Measuring Mercury Trends in Freshwater Fish in Washington State: 2007 Sampling Results

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Measuring Mercury Trends in Freshwater Fish in Washington State: 2007 Sampling Results

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Waterbody Numbers: see Appendix A

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List of Acronyms

Following are acronyms and abbreviations used frequently in this report.

DOC	Dissolved Organic Carbon
DOH	Washington State Department of Health
Ecology	Washington State Department of Ecology
EPA	U.S. Environmental Protection Agency
MEL	Manchester Environmental Laboratory
MQO	Measurement Quality Objectives
NTR	National Toxics Rule
QA/QC	Quality Assurance/Quality Control
RPD	Relative Percent Difference
RSD	Relative Standard Deviation
TOC	Total Organic Carbon

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Abstract

This report presents results from the third year of a long-term monitoring effort by the Washington State Department of Ecology to measure mercury trends in resident freshwater fish tissue. Six sites per year for five years (30 sites total) are assessed to characterize trends over time (temporal trends).

In 2007, 60 individual largemouth bass and 32 composite samples representing eight species were analyzed from Deer Lake, Lake Fazon, Lower Goose Lake, Lake Ozette, Lake Samish, and Lake St. Clair. Water and sediment samples were also collected to evaluate selected parameters that may influence mercury uptake in fish tissues.

Seventy-three percent of individuals and 28% of composites sampled exceeded the Environmental Protection Agency's recommended water quality criterion of 300 ppb. A single four-year-old female bass from Lake Ozette contained a mercury concentration of 1800 ppb. This sample was one of seven exceeding the National Toxics Rule human health criterion of 825 ppb. This sample had the highest concentration recorded in a largemouth bass during the first three years of this long-term monitoring study.

A temporal analysis was performed for three lakes (Deer, Fazon, and Samish) sampled in 2001-2002 and again in 2007. Time between sampling events ranged from 58-72 months. Results from Deer Lake estimated a 15% decrease in mercury concentrations for fish at a given length. Estimated changes in concentration were small at Samish and Fazon Lakes.

Mercury concentrations in standard-sized bass from the first three years of the project were compared through a t-test to determine if concentrations from eastern and western Washington differed. The test showed a significant difference between the two areas with a higher average concentration among western Washington waterbodies (294 ppb to 126 ppb).

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Introduction

Background

While mercury is a naturally occurring substance, human activity has greatly increased the release of mercury into the environment. Consequences of this include increased health risks to humans and animals due to the persistent, bioaccumulative, and toxic nature of this substance. Concerns about these risks have led governments at international, national, state, and local levels to recognize and address the problems associated with the use and disposal of mercury.

Mercury is widespread in the environment, being released to the atmosphere from varied sources and transported globally. Natural sources of mercury include weathering of mercury-bearing rocks and soil, volcanic activity, forest fires, and degassing from water surfaces. Anthropogenic (human-caused) sources include combustion of fossil fuels, metals production, and industrial processes. Lake sediment records show that atmospheric mercury has tripled over the last 150 years suggesting that two thirds of atmospheric mercury is of anthropogenic origin (Morel et al., 1998; Mason et al., 1994).

In humans, mercury can affect the nervous system, with children and developing fetuses being most at risk (EPA, 2000). Concern with these health risks resulted in the 2002 Washington State Legislature funding the Washington State Departments of Ecology (Ecology) and Health (DOH) to develop a plan targeting mercury as the first chemical in the state's *Proposed Strategy to Continually Reduce Persistent, Bioaccumulative Toxins (PBTs) in Washington State* (Gallagher, 2000).

The *Washington State Mercury Chemical Action Plan* (Peele, 2003) was developed in 2003 by Ecology and DOH. This plan summarized current information on mercury in Washington and made recommendations for reducing mercury emissions in Washington.

Previous Studies on Mercury in Washington

Several studies have described the extent and severity of mercury contamination in fish and sediments throughout Washington, many of which led to issuance of fish consumption advisories.

Mercury Trends Monitoring

Furl (2007a) examined individual and composite samples among a variety of fish species during the second year of the current study. A total of 17% of individuals and 3% of composites sampled exceeded the U.S. Environmental Protection Agency (EPA) recommended water quality criterion of 300 ppb. A single nine-year-old female bass from Mason Lake contained a mercury concentration of 952 ppb. It was the only sample exceeding the National Toxics Rule (NTR) of 825 ppb (CFR, 2004; EPA, 2001).

Furl et al. (2007) examined mercury in individual bass as part of the first year of the current study. Mercury levels were within typical ranges (0-300 ppb) of previous fish tissue studies conducted within the state. Less than 10% of samples exceeded the EPA recommended criterion (300 ppb), and no samples exceeded the NTR criterion (825 ppb).

Furl (2007b and 2008) examined mercury concentrations in age-dated sediment cores from Loon Lake, Wannacut Lake, Walupt Lake, Lake Ozette, Lake Sammamish, and Lake St. Clair. Recent flux rates in the upper most horizons of the cores ranged from 3-259 ug/m²/yr with higher fluxes found in western Washington. The studies found flux rates have *generally* declined in the upper most horizons of the sediment cores.

Statewide Bass Study

Fischnaller et al. (2003) examined mercury in 185 bass and sediment from 20 sites across Washington. Samples of muscle tissue from bass confirmed that elevated levels of mercury are prevalent across Washington. Many fish exceeded one or more criteria for protection of human health. About 23% of fish representing 14 of 20 sites exceeded the EPA criterion (300 ppb). A single ten-year old fish from Lake Samish had a muscle tissue mercury level of 1280 ppb, exceeding the NTR criterion (825 ppb). The study recommended implementing a long-term monitoring plan for mercury in fish and was the basis of DOH's issuance of a statewide fish consumption advisory for largemouth and smallmouth bass (McBride, 2003).

Lake Whatcom Studies

Norton (2004) investigated mercury levels in surface water, surficial sediments, and sediment cores of Lake Whatcom, in cooperation with the U.S. Geological Survey and the Whatcom County Health Department (see Paulson 2004, below). Findings suggest that mercury levels began increasing around 1900, may have peaked in the late 1990s, and appear to be declining. This study recommended that mercury levels in fish from Lake Whatcom be monitored periodically to determine if mercury levels decline over time. This study also recommended monitoring bottom waters for methylmercury and total mercury to help evaluate compliance with water quality target concentrations in the lake and to prevent excessive bioaccumulation of mercury in fish.

Paulson (2004) examined sources of mercury in sediments, water, and fish for 8 lakes in Whatcom County. An atmospheric deposition model was developed to allow comparison of deposition patterns in the lakes sampled. Mercury emissions from known sources in the area (e.g., waste incinerators, a sewage-sludge incinerator, a chlor-alkali plant) were modeled as part of this effort. Relationships between point source deposition and mercury concentrations in bass could not be established.

Serdar et al. (2001) examined mercury concentrations in 273 fish from 6 finfish and one crayfish species in Lake Whatcom. Mercury levels were elevated in smallmouth bass. These data were used in development of a fish consumption advisory for Lake Whatcom (Lake Whatcom Cooperative Management Program, 2001). Serdar et al. (2001) recommended a monitoring program to routinely characterize mercury levels in fish throughout Washington.

A frequency distribution of all fish tissue mercury data (n = 1712) located in Ecology's Environmental Information Management (EIM) database is included in Figure 1.



Figure 1. Frequency Distribution of all Mercury Concentrations in Freshwater Fish Available in EIM (accessed July, 28 2008).

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Study Design

Goal and Objectives

In 2005, the Legislature began funding a long-term mercury monitoring program in Washington. This project included two components:

- Determine mercury levels in edible tissue from ten individual fish of the same species (bass and/or walleye) from 6 sites per year for long-term trend characterization. Sampling at each of these sites will be repeated every five years such that a total of 30 sites will be sampled over a five-year period.
- Collect sediment cores from 3 lakes per year to assess depositional history of mercury in Washington.

The sediment coring effort began in 2006.

Additional objectives of the fish tissue component include:

- Collect ancillary data on the sites where fish were collected to better understand patterns, dynamics, and changes in fish tissue mercury levels over space and time. Ancillary data will include:
 - Fish length, weight, sex, and age.
 - Lake morphological and hydrological characteristics.
 - Alkalinity, dissolved organic carbon, and chlorophyll concentrations from top and bottom waters; vertical profiles of temperature, dissolved oxygen, conductivity, and pH.
 - Three surficial sediment grabs analyzed for mercury, total organic carbon, and grain size.
- Determine mercury concentrations in composite samples from two other fish species that are present at the sites where bass and/or walleye are collected. For each species, three composite samples consisting of 3-5 fish will be collected. This objective is intended to aid DOH in crafting more informative recommendations for fish consumption advisories.

Site Information

Figure 2 displays the 2007 study lakes: Deer Lake, Lake Fazon, Lower Goose Lake, Lake Ozette, Lake Samish, and Lake St. Clair. Fish were collected in August and September, 2007.

Lakes were selected considering numerous criteria including: proximity to known mercury sources, popularity among anglers, availability of target fish species, and inclusion in the Fischnaller et al. (2003) mercury screening study.

Table 1 gives more information for each of these sites. The project plan discusses complete site selection considerations (Seiders, 2006).



Figure 2. 2007 Study Lakes.

Name	Deer	Fazon	Lower Goose	Ozette	Samish	St. Clair
County	Stevens	Whatcom	Grant	Clallam	Whatcom	Thurston
Drainage (sq mi)	18.2	0.97		77.5	9.2	14.5
Altitude (ft)	2474	128	856	29	273	73
Surface Area (acres)	1100	31	50	7300	680	88
Lake Volume (acre-ft)	57,000	300	1,300	960,000	24,000	3,600
Maximum depth (ft)	75	17	75	320	75	110
Mean Depth (ft)	52	10	25	130	31	40

Table 1. 2007 Study Lakes Location and Physical Information.

Methods

Sample Collection

In all, 180 fish encompassing 8 different species were collected from the 2007 study lakes. Sixty individual fish along with 32 composite samples were analyzed by Manchester Environmental Laboratory (MEL) for total mercury concentrations. Collection goals for each waterbody, as outlined in the project plan (Seiders, 2006), were 10 individual bass or walleye for individual analysis, 3 composite samples of 3-5 fish for 2 additional species, 2 water samples, and 3 surface sediment grab samples. Collection goals were met at all sites with the exception of composite fish samples from Deer Lake. Only one composite sample of 2 additional species was retained from the lake. Detailed information on all fish collected is included in Appendix C.

Field Procedures

Fish

The collection, handling, and processing of fish tissue samples for analysis were guided by methods described in the EPA's *Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories* (EPA, 2000) and Ecology's Environmental Assessment Program's *Standard Operating Procedures for Resecting Finfish Whole Body, Body Parts or Tissue Samples* (Sandvik, 2006). Fish were collected by Ecology crews using boat electrofishing and gill netting.

Fish were inspected to ensure that they were acceptable for further processing (e.g., no obvious damage to tissues, skin intact). Acceptable fish were euthanized by a blow to the head with a dull object, rinsed in ambient water to remove foreign material from their exterior, weighed to the nearest gram, and their total lengths measured to the nearest millimeter. Individual fish were then double-wrapped in foil and placed in a plastic zip-lock bag along with a sample identification tag. The bagged specimens were placed on ice in the field. Fish remained on ice for a maximum of 24-72 hours and then were frozen and held at -20° C at Ecology facilities in Lacey, Washington for processing at a later date.

For processing, fish were removed from the freezer, partially thawed, slime and scales removed, rinsed in tap water, and followed by a rinse in deionized water. Fish were then filleted with the skin left on and cut into small cubes. The tissue was passed three times through a Kitchen-Aid food grinder and homogenized by stirring to a consistent texture and color. Subsamples from the homogenate were taken and placed into previously cleaned 2 or 4 ounce glass containers (I-Chem 200[®]). Sample jars were assigned a laboratory identification number and transported to the laboratory for analyses. Excess homogenate was placed in an appropriate container, labeled, and archived frozen at -20° C.

After fillets were removed, the sex of the fish was determined, when possible, and recorded. Otoliths and scales were removed from fish that were analyzed individually and sent to Washington Department of Fish and Wildlife (WDFW) biologists to determine age. All utensils used for processing tissue samples were cleaned to prevent contamination of the sample. Utensils included stainless steel bowls and knives and tissue grinding appliances having plastic, wood, bronze, and stainless steel parts. All utensils for fish tissue sampling were cleaned with the following procedure: hand washed with soap (Liquinox) and hot water, hot tap water rinse, 10% nitric acid rinse, and a final deionized water rinse. Utensils were air-dried and wrapped in aluminum foil until used. Fish were filleted and tissues processed on the dull side of heavy-duty aluminum foil covering a nylon cutting board laid on the workbench. Each fish was processed on a new/clean sheet of aluminum foil with cleaned utensils to prevent contamination from one sample to the next.

Sediment

The collection, handling, and processing of sediment samples were guided by Puget Sound Estuary Protocol (PSEP, 1986). Profundal sediment samples were collected with a single grab using a 0.02 m² stainless steel petite ponar. The overlying water was siphoned away, and the top two centimeters were removed with a stainless steel spoon. Sediments coming in contact with the side of the ponar device were not retained.

Sub-samples were homogenized on the boat using stainless steel bowls and spoons and then placed in pre-cleaned jars according to MEL protocol (MEL, 2005). Samples were packed in ice and shipped to MEL within 96 hours. All utensils used to collect and prepare samples were cleaned in the same manner as utensils used in fish tissue processing.

Water

Two water samples were obtained at the deepest part of the lake using a one-liter Kemmerer sampler. The samples were obtained at the mid-points of the hypolimnion¹ and epilimnion² in stratified lakes. At well-mixed 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, chlorophyll, and DOC.

Conductivity, pH, dissolved oxygen, and water temperature were measured at the water sample sites using a Hydrolab[®] following Ecology standard operating procedures (Swanson, 2007). All units were calibrated prior to field use, and Winkler titrations were performed as a measure of quality control for the dissolved oxygen readings.

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

² The uppermost layer of water in a lake where water temperature changes less than 1° C per one meter of depth.

Laboratory Procedures

All samples were analyzed at MEL with the exception of grain size which was performed by Analytical Resources Inc. Table 2 contains information on the analytical methods used to perform laboratory analysis.

Analyte	Matrix	Method
Mercury	Tissue	CVAA, EPA 245.6
Mercury	Sediment	CVAA, EPA 245.5
TOC	Sediment	PSEP-TOC
Grain Size	Sediment	PSEP, Sieve and Pipette
Alkalinity	Water	SM2320B
DOC	Water	EPA 415.1
Chlorophyll	Water	SM10200H3M
TOC - Total Ora	ania Carbon	CUAA - Cold Vener Atom

Table 2. Analytes and Analytical Methods.

TOC = Total Organic CarbonCVAA = Cold Vapor Atomic AbsorptionDOC = Dissolved Organic CarbonPSEP = Puget Sound Estuary Protocol

In 2005, Ecology switched laboratory methods for analyzing mercury in fish tissues from method EPA 245.5 to EPA 245.6. A study was conducted (Furl, 2007c) comparing the two analytical methods, and method 245.5 was found to under report mercury levels by 25 – 38% varying with magnitude of concentration. Data collected for the mercury screening study (Fischnaller et al., 2003) were measured using method EPA 245.5. Results used from Fischnaller et al. (2003) in the current report are adjusted data and qualified as estimates.

Total mercury as opposed to methylmercury has been the target analyte used in other fish tissue studies in Washington due to the relative simplicity and lower cost. Methylmercury, the bioaccumulative and toxic form of mercury in fish tissue, accounts for more than 95% of the mercury in fish tissue where it is associated with muscle proteins (Bloom, 1995; Driscoll et al., 1994).

Data Quality Assessment

Results from MEL included a case narrative (Momohara, 2007) describing results from the quality control and quality assurance procedures used during analyses. These results included: holding times, instrument calibration, method blanks, matrix spikes, laboratory duplicates, laboratory control samples, and Standard Reference Material (SRM) 1946 (Lake Superior fish tissue) from the National Institute of Standards and Technology.

The quality assessment indicated all sediment and fish tissue data met measurement quality objectives outlined by the project plan. Several water samples (DOC) were qualified as estimates due to elevated reporting limits resulting from difficulty during instrument calibration. Data quality summaries describing laboratory duplicates, matrix spikes, and SRM analyses can be found in Appendix B.

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Results

Summary statistics of the fish, sediment, and water samples collected in 2007 are described below. Complete results are located in Appendix C.

Fish

Individual Largemouth Bass

Table 3 contains summary statistics of the physical characteristics for the largemouth bass collected from each lake, and Figure 3 displays mercury concentrations with age noted at the bottom of each bar.

Lake	Statistic	Total Length (mm)	Weight (g)	Age (yr)	Mercury (ppb)
	Mean	383	871	5.8	335
Deer	Std. Dev.	50	358	2.8	141
Deer	Minimum	315	444	3.0	190
	Maximum	454	1445	10.0	586
	Mean	382	855	6.0	386
Foren	Std. Dev.	43	313	0.9	59
Fazon	Minimum	319	372	5.0	317
	Maximum	456	1380	8.0	525
	Mean	400	1047	4.1	319
Lower Coose	Std. Dev.	21	136	0.7	Mercury (ppb) 335 141 190 586 386 59 317 525 319 64 209 389 715 474 350 1800 344 191 130 637 423 226 219 954 420 266 130 130
Lower Goose	Minimum	370	810	3.0	209
	Maximum	435	1245	5.0	389
	Mean	342	594	3.3	715
Ozatta	Std. Dev.	50	261	0.8	474
Ozelle	Minimum	246	207	2.0	350
	Maximum	415	1080	5.0	1800
	Mean	354	791	4.1	344
Samish	Std. Dev.	74	446	2.0	191
Samisii	Minimum	251	228	2.0	130
	Maximum	457	1451	8.0	637
	Mean	345	641	6.4	423
St. Clair	Std. Dev.	71	415	4.8	335 141 190 586 386 59 317 525 319 64 209 389 715 474 350 1800 344 191 130 637 423 226 219 954 420 266 130 1800
St. Clair	Minimum	274	254	3.0	219
	Maximum	452	1257	17.0	954
	Mean	368	799	5.0	420
All Lakas	Std. Dev.	57	357	2.7	266
All Lakes	Minimum	246	207	2.0	130
	Maximum	457	1451	17.0	1800

Table 3. S	Summary	Statistics	for	Individual	Largemouth	Bass (n	= 10 per	lake).
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Std. Dev. - standard deviation



Figure 3. Mercury Concentrations and Age of Individual Largemouth Bass.

Mercury concentrations in largemouth bass ranged from 130 ppb (Lake Samish) to 1800 ppb (Lake Ozette). Seventy-three percent (n = 44) of the individual largemouth bass exceeded EPA's recommended mercury criterion of 300 ppb. Seven percent (n = 4) exceeded the NTR criterion of 825 ppb.



Figure 4 is a boxplot graphically displaying the normality (minimum, 25th percentile, median, 75th percentile, and maximum) of mercury concentrations for the individual bass.

Figure 4. Boxplots of Mercury Concentrations in Individual Bass.

Distribution and variance of concentrations varied widely among lakes, with Lake Ozette and Lake St. Clair containing the widest range of concentrations. Boxplots displaying distribution of weight and length for individual bass are located in Appendix D.

Size Range

Target size ranges for individual bass were determined by considering historical data, usefulness for long-term monitoring, angler-preferred size ranges, and fishing regulations. The target size range is expressed in (1) total length of an individual fish (250 to 460 mm, or about 10 to 18 inches) and (2) terms of the spread or range of the group of fish collected: the length of the smallest fish should be at least 75% the length of the largest fish (Seiders, 2006).

Figure 5 displays the size ranges for the individual bass. Above the bars is the length of the smallest fish expressed as a percentage of the largest fish for each lake.



Figure 5. Total Lengths of Individual Fish Used for Trends Monitoring.

While all fish collected were within the targeted size range (250-460 mm), only Lower Goose Lake met both length criteria. This should be considered when examining summary statistics and box plots for tissue concentrations as mercury has been shown to vary with length (e.g., Furl et al., 2007; Fischnaller et al., 2003; Serdar et al., 2001).

Composite Fish Samples

In addition to individual largemouth bass, composite samples consisting of 2 additional species were collected at each site. Mercury concentrations for the composites along with the species code are graphed in Figure 6. Physical data for the fish used in the composites, along with the number of fish in the composite sample, can be found in Appendix C.



Figure 6. Mercury Concentrations in Composite Samples.

Mercury concentrations in the composite samples varied from 25 ppb (Deer Lake) to 1920 ppb (Lake Ozette). Lower Goose Lake and Lake Samish both contained samples exceeding the EPA recommended criteria along with Lake Ozette which contained samples above the NTR criterion. Excluding northern pike minnow, species examined in the composites were generally lower than largemouth bass samples from the same lake.

Sediment

Three sediment grab samples were obtained from each study lake including 3 additional replicates taken at Ozette and St. Clair. Sediment analysis included: mercury, TOC, and grain size. Figure 7 displays average mercury results in sediments. Grain size and TOC averages are located in Figure 8. Average sediment data for Ozette and St. Clair do not include replicate analyses.

Mercury concentrations, grain size composition, and TOC levels varied widely between lakes. Average mercury concentrations ranged from 25 ppb (Lower Goose Lake) to 297 ppb (Lake St. Clair). TOC averaged 9.5%, and average grain size was 69% fine grained material (< 62u) across all lakes. Statistical information examining variance among sampling sites within each lake are included in Appendix B.



Figure 7. Average Mercury Concentrations in Sediments.



Figure 8. Grain Size (% Fines < 62u) and Total Organic Carbon (%).

Water

Upper and lower water grab samples were taken from each of the study lakes, including replicates from Lake Ozette and Lake St. Clair. Results are located in Table 4. Replicate samples are not included in Table 4.

Lake	Collection Date	Depth (m)	Chl-a (ug/L)	Alkalinity (mg/L)	DOC (mg/L)
Door	7/31/2007	3.5	0.91 U	41	4.0
Deel	7/31/2007	14.0	1.9 U	39	4.0
Foren	7/25/2007	0.5	3.4 J	50.4	18 J
Fazon	1/23/2007	3.0	97.3	52.4	16.0
Lower Coose	7/20/2007	3.0	3.9	86.6	1.6
Lower Goose	7/30/2007	16.0	1.6 U	177	2.5
Ozotto	7/24/2007	10.0	0.89	6.4	3.6
Ozelle	7/24/2007	35.0	0.69	6.2	3.6
Somiah	7/26/2007	3.0	2.2	18	1.9 J
Samish	1/20/2007	14.0	2.9	18	1.7 J
St. Cloin	7/22/2007	2.0	6.9	50.8	4.2
St. Clair	1/25/2007	25.0	1.6	42	4.0 18 J 16.0 1.6 2.5 3.6 3.6 1.9 J 1.7 J 4.2 8.4

Table 4. Upper and Lower Water Grabs.

U = Not detected at detection limits shown

 $\mathbf{J} = \mathbf{Estimated}$

Dissolved oxygen and temperature profiles were measured 1-2 times during a single day at all 6 lakes during the last week of July 2007, using a Hydrolab©. Vertical profiles for both parameters are included in Figure 9.

Temperature profiles revealed distinct thermoclines³ at all 6 study lakes. Low dissolved oxygen levels (< 2 mg/L) existed in bottom waters at Fazon Lake, Lower Goose Lake, and Lake Samish. Dissolved oxygen levels at Lake Ozette were only measured to a depth of 48 meters (maximum depth \approx 100m).

 $^{^{3}}$ Thermocline – a layer of water where there is an abrupt change in temperature that separates the warmer surface water from the colder deep water.

Temperature



Dissolved Oxygen



Figure 9. Dissolved Oxygen and Temperature Profiles for the 2007 Study Lakes.

Discussion

Relationships of Mercury Concentrations and Fish Size and Age

Mercury concentrations were regressed against length, weight, and age using simple linear regression to determine the amount of variability explained by each of the physical characteristics. Results are displayed in Table 5 and scatterplots are located in Appendix D. Positive relationships between mercury concentrations and fish size and age have been well established and previously documented in Washington State mercury reports (Furl et al., 2007; Fischnaller et al., 2003; Serdar et al., 2001).

Lake	Length	r ² Weight	Age
Deer	0.83	0.80	0.87
Fazon	0.17	0.17	0.16
Lower Goose	0.34	0.38	0.54
Ozette	0.49	0.53	0.59
St. Clair	0.74	0.66	0.95
Samish	0.74	0.71	0.75

Table 5. Coefficients of Determination for Linear Regressions (bolded values indicate p > 0.10).

Each of the physical parameters explained at least 30% of the variance in mercury concentrations (generally > 50%) with the exception of Fazon Lake where p values were greater than 0.10. On average, age had the highest coefficient of determination ($r^2 = 0.64$) followed by length and weight respectively.

Standard-Sized Fish and Factors Affecting Bioaccumulation

Multiple regression analysis was used to derive mercury concentrations for a standard-sized fish to allow for direct comparisons between lakes after fish length was considered (Figure 10). Length was used as the predictive variable as opposed to age due to ease of measurement in the field. The same technique was used in previous Washington State mercury reports (Furl et al., 2007; Furl, 2007; and Fischnaller et al., 2003).

A standard-sized (356 mm) fish was estimated by calculating the following multiple regression formula:

$$Log_{10}(Hg) = M + \{B1 * Log_{10}(356 mm)\} + \{B2 * (Log_{10}(356 mm))^2\}$$
$$10^{Log_{10}(Hg)} = Hg Concentration at 356mm$$

The regression formula was also calculated for a fish size of 306 and 406 mm to provide insight on rates of mercury accumulation based on length (slope of the regression line). These values

are represented as the lower (306 mm fish size) and upper (406 mm fish size) tails of error bars in Figure 9.



J = Length did not serve as an adequate predictor of mercury concentration (p > 0.10). Concentrations were estimated from data associated with lengths of 356 mm \pm 20 mm.

Figure 10. Projected Mercury Concentrations for a 356 mm Bass.

Regression coefficients (M, B1, B2), products, and standardized mercury concentrations are listed in Appendix D for each lake. Loon Lake, Long Lake, Liberty Lake, and the Yakima River (2005 study) and Lake Offut (2006 study) were estimated by extrapolating from existing mercury data because fish length did not serve as an adequate independent variable in the regression analysis (p > 0.10).

Estimated mercury concentrations for standard-sized bass were elevated in 2007 lakes when compared to the 2 previous study years. With the exception of Lower Goose Lake, all 2007 study lakes contained higher standard-sized concentrations than any lake examined in 2005 and 2006. Lake Ozette standard-sized bass contained the highest amount of mercury (648 ppb) calculated at mercury trends sites during the first 3 years of monitoring. A dot histogram with a kernel smoother was constructed using the standard-sized concentrations from each of the mercury trends waterbodies (n = 17) to examine the normality of concentrations (Figure 11). Results show a fairly normal distribution amongst standard-sized concentrations with the exception of Lake Ozette.



Figure 11. Dot Histogram with Kernel Smoother Displaying Estimated Mercury (Hg) Concentrations in Standard-sized 356 mm Bass. Lake Ozette is represented by the red circle.

Colman et al., (2008) collected 7 largemouth bass from Lake Dickey (5 miles east of Ozette) in 2007 as part of a nationwide mercury study. Elevated tissue concentrations of similar magnitude to Lake Ozette were recorded. The regression equation was applied to the Lake Dickey fish, and an estimated mercury concentration of 621 ppb was calculated for a 356 mm bass. The anomalous values recorded at Ozette and Dickey are difficult to reconcile considering their remote locations far from point⁴ source pollution. Both watersheds are heavily logged (Ritchie, 2008); therefore high sedimentation rates within the basins may be contributing mercury to the lakes from their catchments.

Correlations

A correlation matrix was produced using Spearman Rank correlation to evaluate relationships between 14 physical and chemical lake variables and mercury concentrations in a standard-sized (356 mm) bass. Spearman Rank is a non-parametrical test (used when normality of the data isn't known) which ranks data in order of increasing value before calculating coefficients. All mercury monitoring lakes displayed in Figure 9 were included in the analysis except for Offut Lake where no ancillary data outside of fish tissue concentrations were measured.

⁴ Sources of pollution that discharge at a specific location from pipes, outfalls, and conveyance channels to a surface water. Power plants releasing mercury to the air are also point sources.

Variables were grouped into sediment chemical composition, water chemical composition, and morphologic characteristics of the lake. Temperature and pH were divided into top and bottom waters due to the effects lake stratification had on results. Table 6 displays correlation coefficients for the lake variables and standard-sized bass concentrations.

	Mercury in a Standard-sized Bass				
Parameter	Sediment Chemistry	Water Chemistry	Morphologic Characteristics		
Mercury in sediment	0.750				
Total Organic Carbon	0.426				
pH - Top Waters		-0.550			
pH - Bottom Waters		-0.854			
Temperature - Top Waters		0.138			
Temperature - Bottom Waters		-0.567			
Conductivity		-0.221			
Dissolved Organic Carbon		0.276			
Alkalinity		-0.493			
Lake Volume			-0.214		
Surface Area			-0.514		
Drainage Area			-0.445		
Maximum Depth			0.560		
Mean Depth			0.443		

Table 6.	Correlation	Matrix	Describing	Relationshir	os with a	Standard-Sized	(356 mm)	Bass.
1 ubic 0.	Conclution	mann	Deserioning	renationsing	b with a	Stunduru Dizeu	(330 mm)	Dubb.

Several lake variables displayed strong correlations ($r > \pm 0.5$) with mercury concentrations in a standard-sized (356 mm) bass. Strong relationships existed between mercury sediment concentrations, pH in top and bottom waters, temperature in bottom waters, lake surface area, and lake maximum depth.

Average sediment mercury concentrations had a strong (0.750) positive correlation with standard-sized fish tissue estimations. Over the first 3 years of monitoring, sediment concentration averages have contained considerable variability between the 3 sediment grabs (6% – 90% RPD) at each site, and may not be representative of true concentrations. Additionally, larger scale studies have not found mercury concentrations in sediments to correlate well with tissue concentrations (Hanten et al., 1998 and Grieb et al., 1990). Additional years of monitoring and increased sediment testing would be needed to support this finding.

Negative correlations between tissue concentrations and pH (-0.550 and -0.854, top and bottom respectively) and alkalinity (-0.493) were recorded. The effects of low pH and alkalinity have been well established with elevated levels of mercury in fish (Hanten et al., 1998; Grieb et al., 1990; and Hrabik and Watras, 2002). The increased accumulation of mercury in low-pH systems is attributed to increased microbial methylation in acidic waters (Xun et al., 1987). The inverse relationship with alkalinity and mercury levels is likely related to a waterbody's inability to neutralize acidic inputs when alkalinity is very low. The correlation between alkalinity and pH revealed a strong positive relation in the study lakes (0.624 and 0.613, top and bottom respectively).

Maximum depth (0.560) and bottom water temperature (-0.567) also displayed strong correlations with fish concentrations. Enhanced methylmercury production and elevated tissue concentrations have been proposed in higher water temperatures (Bodaly et al., 1993). The inverse relationship between bottom water temperature and positive relationship with maximum depth found in this study may be indicative of low dissolved oxygen levels. The enrichment of methylmercury in anoxic hypolimnetic lake volumes has been observed by several researchers (Herrin et al., 1998; Eckley et al., 2005). Oxygen concentrations have been found to vary spatially and temporally with methylmercury buildup in proportion with each other, and de-stratification is believed to be a key entry point of methylmercury to the food chain (Herrin et al., 1998).

In this project, low dissolved oxygen levels (< 2.0 mg/L) have been measured at Meridian, American, Sammamish, Mason, Loon, Potholes Lakes, Lower Goose Lake, Lake Samish, and Fazon Lake.

Trends Assessment

Spatial Analysis

Waterbodies from Figure 9 containing standard-sized (356 mm) bass mercury concentrations were mapped (Figure 12) to examine spatial differences among lakes. Western Washington waterbodies (n = 8) were compared to eastern Washington waterbodies (n = 9) using a student's t-test to see if differences in concentrations exist. The test showed a significant difference (t = -2.7, p < 0.05) between the two groups, with a higher average among western Washington waterbodies (294 ppb to 126 ppb).



Figure 12. Mercury Sample Sites Categorized by Geographical Regions.

Fischnaller et al. (2003) used an ANOVA with a Bonferroni adjustment to determine if differences existed between standard-sized (356 mm) bass at 15 lakes statewide. Comparisons between waterbodies found 3 eastern Washington waterbodies, Moses Lake, Long Lake, and Banks Lake, to have adjusted concentrations significantly lower than the majority of the other waterbodies. The student's t-test approach described above was applied to the Fischnaller et al. (2003) dataset. The test showed a significant difference (t = -4.75, p < 0.05) between eastern (n = 7) and western (n = 8) waterbodies, with a higher average among western Washington lakes (192 to 138 ppb). The t-test was conducted on original unadjusted data measured by EPA Method 245.5.

It should be noted that the selection process from both studies was not random and contained no statistical design. However, the first three years of mercury trends monitoring indicate widespread low tissue concentrations across eastern Washington waterbodies. Sediment cores collected from eastern Washington lakes also displayed low mercury flux rates when compared to cores from western Washington (Furl, 2007b; 2008). Greater mercury concentrations in fish among western Washington lakes may be the result of proximity to point source pollution and high levels of rainfall in the region resulting in elevated wet deposition.

Temporal Analysis

In addition to the current 2007 study, 10 individual largemouth bass were collected from Deer Lake, Fazon Lake, and Lake Samish as part of Ecology's mercury screening study in 2003 (Fischnaller et al.). In order to estimate any shifts in trends and their magnitudes, a generalized linear model of mercury concentrations in tissues as a function of log_{10} transformed lengths and a dummy variable representing collection year was generated.

1.
$$Log_{10}(Hg) = M + B1(log_{10} Length) + B2(Year)$$

Year was assigned a value of 0 (Fischnaller et al., 2003) or 1 (Mercury Trends, 2007) corresponding with the study. The coefficient B2 and standard error associated with the variable were used to estimate the shift for each lake using the following equation:

2.
$$g = 100 \langle \{ \exp[B2(V(B2)/2)] \} - 1 \rangle$$

where V(B2) is the estimated variance of B2 (Halvorsen and Palmquist, 1980; Kennedy, 1981).

Figure 13 displays the slopes of the lines calculated from the multiple regression model (equation 1) using the dummy variable alongside plotted data from both years and the estimated shift (g).



Deer Lake; 58 months; 15% decrease

Fazon Lake; 71 months; 7% decrease



Lake Samish; 72 months; 1% increase

Figure 13. Temporal Analyses of Lakes Sampled during Multiple Years.

Samples from Deer Lake indicated a 15% decrease in mercury concentrations from fish collected in 2002 at a given length. Results from Deer Lake are similar to temporal decreases in 2006 study lakes (Newman, Long, Meridian, and Moses) where estimated downshifts ranged from 13-31%. Differences in mercury concentrations were small between Fazon Lake and Lake Samish fish groups. Considering sample size, small changes in tissue concentrations estimated from the regression model are likely insignificant.
Criteria for Protection of Human Health

Criteria for Mercury

Various criteria have been developed concerning mercury concentrations in fish tissue in order to meet differing needs:

- 1. EPA's recommended criterion of 300 ppb ww (based on 17.5 grams/day fish consumption rate).
- 2. National Toxics Rule: 825 ppb ww (based on 6.5 grams/day fish consumption rate).
- 3. EPA screening values which are 400 ppb ww for recreational fishers and 49 ppb ww for subsistence fishers (based on freshwater fish consumption rates of 17.5 and 142.4 grams/ day, respectively).

These criteria are summarized below and compared with mercury levels found in fish collected in 2007. Appendix E discusses how Ecology and DOH evaluate fish tissue data to meet the different mandates these agencies have.

1. EPA's recommended criterion

The EPA's current recommended water quality criterion for methylmercury is 300 ppb (EPA, 2001). This is the maximum advisable concentration of methylmercury in fish and shellfish to protect consumers among the general population. EPA expects the criterion to be used as guidance by states and authorized tribes, and EPA in establishing or updating water quality standards for waters of the United States.

2. National Toxics Rule

Washington's water quality standards criteria for toxic contaminants were issued to the state in EPA's 1992 National Toxics Rule (40CFR131.36). Washington's water quality standards further state that risk-based criteria for carcinogenic substances be based on a risk level of 10^{-6} . A risk level is an estimate of the number of cancer cases that would be caused by exposure to a specific contaminant. At a risk level of 10^{-6} , one person in a million would be expected to contract cancer due to long-term exposure to a specific contaminant. These risks are upper-bound estimates, while true risks may be as low as zero. Exposure assumptions include an acceptable risk level and the consumer's body weight, length of exposure, and fish consumption rate. The NTR criteria are based on a fish consumption rate of 6.5 grams/day.

3. EPA Screening Values

Screening values (SVs) for carcinogenic and non-carcinogenic substances were developed by EPA in order to aid the prioritization of areas that may present risks to human populations from fish consumption. The EPA SVs are considered guidance only; they are not regulatory thresholds (EPA, 2000).

Assumptions about exposure to contaminants were also used in developing the EPA SVs. The SV approach is similar to that used for developing the NTR with 2 assumptions: the cancer risk level (10^{-5}) and the consumption rate (17.5 grams/day for recreational fishers and 142.4 grams per day for subsistence fishers). Screening values for non-carcinogenic effects are calculated using toxicological data from a variety of tests.

Human Health Criteria Exceedances

While the criterion recommended by EPA in 2001 for mercury in freshwater fish is 300 ppb, the NTR criterion of 825 ppb wet weight remains as the value used in Washington's water quality standards for regulatory purposes.

DOH's process for establishing fish consumption advisories uses an approach similar to the EPA's *Guidance for assessing Chemical Contaminant Data for use in Fish Advisories Vol. 1-4* (EPA, 2000). Information concerning DOH's data evaluation of fish toxics data is detailed in Appendix E.

Summary statistics displaying percentages of fish samples analyzed by this study exceeding various criteria are included in Table 7.

Table 7. Percentage of Individual and Composite Fish Tissue Samples from the 2007 Study Lakes Exceeding Health Criteria.

Critoria	Percent Exceeding Criteria		
	Individual	Composite	
EPA Screening Values for subsistence fisherman (49 ppb)	100	94	
EPA Recommended Criteria (300 ppb)	73	28	
EPA Screening Values for recreational fisherman (400 ppb)	37	19	
National Toxics Criteria (825 ppb)	7	9	

Conclusions

Sixty individual largemouth bass samples, and 32 composite samples from 8 species, were analyzed for total mercury as part of the third year of a five-year study to gather information on mercury trends in resident freshwater fish from Washington State.

In addition to fish tissue, water and sediment samples were collected to evaluate other factors that may influence mercury uptake in fish. The following 6 lakes were sampled in 2007: Deer Lake, Lake Fazon, Lower Goose Lake, Lake Ozette, Lake Samish, and Lake St. Clair.

Consistent with previous Ecology reports documenting mercury in fish tissue, concentrations were generally higher in older and larger fish. Seventy-three percent (44) of individuals and 28% (9) of composites sampled exceeded the EPA's recommended water quality criterion of 300 ppb. A single four-year-old female bass from Lake Ozette contained a mercury concentration of 1800 ppb. This is the highest concentration recorded in a largemouth bass during the first three years of the study. Seven individual and composite samples surpassed the National Toxics Rule of 825 ppb.

Other significant findings included:

- A temporal analysis was performed for 3 lakes (Deer, Fazon, and Samish) sampled in 2001-2002 and again in 2007. Time between sampling events ranged from 58-72 months. Deer Lake results estimated a 15% decrease in mercury concentrations for fish at a given length. Estimated concentration changes were small at Samish and Fazon Lakes and not believed to be significant.
- Mercury concentrations in standard-sized bass from the first 3 years of the project were compared to determine if concentrations from eastern and western Washington differ. The average concentration in western Washington (294 ppb) was significantly higher than the average eastern Washington concentration (126 ppb). Greater mercury concentrations in fish from western Washington lakes may be the result of proximity to point source pollution and high levels of rainfall in the region resulting in elevated wet deposition.
- Correlation matrices were produced examining relationships between water and sediment composition to the standard length bass for the first 3 years of monitoring. Strong relationships existed between mercury concentration in fish and mercury sediment concentrations, pH in top and bottom waters, temperature in bottom waters, lake surface area, and lake maximum depth.

Recommendations

As a result of the study, recommendations for future mercury trends studies include:

- Sample additional sediment at study lakes to more accurately define sediment mercury concentrations.
- Investigate elevated mercury concentrations found in Lake Ozette largemouth bass and northern pike minnow.
- Consider adding methylmercury analysis to the water sampling plan in order to gain knowledge on lake factors affecting methylation.

References

Bloom, N., 1995. Considerations in the Analysis of Water and Fish for Mercury. In National Forum on Mercury in Fish: Proceedings. U.S. Environmental Protection Agency Office of Water, Washington D.C. EPA Publication No. 823-R-95-002.

Bodaly, R.A., J.M., Rudd, R.P. Fudge, C.A. Kelly, 1993. Mercury concentrations in fish related to size of remote Canadian shield lakes. Canadian Journal of Fisheries and Aquatic Sciences, Vol. 50: 980-987.

CFR, 2004. Code of Federal Regulations, Title 40, Part 131, Section 36: Toxics Criteria for Those States Not Complying with Clean Water Act Section 303(3)(2)(B). Revised July 1, 2004. http://edocket.access.gpo.gov/cfr_2005/julqtr/40cfr131.36.htm

Colman, J., K.D. Lee, C.A. Batdorf, and O.C. Pancorbo, 2008. Comparison of Mercury Uptake by Largemouth Bass and Yellow Perch in Upwind and Downwind Locations on the North American Continent. Manuscript in preparation.

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

Eckley, C.S., C.J. Watras, H. Hintelmann, K. Morrison, A.D. Kent, and O. Regnell, 2005. Mercury Methylation in the Hypolimnetic Waters of Lakes with and without Connection to Wetlands in Northern Wisconsin. Canadian Journal of Fisheries and Aquatic Sciences, Vol. 62: 400-411.

EPA, 2000. Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories -Volume 1: Field Sampling and Analysis, Third Edition. U.S. Environmental Protection Agency, Office of Water. Washington, D.C. EPA Publication No. EPA-823-B-00-007. www.epa.gov/ost/fishadvice/volume1/

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

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

Furl, C., K. Seiders, D. Alkire, and C. Deligeannis, 2007. Measuring Mercury Trends in Freshwater Fish in Washington State: 2005 Sampling Results. Washington State Department of Ecology, Olympia, WA. Publication No. 07-03-007. <u>www.ecy.wa.gov/biblio/0703007.html</u>

Furl, C., 2007a. Measuring Mercury Trends in Freshwater Fish in Washington State: 2006 Sampling Results. Washington State Department of Ecology, Olympia, WA. Publication No. 07-03-043. <u>www.ecy.wa.gov/biblio/0703043.html</u>

Furl, C., 2007b. History of Mercury in Selected Washington Lakes Determined from Age-Dated Sediment Cores: 2006 Sampling Results. Washington State Department of Ecology, Olympia, WA. Publication No. 07-03-019. <u>www.ecy.wa.gov/biblio/0703019.html</u>

Furl, C., 2007c. Comparison of Analytical Methods for Measuring Mercury in Fish Tissues. Washington State Department of Ecology, Olympia, WA. Publication No. 07-03-041. www.ecy.wa.gov/biblio/0703041.html

Furl, C., 2008. History of Mercury in Selected Washington Lakes Determined from Age-Dated Sediment Cores: 2007 Sampling Results. Washington State Department of Ecology, Olympia, WA. Publication No. 08-03-012. <u>www.ecy.wa.gov/biblio/0803012.html</u>

Gallagher, M., 2000. Proposed Strategy to Continually Reduce Persistent, Bioaccumulative Toxins (PBTs) in Washington State. Washington State Department of Ecology, Olympia, WA. Publication No. 00-03-054. <u>www.ecy.wa.gov/biblio/0003054.html</u>

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

Halvorsen, R. and P. Palmquist, 1980. The Interpretation of Dummy Variables in Semilogarithmic Equations. American Economic Review, Vol. 70: 474-475.

Hanten, R.P, R.M. Neuman, S.M. Ward, R.J. Carley, C.R. Perkings, and R. Pirrie, 1998. Relationships between Concentrations of Mercury in Largemouth Bass and Physical and Chemical Characteristics of Connecticut Lakes. Transactions of the American Fisheries Society. Vol. 127: 807-818.

Herrin, R.T., R.C. Lathrop, P.R. Gorski, A.W. Andren, 1998. Hypolimnetic Methylmercury and its Uptake by Plankton During Fall Destratification: A Key Entry Point of Mercury Into Lake Food Chains? Limnology and Oceanography, Vol. 43: 1476-1486.

Hrabik, T.R. and C.J. Watras, 2002. Recent Declines in Mercury Concentration in a Freshwater Fishery: Isolating the Effects of De-Acidification and Decreased Atmospheric Mercury Deposition in Little Rock Lake. The Science of the Total Environment, Vol. 297: 229-237.

Kennedy, P., 1981. Estimation with Correctly Interpreted Dummy Variables in Semilogarithmic Equations. American Economic Review, Vol. 71: 801.

Lake Whatcom Cooperative Management Program, 2001. Fish Advisory: Mercury in Lake Whatcom Smallmouth Bass and Yellow Perch. Whatcom County, City of Bellingham, and Water District #10. <u>www.lakewhatcom.wsu.edu/lwhealthadv.html</u>

Mason, R.P., W.F. Fitzgerald, and F.M. Morel, 1994. The Biogeochemical Cycling of Elemental Mercury: Anthropogenic Influences. Geochim. Cosmochim. Acta. Vol. 58 (15): 3191-3198.

McBride, D., 2003. Statewide Bass Advisory. Washington State Department of Health, Olympia, WA. <u>www.doh.wa.gov/ehp/oehas/publications_pdf/03statewidebass.pdf</u>

McBride, D., 2006. Personal communication regarding sampling strategy for this trend monitoring effort. Washington State Department of Health, Olympia, WA. February 23, 2006.

MEL, 2005. Manchester Environmental Laboratory Lab Users Manual, Eight Edition. Manchester Environmental Laboratory, Washington State Department of Ecology, Manchester, WA.

Momohara, D., 2007. Personal communication. Case Narrative for Laboratory Results for the Mercury Trends in Fish 2007 Study.

Morel, F., A. Kraepiel, and M. Amyot, 1998. The Chemical Cycle and Bioaccumulation of Mercury. Annual Reviews of Ecology and Systematics. November 1998, Volume 29: 543-566. Annual Reviews, Palo Alto, CA.

Norton, D., 2004. Mercury in Lake Whatcom Sediments: Spatial Distribution, Depositional History, and Tributary Inputs. Washington State Department of Ecology, Olympia, WA. Publication No. 04-03-019. <u>www.ecy.wa.gov/biblio/0403019.html</u>

Paulson, A. J., 2004. Sources of Mercury in Sediments, Water, and Fish of the Lakes of Whatcom County, Washington. U.S. Geological Survey Scientific Investigations Report 2004-5084. USGS, Reston, VA.

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

PSEP, 1986. Puget Sound Estuary Program: Recommended Protocols for Measuring Selected Variables in Puget Sound. U.S. Environmental Protection Agency, Region 10, Office of Puget Sound, Seattle, WA.

Ritchie, A., 2008. A late quaternary sediment source and deposition history of Lake Ozette, Olympic National Park, Washington. Report to the National Park Service. Department of Earth and Space Sciences, University of Washington.

Sandvik, P., 2006. Standard Operating Procedure for Resecting Finfish Whole Body, Body Parts, or Tissue Samples. Washington State Department of Ecology, Olympia, WA. SOP No. EAP007. www.ecy.wa.gov/programs/eap/quality.html

Seiders, K., 2006. Quality Assurance Project Plan: Measuring Mercury Trends in Freshwater Fish in Washington State. Washington State Department of Ecology, Olympia, WA. Publication No. 06-03-103. <u>www.ecy.wa.gov/biblio/0603103.html</u> Serdar, D., J. Johnston, K. Mueller, and G. Patrick, 2001. Mercury Concentrations in Edible Muscle of Lake Whatcom Fish. Washington State Department of Ecology, Olympia, WA. Ecology Publication No. 01-03-012. <u>www.ecy.wa.gov/biblio/0103012.html</u>

Swanson, T., 2007. Standard Operating Procedures for Hydrolab[®], DataSonde[®], and MiniSonde[®] Multiprobes. Washington State Department of Ecology, Olympia, WA. SOP No. EAP033. <u>www.ecy.wa.gov/programs/eap/quality.html</u>

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

Appendices

- A. Sample Site Descriptions
- B. Quality Assurance Data
- C. Biological, Sediment, and Water Quality Measures
- D. Statistical Analyses
- E. Fish Tissue Data Evaluation by Ecology and DOH

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Appendix A. Sample Site Descriptions

Site Name	Latitude*	Longitude*	WBID	County	EIM "User Location ID"	WRIA
Deer Lake	48.11158	-117.58806	WA-59-9040	Stevens	DEERLK-F	59
Lake Fazon	48.86613	-122.36757	WA-01-9020	Whatcom	FAZONLK-F	1
Lower Goose Lake	46.92399	-119.28944	WA-41-9170	Grant	LGOOSELK-F	41
Lake Ozette	48.09671	-124.63381	WA-20-9040	Clallam	OZETTELK-F	20
Lake Samish	48.66658	-122.38614	WA-03-9160	Whatcom	SAMISHLK-F	3
Lake St. Clair	46.99473	-122.72699	WA-11-9180	Thurston	STCLAIRLK-F	11

Table A1. Sample Site Descriptions for the 2007 Study.

*NAD83 HARN

WBID - Waterbody Identification

EIM - Ecology's Environmental Information Management database

WRIA - Water Resource Inventory Area

Appendix B. Quality Assurance Data

Fish

Fish tissue analyses for mercury were performed by MEL from November 27 to 30, 2007. Samples were received by the laboratory frozen and in good condition. Analyses were performed within EPA established holding times. Measurement quality objectives (MQOs) for fish tissue analysis are described below in Table B1.

Table B1. Measurement Quality Objectives for Fish Tiss	ue Analysis.
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Parameter	Matrix	Reporting Limit	Accuracy	Check Standard (% recovery limit)	Duplicate Sample (RPD)	Matrix Spike (% recovery limit)
Mercury, total	Tissue	0.017 mg/kg, wet	+/- 15% of SRM value	80-120%	<20%	75-125%

RPD – Relative Percent Difference

SRM – Standard Reference Material

Data quality for fish tissue was assessed through matrix spikes, laboratory blanks, standard reference material (SRM 1946), and laboratory control samples. All laboratory control measures met the above MQOs and are recorded in Tables B2 – B5.

Sample Number	Recovery	RPD (%)
07469219 LMX1	100	1.0
07469219 LMX2	101	1.0
07469239 LMX1	96	1.0
07469239 LMX2	97	1.0
07469259 LMX1	99	2.0
07469259 LMX2	101	2.0
07469269 LMX1	97	4.0
07469269 LMX2	101	4.0
07469289 LMX1	90	0.0
07469289 LMX2	90	0.0
Mean	97.2	1.6

Table B2. Matrix Spike Recoveries and Duplicates.

Sample Number	Result (mg/Kg)
MB07325H1	0.017 U
MB07330H1	0.017 U
MB07330H2	0.017 U
MB07332H1	0.017 U
MB07332H2	0.017 U

Table B3. Laboratory Blanks.

U = undetected at the level indicated

Table B4. Standard Reference Material.

Sample Number	Recovery (%)
ML07325H2	114
ML07330H3	107
ML07330H4	106
ML07332H3	108
ML07332H4	107

Table B5. Laboratory Control Samples.

Sample Number	Recovery (%)
ML07325H1	102
ML07330H1	102
ML07330H2	99
ML07332H1	102
ML07332H2	92

Sediment

Sediment analyses were conducted from October – December 2007. Samples were received by the laboratory in proper condition. All analyses were performed by MEL staff except for grain size which was done by Analytical Resources Inc. All sediment analyses were performed within proper holding times. MQOs as outlined by the project plan appear in Table B6.

Parameter	Matrix	Reporting Limit	Accuracy	Check Standard (% recovery limit)	Duplicate Sample (RPD)	Matrix Spike (% recovery limit)
Mercury, Total	Sediment	0.005 mg/kg, dry	N/A	85-115%	<20%	75-125%
Total Organic Carbon	Sediment	0.10%	N/A	80-120%	<20%	75-125%
Grain Size	Sediment	1%	N/A	N/A	<20%	N/A

Table B6.	Measurement	Quality	Objectives	for Sedime	ent Analysis.
			J		<i>.</i>

N/A = Not analyzed for

Quality control for mercury analyses and TOC was assessed by examining matrix spikes, field replicates, laboratory blanks, laboratory control samples, and duplicates (TOC only). Results appear in Tables B7 – B15. Quality assurance for grain size (Table B16) was assessed through a triplicate sample. All quality control guidelines outlined in Table B6 were met for sediment sampling.

Mercury

Sediment sample mercury concentrations varied widely in different locations within the 6 lakes (see Table B11), but the RPD between source sample and replicate samples (taken as successive grabs) was low (see Table B8).

Table B7. Mercury Matrix Spikes.

Sample Number	Recovery (%)	RPD (%)
07304370-LMX1	90	1 1
07304370-LMX2	91	1.1

Sample Number	Field ID	Result (mg/Kg)	Sample Number	Field ID	Result (mg/Kg)	RPD Between Source Sample and Replicate
07304363	OZE-SED1	0.231	07304368	OZE-SED1R	0.247	6.7%
07304364	OZE-SED2	0.163	07304369	OZE-SED2R	0.146	11.0%
07304365	OZE-SED3	0.219	07304370	OZE-SED3R	0.187	15.8%
-	Mean	0.204	-	Mean	0.193	11.2%
-	RPD of results	34.5%	-	RPD of results	51.4%	-
-	-	-	-	RPD of Means	5.5%	-
Sample Number	Field ID	Result (mg/Kg)	Sample Number	Field ID	Result (mg/Kg)	RPD Between Source Sample and Replicate
07304352	SC-SED1	0.296	07304357	SC-SED1R	0.236	22.60%
07304353	SC-SED2	0.252	07304358	SC-SED2R	0.259	2.80%
07304354	SC-SED3	0.272	07304359	SC-SED3R	0.297	8.80%
-	Mean	0.273	-	Mean	0.264	11.40%
-	RPD of results	16.1%	-	RPD of results	22.9%	-
-	-	-	-	RPD of Means	3.5%	-

Table B8. Mercury Field Replicates.

RPD - Relative Percent Difference

Table B9. Mercury Laboratory Blanks.

Sample Number	Result (mg/Kg)
MB07225H1	0.0050 U
MB07225H2	0.0050 U

U - undetected at the level indicated

Table B10. Mercury – Laboratory Control Samples.

Sample Number	Recovery (%)
ML07225H1	102
ML07225H2	102

Deer La	ke	Lake Fa	zon	Lower Goos	se Lake	Lake Oz	ette	Lake Sa	mish	Lake St.	Clair
Field ID	Result (ppb)	Field ID	Result (ppb)	Field ID	Result (ppb)	Field ID	Result (ppb)	Field ID	Result (ppb)	Field ID	Result (ppb)
DEER-SED1	137	FAZ-SED1	236	LG005-SD1	29	OZE-SED1	231	SM-SED1	151	SC-SED1	296
DEER-SED2	100	FAZ-SED2	236	LG005-SD2	30	OZE-SED2	163	SM-SED2	211	SC-SED2	252
DEER-SED3	75.6	FAZ-SED3	254	LG005-SD3	25	OZE-SED3	219	SM-SED3	173	SC-SED3	272
-	-	-		-		OZE-SED1R	247	-		SC-SED1R	236
-	-	-		-		OZE-SED2R	146	-		SC-SED2R	259
-	-	-		-		OZE-SED3R	187	-		SC-SED3R	297
Mean	104.2		242.0		28.0		198.8		178.3		268.7
RPD ¹	57.8%		7.3%		18.2%		51.4%		33.1%		22.9%
2	29.7		4.3		0.1		0.2		0.2		0.1

Table B11. 2007 Mercury in Sediments Results.

¹ Relative Percent Difference = (max - min) / ((mean) * 100) ² Relative Standard Deviation = 100 * (sd / mean)

TOC

Sample Number	Result (%)	RPD (%)
07314386	2.340 2.340	0.0
07314391	4.800 4.880	1.7

Table B12. TOC – Laboratory Duplicates.

Table B13. TOC – Laboratory Matrix Spikes.

Sample Number	Recovery (%)
GL07228T5-ERAS	117
GL07228T6-ERAS	99

Table B14. TOC – Laboratory Blanks.

Sample Number	Result (%)			
GB07228T5	0.1 U			
GB07228T6	0.1 U			
U - undetected at level indicated				

Table B15.	TOC –	Field	Replicates.
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Sample Number	Field ID	Result (%)	Sample Number	Field ID	Result (%)
07304352	SC-SED1	13.2	07304357	SC-SED1R	10.9
07304353	SC-SED2	12.7	07304358	SC-SED2R	12.5
07304354	SC-SED3	11.3	07304359	SC-SED3R	12.4
-	Mean	12.4	-	Mean	11.9
-	RPD of results	15.5%	-	RPD of results	12.9%
-	-	-	-	RPD of means	3.8%
Sample Number	Field ID	Result (%)	Sample Number	Field ID	Result (%)
Sample Number 07304363	Field ID OZE-SED1	Result (%) 4.56	Sample Number 07304368	Field ID OZE-SED1R	Result (%)
Sample Number 07304363 07304364	Field ID OZE-SED1 OZE-SED2	Result (%) 4.56 4.27	Sample Number 07304368 07304369	Field ID OZE-SED1R OZE-SED2R	Result (%) 4.36 4.34
Sample Number 07304363 07304364 07304365	Field ID OZE-SED1 OZE-SED2 OZE-SED3	Result (%) 4.56 4.27 4.13	Sample Number 07304368 07304369 07304370	Field ID OZE-SED1R OZE-SED2R OZE-SED3R	Result (%) 4.36 4.34 4.03
Sample Number 07304363 07304364 07304365 -	Field ID OZE-SED1 OZE-SED2 OZE-SED3 Mean	Result (%) 4.56 4.27 4.13 4.3	Sample Number 07304368 07304369 07304370	Field ID OZE-SED1R OZE-SED2R OZE-SED3R Mean	Result (%) 4.36 4.34 4.03 4.2
Sample Number 07304363 07304364 07304365 - -	Field ID OZE-SED1 OZE-SED2 OZE-SED3 Mean RPD of results	Result (%) 4.56 4.27 4.13 4.3 9.9%	Sample Number 07304368 07304369 07304370 - -	Field ID OZE-SED1R OZE-SED2R OZE-SED3R Mean RPD of results	Result (%) 4.36 4.34 4.03 4.2 7.9%

Grain Size

Sample Number	Result (%)
SC-SED1	91.6
SC-SED1	85.6
SC-SED1	85.5
Mean	87.6
RPD	6.9%
RSD	4.0

Table B16. Grain Size Triplicate.

Water

Measurement quality objectives for water analysis are presented in Table B17.

Parameter	Matrix	Reporting Limit	Accuracy	Check Standard (% recovery limit)	Duplicate Sample (RPD)	Matrix Spike (% recovery limit)
Dissolved Organic Carbon	water	1 mg/L	N/A	80-120%	<20%	75-125%
Alkalinity	water	5 mg/L	N/A	80-120%	<10%	N/A
Dissolved Oxygen	water	0.2 mg/L	+/- 0.2 mg/L	N/A	< 10%	N/A
pН	water	1.0 SU	+/- 0.3 pH units	N/A	< 10%	N/A
Conductivity	water	5 uS/cm	+/- 5 uS/cm	N/A	< 10%	N/A
Temperature	water	0.0 C	+/- 0.2° C	N/A	< 10%	N/A
Secchi Disc (20 cm dia)	water	1/4 foot	+/- 1/4 foot	N/A	< 10%	N/A

Table B17. Measurement Quality Objectives for Water Analysis.

Dissolved Organic Carbon

Quality control for DOC was assessed through laboratory duplicates, laboratory blanks, laboratory control samples, matrix spikes, and field replicates (Tables B18-B22). Dissolved organic carbon results met all laboratory Quality Assurance/Quality Control requirements. Difficulties were encountered during the analyses, and the reporting limit was raised to 2.0 ppm from 0.5 ppm. All values under 2.0 ppm were qualified as an estimate.

Sample Number	Result (mg/L)	RPD (%)
07304371	1.0 UJ 1.0 UJ	0.0
07304362	3.60 3.60	0.0

Table B18. DOC - Laboratory Duplicates.

UJ - undetected at the estimated level indicated

Table B19. DOC – Laboratory Blanks.

Sample Number	Result (mg/L)
GB07212T1	1.0 UJ
GB07212T2	1.0 UJ
GB07224T1	1.0 U

UJ - undetected at the estimated level indicated U - undetected at level indicated

Table B20. DOC – Laboratory Control Samples.

Sample Number	Recovery (%)
GL07212T1	91
GL07212T2	91
GL07224T1	92

Table B21. DOC – Laboratory Matrix Spike.

Sample Number	Recovery (%)
07304372	103

Table B22. DOC – Field Replicates.

Sample Number	Result (mg/L)	Sample Number	Result (mg/L)	RPD (%)
07304361-T1	3.6	07304366-T2	3.7	2.7
07304362- B1	3.6	07304367-B2	3.6	0.0
07304350-T1	4.2	07304355-T2	4.4	4.7
07304351-B1	8.4	07304356-B2	8.6	2.4

Alkalinity

All laboratory QA/QC requirements were met for alkalinity analyses. Results of laboratory duplicates, blanks, laboratory control samples, and field replicates are presented in Tables B 23-26

Sample Number	Result (mg/L)	RPD (%)
06384230	47 J 47 J	0.0
06384231	47 J 47 J	0.0
06394268	86.8 86.7	0.1
06394269	86.7 86.1	0.7
06394284	24.50 24.60	0.4

Table B23. Alkalinity – Laboratory Duplicates.

Table B24. Alkalinity – Laboratory Blanks.

Sample Number	Result (mg/L)
GB07218K1	5 U

U - undetected at level indicated

Table B25. Alkalinity – Laboratory Control Samples.

Sample Number	Recovery (%)
GL07218K1	95

Table B26. Alkalinity – Field Replicates.

Sample Number	Result (mg/L)	Sample Number	Result (mg/L)	RPD (%)
07304350-T1	50.8	07304355-T2	50.4	0.8
07304351-B1	42	07304356-B2	37	12.7
07304361-T1	6.4	07304366-T2	6.4	0.0
07304362-B1	6.2	07304367-B2	6.1	1.6

Chlorophyll

All laboratory QA/QC requirements were met for chlorophyll analyses. One sample leaked during transport and was qualified as an estimate. Tables B27 and B28 display the results of field replicates and blanks analyzed for chlorophyll.

Sample Number	Result (ug/L)	Sample Number	Result (ug/L)	RPD (%)
07304350-T1	6.9	07304355-T2	7.8	12.2
07304351-B1	1.6	07304356-B2	1.8	11.8
07304361-T1	0.89	07304366-T2	1.1	21.1
07304362-B1	0.69	07304367-B2	0.2	103.3

Table B27. Chlorophyll – Field Replicates.

Table B28. Chlorophyll – Blanks.

Sample Number	Result (ug/L)
GB07204Y1	0.050 U
GB07205Y1	0.050 U
GB07212Y1	0.050 U

U - undetected at level indicated

Appendix C. Biological, Sediment, and Water Quality Measures

Lake	Species Code	Collection Date	Total Length (mm)	Weight (gm)	Age	Fulton's Fish Condition Index	Sex	Mercury (ppb)
Deer	LMB	9/18/07	365	723	4	1.49	F	190
Deer	LMB	9/18/07	320	485	3	1.48	Μ	213
Deer	LMB	9/18/07	315	444	3	1.42	Μ	217
Deer	LMB	9/18/07	369	786	4	1.56	F	249
Deer	LMB	9/18/07	363	693	4	1.45	F	259
Deer	LMB	9/19/07	355	562	6	1.26	Μ	273
Deer	LMB	9/18/07	410	1177	5	1.71	Μ	390
Deer	LMB	9/18/07	454	1445	10	1.54	F	486
Deer	LMB	9/19/07	435	1065	9	1.29	Μ	486
Deer	LMB	9/18/07	441	1325	10	1.54	F	586
Fazon	LMB	9/5/07	365	743	6	1.53	Μ	317
Fazon	LMB	9/5/07	381	792	6	1.43	Μ	343
Fazon	LMB	9/5/07	410	979	5	1.42	F	349
Fazon	LMB	9/5/07	320	432	5	1.32	F	350
Fazon	LMB	9/5/07	425	1251	6	1.63	Μ	368
Fazon	LMB	9/5/07	382	819	6	1.47	F	374
Fazon	LMB	9/5/07	390	893	8	1.51	Μ	400
Fazon	LMB	9/5/07	374	884	6	1.69	F	402
Fazon	LMB	9/5/07	319	372	5	1.15	F	428
Fazon	LMB	9/5/07	456	1380	7	1.46	F	525
Lower Goose	LMB	9/19/07	370	810	3	1.60	F	209
Lower Goose	LMB	9/19/07	375	911	3	1.73	Μ	226
Lower Goose	LMB	9/19/07	420	1137	4	1.53	F	276
Lower Goose	LMB	9/19/07	410	1186	4	1.72	Μ	307
Lower Goose	LMB	9/19/07	385	952	5	1.67	М	315
Lower Goose	LMB	9/19/07	419	1140	4	1.55	F	352
Lower Goose	LMB	9/19/07	390	962	4	1.62	F	363
Lower Goose	LMB	9/19/07	400	1069	5	1.67	Μ	373
Lower Goose	LMB	9/19/07	395	1053	5	1.71	M	380
Lower Goose	LMB	9/19/07	435	1245	4	1.51	F	389
Ozette	LMB	9/12/07	298	383	3	1.45	Μ	350
Ozette	LMB	9/12/07	246	207	2	1.39	Μ	351
Ozette	LMB	10/22/07	340	551	3	1.40	F	385
Ozette	LMB	9/12/07	320	440	3	1.34	Μ	474
Ozette	LMB	9/12/07	341	526	3	1.33	F	496
Ozette	LMB	10/22/07	353	624	3	1.42	F	546
Ozette	LMB	10/22/07	324	446	3	1.31	M	617
Ozette	LMB	9/12/07	415	1080	4	1.51	F	864
Ozette		10/22/07	397	77/1	5	1.23	M	1270
Ozette	LMB	9/12/07	390	910	4	1.53	F	1800
Samish	LMB	9/4/07	269	332	2	1.71	M	130
Samish	LMB	9/4/07	254	228	2	1.39	M	150
Samish	LMB	9/4/07	346	657	3	1.59	M	185
Samish	LMB	9/4/07	342	612	3	1.53	F	200

Table C1. Individual Fish Data by Lake.

Lake	Species Code	Collection Date	Total Length (mm)	Weight (gm)	Age	Fulton's Fish Condition Index	Sex	Mercury (ppb)
Samish	LMB	9/4/07	251	243	2	1.54	М	205
Samish	LMB	9/4/07	419	1338	6	1.82	Μ	418
Samish	LMB	9/4/07	403	1096	5	1.67	F	428
Samish	LMB	9/4/07	385	878	5	1.54	Μ	510
Samish	LMB	9/4/07	457	1451	8	1.52	Μ	574
Samish	LMB	9/4/07	410	1070	5	1.55	F	637
St. Clair	LMB	8/23/07	278	254	3	1.18	F	219
St. Clair	LMB	8/23/07	274	274	3	1.33	F	232
St. Clair	LMB	8/23/07	300	380	4	1.41	Μ	301
St. Clair	LMB	8/23/07	287	337	3	1.43	F	315
St. Clair	LMB	8/23/07	291	294	3	1.19	F	331
St. Clair	LMB	8/23/07	325	495	5	1.44	F	357
St. Clair	LMB	8/23/07	381	778	5	1.41	Μ	397
St. Clair	LMB	8/23/07	432	1257	9	1.56	F	458
St. Clair	LMB	8/23/07	429	1171	12	1.48	Μ	662
St. Clair	LMB	8/23/07	452	1171	17	1.27	F	954

Lake	Species Code	Collection Date	Total Length (mm)	Weight (g)	Fulton's Fish Condition Index	Number of Fish in Composite	Mercury (ppb)
Deer	BBH	9/19/07	268	304	1.57	3	25
Deer	RBT	9/18/07	363	472	0.98	5	60
Fazon	BBH	9/5/07	289	296	1.22	3	57
Fazon	BBH	9/5/07	294	293	1.15	3	50
Fazon	BBH	9/5/07	303	315	1.13	3	60
Fazon	BG	9/5/07	191	178	2.54	3	150
Fazon	BG	9/5/07	172	124	2.43	3	96
Fazon	BG	9/5/07	163	104	2.41	3	120
Lower Goose	BC	9/19/07	325	637	1.77	3	378
Lower Goose	BC	9/19/07	197	112	1.46	3	110
Lower Goose	BC	9/19/07	168	69	1.46	4	96
Lower Goose	BG	9/19/07	167	91	1.87	5	75
Lower Goose	BG	9/19/07	153	68	1.89	5	63
Lower Goose	BG	9/19/07	141	49	1.73	5	71
Ozette	NPM	9/12/07	377	433	0.81	5	1920
Ozette	NPM	9/12/07	343	346	0.85	5	1400
Ozette	NPM	9/12/07	318	262	0.81	5	1090
Ozette	YP	9/12/07	257	183	1.08	5	305
Ozette	YP	9/12/07	244	181	1.24	5	248
Ozette	YP	9/12/07	221	137	1.28	5	197
Samish	CTT	9/4/07	277	185	0.86	3	64
Samish	CTT	9/4/07	259	166	0.95	3	562
Samish	CTT	9/4/07	237	124	0.93	3	44
Samish	NPM	9/4/07	438	776	0.91	3	636
Samish	NPM	9/4/07	393	512	0.84	3	701
Samish	NPM	9/4/07	369	394	0.79	3	303
St. Clair	BG	8/23/07	157	90	2.33	3	160
St. Clair	BG	8/23/07	148	72	2.22	4	193
St. Clair	BG	8/23/07	135	53	2.15	4	170
St. Clair	YP	8/23/07	280	296	1.34	3	204
St. Clair	YP	8/23/07	250	238	1.53	3	205
St. Clair	YP	8/23/07	235	187	1.44	4	170

Table C2.Composite Fish Data by Lake.

				Sediment			Water		
Lake	Collection Date	Field ID	Depth (m)	Mercury (ppb)	TOC (%)	Grain Size (% fines)*	Chl-a (ug/L)	Alkalinity (mg/L)	DOC (mg/L)
Deer	7/31/2007	DEER-SED1	22.9	137	5.38	77.3	-	-	-
Deer	7/31/2007	DEER-SED2	15.2	100	6.56	44.3	-	-	-
Deer	7/31/2007	DEER-SED3	9.1	75.6	5.02	63.9	-	-	-
Deer	7/31/2007	DEER-T1	3.5	-	-	-	0.91 U	41	4.0
Deer	7/31/2007	DEER-B1	14.0	-	-	-	1.9 U	39	4.0
Fazon	7/25/2007	FAZ-SED1	17.0	236	24.1	53.4	-	-	-
Fazon	7/25/2007	FAZ-SED2	15.0	236	27.4	46.3	-	-	-
Fazon	7/25/2007	FAZ-SED3	12.0	254	25	50.1	-	-	-
Fazon	7/25/2007	FAZ-T1	0.5	-	-	-	3.4 J	50.4	18 J
Fazon	7/25/2007	FAZ-B1	3.0	-	-	-	97.3	52.4	16.0
Lower Goose	7/30/2007	LG005-SD1	27.4	29	4.54	93.3	-	-	-
Lower Goose	7/30/2007	LG005-SD2	16.2	30	3.24	95.6	-	-	-
Lower Goose	7/30/2007	LG005-SD3	9.1	25	2.33	72.7	-	-	-
Lower Goose	7/30/2007	LG005-T1	3.0	-	-	-	3.9	86.6	1.6
Lower Goose	7/30/2007	LG005-B1	16.0	-	-	-	1.6 U	177	2.5
Ozette	7/24/2007	OZE-SED1	56.1	231	4.56	84.7	-	-	-
Ozette	7/24/2007	OZE-SED2	33.5	163	4.27	76.3	-	-	-
Ozette	7/24/2007	OZE-SED3	23.8	219	4.13	76.9	-	-	-
Ozette	7/24/2007	OZE-SED1R	54.9	247	4.36	-	-	-	-
Ozette	7/24/2007	OZE-SED2R	33.5	146	4.34	-	-	-	-
Ozette	7/24/2007	OZE-SED3R	23.5	187	4.03	-	-	-	-
Ozette	7/24/2007	OZE-T1	10.0	-	-	-	0.89	6.4	3.6
Ozette	7/24/2007	OZE-B1	35.0	-	-	-	0.69	6.2	3.6
Ozette	7/24/2007	OZE-T2	10.0	-	-	-	1.1	6.4	3.7
Ozette	7/24/2007	OZE-B2	35.0	-	-	-	0.22	6.1	3.6 UJ
Samish	7/26/2007	SM-SED1	21.9	151	5.17	63.5	-	-	-
Samish	7/26/2007	SM-SED2	9.1	211	5.76	53.5	-	-	-
Samish	7/26/2007	SM-SED3	15.8	173	6.27	54.5	-	-	-
Samish	7/26/2007	SM-T1	3.0	-	-	-	2.2	18	1.9 J
Samish	7/26/2007	SM-B1	14.0	-	-	-	2.9	18	1.7 J
St. Clair	7/23/2007	SC-SED1	24.4	296	13.2	85.5	-	-	-
St. Clair	7/23/2007	SC-SED2	29.9	252	12.7	68.1	-	-	-
St. Clair	7/23/2007	SC-SED3	21.3	272	11.3	84.6	-	-	-
St. Clair	7/23/2007	SC-SED1R	24.4	236	10.9	-	-	-	-
St. Clair	7/23/2007	SC-SED2R	29.9	259	12.5	-	-	-	-
St. Clair	7/23/2007	SC-SED3R	21.6	297	12.4	-	-	-	-
St. Clair	7/23/2007	SC-T1	2.0	-	-	-	6.9	50.8	4.2
St. Clair	7/23/2007	SC-B1	25.0	-	-	-	1.6	42	8.4
St. Clair	7/23/2007	SC-T2	2.0	-	-	-	7.8	50.4	4.4
St. Clair	7/23/2007	SC-B2	25.0	-	-	-	1.8	37	8.6

Table C3. Water and Sediment Results.

* % fines = < 62 u

		Ecology
Common name	Scientific name	Species
		Code
Black crappie	Pomoxis nigromaculatus	BC
Bluegill	Lepomis macrochirus	BG
Bridgelip sucker	Catostomus columbianus	BLS
Brook trout	Salvelinus fontinalis	BKT
Brown bullhead	Ameiurus nebulosus	BBH
Brown trout	Salmo trutta	BNT
Burbot	Lota lota	BUR
Channel catfish	Ictalurus punctatus	CC
Chiselmouth	Arocheilus alutaceaus	CLM
Common carp	Cyprinus carpio	ССР
Cutthroat trout	Oncorhynchus clarki	CTT
Green sturgeon	Acipenser medirostrus	GST
Green sunfish	Lepomis cyanellus	GS
Kokanee salmon	Oncorhynchus nerka	KOK
Lake trout	Salvelinus namaycush	LT
Lake whitefish	Coregonus clupeaformis	LWF
Largemouth bass	Micropterus salmoides	LMB
Largescale sucker	Catostomus macrochelius	LSS
Longnose sucker	Catostomus catostomus	LNS
Mountain sucker	Catostomus platyrhynchus	MS
Mountain whitefish	Prosopium williamsoni	MWF
Northern pikeminnow	Ptychocheilus oregonensis	NPM
Peamouth	Mylocheilus caurinus	PEA
Pumpkinseed	Lepomis gibbosus	PMP
Rainbow trout	Oncorhynchus mykiss	RBT
Rock bass	Ambloplites rupestris	RKB
Sculpins	Cottus sp.	COT
Smallmouth bass	Micropterus dolomieu	SMB
Starry flounder	Platicthys stellatus	STF
Walleye	Stizostedion vitreum	WAL
Warmouth	Lepomis gulosis	WM
White crappie	Pomoxis annularis	WC
White sturgeon	Acipenser transmontanus	WST
Yellow bullhead	Ameiurus natalis	YBH
Yellow perch	Perca flavescens	YP

Table C4. Species Code List.

Appendix D. Statistical Analyses

			Regression Coefficients			Mercury				м ·	fisher 1
Waterbody	Species	Study	Constant	B1	B2	Concentration at 356mm		р	r ²	sediment	ratio
2002 Study											
Moses Lake	LMB	Fischnaller et al. 2003	-5.289	2.044	0.267	46		0.000	0.905	27	1.71
Long Lake	LMB	Fischnaller et al. 2003	-346.551	266.766	-51.024	85		0.021	0.573	33	2.57
Banks Lake	LMB	Fischnaller et al. 2003	158.016	-125.669	25.311	141		0.012	0.634	12	11.77
Lake Terrell	LMB	Fischnaller et al. 2003	140.926	-111.341	22.323	146		0.014	0.62	177	0.83
Okanogan R.	LMB	Fischnaller et al. 2003	69.509	-54.893	11.191	202		0.000	0.872	7	28.80
Duck Lake	LMB	Fischnaller et al. 2003	114.056	-91.727	18.788	212		0.000	0.905	103	2.06
Palmer Lake	LMB	Fischnaller et al. 2003	55.528	-44.418	9.241	227		0.015	0.614	57	3.98
Lake Samish	LMB	Fischnaller et al. 2003	44.475	-36.499	7.841	248		0.017	0.597	100	2.48
Vancouver Lake	LMB	Fischnaller et al. 2003	-12.586	7.99	-0.825	269		0.000	0.878	61	4.41
Walla Walla R.	LMB	Fischnaller et al. 2003	-44.898	35.237	-6.54	271		0.002	0.772	13	20.83
Black Lake	LMB	Fischnaller et al. 2003	16.325	-12.908	2.929	287		0.000	0.981	23	12.49
Deer Lake	LMB	Fischnaller et al. 2003	-	-	-	293	J	-	-	55	0.00
Kitsap Lake	LMB	Fischnaller et al. 2003	17.415	-14.298	3.308	295		0.008	0.673	147	2.00
Loomis Lake	LMB	Fischnaller et al. 2003	-	-	-	306	J	-	-	149	0.00
Fazon Lake	LMB	Fischnaller et al. 2003	-107.609	81.578	-15.059	317		0.098	0.661	25	12.67
Newman Lake	LMB	Fischnaller et al. 2003	-46.616	36.281	-6.671	335		0.000	0.976	29	11.57
Lake Meridian	LMB	Fischnaller et al. 2003	-81.584	63.255	-11.865	370		0.016	0.729	212	1.74
2005 Study											
Long Lake	SMB	Mercury trends 2005	-	-	-	31	J	0.180	0.212	120	0.26
Silver Lake	LMB	Mercury trends 2005	127.366	-103.162	21.157	76		0.028	0.539	45	1.70
Potholes Res.	SMB	Mercury trends 2005	19.756	-16.15	3.589	82		0.013	0.628	9	9.12
Loon Lake	LMB	Mercury trends 2005	-	-	-	137	J	-	-	96	1.43
Liberty Lake	SMB	Mercury trends 2005	-	-	-	137	J	0.323	0.069	83	1.65
Yakima R.	SMB	Mercury trends 2005	-197.42	154.535	-29.895	180	J	0.341	0.054	33	5.45
2006 Study											
Moses Lake	LMB	Mercury trends 2006	38.322	-31.337	6.62	29		0.001	0.842	18	1.61
Newman Lake	LMB	Mercury trends 2006	-271.292	209.038	-39.92	152		0.009	0.67	71	2.15

Table D1. Adjusted Mercury Levels for a Standardized Length and Weight.

			Regression Coefficients			Mercury				Manageria	C 1 1
Waterbody	Species	Study	Constant	B1	B2	Concentration at 356mm		р	r ²	sediment	ratio
Lake Offut	LMB	Mercury trends 2006	-	-	-	188	J	-	-	-	0.00
Lake Sammamish	LMB	Mercury trends 2006	-11.735	8.868	-1.315	214		0.003	0.752	178	1.20
Lake Meridian	LMB	Mercury trends 2006	17.004	-13.679	3.111	226		0.000	0.898	266	0.85
2007 Study											
Lower Goose Lake	LMB	Mercury trends 2007	-636.051	488.208	-93.307	147		0.048	0.461	28	5.26
Deer Lake	LMB	Mercury trends 2007	110.547	-86.637	17.34	239		0	0.854	104.2	2.29
Lake Samish	LMB	Mercury trends 2007	62.779	-50.346	10.46	261		0.003	0.763	178.3	1.47
Lake Fazon	LMB	Mercury trends 2007	-	-	-	352	J	-	-	242	1.45
Lake St. Clair	LMB	Mercury trends 2007	31.49	-24.765	5.267	390		0.002	0.786	273.3	1.43
Lake Ozette	LMB	Mercury trends 2007	82.929	-66.769	13.862	648		0.016	0.604	204.3	3.17

 $Regression Equation: Log_{10} (Mercury) = Constant + \{B1 * Log_{10} (Length)\} + \{B2 * (Log_{10} (Length))^2\}$

J - Mercury concentrations are not estimated using the multiple regression equation above. Loon Lake did not contain any fish within the specified size range, while positive significant relationships between mercury concentration and size did not exist at Offut, Deer, Fazon, and Loomis. Estimates for Loon Lake were extrapolated from existing data, while Offut, Deer, Fazon, and Loomis estimates were based off of fish as close to 356 mm as allowable.

* - Weight did not serve as an adequate predictor variable for the Yakima R., Okanogan R., and Walla Walla R. datasets. Regression was based off of length as reported in Furl et al. (2007).

		Coefficient		Standard Error		Adjusted	
Waterbody	Constant	B(1)	B(2)	of $B(2)$	р	r ²	
	Constant	Length	Year	OI D(2)		1	
Fazon	-0.773	1.319	-0.049	0.023	< 0.05	0.536	
Deer	-4.65	2.826	-0.148	0.0175	< 0.05	0.761	
Samish	-4.486	2.72	0.049	0.038	< 0.05	0.642	

Table D2. Regression Results using Year as Dummy Variable.

Regression Equation: $Log_{10}(Mercury) = M + B1(Log_{10} Length) + B2(Year)$



Figure D1. Simple Linear Regression Plots for Mercury and Length.



Figure D2. Simple Linear Regression Plots for Mercury and Weight.



Figure D3. Simple Linear Regression Plots for Mercury and Age.



Figure D4. Boxplot of Weight in Individual Bass.



Figure D5. Boxplot of Length in Individual Bass.

Table D3. Standard-Sized (356 mm) Largemouth Bass Concentrations from Fischnaller et al. (2003).

Lake	Mercury (ppb), standard size 356 mm bass	Location				
Moses	36					
Upper Long	64					
Banks	105	Eastern Washington				
Okanogan	150					
Palmer	166					
Walla Walla R.	199					
Newman	245					
Terrell	110					
Duck	147					
Samish	181	Western Washington				
Kitsap	214					
Black	220					
Fazon	232					
Vancouver	242					
Meridian	312					

Table D4. Results of t-test Comparing Eastern and Western Washington Lakes.

Eastern Washington bass mercury ppb, mean $(n = 7)$	137.9	
Western Washington bass	192.3	
mercury ppb, mean $(n = 8)$		
Mean difference	-54.4	
SD Difference	30.3	
95% CL	-82.5 to -26.4	
t =	-4.750	
df =	6	
P =	0.003	

SD – Standard deviation

CL – Confidence level

T - t score df - degrees freedom P - p value

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Appendix E. Fish Tissue Data Evaluation by Ecology and DOH

Several federal and state agencies collect and evaluate fish tissue data in Washington State Ecology, Department of Health, Washington Department of Fish and Wildlife, 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.

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. The 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.

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 (McBride, 2006). DOH uses an approach similar to that in EPA's *Guidance for Assessing Chemical Contaminant Data for use in Fish Advisories Vol. 1-4* 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 document above, incorporate the varied risk management decisions essential to developing fish consumption advisories.

Following are definitions of these terms:

- **Risk Assessment** involves calculating allowable meal limits based on known fish contaminant concentrations. These calculations are conducted for both non-cancer and cancer endpoints 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 endpoints 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 how best to provide guidance to the public to increase consumption of fish low in contaminants to gain the benefits of eating fish, while steering the public away from fish that have high levels of health-damaging contaminants.