NJ	0.0973 UJ	0.0993 U	0.0993 U	0.0993 U	0.486 UJ	13.3 UJ	0.485 UJ	0.558 UJ	1.73
	1412023-32	NPM	F	1011	3.89 UJ	19.7 UJ	0.379 NJ	0.0975 UJ	0.099 U	0.099 U	0.099 U	0.493 UJ	16.4 UJ	0.833 UJ	0.633 UJ	1.54
Snohomish River	1412023-33	MWF	F	5033	3.89 J	47 UJ	0.622 J	0.129 UJ	0.0992 U	0.0992 U	0.0992 U	0.481 UJ	14.6 UJ	1.47 UJ	0.63 UJ	3.16
	1412023-34	MWF	F	36965	3.62 NJ	15.2 NJ	1.34 NJ	0.0974 UJ	0.237	0.0996 U	0.0996 U	0.485 UJ	19.1 UJ	1.44 UJ	0.592 UJ	2.49
	1412023-35	PEA	F	1986	2.52 NJ	6.81 UJ	1.69 NJ	0.198 J	0.0989 U	0.0989 U	0.0989 U	0.485 UJ	15 UJ	0.485 UJ	0.509 UJ	1.84
	1412023-36	PEA	F	2780	18.3 J	14.6 NJ	1.01 J	0.0982 UJ	0.0992 U	0.0992 U	0.0992 U	0.498 UJ	14.1 UJ	0.498 UJ	0.557 UJ	1.72
Lake Stevens	1412023-37	BBH	W	4560	2.42 NJ	19.6 UJ	0.76 NJ	0.1 NJ	0.1 U	0.1 U	0.1 U	1.14 J	24.5 UJ	2.82 J	1.34 J	3.81
	1412023-38	BBH	W	5225	1.38 UJ	11 UJ	0.728 NJ	0.102 NJ	0.0973 U	0.0973 U	0.0973 U	0.638 J	24.4 UJ	0.557 UJ	1.47 J	2.99
	1412023-39	LMB	F	3627	1.21 NJ	9.11 UJ	0.359 J	0.099 UJ	0.0993 U	0.0993 U	0.0993 U	0.498 UJ	16.8 UJ	4.08 J	2.79 J	1.19
	1412023-40	LMB	F	4400	0.682 NJ	16.8 UJ	0.528 NJ	0.0978 UJ	0.0963 U	0.0963 U	0.0963 U	0.5 UJ	15.7 UJ	0.5 UJ	1.08 J	1.79
Lake Whatcom	1412023-41	BBH	W	2534	1.09 UJ	11.7 UJ	0.855 J	0.0988 UJ	0.148	0.0961 U	0.0961 U	1.21 J	18.1 UJ	0.585 UJ	1.06 J	4.94
	1412023-42	BBH	W	2734	2.97 NJ	5.99 UJ	0.903 J	0.0951 UJ	0.171	0.0996 U	0.0996 U	1.72 UJ	30.6 UJ	1.79 UJ	1.05 J	4.49
	1412023-43	SMB	F	3232	1.93 UJ	14.1 J	0.733 NJ	0.0996 UJ	0.362	0.0985 U	0.0985 U	0.488 UJ	21.4 UJ	0.488 UJ	0.514 UJ	2.89
	1412023-44	SMB	F	5507	10.8 NJ	9.07 UJ	0.633 NJ	0.0992 UJ	0.0973 U	0.0973 U	0.0973 U	0.507 J	16.4 UJ	0.628 UJ	0.766 UJ	2.43

Species Codes: BBH: brown bullhead; SMB: smallmouth bass; LMB: largemouth bass; TCH: tench; LSS: largescale sucker;
NPM: northern pikeminnow; MWF: mountain whitefish; PEA: peamouth.
Sample Types: F: fillet; W: whole body.
J: Estimated value.
U: Compound not detected at or above limit of quantitation.
UJ: Compound not detected at or above estimated value.
NJ: Compound was tentatively identified; result is an estimated value.

Appendix C. Glossary, Acronyms, and Abbreviations

Glossary

Diffuse source: Pollution that enters any waters of the state from any dispersed land-based or water-based activities, including but not limited to atmospheric deposition, surface-water runoff from agricultural lands, urban areas, or forest lands, subsurface or underground sources.

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.

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.

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

BFR	Brominated flame retardant
BTBPE	1,2-Bis(2,4,6-tribromophenoxy)ethane
CAP	Chemical action plan
DBDPE	Decabromodiphenylethane
Ecology	Washington State Department of Ecology
EIM	Environmental Information Management database
EPA	U.S. Environmental Protection Agency
EPS	Expanded polystyrene
HBBz	Hexabromobenzene
HBCD	Hexabromocyclododecane
MEL	Manchester Environmental Laboratory
NP	Nonylphenol
NPE	Nonylphenol ethoxylate
NP1EO	Nonylphenol monoethoxylate
NP2EO	Nonylphenol diethoxylate
OP	Octylphenol
PBDE	Polybrominated diphenyl ethers
PBEB	Pentabromoethylbenzene
PBT	Persistent, bioaccumulative, toxic chemical
RPD	Relative percent difference
RSD	Relative standard deviation

SOP	Standard operating procedures
WAC	Washington Administrative Code
WRIA	Water Resource Inventory Area
WWTP	Wastewater treatment plant
XPS	Extruded polystyrene

Units of Measurement

g	gram, a unit of mass
lw	lipid weight
mm	millimeters
ng/g	nanograms per gram (parts per billion)
ng/Kg	nanograms per kilogram (parts per trillion)
ww	wet weight