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Mercury Sources to Lake Ozette and Lake Dickey: Highly Contaminated Remote Coastal Lakes, Washington State, USA

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Abstract Mercury concentrations in largemouth bass and mercury accumulation rates in age-dated sediment cores were examined at Lake Ozette and Lake Dickey in Washington State. Goals of the study were to compare concentrations in fish tissues at the two lakes with a larger statewide dataset and examine mercury pathways to the lakes. After accounting for fish length, tissue concentrations at the lakes were significantly higher than other Washington State lakes. Wet deposition and historical atmospheric monitoring from the area show no indication of enhanced local or regional deposition. Sediment core records from the lakes indicate rising sedimentation rates coinciding with logging in the lakes' drainages has greatly increased the net flux of mercury to the waterbodies.

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1 Introduction

Mercury contamination of aquatic food webs is a widespread global phenomenon with mercury levels found in remote aquatic ecosystems rendering fish unsuitable for consumption (Fitzgerald et al. 1998). Anthropogenic mercury emissions as a result of coal combustion and waste incineration have severely altered the natural mercury cycle. Once mercury is emitted to the atmosphere, it can be transported globally depending on its chemical speciation (Schroeder and Munthe 1998).

The complex biogeochemical cycling characteristics of mercury make it difficult to identify the important factors influencing loading and subsequent biological uptake at any single lake location. Land-use activity leading to increased soil erosion can result in increased mercury export from watersheds, enhancing fluxes to waterbodies (Engstrom et al. 2007; Grigal 2002). Additionally, atmospheric processes influencing mercury speciation may have implications for enhanced loading in coastal settings (Malcolm and Keeler 2003).

Recent mercury monitoring of largemouth bass (*Micropterus salmoides*) in Washington State has resulted in a large database to evaluate differences in

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tissue concentrations among lakes (Fischnaller et al. 2003; Furl et al. 2007; Furl 2007a; Furl and Meredith 2008). Unexpectedly, the highest mercury concentrations were found at Lake Ozette and Lake Dickey located in the remote coastal region of the Olympic Peninsula. We examined available mercury concentrations among largemouth bass from 24 lakes in Washington State, mercury accumulation in age-dated sediment cores from Lake Ozette and Lake Dickey, and wet deposition data from a national Mercury Deposition Network (MDN) station near Lake Ozette and Lake Dickey.

Specific goals of the study are to assess sources and pathways of mercury to Lake Ozette and Lake Dickey and compare their levels of mercury contamination in largemouth bass with other Washington State lakes to determine the extent of tissue contamination.

2 Methods

2.1 Study Area

Located within the coastal strip of the Olympic National Park 5 km from the Pacific Ocean, Lake Ozette is the third largest natural lake in Washington State with a surface area of 29.5 km^2 and an average depth of 40 m (Bortleson et al. 1976; Fig. 1). The National Park Service owns 15% of the 118-km^2 drainage basin, while over 80% of the lake catchment is zoned as commercial forest land. Approximately 60% of the Ozette drainage basin flows to the lake by three large creeks. In addition to three main inflows, numerous unnamed perennial streams contribute surface water to Lake Ozette. The lake is drained by the Ozette River at its north end into the Pacific Ocean. The average lake level is 10 m above sea level; drainage basin elevations range up to 580 m. Watershed geology consists of glacio-fluvial deposits situated between resistant marine deposited sedimentary rocks. Human population of the Lake Ozette watershed is estimated to be less than 100 (Haggerty et al. 2007).

Lake Dickey is located approximately 10 km directly east of Lake Ozette outside of the Olympic National Park at 59 m above sea level (Fig. 1). The lake is considerably smaller than Ozette with an area of 2 km^2 and an average depth of 7.6 m. The lake receives perennial inputs from the 38.1-km^2 drainage basin and is drained by a small outflow at its south end flowing to the Quillayute River (Bortleson et al. 1976).

Coastal temperate rainforests dominated by coniferous species cover the lake catchments. Commercial logging is the largest land-use activity with private timber companies owning the majority of the land.

Fig. 1 Location of study lakes along with major population centers in Oregon, Washington, and British Columbia

Seattle, WA, USA and Vancouver, BC, Canada are the nearest urban population centers located approximately 180 km to the east. Climate in the area can be characterized as temperate coastal marine, resulting in mild winters and cool summers. Average annual precipitation in the area regularly exceeds 250 cm per year with greater than 80% occurring between October and April. Fog drip also contributes a large amount to ground surface precipitation. Air flows from the west occur greater than 50% of the time at the nearest weather station 20 km south of Lake Ozette (Haggerty et al. 2007).

Seventeen largemouth bass were collected from Lake Ozette and Lake Dickey for mercury analysis. In addition to Lake Ozette and Lake Dickey, mercury tissue concentrations were examined among 22 additional Washington State lakes (referred to as statewide lakes) to determine if tissue concentrations at Lake Ozette and Lake Dickey differed significantly from the statewide lakes. The statewide group contains lakes with wide ranging morphology, hydrology, and land uses (Fischnaller et al. 2003; Furl et al. 2007; Furl 2007a; Furl and Meredith 2008; Fig. 1 and Table 1).

Table 1 Lake information

Lake	Surface area (km^2)	Drainage area (km^2)	Max depth (m)	Avg. depth (m)	Collection date	Avg. rainfall 1982–2007 (cm)	Study
Dickey	2.0	38.1	13.7	7.6	8/15/2007	276.7	Colman et al., submitted for publication[a]
Ozette	29.5	118.0	97.5	40.0	9/12/2007	250.0	Furl and Meredith 2008[b]
Deer	4.5	47.1	22.9	15.8	9/18/2007	53.6	Furl and Meredith 2008[b]
Fazon	0.1	2.4	5.2	3.0	9/5/2007	113.1	Furl and Meredith 2008[b]
Lower Goose	0.2	-	22.9	7.6	9/19/2007	21.1	Furl and Meredith 2008[b]
St. Clair	0.4	37.6	33.5	12.2	8/23/2007	141.3	Furl and Meredith 2008[b]
Samish	2.8	23.8	22.9	9.4	9/4/2007	105.7	Furl and Meredith 2008[b]
Moses	27.5	7,976.9	11.6	5.8	10/9/2006	24.6	Furl 2007a, b[b]
Newman	4.9	74.1	9.1	5.8	9/27/2006	47.2	Furl 2007a, b[b]
Offut	0.8	7.0	7.6	4.6	10/30/2006	138.5	Furl 2007a, b[b]
Sammamish	19.8	253.8	32.0	17.7	10/4/2006	110.0	Furl 2007a, b[b]
Meridian	0.6	3.0	27.4	12.5	10/5/2006	136.9	Furl 2007a, b[b]
Loon	4.6	36.5	30.5	14.0	10/26/2005	60.7	Furl 2007a, b[b]
Silver	9.3	101.8	3.0	1.8	9/22/2005	308.0	Furl 2007a, b[b]
Banks	1.1	-	25.9	14.3	11/7/2001	21.8	Fischnaller et al. 2003[c]
Terrell	1.8	7.4	3.0	2.1	9/26/2001	86.2	Fischnaller et al. 2003[c]
Long	-	-	54.9	14.6	6/18/2001	47.4	Fischnaller et al. 2003[c]
Vancouver	9.3	-	4.6	1.0	10/3/2002	105.6	Fischnaller et al. 2003[c]
Black	2.3	26.2	8.8	5.8	10/7/2002	116.0	Fischnaller et al. 2003[c]
Duck	1.1	3.7	9.1	3.4	10/10/2002	193.2	Fischnaller et al. 2003[c]
Loomis	0.7	3.7	2.7	1.5	10/11/2002	204.2	Fischnaller et al. 2003[c]
Palmer	8.5	766.6	24.1	15.5	10/15/2002	36.7	Fischnaller et al. 2003[c]
Kitsap	1.0	7.1	8.8	5.5	10/31/2002	99.2	Fischnaller et al. 2003[c]
Padden	0.6	6.8	18.0	8.2	9/27/2001	100.1	Seiders 2003[c]

[a] Analytical method EPA 7473

[b] Analytical method EPA 245.6

[c] Analytical method EPA 245.5

2.2 Fish Tissue Collection, Processing, and Analysis

Largemouth bass were collected by electroshocking and gillnetting from 2001 to 2007. Fish were measured, double-wrapped in aluminum foil, placed on ice in the field, and frozen (−20°C) within 72 h of collection until further processing. Fish from Lake Dickey were filleted skin-off in the field and shipped on ice overnight to the laboratory of William X. Wall Experiment Station, Massachusetts Department of Environmental Protection, in Lawrence, Massachusetts for analysis.

Fish were prepared for analysis from Lake Ozette and the statewide lakes by filleting with skin-on, passed three times through a Kitchen-Aid food grinder, and homogenized to a uniform color and texture. Utensils contacting the samples were cleaned using sequential rinses with tap water, Liquinox detergent, and hot tap water, 10% nitric acid, and deionized water. Tissues were analyzed using EPA Method 245.5, 245.6, or 7473 (Table 1).

Quality control for tissue analysis included analysis of laboratory control samples (80–120%), standard reference material (±15%), method blanks, matrix spike recoveries (75–125%), and matrix spike duplicates (<25% RPD). Data were generally good across all lakes with the exception of inadequate matrix spike duplicates (>25% RPD) at Duck Lake. Other standards run with samples from Duck Lake were within the typical range of agreement found in other lakes. Detailed methodology descriptions and results for tissue monitoring is included in Fischnaller et al. (2003), Furl et al. (2007), Furl (2007a), Furl and Meredith (2008), and Colman et al. (submitted for publication).

2.3 Wet Deposition Monitoring

Mercury wet deposition measurements were made at two Washington State MDN collection sites. Precipitation samples were collected and analyzed using methods specified by the network protocol (Welker and Vermette 1996). The Makah station is located 15 km from the north end of Lake Ozette and has been operating since March 2007. The second station is located in Seattle approximately 180 km from Lake Ozette and Lake Dickey. Wet deposition measurements have been conducted at the Seattle station since 1996.

2.4 Sediment Core Collection, Processing, and Analysis

Sediment cores were collected using a 13×13×50 cm Wildco box corer containing an acrylic liner. Cores were collected from deep areas of the lake with uniform bathymetry removed from significant surface water inputs. Cores reflecting the least disturbed sediments and a distinct sediment–water interface were immediately sectioned in the field. Subsamples were extruded in 1-cm intervals for the entire length of the core, stored in pre-cleaned 8 oz Nalgene bottles, and placed on ice in the field. One sediment core was collected at Lake Ozette, and two cores from approximately the same location were collected at Lake Dickey.

Sediment cores were analyzed for ^{210}Pb activity in order to assign dates and sedimentation rates over the past 100–150 years. For Lake Ozette, ^{210}Pb activity was determined in selected composites comprised of two to three 1-cm intervals using gamma spectroscopy for 1,000 min per sample. Samples were measured to a method detection limit of at least 0.45 pCi/g. Sample counts were done in one batch, and quality control measures consisted of one control sample, one method blank, and one duplicate. The control samples were recovered at an average of 104%, the method blanks were not detected above 0.300 pCi/g, and duplicates had a relative percent difference of 1.8%. For Lake Dickey, ^{210}Pb activity was determined for each 1-cm horizon at Lake Dickey using planar germanium detectors counting gamma ray emissions for 48–96 h, which provided an average ^{210}Pb counting error of less than 2.6%. A correction for self-absorption was made based on the geometry of the gamma-counted sample (Cutshall et al. 1983). Accuracy was confirmed by analyses of standard reference materials, which yielded agreement within 5% of certified values.

Mercury analyses for selected 1-cm intervals from the Lake Ozette core were conducted by the Washington State Department of Ecology's Manchester Environmental Laboratory using EPA method 245.5. Matrix spikes, blanks, and control samples were included for quality assurance. Two matrix spikes were recovered at 82% and 84%, respectively. A single blank was undetected at 0.0050 ng/g, and two control samples were recovered at 104% and 111%. The data were not adjusted for matrix spike recoveries. Sediment mercury analyses for Lake Dickey were conducted by the Wall Experiment Station where tissue analyses were performed. EPA Method 7473 was used for the determination of mercury concentrations on freeze-dried sediments. Average of 26 measurements of standard reference material was 101% of standard, range of 85 to 115%; spike recoveries ranged from 99% to 102%. Detailed methodology descriptions and results for the sediment cores are included in Furl (2007b, 2008),and Colman et al. (submitted for publication).

2.5 Age and Sedimentation Rate Calculations

The constant rate of supply model was used to estimate dates and varying sedimentation rates throughout the cores (Appleby and Oldfield 1978). For Lake Ozette, supported ^{210}Pb was estimated as the amount of ^{210}Pb present at deep intervals, beyond the zone of decline. Lake Dickey supported ^{210}Pb levels were determined from ^{214}Pb assays in each 1-cm horizon (Joshi 1987). An assumed sediment density of 2.5 g/cm^3 was used to compute dry mass for core dating.

Several horizons were analyzed for mercury without an accompanying ^{210}Pb measurement in the Lake Ozette core. Dates were assigned to these measurements by working back in time from the most recent ^{210}Pb derived date using an estimated interval mass accumulation rate (MAR) modeled from the ^{210}Pb sedimentation curve along with the mass of the interval:

$$\text{Date}_i = \text{Date}_{\text{Pb}} - (\text{cum}_i/\text{MAR}_i)$$

Where

- Date_i deposition date of sample without ^{210}Pb measurements
- Date_{Pb} date assigned to the bottom of the interval last measured for ^{210}Pb
- cum_i cumulative mass from Date_i to midpoint of sample i
- MAR_i interval MAR for sample i estimated from the ^{210}Pb derived MAR curve

Mercury flux rates (micrograms per square meter per year) were calculated as the product of the sedimentation rate and dry weight mercury concentration. The results estimate net deposition to the lake.

2.6 Statistical Calculations

Differences among Lake Ozette and Lake Dickey tissue concentrations were examined using an analysis of covariance (ANCOVA) to test for an effect of lake on fish tissue mercury concentrations. Length was selected as the covariate to account for the effect of fish size on mercury concentrations. Tissue concentrations from each of the 22 statewide lakes were then compared to the combined Ozette and Dickey data using an ANCOVA with a post hoc Dunnett's test. After the model accounted for variability associated with regression of mercury concentration on fish length, mean mercury concentrations for each lake were compared to Lake Ozette and Lake Dickey. Data for both tests were $\log_{10}$-transformed to improve the normality of the data. Statistical calculations were performed with Minitab.

3 Results

3.1 Fish Tissue

Mercury concentrations in fillets from Lake Ozette and Lake Dickey largemouth bass ranged from 190 to 2,500 ng/g ww (n=17). Tissue concentrations averaged 715 and 889 ng/g for Lake Ozette and Lake Dickey, respectively. Average fish length was slightly greater at Lake Dickey (358 mm) than Lake Ozette (342 mm). Length explained 49% and 83% of the variability in mercury concentrations for Ozette and Dickey, respectively (Fig. 2). The ANCOVA p value results indicated that mean mercury concentrations

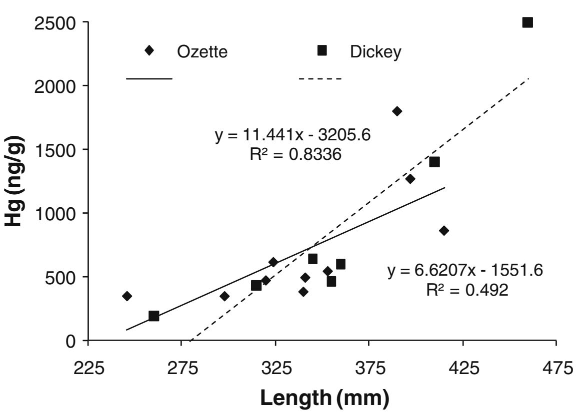

Fig. 2 Fish tissue concentrations vs. length for Lake Ozette and Lake Dickey

did not differ between the lakes after considering length (Table 2).

The ANCOVA and Dunnett's test performed on each of the 22 statewide lakes and the combined data of Lake Ozette and Lake Dickey found significantly lower mean mercury concentrations at each lake than the combined data from Lake Ozette and Lake Dickey (Table 2). Figure 3 displays regression of mercury concentrations against fish length for the combined Lake Ozette and Lake Dickey data along with other statewide lakes.

3.2 Mercury Wet Deposition

Table 3 presents mercury wet deposition and precipitation values for the two Washington State MDN stations. The Makah station measured slightly higher mercury deposition than the Seattle station over the first 12 months of operation. Over the same time period, precipitation at the Makah station was nearly three times greater than the Seattle station. Wet deposition values for the 10-month period starting March 2008 were similar at both stations. Precipitation ratios between sites were similar to the first year of monitoring although the sites received much less precipitation (≈60%). Yearly wet deposition at the Seattle station remained largely unchanged from 2001 to 2006 averaging 6.38 μg/m^2 (SD=0.80).

3.3 Sediment Cores

Sedimentation rates at the three coring locations ranged from 0.02 to 0.12 g/cm^2/year with the highest sedimentation rates occurring at or near the top or the core (Fig. 4). Unsupported atmospheric ^{210}Pb fluxes estimated from core inventories were 0.72, 0.77, and 0.93 pCi/cm^2/year at Dickey 1, Dickey 2, and Ozette, respectively. Typically, unsupported ^{210}Pb fluxes calculated from measured data fall within 0.2–1.0 pCi/cm^2/year (Appleby and Oldfield 1984). The greatest incremental increases in sedimentation rates occur during the last half of the twentieth century. Post-1950 dry weight concentrations varied little at the Dickey cores 200–230 ng/g (n=39) with the exception of a single anomalous value in the 2–3-cm horizon of Dickey 2 (140 ng/g). Dry weight concentrations in the Ozette core experienced a slightly more erratic pattern with a narrow range of concentrations during the same post-1950 time period (170–271 ng/g, n=9). Recent mercury flux rates estimated from the uppermost core horizon ranged from 196 μg/m^2/year at Ozette to 249 μg/m^2/year at Dickey 2.

Post-1950 mercury flux rates correlated strongly with sedimentation rates at Lake Ozette (r=0.925), and sedimentation rates explained 86% of the variance in flux rates (F=23.8, p<0.05; Fig. 5). Mercury concentrations at Lake Ozette had no correlation with flux rates (r=0.032) and explained 0% of the variance in mercury flux (F=0.004, p>0.05). A similar relationship was apparent in the Lake Dickey cores. Flux rates and sedimentation correlated strongly in both Lake Dickey core 1 and core 2 (r=0.995 and r=0.947, respectively), and sedimentation explained 99% and 90% of the variance in flux (F=1,774, p<0.05; F=157, p<0.05, respectively). Concentration had a weak correlation with flux rate at Dickey 1

Table 2 Statistical results

	Source	DF	Adjusted sum of squares	Adjusted mean squares	F	p
Ozette vs. Dickey ANCOVA	Length	1	1.01	1.01	45.93	0.000
	Lake	1	0.00	0.00	0.23	0.639
	Error	14	0.31	0.02		
	Total	16				
	Adjusted r square=0.738					
Statewide Lakes vs. Ozette and Dickey ANCOVA	Length	1	7.20	7.20	435.04	0.000
	Lake	22	18.95	0.86	52.01	0.000
	Error	211	3.49	0.02		
	Total	234				
	Adjusted r square=0.867					

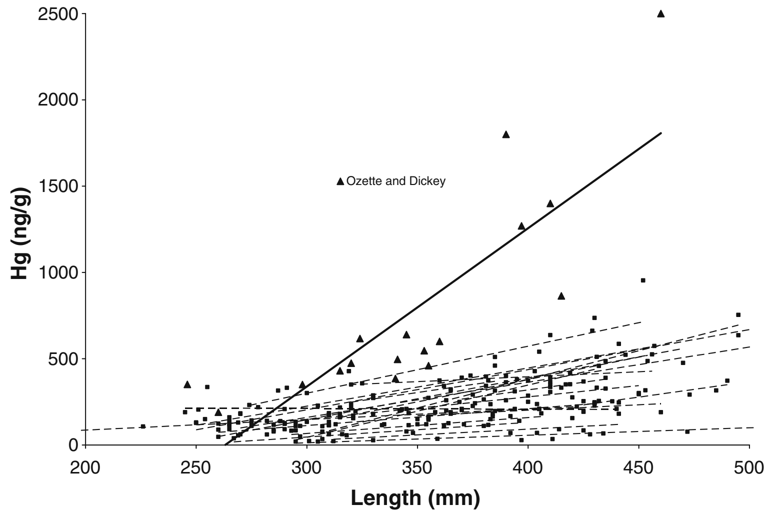

Fig. 3 Mercury regressions against fish length for combined Lake Ozette and Lake Dickey data and all Statewide Lakes

(r=0.412) and no correlation at Dickey 2 (r=0.040). Concentration explained 17% and 0% of the variance in flux rates at Dickey 1 and Dickey 2, respectively (F=3.485, p>0.05; F=0.029, p>0.05).

4 Discussion

4.1 Fish Tissue

High concentrations in fish tissue were not anticipated considering the remote location of Lake Ozette and Lake Dickey. While elevated mercury tissue concentrations in remote locations have been documented (Wiener et al. 2006), it was unexpected to find significant differences when comparing concentrations to lakes spread across the entire state. Average mercury concentrations at Duck Lake and Loomis Lake, included in the statewide dataset, were low (247 and 311 ng/g, respectively). Both of these lakes are located within 3 km of the Pacific Ocean south of Ozette and Dickey and do not have extensive timber harvest in their drainages.

Causes of the elevated concentrations in fish are currently unknown. Researchers have correlated several environmental factors, including pH, sulfate, chloride, and DOC, with elevated tissue concentrations in past investigations due to their effects on mercury methylation (Grieb et al. 1990; Hanten et al. 1998; Hrabik and Watras 2002). These constituents were measured in the water column from Lake Ozette along with 29 other randomly selected statewide lakes as part of the U.S. Environmental Protection Agency's 2007 National Lake Assessment. Lake Ozette concentrations for all four parameters fall within one standard deviation of the dataset mean (Maggie Bell-Mckinnon, personal communication). No data were obtained for Lake Dickey. Production of methylmercury also occurs in wetland areas, which exist within the lakes' drainages due to their low-relief geology. Specific factors mediating methylmercury production and biological uptake were beyond the scope of this study. However, sources and pathways of mercury to the lakes can be assessed through atmospheric monitoring and dated sediment cores.

Table 3 Wet deposition results

MDN station	March 2007–February 2008 (12 months)		March 2008–December 2008 (10 months)	
	Wet deposition (μg/m^2)	Precipitation (cm)	Wet deposition (μg/m^2)	Precipitation (cm)
WA03—Makah	7.59	223	4.31	101
WA13—Seattle	6.55	77	4.88	38

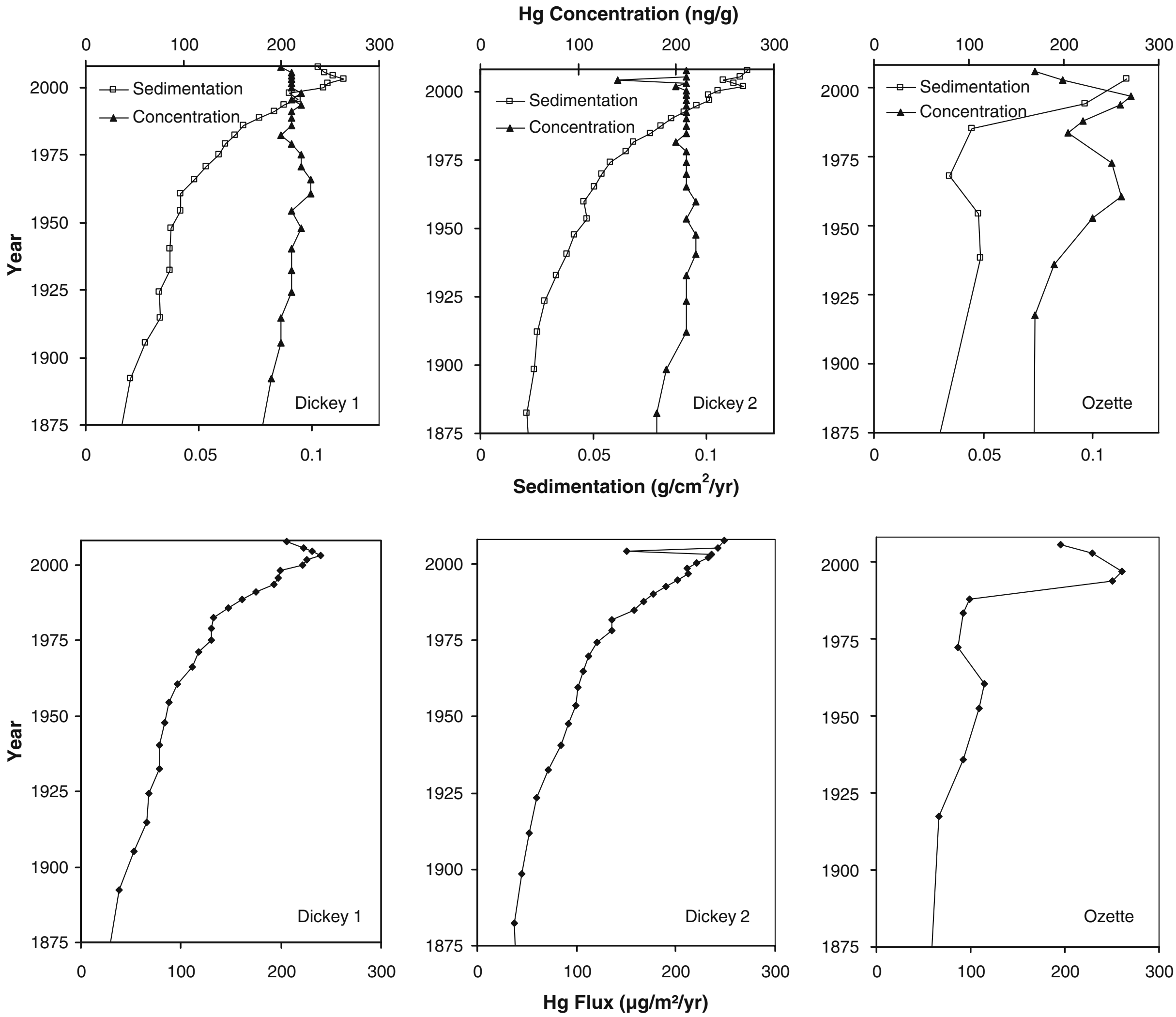

Fig. 4 Sediment core concentrations, sedimentation rates, and flux profiles

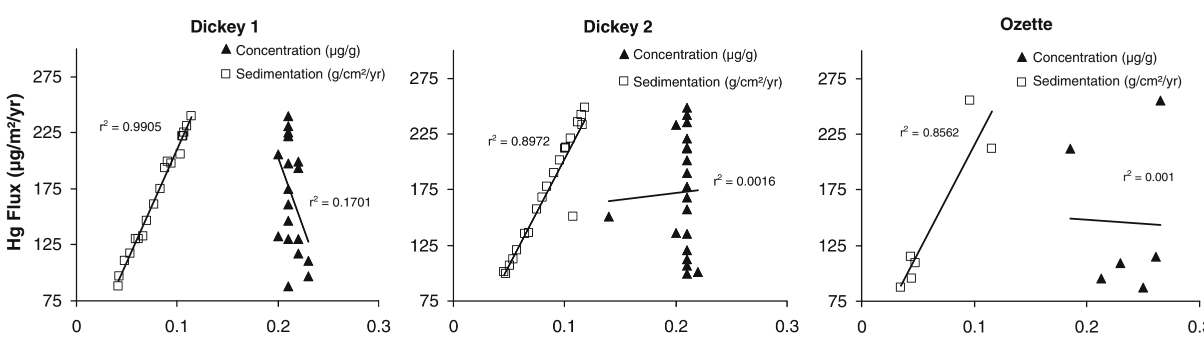

Fig. 5 Post-1950 mercury flux rates plotted with sedimentation rates and mercury concentrations

4.2 Atmospheric Mercury Loading

Atmospheric deposition of mercury is viewed as the dominant source of mercury to waterbodies and their drainages (Fitzgerald et al. 1998). Non-atmospheric point sources such as wastewater discharges and mining wastes can also serve as sources to a waterbody. Mercury mining, which has contaminated many drainages in the western USA, has been prevalent in the central part of Washington state but is absent on the Olympic Peninsula (USGS 2007). Point sources, both atmospheric and non-atmospheric, within the immediate vicinity of Lake Ozette and Lake Dickey are lacking.

Developing economies and increased energy needs resulted in a transfer of dominant global emissions of atmospheric mercury from North America/Europe to Asia (Pacyna et al. 2006; Seigneur et al. 2001). Led by China, Asian sources emitted approximately 54% of the global total in 2000 (Wu et al. 2006). Estimates for total Asian emissions, including contributions from terrestrial surfaces, range from 1,260 to 2,270 Mg/year (Jaffe et al. 2005; Pan et al. 2007; Strode et al. 2007). Increasing Asian mercury emissions have been of particular concern in the western USA due to its position downwind from Asia (e.g., Jaffe et al. 2005; Weiss-Penzias et al. 2003; Jaffe and Strode 2008). The unique far westerly location of Lake Ozette and Dickey places them as the closest lakes to Asia within the conterminous USA.

Asian air masses containing mercury have been documented at Cheeka Peak, 15 km north of Lake Ozette (Jaffe et al. 2005; Weiss-Penzias et al. 2006). The Asian air masses can reach the region in as little as 5 days and bear a similar signal to the global reservoir dominated by elemental mercury with a small percentage of RGM (Jaffe et al. 2005; Weiss-Penzias et al. 2003). Without the benefit of a baseline period and multiple years of recent wet deposition measurements from the Makah station, quantifying changes in wet deposition coinciding with increased Asian emissions cannot be completed. Recent modeling studies examining the Asian contribution to deposition within the USA found Asian deposition to be broadly dispersed due to elemental mercury being the dominant species. Location or orientation to Asia appears to be a less important factor controlling deposition than rates of in situ production of RGM from the Asian elemental mercury pool (Strode et al. 2007; Jaffe and Strode 2008).

Results from wet deposition monitoring found similar yearly deposition rates at the Makah and Seattle sites with disparate precipitation concentrations. Atmospheric concentrations in the Seattle metro area averaged 2.5 ng/m^3 over a 2-year period from 1994 to 1995 (Bloom et al. 1995). Atmospheric monitoring conducted over a 13-month period at Cheeka Peak found concentrations consistent with the northern hemisphere background (1.7 ng/m^3; Weiss-Penzias et al. 2006; Ebinghaus et al. 2002). The background concentrations recorded at Cheeka Peak atmospheric monitoring suggests that deposition in the area is likely dominated by the global mercury reservoir and not influenced by local or regional atmospheric sources.

4.3 Sediment Core Record of Drainage Disturbance

Mercury flux rates measured in the uppermost intervals of sediment cores from Lake Ozette and Dickey Lake (≈200 μg/m^2/year) were greater than fluxes measured in other coring studies in regional remote locations. Landers et al. (2008) found recent mercury fluxes generally less than 50 μg/m^2/year at two lakes located within the interior of the Olympic National Park and two lakes within Mount Rainier National Park, WA, USA. All four lakes are located in undisturbed drainages, and mercury concentrations for largemouth bass are not available for the national park lakes. Colman et al. (submitted for publication) estimated recent (1990–2007) mercury fluxes from 20 to 65 μg/m^2/year in four coastal lakes in Oregon. Average mercury concentrations in largemouth bass across all four lakes were approximately half (443 ng/g) the levels recorded at Ozette and Dickey (Colman et al., submitted for publication).

Sedimentation rates at Ozette and Dickey coring locations largely control mercury flux rates after 1950, explaining greater than 85% of the variance in flux rates for all three cores (Fig. 5). Mercury concentrations during this same time period were contained within a narrow range varying little with sedimentation. These findings are contrary to those of Engstrom et al. (2007) who found a negative correlation between sediment accumulation and dry weight concentrations over a large dataset of 55 Minnesota cores. The authors explained this relationship as increased sedimentation having a diluting effect on atmospheric inputs.

Numerous researchers determined logging in the Lake Ozette catchment to be the source of increased sedimentation to the lake (Haggerty et al. 2007; Ritchie 2009; Herrera 2006). Currently, only 20% of the Lake Ozette catchment remains as primary forest (Ritchie 2009). Herrera (2006) estimated current sedimentation rates at least three times greater than pre-logging levels. Haggerty et al. (2007) attributed elevated sedimentation rates at Ozette to high road density and a large percentage of hydrologically immature forest due to logging. The sediment cores from the present study indicate current sedimentation rates four times higher than average baseline values at both lakes. Figure 6 displays sedimentation rates estimated from the cores plotted with remaining primary forest as a percent of watershed in the Lake Ozette catchment reconstructed from aerial photography (Ritchie 2009).

Increased terrestrial output of mercury to lake ecosystems resulting from clear-cut logging practices have been recorded elsewhere. In a 7-year study, Porvari et al. (2003) found significant increases in the total mercury and methylmercury load in runoff water after clear-cutting in a small forested catchment in Norway. Additionally, similar to the present study, the authors found no concentration increases in runoff samples, but rather an increase in total flux due to elevated flow. The effects of logging practices on mercury concentrations in lake biota were studied in Quebec. Garcia and Carignan (1999) observed significantly higher concentrations of mercury in zooplankton in lakes with recently logged watersheds compared to lakes with undisturbed or recently burned watersheds. The same authors found significantly higher mercury concentrations in northern pike in logged watersheds compared with undisturbed catchments (Garcia and Carignan 2000). No other lake catchments included in the statewide tissue statistical analysis have been as extensively logged in recent times as Lake Ozette and Lake Dickey.

5 Summary and Conclusion

The data presented in this paper document unprecedented concentrations of mercury in Washington State largemouth bass. Atmospheric concentrations and depositional data indicate that the two lakes are relatively unaffected by local and regional point source polluters. The high levels of contamination found in fish tissue at the lakes have occurred under atmospheric conditions near the global background. Sediment core data from both lakes indicate increased sedimentation due to logging and greatly increased net flux of mercury to the waterbodies. Clear-cut logging has been correlated with increased mercury and methylmerucry flux in other investigations. Specific factors influencing methylmercury production and biological uptake were beyond the scope of this study. Additional work is necessary to determine the relative contributions of catchment and in-lake processes resulting in the elevated tissue levels.

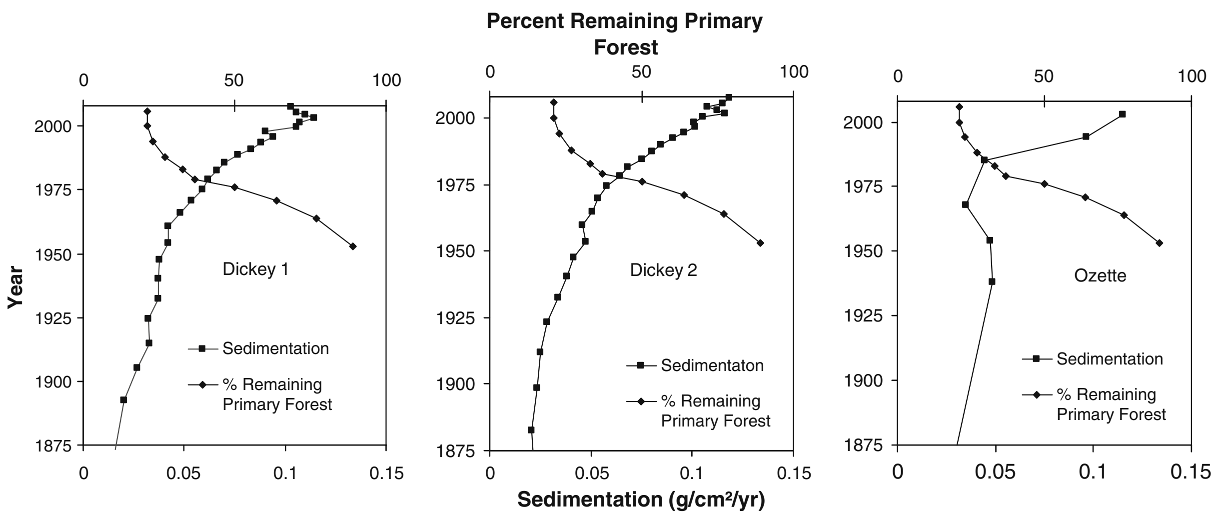

Fig. 6 Estimated sedimentation rates plotted with percent remaining primary forest (Ritchie 2009) in the Lake Ozette drainage basin

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