Freshwater Fish Contaminant Monitoring Program

Annual Report for 2013

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Cover photo: Common carp (*Cyprinus carpio*) and northern pikeminnow (*Ptychocheilus oregonensis*) collected from Lake Pateros on the Columbia River.

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by

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

WRIAs

- 32 - Walla Walla
- 40 - Alkali-Squilchuck
- 41 - Lower Crab
- 44 - Moses Coulee
- 45 - Wenatchee
- 47 - Chelan
- 50 - Foster
- 57 - Middle Spokane

HUC numbers

- 17010305 - Upper Spokane
- 17020005 - Chief Joseph
- 17020010 - Upper Columbia-Entiat
- 17060110 - Lower Snake
- 17070101 - Middle Columbia-Lake Wallula

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Abstract

Results from the Freshwater Fish Contaminant Monitoring Program in 2013 are summarized for three areas in Washington: the mid-Columbia River (Wanapum Dam to Grand Coulee Dam), McNary National Wildlife Refuge, and Liberty Lake. Goals were to (1) characterize contaminant concentrations in fish and (2) determine spatial and temporal patterns for Columbia River fish.

Washington's water quality standards for contaminants in fish tissue were met for most of the chemicals analyzed. However, many samples failed to meet standards for 4,4'-DDE, t-PCBs, and dioxins/furans. The high variability and small sample sizes associated with these data made it difficult to detect spatial and temporal patterns in mid-Columbia River fish, yet some patterns emerged.

Results show that 4,4'-DDE in fish significantly increased in Lake Pateros where the Okanogan River enters the Columbia River, and continued increasing into Lake Entiat before decreasing in the Wenatchee Reach. In general, levels of other organic contaminants in fish increased from upstream to downstream sites. Yet this pattern was weaker and more variable, and the influence of the Okanogan River was less clear, as was seen with 4,4'-DDE. Mercury in fish followed no spatial patterns, and levels appeared to be more dependent on fish species. Temporal trends were not seen in concentrations of 4,4'- DDE between 2004 and 2013 for five cases of comparable data. However, levels of TCDD-TEQ in common carp and channel catfish in Lake Wallula appeared to be lower in 2013 than in 1990.

Samples at McNary National Wildlife Refuge showed elevated levels of t-DDT, t-PCB, and dieldrin in fish from waters directly connected to the Snake and Columbia Rivers, while fish from Burbank Slough had low concentrations of all contaminants.

Walleye from Liberty Lake did not meet standards for t-PCBs, 4,4'-DDE, and dioxins/furans. While all samples met Washington's standard for mercury, two of the samples did not meet other benchmarks.

Introduction

Since 2001, the Washington State Department of Ecology's (Ecology) Freshwater Fish Contaminant Monitoring Program (FFCMP)[1] has characterized persistent, bioaccumulative, and toxic chemicals (PBTs) in freshwater fish statewide with analysis of over 400 fish tissue samples from 150 sites. The FFCMP has two broad goals: (1) long-term monitoring for temporal trends and (2) exploratory monitoring to characterize the extent of contamination in areas of interest.

Results from fish contaminant monitoring are used for a variety of purposes, such as water quality assessments, health risk assessments, determining total maximum daily load (TMDL) effectiveness, and evaluating spatial and temporal trends. Target analytes are most often mercury, polychlorinated biphenyls (PCBs), dioxins and furans (PCDD/Fs), chlorinated pesticides (CPs) – especially dichloro-diphenyl-trichloroethane (DDT) and its breakdown products dichloro-diphenyl-dichloroethane (DDD) and dichloro-diphenyl-dichloroethylene (DDE), and polybrominated diphenyl ethers (PBDEs). More information about these and other chemicals is at www.ecy.wa.gov/programs/eap/toxics/chemicals_of_concern.html.

The accumulation of contaminants can have a variety of health effects on humans and wildlife, such as reproductive abnormalities, neurological problems, and behavioral changes. A primary route of exposure for people is through the consumption of contaminated food, particularly fish. The Washington State Department of Health (Health) currently has a statewide fish consumption advisory (FCA) for mercury in bass and northern pikeminnow. There are also 16 site-specific advisories due to contamination of fish by various chemicals: www.doh.wa.gov/CommunityandEnvironment/Food/Fish.aspx.

Results from the 2013 FFCMP are summarized here for three areas: the mid-Columbia River, McNary National Wildlife Refuge (NWR), and Liberty Lake. Results may be downloaded from Ecology's Environmental Information Management database (EIM) at http://www.ecy.wa.gov/eim.

The project plan for the 2013 monitoring effort gives more detail about the mid-Columbia River and McNary NWR work (Seiders, K., 2013a). The Liberty Lake work was added to address emerging concerns about the lake's walleye fishery. The Washington Department of Fish and Wildlife (WDFW) and Health approached Ecology about measuring contaminant levels in walleye so they could incorporate potential risks to human health in managing this fishery.

Figure 1 shows fish sampling locations for the mid-Columbia River and McNary NWR. Not shown is Liberty Lake, which lies about 15 miles east of downtown Spokane.

[1] www.ecy.wa.gov/programs/eap/toxics/wstmp.html

Columbia River

Fish from the mid-Columbia River have some of the highest concentrations of DDT compounds in Washington. Sources of DDT are related to historical pesticide use in this basin and are being addressed in some areas. The Okanogan River, a major tributary to the upper area of the mid-Columbia River, was the subject of a TMDL study in 2001 (Serdar, 2003) and later, a water cleanup plan (Peterschmidt, 2006) to address high levels of DDT compounds.

Concentrations of PCBs and PCDD/Fs are also elevated in some mid-Columbia River species. There are a number of 303(d) listings for these contaminants in the study area. In 1991, EPA established a TMDL for 2,3,7,8-TCDD (EPA, 1991) because water quality standards were not being met in the Columbia, Snake, and Willamette Rivers. The states of Washington, Oregon, and Idaho requested that EPA establish this TMDL as a federal action to help address multi-state consistency concerns.

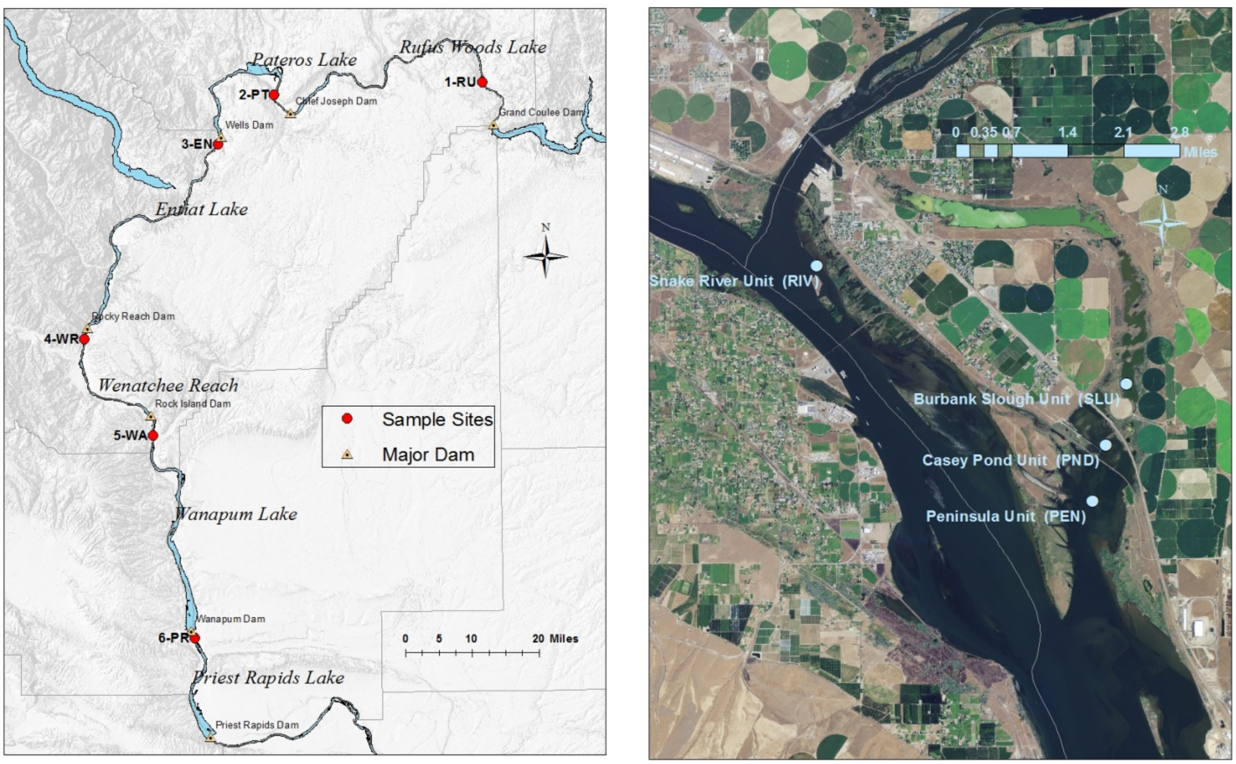

Figure 1. Fish collection areas for the mid-Columbia River (left) and the McNary NWR (right).

At least six fish tissue monitoring efforts have been conducted since the early 1990s that included the Columbia River from the Wallula Gap to Grand Coulee Dam (EPA, 2002; EPA, 2005; EPA, Herger, 2013, Energy, 2012; Seiders et al, 2007; and Serdar et al, 1991). When viewed collectively, these historical efforts reveal a patchwork of sites, species, tissue types, collection seasons, target analytes, and analytical methods.

The 2013 effort targeted sites and species that were sampled historically in order to gain a temporal perspective where possible. We aimed to produce a robust data set to serve as a benchmark for future sampling and comparisons. Ecology's FFCMP plans to sample this area again in about 10 years.

McNary National Wildlife Refuge

The McNary NWR lies adjacent to the mid-Columbia River between the confluence of the Snake and Walla Walla Rivers. This refuge contains up to 2,000 acres of backwater habitats where seven salmonid stocks which are listed under the Endangered Species Act spend portions of their life histories. In planning for an extensive backwater habitat enhancement and restoration project, the U.S. Fish and Wildlife Service (USFWS) needed to characterize the refuge's resources, which included describing the extent of contaminants in water, sediment, and biota (Campbell, 2011; Kisor et al., 2011).

Ecology and the USFWS worked together in collecting fish from four areas or "Units" of the refuge (Figure 1) and the FFCMP analyzed tissues for various contaminants. The USFWS will use Ecology's results to compare fish contaminant burdens with fish health data (histology, parasitology, bacteriology, and virology) to determine if contaminants are affecting the health of refuge fish.

Liberty Lake

Concerns about contamination of fish in Liberty Lake led to a cooperative effort by Ecology, Health, and WDFW to fill data gaps. The levels of mercury in bass sampled in 2010 by Ecology's Mercury Trends in Freshwater Fish program ranged from 180-483 ug/kg with half of the samples exceeding (not meeting) the EPA-Recommended Criterion of 300 ug/kg (Meredith and Friese, 2011). While brown trout and yellow perch were also sampled for mercury in 2010, information about the extent of contaminants in walleye, and potential risks to consumers, was lacking.

Walleye is becoming popular for sport fishing in Liberty Lake. The goal of the monitoring effort was to characterize contaminants in walleye to help inform decisions about the management of this fishery.

Methods

Field and Laboratory Methods

Twelve species of fish were collected from three general areas and used to form composite samples to meet site-specific objectives (Table 1). Fish were collected by staff from Ecology, USFWS, and WDFW. Sample collection, preparation, and analytical methods followed those described in the project plans for the FFCMP (Seiders, 2013a, 2013b). A total of 97 samples of fish tissue were analyzed for some or all of these chemicals: chlorinated pesticides (CPs), mercury, PBDEs, PCBs, and PCDD/Fs. All results were reported on a wet-weight basis.

Table 1. Number of composite samples analyzed per fish species per site, 2013.

Sample Location	LSS	NPM	MWF	CLM	CCP	WAL	RBT,w	RBT,h	BUR	PEA	CAT	SMB	LMB
Columbia River													
Rufus Woods Lake (below Grand Coulee Dam, "1-RU")	5	2	2			3	3	2	1				
Pateros Lake (below Chief Joseph Dam, "2-PT")	5	1			3								
Entiat Lake (below Wells Dam, "3-EN")	5	2		4									
Wenatchee Reach (below Rocky Reach Dam, "4-WR")	5	4	3	3									
Wanapum Lake (below Rock Island Dam, "5-WA")	5	6		3						3			
Priest Rapids Lake (below Wanapum Dam, "6-PR")	5	3	4			2							
McNary National Wildlife Refuge (NWR)													
Burbank Slough Unit					1								1
Casey Pond Unit					1							1	
Peninsula Unit					1						2	1	
Snake River unit					1							1	
Spokane County													
Liberty Lake (near Spokane)						3							

Species codes:
BUR: Burbot, CAT: Channel catfish, CCP: Common carp, CLM: Chiselmouth, LMB: Largemouth bass, LSS: Largescale sucker, MWF: Mountain whitefish, NPM: Northern pikeminnow, PEA: Peamouth, RBT: Rainbow trout (h: hatchery, w: wild), SMB: Smallmouth bass, WAL: Walleye.

Most composite samples consisted of skin-on fillets from five individual fish of the same species per site, with the exception of largescale suckers that were processed as whole fish. For most Columbia River sites, multiple composite samples of the same species were collected in order to address sampling variability and improve the strength of statistical tests to determine spatial or temporal differences. For the Columbia River, historical data informed the selection of species and fish size ranges for collections in order to improve the spatial and temporal comparability of results among sites and studies.

Data Quality Assessment

The quality of data from the 2013 study was assessed by reviewing laboratory case narratives, analytical results, and field replicate data. Quality control procedures included a mixture of analyses such as method blanks, calibration and control standards, matrix spikes, matrix spike duplicates, surrogate recoveries, laboratory duplicates and field replicates. Overall, most of the 2013 data met measurement quality objectives. Result values for DDT compounds in sample 1401003-66 (EN-LSS-1) were inexplicably high and not used in this reporting effort. All other results were deemed usable as qualified.

An important exception to data quality was completeness of the 2013 data set: only 45% of target samples were collected because of challenges in getting enough samples of target species in desired size ranges. This affected the ability of this project to fully meet all of its objectives. In many cases, non-target species and size ranges were substituted in order to provide alternative information.

The quality and comparability of historical data were examined by reviewing the individual study reports with emphasis on field, laboratory, and quality assurance procedures. Most of the historical studies were loaded into EIM. These data were captured to allow easier access and broader use. The loading process involves reviews of data quality and often incorporates the translation of original data qualifiers to data qualifiers that are used in EIM. In most cases, data from historical studies were deemed acceptable as qualified for uses in this report.

Some samples had concentrations of PCBs determined using different analytical methods for Aroclors and congeners. A paired sample t-test showed that t-PCB values from these different methods were comparable, which is consistent with previous comparisons (Seiders and Deligeannis, 2009; Johnson et al., 2010). Yet, exceptions existed for some results. Such exceptions may be associated with the challenges of identifying and interpreting PCB Aroclors in the fish tissue matrix where the target analytes are often weathered and confounded by other matrix interferences like lipids. Where appropriate, results from these two analytical methods for PCBs were pooled for further comparisons.

Further assessment of data quality is beyond the scope of this report. Other quality assurance information is available by contacting the authors of this report.

Data Reduction, Trends Analyses, Water Quality Criteria

Data reduction and management procedures followed practices described in the project plan for the FFCMP (Seiders, 2013b). Results from some groups of target analytes were summed in order to account for their additive effects and simplicity of comparison to various criteria and other data. Summed values in this report are noted using the prefix "t-", as in t-PCB. Procedures for summing followed Ecology guidance for the Water Quality Assessment process (Ecology, 2012).

Contaminant concentrations in fish can be influenced by many factors, such as: species, tissue type, size, age, lipid content, collection location, collection season, and analytical method. These factors were considered while choosing samples for various comparisons. For the mid-Columbia River data, sample results from 2013 were plotted to examine relationships between 4,4'-DDE (DDE) and mercury to fish length, weight, age, and lipids. DDE served as a surrogate for other organic contaminants in the first round of plots. Simple linear regression was used to help determine the existence and strength of relationships. These plots showed that relationships among these parameters were non-existent, inconsistent, or too weak to use in normalizing the data or performing other adjustments using co-variance. Such adjustments could potentially increase the sensitivity of statistical tests for differences among sites or between years.

Data sets representing cases of individual fish species and sites from the mid-Columbia River were examined for spatial (among sites) and temporal (over time) differences. Plots of mean values with 95% confidence intervals suggested that differences among sites or over time would not be discernible in most cases. Yet in some cases, further testing might show differences. Spatial comparisons of the 2013 data were done using the non-parametric Kruskal-Wallis single-factor ANOVA with Dwass-Steel-Critchlow-Fligner tests for pair-wise comparisons (SYSTAT, 2012). Where statistical tests were conducted, the generalized null hypothesis was that data sets did not differ. For these tests, an alpha level of 0.10 was chosen; this means there was a low probability (10%) that the outcome was due to chance. Statistical testing for temporal comparisons was not pursued.

Results were compared to Washington's human-health based water quality standards (Chapter 173-201A WAC) that can be expressed as fish tissue equivalent concentrations (FTEC). The FTEC is the concentration of a contaminant in edible fish tissue that equates to Washington's water quality criterion for the protection of human health from that contaminant. Fish tissue sample concentrations lower than the FTEC indicate that water quality standards are met for that specific contaminant.

The FTEC for PCDD/Fs applies to two expressions of these compounds: the single congener 2,3,7,8-TCDD (the most toxic congener) and TCDD-TEQ, (the toxicity equivalent or TEQ to 2,3,7,8-TCDD. This TCDD-TEQ approach accounts for the cumulative toxicity of all dioxin and furan congeners and follows recommendations by EPA (EPA, 2010) and the World Health Organization (Van den Berg et al., 2006). In Washington's water quality assessment, TCDD-TEQ results that exceed (do not meet) the FTEC for 2,3,7,8-TCDD (0.065 ng/kg) are placed in Category 2 – "Segment is a Water of Concern", rather than in Category 5 – "Segment is on 303(d) List" (Ecology, 2012). For TCDD-TEQ results that exceed 0.065 ng/kg, Category 2 is used because TCDD-TEQ is not specifically listed in the National Toxics Rule (40 CFR 131.36, 2006), the basis for Washington's water quality standards for the protection of human health.

Human-health-based water quality criteria used by Washington are contained in the National Toxics Rule (WAC Chapter 173-201A, 2012) (Ecology, 2011b) and are used in Washington's Water Quality Assessment (WQA) process. The WQA process involves the review of information from sampling efforts in the context of water quality standards. Water bodies are then assigned to one of five categories that help guide the management of pollution problems. This process and categories are described at www.ecy.wa.gov/programs/wq/303d/index.html.

Results and Discussion

Results for the most frequently detected analytes in 2013 are summarized below. All results are available in EIM under the Study ID FFCMP13.

Columbia River

Table 2 summarizes results for fillets from 8 species and whole tissue from largescale suckers. Ancillary data for lipids, fish size, and fish age are given because these can influence contaminant concentrations in fish. The pesticide DDT and its breakdown products were the only chlorinated pesticides detected in fillet tissue. No sites met FTECs for DDE, PCBs, and TCDD-TEQ, except for DDE in fish from Rufus Woods Lake. Common carp from Lake Pateros did not meet the FTEC for 2,3,7,8-TCDD.

Table 2. Summary of results from 2013: Rufus Woods Lake to Priest Rapids Lake.

Tissue Type	Statistic	4,4'-DDE (ug/kg)	t-PCB (ug/kg)	TCDD-TEQ (ng/kg)	2,3,7,8-TCDD (ng/kg)	t-PBDE (ug/kg)	Mercury (ug/kg)	Lipids (%)	Mean* Total Length (mm)	Mean* Weight (g)	Mean* Age (yrs)
Fillet (most species)	count	54	54	33	33	27	54	54	54	54	54
	mean	**161.6**	**33.6**	**0.226**	0.033	10.4	194.1	3.00	392.6	953.7	6.7
	median	**89.0**	**5.3**	**0.162**	0.023	5.4	104.1	1.88	356.8	421.8	6.7
	90th %ile	**371.0**	**79.2**	**0.578**	**0.077**	15.5	486.8	6.71	518.9	1457.9	11.5
	min	1.0	4.8	0.024	0.010	0.6	22.6	0.40	251.8	144.8	1.0
	max	**1000.0**	**118.5**	**0.977**	**0.110**	110.5	620.0	16.4	727.4	6787.6	14.2
	FTEC	31.6	5.3	0.065	0.065		770				
	% > FTEC	56%	89%	82%	12%		0%				
Whole (LSS only)	count	29	30			6	30	30	30	30	30
	mean	282.0	64.3			14.9	62.9	4.45	483.4	1127.7	12.0
	median	200.0	52.5			14.3	63.2	4.45	481.1	1109.4	11.4
	90th %ile	640.0	110.3			22.8	87.5	6.82	510.6	1414.9	14.5
	min	6.8	9.8			3.8	32.1	0.98	448.8	807.2	8.0
	max	850.0	161.0			29.1	103.0	8.84	520.4	1542.2	19.8

Bold values do not meet Washington's water quality standards.
* Mean of the average length, weight, or age of individual fish that were used in each composites sample.

Figure 2 shows the range of DDE and PCBs seen in the 11 species that were sampled from the mid-Columbia River sites. These boxplots show that the highest concentrations of these contaminants in fillet tissue are in common carp, channel catfish, and northern pikeminnow. High levels are also seen in whole largescale suckers. Species codes are shown in Table 1.

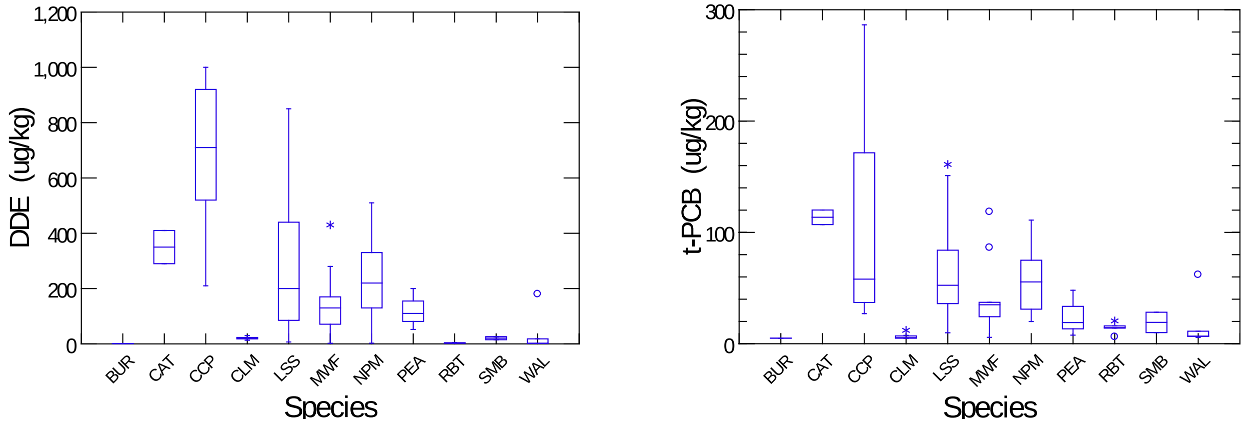

Figure 2. Boxplots of DDE and t-PCB in fillet tissue from ten species and whole tissue from one species (LSS) from the mid-Columbia River in 2013.

Species Codes:
BUR: Burbot, CAT: Channel catfish, CCP: Common carp, CLM: Chiselmouth, LSS: Largescale sucker, MWF: Mountain whitefish, NPM: Northern pikeminnow, PEA: Peamouth, RBT: Rainbow trout, SMB: Smallmouth bass, WAL: Walleye.

Figure 3 shows results for t-DDT, t-PCBs, TCDD-TEQ, and t-PBDEs in fillet tissue from multiple species of fish collected across Washington since 2001. Results from 2013 are also indicated, along with values that can be used for assessing health risks to humans consuming contaminated fish. These are: (1) the U.S. EPA Screening Values (SVs) for Subsistence and Recreational Fishers (EPA, 2000) and (2) Washington's FTECs. Screening values and regulatory thresholds have not yet been established for PBDEs.

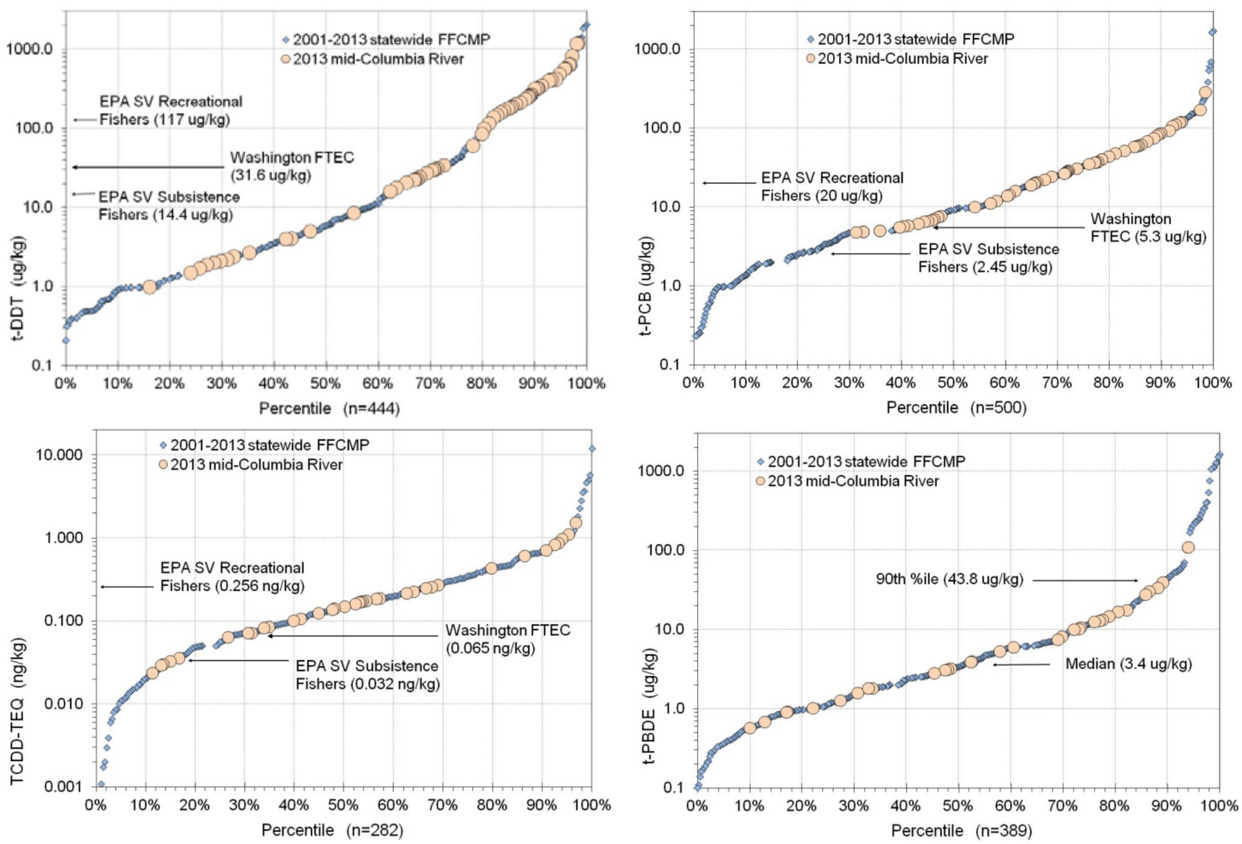

Figure 3. Cumulative frequency distributions for t-DDT, t-PCB, TCDD-TEQ, and t-PBDE in fillet tissue from Washington.

For these four contaminants, the 2013 results are generally representative of statewide values with one clear exception. For t-DDT, the 2013 results that are below the 60th percentile are fish from Rufus Woods Lake, which is above the confluence of the Okanogan River. The 2013 results that are above the 60th percentile are among the highest in the state and are from samples collected below Rufus Woods Lake. For PCBs, many of the 2013 samples are in the upper 60th percentile of statewide values.

Spatial Trends

2013 Ecology Results

Figure 4 shows results for two species from different sites. Results from multiple samples are shown as mean values with 95% confidence limits. Results are ordered by site from upstream (1-RU, Rufus Woods Lake) to downstream (6-PR, Priest Rapids Lake). Sample site and species codes are shown in Table 1.

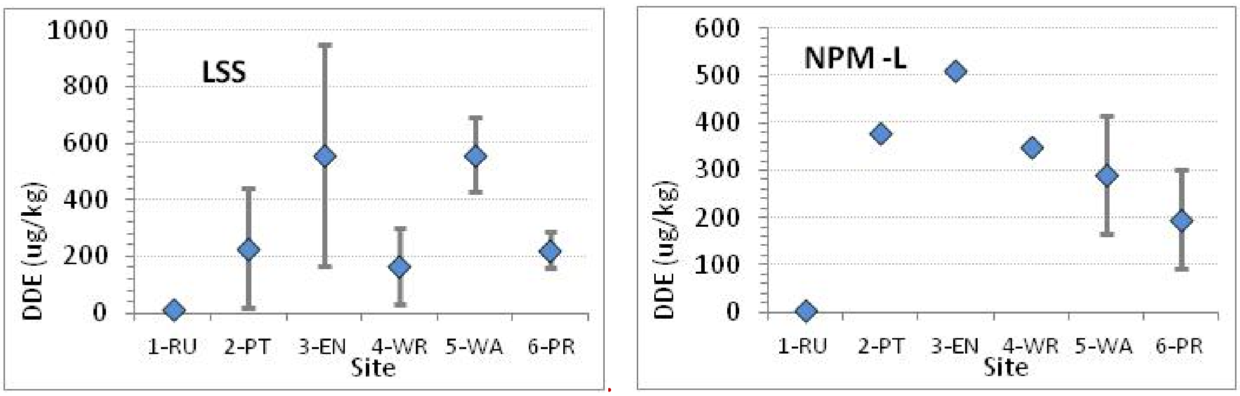

Figure 4. Results for DDE in whole largescale sucker (LSS) and fillet of larger-sized northern pikeminnow (NPM-L) from six sites in the mid-Columbia River.
Vertical bars: 95% confidence interval for mean values.

The large confidence intervals in Figure 4 are consistent with the high variability common to fish tissue samples. Despite the high variability, results show that DDE increases in Lake Pateros where the Okanogan River enters the Columbia River, and continue to increase into Lake Entiat before decreasing in fish from the Wenatchee Reach.

In general, levels of PCBs, PBDEs, and PCDD/Fs in various species also increased from upstream to downstream sites. Yet this pattern for other chemicals was weaker and more variable, and the influence of the Okanogan River was less clear, as is seen with DDE.

Mercury in fish followed no spatial patterns within species. Northern pikeminnow had the highest levels (147-620 ug/kg), which were two to five times higher than in other species regardless of location. Concentrations of mercury in 10 of the 18 samples of northern pikeminnow were greater than 400 ug/kg. This concentration corresponds to Health's statewide consumption advisory for northern pikeminnow: to consume no more than two meals per month.

The data set for DDE in largescale suckers was adequately robust for statistical testing. The Kruskal-Wallis ANOVA test showed that concentrations of DDE in Rufus Woods Lake fish were significantly less than levels seen at other sites. Levels in Wanapum Lake fish were greater than those found in fish from Priest Rapids Lake, the Wenatchee Reach, and Rufus Woods Lake. Similar testing was not pursued for northern pikeminnow or other species, due to small sample sizes.

2013 Ecology and 2009 Energy Results

The U.S. Department of Energy (Energy) sampled many fish during 2009 to address various risks from the Hanford Nuclear Reservation (Energy, 2012). Multiple species of fish were collected from four study areas of the mid-Columbia River from Wanapum Lake to Lake Wallula. Most of these areas were downstream of the FFCMP 2013 sampling. When Energy's results were combined with Ecology's 2013 results, a broader picture of contaminant concentrations in mid-Columbia River fish emerges. In general, levels of DDE and PCBs in fish from the Hanford Reach were similar to levels found at other mid-Columbia River sites during the 2013 study, with the possible exception of higher PCB concentrations in mountain whitefish from the Hanford Reach.

Figure 5 plots mean values of DDE and t-PCB for two species of fish common to both studies at various sites: mountain whitefish and common carp. Results are ordered by site from upstream to downstream and indicated by site abbreviation, river mile, and sample year.

The high variability associated with these results, as indicated by the large 95% confidence intervals, suggests that differences among most sites are not statistically significant. Qualitatively, concentrations of DDE in Rufus Woods Lake mountain whitefish are quite low and then increase downstream of the confluence with the Okanogan River. The pattern for DDE in common carp shows the high levels in Lake Pateros (site of Okanogan River confluence) and then slight decreases in downstream direction. Concentrations of PCBs in mountain whitefish show a general increase in a downstream direction and then a slight decline as the upstream areas of Lake Wallula are reached. Concentrations of PCBs in common carp show a similar pattern, although the fish collected at the McNary NWR show higher concentrations.

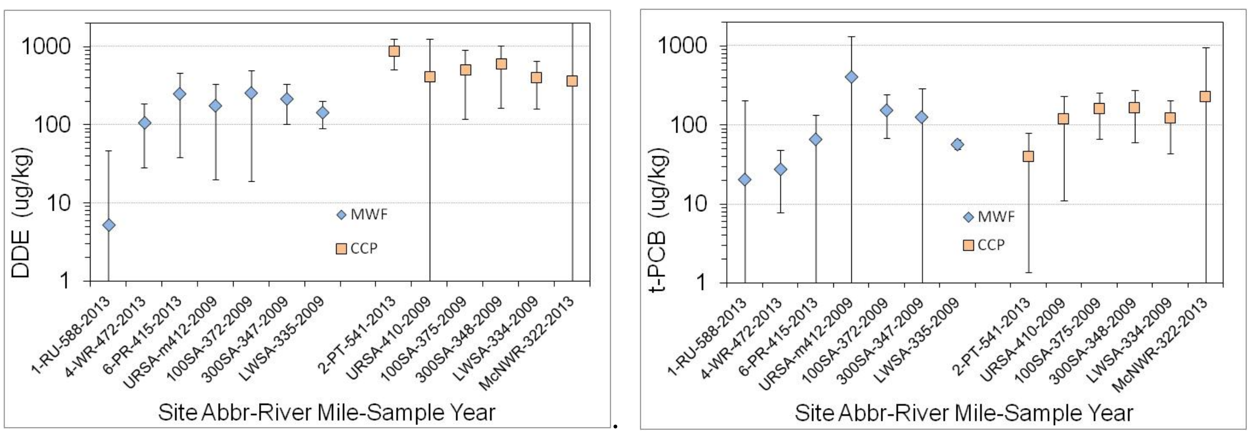

Figure 5. Comparison of mean values for DDE and t-PCB in fillets of mountain whitefish (MWF) and common carp (CCP).

Vertical bars: 95% confidence intervals for mean values.

Site abbreviations:
1-RU: Rufus Woods Lake; 4-WR: Wenatchee Reach; 6-PR: Priest Rapids Lake; URSA: Upriver Sub-Area, I-90 Bridge to Vernita Bridge; 100SA: 100 Area Sub-Area, Vernita Bridge to south end of White Bluffs area; 300SA: 300 Area Sub-Area, South end of White Bluffs area to Richland; LWSA: Lake Wallula Sub-Area, Richland to McNary Dam; McNWR: McNary National Wildlife Refuge.

Temporal Trends

Results from 2013 and historical studies were examined for clues to changes in concentrations of contaminants in fish over time. Few chemicals had data records that could be evaluated, yet DDE showed the most promise for further evaluation, partly because of its relatively high levels. For DDE, five sets of site-species groups of data were found that might show changes over time. For PCDD/Fs, two sets of site-species groups from Lake Wallula were used to show differences between two time periods. While Lake Wallula was not one of the mid-Columbia River sample sites in 2013, some results from the 2013 work in McNary NWR were used to represent Lake Wallula.

Figure 6 shows results for DDE from 2013 and previous years with the site, species, and sample year. Again, the mean values and 95% confidence interval for replicate samples demonstrate the high variability associated with organic contaminants in fish and small sample sizes. Most of the data from historical studies are results from single samples, so variability could not be characterized. No obvious temporal trend was present for any of the site-species groups, so statistical testing was not pursued. Future sampling should provide larger sample sizes to help detect temporal trends in DDE.

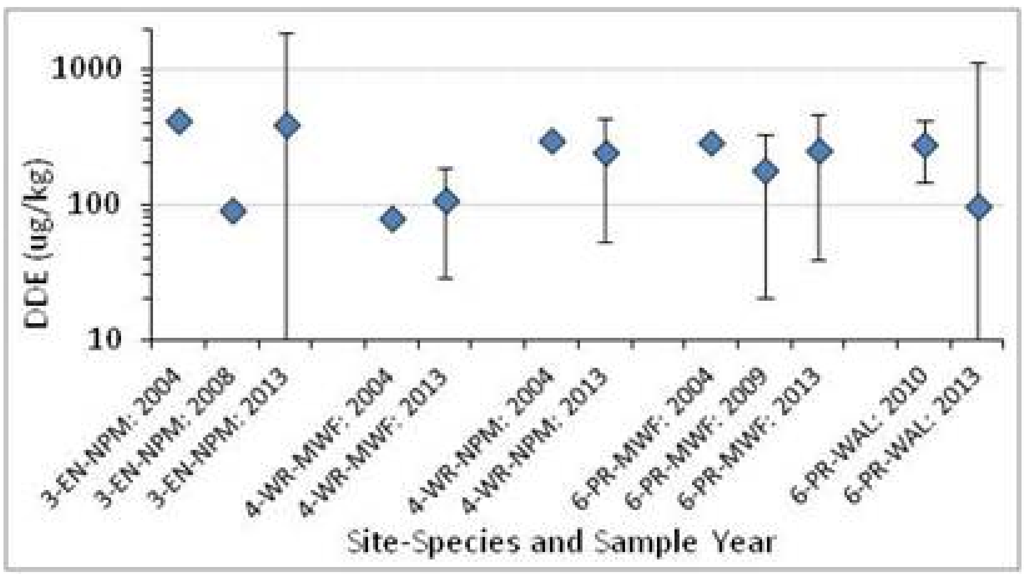

Figure 6. Results for five cases of DDE over time in northern pikeminnow (NPM), mountain whitefish (MWF), and walleye (WAL) from three sites in the mid-Columbia River.
Vertical bars: 95% confidence intervals for mean values.

Figure 7 shows results for TCDD-TEQ in two species from Lake Wallula from 2013 and 1990. The 1990 data are from a study that sampled sport fish in the mid-Columbia River (Serdar et al, 1991). Qualitatively, a change over time in TCDD-TEQ appears to have occurred. Again, the mean values and 95% confidence interval show the high variability associated with these small sample sizes. Future sampling could provide more robust data to help quantitatively detect temporal trends.

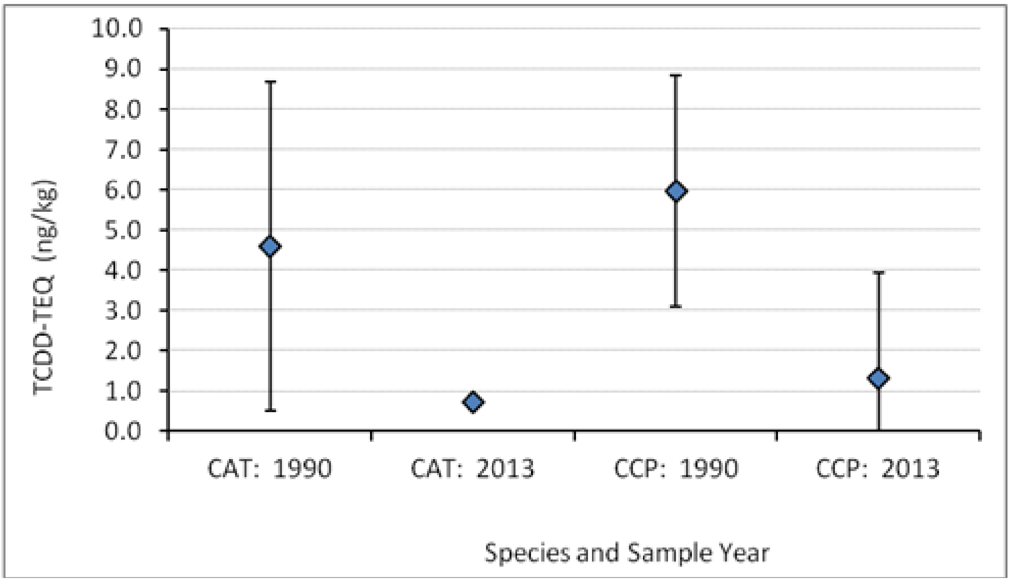

Figure 7. Results for TCDD-TEQ in channel catfish (CAT) and common carp (CCP) from Lake Wallula between 1990 and 2013.

Vertical bars: 95% confidence intervals for mean values.

McNary National Wildlife Refuge

Figure 8 shows results for key analytes along with the site and species codes. Carp and bass from the Burbank Slough Unit (SLU) had low concentrations of all contaminants. The Burbank Slough Unit is a series of four large ponds adjacent to, yet separate from, the confluence of the Columbia and Snake Rivers. Most fish from waters directly connected to the Snake and Columbia Rivers had elevated levels of t-DDT and t-PCB. Many of these samples exceeded FTECs for DDE and t-PCBs. Concentrations of t-PBDE in catfish and carp (16-40 ug/kg) are in the 80th - 90th percentile of levels in fish from across Washington (Figure 3). Bass had lower levels of organic contaminants, likely due to their smaller size, younger age, and lower lipid content.

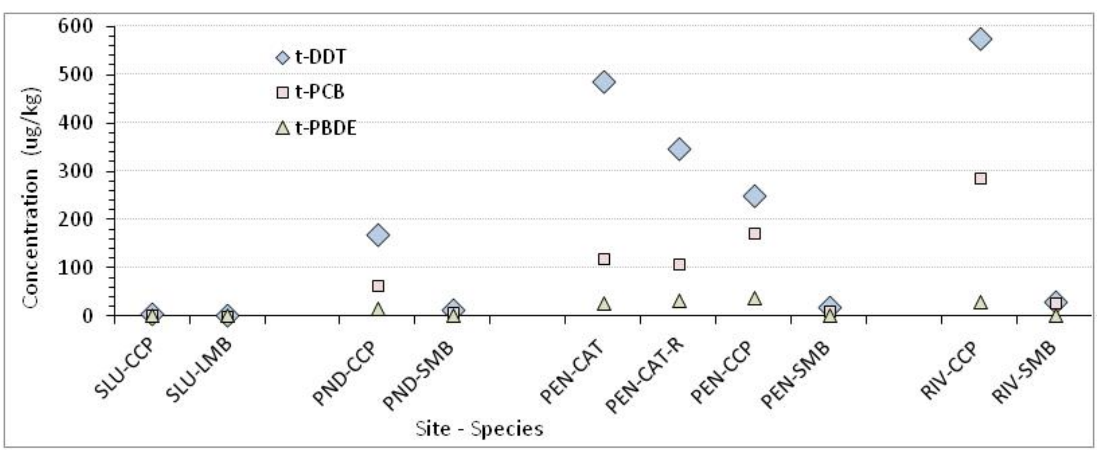

Figure 8. Results for t-DDT, t-PCB, and t-PBDE in common carp (CCP), largemouth bass (LMB), smallmouth bass (SMB), and channel catfish (CAT) from McNary NWR.

Five samples of carp and catfish were analyzed for PCDD/Fs. The mean value for 2,3,7,8-TCDD, the most toxic congener, was 0.227 ng/kg, about 3.5 times higher than Washington's FTEC. The mean value for TCDD-TEQ was 0.764 ng/kg, which is nearly 12 times higher than Washington's FTEC. Most samples that were analyzed for PCDD/Fs did not meet FTECs for 2,3,7,8-TCDD and TCDD-TEQ.

Other pesticides were detected only in common carp and catfish while mercury was found in all samples. Detected pesticides included chlordanes, chlorpyrifos, dacthal, dieldrin, and hexachlorobenzene. Of these pesticides, only dieldrin in common carp from the Snake River Unit did not meet Washington's FTEC (0.65 ug/kg). Levels of mercury were typical of those seen in these species and all were less than Washington's FTEC (770 ug/kg).

Liberty Lake

Table 3 shows results for walleye from Liberty Lake. Samples that did not meet Washington's water quality standards for t-PCB, DDE, and PCDD/Fs are shown in bold. All samples met Washington's FTEC for mercury (770 ug/kg). However, two of the samples exceeded both EPA's Recommended Criterion of 300 ug/kg (EPA, 2001) and the threshold of 400 ug/kg which corresponds to the two-meals-per-month advice that Health established for the statewide fish consumption advisory for bass (McBride, 2003).

Concentrations of mercury, t-PCB, and t-DDT were strongly correlated with age and fish size, even though the sample size was small. The strong correlations between age and contaminant concentrations were influenced by creating composite samples based on age, rather than length: WDFW provided age data along with the fish they collected for chemical analyses.

These results, along with results from statewide contaminant sampling of walleye, were given to WDFW and Health in 2014 so they could begin addressing management of the lake's walleye fishery and assessing health risks to humans consuming walleye.

Table 3. Results for walleye from Liberty Lake.

Sample ID (1401003-)	Mercury (ug/kg)	t-PCB (ug/kg)	t-DDT (ug/kg)	t-PBDE (ug/kg)	TCDD-TEQ (ng/kg)	2,3,7,8-TCDD (ng/kg)	Lipid (%)	Mean* Total Length (mm)	Mean* Weight (g)	Mean* Age (yrs)
13	156	**6.7**	1.8	-	-	-	0.6	471	1018	4.4
14	459	**35**	**35.1**	4.99	**0.122J**	0.033J	0.2	644	2883	9.4
15	615	**118**	**82.5**	-	-	-	2.3	614	2333	17.0
mean	410	**53.2**	**39.8**	4.99	**0.122J**	0.033J	1.0	576	2078	10.3
FTEC	770	5.3	31.6**		0.065	0.065				

Bold values do not meet Washington's water quality standards.
* Mean of the average length, weight, or age of individual fish that were used in each composites sample.
** Value is for DDE, the main contributor to t-DDT.
J: estimated value.

Conclusions

Mid-Columbia River

- Fish from the mid-Columbia River continue to show elevated concentrations of organic contaminants. No sites met Washington's FTECs for DDE, PCBs, and TCDD-TEQ; except for DDE in fish from Rufus Woods Lake. Common carp from Lake Pateros did not meet the FTEC for 2,3,7,8-TCDD.

- Concentrations of DDE in fish significantly increased in Lake Pateros where the Okanogan River enters the Columbia River, and continued increasing into Lake Entiat before decreasing in the Wenatchee Reach.

- In general, levels of other organic contaminants in fish also increased from upstream to downstream sites. Yet this pattern was weaker and more variable, and the influence of the Okanogan River was less clear, as was seen with DDE.

- Mercury levels in fish followed no spatial patterns within species. Northern pikeminnow had the highest concentrations: two to five times higher than in other species, regardless of location.

- The high variability and small sample sizes associated with these data made detection of differences over time difficult. Temporal trends were not seen in levels of DDE between 2004 and 2013 in five cases of comparable data. However, levels of TCDD-TEQ in common carp and channel catfish in Lake Wallula appeared to be lower in 2013 than in 1990.

- Only 45% of target samples were collected because of challenges in getting enough samples of target species in desired size ranges. This reduced the ability of this project to meet its objectives of detecting spatial and temporal changes.

McNary National Wildlife Refuge

- Most fish from waters directly connected to the Snake and Columbia Rivers had elevated concentrations of t-DDT, t-PBDE, and t-PCB, while fish from the pond-like Burbank Slough Unit had low levels of all contaminants.

- Other pesticides, including chlordanes, chlorpyrifos, dacthal, dieldrin, and hexachlorobenzene, were detected only in common carp and catfish. Only dieldrin was above the FTEC. Mercury was detected in all samples at levels typically seen in these species.

Liberty Lake

- Walleye did not meet Washington's water quality standards for t-PCB, DDE, and PCDD/Fs. While all samples met Washington's FTEC for mercury, two of the samples did not meet both EPA's Recommended Criterion of 300 ug/kg (EPA, 2001) and the threshold of 400 ug/kg used in the statewide fish consumption advisory for bass (McBride, 2003).

Recommendations

- Future sampling of mid-Columbia River fish for spatial and temporal trend analyses should focus on the sites, species, and fish size ranges that are comparable to the 2013 samples.
- Species of greatest value would be those that are more widespread and abundant, such as: largescale sucker, northern pikeminnow, and mountain whitefish. The use of multiple species provides "weight of evidence" approach in detecting trends.
- A minimum of seven field replicate composite samples of a single fish species per site should be collected in future efforts. Where data are available, statistical power analyses should be done to help determine appropriate sample sizes needed for trends analyses.
- Based on contaminant concentrations and the ages of fish sampled in 2013, re-sampling fish at a frequency of 5-10 years seems appropriate for temporal trends analyses.
- Results from Liberty Lake sampling should be used by state and local groups to help manage the lake's walleye fishery and address concerns about risks to human health from consumption of walleye.
- Results from multiple recent fish tissue studies in the mid-Columbia River should be included in the next section 303(d) Water Quality Assessment conducted by Ecology. These studies include:
    - Ecology's 2013 Freshwater Fish Contaminant Monitoring Program (FFCMP).
    - EPA's 2008-2009 survey.
    - U.S. Department of Energy's 2009 remedial investigation.

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Appendix. Glossary, Acronyms, and Abbreviations

Glossary

Analyte: A substance or constituent being measured in an analytical procedure (parameter). A physical, chemical, or biological property whose measured value help determine the characteristics of something of interest.

Clean Water Act: A federal act passed in 1972 that contains provisions to restore and maintain the quality of the nation's waters. Section 303(d) of the Clean Water Act establishes the TMDL program.

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

Fish Tissue Equivalent Concentration (FTEC): The concentration of a contaminant in fish tissue that equates to Washington's water quality standard for toxic substances for the protection of human health. Washington uses the National Toxics Rule Water Quality Criteria for the protection of human health. The FTEC is calculated by multiplying the contaminant-specific Bioconcentration Factor (BCF) times the contaminant-specific National Toxics Rule Water Quality Criterion for water.

Spatial: Relating to space, location, and distance, such as between two sampling sites.

Temporal: Relating to time, such as between one year and another.

Total Maximum Daily Load (TMDL): Water cleanup plan. A distribution of a substance in a waterbody designed to protect it from not meeting (exceeding) water quality standards. A TMDL is equal to the sum of all of the following: (1) individual wasteload allocations for point sources, (2) the load allocations for nonpoint sources, (3) the contribution of natural sources, and (4) a Margin of Safety to allow for uncertainty in the wasteload determination. A reserve for future growth is also generally provided.

Trend: A meaningful change or difference that can be measured and differentiated from measurement error. Often used in the context of time (temporal trend) or space (spatial trend).

Water Quality Assessment (WQA): Washington's Water Quality Assessment lists the water quality status for water bodies in the state. This assessment meets the federal requirements for an integrated report under Sections 303(d) and 305(b) of the Clean Water Act. The assessed waters are grouped into categories that describe the status of water quality. The 303(d) list comprises those waters that are in the polluted water category, for which beneficial uses– such as drinking, recreation, aquatic habitat, and industrial use – are impaired by pollution.

303(d) list: Section 303(d) of the federal Clean Water Act requires Washington State to periodically prepare a list of all surface waters in the state for which beneficial uses of the water – such as for drinking, recreation, aquatic habitat, and industrial use – are impaired by pollutants.

These are water quality-limited estuaries, lakes, and streams that fall short of state surface water quality standards and are not expected to improve within the next two years.

Acronyms and Abbreviations

ANOVA	Analysis of Variance
BUR	Burbot (*Lota lota*)
CAT	Channel catfish (*Ictalurus punctatus*)
CCP	Common carp (*Cyprinus carpio*)
CLM	Chiselmouth (*Arocheilus alutaceus*)
DDD	Dichloro-diphenyl-dichloroethane
DDE	Dichloro-diphenyl-dichloroethylene
DDT	Dichloro-diphenyl-trichloroethane
Ecology	Washington State Department of Ecology
EPA	U.S. Environmental Protection Agency
FCA	Fish Consumption Advisory
FFCMP	Freshwater Fish Contaminant Monitoring Program
FTEC	Fish tissue equivalent concentration
Health	Washington State Department of Health
J	Estimated value
LMB	Largemouth bass (*Micropterus salmoides*)
LSS	Largescale sucker (*Catostomus macrocheilus*)
MEL	Manchester Environmental Laboratory
MWF	Mountain whitefish (*Prosopium williamsoni*)
NPM	Northern pikeminnow (*Ptychocheilus oregonensis*)
NWR	McNary National Wildlife Refuge
PBDE	Polybrominated diphenyl ether
PCB	Polychlorinated biphenyl
PCDD/F	Polychlorinated dibenzo-p-dioxin and -furan
PEA	Peamouth (*Mylocheilus caurinus*)
RBT	Rainbow trout (*Oncorhynchus mykiss*)
SMB	Smallmouth bass (*Micropterus salmoides*)
SV	Screening value
t-DDT	Total DDTs
t-PBDEs	Total PBDEs
t-PCBs	Total PCBs
TCDD	2,3,7,8-tetra-chlorinated dibenzo-p-dioxin
TEQ	Toxicity equivalent
TMDL	Total Maximum Daily Load
U	Not detected at the reported value
UJ	Undetected at the estimated reported value
USFWS	U.S. Fish and Wildlife Service
WAC	Washington Administrative Code
WAL	Walleye (*Sander vitreus*)

WDFW	Washington Department of Fish and Wildlife
WQA	Water Quality Assessment
WRIA	Water Resource Inventory Area

Units of Measurement

<	less than
>	greater than
=	equal to
ft	feet
g	gram, a unit of mass
kg	kilograms, a unit of mass equal to 1,000 grams
mg	milligram
mg/kg	milligrams per kilogram (parts per million)
mm	millimeter
ng/kg	nanograms per kilogram (parts per trillion)
ug/g	micrograms per gram (parts per million)
ug/kg	micrograms per kilogram (parts per billion)