History of Mercury in Selected Washington Lakes Determined from Age-Dated Sediment Cores: 2006 Sampling Results



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Cover photo: Lake Sammamish sediment core taken in October 2006.

## History of Mercury in Selected Washington Lakes Determined from Age-Dated Sediment Cores: 2006 Sampling Results

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April 2007

Lake Ozette WA-20-9040 Lake Sammamish WA-08-9270 Lake St. Clair WA-11-9190 This page is purposely left blank for duplex printing.

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## Abstract

In 2005, the Washington State Legislature provided funding to the Washington State Department of Ecology (Ecology) to develop a long-term monitoring program for mercury in freshwater systems. Mercury is the first pollutant to be studied under the *Persistent, Bioaccumulative, and Toxics (PBT) Reduction Strategy*.

This report presents results of the first year of evaluating mercury deposition through the use of age-dated sediment cores.

During the fall of 2006, Ecology collected sediment cores and surface sediments from the following lakes: (1) Lake Ozette, located in the northwest corner of the Olympic Peninsula, (2) Lake Sammamish in east King County, and (3) Lake St. Clair in Thurston County. The selected lakes reflect potential impacts from different sources: trans-Pacific, large urban area, and the only coal-fired power plant in Washington, respectively.

Sediment cores were dated, using the constant rate of supply model, by examining stable lead, <sup>210</sup>lead, and percent solids. Sediments were also analyzed for total mercury, total organic carbon, selenium, and grain size.

Lake Ozette sediments did not show mercury above background (natural) levels until the mid 1900s, and peaked in the mid 1990s. Sediment cores for Lakes Sammamish and St. Clair displayed increased mercury levels beginning in the early 1900s, and peaked during World War II industrialization in the regions.

Mercury concentrations in recently deposited sediments at Lake Ozette have remained steady with possible slight declines. Mercury levels in sediments at Lake Sammamish have steadily declined since reaching maximum levels in the 1940s. Recent trends at Lake St. Clair reveal steadily increasing levels of mercury over approximately the last 20 years.

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## Introduction

Mercury is a powerful neurotoxin that can bioaccumulate in the food chain, leading to dangerous concentrations in some species of fish and rendering them unsuitable for human consumption. In efforts to reduce human exposure to long-lasting toxins that bioaccumulate in tissue, a *Persistent*, *Bioaccumulative Toxins (PBT) Reduction Strategy for Washington State* (Gallagher, 2000) was developed by Ecology in 2000. Mercury was the first priority pollutant chosen by the state to be addressed under the PBT strategy, resulting in development of a *Washington State Mercury Chemical Action Plan* (Peele et al., 2003).

The monitoring project created under the PBT strategy has two components:

- 1. Investigate mercury trends in fish tissue by collecting and analyzing fish from six lakes around the state each year (Seiders, 2006). The study will evaluate temporal and spatial patterns of mercury in fish tissue.
- 2. Evaluate historical and recent mercury deposition in lake sediments by collecting sediment cores and surface sediments from lakes around the state (Coots, 2006).

This report represents the first year of the sediment coring study with the primary objective being to evaluate mercury deposition through the use of age-dated sediment cores.

## Sediment Core Studies in Washington

Limited freshwater sediment coring studies have been conducted for mercury in Washington lakes. Ecology, in cooperation with the U.S. Geological Survey (USGS), conducted a study of Lake Whatcom, collecting one sediment core from each of the lake's three basins. Norton (2004) reported that mercury concentrations began to increase from background (natural) levels around 1900, steadily increasing in the lake until peaking between 1987 and 1995. Results suggest that mercury concentrations in sediments have leveled off or may be decreasing.

USGS (Paulson, 2004) conducted a companion study in cooperation with the Whatcom County Health Department. Sediment cores were taken from five additional Whatcom County lakes. The additional studied lakes included Lake Terrell, Lake Samish, Baker Lake, Wiser Lake, and Fazon Lake.

Paulson reported that increases in mercury loading were largest in the first half of the 20<sup>th</sup> century. Most increases in mercury sedimentation occurred before major facilities emitting mercury to the atmosphere began operating in Whatcom County. Paulson concludes that the global reservoir was responsible for the majority of mercury deposition to the lakes during the first half of the 20<sup>th</sup> century.

The USGS (Van Metre et al., 2004) conducted a national sediment coring study of 56 lakes including Lake Washington and Lake Ballinger. Study data reveal Lake Washington sediment concentrations were largely influenced by local sources. Mercury concentrations increased substantially above background in the early 20<sup>th</sup> century. Between 1930 and 1970, levels appeared to be fairly constant and then began to fall. Lake Ballinger's mercury sediment record was only recovered to the 1960s. The trends data showed increases to the early 1990s and then decreasing values in more recent times.

Ecology (Yake, 2001) conducted a literature review of the use of sediment cores as a means to track persistent pollutants in Washington State. A total of eleven marine and freshwater studies were reviewed for a variety of contaminants.

## **Site Descriptions**

Figure 1 displays the locations of the three study lakes. The Quality Assurance (QA) Project Plan identified Clear Lake as a sample site for year 1, but sediment conditions were not conducive for coring and Lake St. Clair was substituted in its place. Year 1 site selections were made from western Washington lakes, and Year 2 selections will consist of three lakes from eastern Washington. Table 1 contains information concerning physical characteristics and location of the lakes.



Figure 1. Study Site Locations in 2006.

	Lake St. Clair	Lake	Lake
	(South Arm)	Ozette	Sammamish
Length (mi)	0.5	8	8
Width (mi)	0.5	5	0.75
Surface Area (ac)	88	7300	4900
Volume (ac-ft)	3600	960,000	285,000
Drainage Area (ac)	9280	49,600	63,000
Maximum Depth (ft)	110	320	105
Mean Depth (ft)	40	130	58
DA:SA Ratio <sup>1</sup>	105:1	6.8:1	12.9:1
County	Thurston	Clallam	King

 Table 1. Study Site Morphometry and General Information.

<sup>1</sup> Drainage Area to Surface Area Ratio.

#### Lake Ozette

Lake Ozette is the most westerly lake in the continental United States and the third largest natural lake in Washington State. It is located in the northwest corner of the Olympic Peninsula and within the coastal strip of the Olympic National Park. Lake Ozette is in Water Resource Inventory Area (WRIA) 20. Roughly a mile and one half from the Pacific Ocean in western Clallam County, Lake Ozette is about 21 miles from the nearest town, Sekiu. Situated in the remote northwest region of the Olympic Peninsula, the lake is generally isolated from impacts from urban/industrial centers of the Puget Sound basin.

A number of surface water flows contribute to the lake's volume, with the largest including Big River, Crooked Creek, Umbrella Creek, South Creek, and Siwash Creek. Numerous smaller named and unnamed perennial and ephemeral streams also contribute to the total input to the lake. Outflow is to the Pacific Ocean by way of Ozette River discharging from the lake's north end.

Land use in the basin is dominated by forest land at 83%. The lake surface accounts for about 16% of the basin area, residential use is almost non-existent, and agriculture is an estimated 1% (Bortleson et al., 1976a).



Figure 2. Lake Ozette Study Area and Drainage Basin.

#### Lake Sammamish

Lake Sammamish is a natural lake located in King County, within WRIA 8. Situated at the western edge of the Cascade foothills, the lake is surrounded by several suburbs of Seattle including Issaquah, Sammamish, Bellevue, and Redmond. Elliot Bay and central Puget Sound are about 11 miles from the east lakeshore. Impacts to the lake are likely from the well-established urban area surrounding the lake and the historical releases from the ASARCO copper smelter near Tacoma.

Issaquah Creek is the primary tributary to Lake Sammamish, contributing about 70% of the surface water (KCDNR, 1999). Other larger surface water inflows to the lake include Tibbetts Creek and George Davis Creek. A number of smaller named and unnamed perennial and ephemeral streams also contribute to the lake's total input. Outflow from the lake is to the north into the Sammamish River, draining into Lake Washington's north end.

Land use within the basin is estimated at 40.7% for urban development, 55.8% for mixed forest/vegetative land, and the remaining 3.5% in surface water (Moshenberg, 2004).



Figure 3. Lake Sammamish Study Area and Drainage Basin.

### Lake St. Clair (South Arm)

Lake St. Clair is an irregularly shaped natural lake containing distinct north and south arms separated by a narrow strip of water. The waterbody is located in Thurston County in the Deschutes watershed approximately 10 miles southeast of Olympia, within WRIA 11. The Deschutes watershed has contained historical and present logging, grazing, and agricultural activities. As of 1990, the heavily used recreation lake had approximately 300 houses located along the north and south arms of the lake with two storm drains emptying into the lake. The lake is fed by Eaton Creek along with a number of perennial streams (Bortleson et al., 1976b).



Figure 4. St. Clair Study Area and Drainage Basin.

## **Methods**

### **Study Description**

Due to concern over the health risks mercury may cause to the general public, evaluation of mercury levels in sediments were included as part of the PBT strategy (Gallagher, 2000). The primary goals of the study are to determine historical and current trends of mercury deposition in sediments from lakes throughout Washington State. Specific objectives of the study include:

- Evaluate sediment trends in different areas of the state by analysis of age-dated sediment cores from three Washington lakes per year (Coots, 2006).
- Determine recent mercury deposition in lakes by sampling and analyzing surface sediments to help select lakes for the companion study of mercury trends in freshwater fish (Seiders, 2006).

During the fall of 2006, Ecology's Environmental Assessment Program collected one core sample and one grab sample of surface sediments from each of the study lakes. The sediment cores were age dated, and sedimentation rates were estimated using stable lead analysis and <sup>210</sup>Pb radio-dating. The core samples were also analyzed for total organic carbon (TOC) and mercury.

The grab samples were taken to help characterize recent sedimentation by analyzing mercury, TOC, and selenium and also to allow for enough material to measure grain size. Studies have shown selenium to bind methylmercury reducing its bio-availability (Hodson, 1988).

## Sampling Design

Site selections were based on several factors identified in the QA Project Plan (Coots, 2006) including:

- Statewide coverage
- Proximity to known or suspected mercury sources
- Lake depositional patterns
- Developed access for Ecology's sampling platform

The three lakes selected represent diverse scenarios for mercury pollution. Lake Ozette was chosen to represent global/trans-Pacific mercury pollution due to its remote western location in a watershed largely uninfluenced by anthropogenic impacts. Situated adjacent to the Seattle Metropolitan area, Lake Sammamish will provide information on mercury deposition near a large urban/industrial area. Lake St. Clair represents a smaller residential area located near Washington's only coal-fired power plant.

## Sample Collection

Ecology collected sediment cores and grab samples using the 26' Research Vessel *Skookum*. Sampling stations were located with a differentially corrected Global Positioning System (GPS) ( $\pm$  3 meters) and recorded in field logs. Sediment samples were taken at the deepest part of the lake with the exception of Lake Ozette. Ozette's sediments were collected from a deep area (75m) closer to the ocean side of the lake in an attempt to avoid any possible mass wasting events caused by human disturbances.

Prior to field use, utensils along with acrylic core liners were:

- Washed thoroughly with tap water and Liquinox detergent.
- Rinsed repeatedly with hot tap water.
- Rinsed with de-ionized water.
- Washed in 10% nitric acid.
- Re-rinsed with de-ionized water.

Utensils were wrapped in aluminum foil until used in the field. The corer was thoroughly brushed down with on-site water at each sample location prior to collection of the subsequent sample to avoid cross-contamination.

Once collected, samples were placed in pre-cleaned, 8oz I-Chem jars and stored on ice in the field for no longer than 24 hours. Samples were then frozen at Ecology headquarters in Lacey, Washington, at -20°C.

#### **Sediment Cores**

A Wildco stainless steel box corer containing a 13cm x 13cm x 50cm acrylic liner collected sediment cores approximately 45cm deep.

Once retrieved, Ecology staff visually inspected the sample to determine suitability for subsampling. After a useable core was obtained:

- Overlying water was siphoned away and the acrylic liner removed from the corer.
- The sediment-filled liner was then placed on an extruder table.
- 1cm slices of the core were removed with aluminum plates as the core was pushed from the liner.
- Each subsample layer was then retained excluding any portion of the sediment to come in contact with the liner.

### **Surface Sediments**

Field techniques, sample preparation, and analysis of surface sediments followed Puget Sound Estuary Protocols (PSEP, 1996) when applicable. A 0.1m<sup>2</sup> stainless steel van Veen ponar grab sampler was used to obtain surface sediment immediately adjacent to the coring location (< 3m).

Upon retrieval of the grab, overlying water was siphoned off and the sample was inspected prior to removing the sediments. The top 2cms were removed from the grab, excluding sediment that contacted the sides of the grab. Samples from Lake Sammamish and Lake St. Clair were composite samples of equal amounts from three different grabs, while the Lake Ozette sample consisted of a single grab.

## **Sampling Preparation and Analysis**

Sediment samples chosen for analyses were unfrozen, homogenized, placed in proper sample jars, and sent to the lab for analyses. The jar types, along with holding times, are included in Table 2. Table 3 contains information on the analytical methods used for analysis.

Subsamples chosen for coring analysis were dated by analyzing layers for the radioisotope <sup>210</sup>lead and stable lead. Subsamples were selected for analysis that represent (1) recent conditions (top layer), (2) background conditions which are used to calibrate the <sup>210</sup>lead dating (bottom layer), and (3) a more concentrated selection of layers in the upper core, with wider spacing between layers moving down through the core. Additional sampling design for the sediment cores can be found in Appendix B.

Analyte	Container <sup>1</sup>	Preservation	Holding Time
Total Mercury	Certified 2oz Glass, Teflon Lid Liner	Cool to 4° C	28 Days
<sup>210</sup> Lead	Polystyrene	Freeze, -18° C Cool to 4° C	na
Total Lead	2oz Glass	Freeze, -18° C Cool to 4° C	2 Years 6 Months
Total Selenium	2oz Glass	Freeze, -18° C Cool to 4° C	6 Months
Total Organic Carbon	2oz Glass or Polyethylene	Freeze, -18° C Cool to 4° C	6 Months 14 Days
Grain Size	Glass or Polyethylene	Cool to 4° C	6 Months

Table 2. Container and Holding Information for Sediment Analysis.

<sup>1</sup> = Containers obtained from Manchester Laboratory.

Analysis	Number of Samples <sup>1</sup>	Reporting Limit	Method Description	Analytical Method	Laboratory
Total Mercury	52	0.005 (mg/kg dw)	CVAA	EPA 245.5; MEL SOP <sup>2</sup>	MEL
<sup>210</sup> Lead	30	l (dpm/g)	Gamma Detection	EPA 901.1	STL
Total Lead	30	2 (mg/kg dw)	ICP <sup>4</sup> - MS	EPA 200.8	MEL
Total Selenium	3	0.5 (mg/g dw)	ICP - MS	EPA 200.8	MEL
TOC	36	0.1 (%)	Combustion NDIR	PSEP Protocol	MEL
Grain Size	3	1 (%)	Sieve and Pipette	PSEP Protocol	Analytical Resources

Table 3. Analytical Methods Information.

1 = Includes QA samples.

2 = MEL modifications to analytical methods are documented in their Standard Operating Procedures.

3 = Disintegrations per minute/gram.

4 = Inductively coupled argon plasma.

TOC = Total organic carbon.

EPA = U.S. Environmental Protection Agency

MEL = Manchester Environmental Laboratory

PSEP = Puget Sound Estuary Program

STL = Severn Trent Laboratories

### **Quality Assurance**

Manchester Environmental Laboratory followed standard operating procedures as described in the *Quality Assurance Manual for the Washington State Department of Ecology Manchester Environmental Laboratory* (MEL, 2001) during quality control tests. Also, Manchester staff reviewed data produced by contract laboratories before reaching project staff. Table 4 contains Quality Control (QC) procedure for the required analysis.

Analysis	Method Blanks	LCS	MS/MSD <sup>3</sup>	Lab Duplicates
Total Mercury	1/batch	1/batch	1/batch <sup>1</sup>	1/batch
Total Lead	1/batch	1/batch	1/batch	1/batch
Total Selenium	1/batch	1/batch	1/batch <sup>1</sup>	1/batch
Total Organic Carbon	1/batch	1/batch		1/batch
Grain Size <sup>2</sup>				1/batch

Table 4. Quality Assurance/Quality Control (QA/QC) Procedure Information.

1 = ERA Priority Pollutant Inorganic lot #247 - a soils standard reference material (SRM).

One per batch for the surface sediment samples.

2 = Lab Services performed by Analytical Resources, Incorporated

3 = Matrix Spike/ Matrix Spike Duplicate

Field duplicates were not included as part of QC as outlined by the QA Project Plan (Coots, 2006). Laboratory control samples (LCS) were recovered at acceptable rates and relative percent differences (RPDs) were generally low between matrix spikes. Overall data quality was good for the entire project.

Analytical problems attributed to sediment consistency, along with natural variability, inflated RPDs between two lab duplicates. Two of 8 lab duplicates at Lake Ozette and Lake St. Clair exceeded RPD limits of 25% (Appendix A).

Grain size analysis was qualified due to sediment samples being frozen before analysis along with the proper proportion of fines not available in the sample. The expanding and condensing caused by freezing may have affected grain size and distribution.

Lake St. Clair met the 28-day mercury holding time; however, Lake Sammamish and Lake Ozette exceeded holding times but were analyzed within 33 days after collection. All samples were frozen prior to preparation and shipment to the laboratory.

Thirty-six samples were analyzed for <sup>210</sup>Pb by Severn Trent Laboratories (STL). No analytical problems were encountered and only one sample did not meet detection limits. A complete analysis of all QC data is included in Appendix A.

## **Results**

### **Surface Sediments**

Ecology collected grab samples (top 2cm) to examine grain size, mercury, total organic carbon (TOC), and selenium in the most recently deposited sediments (Table 5).

Location	Mercury Grab (mg/Kg dw)	Mercury Core <sup>3</sup> (mg/Kd dw)	RPD (%)	TOC @ 70°C (%)	Selenium (mg/Kg dw)	Grain Size <sup>1</sup> (% < 62 microns)
Lake Ozette	0.224 <sup>2</sup>	.159² J	33.9	4.65	.50u	61.4
Lake Sammamish	0.17	0.15	12.5	4.63	.50u	67.1
Lake St. Clair	0.29	na	-	12.2	.50u	39.1

Table 5. Surface Sediment Analysis.

<sup>1</sup> Analysis performed by Analytical Resources, Inc. Additional information in Quality Assurance section.

<sup>2</sup> Average of lab duplicates.

<sup>3</sup> Average concentration of the top 2cm.

J = Estimated value.

Variability existed between mercury concentrations in the grab sample and an average of the top two centimeters of the core samples from Lake Ozette. All three samples were re-analyzed in an attempt to add certainty to the results, but reanalysis of the samples yielded a wide range of RPD (17% - 55%) between the duplicates (Appendix B).

A difference in mercury concentration is expected due to natural variability in sediments, but analytical difficulties are also believed to have contributed to the incongruous results. The grab sample and uppermost horizons from the sediment core contained large amounts of water ( $\approx 75\%$ ). Manchester Laboratory sediment analysis protocol is not designed for analysis of samples with such high water content (personal communication).

The Lake Ozette sediment core profile was constructed using the averaged grab samples to represent the top two centimeters as they are believed to most closely represent current sediment conditions.

Lake Ozette and Lake Sammamish sediments were fairly similar in respect to grain size and percent TOC. Sediments at Lake St. Clair contained a lower amount of percent fines ( $\approx$  -25% < 62 microns) with TOC almost three times higher than the other lakes. Selenium was not detected in sediments at any of the lakes.

## **Sediment Cores**

Sediment cores were age dated using a weight of evidence approach between stable lead and <sup>210</sup>lead dating markers. TOC and percent solids were also analyzed throughout the core to determine consistency of supply and compaction. Dating reliability for each core was assigned a value of good, fair, or poor based on the overall consistency of dating markers.

All quality assurance criteria were met except for the lab duplicates of the Lake Sammamish peak value (25-26cm) and Lake Ozette horizon 0-1cm (Table A8). Both samples were qualified as estimates. Additional information on the dating analysis is available in Appendix B.

### <sup>210</sup> Lead

One of the last elements created by radioactive decay of uranium-238 (<sup>238</sup>U), <sup>210</sup>lead forms naturally in sediments and rocks containing <sup>238</sup>U, as well as in the atmosphere where it is a byproduct of radon gas (<sup>222</sup>Rn). <sup>210</sup>Lead is present in sediments as either supported or unsupported <sup>210</sup>lead. Supported <sup>210</sup>lead is represented by the small amount of <sup>222</sup>Rn gas captured in soils, and unsupported lead represents atmospherically deposited <sup>210</sup>lead resulting from the decay of <sup>222</sup>Rn that has escaped into the atmosphere.

As sediments are buried and isolated, the amount of <sup>210</sup>lead present above the secular equilibrium expected in the decay series equals unsupported <sup>210</sup>lead. Using the known half-life (22.3 years) of <sup>210</sup>lead and the amount of the unsupported isotope, the rate of sedimentation and the date of formation can be calculated for approximately the last 150 years (Van Metre et al., 2004 and Charles and Hites, 1987).

#### Stable Lead

Elevated concentrations of stable lead in western Washington first appeared in sediments between 1920 and 1940, and the peak for stable lead is typically around 1975 coinciding with the elimination of lead from gasoline (Van Metre reported in Yake, 2001). Accumulation rates were derived by dividing the accumulated mass sediments at the midpoint of the interval at which the peak was found by 31 years (2006-1975).

The stable lead peak is defined as the midpoint of the sections in which values are within 10% of the highest value. Confidence intervals were calculated for stable lead peaks by defining an upper and lower boundary around the horizon(s) containing the peak (Paulson, 2004 and Norton, 2004).

Accumulation rates derived from first appearance of stable lead were calculated in a similar manner. A range of depths was determined in the sediment core where stable lead first appeared in elevated concentrations, and the accumulation rates determined by dividing the midpoints by 66 and 86 (2006-1940 and 2006-1920, respectively).

### Dating Analysis

Table 6 contains the accumulation rates derived from dating analysis along with a reliability score, and Figure 5 displays the age dated sediment cores with mercury concentrations.

Waterbody	Mass Accumulation Weighted Average (g/cm <sup>2</sup> /yr)	Mass Accumulation Range (g/cm <sup>2</sup> /yr)	Linear Accumulation (cm/yr)	Dating Reliabilty <sup>1</sup>
Lake Ozette	0.074	0.053 - 0.099	0.28	good
Lake Sammamish	0.066	0.061 - 0.097	0.31	good
Lake St. Clair	0.081	0.039 - 0.115	0.43	fair

 Table 6. Sediment Accumulation Rates.

<sup>1</sup> Rated Good, Fair, or Poor Based on consistency of dating information.





Figure 5. History of Mercury Concentrations Constructed from Dated Sediment Cores.

## **Discussion**

### **Transport to Sediments**

Mercury from point and nonpoint sources can be delivered to lake sediments either by direct fallout (wet or dry deposition) onto the lake's surface or fallout onto the watershed followed by fluvial transport (Van Metre et al., 2004; Engstrom and Swain, 1997; and Fitzgerald et al., 1998). Mercury has multiple paths to contaminate sediments; therefore, watershed characteristics along with sedimentation rates largely influence deposition.

Lakes with low sedimentation rates, no regional or local mercury sources, and small drainage area-to-surface area ratios accumulate mercury pollution from atmospheric deposition to the lake's surface and drainage area. Urban, agricultural, and deforested settings are influenced greatly by fluvial inputs. Impermeable surfaces, destruction of vegetation, de-forestation, and erosion from anthropogenic influence increase sediment delivery along with chances for mass movement of sediments during major storm events (Grant and Wolff, 1991).

It is well documented (Yang et al., 2002; and Van Metre et al., 1997) that addition of contaminants to lake sediments through fluvial inputs can far exceed atmospheric fallout. Catchment soils exposed to mercury from atmospheric deposition can store mercury and in turn serve as contamination sources to the lakes in which they drain. Caution must be taken when using surface sediments and sediment cores to describe trends and sources involving atmospheric deposition.

#### **Surface Sediments**

No state or national Freshwater Sediment Quality Values (FSQVs) have been mandated requiring mercury in sediments to meet numerical criteria for protecting benthic<sup>1</sup> organisms. Currently, Ecology is undergoing the process of evaluating draft guidelines for future use. Until Ecology adopts criteria, toxicity to benthic communities is evaluated on a site-specific basis.

Numerous FSQVs are currently available in North America. In 1995, Ecology produced a synopsis of the various FSQVs available (Batts and Cubbage, 1995), and in 2002 and 2003 Avocet Consulting recommended numerical criteria for FSQVs in Washington (Avocet, 2003). Listed below are samples of pertinent FSQVs for total mercury.

<sup>&</sup>lt;sup>1</sup> Organisms living at the bottom of, or in the sediments of, a waterbody.

Freshwater Sediment Quality Value	mg/kg dw	Effects	Provided By
Threshold Effects Level (TEL)	0.17	Adverse biological effects are rarely seen below this level	Environment Canada
Probable Effects Level (PEL)	0.49	Adverse biological effects are frequently seen above this level	Environment Canada
Floating Percentile Method (FPM)	0.5	Proposed level based off or Washington data which can be adjusted to optimize sensitivity and reliability	Avocet Consulting

Table 7. Selected Freshwater Sediment Quality Values (FSQVs) from North America.

According to the FSQVs, mercury contamination does not frequently impact benthic communities at the study lakes. Figure 6 displays surface sediment mercury levels along with the Threshold Effects Level (TEL) and Floating Percentile Method (FPM).



Figure 6. Surface Sediment Mercury Levels.

### Sediment Cores

Mercury enrichment ratios for the historically dated sediment cores are graphed in Figure 7. The mercury enrichment ratio measures increase over background concentration and equals mercury concentration divided by background concentration. The ratio allows for contamination comparisons between the three lakes without having to factor sediment characteristics that may affect mercury concentration (i.e. type, grain size, TOC). Table 8 contains information on mercury concentrations and enrichment ratios.

Lake	Maximum Concentration (ug/kg)	Average Background Concentration (ug/kg)	Maximum Enrichment Ratio	Year
Lake Ozette	0.274	0.168	1.63	1990
Lake Sammamish	0.409	0.096	4.26	1942
Lake St. Clair	0.385 J	0.099	3.89	1946

Table 8. Maximum Mercury Enrichment Ratios.

J = Estimated Value.

Mercury enrichment was highest at the two inland lakes located in urbanized watersheds. The sharp increases in mercury concentrations around the same time indicate Lake Sammamish and Lake St. Clair were affected by local sources related to World War II production. Enrichment ratios are likely understated for all three study lakes because baseline levels were not established from the deepest horizons in the core. Increased levels of mercury from the global reservoir have been documented during the early to mid 1800s (Schuster et al., 2002 and Engstrom and Swain, 1997).





Figure 7. Historically Dated Mercury Enrichment Ratios.

#### Lake Ozette

Consistent with other Washington State coring studies, mercury in Lake Ozette sediments began to increase in the early-to-mid 1900s (Van Metre reported in Yake, 2001; Norton, 2004; and Paulson, 2004). The sediment profile displays two distinct peaks with very low enrichment ratios of 1.63 and 1.55.

The two mercury enrichment peaks occur around 1965 and 1994 with corresponding mercury concentrations of 0.261 and 0.274, respectively. The first peak is likely tied to regional deposition coupled with increased sedimentation due to the construction of logging roads and deforestation in the drainage basin. Dlugokenski et al. (1981) found Lake Ozette sockeye to be negatively impacted due to habitat alteration from increased sedimentation in the late 1950s.

The second peak, occurring in the early to mid 1990s, represents the highest mercury value in the core and results from regional and global inputs. Historically, the Olympic Peninsula has been home to several pulp and paper mills over the last half century which may have contributed mercury to Lake Ozette sediments. Limited data are available for amount and type of mercury emitted from pulp and paper mills across the United States; but, the EPA estimates United States emissions to be approximately 1.9 tons per year (EPA, 1997). Two large Rayonier mills were closed down in the 1990s, Rayonier Grays Harbor and Rayonier Port Angeles.

The incongruous pattern of mercury levels between Ozette and the other two study lakes indicate that mercury supply to sediments is from distinctly different sources. The increases that occurred during World War II production did not affect mercury levels in Lake Ozette, and the decreases during the 1970s and 1980s at the two inland lakes were marked by increases at Ozette. Historical and present mercury fluxes in Lake Ozette sediments appear to be the result of watershed activity and regional pollution sources, with minimal contributions from Puget Sound emissions. It is difficult to assess current trends in the mercury deposition due to confounding analyses in the upper core, but mercury levels appear to be holding steady with possible declines attributed to reduced regional pollution.

#### Lake Sammamish

Mercury enrichment levels in the core peaked during the mid 1940s at 4.26. During this time the Seattle, Tacoma, and Bremerton area was one of the nation's major production centers for World War II. By 1942, Washington was responsible for one-third of refined aluminum in the United States, and involved in many other industrial processes (Warren, 2000). Local pollution from World War II production is very likely to have influenced mercury loading to Lake Sammamish. Effects of World War II mercury pollution have also been recorded in other coring projects outside of Washington State (Schuster et al., 2002). Since the peak, mercury levels have steadily declined over the last half century.

Mercury enrichment patterns and timing were similar to those discovered in nearby Lake Washington (Van Metre reported in Yake, 2001). Mercury contamination reached an enrichment ratio of approximately six in the early 1940s and has steadily declined since the mid 1960s.

The declines to mercury levels in Lake Sammamish can be attributed to rapid post World War II declines along with slowly declining local and regional sources affecting the Puget Sound area. Reconstruction of point source mercury emissions is difficult due to lack of good historical documentation, but the use of mercury laden products has been reduced in recent years along with reductions from municipal landfills. Numerous municipal and hospital waste incinerators have also closed in the Seattle area in the last decade (Prestbo et al., 2006 and Peele, 2003):

- Tacoma Steam Plant
- Sand Point Steam Plant
- Northwest Hospital
- Veterans Administration Hospital
- Swedish Hospital
- University of Washington Hospital
- Evergreen Hospital
- Valley General Hospital

The Mercury Deposition Network (MDN) operates a wet deposition sampling station less than 10 miles away from Lake Sammamish near Lake Washington. The MDN station recorded significant decreases in mercury occurring in the mid 1990s. Prestbo et al. (2006) attributes the decreases at the station to closures of the medical incinerators listed above.

#### Lake St. Clair

The mercury enrichment ratio reached a maximum value of 3.89 during the mid 1940s. The maximum value used to compute the enrichment ratio was qualified as an estimated value because lab duplicates on the sample failed to meet quality assurance criteria (Appendix A). The timing and abruptness of the peak signifies sediments were affected by local sources of pollution likely relating to World War II production. After mercury levels peaked, a drop occurred in the 1950s signaling significant local reductions.

Mercury levels remained fairly low and consistent from the 1950s until the mid 1980s. However, the last 20 years of sediment-history records provided from the core reveal an increase from 220 ppb ( $\approx$  1988) to 370 ppb in the uppermost centimeter ( $\approx$  2006).

An increasing trend not identified in other state sediment cores (Norton, 2004; Paulson, 2004; Yake, 2001; and Van Metre et al., 2004) indicates there may be local sources responsible for mercury loading. The chronology of mercury increases does not correlate well with the TransAlta power plant which began operating in 1971, but the plant is the only known significant mercury source in the area.

## Conclusions

This study was the first year of a five-year study on present and historical mercury trends in Washington State. For this first year, sediment cores were collected from three western Washington lakes: Lake Ozette, Lake Sammamish, and Lake St. Clair.

Study results indicate that Lake Ozette has received relatively little mercury impact compared with the other two lakes. Pulp and paper mills on the Olympic Peninsula have potentially contributed to some mercury loading while logging activity has affected mercury levels through increased sedimentation during the mid 1900s.

Difficulty with analyses in the upper sediment core of Lake Ozette complicated current trends analyses, but it appears that recent mercury contamination has leveled off or slightly declined. Current sources of mercury pollution to Lake Ozette are the global mercury reservoir and slight regional effects when winds are from the east.

Lake Sammamish received a large amount of mercury in a short timeframe during the 1940s. Puget Sound's rapid population and industry growth during the World War II years likely contributed to the high enrichment ratio of 4.26. Neighboring Lake Washington exhibited similar mercury contamination patterns. Since the 1940s, mercury levels in Lake Sammamish have steadily decreased. Sources to the lake include the global reservoir and Puget Sound air releases.

Lake St. Clair displayed contamination patterns similar to Lake Sammamish, signifying that Lake St. Clair also was affected by the increased industrialization during World War II. Pollution levels dropped quickly after spiking in the mid 1940s with an enrichment ratio of 3.89. Levels remained low until the mid 1980s; they have steadily increased since then. Abrupt changes in the mercury profile not displayed in the other sediment cores suggest influence by a local source, possibly the TransAlta power plant 17.5 miles to the southwest.

Mercury pollution at Lake Ozette and Lake Sammamish are fairly low and within typical ranges detected during other mercury studies in the state.

Lake St. Clair mercury levels are slightly higher than in the other two study lakes. Although contamination has not reached levels where benthic impairment is expected, levels of impairment (0.5 mg/kg) could occur in the future if the current trend continues.

## **Recommendations**

As a result of this study, the following recommendations are made:

- Continue sampling diverse lakes from different types of watersheds (urban, agricultural, industrial, and pristine) to evaluate the watershed's effect on mercury loading in sediments.
- In the Year 2 study, evaluate spatial geographical effects of mercury loading on eastern Washington lakes.
- Sample another coastal lake in a different drainage area than Lake Ozette to clarify sources of mercury contamination and monitor possible trans-Pacific loading.
- Sample lakes closer to the TransAlta power plant in Centralia to verify if other lakes are experiencing mercury trends similar to those detected at Lake St. Clair.
- Analyze grab samples from Lake St. Clair to determine if mercury loading is still increasing.
- Analyze the deepest horizon as possible in the sediment core to ensure an adequate background (natural) value is obtained. Sediment core researchers have reported rises in mercury levels beginning in the mid 1800s. The earliest mercury values from this current study are from the late 1800s.

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## **Appendices**

## Appendix A. Quality Assurance Data

- Table A1.Measurement Quality Objectives
- Table A2.Total Organic Carbon
- Table A3. Selenium
- Table A4.Mercury
- Table A5.Stable Lead
- Table A6.Grain Size
- Table A7. <sup>210</sup>Lead
- Table A8.Mercury Lab Duplicates

Demonster	Accuracy	Precision	Bias	Lowest
Parameter	(% of True value)	(Duplicate RPD %)	(% of True value)	Concentration
Total Organic				
Carbon	-	25%	-	1%
Grain Size	-	25%	-	0.10%
Total Mercury	<u>+</u> 40% SRM	25%	<u>+</u> 40% LCS	0.005 mg/Kg, dry
Total Lead	<u>+</u> 40% SRM	25%	<u>+</u> 40% LCS	2 mg/Kg, dry
Total Selenium	<u>+</u> 40% SRM	25%	<u>+</u> 40% LCS	0.5 mg/Kg, dry
<sup>210</sup> Lead	-	25%	-	1dpm*/g

#### Table A1. Measurement Quality Objectives.

#### Table A2. Total Organic Carbon (TOC).

Lab Duplicates					
		Result			RPD
Sample #	Field ID	(%)	LDP1	LDP2	(%)
6434300	GRABOZETT	4.65	4.65	4.87	4.66
6434302	GRABCLAIR	12.2	12.8	12.6	7.98
				Mean:	6.32
Lab Method Blar	ık				
		Result			
Sample #	Field ID	(mg/Kg dw)			
GB 06310T1	Lab BLNK	0.10 U			
GB 06310T2	Lab BLNK	0.10 U			
U= Not detected at det	ection limit shown.				

#### Laboratory Control Sample

		%
Sample #	Field ID	Recovered
GL06310T1	Lab ERAN	99
GL06310T2	Lab ERAN	100
	Mean:	99.5

#### Table A3. Selenium.

Matrix Spike			
	LMX1	LMX2	RPD
Sample #	(%)	(%)	(%)
6434302	77	85	8

#### Lab Method Blank

		Result	
Sample #	Field ID	(mg/Kg dw)	
MB06312I1	Lab BLNK	0.50 U	

U= Not detected at detection limit shown.

#### Laboratory Control Sample

		%	
Sample #	Field ID	Recovered	
ML06312I1	Lab LCS	106	
ML06312I2	Lab ERAS	108	
	Mean:	107.00	

#### Table A4. Mercury.

#### Matrix Spike

	LMX1	LMX2	RPD
Sample #	(%)	(%)	(%)
6434251	82	84	2
6434273	93	94	1
6434302	100	97	3
		Mean:	2.00

#### Lab Method Blank

		Result	
Sample #	Field ID	(mg/Kg dw)	
MB06303H1	Lab BLNK	0.0050 U	
MB06300H1	Lab BLNK	0.0050 U	
MB06300H2	Lab BLNK	0.0050 U	

U= Not detected at detection limit shown.

#### Laboratory Control Sample

		%
Sample #	Field ID	Recovered
ML06303H1	Lab LCS	104
ML06303H2	Lab ERAS	111
ML06300H1	Lab LCS	89
ML06300H2	Lab ERAS	100
ML06300H3	Lab LCS	90
ML06300H4	Lab ERAS	101
	Mean:	99.17

#### Table A5. Stable Lead.

#### **Matrix Spike**

	LMX1	LMX2	RPD
Sample #	(%)	(%)	(%)
6434261	95	88	7

#### Lab Method Blank

		Result	
Sample #	Field ID	(mg/Kg dw)	
MB06307I1	Lab BLNK	0.10 U	
MB06312I1	Lab BLNK	0.10 U	

U= Not detected at detection limit. shown.

#### Laboratory Control Sample

		%	
Sample #	Field ID	Recovered	
ML06307I1	Lab LCS	104	
ML06307I2	Lab ERAS	95	
ML06312I1	Lab LCS	108	
ML06312I2	Lab ERAS	96	
	Mean:	100.75	

#### Table A6. Grain Size.

#### **Grain Size Triplicate** % Total Fines Duplicate Duplicate RPD Sample # Field ID (< 62.5 microns)2 (%) 1 43-4302 St. Clair 39.1 39.9 33.2 17.91 Data Pipette QA Ratio Sample # Field ID Qualifiers Portion 43-4302 St. Clair 102.9 SS,F 3.5 43-4302 Dup 1 99.8 SS,F 3.4 43-4302 Dup 2 98.3 SS,F 2.7 43-4300 Ozette 98.4 F 9.8 43-4301 F Sammamish 100.4 6.9

SS = Did not contain portion of fines required to qualify pipette analysis.

F = Sediment was frozen prior to analysis.

### Table A7. <sup>210</sup>Lead.

Lab Method Blank			
Sample #	Field ID	Result	
JJ88H1AA	BLANK QC	-3.56E-01	
MB06300H1	Lab BLNK	0.0050 U	
MB06300H2	Lab BLNK	0.0050 U	

U= Not detected at detection limit shown.

#### Laboratory Control Sample

		%
Sample #	Field ID	Recovered
ML06303H1	Lab LCS	104
ML06303H2	Lab ERAS	111
ML06300H1	Lab LCS	89
ML06300H2	Lab ERAS	100
ML06300H3	Lab LCS	90
ML06300H4	Lab ERAS	101
	Mean:	99.17

Table A8. Mercury Lab Duplicates.

Field ID	Interval	Original Analysis (mg/kg)	Lab Duplicate	RPD	Average
Ozettel	0_1	0.007	0.17	55	0.13/ I
	0-1	0.097	0.17	17	0.134 J
Ozette2	1-2	0.168	0.199	17	0.184
OzetteGrab	0-2	0.25	0.197	24	0.224
OZETTE5	6-7	0.22	0.259	16	0.240
OZETTE6	7-8	0.205	0.25	20	0.228
OZETTE7	9-10	0.25	0.245	2	0.248
CLAIR2	2-3	0.28	0.252	11	0.266
CLAIR11	25-26	0.519	0.251	70	0.385 J

\*Reported as an average of duplicates. \*RPDs exceeding ±25% were reported as estimates (J).

### **Appendix B. Sediment Core and Surface Sediment Data**

- Table B1.Lake St. Clair Sediment Core Analysis
- Table B2. Lake Ozette Sediment Core Analysis
- Table B3.Lake Sammamish Sediment Core Analysis
- Table B4.<sup>210</sup>Lead Laboratory Results
- Table B5.Sediment Core Dating Markers
- Table B6.Sediment Core Mercury Analysis
- Table B7.Surface Sediment Grain Size Results

#### Table B1. Lake St. Clair Sediment Core Analysis.

Section	Dating Analysis	ID	Mercury Analysis	ID
1	<sup>210</sup> Pb and Total Pb	StCL1	Hg and TOC	Clair1
2	<sup>210</sup> Pb and Total Pb	StCL1	-	
3			Hg and TOC	Clair2
4	<sup>210</sup> Pb and Total Pb	StCL 2		
5	<sup>210</sup> Pb and Total Pb	StCL 2	Hg and TOC	Clair3
6			-	
7	<sup>210</sup> Pb and Total Pb	StCL 3	Hg and TOC	Clair4
8	<sup>210</sup> Pb and Total Pb	StCL 3		
9			Hg and TOC	Clair5
10	<sup>210</sup> Pb and Total Pb	StCL 4		
11	<sup>210</sup> Pb and Total Pb	StCL 4	Hg and TOC	Clair6
12	210			
13	<sup>210</sup> Pb and Total Pb	StCL 5		
14	<sup>210</sup> Pb and Total Pb	StCL 5	Hg and TOC	Clair7
15	210			
16	<sup>210</sup> Pb and Total Pb	StCL 6		
17	<sup>210</sup> Pb and Total Pb	StCL 6	Hg and TOC	Clair8
18	210-21 1	a. at =		
19	<sup>210</sup> Pb and Total Pb	StCL 7		
20	<sup>210</sup> Pb and Total Pb	StCL 7	Hg and TOC	Clair9
21	<sup>210</sup> Pb and Total Pb	StCL 7		
22			Hg and TOC	Clair10
25	210 Dh and Tatal Dh	StCI 9	ng and TOC	Clairio
24	<sup>210</sup> Dh and Total Dh	SICL 8	Ца	Clair12
25	<sup>210</sup> Dh and Total Dh	SICL 8	пg Ца and TOC	Clair11
20	FU and Total FU	SICLO	ng and TOC	Clairi
28			Hg	Clair14
29	<sup>210</sup> Pb and Total Pb	StCL 9	0	
30	<sup>210</sup> Pb and Total Pb	StCL 9		
31	<sup>210</sup> Pb and Total Pb	StCL 9		
32		~~~~ /	Hg	Clair15
33			C	
34				
35 36				
37			Hg and TOC	Clair12
38	<sup>210</sup> Pb and Total Pb	StCL 10	-	
39	<sup>210</sup> Pb and Total Pb	StCL 10		
40				
41				
42				
44				

Collected 10/5/06; Depth 100 feet; Latitude/Longitude 46° 59.681/122° 43.635 NAD 83

Sediment was silty, clay-like material. The core was stratified into 1/3s: black (on top), dark-brown, and light-brown.

#### Table B2. Lake Ozette Sediment Core Analysis.

Section	Dating Analysis	ID	Mercury Analysis	ID
1	<sup>210</sup> Pb and Total Pb	Oze1	Hg and TOC	Ozette1
2	<sup>210</sup> Pb and Total Pb	Oze1	Hg and TOC	Ozette2
3			Hg	Ozette13
4	<sup>210</sup> Pb and Total Pb	Oze2	Hg and TOC	Ozette3
5	<sup>210</sup> Pb and Total Pb	Oze2	Hg and TOC	Ozette4
6				
7	<sup>210</sup> Pb and Total Pb	Oze3	Hg and TOC	Ozette5
8	<sup>210</sup> Pb and Total Pb	Oze3	Hg and TOC	Ozette6
9				
10	<sup>210</sup> Pb and Total Pb	Oze4	Hg and TOC	Ozette7
11	<sup>210</sup> Pb and Total Pb	Oze4		
12			Hg and TOC	Ozette8
13	<sup>210</sup> Pb and Total Pb	Oze5		
14	<sup>210</sup> Pb and Total Pb	Oze5	Hg and TOC	Ozette9
15				
16	<sup>210</sup> Pb and Total Pb	Oze6		
17	<sup>210</sup> Pb and Total Pb	Oze6	Hg and TOC	Ozette10
18				
19	<sup>210</sup> Pb and Total Pb	Oze7		
20	<sup>210</sup> Pb and Total Pb	Oze7	Hg and TOC	Ozette11
21	<sup>210</sup> Pb and Total Pb	Oze7		
22				
23				
24	<sup>210</sup> Pb and Total Pb	Oze8		
25	<sup>210</sup> Pb and Total Pb	Oze8		
26	<sup>210</sup> Pb and Total Pb	Oze8		
27				
28	210-4			
29	<sup>210</sup> Pb and Total Pb	Oze9		
30	<sup>210</sup> Pb and Total Pb	Oze9		
31	<sup>210</sup> Pb and Total Pb	Oze9		
32				
33 24			Ug and TOC	0=====12
34			ng and TOC	Ozette12
36				
37				
38				
39	<sup>210</sup> Pb and Total Pb	Oze10		
40	<sup>210</sup> Pb and Total Pb	Oze10		
41				
42				
43				
44			4	
Core color	did not vary past 1-2 cm. ve	ry consistent g	grey/brown.	

Collected 9/28/06; Depth 253 feet; Latitude/Longitude 48° 4.845/124° 39.003 NAD 83

Table B3. Lake Sammamish Sediment Core Analysis.

Section	Dating Analysis	ID	Mercury Analysis	ID
1	<sup>210</sup> Pb and Total Pb	Sam1	Hg and TOC	Sammam1
2	<sup>210</sup> Pb and Total Pb	Sam1	Hg and TOC	Sammam2
3			Hg and TOC	Sammam3
4	<sup>210</sup> Pb and Total Pb	Sam2	-	
5	<sup>210</sup> Pb and Total Pb	Sam2	Hg and TOC	Sammam4
6			Hg and TOC	Sammam5
7	<sup>210</sup> Pb and Total Pb	Sam3		
8	<sup>210</sup> Pb and Total Pb	Sam3	Hg and TOC	Sammam6
9				
10	<sup>210</sup> Pb and Total Pb	Sam4	Hg and TOC	Sammam7
11	<sup>210</sup> Pb and Total Pb	Sam4		
12			Hg and TOC	Sammam8
13	<sup>210</sup> Pb and Total Pb	Sam5		
14	<sup>210</sup> Pb and Total Pb	Sam5	Hg and TOC	Sammam9
15	•			
16	<sup>210</sup> Pb and Total Pb	Sam6		
17	<sup>210</sup> Pb and Total Pb	Sam6	Hg and TOC	Sammam10
18	210			
19	<sup>210</sup> Pb and Total Pb	Sam7		
20	<sup>210</sup> Pb and Total Pb	Sam7	Hg and TOC	Sammam11
21	<sup>210</sup> Pb and Total Pb	Sam7		
22				G 10
23	21000 100 (101	G 0	Hg	Sammam13
24	<sup>210</sup> Pb and Total Pb	Sam8		
25	<sup>210</sup> Pb and Total Pb	Sam8		
26	Pb and Total Pb	Sam8		
27				
20	<sup>210</sup> Ph and Total Ph	Sam0	Нα	Sammam1/
30	<sup>210</sup> Ph and Total Ph	Samo	ng	Sammanni
31	<sup>210</sup> Ph and Total Ph	Samo		
32		Sam		
33				
34			Hg and TOC	Sammam12
35	<sup>210</sup> Pb and Total Pb	Sam10	-	
36	<sup>210</sup> Pb and Total Pb	Sam10		
37				
38				
39				
40				
41				
42				
43 44				
24 25 26 27 28 29 30 31 32 33 34 35 36 37 38 39 40 41 42 43 44	<ul> <li><sup>210</sup>Pb and Total Pb</li> </ul>	Sam8 Sam8 Sam9 Sam9 Sam9 Sam10 Sam10	Hg Hg and TOC	Sammam14 Sammam12

Collected 10/2/06; Depth 90 feet; Latitude/Longitude 48° 35.34/122° 5.955 NAD 83

Core color did not vary past 1-2 cm. very consistent grey/brown.

	Result			Result:	Result:
Field ID	(pCi/g)	Uncertainty	MDA <sup>1</sup>	MDA <sup>2</sup>	Uncertainty <sup>3</sup>
Oze1	9.17	2.24	1.49	6.15	4.09
Oze2	8.39	1.86	1.16	7.23	4.51
Oze3	12.4	2.97	2.12	5.85	4.18
Oze4	9.94	2.34	1.64	6.06	4.25
Oze5	5.48	1.78	1.5	3.65	3.08
Oze6	3.92	1.3	1.11	3.53	3.02
Oze7	2.51	1.17	1.25	2.01	2.15
Oze8	1.93	0.95	0.98	1.97	2.03
Oze9	1.6	0.545	0.523	3.06	2.94
Oze10	1.34	0.511	0.422	3.18	2.62
Sam1	10.10	3.28	2.39	4.23	3.08
Sam2	8.19	2.07	1.81	4.52	3.96
Sam3	5.47	1.53	1.23	4.45	3.58
Sam4	4.19	1.84	1.82	2.30	2.28
Sam5	3.08	0.70	0.62	4.96	4.38
Sam6	2.38	0.60	0.52	4.59	3.99
Sam7	1.74	0.85	0.89	1.96	2.06
Sam8	1.56	0.51	0.55	2.86	3.07
Sam9	1.28	0.50	0.59	2.17	2.57
Sam10	0.84	0.17	0.52	1.62	4.86
St. Clair1	7.28	1.92	2.16	3.37	3.79
St. Clair2	8.42	2.54	2.17	3.88	3.31
St. Clair3	5.92	1.82	1.58	3.75	3.25
St. Clair4	3.23	1.17	1.17	2.76	2.76
St. Clair5	2.15	1.05	1.29	1.67	2.05
St. Clair6	3.83	1.5	1.52	2.52	2.55
St. Clair7	2.59	1.22	0.932	2.78	2.12
St. Clair8	1.51	0.621	0.407	3.71	2.43
St. Clair9	0.6	0.548	0.509	1.18	1.09
St. Clair10	0.587	0.313	0.226	2.60	1.88

Table B4. <sup>210</sup>Lead Laboratory Results.

<sup>1</sup> Minimal Detectable Activity (MDA).

 $^{2}$  Result to MDA ratio: Ratio of the result to the MDC. A value greater than 1 may indicate activity above background at a high level of confidence. Caution should be used when applying this factor and it should be used in concert with the qualifiers associated with the result.

<sup>3</sup> Result to Total Uncertainty: Ratio of the result to the total uncertainty. If the uncertainty has a coverage factor of 2, a value greater than 1 may indicate activity above background at approximately the 95% level of confidence assuming a two-sided confidence interval. Caution should be used when applying this factor and it should be used in concert with the qualifiers associated with the result.

			Percent		
	Collection	Interval	Solids	Lead	<sup>210</sup> Lead
Lake	Date	(cm)	(%)	(mg/kg)	(piC/g)
Lake Ozette	9/28/2006	0-2	26.6	14.5	9.17
		3-5	26.2	12.5	8.39
		6-8	16.6	15.2	12.4
		9-11	19.6	19.1	9.94
		12-14	16.1	16	5.48
		15-17	22.5	15	3.92
		18-21	24.5	12	2.51
		23-26	24.2	12.3	1.96
		28-31	24.8	12.6	1.66
		37-39	24.3	12.4	1.34
Lake					
Sammamish	10/2/2006	0-2	10	29.8	10.1
		3-5	16.6	34.7	8.19
		6-8	19	84.5	5.47
		9-11	20.7	93.8	4.19
		12-14	20.9	55.7	3.08
		15-17	21.2	27.6	2.38
		18-21	22.1	32.2	1.74
		23-26	21.5	30.9	1.56
		28-31	19.1	3.52	1.28
		34-36	21	2.93	0.84
Lake St. Clair	10/5/2006	0-2	11.3	24.6	7.28
		3-5	11.1	27.8	8.42
		6-8	14.3	24.3	5.92
		9-11	14.4	47.7	3.23
		12-14	21.7	33	2.15
		15-17	18.8	36.7	3.83
		18-21	19.4	25.9	2.59
		23-26	21.1	25.1	1.51
		28-31	19.2	41.8	0.6
		37-39	18.7	12.7	0.587

Table B5. Sediment Core Dating Markers.

	Collection	Interval	TOC	Mercury
Lake	Date	(cm)	(70)	(mg/kg)
Lake Ozette	9/28/2006	0-1	5.41	0.134 J
		1-2	5.03	0.154
		2-3	-	0.274
		3-4	5.05	0.271
		4-5	4.94	0.26
		6-7	4.78	0.24
		7-8	4.47	0.228
		9-10	4.84	0.248
		11-12	5.17	0.261
		13-14	4.95	0.23
		16-17	5.6	0.19
		19-20	4.71	0.17
		33-34	4.71	0.166
Lake Sammamish	10/2/2006	0-1	5 14	0.15
Sammannsn	10/2/2000	1_2	J.14 A A1	0.15
		2_3	4.08	0.15
		2-5 4-5	3.83	0.18
		4- <i>5</i> 5-6	3 79	0.10
		5 0 7-8	3.62	0.19
		9-10	3 39	0.221
		11-12	3 37	0.297
		13-14	3.5	0.363
		16-17	4 1	0 405
		19-20	4.6	0.409
		22-23	-	0.356
		28-29	-	0.12
		33-34	4.79	0.096
Lake St. Clair	10/5/2006	0-1	14.5	0.37
		2-3	13.8	0.266
		4-5	13.9	0.33
		6-7	11.9	0.26
		8-9	12.7	0.24
		10-11	12.1	0.22
		13-14	7.41	0.22
		16-17	8.92	0.24
		19-20	8.98	0.251
		22-23	8.23	0.21
		24-25	-	0.226
		25-26	9.92	0.385
		27-28	-	0.21
		31-32	-	0.13
		35-36	8.96	0.099

Table B6. Sediment Core Mercury Analysis.

J: Estimated value

Sample	% Gravel (> 2000 microns)	% Sand (62 - 2000 microns)	% Silt (3.9 - 62 microns)	% Clay (< 3.9 microns)
Lake Ozette	0.1	60.9	23.0	16.0
LDP1	0.0	60.1	24.1	15.9
LDP2	0.0	66.9	17.9	15.3
Lake Sammamish	0.7	37.9	46.8	14.5
Lake St. Clair	0.0	33.0	52.0	15.1

Table B7. Surface Sediment Grain Size Results.