DEPARTMENT OF
ECOLOGY
State of Washington

Determination of PBT Chemical Trends in Selected Washington Lakes Using Age-Dated Sediment Cores

2009 Sampling Results

June 2010
Publication No. 10-03-035

Publication and Contact Information

This report is available on the Department of Ecology's website at
www.ecy.wa.gov/biblio/1003035.html.

Data for this project are available at Ecology's Environmental Information Management (EIM) website www.ecy.wa.gov/eim/index.htm. Search User Study ID, CFUR0007.

Project Tracker Code for this study is 06-513.

For more information contact:

Publications Coordinator
Environmental Assessment Program
P.O. Box 47600, Olympia, WA 98504-7600
Phone: (360) 407-6764

Washington State Department of Ecology - www.ecy.wa.gov/

o Headquarters, Olympia	(360) 407-6000
o Northwest Regional Office, Bellevue	(425) 649-7000
o Southwest Regional Office, Olympia	(360) 407-6300
o Central Regional Office, Yakima	(509) 575-2490
o Eastern Regional Office, Spokane	(509) 329-3400

Determination of PBT Chemical Trends in Selected Washington Lakes Using Age-Dated Sediment Cores

2009 Sampling Results

by
Chad Furl and Tanya Roberts

Toxics Studies Unit
Environmental Assessment Program
Washington State Department of Ecology
Olympia, Washington 98504-7710

Waterbody Numbers:
American Lake: WA-12-9010
Black Lake: WA-59-9010
Upper Twin Lake: WA-43-3500

This page is purposely left blank

Table of Contents

Page

List of Figures and Tables

Page

Figures

Tables

This page is purposely left blank

Abstract

This study reports results from the fourth year of a monitoring program to evaluate contaminant trends in lakes using age-dated sediment cores. The Washington State Department of Ecology collected deep sediment cores in August and September 2009 from American Lake (Pierce County), Black Lake (Stevens County), and Upper Twin Lake (Lincoln County). Contaminants of interest included mercury, lead, and polycyclic aromatic hydrocarbons (PAHs). Dates were applied to sediment intervals based on radioactive lead (^{210}Pb) measurements.

American Lake contained the greatest historical contamination of all three lakes. Mercury, lead, and PAH concentrations were all highly elevated over background concentrations by 1900 indicating the Asarco plant as a major area source of metals. All three contaminants displayed similar profiles marked by substantial declines over the latter half of the 1900s through recent times.

The Black Lake contaminant profile displayed an erratic mercury and lead pattern post-1950. Concentrations for both contaminants peaked much more recently than the other two lakes. The most recent sediments have displayed an overall declining trend. PAH concentrations ranged from 45 – 236 µg/Kg in four intervals from the Black Lake core. There were not enough PAH detections to describe trends.

Mercury and lead concentrations at Upper Twin Lake were relatively low when compared to the other two lakes. Mercury and lead concentrations have remained within a narrow range over the entire length of the core showing overall declines. No PAHs were found above detection limits in core samples.

Acknowledgements

The authors of this report thank the following Washington State Department of Ecology staff for their contribution to this study:

- Callie Meredith and Keith Seiders for help with sample collection.
- Pam Marti for help with geologic site descriptions.
- Manchester Environmental Laboratory staff for their help and dedication to analytical services: Leon Weiks, Nancy Rosenbower, Dean Momohara, Dickey Huntamer, Dolores Montgomery, Jeff Westerlund, Karin Feddersen, Stuart Magoon, and others.
- Dale Norton for project guidance and review of the report drafts.
- Dave Serdar for reviewing the draft report.
- Joan LeTourneau and Cindy Cook for formatting and editing the final report.

Introduction

In 2000, a *Proposed Strategy to Continually Reduce Persistent, Bioaccumulative Toxics in Washington State* (Gallagher, 2000) was developed by the Washington State Department of Ecology (Ecology). The plan seeks to reduce, and where possible, eliminate persistent, bioaccumulative, and toxic (PBT) chemicals through phasing out use and production.

Mercury was the first chemical 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). A chemical action plan for polybrominated diphenyl ethers was finalized in 2006 (Geller, 2006), and a chemical action plan was issued for lead in 2009 (Davies et al., 2009). A chemical action plan examining polycyclic aromatic hydrocarbons (PAHs) will be drafted in 2011.

A portion of the PBT strategy is aimed at monitoring spatial and temporal trends of PBTs in Washington lake sediments (Gallagher, 2000). Paleolimnological studies have been a useful way of investigating historical rates of contaminant deposition to lake sediments (Yake, 2001; Gallagher et al., 2004; Kamman and Engstrom, 2002; and Engstrom et al., 2007). Coring studies allow for examination of trends and evaluation of current contaminant levels over pre-industrial levels.

This is the fourth year of a statewide study. The goal of the study is to evaluate historical trends in contaminant accumulation statewide through the use of age-dated sediment cores. A complete project plan is described in detail by Coots (2006) and Meredith and Furl (2008). The first two years were aimed at quantifying mercury concentrations only (Furl, 2007; 2008). Currently, contaminants of interest are mercury, lead, and PAHs. This report presents results from three single deep sediment cores collected from American Lake, Black Lake, and Upper Twin Lake.

Contaminant Descriptions

Mercury

Mercury contamination is a widespread global phenomenon with toxic levels found in remote aquatic ecosystems rendering fish unsuitable for consumption. Mercury is transported and deposited to all reaches of the earth via the atmosphere where it can undergo transformation to methylmercury (Rudd, 1995). Anthropogenic releases of mercury have severely altered the natural mercury cycle. Sediment core records have indicated that anthropogenic mercury inputs to sediments have increased by a factor of 2-8 (Engstrom and Swain, 1997).

Natural sources of mercury include weathering of mercury-bearing rocks and soil, volcanic activity, forest fires, and degassing from the earth's surface. Anthropogenic sources include combustion of fossil fuels, metals production, and industrial processes. Currently, the largest point source of anthropogenic mercury in Washington State is from the TransAlta coal-fired power plant in Centralia (USEPA, 2006).

Lead

Lead is a naturally occurring element whose anthropogenic distribution has greatly increased environmental exposure. Lead is considered a metal of concern because it is widespread and persistent in the environment, bioaccumulative, and highly toxic to humans and wildlife.
It is a powerful neurotoxin and children are especially at risk from exposure. The use of alkyl-lead additives in gasoline since the 1920s resulted in widespread lead pollution in the environment. With U.S. restrictions on leaded gasoline, environmental lead concentrations have declined considerably (ATSDR, 2007).

Historically, lead was emitted from the Asarco smelter located in Ruston, Washington. The plant operated as a lead smelter from 1890 - 1905. From 1905 until its closure in 1985, the smelter primarily refined copper (Glass, 2003). Today, point sources of lead to the environment in Washington State include industrial releases such as mining, the Hanford nuclear reservation, military bases, and large energy users. Nonpoint sources of lead to the environment occur through product use, ammunition, aviation fuel, and road dust (Davies et al., 2009). Low concentrations of lead naturally enter surface waters through weathering of bedrock and soils.

PAHs

PAHs are organic, lipophilic contaminants with widespread environmental prevalence. The compounds are formed primarily from the incomplete combustion of carbon-containing materials. PAHs also occur in natural deposits of oil, coal, and tar. Several PAHs are known carcinogens (ATSDR, 1995). Anthropogenic sources of PAHs such as vehicles, heating and power plants, industrial processes, and open burning are believed to be the principal sources to the environment (Van Metre and Callendar, 2000).

The 22 PAHs and associated compounds measured as part of the current 2009 study are shown in Table 1.

Table 1. PAHs and associated compounds measured in sediments.

Analyte
Naphthalene
2-Methylnaphthalene
1-Methylnaphthalene
2-Chloronaphthalene
Acenaphthylene
Acenaphthene
Dibenzofuran
Fluorene
Phenanthrene
Anthracene
Carbazole
Fluoranthene
Pyrene
Retene
Benzo(a)anthracene
Chrysene
Benzo(b)fluoranthene
Benzo(k)fluoranthene
Benzo(a)pyrene
Indeno(1,2,3-cd)pyrene
Dibenzo(a,h)anthracene
Benzo(ghi)perylene

This page is purposely left blank

Methods

Site Descriptions

Site selection criteria were guided by the project plan. In the current study, the eastern Washington sites were selected to provide broad coverage of the state where data do not exist. American Lake was chosen to further investigate higher contaminant levels generally found in western Washington (Furl, 2007; 2008; Furl et al., 2009).

Figure 1 displays the sediment coring locations and lakes. The dot (●) represents the location of core collection within the lake. Locations of all sediment cores sampling sites through the first four years can be found in Appendix B.

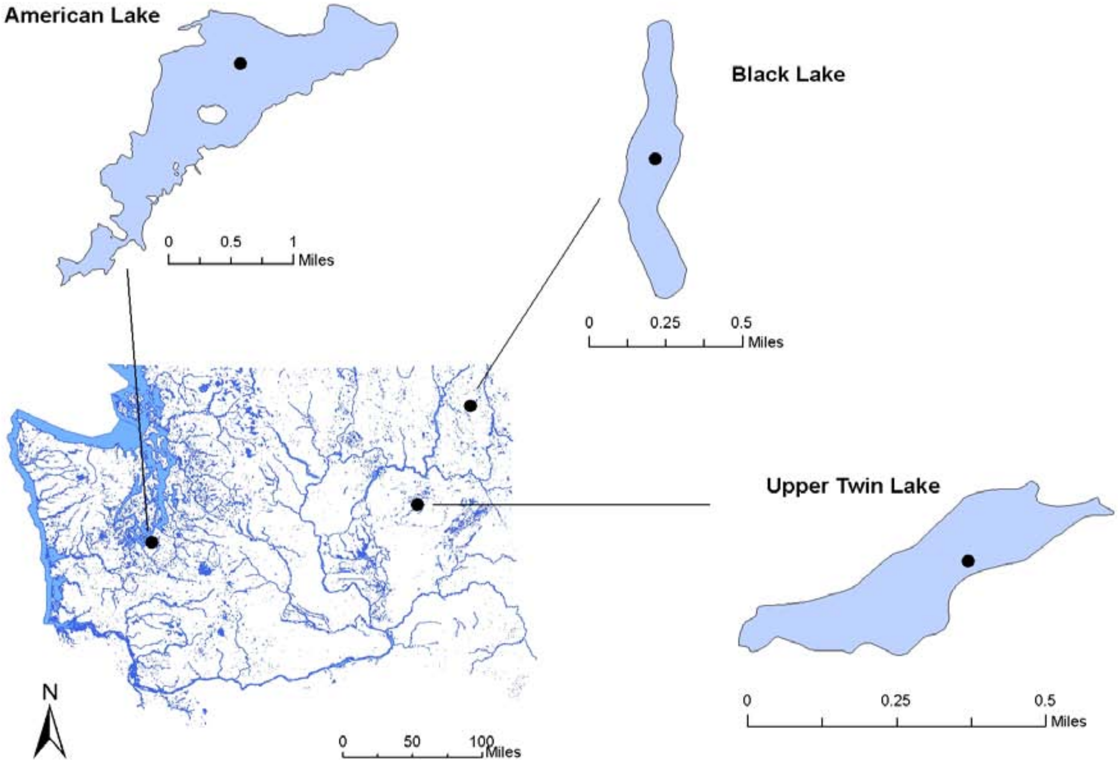

Figure 1. Sediment coring locations, 2009.

Table 2 contains information on the catchment area, lake morphology, and precipitation.

Table 2. Study lakes site data, 2009.

Lake	American	Black	Upper Twin
Surface Area (acre)	1100	70	42
Drainage Area (acre)	16256	576	55104
Volume (acre-ft)	60000	1900	1300
Coring Site Depth (ft)	79	42	64
Maximum Depth (ft)	90	45	64
Mean Depth (ft)	53	27	31
DA:SA Ratio[1]	15	8	1312
Precipitation (in) [2]	42	28	12
Altitude (ft)	235	3701	1877
Coring Site Latitude/ Longitude (dd)[3]	47.1348 / -122.5599	48.5616 / -117.6259	47.5324 / -118.4975

[1] Drainage-area to surface-area ratio.
[2] 1984-2008 average. Precipitation data provided by PRISM: www.prism.oregonstate.edu.
[3] Decimal degrees.

American Lake

American Lake, located in Lakewood, is two miles southeast of the southern end of the Puget Sound. It is the largest naturally occurring lake in Pierce County. Murray Creek, the main surface water inflow, flows into the lake through the Fort Lewis military reservation. Additional inflow is received from groundwater, precipitation, and local run-off. No natural outlet exists; however, a culvert installed in 1956 now transports seasonal overflow from the lake into Sequalitchew Creek, which flows into Puget Sound at the Nisqually Delta (Bortleson, 1976).

American Lake is one of a series of kettle lakes formed on a glacial outwash plain composed of a series of deposits. The Steilacoom Gravel, a glacial recessional outwash, mantels the area and is underlain by the Vashon till and the Vashon advance outwash. Colvos sand and Salmon Springs drift underlie the region near sea level (Walters and Kimmel, 1968). Soils are a gravelly sandy loam on a gently undulating slope (Anderson et al., 1955). Bordered by several small cities, Interstate 5, and a military base, developed land use in the area is urban or suburban, with about half of the drainage basin remaining as undeveloped woodlands or fields.

Black Lake

Black Lake is located in Stevens County in the northeast part of the State, about 15 miles east of Colville. The lake has intermittent surface inflow and outflow (Dion et al., 1976b). Geology of this mountainous region is complex and is characterized by several discrete periods of igneous intrusion, folding, and faulting. Glacial deposits, including outwash, till, and drift, underlay the lake valley. Area soils include stony silt loam and rock outcrop complexes developed in volcanic ash over the glacial till (NRCS, 2006). Current land use in the watershed is largely undeveloped forestland, with some vacation homes and a small resort (Jack, 2003).

Upper Twin Lake

Upper Twin Lake is one of a series of enlargements of Lake Creek, an intermittent stream, located near Odessa in rural Lincoln County (Dion et al., 1976a). Formerly a series of perennial wetlands, channelizing efforts prior to the 1950s aimed at improving cattle pasture heavily altered the Lake Creek drainage. Recent efforts have restored two miles of perennial wetlands immediately upstream of the lake (Lowe, 2010).

Upper Twin Lake lies within the channeled scablands, a unique geologic setting created by massive ice age floods. Geology of the Scablands consists of the Miocene Columbia flood basalts covered with loess. Area soils consist primarily of Cocolalla silt loam, a mix of volcanic ash and loess. Rangeland covers most of the area (BLM, 2008).

Field Methods

Sediment cores were collected from deep, flat areas of the lakes using a Wildco stainless steel box corer containing a 13 cm x 13 cm x 50 cm acrylic liner. Collection efforts were guided by the Ecology Environmental Assessment Program's (EAP) standard operating procedure for *Collection of Freshwater Sediment Core Samples using a Box or KB Corer* (Furl and Meredith, 2008). Cores were determined suitable for subsampling if the sediment-water interface was intact and the core was adequately deep (approximately 35-45 cm). Overlying water was siphoned off the top of the core, and 1-cm section intervals were removed using an extruding device.

Surface sediment grabs were collected using a stainless steel ponar at the coring location (within 3 meters) for grain size analyses. Surface sediment sampling was conducted in accordance with the EAP's standard operating procedure for *Freshwater Sediment Sampling* (Blakley, 2008). Sediment grab samples were kept if the sediment-water interface was preserved. Overlying water was siphoned out of the ponar grab, and the top 2 cm of the sediments were removed using a stainless steel spoon.

The corer, ponar grab, and all equipment coming into contact with sediments were cleaned prior to sampling using the following procedure: Liquinox detergent and hot water wash, nitric acid rinse, de-ionized water rinse, and acetone rinse. The equipment was air dried and wrapped in foil until used in the field. The cleaning procedure was followed between lakes. If an initial core or grab proved unsuitable, the sampling equipment was washed with ambient lake water between samples until a suitable sample was obtained.

Core sections and surface sediment grab samples were placed in 8-oz I-Chem jars with Teflon-lined lids, placed in plastic bags, and put on ice in coolers until transport to Ecology headquarters. Core samples remained in coolers for no more than 48 hours before being frozen at Ecology headquarters at -20° C. Grab samples were stored at 4º C until undergoing grain size analysis.

Field sampling was conducted during August and September 2009.

Sample Preparation and Analysis

Core sediment samples were processed at Ecology headquarters by homogenizing individual 1-cm horizons until the sediments were uniform in color and texture. Sub-samples were taken from the sediment homogenate and placed in the proper pre-cleaned jars for chemical analysis. All chemical analyses for the sediment core were performed on 1-cm intervals. The grab samples were analyzed for grain size only. Appendix C includes detailed information on the chemical analyses performed for each core.

Mercury

Samples were shipped frozen to MEL in pre-cleaned 2-oz I-Chem jars. Sediments were digested with aqua regia for two minutes at 95ºC. Potassium permanganate was then added, and the sample was oxidized for an additional 30 minutes at 95ºC. Mercury was then reduced and measured with conventional cold vapor atomic absorption. Digestion and analysis was conducted according to EPA Method 245.5.

Lead

Samples were shipped frozen to Ecology's Manchester Environmental Laboratory (MEL) in pre-cleaned 2-oz I-Chem jars. Sediments were digested with nitric acid and hydrogen peroxide (EPA Method 3050B) and analyzed by ICP-MS (EPA Method 200.8).

PAHs

MEL measured 30 samples for 22 PAHs and associated compounds. Sediments were shipped frozen in pre-cleaned 4-oz I-Chem jars. Samples were extracted on an accelerated solvent extractor with methylene chloride and analyzed with capillary GC/MS (MEL modification of EPA SW 846 8270). Silica gel cleanup was used following EPA Method 3630C.

^{210}Pb

Forty-five samples were measured for ^{210}Pb radioactivity by counting beta emissions of the daughter product ^{210}Bi. Samples were shipped in nitric rinsed polystyrene jars provided by the laboratory. Samples near the top of the core were tested at a higher density where ^{210}Pb is at its greatest and is spread out deeper in the core. The same intervals measured for ^{210}Pb were also analyzed for stable lead and total mercury.

Total Organic Carbon

MEL measured samples for TOC. Samples were shipped frozen and measured according to Puget Sound Estuary Protocol (PSEP) methodology (USEPA, 1986). Sediment aliquots were acidified to remove inorganic carbon. The sample was then combusted at 900ºC, and carbon dioxide was measured for TOC calculations.

Grain Size

Surface grabs were measured for grain size (gravel, sand, silt, clay) at each of the coring locations. Sediments were cooled to 4ºC, remaining unfrozen until analysis. Analysis was completed using sieving and pipet techniques according to PSEP protocol (USEPA, 1986).

Data Quality

MEL prepared case narratives describing the quality of the analytical data. The narratives include a description of results, laboratory quality assurance, and special issues encountered during analysis. Case narratives are available upon request.

Data for each of the six analytes were generally good. Brief descriptions of the results are included below. Quality assurance tests, along with the project's measurement quality objectives (MQOs), are included in Appendix D.

Mercury and Lead

Sediment intervals were measured in 3 batches each for mercury and lead. Quality control consisted of method blanks, control samples, matrix spikes, and matrix spike duplicates.

For mercury, all blank samples were non-detects (< 0.0050 mg/Kg). Control samples and matrix spikes were recovered at an average of 107% and 91%, respectively. Duplicate precision among matrix spikes averaged 9% difference with one of the duplicates violating MQOs (> 20% RPD). Several mercury samples in the upper portions of the American Lake and Upper Twin Lake cores contained concentrations less than the limit of quantitation (LOQ). Values below the reporting limit were used since the concentrations seemed reasonable based on surrounding intervals. Concentrations reported outside the calibration range were qualified as estimates (J).

For lead, all blank samples were non-detects (< 0.10 mg/Kg). Control samples and matrix spikes were each recovered at an average of 100%. Duplicate precision among matrix spikes averaged 3% difference.

PAHs

Quality control for PAH samples consisted of surrogate recoveries (2-Fluorobiphenyl, Pyrene-D10, and Terphenyl-D14), method blanks, laboratory control samples, matrix spikes, matrix spike duplicates, laboratory duplicates, and standard reference material.

Surrogate recoveries, method blanks, laboratory control samples, and matrix spike duplicates all met MQOs. Several analytes were qualified as estimates in one of the sample duplicates. Benzo(g,h,i)perylene was recovered low in one matrix spike; no action was taken since the recovery for the compound was acceptable in the duplicate. Several analytes were recovered low in the standard reference material. No action was taken due to acceptable recoveries in matrix and control spikes.

^{210}Pb

Quality control for ^{210}Pb samples consisted of blanks, control samples, and duplicates. All blank samples were less than the LOQ, laboratory control samples were recovered at an average of 95%, and duplicates were within MQOs.

Total Organic Carbon

Quality control for TOC samples consisted of blanks, duplicates, and analysis of standard reference material. All blank samples were less than the LOQ, duplicates varied on average 3%, and recovery of standard reference material averaged 93%.

Grain Size

Quality control for grain size samples consisted of a single triplicate. Results were within specified MQOs with the exception of the gravel fraction.

Data Processing

Sedimentation Modeling and Data Calculations

The constant rate of supply (CRS) model was applied to the ^{210}Pb measurements to estimate dates and varying sedimentation rates throughout the core (Appleby and Oldfield, 1978). The model works by measuring the difference in supported and unsupported ^{210}Pb in sediment horizons. Supported ^{210}Pb is represented by the small amount of the precursor gas ^{222}Rn that is captured in soils. Unsupported ^{210}Pb represents atmospherically deposited ^{210}Pb resulting from the decay of ^{222}Rn that escapes into the atmosphere and is estimated by subtracting supported ^{210}Pb from total ^{210}Pb. Using the known half-life (22.3 years) of ^{210}Pb 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; Charles and Hites, 1987).

Supported ^{210}Pb present in the study cores was estimated as the amount present at deep intervals where it appeared that ^{210}Pb no longer declined (Figure 2). Sediment dry mass (g/cm^2) was calculated from percent solids data obtained from total lead and mercury measurements made at MEL. Estimates on percent solids were extrapolated where data did not exist. An assumed sediment density of 2.7 g/cm^3 was used based on other Washington State coring studies (Paulson, 2004).

The amount of sediment focusing at the coring site was judged by unsupported yearly flux estimates from the sediment core. Sediment focusing refers to the deposition of sediments and associated contaminants from one part of the lake to another as the result of morphometry, water current, sediment cohesiveness, and other physical and chemical processes (Blais and Kalff, 1995). A core estimate greater than expected values indicates focusing at the site.

Mercury Flux Calculations

Mercury flux rates (µg/m^2/yr) were calculated as the product of sediment mass accumulation rates (SMARs) and dry weight mercury concentrations. Mercury flux rate results are used to estimate net deposition to the lake. Flux rates normalize the variance involved with interpreting dry weight concentrations under varying sedimentation rates.

PAH Data Processing

Summing procedures were conducted according to the Washington State Sediment Management Standards rule (Washington State Department of Ecology, 1995). Only detected concentrations were included in total PAH sums. When no PAHs were detected in a sample, the single highest detection limit, appropriately qualified, represented the total.

This page is purposely left blank

Results and Discussion

Core Dates and Sedimentation Rates

^{210}Pb concentrations along with sediment depth for each profile are displayed in Figure 2.

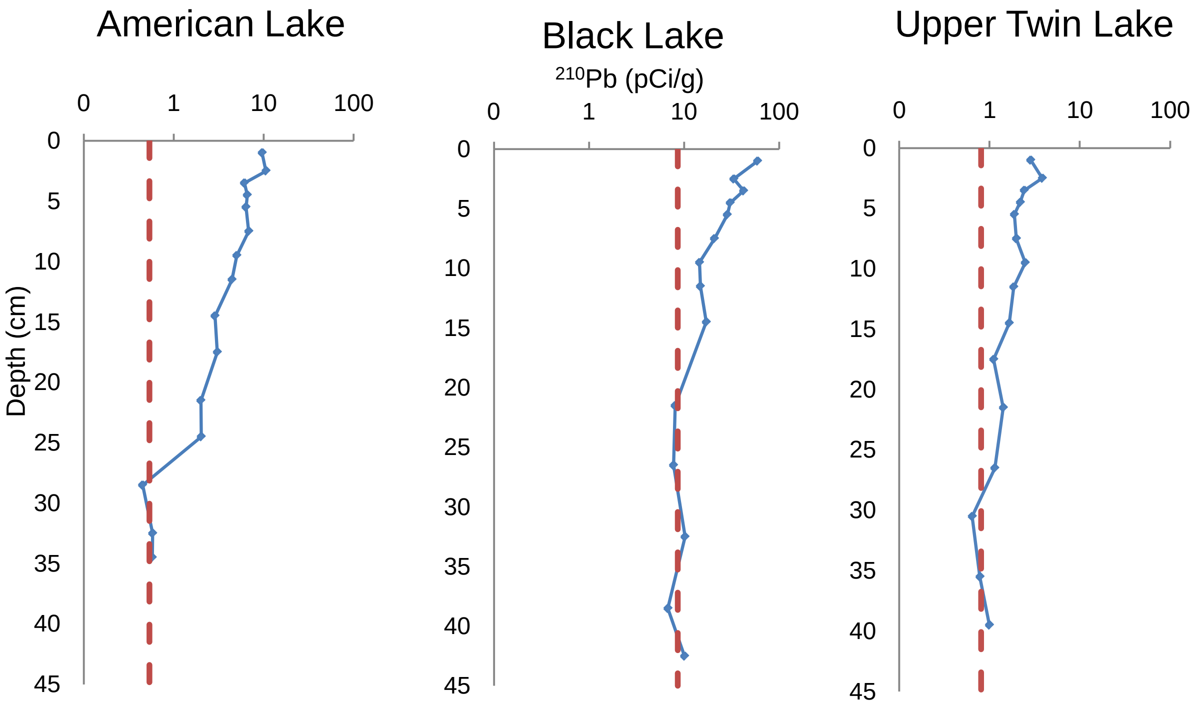

Figure 2. ^{210}Pb measurements plotted with depth.
Dashed line represents supported ^{210}Pb concentrations.

^{210}Pb concentrations in the sediment cores did not display perfect monotonic declines. Supported ^{210}Pb levels were reached at approximately 20 cm for Black Lake and 30 cm for American and Upper Twin Lakes. Estimated supported ^{210}Pb concentrations and unsupported yearly flux values are shown in Table 3.

Table 3. Supported ^{210}Pb concentrations (n) and yearly unsupported ^{210}Pb flux.

Lake	Supported ^{210}Pb (pCi/g)	Unsupported ^{210}Pb Yearly Flux (pCi/cm^2/yr)
American	0.54 (3)	0.231
Black	8.56 (5)	0.746
Upper Twin	0.81 (3)	0.130

Unsupported ^{210}Pb fluxes calculated from measured data normally fall within 0.2 – 1.0 pCi/cm^2/yr (Oldfield and Appleby, 1984). Atmospheric ^{210}Pb deposition measured over a 7-year period in Seattle were at the lower end of this range (0.198 pCi/cm^2; SD = 0.108) (Nevissi, 1985). Unsupported yearly fluxes estimated from statewide cores over the first three years of the project were 0.50 pCi/cm^2 (SD = 0.26; n = 8) (Furl, 2007; 2008; Furl et al., 2009).

The estimates for unsupported ^{210}Pb yearly flux at American Lake (0.231 pCi/cm^2/yr) closely correspond with the measured values collected by Nevissi (1985) 40 miles north of the coring location.

Unsupported yearly ^{210}Pb fluxes at Upper Twin Lake (0.130 pCi/cm^2/yr) were low but within the estimated range found at other Washington State coring locations. Similarly, unsupported fluxes at Black Lake are higher than the average, but fall within the range estimated from previous Washington State sediment cores. Based on the estimated fluxes from the three sediment cores, no adjustments were made for sediment focusing.

Sedimentation rates along with their estimated dates are shown in Figure 3.

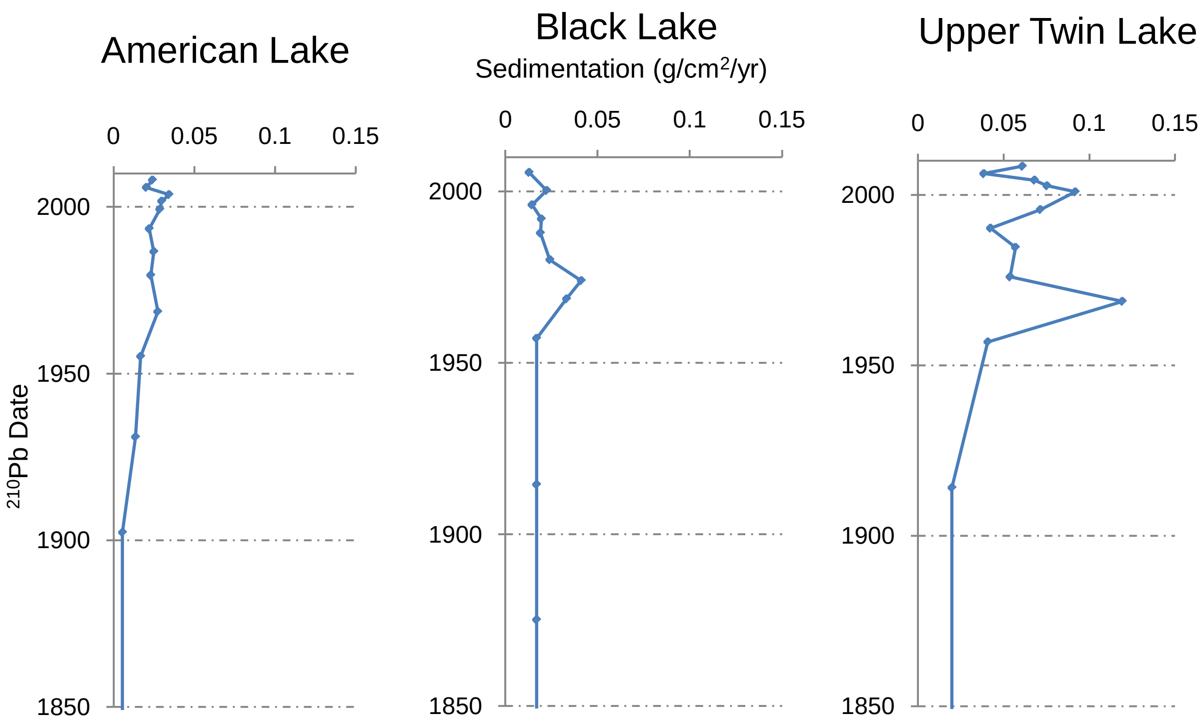

Figure 3. Sediment core sedimentation rates with dates.

Estimated sedimentation rates ranged from 0.005 – 0.12 g/cm^2/yr. Sedimentation ranges from previous coring reports are shown in Table 4. Sedimentation rates were lowest at American and Black Lakes where they did not exceed 0.05 g/cm^2/yr.

American Lake sedimentation rates increased modestly from 1900 to approximately 1970. After 1970, sedimentation rates were fairly stable, remaining within a narrow range.

Variable sedimentation rates at Black Lake could only be estimated to the mid-1950s from which rates increased by small amounts until their maximum during the mid-1970s. After this point, sedimentation rates decreased over 3-fold to their present values.

Sedimentation rates at Upper Twin Lake ranged from 0.02 - 0.12 g/cm^2/yr and were highly variable relative to the other two lakes. The highest rates were recorded near 1970 and were elevated over the surrounding horizons. It should be noted that total ^{210}Pb concentrations were low at Upper Twin Lake (0.642 – 3.840 pCi/g), increasing the chance for large swings in modeled sedimentation rates from small changes in ^{210}Pb counts.

Table 4 displays sedimentation rate ranges along with the peak year from previous years of the project.

Table 4. Sedimentation ranges and peak sedimentation years from previous coring reports.

Lake	Sedimentation Range (g/cm^2/yr)	Peak Year	Study
American Lake	0.0055 - 0.0343	2004	Present study
Black Lake	0.0129 – 0.0411	1974	
Upper Twin Lake	0.0198 – 0.1193	1969	
Lacamas Lake	0.0107 - 0.0674	1966	Furl et al., 2009
Lake Offutt	0.0216 - 0.0390	2001	
Lake Washington	0.0177 - 0.1121	1977	
Loon Lake	0.0090 - 0.0248	1993	Furl, 2008
Wannacut Lake	0.0094 - 0.0229	1937	
Lake Ozette[1]	0.0223 - 0.1153	2004	Furl, 2007
Lake Sammamish[1]	0.0188 - 0.0637	1950	
Lake St. Clair[1]	0.0170 - 0.0895	1983	

[1] Furl 2007 - ^{210}Pb values are included in report.
Sedimentation rates and associated year are not included in the 2007 report.

Total Organic Carbon and Grain Size

Percent TOC in core intervals ranged from 3.34 – 17.10. Average (n = 10) percent TOC concentrations for each of the cores was 14.35, 8.89, and 4.92 at Black, American, and Upper Twin, respectively. Within each core, concentrations varied little (Appendix C).

Percent fines (clay + silt) measured in sediment grabs near the coring site were 51.4, 60.7, and 64.3 at American, Black, and Upper Twin, respectively.

Contaminant Profiles

Mercury

Figure 4 displays mercury flux and dry weight concentration profiles for each of the lakes.

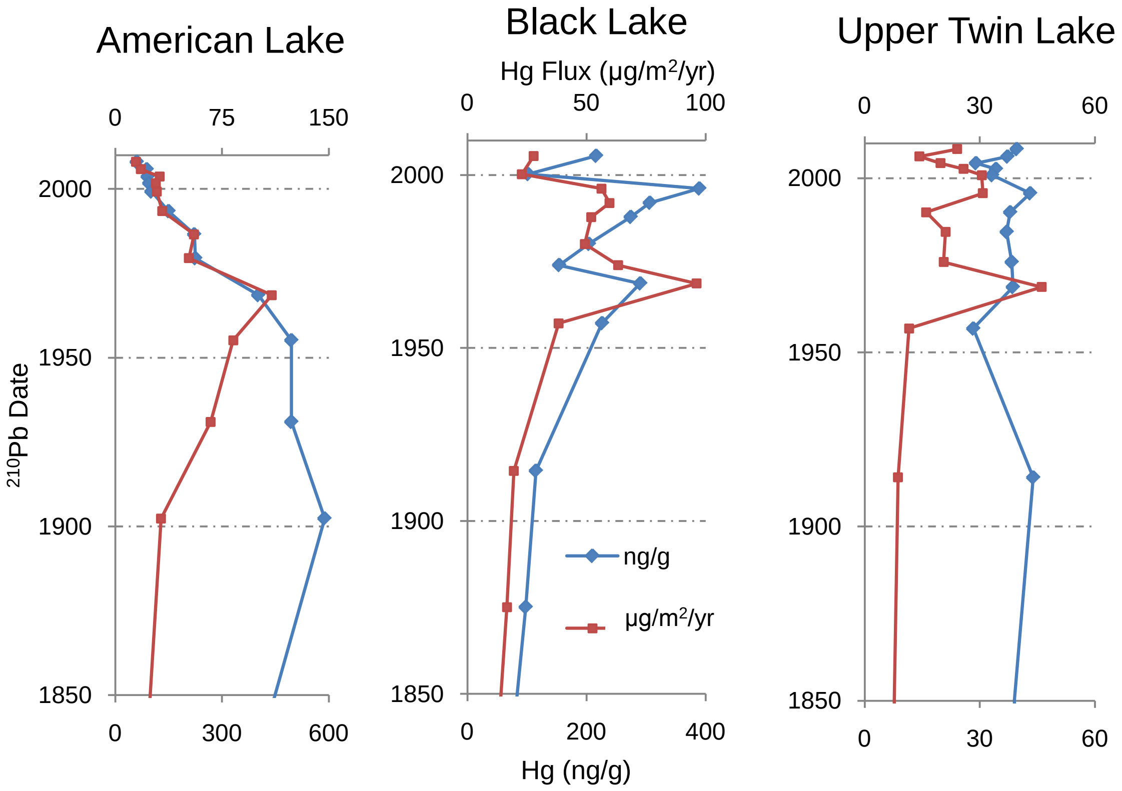

Figure 4. Mercury flux and dry weight concentration profiles with dates.
(note scale differences)

Concentrations

Mercury dry weight concentrations ranged from 28 – 588 ng/g, with the highest concentrations found at American Lake followed by Black Lake and Upper Twin Lake. Several sediment intervals contained concentrations less than the LOQ in the upper portion of the sediment core at American Lake (intervals 0-2, 2-3, and 3-4) and Upper Twin Lake (intervals 0-2, 2-3, 3-4, 4-5, 5-6, 9-10, and 11-12). Results from those intervals outside of the calibration range are reported as estimates.

Mercury concentration profiles displayed dissimilar shapes between the three lakes. Concentrations at American Lake were highly elevated deep in the core reaching their maximum (588 ng/g) by 1900. The elevated concentrations near the turn of the century are likely heavily influenced by metals smelting at the Asarco plant 15 miles north of the lake. The plant began smelting lead in 1890, and numerous studies have documented local metals contamination attributed to atmospheric deposition from the plant (Glass, 2003 and references therein).

After 1900, dry weight concentrations remained elevated through the mid-1950s before experiencing steady declines through recent times. The most recent intervals dated from 2000 – present contained an average concentration of 84 ng/g. The shape of the concentration profile is very similar to mercury profiles found at Lake Washington and Lake Sammamish approximately 35 miles to the north (Furl, 2007; 2008).

The Black Lake mercury concentration profile displayed an erratic pattern with several large concentration changes over a short time period. Mercury concentrations showed significant elevation over background levels by the mid-1950s reaching a local maximum near 1970 (290 ng/g). Concentrations declined briefly before a steady rise to the overall maximum value (389 ng/g) in the late 1990s. Concentrations in the uppermost horizon continued the erratic pattern dropping nearly four-fold from the maximum before increasing in the 0-2 cm horizon. The changes in concentration appeared to be strongly affected by sedimentation rates. Periods of increasing mercury concentrations were marked by falling sedimentation rates and vice versa. The diluting effect sedimentation can have on metals concentration has been observed in other coring studies (Engstrom et al., 2007).

Mercury concentrations throughout the Upper Twin Lake sediment core were found in a tight range (28.3 – 43.9 ng/g) varying relatively little compared to the other two lakes. Little trends information could be definitively discerned from the Upper Twin Lake sediment core due to very low concentrations. Peak concentrations occurred just after the 1900s and again in the mid 1990s. Recent concentrations displayed an overall declining trend with the upper two horizons indicating increases.

Fluxes

Mercury flux values ranged from 4 – 110 $\mu g/m^2/yr$. Flux profiles at American and Black Lake displayed surprisingly similar flux profiles considering their distinct locations, drainage land uses, and climate. Fluxes at both lakes peaked in the late 1960s (American = 110 $\mu g/m^2/yr$; Black = 96 $\mu g/m^2/yr$) and displayed a downward trend through recent times. Modern fluxes (after 2000) at Black Lake are estimated at 25.3 $\mu g/m^2/yr$ (n = 2). Modern fluxes at American Lake over the same time period are 23.1 $\mu g/m^2/yr$ (n = 4).

Fluxes at Upper Twin Lake displayed a similar pattern to the concentration profile. As with concentrations, the most recent fluxes display an overall declining trend with a recent increase in the uppermost horizon. Modern fluxes from 2000 – present are 22.9 $\mu g/m^2/yr$.

Table 5 presents flux rate ranges from 2006-09 coring reports along with estimates of modern fluxes.

Table 5. Mercury flux estimations from all coring reports as part of the 2006-09 studies.

Lake	Flux Range (µg/cm^2/yr)	Peak Year	Average Flux 2000 - present (µg/cm^2/yr)	n	Study
American	4 - 110	1969	23.1	4	Present study
Black	12 - 96	1969	25.3	2	
Upper Twin	8 - 46	1969	22.9	5	
Lacamas	14 - 77	1966	63.6	3	Furl et al., 2009
Offutt	14 - 77	1998	70.0	3	
Washington	12 - 386	1940	144.4	4	
Loon	2 - 22	1995	15.1	1	Furl, 2008
Wannacut	2 - 361	1932	10.5	1	
Ozette[2]	37 - 261	1997	212.8	2	Furl, 2007
Sammamish[2]	18 - 273	1934	94.0	3	
St. Clair[2]	17 - 271	1975	214.4	2	

[1] Number of measurements used in 2000 - present flux average.
[2] Fluxes not reported in Furl, 2007.

Lead

Figure 5 displays lead dry weight concentration profiles along with dates.

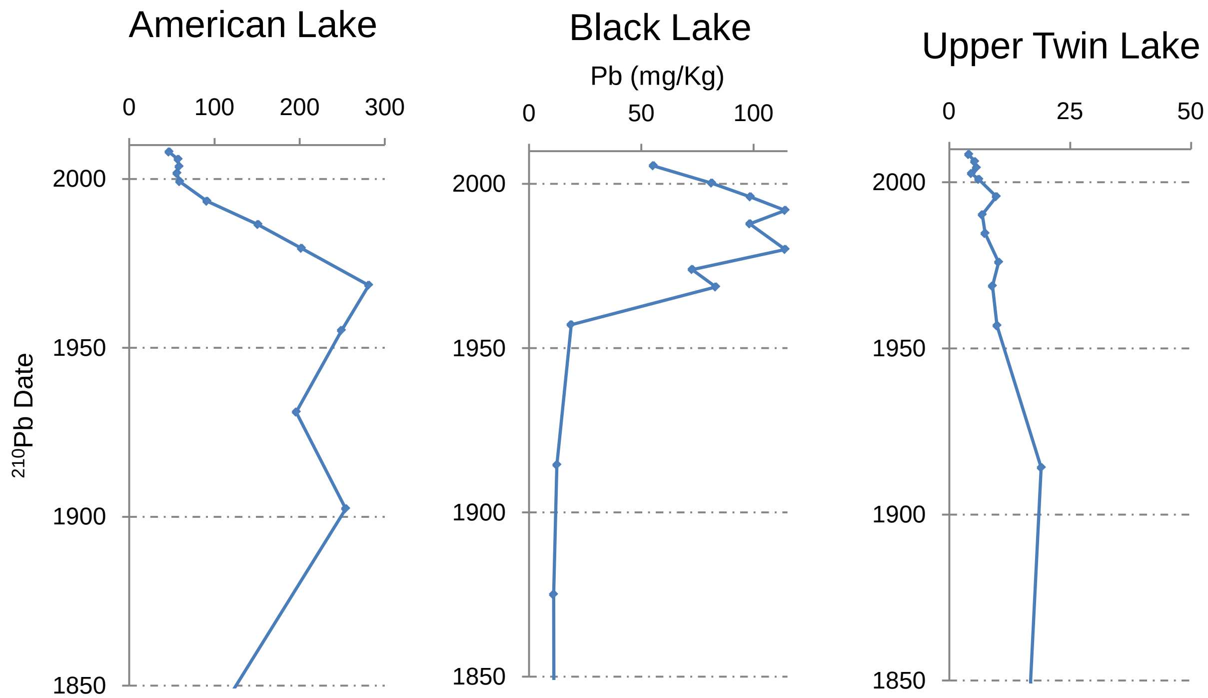

Figure 5. Lead concentration profiles with dates.
(note scale differences)

Lead concentrations in the three sediment cores ranged from 4.0 – 281 mg/kg. The highest concentrations were found at American Lake followed by Black Lake and Upper Twin Lake. Lead concentration profiles at each of the three lakes shared a similar shape with their mercury profiles. Concentrations at each of the three lakes are declining through recent times.

Lead concentrations at American Lake were highly elevated over background levels by 1900 due to contributions from the Asarco plant as discussed above. Lead levels remained elevated in the core through the 1960s. From 1970 onward, lead concentrations consistently declined until stabilizing over the last decade. Concentrations from 2000 - present range from 40 – 60 mg/kg.

At Black Lake, lead concentrations remained low, near background levels, through the 1950s. Concentrations increased until reaching their maximum in the latter half of the 20th century. Since the early 1990s, lead concentrations have consistently decreased to the present time. The most recent sediments contained concentrations near 50 mg/kg.

Lead concentrations at Upper Twin Lake are relatively low compared to the other two study lakes. The highest concentrations were near 20 mg/kg and found deep within the core. Concentrations have steadily declined through recent times. The higher sedimentation rates during the latter half of the 20th century seem to have a diluting effect on concentrations. Concentrations in the upper portion of the core range from 4 – 6 mg/kg.

Table 6 presents data related to lead concentrations found in the 2008 sediment core study.

Table 6. Lead concentrations from all coring reports as part of the 2008 and 2009 studies.

Lake	Concentration (mg/Kg)	Peak Year	Average Concentration 2000 – present (mg/Kg)	n[1]	Study
American	41.6-281	1969	54.4	4	Present study
Black	10.9-114	1980, 1992	68.3	2	
Upper Twin	4.03-21.7	< 1850	5.1	5	
Washington	5.7-200	1977	70.9	4	Furl et al., 2009
Offutt	6.28-49.5	1988	49.1	3	
Lacamas	13.5-50.7	1953	24.1	3	

[1] Number of measurements used in 2000 - present flux average.

PAHs

Figure 6 displays summed PAH totals (µg/kg ww) in the American Lake sediment core. Concentrations greater than the LOQ were found in every interval examined except the uppermost horizon (290U).

PAHs were infrequently detected in the two eastern Washington sediment cores. Concentrations ranging from 45 – 236 µg/kg were found in four intervals from the Black Lake core. Only two values were greater than the LOQ post-1950 at Black Lake (Appendix C). No PAHs were found greater than the LOQ (36 – 150 µg/kg) in the Upper Twin Lake core.

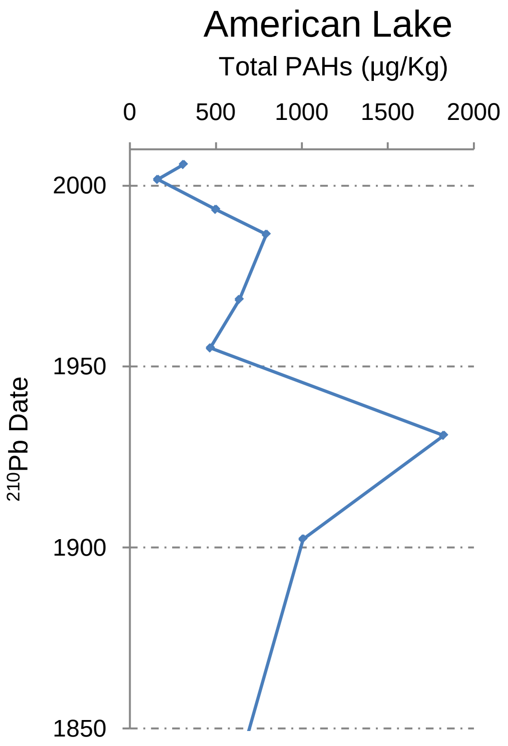

Figure 6. PAH concentrations in American Lake sediment intervals with dates.

As with the other contaminants analyzed, PAHs were highly elevated over background levels by 1900 at American Lake. The maximum concentration (1825 µg/kg) was found during the first half of the 20th century. After peaking, concentrations displayed an erratic pattern with overall substantial declines. Concentrations in the 2-3 and 0-2 interval were 311 and 290U µg/kg, respectively.

Lake Washington and American Lake sediment cores have contained the highest PAH concentrations from cores collected as part of this study. TOC-normalized concentrations (µg/kg organic carbon) for both cores are presented in Figure 7.

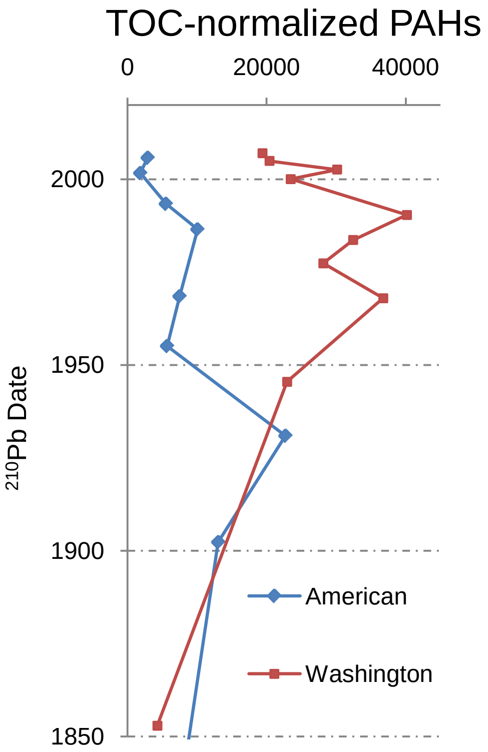

Figure 7. TOC-normalized PAH concentrations in sediment cores from American Lake and Lake Washington.

The PAH TOC-normalized concentration profiles are dissimilar in magnitude. Normalized concentrations were much higher at Lake Washington partly due to low TOC and finer sediment (TOC average = 3.0%; silt+clay = 75%) compared to American Lake (TOC average = 8.9%; silt+clay = 51.4%). PAH concentrations in American Lake reached their maximum much earlier (approximately 50 years) than Lake Washington. While both cores are experiencing downward trends, concentrations have fallen much more at American Lake.

Parent Ratios

Several parent PAH ratios have been proposed to discriminate between sources in environmental samples (Yunker et al., 2002; Budzinski et al., 1997). The ratio using fluoranthene and pyrene has been used to distinguish between petroleum, petroleum combustion, and grass, wood, and coal combustion. Yunker et al. (2002) proposes a fluoranthene / (fluoranthene + pyrene) (Fl/(Fl + Py)) ratio of 0.40 as the petroleum/combustion transition point. Additionally, Yunker et al. (2002) propose ratios between 0.40 – 0.50 to represent liquid fossil fuel combustion whereas values greater than 0.50 are characteristic of grass, wood, or coal combustion. Fl/(Fl+Py) ratios for American Lake are shown in Figure 8 along with ratios from the 2008 study lakes (Lacamas Lake, Offutt Lake, and Lake Washington).

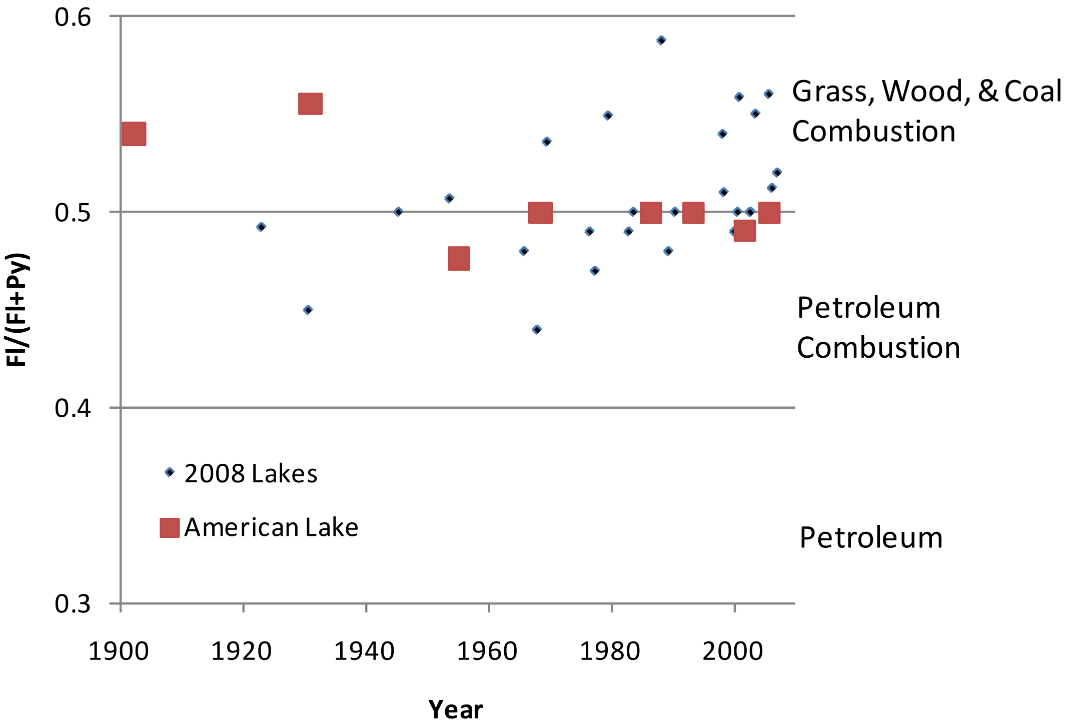

Figure 8. Fluoranthene / (Fluoranthene + Pyrene) ratios at American Lake and the 2008 study lakes.

Ratios from American Lake indicate mixed combustion sources are dominant in the lake. PAH ratios prior to 1950 suggested the main sources were grass, wood, and coal combustion. After 1950, the ratio decreased towards petroleum combustion associated with vehicular traffic.

Conclusions

This study was the fourth year of a five-year study on present and historical contaminant trends in Washington State lake sediments. Deep sediment cores were collected from American Lake, Black Lake, and Upper Twin Lake and analyzed for mercury, lead, and PAHs. Dates and sedimentation rates were assigned to sediment intervals from ^{210}Pb measurements using the constant rate of supply model.

American Lake contained the greatest historical contamination of all three contaminants. Lead, mercury, and PAH concentrations were all highly elevated over background concentrations by 1900 indicating the Asarco plant as a major area source of metals. All three contaminants displayed similar profiles marked by substantial declines over the latter half of the 1900s through recent times to near background levels. Mercury flux rates from 2000 – present averaged 23.1 μg/m^2/yr.

The Black Lake contaminant profile displayed an erratic mercury and lead pattern post-1950. Mercury concentrations in the upper 3 cm of the core have fallen substantially since the maximum value in the mid-1990s. Current mercury fluxes to the lake since 2000 average 25.3 μg/m^2/yr. Lead concentrations have displayed constant declines since reaching their maximum in the early 1990s. Concentrations before 1990 were characterized by an erratic pattern with an overall increasing trend. PAH concentrations ranged from 45 – 236 µg/Kg in four intervals from the Black Lake sediment core. There were not enough detections to describe PAH trends.

Mercury and lead concentrations at Upper Twin Lake were relatively low when compared to the other two lakes. Mercury concentrations remained within a narrow range over the entire length of the sediment core. Recent concentrations displayed an overall declining trend with the upper two horizons indicating increases. Average mercury flux over the last decade is 22.8 μg/m^2/yr. Lead concentrations have displayed near perfect monotonic declines through recent times since the maximum shortly after 1900. No PAHs were detected above the limit of quantification in sediment intervals.

Recommendations

The results of this study support the following recommendations.

- Sample suburban and urban lakes to determine trends in PAH contamination.
- Analyze mercury samples at a greater mass to lower detection limits in upper core intervals containing low percent solids.
- Consider other analytes (e.g., levoglucosan) that may be used to evaluate sources of PAHs.
- Lower detection limits in PAH samples.
- Obtain a longer sediment coring device so reservoirs and other high sedimentation systems can be cored.

References

Agency for Toxic Substances and Disease Registry (ATSDR), 1995. Toxicological Profile for Polycyclic Aromatic Hydrocarbons (PAHs). U.S. Department of Health and Human Services.

Agency for Toxic Substances and Disease Registry (ATSDR), 2007. Toxicological Profile for Lead. U.S. Department of Health and Human Services.

Anderson, W.W., A.O. Ness, and A.C. Anderson, 1955. Soil Survey of Pierce County, Washington. United States Department of Agriculture, Soil Conservation Service.

Appleby, P.G. and F. Oldfield, 1978. The Calculation of Lead-210 Dates Assuming a Constant Rate of Supply of Unsupported ^{210}Pb to the Sediment. Catena, Vol. 5: 1-8.

Blais J. and J. Kalff, 1995. The influence of lake morphometry on sediment focusing. Limnology and Oceanography 40:582-588.

Blakley, N., 2008. Standard Operating Procedure for Obtaining Freshwater Sediment Samples. Washington State Department of Ecology, Olympia, WA. SOP Number EAP040. www.ecy.wa.gov/programs/eap/quality.html.

Bortleson, G.C., N.P. Dion, J.B. McConnell and L.M. Nelson, 1976. Reconnaissance Data on Lakes in Washington, Volume 3 Kitsap, Mason, and Pierce Counties. Water-Supply Bulletin 43, Volume 3. United States Geological Survey in Cooperation with the Washington State Department of Ecology.

Budzinski, H., I. Jones, J. Bellocq, and others, 1997. Evaluation of sediment contamination by polycyclic aromatic hydrocarbons in the Gironde estuary. Marine Chemistry. Vol. 58: 85-97.

BLM (Bureau of Land Management), 2008. Sandygren Wetland Restoration Environmental Assessment: EA#OR135-FY07-EA-015. Bureau of Land Management, Spokane, WA. www.blm.gov/or/districts/spokane/plans/files/Sandygren%20EA.Maps.pdf.

Charles, M.J. and R.A. Hites, 1987. Paper/book chapter: Sediments as Archives of Environmental Pollution Trends. Copyright 1987. American Chemical Society. Book Title: Sources and Fates of Aquatic Pollutants. Editors: Ronald A. Hites and S.J. Eisenreich.

Coots, R., 2006. Quality Assurance Project Plan: Depositional History of Mercury in Selected Washington Lakes Determined from Sediment Cores. Washington State Department of Ecology, Olympia, WA. Publication No. 06-03-113. www.ecy.wa.gov/biblio/0603113.html.

Davies, H. and others, 2009. Washington State Lead Chemical Action Plan. Washington State Department of Ecology, Olympia, WA. Publication No. 09-07-008. www.ecy.wa.gov/pubs/0907008.pdf.

Dion, N.P., G.C. Bortleson, J.B. McConnell, and L.M. Nelson, 1976a. Reconnaissance Data on Lakes in Washington, Volume 4 Adams, Benton, Douglas, Franklin, Grant, Lincoln, Walla Walla, and Whitman Counties. Water-Supply Bulletin 43, Volume 4. United States Geological Survey in Cooperation with the Washington State Department of Ecology.

Dion, N.P., G.C. Bortleson, J.B. McConnell, and L.M. Nelson, 1976b. Reconnaissance Data on Lakes in Washington, Volume 7 Pend Oreille, Spokane, and Stevens Counties. Water-Supply Bulletin 43, Volume 7. United States Geological Survey in Cooperation with the Washington State Department of Ecology.

Engstrom, D.R. and E.B. Swain, 1997. Recent Declines in Atmospheric Mercury Deposition in the Upper Midwest. Environmental Science and Technology, 31: 960-967.

Engstrom, D.R., S. J. Balogh, and E. B. Swain, 2007. History of mercury inputs to Minnesota lakes: Influences of watershed disturbance and localized atmospheric deposition. Limnology and Oceanography, 52(6): 2467-2483.

Furl, C., 2007. 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. www.ecy.wa.gov/biblio/0703019.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. www.ecy.wa.gov/biblio/0803012.html.

Furl, C. and C. Meredith, 2008. Standard Operating Procedure for Collection of Freshwater Sediment Core Samples Using a Box or KB Corer. Washington State Department of Ecology, Olympia, WA. SOP Number EAP038. www.ecy.wa.gov/programs/eap/quality.html.

Furl, C., C. Meredith and M. Friese, 2009. Determination of PBT Chemical Trends in Selected Washington Lakes Using Age-Dated Sediment Cores: 2008 Sampling Results. Washington State Department of Ecology, Olympia, WA. Publication No. 09-03-030. www.ecy.wa.gov/biblio/0903030.html.

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

Gallagher, L., R.W. MacDonald, and D.W. Paton, 2004. The Historical Record of Metals in Sediments from Six Lakes in the Fraser River Basin, British Columbia. Water, Air, and Soil Pollution, 152: 257-278.

Geller, L., 2006. Washington State Polybrominated Diphenyl Ether (PBDE) Chemical Action Plan: Final Report. Washington State Department of Ecology, Olympia, WA. Publication No. 05-07-048. www.ecy.wa.gov/biblio/0507048.html.

Glass, G., 2003. Credible Evidence Report: The ASARCO Tacoma Smelter and Regional Soil Contamination in Puget Sound, Final Report. Tacoma Pierce County Health Department and Washington State Department of Ecology. www.ecy.wa.gov/programs/tcp/sites/tacoma_smelter/Sources/Credible_Evidence/web%20pieces/Credfinl.pdf.

Jack, Richard, 2003. Investigation of Background Inorganic and Organic Arsenic in Four Washington Lakes. Washington State Department of Ecology, Olympia, WA. Publication No. 0303024. www.ecy.wa.gov/pubs/0303024.pdf.

Kamman, N.C. and D.R. Engstrom, 2002. Historical and present fluxes of mercury to Vermont and New Hampshire lakes inferred from ^{210}Pb dated sediment cores. Atmospheric Environment 36: 1599 – 1609.

Lowe, J., 2010. Personal communication. United States Bureau of Reclamation, Spokane, WA.

Meredith, C. and C. Furl, 2008. Addendum #1 to Quality Assurance Project Plan: Depositional History of Mercury in Selected Washington Lakes Determined from Sediment Cores. Washington State Department of Ecology, Olympia, WA. Publication No. 06-03-113ADD. www.ecy.wa.gov/biblio/0603113add1.html.

NRCS (National Resource Conservation Service), 2006. Rapid Watershed Assessment HUC 170200003: Colville Watershed. ftp-fc.sc.egov.usda.gov/WA/Tech/RWA/Profiles/colville_FINAL_062707.pdf.

Nevissi, A.E., 1985. Measurement of ^{210}Pb Atmospheric flux in the Pacific Northwest. Health Physics. 45(2): 169-174.

Oldfield, F. and P.G. Appleby, 1984. Empirical testing of ^{210}Pb dating models. In Haworth, E.Y. & J.G. Lund (eds.). Lake Sediments and Environmental History. Leicester Univ. Press: 93-124.

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, 98 pages. Prepared in Cooperation with the Whatcom County Health Department and the Washington State Department of Ecology.

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

Rudd, J.W.M., 1995. Sources of methylmercury to freshwater ecosystems: a review. Water, Air, Soil Pollution. 80: 697 – 713.

USEPA (United States Environmental Protection Agency). Toxics Release Inventory, 2006. www.epa.gov/triexplorer. Accessed 10 February 2008.

USEPA (United States Environmental Protection Agency), 1986. Recommended Protocols for Measuring Conventional Sediment Variables in Puget Sound. 49 p.

Van Metre, P. and E. Callendar, 2000. Urban Sprawl Leaves its PAH Signature. Environmental Science and Technology. Vol. 34: 4064-4070.

Van Metre, P., J. Wilson, C. Fuller, E. Callendar, and B. Mahler, 2004. Collection, Analysis and Age-Dating of Sediment Cores From 56 U.S. Lakes and Reservoirs Sampled by the U.S. Geological Survey, 1992–2001. United States Geological Survey. http://pubs.usgs.gov/sir/2004/5184/.

Walters, K.L. and G.E. Kimmel, 1968. Ground-water occurrence and stratigraphy of unconsolidated deposits, central Pierce County, Washington. Water Supply Bulletin 22. United States Geological Survey in cooperation with the Washington Department of Water Resources.

Washington State Department of Ecology, 1995. Sediment Management Standards, Chapter 173-204 WAC. Washington State Department of Ecology. Publication No. 96-252. Revised December, 1995. www.ecy.wa.gov/pubs/96252.pdf.

Yake, B., 2001. The Use of Sediment Cores to Track Persistent Pollutants in Washington State - A Review. Washington State Department of Ecology, Olympia, WA. Publication No. 01-03-001. www.ecy.wa.gov/biblio/0103001.html.

Yunker, M.B, R.W. Macdonald, R. Vingarzan, and others, 2002. PAHs in the Fraser River basin: a critical appraisal of PAH ratios as indicators of PAH source and composition. Organic Geochemistry. Vol. 33: 489-515.

Appendices

This page is purposely left blank

Appendix A. Glossary, Acronyms, and Abbreviations

Glossary

Analyte: Constituent being measured (parameter).

Anthropogenic: Human-caused.

Bioaccumulative: A substance that, once introduced, remains, or accumulates, in an organism.

Methylmercury: Mercury, Hg, with a methyl atom attached, CH_3Hg^+, or more commonly MeHg. Methylmercury is an organic and more toxic and bioavailable form of mercury than inorganic, or elemental, mercury.

Lipophilic: Readily absorbs to oils.

Nonpoint source: Unconfined and diffuse sources of contamination. Pollution that enters water from dispersed land-based or water-based activities. This includes, but is not limited to, atmospheric deposition, surface water runoff from agricultural lands, urban areas, or forest lands, subsurface or underground sources, or discharges from boats or marine vessels not otherwise regulated under the National Pollutant Discharge Elimination System program.

Parameter: Water quality constituent being measured (analyte). A physical, chemical, or biological property whose values determine environmental characteristics or behavior.

Point source: Source of pollution that discharge at a specific location from pipes, outfalls, and conveyance channels to a surface water. Examples of point source discharges include municipal wastewater treatment plants, municipal stormwater systems, industrial waste treatment facilities, and construction sites that clear more than 5 acres of land.

Pollution: Such contamination, or other alteration of the physical, chemical, or biological properties, of any waters of the state. This includes change in temperature, taste, color, turbidity, or odor of the waters. It also includes discharge of any liquid, gaseous, solid, radioactive, or other substance into any waters of the state. This definition assumes that these changes will, or are likely to, create a nuisance or render such waters harmful, detrimental, or injurious to (1) public health, safety, or welfare, or (2) domestic, commercial, industrial, agricultural, recreational, or other legitimate beneficial uses, or (3) livestock, wild animals, birds, fish, or other aquatic life.

Sediment: Solid fragmented material (soil and organic matter) that is transported and deposited by water and covered with water (example, river or lake bottom).

Sediment core: Vertical sediment sample.

Sediment horizon: Vertical sub-section of a sediment core.

Spatial trends: How contaminant trends differ among various geographical regions.

Surface waters of the state: Lakes, rivers, ponds, streams, inland waters, salt waters, wetlands and all other surface waters and watercourses within the jurisdiction of Washington State.

Temporal trends: Characterize trends over time.

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

^{210}Pb: Isotopic lead with a 22.3 year half-life. Part of the uranium-238 decay chain.

Acronyms and Abbreviations

Following are acronyms and abbreviations used frequently in this report.

CRS	Constant Rate of Supply Model
Ecology	Washington State Department of Ecology
EIM	Environmental Information Management database
EPA	U.S. Environmental Protection Agency
MEL	Manchester Environmental Laboratory
LOQ	Limit of quantification
MQO	Measurement quality objective
PAHs	Polycyclic aromatic hydrocarbons
PBDE	Polybrominated diphenyl ethers
^{210}Pb	Lead ^{210} (See Glossary above)
PBT	Persistent, bioaccumulative, and toxic substance
PSEP	Puget Sound Estuary Protocol
RPD	Relative percent difference
RSD	Relative standard deviation
SD	Standard deviation
SMARs	Sediment mass accumulation rates
SOP	Standard operating procedures
SRM	Standard reference materials
TOC	Total organic carbon
USGS	U.S. Geological Survey

Units of Measurement

°C	degrees centigrade
cm	centimeter, a unit of distance
Ci	curie, a unit of radioactivity equivalent to 3.7×10^{10} decays per second
dw	dry weight
ft	feet

g	gram, a unit of mass
g/cm^2	grams per square centimeter per year
g/cm^2/yr	gram per square centimeter per year
in	inch, a unit of distance
kg	kilograms, a unit of mass equal to 1,000 grams.
m	meter
m^2	square meter
ng/g	nanograms per gram (parts per billion)
ng/Kg	nanograms per kilogram (parts per trillion)
oz.	ounce, a unit of volume
p	pico, or 10^{-12}
pCi/cm^2	picocurie per square centimeter
pCi/cm^2/yr	picocurie per square centimeter per year
pCi/g	picocuries per gram
pg/g	picograms per gram (parts per trillion)
µ	micro, or 10^{-6}
µg/cm^2/yr	micrograms per square centimeter per year
µg/g	micrograms per gram (parts per million)
µg/Kg	micrograms per kilogram (parts per billion)
µg/m^2/yr	micrograms per square meter per year
ww	wet weight

Appendix B. PBT Trends Sediment Coring Locations

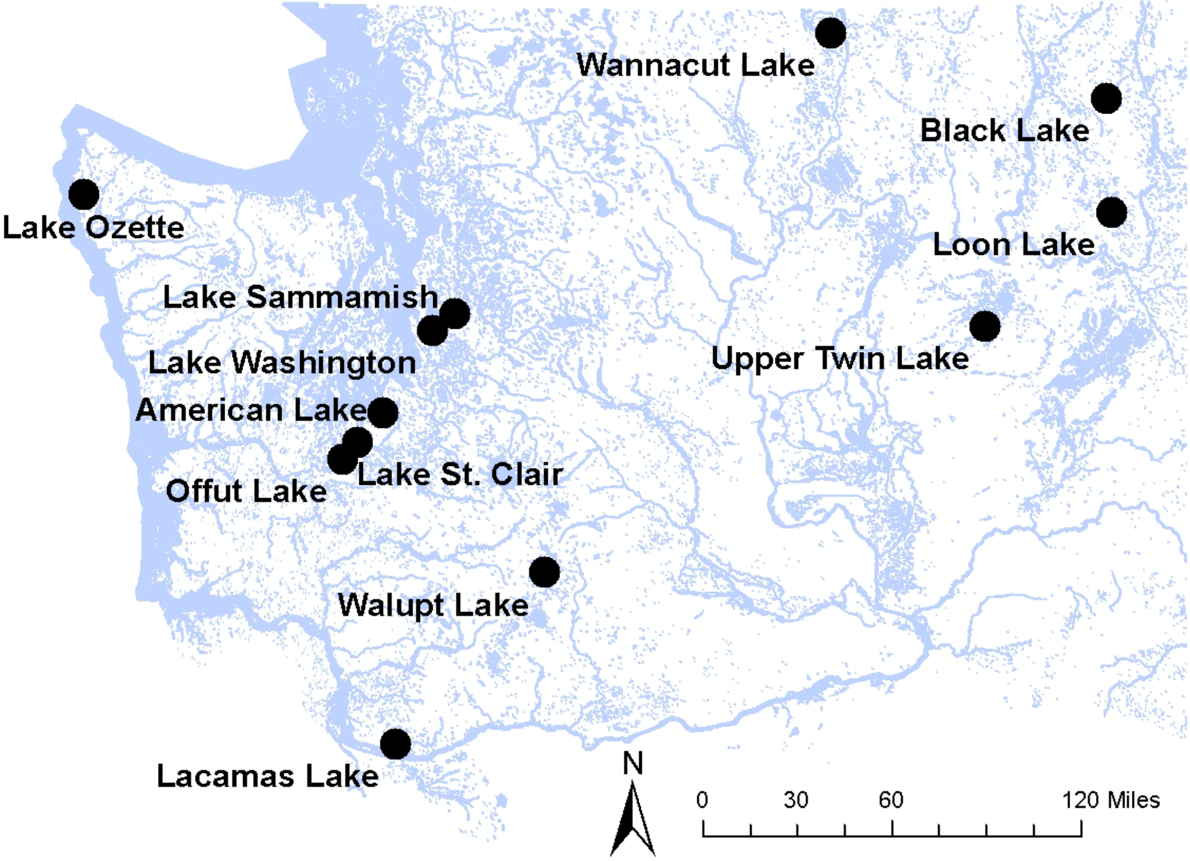

Figure B-1. Locations of sediment coring sampling stations, 2006-09.

Appendix C. Sediment Core and Surface Sediment Data

Table C-1. Black Lake core sectioning, collected September 16, 2009

Table C-2. Upper Twin Lake core sectioning, collected September 15, 2009

Table C-3. American Lake core sectioning, collected August 28, 2009

Table C-1. Black Lake core sectioning, collected September 16, 2009.

Section (cm intervals)	Analyte					Field ID	Sample No.
	Mercury (ng/g)	Lead (mg/Kg)	Total PAHs (ug/Kg)	TOC (%)	^{210}Pb (pCi/g)		
0-2	216	55.3	210U	14.1	59.2	BLK01	0908050-01
2-3	102	81.3	150U	12.3	33.3	BLK02	0908050-02
3-4	389	98.5	----	----	42.2	BLK03	0908050-03
4-5	306	114	190U	15.5	30.5	BLK04	0908050-04
5-6	274	98.3	----	----	28.4	BLK05	0908050-05
6-7	----	----	----	----		----	----
7-8	204	114	70J	15.8	20.8	BLK06	0908050-06
8-9	----	----	----	----		----	----
9-10	154	72.6	236J	15.3	14.5	BLK07	0908050-07
10-11	----	----	----	----		----	----
11-12	290	83.1			14.8	BLK08	0908050-08
12-13	----	----	----	----		----	----
13-14	----	----	----	----		----	----
14-15	226	18.7	150UJ	19.2	17.1	BLK09	0908050-09
15-16	----	----	----	----		----	----
16-17	----	----	----	----		----	----
17-18	----	----	----	----		----	----
18-19	----	----	----	----		----	----
19-20	44.4	4.14	41U	4.83		BLK10	0908050-10
20-21	----	----	----	----		----	----
21-22	115	12.4	45J	16.2	8.02	BLK11	0908050-11
22-23	----	----	----	----		----	----
23-24	----	----	----	----		----	----
24-25	----	----	----	----		----	----
25-26	----	----	----	----		----	----
26-27	98	10.9	110UJ	13.2	7.72	BLK12	0908050-12
27-28	----	----	----	----		----	----
28-29	----	----	----	----		----	----
29-30	----	----	----	----		----	----
30-31	----	----	----	----		----	----
31-32	----	----	----	----		----	----
32-33	72	11.1	----	----	10.2	BLK13	0908050-13
33-34	----	----	----	----		----	----
34-35	----	----	----	----		----	----
35-36	----	----	----	----		----	----
36-37	----	----	----	----		----	----
37-38	----	----	----	----		----	----
38-39	112	12.3	93J	17.1	6.75	BLK14	0908050-14
39-40	----	----	----	----		----	----
40-41	----	----	----	----		----	----
41-42	----	----	----	----		----	----
42-43	114	13.0	----	----	10.1	BLK15	0908050-15

J - Analyte positively identified, approximate concentration.
U – Analyte not detected at detection limit shown.

Table C-2. Upper Twin Lake core sectioning, collected September 15, 2009.

Section (cm intervals)	Analyte					Field ID	Sample No.
	Mercury (ng/g)	Lead (mg/Kg)	Total PAHs (ug/Kg)	TOC (%)	^{210}Pb (pCi/g)		
0-2	39.6J	4.03	150U	7.19	2.850	UT01	0908050-17
2-3	37.2J	5.25	130UJ	5.94	3.840	UT02	0908050-18
3-4	29J	5.61	----	----	2.420	UT03	0908050-19
4-5	34.2J	4.59	110U	5.46	2.190	UT04	0908050-20
5-6	33.2J	6.10	----	----	1.880	UT05	0908050-21
6-7	----	----	----	----		----	----
7-8	43.1	9.71	62U	5.85	1.980	UT06	0908050-22
8-9	----	----	----	----		----	----
9-10	37.9J	6.82	110U	4.80	2.480	UT07	0908050-23
10-11	----	----	----	----		----	----
11-12	37J	7.39	----	----	1.850	UT08	0908050-24
12-13	----	----	----	----		----	----
13-14	----	----	----	----		----	----
14-15	38.3	10.2	82U	3.99	1.650	UT09	0908050-25
15-16	----	----	----	----		----	----
16-17	----	----	----	----		----	----
17-18	38.6	8.91	88U	4.62	1.110	UT10	0908050-26
18-19	----	----	----	----		----	----
19-20	----	----	----	----		----	----
20-21	----	----	----	----		----	----
21-22	28.3	9.86	66U	3.34	1.420	UT11	0908050-27
22-23	----	----	----	----		----	----
23-24	----	----	----	----		----	----
24-25	----	----	----	----		----	----
25-26	----	----	----	----		----	----
26-27	43.9	19.0	53U	4.31	1.140	UT12	0908050-28
27-28	----	----	----	----		----	----
28-29	----	----	----	----		----	----
29-30	----	----	----	----		----	----
30-31	39	16.8	----	----	0.642	UT13	0908050-29
31-32	----	----	----	----		----	----
32-33	----	----	----	----		----	----
33-34	----	----	----	----		----	----
34-35	----	----	----	----		----	----
35-36	46	21.7	36U	3.66	0.782	UT14	0908050-30
36-37	----	----	----	----		----	----
37-38	----	----	----	----		----	----
38-39	----	----	----	----		----	----
39-40	42.5	19.8	----	----	0.995	UT15	0908050-31

J - Analyte positively identified, approximate concentration
U – Analyte not detected at detection limit shown

Table C-3. American Lake core sectioning, collected August 28, 2009.

Section (cm intervals)	Analyte					Field ID	Sample No.
	Mercury (ng/g)	Lead (mg/Kg)	Total PAHs (ug/Kg)	TOC (%)	^{210}Pb (pCi/g)		
0-2	60.7J	46.5	290U	10.9	9.61	AMER01	0908050-33
2-3	88.7J	57.2	311J	10.7	10.60	AMER02	0908050-34
3-4	91.3J	58.3	----	----	6.09	AMER03	0908050-35
4-5	95.9	55.8	161J	8.79	6.56	AMER04	0908050-36
5-6	102	59.1	----	----	6.34	AMER05	0908050-37
6-7	----	----	----	----		----	----
7-8	150	91.0	499	9.06	6.82	AMER06	0908050-38
8-9	----	----	----	----		----	----
9-10	222	151	793	7.89	5.02	AMER07	0908050-39
10-11	----	----	----	----		----	----
11-12	225	202	----	----	4.44	AMER08	0908050-40
12-13	----	----	----	----		----	----
13-14	----	----	----	----		----	----
14-15	402	281	636	8.50	2.87	AMER09	0908050-41
15-16	----	----	----	----		----	----
16-17	----	----	----	----		----	----
17-18	495	249	468	8.23	3.05	AMER10	0908050-42
18-19	----	----	----	----		----	----
19-20	----	----	----	----		----	----
20-21	495	196	1825	8.04	2.00	AMER11	0908050-43
21-22	----	----	----	----		----	----
22-23	----	----	----	----		----	----
23-24	----	----	----	----		----	----
24-25	588	254	1007	7.72	2.02	AMER12	0908050-44
25-26	----	----	----	----		----	----
26-27	----	----	----	----		----	----
27-28	----	----	----	----		----	----
28-29	417	95.7	----	----	0.45	AMER13	0908050-45
29-30	----	----	----	----		----	----
30-31	----	----	----	----		----	----
31-32	----	----	----	----		----	----
32-33	158	52.9	201	9.02	0.58	AMER14	0908050-46
33-34	----	----	----	----		----	----
34-35	81.8	41.6	----	----	0.58	AMER15	0908050-47

J - Analyte positively identified, approximate concentration.
U – Analyte not detected at detection limit shown.

Appendix D. Quality Assurance Data

Table D-1. Measurement Quality Objectives.

Parameter	Surrogate % Recovery	SRM % Recovery	MS % Recovery	MSD RPD	Duplicate RPD	LCS % Recovery	Blank
Total Organic Carbon	---	75-125	---	---	< 20%	----	< LOQ
Grain Size	---	----	----	---	< 25%	----	---
Total Mercury	---	----	75 - 125	< 20%	---	85 - 115	< LOQ
Total Lead	---	----	75 - 125	< 20%	---	85 - 115	< LOQ
PAHs	2-Fluorobiphenyl - 30-115% Pyrene-D-10 - 50-150% Terphenyl-D-14- 18-137%	65-135	50-15	<40%	< 40%	50-150	< LOQ
Lead^{210}	---	----	---	---	<30%	80-120	< LOQ

SRM – Standard Reference Material.
MS/MSD – Matrix Spike/ Matrix Spike Duplicate.
RPD – Relative Percent Difference.
LCS – Laboratory Control Sample.
LOQ – Limit of Quantitation.

Table D-2. Total Organic Carbon.

Lab Duplicate

Sample	Result	Source Sample Result	RPD (%)
	(mg/kg dw)	(mg/kg dw)	
B09I238-DUP1	13.8	14.1	2
B09I238-DUP2	13.6	14.1	3
B09I239-DUP1	11.4	10.9	4
B09I239-DUP2	11.4	10.9	4

Lab Method Blanks

Sample	Field ID	Result
		(mg/kg dw)
B09I239-BLK1	Lab BLNK	0.10 U

Reference Sample

Reference	% Recovery
B09I238-SRM1	96
B09I239-SRM1	90

RPD – Relative Percent Difference.
U – Analyte not detected at detection limit shown.

Table D-3. Mercury.

Lab Duplicate

Sample	Result	Source Sample Result	RPD (%)
	(mg/kg dw)	(mg/kg dw)	
B09I244-DUP1	.0060 U	.0059 U	NC

Matrix Spikes (% Recovery)

Sample	LMX1	LMX2	RPD (%)
B09I235-MS1/MSD1	97	97	3
B09I244-MS1/MSD1	75	93	21
B09I201-MS1/MSD1	91	93	2

Lab Method Blanks

Sample	Field ID	Result
		(mg/kg dw)
B09I235-BLK1	Lab BLNK	0.0050U
B09I244-BLK1	Lab BLNK	0.0050U
B09I244-BLK2	Lab BLNK	0.0050U
B09I201-BLK1	Lab BLNK	0.0050U
B09I261-BLK1	Lab BLNK	0.10U
B09J049-BLK1	Lab BLNK	0.10U

Lab Control Samples

Sample	Field ID	% Recovery
B09I235-BS1	Lab LCS	108
B09I244-BS1	Lab LCS	109
B09I201-BS1	Lab LCS	105

RPD – Relative Percent Difference.
U – Analyte not detected at detection limit shown.
NC – Not calculated, both results undetected at level shown.

Table D-4. Lead.

Matrix Spikes (% Recovery)

Sample	LMX1	LMX2	RPD (%)
B09I261-MS1/MSD1	98	97	0.9
B09J049-MS1/MSD1	98	109	8
B09J066-MS1/MSD1	99	97	0.8

Lab Method Blanks

Sample	Field ID	Result
		(mg/kg dw)
B09I261-BLK1	Lab BLNK	0.10U
B09J049-BLK1	Lab BLNK	0.10U
B09J066-BLK1	Lab BLNK	0.10U

Lab Control Samples

Sample	Field ID	% Recovery
B09I261-BLK1	Lab LCS	101
B09J049-BS1	Lab LCS	103
B09J066-BS1	Lab LCS	95

RPD – Relative Percent Difference.
U – Analyte not detected at detection limit shown.

Table D-5. ^{210}Lead.

Lab Method Blanks

Sample	Result (pCi/total)
WG8648-1	3.08U
WG8663-1	1.36U
WG8666-1	1.36U

Lab Control Samples

Sample	% Recovery
WG8648-2	102.0
WG8648-3	90.7
WG8648-4	85.7
WG8648-5	104.6
WG8648-6	86.3
WG8663-2	97.3
WG8666-2	97.5

Lab Duplicates

Sample	Original (pCi/g)	Duplicate (pCi/g)	RPD (%)
WG8648-7	3.03	4.09	29.8
WG8663-3	< 2.78	< 2.82	*
WG8666-3	0.782	0.965	**

* – Not calculated.
** – Not calculated since results are < 5x method detection limits. .
U – Analyte not detected at detection limit shown.

Table D-6. PAHs.

Lab Duplicates

Analyte	Sample No. 0908050-12			Sample No. 0908050-13		
	Result	LDP1	RPD	Result	LDP1	RPD
	(µg/kg)		(%)	(µg/kg)		(%)
1-Methylnaphthalene	110U	110U		130U	130U	
2-Chloronaphthalene	110U	110U		130U	130U	
2-Methylnaphthalene	110U	110U		130U	130U	
Acenaphthene	110U	110U		130U	130U	
Acenaphthylene	110U	110U		130U	130U	
Anthracene	110U	110U		130U	130U	
Benzo(a)anthracene	110U	110U		130U	130U	
Benzo(a)pyrene	110UJ	110UJ		130U	130U	
Benzo(b)fluoranthene	110UJ	110UJ		**34NJ**	**62NJ**	59
Benzo(ghi)perylene	110UJ	110UJ		130U	130U	
Benzo(k)fluoranthene	110UJ	110UJ		130U	130U	
Carbazole	110U	110U		130U	130U	
Chrysene	110U	110U		130U	130U	
Dibenzo(a,h)anthracene	110UJ	110UJ		130U	130U	
Dibenzofuran	110U	110U		130U	130U	
Fluoranthene	110U	110U		**69J**	**140J**	70
Fluorene	110U	110U		130U	130U	
Indeno(1,2,3-cd)pyrene	110UJ	110UJ		130U	130U	
Naphthalene	110U	110U		130U	130U	
Phenanthrene	110U	110U		**52J**	**100J**	66
Pyrene	110U	110U		**46J**	**92J**	67
Retene	110U	110U		130U	130U	

Table D-6 (continued). PAHs.

Matrix Spikes

Analyte	Sample No. 0908050-10			Sample No. 0908050-44		
	LMX1	LMX2	RPD	LMX1	LMX2	RPD
	% Recovery		(%)	% Recovery		(%)
1-Methylnaphthalene	89	88	7	86	86	1
2-Chloronaphthalene	89	87	5	85	82	3
2-Methylnaphthalene	95	95	7	92	93	2
Acenaphthene	94	93	6	90	88	2
Acenaphthylene	94	92	6	91	88	2
Anthracene	101	100	7	101	98	2
Benzo(a)anthracene	98	97	7	99	96	3
Benzo(a)pyrene	97	95	6	95	92	3
Benzo(b)fluoranthene	106	96	2	99	94	5
Benzo(ghi)perylene	41	51	29	78	81	5
Benzo(k)fluoranthene	111	108	4	105	90	14
Carbazole	101	100	7	101	98	2
Chrysene	98	98	7	100	97	3
Dibenzo(a,h)anthracene	55	69	30	92	95	5
Dibenzofuran	94	93	6	92	89	3
Fluoranthene	99	98	7	99	95	3
Fluorene	98	96	6	96	92	3
Indeno(1,2,3-cd)pyrene	53	66	29	91	94	4
Naphthalene	89	89	8	82	82	0.6
Phenanthrene	99	98	6	98	95	3
Pyrene	100	100	8	101	97	3
Retene	101	101	7	103	99	3

Table D-6 (continued). PAHs.

Lab Method Blanks

Analyte	Sample No. B09I217-BLK1	Sample No. B09I227-BLK1
	(µg/kg dw)	
1-Methylnaphthalene	25U	25U
2-Chloronaphthalene	25U	25U
2-Methylnaphthalene	25U	25U
Acenaphthene	25U	25U
Acenaphthylene	25U	25U
Anthracene	25U	25U
Benzo(a)anthracene	25U	25U
Benzo(a)pyrene	25U	25U
Benzo(b)fluoranthene	25U	25U
Benzo(ghi)perylene	25U	25U
Benzo(k)fluoranthene	25U	25U
Carbazole	25U	25U
Chrysene	25U	25U
Dibenzo(a,h)anthracene	25U	25U
Dibenzofuran	25U	25U
Fluoranthene	25U	25U
Fluorene	25U	25U
Indeno(1,2,3-cd)pyrene	25U	25U
Naphthalene	25U	25U
Phenanthrene	25U	25U
Pyrene	25U	25U
Retene	25U	25U

Table D-6 (continued). PAHs.

Lab Control Samples

Analyte	Sample No. B09I217-BS1	Sample No. B09I227-BS1
	% Recovery	
1-Methylnaphthalene	77	83
2-Chloronaphthalene	77	79
2-Methylnaphthalene	82	89
Acenaphthene	80	84
Acenaphthylene	81	84
Anthracene	89	96
Benzo(a)anthracene	91	96
Benzo(a)pyrene	87	92
Benzo(b)fluoranthene	87	95
Benzo(ghi)perylene	94	100
Benzo(k)fluoranthene	92	94
Carbazole	92	98
Chrysene	92	96
Dibenzo(a,h)anthracene	96	103
Dibenzofuran	81	85
Fluoranthene	91	97
Fluorene	85	90
Indeno(1,2,3-cd)pyrene	98	104
Naphthalene	77	80
Phenanthrene	88	94
Pyrene	92	97
Retene	92	97

Table D-6 (continued). PAHs.

Lab Reference Samples

Analyte	Sample No. B09I217-SRM1	Sample No. B09I227-SRM1
	% Recovery	
1-Methylnaphthalene	52	52
2-Methylnaphthalene	45	44
Acenaphthene	46	43
Anthracene	53	50
Benzo(a)anthracene	88	82
Benzo(a)pyrene	84	78
Benzo(b)fluoranthene	127	99
Benzo(ghi)perylene	24	22
Benzo(k)fluoranthene	118	90
Chrysene	116	107
Dibenzo(a,h)anthracene	53	51
Fluoranthene	92	87
Fluorene	41	39
Indeno(1,2,3-cd)pyrene	26	25
Naphthalene	44	42
Phenanthrene	91	84
Pyrene	85	83

RPD – Relative Percent Difference.
U – Analyte not detected at detection limit shown.
NJ – Analyte tentatively identified, concentration is estimate.
J – Analyte positively identified, concentration is estimate.

Table D-7. Grain Size.

	0908050-48 (grams)	0908050-48DUP (grams)	0908050-48TRIP (grams)	RPD (%)
Gravel	0.0287	0.0695	0.0271	101.5
Sand	0.958	1.0178	1.0075	6.0
Silt	0.725	0.655	0.68	10.2
Clay	0.57	0.525	0.605	14.1

RPD – Relative Percent Difference.