PBT Monitoring:
PBDE Flame Retardants in Spokane River Fish, 2009

DEPARTMENT OF ECOLOGY
State of Washington

March 2010
Publication No. 10-03-015

Publication and Contact Information

This report is available on the Department of Ecology's website at
www.ecy.wa.gov/biblio/1003015.html

Data for this project are available at Ecology's Environmental Information Management (EIM) website www.ecy.wa.gov/eim/index.htm. Search User Study ID, CFUR0005.

Ecology's Activity Tracker Code for this study is 09-502.

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Cover photo: Spokane River upstream of Upriver Dam.

PBT Monitoring:
PBDE Flame Retardants
in Spokane River Fish, 2009

by
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Waterbody Numbers:

Rock Lake:	WA-34-9290
Spokane River:	WA-57-1010, WA-54-1020, WA-54-9040
Williams Lake:	WA-34-9480

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Abstract

The Washington State Department of Ecology (Ecology) analyzed 13 polybrominated diphenyl ethers (PBDEs) in fish tissue samples from the Spokane River to characterize contaminant concentrations in the diet of osprey (*Pandion haliaetus*) nesting along the river. The project also assessed spatial and temporal trends in fish tissue PBDE concentrations since the river was last sampled in 2005. Sampling was conducted as part of a larger cooperative study with the U.S. Geological Survey (USGS) investigating PBDE concentrations in osprey eggs along the Spokane River.

Fish were collected from six locations from the Idaho border through Long Lake during spring 2009. Reference samples were also collected from Rock Lake and Williams Lake. In total, 6 species were analyzed including 27 whole body, 9 fillet, and 8 carcass composite samples. Total PBDE concentrations in whole fish composites from the Spokane River ranged from 30.6 – 2,531 µg/Kg wet weight. Concentrations were generally higher in sportfish than in bottom dwellers. The highest concentrations were found in the lower sections of the river from the Nine Mile reach through Lower Long Lake.

Temporal trends in Spokane River PBDE concentrations were assessed by comparing PBDE results to an Ecology survey conducted in fall 2005. Largescale sucker (*Catostomus macrocheilus*) and mountain whitefish (*Prosopium williamsoni*) samples were paired between the two studies based on sampling location and fish composite length. Wet weight PBDE concentrations from the 2009 survey were lower in 21 of 23 paired samples, often by greater than 50%. The apparent decline in PBDE concentrations was hypothesized to be associated with seasonal differences in fish physiology and river hydrology.

Acknowledgements

The authors of this report would like to thank the following people for their contribution to this study:

- Chuck Henny, James Kaiser, and USGS staff for collaboration with osprey egg collection.
- Rene Wiley for help with site access.
- Washington State Department of Ecology staff:
  - Michael Friese and Casey Deligeannis for help with sample collection and processing.
  - Manchester Environmental Laboratory (MEL) staff: Dolores Montgomery, Stuart Magoon, and Leon Weiks for analysis of the samples and assistance with this project.
  - Dale Norton for project guidance and review of the Quality Assurance Project Plan and draft reports.
  - Dave Serdar for reviewing the draft report.
  - Joan LeTourneau and Jean Maust for formatting and editing the final report.

Introduction

Background on PBDEs

Polybrominated diphenyl ethers (PBDEs) are hydrophobic, lipophilic contaminants composed of a diphenyl ether molecule with up to 10 bromine atoms attached to the rings allowing for 209 possible congeners (Figure 1). PBDEs have been manufactured since the 1970s as fire-retarding additives in numerous products including polyurethane foams, plastics, paints, textiles, and electronics (Rahman et al., 2001).

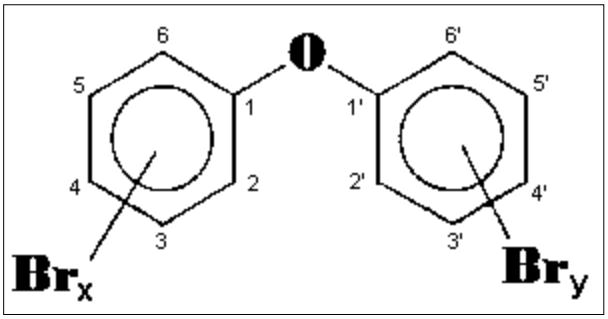

Figure 1. PBDE General Structure.

PBDEs are a high-volume production chemical with the majority of its use in North America (Alaee et al., 2003). Commercial production of PBDEs has primarily consisted of penta-BDE, octa-BDE, and deca-BDE formulations. Production of penta-BDE and octa-BDE was voluntarily phased out in the United States in 2004 due to concerns over their toxicity. The Washington State legislature passed legislation banning products containing penta- and octa-BDEs (PBDE Rule, Chapter 70.76 RCW) in 2007 and deca-BDEs in upholstery and electronics in 2011 (Ecology and DOH, 2008). Recently, the two U.S. producers of deca-BDE and the largest importer agreed to end all production, importation, and sale of deca-BDE by 2013 (EPA, 2009).

Environmental release of PBDEs can occur during initial synthesis, incorporation into products, use and disposal via direct discharge, leaching, volatilization, incineration, wastewater treatment plants, and other sources (Hale et al., 2003). Once in the environment, PBDEs enter the aquatic food chain where they bioaccumulate. Deca-BDE is debrominated to lower brominated congeners through exposure to UV light, microorganisms, and metabolic processes in some fish species (Eriksson et al., 2004; Gerecke et al., 2005; Stapleton et al., 2006). This is of particular interest since lower brominated congeners are more toxic (Tomy et al., 2004; Kelly et al., 2008).

There is increasing experimental evidence that PBDE exposure may be detrimental to wildlife health, affecting sex and thyroid hormones, modulation of liver enzymes, immune system function, and neurological development (Kierkegaard et al., 2006; Darnerud et al., 2001; Birnbaum and Staskal, 2004). In order to address these concerns, the Washington State Department of Ecology (Ecology) and Washington State Department of Health (DOH) finalized

a Chemical Action Plan for PBDEs in 2006. The plan outlines future steps to reduce the threat of PBDEs in the environment (Ecology and DOH, 2006).

PBDEs in Washington State

Several studies have examined PBDEs in Washington State fish (Johnson and Olson, 2001; Seiders and Yake, 2002; Johnson et al., 2006), surface waters (Johnson et al., 2006; Sandvik, 2009), stormwater (Lubliner, 2009), wastewater treatment plant effluent (EIM Project ID YAKR37TX), and osprey eggs (Henny et al., 2009). In 2005, Ecology conducted a statewide PBDE survey along with a more intensive study of fish tissues from 6 reaches of the Spokane River (Johnson et al., 2006; Serdar and Johnson, 2006). Results from these studies have identified the Spokane River as having the highest PBDE levels in both water and fish tissue samples statewide.

Figure 2 displays a cumulative frequency graph of total PBDEs in Washington freshwater fish samples from various Ecology monitoring efforts. The Spokane River values are from sampling in 1999 and 2005. Currently, sources and causes of elevated PBDE concentrations in the Spokane River are unknown.

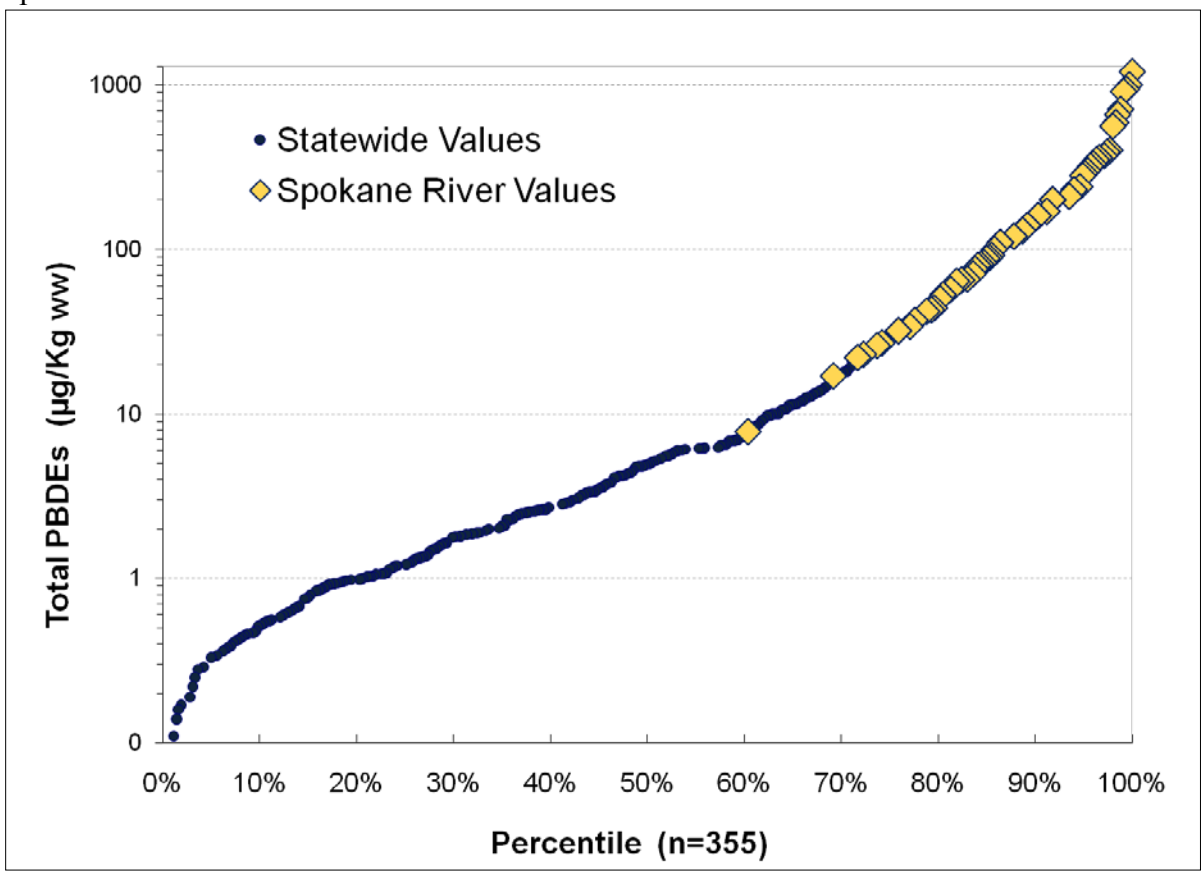

Figure 2. Cumulative Frequency Graph of Total PBDEs in Freshwater Fish Tissue Samples from Selected Ecology Studies.

The high levels of PBDE contamination in the Spokane River are relevant in a national context as well. Total concentrations in Spokane River fish measured by Serdar and Johnson (2006) were elevated compared to many previously reported values in North American rivers (Hites, 2004; Hale et al., 2003; Rayne et al., 2003; Xia, 2008). Hites (2004) reviewed PBDE concentrations of several studies nationwide conducted from 1994 – 2000. Mean concentrations of lipid-normalized PBDEs in suckers (largescale and bridgelip) and mountain whitefish collected by Serdar and Johnson (2006) were roughly 5 and 11 times greater, respectively, than the mean PBDE fish value calculated by Hites.

Study Goals and Objectives

In view of the elevated water and fish tissue concentrations recorded along the Spokane River and the potential deleterious effects on wildlife, Ecology, in cooperation with USGS, conducted a one-time survey measuring PBDE concentrations in fish tissues and osprey eggs along the river during 2009.

This report details findings from the fish portion of the project. The primary goal of the fish tissue study is to assess PBDE exposure in the osprey diet. Secondary objectives of the fish tissue study are to:

- Identify spatial and fish species patterns in the environmental distribution and accumulation of PBDEs.
- Identify temporal trends in PBDE concentrations since the river was last sampled in 2005.

The primary objective of the USGS egg study is to determine if reproductive success of ospreys nesting on the Spokane River is negatively affected by PBDE exposure. Results from the osprey egg portion of the study will be reported by the USGS in 2010.

Methods

Study Design

Ecology collected fish samples from six reaches of the Spokane River from the Idaho border downstream through Long Lake (Figure 3). Fish collection sites were chosen to:

- Provide broad spatial coverage of the river upstream and downstream of the city of Spokane.
- Correspond with suspected feeding locations of osprey nesting along the river.

A secondary consideration for site selection was to match previous Ecology fish collection efforts from 2005. Fish and osprey eggs were also collected from Rock and Williams Lakes southwest of the city of Spokane to serve as reference values for the area. Detailed descriptions of sampling stations are in Appendix B.

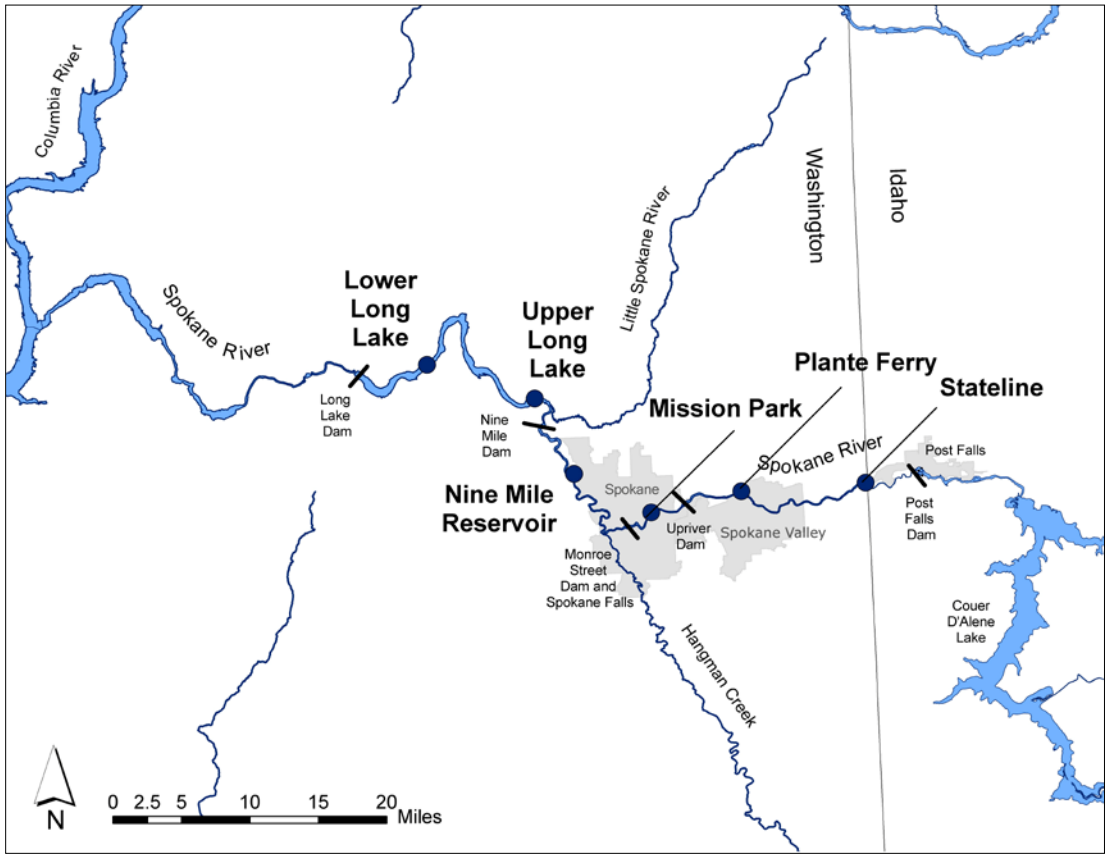

Figure 3. Spokane River Fish Collection Locations.

Species selection at each Spokane River station was guided by the previous PBDE survey and fish suspected to be part of the osprey diet. Ospreys are opportunistic foragers; their diet often consists of 2 to 3 species, regardless of fish communities (Poole et al., 2002). Johnson et al. (2008) found largescale suckers to be the major component of osprey diets along the Columbia and Willamette Rivers representing 84.3% and 92.7% of biomass consumed, respectively. Largescale suckers are abundant in the Spokane River, and three composite samples from each river reach were collected to provide more data on this species.

Sample types consisted of whole body, fillet, or carcass composites. *Carcass samples* are the remainders of the fish after the fillet has been removed for processing. Whole fish PBDE concentrations were calculated from the relative weights of the fillet and carcass samples. Fillet and carcass samples were analyzed for mountain whitefish, rainbow trout, and smallmouth bass. A combination of samples was prepared for these species in order to (1) match sample type (fillet or whole body) from the 2005 study and (2) mimic osprey feeding habits (whole body). All largescale sucker and northern pikeminnow samples were analyzed as whole body.

All fish samples were collected during March and April 2009 to assess contaminant concentrations in the osprey diet prior to the egg laying period. Samples were analyzed for percent lipids and the following 13 PBDE congeners: -47, -49, -66, -71, -99, -100, -138, -153, -154, -183, -184, -191, -209.

The study was conducted according to a Quality Assurance Project Plan (Furl et al., 2009).

Study Area

Spokane River

The Spokane River drainage area covers approximately 6,000 square miles in northeastern Washington and northern Idaho (Figure 4). Boundaries for the drainage area are created by the Bitterroot Mountains to the east, the Selkirk Mountains to the north, and the Columbia Plateau to the south and west (Konrad, 2006). The river originates in northern Idaho where it drains Lake Coeur d'Alene and flows westerly approximately 110 miles before entering the Columbia River 30 miles upstream of Grand Coulee Dam.

Streamflow in the Spokane River is regulated by the Post Falls Dam in Idaho. Typically water is allowed to flow through the Post Falls Dam from December to June, and the gates are set to maintain a specific water level for Lake Coeur d'Alene during the summer months (Hortness and Covert, 2005). Several other dams along the Spokane River restrict upstream movement of fish, resulting in segmented fish populations (Figure 3).

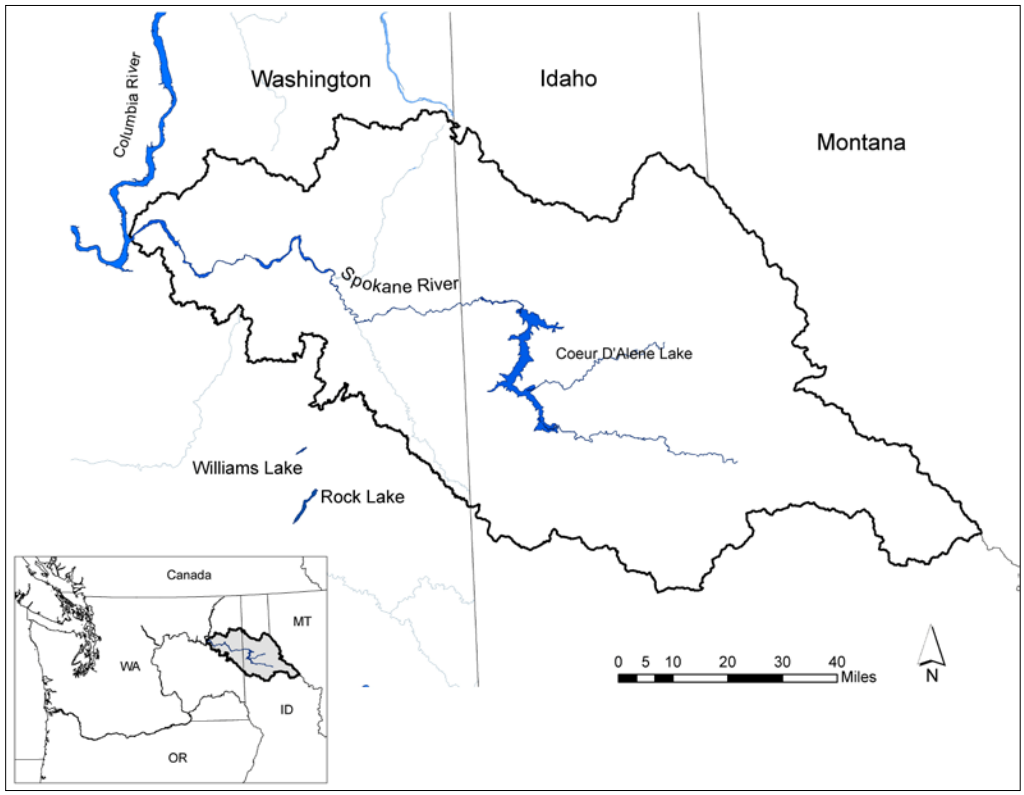

Figure 4. Spokane River Drainage Area.

Reference Sites

Rock and Williams Lakes were chosen to serve as local reference sites for fish tissue and osprey egg concentrations. The lakes are located approximately 30 miles southwest of the city of Spokane, and their drainage basins have experienced little development (Figure 4).

Field Procedures

The collection, handling, and processing of fish tissue samples were guided by methods described in EPA's *Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories* (EPA, 2000) and Ecology's *Standard Operating Procedures for Field Collection, Processing, and Preservation of Finfish Samples at the Time of Collection in the Field* (Sandvik, 2006a). Fish were collected using boat electrofishing during March and April 2009.

Once captured, fish were inspected to ensure that they were acceptable for further processing (e.g., no obvious damage to tissues; skin intact). Acceptable fish were euthanized by a blow to the head with a dull object, rinsed in ambient water to remove foreign material from their exterior, weighed to the nearest gram, and their total lengths measured to the nearest millimeter. Individual fish were double-wrapped in foil and placed in a plastic zip-lock bag along with a sample identification tag. The bagged specimens were placed on ice in the field. Fish remained on ice until frozen (–20°C) at Ecology headquarters.

Laboratory Procedures

Sample Preparation

Fish tissue samples were prepared following adapted guidelines from Ecology's *Standard Operating Procedures for Resecting Finfish Whole Body, Body Parts or Tissue Samples* (Sandvik, 2006b). Fish were removed from the freezer and partially thawed before processing. Slime and scales were removed from specimens being prepared for fillet analysis, with a subset of scales retained for age determination. Fish were then rinsed with tap water followed by a deionized water rinse. Skin-on fillets were removed from the fish in small cubes.

Fillet tissues from individual fish were passed through a Kitchen-Aid food grinder two times. For compositing, equal weights of fillet tissue were mixed together and homogenized a third time using a sonicator. Composite samples consisted of 3 to 5 individual fish. Whole fish and carcass composite samples were prepared the same as fillet composites using a Hobart commercial meat grinder instead of a Kitchen-Aid. The weight of the whole fish, fillet, and carcass were recorded to estimate whole fish concentrations when necessary. Subsamples of the homogenate were placed into laboratory-provided clean glass jars. Samples were assigned a Manchester Environmental Laboratory (MEL) identification number, refrozen, and shipped to the laboratory for analysis. Excess homogenate was labeled and archived at -20° C at Ecology Headquarters.

The sex of the fish was determined after tissue sample processing. Washington Department of Fish and Wildlife biologists determined the age of the fish using otoliths and scales. Table 1 displays species retained from each reach along with sample processing information.

Table 1. Number of Composite Samples Analyzed by Species and Reach.
All samples are whole body except where indicated differently.

Species	Species (Common Name)	Species (Acronym)	State-line	Plante Ferry	Mission Park	Nine Mile Res.	Upper Long Lake	Lower Long Lake	Rock Lake	Williams Lake
Catostomus macrocheilus	Largescale sucker	LSS	3	3	3	3	3	3		
Prosopium williamsoni	Mountain whitefish	MWF			1*	3*	2*			
Ptychocheilus oregonensis	Northern pikeminnow	NPM					3	3		
Micropterus dolomieu	Smallmouth bass	SMB					2*			
Oncorhynchus mykiss	Rainbow trout	RBT		1*					1	1
Salmo trutta	Brown trout	BNT							1	

All sample composites include 3 to 5 fish.
* fillet plus carcass.

Sample Analysis

The tissue samples were extracted by Soxtherm following MEL Standard Operating Procedure (MEL SOP) 730101 (adapted from EPA method SW846 3541). Following extraction, 10% of the sample was split off and used for lipid analysis. The remainder of the sample received micro florisil column cleanup following MEL SOP 730091 and was concentrated to a final volume of 2 mL. The extracts were then treated with acid for further cleanup.

PBDEs were determined by EPA method 8270 for semi-volatile analysis in SIM mode following MEL SOP 730104. Percent lipid was determined gravimetrically according to MEL SOP 730009.

Data Quality

MEL provided case narratives assessing the analytical quality of the data. Case narratives are available upon request. These include summaries of analytical methods, assessment of holding times, instrument tuning, initial and continuing calibrations, method blanks, matrix spikes, laboratory control samples, surrogate spikes, internal standards, duplicate spikes, and standard reference material (SRM).

All 44 samples were run in three analytical batches. Quality control tests along with the rates at which they were conducted are included in Table 2.

Table 2. Quality Control Tests and Measurement Quality Objectives.

Quality Control Test	Measurement Quality Objective	Test Rate
Surrogate Recovery	50-150% recovery	every sample
Laboratory Control Sample	50-150% recovery	1/batch
Method Blank	non-detect	1/batch
Matrix Spike	50-150% recovery	1/batch
Matrix Spike Duplicate	< 40% RPD	1/batch
Duplicate	< 40% RPD	1/batch
Standard Reference Material	no criteria	1/batch

RPD = Relative Percent Difference.

Data quality for PBDE analyses was generally good across the entire project. Below is a description of results outside of the laboratory's guidelines. Complete results for all Quality Control tests are located in Appendix C.

No recovery of surrogate spikes occurred in two samples (#0905034-01 and -02; Stateline LSS 1 and 2) during the original analysis, requiring the samples to be rerun (#0905034-01RE1 and

-02RE1). Quality control tests on the two samples included a method blank, laboratory control sample, surrogate spikes, and SRM. Quality control results for the two sample batch are included in Appendix C.

Instrument tuning, initial calibrations, and continuing calibrations were all met within laboratory guidelines with the exception of a few continuing calibration verifications (CCVs) for PBDE 209. No action was required since PBDE 209 was not detected in any of the samples on days the CCV was outside of laboratory limits.

Low levels of PBDEs 47, 99, and 209 were detected in two of the method blanks (Table 3). Concentrations were considered native to the sample if they were 5 times greater than the sample blank. Results for these congeners less than 5 times blank contamination were qualified as estimates (J).

Table 3. Results (µg/kg) of Laboratory Method Blank Analyses.

Sample Number	PBDE		
	47	99	209
B09E072-BLK1	< MDL	-	12.8 J
B09E075-BLK1	< MDL	< MDL	1.38

Method detection limit = 0.22.
J = Estimated value

Hexabromobiphenyl was spiked into all samples prior to extraction and served as a surrogate for estimating recovery of the target compounds. Recoveries were high (> 150%) in four samples (0905034- 02RE1, 03, 15, 17), and results for those samples were qualified as estimates (J).

Recovery of PBDE 209 was low in both matrix spike #B09E135-MS1 and matrix spike duplicate #B09E135-MSD1. Results for PBDE 209 were already qualified in native sample #0905034-44 based on low continuing calibration response. Recoveries for PBDE 138, 183, 191, and 209 were high in matrix spike #B09E072-MS1 and matrix spike duplicate #B09E072-MSD1. None of the compounds were detected in the native sample.

Data Processing

Summing

Total PBDE values are reported as the sum of detected values for 13 target congeners. Values qualified as estimates (J and NJ) by the laboratory are treated as detected values. Non-detect values (U and UJ) are assigned a value of zero when other congeners making up the sum are detected. If qualified congeners (J and/or NJ) comprise greater than 10% of the total summed concentration, the total concentration is qualified. When all individual congeners are reported as non-detects (U and/or UJ), the highest reporting limit, appropriately qualified, represents the sum.

Whole Fish Calculations

Whole fish results were determined in 8 samples by analyzing a fillet sample and the remaining carcass. A whole body concentration was calculated from the concentrations and relative weights of the sample using the following calculation:

$$C_w = [(C_f * M_f) + (C_r * M_r)] / (M_f + M_r)$$

Where:
C = concentration (µg/Kg ww)
M = weight (Kg)
w = whole fish
f = fillet
r = carcass

Trends Analysis

Changes in largescale sucker and mountain whitefish PBDE concentrations were examined visually by graphing total and lipid-normalized concentrations with those reported by Serdar and Johnson (2006). Samples were paired according to composite length at each location. Changes in concentrations were further assessed by calculating percent change in total and lipid-normalized values by:

$$\%_{\blacktriangle} = [(\text{abs} (C_{2005} - C_{2009})) / C_{2005}] * 100$$

Where:
$\%_{\blacktriangle}$ = percent change
abs = absolute value
C_{2005} = concentration from 2005 study
C_{2009} = concentration from 2009 study

Fish from the current 2009 study were retained within 1 river mile of the previous investigation. In 2005, bridgelip suckers were collected from the Nine Mile reach. Fish were collected for the initial study during the fall of 2005. Time between sample collections for the two studies was approximately 3.5 years.

Results and Discussion

Occurrence of PBDEs in Spokane River Fish

PBDE and lipid results from the Spokane River and the 2 reference lakes are shown in Table 4. Complete ancillary fish data are included in Appendix D.

Table 4. PBDE and Lipid Concentrations in Fish Collected from the Spokane River and Two Reference Waterbodies, 2009.

Spokane River Reach	Collection Date	Sample ID	Species	Tissue	PBDEs (µg/Kg ww)													Lipids (%)	Total PBDEs (µg/Kg ww)	Lipid-normalized Total PBDEs (µg/Kg lipid)
					47	49	66	71	99	100	138	153	154	183	184	191	209			
Stateline	3/4/2009	0905034-01	LSS	Whole	66	0.79 J	1.6 U	1.6 U	1.6 U	10	3.2 U	1.6 J	3.2 J	3.2 U	3.2 U	3.2 U	8 U	6.98	81.6	1169
"	"	0905034-02	LSS	Whole	87 J	1.7 J	1.5 U	1.5 U	1.5 U	13 J	3 U	2.2 J	3.7 J	3 U	3 U	3 U	7.5 U	6.82	107.6 J	1578
"	"	0905034-03	LSS	Whole	120 J	2.1 J	0.88 U	0.88 U	0.88 U	23 J	1.8 U	4 J	6.8 J	1.8 U	1.8 U	1.8 U	15 UJ	4.45	155.9 J	3503
Plante Ferry	3/4/2009	0905034-04	LSS	Whole	57	4.4 U	4.4 U	4.4 U	4.4 U	11	8.8 U	8.8 U	4.6 J	8.8 U	8.8 U	8.8 U	22 U	5.91	72.6	1228
"	"	0905034-05	LSS	Whole	77	4.4 U	4.4 U	4.4 U	4.4 U	14	8.8 U	2.7 J	5.3 J	8.8 U	8.8 U	8.8 U	22 U	6.69	99.0	1480
"	"	0905034-06	LSS	Whole	74	2.2	0.44 U	0.44 U	0.44 U	14	0.88 U	2.5	5.3	0.88 U	0.88 U	0.88 U	21 UJ	4.65	98.0	2108
"	"	0905034-19/20	RBT	Whole*	83	3	2	0.43 UJ	47	17	0.87 UJ	5	4	0.87 UJ	0.87 UJ	0.87 UJ	2.20 UJ	4.58	162	3542
"	"	0905034-19	RBT	Fillet	47	1.7	1.1	0.43 U	28	10	0.87 U	3.1	2.6	0.87 U	0.87 U	0.87 U	2.2 U	2.34	93.5	3996
Mission Park	3/3/2009	0905034-07	LSS	Whole	24	0.44 U	0.44 U	0.44 U	0.44 U	4.4	0.87 U	0.6 J	1.6	0.87 U	0.87 U	0.87 U	12 UJ	4.3	30.6	712
"	"	0905034-08	LSS	Whole	23	1.7	0.44 U	0.44 U	0.44 U	8	0.88 U	0.6 J	2.7	0.88 U	0.88 U	0.88 U	2.2 U	3.23	36.0	1115
"	"	0905034-09	LSS	Whole	26	0.43 U	0.43 U	0.43 U	0.43 U	5.2	0.85 U	0.31 J	1.9	0.85 U	0.85 U	0.85 U	21 UJ	2.77	33.4	1206
"	"	0905034-21/22	MWF	Whole*	139	10	7	0.44 UJ	165	41	0.87 UJ	18	13	0	0.87 UJ	0.87 UJ	2.20 UJ	3.16	392	12420
"	"	0905034-21	MWF	Fillet	90	6.4	4.4	0.43 U	100	28	0.86 U	12	8.5	0.86 U	0.86 U	0.86 U	2.1 U	1.72	249.3	14494
Nine Mile	4/7/2009	0905034-10	LSS	Whole	100	2.3	0.44 U	0.44 U	0.44 U	28	0.87 U	7.5	7.4	0.87 U	0.87 U	0.87 U	20 UJ	4.54	145.2	3198
"	"	0905034-11	LSS	Whole	100	0.52	0.43 U	0.43 U	0.2 J	20	0.85 U	3.7	5.6	0.85 U	0.85 U	0.85 U	2.1 U	3.02	130.0	4305
"	"	0905034-12	LSS	Whole	230	2.6	0.44 U	0.44 U	0.89	88	0.88 U	8.8	18	0.88 U	0.88 U	0.88 U	2.2 U	1.82	348.3	19137
"	"	0905034-26/27	MWF	Whole*	358	10	12	0.43 UJ	287	72	0.86 UJ	19	21	0.61	0.86 UJ	0.86 UJ	2.10 UJ	2.61	780	29893
"	"	0905034-26	MWF	Fillet	180	3.9	5.4	0.44 U	150	36	0.88 U	12	10	0.29 J	0.88 U	0.88 U	2.2 U	1.08	397.6	36814
"	"	0905034-28/29	MWF	Whole*	572	11	17	0.44 UJ	564	144	0.88 UJ	40	34	0.79	0.88 UJ	0.88 UJ	2.20 UJ	2.88	1382	47976
"	"	0905034-28	MWF	Fillet	240	7.1	10	0.44 U	240	74	0.89 U	20	24	0.45 J	0.89 U	0.89 U	2.2 U	1.36	615.6	45261
"	"	0905034-30/31	MWF	Whole*	943	42	44	0.44 UJ	943	368	0.88 UJ	108	81	1.36	0	0.87 UJ	2.20 UJ	3.14	2531	80602
"	"	0905034-30	MWF	Fillet	700	40	40	0.43 U	700	360	0.85 U	100	78	0.75 J	0.85 U	0.85 U	2.1 U	1.66	2018.8	121611
Upper Long Lake	4/7/2009	0905034-13	LSS	Whole	110	3.1	0.43 U	0.43 U	0.43 U	23	0.85 U	1.6	5.1	0.29 J	0.85 U	0.85 U	4.3 UJ	4.11	143.1	3482
"	"	0905034-14	LSS	Whole	200	3.5	0.42 U	0.42 U	0.42 U	38	0.85 U	4	8	0.85 U	0.85 U	0.85 U	9.8 UJ	4.7	253.5	5394
"	"	0905034-15	LSS	Whole	250 J	0.44 U	0.44 U	0.44 U	0.31 J	45 J	0.88 U	6.6 J	12 J	0.88 U	0.88 U	0.88 U	17 UJ	7.1	131.9 J	1858
"	"	0905034-35/36	MWF	Whole*	127	7	4	0.43 UJ	81	26	0.87 UJ	6	7	0.87 UJ	0.87 UJ	0.87 UJ	2.20 UJ	0.68	258	37991
"	"	0905034-35	MWF	Fillet	72	4.6	2.4	0.41 U	47	15	0.81 U	4.1	4.7	0.81 U	0.81 U	0.81 U	2 U	0.28	149.8	53500
"	"	0905034-37/38	MWF	Whole*	156	8	3	0.40 UJ	85	41	0.80 UJ	9	12	0.80 UJ	0.80 UJ	0.80 UJ	4.20 UJ	3.61	315	8739
"	"	0905034-37	MWF	Fillet	99	4.8	2.2	0.4 U	53	25	0.8 U	5.2	7	0.8 U	0.8 U	0.8 U	2 UJ	0.14	196.2	140143
"	"	0905034-39	NPM	Whole	110	6.5	0.39 U	0.39 U	0.39 U	30	0.78 U	3.6	8.8	0.78 U	0.78 U	0.78 U	2.3 UJ	4.56	158.9	3485
"	"	0905034-40	NPM	Whole	160	9.6	0.4 U	0.4 U	0.4 U	36	0.79 U	2.6	8.3	0.79 U	0.79 U	0.79 U	2 UJ	3.18	216.5	6808
"	"	0905034-41	NPM	Whole	100	8.6	0.4 U	0.4 U	0.4 U	29	0.79 U	0.34 J	7.1	0.79 U	0.79 U	0.79 U	4 UJ	2.44	145.0	5944
"	"	0905034-32/33	SMB	Whole*	114	10	2	0.44 UJ	32	19	0.89 UJ	5	5	0.89 UJ	0.89 UJ	0.89 UJ	2.20 UJ	4.34	187	4298
"	"	0905034-32	SMB	Fillet	46	4.1	0.81	0.43 U	13	8.3	0.87 U	1.8	1.9	0.87 U	0.87 U	0.87 U	2.2 U	1.64	75.9	4629
"	"	0905034-34	SMB	Fillet	87	3.8	1.1	0.44 U	58	8.7	0.88 U	2	2	0.88 U	0.88 U	0.88 U	2.2 U	1.45	162.6	11214
Lower Long Lake	4/8/2009	0905034-16	LSS	Whole	96	5.6	0.44 U	0.44 U	0.44 U	17	0.87 U	2.4	3.7	0.87 U	0.87 U	0.87 U	4.8 UJ	5.75	124.7	2169
"	"	0905034-17	LSS	Whole	270 J	8 J	0.44 U	0.44 U	0.44 U	31 J	0.88 U	2.8 J	8.1 J	0.88 U	0.88 U	0.88 U	9.7 UJ	4.7	319.9 J	6806
"	"	0905034-18	LSS	Whole	130	5	0.43 U	0.43 U	0.43 U	22	0.86 U	1.6	4.2	0.86 U	0.86 U	0.86 U	2.1 U	4.24	162.8	3840
"	"	0905034-42	NPM	Whole	150	11	0.39 U	0.39 U	0.39 U	47	0.79 U	4	13	0.79 U	0.79 U	0.79 U	2 UJ	5.48	225.0	4106
"	"	0905034-43	NPM	Whole	70	5.3	0.4 U	0.4 U	0.4 U	17	0.8 U	1.1	4.6	0.8 U	0.8 U	0.8 U	4.8 UJ	4.21	98.0	2328
"	"	0905034-44	NPM	Whole	59	6.2	0.4 U	0.4 U	0.25 J	19	0.8 U	1.7	4.8	0.8 U	0.8 U	0.8 U	4 UJ	1.73	91.0	5257

Reference Waterbody	Collection Date	Sample ID	Species	Tissue	PBDEs (µg/Kg ww)													Lipids (%)	Total PBDEs (µg/Kg ww)	Lipid-normalized Total PBDEs (µg/Kg lipid)
					47	49	66	71	99	100	138	153	154	183	184	191	209			
Rock Lake	4/6/2009	0905034-24	BNT	Whole	0.29 J	0.43 U	0.43 U	0.43 U	0.43 U	0.43 U	0.87 U	0.87 U	0.87 U	0.87 U	0.87 U	0.87 U	2.2 U	0.65	0.3	45
Rock Lake	4/6/2009	0905034-23	RBT	Whole	0.43 U	0.43 U	0.43 U	0.43 U	0.43 U	0.43 U	0.85 U	0.85 U	0.85 U	0.85 U	0.85 U	0.85 U	2.1 U	2.21	2.1 U	95
Williams Lake	4/8/2009	0905034-25	RBT	Whole	48	4.1	2	0.43 U	89	19	0.86 U	14	10	0.19 J	0.86 U	0.86 U	2.1 U	1.44	186.3	12937

U = Analyte not detected at or above reported quantitation limit.
UJ = Analyte not detected at or above reported quantitation limit. Quantitation limit is approximate.
J = Estimated value.
* = Whole body value calculated using carcass + fillet.

Total PBDE concentrations in whole fish composites from the Spokane River ranged from 30.6 – 2,531 µg/Kg wet weight (ww). Figure 5 presents a boxplot of PBDE concentrations by species. Table 5 provides a statistical summary of individual congeners from the three most prevalent species: northern pikeminnow, mountain whitefish, and largescale suckers. For summary statistics, the reporting limit was used to calculate values when non-detects were present.

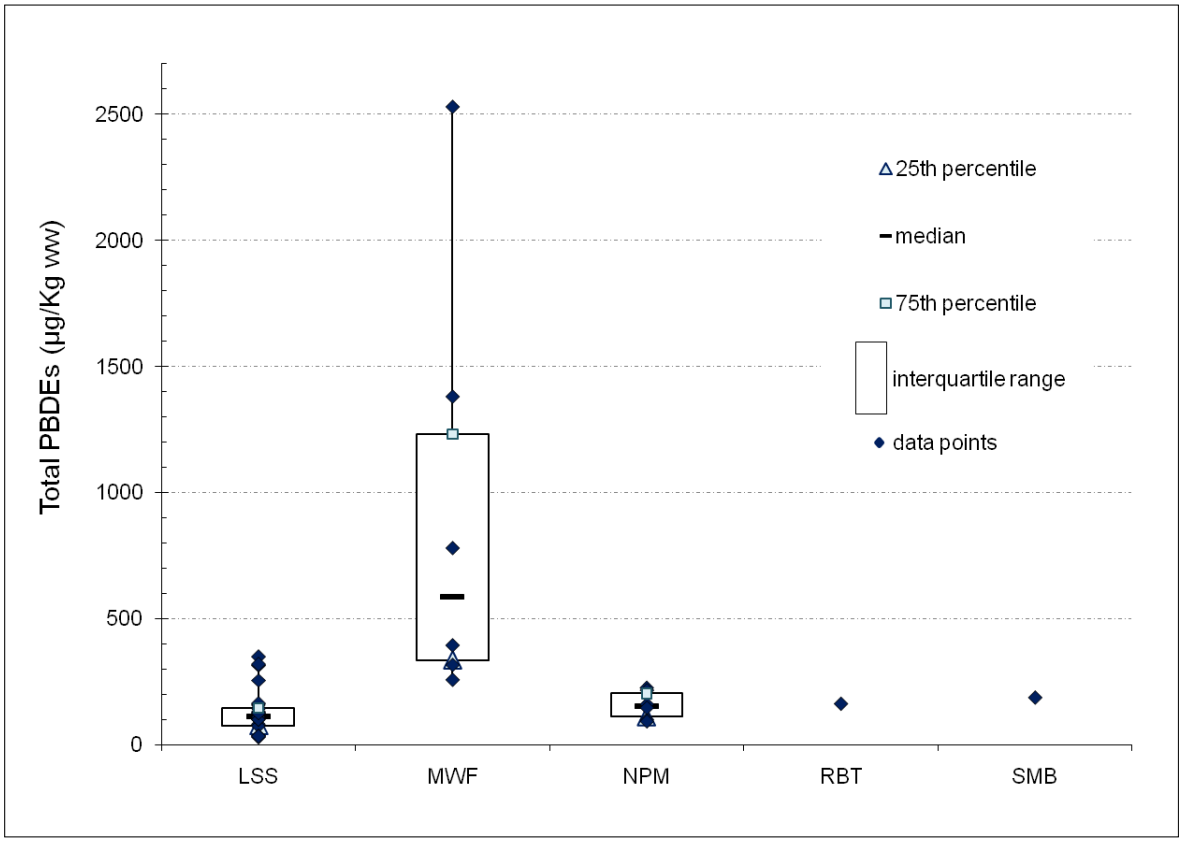

Figure 5. Boxplot of Total PBDE Concentrations by Fish Species.

Table 5. Statistical Summary of PBDE Concentrations (µg/Kg ww) in Northern Pikeminnow, Mountain Whitefish, and Largescale Sucker Composites Collected from the Spokane River in Spring 2009.

NPM (n = 6)					
PBDE	Detection Frequency (%)	Minimum	Median	Mean	Maximum
47	100	59.0	105.0	108.2	160.0
49	100	5.3	7.6	7.9	11.0
66	0	< 0.39	---	---	< 0.40
71	0	< 0.39	---	---	< 0.40
99	17	0.3	< 0.40	< 0.37	< 0.40
100	100	17.0	29.5	29.7	47.0
138	0	< 0.78	---	---	< 0.80
153	100	0.3	2.2	2.2	4.0
154	100	4.6	7.7	7.8	13.0
183	0	< 0.78	---	---	< 0.80
184	0	< 0.78	---	---	< 0.80
191	0	< 0.78	---	---	< 0.80
209	0	< 2	---	---	< 4.8

MWF (n = 6)					
PBDE	Detection Frequency (%)	Minimum	Median	Mean	Maximum
47	100	127.0	257.2	382.4	942.6
49	100	7.3	9.9	14.8	42.4
66	100	3.5	9.4	14.5	44.0
71	0	< 0.40	---	---	< 0.44
99	100	81.0	226.1	354.1	942.6
100	100	26.3	56.8	115.4	368.1
138	0	< 0.80	---	---	< 0.88
153	100	6.3	18.5	33.4	108.1
154	100	7.0	17.1	28.0	81.2
183	67	0.3	< 0.79	< 0.79	1.4
184	17	0.4	< 0.87	< 0.78	< 0.88
191	0	< 0.80	---	---	< 0.88
209	0	< 2.1	---	---	< 4.2

LSS (n = 18)					
PBDE	Detection Frequency (%)	Minimum	Median	Mean	Maximum
47	100	87.0	185.0	182.0	270.0
49	72	< 0.43	< 1.7	< 2.5	8.0
66	0	< 0.42	-	-	< 4.4
71	0	< 0.42	-	-	< 4.4
99	17	0.2	< 0.44	< 1.0	< 4.4
100	100	13.0	27.0	28.0	45.0
138	0	< 0.85	-	-	< 8.8
153	94	0.3	< 2.45	< 3.02	8.8
154	100	3.2	5.3	6.2	12.0
183	6	0.3	< 0.88	< 2.02	< 8.8
184	0	< 0.85	-	-	<8.8
191	0	< 0.85	-	-	<8.8
209	0	< 2.1	-	-	< 22

On average, PBDE 47 was the largest contributor to total concentrations across all species. Figure 6 displays the average percent contribution of each individual congener to the total PBDE sum (whole fish) for each of the 5 species encountered.

Largescale suckers and northern pikeminnow contained larger percentages of PBDE 47 than other species and accumulated very little PBDE 99. PBDE 100 represented the second largest contributor to the total sum in suckers and northern pikeminnow. PBDE 47 and 100 together accounted for 88% and 92% of total PBDEs in northern pikeminnow and largescale suckers, respectively.

Congener patterns in mountain whitefish, rainbow trout, and smallmouth bass whole body samples were similar to suckers and northern pikeminnow with the exception of significant accumulation of PBDE 99. PBDEs 47, 99, and 100 accounted for greater than 88% of total PBDEs in those species following the pattern of 47>99>100.

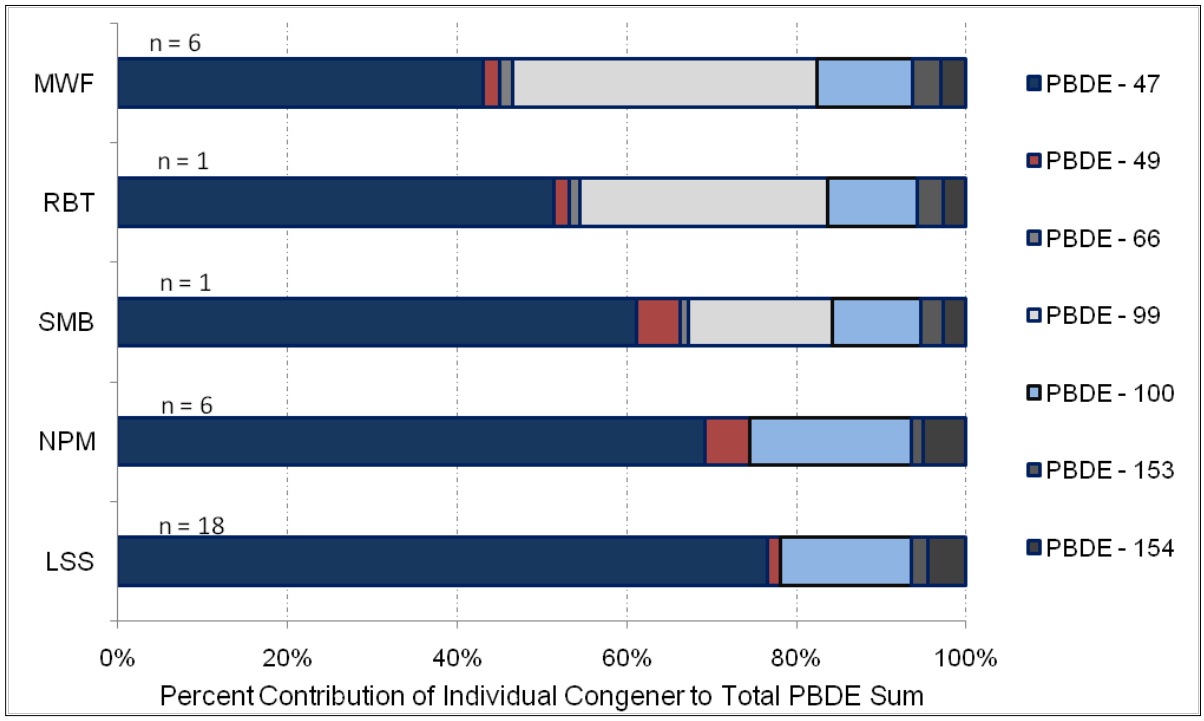

Figure 6. Mean Percent Contribution of Individual PBDE Congeners to Total PBDE Sums Measured in Whole Fish Collected from the Spokane River, 2009.

PBDEs in Fillet vs. Whole Fish

Fillet and carcass tissues were analyzed from several sportfish samples (Table 4). Whole fish concentrations were calculated using their relative weights and concentrations. A combination of sample types was used to (1) match sample type from the 2005 Spokane River PBDE study and (2) mimic osprey feeding habits. Fillet PBDE concentrations were less than whole fish in all cases. Whole fish concentrations were approximately 25 – 150% higher than fillets. Results are located in Table 6.

Table 6. PBDE Concentrations (µg/Kg ww) in Rainbow Trout, Mountain Whitefish, and Smallmouth Bass Fillet and Whole Fish Composites.

Reach	Species	Fillet ID	Fillet Concentration	Whole Fish Concentration	Percentage Increase over Fillet Concentration
Plante Ferry	RBT	0905034-19	93.5	162	73.3%
Mission Park	MWF	0905034-21	249.3	392	57.2%
Nine Mile	MWF	0905034-26	397.6	780	96.2%
		0905034-28	615.6	1382	124.5%
		0905034-30	2018.8	2531	25.4%
Upper Long Lake	SMB	0905034-32	75.9	187	146.4%
	MWF	0905034-35	149.8	258	72.2%
		0905034-37	196.2	315	60.6%

Correlation of Total PBDEs with Fish Characteristics

Pearson correlations were computed to examine relationships between whole fish wet weight PBDE concentrations and fish physical characteristics (Table 7). Correlations were calculated at each reach where 3 composites of a single species were retained. Simple linear regressions are displayed in Appendix E.

Length, weight, and age generally displayed a strong positive relationship with PBDEs at all reaches. Positive correlations between PBDEs and fish age and size have been documented in Washington and elsewhere (Johnson et al., 2006; Hale et al., 2001; Loganathan et al., 1995). Northern pikeminnow from Upper and Lower Long Lake were the only samples where size and age were negatively correlated with PBDEs.

Percent lipids displayed a wide ranging relationship with PBDE concentrations. In largescale suckers, the correlation ranged from -0.957 to 0.846 with an average correlation coefficient from the six reaches of -0.345. Strong positive relationships were found in sportfish at Nine Mile and Lower Long Lake. Inconsistent relationships between lipids and PBDEs were not expected due to their lipophilic properties. Johnson et al. (2006) noted a positive relationship between PBDEs and lipids in multiple Washington State waterbodies.

Table 7. Correlations between Total PBDEs (µg/Kg ww) in Whole Fish and Fish Physical Characteristics.

Reach	Species	Correlation Coefficient (r)			
		Percent Lipids	Length	Weight	Age
Stateline	LSS	-0.957	1	0.948	0.988
Plante Ferry	LSS	-0.101	0.712	0.756	0.723
Mission Park	LSS	-0.699	0.527	0.496	0.954
Nine Mile	LSS	-0.794	0.734	0.869	0.876
Upper Long Lake	LSS	0.876	0.999	1	0.822
Lower Long Lake	LSS	-0.396	0.319	0.285	0.634
Average:	LSS	-0.345	0.715	0.726	0.833

Reach	Species	Correlation Coefficient (r)			
		Percent Lipids	Length	Weight	Age
Nine Mile	MWF	0.982	0.928	0.908	0.933
Upper Long Lake	NPM	0.011	-0.143	-0.148	-0.002
Lower Long Lake	NPM	0.789	-0.906	-0.901	-0.713

PBDE Concentrations in Statewide and Reference Waterbodies

As anticipated, PBDE concentrations in Spokane River fish are highly elevated over other freshwater areas across the state. Figure 7 displays a cumulative frequency graph of PBDE concentrations in selected freshwater fish fillets from other areas in Washington (n = 309). Data collected from the current 2009 study are graphed along with the statewide concentrations. Fillet concentrations are used where available (sportfish) for the Spokane River in order to match the statewide data pool.

Fish tissue samples were also collected from Rock and Williams Lakes to serve as reference values for the area. Rock Lake PBDE concentrations in rainbow and brown trout whole body samples were low (2.1 U and 0.3 µg/Kg ww, respectively).

A single whole body rainbow trout sample from Williams Lake contained a concentration previously unseen in an undeveloped area (186.3 µg/Kg ww). The tissue sample was re-extracted and reanalyzed, confirming the original analysis. Aging of the trout indicated they were hatchery fish (personal communication, Lucinda Williams WDFW). Over 10,000 catchable rainbow trout were planted in Williams Lake within 2 weeks of fish collections. Fish were collected from the lake after stocking due to local knowledge indicating osprey nesting on the lake fed primarily on hatchery rainbow trout.

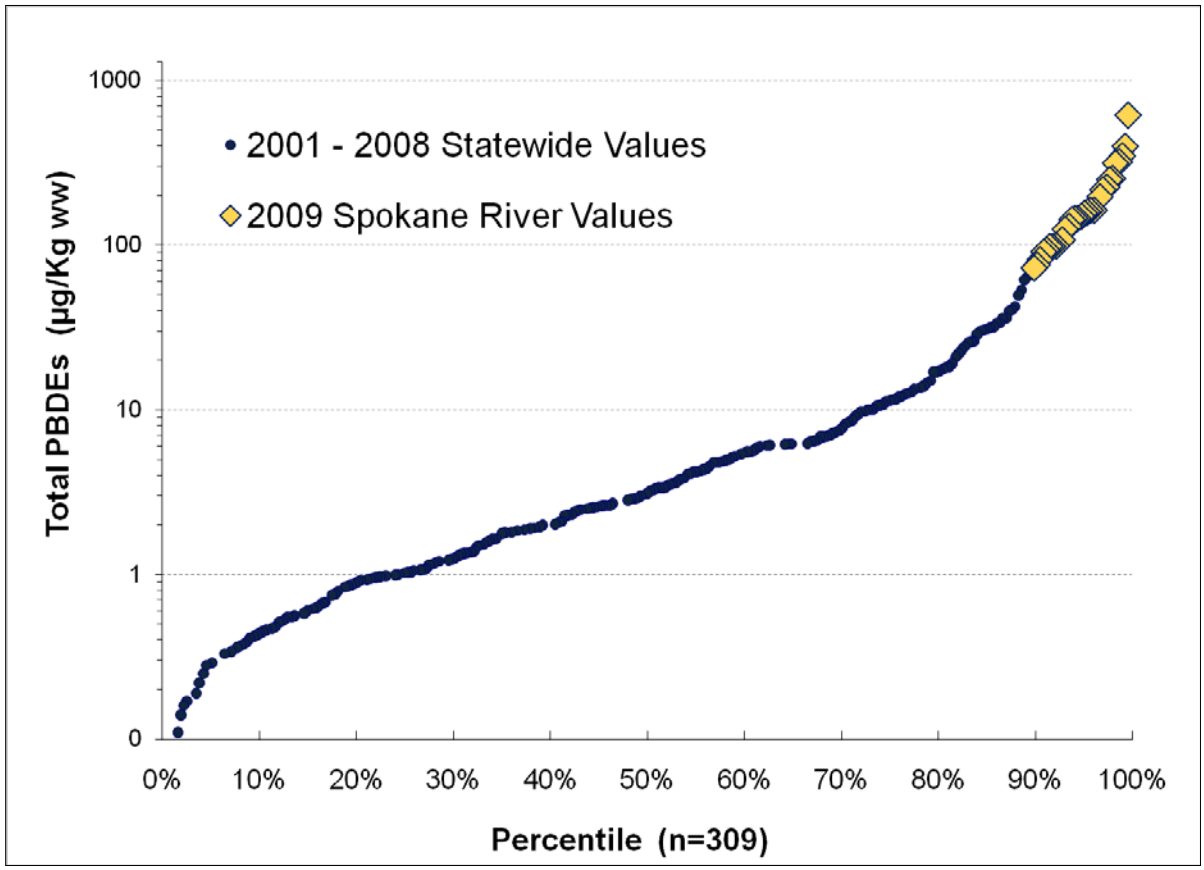

Figure 7. Cumulative Frequency of Fish Tissue PBDE Values in Statewide Waterbodies (2001-2008) and the Spokane River (2009).

Causes for the elevated concentrations in Williams Lake hatchery rainbow trout are unknown. Serdar et al. (2006) examined PBDE concentrations in hatchery rainbow trout and their feed at 10 hatcheries around the state. Concentrations in catchable-sized rainbow trout collected from the hatcheries ranged from 0.24 – 1.10 J µg/Kg ww, consistent with values for Rock Lake. PBDE burdens from hatchery environments are not applicable to Spokane River trout. Trout captured from the Spokane River were native redband rainbow trout (*Oncorhynchus mykiss gairdneri*).

PBDE Trends in the Spokane River

Spatial Distribution

The 6 sampling locations along the Spokane River cover approximately 60 river miles. Within this stretch, 5 hydroelectric dams (Upriver Dam RM 79.9, Monroe Street Dam/Upper Falls Dam RM 73.4, Nine Mile Dam RM 57.6, and Lake Spokane Dam RM 33.9) exist, resulting in segmented fish populations (Figure 3). Fish movement between reaches is obstructed with the exceptions of Stateline/Plante Ferry and Upper/Lower Long Lake. These stations are separated by > 10 miles.

Figures 8 and 9 display mean total PBDE concentrations in whole body samples at each reach for largescale suckers and sportfish, respectively.

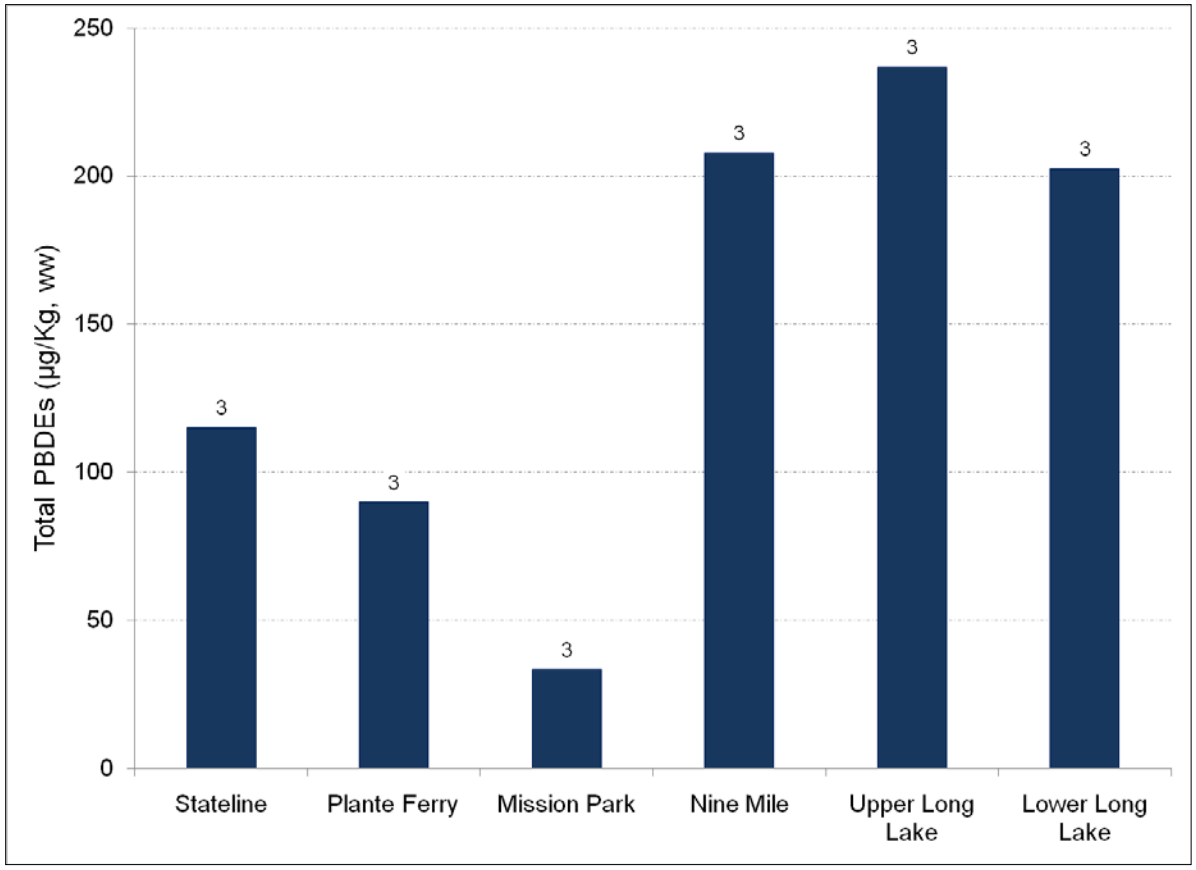

Figure 8. Mean Total PBDE Concentrations in Spokane River Largescale Suckers (whole body) Collected in 2009.

Number of composite samples included in average concentrations is indicated above bars.

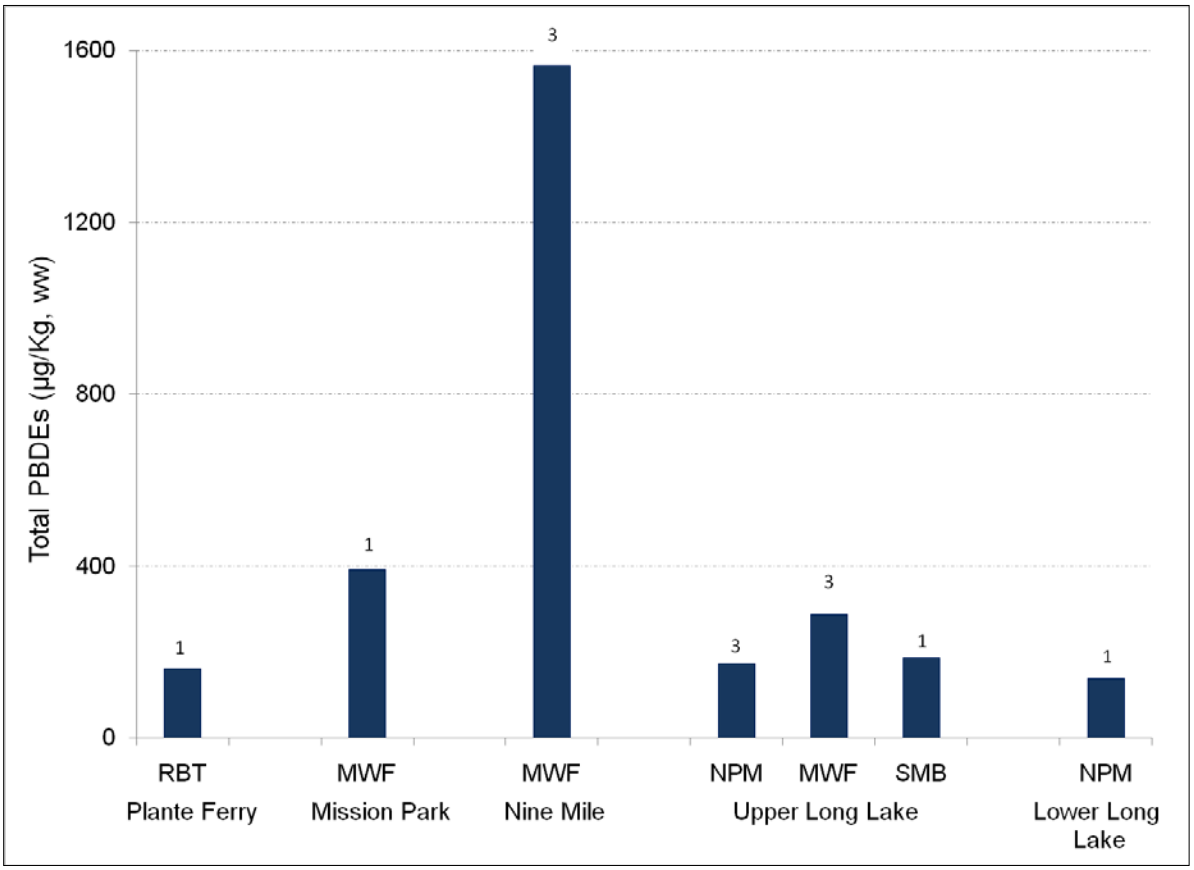

Figure 9. Mean Total PBDE Concentrations in Spokane River Sportfish (whole body) Collected in 2009.

Number of composite samples included in average concentrations is indicated above bars

The highest PBDE concentrations in the study were found in the three downstream reaches of the river, particularly the Nine Mile stretch. Concentrations in largescale suckers display a decreasing trend from the border through Mission Park and are elevated from Nine Mile through Lower Long Lake. The spatial trend in largescale suckers closely matches findings by Serdar and Johnson (2006).

The same broad spatial pattern could not be confirmed in sportfish due to species changes among reaches. However, the highest sportfish concentrations (mountain whitefish) in the study were again from the Nine Mile reach.

The Riverside Park Water Reclamation Facility (WWTP) discharges effluent three river miles upstream of the Nine Mile sampling station. While the causes of the fish contamination are unknown, the large disparity between the Mission Park and Nine Mile stations indicates a new source of PBDEs between the sample stations. WWTPs have been shown to be sources of PBDEs to aquatic environments (Song et al., 2006). In addition to the WWTP, Latah (Hangman) Creek enters the Spokane River between the sample stations. The Latah Creek watershed is largely rural and can deliver substantial sediment loads to the river.

Temporal Trends

Changes in PBDE concentrations over time were assessed by comparing data from the 2005 Spokane River study (Serdar and Johnson, 2006) and the current 2009 survey.

Largescale Suckers

Largescale suckers were the most abundant species encountered along the Spokane River and provide the best data set to examine temporal trends. A total of 18 whole body composites were analyzed from 6 reaches in both the 2005 study and the 2009 study. Samples from the two studies were paired based on sample length at each reach. Relative percent difference (RPD) between composite lengths was less than 5% in all cases except the largest sucker composite from the Nine Mile station (13.7%).

Figures 10 and 11 present wet weight and lipid-normalized PBDE concentrations, respectively, in paired sucker samples from both studies.

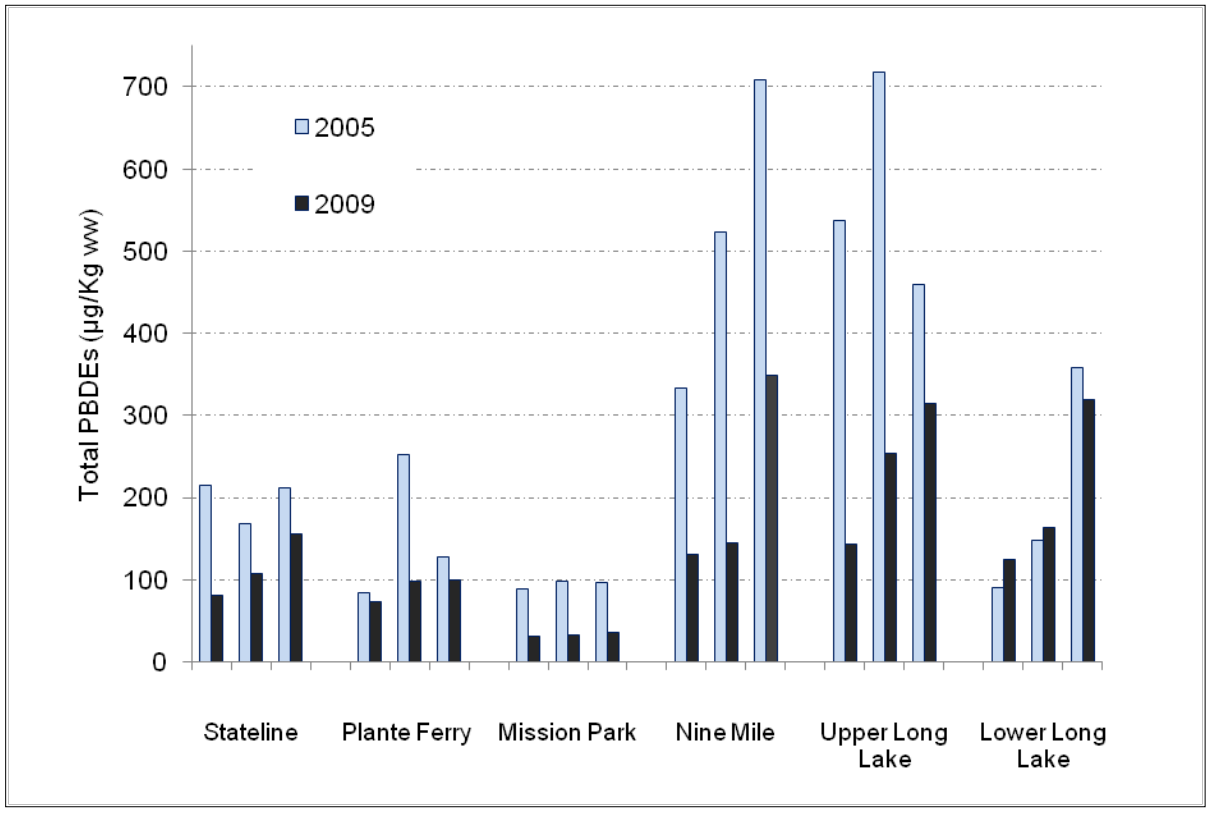

Figure 10. Total PBDE Concentrations in Largescale Suckers (whole body) Collected from the Spokane River in 2005 and 2009.

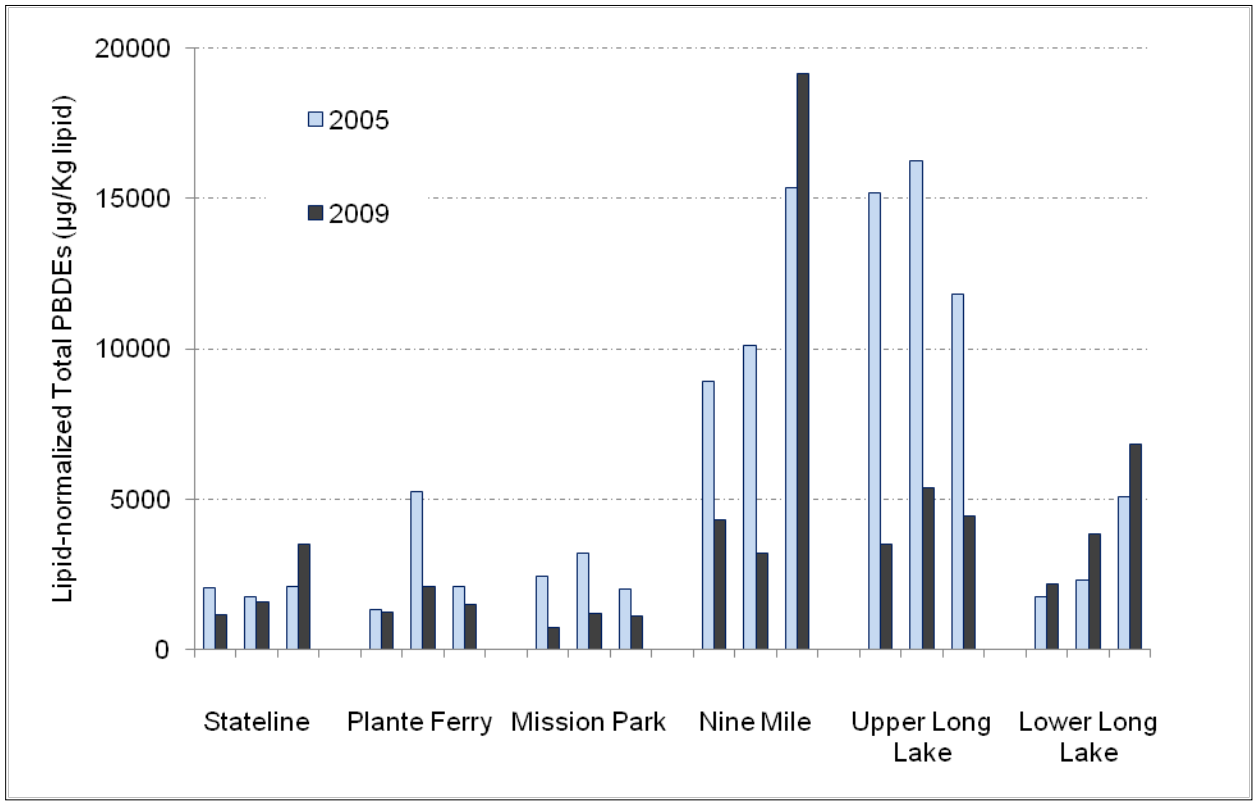

Figure 11. Total Lipid-normalized PBDE Concentrations in Largescale Suckers (whole body) Collected from the Spokane River in 2005 and 2009.

Wet weight PBDE concentrations were lower in all 2009 samples except the two smallest (length) composites from Lower Long Lake. The general decrease in concentrations could not be reconciled by changes in fish size or lipid concentrations alone. The RPD in lengths of the paired samples was low (average 2%). Average percent lipids in suckers from the fall 2005 collection effort was 5.71%, compared to a 4.77% average during the spring 2009 collection. Lipid-normalized concentrations were lower in 13 of the 18 paired samples, reducing the strength of the declining trend.

Figure 12 presents percent change in total and lipid-normalized concentrations for paired largescale sucker samples at each reach.

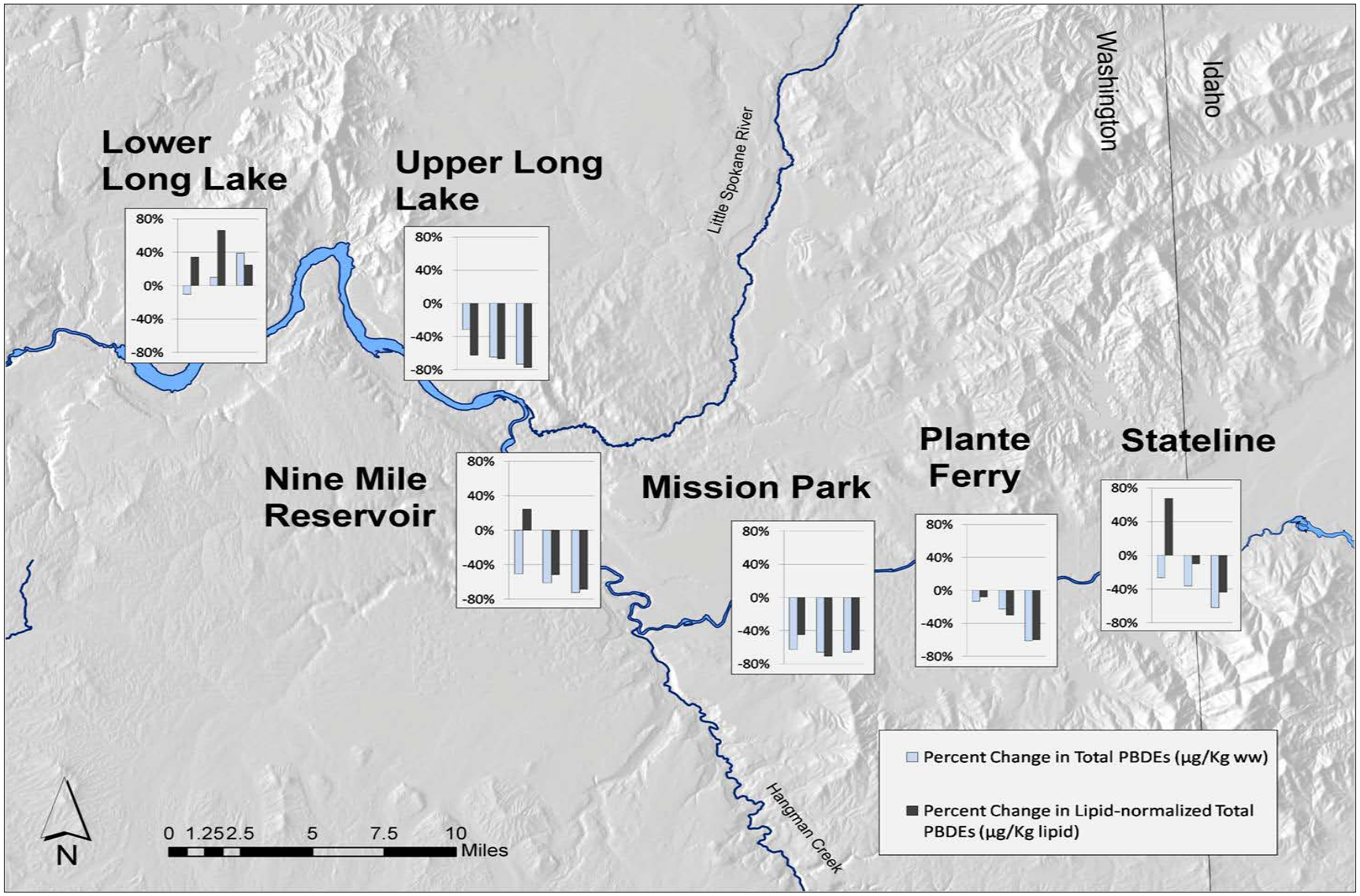

Figure 12. Percent Change in Total PBDEs and Lipid-normalized Total PBDEs in Suckers at Six Spokane River Reaches between 2005 and 2009.

On average, wet weight PBDEs in paired samples were approximately 40% lower in the 2009 samples. Lipid-normalized concentrations decreased, on average, 24% in the paired samples. Lower Long Lake was the only reach that did not display consistent declines in total and lipid-normalized concentrations. Concentrations at this reach displayed a 22% and 42% average increase in wet weight and lipid-normalized concentrations, respectively.

Conflicting trend directions were observed between wet weight and lipid-normalized concentrations in single composites from Lower Long Lake, Nine Mile, and Stateline. Percent lipids in these composites were lower than the group mean in all three instances, resulting in elevated normalized concentrations.

Mountain Whitefish

Changes in PBDE concentrations were also examined in 5 mountain whitefish fillet composites from 3 reaches. Figures 13 and 14 present wet weight and lipid-normalized concentrations from both study years.

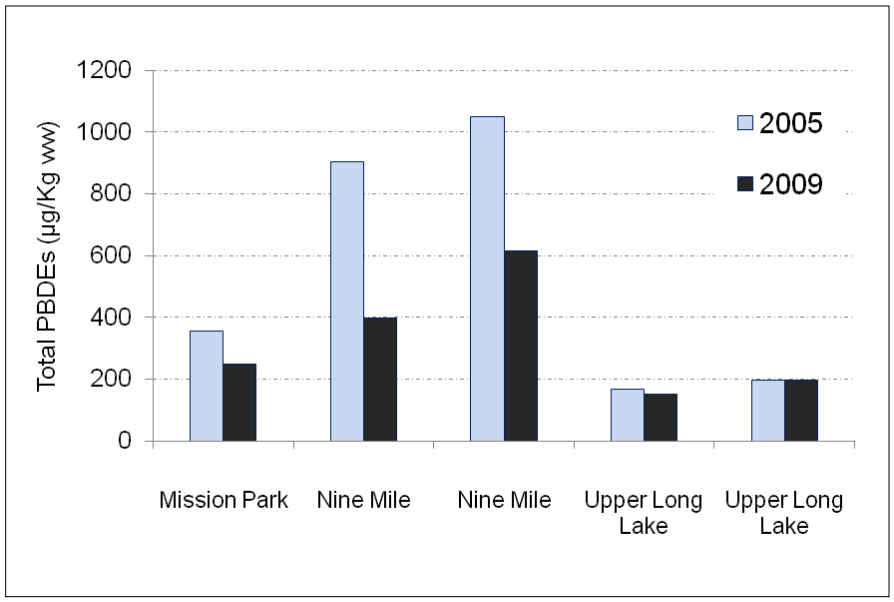

Figure 13. Total PBDE Concentrations in Mountain Whitefish (fillet) Collected from the Spokane River in 2005 and 2009.

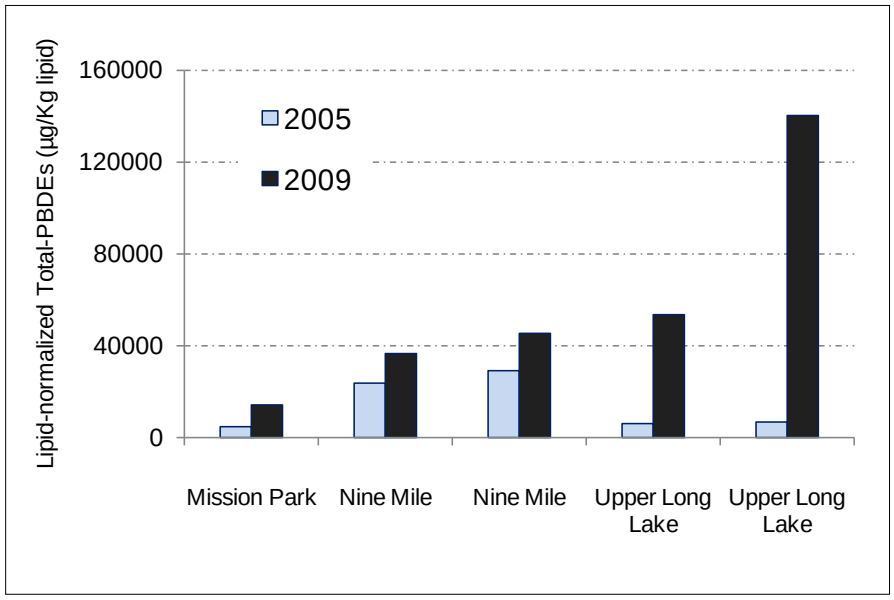

Figure 14. Total Lipid-normalized PBDE Concentrations in Mountain Whitefish (fillet) Collected from the Spokane River in 2005 and 2009.

Wet weight values indicated a decreasing trend while lipid-normalized values show PBDE concentrations increasing. Changes in fillet concentrations appeared to be strongly affected by percent lipids. Fillet samples taken during the current 2009 survey were severely depleted of lipids when compared to the 2005 study.

Reduced PBDE Tissue Concentrations

Large decreases in concentrations for both species were not expected over such a short time period. Studies assessing biotic trends in PBDE concentrations have overwhelmingly reported rapidly increasing rates (Ikonomou et al., 2002; Lebeuf et al., 2004; Norstrom et al., 2002; Rayne et al., 2003). Gauthier et al. (2008) described a leveling trend in PBDE 47, 99, and 100 since 2000 in herring gull eggs. Decreasing trends (1998-2004) in Lake Ontario lake trout were recently reported by Ismail et al. (2009).

Causes behind the large change in fish tissue concentrations are unknown. Seasonal variation due to changing hydrology and fish physiology may be partly responsible for the declines in wet weight concentrations. On average, percent lipids were lower in both species during the March to April 2009 collection effort. Depleted energy stores at the end of winter have been documented in several fish species (Biro et al., 2004; Cunjak and Power, 1986). Studies examining PBDE elimination under increased lipid metabolism are lacking.

Evidence exists suggesting changing river flows have a large effect on PBDE concentrations in the Spokane River water column. Dissolved PBDE concentrations have been monitored intermittently since 2005 at the Nine Mile Dam, using semi-permeable membrane devices (SPMDs) (Johnson et al., 2006; Sandvik, 2009; Sandvik in prep.) The devices are deployed for approximately 1 month during high and low river flows and can be used to estimate water column concentrations. During fall 2005 and spring 2006, estimated dissolved PBDE water

column concentrations were approximately 6 times greater during the fall (926 and 146 pg/L fall and spring, respectively) (Johnson et al., 2006). The large change in water column concentrations were interpreted as a dilution of local sources in the spring from increased flow due to snowmelt in the upper watershed. SPMD monitoring during 2008 and 2009 showed a similar relationship between season and dissolved water concentrations at Nine Mile Dam (personal communication, Patti Sandvik, ECY).

Tissue collections for the two studies occurred during different seasons and dissimilar river flows. The majority of the 2005 collection effort occurred during August and September. During those two months, flows were low, ranging from 600 – 1900 cfs at the Mission Park site. The 2009 sample collection took place during March and April when flows at the same gage were greater than 5,000 cfs (Figure 15). The effect season and river flow have on tissue PBDE concentration is unknown.

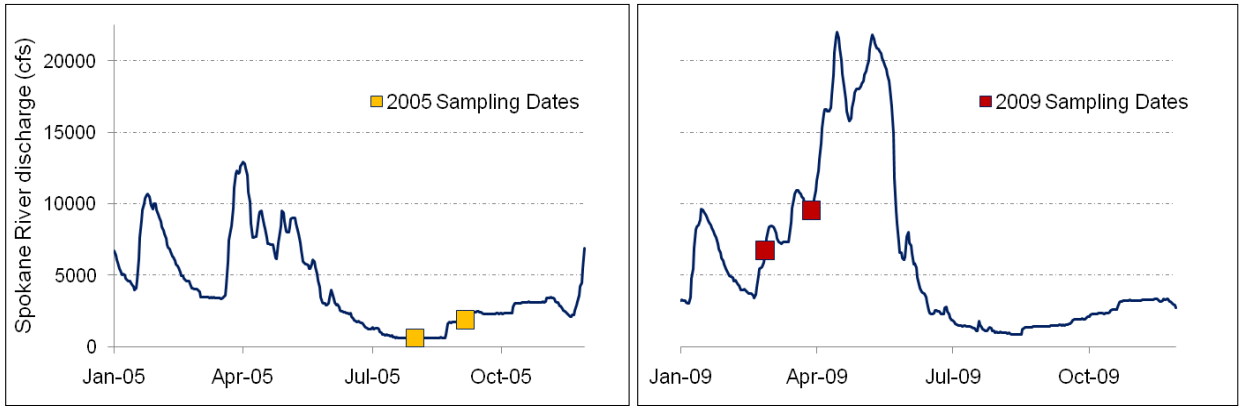

Figure 15. Spokane River Flows during 2005 and 2009 Sampling.

Conclusions

Results from this 2009 study present one part of a joint effort between Ecology and USGS to examine PBDE concentrations in osprey eggs along the Spokane River. During this study, Ecology measured PBDE concentrations in sportfish and largescale suckers from the Spokane River in order to characterize contaminant concentrations in the diet of osprey nesting along the river. Results from the osprey egg portion of the study will be reported by USGS in 2010.

Ecology sampled 6 river reaches from the Idaho border through Long Lake. Total PBDE concentrations in whole fish composites ranged from 30.6 – 2,531 µg/Kg wet weight. PBDE concentrations were strongly correlated with fish size and age within a specific reach. Percent lipids had a highly variable correlation with PBDE concentrations in whole body largescale sucker samples. Concentrations were generally higher in sportfish than largescale suckers.

The highest PBDE concentrations were found in the lower three stretches beginning at Nine Mile. PBDE levels in largescale sucker composites from Nine Mile were approximately 6 times greater than those from Mission Park, the nearest upstream reach. The large change in concentrations between the two reaches was also noted by Serdar and Johnson (2006) during a 2005 survey. The data suggest major PBDE sources are located in the city of Spokane.

Temporal trends in PBDEs were examined by comparing mountain whitefish and largescale sucker concentrations to those recorded in the 2005 survey. Samples were paired between the two studies according to location and fish composite length. Wet weight concentrations from the 2009 survey were lower in 21 of 23 paired samples, often by greater than 50 percent. The apparent declines in PBDE levels may be associated with seasonal differences in fish physiology and river hydrology during fall and spring sampling events. Despite the lower PBDE levels recorded in 2009, concentrations in Spokane River fish are highly elevated compared to other areas of Washington State.

Recommendations

As a result of this 2009 study, the following recommendations are made:

- Conduct an investigation to determine PBDE sources to the Spokane River in Idaho and Washington. Source identification should include PBDE analysis of effluent discharged from the Riverside Park Water Reclamation Facility above the Nine Mile stretch.

- Continue to use largescale suckers in fish PBDE monitoring investigations for the Spokane River. The species are abundant throughout the river and represent the largest historical data set for any species. Fish should be collected during the fall low-flow period to determine whether declining contaminant concentrations are the result of environmental conditions.

- Continue SPMD monitoring for PBDEs at the Nine Mile station. Consider adding additional stations upstream.

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Appendices

Appendix A. Glossary, Acronyms, and Abbreviations

Glossary

Bioaccumulation: Progressive increase in the amount of a substance in an organism or part of an organism which occurs because the rate of intake exceeds the organism's ability to remove the substance from the body.

Boxplot: A graphical depiction of a data set showing the 25th percentile, 50th percentile or median, the 75th percentile, range of data, and outliers.

Congener: In chemistry, congeners are related chemicals. For example, polychlorinated biphenyls (PCBs) are a group of 209 related chemicals that are called congeners.

Contaminant: Any physical, chemical, biological, or radiological substance or matter that has an adverse effect on air, water, or soil.

Hydrophobic: Describing the character of a molecule or atomic group which is insoluble in water, or resistant to wetting or hydration.

Lipophilic: Having an affinity for, tending to combine with, or capable of dissolving in lipids.

Octanol-water partition coefficient (K_{OW}): Ratio of the concentration of a chemical in octanol and in water at equilibrium and at a specified temperature. The ratio is often used to help predict the extent a contaminant will bioaccumulate in fish.

PBDE flame retardant: A group of organohalogen chemicals added to consumer products so the products will not catch on fire or will burn more slowly if exposed to flame or high heat.

Persistent, bioaccumulative, toxic substance (PBT): A distinct group of chemicals that threaten the health of people and the environment. They 1) remain in the environment for a long time without breaking down (persist), 2) are accumulated by animals and humans and increase in concentration up the food chain (bioaccumulate), and 3) are linked to toxic effects in fish, wildlife, and humans (toxic).

Reach: A specific portion or segment of a stream.

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

The following are acronyms and abbreviations used frequently in this report.

BNT	Brown trout
Ecology	Washington State Department of Ecology
EIM	Environmental Information Management database
EPA	U.S. Environmental Protection Agency
LCS	Laboratory control sample
LSS	Largescale sucker
MEL	Manchester Environmental Laboratory
MWF	Mountain whitefish
NAD	North American Datum
NPM	Northern pikeminnow
PBDE	Polybrominated Diphenyl Ethers
PBT	Persistent, Bioaccumulative, and Toxic substance
RBT	Rainbow trout
RCW	Revised Code of Washington
RM	River Mile
RPD	Relative Percent Difference
SMB	Smallmouth bass
SOP	Standard Operating Procedures
SRM	Standard Reference Materials
USGS	U.S. Geological Survey
UV	Ultraviolet
WRIA	Water Resources Inventory Area
WWTP	Wastewater Treatment Plant

Units of Measurement

°C	degrees centigrade
cfs	cubic feet per second
ft	feet
g	gram, a unit of mass
kg	kilograms, a unit of mass equal to 1,000 grams.
m	meter
pg/L	picograms per liter (parts per quadrillion)
µg/Kg	micrograms per kilogram (parts per billion)
ww	wet weight

Appendix B. Sampling Locations.

Table B-1. Spokane River Sampling Station Descriptions for the 2009 PBDE Study.

Spokane River Reach	Collection Date	Description	River Mile	Latitude[a]	Longitude[a]	Species
Stateline	3/4/2009	Spokane River near Idaho/Washington border	96.2	47.69720	-117.04133	Largescale sucker
Plante Ferry	3/4/2009	Spokane River near Plante's Ferry Park	84.0 - 84.2	47.69132	-117.25306	Largescale sucker Rainbow trout
Mission Park	3/3/2009	Spokane River near Mission Park	75.1 - 78.0	47.66461	-117.4047	Largescale sucker Mountain whitefish
Nine Mile	4/7/2009	Spokane River near Plese Flats at Riverside State Park	63.1 - 64.0	47.73073	-117.51046	Largescale sucker Mountain whitefish
Upper Long Lake	4/7/2009	Spokane River near Upper Long Lake	56.5 - 57.1	47.79274	-117.53435	Mountain whitefish
			54.0 - 54.1	47.79447	-117.57636	Largescale sucker Northern pikeminnow Smallmouth bass
Lower Long Lake	4/8/2009	Spokane River near Lower Long Lake	38.8 - 39.6	47.83137	-117.76374	Largescale sucker Northern pikeminnow

[a] NAD83 Datum.

Table B-2. Sampling Station Descriptions of Reference Waterbodies for the 2009 PBDE Study.

Reference Waterbody	Collection Date	Description	Latitude[a]	Longitude[a]	Species
Rock Lake	4/6/2009	Rock Lake near west shore of lake	47.18209	-117.68518	Brown trout
					Rainbow trout
Williams Lake	4/8/2009	Williams Lake near southwest shoreline of lake	47.32234	-117.69549	Rainbow trout

[a] NAD83 Datum.

Appendix C. Data Quality

Table C-1. PBDE Laboratory Method Blanks. Bolded values indicate detections.

B09E072-BLK1		
Congener	Result (µg/Kg)	Qualifier
PBDE-047	0.22	U
PBDE-049	0.22	U
PBDE-066	0.22	U
PBDE-071	0.22	U
PBDE-099	0.22	U
PBDE-100	0.22	U
PBDE-138	0.44	U
PBDE-153	0.44	U
PBDE-154	0.44	U
PBDE-183	0.44	U
PBDE-184	0.44	U
PBDE-191	0.44	U
PBDE-209	**12.8**	**J**

B09E075-BLK1		
Congener	Result (µg/Kg)	Qualifier
PBDE-047	0.22	U
PBDE-049	0.22	U
PBDE-066	0.22	U
PBDE-071	0.22	U
PBDE-099	0.22	U
PBDE-100	0.22	U
PBDE-138	0.44	U
PBDE-153	0.44	U
PBDE-154	0.44	U
PBDE-183	0.44	U
PBDE-184	0.44	U
PBDE-191	0.44	U
PBDE-209	**1.38**	

B09E135-BLK1		
Congener	Result (µg/Kg)	Qualifier
PBDE-047	0.20	U
PBDE-049	0.20	U
PBDE-066	0.20	U
PBDE-071	0.20	U
PBDE-099	0.20	U
PBDE-100	0.20	U
PBDE-138	0.40	U
PBDE-153	0.40	U
PBDE-154	0.40	U
PBDE-183	0.40	U
PBDE-184	0.40	U
PBDE-191	0.40	U
PBDE-209	1.00	UJ

B09F210-BLK1		
Congener	Result (µg/Kg)	Qualifier
PBDE-047	0.44	U
PBDE-049	0.44	U
PBDE-066	0.44	U
PBDE-071	0.44	U
PBDE-099	0.44	U
PBDE-100	0.44	U
PBDE-138	0.89	U
PBDE-153	0.89	U
PBDE-154	0.89	U
PBDE-183	0.89	U
PBDE-184	0.89	U
PBDE-191	0.89	U
PBDE-209	2.20	UJ

U - undetected at the level indicated.
UJ – undetected; detection limit is estimated.
J – estimated value.

Table C-2. PBDE Laboratory Control Samples.

B09E072-BS1	
Congener	Recovery (%)
PBDE-047	69
PBDE-049	67
PBDE-066	74
PBDE-071	68
PBDE-099	82
PBDE-100	79
PBDE-138	102
PBDE-153	94
PBDE-154	74
PBDE-183	99
PBDE-184	89
PBDE-191	109
PBDE-209	127

B09E075-BS1	
Congener	Recovery (%)
PBDE-047	98
PBDE-049	99
PBDE-066	98
PBDE-071	100
PBDE-099	100
PBDE-100	99
PBDE-138	102
PBDE-153	102
PBDE-154	101
PBDE-183	105
PBDE-184	101
PBDE-191	104
PBDE-209	77

B09E135-BS1	
Congener	Recovery (%)
PBDE-047	76
PBDE-049	78
PBDE-066	76
PBDE-071	77
PBDE-099	75
PBDE-100	75
PBDE-138	74
PBDE-153	77
PBDE-154	80
PBDE-183	77
PBDE-184	78
PBDE-191	73
PBDE-209	51

B09F210-BS1	
Congener	Recovery (%)
PBDE-047	86
PBDE-049	86
PBDE-066	86
PBDE-071	84
PBDE-099	90
PBDE-100	85
PBDE-138	105
PBDE-153	106
PBDE-154	85
PBDE-183	100
PBDE-184	95
PBDE-191	105
PBDE-209	77

Table C-3. PBDE Standard Reference Material Samples.

B09E072-SRM1	
Congener	Recovery (%)
PBDE-047	79
PBDE-099	105
PBDE-153	96

B09E075-SRM1	
Congener	Recovery (%)
PBDE-047	104
PBDE-099	111
PBDE-153	74

B09E135-SRM1	
Congener	Recovery (%)
PBDE-047	74
PBDE-099	76
PBDE-153	52

B09F210-SRM1	
Congener	Recovery (%)
PBDE-047	96
PBDE-099	110
PBDE-153	93

Table C-4. PBDE Laboratory Matrix Spikes.

B09E072-MS1	
Congener	Recovery (%)
PBDE-047	NC
PBDE-049	77
PBDE-066	110
PBDE-071	81
PBDE-099	112
PBDE-100	NC
PBDE-138	156
PBDE-153	135
PBDE-154	79
PBDE-183	163
PBDE-184	138
PBDE-191	194
PBDE-209	237

B09E075-MS1	
Congener	Recovery (%)
PBDE-047	99
PBDE-049	100
PBDE-066	103
PBDE-071	98
PBDE-099	110
PBDE-100	104
PBDE-138	109
PBDE-153	107
PBDE-154	101
PBDE-183	110
PBDE-184	102
PBDE-191	112
PBDE-209	98

B09E135-MS1	
Congener	Recovery (%)
PBDE-047	NC
PBDE-049	82
PBDE-066	103
PBDE-071	105
PBDE-099	75
PBDE-100	NC
PBDE-138	85
PBDE-153	87
PBDE-154	84
PBDE-183	85
PBDE-184	82
PBDE-191	87
PBDE-209	46

NC = Not Calculated. Native sample contained high level of analyte (3X greater or more); the difference between spiked and native values was not great enough to determine an accurate recovery.

Table C-5. PBDE Laboratory Duplicates. Bolded values indicated detections.

0905034-05			B09E072-DUP1			RPD (%)
Congener	Result (µg/Kg)	Qualifier	Congener	Result (µg/Kg)	Qualifier	
PBDE-047	**77**		PBDE-047	**68**	J	12
PBDE-049	4.4	U	PBDE-049	**0.89**	J	---
PBDE-066	4.4	U	PBDE-066	0.89	U	0
PBDE-071	4.4	U	PBDE-071	0.89	U	0
PBDE-099	4.4	U	PBDE-099	0.89	U	0
PBDE-100	**14**		PBDE-100	**13.7**	J	2
PBDE-138	8.8	U	PBDE-138	1.8	U	0
PBDE-153	**2.7**	J	PBDE-153	**1.9**	J	37
PBDE-154	**5.3**	J	PBDE-154	**4.3**	J	20
PBDE-183	8.8	U	PBDE-183	1.8	U	0
PBDE-184	8.8	U	PBDE-184	1.8	U	0
PBDE-191	8.8	U	PBDE-191	1.8	U	0
PBDE-209	22	U	PBDE-209	13	UJ	0

0905034-43			B09E135-DUP1			RPD (%)
Congener	Result (µg/Kg)	Qualifier	Congener	Result (µg/Kg)	Qualifier	
PBDE-047	**70**		PBDE-047	**71**		2
PBDE-049	**5.3**		PBDE-049	**5.3**		1
PBDE-066	0.4	U	PBDE-066	0.39	U	0
PBDE-071	0.4	U	PBDE-071	0.39	U	0
PBDE-099	0.4	U	PBDE-099	0.39	U	0
PBDE-100	**17**		PBDE-100	**17**		2
PBDE-138	0.8	U	PBDE-138	0.78	U	0
PBDE-153	**1.1**		PBDE-153	**1.1**		2
PBDE-154	**4.6**		PBDE-154	**4.7**		2
PBDE-183	0.8	U	PBDE-183	0.78	U	0
PBDE-184	0.8	U	PBDE-184	0.78	U	0
PBDE-191	0.8	U	PBDE-191	0.78	U	0
PBDE-209	4.8	UJ	PBDE-209	2.0	UJ	0

0905034-25			B09E075-DUP1			RPD (%)
Congener	Result (µg/Kg)	Qualifier	Congener	Result (µg/Kg)	Qualifier	
PBDE-047	**48**		PBDE-047	**36**		29
PBDE-049	**4.1**		PBDE-049	**3.0**		32
PBDE-066	**2.0**		PBDE-066	**1.6**		22
PBDE-071	0.43	U	PBDE-071	0.43	U	0
PBDE-099	**89**		PBDE-099	**66**		30
PBDE-100	**19**		PBDE-100	**14**		32
PBDE-138	0.86	U	PBDE-138	0.87	U	0
PBDE-153	**14**		PBDE-153	**12**		18
PBDE-154	**10**		PBDE-154	**8.9**		12
PBDE-183	**0.19**	J	PBDE-183	0.87	U	---
PBDE-184	0.86	U	PBDE-184	0.87	U	0
PBDE-191	0.86	U	PBDE-191	0.87	U	0
PBDE-209	2.1	U	PBDE-209	2.2	U	0

RPD – Relative percent difference.
U - undetected at the level indicated.
J – estimated value.

Table C-6. Lipid Laboratory Method Blanks. Bolded values indicate detections.

Sample Number	Result (%)	Qualifier
B09E074-BLK1	0.01	U
B09E076-BLK1	**0.02**	
B09E134-BLK1	0.01	U

U - undetected at the level indicated.

Table C-7. Lipid Laboratory Duplicates.

Sample Number	Result (%)	RPD (%)
0905034-05	6.69	9.0
B09E074-DUP1	6.12	

Sample Number	Result (%)	RPD (%)
0905034-43	4.21	4.0
B09E134-DUP1	4.36	

Sample Number	Result (%)	RPD (%)
B09E076-DUP1	1.3	10
0905034-25	1.44	

RPD = Relative Percent Difference

Appendix D. Fish Biological Data

Table D-1. Composite Fish Data by Spokane River Reach, 2009.

Spokane River Reach	River Mile	Sample ID	Species	No. of Individuals per Sample	Mean Length (mm)	Mean Weight (g)	Mean Age	Tissue Type
Stateline	96.2	0905034-01	LSS	3	451	1112	8.67	Whole
		0905034-02	LSS	3	476	1127	9.67	Whole
		0905034-03	LSS	4	520	1565	13.5	Whole
Plante Ferry	84.0 - 84.2	0905034-04	LSS	3	470	1139	8.33	Whole
		0905034-05	LSS	4	488	1322	9.5	Whole
		0905034-06	LSS	5	529	1643	12	Whole
		0905034-19	RBT	3	400	578	3	Fillet
		0905034-20	RBT	3	400	578	3	Carcass
Mission Park	75.1 - 78.0	0905034-07	LSS	3	421	850	8	Whole
		0905034-08	LSS	5	456	1029	12.4	Whole
		0905034-09	LSS	4	490	1226	11.5	Whole
		0905034-21	MWF	4	370	434	9.75	Fillet
		0905034-22	MWF	4	370	434	9.75	Carcass
Nine Mile	63.1 - 64.0	0905034-10	LSS	3	398	832	7	Whole
		0905034-11	LSS	3	455	959	9	Whole
		0905034-12	LSS	3	487	1120	11.67	Whole
		0905034-26	MWF	3	280	195	4	Fillet
		0905034-27	MWF	3	280	195	4	Carcass
		0905034-28	MWF	3	322	284	6	Fillet
		0905034-29	MWF	3	322	284	6	Carcass
		0905034-30	MWF	3	341	318	7	Fillet
		0905034-31	MWF	3	341	318	7	Carcass
Upper Long Lake	56.5 - 57.1	0905034-13	LSS	3	438	955	11	Whole
		0905034-14	LSS	4	468	1218	14.5	Whole
		0905034-15	LSS	5	487	1365	13.6	Whole
		0905034-32	SMB	3	347	623	5.67	Fillet
		0905034-34	SMB	2	478	1886	11	Fillet
		0905034-35	MWF	4	278	175	2.5	Fillet
		0905034-36	MWF	4	278	175	2.5	Carcass
		0905034-37	MWF	5	320	253	6	Fillet
		0905034-38	MWF	5	320	253	6	Carcass
		0905034-39	NPM	5	381	488	11.8	Whole
		0905034-40	NPM	4	418	661	13.25	Whole
		0905034-41	NPM	4	449	813	14	Whole
Lower Long Lake	38.8 - 39.6	0905034-16	LSS	3	411	847	6	Whole
		0905034-17	LSS	5	462	1131	13.8	Whole
		0905034-18	LSS	5	493	1328	14	Whole
		0905034-42	NPM	4	378	498	11.25	Whole
		0905034-43	NPM	4	404	601	11.5	Whole
		0905034-44	NPM	3	427	695	12.33	Whole

Table D-2. Composite Fish Data by Reference Waterbody, 2009.

Reference Waterbody	Sample ID	Species	No. of Individuals per Sample	Mean Length (mm)	Mean Weight (g)	Mean Age	Tissue Type
Rock Lake	0905034-23	RBT	4	393	644	2	Whole
	0905034-24	BNT	5	366	398	2	Whole
Williams Lake	0905034-25	RBT	5	270	213	1.4	Whole

Appendix E. Linear Regressions

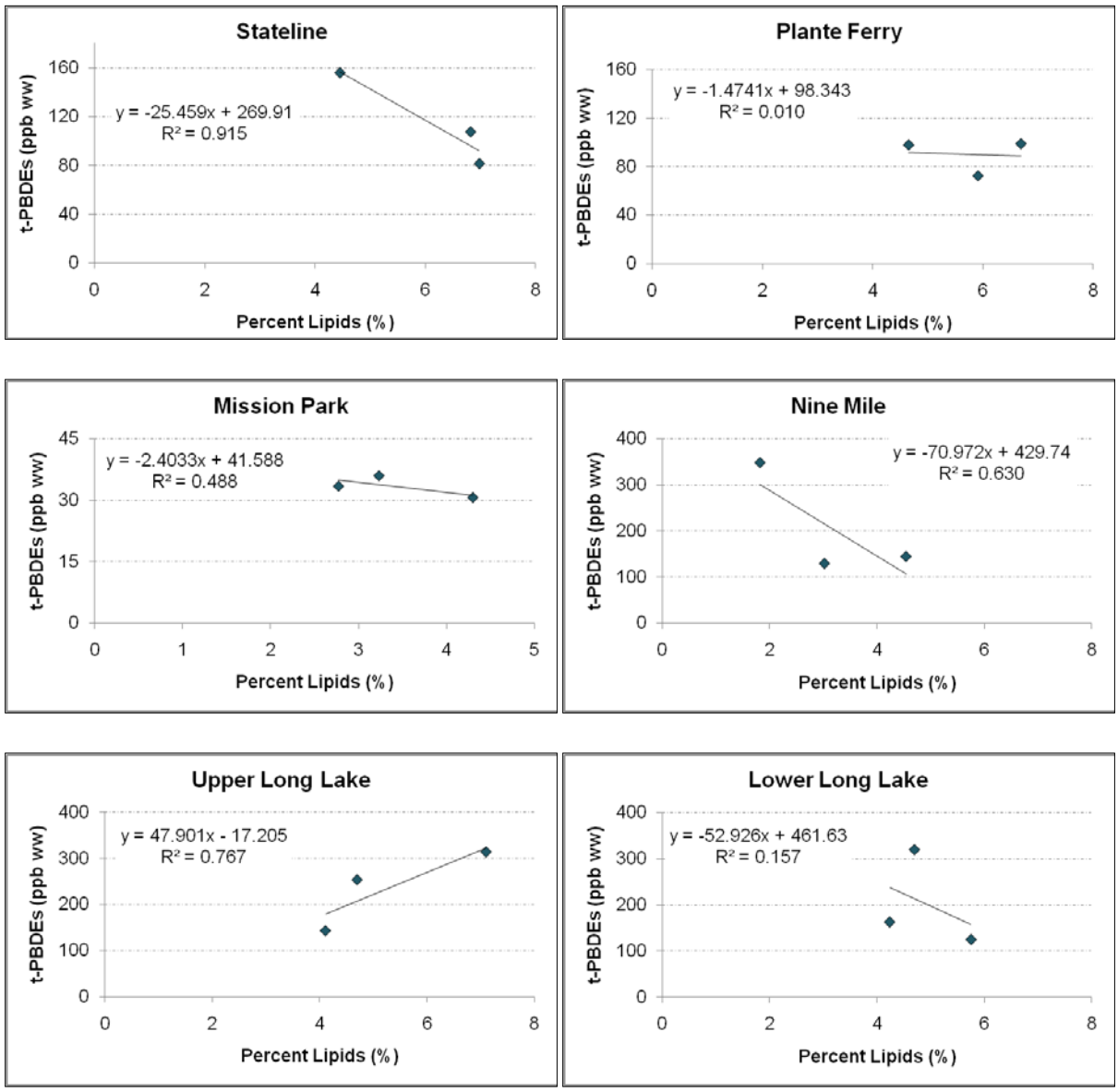

Figure E-1. Simple Linear Regression Plots for Total PBDEs (ppb ww) and Percent Lipids in Largescale Sucker Tissue.

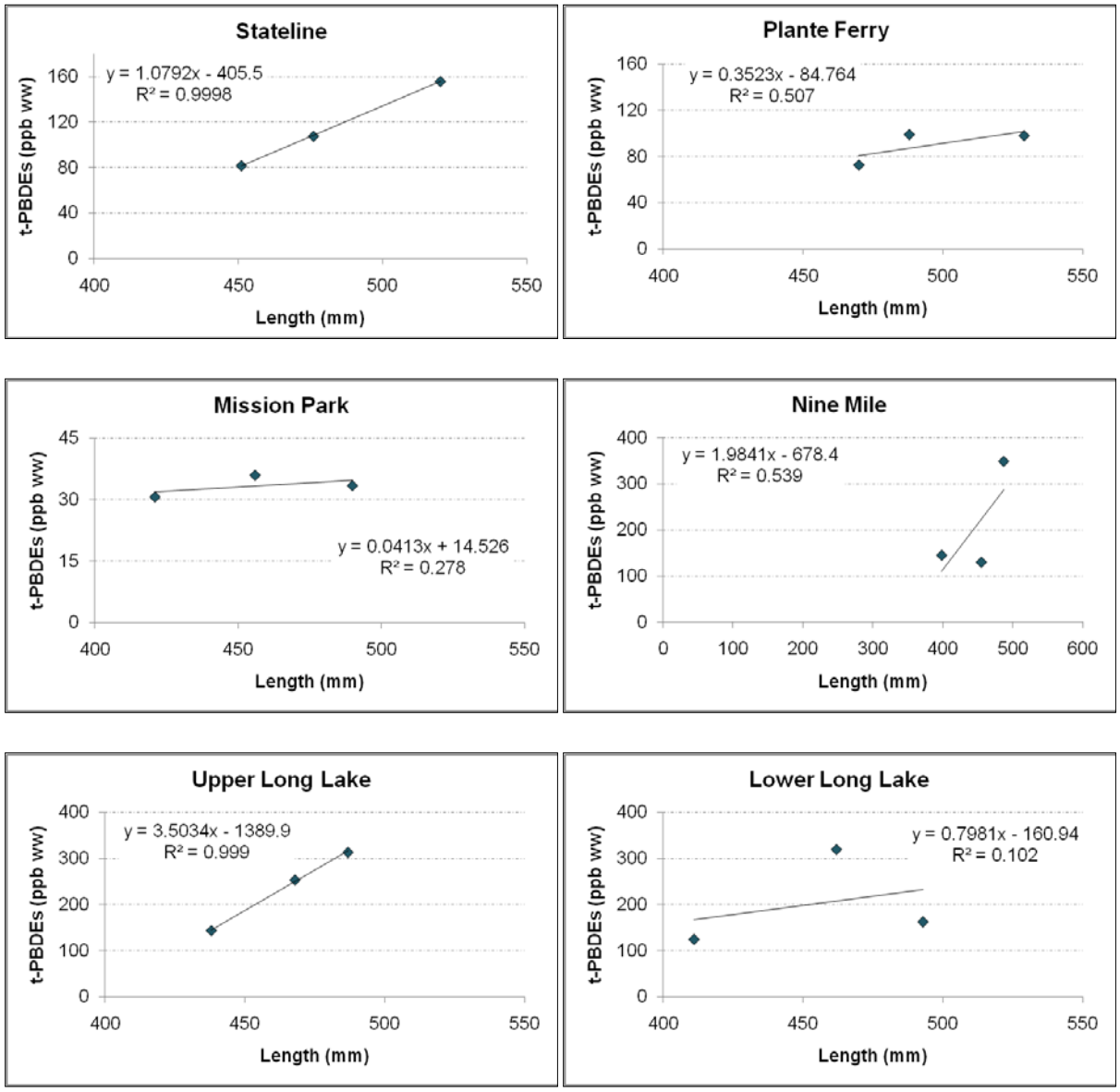

Figure E-2. Simple Linear Regression Plots for Total PBDEs (ppb ww) and Largescale Sucker Total Length (mm).

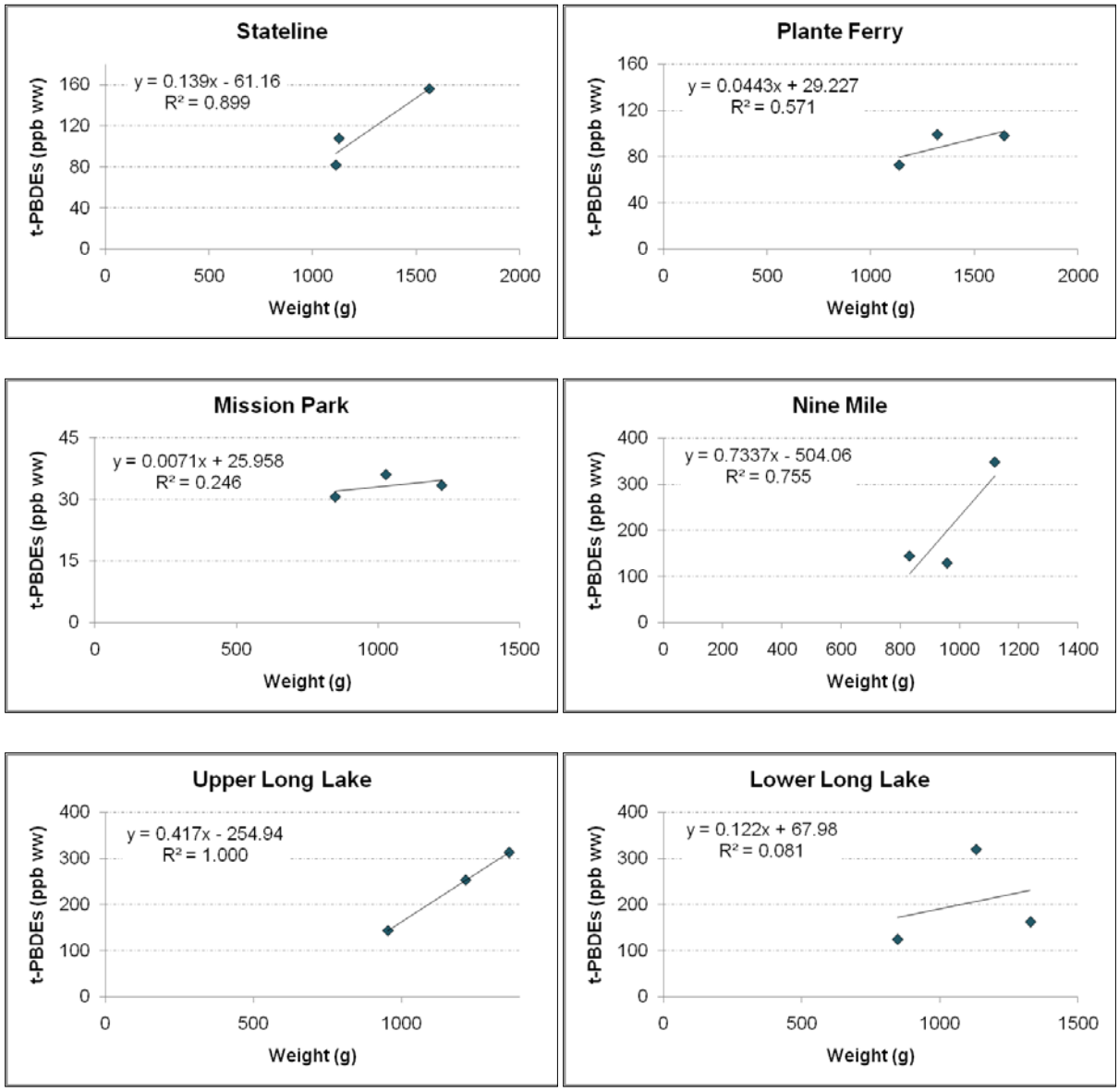

Figure E-3. Simple Linear Regression Plots for Total PBDEs (ppb ww) and Largescale Sucker Total Weight (g).

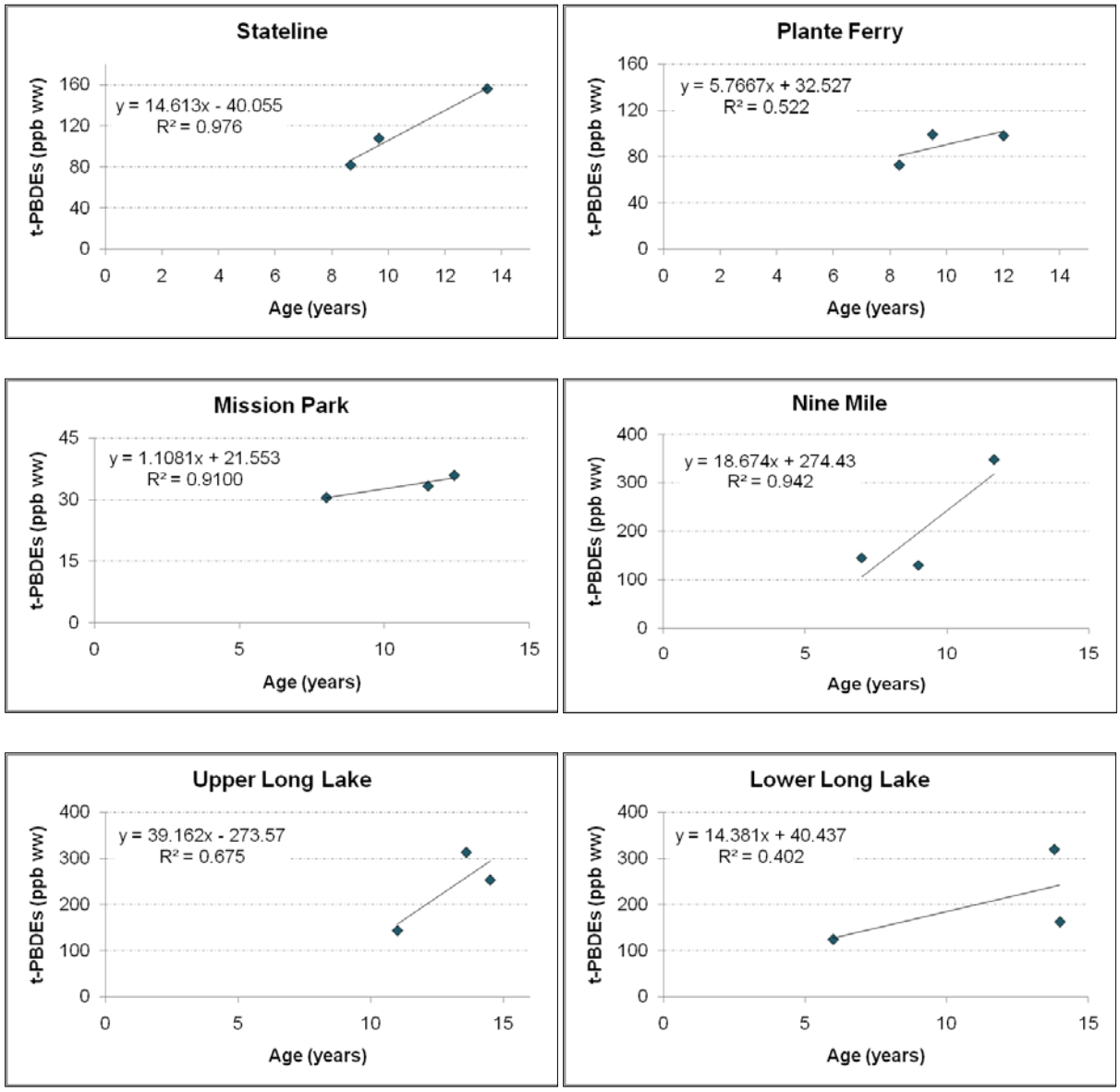

Figure E-4. Simple Linear Regression Plots for Total PBDEs (ppb ww) and Largescale Sucker Age (years).

Appendix F. Temporal Trends

Table F-1. Summary of Differences in Largescale Sucker PBDE Concentrations between 2005 and 2009 (Current) Studies.

Station	2005 Study					Current Study (2009)					% Change in Total PBDEs	Increase (+) or Decrease (-)	% Change in Lipid-normalized PBDEs	Increase (+) or Decrease (-)	RPD in Length
	Sample ID	Fish Length (mm)	Fish Lipids (%)	Total PBDEs (µg/Kg ww)	Lipid-normalized Total PBDEs (µg/Kg lipid)	Sample ID	Fish Length (mm)	Fish Lipids (%)	Total PBDEs (µg/Kg ww)	Lipid-normalized Total PBDEs (µg/Kg lipid)					
Stateline	05494247	444	10.36	214.4	2069.4	0905034-01	451	6.98	81.6	1168.9	61.9%	-	43.5%	-	1.5%
	05494246	470	9.63	168.7	1752.0	0905034-02	476	6.82	107.6	1577.7	36.2%	-	9.9%	-	1.2%
	05494245	516	10.13	212.1	2093.6	0905034-03	520	4.45	155.9	3503.4	26.5%	-	67.3%	+	0.7%
Plante Ferry	05494250	451	6.25	83.5	1336.0	0905034-04	470	5.91	72.6	1228.4	13.1%	-	8.1%	-	4.2%
	05494248	532	4.80	252.2	5254.2	0905034-06	529	4.65	98.0	2107.5	61.1%	-	59.9%	-	0.6%
	05494249	484	6.06	127.7	2107.3	0905034-05	488	6.69	99.0	1479.8	22.5%	-	29.8%	-	0.7%
Mission Park	05494253	414	3.69	89.5	2425.5	0905034-07	421	4.30	30.6	711.6	65.8%	-	70.7%	-	1.8%
	05494251	501	3.04	97.8	3217.1	0905034-09	490	2.77	33.1	1194.9	66.2%	-	62.9%	-	2.3%
	05494252	455	4.76	96.6	2029.4	0905034-08	456	3.23	36.0	1114.6	62.7%	-	45.1%	-	0.2%
Nine Mile^	05494257/58*	441	3.75	333.7	8898.7	0905034-11	455	3.02	130.0	4305.3	61.0%	-	51.6%	-	3.2%
	05494260	405	5.17	522.9	10114.1	0905034-10	398	4.54	145.2	3198.2	72.2%	-	68.4%	-	1.7%
	05494259	425	4.61	708.3	15364.4	0905034-12	487	1.82	348.3	19136.8	50.8%	-	24.6%	+	13.7%
Upper Long Lake	05494254	438	3.54	537.9	15194.9	0905034-13	438	4.11	143.1	3481.5	73.4%	-	77.1%	-	0.1%
	05494255	461	4.42	718.0	16244.3	0905034-14	468	4.70	253.5	5393.6	64.7%	-	66.8%	-	1.5%
	05494256	487	3.88	458.9	11827.3	0905034-15	487	7.10	313.9	4421.3	31.6%	-	62.6%	-	0.1%
Lower Long Lake	05494243	407	5.16	89.8	1740.3	0905034-16	411	5.75	124.7	2168.7	38.9%	+	24.6%	+	1.0%
	05494242	491	6.40	147.9	2310.9	0905034-18	493	4.24	162.8	3839.6	10.1%	+	66.1%	+	0.3%
	05494244	460	7.05	357.4	5069.5	0905034-17	462	4.70	319.9	6806.4	10.5%	-	34.3%	+	0.4%

* calculated whole fish value.
^ bridgelip suckers were collected for the 2005 study.
RPD = relative percent difference.

Table F-2. Summary of Differences in Mountain Whitefish PBDE Concentrations between 2005 and 2009 (Current) Studies.

Station	2005 Study					Current Study (2009)					% Change in Total PBDEs	Increase (+) or Decrease (-)	% Change in Lipid-normalized PBDEs	Increase (+) or Decrease (-)	RPD in Length
	Sample ID	Fish length (mm)	Fish lipids (%)	Total PBDEs (µg/Kg ww)	Lipid-normalized Total PBDEs (µg/Kg lipid)	Sample ID	Fish Length (mm)	Fish lipids (%)	Total PBDEs (µg/Kg ww)	Lipid-normalized Total PBDEs (µg/Kg lipid)					
Mission Park	5494266	374	6.69	355	5306.4	0905034-21	370	1.72	249.3	14494.2	29.8%	-	173.1%	+	1.0%
Nine Mile	5494267	292	3.80	905.1	23818.4	0905034-26	280	1.08	397.59	36813.9	56.1%	-	54.6%	+	4.1%
	5494268	321	3.59	1049.1	29222.8	0905034-28	322	1.36	615.55	45261.0	41.3%	-	54.9%	+	0.2%
Upper Long Lake	5494240	282	2.56	167	6523.4	0905034-35	278	0.28	149.8	53500.0	10.3%	-	720.1%	+	1.6%
	5494239	318	2.90	198.2	6834.5	0905034-37	320	0.14	196.2	140142.9	1.0%	-	1950.5%	+	0.6%

RPD - relative percent difference.