# PBT Chemical Trends Determined from Age-Dated Lake Sediment Cores, 2016 Results



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#### **Key Findings**

- Polychlorinated biphenyls (PCBs) were analyzed in sediment cores collected from Deep Lake, Spanaway Lake, and Lake Spokane in 2016.
- Total (T-) PCB concentrations in Deep and Spanaway Lakes sediments have declined since peak levels and recent concentrations appear to have stabilized.
- T-PCBs were fairly stable in Lake Spokane sediments between the early 2000s and early 2016, but increased in the most recently deposited sediment layer. Current T-PCB levels were much less than historical peaks observed in the 1960s.
- PCB congener profiles revealed no consistent temporal patterns in individual congeners across sites. Shifts in congener patterns varied among cores, but PCBs associated with Aroclor sources and resistant to degradation were dominant in all three cores.

**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. A new organic PBT is rotated into the target analyte list each year, which over time provides data on a range of PBTs.

In 2016, the target analyte for 2016 was polychlorinated biphenyls (PCBs). PCBs are highly persistent in the environment, bioaccumulative, and have toxicity concerns including cancer and harm to the immune, nervous, and reproductive systems, as well as thyroid disruption. Ecology and the Department of Health (DOH) developed a chemical action plan (CAP) for PCBs in 2015 and recommended actions to protect human health and the environment from PCBs (Davies, 2015).

Ecology collected sediment cores from Deep Lake, Spanaway Lake, and Lake Spokane



Figure 1. 2016 Sediment Core Locations.

(Figure 1). Eight samples near the top of each core were analyzed for PCB congeners. Samples selected for analysis were weighted toward the top of the core to evaluate patterns in the most recent decades of deposition. Information provided by this program supports the development of CAPs and is used to track progress on existing actions to reduce PBT levels in the environment.

#### For More Information

#### **PBT** Monitoring Program website:

https://ecology.wa.gov/Research-Data/Monitoring-assessment/toxics-monitoring/Contaminants-in-sediment-cores

**Chemical Action Plan website:** 

https://ecology.wa.gov/Waste-Toxics/Reducing-toxic-chemicals/Addressing-priority-toxic-chemicals

### **Methods and Data Quality**

Ecology collected three sediment cores 35-45 cm in length using a Wildco<sup>©</sup> box corer following Ecology standard operating procedures (Furl and Meredith, 2008) and the Quality Assurance Project Plan (QAPP) (Mathieu, 2016). Surface sediments were collected using a standard ponar for grain size analysis.

Manchester Environmental Laboratory (MEL) and contract laboratories analyzed sediment samples for total lead, <sup>210</sup>Pb, total organic carbon (TOC), grain size, and PCB congeners using methods described in the QAPP. Pacific Rim Laboratory analyzed PCB congeners by HR GC/MS, following EPA Method 1668C. Total lead and grain size data were used to help interpret contaminant concentrations but are not summarized in this report. All data, including total lead and grain size, are available for download in Ecology's EIM database (<u>http://www.ecy.wa.gov/eim/</u>) by searching Study ID: SEDCORE16.

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. Measurement quality objectives (MQOs) were generally met for analyses of total lead, <sup>210</sup>Pb, grain size and TOC. Results for one matrix spike and matrix spike duplicate analyzed for total lead were outside of MQOs. The standard spiking level was insufficient for the elevated concentration in the source sample and therefore recoveries were not evaluated. For <sup>210</sup>Pb analyses, one of the three lab duplicates exceeded MQOs. The associated field sample was qualified "J" as an estimate. The average of the field value and lab duplicate value was used in the age dating model. Results are unlikely to be affected by this MQO exceedance.

PCB congener results below the EQL (estimated quantitation limit) but above the instrument detection limit (IDL) were qualified "J" as estimates. Laboratory control samples (LCS) were generally within method acceptance limits, with the exception of one batch for PCB-018 and PCB-093/098/095 (which had low recoveries) and PCB-040, PCB-044, and PCB-139/149 (which had high recoveries). Associated samples were qualified "J" as estimates. Two lab duplicate results (#1608047-03 for PCB-209 and #1608047-37 for PCB-027) had a relative percent difference outside of MQOs; sample results for those analytes were qualified "J" as estimates. Surrogate results showing poor recovery (0%) were rejected and re-analyzed by the laboratory. One sample surrogate recovery (#1608047-33 for PCB-126) was low - 15% - and the result changed from "U" to "UJ" to indicate that the non-detect was an estimate.

PCB congener results less than 10 times method blank contamination were censored as non-detects for this report and are reported as such in EIM. Total PCB (T-PCB) concentrations on pages 6-7 of this report include only detected congener results that were unqualified or were qualified "J" (indicating that the analyte was positively identified and the associated value is an estimate). Data qualified as "NJ" (indicating that the analyte has been "tentatively identified" and the associated value represents its approximate concentration) were not included in T-PCB sums. In accordance with the QAPP, data was also re-censored at 3 times the method blank for PCB congener profile analysis, reported on pages 8-9 of this report. Data qualified as "NJ" were included in the PCB congener profile analysis.

## **Study Locations**

The 2016 study locations were selected based on criteria outlined in the QAPP. Deep Lake was selected to represent PCB concentrations in a primarily undeveloped, forested waterbody with no known PCB contamination issues. Spanaway Lake is an urban waterbody selected to reflect urban inputs of PCBs, such as through stormwater. Lake Spokane is a waterbody with well-documented PCB contamination issues (Serdar et al., 2011; Limnotech, 2016).

Deep Lake is located in the northeastern part of the state, surrounded by a primarily forested watershed. There are residences along the shoreline and livestock grazing in the watershed for part of the year. Water flows into the lake from the north through Deep Creek and out through the south end of the lake. Previous work by Ecology has shown that total suspended sediment (TSS) levels entering Deep Lake from Deep Creek in the spring can be elevated (Stuart, 2015). Annual precipitation levels at Deep Lake average 28.3". Spanaway Lake is located in Pierce County within a watershed dominated by residential. commercial, and military base land types. Major hydrological inputs to the lake include groundwater discharge and inflow from Coffee Creek on the southern shoreline (Pierce Coun- WA:LA = watershed area to lake area ratio ty, 2017). Surface water drains from the lake to the north

Waterbody	County	Elevation (ft)	Max Depth (ft)	Mean Depth (ft)	Lake Area (ac)	Watershed Area (ac)	WA:LA
Deep Lake	Stevens	2,025'	49'	34'	210	30,784	147
Lower Lake Spokane	Spokane	1,536'	180'	50'	45,227	4,249,600	94
Spanaway Lake	Pierce	320'	28'	16'	280	10,880	39

 Table 1. Physical Descriptions of 2016 Study Locations.

through Spanaway Creek. Average annual precipitation to the lake is 41". Spanaway Lake's watershed is composed of highly permeable glacial till and outwash soils overlying bedrock deposits, resulting in very little surface water runoff (Pierce County, 2017; Johnson et al., 2011).

Lake Spokane is a 24-mile long reservoir in Spokane County formed by Long Lake Dam in 1915. Upstream of the reservoir, the Spokane River runs through the city of Spokane draining many different land types throughout the large watershed. The Spokane River has been identified as a hot spot of PCB contamination since the 1990s (Serdar et al., 2011; Limnotech, 2016). Hydrological inflow and outflow to Lake Spokane are dominated by surface water and water residence time in the reservoir is relatively short - around 16 days (Cusimano, 2004). Lower Lake Spokane receives an average annual precipitation of 16".

## **Core Dating**

#### **Deep Lake**

Dates were calculated for the Deep Lake core using <sup>210</sup>Pb activities and the constant rate of supply (CRS) model (Appleby and Oldfield, 1978) (Figure 2). Percent solids were used to calculate dry mass for all cores. The supported <sup>210</sup>Pb value for the Deep Lake core (0.23 pCi/g) was estimated from a nearby sediment core in the region (Williams Lake; Mathieu and McCall, 2016), as the core did not penetrate deep enough to reach supported <sup>210</sup>Pb. Alternative dating models were considered, but a clear increase in dry bulk density and decrease in the <sup>210</sup>Pb activity profile at the 20-25cm depth indicated a large spike in sedimentation during that time. The CRS model allows for varying sedimentation rates and was determined to be the most appropriate model.

Because the core did not capture the entire inventory of unsupported <sup>210</sup>Pb, there is a higher degree of uncertainty around the assigned dates, particularly in the lower layers of the core. Figure 2b presents the estimated error, or uncertainty, associated with each dated sediment layer.



Figure 2. Deep Lake Dating Results: a) <sup>210</sup>Pb Activity Plotted Against Sediment Core Depth and b) Modeled Age of Sediment Core Depth with Associated Uncertainty/Error. Dates assigned to sediment layer midpoint are included in graph a.

## **Core Dating**

#### Spanaway Lake

Spanaway Lake sediment core dates were calculated using <sup>210</sup>Pb activities and the CRS model (Appleby and Oldfield, 1978). Supported <sup>210</sup>Pb was estimated as the activity present at the deepest interval where there was no further decline (0.17 pCi/g). The mean yearly unsupported <sup>210</sup>Pb flux estimated by the CRS model was 0.15 pCi/cm<sup>2</sup>/yr.

A focus factor correction was applied to contaminant fluxes for Spanaway Lake in this report. This is to correct for the focusing of fine-grained material to coring locations or the transport of sediments away from coring sites. Yearly unsupported <sup>210</sup>Pb flux values were divided by estimated total unsupported fluxes calculated using a lake-specific precipitation value and atmospheric <sup>210</sup>Pb deposition measured in Washington State (Lamborg et al., 2013). The focus factor for Lake Spanaway was 0.69. No focus factors were applied to the other two lake sediment cores because neither captured the entire inventory of excess <sup>210</sup>Pb necessary for the calculation.



Figure 3. Spanaway Lake Dating Results: a) <sup>210</sup>Pb Activity Plotted Against Sediment Core Depth and b) Modeled Age of Sediment Core Depth with Associated Uncertainty/Error. Dates assigned to sediment layer midpoint are included in graph

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#### Lake Spokane

The Lake Spokane sediment core did not reach supported <sup>210</sup>Pb activities necessary for the CRS model. The CF:CS (constant flux: constant sedimentation) model was used to assign sediment dates instead. The CF:CS model can be used when unsupported <sup>210</sup>Pb concentrations plotted on a logarithmic scale against cumulative dry mass appears linear; the slope of this least squares fit relationship is used to calculate the sediment mass accumulation rate (MAR) for the core (Appleby and Old-field, 1992). The supported <sup>210</sup>Pb value used in the model was taken from core data collected by Serdar et al. (2011) at the same location in 2004. Serdar et al. (2011) analyzed three core samples (top, middle, and bottom) for Radium 226 (<sup>226</sup>Ra), which is assumed to be in equilibrium with supported <sup>210</sup>Pb.

Errors were not calculated for the CF:CS modeled dates. However, given that the core did not capture the entire inventory of <sup>210</sup>Pb, uncertainty is likely to be high for the given dates. Uncertainties for the <sup>210</sup>Pb activities were similar to uncertainties for the Deep Lake <sup>210</sup>Pb activities.



Figure 4. Lake Spokane Dating Results: <sup>210</sup>Pb Activity Plotted Against Sediment Core Depth.

Dates assigned to sediment layer midpoint are included in graph.

## **Sediment Accumulation Rates**

### **Profiles**

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Sediment MARs for Deep and Spanaway Lakes were calculated using the CRS model. MAR profiles and associated uncertainties are presented in Figure 5.

Deep Lake MARs were fairly low and steady between the 1950s and 1970, started rising in the late 1970s, and peaked at 0.36 g/cm<sup>2</sup>/yr around 1987. After a consistent decline from the peak through the 1990s, sediment accumulation rates have been relatively constant, ranging between 0.16 - 0.21 g/ cm<sup>2</sup>/yr from 2001 to the present. Sediment MARs from the 1970s to the top of the core were quite high compared to other lakes cored in the region. Peak MARs in the Deep Lake core were 5 and 10 times higher than max MAR values from nearby Williams and Cedar Lakes (Mathieu and McCall, 2016; Johnson et al., 2013). Monitoring of TSS in the surface water inlet to the lake has shown elevated suspended sediment loadings during certain times of the year (Stuart, 2015). Loss on ignition analyses of the sediment core samples confirmed the MAR spike in the late 20<sup>th</sup> century to be a result of increased mineral sediment transported to



the coring site, and not an increase in organic mat- Figure 5. Sediment Mass Accumulation Rate (MAR) Profiles.

The Spanaway Lake sediment core showed low

MARs from the early 1900s through 1960. Three periods of large increases in sediment accumulation were evident: in the late 1960s, early 1990s, and around 2011. These three periods of high sediment accumulation rates were all in the range of 0.05 - 0.07 g/cm<sup>2</sup>/yr. Sediment MARs had returned to a lower rate of 0.02 g/cm<sup>2</sup>/yr in the most recent sediment layer (2014).

The CF:CS model for Lake Spokane resulted in one sediment MAR for the entire core: 0.35 g/cm<sup>2</sup>/yr. The CRS model fit to the Lake Spokane core data showed a fairly consistent sediment accumulation rate, ranging between 0.14 - 0.37 g/ cm<sup>2</sup>/yr. The similarity of MARs between the two dating models, along with a fairly consistent range, supported the use of the CF:CS model, which uses one MAR for the core.

Table 2.	Sediment Mass Acc	umulation Rates	(MAR	) Results.
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Range in MAR (g/cm²/yr)	Peak year	Modern MAR (g/cm²/yr)	Modern year
0.104 - 0.356	1987	0.199	2016
0.007 - 0.067	1968	0.022	2014
0.348		0.348	2016
	Range in MAR (g/cm²/yr) 0.104 - 0.356 0.007 - 0.067 0.348	Range in MAR (g/cm²/yr)Peak year0.104 - 0.35619870.007 - 0.06719680.348	Range in MAR (g/cm²/yr)Peak yearModern MAR (g/cm²/yr)0.104 - 0.35619870.1990.007 - 0.06719680.0220.3480.348

\*Lake Spokane dating model used one average mass accumulation rate for the entire core.

*Modern* = *upper-most sediment interval measured for the core.* 

## **Total Polychlorinated Biphenyls (T-PCBs)**

Polychlorinated biphenyls (PCBs) are a group of man-made chemicals that were manufactured and used in the U.S. from the 1930s through the 1970s. Production stopped after regulations in the 1970s prohibited the manufacture, processing, and distribution of PCBs, but some legacy uses were allowed to continue (Davies, 2015). PCBs are highly persistent in the environment, bioaccumulative, and have toxicity concerns including cancer and harm to the immune, nervous, and reproductive systems. Washington's PCB CAP contains detailed information on major sources and pathways, exposure routes, toxic effects, and recommendations for actions to protect human health and the environment (Davies, 2015).

Eight sections from the Deep Lake, Spanaway Lake, and Lake Spokane sediment cores were analyzed for up to 209 different PCB congeners. Total PCB concentrations (T-PCBs) were calculated according to the procedure outlined in the QAPP, using a 10x blank censoring rule and including only detected values. All PCB concentrations are reported on a dry weight basis unless otherwise noted.

#### Table 3. Deep Lake Sediment Core PCB Data.

#### **Deep Lake**

Sample ID	Core section (cm)	TOC (%)	T-PCBs (ug/kg dw)	T-PCB flux (ng/cm <sup>2</sup> /yr)	Year
	0-2	5.64	0.796	0.159	2016
	2-3	5.61	0.934	0.175	2014
	3-4		1.03	0.184	2013
Deep Lake	5-6	4.91	0.921	0.154	2011
Deep Lake	7-8		0.985	0.166	2007
	9-10	5.13	1.23	0.249	2004
	11-12		2.17	0.460	2001
	21-22	4.07	0.157	0.056	1987

T-PCB concentrations were low in the Deep Lake sediment core, ranging from 0.16 - 2.17 ug/kg with a median of 0.96 ug/kg (Figure 6 and Table 3). T-PCB fluxes were also low and displayed a very similar pattern to concentrations, ranging from 0.056 - 0.460 ng/cm<sup>2</sup>/yr. The peak T-PCB concentration (2.17 ug/kg) and flux (0.460 ng/cm<sup>2</sup>/yr) occurred in 2001, then concentrations and fluxes decreased through the early 2000s and maintained a steady concentration and flux of around 1.0 ug/kg and 0.17 ng/cm<sup>2</sup>/yr, respectively, through the most recent sediment layer. The correlated concentration and flux profile indicate

that PCBs predominantly enter Deep Lake through fluvial transport of watershed sediments, instead of direct atmospheric fallout onto the lake surface.

#### Spanaway Lake

T-PCB concentrations in the Spanaway Lake core ranged from 16.8 - 69.3 ug/kg (median = 42.1 ug/kg)(Figure 3 and Table 4). The earliest sampled layer (1968) contained the lowest concentration, then levels increased through the early 1990s to a peak in 1992. After the peak, T-PCB concentrations declined consistently through the early 2000s and displayed a leveling out in the most recent sediments (between 2008 and 2014). Fluxes followed a similar pattern, with rising fluxes until the 1990s followed by declines.



Figure 6. Profiles of T-PCB Concentrations (ug/kg) and Fluxes (ng/cm<sup>2</sup>/yr) in 2016 Sediment Cores.

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## **Total Polychlorinated Biphenyls (T-PCBs)**

However, in the flux profile a spike is evident in the early 2010s, suggesting that transport of PCB-containing sediments increased during that time. T-PCB fluxes ranged from 0.711 - 3.17 ng/cm<sup>2</sup>/yr.

### Lake Spokane

The Lake Spokane sediment core captured a relatively short time frame due to high sedimentation rates, with measured samples spanning 2001 to 2016. In this time frame, T-PCB concentrations in the core sediments ranged from 18.0 - 144 ug/kg (median = 32.5 ug/kg). For most of the core, T-PCB concentrations were within a fairly narrow range of 18.0 - 32.6

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Sample ID	Core section (cm)	TOC (%)	T-PCBs (ug/kg dw)	T-PCB flux (ng/cm²/yr)	Year	
Spanaway Lake	0-2	13.2	35.4	1.10	2014	
	2-3	12.1	38.3	3.17	2011	
	3-4		32.4	1.02	2008	
	5-6	12.5	45.9	1.28	2001	
	7-8	13.2	59.3	4.45	1995	
	9-10		69.3	4.57	1992	
	12-13		47.0	3.55	1987	
	18-19	16.1	16.8	1.62	1968	

#### Table 4. Spanaway Lake Sediment Core PCB Data.

ug/kg. Two spikes in concentrations and fluxes occurred: in 2007 (81.7 ug/kg) and 2016 (144 ug/kg). The surface layer (2016) in the Spokane core had the highest-measured T-PCB concentration and was the only sample above Washington's Freshwater Sediment Cleanup Objective (*based on total PCB Aroclors*) of 110 ug/kg (WAC 173-204).

Because PCBs were only measured in the Lake Spokane sediment core as far back as 1997, we compared the recent profile to data measured in an earlier core collected by Serdar et al. (2011) in 2004. The Serdar et al. (2011) study analyzed sections of the core that dated back to 1950, thus reaching peak PCB concentrations. Figure 7 shows T-PCB concentrations in the Serdar et al. core that have been normalized to an organic carbon basis and corrected from a total Aroclor sum to a T-PCB congener sum based on a formula presented by Coots (2014). T-PCB concentrations on an organic car-

bon basis measured in the current core collected for this 2016 study are included alongside the historical data in Figure 7 to show how the current sediment core PCB profile fits into a historical perspective.

T-PCB concentrations in lower Lake Spokane appear to have peaked in the early 1960s at 1,950 ug/kg (72,300 ug/kg OC), and declined until the late 1980s. Between the late 1980s and modern sediments (2016), T-PCB concentrations have remained within the range of 18.0 - 144 ug/kg (564 - 4,900 ug/kg OC). Other surface sediments collected from Lake Spokane (upper, mid, and lower) in 2000 and 2003/2004 showed a similar range and variability in T-PCB concentrations (2.0 - 99 ug/kg; 400 - 3808 ug/kg OC) (Serdar et al., 2011; Johnson and Norton, 2001).

	Sample ID	Core section (cm)	TOC (%)	T-PCBs (ug/kg dw)	T-PCB flux (ng/cm <sup>2</sup> /yr)	Year
Ī	Lake Spokane	0-2	3.69	144	50.2	2016
		2-3	3.74	21.6	7.51	2015
		3-4	3.49*	32.7	11.4	2014
		5-6	3.23	18.2	6.34	2013
		7-8	3.19	18.0	6.28	2011
		9-10	3.12	32.4	11.3	2009
		11-12	2.89*	81.7	28.4	2007
		18-19	2.54	32.5	11.3	2001

 $^{*}\mbox{TOC}$  not analyzed for this interval. Value is an average of the TOC measurements in the above and below interval.



Figure 7. Profile of T-PCB Concentrations (ug/kg OC) Measured in Lake Spokane Sediment Cores Collected in 2016 (red) and 2004 (grey).

### **PCB Congener Profiles**

The sediment cores were also analyzed for patterns in PCB congener make up over the years. For this congener analysis, data were censored for blank contamination using a 3 times rule and tentatively qualified data ("NJ") were included. The following sections describe PCB profiles as relative abundance of summed homolog groups (Figure 8) and individual congeners (Figure 9).

#### **Deep Lake**

Pentachlorobiphenyls were the most abundant homolog group in the Deep Lake core from 1985 through the late 2000s, contributing greater than 20% of the total PCB concentration. The percent composition of hexachlorobiphenyls increased over the length of the core, with a relative abundance similar to penta-PCBs in the most recent sediments (2012 through 2016). The decline of the pentachlorobiphenyl homolog group in the upper sediments was driven by decreased relative contributions of PCB-093/098/095 and PCB-110. Whereas, in the hexachlorobiphenyl homolog group the contribution of congeners PCB-138 and PCB-153 increased between the early 2000s and current sediments. However, PCB concentrations were very low in Deep Lake and differences in relative abundance reflect small changes in concentrations.

#### Spanaway Lake

In the Spanaway Lake core, hexachlorobiphenyls were the most dominant homolog group for all sediment layers analyzed. Hexachlorobiphenyls started out at 25% of the PCB total in the mid-1960s, then increased in the early 1990s to around 40% of the total. An increase in PCB-153 over the length of the core is primarily responsible for the rise in the hexachlorobiphenyl group. The heptachlorobiphenyl profile mirrored the hexachlorobiphenyl profile, with a slight increase in 2010 where the hexa group declined briefly. The mono through penta homolog profiles remained very consistent throughout the core.

No monotonic patterns in the core were observed for individual PCB congeners, with the exception of the increase in percent contribution of PCB-153 through the most recent sediments. The generally consistent congener profile throughout the core suggests there has been little change in sources or pathways of PCBs to the lake over the time period studied.



Figure 8. Relative Abundance of PCB Homologue Totals in 2016 Sediment Cores.

## **PCB Congener Profiles**

### Lake Spokane

Hexachlorobiphenyls were the dominant homolog group found in the Lake Spokane core through most of the measured sediment layers. In the most recent sediments (2015-2016), the congener make-up shifted from higher chlorinated groups to lighter homologs, with a decrease in hexa and hepta homologs and an increase in the tetra and penta homolog groups. The dominant homolog group in the most-recent sediments was the pentachlorobiphenyl group (33%). This rise in the penta homolog group was driven by an increase in PCB-110 in the most recent sediments; whereas the decline in the hexachlorobiphenyl group reflects decreasing percent contributions of PCB-138 and PCB-153.





### Conclusions

Ecology collected sediment cores from Deep Lake, Spanaway Lake, and Lake Spokane in 2016 to evaluate recent trends in PCB deposition profiles. The following conclusions can be made:

#### **Temporal trends in T-PCBs**

- Sediment core profiles of Deep and Spanaway Lakes show T-PCB concentrations and fluxes have declined since peak levels, with a leveling off of T-PCB concentrations in the most-recent sediments. Maximum T-PCB concentrations occurred around 2001 and 1992 in the Deep and Spanaway cores, respectively.
- T-PCB concentrations and fluxes in the Lake Spokane core showed fairly stable levels between the late 2000s and early 2016. T-PCBs increased in the most-recent layer (2016), though this may reflect inherent variability in the sediments. When compared to a longer period of record (core data obtained from Serdar et al., 2011), T-PCB concentrations in the recent sediments were similar to concentrations in sediments deposited since approximately 1985, following peak concentrations occurring in the 1960s.

#### **PCB** congener profiles

- No obvious temporal patterns emerged in PCB congener profiles across all cores.
- There was an increase in the percent of hexachlorobiphenyls in the Deep Lake core through recent sediments and a slight decrease in pentachlorobiphenyls at the top of the core. However, differences in congener profiles in the Deep Lake core are magnified by low PCB concentrations in the lake.
- The Spanaway Lake core showed similar congener profiles over the years, suggesting little change in PCB sources or pathways to the lake over the last several decades. The only consistent trend was an increase in PCB-153 over the length of the core.
- The most-recent sediments of Lake Spokane (2015-2016) were dissimilar to the rest of the core, with a shift away from the more highly chlorinated congeners to lighter congeners. This pattern could reflect dissolution or leaching of the lighter congeners in down-core sediments after burial, or a recent shift in sources away from a heavier congener make-up.
- Across the three cores, congeners associated with Aroclor sources were dominant. The individual congeners PCB-110, PCB-138, PCB-139/149, and PCB-153 had the highest relative abundance of all congeners analyzed. These congeners are generally recalcitrant to degradation and metabolism (Buckman et al., 2007; Boon et al., 1997). Non-Aroclor congeners attributed to inadvertent production were generally not significant in the overall PCB composition. The exception to this was a higher relative concentration of PCB-209 in Deep and Spanaway Lakes. Congeners associated with microbial dechlorination were also not abundant in the cores (Brown et al., 1987; Rodenburg et al., 2010).

### Recommendations

- This sediment core monitoring program should continue to evaluate how levels of PCBs in the environment are changing. Based on the data presented in this report, it is likely that PCB concentrations have stabilized after their initial decline following the end of manufacturing in the 1970s. It would be helpful to know if additional work on controlling sources of PCBs could result in reducing current levels of PCB. On the current rotation basis, the next sampling for PCBs is scheduled for 2022.
- Additional investigation into sediment-bound PCBs in Lake Spokane should be considered. Suspended sediment sampling would help to confirm current concentrations and assess sources and/or pathways to the planktonic and benthic food web of Lake Spokane.

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### **Glossary of Terms**

**CF:CS Model:** Constant Flux: Constant Sedimentation Model. A model applied to <sup>210</sup>Pb measurements in sediment core samples to estimate dates with a constant sedimentation rate (Appleby and Oldfield, 1992). The CF:CS model can be used when unsupported 210Pb concentrations plotted on a logarithmic scale against cumulative dry mass appears linear. The slope of this relationship is used to calculate the sediment mass accumulation rate for the core.

**CRS Model:** Constant Rate of Supply Model. A model applied to <sup>210</sup>Pb measurements in sediment core samples to estimate dates and varying sedimentation rates (Appleby and Oldfield, 1978). The model works by measuring the difference in supported and unsupported <sup>210</sup>Pb in sediment horizons, and the relation of that difference to the inventory of unsupported <sup>210</sup>Pb of the whole core. Using the known half-life (22.3 years) of <sup>210</sup>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.

**Flux:** An estimated rate of net deposition of a contaminant 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.

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

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

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This report is available on the Department of Ecology's website at

Data for this project are available at Ecology's Environmental Information Management (EIM) website <u>www.ecy.wa.gov/eim/index.htm</u>. Search Study ID, SEDCORE16.

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