Measuring Mercury Trends in Freshwater Fish, 2015 Sampling Results

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2015 Highlighted Findings

Statistical tests showed the following trends in mercury levels of bass collected in 2015 compared to 2005/2010:

- Levels **increased** 73% in Silver Lake between 2005 and 2015, but there was no difference between 2010 and 2015.
- **No change** was found in 4 of the 6 waterbodies between 2015 and 2005/2010.
- 98% of bass samples exceeded the Washington State Water Quality Standard for methylmercury.

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 fish-eating wildlife and humans.

Overview

In 2005, the Washington State Department of Ecology (Ecology) began a monitoring program with the primary goal of assessing temporal trends in mercury levels of freshwater fish throughout the state. Each year, Ecology collects 10 individual largemouth or smallmouth bass from 6 lakes for analysis of total mercury. Ecology returns to each set of lakes every 5 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 2015, Ecology collected fish from Liberty Lake, Loon Lake, Potholes Reservoir, Silver Lake (Cowlitz County), Lake Spokane, and the Yakima River near Horn Rapids Park (Figure 1). Fillet tissue was analyzed for total mercury to compare to data collected by the program in 2005 and 2010.

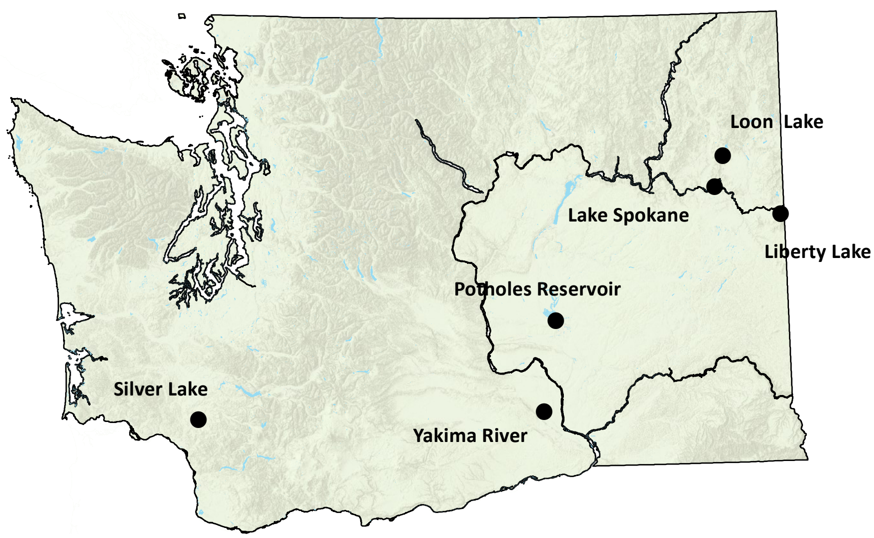


Figure 1. Locations of 2015 Sampling Sites.

This monitoring program supports Ecology's efforts to track mercury reductions in Washington State. Ecology and the state Department of Health (DOH) developed a chemical action plan (CAP) for mercury in 2003 (Peele et al., 2003) to address the threat of mercury in Washington and made recommendations for state actions to reduce mercury.

For More Information

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

Chemical Action Plan website: http://www.ecy.wa.gov/programs/hwtr/RTT/pbt/caps.html

Publication No. 17-03-006 April 2017

Methods

In the fall of 2015, Ecology collected a total of 57 individual bass from 6 waterbodies. Bass were analyzed individually to achieve statistical power in detecting trends. Additional fish species were also collected, forming 15 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). Washington Department of Fish and Wildlife (WDFW) biologists determined ages of the bass. Edible muscle tissue from fish were analyzed individually, or as 3-5 fish composite samples, for total mercury using cold vapor atomic absorption (EPA Method 245.6). All mercury data presented in this report are expressed on a ug/kg (ppb) wet weight basis.

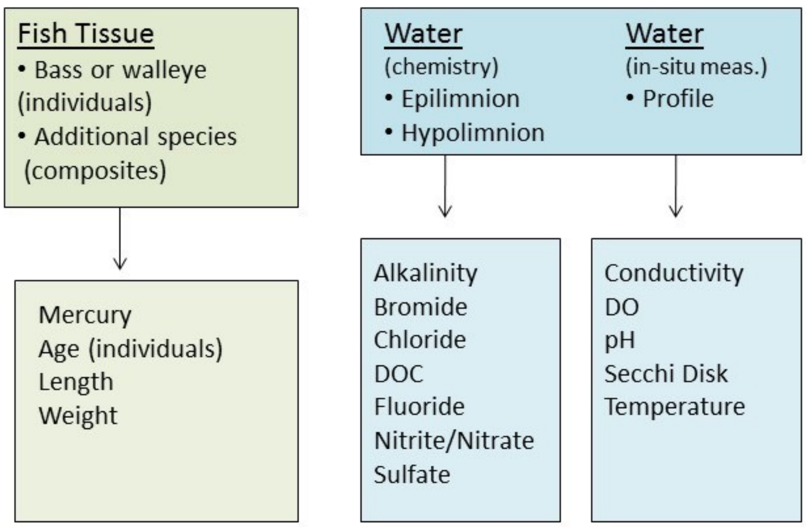


Figure 2. Analytes and Measurements Recorded at each Water-body. *DOC: dissolved organic carbon; DO: dissolved oxygen.*

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 2015. 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 collection goals met in 2015 is provided in Table 1. Fish collection efforts did not meet goals at Liberty Lake and Potholes Reservoir, where only 9 and 8 smallmouth bass were encountered, respectively.

All quality control (QC) tests for fish-tissue and water-chemistry laboratory data were within measurement quality objectives (MQOs) with the following exceptions. Three laboratory control samples for the fish tissue mercury analysis had recoveries lower than MQOs outlined in the QAPP (90-110%) (Seiders, 2006) but were within Ecology's Manchester Environmental Laboratory's (MEL's) method acceptance limits (85-115%). Results were deemed usable without qualification. Also, one field blank contained chloride above reporting limits; however, all field samples associated with the blank were 10 times greater than the blank concentration. Quality assurance (QA) data and laboratory case narratives are available upon request.

Table 1. Summary of Sampling Conducted in 2015.

Collection Goal	Waterbody					
	Liberty Lake	Loon Lake	Potholes Reservoir	Silver Lake	Lake Spokane	Yakima River
10 individual bass	NA*	+	NA*	+	+	+
Species collected for composites	YP	PMP	BBH, LWF, YP	BC	LSS, NPM	NPM
2 water samples	+	+	+	+	+	+
Hydrolab profile	+	+	+	+	+	+

NA = Not Achieved. "+": goal achieved. *9 individual bass collected from Liberty and 8 individual bass from Potholes. YP: yellow perch; PMP: pumpkinseed; BBH: brown bullhead; LWF: lake whitefish; BC: black crappie; LSS: largescale sucker; NPM: northern pikeminnow.

Waterbody Descriptions

Location and physical data for the 2015 study waterbodies are included in Table 2 and Figure 1. Ecology previously sampled the 6 waterbodies in 2005 (Furl et al., 2007) and 2010 (Meredith and Friese, 2011). Site selection in 2005 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). The QAPP contains detailed information on site selection (Seiders, 2006).

Table 2. Location and Physical Data for 2015 Waterbodies.

Waterbody	Liberty	Loon	Potholes	Silver	Spokane	Yakima River
County	Spokane	Stevens	Grant	Cowlitz	Spokane	Benton
Drainage Basin (sq mi)	13.3	14.1	3,920	39.3	6,168	6,120
Elevation (ft)	2,053	2,381	1,046	484	1,536	410
Surface Area (ac)	713	1,130	28,000	2,300	45,000	-
Lake Volume (ac-ft)	16,300	51500	500,000	13,000	243,000	-
Maximum Depth (ft)	30	100	140	10	180	10
Mean Depth (ft)	23	46	18	6	50	6
Dominant Land Type	residential, forest	forest, residential	shrubland, agriculture	forest, residential	mixed	shrubland, agriculture

Water Sample Results

Multi-parameter water profiles indicated thermal stratification in the 5 lakes surveyed. The exception to this was Liberty Lake, a large but shallow lake with a maximum depth of only 10 feet. Profiles also showed a drop in dissolved oxygen near the bottom at all 6 waterbodies with the exception of Silver Lake and Yakima River. Generally, pH decreased with depth in all waterbodies.

The waterbodies on the east side of the state, particularly Potholes Reservoir, Yakima River, and Lake Spokane, had the highest concentrations of alkalinity, chloride, and sulfate. Silver Lake, on the west side, had the lowest alkalinity and sulfate levels. Liberty Lake was also relatively low in alkalinity and sulfate.

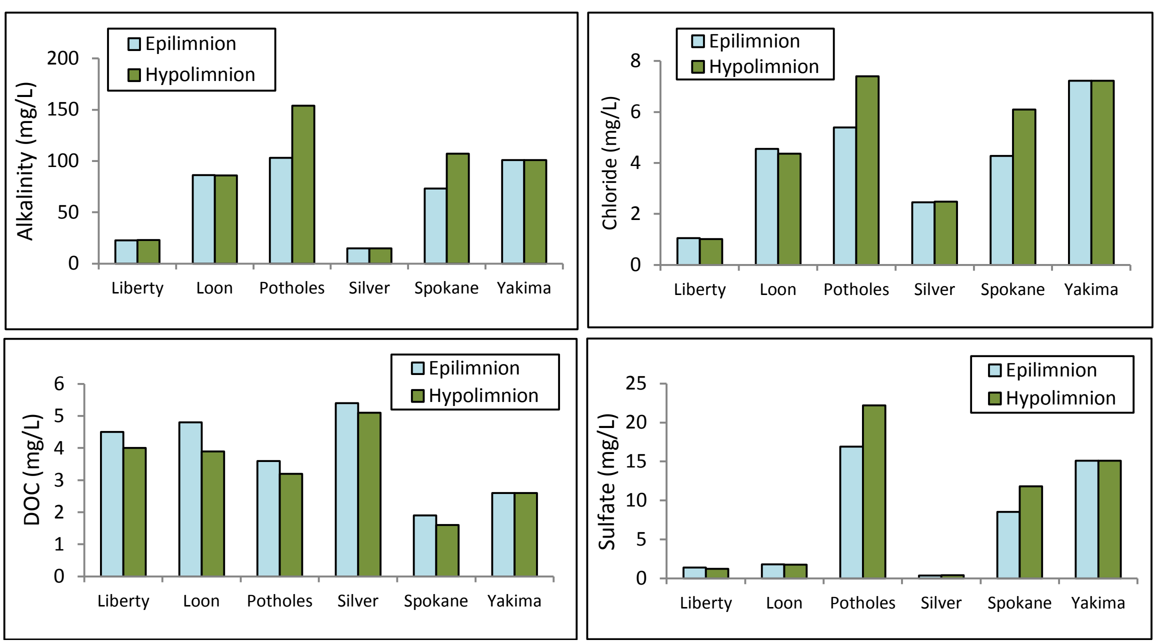


Figure 3. Alkalinity, Chloride, DOC, and Sulfate Concentrations in Water Samples.

2015 Fish Tissue Results

Numerical Thresholds for Mercury in Fish Tissue

Data in this report are compared to 2 methylmercury thresholds: Washington State's new Water Quality Standard (WQS) for human health (40 CFR 131.45), which went into effect on December 28, 2016, and DOH's Screening Level (DOH 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. This study analyzes total mercury as a surrogate for methylmercury, as methylmercury makes up more than 95% of the total mercury in fish tissue (Bloom, 1995; Driscoll et al., 1994).

Washington State's methylmercury WQS of 30 ppb is a tissue-based human health criterion based on a fish consumption rate of 175 g/day. This rate is representative of the average fish consumption of all fish and shellfish consumed (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.

The DOH Screening Level is a threshold DOH uses when developing fish consumption advisories, in addition to other factors. The DOH SL - 101 ppb - is based on a general population consumption rate of 59.7 g/day, which the American Heart Association recommends for a healthy diet (2 meals per week). DOH uses the SL to provide advice to fish consumers in Washington, while the WQS 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 (not meeting) these thresholds do not necessarily represent an impaired use or a fish consumption advisory.

Individual Largemouth and Smallmouth Bass

Fillet tissue samples from a total of 57 individual bass were analyzed for mercury in 2015. Mercury concentrations of smallmouth and largemouth bass are shown in Figure 4, and summary statistics are presented in Table 3. Mercury concentrations in individual bass ranged from 27.7 to 572 ppb, with a median of 105 ppb. All but one sample (98%) exceeded the state's WQS for human health of 30 ppb. Thirty-one bass samples (54%) exceeded the DOH SL. All waterbodies except for Lake Spokane had at least one exceedance of the DOH SL. Bass collected from Liberty and Loon Lakes contained the highest levels of mercury. These lakes also had some of the oldest (9 -14 years) and longest (459-547 mm) bass collected in 2015.

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)	Median
Liberty	SMB	9	217 - 496	351 (114)	103 - 2129	860 (765)	3 - 11	5.6 (3.3)	50.7 - 572	223 (192)	105
Loon	LMB	10	301 - 547	410 (78)	352 -2876	1186 (760)	3 - 14	9.0 (4.0)	104 - 470	234 (120)	223
Potholes	SMB	8	286 - 429	344 (52)	285 - 1090	564 (278)	2 - 5	3.1 (1.0)	60.7 - 131	87.3 (26.9)	72.4
Silver	LMB	10	230 - 462	330 (77)	157 - 1696	620 (504)	2 - 9	3.7 (2.2)	42.9 - 243	130 (77.9)	111
Spokane	SMB	10	246- 452	350 (88)	183 - 1397	694 (493)	2 - 7	4.1 (2.0)	27.7 - 96.5	65.3 (21.6)	75.2
Yakima	SMB	10	232 - 351	296 (36)	155 - 604	358 (138)	1 - 2	1.8 (0.4)	87.8 - 250	148 (48.8)	140
All Sites	---	57	217 - 547	347 (83)	103 - 2876	716 (583)	1-14	5 (3.3)	27.7 - 572	149 (115)	105

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

2015 Fish Tissue Results

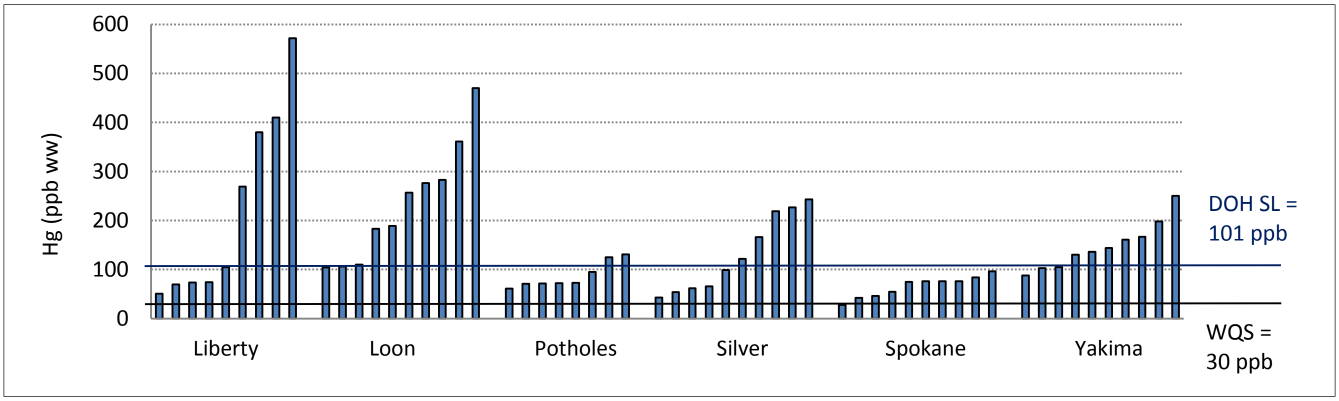


Figure 4. Mercury Concentrations of Individual Bass Samples.
DOH SL = WA Department of Health Screening Level; WQS = Washington State Water Quality Standard for methylmercury.

Ancillary Species

Fifteen fish composite samples were analyzed for total mercury in 2015. Mercury concentrations ranged from 16.8 to 439 ppb and the median concentration was 109 ppb. Eighty percent of samples (12 out of 15) contained mercury levels above Washington's WQS of 30 ppb. Fifty-three percent of composites (8 of 15) were above the DOH SL of 101 ppb. Northern Pikeminnow collected from Yakima River had the highest mercury concentrations. The average length of northern pikeminnow in the sample with the highest concentration was twice that of the other 2 samples from Yakima River, although a similar sized northern pikeminnow from Lake Spokane had much lower levels.

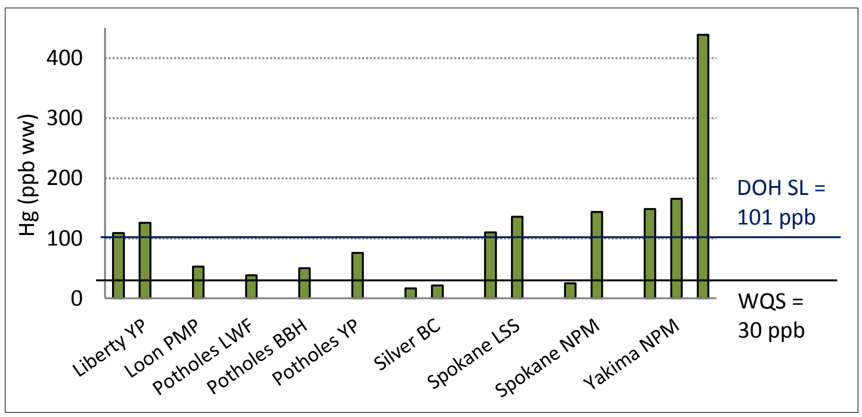


Figure 5. Mercury Concentrations of Composite Fish Samples.
DOH SL = WA Department of Health Screening Level; WQS = Washington State Water Quality Standard for methylmercury; YP = yellow perch; PMP = pumpkinseed; LWF = lake whitefish; BBH = brown bullhead; BC = black crappie; LSS = largescale sucker; NPM = northern pikeminnow.

Table 4. Linear Regression Coefficients for Log_{10} Mercury: Size/Age Relationships.

Lake	r^2		
	Length	Weight	Age
Liberty	**0.93**	**0.92**	**0.95**
Loon	**0.91**	**0.87**	**0.84**
Potholes	0.49	0.48	**0.57**
Silver	**0.47**	**0.51**	**0.55**
Spokane	**0.78**	**0.76**	**0.69**
Yakima	0.12	0.11	0.15

***Bolded** values indicate statistical significance ($p < 0.05$).*

Fish Tissue Mercury Relationships

Linear regressions between $\log_{10}$ mercury concentrations and $\log_{10}$ fish size or age are presented in Table 4. Regressions were conducted on $\log_{10}$ data to make the relationship linear and improve normality. Mercury concentrations in bass significantly increased with fish size and age at Liberty Lake, Loon Lake, Silver Lake, and Lake Spokane. Bass collected from Potholes Reservoir did not show a significant relationship between mercury and fish size, but a significant relationship was apparent between mercury and fish age. No relationships were apparent for Yakima River bass.

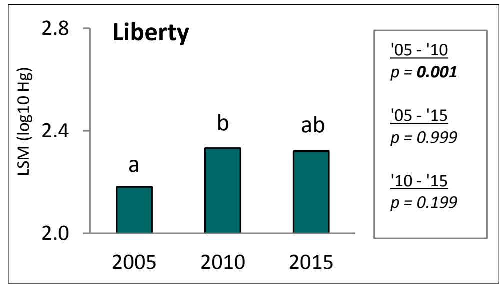
Temporal Trends

Individual bass were analyzed for mercury in 2005, 2010, and 2015 for this program. Temporal trends in bass mercury levels were assessed using analysis of covariance (ANCOVA). To control for the effect of fish size on mercury accumulation, a covariate of fish length was used for Liberty, Loon, and Silver Lakes datasets, while age was used for the Potholes Reservoir data. No covariate was used for the Lake Spokane and Yakima River datasets, as mercury-to-size relationships were not significant. Data were $\log_{10}$-transformed to make the relationships linear and to improve normality. All datasets met test assumptions except for Liberty and Loon Lake datasets, where homogeneity of variances was not met. However, sample sizes were equal, and the ANCOVA test is robust to heterogeneous variances when sample sizes are equal and data are normally distributed (Huitema, 2011).

Least squares means were back-transformed to provide an estimate of length-adjusted bass mercury concentrations. A Duan's Smearing estimator was applied to estimated means to correct for back-transformation bias (Duan, 1983; Helsel and Hirsch, 2002). Percent change between collection years was calculated using the corrected estimated mercury means.

ANCOVA

ANCOVA results showed a significant difference in least squares means for at least one of the collection years in two out of the six waterbody datasets. Mercury levels in Silver Lake bass collected in 2015 were 72.6% higher than bass collected in 2005 (Figure 6 and Table 5). No significant difference was found in Silver Lake between 2010 and 2015. Mercury concentrations in Liberty Lake bass collected in 2010 were 41.6% higher compared to 2005, but no change was found in 2015 from either 2005 or 2010.



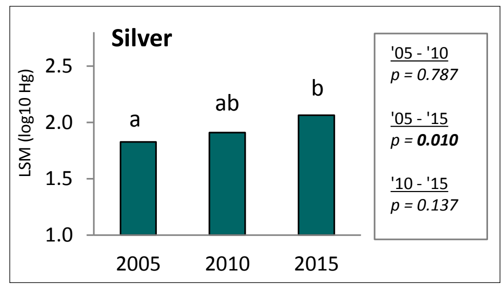


Figure 6. Post-hoc Test Results. *Post-hoc tests performed were Games Howell (Liberty) and Bonferroni (Silver). A difference in letters indicates significant difference between collection years.*

Table 5. Results of ANCOVA Comparing Mercury Levels in Bass Between 2005, 2010, and 2015 Collections.

Waterbody	Species	Co-variate	Sum of Squares	df	Mean Squares	F-Ratio	p-Value	2005 Hg_{bass}	2010 Hg_{bass}	2015 Hg_{bass}	Mean Fish Length (mm)
Liberty	SMB	length	0.166	2	0.083	12.263	**< 0.001**	154	219	213	382
Loon	LMB	length	0.004	2	0.002	0.439	0.649	234	248	234	425
Potholes	SMB	age	0.039	2	0.020	1.860	0.177	106	122	98.5	375
Silver	LMB	length	0.285	2	0.142	5.388	**0.011**	71.1	86.3	123	338
Spokane	SMB	none	0.049	1	0.049	1.136	0.312	64.8	---	87.0	416
Yakima	SMB	none	0.003	2	0.001	0.062	0.940	153	145	150	314

All variables were log10 transformed prior to ANCOVA. Hg_{bass} = back-transformed least squares means with Duan's Smearing estimator applied to correct for back-transformation bias (Duan, 1983; Helsel and Hirsch, 2002).
df = degrees of freedom; SMB = smallmouth bass; LMB = largemouth bass.

Temporal Trends

Discussion

Only one waterbody showed a significant change in 2015 bass mercury levels compared to previous collection years, and that change was from 2005 to 2015. No trends between 2010 and 2015 were detected. A lack of recent temporal trends in fish tissue mercury levels has been reported from waterbodies across North America (Eagles-Smith et al., 2016; Tang et al., 2013). Decreases in fish tissue mercury levels on a broad scale typically occurred following dramatic mercury emissions reductions in the 1970s and 1980s (e.g. Chalmers et al., 2011) and in areas of hotspots following improvements to local emissions control (e.g. Hutcheson et al., 2014).

In Washington State, mercury concentrations in precipitation and wet deposition decreased following the closure of medical waste incinerators in the 1990s (Prestbo and Gay, 2009) and have remained moderately low with no significant increasing or decreasing trends in the past decade (Weiss-Penzias, 2016). Though the response of fish tissue mercury levels to mercury loading is complex and can vary depending on many factors, it is worth noting here because the waterbodies in this study lack direct point sources. As mercury loading from the atmosphere remains stable, trends found in this study's waterbodies are likely to reflect local effects from watershed changes and in-lake processes.

While the increase in Silver Lake bass mercury concentrations in 2015 over 2005 levels was substantial (73%), mercury concentrations are still relatively low in the lake compared with other waterbodies sampled for this program over the last 5 years. The length-adjusted estimated mercury mean (at 332 mm) in 2015 was 123 ppb, compared to 71.1 ppb in 2005. This program did not collect water data from Silver Lake in 2005, but samples taken in 2010 were similar to 2015, with slightly lower alkalinity values. The lake has experienced increasing eutrophication and frequent algal blooms over the last 2 decades (WA State Toxic Algae database, accessed 3/15/17); however, we do not have sufficient data available to assess what effect, if any, this has had on mercury accumulation in the bass.

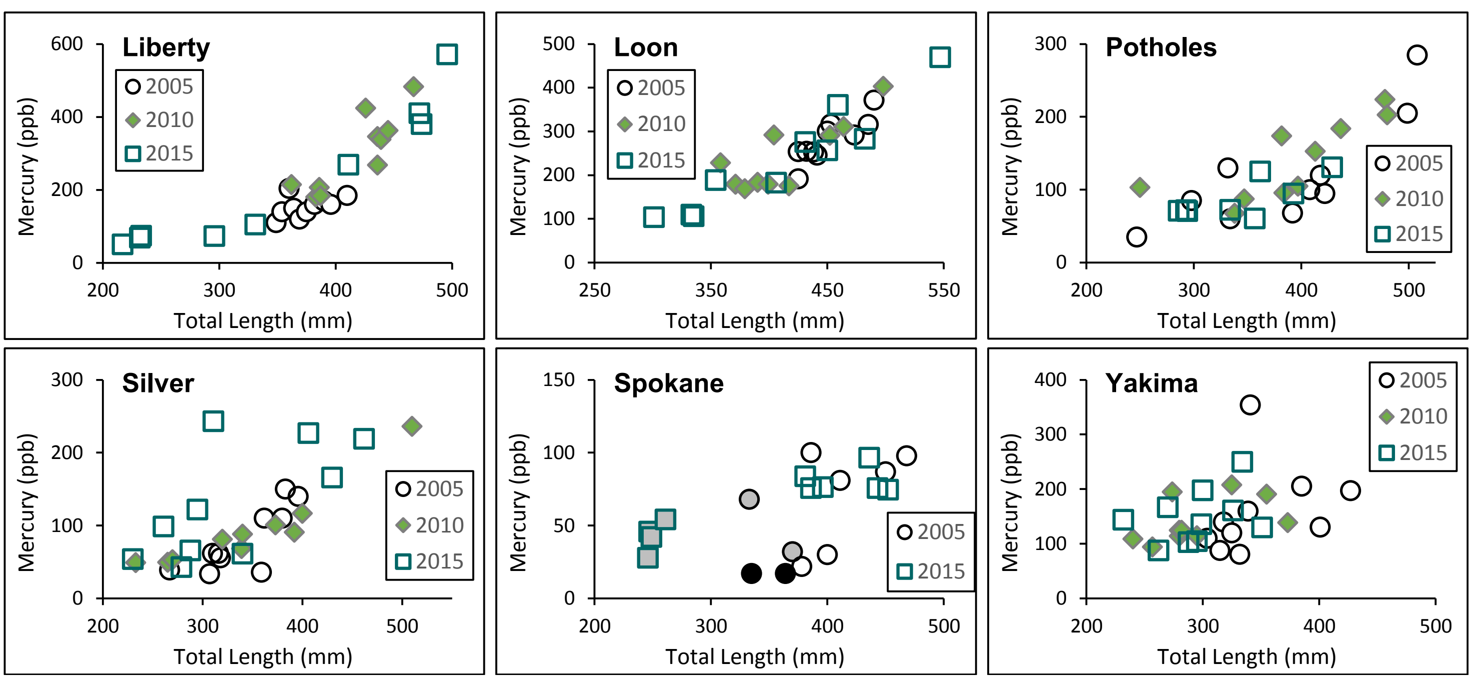


Figure 7. Mercury and Fish Length Data Sets for All Bass Collection Years.

Grey shapes= datapoint excluded from analysis; solid black = non-detect value and excluded from analysis.

Temporal Trends

Composite Fish Samples

In 2010 and 2015, additional species were collected for analysis of composite samples to support fish consumption advisories by DOH, not to detect trends. Due to smaller sample sizes, statistical trends are not possible with the composite samples dataset. The data presented in Figure 7 provides a qualitative view of mercury levels in species other than bass collected by this program. Samples were paired based on average composite fish length (< 10% relative percent difference between collection years). Not all composite samples were able to be paired with the 2010 collection year based on length and, therefore, are not included in the graph. No composite samples were collected in 2005.

Mercury concentrations showed mixed results in Potholes Reservoir composites, with one brown bullhead sample higher in 2015 compared to 2010 and a lake whitefish sample slightly lower in 2015. However, both composite pairs were low in mercury. Black crappie collected from Silver Lake were also very low in mercury and similar between collection years.

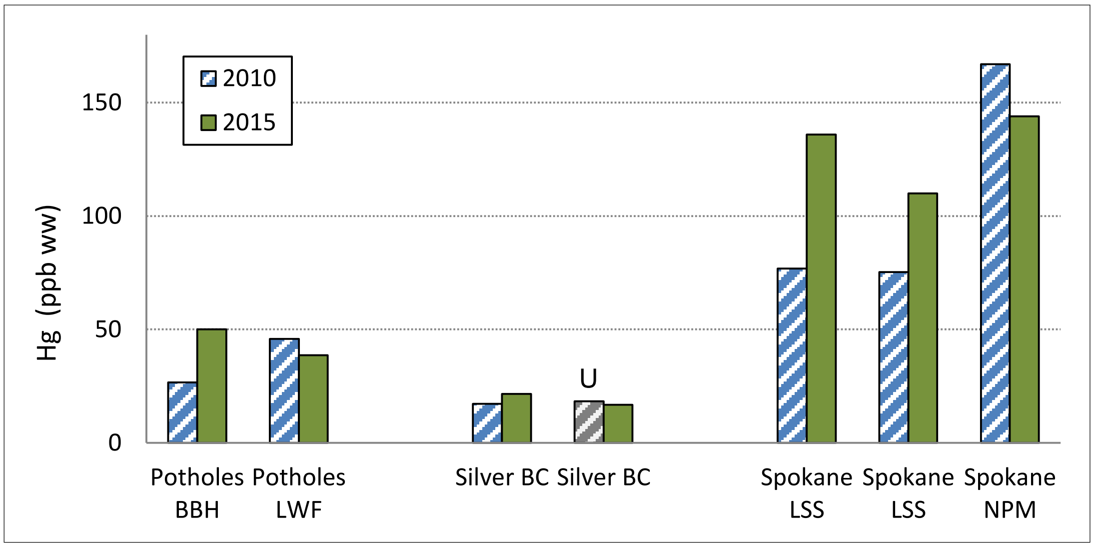


Figure 7. 2010 and 2015 Mercury Concentrations in Composite Samples.
BBH = brown bullhead; LWF = lake whitefish; BC = black crappie; LSS = largescale sucker; NPM = northern pikeminnow.
U = Mercury not detected at level indicated.

Two composites of Lake Spokane largescale suckers contained higher mercury concentrations in 2015 compared to 2010, while the northern pikeminnow sample was slightly lower in 2015.

Conclusions

In the fall of 2015, Ecology collected largemouth and smallmouth bass from 6 waterbodies as part of a long-term monitoring study to assess mercury trends. A total of 57 individual bass from Liberty Lake, Loon Lake, Potholes Reservoir, Silver Lake, Lake Spokane, and the Yakima River were analyzed for total mercury. This report compares 2015 fish tissue mercury concentrations to mercury concentrations in fish previously sampled from the waterbodies in 2005 and 2010. Mercury was also analyzed in 15 composite samples of other fish species.

Results of this study include the following:

- Statistical tests revealed a significant increase in estimated mean mercury concentrations in Silver Lake largemouth bass between 2015 and 2005. The length-adjusted estimated mercury concentration for Silver Lake was 123 ppb in 2015 compared to 71.1 ppb in 2005, a 72.6% increase. While this change is substantial, mercury concentrations are still fairly low in the lake compared to other waterbodies sampled for this program over the last 5 years.
- No other waterbodies tested in 2015 showed a significant change in bass mercury concentrations from previous sampling years. This lack of trends is consistent with other reports across North America on recent temporal trends in mercury levels in fish and also consistent with trends in measured regional mercury deposition in precipitation.
- Composite samples collected from Potholes Reservoir and Silver Lake had similar concentrations between 2010 and 2015 (no composites were collected in 2005). Two composites of Lake Spokane largescale suckers contained higher mercury concentrations in 2015 compared to 2010, while a Lake Spokane northern pikeminnow sample was slightly lower in 2015. Composite samples are not collected for statistical trend analysis.
- Ninety-eight percent of individual bass samples (56 out of 57) analyzed in 2015 were above (did not meet) Washington State's new Water Quality Standard for methylmercury of 30 ppb. Eighty percent of composites exceeded this criterion as well. The number of samples exceeding the DOH Screening Level were lower, with 54% of individual bass and 53% of composites containing mercury levels above 101 ppb.

Recommendations

The data presented in this report should be reviewed by the following agencies:

- Ecology should assess the data during the next Water Quality Assessment.
- The Washington State Department of Health (DOH) should review the results and consider the data when making or updating fish consumption advisories.

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