PBT Chemical Trends Determined from Age-Dated Lake Sediment Cores, 2015 Results

DEPARTMENT OF ECOLOGY State of Washington

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Key Findings

- Recent lead concentrations declined in the Meridian and Williams Lake cores and stabilized in the Lake Whatcom core.
- Mercury concentrations peaked in 1956, 1970, and 1990 in the Meridian, Whatcom, and Williams cores. Recent concentrations declined in Lake Meridian and leveled off in the Whatcom and Williams cores.
- PBDEs started increasing between 1970 and 2000 in the cores and continued to rise through recent sediments. Meridian was the only core to show a leveling off of concentrations.
- BTBPE was detected inconsistently, with maximum concentrations appearing in 2009 (Meridian) and 2013 (Whatcom). DBDPE was not detected in any of the three lakes, while HBBz and PBEB were present at low levels.

Overview

The Washington State Department of Ecology's (Ecology's) Persistent, Bioaccumulative, and Toxic (PBT) Monitoring Program conducts long-term monitoring of freshwater sediment cores to help characterize the occurrence and temporal trends of PBTs in Washington State. A single sediment core is collected each year from three waterbodies and age-dated in order to reconstruct contaminant deposition profiles.

In 2015, sediment cores collected from Meridian, Whatcom, and Williams Lakes (Figure 1) were analyzed for lead, mercury, and brominated flame retardants (BFRs). BFRs are a broad class of chemicals used in consumer products, such as furniture and electronics, to prevent or slow the spread of fire. BFRs analyzed in the sediment cores included polybrominated diphenyl ethers (PBDEs), 1,2-Bis(2,4,6-tribromophenoxy) ethane (BTBPE), decabromodiphenylethane (DBDPE), hexabromobenzene (HBBz), and pentabromoethylbenzene (PBEB).

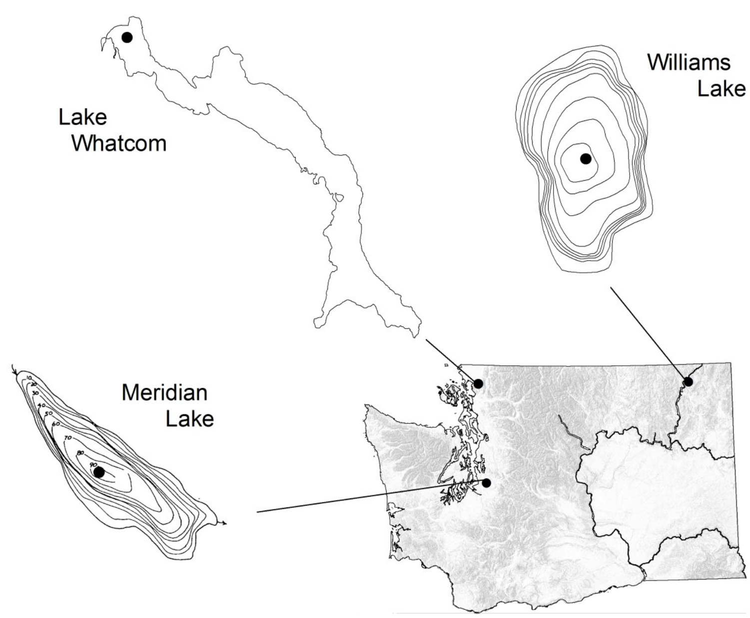

Figure 1. Lake Sediment Core Locations, 2015.

Information provided by this sediment coring program supports the development of agency Chemical Action Plans (CAPs) and is used to track progress on existing CAP recommendations and actions to reduce PBT levels in the environment. A CAP for PBDEs was developed in 2006 (Ecology, 2006), but there are currently no CAPs for other brominated flame retardants.

For More Information

PBT Monitoring Program website: http://www.ecy.wa.gov/programs/eap/toxics/pbt.html

Chemical Action Plan website: http://www.ecy.wa.gov/programs/hwtr/RTT/pbt/caps.html

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Methods and Data Quality

This study followed a Quality Assurance Project Plan (QAPP) and QAPP addendum (Coots, 2006; Mathieu, 2015). Ecology collected three sediment cores 35-45 cm in depth with a Wildco© box corer, following Ecology standard operating procedures (Furl and Meredith, 2008). Surface sediments were collected with a standard ponar for grain size analysis. Sediment core collection efforts were unsuccessful at Pierre Lake due to high water content of the sediments. An alternative nearby lake - Williams Lake - replaced Pierre Lake for this study.

Manchester Environmental Laboratory (MEL) and contract laboratories analyzed samples for total lead, mercury, ^{210}Pb, total organic carbon (TOC), grain size, and BFRs using methods described in the QAPP and QAPP addendum. AXYS Analytical Services Ltd. analyzed BFRs by HR GC/MS, following AXYS method MLA-033, which is in general accordance with EPA Method 1614A. TOC and grain size data were used to help interpret contaminant concentrations and are not presented in this report. All data, including TOC and grain size, are available for download in Ecology's EIM database (http://www.ecy.wa.gov/eim/) by searching Study ID: SEDCORE15.

Measurement quality objectives (MQOs) were met for analyses of metals, ^{210}Pb, and TOC. For grain size analysis, the relative standard deviation of the gravel fraction analysis exceeded 25%. All other size fractions were within MQOs.

MEL's QA coordinator reviewed and verified that contract laboratory data were generated following the analytical method with no omissions or errors. All data were reviewed by the project manager and deemed usable as qualified.

BFR results were qualified for the following reasons. Several BFRs were detected in the method blanks. In samples where the detected concentration of the affected sample results was less than ten times the blank contamination, the result was qualified as a non-detect at the estimated quantitation limit (EQL) or at the level of detection if greater than the EQL. Three surrogate recoveries were outside of method acceptance limits, and affected results were qualified as estimates. Lab duplicate results exceeded MQOs for relative percent difference for several compounds. Because all other sample and batch QC indicators appeared acceptable, the differences were attributed to sample inhomogeneity and results were used as qualified. Laboratory control sample recoveries for PBEB analyses were low. Affected samples were either rejected and reanalyzed or qualified as estimates. Samples that met all identification criteria except the isotope abundance ratio were qualified "NJ" to indicate that the compound was tentatively identified and the value is an estimate. All samples were reported down to the instrument's estimated detection limit (EDL); results reported below the EQL were qualified as estimates.

Study Locations

Table 1. Physical Descriptions of 2015 Study Lakes.

Waterbody	Elevation (ft)	Max Depth (ft)	Lake Area (ac)	Watershed Area (ac)	WA:LA
Meridian	370	90	150	742	4.9
Whatcom	315	330	5,000	35,776	7.2
Williams	1,980	47	38	3,878	102

WA:LA = watershed area to lake area ratio.

Lake Meridian lies within the city limits of Kent. The small watershed is highly developed with primarily residential properties, and the lake shoreline is densely populated. Basin geology consists of predominantly fine-grained Vashon till deposits. Meridian has an average annual rainfall of 45". Intermittent surface-water inflow is at the northwest end and flows out to the east toward Soos Creek.

Lake Whatcom is a large lake made up of three distinct basins. Basin 1, where the sediment core was collected, lies within the city of Bellingham. The entire watershed consists of primarily forestland, with smaller portions of residential and developed areas concentrated in the region surrounding Basin 1. Vashon recessional outwash and bedrock deposits make up the basin geology. With an annual precipitation of 44", the lake is fed through many perennial streams and drains to Whatcom Creek at the western shore of Basin 1.

Williams Lake is located in Stevens County in northeastern Washington. The lake's watershed consists of undeveloped forestland and agriculture, underlaid by continental glacial outwash and marine metasedimentary rocks. There is no development along the shoreline, aside from a road close to the eastern shore. The lake has an average annual precipitation of 22". There are no surface inlets and only an intermittent outlet drains to the south toward Colville River.

Core Dating

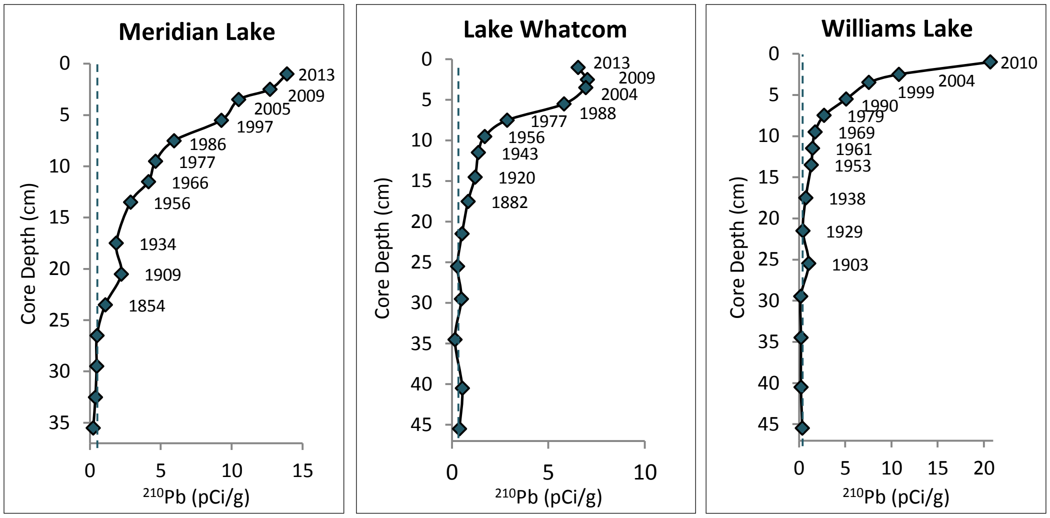

Figure 2. ^{210}Pb Activity Plotted Against Sediment Core Depth.
Dates assigned to sediment layer midpoint using the CRS model are included in graph.

Dates were calculated for the three cores using ^{210}Pb activities and the constant rate of supply (CRS) model (Appleby and Oldfield, 1978) (Figure 2). Percent solids were used to calculate dry mass. Supported ^{210}Pb was estimated as the average activity present at deep intervals where there was no further decline. Average supported ^{210}Pb activities in Meridian, Whatcom, and Williams Lakes were estimated as 0.40, 0.39, and 0.23 pCi/g (picocurie per gram), respectively.

Yearly unsupported ^{210}Pb fluxes in pCi/cm^2/yr were 0.29, 0.30, and 0.18 for Meridian, Whatcom, and Williams, respectively. This report uses focus factors to correct for the focusing of fine-grained material to coring locations or the transport of sediments away from coring sites. The unsupported ^{210}Pb flux values were divided by estimated total unsupported fluxes calculated using lake-specific precipitation values and atmospheric ^{210}Pb deposition measured in Washington State (Lamborg et al., 2013). Focus factors for Meridian, Whatcom, and Williams Lakes were 1.26, 1.07, and 1.58, and they are applied to contaminant fluxes throughout this report to normalize the data.

Sediment Accumulation Rates

Sediment accumulation rates in Lake Meridian were low and steady throughout most of the 1900s and early 2000s, ranging from 0.003 - 0.021 g/cm^2/yr. This is consistent with other lakes in the lower Puget Sound area with similar watershed area to lake area ratios.

Lake Whatcom sediment accumulation rates ranged from 0.011 - 0.045 g/cm^2/yr. Previous sediment core work on Lake Whatcom estimated an average rate of 0.045 g/cm^2/yr for Basin 1 (Norton et al., 2004). Increases began over the first half of the 20th century and rates appeared to be increasing again in the most recent decade at Lake Whatcom.

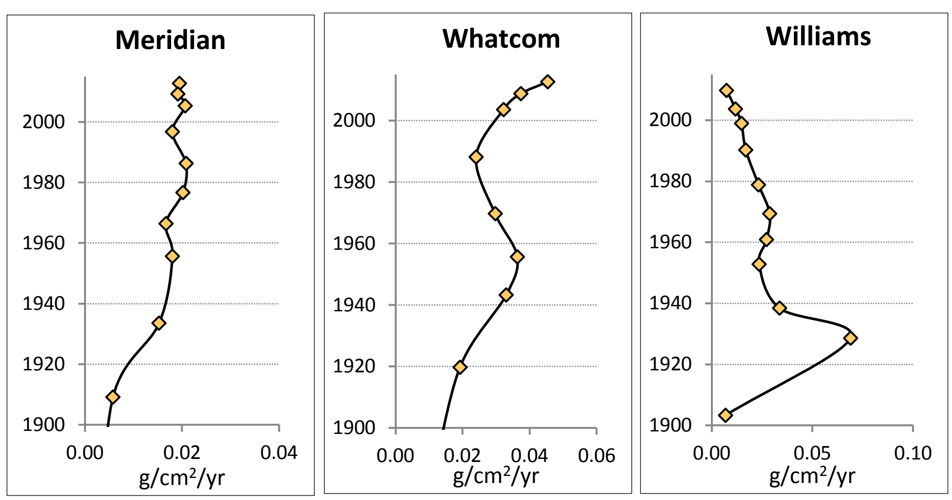

Figure 3. Estimated Sediment Accumulation Rates (g/cm^2/yr).

Williams Lake sediment accumulation rates ranged from 0.007 - 0.069 g/cm^2/yr. A spike in sedimentation occurred around 1930 and has steadily declined since then. TOC also peaked to 27% around the same time.

Lead

Profiles

Lead concentrations in Lake Meridian sediments increased throughout the 1900s until a peak in 1977 of 384 ug/g, consistent with peak leaded gasoline use. Lead concentrations have steadily decreased since 1977 to 94 ug/g in 2013. Lead fluxes (an estimate of the rate of net deposition to the coring site) in Lake Meridian exhibits the same trend with peak flux in the same year (6.13 ug/cm^2/year).

Lake Whatcom lead concentrations began to increase above baseline in the early 1900s and continued to increase consistently until 1977 with a peak lead concentration of 111 ug/g. Since then lead concentrations declined to 75 ug/g in 2013. Lead flux in Lake Whatcom in 2013 was 3.17 ug/cm^2/year, the highest rate since 1977.

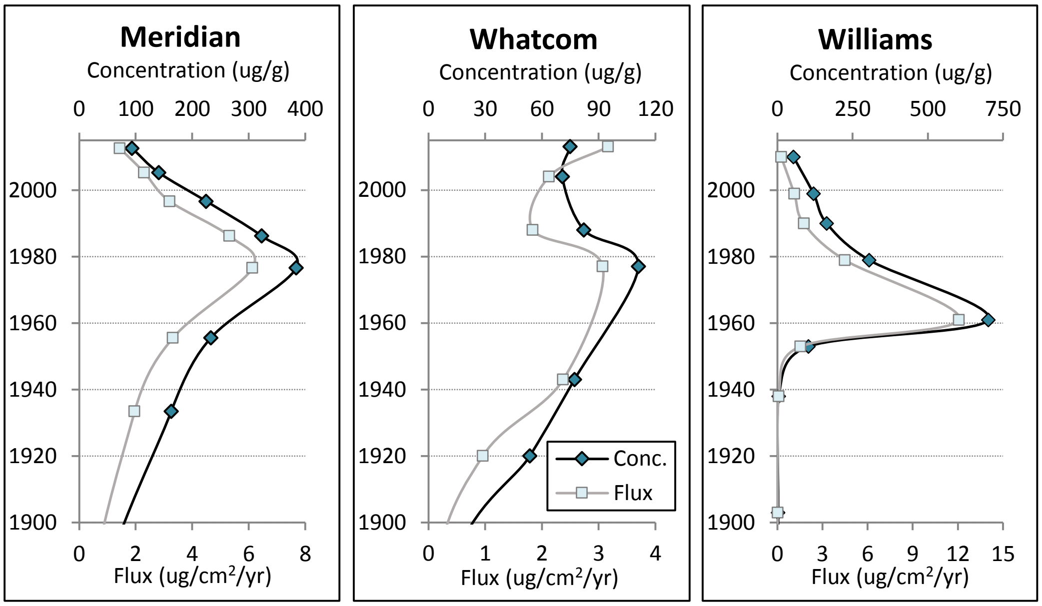

Figure 4. Lead Concentration (ug/g) and Focus Factor-corrected Flux (ug/cm^2/yr) Profiles in Sediment Cores.

Williams Lake lead concentrations and fluxes remained low until a large spike in the mid-1900s. Lead concentrations increased from 103 ug/g in 1953 to 701 ug/g in 1961. Concentrations have steadily declined in the subsequent years and were 54 ug/g in 2010. Lead fluxes showed a similar pattern, with a peak of 12.07 ug/cm^2/yr in 1961 and steady declines to 0.25 ug/cm^2/yr in 2010.

Enrichment

Enrichment factors (EFs) compare the relative contaminant concentration of a sediment core interval to a baseline level. We calculated EFs as individual lead concentrations divided by the average stable pre-industrial lead concentrations at the bottom of the core (referred to as "baseline" in this report). "Modern" sediment refers to the top-most horizon of the sediment core (0 - 2 cm). Table 3 displays lead enrichment factors for the three lakes.

Williams Lake contained the highest lead concentrations and peak enrichment of all three lakes (Table 3) with maximum lead concentration more than 400 times above baseline conditions. Modern sediments have an EF of 32.

Lake Whatcom had the lowest lead concentrations and EFs. Enrichment factors peaked at 20 in 1977, declined to 12.7 in 2004 and had a slight uptick in modern sediments at 13.5.

Modern sediment concentrations from Lake Meridian were 12.8 times greater than baseline and 52 times greater at peak in 1977.

Table 2. Lead Concentrations and Enrichment Results.

Lake	Range (ug/g)	Baseline (ug/g)	Peak Enrichment	Modern Enrichment
Meridian	5.85 - 384	7.3	52 (1977)	13 (2013)
Whatcom	5.23 - 111	5.6	20 (1977)	14 (2013)
Williams	1.59 - 701	1.7	419 (1961)	32 (2010)

Mercury

Profiles

Mercury concentrations in Lake Meridian increased over the first half of the 1900s until peak concentration in 1956 (663 ng/g) and then steadily declined to 118 ng/g in 2013. Mercury fluxes followed this trend with a modern flux of 18.15 ug/m^2/year.

Lake Whatcom sediments reached a peak mercury concentration in 1970 (240 ng/g), then declined through 2004 to 140 ng/g. The most recent sediment layer (147 ng/g) indicated that modern concentrations have remained stable. However, mercury fluxes showed an increase from 42.0 ug/m^2/year in 2004 to 62.1 ug/m^2/year in 2013.

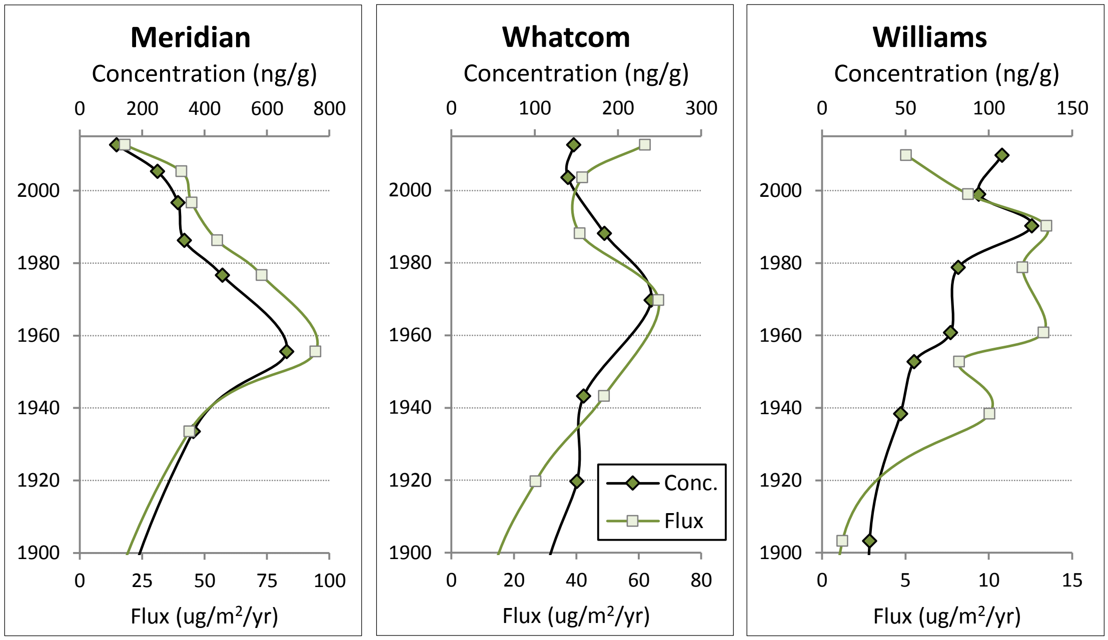

Figure 5. Mercury Concentration (ng/g) and Focus Factor-corrected Flux (ug/m^2/yr) Profiles in Sediment Cores.

Mercury sediment concentrations in Williams Lake have gradually increased from baseline concentrations throughout the core. Williams had a peak mercury concentration of 126 ng/g in 1990 with a modern sediment concentration of 108 ng/g. Mercury fluxes increased over the first half of the 20th century, remained steady between 1960 and 1990, and then decreased steadily to a flux rate of 5.04 ug/m^2/year in the top-most sediment.

Enrichment

Meridian and Whatcom Lakes contained relatively high baseline mercury concentrations (62.0 and 77.1, respectively). The Meridian core had the highest peak concentration (663 ng/g) and EF (11) of the three lakes.

Mercury enrichment throughout the Lake Whatcom core was fairly low, with the peak EF of 3.1 occurring in 1970 and a modern EF of 1.9 measured in 2013.

Table 3. Mercury Concentration and Enrichment Results.

Lake	Range (ng/g)	Baseline (ng/g)	Peak Enrichment	Modern Enrichment
Meridian	51.2 - 663	62.0	11 (1956)	1.9 (2013)
Whatcom	71.0 - 240	77.1	3.1 (1970)	1.9 (2013)
Williams	14.1 - 126	18.9	6.7 (1990)	5.7 (2010)

Williams Lake sediment mercury concentrations were fairly low, but EFs were comparable to the other lakes, due to the low baseline concentration of 18.9 ng/g. Williams Lake sediments had the highest modern EF value of the three lakes (5.7) in 2010, and a peak EF in 1990 of 6.7.

Brominated Flame Retardants

T-PBDEs

Forty PBDE congeners were analyzed in the Meridian, Whatcom, and Williams Lakes sediment cores. PBDEs were detected in 93% of samples, with the number of detected congeners ranging from 2 to 31. Concentrations of total PBDEs (T-PBDEs, sum of detected congeners) in detected samples ranged from 0.0009 - 32.0 ng/g. Focus-corrected fluxes ranged from 0.003 - 487 pg/cm^2/yr.

T-PBDE concentrations and fluxes began increasing gradually in the late 1960s in the Meridian core and then increased sharply from the 1990s through 2009. T-PBDEs appear to have leveled off in the most recent sediments, with concentrations of 32.0 ng/g and 31.7 ng/g in 2009 and 2013, respectively. In Lake Whatcom, T-PBDE concentrations began increasing after 1970. By the late 1980s, the concentration was 4.71 ng/g, and then doubled to 8.23 ng/g in 2013. T-PBDEs in Williams Lake did not start rising until the late 1990s. A sharp increase in concentrations and fluxes occurred in the 2000s, as concentrations rose from 0.63 ng/g in 2004 to 2.98 ng/g in 2010, though levels remained lower than those of the more developed lakes.

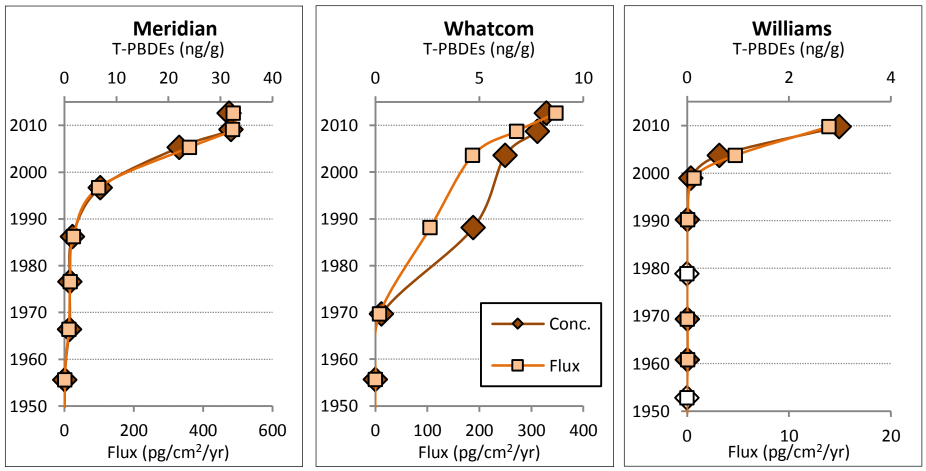

Figure 6. T-PBDE Concentration (ng/g) and Focus Factor-corrected Flux (pg/cm^2/yr) Profiles in Sediment Cores.
White squares and diamonds indicate that no PBDEs were detected at or above the

PBDE Congeners

To evaluate temporal trends in the deposition of commercial flame retardant mixtures, PBDE congeners are presented in Figure 7 as indicators of the Penta-BDE (sum of -47, -85, -99, 100, -153, and -154), Octa-BDE (-183 only), and Deca-BDE (-209 only) commercial flame retardants. Summed nona-BDE congeners (-206, -207, -208) are also shown in Figure 7. Nona-BDEs are breakdown products of Deca-BDE, but they can also be present in Octa-BDE (La Guardia et al., 2006).

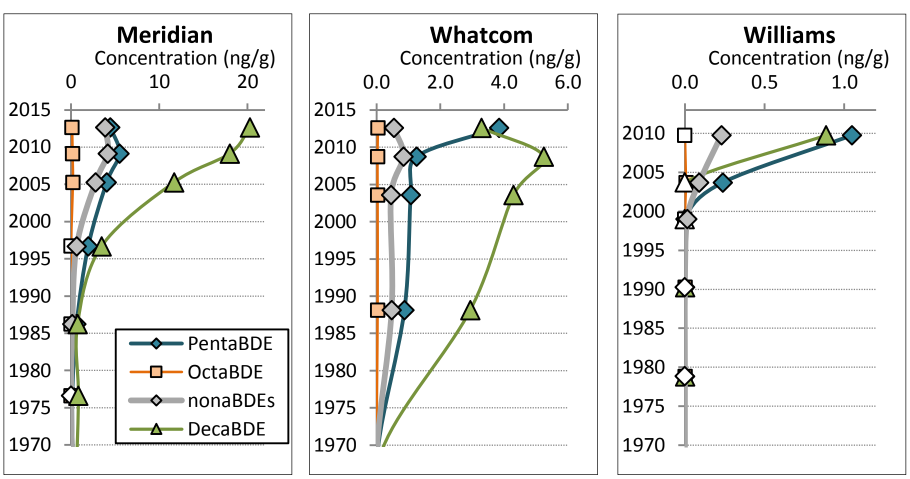

Figure 7. Concentrations of PBDE Congeners Summed as Commercial Formulation Indicators.
PentaBDE = sum of BDE-47, -85, -99, -100, -153, and -154; OctaBDE = BDE-183; nonaBDEs = sum of BDE-206, -207, and -208. DecaBDE = BDE209.

Indicators of the Penta- and Octa-BDE mixtures rose gradually in the Lake Meridian core from the mid-1990s until peaking in the mid-2000s, whereas Deca-BDE increased sharply and consistently from the 1990s through the most recent sediments. Surface concentrations of Deca-BDE (20.3 ng/g) were 1-2 orders of magnitude higher than those of Penta- and Octa-BDE (4.46 and 0.112 ng/g, respectively).

Trends in recent sediments of the Lake Whatcom core were dissimilar. Deca-

Brominated Flame Retardants

BDE peaked at 5.25 ng/g in 2009 and then declined, while Penta-BDE spiked in the late 2000s to a maximum concentration (3.85 ng/g) in the top layer, 2013. Concentrations in the Williams Lake core were low, with primarily non-detects until 1999. Deca- and Penta-BDE both peaked in the most recent Williams Lake sediments (2010) at 0.885 and 1.05 ng/g, respectively. Octa-BDE concentrations in all three cores were low, with detections below 0.2 ng/g and no apparent temporal patterns.

U.S. manufacturers phased out Penta- and Octa-BDE flame retardants in the mid-2000s and stopped most uses of Deca-BDE by the end of 2012. The Lake Meridian core reflects this usage pattern, with Deca-BDE continuing to increase after Penta-BDE peaked in the late 2000s. Lake Meridian has a relatively small watershed area, and a quicker response to contaminant deposition in the watershed could be expected. Both Whatcom and Williams Lakes show Penta-BDE continuing to increase through modern sediments. This could be a function of the lag time between contaminant deposition in a watershed and the accumulation of those contaminated sediments to the lake bottom. It could also reflect the continued use of products containing these flame retardants, which may continue to be a source to the environment.

Table 4. BTBPE, DBDPE, HBB, and PBEB Concentrations (pg/g) in 2015 Sediment Cores.

Waterbody	Year	BTBPE (pg/g)	DBDPE (pg/g)	HBBz (pg/g)	PBEB (pg/g)
Meridian Lake	2013	201 UJ	475 UJ	6.25 NJ	2.67 NJ
	2009	**1,110**	342 UJ	**7.26**	**1.73**
	2005	**149**	280 UJ	**3.58**	2.9 NJ
	1997	273 UJ	345 UJ	**9.10**	4.34 NJ
	1986	**45.3**	308 UJ	9.52 UJ	0.797 UJ
	1977	255 UJ	221 UJ	**2.16**	**1.43**
	1966	296 UJ	218 UJ	**1.25**	0.573 UJ
	1956	747 UJ	251 UJ	0.752 NJ	0.831 UJ
	1909	9.86 UJ	238 UJ	**0.414**	0.221 UJ
Lake Whatcom	2013	**320**	156 UJ	1.32 NJ	0.518 NJ
	2009	153 UJ	206 UJ	1.49 NJ	REJ
	2004	110 UJ	158 UJ	**3.15**	**0.491**
	1988	**135**	247 UJ	1.56 NJ	**1.02**
	1970	82.1 UJ	154 UJ	**30.3**	1.22 NJ
	1956	10.9 UJ	94 UJ	0.402 NJ	0.101 UJ
	1943	123 UJ	91.6 UJ	0.182 UJ	0.0991 UJ
	1920	286 UJ	492 UJ	0.634 NJ	0.0991 UJ
	1882	193 UJ	278 UJ	0.561 NJ	0.1 UJ
Williams Lake	2010	340 UJ	1780 UJ	**5.07**	0.207 UJ
	2004	17.7 UJ	526 UJ	**0.807**	**0.192 J**
	1999	39.6 UJ	349 UJ	**1.00**	0.196 UJ
	1990	202 UJ	899 UJ	0.998 UJ	0.174 UJ
	1979	87 UJ	728 UJ	0.914 NJ	0.134 UJ
	1969	58.7 UJ	631 UJ	0.679 NJ	0.106 UJ
	1961	53.9 UJ	339 UJ	**0.99**	0.141 UJ
	1953	62 UJ	689 UJ	0.697 UJ	0.172 UJ
	< 1850	30.6 UJ	658 UJ	1.26 NJ	0.13 UJ

UJ: Not detected at or above reported estimate. NJ: Analyte tentatively identified and the reported result is an estimate. J: Result is an estimate. ***Bold*** *indicates analyte was detected.*

Non-BDE Flame Retardants

As commercial uses of PBDEs were phased out, manufacturers used alternative flame retardants as replacements to meet flammability standards. BTBPE and DBDPE are brominated flame retardants currently used as replacements for PBDEs (Octa- and Deca-BDE formulations, respectively) (Vorkamp and Riget, 2014). HBBz and PBEB are no longer produced in the U.S., but minor production or usage may occur in other countries (Covaci et al., 2011; Vorkamp and Riget, 2014). Table 4 presents concentrations of these four alternative flame retardants in the 2015 sediment cores.

BTBPE was detected in the Meridian and Whatcom cores at concentrations between 45.3 - 1,110 pg/g. BTBPE first appeared after 1980 in both cores, but detections were not consistent, nor was there an apparent temporal pattern. The peak concentration occurred in 2009 for Lake Meridian (1,110 pg/g) and 2013 in Lake Whatcom (320 pg/g). DBDPE was not detected in any of the cores at quantitation limits ranging from 91.6 - 1,780 pg/g.

HBBz and PBEB were detected more frequently (44% and 15% of samples, respectively) than BTBPE but at much lower concentrations and with lower quantitation limits. HBBz detections ranged from 0.414 to 30.3 pg/g, with a median of 2.66 pg/g. Detected concentrations of PBEB ranged from 0.491 to 1.73 pg/g (median = 1.23 pg/g).

Our results are somewhat lower in detection frequency and concentrations than sediment cores collected from the Great Lakes (Yang et al., 2012). BTBPE and DBDPE ranged from 130 - 8,300 pg/g and 110-2,800 pg/g, respectively in the upper sediments of Great Lakes cores (Yang, 2012). However, concentrations and detection frequencies were similar to surficial sediments collected from the San Francisco Bay (Klosterhaus et al., 2012) and Swedish Lakes (Ricklund et al., 2012).

Conclusions

Ecology collected sediment cores from Meridian, Whatcom, and Williams Lakes in 2015 for analysis of lead, mercury, and brominated flame retardants. Contaminant deposition profiles showed the following trends:

- Lead concentrations increased in Meridian and Whatcom sediments until a peak in 1977 for both lakes. Concentrations and fluxes declined consistently since then in the Meridian core to a modern concentration of 94 ug/g. Lake Whatcom concentrations decreased initially from the peak and then stabilized to a concentration of 75 ug/g in 2013, while lead fluxes increased in the top layer. In the Williams Lake core, lead concentrations and fluxes showed a large spike in 1961 (701 ug/g) and then steadily declined to a concentration of 54 ug/g in 2010.

- Mercury concentrations peaked in 1956, 1970, and 1990 in the Meridian, Whatcom, and Williams cores. Meridian and Whatcom profiles were similar to patterns in lead, where Lake Meridian concentrations and fluxes declined steadily after maximum and Lake Whatcom concentrations appeared to plateau in recent sediments, while fluxes increased. Mercury concentrations were 118, 147, and 108 ng/g in the top sediment layers of Meridian, Whatcom, and Williams, respectively.

- PBDEs were detected in all three cores, with T-PBDE surface sediment concentrations of 2.98 ng/g (Williams Lake), 8.23 ng/g (Lake Whatcom), and 31.7 ng/g (Lake Meridian). Surficial fluxes of T-PBDEs ranged from 13.9 pg/cm^2/yr in Williams Lake to 487 pg/cm^2/yr in Lake Meridian. T-PBDEs rose sharply in Lake Meridian beginning in the 1990s through the late 2000s, and then leveled off between 2009 and 2013. T-PBDEs in Lake Whatcom began increasing after 1970, doubling in concentration between the 1980s and 2013. In Williams Lake, T-PBDEs were lower and did not start rising until the late 1990s and then increased sharply through the top of the core.

- PBDE congeners summed as indicators of the commercial flame retardant mixtures Penta-, Octa-, and Deca-BDE indicated a leveling off of Penta-BDE in recent sediments of Lake Meridian, whereas Deca-BDE continued to rise through the top of the core. In Lake Whatcom, Deca-BDE peaked in 2009 and then declined, while Penta-BDE spiked between 2009 and 2013. Deca- and Penta-BDE both increased through the top of the core in Williams Lake. Octa-BDE concentrations were low in all three cores (< 0.2 ng/g) and showed no apparent temporal patterns.

- Alternative brominated flame retardants were detected infrequently and inconsistently in the three cores. BTBPE was measured in the Meridian and Whatcom cores at concentrations ranging from 45.3 to 1,110 pg/g. BTBPE first appeared after 1980 in both cores, with maximum concentrations in 2009 for Lake Meridian and 2013 in Lake Whatcom. BTBPE was not detected in the Williams Lake core, and DBDPE was not detected in any of the cores at quantitation limits ranging from 91.6 to 1,780 pg/g. HBBz and PBEB were detected more frequently than BTBPE and in all three cores, but at much lower concentrations and quantitation limits than BTBPE.

Recommendations

- The rising trend of PBDEs in lake sediments warrants continued monitoring of these contaminants in the environment. Ecology currently monitors PBDEs in fish tissue; additional media should be considered.
- While detections were not consistent, the BTBPE results suggest that alternative brominated flame retardants are persisting in the environment and could be increasing. Further investigation is recommended to help characterize the occurrence of additional alternative flame retardants in Washington's environment.

References

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

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. https://fortress.wa.gov/ecy/publications/summarypages/0603113.html

Covaci, A., S. Harrad, M.A.-E. Abdallah, N. Ali, R.J. Law, D. Herzke, and C.A. de Wit, 2011. Novel brominated flame retardants: A review of their analysis, environmental fate and behaviour. Environment International, Vol. 37: 532-556.

Ecology, 2006. Washington State Polybrominated Diphenyl Ether (PBDE) Chemical Action Plan: Final Plan. Washington State Department of Ecology, Olympia, WA. Publication No. 05-07-048. https://fortress.wa.gov/ecy/publications/summarypages/0507048.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. Publication No. EAP038. http://www.ecy.wa.gov/programs/eap/quality.html

Klosterhaus, S.L., H.M. Stapleton, M.J. La Guardia, and D.J. Greig, 2012. Brominated and chlorinated flame retardants in San Francisco Bay sediments. Environment International, 47: 56-65.

La Guardia, M.J., R.C. Hale, and E. Harvey, 2006. Detailed Polybrominated Diphenyl Ether (PBDE) Congener Composition of the Widely Used Penta-, Octa-, and Deca-PBDE Technical Flame-retardant Mixtures. Environmental Science and Technology, Vol. 40: 6247-6254.

Lamborg, C.H., D.R. Engstrom, W.F. Fitzgerald, and P.H. Balcom, 2013. Apportioning global and non-global components of mercury deposition through ^{210}Pb indexing. Science of the Total Environment, Vol. 448: 132-140.

Mathieu, C. and M. Friese, 2012. PBT Chemical Trends in Washington State Determined from Age-Dated Lake Sediment Cores, 2011 Sampling Results. Washington State Department of Ecology, Olympia, WA. Publication Number 12-03-045. https://fortress.wa.gov/ecy/publications/summarypages/1203045.html

Mathieu, C., 2013. PBT Chemical Trends Determined from Age-Dated Lake Sediment Cores, 2012 Results. Washington State Department of Ecology, Olympia, WA. Publication Number 13-03-036. https://fortress.wa.gov/ecy/publications/SummaryPages/1303036.html

Mathieu, C., 2015. Addendum 5 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. 15-03-113. https://fortress.wa.gov/ecy/publications/SummaryPages/1503113.html

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

Ricklund, N., Am. Kierkegaard, and M.S. McLachlan. Levels and Potential Sources of Decabromodiphenyl Ethan (DBDPE) and Decabromodiphenyl Ether (DecaBDE) in Lake and Marine Sediments in Sweden. Environmental Science and Technology, Vol. 44: 1987-1991.

Schroeder, M.C., 1952. Geology of the Bead Lake District Pend Oreille County, Washington. Division of Mines and Geology. State of Washington Department of Conservation and Development Bulletin No. 40.

Vorkamp, K. and F.F. Riget, 2014. A review of new and current-use contaminants in the Arctic environment: Evidence of long-range transport and indications of bioaccumulation. Chemosphere, Vol. 111: 379-3.

Yang, R., H. Wei, J. Guo, and A. Li, 2012. Emerging Brominated Flame Retardants in the Sediment of the Great Lakes. Environmental Science and Technology, Vol. 46: 3119-3126.

Glossary of Terms

CRS Model: A model developed by Appleby and Oldfield (1978) applied to ^{210}Pb measurements in a sediment core to estimate dates and varying sedimentation rates. The model works by measuring the difference in supported and unsupported ^{210}Pb in sediment horizons, and the relation of that difference to the inventory of unsupported ^{210}Pb of the whole core. 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.

Enrichment Factor: The enrichment factor is a unitless value describing the amount concentrations have increased/decreased from the baseline concentration. Enrichment factors were calculated for the contaminants by dividing concentrations in the interval of interest by the baseline value. Baseline estimates were calculated by taking the average of the lowest contaminant values at the bottom of the core where concentrations were stable.

Flux: An estimated rate of net deposition to the lake. Flux rates normalize the variance involved with interpreting dry weight concentrations under varying sedimentation rates. Contaminant flux rates were calculated as the product of the sediment mass accumulation rate and dry weight contaminant concentration.

Focus Factor: A focus factor corrects for the focusing of fine-grained sediments to the coring location or the transport of sediments away from coring sites. Sediment cores for this study are often collected in the deepest part of the lake, and fine-grained sediments preferentially deposit in these areas.

Modern: Refers to the top-most sediment horizon of the sediment core (0 - 2cm).

Supported ^{210}Pb: Supported ^{210}Pb is represented by the small amount of the precursor gas ^{222}Rn (radon) that is captured in soils. Supported ^{210}Pb in this study was estimated as the average ^{210}Pb value at deep intervals where it appeared to no longer decline.

Unsupported ^{210}Pb: Unsupported ^{210}Pb represents the atmospherically deposited ^{210}Pb resulting from the decay of ^{222}Rn that escapes into the atmosphere. Unsupported ^{210}Pb in this study was estimated by subtracting supported ^{210}Pb from total ^{210}Pb at a given depth.

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