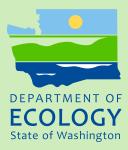


Measuring Mercury Trends in Freshwater Fish in Washington State

2014 Sampling Results



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Measuring Mercury Trends in Freshwater Fish in Washington State

2014 Sampling Results

by

Callie Mathieu and Melissa McCall

Toxics Studies Unit Environmental Assessment Program Washington State Department of Ecology Olympia, Washington 98504-7710

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

WRIAs

- 42 Grand Coulee
- 22 Lower Chehalis
- 15 Kitsap
- 60 Kettle
- 1 Nooksack
- 35 Middle Snake

HUC numbers

- 17020014 Banks Lake
- 17100105 Grays Harbor
- 17110019 Puget Sound
- 17020003 Colville
- 17110004 Nooksack
- 17060103 Lower Snake-Asotin

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Abstract

In 2005, the Washington State Department of Ecology (Ecology) began a long-term monitoring program to assess temporal trends in mercury levels of freshwater fish throughout the state. Each year Ecology collects ten individual largemouth or smallmouth bass from six waterbodies for analysis of total mercury. Ecology returns to each set of waterbodies every five years to assess trends.

In 2014, Ecology collected a total of 60 bass from Banks Lake, Failor Lake, Kitsap Lake, Pierre Lake, the Snake River, and Lake Whatcom. Mercury concentrations ranged from 42 - 1,240 ppb across the waterbodies. The maximum concentration came from an 8-year-old smallmouth bass from Lake Whatcom and was the only sample to exceed (not meet) the Fish Tissue Equivalent Concentration used in the state's Water Quality Assessment (770 ppb). Sixteen bass samples exceeded the EPA Recommended Criterion (300 ppb).

Statistical tests showed a significant difference in mercury levels in bass between 2009 and 2014 for one out of five waterbodies. Statistical tests were not possible for the Kitsap Lake dataset. Bass from Failor Lake increased 44% in estimated mercury levels between the 2009 and 2014 collection years. No change over the five-year period was found in bass from Banks Lake, Pierre Lake, the Snake River, or Lake Whatcom. However, mercury levels in Lake Whatcom bass collected in 2014 were significantly lower than in bass collected in 2000. Mercury levels in the 2014 bass were 60% lower than the historical data from 2000.

This report also summarizes temporal trends from all waterbodies re-sampled after a five-year period for this program. Statistical analysis was possible for 26 of the 30 waterbodies. Tests showed no difference in bass mercury concentrations between the first and second sampling visits for over half of the waterbodies (54%). Mercury levels in bass increased in 35% of waterbodies (9 out of 26). Three lakes (12%) showed decreases in mercury concentrations.

Acknowledgements

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 - Jean Maust, Joan LeTourneau, and Cindy Cook for formatting and editing the final report.

Introduction

Background

In 2000 Washington State began an effort to reduce human sources of several toxic chemicals called persistent, bioaccumulative toxics - or PBTs. PBTs are not easily broken down. They build up through the food chain and cause adverse biological effects.

Mercury is a PBT. Though it occurs naturally in certain hard-rock and metallic ores, it can enter the environment from both natural emissions and human activities. 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). Natural sources of mercury 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, including mining.

Mercury discharged to land, air, or water can eventually find its way to lakes, rivers, and the ocean, where it settles into sediments. All forms of mercury can be harmful to humans and other animals, depending on the route and amount of exposure. For most people, fish consumption is the primary source of exposure. Mercury has highly toxic effects in humans, with developing fetuses and young children most at risk (Peele, 2003). It predominantly affects the central nervous system and can lead to problems within cardiovascular systems.

The Washington State Departments of Ecology (Ecology) and Health (DOH) developed a Chemical Action Plan (CAP) for mercury in 2003 (Peele, 2003) detailing natural and anthropogenic sources, occurrence and biogeochemical cycling in the environment, mercury use and emissions in Washington, a summary of health effects and concerns, and fish consumption advisories in Washington due to mercury-contaminated fish. Several early studies (Serdar et al., 2001; Norton, 2004) helped to characterize the extent and severity of mercury contamination in fish throughout Washington State. Fischnaller et al. (2003) found elevated levels of mercury in bass muscle tissue from 20 sites across the state and recommended a statewide monitoring program to address this issue.

In 2005 Ecology began long-term monitoring of mercury in fish. The main goal of this effort is to monitor mercury levels in edible (fillet) tissue from freshwater fish over time to understand the scope of fish tissue contamination levels in Washington. Information gained from this long-term monitoring effort is useful in understanding the fate of mercury in our environment and will be useful in future efforts that may be developed for determining the effectiveness of Washington's Mercury CAP. This report summarizes results from the tenth year of sampling.

Assessment of Mercury in Fish Tissue

Numerical thresholds for mercury are based on the toxicological effects of methylmercury. Methylmercury is the bioaccumulative and toxic form of mercury in fish tissue, which accounts for more than 95% of the total mercury in fish tissue (Bloom, 1995; Driscoll et al., 1994; Grieb et al., 1990). Total mercury is the target analyte used in this study as a surrogate for methylmercury due to the comparative simplicity and lower cost of analysis for total mercury.

Ecology assesses fish tissue data to help determine whether designated uses of fish/shellfish and drinking surface waters are being met in ambient water of Washington State. For mercury, Ecology uses a fish tissue equivalent concentration (FTEC) of 770 ppb for its Water Quality Assessment. This assessment level for the protection of human health is based on the National Toxics Rule (NTR) (40CFR131.36) criterion for mercury in water. The FTEC is calculated by multiplying the mercury-specific bioconcentration factor by the mercury-specific Water Quality Criterion found in the NTR.

Another helpful guideline for putting fish tissue mercury concentrations into context is the U.S. Environmental Protection Agency (EPA) Recommended Criterion. The EPA issued a Recommended Criterion for methylmercury (300 ppb) as guidance to states and tribes for the protection of human health (EPA, 2001). This guidance describes the level of methylmercury in freshwater fish that should not be exceeded in order to protect the general fish-consuming population. This is not a regulatory criterion in Washington State.

Washington's DOH assesses mercury levels in fish tissue to evaluate the risks of human consumption of contaminated fish. DOH uses an approach similar to that in EPA's *Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories Vol. 2* (EPA, 2000a). The EPA suggests using these documents as a framework from which states can develop fish consumption advisories. Washington State currently has a statewide consumption advisory based on mercury levels and recommends guidelines for consumption of largemouth and smallmouth bass (McBride, 2003).

Study Design

The primary goal of this project is to characterize temporal trends in mercury levels of uppertrophic level freshwater fish in Washington State. To meet this goal, Ecology analyzes fish and water samples collected from six waterbodies each year and then re-samples those waterbodies every five years.

Specific objectives of the study are to:

- Measure mercury levels in fillets of 10 individual bass from six waterbodies per year for five years, for a total of 30 sites. Sampling is 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 include:
 - Fish length, weight, sex, and age.
 - Alkalinity, bromide, chloride, dissolved organic carbon (DOC), fluoride, nitrate/nitrite and sulfate concentrations from the top and bottom of the water column.
 - Vertical water column profiles of temperature, dissolved oxygen, conductivity, and pH.
- Determine mercury concentrations in composite fillet samples of three to five individual fish from two other fish species present at sampling sites where bass are collected.

Largemouth and smallmouth bass are the target species for this project because of their known propensity to accumulate mercury, their widespread occurrence in Washington waterbodies, and because they are frequently targeted by recreational anglers. Detailed study design information can be found in the Quality Assurance (QA) Project Plan (Seiders, 2006) and addendum (Meredith and Furl, 2010).

Study Locations

Five lakes and one river site were sampled in 2014: Banks, Kitsap, Failor, Pierre, and Whatcom Lakes, and the Snake River near Clarkston (Figure 1). All sites were first visited in 2009 (Meredith et al., 2010), with the exception of Kitsap Lake. Site selection in 2009 was 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). Kitsap Lake was added to the program in 2014 to replace Vancouver Lake, where bass collection efforts were unsuccessful in 2009. Kitsap Lake was selected as a back-up waterbody because mercury data were available from the 2001/2002 survey of mercury in bass tissue. The QA Project Plan contains detailed information on site selection considerations (Seiders, 2006). Physical information for each waterbody sampled in 2014 is presented in Table 1.

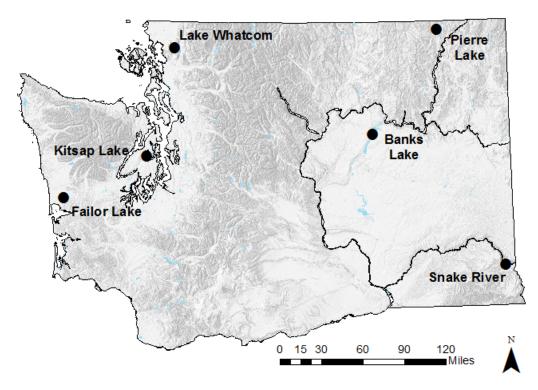


Figure 1. 2014 Sample Locations for Mercury Trends in Fish.

Table 1	Physical Characteristic	s of Study Sites Sampled in 2014.
	Thysical Characteristic	s of Study Sites Sampled in 2014.

	Banks	Failor	Kitsap	Pierre	Snake*	Whatcom
County	Grant	Grays Harbor	Kitsap	Stevens	Asotin	Whatcom
Drainage Basin (ac)		3,130	1,750	17,152		35,776
Elevation (ft)	1,570	117	127	2,005	700	315
Surface Area (ac)	27,000	65	248	110		5,000
Lake Volume (ac-ft)	1,300,000	500	4,480	3,000		770,000
Maximum Depth (ft)	85	22	29	75	50	330
Mean Depth (ft)	47	8	18	28	12	150
Dominant Land Type	barren/ agricultural	forested	residential	forested	barren/ agricultural	forested/ residential

*Snake River near Clarkston, WA.

Collection Goals

Table 2 displays a summary of the collection goals met in 2014. Ecology and the Washington Department of Fish and Wildlife (WDFW) collected fish from the six sampling sites in September and October of 2014. A total of 60 individual largemouth or smallmouth bass and 72 fish of additional species were collected, encompassing eight species. Ten bass were collected at all six sites. Additional species of fish were collected from all sites with the exception of Failor and Pierre Lakes. No other species were encountered at these waterbodies. Water samples were collected from each of the locations during July of 2014 in an attempt to capture lake-stratified conditions. No water profile was taken at Banks Lake due to weather constraints.

Collection Goal	Waterbody									
	Banks Lake	Failor Lake	Kitsap Lake	Pierre Lake	Snake River	Lake Whatcom				
10 individual bass	+	+	+	+	+	+				
Species collected for composites	BBH, CCP, LWF	NA	BBH	NA	NPM, LSS	ВВН, КОК				
2 water samples	+	+	+	+	+	+				
Hydrolab profile	NA	+	+	+	+	+				

Table 2. Summary of Collection goals for 2014.

NA: Collection goal not attained

+ Collection goal met

BBH: Brown Bullhead, CCP: Common Carp, KOK: Kokanee,

LSS: Largescale Sucker, LWF: Lake Whitefish, NPM: Northern Pikeminnow

Methods

Field Procedures

Fish

Methods for the collection and handling of fish tissue samples are described in EPA (2000b) and Sandvik (2014a). Fish were collected using gill nets and Ecology's electrofishing boat. Field crews inspected fish 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.

Fish samples were processed at Ecology headquarters following standard operating procedures (Sandvik, 2014b). 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 pre-cleaned 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. For aging, otoliths were extracted from bass >300 mm in length and scale samples were removed from fish <300 mm in length. These age structures were 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 hot 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.

Water

Water column conductivity, pH, dissolved oxygen, and temperature were measured using a Hydrolab multi-parameter probe following standard operating procedures (Swanson, 2010). All instruments were calibrated prior to field use, and Winkler titrations performed as a measure of quality control for dissolved oxygen readings. Water transparency was measured using a Secchi disc following field protocols described in EPA (2007).

Two water samples were obtained from different depths at the deepest part of each waterbody 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 temperature 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, bromide, chloride, dissolved organic carbon (DOC), fluoride, nitrite/nitrate, and sulfate. DOC was filtered in the field prior to preservation with hydrochloric acid. Nitrite/nitrate was also preserved with hydrochloric acid in the field. Samples were stored on ice in the field until shipment to the laboratory.

Laboratory Procedures

Ecology's Manchester Environmental Laboratory (MEL) performed all laboratory analysis. Table 3 provides a list of matrices, analytes, and analytical methods.

Matrix	Analyte	Method		
Fish Tissue	Mercury	CVAA, EPA 245.6		
	Alkalinity	SM2320B		
	DOC	SM5310B		
	Bromide	EPA300.0		
Water	Chloride	EPA300.0		
	Fluoride	EPA300.0		
	Sulfate	EPA300.0		
	Nitrate-Nitrite	SM4500NO3I		

 Table 3. Analytes and Analytical Methods

DOC: dissolved organic carbon CVAA: Cold Vapor Atomic Absorption

Data Quality

MEL received all samples in good condition and within the proper temperature range. All laboratory analyses were conducted within their method holding times. Instrument calibration checks were within acceptance limits, and no analytically significant levels of analytes were detected in the method blanks. MEL provided case narratives describing instrument calibrations, method blank analyses, and QA tests. These narratives are available from the project manager upon request.

MEL met all measurement quality objectives (MQOs) for mercury analysis of fish tissue, with the following exception. One standard reference material (SRM) recovery was outside of MQOs outlined in the project plan (117%). However, because MEL does not have enough data points on record for SRM recoveries, MEL does not have QC limits for this type of SRM and therefore the data were not qualified. Average recoveries for laboratory control samples (LCS), SRM, and matrix spikes were 105%, 105%, and 88%, respectively.

Laboratory duplicates were performed on matrix spike samples, and the average relative percent difference (RPD) was 6.3%. Laboratory blanks were not detected above 17 ug/Kg (ppb) wet weight.

Quality Control for water chemistry analyses included LCS, matrix spikes, laboratory duplicates, and laboratory blanks. All were within MQOs. Quality Control data for water analyses are available upon request.

Results

Fish

Ecology analyzed a total of 60 individual bass and 17 composites of additional species for mercury in 2014. The following sections summarize individual bass and composite fish tissue results. All results are available from Ecology's Environmental Information Management (EIM) database at http://www.ecy.wa.gov/eim under the Study ID: HgFish14.

Individual Bass

Largemouth and smallmouth bass were collected from Banks, Failor, Kitsap, Pierre and Whatcom Lakes, as well as the Snake River. Table 4 displays statistical summaries of ancillary data and mercury concentrations measured in the individual bass collected in 2014.

Table 4. Statistical Summary of 2014 Individual Bass Lengths, Weights, Ages, and Mercury Concentrations.

Waterbody	Species	Length (TL mm)		Weigł (g)	Age (yr)		Hg (ppb ww)		
Waterbody	Opecies	Range	Med.	Range	Med.	Range	Med.	Range	Med.
Banks	SMB	288 - 430	401	289 - 1171	803	2 - 10	6	66.5 - 303	187
Failor	LMB	119 - 480	216	110 -2034	151	1 - 12	1	41.3 - 398	60
Kitsap	LMB	251 - 497	307	254 - 2420	474	1 - 10	2	121 - 480	179
Pierre	SMB	294 - 381	328	328 - 813	472	5 - 8	8	136 - 223	176
Snake	LMB	270 - 462	354	320 - 1679	700	1 - 7	3	55.5 - 372	213
Whatcom	SMB	290 - 440	410	369 - 1544	1098	2 - 9	5	162 - 1240	377
All Sites		119 - 497	351	110 - 2420	625	1-12	5	41 - 1240	191

Summaries are based on ten samples per site.

TL: total length; ww: wet weight; Med.: median SMB: smallmouth bass; LMB: largemouth bass

Bass Size

Figure 2 displays the length-to-weight relationship for individual bass collected from the six waterbodies. Eighty-two percent (49 out of 60) of individual bass lengths met the target size range of 250 - 460 mm.

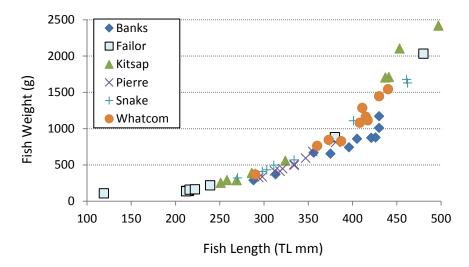


Figure 2. Relationship of Individual Bass Length and Weight Collected at Each of the Waterbodies.

Bass Mercury Results

All individual bass samples contained mercury levels above the reporting limit of 17 ug/Kg ww (referred to throughout this report as ppb). Concentrations ranged from 41 - 1,240 ppb across waterbodies. Lake Whatcom contained the highest levels of mercury in bass, with an average concentration of 507 ppb and a median of 377 ppb. These fish were also the largest (median weight = 1,098 grams, median length = 410 mm), but not the oldest, of the six waterbodies. The lowest mercury levels were found in Failor Lake (mean = 93.9 ppb, median = 60 ppb), where all but two of the bass collected were only 1 year old, making them younger than the other fish collected. Figure 3 displays mercury concentrations measured in individual bass.

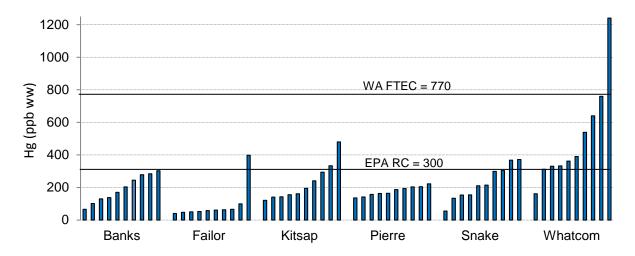
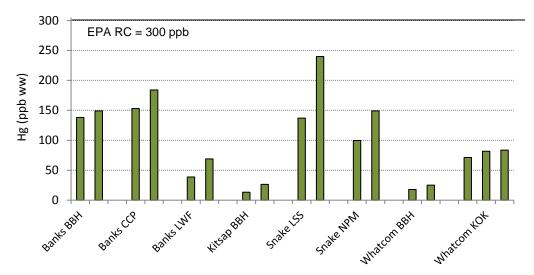


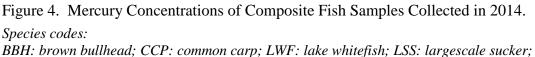
Figure 3. Mercury Concentrations of Individual Bass Collected in 2014. WA FTEC: Washington Fish Tissue Equivalent Concentration EPA RC: Environmental Protection Agency Recommended Criterion

One individual bass sample from Lake Whatcom – an eight-year-old male – exceeded the FTEC for mercury. Twenty-seven percent of individual bass samples (16 out of 60) had mercury concentrations above the EPA Recommended Criterion for methylmercury of 300 ppb. More than half of these bass came from Lake Whatcom. All lakes except for Pierre had at least one sample above the EPA Recommended Criterion.

Fish Composites

Field crews collected additional fish species from Banks, Kitsap, and Whatcom Lakes and the Snake River. Seventeen composites, including six species, were analyzed for mercury. Figure 4 displays mercury concentrations measured in the composite samples.





BBH: brown bullhead; CCP: common carp; LWF: lake whitefish; LSS: largescale suck NPM: northern pikeminnow; KOK: kokanee

Mercury concentrations in ancillary species ranged from 13.3 – 240 ppb with a mean of 98.6 ppb. Snake River largescale suckers had the highest concentrations (mean 189 ppb) and Kitsap Lake brown bullhead contained the lowest levels of mercury (mean 20 ppb). No composite samples exceeded the state FTEC assessment level of 770 ppb or the EPA Recommended Criterion of 300 ppb.

Water

Water samples were collected from the epilimnion and hypolimnion of all six waterbodies. Analytes included alkalinity, sulfate, dissolved organic carbon (DOC), bromide, chloride, fluoride, and nitrite/nitrate. Field crews also measured vertical profiles of dissolved oxygen, temperature, pH, and conductivity at each waterbody, except for Banks Lake. The following figure (5) displays the alkalinity, sulfate, and DOC results.

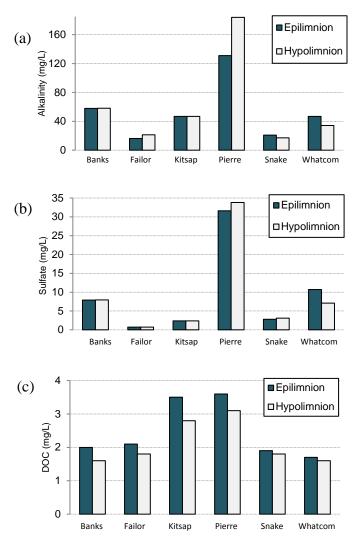


Figure 5. Concentrations of (a) Alkalinity, (b) Sulfate, and (c) Dissolved Organic Carbon Measured from the Epilimnion and Hypolimnion of the 2014 Waterbodies.

Across the six waterbodies alkalinity values ranged from 16 - 184 mg/L. The highest and lowest concentrations were found at Pierre Lake and Failor Lake respectively. Concentrations of Sulfate ranged from 0.7 - 33.8 mg/L. Similar to alkalinity, Pierre Lake contained the greatest sulfate concentrations and Failor Lake the lowest. Dissolved organic carbon concentrations were less variable, all concentrations were below 4 mg/L, with a range of 1.6 - 3.6 mg/L.

Chloride was detected in all samples at low concentrations, while fluoride and nitrite/nitrate were detected in less than 50% of samples, also at low concentrations. Bromide was not detected above 0.2 mg/L in any of the samples.

Vertical temperature profiles indicated Failor, Kitsap, and Pierre Lakes were stratified at the time of water sample collections. Lake Whatcom and Snake River did not show stratification and field crews were unable to obtain a hydrolab profile of Banks Lake due to weather. Dissolved oxygen levels dropped below 1 mg/L in the bottom waters of Failor, Kitsap, and Pierre Lakes.

Fish Tissue - Mercury Relationships

Simple linear regressions were used to evaluate mercury concentration against fish length, weight, and age. Table 5 displays coefficients of determination (r^2). All data were log₁₀ transformed prior to regression analysis to improve normality which was assessed through Shapiro-Wilks normality test.

Pierre Lake mercury concentrations in bass had no statistically significant relationship with length, weight, or age. Mercury concentrations in bass significantly increased with fish length, weight, and age for all other waterbodies, with the exception of age in the Snake River dataset. For Banks, Failor, Kitsap, and Whatcom Lakes, age had the strongest relationship with mercury concentration.

XX7 / 1 1	r ²					
Waterbody	Length	Weight	Age			
Banks	0.88	0.84	0.93			
Failor	0.69	0.87	0.94			
Kitsap	0.84	0.84	0.91			
Pierre	0.02	0.02	0.00			
Snake	0.71	0.72	0.60			
Whatcom	0.60	0.55	0.80			

Table 5. Linear Regression Coefficients of Determination for Mercury Relationships.

Bolded values indicate statistical significance at p < 0.05.

2009-2014 Temporal Trends

This study collected bass from Banks, Failor, Pierre and Whatcom Lakes, as well as from the Snake River, in 2009. Largemouth bass was collected from Kitsap Lake in 2002 (Fischnaller, 2003). Results from prior years were compared to the 2014 sample results to examine statistically significant differences in mercury concentrations using analysis of covariance (ANCOVA).

All variables were log₁₀ transformed prior to analysis to achieve normality and homogeneity of variance. Fish length was used as the covariate in all lakes except Pierre, to control for the effect of fish size on mercury accumulation in bass. Mercury concentrations in bass from Pierre Lake were not significantly correlated with fish length, weight, or age. As a result, an analysis of variance (ANOVA) was used for Pierre Lake using only samples within the same size range. Data from Kitsap Lake did not meet requirements for ANCOVA and there were too few samples of similar sized fish from the two collection periods to do an ANOVA. Therefore, no statistical test was run for the Kitsap Lake dataset.

Analysis of Covariance

ANCOVA results showed a significant difference in bass mercury levels between 2009 and 2014 for one out of the five waterbodies. Bass from Failor Lake had a significant increase in estimated mercury levels (least squares means). Statistical tests for all other waterbodies revealed no significant change in estimated bass mercury levels between 2009 and 2014. Additional smallmouth bass data from a 2000 survey were available for Lake Whatcom (Serdar et al., 2001). Lake Whatcom 2014 mercury levels in bass were significantly lower than levels in the samples collected in 2000, but not significantly different from 2009 concentrations. The 2009 mercury values were also lower than the 2000 survey. Results of statistical tests for the five waterbodies are presented in Table 6. Figure 6 displays mercury and length data of individual bass included in the ANCOVA.

		ANCOVA							Post-Hoc		
Waterbody	Species	Co- variate	Sum of Squares	df	Mean Squares	F- Ratio	p-Value	2000 Hg _{bass}	2009 Hg _{bass}	2014 Hg _{bass}	
Banks	SMB	length	0.021	1	0.021	1.004	0.330		131	154	
Failor	LMB	length	0.112	1	0.112	5.500	0.031		61 ^a	88 ^b	
Pierre	SMB	none	0.010	1	0.010	0.606	0.450		201	179	
Snake	LMB	length	0.047	1	0.047	1.802	0.198		151	193	
Whatcom	SMB	length	1.320	1	1.320	51.38	<0.001	755 ^a	303 ^b	299 ^b	

Bolded values indicate statistical significance at p <0.05.

df: degrees of freedom

Hg_{bass}: value in ppb, back-transformed least squares means from Bonferroni post hoc tests, with Duan's Smearing estimator applied to correct for back-transformation bias (Helsel and Hirsch, 2002; Duan, 1983).

A difference in letters (a,b) indicates statistically significant difference in estimated least squares means for those collection dates.

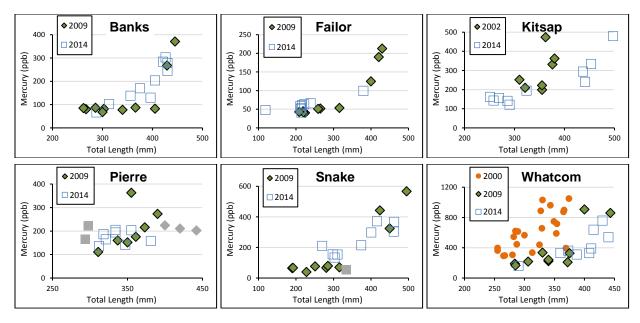


Figure 6. Mercury to Fish Length Relationship of Individual Bass Collected in 2009* and 2014. *Kitsap Lake bass were collected in 2002; Lake Whatcom includes bass from 2000. Grey diamond: 2009 datapoint excluded for analysis; grey square: 2014 datapoint excluded for analysis.

Discussion

Failor is the only lake to have a significant change in bass mercury concentrations between 2009 and 2014, with back-transformed estimated mercury levels increasing 44% from 2009 to 2014. Though mercury levels increased in Failor Lake bass, it should be noted that concentrations were still quite low compared to those recorded for the other waterbodies. The back-transformed estimated mercury levels from 2009 and 2014 were 61 ppb and 88 ppb, respectively, at a length of 259 mm. Both values are well below the EPA Recommended Criterion of 300 ppb and the state Water Quality assessment level of 770 ppb.

To put the ANCOVA results into context, the authors qualitatively examined factors that may influence changing mercury levels in bass tissue: differences in water chemistry between the two collection years (alkalinity, DOC, pH, DO, and conductivity), average precipitation for the five-year period preceding fish collections, landscape changes identified by aerial photo-imagery, and fish size to fish age relationships (data in Appendix A). The pH levels and annual precipitation were the only identified variables that may have had an influence on the increase in mercury concentrations in Failor Lake bass.

Compared to 2009 sampling, Failor Lake pH values in 2014 were notably more acidic. Epilimnion pH decreased from 8.7 in 2009 to 7.2 in 2014, and hypolimnion pH dropped from 8.3 to 6.5. Lower pH has been correlated with higher mercury concentrations in bass in Eastern Washington lakes (Mathieu et al., 2013b). Researchers have proposed that mercury methylation by microbial processes is increased in acidic waters (Xun et al., 1987; Kelly et al., 2003). Annual watershed precipitation has also been positively correlated to mercury concentrations in bass in Washington State lakes (Mathieu et al., 2013b). The five-year precipitation average at Failor Lake increased five inches from 2009 to 2014 averages (103" to 108") (PRISM Climate Group, accessed on 3/4/2016).

Back-transformed estimated mercury levels of the 2014 Lake Whatcom bass samples were 60% lower than mercury levels of bass collected in 2000. However, bass from 2000 were collected in the spring, while 2009 sampling occurred in the fall. Therefore, the difference in mercury concentrations could reflect seasonal variation. A recent sediment core collected in Basin 1 of Lake Whatcom shows mercury concentrations peaking in the 1970s, declining through the early 2000s, and remaining consistent around 0.14 mg/Kg between the early 2000s and 2013 (Mathieu and McCall, in prep).

Statewide Temporal Trends

Sampling in 2014 completed a cycle of re-visiting all waterbodies for the Mercury Trends in Fish Tissue project. Ten bass were analyzed for mercury from six waterbodies annually, from 2005 to 2009, and re-sampled five years later, between 2010 and 2014. Annual reports for this project have included temporal trends assessed using ANCOVA to identify statistically significant differences in mercury concentrations between collection years (Meredith and Friese, 2011; Mathieu and Friese, 2012; Mathieu et al., 2013b; Mathieu and McCall, 2015a).

Results for all trend tests are displayed in Table 7. Percent change in bass mercury concentrations was calculated as the percent change between back-transformed least squares means, corrected for transformation bias with Duan's Smearing estimator (Duan, 1983).

Table 7. Analysis of Covariance (ANCOVA) Results of All Mercury Trends Calculated between 2010 and 2014.

Date of First Sampling Visit	Date of Second Sampling Visit	Waterbody	Species	Trend in Hg _{bass}	Percent Change in Hg _{bass}	Co- variate	First Visit Hg _{bass}	Second Visit Hg _{bass}	Mean Fish Length (mm)
2005	2010	Liberty Lake	SMB	\uparrow	34%	L	182	244	394
2005	2010	Loon Lake	LMB	=		L	249	260	430
2005	2010	Potholes Res.	SMB	=		L	107	134	381
2005	2010	Silver Lake	LMB	=		L	72	87	337
2005	2010	Yakima River	SMB	=		L	136	161	319
2006	2011	Meridian Lake	LMB	=		L	211	195	333
2006	2011	Moses Lake	SMB	=		L	28	29	337
2006	2011	Newman Lake	LMB	=		L	199	241	391
2006	2011	Offutt Lake	LMB	=		none	210	179	294
2006	2011	Lake Sammamish	LMB	\uparrow	34%	L	247	330	380
2007	2012	Deer Lake	LMB	\uparrow	22%	L	318	390	382
2007	2012	Lake Fazon	LMB	\uparrow	25%	none	384	479	403
2007	2012	Lower Goose Lake	LMB	\checkmark	-30%	L	322	225	402
2007	2012	Lake Ozette	LMB	=		L	526	470	317
2007	2012	Lake Samish	LMB	\uparrow	46%	L	235	343	305
2007	2012	Lake St. Clair	LMB	\uparrow	25%	А	422	526	362
2008	2013	Lake Goodwin	SMB	\uparrow	49%	none	117	174	247
2008	2013	Leland Lake	LMB	\checkmark	-34%	L	506	335	358
2008	2013	Loomis Lake	LMB	\uparrow	55%	L	119	185	216
2008	2013	McIntosh Lake	LMB	=		А	129	101	301
2008	2013	Lake Nahwatzel	LMB	\checkmark	-44%	L	353	197	255
2009	2014	Banks Lake	SMB	=		L	131	154	357
2009	2014	Failor Lake	LMB	\uparrow	44%	L	61	88	259
2009	2014	Pierre Lake	SMB	=		none	201	179	345
2009	2014	Snake River	LMB	=		L	151	193	324
2009	2014	Lake Whatcom	SMB	=		L	403	370	367

L: length; A: age

Hgbass: back-transformed least squares means from Bonferroni post-hoc tests, with Duan's Smearing estimator applied to correct for back-transformation bias (Helsel and Hirsch, 2002; Duan, 1983).

All variables were log10 transformed prior to analysis to achieve normality and homogeneity of variance.

In total, collection goals were met and statistical analysis was possible for 26 of the 30 waterbodies. Statistical tests showed no difference in bass mercury concentrations between the first and second sampling visits for over half of the waterbodies (54%). Mercury levels in bass increased in 35% of waterbodies (9 out of 26). Three lakes (12%) showed decreases in mercury concentrations. The average percent change in estimated mercury values was 37.1% for sites showing an increase in mercury concentrations and -36.0% for the sites with decreases.

The lack of trends in half of the waterbodies is consistent with other North American reports on recent temporal trends in mercury concentrations in fish (Eagles-Smith et al., 2016; Tang et al., 2013; Chalmers et al., 2011). Dramatic decreases in fish mercury concentrations typically occurred following reductions in mercury emissions in the 1970s and 1980s. In Washington State, mercury concentrations in precipitation and wet deposition have remained moderately low since the closure of medical waste incinerators in the 1990s (Prestbo and Gay, 2009).

Mercury accumulation in bass is determined by a complex set of factors, including the amount of mercury loading to the waterbody, the availability of that mercury to the trophic system (i.e., methylation), and food web dynamics. The following sections discuss the trends results in relation to these factors for the statewide dataset as a whole. Annual reports provide a more detailed discussion of trends results for individual waterbodies.

Mercury Loading

While mercury concentrations and wet deposition in the Northwest U.S. have remained relatively unchanged over the last two decades (Weiss-Penzias et al., 2016; Prestbo and Gay, 2009), the amount of mercury that reaches a waterbody is controlled by watershed-specific factors. Watershed development can result in increased lake sediment mercury accumulation rates (Drevnick et al., 2016) through mobilization of water and mercury-bound sediment to the waterbody (Domagalski et al., 2016). Watershed land uses and degree of development did not appear to explain mercury trends seen in the Washington State bass. Landscape changes assessed through GIS photo-imagery between the collection periods were qualitatively examined and did not reveal any apparent contributing factors.

Ecology has analyzed mercury in sediment cores collected from nine of the lakes in this study. Sediment cores from Lake St. Clair and Lake Goodwin showed increases in mercury concentrations and fluxes since the 1990s (Furl, 2007; Mathieu and McCall, 2015b). Mercury levels in bass also increased in these lakes: by 33% in St. Clair and 52% in Goodwin. The consistent trend in both fish tissue and sediment fluxes suggests that recent increases in mercury loading may be at least partly responsible for the increase seen in bass concentrations in these two lakes. The trend in mercury concentrations in bass from Lake Offutt was also consistent with that of the sediment core collected there (Furl et al., 2009). Both fish tissue mercury levels and recent (since 1990s) sediment mercury concentrations and fluxes were unchanged in Lake Offutt.

Trend direction was inconsistent at the other six lakes with sediment core data. Sediment mercury concentrations and fluxes decreased since the 1990s at Loon and Ozette Lakes, yet mercury levels in bass showed no change (Furl, 2008; Furl, 2007). Decreases were also seen in

Lake Sammamish sediment concentrations and fluxes (Furl, 2007), whereas bass mercury concentrations increased over the recent five-year period. Conflicting trends were also seen at Deer, Samish, and Nahwatzel Lakes. Sediment mercury loading appears not to have been a key factor affecting fish tissue levels in these lakes. However, variation in mercury mobilization rates from the watershed can result in time lags ranging up to decades or longer until a response in fish mercury levels are seen (Munthe et al., 2007).

Water Chemistry and Waterbody Physical Factors

Correlations between percent change in bass mercury levels and variables potentially affecting mercury methylation were examined to explore patterns that may explain trends in fish tissue concentrations across the 26 waterbodies. Summer water samples collected in the corresponding years of fish collections at the waterbodies have been analyzed for dissolved organic carbon (DOC) and alkalinity. Water profile measurements of temperature, pH, conductivity, and dissolved oxygen (DO) were also taken. No relationships were found between the percent change in bass mercury levels and the difference in first and second visit water chemistry values, on a statewide scale. However, it should be noted that water sampling occurred as a discrete sampling event in one year, whereas fish tissue samples are an integration of multiple years of exposure.

The difference in average annual precipitation values for the five-year period preceding fish collections did not correlate with percent change in bass mercury concentrations. Other factors, such as physical features of the waterbody (i.e., drainage area to surface area ratios, lake volume, elevation, etc.), also did not reveal any patterns related to mercury trends. Furthermore, geographic location did not appear to influence percent change in mercury levels, as correlations between percent change and latitude or longitude showed no relationship. However, a greater proportion of waterbodies on the west side of the state (7 out of 15) showed increased mercury concentrations in bass compared to the east side (2 out of 11). Figure 7 shows the trend in bass mercury concentrations across the statewide dataset.

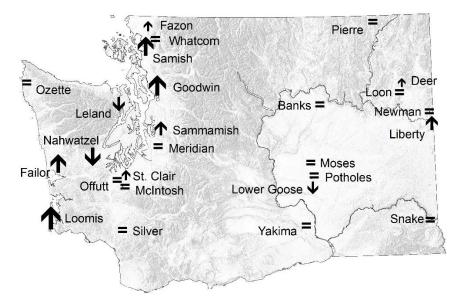


Figure 7. Temporal Trends of Bass Mercury Concentrations in a Five-Year Time Period. Arrows indicate increase or decrease, with size of arrow proportional to the percent change. An equals sign represents waterbodies where no significant difference was found.

Food Web Dynamics

Data useful for evaluation of changes in food web dynamics was not available for the two time periods. Indicators of trophic level, such as stable carbon and nitrogen isotope analyses, or bass diet assemblage data would be useful to help determine whether changes in mercury concentrations are influenced by increases or decreases in trophic chain lengths and/or the position of the bass in that trophic chain.

Changes in bass growth rates could influence trends in mercury concentrations. Relationships between fish size and age were evaluated through correlations. Fish size (total length and weight) to fish age relationships among the two sampling periods were very similar for most of the waterbodies. However, bass from Sammamish and Loomis Lakes were generally older at the same lengths and weights in the second visit compared to the first visit. This may help explain why mercury concentrations were higher in the bass collected in the second visit, as older fish have more time to accumulate mercury. Using age as the covariate for the Sammamish dataset resulted in no change in mercury concentrations. ANCOVA was not possible using age as the covariate for the Loomis dataset.

Leland Lake fish collected in the second visit were younger at the same lengths and weights than those collected in the first, particularly the bass at lengths above 400 mm. Replacing length with age as the covariate for ANCOVA with the Leland Lake fish resulted in no significant difference between the collection years. Growth dilution (the dilution of mercury in an organism as biomass increases) in the bass collected in the second visit may help explain the lower mercury concentrations found in those fish compared to those collected in the first visit.

Conclusions

Ecology collected largemouth and smallmouth bass from six waterbodies in the fall of 2014 as part of a long-term monitoring study to assess mercury trends in fish tissue. Fish were collected from Banks Lake, Failor Lake, Kitsap Lake, Pierre Lake, Snake River, and Lake Whatcom. A total of 60 individual bass were analyzed for total mercury. Results were compared to bass previously sampled from the waterbodies in 2009. Mercury was also analyzed in 17 composite samples of other fish species from the waterbodies. Results from the 2014 sampling include the following:

- Mercury concentrations of largemouth and smallmouth bass collected in 2014 ranged from 41.3 1,240 ppb across the waterbodies. Lake Whatcom bass contained the highest concentrations and Failor Lake the lowest. Mercury in composite samples of additional species ranged from 13.3 240 ppb.
- One sample an 8-year-old smallmouth bass from Lake Whatcom was above (did not meet) the state FTEC for Washington's Water Quality Assessment of 770 ppb.
- Sixteen bass samples were above (did not meet) the EPA Recommended Criterion of 300 ppb. All of the waterbodies except for Failor Lake had at least one sample with a mercury concentration above the EPA Recommended Criterion. However, this is not a regulatory threshold for Washington State.
- Statistical tests showed a significant difference in bass mercury levels between the two collection periods for one out of the five waterbodies. Bass from Failor Lake increased 44% in estimated mercury levels between the 2009 and 2014 collection years. No change was found in bass collected from Banks Lake, Pierre Lake, Snake River, or Lake Whatcom. However, Lake Whatcom bass collected in 2014 had significantly lower levels than bass collected in 2000.

Sampling in 2014 completed a cycle of re-visiting all waterbodies for the Mercury Trends in Fish Tissue project. Ten bass were analyzed for mercury from six waterbodies annually between 2005 through 2009, and these waterbodies were re-sampled after a five-year-period. This report summarized trend results from all waterbodies sampled in the last ten years with the following conclusions:

• Statistical analysis was possible for 26 of the 30 waterbodies. Results showed no difference in bass mercury concentrations between the first and second sampling visits for over half of the waterbodies (54%). Bass mercury levels increased in 35% of waterbodies (9 out of 26). Three lakes (12%) showed decreases in mercury concentrations. The lack of trends in the statewide dataset is consistent with other reports across North America on recent temporal trends in mercury concentrations in fish.

Recommendations

Results of this study support the following recommendations:

- The mercury concentrations in fish tissue reported for the 2014 sampling effort should be reviewed by Ecology during the next Water Quality Assessment and by DOH for assessment and updating of fish consumption advisories.
- Sampling for this project should continue until at least all of the waterbodies have been sampled a total of 3 times, which will be completed in 2019. At that time, another report summarizing trends from all of the waterbodies together should be written and goals/objectives of the project re-assessed.
- Sampling should be added to the project that allows for characterizing the trophic level of the bass being analyzed for mercury.
- Ecology should consider conducting a study to investigate mercury concentrations in surface water of the waterbodies that showed significant trends in fish mercury levels. The current study is limited to trends on a broader statewide scale, but a more detailed examination of the mercury budget in individual waterbodies or watersheds would help determine causes of increases or decreases in fish mercury concentrations.

References

Bloom, N., 1995. Considerations in the Analysis of Water and Fish for Mercury. In National Forum on Mercury in Fish: Proceedings. U.S. Environmental Protection Agency Office of Water, Washington D.C. EPA Publication 823-R-95-002.

CFR, 2004. Code of Federal Regulations, Title 40, Part 131, Section 36: Toxics Criteria for Those States Not Complying with Clean Water Act Section 303(3)(2)(B). Revised July 1, 2004.

Chalmers, A.T., D.M. Argue, D.A. Gay, M.E. Brigham, C.J. Schmitt, and D.L. Lorenz, 2011. Mercury Trends in Fish from Rivers and Lakes in the United States, 1969-2005. Environmental Monitoring and Assessment, Vol. 175: 175-191.

Domagalski, J., M.S. Majewski, C.N. Alpers, C.S. Eckley, C.A. Eagles-Smith, L. Schenk, and S. Wherry, 2016. Comparison of Mercury Mass Loading in Streams to Atmospheric Deposition in Western North America: Evidence for Non-Atmospheric Mercury Sources. Science of the Total Environment, doi: 10.1016/j.scitotenv.2016.02.112.

Drevnick, P.E., C.A. Cooke, D. Barraza, J.M. Blais, K.H. Coale, B.F. Cumming, C.J. Curtis, B. Das, W.F. Donahue, C.A. Eagles-Smith, D.R. Engstrom, W.F. Fitzgerald, C.V. Furl, J.E. Gray, R.I. Hall, T.A. Jackson, K.R. Laird, W.L. Lockhart, R.W. Macdonald, M.A. Mast, C. Mathieu, D.C.G. Muir, P.M. Outridge, S.A. Reinemann, S.E. Rothenberg, A.C. Ruiz-Fernandez, V.L. St. Louis, R.D. Sanders, H. Sanei, E.K. Skierszkan, P.C. Van Metre, T.J. Veverica, J.A. Wiklund, and B.B. Wolfe, 2016. Spatiotemporal Patterns of Mercury Accumulation in Lake Sediments of Western North America. Science of Total Environment, doi: 10.1016/j.scitotenv.2016.03.167.

Driscoll, C., C. Yan, C. Schofield, R. Munson, and J. Holsapple, 1994. The Mercury Cycle and Fish in the Adirondack Lakes. Environment Science and Technology, Volume 28, No. 3. American Chemical Society.

Duan, N., 1983. Smearing Estimate: A nonparametric retransformation method. Journal of American Statistical Association, Vol. 78: 605-610.

Eagles-Smith, C., J.T. Ackerman, J.J. Willacker, M.T. Tate, M.A. Lutz, J.A. Fleck, A.R. Stewart, J.G. Wiener, D.C. Evers, J.M. Lepak, J.A. Davis, and C. Flanagan Pritz, 2016. Spatial and Temporal Patterns of Mercury Concentrations in Freshwater Fish across the Western United States and Canada. Science of the Total Environment, doi: 10.1016/j.scitotenv.2016.03.229.

EPA, 2000a. Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories. Volume II – Risk Assessment and Fish Consumption Limits, Third Edition. U.S. Environmental Protection Agency, Office of Water, Washington D.C. Publication Number EPA-823-B-00-008.

EPA, 2000b. Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories – Volume 1: Field Sampling and Analysis, Third Edition. U.S. Environmental Protection Agency, Office of Water, Washington D.C. Publication No. EPA-823-B-00-007.

EPA, 2001. Water Quality Criterion for the Protection of Human Health: Methylmercury. U.S. Environmental Protection Agency, Office of Science and Technology, Washington D.C. Publication No. EPA-823-R-01-001.

Fischnaller, S., P. Anderson, and D. Norton, 2003. Mercury in Edible Fish Tissue and Sediments from Selected Lakes and Rivers of Washington State. Washington State Department of Ecology, Environmental Assessment Program. Olympia, WA. Ecology Publication No. 03-03-026 www.ecy.wa.gov/biblio/0303026.html

Helsel, D.R. and R.M. Hirsch, 2002. Statistical Methods in Water Resources, Techniques of Water Resources Investigations, Book 4, chapter A3. U.S. Geological Survey. 522 pages.

Grieb, T.M., C.T. Driscoll, S.P. Gloss, C. L. Schofield, G.L. Bowie, and D.B. Porcella, 1990. Factors Affecting Mercury Accumulation in Fish in the Upper Michigan Peninsula. Environmental Toxicology and Chemistry. Vol. 9: 919-930.

Kelly, C.A., J.W.M. Rudd, and M.H. Holoka, 2003. Effect of pH on Mercury Uptake by an Aquatic Bacterium: Implications for Hg Cycling. Environmental Science and Technology, Vol. 37: 2941-2946.

Mathieu, C. and M. Friese, 2012. Mercury Trends in Freshwater Fish, 2011 Sampling Results. Washington State Department of Ecology, Olympia, WA. Publication No. 12-03-051. https://fortress.wa.gov/ecy/publications/summarypages/1203051.html

Mathieu, C., M. Friese, and A. Bookter, 2013a. Mercury Trends in Freshwater Fish, 2012 Sampling Results. Washington State Department of Ecology, Olympia, WA. Publication No. 13-03-044. <u>https://fortress.wa.gov/ecy/publications/SummaryPages/1303044.html</u>

Mathieu, C., C. Furl, T. Roberts, M. Friese, 2013b. Spatial Trends and Factors Affecting Mercury Bioaccumulation in Freshwater Fishes of Washington State, USA. Archives of Environmental Contamination and Toxicology, Volume 65: 122-13.

Mathieu, C. and M. McCall, 2015a. Mercury Trends in Freshwater Fish, 2013 Sampling Results. Washington State Department of Ecology, Olympia, WA. Publication No. 15-03-010. <u>https://fortress.wa.gov/ecy/publications/SummaryPages/1503010.html</u>

Mathieu, C. and M. McCall, 2015b. PBT Chemical Trends Determined from Age-Dated Lake Sediment Cores, 2014 Results. Washington State Department of Ecology, Olympia, WA. Publication No. 15-03-027. https://fortress.wa.gov/ecy/publications/SummaryPages/1503027.html

Meredith, C., C. Furl, M. Friese, 2010. Measuring Mercury Trends in Freshwater Fish in Washington State, 2009 Sampling Results. Washington State Department of Ecology, Olympia, WA. Publication No. 10-03-058 <u>http://www.ecy.wa.gov/biblio/1003058.html</u>

Meredith, C. and M. Friese, 2011. Measuring Mercury Trends in Freshwater Fish in Washington State, 2010 Sampling Results. Washington State Department of Ecology, Olympia, WA. Publication No. 11-03-053.

https://fortress.wa.gov/ecy/publications/summarypages/1103053.html

Meredith, C. and C. Furl, 2010. Quality Assurance Project Plan Addendum: Measuring Mercury Trends in Freshwater Fish in Washington State. Washington State Department of Ecology, Olympia, WA. Publication No. 06-03-103Addendum1. www.ecy.wa.gov/biblio/0603103Addendum1.html

Munthe, J., R.A. Bodaly, B.A. Branfireun, C.T. Driscoll, C.C. Gilmour, R. Harris, M. Horvat, M. Lucotte, and O. Malm, 2007. Recovery of Mercury-Contaminated Fisheries. Ambio. Vol. 36: 33-44.

Norton, D., 2004. Mercury in Lake Whatcom Sediments: Spatial Distribution, Depositional History, and Tributary Inputs. Department of Ecology, Environmental Assessment Program, Olympia, WA. Ecology Publication No. <u>www.ecy.wa.gov/biblio/0403019.html</u>

Peele, C., 2003. Washington State Mercury Chemical Action Plan. Washington State Departments of Ecology and Health, Olympia, WA. Ecology Publication No. 03-03-001. www.ecy.wa.gov/biblio/0303001.html

Prestbo, E.M. and D.A. Gay, 2009. Wet Deposition of Mercury in the U.S. and Canada, 1996-2005: Results and Analysis of the NADP Mercury Deposition Network (MDN). Atmospheric Environment, Vol. 43: 4223-4233.

Sandvik, P., 2014a. Standard Operating Procedure for Field Collection, Processing and Preservation of Finfish Samples at the Time of Collection in the Field, Version 1.0. Washington State Department of Ecology, Olympia, WA. SOP No. EAP009. www.ecy.wa.gov/programs/eap/quality.html

Sandvik, P., 2014b. Standard Operating Procedure for Resecting Finfish Whole Body, Body Parts, or Tissue Samples, Version 1.0. Washington State Department of Ecology, Olympia, WA. SOP No. EAP007. <u>www.ecy.wa.gov/programs/eap/quality.html</u>

Seiders, K., 2006. Measuring Mercury Trends in Freshwater Fish in Washington State. Quality Assurance Project Plan. Washington State Department of Ecology, Olympia, WA. Publication No. 06-03-103. <u>www.ecy.wa.gov/biblio/0603103.html</u>

Selin, N., 2009. Global Biogeochemical Cycling of Mercury: A Review. Annual Review of Environmental and Resources. Vol 34: 43-63.

Serdar, D., J. Johnston, K. Mueller, and G. Patrick, 2001. Mercury Concentrations in Edible Muscle of Lake Whatcom Fish. Washington State Departments of Ecology and Health, Olympia, WA. Ecology Publication No. 01-03-012. <u>www.ecy.wa.gov/biblio/0103012.html</u> Swanson, T., 2010. Standard Operating Procedures for Hydrolab DataSonde® and MiniSonde® Multiprobes. Washington State Department of Ecology, Olympia, WA. SOP No. EAP033. www.ecy.wa.gov/programs/eap/quality.html

Tang, R.W.K., T.A. Johnston, J.M. Gunn, and S.P. Bhavsar, 2013. Temporal Changes in Mercury Concentrations of Large-Bodied Fishes in the Boreal Shield Ecoregion of Northern Ontario, Canada. Science of the Total Environment, Vol. 444: 409-416.

Weiss-Penzias, P.S., D.A. Gay, M.E. Brigham, M.T. Parsons, M.S. Gustin, and A. Schure, 2016. Trends in Mercury Wet Deposition and Mercury Air Concentrations across the U.S. and Canada. Science of the Total Environment, doi:10.1016/j.scitotenv.2016.01.061.

Xun, L., N.E.R. Campbell, and J.W.M. Rudd, 1987. Measurements of Specific Rates of Net Methyl Mercury Production in the Water Column and Surface Sediments of Acidified and Circumneutral Lakes. Canadian Journal of Fisheries and Aquatic Sciences, Vol. 44: 750-757.

Appendices

Appendix A. Water Sample Results from 2009 and 2014

Waterbody	Depth (m)	2009 Alkalinity (mg/L)	2014 Alkalinity (mg/L)		
Banks	6	57.4	57.9		
Dariks	14.5	61.1	58.2		
Failor	1.2	17.4	16.3		
FallOI	3.0	20.6	21.4		
Pierre	3.0	135	131		
Pierre	12	180	184		
Whatcom	6.0	21.2	46.9		
whatcom	27	17.9	34.2		

Table A-1. Alkalinity in Surface Waters Collected in 2009 and 2014.

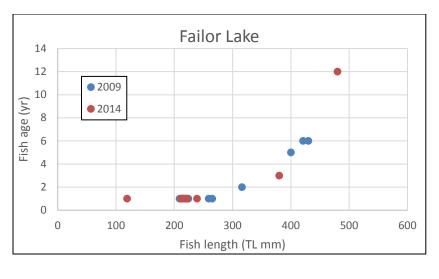
Table A-2. Dissolved Organic Carbon (DOC) in Surface Waters Collected in 2009 and 2014.

Waterbody	Depth (m)	2009 DOC (mg/L)	2014 DOC (mg/L)	
Banks	6	1.4	2	
Daliks	14.5	1.3	1.6	
Failor	1.2	1.6	2.1	
FallOI	3.0	1.3	1.8	
Pierre	3.0	3.8	3.6	
Fielle	12	2.9	3.1	
Whatcom	6.0	1.8	1.7	
Whatcom	27	1.6	1.6	

Table A-3. pH, Dissolved Oxygen (DO), and Conductivity Measured In-Situ in 2009 and 2014.

	Depth	2009	2014	2009	2014	2009	2014	
Waterbody	(m)	рН	рН	DO (mg/L)	DO (mg/L)	Cond (uS/cm)	Cond (uS/cm)	
Failor	1.2	7.80	8.56	7.50	8.31	111.5	128	
FallOI	3.0	7.12	8.12	2.98	7.11	120.7	130	
Whatcom	6.0	9.56	9.63	9.56	11.22	161.0	160	
Whatcom	27	9.67	9.71	9.55	11.52	161.0	160	

Water profiles not available for Banks or Pierre Lakes.



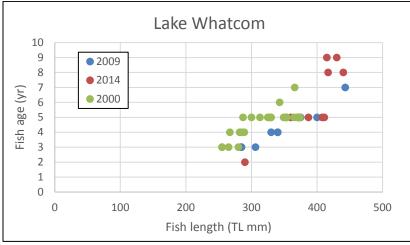


Figure A-1. Fish Length to Age Relationships for Failor and Whatcom Lakes.

Appendix B. Statewide Dataset

Date of first			Dominant	Percent	Drainage	Elevation	Surface	DA:SA	Difference in values for first and second sampling collections:							
Waterbody	sampling visit	sampling visit	land type	change in Hg _{bass}	area (ac)	(ft)		ratio	annual precip.*	Hg:mm slopes	FFI	mean alkalinity	mean DOC	epi. pH	hypo. pH	hypo. DO
Liberty	2005	2010	R, F	34%	8512	2,053	713	12	1.6	1.841	0.01	-1.55	-0.7	-2.1	-1.8	1.1
Loon	2005	2010	R, F		9024	2,381	1,130	8	1.3	-0.665	-0.26	0.9	0.9	-0.6	-0.3	-0.3
Potholes	2005	2010	В, А		25,008,800	1,046	28,000	893	0.2	-0.728	0.03	-14.5	0.1	-1	-0.1	-0.2
Silver	2005	2010	F		25152	484	2,300	11	2.6	0.057	0.02	-17.7	6.2	0.2	-0.1	2.7
Yakima	2005	2010	В, А		6120	410	-	-	-0.2	-0.516	0.00	-15	0.1			-1.6
Meridian	2006	2011	R, U		742	370	150	5	3	-0.247	-0.16	0.7	0.4	0.2	0.17	0
Moses	2006	2011	U, B		1,971,200	1,046	6,800	290	0.4	0.812	-0.03	6	0.1	-0.18	-1.39	-10.16
Newman	2006	2011	F, R		18,304	2,124	1,200	15	0.2	1.1	-0.02	-1.5	10.9	-0.37	-0.51	-6.82
Offutt	2006	2011	R, O		1,728	230	200	9	6.8	0.624	-0.03					
Sammamish	2006	2011	R, F	34%	62,720	26	4,900	13	5.1	-0.36	-0.10	5.1	0.2	-0.04	-0.04	0.78
Deer	2007	2012	F, R	22%	11,648	2,474	1,100	11	0.1	-0.957	0.13	-0.3	1.55	0.25	0.43	0.34
Fazon	2007	2012	F	25%	621	128	31	20	-0.9		0.08	7.7	-2.5	0.64	0.11	-1.65
Lower Goose	2007	2012	В	-30%	-	856	50	-	-1	1.683	0.09	-7.6	0.25	0.39	0.68	-1.98
Ozette	2007	2012	F		49,600	29	7,300	7	1.3	0.075	0.15	-6.3	0.85	0.19	0.29	-0.97
Samish	2007	2012	F, R	46%	5,888	273	680	9	-2.9	-0.328	-0.24	6.5	0.85	-0.49	0.18	2.31
St. Clair	2007	2012	R, F	25%	9,280	73	88	105	0.8	-0.854	0.10	2	0.8	0.35	0.31	-3.35
Goodwin	2008	2013	R, F	49%	3,310	324	560	6	3.5		0.01	-4.85	-0.1	0.75	0.08	-0.14
Leland	2008	2013	F	-34%	3,650	190	110	33	4.3	1.244	-0.06	-0.6	-3.45	-0.34	-0.11	-0.07
Loomis	2008	2013	W <i>,</i> F	55%	922	17	170	5	0.3	-0.157	-0.18	5.6	1.35	0.07	0.04	1.97
McIntosh	2008	2013	F, R		1,450	336	93	16	1.3	-1.194	-0.08	5.35	0.7	-0.22	-0.47	-0.76
Nahwatzel	2008	2013	F	-44%	3,970	440	270	15	-0.5	-1.559	0.06	-1.8	0.1	0.17	0.58	0.58
Banks	2009	2014	В, А		-	1,570	27,000	-	0.8	1.202	-0.16	-1.2	0.45			
Failor	2009	2014	F	44%	3,130	117	65	48	4.8	-0.693	0.30	-0.15	0.5	-1.54	-1.82	-0.64
Pierre	2009	2014	F		17,152	2,005	110	156	0.8		-0.05	0.0	0.0			
Snake	2009	2014	В, А		-	700	-	-	2.5	-0.813	0.12					
Whatcom	2009	2014	F, R		35,776	315	5,000	7	-7	0.161	0.12	21	-0.05	-0.15	-0.67	0.36

Table B-1. Select Data Examined for Statewide Trends in Bass Mercury Levels.

A: agriculture, B: barren, F: forested, O: open space, R: residential, U: urban, W: wetlands *Mean annual precipitation values for five-year period preceding fish collections.

Appendix C. Glossary, Acronyms, and Abbreviations

Glossary

Alkalinity: The quantitative capacity of water to neutralize an acid.

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.

Dissolved organic carbon (DOC): The fraction of total organic carbon in water that passes through a 0.45 micron pore-diameter filter.

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

Fish Tissue Equivalent Concentration (FTEC): The FTECs is a tissue contaminant concentration used by Ecology to determine whether the designated uses of fishing and drinking from surface waters are being met. The FTEC is an interpretation of Washington's water quality criterion for a specific chemical for the protection of human health: the National Toxics Rule (40 CFR 131.36). Fish tissue sample concentrations that are lower than the FTEC suggest that the uses of fishing and drinking from surface waters are being met for that specific contaminant. Where a FTEC is not met (i.e., concentration of a chemical in fish tissue is greater than the FTEC), that water body is then placed into Category 5 during Washington's periodic Water Quality Assessment (www.ecy.wa.gov/programs/wq/303d/index.html). Category 5 listings become part of Washington's 303(d) list during the assessment process. The FTEC is calculated by multiplying the contaminant-specific Bio-Concentration Factor (BCF) times the contaminant-specific Water Quality Criterion found in the National Toxics Rule.

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

Growth dilution: The dilution of a contaminant in an organism as biomass increases from growth, resulting in a lower contaminant concentration.

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

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 trends: Characterization of trends 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.

Water Quality Assessment: 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.

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

ANCOVA	Analysis of Covariance
ANOVA	Analysis of Variance
DOC	Dissolved organic carbon
Ecology	Washington State Department of Ecology
EIM	Environmental Information Management database
EPA	U.S. Environmental Protection Agency
FTEC	Fish Tissue Equivalent Concentration
GIS	Geographic Information System software
Hg	Mercury
MEL	Manchester Environmental Laboratory
NTR	National Toxics Rule
PBT	persistent, bioaccumulative, and toxic substance
RPD	Relative percent difference
SOP	Standard operating procedures
SRM	Standard reference materials
WAC	Washington Administrative Code
WRIA	Water Resource Inventory Area

Units of Measurement

°C	degrees centigrade
ft	feet
g	gram, a unit of mass
mg/L	milligrams per liter (parts per million)

mm	millimeters
ng/L	nanograms per liter (parts per trillion)
ppb	parts per billion
ppm	parts per million
ug/Kg	micrograms per kilogram (parts per billion)
ug/L	micrograms per liter (parts per billion)