

# Measuring Mercury Trends in <br> Freshwater Fish in Washington State 

## 2010 Sampling Results

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# Measuring Mercury Trends in Freshwater Fish in Washington State 

## 2010 Sampling Results

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Waterbody Numbers: See Table B-1

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## Abstract

The Washington State Department of Ecology (Ecology) has conducted a long-term trend monitoring program for mercury in freshwater fish since 2005. This report summarizes findings from the sixth year (2010) of monitoring.

Ecology collected a total of 50 individual bass and 27 composite fish samples for mercury analyses in the fall of 2010. Waterbodies sampled were Liberty Lake, Loon Lake, Potholes Reservoir, Silver Lake, Lake Spokane, and Yakima River. Bass collection efforts were unsuccessful at Lake Spokane. Water chemistry samples were also collected from the six waterbodies during the summer of 2010.

Mercury concentrations in individual bass ranged from 49.3 to 483 ppb across the waterbodies. Liberty Lake bass contained the highest levels (mean $=301 \mathrm{ppb}$ ) and Silver Lake the lowest (mean = 94 ppb ). Mercury concentrations in composite samples of species other than bass ranged from not detected (18.3U) to 167 ppb . Ninety-six percent of composites had mercury levels above 18.3 ppb .

All fish tissue samples were below (met) the Washington State Water Quality Standard for mercury of 770 ppb . Seven out of 50 individual bass samples contained mercury levels above the U.S. Environmental Protection Agency (EPA) Recommended Criterion of 300 ppb . All composite samples were below the EPA Recommended Criterion.

Results from statistical tests showed no significant differences in bass mercury levels between 2005 and 2010 at four of the five waterbodies evaluated. A significant difference was found at Liberty Lake, where the percent change in estimated $\log _{10}$ mercury means was $+5.7 \%$ from 2005 to 2010 .

This report recommends at least three 5-year cycles of monitoring at Mercury Trends sites.

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## Introduction

## Background

Mercury is a naturally occurring element that exhibits persistent, bioaccumulative, and toxic (PBT) properties. Sources of mercury in the environment are both natural and anthropogenic, but human actions have increased the amount of mercury cycling in the environment three- to five-fold since the beginning of the industrial age (Selin, 2009). Human-caused releases occur from combustion of fossil fuels, metals production, and industrial processes. Mercury enters aquatic systems primarily through deposition from the atmosphere.

Mercury in the environment is a concern because of its highly toxic and bioaccumulative nature. Levels increase up food chains, resulting in concentrations among upper trophic level fish that can be harmful to piscivorous wildlife and humans.

The Washington State Departments of Ecology (Ecology) and Health (DOH) developed a chemical action plan (CAP) for mercury in 2003 (Peele, 2003). The CAP addressed the threat of mercury in Washington State and made recommendations on mercury reductions to be taken by the state.

In 2005, Ecology began monitoring largemouth and smallmouth bass from waterbodies across the state in order to assess temporal changes in environmental mercury levels. This report represents the sixth year of the monitoring program. Ecology studies leading up to this project are summarized in the next section.

## Previous Fish Tissue Mercury Studies

Elevated concentrations of mercury in Washington State freshwater fish were first highlighted by a study conducted in 2000 on Lake Whatcom (Serdar et al., 2001). This study analyzed 273 fish and signal crayfish and found 12 bass and one yellow perch had mercury levels that exceeded (did not meet) the state regulatory criterion, which was 825 parts per billion ( ppb ) at the time of publication. Smallmouth bass contained the greatest mercury concentrations of all species analyzed. The authors recommended statewide monitoring of bass mercury levels.

In response to the Lake Whatcom study recommendations, Ecology conducted a one-time survey to assess mercury concentrations in bass across Washington State during 2001-2002 (Fischnaller et al., 2003). The study collected a total of 185 fish (largemouth and smallmouth bass) from 18 lakes and two rivers. Mercury levels exceeded the EPA Recommended Criterion ( 300 ppb ) in $23 \%$ of bass analyzed, and a single ten-year-old largemouth bass from Samish Lake exceeded the National Toxics Rule (NTR) criterion ( 825 ppb at time of publication). This study confirmed that elevated mercury levels were prevalent in Washington State bass and recommended that Ecology develop a monitoring plan for mercury in freshwater bass.

Ecology designed and implemented a long-term trend monitoring program in 2005 to assess changes in mercury levels of bass over time. Results from the first five years of monitoring were reported by Furl et al. (2007), Furl (2007a), Furl and Meredith (2008), Furl et al. (2009), and Meredith et al. (2010). The project goals included assessment of temporal changes in bass levels by re-sampling lakes every five years on a rotating basis. This report summarizes results from the sixth year of sampling, which is the first year of trends data and compares the current results (2010) to the 2005 study (Furl et al., 2007).

## Mercury Criteria for Human Health

The NTR and the EPA Recommended Criterion provide numerical thresholds useful in assessing mercury levels of waterbodies. However, Ecology has adopted the NTR Rule as the state's regulatory criterion to determine whether a waterbody meets water quality standards in Washington State. The following sections provide more information on these two criteria. Appendix F describes how Ecology and DOH evaluate mercury levels in fish tissue differently.

## National Toxics Rule (NTR)

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

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

## EPA Recommended Criterion

In 2001, EPA published a recommended water quality criterion for methylmercury to be used as guidance by states and tribes in the protection of human health (EPA, 2001). EPA based this value ( 300 ppb ) on a fish consumption rate of $17.5 \mathrm{~g} / \mathrm{d}$ for the general human adult population. This Recommended Criterion describes the level of mercury in freshwater fish that should not be exceeded in order to protect the general fish-consuming population.

## Study Design

The primary goal of the project is to characterize temporal trends in mercury levels of uppertrophic level freshwater fish in Washington State. To meet this goal, Ecology analyzes mercury levels in fillets of 10 individual bass from six waterbodies per year for five years. Sampling is to be repeated at each site every five years for long-term trend assessments.

A secondary goal of the project is to better understand patterns, dynamics, and changes in fish mercury accumulation. Ecology collects ancillary data, such as water chemistry and physical measurements, from each site for analysis of parameters that may affect mercury accumulation in fish. Figure 1 displays the ancillary data collected or measured at each site.

This project also helps DOH determine fish consumption advisories by providing mercury data on additional fish species. When available, Ecology collects two fish species in addition to bass from each study site and analyzes them as composites for mercury. Composite fillet samples consist of three to five individual fish per composite.


Figure 1. Analytes and Measurements Taken for each Waterbody as Part of the Mercury Trends Sampling Design.
DOC: dissolved organic carbon
DO: dissolved oxygen

Several changes were made to the study design in 2010. A Quality Assurance (QA) Project Plan Addendum describes these changes in detail (Meredith and Furl, 2010). Briefly, sediment sampling was discontinued since mercury concentrations in the top 2 cm of lake sediments are not expected to significantly change within such a short period (five years). Analyses of chlorophyll in water were discontinued because data from the baseline period did not show a significant relationship with fish tissue mercury levels. The following analytes were added to water analyses: bromide, chloride, fluoride, nitrate/nitrite, and sulfate. These analytes will provide information on potential factors influencing fish tissue mercury levels. More information on the study design can be found in the QA Project Plan (Seiders, 2006).

## Site Descriptions

Figure 2 displays the location of waterbodies sampled in 2010. These sites were targeted because bass mercury data are available at all six sites from the 2005 study (Furl et al., 2007). The project plan discusses original site selection criteria (Seiders, 2006). Table 1 displays physical characteristics of the lakes. Other site information, such as geographical coordinates, is available in Appendix B.


Figure 2. Fish Collection Sites for Mercury Trends Study Years 2005 and 2010.
Black circles indicate bass collected from site in both years; hollow circles indicate bass collected only in 2005.

Table 1. Physical Characteristics of Study Sites Sampled in 2005 and 2010.

| Name | Liberty <br> Lake | Loon <br> Lake | Potholes <br> Reservoir | Silver <br> Lake | Lake <br> Spokane | Yakima <br> River* |
| :--- | :---: | :---: | :---: | :---: | :---: | :---: |
| County | Spokane | Stevens | Grant | Cowlitz | Spokane | Benton |
| Drainage area (sq mi) | 13.3 | 14.1 | 3920 | 39.3 | 6168 | 6120 |
| Altitude (ft) | 2053 | 2381 | 1046 | 484 | 1536 | 410 |
| Surface area (acres) | 713 | 1130 | 28,000 | 2,300 | 45,000 | - |
| Lake volume (acre-ft) | 16,300 | 51,500 | 500,000 | 13,000 | 243,000 | - |
| Mean depth (ft) | 23 | 46 | 18 | 6 | 50 | 6 |
| Max depth (ft) | 30 | 100 | 140 | 10 | 180 | 10 |

* Yakima River above Wanawish Dam (Horn Rapids Dam)


## Collection Goals

A total of 50 individual bass and 109 fish of additional species were collected in 2010. Field crews obtained bass from all six sites except for Lake Spokane, where efforts were unsuccessful. Fish species in addition to bass were collected from all six sites. Fish collections occurred in September and October in order to match the timeframe of 2005 fish collections. Fall sampling was targeted in 2005 to coincide with other fish collection efforts and to have historical fall data to compare to (e.g., Fischnaller et al., 2003).

Field crews collected water samples and recorded multi-parameter vertical profiles from each of the six waterbodies in August 2010. Vertical profiles of pH were not obtained from Liberty Lake or Yakima River due to calibration problems. August was chosen for water sampling to match previous water collections and to capture lake conditions during thermal stratification. Previous water samples were collected in 2006 at the six waterbodies, one year after the 2005 fish collections, due to limited resources.

## Methods

## Field Procedures

## Fish

Methods for the collection and handling of fish tissue samples are described in EPA (2000) and Sandvik (2010). All fish were collected using gill nets or electrofishing. Fish were inspected to ensure there was no visible damage to skin or tissue. After positive identification, fish selected for sampling were euthanized by blunt force to the head. Fish were rinsed in ambient water, weighed to the nearest gram, and total length was measured to the nearest millimeter. Specimens were individually wrapped in foil (dull side in) and packaged in zipper-lock bags with identification labels. Packaged specimens were immediately packed in ice and held for a maximum of 72 hours during transport to Ecology Headquarters in Lacey, WA. Specimens were stored frozen until later processing.

Fish samples were processed at Ecology Headquarters following standard operating procedures (Sandvik, 2006). During processing, partially thawed fish were cleaned of slime and scales, rinsed in tap water, de-scaled, and rinsed with deionized water. Skin-on fillets were removed from one or both sides of the fish and cut into smaller sections. Tissue was ground three times using a Kitchen-Aid® food grinder and homogenized after each run through the grinder. After samples were a uniform color and texture, subsamples were removed and stored in clean (I-Chem 200®) glass jars. Jars labeled with laboratory identification numbers were transported to Ecology's Manchester Environmental Laboratory (MEL) for analysis. Remaining homogenized tissue was archived in clean jars, labeled, and placed in cold storage at $-20^{\circ} \mathrm{C}$.

After tissue samples were removed, sex of the fish was determined. The appropriate age structures (either scales or otoliths) were removed from largemouth and smallmouth bass and sent to Washington Department of Fish and Wildlife (WDFW) biologists for age determination.

All utensils were cleaned prior to use and after each sample was processed. Utensils were cleaned with Liquinox ${ }^{\circledR}$ and tap water, rinsed with $10 \%$ nitric acid, and rinsed with deionized water.

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

## Water

Water column conductivity, pH , dissolved oxygen, and temperature were measured at the water sample sites using a Hydrolab ${ }^{\circledR}$ following the Environmental Assessment Program's Standard Operating Procedures for Hydrolab ${ }^{\circledR}$ Datasonde ${ }^{\circledR}$ MiniSonde ${ }^{\circledR}$ Multiprobes (Swanson, 2007). All instruments were calibrated prior to field use, and Winkler titrations were performed as a measure of quality control for the dissolved oxygen readings. Water transparency was measured using a Secchi disc following field protocols described in EPA (2007).

Two water samples were obtained from different depths at the deepest part of the lake using a one-liter Kemmerer sampler. The samples were obtained at the mid-points of the hypolimnion and epilimnion in stratified lakes. The depth of the hypolimnion and epilimnion were determined using a temperature profile to locate the thermocline. At unstratified lakes the samples were obtained at $10-15 \%$ and $85-90 \%$ of total depth. Samples were retrieved and placed in the proper pre-cleaned bottles for analysis of alkalinity, bromide, chloride, dissolved organic carbon (DOC), fluoride, nitrite/nitrate, and sulfate. DOC and nitrite/nitrate were preserved with hydrochloric acid in the field. Samples were stored on ice in the field until shipment to MEL.

## Laboratory Procedures

MEL conducted all laboratory analyses. Table 2 provides a list of matrices, analytes, and analytical methods.

Table 2. Analytes and Analytical Methods.

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

CVAA: cold vapor atomic absorption; EPA: U.S. Environmental Protection Agency; SM: Standard Methods

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

## Data Quality

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

MEL met all quality control (QC) tests for fish tissue analyses. Average recoveries for laboratory control samples (LCS), standard reference materials (SRM), and matrix spikes were $111 \%, 115 \%$, and $95 \%$, respectively. One SRM recovery was outside of measurement quality objectives (MQOs) outlined in the project plan (117\%) (Seiders, 2006). However, because MEL does not have enough data points on record for SRM recoveries, MEL does not have QC limits for this type of SRM and the data were not qualified.

High recoveries of the LCS and SRM samples may indicate that the laboratory analyses were biased slightly high. Because the LCS recoveries were within MQOs, no corrective action was taken and mercury values are reported unqualified in this report.

Laboratory duplicates were performed on matrix spike samples, and the average relative percent difference (RPD) was $6.6 \%$. Laboratory blanks were not detected above $0.017 \mathrm{mg} / \mathrm{kg}$.

QC for water chemistry analyses included LCS, matrix spikes, laboratory duplicates, and laboratory blanks. All were within MQOs except for one DOC matrix spike recovery. The recovery for this sample was $214 \%$. MEL reported that the source sample was from a different project, so no evaluation of the recovery was made.

Complete results of QC tests for fish tissue are provided in Appendix D. QC data for water sample analyses are available upon request.

## Statistical Analysis

To test for significant differences in fish mercury levels between years, we used an analysis of covariance (ANCOVA) with fish length as the covariate. Because bass mercury concentrations increased with size, we chose a statistical test that removes this effect before comparing mercury levels. A Bonferroni post-hoc test was then used to evaluate differences in means.

To satisfy assumptions of ANCOVA, we first tested for normal distributions of the data, homogeneity of variances, and equality of the $\log _{10}$ length to $\log _{10}$ mercury regression slopes. All length and mercury data were $\log _{10}$-transformed to improve normality of the data and to make the relationship linear. Normality was assessed through Shapiro-Wilks tests ( $\alpha=0.05$ ), and all mercury and length data were normally distributed after transformations. We used the Levene's Test to assess homogeneity of variances ( $p>0.05$ ). All datasets met this assumption. The homogeneity of slopes assumption was also met, as the year + length interaction term was not significant at any of the lakes ( $\mathrm{p}>0.05$ ), indicating that the year-specific slopes do not statistically differ.

## Results

## Fish

Ecology analyzed a total of 50 individual bass and 27 composite samples for mercury in 2010. The following sections summarize individual bass and composite fish results.

## Individual Bass

Largemouth and smallmouth bass were collected from Liberty, Loon, Potholes, and Silver Lakes, as well as the Yakima River. Bass collection efforts at Lake Spokane were unsuccessful. Table 3 presents statistical summaries of individual bass collected from the sites. Complete biological data for each individual bass are provided in Appendix C.

Table 3. Statistical Summary of 2010 Individual Bass Lengths, Weights, Ages, and Mercury Concentrations.

| Waterbody | Species | Length (TL mm) |  | Weight (g) |  | Age (yr) |  | Hg (ppb ww) |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  | Range | Mean $( \pm S D)$ | Range | $\begin{aligned} & \text { Mean } \\ & ( \pm S D) \end{aligned}$ | Range | $\begin{aligned} & \text { Mean } \\ & ( \pm S D) \end{aligned}$ | Range | $\begin{aligned} & \text { Mean } \\ & ( \pm S D) \end{aligned}$ |
| Liberty | SMB | 362-467 | 417 (34) | 698-1578 | 1072 (292) | 4-8 | 5.6 (1.5) | 180-483 | 301 (106) |
| Loon | LMB | 358-498 | 413 (45) | 655-2147 | 1198 (456) | 6-9 | 7.2 (1.0) | 169-403 | 241 (79) |
| Potholes | SMB | 250-480 | 391 (69) | 181-1668 | 907 (457) | 2-9 | 4.7 (2.7) | 67.8-224 | 140 (55) |
| Silver | LMB | 233-510 | 344 (81) | 187-2445 | 735 (666) | 2-11 | 4.3 (2.8) | 49.3-236 | 94 (55) |
| Yakima | SMB | 240-373 | 296 (42) | 166-747 | 388 (191) | 2-4 | 2.4 (0.7) | 94.5-208 | 142 (41) |
| All Sites | --- | 233-510 | 372 (72) | 166-2445 | 860 (512) | 2-11 | 4.8 (2.4) | 49.3-483 | 183 (103) |

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

## Bass Size

Figure 3 displays the length-to-weight relationship for individual bass collected from the five waterbodies. Eighty-four percent ( 42 out of 50 ) of individual bass lengths met the target size range of $250-460 \mathrm{~mm}$.


Figure 3. Relationship of Individual Bass Length and Weight Collected at Each of the Waterbodies.

## Bass Mercury Level

All individual bass samples contained mercury levels above detection limits. Concentrations ranged from 49.3-483 ppb across the waterbodies. Liberty Lake contained the highest levels of mercury in bass, with an average of 301 ppb . These fish were also the longest (mean total length $=417 \mathrm{~mm}$ ). The lowest mercury levels were measured from Silver Lake bass (mean $=94 \mathrm{ppb})$. Figure 4 displays mercury concentrations measured in individual bass.


Figure 4. Mercury Concentrations of Individual Bass Collected in 2010.

Fourteen percent of individual bass samples (7 out of 50) were above (did not meet) the EPA Recommended Criterion of 300 ppb . These bass came from Liberty and Loon Lakes. No individual bass samples exceeded the state water quality standard of 770 ppb .

## Fish Composites

Field crews collected additional fish species from Liberty, Loon, Potholes, Silver, and Spokane Lakes, and the Yakima River. Twenty-seven composites encompassing 11 species were analyzed for mercury. Figure 5 displays mercury concentrations measured in the composite samples. Complete composite fish data is included in Appendix C.


Figure 5. Mercury Concentrations of Composite Fish Samples Collected in 2010.
ND: Not detected at or above 18.3 ppb .
Species codes:
BBH: brown bullhead; BC: black crappie; BG: blue gill; BNT: brown trout;
LSS: largescale sucker; LWF: lake whitefish; MWF: mountain whitefish;
NPM: northern pikeminnow; TT: tiger trout; YP: yellow perch.

Ninety-six percent of composite samples contained quantifiable amounts of mercury. Only one sample, from Silver Lake, was below detection limits. Concentrations ranged from 18.3 U - 167 ppb. Lake Spokane northern pikeminnow had the highest mercury level among the composites. Black crappie and yellow perch from Silver Lake contained the lowest levels of mercury.

No composite samples exceeded the state regulatory criterion of 770 ppb or the EPA Recommended Criterion (300 ppb).

## Water

Water samples were collected from the epilimnions and hypolimnions of all six waterbodies. Analytes included alkalinity, sulfate, DOC, bromide, chloride, fluoride, and nitrite/nitrate. Field crews measured vertical profiles of dissolved oxygen, temperature, pH , and conductivity at each waterbody as well. The following figures display alkalinity, sulfate, and DOC results. Appendix C contains all water chemistry data, along with temperature, dissolved oxygen, and pH profiles.


Figure 6. Alkalinity Values Measured in the Epilimnion and Hypolimnion of the 2010 Waterbodies.


Figure 7. Sulfate Concentrations Measured from the Epilimnion and Hypolimnion of the 2010 Waterbodies.


Figure 8. Dissolved Organic Carbon (DOC) Concentrations Measured from the Epilimnion and Hypolimnion of the 2010 Waterbodies.
ND: Not detected above $1 \mathrm{mg} / \mathrm{L}$.

Alkalinity values ranged from $16-152 \mathrm{mg} / \mathrm{L}$ across the six waterbodies. The highest concentrations were found at Potholes Reservoir and the lowest at Silver Lake. Sulfate levels ranged from $0.45-20.5 \mathrm{mg} / \mathrm{L}$. Similar to alkalinity, Potholes Reservoir contained the greatest sulfate concentrations and Silver Lake the lowest. DOC concentrations were less variable, and most concentrations were below $5 \mathrm{mg} / \mathrm{L}$, with a range of $1 \mathrm{U}-22.2 \mathrm{mg} / \mathrm{L}$. The hypolimnion sample from Silver Lake had a particularly high DOC value, which appears to be an outlier.

Chloride was detected in all samples at low levels, while fluoride and nitrite/nitrate were detected in $50 \%$ of samples at low levels. Bromide was not detected above $0.2 \mathrm{mg} / \mathrm{L}$ in any of the samples.

Vertical temperature profiles indicated that Loon Lake, Lake Spokane, and Potholes Reservoir were stratified at the time of water sample collections (see Appendix C; Figure C-1). Liberty and Silver Lakes did not show stratification. Dissolved oxygen levels dropped below $1 \mathrm{mg} / \mathrm{L}$ in the bottom waters of Liberty Lake, Loon Lake, and Potholes Reservoir.

## Correlations

Simple linear regression coefficients were calculated to examine the relationship between bass mercury concentrations and fish size and age at the five waterbodies where bass were collected. All data were $\log _{10}$ transformed to improve normality. Table 4 presents the regression coefficients.

Table 4. Coefficients of Determination $\left(R^{2}\right)$ for Simple Linear Regressions of Mercury Concentrations with Fish Size and Age.

| Waterbody | Length | Weight | Age |
| :--- | :---: | :---: | :---: |
| Liberty | $\mathbf{0 . 7 6 5}$ | $\mathbf{0 . 7 9 7}$ | $\mathbf{0 . 9 3 2}$ |
| Loon | $\mathbf{0 . 6 0 8}$ | $\mathbf{0 . 5 8 0}$ | $\mathbf{0 . 7 0 4}$ |
| Potholes | $\mathbf{0 . 4 8 2}$ | 0.378 | $\mathbf{0 . 7 6 9}$ |
| Silver | $\mathbf{0 . 9 0 7}$ | $\mathbf{0 . 9 4 2}$ | $\mathbf{0 . 8 3 5}$ |
| Yakima | 0.326 | 0.3201 | 0.254 |

Bold values indicate statistically significant relationships ( $p<0.05$ ).

Bass mercury levels increased with fish length, weight, and age at all sites. Relationships were statistically significant at all sites except the Yakima River (length, weight, or age) and Potholes Reservoir (weight only).

Correlations were not calculated for water chemistry parameters collected in 2010, as only five data points (waterbodies) were available. Five data points lacks statistical power for a test such as correlation. In future years, correlations will be possible and these relationships can be evaluated. Correlations between fish mercury levels and water chemistry, lake morphology, and sediment mercury collected over the years 2005 - 2009 as part of this project are reported by Meredith et al. (2010).

## Temporal Trends

Data from the 2005 study were compared to 2010 data to determine differences between the collection years. The following sections discuss differences in bass lengths, bass mercury levels, and water chemistry.

## Bass Length and Mercury Values

Figure 9 displays individual bass mercury and length data from 2005 and 2010 at the five waterbodies where bass were collected.


Figure 9. Mercury to Fish Length Relationships of Individual Bass Collected 2005 and 2010.

Bass lengths captured in 2010 were similar to 2005 lengths, although 2010 Liberty Lake fish were generally longer than those caught in 2005. Length ranges overlapped at all waterbodies, which is important when comparing fish mercury levels. Length-to-weight relationships were similar between the two years at all waterbodies. Mercury concentrations increased with fish length at all waterbodies for both collection years.

## Analysis of Covariance

Figure 10 graphically displays results of the ANCOVA with Bonferroni post-hoc test. The bars in the graphs represent the estimated mean of $\log _{10}$ bass mercury evaluated at the mean $\log _{10}$ fish length of all data points for each waterbody. Complete statistical results from the ANCOVA and post-hoc test are provided in Appendix E.


Figure 10. Results of Analysis of Covariance and Post-Hoc Test Comparing Mercury Levels in Bass Between Years 2005 and 2010 Using Fish Length as a Covariate.
Bars represent the estimated marginal means for $\log _{10}$ bass mercury evaluated at the $\log _{10}$ length specified in each graph. Error bars represent $95 \%$ confidence intervals.

The ANCOVA results showed no significant change in bass mercury levels between 2005 and 2010 at four of the five waterbodies and a significant difference in Liberty Lake bass.

Liberty Lake mercury levels were significantly higher in 2010 than 2005, with a percent change between estimated mercury means of $+5.7 \%$. The higher mercury values could be reflecting higher laboratory bias in the 2010 analyses. As stated in the Data Quality section, LCS and SRM recoveries averaged $111 \%$ and 113\%, respectively, whereas LCS and SRM recoveries in 2005 averaged $98 \%$ and $102 \%$, respectively. Although the recoveries for the 2010 analysis were within quality objectives and the data were accepted without qualification, it is worth noting here because this type of statistical analysis is sensitive to differences in laboratory performance. Other factors, such as changes in mercury deposition to Liberty Lake's watershed or factors affecting methylation, could be responsible for the difference, but are outside the scope of this report.

The lack of significant differences at four of the lakes is not surprising given the relatively short time period between fish collections and the low mercury concentrations of the bass. The response time between reductions in mercury releases or emissions and decreased levels in fish tissue can vary greatly. Environmental factors such as mercury mobilization rates can result in time lags ranging up to decades or longer until a response in fish mercury levels are seen (Munthe et al., 2007). In a national workshop organized by EPA, mercury research scientists determined that 10-40 years of monitoring would be necessary to quantify mercury trends in aquatic biota across a range of ecosystems (EPA, 2008).

National studies have found very few temporal trends in fish mercury levels and in wet deposition over the recent time period of 1996-2005 (Chalmers et al., 2010; Prestbo and Gay, 2009). For most regions of the U.S., the greatest decreases in fish mercury levels were seen in the 1970s following sharp reductions in mercury emissions (Chalmers et al., 2010). In Washington, a significant decrease in mercury concentrations in rainwater and wet deposition was seen at a Seattle monitoring station after 1997 due to the closure of nearby medical waste incinerators (Prestbo and Gay, 2009). Since then levels have remained moderately low.

The waterbodies sampled in 2005 and 2010 contained bass with relatively low levels of mercury. Sixty-eight percent of the bass measured in 2005 were below the median mercury value (191 ppb) of all bass analyzed by the Mercury Trends project during the baseline period (2005 2009). Because of the low mercury levels in these waterbodies to begin with, substantial changes may not occur or it may take longer to see quantifiable differences. The detection of temporal trends may be more likely at other sites, as mercury levels were generally higher at the waterbodies sampled during 2006-2009.

## Water Samples

Water samples collected from the five waterbodies where bass were retained are compared in this section. Alkalinity and DOC were chosen as target analytes in 2006 and 2010 because of their potential relation to mercury bioaccumulation.

An inverse relationship between alkalinity and bass mercury levels was found among the 25 Mercury Trends sites over the first five years of monitoring (Meredith et al., 2010). This finding is consistent with other studies that have found elevated bass mercury levels associated with low alkalinity and pH (Hanten et al., 1998; Lange et al., 1993; Simonin et al., 2008). Increases in mercury levels under low-pH conditions have been explained by enhanced microbial production in acidic waters and/or increased bioavailability of mercury in low-pH conditions (Xun et al., 1987; Wiener et al., 1990).

Meredith et al. (2010) did not find a relationship between DOC and bass mercury levels, but other studies have shown that DOC levels can influence the transport and bioavailability of mercury (Driscoll et al., 1994).

Table 5 compares alkalinity and DOC values between the two sampling years. In general, most of the waterbodies had similar values for alkalinity and DOC in 2006 and 2010. Alkalinity values were slightly lower in the 2010 samples at all five sites except for Loon Lake. The Silver Lake epilimnion DOC value was slightly lower in 2010 as well.

Table 5. Water Sample Results from 2006 and 2010 Collections.

| Waterbody | Depth (m) |  | Alkalinity (mg/L) |  | DOC (mg/L) |  |
| :--- | :---: | :---: | :---: | :---: | :---: | :---: |
|  | '06 | '10 | '06 | '10 | '06 | '10 |
| Liberty | 2.5 | 2.5 | 24.5 | 22.8 | 3.9 | 3.2 |
|  | 6.5 | 6.5 | 24.3 | 22.9 | 3.8 | 3.2 |
| Loon | 4.0 | 4.0 | 86.8 | 86.4 | 3.4 | 4.4 |
|  | 20.0 | 22.0 | 86.7 | 89 | 3.1 | 3.9 |
| Potholes | 4.0 | 4.0 | 153 | 134 | 2.9 | 3.2 |
|  | 16.0 | 16.0 | 162 | 152 | 2.7 | 2.6 |
| Silver | 1.5 | 1.0 | 34 | 16.6 | 7.2 J | 4.5 |
|  | --- | 2.0 | --- | 16.0 | --- | 22.2 |
| Yakima | 1.5 | 0.5 | 119 | 104 | 1.5 | 1.5 |
|  | --- | 1.5 | --- | 104 | --- | 1.6 |

DOC: Dissolved organic carbon.

## Conclusions

Ecology analyzed 50 individual bass and 27 composites of additional fish species in 2010 as part of the sixth year of the Mercury Trends project. Fish were collected from the six waterbodies previously sampled in 2005 in order to determine changes in mercury levels over time. Targeted waterbodies were Liberty, Loon, Potholes, Silver, and Spokane Lakes, as well as the Yakima River. No bass were found at Lake Spokane. Water samples were also collected from each of the waterbodies and analyzed for parameters that may influence mercury bioaccumulation.

Results of this 2010 study support the following conclusions:

- Statistical analysis results showed no difference in bass mercury levels between 2005 and 2010 for four of the five waterbodies evaluated. Reasons for the lack of trends may include the short time period between sampling and the low mercury levels of the bass at these waterbodies.
- Results showed Liberty Lake bass collected in 2010 contained higher mercury levels than 2005 bass. The percent change in estimated $\log _{10}$ mercury means was $+5.7 \%$ from 2005 to 2010. Higher laboratory control sample (LCS) recoveries in the 2010 analysis may partially explain the difference between years.
- Individual bass mercury concentrations ranged from 49.3 to 483 ppb across the waterbodies. Liberty Lake contained the highest levels of mercury in bass, with an average of 301 ppb . These fish were also the longest. The lowest mercury levels were measured in Silver Lake bass (mean $=94 \mathrm{ppb}$ ).
- No bass samples exceeded the Washington State regulatory mercury criterion of 770 ppb . Fourteen percent of individual bass samples (7 out of 50) contained mercury levels above the EPA Recommended Criterion of 300 ppb . These fish came from Liberty and Loon Lakes. Composite fish samples did not exceed either criteria.
- Mercury levels in composite fish ranged from 18.3U - 167 ppb. Lake Spokane northern pikeminnow had the highest mercury level among the composites. Black crappie and yellow perch from Silver Lake contained the lowest levels of mercury.


## Recommendations

As a result of this 2010 study, the authors of this report recommend the following:

1. Continue the sampling regime of measuring bass mercury levels from waterbodies every five years. As no significant changes were found at four of the five waterbodies in 2010, we recommend at least three sampling cycles. Temporal trends in mercury levels of fish can take decades to detect. Three sampling events spaced five years apart may show differences that are not detectable with only two datasets.
2. Aim for tighter recoveries in laboratory quality control analyses. Differences in quality control test recoveries can indicate a laboratory bias that may influence temporal trend results. Consider changing the measurement quality objectives for laboratory control samples from $\pm 20 \%$ to $\pm 10 \%$.

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## Appendices

## Appendix A. Glossary, Acronyms, and Abbreviations

## Glossary

Alkalinity: The quantitative capacity of water to neutralize an acid.
Analyte: Water quality constituent being measured (parameter).
Anthropogenic: Human-caused.
Bioaccumulation: Progressive increase in the amount of a substance in an organism or part of an organism which occurs because the rate of intake exceeds the organism's ability to remove the substance from the body.

Biota: Flora (plants) and fauna (animals).
Bromide: The measure of the bromide ion in water.
Chloride: The measure of the chloride ion in water.
Clean Water Act: A federal act passed in 1972 that contains provisions to restore and maintain the quality of the nation's waters. Section 303(d) of the Clean Water Act establishes the TMDL program.

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

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

Dissolved oxygen (DO): A measure of the amount of oxygen dissolved in water.
Epilimnion: The uppermost layer of water in a lake where water temperature changes less than $1^{\circ} \mathrm{C}$ per one meter of depth.

Exceeded criterion: Did not meet or violated the criterion.
Fluoride: The measure of the fluoride ion in water.
Grab sample: A discrete sample from a single point in the water column or sediment surface.
Hypolimnion: The deepest layer of water in a lake where water temperature changes less than $1^{\circ}$ C per one meter of depth.

Morphology: Shape (e.g., channel morphology).
Nitrite/Nitrate: The measure of nitrite plus nitrate as nitrogen in water.
Otolith: Part of the inner ear of a fish. This structure is used to determine the age of a fish.
Parameter: Water quality constituent being measured (analyte).
$\mathbf{p H}$ : A measure of the acidity or alkalinity of water. A low $\mathbf{p H}$ value ( 0 to 7 ) indicates that an acidic condition is present, while a high $\mathrm{pH}(7$ to 14 ) indicates a basic or alkaline condition. A pH of 7 is considered to be neutral. Since the pH scale is logarithmic, a water sample with a pH of 8 is ten times more basic than one with a pH of 7 .

Sulfate: One of several minerals containing positive sulfur ions bonded to negative oxygen ions.
Temporal trends: Characterization of trends over time.
Thermocline: A temperature gradient in a thermally stratified, or temperature divided, body of water. Commonly associated with solar heating of the upper layers of a waterbody while the cooler layers remain on the bottom.

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

## Acronyms and Abbreviations

| ANCOVA | Analysis of covariance |
| :--- | :--- |
| CFR | Code of Federal Regulations |
| DOC | Dissolved organic carbon |
| DOH | Washington State Department of Health |
| EAP | Environmental Assessment Program |
| Ecology | Washington State Department of Ecology |
| EIM | Environmental Information Management database |
| EPA | U.S. Environmental Protection Agency |
| Hg | Mercury |
| LCS | Laboratory control sample |
| MEL | Manchester Environmental Laboratory |
| MQO | Measurement quality objectives |
| NTR | National Toxics Rule |
| PBT | Persistent, bioaccumulative, and toxic substance |
| RM | River mile |
| QA | Quality assurance |
| QC | Quality control |
| RPD | Relative percent difference |
| RSD | Relative standard deviation |
| SOP | Standard operating procedures |
| SRM | Standard reference materials |
| U | Not detected at the level indicated. |
| USGS | U.S. Geological Survey |
| WDFW | Washington Department of Fish and Wildlife |
| WRIA | Water Resource Inventory Area |

## Units of Measurement

| ${ }^{\circ} \mathrm{C}$ | degrees centigrade |
| :--- | :--- |
| ft | feet |
| g | gram, a unit of mass |


| $\mathrm{g} / \mathrm{d}$ | grams per day |
| :--- | :--- |
| m | meter |
| $\mathrm{mg} / \mathrm{Kg}$ | milligrams per kilogram (parts per million) |
| $\mathrm{mg} / \mathrm{L}$ | milligrams per liter (parts per million) |
| mL | milliliters |
| mm | millimeters |
| $\mathrm{ng} / \mathrm{g}$ | nanograms per gram (parts per billion) |
| ppb | parts per billion |
| $\mathrm{ug} / \mathrm{g}$ | micrograms per gram (parts per million) |
| $\mathrm{ug} / \mathrm{Kg}$ | micrograms per kilogram (parts per billion) |
| $\mathrm{ug} / \mathrm{L}$ | micrograms per liter (parts per billion) |
| $\mathrm{uS} / \mathrm{cm}$ | microsiemens per centimeter, a unit of conductivity |
| ww | wet weight |

## Appendix B. Site Descriptions

Table B-1. Detailed Site Descriptions, 2010 Waterbodies.

| Site Name | Latitude ${ }^{1}$ | Longitude ${ }^{1}$ | WBID ${ }^{2}$ | County | EIM "User Location" ID | WRIA | Location Description |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Liberty Lake | 47.6459 | -117.0776 | WA-57-9010 | Spokane | LIBERTY-F | 57 | Approx. 15 mi . E of Spokane |
| Lake Spokane | 47.8415 | -117.7249 | WA-54-9040 | Spokane | SPK 40.8 | 54 | Formerly "Long Lake", Spokane River (RM 40.8) |
| Loon Lake | 48.0536 | -117.6319 | WA-59-9130 | Stevens | LOON-F | 59 | 20 mi . S of Chewelah off Hwy 395 |
| Potholes Reservoir | 46.9813 | -119.3144 | WA-41-9280 | Grant | POTHOLES-F | 41 | 10 mi . SW of Moses Lake |
| Silver Lake | 46.2991 | -122.7702 | WA-26-9110 | Cowlitz | SILVERLNRCR-F | 26 | 12 mi . NE of Longview and 5 mi . E of Castle Rock |
| Yakima River | 46.3711 | -119.4364 | WA-37-1010 | Benton | YAKIMARABVHR-F | 37 | 12 mi . NW of Richland, above Horn Rapids Dam |

[^0]
## Appendix C. Data

Table C-1. Individual Bass Data.

| Lake | Species Code | Sample ID | Collection <br> Date | Total <br> Length (mm) | Weight <br> (g) | Age | Fulton's Fish Condition Index | Sex | Mercury (ppb) |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Loon <br> Lake | LMB | 1012025-01 | 10/25/10 | 379 | 918 | 6 | 1.69 | F | 169 |
|  |  | 1012025-02 |  | 498 | 2147 | 8 | 1.74 | F | 403 |
|  |  | 1012025-03 |  | 464 | 1680 | 9 | 1.68 | M | 311 |
|  |  | 1012025-04 |  | 399 | 1062 | 7 | 1.67 | M | 179 |
|  |  | 1012025-05 |  | 404 | 1275 | 8 | 1.93 | M | 292 |
|  |  | 1012025-06 |  | 452 | 1391 | 8 | 1.51 | F | 291 |
|  |  | 1012025-07 |  | 417 | 1205 | 7 | 1.66 | F | 176 |
|  |  | 1012025-08 |  | 390 | 940 | 6 | 1.58 | F | 184 |
|  |  | 1012025-10 |  | 358 | 655 | 7 | 1.43 | M | 228 |
|  |  | 1012025-11 |  | 371 | 708 | 6 | 1.39 | M | 179 |
| Liberty Lake | SMB | 1012025-12 | 10/26/10 | 445 | 1357 | 7 | 1.54 | M | 363 |
|  |  | 1012025-13 |  | 436 | 1223 | 6 | 1.48 | F | 346 |
|  |  | 1012025-14 |  | 439 | 1254 | 6 | 1.48 | M | 338 |
|  |  | 1012025-15 |  | 362 | 698 | 4 | 1.47 | M | 215 |
|  |  | 1012025-16 |  | 386 | 762 | 4 | 1.32 | F | 207 |
|  |  | 1012025-17 |  | 436 | 1119 | 6 | 1.35 | F | 268 |
|  |  | 1012025-18 |  | 387 | 772 | 4 | 1.33 | M | 184 |
|  |  | 1012025-19 |  | 383 | 854 | 4 | 1.52 | M | 180 |
|  |  | 1012025-20 |  | 426 | 1104 | 7 | 1.43 | F | 424 |
|  |  | 1012025-22 |  | 467 | 1578 | 8 | 1.55 | F | 483 |
| Yakima River | SMB | 1012025-23 | 10/27/10 | 325 | 518 | 3 | 1.51 | M | 208 |
|  |  | 1012025-24 |  | 282 | 345 | 2 | 1.54 | M | 125 |
|  |  | 1012025-25 |  | 274 | 262 | 2 | 1.27 | M | 195 |
|  |  | 1012025-26 |  | 355 | 655 | 3 | 1.46 | F | 191 |
|  |  | 1012025-27 |  | 240 | 166 | 2 | 1.20 | M | 109 |
|  |  | 1012025-28 |  | 257 | 219 | 2 | 1.29 | M | 94.5 |
|  |  | 1012025-29 |  | 280 | 311 | 2 | 1.42 | F | 125 |
|  |  | 1012025-30 |  | 373 | 747 | 4 | 1.44 | F | 139 |
|  |  | 1012025-31 |  | 295 | 360 | 2 | 1.40 | M | 115 |
|  |  | 1012025-32 |  | 280 | 294 | 2 | 1.34 | M | 114 |


| Lake | Species Code | Sample ID | Collection <br> Date | Total <br> Length (mm) | Weight <br> (g) | Age | Fulton's <br> Fish <br> Condition Index | Sex | Mercury (ppb) |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Potholes Reservoir | SMB | 1012025-33 | 10/19/10 | 250 | 181 | 2 | 1.16 | U | 103 |
|  |  | 1012025-34 | 10/20/10 | 347 | 605 | 2 | 1.45 | M | 87.4 |
|  |  | 1012025-35 |  | 338 | 575 | 2 | 1.49 | F | 67.8 |
|  |  | 1012025-36 |  | 478 | 1508 | 9 | 1.38 | M | 224 |
|  |  | 1012025-37 |  | 397 | 820 | 4 | 1.31 | F | 105 |
|  |  | 1012025-38 |  | 382 | 627 | 4 | 1.12 | F | 174 |
|  |  | 1012025-39 |  | 384 | 856 | 4 | 1.51 | M | 95.9 |
|  |  | 1012025-40 |  | 437 | 1255 | 8 | 1.50 | M | 184 |
|  |  | 1012025-41 |  | 413 | 974 | 4 | 1.38 | F | 153 |
|  |  | 1012025-42 |  | 480 | 1668 | 8 | 1.51 | F | 203 |
| Silver Lake | LMB | 1012025-43 | 9/13/10 | 510 | 2445 | 11 | 1.84 | F | 236 |
|  |  | 1012025-44 |  | 392 | 903 | 6 | 1.50 | F | 90.9 |
|  |  | 1012025-45 |  | 339 | 443 | 3 | 1.14 | M | 68.4 |
|  |  | 1012025-46 |  | 400 | 1057 | 6 | 1.65 | M | 117 |
|  |  | 1012025-47 |  | 373 | 763 | 4 | 1.47 | F | 101 |
|  |  | 1012025-48 |  | 320 | 413 | 3 | 1.26 | F | 81.4 |
|  |  | 1012025-49 |  | 233 | 187 | 2 | 1.48 | M | 49.3 |
|  |  | 1012025-50 |  | 265 | 263 | 3 | 1.41 | M | 49.7 |
|  |  | 1012025-51 |  | 270 | 280 | 2 | 1.42 | F | 53.3 |
|  |  | 1012025-52 |  | 340 | 599 | 3 | 1.52 | M | 88.1 |

LMB: largemouth bass.
SMB: smallmouth bass.

Table C-2. Composite fish data.

| Lake | Species Code | Sample ID | Collection Date | Average Total Length (mm) | Average Weight (g) | Fulton's Fish Condition Index | Number of Fish per Composite | Mercury (ppb) |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Loon Lake | BG | 1012025-55 | 10/25/10 | 174 | 101 | 1.87 | 3 | 70.6 |
|  | BG | 1012025-56 |  | 193 | 131 | 1.81 | 4 | 155 |
|  | TT | 1012025-53 |  | 392 | 471 | 0.78 | 3 | 27.5 |
|  | TT | 1012025-54 |  | 413 | 582 | 0.83 | 4 | 26.3 |
| Liberty Lake | BNT | 1012025-59 | 10/26/10 | 436 | 840 | 0.99 | 3 | 92.1 |
|  | BNT | 1012025-60 |  | 475 | 1019 | 0.95 | 3 | 131 |
|  | YP | 1012025-57 |  | 96 | 199 | 22.51 | 4 | 118 |
|  | YP | 1012025-58 |  | 114 | 209 | 14.91 | 4 | 98.1 |
| Yakima River | LSS | 1012025-61 | 10/27/10 | 290 | 259 | 1.06 | 4 | 68.8 |
|  | LSS | 1012025-62 |  | 310 | 309 | 1.04 | 4 | 68.8 |
|  | LSS | 1012025-63 |  | 326 | 366 | 1.05 | 4 | 66.8 |
| Lake Spokane | LSS | 1012025-65 | 10/26/10 | 453 | 1024 | 1.10 | 4 | 75.3 |
|  | LSS | 1012025-66 |  | 507 | 1507 | 1.16 | 5 | 76.8 |
|  | MWF | 1012025-64 |  | 268 | 186 | 0.96 | 3 | 23.9 |
|  | NPM | 1012025-67 |  | 387 | 500 | 0.86 | 3 | 106 |
|  | NPM | 1012025-68 |  | 431 | 762 | 0.92 | 4 | 167 |
| Potholes Reservoir | BBH | 1012025-72 | 10/20/10 | 254 | --- | --- | 5 | 30.6 |
|  | BBH | 1012025-73 |  | 318 | --- | --- | 4 | 26.7 |
|  | LWF | 1012025-69 |  | 358 | 500 | 1.09 | 4 | 46.9 |
|  | LWF | 1012025-70 |  | 475 | 1201 | 1.12 | 4 | 45.8 |
|  | LWF | 1012025-71 |  | 535 | 1928 | 1.26 | 3 | 55.4 |
| Silver Lake | BC | 1012025-77 | 9/13/10 | 174 | 81 | 1.52 | 5 | 18.7 |
|  | BC | 1012025-78 |  | 181 | 88 | 1.48 | 5 | 18.3 U |
|  | BC | 1012025-79 |  | 186 | 92 | 1.43 | 5 | 17.2 |
|  | YP | 1012025-74 |  | 153 | 44 | 1.22 | 5 | 25.0 |
|  | YP | 1012025-75 |  | 161 | 52 | 1.24 | 5 | 21.0 |
|  | YP | 1012025-76 |  | 172 | 58 | 1.14 | 5 | 20.0 |

U : Not detected at the level indicated.
Species:
BG: bluegill
TT: tiger trout
BNT: brown trout
YP: yellow perch
LSS: largescale sucker
MWF: mountain whitefish
NPM: northern pikeminnow
BBH: brown bullhead
LWF: lake whitefish
BC: black crappie


Figure C-1. Temperature Profiles for 2010 Lakes.


Figure C-2. Dissolved Oxygen Profiles for 2010 Lakes.


Figure C-3. pH Profiles for 2010 Lakes. No pH data available for Liberty Lake or Yakima River.

Table C-3. Water Data.

| Lake | Collection Date | Sample ID | Depth <br> (m) | Alkalinity (mg/L) | Bromide (mg/L) | Chloride (mg/L) | $\begin{gathered} \mathrm{DOC} \\ (\mathrm{mg} / \mathrm{L}) \end{gathered}$ | Fluoride (mg/L) | Nitrate/ Nitrite (mg/L) | Sulfate <br> (mg/L) |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Loon Lake | 8/10/2010 | $\begin{aligned} & 1008049-06 \\ & 1008049-07 \end{aligned}$ | $\begin{gathered} \hline 4.0 \\ 22.0 \end{gathered}$ | $\begin{gathered} \hline 86.4 \\ 89 \end{gathered}$ | $\begin{aligned} & 0.2 \mathrm{U} \\ & 0.2 \mathrm{U} \end{aligned}$ | $\begin{aligned} & 3.85 \\ & 3.75 \end{aligned}$ | $\begin{aligned} & 4.4 \\ & 3.9 \end{aligned}$ | $\begin{gathered} \hline 0.18 \\ 0.2 \end{gathered}$ | $\begin{gathered} \hline 0.01 \mathrm{U} \\ 0.067 \end{gathered}$ | $\begin{aligned} & 2.03 \\ & 1.97 \end{aligned}$ |
| Lake Spokane | 8/10/2010 | $\begin{aligned} & 1008049-01 \\ & 1008049-02 \end{aligned}$ | $\begin{gathered} \hline 2.5 \\ 20.0 \end{gathered}$ | $\begin{aligned} & 59.7 \\ & 93.8 \end{aligned}$ | $\begin{aligned} & 0.2 \mathrm{U} \\ & 0.2 \mathrm{U} \end{aligned}$ | $\begin{aligned} & \hline 3.08 \\ & 4.95 \end{aligned}$ | $\begin{aligned} & 1.2 \\ & 1 \mathrm{U} \end{aligned}$ | $\begin{aligned} & 0.1 \mathrm{U} \\ & 0.1 \mathrm{U} \end{aligned}$ | $\begin{gathered} 0.372 \\ 1.20 \end{gathered}$ | $\begin{aligned} & 7.22 \\ & 11.0 \end{aligned}$ |
| Potholes Reservoir | 8/12/2010 | 1008049-19 1008049-20 1008049-21 REP 1008049-22 REP | $\begin{gathered} \hline 4.0 \\ 16.0 \\ 4.0 \\ 16.0 \end{gathered}$ | $\begin{aligned} & 134 \\ & 152 \\ & 133 \\ & 151 \end{aligned}$ | $\begin{aligned} & 0.2 \mathrm{U} \\ & 0.2 \mathrm{U} \\ & 0.2 \mathrm{U} \\ & 0.2 \mathrm{U} \end{aligned}$ | $\begin{gathered} 6.89 \\ 6.94 \\ 6.9 \\ 6.79 \end{gathered}$ | $\begin{aligned} & 3.2 \\ & 2.6 \\ & 3.1 \\ & 2.5 \end{aligned}$ | $\begin{gathered} 0.33 \\ 0.28 \\ 0.29 \\ 0.3 \end{gathered}$ | $\begin{gathered} \hline 0.2 \\ 0.01 \mathrm{U} \\ 0.198 \\ 0.01 \mathrm{U} \end{gathered}$ | $\begin{aligned} & 20.4 \\ & 20.5 \\ & 20.5 \\ & 20.4 \end{aligned}$ |
| Silver Lake | 8/3/2010 | $\begin{aligned} & 1008048-01 \\ & 1008048-02 \end{aligned}$ | $\begin{aligned} & 1.0 \\ & 2.0 \end{aligned}$ | $\begin{aligned} & 16.6 \\ & 16.0 \end{aligned}$ | $\begin{aligned} & 0.2 \mathrm{U} \\ & 0.2 \mathrm{U} \end{aligned}$ | $\begin{aligned} & 2.71 \\ & 2.77 \end{aligned}$ | $\begin{gathered} 4.5 \\ 22.2 \end{gathered}$ | $\begin{aligned} & 0.1 \mathrm{U} \\ & 0.1 \mathrm{U} \end{aligned}$ | $\begin{aligned} & 0.01 \mathrm{U} \\ & 0.01 \mathrm{U} \end{aligned}$ | $\begin{aligned} & 0.46 \\ & 0.45 \end{aligned}$ |
| Liberty Lake | 8/11/2010 | 1008049-17 1008049-18 1008049-11 REP 1008049-12 REP | $\begin{aligned} & 2.5 \\ & 6.5 \\ & 2.5 \\ & 6.5 \end{aligned}$ | $\begin{aligned} & 22.8 \\ & 22.9 \\ & 23.1 \\ & 23.3 \end{aligned}$ | $\begin{aligned} & \hline 0.2 \mathrm{U} \\ & 0.2 \mathrm{U} \\ & 0.2 \mathrm{U} \\ & 0.2 \mathrm{U} \end{aligned}$ | $\begin{gathered} \hline 1.29 \\ 1.31 \\ 1.3 \\ 1.3 \end{gathered}$ | $\begin{aligned} & 3.2 \\ & 3.2 \\ & 3.2 \\ & 3.2 \end{aligned}$ | $\begin{aligned} & \hline 0.1 \mathrm{U} \\ & 0.1 \mathrm{U} \\ & 0.1 \mathrm{U} \\ & 0.1 \mathrm{U} \end{aligned}$ | $\begin{aligned} & 0.01 \mathrm{U} \\ & 0.01 \mathrm{U} \\ & 0.01 \mathrm{U} \\ & 0.01 \mathrm{U} \end{aligned}$ | $\begin{aligned} & 2.29 \\ & 2.28 \\ & 2.28 \\ & 2.31 \end{aligned}$ |
| Yakima River | 8/17/2010 | $\begin{aligned} & 1008050-12 \\ & 1008050-13 \end{aligned}$ | $\begin{aligned} & 0.5 \\ & 1.5 \end{aligned}$ | $\begin{aligned} & 104 \\ & 104 \end{aligned}$ | $\begin{aligned} & 0.2 \mathrm{U} \\ & 0.2 \mathrm{U} \end{aligned}$ | $\begin{aligned} & 6.64 \\ & 6.87 \end{aligned}$ | $\begin{aligned} & 1.5 \\ & 1.6 \end{aligned}$ | $\begin{aligned} & 0.18 \\ & 0.16 \end{aligned}$ | $\begin{aligned} & 0.921 \\ & 0.935 \end{aligned}$ | $\begin{aligned} & 14.3 \\ & 14.2 \end{aligned}$ |

U: Not detected at the level indicated.
REP: Replicate sample.

## Appendix D. Quality Control Data

MEL conducted mercury analyses of fish tissue samples from 12/21/10 through 1/07/11. All analyses were performed within EPA-established holding times. Table D-1 outlines the measurement quality objectives (MQOs) from the project plan (Seiders, 2006).

Table D-1. Measurement Quality Objectives for Fish Tissue Analyses.

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

## Fish Tissue Samples

Tables D-2 through D-5 present the results of quality control (QC) tests for fish tissue mercury analyses.

Table D-2. Mercury Matrix Spikes.

| Sample Number | Recovery <br> $(\%)$ | RPD <br> $(\%)$ |
| :--- | :---: | :---: |
| B10L142-MS1 | 88 | 13.8 |
| B10L142-MSD1 | 101 |  |
| B10L153-MS1 | 101 | 5.1 |
| B10L153-MSD1 | 96 | 88 |
| B11A010-MS1 | 8.4 |  |
| B11A010-MSD1 | 91 | 9.3 |
| B11A009-MS1 | 95 | 4.1 |
| B11A009-MSD1 | 99 |  |
| Mean | 94.9 | 6.6 |

Table D-3. Laboratory Control Samples.

| Sample Number | Recovery <br> $(\%)$ |
| :--- | :---: |
| B10L142-BS1 | 114 |
| B10L153-BS1 | 109 |
| B11A010-BS1 | 112 |
| B11A009-BS1 | 110 |

Table D-4. Standard Reference Materials.

| Sample Number | Recovery <br> (\%) |
| :---: | :---: |
| B10L142-SRM1 | 110 |
| B10L153-SRM1 | 117 |
| B11A010-SRM1 | 118 |
| B11A009-SRM1 | 113 |

Table D-5. Laboratory Blanks.

| Sample Number | Result <br> $(\mathrm{mg} / \mathrm{Kg})$ |
| :---: | :---: |
| B10L142-BLK1 | 0.0170 U |
| B10L153-BLK1 | 0.0170 U |
| B11A010-BLK1 | 0.0170 U |
| B11A009-BLK1 | 0.0170 U |

$U$ - undetected at the level indicated

## Water Samples

MEL conducted analyses of water samples in August 2010. All samples were received by MEL in good condition and analyzed within established holding times. Measurement quality objectives for water analyses are presented in Table D-6. Case narratives containing quality control test results for water analyses are available from the project manager by request.

Table D-6. Measurement Quality Objectives for Water Sample Analyses.

| Analyte | Matrix | LCS <br> (\% recov.) | Lab <br> Duplicates <br> (RPD) | Method <br> Blanks | Matrix <br> Spikes <br> (\% recov.) |
| :--- | :---: | :---: | :---: | :---: | :---: |
| DOC | water | $80-120 \%$ | $\leq 20$ | $<$ LOQ | $75-125 \%$ |
| Alkalinity | water | $80-120 \%$ | $<10$ | $<$ LOQ | N/A |
| Bromide | water | $90-110$ | $\leq 20$ | $<$ LOQ | $75-125$ |
| Chloride | water | $90-110$ | $\leq 20$ | $<$ LOQ | $75-125$ |
| Fluoride | water | $90-110$ | $\leq 20$ | $<$ LOQ | $75-125$ |
| Sulfate | water | $90-110$ | $\leq 20$ | $<$ LOQ | $75-125$ |
| Nitrite+Nitrate | water | $90-110$ | $\leq 20$ | $<$ LOQ | $75-125$ |

## Appendix E. Statistical Analyses

Table E-1 presents results for the analysis of covariance with Bonferroni post-hoc test results.
Table E-1. Analysis of Covariance Results.

| Waterbody | F-ratio | P value | Log $_{10}$ Fish <br> Length $^{\text {a }}$ | 2005 <br> Log $_{10} \mathrm{Hg}$ | 2010 <br> Log $_{10} \mathrm{Hg}$ | Standard <br> Error |
| :--- | :---: | :---: | :---: | :---: | :---: | :---: |
| Liberty | 8.0741 | 0.011 | 2.596 | 2.253 | 2.381 | 0.0285 |
| Loon | 0.2677 | 0.612 | 2.634 | 2.392 | 2.411 | 0.0237 |
| Potholes | 2.1878 | 0.157 | 2.581 | 2.010 | 2.106 | 0.0457 |
| Silver | 1.9525 | 0.180 | 2.527 | 1.840 | 1.923 | 0.0423 |
| Yakima River | 0.8634 | 0.366 | 2.504 | 2.111 | 2.184 | 0.0510 |

${ }^{\text {a }}$ Log10 fish length that the mercury value was evaluated at.
Hg : mercury.

## Appendix F. Fish Tissue Data Evaluation by Ecology and DOH

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

## Ecology

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

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

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

## DOH

While DOH supports Ecology's use of the NTR criteria for identifying problems and controlling pollutant sources so that water quality will meet standards, DOH does not use the NTR criteria to establish fish consumption advisories (Furl et al., 2007). DOH uses an approach similar to that in EPA's Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories Vol. 14 for assessing mercury, PCBs, and other contaminants (EPA, 2000). These guidance documents provide a framework from which states can evaluate fish tissue data to develop fish consumption
advisories based on (1) sound science and (2) established procedures in risk assessment, risk management, and risk communication. Neither the NTR criteria, nor the screening values found in the EPA guidance documents above, incorporate the varied risk management decisions essential to developing fish consumption advisories.

- Risk Assessment involves calculating allowable meal limits based on known fish contaminant concentrations. These calculations are conducted for both non-cancer and cancer criteria using the appropriate Reference Dose (RfD) or Cancer Slope Factor (CSF), if available. These initial calculations are the starting point for evaluating contaminant data to determine whether a fish advisory is warranted. Additionally, known or estimated consumption rates help determine the potential magnitude of exposure and highlight the sensitive groups or populations that may exist due to elevated consumption rates.
- Risk Management includes (but is not limited to) consideration of contaminant background concentrations, reduction in contaminant concentrations through preparation and cooking techniques, known health benefits from fish consumption, contaminant concentrations or health risks associated with replacement foods, and cultural importance of fish. Other considerations are the possible health criteria associated with a contaminant, the strength or weaknesses of the supporting toxicological or sampling data, and whether effects are transient or irreversible.
- Risk Communication is the outreach component of the fish advisory. The interpretation of the data from the risk assessment and risk management components drives how and when the fish advisory recommendations are issued to the public dependent on whether the message is targeted toward a sensitive group or a population or the general public. DOH's dual objective is (1) how best to provide guidance to the public to increase consumption of fish low in contaminants to gain the benefits of eating fish, while (2) steering the public away from fish that have high levels of health-damaging contaminants.


[^0]:    ${ }^{1}$ North American Datum 1983. Coordinates for fish tissue samples are in the central part of the lake, while fish were usually collected from many areas of the lake.
    ${ }^{2}$ Ecology's Water Body Identification Number (WBID)
    ${ }^{3}$ Site identification as used in Ecology's Environmental Information Management (EIM) system.

