

Measuring Mercury Trends in Freshwater Fish, 2013 Sampling Results



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2013 Highlighted Findings

Statistical tests showed the following trends in mercury levels of bass between 2008 and 2013:

- Levels decreased in Leland (34%) and Nahwatzel (44%) Lakes.
- Levels increased in Goodwin (49%) and Loomis (55%) Lakes.
- No change was found in McIntosh Lake.
- Trends could not be assessed for Horsethief Lake.

Why Does it Matter?

Mercury is highly toxic and bioaccumulative. Levels increase up aquatic food chains, resulting in concentrations among top predator fish that can be harmful to them, fish-eating wildlife, and humans. As a neurotoxin, mercury can harm the brain and nervous system.

Overview

The Washington State Department of Ecology (Ecology) began a monitoring program in 2005 with the primary goal of assessing temporal trends in mercury levels in freshwater fish throughout the state. Each year, Ecology collects ten individual largemouth or smallmouth bass from six lakes for analysis of total mercury. Ecology returns to each set of lakes every five years to assess trends and determine if mercury levels are changing. Additional species are also collected and analyzed as composites for a secondary goal of supporting fish consumption advisories.

In 2013, Ecology analyzed fish fillets from Lake Goodwin, Horsethief Lake, Leland Lake, Loomis Lake, McIntosh Lake, and Lake Nahwatzel for mercury (Figure 1). Ecology previously tested these lakes in 2008.

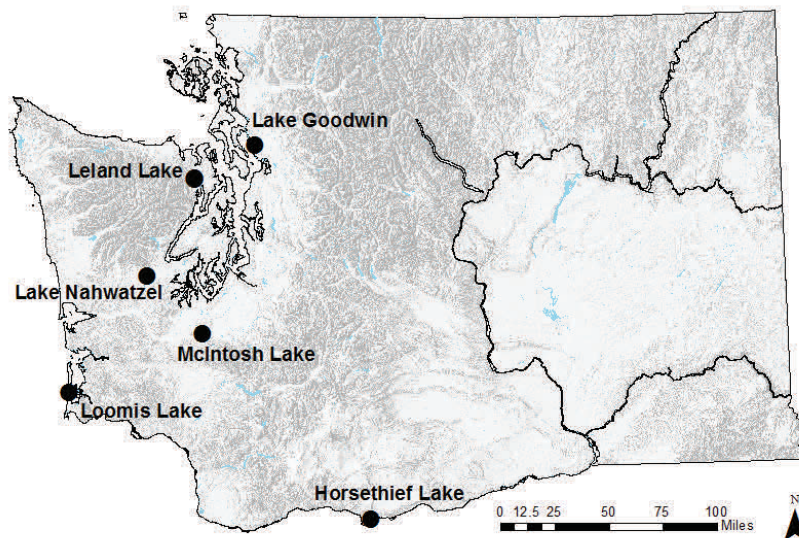


Figure 1. Locations of 2013 Sampling Sites.

This monitoring program supports Ecology's efforts to reduce mercury releases and track trends in Washington. Ecology and the Washington State Department of Health (DOH) developed a Chemical Action Plan (CAP) for mercury in 2003 to address the threat of mercury in Washington and make recommendations for statewide actions to reduce mercury releases (Peele, 2003).

For More Information

PBT Monitoring Program website: www.ecy.wa.gov/programs/eap/toxics/pbt.html

Chemical Action Plan website: www.ecy.wa.gov/programs/swfa/pbt/caps.html

Methods

In the fall of 2013, Ecology collected a total of 54 individual bass from six waterbodies. Bass were analyzed individually to achieve statistical power in detecting trends. Ninety-four fish of additional species were also collected, to form 22 composite samples. Composites of additional species help inform DOH’s fish consumption advisories. Fish were collected and processed following Ecology standard operating procedures (Sandvik, 2014a and 2014b). Edible muscle tissue from fish were analyzed individually or as three to five fish composite samples for total mercury, using cold vapor atomic absorption (EPA Method 245.6). All mercury concentrations in this report are expressed on a ug/kg (ppb) wet weight basis. Washington State Department of Fish and Wildlife (WDFW) biologists determined ages of the bass.

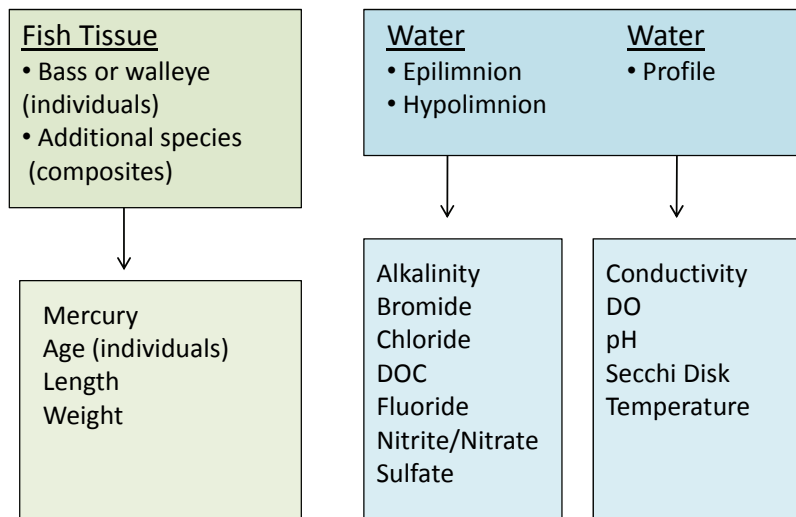


Figure 2. Analytes and Measurements Recorded at each Waterbody. DOC: dissolved organic carbon; DO: dissolved oxygen.

intention of collecting walleye from the waterbody in future years for trend tests. The QAPP specifies that walleye may be collected as a secondary target species at sites where bass are limited (Seiders, 2006).

All quality control (QC) tests for fish tissue and water chemistry laboratory data were within measurement quality objectives outlined in the QAPP (Seiders, 2006) and data were deemed usable without qualification. Detailed quality assurance data and laboratory case narratives are available upon request.

To better understand patterns, dynamics, and changes in mercury accumulation in fish, Ecology also collects water chemistry and in-situ measurements from each site. Ecology measured the water chemistry parameters listed in Figure 2 during the summer of 2013. More detailed information on the study design of this project can be found in the Quality Assurance Project Plan (QAPP)(Seiders, 2006) and QAPP Addendum (Meredith and Furl, 2010).

A summary of sampling conducted and collection goals met in 2013 is provided in Table 1. All collection goals were met with the exception of bass at Horsethief Lake.

Fish collection efforts at Horsethief Lake yielded only 4 smallmouth bass in 2013. Due to the lack of bass observed in the lake, the field crew obtained 10 walleye. The 10 walleye were analyzed for mercury as individuals with the

Table 1. Summary of Sampling Conducted in 2013.

Collection Goal	Horsethief	Goodwin	Leland	Loomis	McIntosh	Nahwatzel
10 individual bass	*	+	+	+	+	+
Additional species collected	WAL, YP	YP	BC, YP	YP	BG, YP	RBT
2 water samples	+	+	+	+	+	+
Hydrolab profile	+	+	+	+	+	+

“+”: goal achieved. *4 individual smallmouth bass collected from Horsethief, 10 walleye collected and analyzed as individuals. WAL: walleye; YP: yellow perch; BC: black crappie; BG: blue gill; RBT: rainbow trout.

Waterbody Descriptions

Ecology previously sampled the six waterbodies in 2008 (Furl et al., 2009). Lake selection in 2008 considered the following criteria: popularity among anglers, availability of target fish species, and inclusion in a 2001-2002 statewide bass survey (Fischnaller et al., 2003). Location and physical data for the 2013 study lakes is included in the table below and in Figure 1.

Table 2. Location and Physical Data for 2013 Study Lakes.

Lake	Goodwin	Horsethief	Leland	Loomis	McIntosh	Nahwatzel
County	Snohomish	Klickitat	Jefferson	Pacific	Thurston	Mason
Drainage Basin (ac)	3310	----	3650	922	1450	3970
Elevation (ft)	324	160	190	17	336	440
Surface Area (ac)	560	92	110	170	93	270
Lake Volume (ac-ft)	13,000	----	1400	830	700	4600
Maximum Depth (ft)	50	----	20	9	11	25
Mean Depth (ft)	23	----	13	5	8	17
Dominant Land Type	residential/ forest	barren	forest	wetland/ forest	forest/ residential	forest

Water Sample Results

Water temperature profiles indicated thermal stratification in all of the lakes, with the exception of McIntosh. Profiles also showed a drop in dissolved oxygen near the bottom at all sites except Nahwatzel. Dissolved oxygen and pH increased with depth in Lake Nahwatzel.

Epilimnion and hypolimnion water samples showed the highest levels of alkalinity and sulfate in Horsethief Lake.

Nahwatzel had the lowest alkalinity, and both Nahwatzel and Loomis were low in sulfate. The greatest concentrations of chloride and DOC were seen in Loomis Lake.

Fluoride and nitrite/nitrate results were low or not detected.

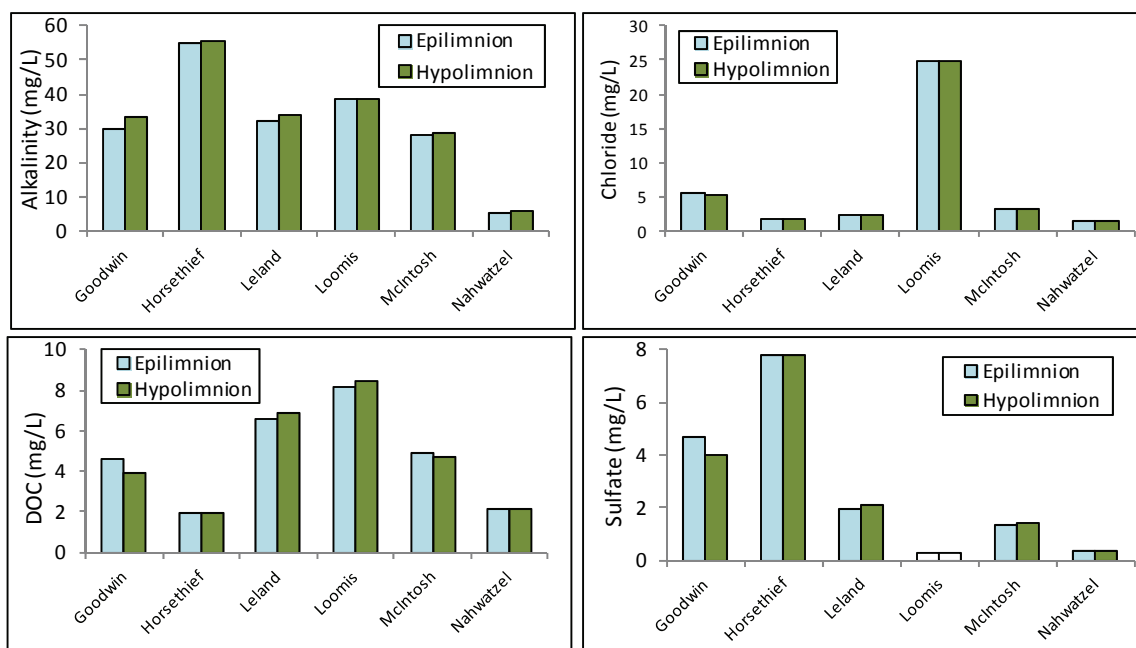


Figure 3. Alkalinity, Chloride, DOC, and Sulfate Concentrations in Water Samples.
Unfilled bars (Loomis Lake sulfate) indicate parameter was not detected at that level.

2013 Fish Tissue Results

Individual Largemouth and Smallmouth Bass

Results of 2013 fish tissue and water chemistry data are available for download at www.ecy.wa.gov/eim by searching Study ID: HgFish13. Summary statistics of bass lengths, weights, ages, and mercury concentrations are shown in Table 3. Mercury concentrations of individual smallmouth and largemouth bass are shown in Figure 4.

Fillet tissue samples from a total of 54 individual bass were analyzed for mercury in 2013. Mercury concentrations ranged from 65 - 592 ppb. The highest mercury levels were measured in Leland Lake followed by Loomis Lake. Seventeen percent (9 out of 54) of the samples exceeded the EPA’s recommended criterion for human health (300 ppb). These samples were collected at Leland, Loomis, McIntosh, and Nahwatzel Lakes. Fish from these lakes ranged in age from four to ten years old. No samples contained mercury levels higher than the Washington State regulatory criterion Fish Tissue Equivalent Concentration (FTEC) of 770 ppb. The FTEC is the threshold used by Washington State to determine whether a waterbody is meeting water quality standards for mercury. DOH uses a different set of criteria when determining fish consumption advisories. For a detailed explanation of how Ecology and DOH evaluate fish tissue mercury data, see Meredith and Friese (2012) Appendix F.

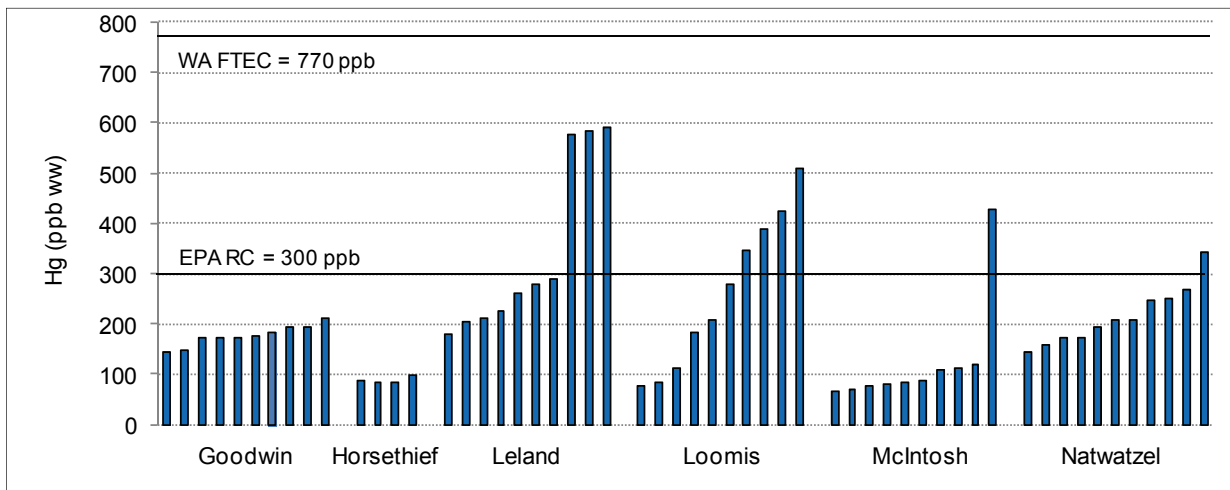
Table 3. Summary Statistics of Bass Lengths, Weights, Ages, and Mercury Concentrations.

Waterbody	Species	n	Length (TL mm)		Weight (g)		Age (yr)		Hg (ppb ww)	
			Range	Mean (±SD)	Range	Mean (±SD)	Range	Mean (±SD)	Range	Mean (±SD)
Goodwin	SMB	10	237 - 313	259 (21)	165 - 410	236 (70)	2 - 3	2.1 (0.3)	145 - 213	176 (21)
Horsethief	SMB	4	282 - 288	285 (3)	259 - 290	279 (14)	4 - 4	4 (0)	83.8 - 98.8	88.6 (6.9)
Leland	LMB	10	271 - 467	350 (62)	253 - 1789	765 (528)	2 - 6	3.6 (1.3)	181 - 592	340 (172)
Loomis	LMB	10	167 - 448	253 (90)	53 - 1434	318 (425)	2 - 7	3.8 (1.5)	77 - 509	261 (152)
McIntosh	LMB	10	195 - 506	318 (91)	97 - 2639	635 (740)	1 - 10	3.4 (2.5)	64.6 - 429	124 (109)
Nahwatzel	LMB	10	235 - 350	265 (33)	163 - 635	263 (142)	4 - 5	4.2 (0.4)	145 - 344	216 (61)
All Sites	---	---	167 - 506	289 (71)	53 - 2639	431 (470)	1-10	3 (1.5)	64.6 - 592	214 (134)

LMB: largemouth bass; SMB: smallmouth bass; SD: standard deviation; TL: total length.

Figure 4. Mercury Concentrations in Individual Fish Collected in 2013.

WA FTEC: Washington Fish Tissue Equivalent Concentration; EPA RC: EPA Recommended Criterion.



2013 Fish Tissue Results

Ancillary Species

Twenty-two composite samples were analyzed for total mercury in 2013. Composite mercury concentrations ranged from 15.1 to 126 ppb, with a median of 66.5 ppb (Figure 5). Leland Lake yellow perch contained the highest levels of mercury and fish from Nahwatzel (rainbow trout) and Horsethief (yellow perch) had the lowest levels. However, all composite fish analyzed in 2013 were quite low and no samples exceeded either the EPA recommended criterion of 300 ppb or the Washington State regulatory criterion (770 ppb).

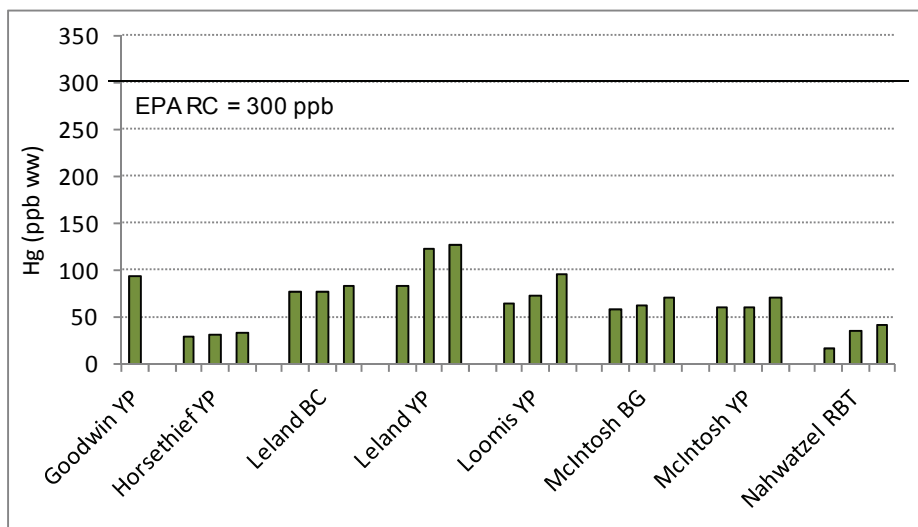


Figure 5. Mercury Concentrations of Composite Fish Samples Collected in 2013.

EPA RC: EPA Recommended Criterion; YP: yellow perch; BC: black crappie; BG: bluegill; RBT: rainbow trout; WAL: walleye.

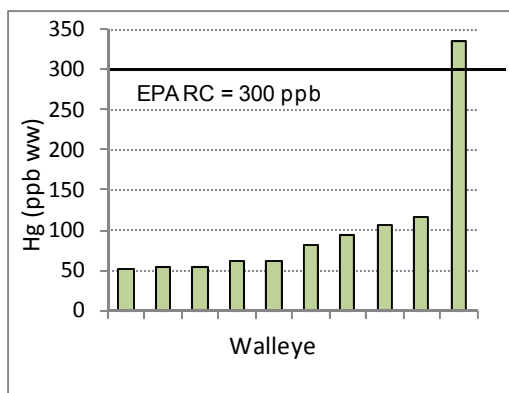


Figure 6. Mercury Concentrations of Individual Walleye from Horsethief Lake.

The field collection crew only encountered four bass at Horsethief Lake. Due to this insufficient sample size for mercury trends, the crew collected ten walleye to analyze as individuals, with the intention of collecting walleye again when the lake is revisited in five years.

Walleye from Horsethief Lake contained generally low mercury concentrations, ranging from 51.6 to 117 ppb in nine of the samples. One walleye had a much higher mercury level, 336 ppb. This sample came from the largest walleye caught (total length = 546 mm, weight = 1,692 grams) and was the only ancillary species sample to exceed the EPA recommended criterion.

Fish Tissue Mercury Relationships

Table 4. Linear Regression Coefficients of determination for Mercury Relationships.

Lake	r ²		
	Length	Weight	Age
Goodwin	0.24	0.27	0.34
Leland	0.82	0.82	0.89
Loomis	0.67	0.66	0.83
McIntosh	0.44	0.47	0.65
Nahwatzel	0.60	0.62	0.54

Bolded values indicate significance at $p < 0.05$.

Mercury concentrations were regressed against fish size and age using simple linear regression. Table 4 displays coefficients of determination (r^2). All data were \log_{10} transformed prior to regression. Normality was assessed through Shapiro-Wilks normality tests.

Mercury concentrations in bass significantly increased with fish size and age at all waterbodies except for Goodwin Lake. Bass collected from Goodwin Lake did not show a relationship between mercury and fish size. All but one bass collected from this lake were of the same age (2 years old).

Linear regressions revealed a stronger relationship between age and mercury concentrations for McIntosh Lake bass than either length or weight.

Temporal Trends

Temporal trends in bass mercury levels were assessed using analysis of covariance (ANCOVA) to identify statistically significant differences between collection years. Fish length (or age) was used as the covariate to control for the effect of fish size on mercury accumulation in bass.

Individual bass were analyzed for mercury in 2008 and 2013 at all 6 lakes. Temporal trends could not be evaluated for Horsethief Lake, as only four smallmouth bass were obtained in 2013. The Horsethief data set is included in Figure 7 for qualitative presentation only. Additional data from a 2002 survey were available for Loomis Lake (Fischnaller et al., 2003) but not included in this temporal trend analysis because fish lengths were dissimilar to the 2008 and 2013 data sets, and the 2002 data did not display a significant mercury/size relationship to control for this difference.

The McIntosh Lake data set exhibited a poor fish length-to-mercury concentration relationship. Age was found to be the strongest predictor of mercury for both 2008 and 2013 McIntosh data sets, and therefore was used as the covariate for the analysis.

Fish size and age were not significant predictors of mercury accumulation in Goodwin Lake bass for either 2008 or 2013 collection years; therefore, no covariate was used for analysis. All but one fish from Lake Goodwin were of the same age (two years); the exception was a three-year-old bass collected in 2013. Its length was outside the range of the rest of the data set and the sample was excluded from analysis.

ANCOVA

Results showed a significant difference in estimated mercury levels (least squares means) between the collection years for four out of the five lakes. Estimated bass mercury levels decreased 34% and 44% in Leland and Nahwatzel Lakes, respectively, between 2008 and 2013. Estimated bass mercury levels increased 49% and 55% at Goodwin and Loomis, respectively, from 2008 to 2013. Statistical tests did not identify a significant change in McIntosh Lake estimated bass mercury levels between 2008 and 2013. Results of the ANCOVA and post hoc tests are included in Table 5. Figure 7 displays the raw bass mercury concentration and fish length data.

Table 5. Results of ANCOVA Comparing Mercury Levels in Bass Between 2008 and 2013 Collection Years.

Waterbody	Species	ANCOVA						post-hoc		
		Co-variate	Sum of Squares	df	Mean Squares	F-Ratio	p-Value	2008 Hg _{bass}	2013 Hg _{bass}	% change
Goodwin	SMB	none	0.142	1	0.142	24.5	< 0.001	117	174	49%
Leland	LMB	length	0.156	1	0.156	10.6	0.005	506	335	-34%
Loomis	LMB	length	0.151	1	0.151	8.57	0.009	119	185	55%
McIntosh	LMB	age	0.058	1	0.058	2.78	0.114	---	---	---
Nahwatzel	LMB	length	0.271	1	0.271	65.6	< 0.001	353	197	-44%

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

Df = degrees of freedom.

Hg_{bass} = 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).

Temporal Trends

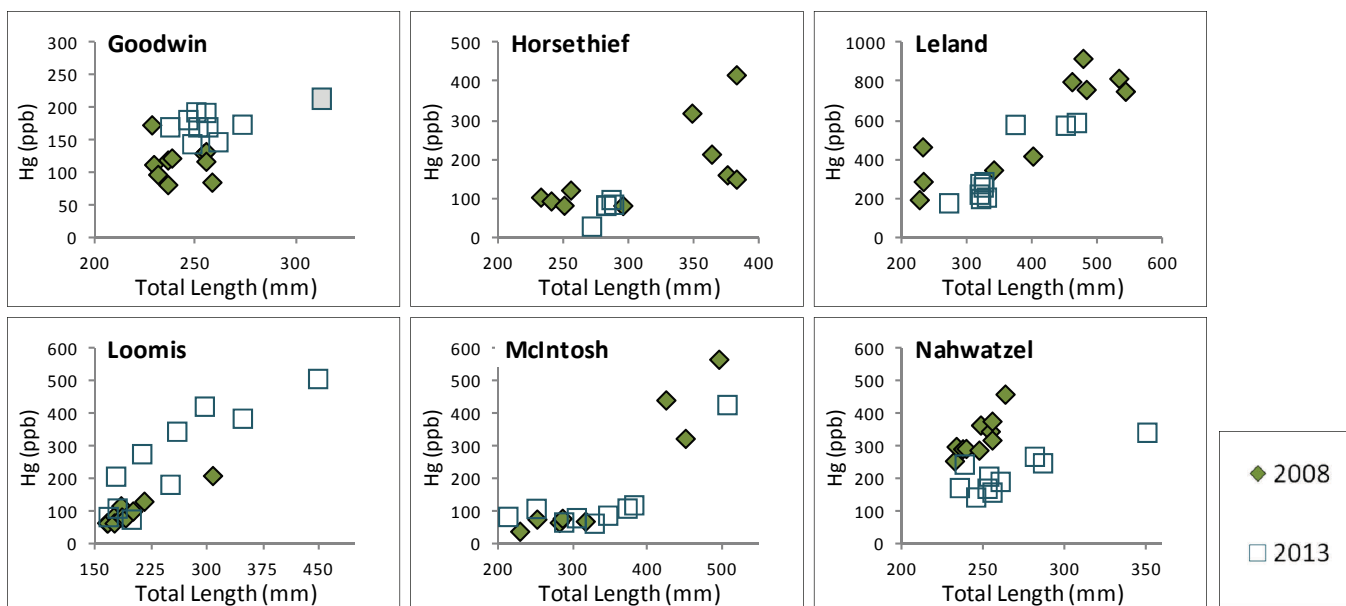


Figure 7. Mercury and Fish Length Data Sets for All Bass Collection Years.

Green diamonds = 2008 data; hollow blue squares = 2013 data; grey square = datapoint excluded from analysis.

Discussion

Bass mercury accumulation is determined by a number of factors, including the amount of mercury loading to the lake, the availability of that mercury to the trophic system (i.e., methylation), and food web dynamics.

No data were available to assess changes in mercury loading to the lakes, with the exception of Nahwatzel. An undated sediment core collected in 2011 from Lake Nahwatzel showed a steady increase in sediment mercury concentrations throughout the core to the uppermost level (Mathieu and Friese, 2012). However, sedimentation rates and mercury fluxes could not be calculated for this core. The increase in mercury concentrations is in contrast to the decrease seen in fish mercury levels at the lake, indicating that factors other than mercury loading may be responsible for the decline.

Water samples were analyzed for parameters that have been shown to influence mercury methylation during both collection periods. Water samples collected in 2013 revealed higher levels of alkalinity in Loomis and McIntosh Lake, compared to 2008 samples. Alkalinity was lower in the 2013 Nahwatzel water samples than in the 2008 samples. The first five years of sampling for this program found lower alkalinity levels correlated with higher bass mercury concentrations (Mathieu et al., 2013).

Positive correlations have been found between DOC (at low to moderate levels) and fish mercury concentrations by facilitating transport of mercury to lakes from nearby wetlands (Driscoll et al., 1995). DOC levels in water samples collected from Leland Lake decreased by an average of 34% between 2008 and 2013. Modest increases in DOC were seen at Loomis Lake. Both are consistent with the trend seen in bass mercury concentrations.

No large-scale changes in landscape, such as wetland size, were identified for any of the study lakes through geographic information system (GIS) exercises during the time period of interest. Average annual precipitation values were slightly higher in the four-year period preceding the 2013 collection for all lakes, compared to the antecedent four-year period for 2008 (PRISM Climate Group, accessed 1/27/15).

Relationships between fish lengths, weights, and ages were similar between collection years at each lake, indicating that differences in mercury are not likely attributable to changes in growth patterns. However, changes in fish communities and bass diets, which were not assessed by this study, may have influenced mercury levels.

Temporal Trends

Composite Fish Samples

Mercury levels in composite fish samples collected in 2008 and 2013 are presented in Figure 8. Composite samples are collected and analyzed to support fish consumption advisories by DOH, not to detect trends. Because of smaller sample sizes, statistical trends are not possible with the composite data set. The data presented in Figure 8 provide a qualitative view of mercury levels in species other than bass collected by this program. Samples were paired based on fish length (< 5% RPD).

Composite samples collected from Horsethief and Leland Lakes in 2013 had lower mercury concentrations than composites analyzed in 2008. This pattern was particularly consistent among Leland Lake composites, as all three composites contained about half the mercury in 2013 compared to 2008. Leland Lake largemouth bass samples were also lower in mercury in 2013 than in 2008.

Mercury concentrations were higher in 2013 yellow perch samples collected from Loomis Lake. No change was found in mercury levels of largemouth bass collected during the same period at Loomis Lake.

Composite samples from McIntosh Lake (bluegill and yellow perch) and Lake Nahwatzel (rainbow trout) contained similar mercury levels in 2013 and 2008. WDFW staff regularly stock Lake Nahwatzel with rainbow trout in the spring and fall.

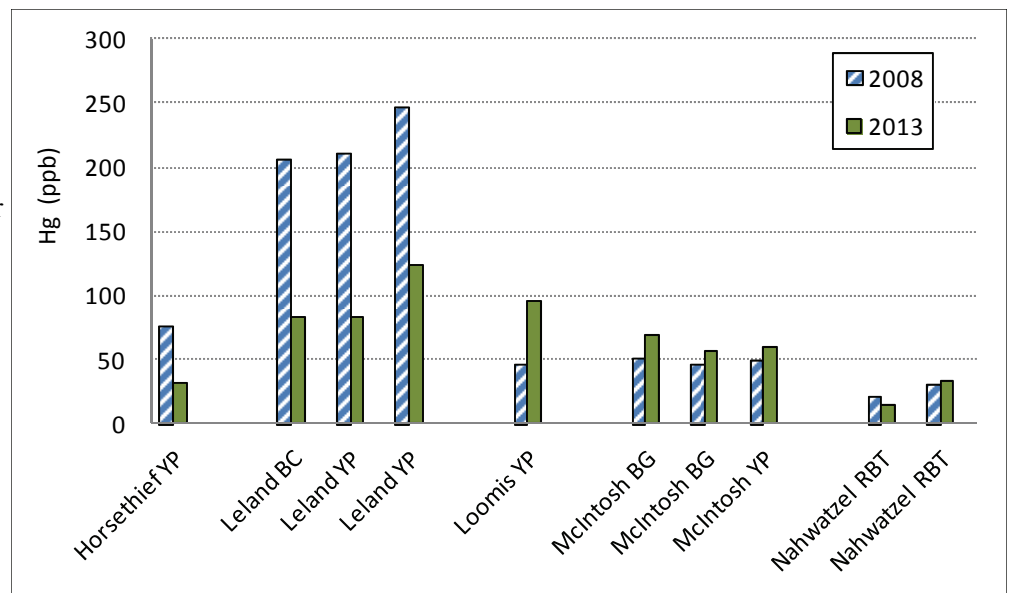


Figure 8. 2008 and 2013 Mercury Concentrations in Composite Samples Grouped by Similar Length.

YP: yellow perch; BC: black crappie; BG: bluegill; RBT: rainbow trout.

Conclusions

Ecology collected largemouth and smallmouth bass from six lakes in the fall of 2013 as part of a long-term monitoring study to assess mercury trends. A total of 54 individual bass were analyzed for total mercury. Results were compared to fish previously sampled from the waterbodies in 2008. Mercury was also analyzed in 22 composite samples of other fish species from the study lakes.

Results of this study include the following:

- No fish samples exceeded the Washington state regulatory criterion of 770 ppb. Ten samples of largemouth bass and walleye were above the EPA-recommended criterion for human health of 300 ppb. All lakes except for Goodwin contained at least one sample above 300 ppb.
- Statistical tests revealed a significant decrease in mercury concentrations between 2013 and 2008 for Leland and Nahwatzel Lake bass. Estimated mercury means decreased 34% and 44% in Leland and Nahwatzel Lakes, respectively.
- Mercury concentrations increased significantly between 2008 and 2013 at Goodwin and Loomis Lakes. Estimated bass mercury levels increased 49% and 55% at Goodwin and Loomis, respectively, from 2008 to 2013.
- No significant change was found in McIntosh Lake bass mercury levels between 2008 and 2013. Trends could not be assessed for Horsethief Lake, as only four smallmouth bass were encountered in 2013.
- Composite samples collected from Horsethief and Leland Lakes in 2013 had lower mercury concentrations than composites analyzed in 2008.
- A 2013 yellow perch composite sample collected from Loomis Lake contained higher mercury levels compared to a composite analyzed in 2008. Composite samples collected from McIntosh and Nahwatzel Lakes contained similar mercury levels in 2013 and 2008.

Recommendations

- This report summarizes the ninth year of sampling for the Mercury Trends in Fish Tissue study. The first five years of this study established baseline mercury levels, and lakes have been re-sampled over the last four years to assess trends. Sampling conducted in 2014 will conclude the first cycle of trends analysis for the full set of study lakes (n = 30). The 2014 report should include an in-depth analysis of trends at the 30 study lakes.
- The 2014 report should also recommend changes to the sampling plan for the next five-year cycle of this study. While reviewing the full data set, the authors should consider whether all ancillary and water chemistry parameters are necessary for the study objectives. The authors should also research techniques to characterize food web structure, such as stable carbon and nitrogen isotopes, and determine whether this would be a feasible addition to the monitoring program.
- The Washington State Department of Health should review the results of this report and consider the information when making or updating freshwater fish consumption advice.

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Data for this project are available at Ecology's Environmental Information Management (EIM) website www.ecy.wa.gov/eim/index.htm. Search Study ID, HgFish13.

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