

Measuring Mercury Trends in Freshwater Fish in Washington State

2009 Sampling Results



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Measuring Mercury Trends in Freshwater Fish in Washington State

2009 Sampling Results

by

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Waterbody Numbers: see Appendix A

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Table of Contents

	Page
List of Figures and Tables	5
Abstract	7
Acknowledgements	8
Introduction	9
Background	9
Criteria for Human Health	9
National Toxics Rule	9
EPA Recommended Criterion	10
Previous Ecology Studies on Mercury in Freshwater Fish from Washington	10
Methods	13
Study Design	13
Site Information	13
Sample Collection	15
Field Procedures	15
Fish	15
Sediment	16
Water	1/
Data Quality Assessment	17
	10
Results	19
Fish	19
Individual Bass	19
Composite Fish	24
Water	23
Discussion	
Mercury Levels and Human Health Criteria Exceedance	
Standard-Size Bass Mercury Concentrations and Comparison to Previous Mer	20
Trends Years	28
Correlations between Mercury and Environmental Variables.	
Trends Assessment.	
Temporal	31
Spatial	32
Conclusions	35
Recommendations	36
References	37

Appendices	41
Appendix A. Sample Site Descriptions	42
Appendix B. Quality Assurance Data	43
Fish Tissue Quality Control (QC)	43
Sediment QC	45
Water QC	49
Appendix C. Biological, Sediment, and Water Quality Data	53
Appendix D. Statistical Analyses	59
Appendix E. Fish Tissue Data Evaluation by Ecology and DOH	63
Ecology	63
DOH	63
Appendix F. Glossary, Acronyms, and Abbreviations	65

List of Figures and Tables

Figures

Figure 1.	Frequency Distribution of Mercury in Washington State Freshwater Fish Tissue	11
Figure 2.	Mercury Trends Study Locations for 2009	14
Figure 3.	Total Lengths of Individual Bass Collected in 2009.	20
Figure 4.	Weights of Individual Bass Collected in 2009.	21
Figure 5.	Mercury Concentrations of Individual Bass Collected in 2009.	22
Figure 6.	Boxplot of Mercury Concentrations in Individual Bass	23
Figure 7.	Mercury Concentrations in Composite Fish Samples.	24
Figure 8.	Average Mercury Concentrations in Sediments	25
Figure 9.	Average Percent Fines and Average Percent Total Organic Carbon in Sediments	25
Figure 10	. Frequency Distribution of All Data for Mercury in Largemouth and Smallmouth Bass Tissue in Ecology's EIM Database (1984 – 2009).	27
Figure 11	. Estimated Standard-Size (356-mm) Bass Mercury Concentrations from Mercury Trends Waterbodies Sampled during 2005 – 2009	29
Figure 12	. Temporal Analysis of Mercury Concentrations in Lake Whatcom Bass	32
Figure 13	. Mercury Concentrations in Standard-Size Bass from Study Lakes during 2005 – 2009.	33

Tables

Table 1.	Physical Information on Study Locations for 2009.	. 14
Table 2.	Summary of Collection Goals for 2009	. 15
Table 3.	Analytes and Analytical Methods	. 17
Table 4.	Summary Statistics for Individual Bass Lengths, Weights, Ages, and Mercury Concentrations.	. 19
Table 5.	Water Sample Data from Upper and Lower Water Column Depths.	. 26
Table 6.	Coefficients of Determination for Simple Linear Regressions of Mercury Concentrations with Fish Size and Age.	28
Table 7.	Matrix Displaying Pearson Correlation Coefficient Values for Relationships between Standard-Size Bass Mercury Levels and Environmental Parameters	. 30

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Abstract

This report summarizes findings from the fifth year of a long-term monitoring program by the Washington State Department of Ecology to assess mercury levels in freshwater fish tissue statewide. The monitoring program began in 2005 with the purpose of identifying temporal changes in environmental mercury levels as state reduction strategies are implemented.

A total of 50 individual bass and 25 composite fish samples of seven different species were collected and analyzed for total mercury in the fall of 2009. Fish were obtained from Banks, Failor, Pierre, Vancouver, and Whatcom Lakes, as well as the Snake River. Sediment and water samples were also collected and analyzed for parameters that may affect mercury accumulation in fish.

Mercury was detected in all individual bass samples and in 92% of composite samples. Mercury concentrations in individual bass ranged from 40 - 907 ppb, with a median of 163 ppb. The highest concentrations were found in Lake Whatcom fish.

Mercury concentrations were higher than Washington's water quality standard of 770 ppb in 4% of individual bass (2 of 50 samples). Concentrations were higher than the human-health-based EPA recommended criterion of 300 ppb in 14% of individual bass (7 of 50 samples) and 4% of composite samples (1 of 25 samples).

 Log_{10} mercury levels in Lake Whatcom bass collected in 2009 were 35% lower than fish collected from the lake in 2000, at a given log_{10} length. However, temporal differences in Lake Whatcom bass may be a result of seasonal variation, as fish from 2000 were collected in the spring.

Mercury levels in bass measured over the last five years of sampling (2005 - 2009) were significantly correlated with sediment mercury (+), water temperature (+), water pH and alkalinity (-), and lake volume, surface area, and drainage area (-).

As previously reported, mercury levels in standard-size bass from Western Washington were significantly higher than from Eastern Washington waterbodies, with a mean difference of 176 ppb.

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Introduction

Background

Mercury is a naturally occurring element, but its cycle has been greatly altered by humans. The persistent, bioaccumulative, and toxic (PBT) properties of mercury have led to increased concern over its present levels in the environment.

Environmental releases of mercury occur through natural processes and human actions. Natural sources include weathering of mercury-bearing rocks and soil, volcanic activity, forest fires, and degassing from aquatic and terrestrial surfaces. Anthropogenic releases come from combustion of fossil fuels, metals production, and industrial processes. Human actions have increased the amount of mercury cycling in the environment three-to five-fold since the beginning of the industrial age (Selin, 2009).

Mercury has highly toxic effects in humans, with developing fetuses and young children most at risk. It predominantly affects the central nervous system and can lead to problems within cardiovascular systems.

To address concerns over mercury in Washington State, the *Washington State Mercury Chemical Action Plan* (Peele, 2003) was developed in 2003 by the Washington State Departments of Ecology (Ecology) and Health (DOH). The mercury Chemical Action Plan identifies current sources of mercury in Washington State and recommends strategies for reducing anthropogenic mercury occurrences and exposures.

Ecology's Environmental Assessment Program began monitoring mercury in freshwater fish tissue (2005) and sediment cores (2006) in order to identify temporal changes in environmental mercury levels throughout the state as chemical action plan reduction strategies are implemented. This report summarizes the fifth year of mercury monitoring in freshwater fish tissue.

Criteria for Human Health

There are several fish tissue criteria that are used to evaluate mercury levels in fish. The National Toxics Rule (NTR) criterion and the Environmental Protection Agency recommended criterion are both useful in assessing mercury levels of waterbodies. However, Ecology has adopted the NTR as Washington State's water quality standard and thus is the criterion used to determine whether a waterbody meets water quality standards. The following sections provide more information on these two criteria. Appendix E describes how Ecology and DOH evaluate mercury levels in fish tissue differently.

National Toxics Rule

The National Toxics Rule (40CFR131.36) issued human-health-based water quality criteria to states in 1992 (CFR, 2004), and Ecology adopted the NTR criterion as the state's water quality standard for mercury. The NTR criterion for mercury in freshwater fish tissue is 770 ppb.

Ecology's interpretation of the NTR for mercury in freshwater fish tissue was updated in 2010 to include human exposure from mercury in drinking water and fish tissue. The previous interpretation (825 ppb) was based on exposure via fish consumption only. The freshwater mercury NTR criterion is based on a practical bioconcentration factor of 5,500 and a fish consumption rate of 18.7 grams/day (g/d).

EPA Recommended Criterion

In 2001, EPA published a recommended water quality criterion for methylmercury to be used as guidance by states and tribes in the protection of human health (EPA, 2001). This value was determined to be 0.3 mg methylmercury/kg freshwater fish tissue (300 ppb), based on a fish consumption rate of 17.5 g/d for the general human adult population. This recommended criterion describes the level of mercury in freshwater fish that should not be exceeded in order to protect the general fish-consuming population. Although values are expressed as methylmercury, EPA recommended states and tribes analyze total mercury in fish tissue and make the conservative assumption that all mercury is present as methylmercury.

Previous Ecology Studies on Mercury in Freshwater Fish from Washington

Serdar et al. (2001) conducted a study of mercury levels in 273 fish and signal crayfish from Lake Whatcom in 2000. Smallmouth bass had the greatest mercury levels of the species analyzed, with an overall average concentration of 490 ppb. Twelve of the bass and one yellow perch exceeded the NTR criterion, which was 825 ppb at the time of publication. This study recommended a statewide monitoring program to routinely assess mercury levels in fish from Washington State lakes.

In response to the Lake Whatcom results, Ecology conducted a statewide study on mercury levels in bass during 2001-02. A total of 185 fish (largemouth and smallmouth bass) from 18 lakes and two rivers across Washington State were evaluated for mercury as part of the screening survey (Fischnaller et al., 2003). Twenty-three percent of fish contained mercury levels exceeding the 300 ppb EPA recommended criterion. Only one fish, a ten-year-old largemouth bass from Samish Lake, exceeded the NTR criterion, with a mercury level of 1280 ppb.

The 2001-02 statewide screening survey was the first study to extensively characterize mercury concentrations throughout freshwater areas of Washington. DOH issued a statewide fish consumption advisory in 2003 for largemouth and smallmouth bass as a result of this study (McBride, 2003).

In 2005, Ecology began a long-term monitoring program for mercury in freshwater fish tissue. The current report summarizes the fifth year of sampling results from this program. Results from the first four years of the Mercury Trends monitoring program were reported by Furl et al. (2007), Furl (2007a), Furl and Meredith (2008), and Furl et al. (2009). Mercury concentrations were determined in individual bass and composite samples of other fish species from approximately six waterbodies per year. Mercury levels in bass ranged from 17 - 1,800 ppb

over the course of the first four years of sampling. The maximum concentration came from a four-year-old bass specimen collected in 2007 from Lake Ozette. Concentrations in individual bass infrequently exceeded the NTR criterion (825 ppb at the time of the publications), ranging from 0 - 7% of the individual bass analyzed. Concentrations in individual bass exceeding EPA's recommended criterion (300 ppb) varied among the years, from 8 - 73% of individual bass samples.

Figure 1 displays a frequency distribution graph generated from all Washington freshwater fish tissue mercury data available in Ecology's Environmental Information Management (EIM) database. Data were retrieved in May 2010 and include only general environmental study results (n = 1876). Samples qualified as undetected (U) were set to zero for the frequency distribution graph.



Figure 1. Frequency Distribution of Mercury in Washington State Freshwater Fish Tissue. (Source: Ecology's EIM database as of May 2010).

Washington mercury values ranged from non-detect – 1,920 ppb, with a median of 115 ppb. A total of 97% of the samples were below the NTR criterion of 770 ppb. Approximately 82% of the samples were below EPA's recommended criterion of 300 ppb.

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Methods

Study Design

Fish, sediment, and water samples were collected from six waterbodies during the fifth year of this project.

Specific goals of the study were to:

- Measure mercury levels in fillets of 10 individual bass and/or walleye from six waterbodies per year for five years, for a total of 30 sites. Sampling is to be repeated at each site every five years for long-term trend assessments.
- Collect ancillary data at each site to better understand patterns, dynamics, and changes in fish tissue mercury levels over space and time. Ancillary data includes:
 - Fish length, weight, sex, and age.
 - Alkalinity, dissolved organic carbon (DOC), and chlorophyll concentrations from the top and bottom of water column; vertical profiles of temperature, dissolved oxygen, conductivity, and pH.
 - Three surficial sediment grabs analyzed for mercury, total organic carbon (TOC), and grain size.
 - Lake morphological and hydrological characteristics.
- Determine mercury concentrations in composite fillet samples of three to five individual fish from two other fish species present at sampling sites where bass/walleye are collected.

Detailed study design information can be found in the Quality Assurance (QA) Project Plan (Seiders, 2006).

Site Information

Five lakes and one river site were sampled in 2009: Banks, Failor, Pierre, Vancouver, and Whatcom Lakes, and the Snake River near Clarkston (Figure 2). Waterbodies were selected for sampling based on several criteria, including: popularity among anglers, availability of target fish species, historical contamination issues, and inclusion in Ecology's 2001/2002 screening survey for mercury (Fischnaller, 2003). The QA Project Plan contains detailed information on site selection considerations (Seiders, 2006). Physical information for each waterbody sampled in 2009 is presented in Table 1.



Figure 2. Mercury Trends Study Locations for 2009.

Name	Banks Lake	Failor Lake	Pierre Lake	Snake River*	Vancouver Lake	Lake Whatcom
County	Grant	Grays Harbor	Stevens	Asotin	Clark	Whatcom
Drainage (sq mi)		4.89	26.8			55.9
Altitude (ft)	1,570	117	2,005	700	9	315
Surface Area (acres)	27,000	65	110		2,300	5,000
Lake Volume (acre-ft)	1,300,000	500	3,000			770,000
Maximum Depth (ft)	85	22	75	77	12	330
Mean Depth (ft)	47	8	28	50	3	150

Table 1. Physical Information on Study Locations for 2009.

*Snake River near Chief Timothy Park, WA.

Sample Collection

Table 2 displays a summary of the fish, sediment, and water collection goals met for 2009. Ecology and the Washington Department of Fish and Wildlife (WDFW) collected fish from the six sampling sites in September and October of 2009. In all, 153 fish were retained, encompassing nine species. Bass were collected from five sites, but crews were unable to collect bass from Vancouver Lake. The Snake River site was added to the sampling plan after fishcollection efforts at one of the original sites, Conconully Reservoir, were unsuccessful.

Sediment and water samples were collected from the five lakes during July and August of 2009 in an attempt to capture lake-stratified conditions. No sediment or water samples were collected from the Snake River due to the late addition of the site to the sampling plan.

	Waterbody							
Collection Goal	Banks Lake	Failor Lake	Pierre Lake	Snake River	Vancouver Lake	Lake Whatcom		
10 individual bass or walleye	+	+	+	+	NA	+		
3 composites (3-5 fish) for each of 2 different species	+	+^	NA	NA	+	+		
3 sediment samples	+	+	+	NA	+	+		
2 water samples	+	+	+	NA	+	+		
1-2 Hydrolab profile	NA	+	NA	NA	+	+		

Table 2. Summary of Collection Goals for 2009.

NA = Collection goal not attained.

+ = Collection goal met.

 $+^{\wedge}$ = Three composites of one species, one composite of the other.

Field Procedures

Fish

Methods for collection, handling, and processing fish tissue samples are described in:

- EPA's Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories (EPA, 2000).
- Ecology's Environmental Assessment Program's (EAP) Standard Operating Procedures for *Resecting Finfish Whole Body, Body Parts or Tissue Samples* (Sandvik, 2006).

Fish were collected using gill nets and an electrofishing boat. Fish were inspected to ensure there was no visible damage to skin or tissue. After positive identification, fish selected for sampling were euthanized by blunt force to the head. Fish were rinsed in ambient water, weighed to the nearest gram, and total length was measured to the nearest millimeter. Specimens were individually wrapped in foil (dull side in) and packaged in zipper-lock bags with identification labels. Packaged specimens were immediately packed in ice and held for a maximum of 72 hours during transport to Ecology Headquarters in Lacey, Washington. Specimens were stored frozen until later processing.

During processing, partially thawed fish were cleaned of slime and scales, rinsed in tap water, and rinsed with deionized water. Skin-on fillets were removed from one or both sides of the fish and cut into smaller sections. Tissue was ground three times using a Kitchen-Aid® food grinder and homogenized after each run through the grinder. After samples were a uniform color and texture, subsamples were removed and stored in clean (I-Chem 200®) glass jars. Jars labeled with laboratory identification numbers were transported to Manchester Environmental Laboratory (MEL) for analysis. Remaining homogenized tissue was archived in clean jars, labeled, and placed in cold storage at -20° C.

After tissue samples were removed, sex of the fish was determined. The appropriate age structures (either scales or otoliths) were removed from fish analyzed individually and sent to WDFW biologists for age determination.

All utensils were cleaned prior to use and after each sample was processed. Utensils were cleaned with Liquinox® and tap water, a deionized water rinse, and a rinse with 10% nitric acid. After a final deionized water rinse, utensils were dried in a fume hood.

Fish were filleted on a nylon cutting board covered with heavy-duty aluminum foil, dull side out. New foil was used after each fish to prevent cross-contamination of samples.

Sediment

The collection, handling, and processing of sediment samples was guided by *Recommended Protocols for Measuring Conventional Sediment Variables in Puget Sound* (EPA, 1986) and EAP's *Standard Operating Procedures for Freshwater Sediment Sampling* (Blakley, 2008). Three surficial sediment samples were taken from distinct areas of the waterbody.

Sediment samples were collected using a 0.02 m² stainless-steel, petite ponar grab dredge. The surface layer of water was siphoned from the sample. The top two centimeters of sediment were removed, homogenized with a stainless steel spoon, and stored in factory-provided, certified clean jars. Sediments coming in contact with the side of the ponar dredge were not retained. Samples were packed in ice and sent to MEL within 96 hours for analysis. All utensils used to collect and process sediment were cleaned using the process described above for fish processing.

Water

Two water samples were obtained from different depths at the deepest part of each lake using a one-liter Kemmerer sampler. The samples were obtained at the mid-points of the hypolimnion and epilimnion in stratified lakes. The depth of the hypolimnion and epilimnion were determined using a Hydrolab® profile to locate the thermocline. At unstratified lakes the samples were obtained at 10-15% and 85-90% of total depth. Samples were retrieved and placed in the proper pre-cleaned bottles for analysis of alkalinity, chlorophyll, and DOC. Samples were stored on ice in the field until shipment to the laboratory.

Conductivity, pH, dissolved oxygen, and water temperature were measured at the water sample sites using a Hydrolab[®] following EAP's *Standard Operating Procedures for Hydrolab*[®] *Datasonde*[®] *Multiprobes* (Swanson, 2007). All instruments were calibrated prior to field use, and Winkler titrations were performed as a measure of quality control for the dissolved oxygen readings. Water transparency was measured using a Secchi disc.

Laboratory Procedures

MEL performed all laboratory analyses with the exception of grain size, which was conducted by Columbia Analytical Services. A list of analytes and analytical methods are provided in Table 3.

Analyte	Matrix	Method
Mercury	Fish Tissue	CVAA, EPA 245.6
Mercury		CVAA, EPA 245.5
Total Organic Carbon	Sediment	PSEP-TOC
Grain Size		PSEP, Sieve and Pipette
Alkalinity		SM2320B
Dissolved Organic Carbon	Water	SM5310B
Chlorophyll		SM10200H3M

Table 3. Analytes and Analytical Methods.

CVAA = Cold Vapor Atomic Absorption.

PSEP = Puget Sound Estuary Protocol.

Total mercury is the target analyte used in this study as a surrogate for methylmercury due to the comparative simplicity and lower cost of analyzing for total mercury. Methylmercury, the bioaccumulative and toxic form of mercury in fish tissue, accounts for more than 95% of the mercury in fish tissue, where it is associated with muscle proteins (Bloom, 1995; Driscoll et al., 1994; Grieb et al., 1990).

Data Quality Assessment

Data quality was assessed by examining quality control and quality assurance procedures used during analyses. MEL provided case narratives documenting holding times, instrument calibration, method blanks, matrix spikes, laboratory duplicates, laboratory control samples, and standard reference material analyses. Case narratives are available upon request.

Assessment of the data indicated that all of the fish tissue and most of the water and sediment analyses were within measurement quality objectives (MQOs) outlined by the project plan (Seiders, 2006). The only exception to this was a laboratory duplicate sample for chlorophyll, which had a relative percent difference outside of MQOs. The native sample was qualified "J," as an estimate. A complete description of MQOs and quality control data is available in Appendix B.

Results

Fish

A total of 50 individual bass and 25 composite fish samples were analyzed for mercury in 2009. Results of fish size, age, and mercury concentrations are described in the following sections.

Individual Bass

Largemouth and smallmouth bass were collected from the Snake River and Banks, Failor, Pierre, and Whatcom Lakes and analyzed individually for total mercury. No bass were found at Vancouver Lake. Summary statistics of bass lengths, weights, ages, and mercury concentrations are presented in Table 4. Complete biological data for individual bass are presented in Appendix C.

Watarbody	Total Length (mm)		Weight (g)		Age (yr)		Mercury (ppb ww)	
waterbody	Min - Max	Mean (±SD)	Min - Max	Mean (±SD)	Min - Max	Mean (±SD)	Min – Max	Mean (±SD)
Banks	262 - 445	340 (68)	238 - 1216	625 (361)	3 - 11	6 (3)	68.2 - 371	129 (104)
Failor	209 - 430	296 (89)	141 - 1487	602 (557)	1 - 6	3 (2)	40.2 - 213	85.4 (66)
Pierre	311 - 442	374 (40)	468 - 1411	820 (312)	5 - 13	9 (3)	111 – 363	209 (70)
Snake	225 - 450	330 (68)	137 - 1797	672 (486)	2 - 6	3 (2)	67.6 – 337	153 (90)
Whatcom	284 - 443	348 (51)	314 - 1420	693 (359)	3 - 7	4 (1)	167 - 907	369 (277)
All Sites	209 - 450	338 (67)	137 - 1797	682 (415)	1 - 13	5 (3)	40.2 - 907	189 (171)

Table 4. Summary Statistics for Individual Bass Lengths, Weights, Ages, and Mercury Concentrations.

SD = Standard Deviation.

Bass Size

Total lengths and weights of individual bass ranged from 209 - 450 mm and from 137 - 1,797 g, respectively. Lengths and weights of individual bass are presented in Figures 3 and 4.







Figure 4. Weights of Individual Bass Collected in 2009. LMB = largemouth bass, SMB = smallmouth bass.

Fish size ranges were similar among the waterbodies. The project plan outlined target fish collection lengths of 250 - 460 mm and recommended the use of EPA's 75% *rule* (the length of the smallest fish should be at least 75% the length of the largest fish) as a rough guide in selecting fish to retain from each waterbody (Seiders, 2006; EPA, 2000). Bass were generally within target lengths outlined in the project plan; however, the 75% *rule* was not met for any of the waterbodies. The smallest fish lengths were between 45% and 70% of the largest fish.

Weight ranges of individual bass at each site varied considerably, as well. However, weight ranges were similar among waterbodies. Boxplots displaying the spread of fish length and weight data are located in Appendix C.

Bass Mercury Levels

Figure 5 displays mercury levels measured in individual bass from the 2009 waterbodies.



Figure 5. Mercury Concentrations of Individual Bass Collected in 2009.

LMB = largemouth bass, SMB = smallmouth bass. NTR = National Toxics Rule. EPA RC = EPA's Recommended Criterion. Mercury was detected in all individual bass fillet samples. Concentrations ranged from 40 - 907 ppb ww, with a mean of 163 ppb. Fish age ranged from 1 - 13 years. The highest mercury concentrations were found in two 7- and 5-year-old smallmouth bass from Lake Whatcom.

Bass collected from Pierre Lake were generally older than those collected from other waterbodies. The youngest fish and the lowest mercury concentrations were found in Failor Lake.

Figure 6 displays the minimum, maximum, and inter-quartile range of bass mercury concentrations from each of the waterbodies.



Figure 6. Boxplot of Mercury Concentrations in Individual Bass. $(q1 = 25^{th} percentile; q3 = 75^{th} percentile; LMB = largemouth bass; SMB = smallmouth bass)$

Composite Fish

Composite fish samples of species other than bass were retained from Banks, Failor, Vancouver, and Whatcom Lakes. No additional species were collected from Pierre Lake or the Snake River. Mercury concentrations measured in composite fish samples are displayed in Figure 7. Length, weight, and age data for fish included in composites are presented in Appendix C.



Figure 7. Mercury Concentrations in Composite Fish Samples.

"ND" indicates the sample result was below detection limits. CTT = cutthroat trout; LWF = lake whitefish; PEA = peamouth; RBT = rainbow trout; SHAD = shad; WAL = walleye; YP = yellow perch; EPA RC = EPA's Recommended Criterion.

Mercury was detected in 92% of composite samples. Mercury concentrations ranged from <20 - 320 ppb ww. The mean of all composite samples was 103.5 ppb. Lake Whatcom peamouth and yellow perch generally contained the highest concentrations. One large yellow perch from Lake Whatcom (not included in graph) was analyzed individually and contained a mercury concentration of 802 ppb. This perch was at least twice as large (total length = 356-mm; weight = 702 g) as other perch analyzed in composites and likely much older.

Sediment

Sediment samples were collected from each of the waterbodies except for the Snake River. Figure 8 displays average sediment mercury results. Average percent fines and TOC of sediment grabs are presented in Figure 9. Replicates are not included in averaged values.



Figure 8. Average Mercury Concentrations in Sediments. *Error bars represent one standard error.*



Figure 9. Average Percent Fines (< $62\mu)$ and Average Percent Total Organic Carbon in Sediments.

Error bars represent one standard error.

Average sediment mercury concentrations ranged from 48.7 - 162 ppb dw across the five waterbodies. Relative standard deviations of mercury averages within waterbodies ranged from 12 - 42, with the greatest variation found at the largest lakes.

Water

Water samples were taken from the epilimnion and hypolimnion of each lake. Chlorophyll, alkalinity, and DOC measured in water samples are presented in Table 5. Replicates are not included in the table below. Lake pH, dissolved oxygen, temperature, and conductivity profiles are presented in Appendix C.

Lake	Collection Date	Depth (m)	Chl-a (µg/L)	Alkalinity (mg/L)	DOC (mg/L)
Bonks	7/28/2000	6.0	3.3	57.4	1.4
Daliks	1/28/2009	16.0	2.1	61.1	1.3
Failor	7/20/2009	1.5	1.5	17.4	1.6
		4.8	6.7	20.6	1.3
Pierre	7/27/2009	4.0	2.8	135	3.8
		16.0	4.8	180	2.9
Whataom	7/22/2009	6.0	3.4	21.2	1.8
whatcom		27.0	0.6	17.9	1.6
Variation	7/21/2000	0.5	141	65.2	3.0
vancouver	7/21/2009	1.0	73.8	64.9	2.9

Table 5. Water Sample Data from Upper and Lower Water Column Depths.

Chl-a = chlorophyll-a.

DOC = dissolved organic carbon.

Discussion

Mercury Levels and Human Health Criteria Exceedance

Mercury concentrations measured in 2009 were within the range of previous Mercury Trends reports, (Furl et al., 2007; Furl 2007a; Furl and Meredith, 2008; Furl et. al., 2009) as well as statewide and national levels. The median 2009 bass mercury value (163 ppb) was lower than the median of statewide mercury in bass tissue data from Ecology studies (197 ppb; n = 646) and EPA's national median for mercury in predator fish (284.6 ppb; n = 486) (EPA, 2009).

Figure 14 displays a cumulative frequency distribution of all mercury in largemouth and smallmouth bass data available in Ecology's EIM database, along with the 2009 Mercury Trends data highlighted.





n = 699 including results from the present study, accessed May 2010, with 2009 Mercury Trends bass data represented by light orange squares. NTR = National Toxics Rule. EPA RC = EPA's Recommended Criterion. Two smallmouth bass from Lake Whatcom (4% of individual bass sampled in 2009) exceeded the NTR criterion of 770 ppb. No composite samples exceeded the NTR criterion.

Seven bass (14% of samples) were above EPA's recommended criterion of 300 ppb. One fish each from Banks Lake, Pierre Lake, and the Snake River, and four of the Lake Whatcom bass, exceeded this value. One composite fish sample (4% of samples) exceeded the EPA recommended guideline.

Relationships between Mercury and Fish Size and Age

Simple linear regressions were conducted to examine the effect of fish length, weight, and age on bass mercury concentrations. Table 6 displays linear regression coefficients of determination for those relationships. Mercury values plotted against length, weight, and age are displayed in Appendix D.

Table 6. Coefficients of Determination (r^2) for Simple Linear Regressions of Mercury Concentrations with Fish Size and Age.

Site	Length	Weight	Age
Banks	0.559	0.536	0.791
Failor	0.862	0.929	0.943
Pierre	0.098	0.034	0.074
Snake	0.586	0.618	0.894
Whatcom	0.658	0.828	0.564

Bolded values indicate statistically significant relationships (p < 0.05).

Strong positive relationships between bass mercury concentrations and fish length, weight, and age were found at all sites except for Pierre Lake. The increase of mercury levels with size and age of bass has been well documented in previous Mercury Trends reports (Furl et al., 2009; Furl and Meredith, 2008; Furl, 2007a; Furl et al., 2007).

Standard-Size Bass Mercury Concentrations and Comparison to Previous Mercury Trends Years

Mercury concentrations in bass were normalized to a standard length (356-mm) using a regression formula in order to make comparisons between waterbodies. This formula is described in detail by Fischnaller et al. (2003) and used in previous Mercury Trends reports to estimate standardized mercury concentrations. The regression formula can be used for waterbodies where total fish length displayed a significant relationship with fish mercury levels. Total length was selected over weight and age because of the ease with which length can be measured in the field (Fischnaller et al., 2003).

The following multiple regression formulas were used:

$$Log_{10} (Hg) = M + \{B1*Log_{10} (356\text{-mm})\} + \{B2*(Log_{10} (356\text{-mm}))^2\}$$
$$10^{Log_{10}(Hg)} = Hg \text{ concentration at } 356\text{-mm}$$

where: M = constant; B1 = coefficient of bass length (log₁₀ mm); and B2 = coefficient of bass length squared (log₁₀ mm²).

No relationship was found between mercury level and fish length at Pierre Lake and therefore regression coefficients were not used to calculate a standardized mercury value. A value of 190 ppb was assigned to Pierre Lake for a 356-mm bass, based on mercury concentrations in similar-sized bass at the site.

Estimated mercury concentrations for 356-mm bass from Mercury Trends waterbodies (study years 2005 – 2009) are graphed in Figure 10. Regression coefficients, products, and standard-size mercury concentrations are provided in Appendix D.



Figure 11. Estimated Standard-Size (356-mm) Bass Mercury Concentrations from Mercury Trends Waterbodies Sampled during 2005 – 2009.

* = concentrations were not estimated using multiple regression, see Appendix D for description.

Standard-size bass mercury concentrations ranged from 80.7 - 298 ppb in the 2009 waterbodies. Lake Whatcom had the highest standardized mercury level of the 2009 waterbodies, and the sixth highest standardized concentration across all sites sampled from 2005 - 2009.

Correlations between Mercury and Environmental Variables

Relationships between standardized (356-mm) bass mercury levels and sediment, water, and lake morphology variables were examined to identify possible factors influencing mercury bioaccumulation in fish.

Pearson correlations were performed on data from all Mercury Trends years combined (2005 - 2009), for a total of 25 sites. Temperature, pH, and conductivity values were taken from the bottom (0.5 - 2-m above the bottom substrate) and top (1 - 3-m below water surface) water column measurements. The epilimnion and hypolimnion samples for DOC and alkalinity were averaged together, as these values were not affected by depth. All variables were log_{10} -transformed, with the exception of bottom temperature, bottom pH, and bottom conductivity values, to improve the normality of the data.

Data were grouped three ways in consideration of spatial differences between the waterbodies: statewide, Eastern Washington sites, and Western Washington sites. Correlation coefficients of relationships between standard-size bass mercury levels and environmental variables are presented in Table 7.

Variable Parameter		Statewide Eastern Western					
Grouping	i urumeter	Сог	Correlation Coefficient				
Sadimant	Mercury	0.585	0.210	0.671			
Seament	Total Organic Carbon	0.358	0.225	-0.032			
	Alkalinity	-0.514	-0.256	-0.332			
	Chlorophyll - Top	0.137	0.135	-0.379			
	Chlorophyll - Bottom	0.170	0.314	-0.171			
	Conductivity - Top	-0.278	-0.277	-0.025			
Wator	Conductivity - Bottom	0.106	0.342	0.313			
water	Dissolved Organic Carbon	0.323	0.296	0.154			
	рН - Тор	-0.406	-0.619	-0.271			
	pH - Bottom	-0.341	-0.720	-0.019			
	Temperature - Top	0.450	0.446	0.250			
	Temperature - Bottom	-0.304	-0.387	-0.312			
	Lake Volume	-0.367	-0.658	0.232			
Morphology	Surface Area	-0.497	-0.794	0.077			
	Drainage Area	-0.493	-0.809	0.065			
	Maximum Depth	0.011	-0.315	0.436			
	Mean Depth	0.206	-0.167	0.529			

Table 7. Matrix Displaying Pearson Correlation Coefficient Values for Relationships between Standard-Size Bass Mercury Levels and Environmental Parameters.

Bolded values indicate statistically significant relationship (p < 0.05).

No environmental variable showed a consistent relationship to standardized bass mercury levels across all three datasets. Parameters showing significant relationships with bass mercury levels for at least one of the datasets were sediment mercury (+), lake alkalinity (-), hypolimnion pH (-), epilimnion temperature (+), lake volume (-), lake surface area (-), and lake drainage area (-).

Average sediment mercury values were positively correlated with bass mercury concentrations for the statewide and Western Washington datasets. Western Washington sites showed less variability in the three sediment grabs than did Eastern Washington sites, with average relative standard deviations of 16 and 27, respectively. Other studies examining mercury bioaccumulation factors have not found a consistent relationship between resident fish and sediment mercury levels (Rose et al., 1999; Hanten et al., 1998).

A negative relationship was found between water pH and fish mercury levels at the Eastern Washington sites only. Water alkalinity showed a negative relationship for the statewide dataset. Low pH and alkalinity have been associated with elevated bass mercury levels by several studies (Hanten et al., 1998; Lange et al., 1993; Simonin et al., 2008). Increases in mercury levels under low pH have been explained by enhanced microbial production in acidic waters and/or increased bioavailability of mercury in low-pH conditions (Xun et al., 1987; Wiener et al., 1990).

Lake volume, surface area, and drainage area showed inverse relationships with fish mercury levels for the statewide and Eastern Washington sites. Bodaly et al. (1993) found higher mercury bioaccumulation in smaller-sized lakes, likely as a function of warmer water temperatures. In contrast, Rose et al. (1999) reported a positive correlation between lake size and drainage area with mercury levels in bass. Inconsistent findings may reflect differences in the topography or hydrological systems, such as amount of wetlands, within a lake's watershed.

Trends Assessment

Temporal

Temporal differences in mercury levels in Lake Whatcom bass between 2000 and 2009 were examined using data from Serdar et al. (2001) and the current study. Because mercury was analyzed using a different method in 2000, the data were transformed using a formula developed by Furl (2007b) to compare to 2009 values. For more accurate values, only bass measuring \leq 400 mm in length were included in the analysis, as the correction equation is robust until that point. From the 2000 data set, only bass collected from the lake's main basin were used in order to match the 2009 collection area.

Figure 12 presents log-normalized mercury and length data for 2000 and 2009 bass collected from Lake Whatcom. Solid lines represent regressed mercury at a given length calculated from a generalized linear model using log-normalized length and a dummy variable (collection year) (Hrabik and Watras, 2002). Percent change (Δ) in mercury levels was estimated by a formula using the coefficient of the dummy variable (collection year) and standard error of that coefficient (Halvorsen and Palmquist, 1980; Kennedy, 1981).



Figure 12. Temporal Analysis of Mercury Concentrations in Lake Whatcom Bass. Solid lines represent regressed mercury slopes. Time between sampling periods = 113 months.

Log₁₀ mercury levels in 2009 Lake Whatcom bass were 35% lower than in 2000 bass at a given log₁₀ length. Bass collected from the two studies were similar in age, size, and location caught. However, bass from 2000 were collected in May, while 2009 sampling occurred in October. Therefore, temporal differences in Lake Whatcom bass may be a result of seasonal variation and not necessarily a decline in mercury levels. Conflicting information exists on the seasonality of mercury concentrations in bass. Ward and Neumann (1999) found significantly higher mercury levels in the spring compared to fall, which they attributed to increased feeding rates, higher protein content, or elevated mercury inputs in run-off during the spring. However, Slotton et al. (1995) found that bass mercury levels were greatest in the fall and winter, with higher levels of bioavailable mercury in the lake after destratification.

Spatial

Waterbodies in this study were equitably distributed across the state. Spatial trends were examined by comparing standard-size (356-mm) mercury levels in bass tissue between Eastern and Western Washington sites. Figure 13 displays standardized bass mercury concentrations from Mercury Trends years 2005 – 2009, throughout the state.

An independent samples t-test was conducted to compare differences in standardized bass mercury levels between Eastern and Western Washington waterbodies. Western Washington mercury levels (mean = 315 ppb; n = 14) were significantly higher than Eastern Washington levels (mean = 139 ppb; n = 13), with a mean difference of 176 ppb (t = -2.923; p = 0.010). This same pattern was reported in the 2007 and 2008 Mercury Trends reports (Furl and Meredith, 2008; Furl et al., 2009). Waterbodies were not randomly selected for assessment of spatial trends.



Figure 13. Mercury Concentrations in Standard-Size (356-mm) Bass from Study Lakes during 2005 – 2009.

Western Washington sites, on average, receive significantly greater annual rainfall, have lower alkalinity values, and have higher percentages of wetlands within their drainage basins than Eastern Washington sites (independent samples t-tests, p < 0.05). The higher mercury values in Western Washington may be a function of these differences. Lakes in areas of high rainfall and with low pH buffering capacity may have higher mercury burdens in biota due to the greater wet deposition inputs of mercury (Downs et al., 1998). The abundance of wetlands in a lake watershed has also been well documented as an important factor in fish mercury levels, as mercury methylation occurs in wetlands and is hydrologically transported to the lake (St. Louis et al., 1994; Rose et al., 1999; Simonin et al., 2008; and Chen et al., 2005).

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Conclusions

A total of 50 individual bass and 25 composite fish samples were analyzed for mercury in 2009, as part of the fifth year of Mercury Trends sampling. Fish were collected from Banks, Failor, Pierre, Vancouver, and Whatcom Lakes, as well as the Snake River near Clarkston, Washington. No bass were found at Vancouver Lake. Sediments and water were also collected at the sites and analyzed for parameters that may affect mercury accumulation in fish.

Mercury was detected in all individual bass samples, with concentrations ranging from 40-907 ppb ww. The median bass mercury concentration, 163 ppb, was lower than statewide and national medians. In fish composite samples, mercury detection frequency was 92%, with a range of <20-320 ppb ww. Lake Whatcom fish contained the highest mercury levels.

Additional significant findings from this study are summarized below.

- Four percent of individual bass (2 of 50 samples) did not meet (exceeded) Washington State's water quality standard (NTR criterion) of 770 ppb. Both fish were collected from Lake Whatcom. No composite samples exceeded this level. Seven bass (14% of samples) and one composite sample (4%) exceeded EPA's recommended criterion of 300 ppb.
- Mercury concentrations were positively correlated with bass length, weight, and age. Mercury concentrations in standard-size bass (356-mm) ranged from 80.7 – 298 ppb in the 2009 waterbodies. Lake Whatcom had the highest standardized mercury level of the 2009 waterbodies, and the sixth highest standardized concentration across all sites sampled from 2005 – 2009.
- Log₁₀ mercury values from Lake Whatcom bass collected in 2009 were 35% lower than bass collected from the lake in 2000, at a given log₁₀ length. However, temporal differences in Lake Whatcom bass may be a result of seasonal variation, as fish from 2000 were collected in the spring and the 2009 fish were collected in the fall.
- Bass mercury levels measured over the last five years of trends sampling (2005 2009) were significantly correlated with sediment mercury (+), water temperature (+), water pH and alkalinity (-), and lake volume, surface area, and drainage area (-) for at least one of the datasets analyzed: statewide, Eastern Washington, and Western Washington.
- In a test for spatial trends, Western Washington mercury levels in bass were significantly higher than Eastern Washington levels, with a mean difference of 176 ppb. This trend was noted in the 2007 and 2008 mercury reports, as well.

Recommendations

Ecology has completed the first five-year cycle (30 sites) of the Mercury Trends project and will begin revisiting those sites in 2010 to assess temporal changes in bass mercury levels.

Based on the first five years of data, the following changes to the sampling plan are proposed:

- Continue taking Hydrolab lake profile measurements (dissolved oxygen, temperature, conductivity, and pH), secchi disk measurements, and water collections for analysis of alkalinity and dissolved organic carbon.
- Discontinue chlorophyll-a analyses in water samples and add sulfate and major ion analyses. Data from the baseline period of the project do not support continued monitoring of chlorophyll-a, as it appears to have little effect on mercury bioaccumulation in fish. Other studies have shown sulfate and other ions to have an important control on methylation rates in freshwater lakes.
- Discontinue sediment sampling for mercury, total organic carbon, and grain size analysis. Sediment data collected during the 2005-09 baseline period provide adequate data for characterizing near-future conditions, and surficial sediments are not expected to change substantially in five years time.
- Continue collection of individual bass and/or walleye, as well as two additional species for composite analyses, at the 30 waterbodies.

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Appendices

Appendix A. Sample Site Descriptions

Site Name	Latitude*	Longitude*	WBID	County	EIM User Location ID	WRIA
Banks L	47.87904	-119.16282	WA-42-9020	Grant	Grant BANKS-F2	
Failor L	47.10894	-123.95865	WA-22-9040	Grays Harbor	FAILOR-F	22
Pierre L	48.90639	-118.13785	WA-60-9040	Stevens	PIERRE-F	60
Snake River	46.42940	-117.15210	WA-35-1010	Asotin	SNAKERDSCLARK-F	35
Vancouver L	45.67983	-122.72094	WA-28-9090	Clark	VANCOUVER-F	28
Whatcom L	48.73104	-122.32514	WA-01-9170	Whatcom	WHATCOM-F	1

Table A-1. Sampling Site Locations for the 2009 Study.

*NAD83 HARN.

WBID = Waterbody Identification Number.

EIM = Ecology's Environmental Information Management database.

WRIA = Water Resource Inventory Area.

Appendix B. Quality Assurance Data

Fish Tissue Quality Control (QC)

MEL conducted mercury analyses of fish tissue samples from December 17, 2009, through January 7, 2010. MEL received samples frozen and in good condition. All analyses were performed within EPA-established holding times. Table B-1 presents MQOs used to assess quality of the data, as outlined in the project plan (Seiders, 2006).

Table B-1.	Measurement	Ouality	Objectives	for Fish	Tissue /	Analysis.
	measurement	Quanty	Objectives	101 1 1511	1100000	. mary 515.

Parameter	Matrix	Reporting Limit	Accuracy	Check Standard (% recovery)	Duplicate Sample (RPD)	Matrix Spike (% recovery)
Mercury, total	tissue	0.017 mg/kg, wet	+/- 15% of SRM value	80-120%	<20%	75-125%

Data quality was assessed through analyses of standard reference materials, laboratory control samples, matrix spikes, and laboratory blanks. All QC tests were within MQOs for fish tissue analyses. Tables B-2 through B-5 display results of the QC tests.

Table B-2. Standard Reference Material.

Sample Number	Recovery (%)
B09L135-SRM1	109
B09L190-SRM1	112
B10A010-SRM1	111
B10A017-SRM1	111

Table B-3. Laboratory Control Samples.

Sample Number	Recovery (%)
B09L135-BS1	105
B09L190-BS1	98
B10A010-BS1	109
B10A017-BS1	107

Sample Number	Recovery	RPD
1	(%)	(%)
B09L135-MS1	100	2.0
B09L135-MSD1	97	3.0
B09L190-MS1	94	2.2
B09L190-MSD1	92	2.2
B10A010-MS1	103	2.0
B10A010-MSD1	100	3.0
B10A017-MS1	106	174
B10A017-MSD1	89	17.4
Mean	97.6	6.4

 Table B-4. Matrix Spike Recoveries and Matrix Spike Duplicates.

Table B-5. Laboratory Blanks.

Sample Number	Result (mg/Kg)
B09L135-BLK1	0.0170 U
B09L190-BLK1	0.0170 U
B10A010-BLK1	0.0170 U
B10A017-BLK1	0.0170 U

Sediment QC

Sediment samples were analyzed during July, August, and September, 2009. MEL performed mercury and TOC analyses and Columbia Analytical Services conducted grain size analyses. Both laboratories received samples in good condition and in proper preservation. Analyses were conducted within established holding times. MQOs for sediment analyses are presented in Table B-6.

Parameter	Matrix	Reporting Limit	Check Standard (% recovery)	Duplicate Sample (RPD)	Matrix Spike (% recovery)
Mercury, total		0.005 mg/kg, dry	85-115%	<20%	75-125%
Total Organic Carbon	Sediment	0.10%	80-120%	<20%	75-125%
Grain Size		1%	N/A	<20%	N/A

 Table B-6. Measurement Quality Objectives for Sediment Analysis.

Mercury QC was assessed by examining laboratory control samples, field replicates, matrix spikes, and laboratory blanks. Quality of TOC analyses were assessed through field replicates, laboratory duplicates, standard reference material, and laboratory blanks. Tables B-7 through B-15 present results of the sediment QC tests.

All mercury QC results were within MQOs. Mercury concentrations in sediment samples varied between different locations of lakes, but the RPD between replicates (taken from the same sampling location as the field sample) was low (Table B-8).

Two TOC laboratory duplicates exceeded the MQO RPD, but both were within 20% RPD of the source sample, so no action was taken. All other TOC tests were within MQOs.

Six grain size samples were qualified as estimates ("J") due to high moisture content, resulting in increased chance of error in measurement.

Mercury

Table B-7. Mercury Laboratory Control Samples.

Sample Number	Recovery (%)
B09H024-BS1	99
B09G243-BS1	104
B09I002-BS1	103

Table B-8. Mercury Field Replicates.

Sample Number	Field ID	Result (mg/Kg)	Sample Number	Field ID	Result (mg/Kg)
0907052-11	PIE-SED1	0.091	0907052-12	PIE-SED1R	0.091
0907052-13	PIE-SED2	0.089	0907052-14	PIE-SED2R	0.089
0907052-15	PIE-SED3	0.066	0907052-16	PIE-SED3R	0.066
	Mean	0.082		Mean	0.082
	RPD of results	32.6		RPD of results	31.8
				RPD of Means	0.12

Sample Number	Field ID	Result (mg/Kg)	Sample Number	Field ID	Result (mg/Kg)
0907052-17	WHA-SED1	0.143	0907052-18	WHA-SED1R	0.154
0907052-19	WHA-SED2	0.151	0907052-20	WHA-SED2R	0.143
0907052-21	WHA-SED3	0.189	0907052-22	WHA-SED3R	0.189
	Mean	0.161		Mean	0.162
	RPD of results	27.7		RPD of results	27.7
				RPD of Means	0.62

Table B-9. Mercury Matrix Spikes.

Sample Number	Recovery (%)	RPD (%)
B09G243-MS1	89	0.6
B09G243-MSD1	98	9.0
B09H024-MS1	90	12
B09H024-MSD1	94	4.3

Table B-10.	Mercury Laboratory Blanks	5.
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Sample Number	Result (mg/Kg)
B09G243-BLK1	0.0050 U
B09H024-BLK1	0.0050 U
B09I002-BLK1	0.0050 U

Table B-11. Mercury in Sediments Results.

Banks I	Lake	Failor	Lake	Lake Go	odwin*	Pierre	Lake	Vancouver	Lake	Lake Wha	atcom
Field ID	Result (mg/Kg)	Field ID	Result (mg/Kg)	Field ID	Result (mg/Kg)	Field ID	Result (mg/Kg)	Field ID	Result (mg/Kg)	Field ID	Result (mg/Kg)
BAN-SED1	0.058	FAL-SED1	0.124	GWN-SED1	0.187	PIE-SED1	0.091	VAN-SED1	0.095	WHA-SED1	0.143
BAN-SED2	0.063	FAL-SED2	0.103	GWN-SED2	0.281	PIE-SED2	0.089	VAN-SED2	0.100	WHA-SED2	0.151
BAN-SED3	0.025	FAL-SED3	0.129	GWN-SED3	0.106	PIE-SED3	0.066	VAN-SED3	0.079	WHA-SED3	0.189
						PIE-SED1R	0.091			WHA-SED1R	0.154
						PIE-SED2R	0.089			WHA-SED2R	0.143
						PIE-SED3R	0.066			WHA-SED3R	0.189
Mean	0.049		0.119		0.191		0.082		0.091		0.162
RPD ¹	86.4		22.4		90.4		31.8		23.5		27.7
RSD ²	42.4		11.6		45.8		15.2		12.0		13.5

*Lake Goodwin fish were sampled as part of the 2008 Mercury in Freshwater Fish study. Sediments and water for Lake Goodwin were sampled in 2009.

Total Organic Carbon (TOC)

Sample Number	Field ID	Result (%)	Sample Number	Field ID	Result (%)
0907052-17	WHA-SED1	2.76	0907052-18	WHA-SED1R	2.61
0907052-19	WHA-SED2	4.23	0907052-20	WHA-SED2R	2.61
0907052-21	WHA-SED3	3.71	0907052-22	WHA-SED3R	3.63
	Mean	3.6		Mean	3.0
	RPD of results	42.1		RPD of results	32.7
				RPD of Means	18.9

Table B-12. TOC Field Replicates.

Sample Number	Field ID	Result (%)	Sample Number	Field ID	Result (%)
0907052-11	PIE-SED1	11.7	0907052-12	PIE-SED1R	11.8
0907052-13	PIE-SED2	15.7	0907052-14	PIE-SED2R	16.1
0907052-15	PIE-SED3	13.9	0907052-16	PIE-SED3R	12.8
	Mean	13.8		Mean	13.6
	RPD of results	29.2		RPD of results	30.8
				RPD of Means	1.5

Table B-13. TOC Laboratory Duplicates.

Sample Number	Result (%)	RPD (%)
B09G269-DUP1	0.469	20.7
B09G269-DUP2	0.381	20.7
B09H152-DUP1	0.496	2.2
B09H152-DUP2	0.485	2.2
B09H226-DUP1	0.313	26
B09H226-DUP2	0.302	5.0

Table B-14. TOC Standard Reference Material.

Sample Number	Result (%)
B09G269-SRM1	86
B09H152-SRM1	83
B09H226-SRM1	92

Table B-15. TOC Laboratory Blanks.

Sample Number	Result (%)
B09G269-BLK1	0.1 U
B09H152-BLK1	0.1 U
B09H226-BLK1	0.1 U

Grain Size

Table B-16. Grain Size Triplicate (% Solids).

Sample Number	Result (%)	RPD (%)
	94.3	
0907051-09	95.5	1.5
	94.1	
	100.5	
0907052-04	90.7	10.3
	96.1	

Water QC

MEL analyzed water samples for DOC, alkalinity, and chlorophyll in July and August, 2009. All samples were received by MEL in good condition and were analyzed within established holding times. MQOs for water analyses are presented in Table B-17.

Check Duplicate Reporting Matrix Spike Parameter Matrix Standard Sample Limit (% recovery) (RPD) (% recovery) DOC 80-120% <20% 75-125% 1 mg/LAlkalinity Water 5 mg/L80-120% <10% N/A Chlorophyll $0.05 \,\mu g/L$ 81 - 120 % <10% N/A

Table B-17. Measurement Quality Objectives for Water Analyses.

Quality of water sample data was assessed by examining laboratory control samples, laboratory duplicates, field replicates, and laboratory blanks. All QC tests were within MQOs except for the chlorophyll replicate sample, which resulted in qualification (J = estimated value) of the native sample. Results of water QC tests are displayed in Tables B-18 through B-27.

Dissolved Organic Carbon (DOC)

Sample	Recovery	RPD
Number	(%)	(%)
B09H066-BS1	100	1.0
B09H066-BSD1	99	1.0
B09H228-BS1	100	0.0
B09H228-BSD1	100	0.0
B09H065-BS1	99	1.0
B09H065-BSD1	98	1.0

Table B-18. DOC Laboratory Control Samples and Duplicates.

Table B-19. DOC Field Replicates.

Sample Number	Result (mg/L)	RPD (%)
0907051-11	1.8	0.0
0907051-13	1.8	0.0
0907051-12	1.6	65
0907051-14	1.5	0.3
0907052-06	3.8	27
0907052-08	3.7	2.7
0907052-07	2.9	3.4
0907052-09	3.0	5.4

Table B-20. DOC Laboratory Blanks.

Sample	Result
Number	(mg/L)
B09H065-BLK1	1.0 U
B09H228-BLK1	1.0 U
B09H066-BLK1	1.0 U

Alkalinity

Table B-21.	Alkalinity	Laboratory	Control	Samples.
14010 2 211			00111101	~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~

Sample	Recovery
Number	(%)
B09H130-BS1	98
B09G279-BS1	98
B09G247-BS1	91
B09G248-BS1	96

Table B-22. Alkalinity Field Replicates.

Sample Number	Result (mg/L)	RPD (%)
0907051-12	17.9	1 1
0907051-14	17.7	1.1
0907051-11	21.2	0.0
0907051-13	21.2	0.0
0907052-07	180	0.0
0907052-09	180	0.0
0907052-06	135	15
0907052-08	133	1.3

Table B-23. Alkalinity Laboratory Duplicates.

Sample	Result	RPD
Number	(mg/L)	(%)
0008040 01	34.7	22
0908049-01	33.9	2.5
0007051 01	17.4	1 1
0907031-01	17.6	1.1
0007064 52	148	0.7
0907004-52	149	0.7

Table B-24. Alkalinity Laboratory Blanks.

Sample	Result
Number	(mg/L)
B09H130-BLK1	5 U
B09G248-BLK1	5 U
B09G247-BLK1	5 U
B09G279-BLK1	5 U

Chlorophyll

Sample	Result	RPD
Number	$(\mu g/L)$	(%)
0907051-12	0.6	0.0
0907051-14	0.6	0.0
0907051-11	3.4	2.0
0907051-13	3.5	2.9
0907052-06	2.8	10.2
0907052-09	3.1	10.2
0907052-07	4.8	2.1
0907052-09	4.7	2.1

Table B-25. Chlorophyll Field Replicates.

Table B-26. Chlorophyll Laboratory Duplicates.

Sample	Result	RPD
Number	(µg/L)	(%)
0907052-01	3.3 2	49.1

Table B-27. Chlorophyll Laboratory Blank.

Sample	Result
Number	(ug/L)
B09H088-BLK1	0.1 U

RPD = Relative Percent Difference [(max - min)/(mean)]*100.

SRM = Standard Reference Material.

 $\mathbf{U}=\mathbf{U}\mathbf{n}\mathbf{d}\mathbf{e}\mathbf{t}\mathbf{c}\mathbf{t}\mathbf{d}$ at or above the reported result.

Appendix C. Biological, Sediment, and Water Quality Data

Lake	Species Code	Sample ID	Collection Date	Total Length (mm)	Weight (g)	Age (yrs)	Fulton's Fish Condition Index	Sex	Mercury (ppb)
		1001012-28		340	671	3	1.71	U	77.2
		1001012-29		303	429	5	1.54	F	81.7
		1001012-30		405	1031	7	1.55	М	82.1
		1001012-31		366	707	5	1.44	М	86.9
Banks Lake	CMD	1001012-32	10/12/00	300	390	4	1.44	М	68.2
	SMD	1001012-33	10/15/09	445	1216	11	1.38	F	371
		1001012-34		429	1023	11	1.30	М	268
		1001012-35		286	303	3	1.30	М	86.2
		1001012-36		267	238	3	1.25	М	80.2
		1001012-37		262	243	3	1.35	М	84.9
		1001012-11		430	1455	6	1.83	F	213
		1001012-12		421	1487	6	1.99	F	190
	LMB	1001012-13		400	1205	5	1.88	F	125
		1001012-14		316	568	2	1.80	F	53.8
Failor Lake		1001012-15	9/22/09	265	359	1	1.93	М	52.0
		1001012-16		259	296	1	1.70	F	50.9
		1001012-17		219	177	1	1.69	F	44.8
		1001012-18		224	167	1	1.49	F	40.2
		1001012-19		220	166	1	1.56	F	41.7
		1001012-20		209	141	1	1.54	F	42.8
		1001012-01		442	1411	12	1.63	F	202
		1001012-02		400	1071	10	1.67	М	224
		1001012-03		420	1210	13	1.63	М	210
		1001012-04		361	692	9	1.47	F	176
Diarra Laka	SMD	1001012-05	0/20/00	390	840	9	1.42	М	273
FIEITE Lake	SMB	1001012-06	9/29/09	373	737	8	1.42	F	216
		1001012-07		355	628	6	1.40	F	363
		1001012-08		337	566	5	1.48	F	160
		1001012-09		350	574	12	1.34	М	152
		1001012-10		311	468	9	1.56	М	111
		1001012-69		225	137	2	1.20	F	116
		1001012-70		243	217	2	1.51	F	67.6
		1001012-71		299	452	2	1.69	F	94.8
		1001012-72		320	507	2	1.55	М	105
Snake	I MD	1001012-73	10/20/00	310	484	2	1.62	М	75.0
River	LIVIB	1001012-74	10/20/09	339	639	3	1.64	М	193
		1001012-75		349	656	2	1.54	F	90.2
		1001012-76		359	654	3	1.41	М	185
		1001012-77		403	1175	6	1.80	М	337
		1001012-78		450	1797	6	1.97	F	263

Table C-1. Individual Fish Data by Waterbody.

Lake	Species Code	Sample ID	Collection Date	Total Length (mm)	Weight (g)	Age (yrs)	Fulton's Fish Condition Index	Sex	Mercury (ppb)
		1001012-44		443	1420	7	1.63	F	861
	SMB	1001012-45		372	764	5	1.48	F	210
		1001012-46		375	790	5	1.50	F	329
		1001012-47		340	588	4	1.50	М	224
Whatcom		1001012-48	10/6/09	330	573	4	1.59	М	336
Lake		1001012-49		284	337	3	1.47	М	191
		1001012-50		400	1166	5	1.82	М	907
		1001012-51		340	565	4	1.44	F	241
		1001012-52		306	411	3	1.43	F	219
		1001012-53		285	314	3	1.36	U	167

Lake	Species Code	Sample ID	Collection Date	Average Total Length (mm)	Average Weight (g)	Fulton's Fish Condition Index	Number of Fish in Composite	Mercury (ppb)
	WAL	1001012-41		402	606	0.88	5	45.3
	WAL	1001012-42		461	1077	1.11	4	86
D 1	WAL	1001012-43	10/12/00	342	408	0.92	5	86.7
Banks	LWF	1001012-38	10/13/09	535	1667	1.08	3	48.6
	LWF	1001012-39		498	1280	1.04	4	93.1
	LWF	1001012-40		397	670	0.94	4	104
	CTT	1001012-24		249	138	0.9	3	63.2
E-il-a	CTT	1001012-25	0/22/00	226	103	0.89	4	55.8
Failor	CTT	1001012-26	9/22/09	191	58	0.82	3	41.3
	RBT	1001012-27		372	787	0.96	3	26.5
	SHAD	1001012-63		240	126	0.91	5	45.1
	SHAD	1001012-64		234	116	0.86	5	22.7 U
Vancouver	SHAD	1001012-65	10/13/09	190	66	0.89	5	20.2 U
v ancouver	YP	1001012-66		192	84	1.19	4	73.5
	YP	1001012-67		177	60	1.09	3	88
	YP	1001012-68		162	44	1.03	3	66.9
	CTT	1001012-58		199	68	0.84	4	81.5
	CTT	1001012-59		260	149	0.84	4	94.3
	PEA	1001012-60		225	105	0.92	5	222
	PEA	1001012-61	10/5/09	232	111	0.88	5	320
Whataom	PEA	1001012-62		212	97	0.99	5	243
whatcom	YP	1001012-54		164	48	1.05	5	84.1
	YP	1001012-55		199	91	1.14	5	164
	YP	1001012-56	10/6/09	294	387	1.42	5	180
	YP	1001012-57	10/6/09	223	151	1.17	4	275
	YP	1001012-79	10/5/09	356	702	1.56	1	802

Table C-2. Composite Fish Data by Lake.

U = Not detected at indicated level.

Species codes used in Appendix C

Smallmouth bass
Walleye
Lake whitefish
Cutthroat trout
Rainbow trout
Shad
Yellow perch
Peamouth

			_		Sediment		Water		
Lake	Collection Date	Sample ID	Depth (m)	Mercury (ppb)	TOC (%)	Grain Size (% fines)*	Chl-a (ug/L)	Alkalinity (mg/L)	DOC (mg/L)
		0907052-03	-	57.6	1.00	96.6	-	-	-
		0907052-04	-	63.4	1.01	93.6	-	-	-
Banks	7/28/09	0907052-05	-	25.1	0.91	70.5	-	-	-
		0907052-01	6.0	-	-	-	3.3 J	57.4	1.4
		0907052-02	16.0	-	-	-	2.1	61.1	1.3
Failor		0907051-03	-	124	5.66	79.8	-	-	-
		0907051-04	-	103	6.50	57.3	-	-	-
	7/20/09	0907051-05	-	129	5.84	72.4	-	-	-
		0907051-01	1.5	-	-	-	1.5	17.4	1.6
		0907051-02	5.8	-	-	-	6.7	20.6	1.3
		0907052-11	-	91.0	11.7	53.5J	-	-	-
		0907052-13	-	89.2	15.7	42.49J	-	-	-
		0907052-15	-	65.5	13.9	58.4J	-	-	-
		0907052-12	-	91.0	11.8	-	-	-	-
Diarra	7/27/00	0907052-14	-	89.0	16.1	-	-	-	-
Pierre	//2//09	0907052-16	-	66.0	12.8	-	-	-	-
		0907052-06	4.0	-	-	-	2.8	135	3.8
		0907052-07	16.0	-	-	-	4.8	180	2.9
		0907052-08	4.0	-	-	-	3.1	133	3.7
		0907052-09	16.0	-	-	-	4.7	180	3.0
		0907051-08	-	94.6	1.40	91.5	-	-	-
		0907051-09	-	100	1.72	93.7	-	-	-
Vancouver	7/21/09	0907051-10	-	79.4	1.77	98.8	-	-	-
		0907051-06	0.5	-	-	-	141	65.2	3.0
Vancouver		0907051-07	1.0	-	-	-	73.8	64.9	2.9
		0907052-17	-	143	2.76	83.8	-	-	-
		0907052-19	-	151	4.2	80.3	-	-	-
		0907052-21	-	189	3.71	83.2	-	-	-
		0907052-18	-	154	2.61	-	-	-	-
Whataom	7/20/00	0907052-20	-	143	2.61	-	-	-	-
whatcom	1/29/09	0907052-22	-	189	3.63	-	-	-	-
		0907051-11	6.0	-	-	-	3.4	21.2	1.8
		0907051-12	27.0	-	-	-	0.6	17.9	1.6
		0907051-13	6.0	-	-	-	3.5	21.2	1.8
		0907051-14	27.0	-	-	-	0.6	17.7	1.5
		0908049-03	-	187	18.3	35.88J	-	-	-
		0908049-04	-	281	19.6	28.3J	-	-	-
Goodwin	8/6/09	0908049-05	-	106	20.5	29.82J	-	-	-
		0908049-01	3.0	-	-	-	1.7	34.7	5.1
		0908049-02	10.0	-	-	-	3.1	38.1	3.6

Table C-3. Water and Sediment Results.

*% fines = $< 62 \mu m$.

Note: Goodwin Lake sediments and water were collected in 2009. Fish from Goodwin Lake were collected in 2008 and reported in 2008 results (Furl et al., 2009).



Figure C-1. Temperature Profile for the 2009 Study Lakes.



Figure C-2. Dissolved Oxygen Profile for 2009 Study Lakes.



Figure C-3. Boxplot of Weight for Individual Bass. ($q1 = 25^{th}$ percentile; $q3 = 75^{th}$ percentile.)







Appendix D. Statistical Analyses

Figure D-1. Simple Linear Regression Plots for Mercury and Bass Length.



Figure D-2. Simple Linear Regression Plots for Mercury and Bass Weight.



Figure D-3. Simple Linear Regression Plots for Mercury and Bass Age.

Table D-1. Regression Coefficients, Products, and Standardized (356-mm) Mercury Concentrations for Bass from Mercury Trends Study Years 2005 – 2009.

Waterbody	Species	Mercury Trends Study Year	Constant	B1	B2	356-mm Hg (ppb)	Note	р	R ²
Long Lake	SMB	2005	177.07	-139.10	27.55	31		0.180	0.212
Silver Lake	LMB		127.37	-103.16	21.16	76		0.028	0.539
Potholes Res.	SMB		19.76	-16.15	3.59	82		0.013	0.628
Loon Lake	LMB		-531.81	1.80	-	109	L	0.002	0.702
Liberty Lake	SMB		-41.80	32.24	-5.89	137		0.323	0.069
Yakima R.	SMB		-197.42	154.54	-29.90	180		0.341	0.054
	-	-	-	-					-
Moses Lake	LMB	2006	38.32	-31.34	6.62	29		0.001	0.842
Newman Lake	LMB		-271.29	209.04	-39.92	152		0.009	0.670
Lake Offutt	LMB		-	-	-	188	S	-	-
Lake Sammamish	LMB		-11.74	8.87	-1.32	214		0.003	0.752
Lake Meridian	LMB		17.00	-13.68	3.11	226		0.000	0.898
L. Goose Lake	LMB		-391.47	1.78	-	241	L	0.080	0.252
Deer Lake	LMB	2007	110.55	-86.64	17.34	239		0.000	0.854
Samish Lake	LMB		62.78	-50.35	10.46	261		0.003	0.763
Lake Fazon	LMB		100.96	-76.87	15.01	352		0.075	0.387
Lake St. Clair	LMB		31.49	-24.77	5.27	390		0.002	0.786
Lake Ozette	LMB		82.93	-66.77	13.86	648		0.016	0.604
		•							
McIntosh Lake	LMB	-	50.34	-41.41	8.82	133		0.000	0.914
Horsethief Lake	SMB	2008	55.23	-44.86	9.45	193		0.049	0.458
Loomis Lake	LMB		-100.19	1.03	-	265	L	0.000	0.905
Leland Lake	SMB		14.90	-10.97	2.41	428		0.007	0.694
Lake Nahwatzel	LMB		-822.24	4.68	-	500	*	0.003	0.659
		•							
Banks Lake	SMB		176.44	-139.97	28.05	81		0.002	0.775
Failor Lake	LMB		53.09	-43.53	9.21	89		0.000	0.971
Snake River	LMB	2009	58.24	-46.91	9.77	143		0.022	0.566
Pierre Lake	SMB		-	-	-	190	S	-	-
Lake Whatcom	SMB		106.24	-85.10	17.41	298		0.008	0.681

LMB: Largemouth bass.

SMB: Smallmouth bass.

L = Size range not captured; used simple linear regression without log-transformation.

S = Length did not serve as a good predictor and mercury concentrations were estimated from fish near the same size.

* = Size range not captured and extrapolation using simple linear regression overestimated concentration; mercury concentration assigned by author.

 $Regression \ Equation: \ Log10 \ (Mercury) = Constant + \{B1 * Log10 \ (Length)\} + \{B2 * (Log10 \ (Length))^2\}.$

Appendix E. Fish Tissue Data Evaluation by Ecology and DOH

Several state and federal agencies collect and evaluate fish tissue data in Washington State: Ecology, DOH, WDFW, EPA, and USGS. Tissue data are evaluated differently by these agencies because their mandates and roles are varied. These multiple evaluations often lead to confusion and misunderstanding among agencies and the public on how fish tissue data are used and interpreted. Most fish tissue contaminant data from Washington fish, regardless of who conducted the study, make their way to DOH for evaluation regarding the safety of consuming contaminated fish.

The following is an overview of how Ecology and DOH evaluate fish tissue data to meet different needs.

Ecology

For many Ecology studies, fish tissue data are evaluated primarily to determine if (1) water quality standards are being met, and (2) potential risks to human health from consuming contaminated fish warrant further study and/or development of a fish consumption advisory. Ecology's role is to determine whether water quality standards are met and to begin the process to correct problems where standards are not met. DOH and local health departments are responsible for developing fish consumption advisories in Washington. There is some overlap in these evaluations because the water quality standards that fish tissue data are compared to were developed for the protection of human health.

Washington's water quality standards criteria for toxic contaminants were issued to the state in EPA's 1992 National Toxics Rule (NTR) (40CFR131.36). The human-health-based NTR criteria are designed to minimize the risk of effects occurring to humans from chronic (lifetime) exposure to substances through the ingestion of drinking water and consumption of fish obtained from surface waters. The NTR criteria, if met, will generally ensure that public health concerns do not arise and that fish advisories are not needed.

The NTR criteria are thresholds that, when exceeded, may lead to regulatory action. When water quality criteria are exceeded, the federal Clean Water Act requires that the waterbody be put on a list and a water cleanup plan be developed for the pollutant causing the problem. This list is known as the "303(d) list," and the water cleanup plan results from a Total Maximum Daily Load (TMDL) study and public involvement process. Ecology uses the TMDL program to control sources of the particular pollutant in order to bring the waterbody back into compliance with the water quality standards.

DOH

While DOH supports Ecology's use of the NTR criteria for identifying problems and controlling pollutant sources so that water quality will meet standards, DOH does not use the NTR criteria to establish fish consumption advisories (Furl et. al., 2007). DOH uses an approach similar to that

in EPA's *Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories Vol. 1-4* for assessing mercury, PCBs, and other contaminants (EPA, 2000). These guidance documents provide a framework from which states can evaluate fish tissue data to develop fish consumption advisories based on (1) sound science and (2) established procedures in risk assessment, risk management, and risk communication. Neither the NTR criteria, nor the screening values found in the EPA guidance documents above, incorporate the varied risk management decisions essential to developing fish consumption advisories.

- *Risk Assessment* involves calculating allowable meal limits based on known fish contaminant concentrations. These calculations are conducted for both non-cancer and cancer criteria using the appropriate Reference Dose (RfD) or Cancer Slope Factor (CSF), if available. These initial calculations are the starting point for evaluating contaminant data to determine whether a fish advisory is warranted. Additionally, known or estimated consumption rates help determine the potential magnitude of exposure and highlight the sensitive groups or populations that may exist due to elevated consumption rates.
- *Risk Management* includes (but is not limited to) consideration of contaminant background concentrations, reduction in contaminant concentrations through preparation and cooking techniques, known health benefits from fish consumption, contaminant concentrations or health risks associated with replacement foods, and cultural importance of fish. Other considerations are the possible health criteria associated with a contaminant, the strength or weaknesses of the supporting toxicological or sampling data, and whether effects are transient or irreversible.
- *Risk Communication* is the outreach component of the fish advisory. The interpretation of the data from the risk assessment and risk management components drives how and when the fish advisory recommendations are issued to the public dependent on whether the message is targeted toward a sensitive group or a population or the general public. DOH's dual objective is how best to provide guidance to the public to increase consumption of fish low in contaminants to gain the benefits of eating fish, while steering the public away from fish that have high levels of health-damaging contaminants.

Appendix F. Glossary, Acronyms, and Abbreviations

Glossary

Analyte: Water quality constituent being measured (parameter).

Anthropogenic: Human-caused.

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.

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.

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

Epilimnion: The uppermost layer of water in a lake where water temperature changes less than 1° C per one meter of depth.

Exceeded criterion: Did not meet or violated the criterion.

Grab sample: A discrete sample from a single point in the water column or sediment surface.

Hypolimnion: The deepest layer of water in a lake where water temperature changes less than 1° C per one meter of depth.

Morphology: Shape (e.g., channel morphology).

Otolith: Part of the inner ear of a fish. This structure is used to determine the age of a fish.

Parameter: Water quality constituent being measured (analyte).

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

Temporal trend: Trend or pattern over time.

Thermocline: A temperature gradient in a thermally stratified, or temperature divided, body of water. Commonly associated with solar heating of the upper layers of a waterbody while the cooler layers remain on the bottom.

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

DOC	Dissolved organic carbon
DOH	Washington State Department of Health
EAP	Environmental Assessment Program
Ecology	Washington State Department of Ecology
EIM	Environmental Information Management database
EPA	U.S. Environmental Protection Agency
GIS	Geographic Information System software
Hg	Mercury
MEL	Manchester Environmental Laboratory
MQO	Measurement quality objectives
NTR	National Toxics Rule
PBT	Persistent, bioaccumulative, and toxic substance
RM	River mile
QC	Quality control
RPD	Relative percent difference
RSD	Relative standard deviation
SOP	Standard operating procedures
SRM	Standard reference materials
TOC	Total organic carbon
USGS	U.S. Geological Survey
WAC	Washington Administrative Code
WDFW	Washington Department of Fish and Wildlife
WRIA	Water Resources Inventory Area
WWTP	Wastewater treatment plant

Units of Measurement

°C	degrees centigrade
dw	dry weight
ft	feet
g	gram, a unit of mass
g/d	grams per day
kcfs	1000 cubic feet per second
m	meter
mg/Kg	milligrams per kilogram (parts per million)
mg/L	milligrams per liter (parts per million)
mL	milliliters
mm	millimeters
ng/g ppb	nanograms per gram (parts per billion) parts per billion

µg/g	micrograms per gram (parts per million)
ug/Kg	micrograms per kilogram (parts per billion)
μg/L	micrograms per liter (parts per billion)
µS/cm	microsiemens per centimeter, a unit of conductivity
WW	wet weight