PBT Trend Monitoring: Measuring Lead in Suspended Particulate Matter, 2015 Results

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2015 Highlighted Findings

- The highest lead concentrations in suspended particulate matter (SPM) were found at the Spokane River sites.
- No significant temporal trends were detected at any of the monitoring sites for the past five to eight years, except for a significant decreasing trend at Thornton Creek.
- SPM-bound lead loads at most sampled locations appear to have been diminished by drought conditions during 2015.

Why Monitor Lead?

Ecology monitors lead in Washington rivers and streams because of concern over its toxic and persistent properties. While lead is a naturally occurring element, human activities have resulted in widespread environmental contamination.

Lead affects humans and wildlife by harming developing nervous systems and other bodily systems. There are many sources of lead from the indoor and outdoor environment, and even minor exposures may cause harm.

Project Overview

In 2009, the state Department of Ecology (Ecology) and the state Department of Health developed a chemical action plan (CAP) for lead (Davies, 2009). The plan identified the toxic effects of lead, described lead's occurrence in the environment, and recommended ways to reduce human and wildlife exposure to lead. Roberts et al. (2011) also discusses potential lead sources in the Puget Sound region.

In 2008, Ecology began a long-term monitoring program to assess temporal changes in lead levels in Washington rivers and streams. Suspended particulate matter (SPM) samples were collected from 15 sites for analysis of total lead through 2014. In 2015, the number of sites was reduced to seven to focus on small urban streams and large rivers with substantial lead contamination (Mathieu, 2015). Samples were collected twice in the spring and twice in the fall at each site. In addition, sediment traps were deployed in the beds of two small streams during 2015 in order to evaluate lead concentrations in accumulated trapped particles.

The primary goal of this program is to provide a baseline for lead concentrations in the environment and evaluate temporal trends as CAP reduction strategies are implemented.

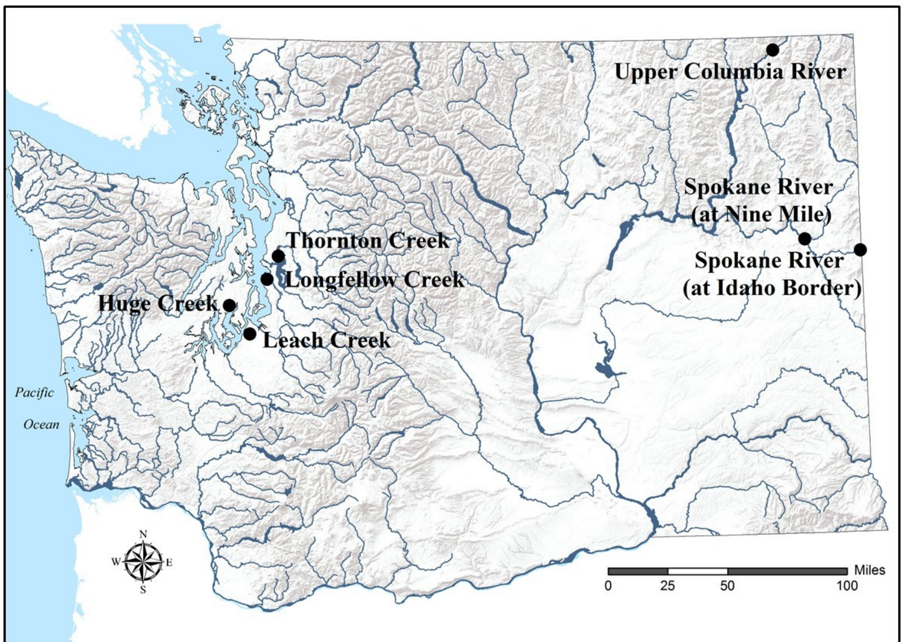

Figure 1. SPM Lead Monitoring Sites in Washington, 2015

For More Information

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

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

Complete 2015 data set in EIM (search Study ID: PbTrends15): www.ecy.wa.gov/eim

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Methods and Data Quality

SPM samples were collected following Ecology's standard operating procedure (SOP) for collection of suspended particles using in-line filtration (Meredith, 2008). Ambient water temperature, pH, and conductivity were measured following Ecology's SOP for collection and analysis of pH (Ward, 2007). Accumulated trapped sediments were collected using a Hamlin sediment trap as described in Lubliner (2012). Ecology's Manchester Environmental Laboratory analyzed total lead in the SPM and trapped sediments following EPA Methods 3050B and 200.8. Grain size was analyzed by Materials Testing & Consulting, Inc. using PSEP methodology (PSEP, 1986).

All quality assurance samples and analyses met measurement quality objectives (MQOs) set for this project (Meredith and Furl, 2008; Mathieu, 2015).

Results

Twenty-eight SPM samples were analyzed for lead in 2015. Lead concentrations ranged from below reporting limits (9-31 mg/kg) to 770 mg/kg. Samples collected from the Spokane River at the Idaho border contained the highest lead levels, followed by the Spokane River at Nine Mile. Lead concentrations were consistently higher than 100 mg/kg at the three urban streams and the Upper Columbia River. These six sites have consistently had the highest lead concentrations since their inclusion in the study.

Table 1. Statistical Summary of Lead in Suspended Particulate Matter, 2015 (mg/kg).

Season	n	FOD	Min	Max	Median	Mean	SD
Spring	14	86%	ND	750	132	259	228
Fall	14	86%	ND	770	184	305	251
2015	28	86%	ND	770	177	282	236

n = number of samples, FOD = frequency of detection
ND = not detected above reporting limit of 0.05 µg lead/filter,
SD = standard deviation

None of the four samples from Huge Creek, the urban reference stream, were detected at a reporting limit of 0.05 µg lead per filter. However, lead from Huge Creek was detected above the method detection limit of 0.003 µg per filter.

A summary of the 2015 lead results is shown in Table 1, and individual concentrations are displayed in Figure 2.

Comparison to Guidelines

Currently, no regulatory criteria exist for lead in SPM. The Sediment Management Standard rule, which includes a freshwater Sediment Cleanup Objective (SCO) for lead, took effect September 1, 2013 (WAC 173-204). Although the SCO is applicable to bottom sediments only, it is a useful threshold to assess levels of lead in SPM since some of the particulate matter will presumably settle to the bottom of the sampled stream or a receiving waterbody.

Twenty-one percent of samples (six of 28) contained lead concentrations above Washington's freshwater SCO of 360 mg/kg (Figure 2). All four samples collected from the Spokane River at the Idaho border and one each of the spring and fall samples from the Spokane River at Nine Mile were above this threshold.

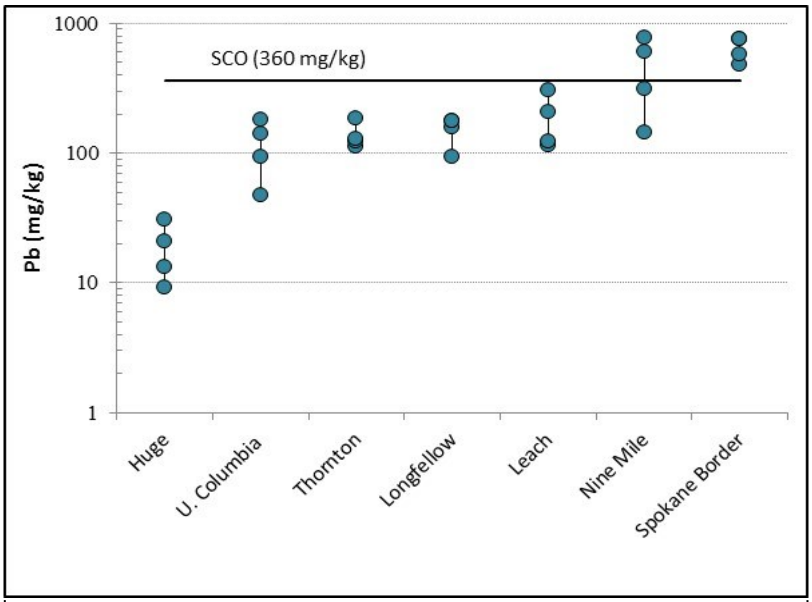

Figure 2. SPM Monitoring Locations Ranked by Mean 2015 Lead Concentrations.
SCO = Sediment Cleanup Objective.

Loading

Daily particulate lead loads were estimated for each sampling site, using lead concentrations calculated from SPM sampling and daily mean streamflow. Streamflow data were provided by the U.S. Geological Survey and the City of Seattle. Figure 3 displays the seasonal mean lead loads for the Spokane River, Upper Columbia River, and small stream monitoring sites.

The Upper Columbia River site contained the highest lead loads (22 - 79 kg/day), followed by the Spokane River Border (2.3 - 13 kg/day) and Ninemile (0.3 - 8 kg/day) sites. Higher loads during spring sampling corresponded with the much higher flows during the spring compared with fall.

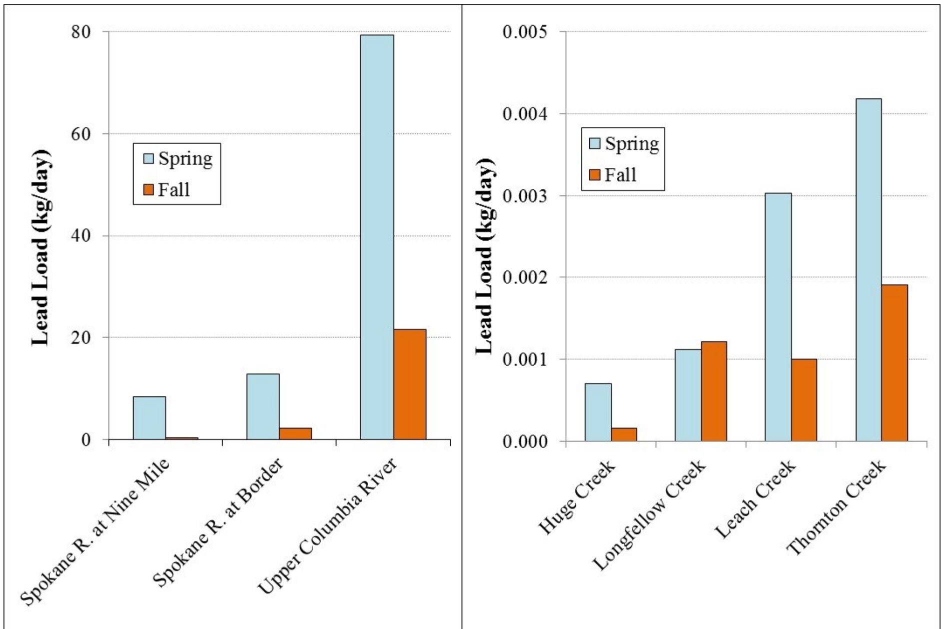

Figure 3. Estimated Mean Particle-bound Lead Loading at SPM Monitoring Sites, Spring and Fall 2015 (kg/day).
Values show the average of two samples per season.

Small streams SPM lead loads were 0.0002 - 0.004 kg/day, with the highest loads at Thornton Creek (0.002 - 0.004 kg/day) and the lowest at the rural reference stream, Huge Creek (0.0002 - 0.0007 kg/day). As in the larger rivers, seasonal lead loads within each stream corresponded to changes in discharge volume (i.e. streamflow).

Since loads of SPM lead reported here are instantaneous loads, they are strongly influenced by streamflow on the day of sampling. Therefore, caution should be used in comparing year-on-year loads.

Drought years such as the 2015 calendar year deliver especially low lead loads due to diminished runoff from melting snowpacks and unusually low rainfall affecting the Columbia and Spokane Rivers. Figure 4 plots flows for each sampling event as percentage of the average across all sampling events. Flows measured during 2015 sampling events were often lower than those during any previous sampling event. The Spokane River flows were particularly low, with spring 2015 flows only 20-30% of average.

Another factor in SPM lead loads are TSS (total suspended solids) levels which are positively correlated with flows (Spearman rank correlation; rho = 0.16—0.76). TSS levels during 2015 sampling were among the lowest for this project and was most pronounced for the small streams. The compounded effect of low TSS (to which lead is bound) and low flows (which convey the TSS) resulted in especially low SPM lead loads in 2015. These are factors to consider when analyzing lead loads in future monitoring.

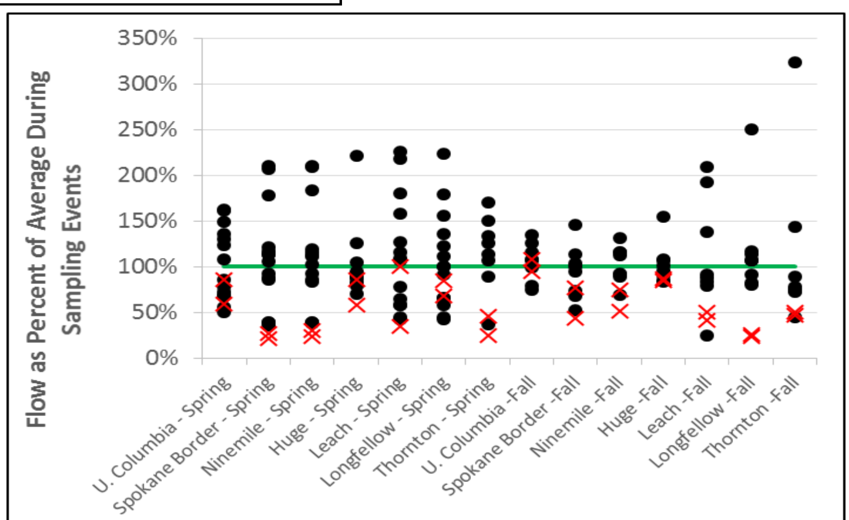

Figure 4. Flows During All Sampling Events Since 2008 as a Percentage of the Average Flow During All Sampling Events.
Black circles = flows during previous sampling events; red X's = flow during 2015; green line (100%) = average of all flows during sampling.

Temporal Trends

Sampling conducted in 2015 was the eighth year (2008-2015) of monitoring at the Columbia and Spokane River study sites and the fifth year (2011-2015) for those added to the program later (Thornton, Longfellow, Leach, and Huge Creeks). The Seasonal Kendall Test — a non-parametric rank test for identifying trends in seasonal time series data — was used to evaluate the SPM lead data for trends during these periods. Separate slope estimates were also calculated for spring and fall seasons. Huge Creek was excluded from this analysis since 90% of the results were below reporting limits; all other data were included.

Table 2. Temporal Trend Statistics for All SPM Lead Data, 2008-2015.

Waterbody	n	Seasonal Kendall Slope Estimate (mg/kg/yr)		Seasonal Kendall Pooled Season p-value
		Spring	Fall	
Spokane River				
Spokane River at Idaho Border	32	-30	-8.4	0.54
Spokane River at Nine Mile	32	1.3	15	0.54
Columbia River				
Columbia River, Upper	32	-3.0	-2.7	0.43
Small Streams				
Thornton Creek	20	-11	-17	**0.009**
Longfellow Creek	20	-5.7	9.0	0.39
Leach Creek	20	-5.6	4.7	0.86

Bold values indicate significance at $p<0.05$.

Table 2 shows results of the Seasonal Kendall Test and seasonal slope estimate at each site. Estimates of slopes were generally downward during the spring, whereas fall results showed a mix of upward and downward slopes. Thornton Creek, showing relatively large downward slope estimates for both spring and fall, was the only location demonstrating a significant overall trend ($p<0.05$). This site also showed a significant downward trend for 2011-2014.

The following sections describe general trends in lead concentrations observed at the monitoring sites. Spring and fall data are separated and fitted with simple (least squares) regressions lines as a visual tool to display patterns in the data.

Spokane River

Figure 5 shows SPM lead concentrations and streamflow at the Spokane River sites from 2008 to 2015. Opposite trends in lead concentrations appear to be occurring at the two sites; decreasing during both spring and fall at the border reach, and increasing during both seasons at Nine Mile. Maximum lead concentrations for both sites occurred in the spring of 2008 and 2012 during the highest flows, and minimum spring concentrations occurred during low-flow years (2009, 2010, 2015). Hallock (2010) found that total lead in the Spokane River was significantly ($p<0.05$) correlated with flow. His analysis also revealed a significant ($p<0.10$) decreasing trend in total lead concentrations in Spokane River water samples collected 1994 through 2009, although these decreases were not significant when data were flow-adjusted or when only data from 2001-2009 were evaluated.

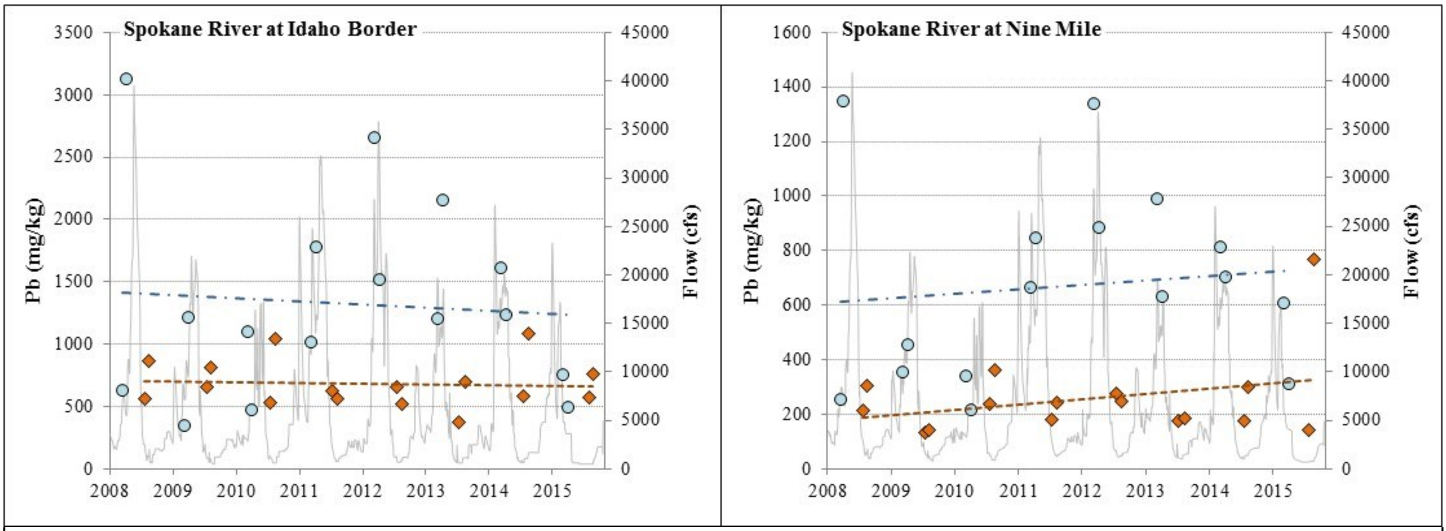

Figure 5. Lead Concentrations in SPM and Streamflow at Spokane River Sites, 2008 to 2015.

Note the different y-axes units and scales. Blue Circles=Spring Samples, Orange Diamonds=Fall Samples, Gray Line=Streamflow, Blue Dot Dash Line=Spring Trendline, Orange Dashed Line=Fall Trendline. Spokane River flow data provided by the U.S. Geological Survey.

Small Streams

Lead concentrations in SPM from Leach, Longfellow, and Thornton Creeks were comparable to those in the Upper Columbia River but much lower than lead concentrations in the Spokane River. These three urban streams continue to have SPM lead an order of magnitude higher than Huge Creek, the small stream reference site.

Spring SPM lead was generally lower than during previous years, while fall concentrations were generally higher (Figure 6). Downward spring trends in lead concentrations were observed at all urban stream locations, while fall concentrations trended upward except at Thornton Creek. As mentioned previously, overall SPM lead concentrations at Thornton Creek show a significant downward trend when analyzed using the Seasonal Kendall test ($p<0.05$).

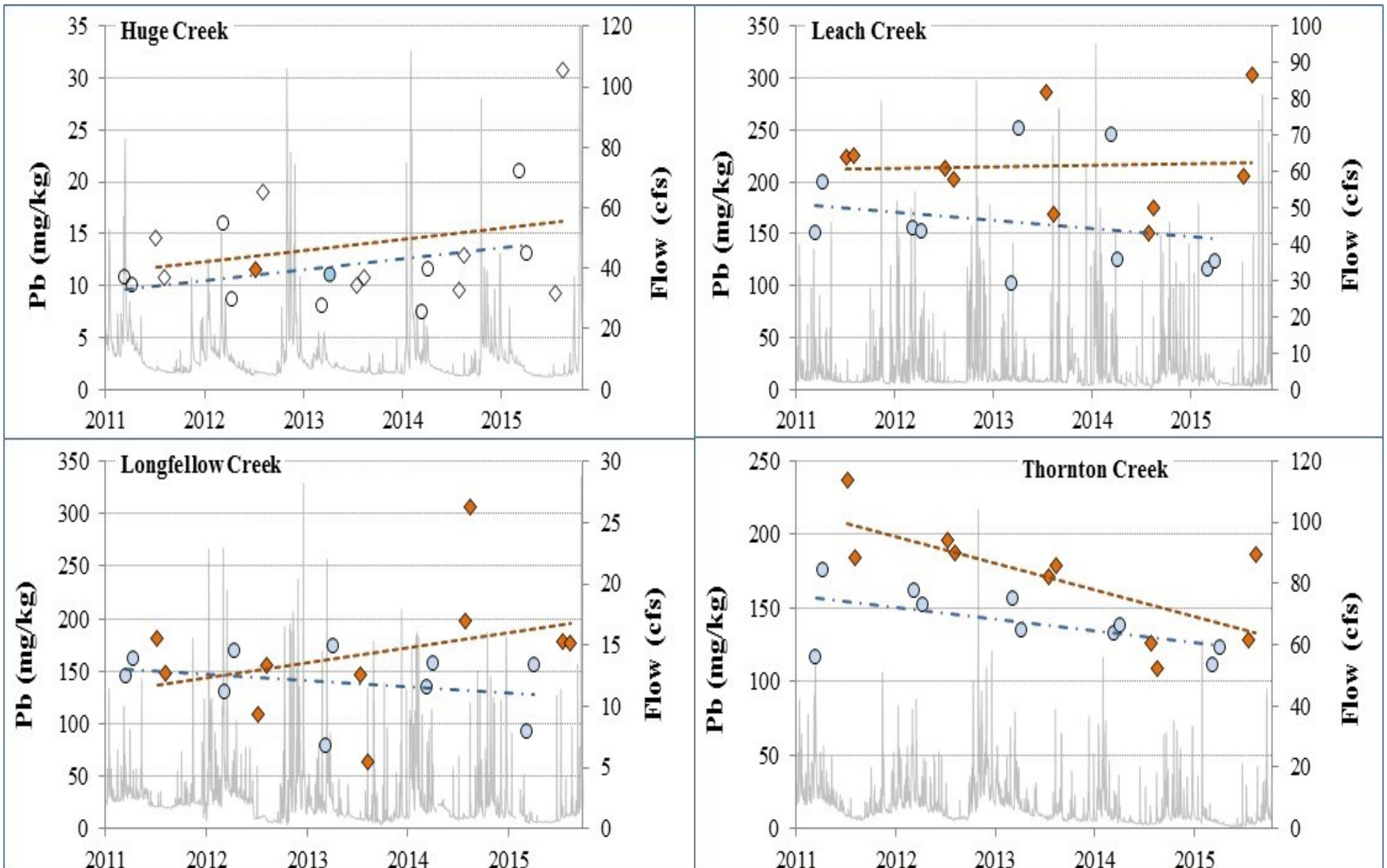

Figure 6. Lead Concentration in SPM and Streamflow at Small Stream Sites, 2008 to 2015.

Note the different y-axes units and scales. Blue Circles=Spring Samples, Orange Diamonds=Fall Samples, Gray Line=Streamflow, Blue Dot-Dash Line=Spring Trendline, Orange Dashed Line=Fall Trendline. Hollow Symbols=Values Above Detection Limit and Below Reporting Limit. Flow data for Huge and Leach Creeks provided by the U.S. Geological Survey. Flow data for Longfellow and Thornton Creeks provided by Seattle Public Utilities.

Upper Columbia River

Figure 7 displays the Columbia River SPM lead concentrations and streamflow from 2008 to 2015. Spring lead concentrations are generally more variable than fall concentrations, but the overall tend is nearly flat among years. Lead concentrations in fall samples appear to show a slight downward trend. Like the Spokane River, the highest lead concentrations at the Upper Columbia River site were measured in the spring of 2008 and 2012.

Additional Metals in Upper Columbia River SPM

In addition to lead, SPM samples from the Upper Columbia River were analyzed for cadmium, copper, and zinc. The Upper Columbia site is downstream from the Teck lead-zinc smelter in Trail, British Columbia. This smelter has been responsible for historically large discharges of these metals as well as arsenic and mercury (Cox et al., 2005).

Results (Table 3) do not appear to show seasonal differences for any of these metals, although many additional rounds of sampling will be required before seasonal patterns are evident. Zinc concentrations are the most variable among the metals, ranging by an order-of magnitude in the four samples analyzed.

Metals in Columbia River border reach SPM have been analyzed intermittently since the early 1990s. Samples collected in the fall of 1992 and 1993 (Bortleson et al., 1994; Serdar et al., 1994) had much higher levels of cadmium, copper, and lead than those found in 2015. Median concentrations of copper and lead remained elevated in the 1996-2000 period (Kelly et al., 2001) even though the Teck smelter had ceased routine discharge of metallurgical slag in 1995. However, the concentrations of these metals appear to have declined since 2000. Caution should be used when making comparisons among studies since collection and analysis methods may be different.

Cadmium concentrations in Upper Columbia River SPM consistently exceed the SCO for cadmium (2.1 mg/kg). Copper and lead are well below their respective SCOs, and it appears that zinc concentrations continue to occasionally exceed the SCO (3,200 mg/kg).

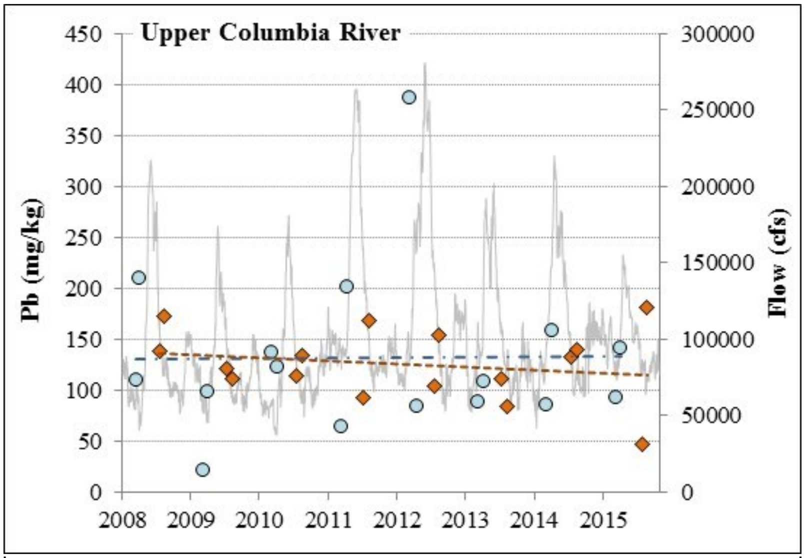

Figure 7. Lead Concentrations in SPM and Streamflow at the Upper Columbia River Site, 2008 to 2015.

Note the different y-axes units and scales. Blue Circles=Spring Samples, Orange Diamonds=Fall Samples, Gray Line=Streamflow, Blue Dot-Dash Line=Spring Trendline, Orange Dashed Line=Fall Trendline. Columbia River flow data provided by the U.S. Geological Survey.

Table 3. Current and Historical Concentrations of Metals in Upper Columbia River Suspended Particulate Matter (mg/kg).

Season/Year	Cadmium	Copper	Lead	Zinc
Spring 2015	5.3 J	63.3 J	92.7	3,990
	4.3 J	83.1	141	2,320
Fall 2015	2.6 J	66.3	46.8	232 J
	8.0 J	179	181	1,600 J
Year-Round 1996-2000	1.3 - 6.8 Med.=3.4	48 - 1,900 Med.=300	89 - 500 Med.=190	670 - 9,300 Med.=2,200
Fall 1992 & 1993	10-16	280-360	490-560	1,100-1,500
SCO	2.1	400	360	3,200

J=result was below reporting limit of 0.05 µg/filter for cadmium, 0.25 µg/filter for copper, and 2.5 µg/filter for zinc. Med.=median. SCO=Sediment Cleanup Objective.

Sediment Traps

Table 4. Lead Concentration and Grain Size in Material Collected from Sediment Traps During 2015

Waterbody	Season	Sediment Traps					SPM from Filters
		Lead (md/kg dw)	Gravel (%)	Sand (%)	Silt (%)	Clay (%)	Lead (md/kg dw)
Longfellow Creek	Spring	112	6.8	11.5	75.4	6.3	93 - 157
	Fall	67.1	NA	NA	NA	NA	177 - 178
Thornton Creek	Spring	71.0	1.6	55.6	40.3	2.5	112 - 123
	Fall	53.7	2.1	68.3	28.1	1.5	129 - 186

NA=not analyzed due to insufficient volume of sample material.

Hamlin-style sediment traps were deployed in two of the urban streams—Longfellow and Thornton Creeks—to capture particulate matter from the water column. These sediment traps were deployed on the bottom of a large culvert in Longfellow Creek and in the streambed of Thornton Creek for four-week durations in the spring and fall. Additional information about the sampling methods can be found in Mathieu (2015). Lubliner (2012) also provides extensive information on the use and efficacy of the Hamlin traps.

Table 4 shows concentrations of lead and grain size analysis of sediments recovered from the traps. Lead concentrations in accumulated sediments from Longfellow and Thornton Creeks were about 30-120% of the SPM concentrations from their respective locations and seasons. Lead concentrations in finer (silt + clay) sediment trap material more closely resemble concentrations in SPM. A greater percentage of fines accumulated at Longfellow Creek (81%) compared with Thornton Creek (avg. of 36%), although no grain size data were collected in the fall at Longfellow Creek due to an insufficient volume of sample material. The grain size results are consistent with the deployment locations at each creek. The location at Longfellow Creek had low velocity water with a soft substrate while the Thornton Creek location had higher velocity water with gravel substrate.

Conclusions

This report summarizes results from the eighth year of a long-term study to evaluate temporal trends in environmental lead concentrations in selected Washington State rivers and streams. A total of 28 suspended particulate matter (SPM) samples from seven sites were analyzed for lead in 2015 and sediment trap material was analyzed at two sites. Results include the following:

- Lead concentrations in SPM ranged from below reporting limits (9-31 mg/kg) at the small stream reference site to 750-770 mg/kg in the Spokane River.
- Samples collected from the Spokane River at the Idaho border contained the highest overall concentrations of SPM lead (mean of 640 mg/kg) followed by the Spokane River at Nine Mile (mean of 460 mg/kg). Lead concentrations in the Upper Columbia River and the three urban streams were comparable, with average concentrations of 120-190 mg/kg
- Twenty-one percent of samples (six of 28) contained lead concentrations above Washington's freshwater Sediment Cleanup Objective (SCO) of 360 mg/kg. All four samples collected from the Spokane River at the Idaho border and one each of the spring and fall samples from the Spokane River at Nine Mile were above this threshold. Upper Columbia River samples analyzed for cadmium consistently exceeded the SCO of 2.1 mg/kg, and zinc exceeded the SCO of 3,200 mg/kg in one of four samples analyzed.
- SPM lead concentrations at Thornton Creek appear to be steadily declining.
- The Upper Columbia River site had the highest SPM lead loading (22 -79 kg/day), due to the comparatively high discharge at this location. SPM lead loads at all locations were generally much higher in the spring than in fall. They appeared to be lower than in previous years due to the drought conditions of 2015.
- Lead concentrations in accumulated sediments from Longfellow and Thornton Creeks were about 30-120% of the SPM concentrations from their respective locations and seasons. Lead concentrations in finer (silt + clay) sediment trap material more closely resembles concentrations in SPM.

Recommendations

As stated in the Quality Assurance Project Plan, the primary goal of this program is to *provide a baseline for the contaminant [lead] in the environment and measure trends over time as chemical action plan [CAP] reduction strategies are implemented* (Meredith and Furl, 2008). This is a fairly broad goal that allows a flexible approach in designing the study, and it appears that the core of the current design is consistent with this goal.

Any changes to the study design should also reflect the program goal. Major changes made after 2014—dropping seven of the mid-sized to large river sites from the monitoring—were consistent with the program goal since these sites generally had low lead levels and were unlikely to respond to CAP implementation. The following recommendations are also consistent with the overall goal to establish a baseline and measure temporal trends:

- Continue to monitor lead in SPM from the urban streams and reference stream.
- Add more urban streams to the SPM monitoring site list. The lead concentrations in small, urban streams are most likely to respond to implementation of CAP reduction strategies.
- Increase the frequency of small stream sampling to provide a larger data set. Also consider sampling SPM under storm conditions and seasonal high flows. SPM data collected under different conditions could provide information to explain intra- and inter-seasonal variability.
- Continue to monitor lead in SPM at the Upper Columbia River and Spokane River sites because lead levels continue to be elevated at these locations. Also, continue to monitor SPM-associated cadmium, copper, and zinc at these locations.
- Sediment trap trials conducted during 2015 showed that Hamlin-style traps were successful at accumulating a large amount of material, but not the high percentage of silt and clay that is desirable. Since the ultimate goal of sediment traps is to collect a large volume of the same type of material as collected via filtration (i.e., very fine material), the Hamlin traps were not considered successful for this application. At present, there are no passive trap designs that are likely to yield a large volume of fines in small streams at low-moderate flow conditions, and active sampling procedures (e.g., multi-day centrifugation) are too labor-intensive. Therefore, the current recommendation is to cease deployment of Hamlin sediment traps but continue to investigate other options to reach the sample collection goal.

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This report is available on the Department of Ecology's website at
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Data for this project are available at Ecology's Environmental Information Management (EIM) website www.ecy.wa.gov/eim/index.htm. Search Study ID, PbTrends15.

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