

---

# PBT Trends in Lake Sediment Cores: 2017 Results

Environmental Assessment Program

Publication: 20-03-002

Authored by: Callie Mathieu

---



## Abstract

---

Since 2006, the Washington State Department of Ecology (Ecology) has conducted long-term monitoring of persistent, bioaccumulative, and toxic (PBT) chemicals through freshwater sediment cores. Ecology collects sediment cores from three lakes each year to help characterize temporal trends of PBTs in the state. In 2017, Ecology collected sediment cores from Bosworth, Martha, and Wilderness Lakes in Western Washington for analysis of polycyclic aromatic hydrocarbons (PAHs).

In all three lakes, total (T-) PAH concentrations began rising in the early 1900s and peaked around mid-century. Concentrations then steadily declined between the 1950s and 2010s. In Martha Lake there was an increase in T-PAH concentrations in the top-most sediment layer, coinciding with a decrease in sediment accumulation rates.

Fluxes of T-PAHs to the coring locations exhibited similar profiles to concentrations in both the Bosworth and Wilderness cores. At Martha Lake, T-PAH fluxes did not decline between the late 1950s and 2010s like concentrations did.

Naturally occurring PAHs dominated the compound makeup profiles in pre-development sediments (<1900) in all three lakes. The stability of the compound profiles between the late 1920s and 2010s indicated little change in major watershed sources throughout the post-development period. High molecular weight PAHs dominated the profiles after the 1920s, suggesting pyrogenic sources (i.e., woodstove and vehicle emissions) were predominant. The exception to this was a spike in retene occurring in the last two decades in Wilderness Lake.

Of the ten sediment cores collected for this monitoring program since 2008, T-PAH concentrations in the majority of lakes peaked between the 1950s and 1990s. In general, sites with the highest concentrations showed declines in PAHs between the 1970s and the top of the core (most recent deposition; collected in 2017). Other lakes exhibited no consistent temporal trend over the last few decades, while Angle Lake was the only site where T-PAHs increased.

---

## Background

---

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 to reconstruct contaminant deposition profiles.

The target PBT contaminants monitored change each year on a rotating basis. In 2017, polycyclic aromatic hydrocarbons (PAHs) were analyzed in the sediment cores. PAHs are formed primarily from the incomplete combustion of carbon-containing materials and also occur in natural deposits of oil, coal, and tar. In the Puget Sound area, the main releases of PAHs to the environment are thought to be from woodstove and fireplace use, combustion of gasoline by vehicles, leaching of creosote-treated wood such as pilings and railroad ties, and petroleum spills (Ecology and King County, 2011). Major environmental pathways include groundwater, surface runoff, air deposition, and wastewater treatment plant effluent (Ecology and King County, 2011).

In 2012, Ecology and the state Department of Health (DOH) published a Chemical Action Plan (CAP) for PAHs and identified several actions to reduce PAH exposures in the state that were either currently being carried out or could be implemented (Ecology and Health, 2012). Priority recommendations included reducing residential wood-smoke emissions, developing outreach programs to reduce exposure from vehicles (e.g., eliminating drips and leaks and implementing anti-idling campaigns), and investigating the removal of creosote-containing products such as pilings and roofing materials.

Studies by the United States Geological Survey (USGS) in the 2000s identified coal tar sealcoats as a major source of PAHs to urban lake sediments (Mahler et al., 2005). PAH concentrations found in Washington State water bodies are more in alignment with asphalt-based sealcoats, which are much lower in PAHs. However, modeling of lake sediment PAH concentrations by Van Metre and Mahler (2010) showed a coal tar sealcoat signature in one small, urban Washington Lake – Lake Ballinger. Estimates from the state Department of Transportation (WDOT) suggested that coal tar based sealcoats have been used in Washington, but public and private use moved toward a blended product (20% coal tar pitch and 80% asphalt emulsion) in the 2000s (Ecology and Health, 2012). In 2011, Washington State passed a coal tar sealant ban (RCW 70.295), which prohibited all use or application of the products by 2013.

## Study Locations

In 2017, Ecology collected sediment cores from Lake Bosworth (Snohomish County), Martha Lake (Snohomish County), and Lake Wilderness (King County). Physical characteristics of the study sites are included in Table 1. Figure 1 displays the locations and coring sites for each lake.

Lake Bosworth is located in a rural area five miles northwest of Lake Stevens. The land surrounding the lake consists primarily of forested areas, with residences along the shoreline. Lake Bosworth is fed from a small stream in the southwest corner and drains to the north via Bosworth Lake Creek toward Pilchuck Creek. Precipitation in the area averages 61" per year. Watershed soils include a combination of gravelly loam and volcanic ash over glacial till.

Martha Lake lies in a heavily urbanized area between Lynnwood and Mill Creek. The watershed changed from 25% residential/commercial in the early 1970s to 80% by the mid-1990s (Snohomish

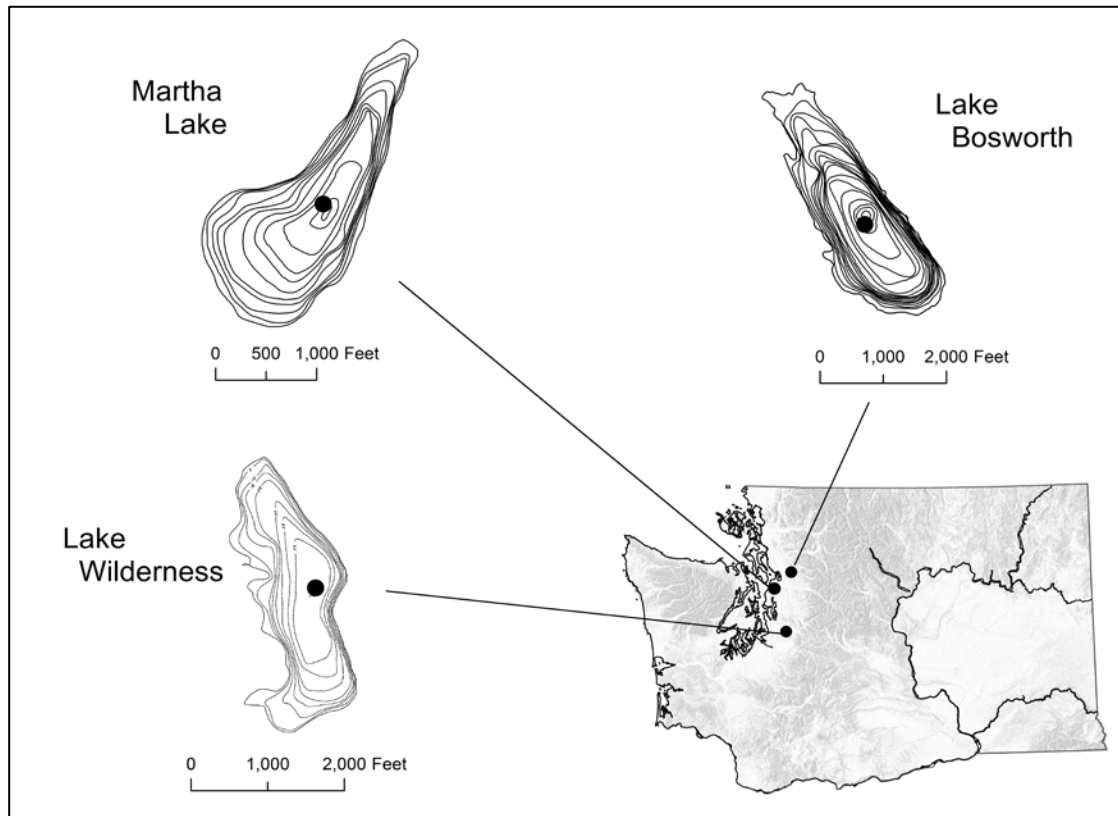
County, 2003). The lake is fed by several small streams to the north and west, including one that drains a portion of a major interstate highway. Water then flows out toward the south through Martha Lake Creek. The lake’s watershed receives an average precipitation of 39” annually and basin soils consist of gravelly sandy loam underlain by compact glacial till.

Lake Wilderness is a small, shallow lake within the city of Maple Valley. The shoreline and watershed of the lake consists of residential suburban land and a large public park along the northwest shore. Lake Wilderness is not fed by perennial streams, but by groundwater seeps, direct precipitation, and stormwater runoff (Aquatechnex, 2004). Water from the lake flows out to the west through Jenkins Creek. Average precipitation at Lake Wilderness is 51” per year. Watershed soils in the Lake Wilderness area consist of gravelly sandy loam.

**Table 1. Physical descriptions of 2017 study locations.**

Waterbody	County	Elevation (ft)	Max depth (ft)	Mean depth (ft)	Lake area (ac)	Watershed area (ac)	WA:LA
Lake Bosworth	Snohomish	563	79	35	110	902	8
Martha Lake	Snohomish	455	48	24	57	512	9
Lake Wilderness	King	470	38	21	69	422	6

WA:LA = watershed area to lake area ratio.



**Figure 1. Locations of 2017 sampling sites. Black circles indicate coring location.**

---

## Methods and Data Quality

---

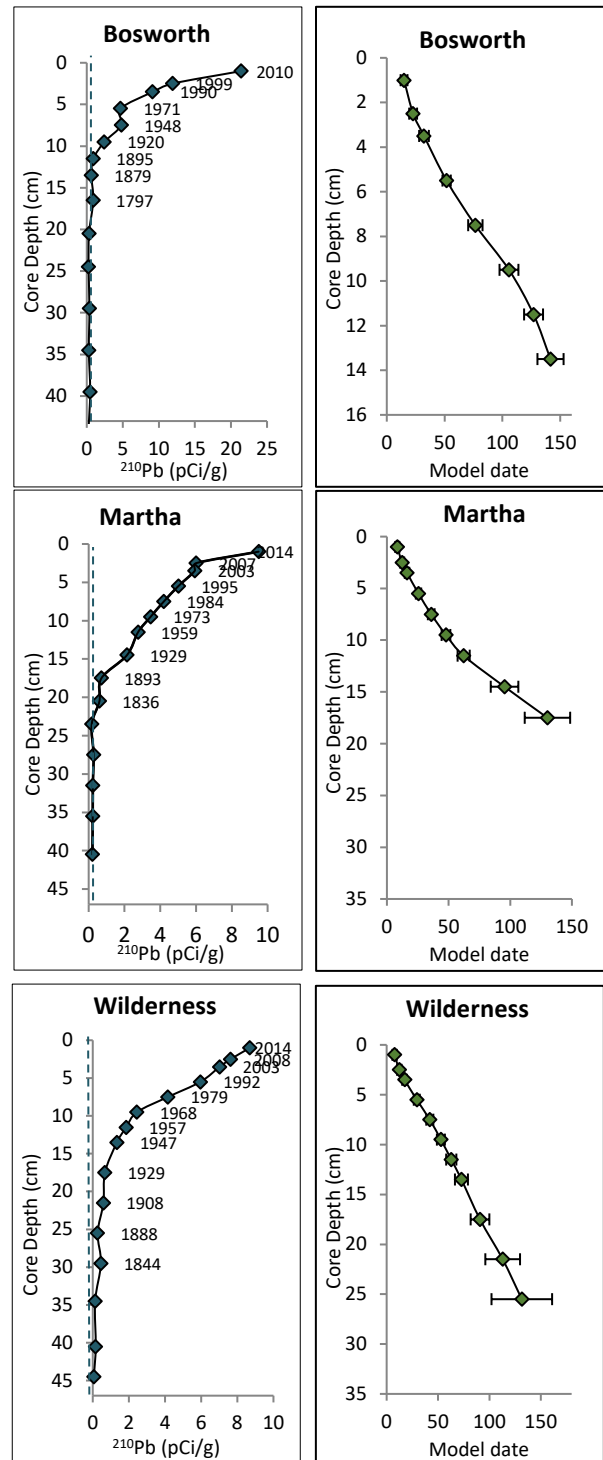
Ecology collected three sediment cores 35 – 45 cm in length using a Wildco© box corer following Ecology standard operating procedures (Furl and Meredith, 2008), the Quality Assurance Project Plan (QAPP) (Mathieu, 2016), and the QAPP addendum (Clinton, 2017). Surface sediments were collected using a standard PONAR for grain size analysis. Manchester Environmental Laboratory (MEL) analyzed or contracted analysis of sediment samples for total lead,  $^{210}\text{Pb}$ , total organic carbon (TOC), grain size, and PAHs using methods described in the QAPP and QAPP addendum. All data are available for download in Ecology's [EIM database](#) by searching Study ID: SEDCORE17.

Measurement quality objectives (MQOs) were met for all analyses, with the following exceptions. Two  $^{210}\text{Pb}$  laboratory duplicate relative percent differences (RPDs) were outside of MQOs (> 20%). Both samples were collected deep in the sediment core where activity was low. Associated samples were qualified "J" as estimated values. Two PAH matrix spike recoveries were outside of MQOs; dibenzothiophene was slightly lower than the 50% acceptance limit (48%) and 1-methylphenanthrene was slightly higher (152%) than the MQO upper limit of 150%. Five PAH surrogate results showed a high bias (>200%). The associated PAH samples were qualified "J" as estimates based on the matrix spike and surrogate exceedances.

# Core Dating

Dates were calculated for the three cores using  $^{210}\text{Pb}$  activities and the constant rate of supply (CRS) model (Appleby and Oldfield, 1978) (Figure 2). Percent solids were used to calculate dry mass. Supported  $^{210}\text{Pb}$  was estimated as the average activity present at deep intervals where there was no further apparent decline (represented as a dashed vertical line in Figure 2). Average supported  $^{210}\text{Pb}$  activities in Bosworth, Martha, and Wilderness Lakes were estimated as 0.32, 0.22, and 0.15 pCi/g (picocurie per gram), respectively.

Yearly unsupported  $^{210}\text{Pb}$  fluxes in pCi/cm<sup>2</sup>/yr were 0.38, 0.23, and 0.15 for Bosworth, Martha, and Wilderness Lakes, respectively. This report applies 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  $^{210}\text{Pb}$  flux values were divided by estimated total unsupported fluxes calculated using lake-specific precipitation values and atmospheric  $^{210}\text{Pb}$  deposition measured in Washington State (Lamborg et al., 2013). Focus factors for Bosworth, Martha, and Wilderness Lakes were 3.1, 2.9, and 1.5, respectively, and they are applied to contaminant fluxes throughout this report.



**Figure 2.**  $^{210}\text{Pb}$  Activity plotted against sediment core depth (left) and modeled age of sediment core depth with associated uncertainty/error (right).

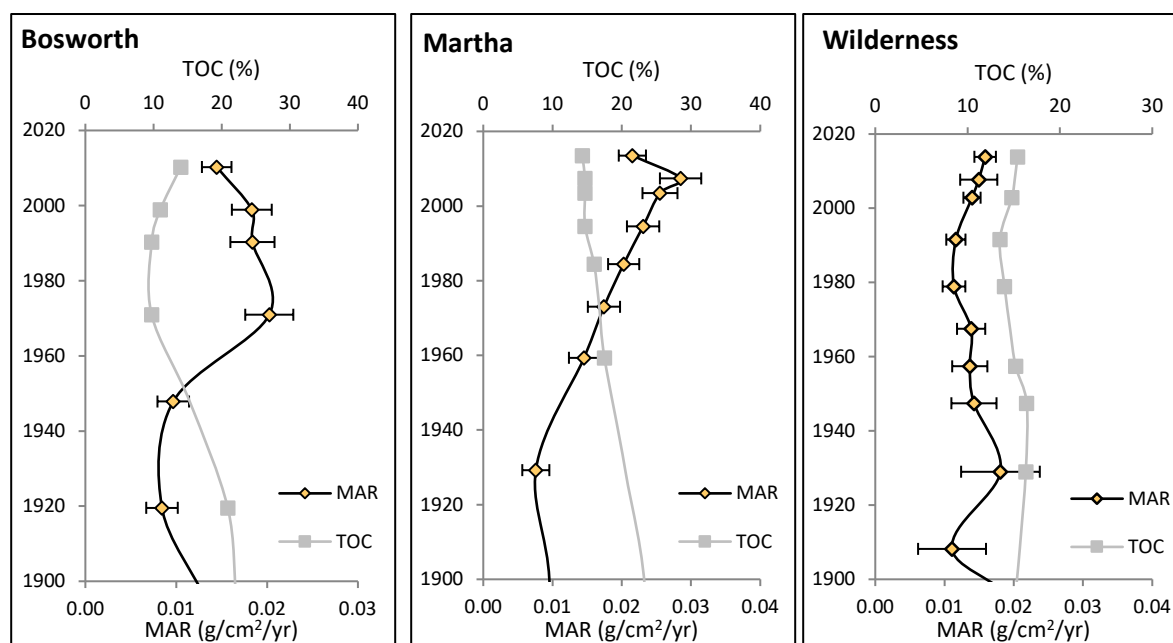
## Sediment Accumulation Rates

Sediment mass accumulation rates (MARs) were calculated using the CRS model for all three lakes. Figure 3 presents profiles of sediment MARs and associated MAR uncertainties, as well as total organic carbon (TOC). Among all three lakes, sediment MARs were low and similar to rates observed for other small urban and suburban lakes in the Puget Sound area (e.g., Angle Lake, Goodwin Lake, and Lake Meridian; Mathieu and Friese, 2012; Mathieu and McCall, 2015; Mathieu and McCall, 2016).

In the Bosworth Lake core, sediment MARs were in the range of 0.001 – 0.02 g/cm<sup>2</sup>/yr. An increase in MARs occurred in the 1960s, and MARs have remained just under 0.02 g/cm<sup>2</sup>/yr since the peak in the early 1970s.

Martha Lake accumulation rates increased steadily from 0.008 g/cm<sup>2</sup>/yr in the 1920s to a maximum of 0.03 g/cm<sup>2</sup>/yr in 2007. The most recent layer (2014) decreased in sediment MAR to a rate of 0.022 g/cm<sup>2</sup>/yr.

Lake Wilderness sediment MARs remained very consistent throughout the core. Between 1900 and the top of the core, MARs ranged from 0.01 to 0.02 g/cm<sup>2</sup>/yr.



**Figure 3. Profiles of estimated sediment accumulation rates (g/cm<sup>2</sup>/yr) and total organic carbon (TOC; %).**

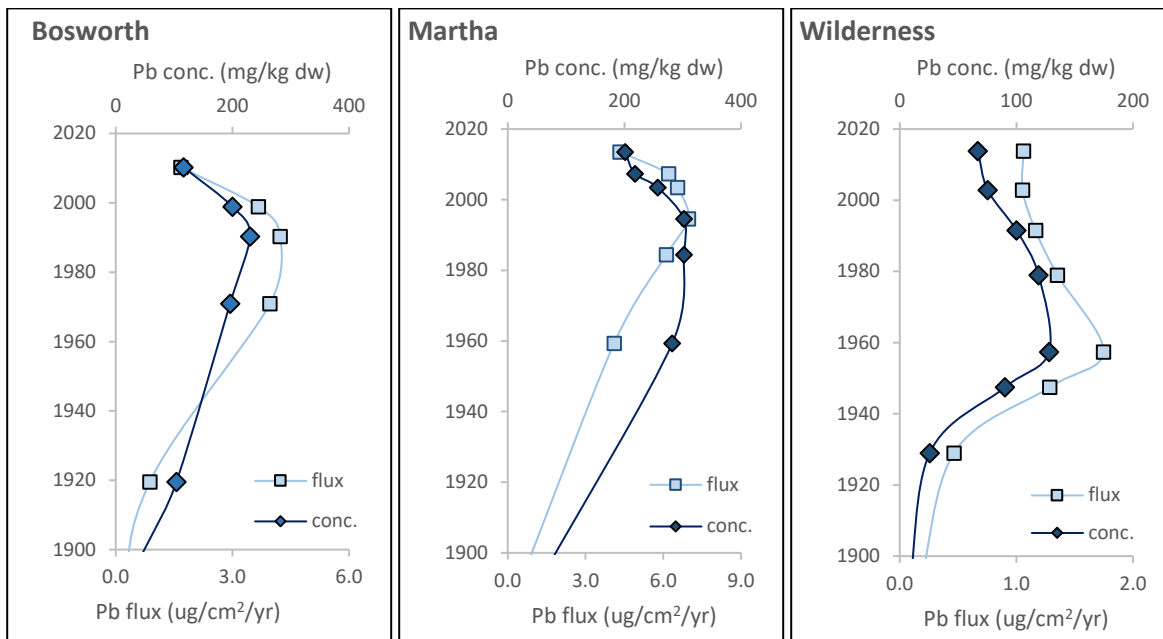
## Lead Results

For each site, ten intervals throughout the sediment core were analyzed for total lead. Figure 4 presents the total lead concentrations and fluxes for each sediment core. Fluxes are given with focus factors applied (as described in the “Core Dating” section) to provide an estimate of the rate of net deposition to the coring site within the lake.

Total lead concentrations and fluxes in the Bosworth Lake core increased steadily from the early 1900s to a peak of 230 mg/kg in 1990. Concentrations then declined through the most recent sediments to 116 mg/kg. The lead flux profile appeared similar to the concentration pattern, with fluxes peaking in 1990 and then declining.

In Martha Lake, lead concentrations increased over the first half of the 1900s, remained consistent around 300 mg/kg between the 1960s and 1990s, and then decreased to 201 mg/kg in the most recent sediment interval. The peak lead concentration for Martha Lake occurred in both 1984 and 1995. Lead fluxes in Martha Lake sediments showed a steady incline over the 1900s to a pronounced peak around 1995. Lead fluxes also declined in the most recent sediments collected.

Total lead concentrations and fluxes in the Lake Wilderness core increased sharply between the late 1920s and the 1950s. After the peak lead concentration in 1957 (128 mg/kg), concentrations remained in the low 100 mg/kg range until the early 1990s, then decreased to a concentration of 66.9 mg/kg in the top sediment horizon. Lead fluxes leveled off between the 1990s and 2010s.



**Figure 4. Profiles of lead (Pb) concentrations (mg/kg dw) and fluxes (ug/cm<sup>2</sup>/yr) in 2017 sediment cores.**

# Polycyclic Aromatic Hydrocarbon (PAH) Results

## T-PAHs

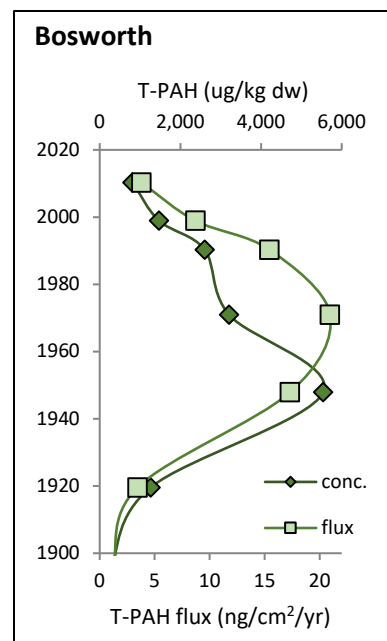
Ten horizons from each of the three lake sediment cores were analyzed for PAH compounds. Appendix A provides a complete list of the PAHs analyzed. Total PAH concentrations (T-PAHs) were calculated as the sum of 16 priority pollutant compounds, according to the procedure outlined in the QAPP (Clinton, 2017), which includes only detected values. Results reported as estimates (“J” qualified) were included in sums, but results reported as tentatively identified (“NJ” qualified) were not. All PAH concentrations are reported on a dry-weight basis unless otherwise noted.

### Lake Bosworth

T-PAH concentrations in Lake Bosworth ranged from 76 - 5,538 ug/kg, with a median of 1,036 ug/kg. Concentrations increased sharply from the beginning of the 1900s to a peak around 1948 and then declined throughout the second half of the 1900s and into the 2000s. The most recent section (2010) had the lowest T-PAH concentration (812 ug/kg) since pre-development. T-PAH fluxes in Lake Bosworth followed a similar pattern to the concentration profile, except the flux of PAHs coming into the lake remained high into the 1970s, before declining. T-PAH fluxes were in the range of 0.02 – 20.9 ng/cm<sup>2</sup>/yr.

**Table 2. Lake Bosworth sediment core PAH data.**

Lake	Core section (cm)	Estimated Year	TOC (%)	T-PAHs (ug/kg dw)	T-PAH flux (ng/cm <sup>2</sup> /yr)
Lake Bosworth	0-2	2010	14	812	3.79
	2-3	1999	11	1,473	8.71
	3-4	1990	9.74	2,602	15.4
	5-6	1971	9.72	3,207	20.9
	7-8	1948	---	5,538	17.3
	9-10	1920	20.9	1,261	3.43
	11-12	1895	---	290	1.23
	13-14	1879	23.4	127	0.61
	16-17	< 1850	21.8	109	0.03
	20-21	< 1850	---	76	0.02



**Figure 5. Profile of T-PAHs in Bosworth Lake sediment core.**

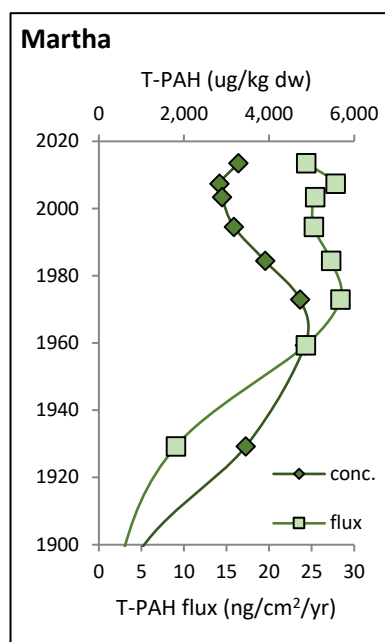


## Martha Lake

Martha Lake T-PAH concentrations ranged from a low of 271 ug/kg in pre-industrial sediments to a peak of 4,841 ug/kg occurring in the late 1950s. Concentrations declined modestly after the mid-century peak, but then showed a slight uptick in the most recent sediment layer, with the top of the core (2014) containing a T-PAH concentration of 3,279 ug/kg. This slight rise in T-PAH concentrations in the most recent sediments coincided with a drop in sediment accumulation rates in the top-most core layer. Unlike the concentration profile, T-PAH fluxes did not decline after the rise to mid-century levels. Instead, T-PAH fluxes continued to rise to a peak of 28.4 ng/cm<sup>2</sup>/yr in the early 1970s and then remained close to 25 ng/cm<sup>2</sup>/yr through the early 2010s.

**Table 3. Martha Lake sediment core PAH data.**

Lake	Core section (cm)	Estimated Year	TOC (%)	T-PAHs (ug/kg dw)	T-PAH flux (ng/cm <sup>2</sup> /yr)
Martha Lake	0-2	2014	14.3	3,279	24.3
	2-3	2007	14.7	2,830	27.8
	3-4	2003	14.7	2,889	25.4
	5-6	1995	14.7	3,173	25.2
	7-8	1984	16	3,904	27.3
	9-10	1973	---	4,725	28.4
	11-12	1959	17.5	4,841	24.3
	14-15	1929	---	3,452	9.03
	17-18	1893	23.5	721	2.35
	20-21	<1850	---	271	0.21



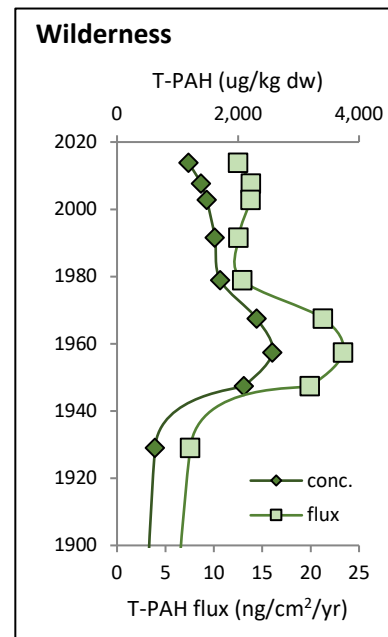
**Figure 6. Profile of T-PAHs in Martha Lake sediment core**

## Wilderness Lake

T-PAH concentrations in the Wilderness Lake core ranged from 37 – 2,567 ug/kg. The lake saw a rise in T-PAH concentrations and fluxes from the 1920s through the 1950s. T-PAH concentrations then declined over the rest of the core to a modern concentration of 1,182 ug/kg. T-PAH fluxes initially decreased after the mid-century peak through the 1980s, and then remained fairly consistent between 1980 and the 2010s.

**Table 4. Wilderness Lake sediment core PAH data.**

Lake	Core section (cm)	Estimated Year	TOC (%)	T-PAHs (ug/kg dw)	T-PAH flux (ng/cm <sup>2</sup> /yr)
Wilderness Lake	0-2	2014	15.4	1,182	12.5
	2-3	2008	---	1,389	13.8
	3-4	2003	14.8	1,482	13.8
	5-6	1992	13.5	1,618	12.6
	7-8	1979	14	1,706	12.9
	9-10	1968	---	2,307	21.3
	11-12	1957	15.2	2,567	23.4
	13-14	1947	16.4	2,097	19.9
	17-18	1929	16.3	628	7.56
	40-41	< 1850	---	36.9	---



**Figure 7. Profile of T-PAHs in Wilderness Lake sediment core.**

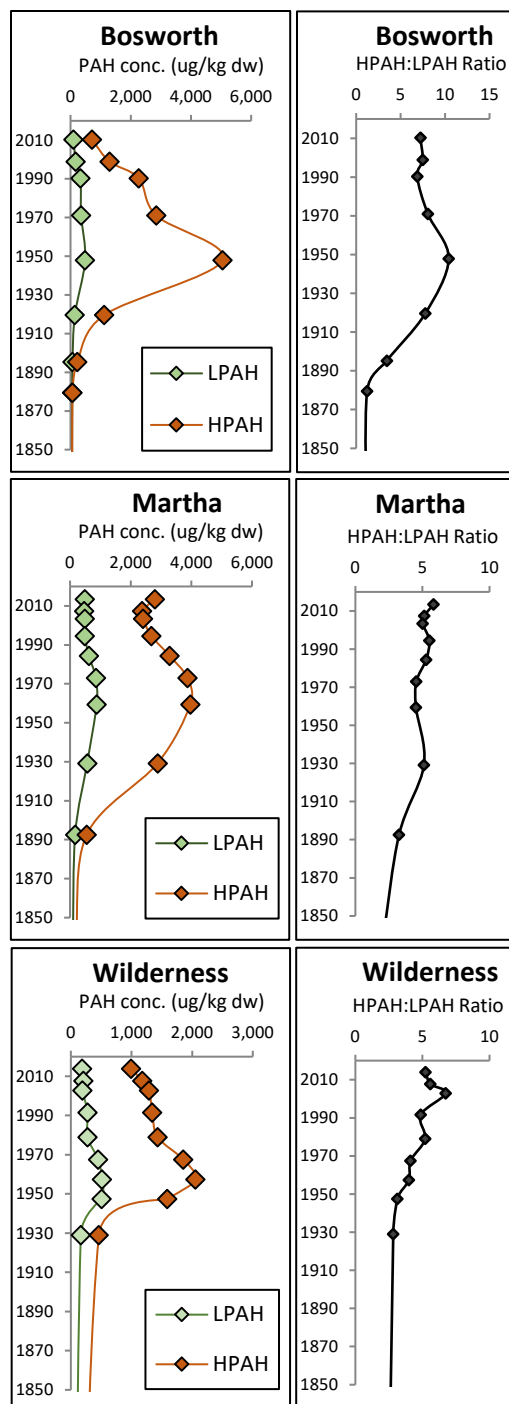
## Low/high molecular weight PAHs

Figure 8 displays profiles of low and high molecular weight PAH sums, as well as the ratio of the two. Low molecular weight PAH (LPAH) totals include compounds containing two to three aromatic rings<sup>1</sup> and high molecular weight PAH (HPAH) totals include compounds with four or more aromatic rings<sup>2</sup>. The LPAH compounds are predominantly derived from uncombusted sources such as oil and petroleum spills, while HPAHs are predominantly derived from combustion sources such as the burning of wood for heat or vehicle exhaust (Hites et al., 1980). Therefore, the HPAH:LPAH ratio can indicate the importance of combustion PAH sources (Van Metre et al., 2000). Other PAH ratios proposed by Yunker et al. (2002) and Yan et al. (2005) were explored for diagnostic purposes, but did not prove sensitive enough to differentiate pyrogenic sources.

Ratios of HPAH:LPAH were close to one in the pre-industrial sediments of all three lakes. In the Bosworth and Martha Lake cores, HPAH was elevated over LPAH beginning in the 1920s/1930s. The largest ratio was observed around the 1950s in the Bosworth core, when HPAH was 10 times higher than LPAH.

In Martha Lake, HPAH was around 5 times higher than LPAH for all of the post-1900 sediment intervals analyzed. The largest ratio occurred in the most recent sediment (2014), with HPAH 5.8 times greater than LPAH. The Wilderness Lake core showed a fairly close ratio of HPAH:LPAH into the late 1920s, and then HPAH rose above LPAH to a maximum ratio of 6.7 in 2003. In the most recent sediment intervals, HPAH:LPAH ratios for Bosworth, Martha, and Wilderness were 7.2, 5.8, and 5.2, respectively.

The HPAH:LPAH profiles for all three sediment cores suggest that combustion sources have been the dominant contributor to total PAHs in the lake sediments deposited after development of the watershed. Increasing vehicle traffic has been suggested as a major PAH source to lake sediments within the last few decades (Van Metre et al., 2000). In a 2007 assessment, wood-burning stoves and fireplaces accounted for the largest airborne PAH releases in the Puget Sound area, followed by vehicle traffic (Ecology, 2007 referenced by Roberts et al., 2011).



**Figure 6. Profiles of low and high molecular weight PAH sums and ratios.**

<sup>1</sup> LPAH sums include: naphthalene, acenaphthylene, acenaphthene, fluorene, anthracene, and Phenanthrene.

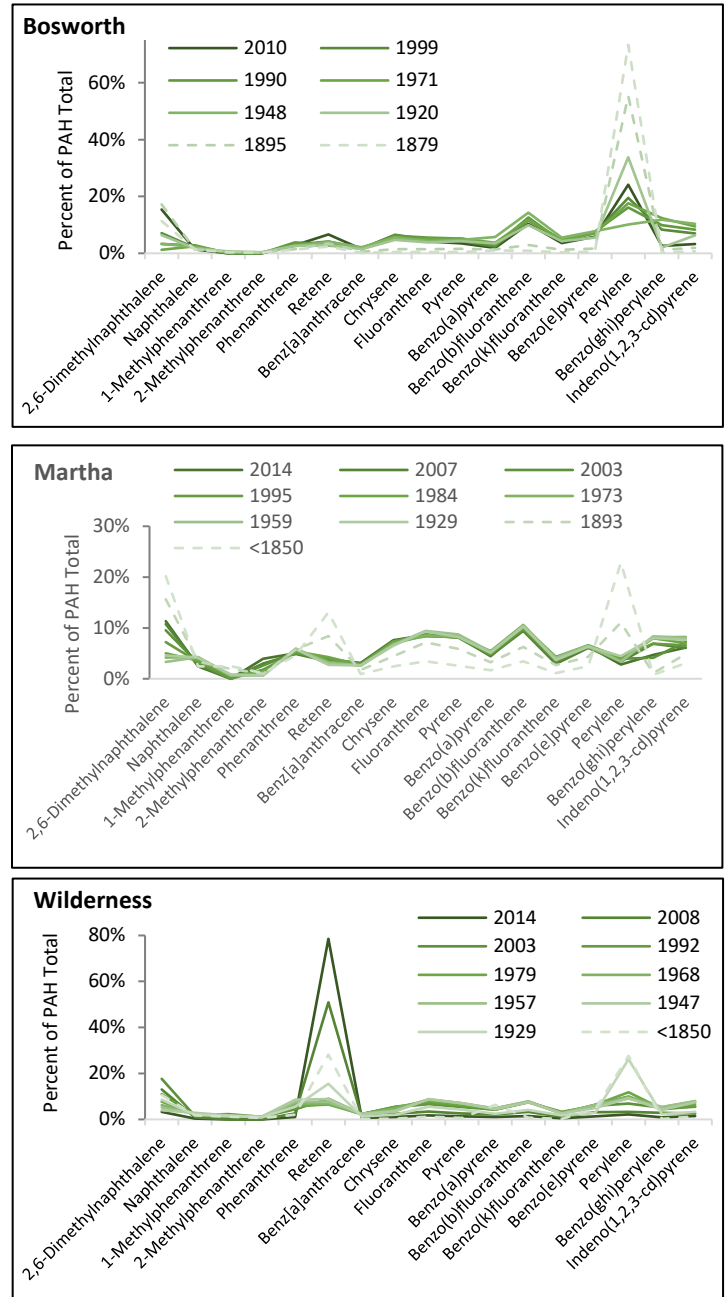
<sup>2</sup> HPAH sums include: chrysene, benzo(a)anthracene, fluoranthene, pyrene, benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(ghi)perylene, Indeno(1,2,3-cd)pyrene, and dibenzo(a,h)anthracene.

## PAH Compound Profiles

In 2017, 35 individual PAH compounds were analyzed in the sediment cores. Profiles showing the relative abundance of each compound are included in Figure 9. One goal of the 2017 sampling year was to assess whether PAHs have declined following a 2013 state ban on coal-tar sealants. However, due to the low sedimentation rates of the three lakes, the sediment cores did not have a high enough resolution to observe a post-ban effect.

For the Bosworth and Martha cores, the pre-1900s profiles were dissimilar to the rest of the core. Perylene – which can be formed naturally via sediment diagenesis (Wakeham et al., 1980) – was abundant in the sediments deposited before 1900. The make-up of PAH compounds was mostly unchanged between the 1920s and the 2010s in the Bosworth and Martha cores, suggesting PAH sources have remained stable over this time period. Slight changes observed during the last few decades tended to occur from variations in the percent contribution of 2,6-dimethylnaphthalene and benzo(ghi)perylene.

In Lake Wilderness, perylene and retene were dominant in pre-1900 sediments. A spike in retene also occurred in recent sediment accumulated (2008-2014). Retene can be formed by wood combustion, such as through forest fires or wood burning for heat (Ramdahl, 1983), or naturally through the breakdown of softwood resins (Hites et al., 1980). With retene excluded, the PAH compound profile trend was similar to the Bosworth and Martha cores; perylene was dominant in the early sediment layers (1929 and <1850), the profile was relatively stable from the late 1940s through the most recent sediments, and 2,6-dimethylnaphthalene was variable throughout.



**Figure 7. PAH Compound Profiles showing Percent of T-PAH Total in Sediment Cores.**

PAH Total includes sum of 35 compounds listed in Appendix A. Compounds making up less than 1% of any sample PAH total were excluded from the graph.

## Statewide Trends

This long-term monitoring study has evaluated PAHs in sediment cores collected in 2008, 2009, 2010, 2011, and 2017. Ten cores over this sampling period were successfully age-dated and contained consistent PAH detections throughout the respective cores. T-PAH concentrations, fluxes, and peak years for all ten lakes are presented in Table 5.

Across the ten lakes, peak years for T-PAH concentrations and fluxes varied widely. T-PAH concentrations peaked as early as in pre-industrial sediments of Lacamas Lake and as late as 2004 in Offutt Lake. The majority of lakes had peak T-PAH concentrations occurring between the 1950s and 1990s. The sites with relatively high peak T-PAH concentrations showed decreased levels between the 1970s and the top of the core. The exception to this was Angle Lake, where T-PAH concentrations increased between the 1970s and early 2000s. All other sites had either no consistent pattern or a decrease in PAH concentrations between the 1970s and the top of the core.

In other areas of the United States, PAH trends in sediment cores have also varied, with cores collected from the Great Lakes showing PAH levels peaking around the mid-20<sup>th</sup> century and a subsequent decrease after reductions in coal emissions (Schneider et al., 2001; Simcik et al., 1996). Cores collected across the U.S. from newly urbanized areas have showed PAHs on a recent increasing trend due to urban sprawl (increased traffic and impervious surfaces) and coal-tar sealcoat use (Van Metre and Mahler, 2005; Van Metre et al., 2000; Van Metre and Mahler, 2010). A pattern of no-trend over the last few decades was reported for PAHs in non-urban lakes across the U.S. (Van Metre and Mahler, 2005).

**Table 5. PAH Concentrations and fluxes in sediment cores collected statewide.**

Year collected:	Lake	Max T-PAH conc. (ug/kg)	Peak year (conc.)	T-PAH conc. in 0-2 cm (ug/kg)	Peak T-PAH flux (ng/cm <sup>2</sup> /yr)	Peak year (flux)	T-PAH flux in 0-2 cm (ng/cm <sup>2</sup> /yr)
2008*	Lacamas	577	< 1850	33	13	1931	2.2
2008*	Washington	1117	1990	638	89	1990	58
2008*	Offutt	219	2004	82	7.9	2004	2.8
2009†	American	1825	1931	290 U	25	1931	7
2010‡	Wenatchee	347	1963	140	12	1999	7.5
2011§	Samish	2536	1923	1203	93	2005	73
2011§	Angle	7606	1999	7110	207	1999	176
2017#	Bosworth	5538	1948	812	65	1971	12
2017#	Wilderness	2567	1957	1182	35	1957	19
2017#	Martha	4841	1959	3279	82	1973	71

U = not detected at or above the indicated concentration. T-PAH sum includes only the 16 priority pollutant PAHs.

\* Furl et al., 2009;

† Furl and Roberts, 2010;

‡ Furl and Roberts, 2011;

§ Mathieu and Friese, 2012;

# Current study.

---

## Conclusions

---

In 2017 Ecology collected sediment cores from Bosworth, Martha, and Wilderness Lakes to evaluate temporal trends in PAH sediment deposition. Sediment core profiles showed the following trends:

- ◆ **Sediment mass accumulation rates (MARs):** All three lake cores had low sediment MARs of less than 0.03 g/cm<sup>2</sup>/yr. These rates are similar to those observed for other small urban and suburban lakes in the Puget Sound area. Profiles of sediment MARs varied among the three lakes.
- ◆ **Lead concentrations and profiles:** All three lakes showed increasing trends in total lead concentrations and fluxes over the first half of the 1900s, coincident with rising leaded gasoline use. Peak lead concentrations were observed around 1990, 1984/1995, and 1957 in the Bosworth, Martha, and Wilderness Lake cores, respectively. Lead concentrations and fluxes declined since their peaks in all three sediment cores.
- ◆ **Total PAH concentrations and profiles:** T-PAH concentrations began rising in the 1920s and peaked by mid-century in all three sediment cores. In Bosworth and Wilderness Lakes, T-PAH concentrations consistently declined after a maximum around 1948 and 1957, respectively. Martha Lake T-PAH concentrations declined after peaking around 1959 until about 2007 and then showed a slight uptick in PAHs in the most recent sediment as sediment accumulation rates dropped. Fluxes of T-PAHs to the coring locations exhibited similar profiles to PAH concentrations, with the exception of fluxes remaining stable (not declining) between the late 1950s and the 2010s in Martha Lake.
- ◆ **Individual PAH Compounds:** Naturally occurring PAHs dominated the compound makeup of core sediments deposited before the 1900s. The compound makeup was fairly stable between the late 1920s and the most recent sediments analyzed, indicating little change in watershed sources over most of the post-development period. High molecular weight PAHs dominated the profiles after the 1920s, suggesting pyrogenic sources (i.e., woodstove and vehicle emissions) were the primary contributors. The possible exception to this was a spike in retene occurring in the last two decades in Wilderness Lake, which may be a marker of the natural breakdown of softwood resins.
- ◆ **Statewide Trends:** This monitoring program has analyzed PAHs in sediment cores from ten lakes since 2008. T-PAHs peaked between the 1950s and 1990s in the majority of lakes. Sites with the highest peak T-PAH concentrations saw decreases between the 1970s and the top of the core (most recent sample), with the exception of Angle Lake (King County) where concentrations increased. The other nine lakes exhibited no consistent temporal pattern in T-PAH concentrations or fluxes over the last few decades.

---

## Recommendations

---

Results from this 2017 study support the following recommendations:

- ◆ The sediment core monitoring program should continue analyzing PAHs in sediment cores on the current rotation schedule. The next scheduled sampling year for PAHs is 2023. While the sediment cores collected in 2017 did not contain a high enough resolution to capture changes in PAH levels after the state's ban on coal-tar sealants, cores collected during 2023 are likely to capture the correct time period. Future site selection should focus on targeting lakes with higher sedimentation rates.
- ◆ For the 2023 round of PAH sampling, the monitoring program should consider adding PAH analytes that would help identify and differentiate pyrogenic sources (i.e., 1,7-dimethylphenanthrene and methylphenanthrenes).

---

## 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.
- Aquatechnex, 2004. Lake Wilderness: Draft Integrated Aquatic Vegetation Management Plan. Prepared for Lake Wilderness Lake Management District Citizens Advisory Committee. Maple Valley, WA.
- Clinton, C., 2017. Addendum 1 to Quality Assurance Project Plan: Long-Term Monitoring of Persistent, Bioaccumulative, and Toxic Chemicals using Age-Dated Lake Sediment Cores. Washington State Department of Ecology, Olympia WA. Ecology Publication 17-03-113. <https://fortress.wa.gov/ecy/publications/SummaryPages/1703113.html>
- Ecology, 2007. Washington State Base Year 2005 County Inventories. Prepared by Sally Otterson, Washington State Department of Ecology, Air Quality Program, Olympia, WA.
- Ecology and Health, 2012. PAH Chemical Action Plan. Washington State Department of Ecology, Olympia, WA. Ecology Publication 12-07-048. <https://fortress.wa.gov/ecy/publications/SummaryPages/1207048.html>
- Ecology and King County, 2011. Control of Toxic Chemicals in Puget Sound: Assessment of Selected Toxic Chemicals in the Puget Sound Basin, 2007-2011. Washington State Department of Ecology, Olympia, WA and King County Department of Natural Resources, Seattle, WA. Ecology Publication 11-03-055. <https://fortress.wa.gov/ecy/publications/SummaryPages/1103055.html>
- Furl, C., C. Meredith, and M. Friese, 2009. Determination of PBT Chemical Trends in Selected Washington Lakes Using Age-Dated Sediment Cores, 2008 Results. Washington State Department of Ecology, Olympia, WA. Publication 09-03-030. <https://fortress.wa.gov/ecy/publications/summarypages/0903030.html>
- Furl, C. and T. Roberts, 2010. Determination of PBT Chemical Trends in Selected Washington Lakes Using Age-Dated Sediment Cores, 2009 Sampling Results. Washington State Department of Ecology, Olympia, WA. Publication 10-03-035. <https://fortress.wa.gov/ecy/publications/summarypages/1003035.html>
- Furl, C. and T. Roberts, 2011. PBT Chemical Trends in Washington State Determined from Age-Dated Lake Sediment Cores, 2010 Sampling Results. Washington State Department of Ecology, Olympia, WA. Publication 11-03-045. <https://fortress.wa.gov/ecy/publications/summarypages/1103045.html>
- Harrison, R.M., D.J.T. Smith, and L. Luhana, 1996. Source Apportionment of Atmospheric Polycyclic Aromatic Hydrocarbons Collected from an Urban Location in Birmingham, U.K. *Environmental Science and Technology*, Vol. 30(3): 825-832.
- Hites, R.A., R.E. LaFlamme, and J.G. Windsor Jr., 1980. Polycyclic Aromatic Hydrocarbons in an Anoxic Sediment Core from the Pettaquamscutt River (Rhode Island, U.S.A.). *Geochimica et Cosmochimica Acta*, Vol. 44: 873-878.
- Lamborg, C.H., D.R. Engstrom, W.F. Fitzgerald, and P.H. Balcom, 2013. Apportioning global and non-global components of mercury deposition through 210Pb indexing. *Science of the Total Environment*, Vol. 448: 132-140.



- 
- Mahler, B.J., P.C. Van Metre, T.J. Bashara, J.T. Wilson, and D.A. Johns, 2005. Parking Lot Sealcoat: An Unrecognized Source of Urban Polycyclic Aromatic Hydrocarbons. *Environmental Science and Technology*, Vol. 39: 5560-5566.
- Mathieu, C., 2016. Quality Assurance Project Plan: Long-Term Monitoring of Persistent, Bioaccumulative, and Toxic Chemicals using Age-Dated Lake Sediment Cores. Washington State Department of Ecology, Olympia, WA. Ecology Publication 16-03-118. <https://fortress.wa.gov/ecy/publications/SummaryPages/1603118.html>
- 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. Ecology Publication 12-03-045. <https://fortress.wa.gov/ecy/publications/summarypages/1203045.html>
- Mathieu, C. and M. McCall, 2015. PBT Chemical Trends Determined from Age-Dated Lake Sediment Cores, 2014 Results. Washington State Department of Ecology, Olympia, WA. Ecology Publication 15-03-027. <https://fortress.wa.gov/ecy/publications/SummaryPages/1503027.html>
- Mathieu, C. and M. McCall, 2016. PBT Chemical Trends Determined from Age-Dated Lake Sediment Cores, 2015 Results. Washington State Department of Ecology, Olympia, WA. Ecology Publication 16-03-046. <https://fortress.wa.gov/ecy/publications/SummaryPages/1603046.html>
- Norton, D., D. Serdar, J. Colton, R. Jack, and D. Lester, 2011. Control of Toxic Chemicals in Puget Sound: Assessment of Selected Toxic Chemicals in the Puget Sound Basin, 2007-2011. Washington State Department of Ecology, Olympia, WA. Ecology Publication 11-03-055. <https://fortress.wa.gov/ecy/publications/SummaryPages/1103055.html>
- Ramdahl, T., 1983. Retene – A Molecular Marker of Wood Combustion in Ambient Air. *Nature*, 306: 580-582.
- Roberts, T., D. Serdar, J. Maroncelli, and H. Davies, 2011. Control of Toxic Chemicals in Puget Sound – Phase 3: Primary Sources of Selected Toxic Chemicals and Quantities Released in the Puget Sound Basin. Washington State Department of Ecology, Olympia, WA. Ecology Publication 11-03-024. <https://fortress.wa.gov/ecy/publications/summarypages/1103024.html>
- Schneider, A.R., H.M. Stapleton, J. Cornwell, and J.E. Baker, 2001. Recent Declines in PAH, PCB, and Toxaphene Levels in the Northern Great Lakes as Determined from High Resolution Sediment Cores. *Environmental Science and Technology*, Vol. 35(19): 3809-3815.
- Simcik, M.F., S.J. Eisenreich, K.A. Golden, S.-P. Liu, E. Lipiatou, D.L. Swackhamer, and D.T. Long., 1996. Atmospheric Loading of Polycyclic Aromatic Hydrocarbons to Lake Michigan as Recorded in the Sediments. *Environmental Science and Technology*, Vol. 30(10): 3039-3046.
- Snohomish County, 2003. State of the Lakes Report, March 2003. Snohomish County Public Works Surface Water Management. [https://snohomishcountywa.gov/DocumentCenter/View/62606/martha\\_s](https://snohomishcountywa.gov/DocumentCenter/View/62606/martha_s)
- Van Metre, P.C., B.J. Mahler, and E.T. Furlong, 2000. Urban Sprawl Leaves Its PAH Signature. *Environmental Science and Technology*, Vol. 34: 4064-4070.
- Van Metre, P.C. and B.J. Mahler, 2005. Trends in Hydrophobic Organic Contaminants in Urban and Reference Lake Sediments across the United States, 1970-2001. *Environmental Science and Technology*, Vol. 39: 5567-5574.
-

- 
- Van Metre, P.C. and B.J. Mahler, 2010. Contribution of PAHs from coal-tar pavement sealcoat and other sources to 40 U.S. Lakes. *Science of the Total Environment*, Vol. 409(2): 334-344.
- Wakeham, S.G., C. Schaffner, and W. Giger, 1980. Polycyclic aromatic hydrocarbons in recent lake sediments – II. Compounds derived from biogenic precursors during early diagenesis. *Geochemical et Cosmochimica Acta*, Vol. 44: 415-429.
- Yan, B., T.A. Abrajano, R.F. Bopp, D.A. Chaky, L.A. Benedict, and S.N. Chillrud, 2005. Molecular Tracer of Saturated and Polycyclic Aromatic Hydrocarbon Inputs into Central Park Lake, New York City. *Environmental Science and Technology*, Vol. 39: 7012-7019.
- Yunker, M.B., R.W. Macdonald, R. Vingarzan, R.H. Mitchell, D. Goyette, and S. Sylvestre, 2002. PAHs in the Fraser River Basin: A Critical Appraisal of PAH Ratios as Indicators of PAH Source and Composition. *Organic Geochemistry*, Vol. 33: 489-515.

# Appendix A. PAH Compounds Analyzed

Table A-1. PAH Compounds analyzed in 2017 sediment cores.

Analyte	PBT List	CWA PP
1,6,7-trimethylnaphthalene		
1'1 biphenyl		
1-Methylnaphthalene		
1-Methylphenanthrene		
2,6-dimethylnaphthalene		
2-Chloronaphthalene		
2-Methylfluoranthene		
2-Methylnaphthalene		
2-Methylphenanthrene		
4-Methyldibenzothiophene		
5-Methylchrysene		
9H-Fluorene, 1-methyl-		
Acenaphthene		x
Acenaphthylene		x
Anthracene		x
Benzo(a)anthracene		x
Benzo(a)pyrene		x
Benzo(b)fluoranthene	x	x

Analyte	PBT List	CWA PP
Benzo(ghi)perylene	x	x
Benzo(k)fluoranthene	x	x
Benzo[e]pyrene		
Carbazole		
Chrysene		x
Dibenzo(a,h)anthracene	x	x
Dibenzofuran		
Dibenzothiophene		
Fluoranthene	x	x
Fluorene		x
Indeno(1,2,3-cd)pyrene	x	x
Naphthalene		x
Perylene		
Phenanthrene		x
Phenanthrene, 3,6-dimethyl-		
Pyrene		x
Retene		

PBT List = Washington's Persistent, Bioaccumulative, and Toxic Chemical List

CWA PP = Clean Water Act Priority Pollutant

---

## Appendix B. Glossary of Terms

---

**CRS Model:** The Constant Rate of Supply (CRS) Model is applied to  $^{210}\text{Pb}$  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  $^{210}\text{Pb}$  in sediment horizons, and the relation of that difference to the inventory of unsupported  $^{210}\text{Pb}$  of the whole core. Using the known half-life (22.3 years) of  $^{210}\text{Pb}$  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:** Flux is 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 because fine-grained sediments preferentially deposit in these areas.

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

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

---

## Publication Information

---

This report is available on the Department of Ecology's website at:  
<https://fortress.wa.gov/ecy/publications/SummaryPages/2003002.html>.

Data for this project are available in Ecology's [EIM Database](#). Search study ID: SEDCORE17.

The Activity Tracker Code for this study: 06-513.

### Suggested Citation:

Mathieu, C., 2020. PBT Trends in Lake Sediment Cores: 2017 Results. Washington State Department of Ecology, Olympia. <https://fortress.wa.gov/ecy/publications/SummaryPages/2003002.html>.

Water Resource Inventory Area (WRIA) and 8-digit Hydrologic Unit Code (HUC) numbers for the study area:

- WRIAs: 07, 08, 09
- HUC numbers: 17020011, 17110012, 17110013

---

## Contact Information

---

Publication Coordinator  
Environmental Assessment Program  
Washington State Department of Ecology  
P.O. Box 47600  
Olympia, WA 98504-7600  
Phone: 360-407-6764

Washington State Department of Ecology – <https://ecology.wa.gov>

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

Any use of product or firm names in this publication is for descriptive purposes only and does not imply endorsement by the author or the Department of Ecology.

To request ADA accommodation for disabilities, or printed materials in a format for the visually impaired, call the Ecology ADA Coordinator at 360-407-6831 or visit <https://ecology.wa.gov/accessibility>.

People with impaired hearing may call Washington Relay Service at 711.

People with speech disability may call 877-833-6341.