Measuring Mercury Trends in Freshwater Fish in Washington State

2017-2019 Sampling Results

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by

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Abstract

Since 2005, the Washington State Department of Ecology (Ecology) has carried out a long-term monitoring study of mercury levels in freshwater fish. Each year Ecology collects ten individual largemouth or smallmouth bass from six waterbodies and analyzes total mercury in the fillet tissue. Sites are re-sampled every five years to evaluate trends in freshwater fish mercury concentrations over time.

Results from the 2017, 2018, and 2019 sampling years are presented in this report. Mercury was detected in all individual bass analyzed, with concentrations ranging from 35-1,100 ppb wet weight (ww). All individual bass fillets contained total mercury levels higher than the state human health water quality criterion for methylmercury (30 ppb), and 75% exceeded the Washington State Department of Health screening level of 101 ppb.

As of 2019, all 30 sites in the Mercury Trends study have been sampled three times, five years apart. This report summarizes statewide trends over the 15 years of sampling since the start of the study. Most of the sites showed no change in bass mercury levels over both a five-year and ten-year period (65% and 63%, respectively). Mercury concentrations decreased in one-third of the sites over a five-year period and in 19% of sites over a ten-year period. Few waterbodies saw increases in bass mercury concentrations, with increases in 7% and 15% of the monitoring sites over the five-year and ten-year period, respectively.

The general spatial pattern of mercury accumulation in bass across the state has remained consistent over the course of 15 years of the long-term monitoring study. Bass concentrations remain highest on the coastal west side of the state and lowest in the arid central and eastern areas of the state.

Introduction

The Washington State Departments of Ecology (Ecology) and Health (DOH) develop chemical action plans (CAPs) to address the public health threat of toxic chemicals (toxics) in the environment. In the early 2000s, the state agencies identified chemicals that were persistent, bioaccumulative, and toxic (PBT) as priorities for toxics reductions efforts. PBTs are a concern because they do not break down easily in the environment and can enter the food web. Once in the food web, PBTs accumulate in organisms and increase along the trophic system, resulting in upper trophic level fish concentrations that can cause harm to people and animals. The first CAP was developed for mercury in 2003 (Peele et al. 2003).

Human activity has significantly increased the release and circulation of mercury in the environment (Selin 2009). Washington's CAP for mercury led to some controls preventing pathways of mercury into the environment. However, contamination continues to occur through global industrial processes and burning of fossil fuels, as well as natural processes. When mercury is released into the environment, it can enter the food web via a biogeochemical process called methylation. Methylmercury is a highly toxic and bioavailable form of mercury.

Mercury increases in concentration as it moves up the food web and bioaccumulates in higher trophic levels, including humans. In humans, mercury can have serious effects on the nervous system. Children and developing fetuses are most at risk for these effects. A major source of exposure to mercury is through eating fish.

In the early 2000s, Ecology studies first reported mercury contamination in Washington state fish, beginning with Lake Whatcom (Serdar et al. 2001) and then statewide (Fischnaller et al. 2003). The statewide survey found elevated levels of mercury in bass fillet tissue from 20 waterbodies and recommended a statewide monitoring program (Fischnaller et al. 2003). In response to that recommendation, and with legislative funding, Ecology began statewide monitoring of mercury in freshwater fish in 2005. As of 2019, the Mercury Trends study has completed 15 years of monitoring.

Methods

Study Design

Ecology's Mercury Trends study was created with the primary goal of characterizing temporal trends in mercury levels in upper trophic level fish in Washington state. We collect ten individual largemouth or smallmouth bass from six waterbodies each year for analysis of total mercury in the fillet tissue. We revisit the same waterbodies every five years to determine whether mercury concentrations are increasing, decreasing, or staying the same.

We also collect up to three ancillary fish species at each site, when encountered, for analysis as composite samples. The composite samples consist of 3-5 individual fish of similar size. Data from the ancillary species are used by DOH to inform public health guidance for fish consumption.

Study Locations

Fish collection locations for the Mercury Trends study include 30 sites statewide. Figure 1 shows the locations sampled during the 2017-2019 sampling period. All sites were selected for this study based on criteria outlined in the original Quality Assurance Project Plan (QAPP; Seiders 2005).

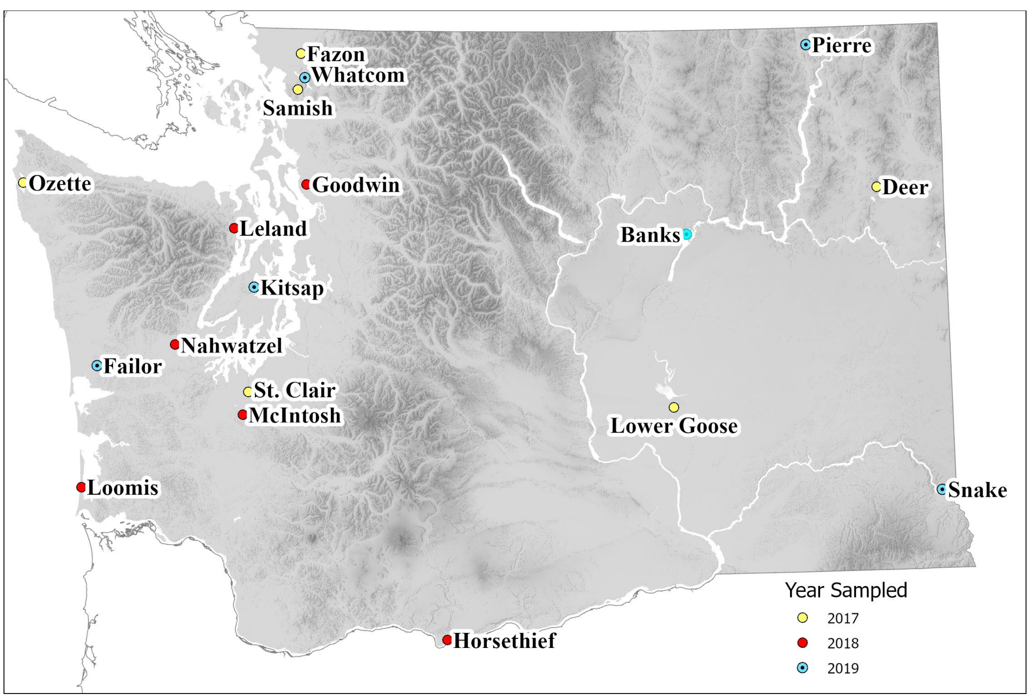

Figure 1. Map of 2017-2019 sampling locations.

Surrounding land uses of the monitoring sites include forests, wetlands, grasslands, as well as human-dominated landscapes such as residential, urban, or agricultural development. Waterbodies are located on both sides of the Cascade Mountains to include coastal and inland arid climates. Table 2 shows the dominant characteristics of each site sampled from 2017 through 2019.

Table 1. Climate and land-use characteristics of sites sampled in 2017-2019.

Location	Drainage area (sq mi)	Elevation (ft)	Annual precip. (in)	Surface area (acres)	Lake volume (acre-ft)	Mean depth (ft)	Max depth (ft)	Dominant Land Type
Deer Lake	18	2,474	26	1,110	57,000	52	75	F, R
Fazon Lake	0.9	128	42	31	300	10	17	F, A
Lower Goose Lake	---	856	8	50	1,300	25	75	G, A
Ozette Lake	78	29	105	7,300	960,000	130	320	F
Lake Samish	13	273	56	810	33,900	51	140	F, R
Lake St. Clair	21	73	52	268	8,700	34	110	F,R
Lake Goodwin	5.2	324	37	560	13,000	23	50	F, R
Horsethief Lake	---	160	14	92	---	---	---	G
Leland Lake	5.7	190	39	110	1,400	13	20	F
Loomis Lake	1.4	17	79	170	830	5	9	F, W
McIntosh Lake	2.2	336	52	93	700	8	11	F, R
Nahwatzel Lake	6.2	440	100	270	4,600	17	25	F
Banks Lake	---	1,570	11	27,000	1,300,000	47	85	G, A
Failor Lake	4.9	117	110	65	500	8	22	F
Kitsap Lake	2.7	127	54	248	4,480	18	29	R
Pierre Lake	27	2,005	27	110	3,000	28	75	F
Snake River	---	700	13	---	---	12	50	G, A
Lake Whatcom	56	315	49	5,000	770,000	150	330	F, R

F = Forest; G = Grassland; A = Agricultural; R = Residential; W = wetland

Field Methods

We collected samples primarily by electrofishing boat. In 2018, gillnets were used for sample collection at Lake Meridian to obtain the collection target. In 2019, WDFW collected samples from Banks Lake as part of their walleye population survey. At each site, field crews made efforts to match size classes of fish to those of previous sampling years. Matching size classes is important for determining long-term trends because size is a covariate to mercury concentrations.

We took several measures to prevent contamination of field samples. Field staff wore nitrile gloves while handling fish. Fish were double wrapped in aluminum foil, sealed in durable plastic bags, and stored on ice until transport back to Ecology Headquarters, where they were stored frozen at -20 degrees Celsius.

Field crews documented all field activities in a logbook. Locations of fishing activity were marked on a map or described in a narrative. Electrofishing settings and duration of current used in the water were logged as required by electrofishing permits. Fish were collected until ten samples that met criteria for analysis were obtained. Fish not meeting size criteria were released. Fish that did meet criteria were euthanized, weighed, and measured. Field lengths and weights were recorded in a logbook. Samples were carefully and meticulously labelled. Field methods are described in detail in Ecology's Standard Operating Procedure (SOP) EAP009 (Sandvik, 2023a). Any deviance from the SOPs were noted and justified in the field logbooks.

Laboratory Analysis

We followed Ecology SOP EAP007 (Sandvik, 2023b) for fish resection and sample preparation. The procedure was conducted at Ecology headquarters several weeks after sample collection. During resection, we collect fish aging structures for analysis by the Washington Department of Fish and Wildlife (WDFW) aging lab. Fish were descaled and filleted. Edible portions of the fish with skin-on were passed through a Kitchen-Aid meat grinder three times to fully homogenize the sample. Composite samples were grouped so that the smallest fish is no more than 25% smaller than the largest fish in the sample. We archived excess portions of the samples until results from the lab were evaluated.

Samples were delivered to Ecology's Manchester Environmental Laboratory (MEL) under chain of custody procedures. MEL performed the analysis using cold vapor atomic absorption (EPA Method 245.6) and reported mercury concentrations in ug/Kg wet weight (ww), which is equivalent to parts per billion (ppb). MEL provided case narratives documenting the method used, quality control (QC) test results, and sample results. The project manager reviewed and accepted all data.

Data Quality

The QAPP (Seiders 2005) describes measurement quality objectives (MQOs) to ensure credibility of data we collect. Most of these objectives were met or fell just outside of established limits. MEL case narratives describe the details of whether data met MQOs. MEL reported that all samples were received frozen and in good condition upon delivery. Lab analyses were conducted within method holding times.

Some MQOs exceeded the limits provided in the QAPP but did not require qualification to the data. Reporting limits were higher than the target of 0.017 mg/Kg. However, all results were well above reporting limits, which adds certainty to the interpretation of results. Standard reference material (SRM) recoveries were 4% over target in 2017 and 2019. However, MEL does not have QC acceptance limits for this type of SRM and does not qualify results based on mercury SRMs due to observed variability in SRM recoveries. All laboratory control samples, matrix spikes, and laboratory duplicates were well within quality objectives.

Collection Goals

We met collection goals for trends analysis with one exception: Eight bass were collected at Horsethief lake. We reported trends for this site regardless of the lower number under the provision that statistical power may be limited for this observation. We did not meet ancillary species collection goals for Deer Lake in 2017, nor at any site in 2018 and 2019, due to limited staff resources.

We met water sampling goals for 2017. However, water sampling was discontinued for this project in 2018 because relationships between water quality characteristics and mercury in fish trends were not strong enough to justify the continued effort. Water quality data are not reported in this report. The data are available through Ecology's Environmental Information Management (EIM) database and used potentially for other assessments such as the Water Quality Assessment under the Clean Water Act

Table 2. Sample collection goals by year, 2017-2019.

Year	Site	10 individual bass	Ancillary species	2 water samples	Hydrolab profile
2017	Deer Lake	+	NA	+	+
2017	Fazon Lake	+	BG	+	+
2017	Lower Goose Lake	+	YP	+	+
2017	Ozette Lake	+	BBH, NPM, YP	+	+
2017	Samish Lake	+	BBH, KOK	+	+
2017	St. Clair Lake	+	YP	+	+
2018	Horsethief Lake*	8	NA	NA	NA
2018	Lake Goodwin	+	NA	NA	NA
2018	Leland Lake	+	NA	NA	NA
2018	Loomis Lake	+	NA	NA	NA
2018	McIntosh Lake	+	NA	NA	NA
2018	Nahwatzel Lake	+	NA	NA	NA
2019	Banks Lake	+	NA	NA	NA
2019	Failor Lake	+	NA	NA	NA
2019	Kitsap Lake	+	NA	NA	NA
2019	Lake Whatcom	+	NA	NA	NA
2019	Pierre lake	+	NA	NA	NA
2019	Snake River	+	NA	NA	NA

BG = Bluegill; NPM = northern pikeminnow; YP = yellow perch; KOK = kokanee; BBH = brown bullhead

+ = Collection goal met; NA = Goal not attained; + = Goal sufficiently met

Statistical Analysis

Statistical summaries of fish and mercury concentrations are calculated for each site and presented in the following section. Mercury concentrations are reported for individual fish and compared to the Washington State water quality criterion (WQC) and the Washington State Department of Health (DOH) screening levels (SLs) for methylmercury. See Appendix A for a description of these thresholds and how they are used.

We used analysis of covariance (ANCOVA) to assess trends in individual bass mercury concentrations over time. Fish length was the most consistent covariate to control for the effect of fish size on mercury accumulation in fish. However, weight and age are closely related covariates that were used if length relationships were found to be weak. We chose covariates based on the strength of the relationship and the ability to meet ANCOVA model assumptions. All data analysis was performed using R (R Core Team 2021).

Data were log_{10}-transformed to increase the likelihood of meeting assumptions of normal distribution. We tested model assumptions with a generalized linear model to test relationships between fish size and mercury concentrations. Visual inspection of Q-Q plots and Shapiro-Wilks tests confirmed normality of the datasets. Outliers were evaluated qualitatively and removed with justification as documented in the results.

We analyzed trends over three observation periods for all waterbodies. We quantified significant differences using Bonferroni adjusted pairwise comparison and least square means comparison. Least squares means from post-hoc tests were back transformed and corrected using Duan's smearing estimator to adjust for log transformation bias (Helsel and Hirsch 2002; Duan 1983). In the following sections, we report trends as significant differences between the most recent reporting period, compared to past sample collections.

Studies conducted by Fischnaller (2003) and Serdar (2001) were used to add additional context to trends for three sites where baseline monitoring was conducted. In 2005, Ecology adopted U.S. Environmental Protection Agency (EPA) method 245.6 for analysis of mercury in fish tissue. Ecology found that EPA 245.5 method underreported mercury by 25 – 38 % (Furl 2007). Furl (2007) developed a conversion method using regression to account for differences between methods. We applied the conversion factor to data collected before 2005 to predict mercury values that could be compared to post-2005 values. Note that using a predictive relationship to estimate values, and then using those values in the ANCOVA, would result in propagation of errors. However, the impact of this may be minimal due to the strength of the regression ($R^2 = 0.977$, $F(1, 85) = 3679.38$, $p < 0.001$) and low mean square error of the residuals (0.003).

2017-2019 Results

From 2017-2019, we analyzed 188 individual bass and walleye, as well as 14 composite samples of additional species, for mercury. The following sections summarize findings of individual fish sizes, mercury concentrations, relationships, and ancillary composite samples. Table 3 displays summary statistics for individual fish collected during this reporting period.

Table 3. Statistical summary of individual fish length, weight, age, and mercury concentrations, 2017-2019.

Waterbody	Species	Length (TL mm, range)	Weight (g, range)	Age (yr, range)	Mercury (ppb, range)	Mercury (ppb, median)
Banks Lake	SMB	229 - 450	331 - 1225	1 - 10	73.6 - 194	129
Deer Lake	LMB	293 - 472	352 - 1834	2 - 14	98.4 - 425	216
Failor Lake	LMB	198 - 485	104 - 2311	1 - 7	46.5 - 365	61.6
Fazon Lake	LMB	261 - 456	215 - 1821	2 - 9	199 - 524	281
Horsethief Lake	LMB	245 - 260	224 - 269	2 - 2	35.6 - 36.5	36.1
Horsethief Lake	SMB	231 - 487	163 - 784	1 - 9	40 - 307	104
Horsethief Lake	WAL	284 - 615	177 - 2123	1 - 4	36.3 - 231	54.9
Kitsap Lake	LMB	230 - 440	192 - 1642	1 - 4	41.9 - 240	66.2
Lake Goodwin	SMB	203 - 295	108 - 324	1 - 3	135 - 202	148
Lake Whatcom	SMB	330 - 433	534 - 1263	4 - 6	251 - 680	471
Leland Lake	LMB	247 - 426	195 - 1336	3 - 5	172 - 569	223
Loomis Lake	LMB	189 - 413	97 - 1102	2 - 8	63 - 343	94.7
L. Goose Lake	LMB	250 - 412	216 - 1174	1 - 3	74 - 134	96.7
McIntosh Lake	LMB	176 - 377	65 - 716	1 - 6	103 - 227	197
Nahwatzel Lake	LMB	238 - 390	172 - 964	3 - 9	193 - 547	278
Ozette Lake	LMB	275 - 430	295 - 1201	2 - 4	257 - 1060	474
Pierre lake	LMB	275 - 400	275 - 1053	2 - 7	99.8 - 274	160
Samish Lake	LMB	227 - 526	165 - 2930	1 - 13	56.5 - 1100	313
Snake River	LMB	235 - 467	225 - 1634	1 - 6	52 - 413	177
St. Clair Lake	LMB	309 - 498	385 - 2114	4 - 11	321 - 793	549

Individual and Composite Results

Individual Fish Sizes

A total of 81% of bass met the target size range of 250 – 460 mm (145 of 180 bass). Bass length to weight ratios showed a consistent pattern with few outliers. Figure 2 shows the relationship between length and weight for individual samples collected in this reporting period. Notable outliers occurred in two fish, one collected from Banks Lake and one from Horsethief Lake.

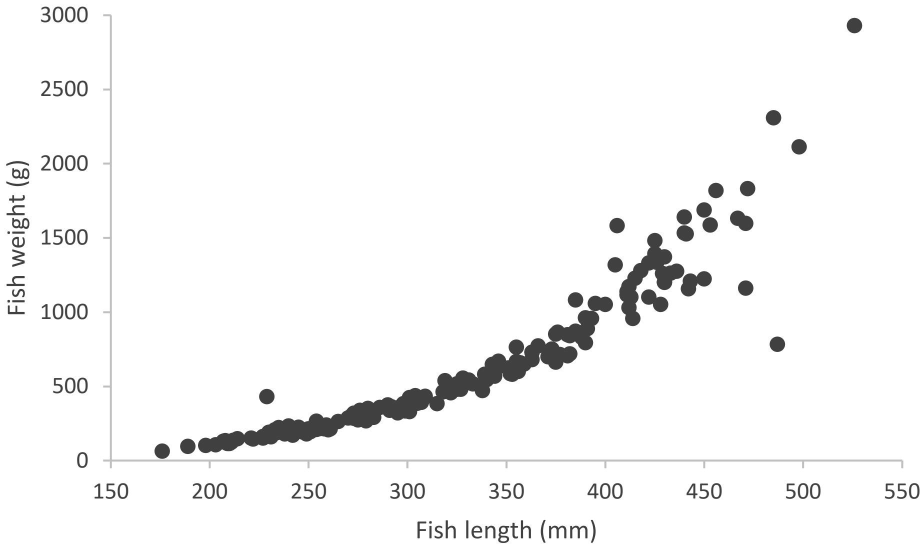

Figure 2. Length to weight relationship of individually analyzed bass.

Individual Mercury Concentrations

Figure 3 displays the mercury concentrations of individual bass and walleye collected during the 2017-2019 reporting period. Mercury was detected in all individual samples higher than the reporting limit. Mercury concentrations ranged from 35 to 1,100 ppb.

The highest individual mercury concentration was observed in a 13-year-old female largemouth bass collected from Lake Samish (1,100 ppb). Lake St. Clair bass contained the highest mercury concentrations (median = 549 ppb) and Horsethief Lake had the lowest concentrations (median = 55 ppb). Bass collected at Lake St. Clair were older on average than any other site (average of 6.7 years). Fish from Horsethief Lake were among the youngest on average (2.6 years) but not the youngest of all groups. Age of individual fish ranged from one to 14 years. The youngest fish were collected from Lake Goodwin.

Two methylmercury thresholds for human health – the state WQC and DOH SL – are shown in Figure 3 to provide context for the total mercury concentrations observed in fish tissue across the waterbodies. Appendix A describes the two thresholds in detail. It should be noted that not all of the total mercury in fish tissue is comprised of methylmercury and the percentage of methylmercury that makes up total mercury in bass varies greatly (Foster and Mathieu, 2023). Total mercury values presented in this report are compared to the methylmercury thresholds to provide context for the data and do not necessary indicate an exceedance. All individual samples had total mercury concentrations above the state WQC of 30 ppb. Seventy-five percent of samples were above the DOH SL of 101 ppb. For several waterbodies, all samples were above the DOH SL, including Fazon, Goodwin, Whatcom, Leland, McIntosh, Nahwatzel, Ozette, and St. Clair.

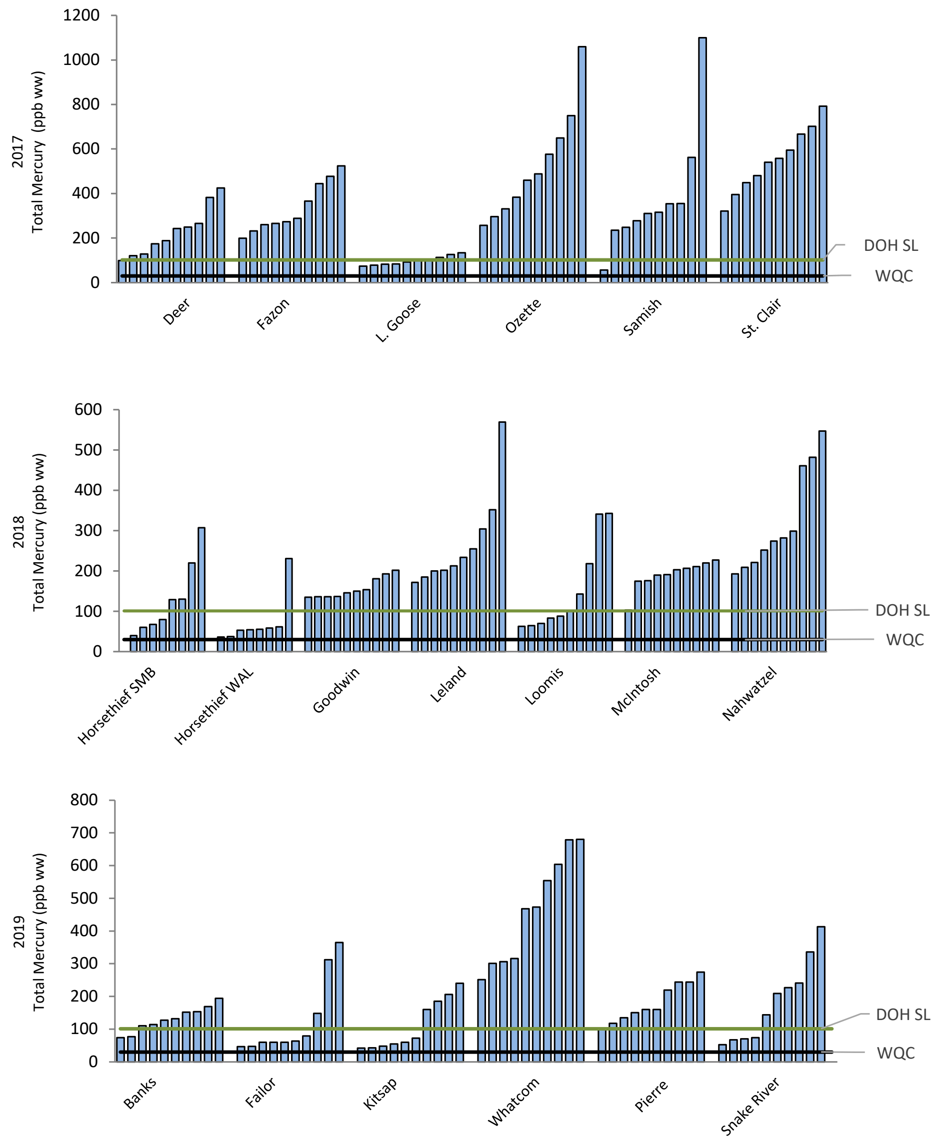

Figure 3. Total mercury concentrations in 2017-2019 individual fish relative to Washington's methylmercury water quality criterion (WQC) and Department of Health screening levels (DOH SL) for human health.

Individual Fish Size and Mercury Relationships

Simple linear regression showed mercury concentrations increased significantly with fish size and age in all lakes except in smallmouth bass at Lake Goodwin and Pierre Lake and largemouth bass at Lake McIntosh. Fish from these lakes showed weak relationships and no statistical significance. Fish from Lake Goodwin were younger on average than at other lakes. Banks Lake smallmouth bass mercury concentrations had a weak relationship with fish length, but a strong relationship with fish weight.

Table 4. Linear regression coefficients used for testing relationships between mercury concentrations and fish size or age, 2017-2019.

Year	Lake	Species	Length	Weight	Age
2017	Deer	LMB	**0.66**	**0.589**	**0.893**
2017	Fazon	LMB	**0.628**	**0.611**	**0.765**
2017	L. Goose	LMB	**0.455**	0.382	**0.563**
2017	Ozette	LMB	**0.79**	**0.783**	**0.788**
2017	Samish	LMB	**0.855**	**0.893**	**0.887**
2017	St Clair	LMB	**0.49**	**0.465**	**0.817**
2018	Goodwin	SMB	0.042	0.032	0.112
2018	Horsethief	SMB	**0.639**	**0.488**	0.824
2018	Horsethief	WAL	**0.738**	**0.706**	**0.882**
2018	Leland	LMB	**0.734**	0.781	**0.477**
2018	Loomis	LMB	**0.621**	0.612	**0.562**
2018	McIntosh	LMB	0.198	0.156	0.207
2018	Nahwatzel	LMB	**0.514**	**0.577**	**0.561**
2019	Banks	SMB	0.372*	**0.703**	**0.442**
2019	Failor	LMB	**0.824**	**0.801**	**0.886**
2019	Kitsap	LMB	**0.906**	**0.91**	**0.942**
2019	Pierre	SMB	0.092	0.093	0.018
2019	Snake	LMB	**0.895**	**0.883**	**0.891**
2019	Whatcom	LMB	**0.436**	**0.412**	0.154

LMB: largemouth bass; SMB: smallmouth bass; WAL: walleye.
Bold values indicate significance at $\alpha = 0.05$.
Asterisk (*) indicates significance at $\alpha = 0.1$.

Ancillary Species Composite Mercury Concentrations

We collected additional species in 2017 for composite sample analysis. Fourteen composite samples composed of five different species were analyzed for mercury. Mercury concentrations in composites ranged from 30.5 ppb to 1,160 ppb. All sample concentrations exceeded the state WQC. Nine samples were above the DOH SL. We observed the highest concentrations in yellow perch and northern pikeminnow from Lake Ozette. The lowest concentrations were found in brown bullhead from Lake Samish.

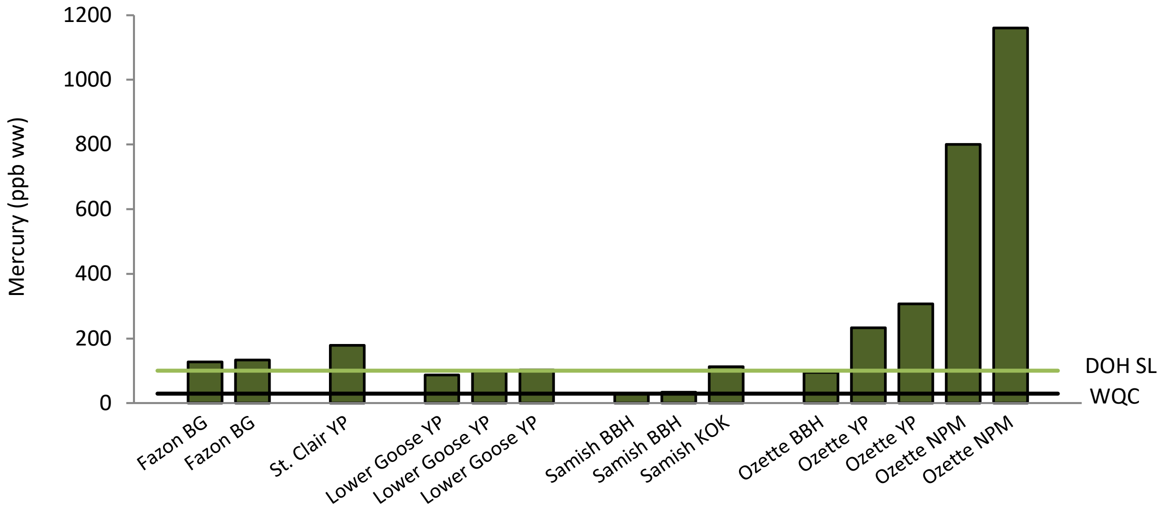

Figure 4. Mercury concentrations in 2017 composite samples of ancillary species relative to Washington's Water Quality Criterion (WQC) and Department of Health screening levels (DOH SL).

Temporal Trends

2017 Mercury Trends

Figure 5 presents mercury and length data of all individual bass samples collected in 2017 and previous years that were used in the trend analysis. We assessed trends for five of six lakes sampled in 2017. Recent trends at Lower Goose Lake could not be identified because of the large difference in size classes between 2017 samples and previously collected data.

We excluded the 2007 Lake Fazon dataset from analysis to meet all assumptions of the ANCOVA. The 2007 fish were much larger and outside of the range of the rest of the dataset. The 2012 and 2017 size classes were similar and allowed us to avoid constraining the data, meet all the assumptions for ANCOVA, and give us more confidence in trend reporting for this period.

We used length as a covariate for all lakes except for Deer Lake. Length was not a significant predictor of mercury in the Deer Lake 2012 dataset. One Deer Lake sample in 2012 had a length outside of the normal length-weight relationship. Fish weight was used as a covariate instead as it displayed significant relationships for all sampling years.

Fish collection efforts at Lake Ozette in 2012 yielded only seven bass. Three bass had been collected and analyzed for mercury in 2011. These samples were included in the analysis as part of the 2012 dataset and deemed representative of the same time frame.

For Samish Lake we include data collected in 2001 (Fischnaller, 2003). The regression formula used to correct for the older mercury analytical method used is limited to fish less than 400 mm, above which the correction loses confidence. As a result of this limitation, three fish greater than 400 mm were removed from the 2001 dataset.

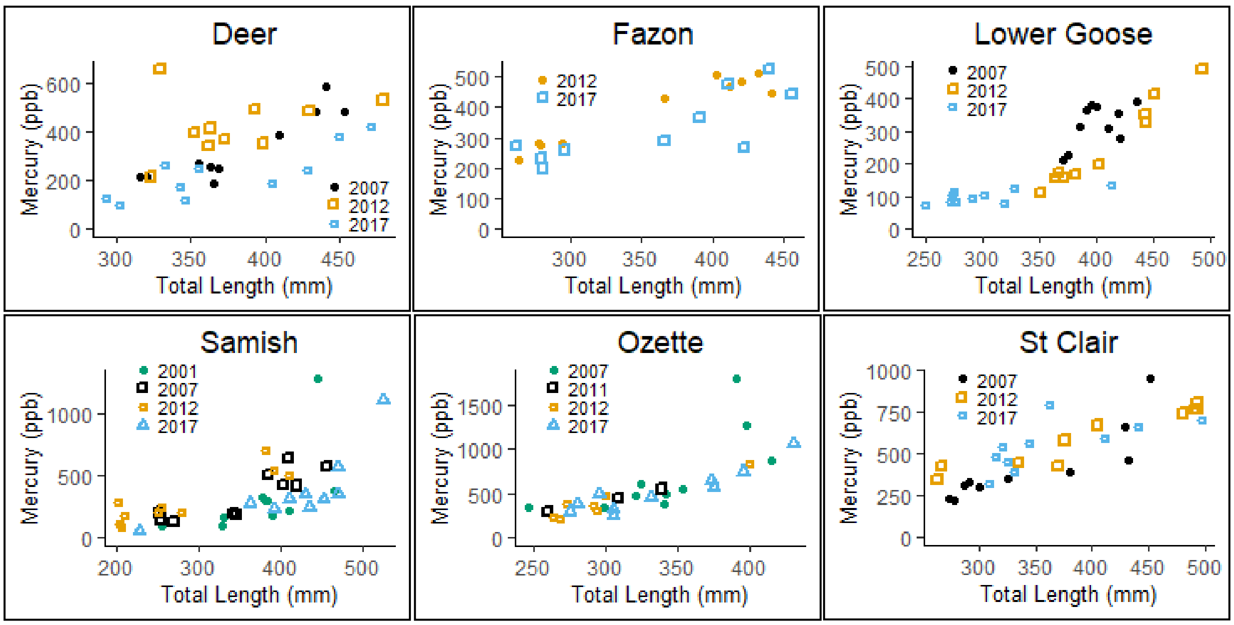

Figure 5. Mercury to fish length relationship of 2017 bass.

Figure 6 presents back-transformed least squares mean mercury concentrations for sites that had a significant difference between sampling years. In 2017, we saw a significant decrease in mercury concentrations at Deer, Fazon, and Samish Lakes. Back-transformed estimated mercury means decreased by 44% in 2017 at Deer Lake compared to 2012 and by 34% compared to 2007. Lake Fazon showed a decrease of 16% in estimated mercury means from 2012 to 2017. Mercury in Samish Lake bass decreased by 56% from 2012 to 2017, reversing a previous increasing trend since 2001.

Mercury concentrations in Lake St Clair bass increased since 2007. Pairwise comparison shows there is no significant difference since the last reporting period in 2012. Mercury Concentrations remain elevated at levels similar to 2012 after increasing significantly from 2007.

No significant change was detected at Ozette Lake. Ozette mercury levels remain relatively high compared to other sites throughout the state.

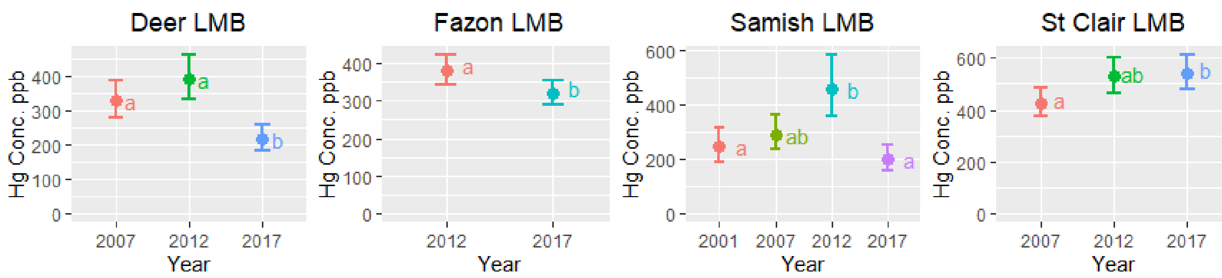

Figure 6. Back-transformed least squares means from 2017 post-hoc tests.
A difference in letters indicates a significant difference between collection years for log_{10} Hg.

2018 Mercury Trends

For the 2018 monitoring sites, Figure 6 presents the mercury and length data of all individual bass included in the trends analysis. We assessed trends using length as a covariate for Leland, Loomis, Nahwatzel, and Horsethief lakes. Size relationships were not a significant predictor at Goodwin and McIntosh Lakes. Instead, we used ANOVA to evaluate trends for Goodwin and McIntosh. Trends for Horsethief Lake were only examined between 2008 and 2018, as the 2013 sample collection did not produce enough data to include for analysis (n = 4). Additionally, two fish were misidentified at Horsethief lake in 2018 leading to reduced sample size in 2018 (n = 8).

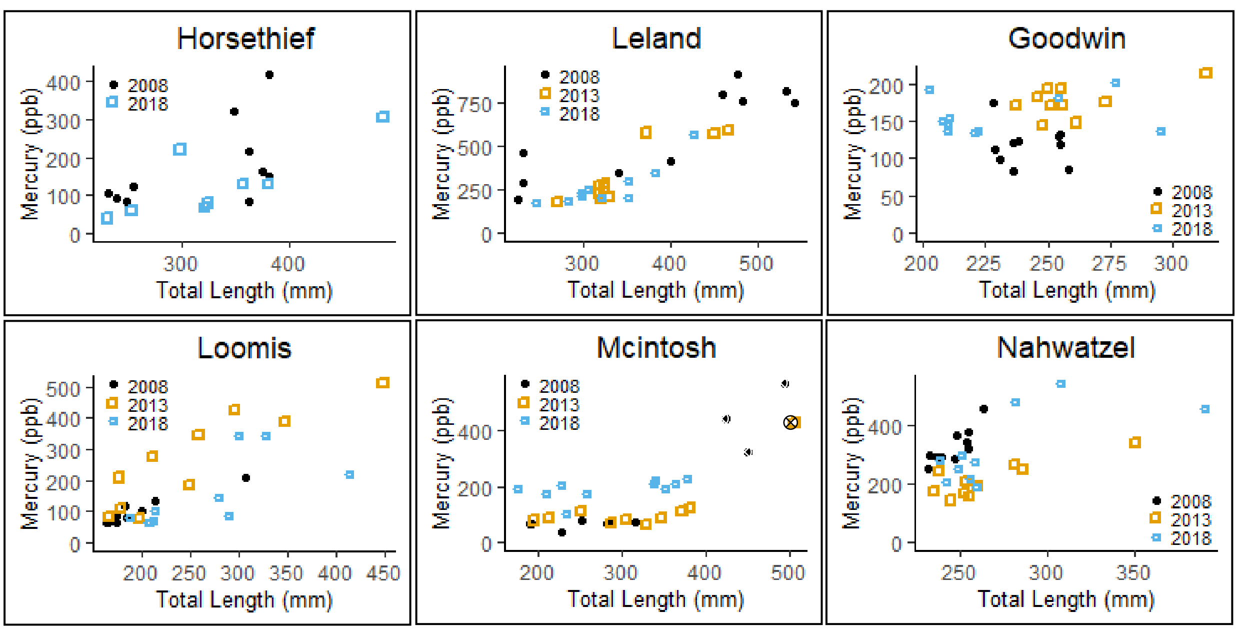

Figure 7. Mercury to fish length relationship of 2018 individual bass.
*Lake McIntosh data constrained to fish size < 400 mm.

ANCOVA results showing a significant difference between sampling years are presented in Figure 8. We saw a significant difference in five out of six lakes.

Lake Goodwin bass were higher in mercury in both 2013 and 2018 compared to 2008. Levels between 2013 and 2018 did not change. Estimated mercury means increased 115% at McIntosh Lake between 2013 to 2018. Data from Lake McIntosh were constrained to ensure that fish size ranges were comparable between years. Fish greater than 400 millimeters were removed. Model assumptions for ANOVA were met after constraining the data. A covariate was not used at McIntosh due to lack of relationship between mercury and size.

Lake Nahwatzel had a 38% increase in mercury concentration in largemouth bass between the 2013 and 2018 collection years. This reverses a previously measured negative trend in mercury levels between the 2008 and 2013 period.

At Loomis Lake, we saw a 49% decrease in mercury concentrations in 2018 compared to 2013. This reverses an increasing trend measured in 2013 from 2008. With the addition of 2018 data, the evidence for significant difference between 2008 and 2013 is now weaker but still meaningful at the $\alpha = 0.1$ level. This change in significance from the last reporting period is likely due to the pooling of variance after adding the 2018 data.

Leland Lake bass collected in 2013 and 2018 contained lower mercury levels than bass collected in 2008. Estimated mercury means in 2018 were 40% lower compared to 2008. In Horsethief Lake, neither smallmouth bass or walleye showed any change in mercury between sampling years.

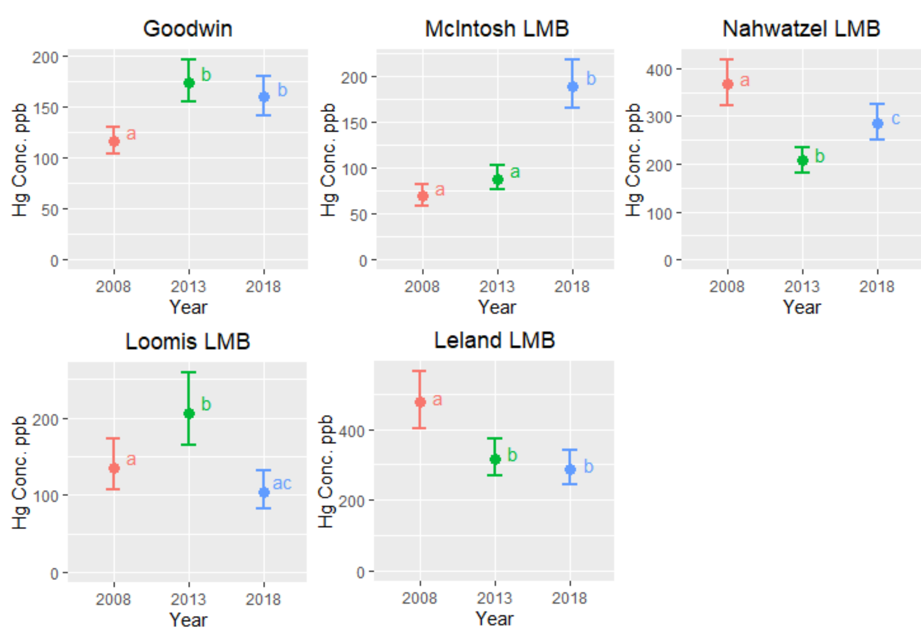

Figure 8. Back-transformed least squares means from 2018 post-hoc tests.

A difference in letters indicates a significant different between collection years for $\log_{10}$ Hg. Difference between 2008 – 2013 at Loomis Lake is significant at alpha = 0.10.

2019 Mercury Trends

Figure 9 presents the mercury concentrations and lengths of all individual bass samples used in trends analyses for the 2019 sampling year. We assessed trends using ANCOVA at five of the sites for the 2019 reporting period. We used weight as a covariate at Banks Lake instead of length. At Pierre Lake we used ANOVA to assess trends because no relationship between mercury and size was observed. Data collected in 2000 by Serdar (2001) was available for Lake Whatcom and included in the analysis.

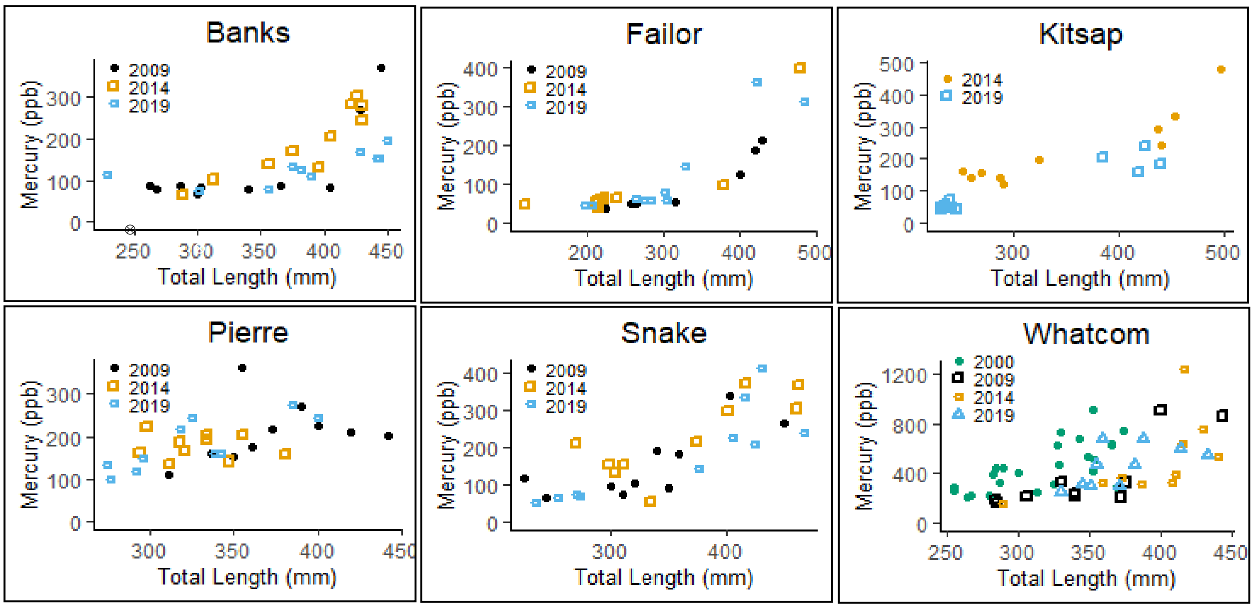

Figure 9. Mercury to fish length relationship of 2019 individual bass.

Figure 10 presents back-transformed least squares mercury means for sites where we found significant differences between sampling years. We detected significant trends at four of six lakes. Three of these lakes exhibited new trends in the 2019 reporting period based on post-hoc analysis.

Mercury concentrations decreased by 33% at Banks Lake in 2019 compared to 2014. This reversed an insignificant trend upward from previous observations. Kitsap Lake mercury concentrations decreased by 44% from 2014 to 2019. This is the first measured trend because 2002 data (Fischnaller, 2003) were not considered comparable based on fish size differences and Kitsap Lake was not sampled in 2009.

Bass mercury concentrations collected in 2000 from Lake Whatcom were significantly higher than from all sampling conducted thereafter. Fish collected in 2009, 2014, and 2019 contained less than one-half the mercury recorded in 2000.

Bass mercury concentrations did not show significant trends at Pierre Lake, Snake River, and Failor Lake. Previous analyses showed an increase in mercury from 2009 to 2014 at Failor Lake. The previous trend is still detectable, although significance is reduced to $\alpha = 0.1$.

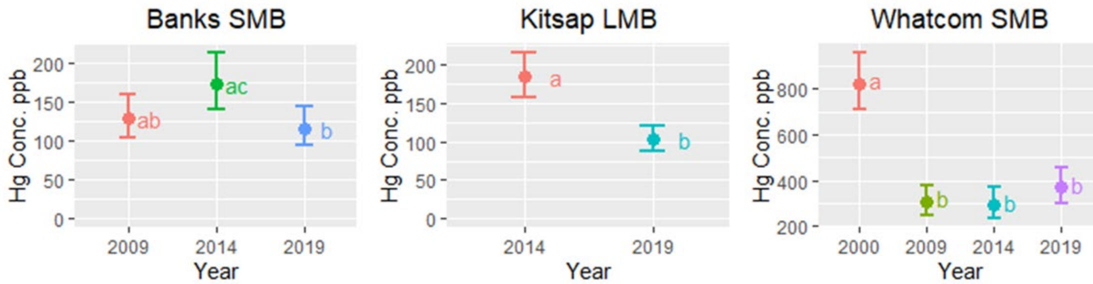

Figure 10. Back-transformed least squares means from 2019 post-hoc tests.

A difference in letters indicates a significant different between collection years for $\log_{10}$ Hg.

Statewide Trends

The Mercury Trends long-term monitoring study completed a third round of sampling at all sites during 2015-2019. Mercury in the fillets of ten largemouth or smallmouth bass has been analyzed from the sites listed in Table 5 on a five-year rotation since 2005. Table 5 displays the trend for each site as determined by ANCOVA results and Figures 11 and 12 display the direction of trends for two time periods mapped across the state. Full statistical results are included in previously published reports (Mathieu and McCall, 2017; Mathieu, 2019) and earlier sections of this report.

The first round of sampling at each of the sites occurred during 2005-2009, the second during 2010-2014, and the third during 2015-2019. Statistical tests show:

- No change in bass mercury levels between the most recent sampling event and the two previous sampling rounds for most sites (65% and 63% of sites for the five-year and ten-year periods, respectively).
- Mercury decreased at eight sites (30%) over a five-year period and decreased at five sites (19%) over a ten-year period.
- Few sites saw increases in mercury; with levels increasing over a five-year period at 7% of the sites and at 15% of the sites over a ten-year period.

Monitoring programs in other areas of the United States have found similar stability in fish mercury levels over the past two decades. Long-term monitoring of fish from the Great Lakes in New York state showed that most of the change in fish mercury levels occurred as declines from the 1970s through 1990 following major pollution control policies, and then the decline slowed or stopped, and inconsistent trends emerged (Richter and Skinner 2020). Largemouth bass collected from the Everglades had decreases in mercury across the 1990s until 2000, and stable trends since then (Lange et al. 2020). A review of fish monitoring programs in North America by Grieb et al. (2019) found overall decreasing trends reported during 1972–2016, with a leveling off in recent years.

Small changes in bass mercury levels may not be captured by this study's sample design. The ability to detect trends was estimated for several scenarios when developing the Mercury Trends study design, and the decision to collect 10 individual fish per site was based on several considerations: trend power, sampling difficulty, and analytical cost. It was estimated that a mean (average of decrease and increase) minimum detectable change using 10 individual fish with adjustment for fish length was 31% (Seiders 2006). Consequently, there may be more subtle changes in the waterbodies that we are not observing. The sensitivity for detecting trends should increase with additional sampling events over time.

As mercury loading from bulk atmospheric deposition has remained relatively unchanged over the last 15 years in Washington state (Weiss-Penzias et al. 2016), mixed trends in fish mercury concentrations appear to be related to watershed and in-lake processes specific to the waterbody. Suggested drivers of divergent trends in fish mercury levels within a region include food web changes (Millard et al. 2020; Taylor et al. 2019), weather events such as flooding (Swinton and Nierzwicki-Bauer 2020), and climate-induced variability (Wang et al. 2019). Many North American lake studies have pointed to climate change as affecting fish tissue mercury trends through increases in waterbody temperatures, increased rainfall transporting more organic matter to waterbodies, and fluctuating water levels (i.e., the reservoir effect) (Grieb et al. 2019).

Table 5. Statewide trends observed in bass mercury levels, 2005-2019.

Date of First Sampling	Date of Second Sampling	Date of Third Sampling	Waterbody	Species	Hg_{bass} 10 Year Trend*	Hg_{bass} 5 Year Trend**	Co-variate
2005	2010	2015	Liberty Lake	SMB	=	=	L
2005	2010	2015	Loon Lake	LMB	=	=	L
2005	2010	2015	Potholes Res.	SMB	=	=	age
2005	2010	2015	Silver Lake	LMB	↑ (73%)	=	L
2005	2010	2015	Lake Spokane	SMB	=	=	none
2005	2010	2015	Yakima River	SMB	=	=	none
2006	2011	2016	Meridian Lake	LMB	=	=	none
2006	2011	2016	Moses Lake	SMB	=	=	L
2006	2011	2016	Newman Lake	LMB	=	=	L
2006	2011	2016	Offutt Lake	LMB	↓ (-50%)	↓ (-42%)	none
2006	2011	2016	Lake Sammamish	LMB	↓ (-37%)	↓ (-53%)	L
2007	2012	2017	Deer Lake	LMB	↓ (-34%)	↓ (-44%)	W
n/a	2012	2017	Lake Fazon	LMB	n/a	↓ (-16%)	L
2007	2012	2017	Lake Ozette	LMB	=	=	L
2007	2012	2017	Lake Samish	LMB	=	↓ (-56%)	L
2007	2012	2017	Lake St. Clair	LMB	↑ (27%)	=	L
2008	2013	2018	Lake Goodwin	SMB	↑ (37%)	=	none
2008	2013	2018	Leland Lake	LMB	↓ (-39%)	=	L
2008	2013	2018	Loomis Lake	LMB	=	↓ (-49%)	L
2008	2013	2018	McIntosh Lake	LMB	↑ (174%)	↑ (115%)	none
2008	2013	2018	Lake Nahwatzel	LMB	↓ (-22%)	↑ (38%)	L
2008	---	2018	Horsethief Lake	SMB	=	n/a	L
2009	2014	2019	Banks Lake	SMB	=	↓ (-33%)	W
2009	2014	2019	Failor Lake	LMB	=	=	L
n/a	2014	2019	Kitsap Lake	LMB	n/a	↓ (-44%)	L
2009	2014	2019	Pierre Lake	SMB	=	=	none
2009	2014	2019	Snake River	LMB	=	=	L
2009	2014	2019	Lake Whatcom	SMB	=	=	L

SMB = smallmouth bass; LMB = largemouth bass; L = length; W = weight; n/a = not available

* Change in mercury between first sampling visit and third sampling visit.

** Change in mercury between second sampling visit and third sampling visit.

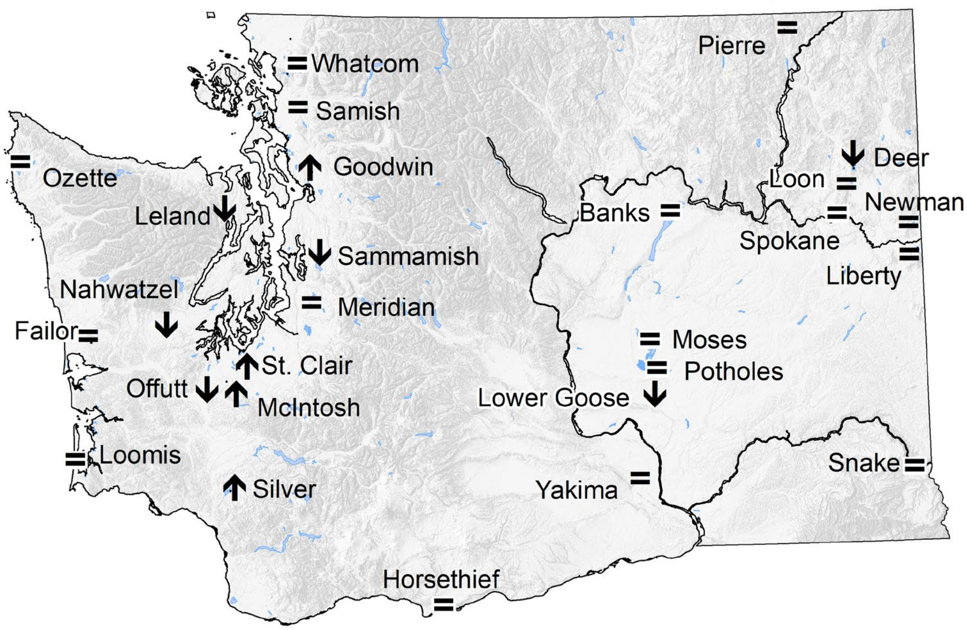

Figure 11. Temporal trends of bass mercury concentrations over a ten-year period (first vs. third visit).

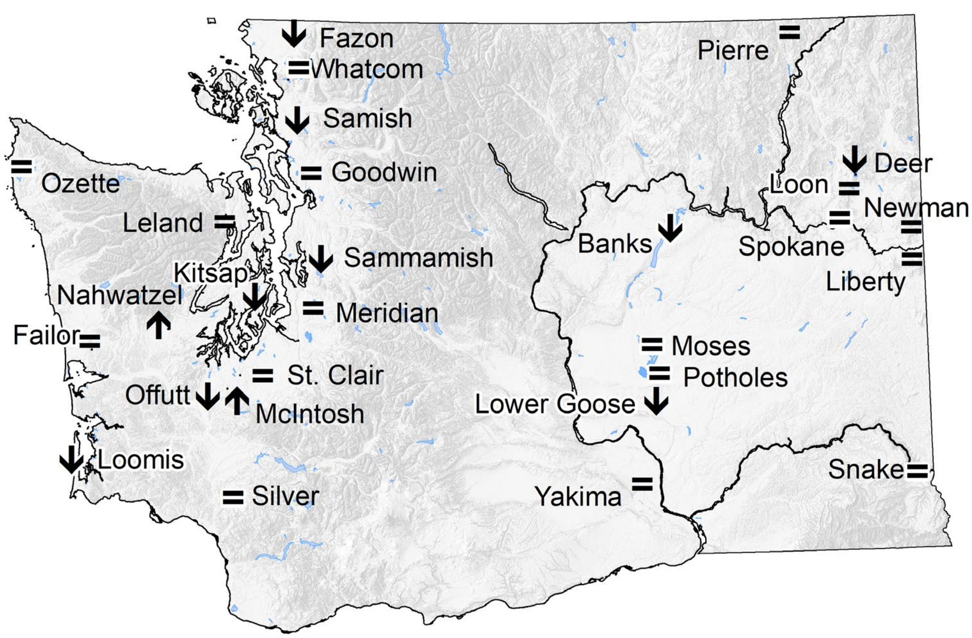

Figure 12. Temporal trends of bass mercury concentrations over a five-year period (second vs. third visit).

Trends in Spatial Pattern

Over the course of the three sampling periods, the general spatial pattern of mercury contamination in bass across Washington state has remained constant. Figure 13 displays estimated mercury concentrations for a 350mm bass calculated using $\log_{10}$ mercury:length linear regressions for each waterbody dataset. In general, standard-length (350mm) mercury concentrations have remained elevated in western Washington compared to eastern, central, and southwestern areas of the state.

After the first round of sampling (2005-2009) at all study locations, we found that mercury bioaccumulation closely followed rainfall patterns in the state, with sites receiving the most rain containing the highest levels of mercury in bass (Mathieu et al. 2013). Most of the spatial variance in the statewide dataset was explained by annual watershed precipitation (increase in bass mercury) and lake alkalinity (decrease in bass mercury). This pattern remained consistent throughout all three study periods, with the wet forested sites containing the highest bass mercury concentrations and the alkaline lakes in agricultural or arid open spaces containing the least.

Fish mercury levels have been tied to land cover in other areas of the U.S., as well. In southeastern states, the variance in largemouth bass mercury concentrations was recently explained by the coverage of evergreen forests, emergent herbaceous wetlands, and pasture/hay (Drenner et al. 2022). A synthesis of northeastern states concluded that forest regions are especially sensitive to mercury inputs, resulting in higher concentrations of fish mercury levels due to the canopy acting as a filter for atmospheric mercury, wetland abundance in forests, and the transport of mercury through organic carbon (Driscoll et al. 2007). Forest or canopy densities have also been correlated with increased fish mercury levels in Pacific Northwest subalpine lakes (Eagles-Smith et al. 2016).

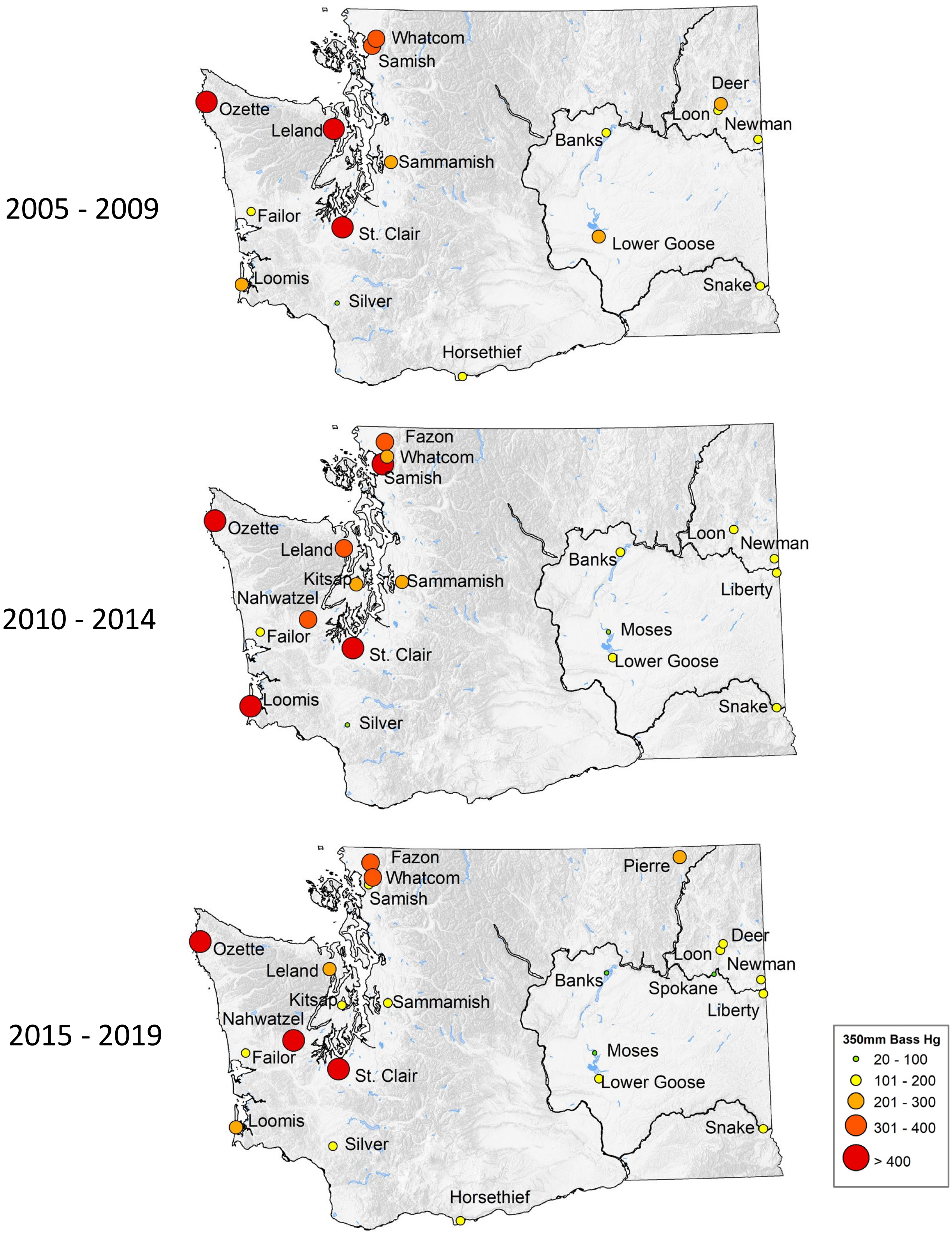

Figure 13. Standard-length (350mm) bass mercury concentrations estimated for the first sampling visit (2005-2009), second visit (2010-2014), and third visit (2015-2019).

Conclusions

This report summarizes the 2017, 2018, and 2019 sampling results of a long-term monitoring study to characterize mercury in freshwater fish and mercury trends over time. Results were compared to previous sampling years at the waterbodies.

- During 2017-2019, mercury was detected in all individual fish (largemouth bass, smallmouth bass, and walleye) with concentrations ranging from 35 – 1,100 ppb ww. The highest mercury concentration in an individual sample came from a 13-year-old largemouth bass from Lake Samish (1,100 ppb), and Lake St. Clair had the highest mercury concentrations over a lake dataset (median = 549 ppb).
- All individual samples exceeded the state water quality criterion (WQC) of 30 ppb, and 75% exceeded the DOH SL of 101 ppb. All samples collected from the following lakes exceeded the DOH SL: Fazon, Goodwin, Whatcom, Leland, McIntosh, Nahwatzel, Ozette, and St. Clair.
- In 2017, significant decreases in bass mercury levels were found at Deer, Fazon, and Samish Lakes compared to 2012. Deer Lake levels were also lower in 2017 compared to 2007. Lake St. Clair was the only lake with an increase, with 2017 mercury levels higher than detected in 2007.
- In 2018, bass from Leland and Loomis Lakes had lower mercury levels than in previous collection years, and Goodwin and McIntosh had higher levels than in previous sampling years. Bass mercury levels from Lake Nahwatzel were lower than in bass collected in 2008, but higher than in bass collected in 2013.
- In 2019, bass mercury levels were lower in Banks and Kitsap Lakes compared to 2014. Lake Whatcom had high concentrations in 2000 and then decreased to consistently lower concentrations in 2009, 2014, and 2019.

As of 2019, all 30 sites in the monitoring study have been sampled three times, five years apart. The first round of sampling occurred during 2005-2009, the second round during 2010-2014, and the third round during 2015-2019. Statewide trends over a five-year period and a ten-year period were summarized in this report, with the following conclusions:

- Most sites showed no change in bass mercury levels over both five-year and ten-year periods (65% and 63%, respectively).
- Mercury decreased at eight of the sites (30%) over a five-year period and at five of the sites (19%) over a ten-year period.
- Few sites showed increases in mercury. Bass mercury levels increased over a five-year period at 7% of sites and at 15% of sites over a ten-year period.
- The general spatial pattern of mercury accumulation in bass across Washington state has remained consistent throughout the 15 years of monitoring. The highest concentrations of mercury are detected in bass collected from lakes on the coastal west side of the state and lowest in the drier central and eastern parts of the state.

Recommendations

Results of this 2017-2019 study support the following recommendations.

- Data presented in this report should be reviewed by (1) the Washington State Department of Health (DOH) when making or updating fish consumption advisories and (2) the Department of Ecology (Ecology) when assessing waterbodies for the next Water Quality Assessment cycle.
- This report marks the second time we have summarized statewide trends for the Mercury Trends long-term monitoring study. We recommend the monitoring project move towards written reports every five years detailing trends after all waterbodies have been re-visited. The next report would be written after the 2024 sampling year. Annual data should be publicly available in Ecology's online database, and trends will be reported annually through visual online mapping software, following an interval review process.
- Ecology's PBT Monitoring Program should consider adding other PBTs as target analytes when analyzing fish collected for this study. This would leverage the collection efforts involved with the Mercury Trends study and could involve special studies (for example, to gain more information on the occurrence of PBTs where state data are still lacking, such as per- and polyfluoroalkyl substances, hexabromocyclododecane, and flame retardants).

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Glossary, Acronyms, and Abbreviations

Glossary

Anthropogenic: Human-caused.

Methylmercury: A highly toxic and bioavailable form of mercury.

Nonpoint source: Pollution that enters any waters of the state from any dispersed land-based or water-based activities, including but not limited to atmospheric deposition, surface-water runoff from agricultural lands, urban areas, or forest lands, subsurface or underground sources, or discharges from boats or marine vessels not otherwise regulated under the NPDES program. Generally, any unconfined and diffuse source of contamination. Legally, any source of water pollution that does not meet the legal definition of "point source" in section 502(14) of the Clean Water Act.

Parameter: Water quality constituent being measured (analyte). A physical, chemical, or biological property whose values determine environmental characteristics or behavior.

Point source: Sources of pollution that discharge at a specific location from pipes, outfalls, and conveyance channels to a surface water. Examples of point source discharges include municipal wastewater treatment plants, municipal stormwater systems, industrial waste treatment facilities, and construction sites where more than 5 acres of land have been cleared.

Stormwater: The portion of precipitation that does not naturally percolate into the ground or evaporate but instead runs off roads, pavement, and roofs during rainfall or snow melt. Stormwater can also come from hard or saturated grass surfaces such as lawns, pastures, playfields, and from gravel roads and parking lots.

Watershed: A drainage area or basin in which all land and water areas drain or flow toward a central collector, such as a stream, river, or lake at a lower elevation.

303(d) list: Section 303(d) of the federal Clean Water Act requires Washington State to periodically prepare a list of all surface waters in the state for which beneficial uses of the water – such as for drinking, recreation, aquatic habitat, and industrial use – are impaired by pollutants. These are water quality limited estuaries, lakes, and streams that fall short of state surface water quality standards and are not expected to improve within the next two years.

Acronyms and Abbreviations

DOH	Washington State Department of Health
Ecology	Washington State Department of Ecology
EIM	Environmental Information Management database
EPA	U.S. Environmental Protection Agency
Hg	mercury
MEL	Manchester Environmental Laboratory
PBT	persistent, bioaccumulative, and toxic substance
QAPP	Quality Assurance Project Plan
SL	Screening Level
SOP	standard operating procedure

SRM	standard reference materials
WDFW	Washington Department of Fish and Wildlife
WQC	Water Quality Criterion

Units of Measurement

ft	feet
in	inch
ppb	parts per billion
sq mi	square mile
µg/kg	micrograms per kilogram (parts per billion)
ww	wet weight

Appendices

Appendix A. Numerical Thresholds for the Protection of Human Health from Mercury in Fish Tissue

To provide context for the mercury results in this 2017-2019 study, data are compared to two methylmercury thresholds:

- Washington State's Water Quality Criterion (WQC) for human health (40 CFR 131.45) that went into effect in 2016.
- Washington State Department of Health's (DOH's) screening level (SL) for fish consumption advisories.

Both thresholds are based on the toxicological effects of methylmercury, the bioaccumulative and toxic form of mercury in fish tissue, while values in this report reflect total mercury. EPA guidance for fish contaminant monitoring programs recommends analyzing mercury as a surrogate for methylmercury as a conservative approach to be most protective of human health (EPA, 2000).

Washington State's methylmercury WQC of 30 ppb (tissue) is a human health criterion based on a fish consumption rate of 175 g/day over a 70-year lifespan. This rate is representative of the average consumption of all fish and shellfish (including salmon and fish/shellfish eaten at restaurants, locally caught, imported, or obtained from other sources) for highly exposed populations that consume both fish and shellfish from Puget Sound waters. Washington state assesses waterbodies for impairment using all data collected from a waterbody over the period of time that the assessment cycle is addressing, using median concentrations of fish tissue composite samples (Ecology 2018).

The DOH SL is a threshold DOH toxicologists use when developing fish consumption advisories, in addition to other factors. The DOH SL of 101 ppb (tissue) is based on a general population consumption rate of 59.7 g/day, which the American Heart Association recommends for a healthy diet (two 8 oz fish meals per week). DOH uses the SL to provide advice to fish consumers in Washington, while the WQC is used to set National Pollutant Discharge Elimination System (NPDES) permit limits and assess waters and represents full protection of the designated use of harvest.

Data exceeding these thresholds do not necessarily represent an impaired use or a fish consumption advisory. State agencies use fish tissue mercury data, including data provided in this report, as part of an overall assessment of a waterbody, using an approach to address average exposures over a period of time.

Appendix B. ANCOVA Results for 2017-2019 Sampling Years

Table B-1. 2017 ANCOVA Results Comparing Mercury Levels in Bass.

Waterbody	Species	Co-variate	Sum of Squares	df	Mean Squares	F-ratio	p-value	2001 Hg_{bass}	2007 Hg_{bass}	2012 Hg_{bass}	2017 Hg_{bass}
Deer	LMB	W	0.338	2	0.169	14.079	**< 0.001**	--	330^{a}	394^{a}	219^{b}
Fazon	LMB	L	0.029	1	0.029	6.16	**0.024**	--	--	381^{a}	320^{b}
L. Goose	LMB	--	--	--	--	--	--	--	--	--	--
Ozette	LMB	L	0.036	2	0.018	1.387	0.268	--	--	--	--
Samish	LMB	L	0.461	3	0.154	7.284	**<0.001**	247^{a}	293^{ab}	459^{b}	201^{a}
St. Clair	LMB	L	0.06	2	0.03	4.335	**0.024**	--	428^{a}	528^{b}	542^{b}

Table B-2. 2018 ANCOVA Results Comparing Mercury Levels in Bass and Walleye.

Waterbody	Species	Co-variate	Sum of Squares	df	Mean Squares	F-Ratio	p-Value	2008 Hg_{bass}	2013 Hg_{bass}	2018 Hg_{bass}
Goodwin	SMB	--	0.176	2	0.088	14.66	**< 0.001**	117^{a}	175^{b}	160^{b}
Leland	LMB	L	0.255	2	0.127	10.404	**< 0.001**	476^{a}	317^{b}	288^{b}
Loomis	LMB	L	0.445	2	0.222	9.895	**< 0.001**	136^{a}	207^{b}	105^{ac}
McIntosh	LMB	--	0.969	2	0.485	50.663	**< 0.001**	69^{a}	88^{a}	189^{b}
Nahwatzel	LMB	L	0.294	2	0.147	20.824	**< 0.001**	367^{a}	207^{b}	285^{c}
Horsethief	SMB	L	0.155	1	0.155	4.033	0.063	--	--	--
Horsethief	WAL	L	0.009	1	0.009	0.717	0.412	--	--	--

Table B-3. 2019 ANCOVA Results Comparing Mercury Levels in Bass.

Waterbody	Species	Co-variate	Sum of Squares	df	Mean Squares	F-value	p-value	2000 Hg_{bass}	2009 Hg_{bass}	2014 Hg_{bass}	2019 Hg_{bass}
Banks	SMB	W	0.162	2	0.081	4.097	**0.028**	--	129^{ab}	174^{ac}	117^{b}
Failor	LMB	L	0.142	2	0.071	3.162	**0.059**	--	66^{a}	98^{b}	81^{b}
Kitsap	LMB	L	0.297	1	0.297	29.60	**< 0.001**	--	--	184^{a}	103^{b}
Pierre	SMB	--	0.021	2	0.011	0.707	0.507	--	--	--	--
Snake	LMB	L	0.059	2	0.029	1.042	0.366	--	--	--	--
Whatcom	SMB	L	1.403	3	0.468	22.64	**< 0.001**	823^{a}	308^{b}	294^{b}	369^{b}

Notes for above tables:

Bolded values indicate statistical significance at $p < 0.05$.

df = degrees of freedom; SMB = smallmouth bass; LMB = largemouth bass; WAL = walleye; L = length

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,c) indicates statistically significant difference in estimated least squares means for those collection dates.