PBT Trend Monitoring: Measuring Lead in Suspended Particulate Matter from Washington State Rivers and Lakes, 2011 Results

DEPARTMENT OF ECOLOGY
State of Washington

June 2011

Publication No. 12-03-031

Publication and Contact Information

This report is available on the Department of Ecology's website at
www.ecy.wa.gov/biblio/1203031.html

Data for this project are available at Ecology's Environmental Information Management (EIM) website www.ecy.wa.gov/eim/index.htm. Search User Study ID, PbTrends11.

The Activity Tracker Code for this study is 08-538.

For more information contact:

Publications Coordinator
Environmental Assessment Program
P.O. Box 47600, Olympia, WA 98504-7600
Phone: (360) 407-6764

Washington State Department of Ecology - www.ecy.wa.gov/

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

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by

Callie Mathieu and Michael Friese

Environmental Assessment Program
Washington State Department of Ecology
Olympia, Washington 98504-7710

Water Resource Inventory Area (WRIA) Numbers: See Table A-1

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Abstract

The Washington State Department of Ecology began a long-term study on lead in suspended particulate matter (SPM) as a component of Ecology's persistent, bioaccumulative, and toxic chemical (PBT) monitoring program in 2008. The goal of this study is to evaluate spatial and temporal trends in environmental levels of lead in Washington State waterbodies. This report summarizes results from the fourth year (2011) of the long-term study.

A total of 59 SPM samples from 15 river and stream monitoring sites were analyzed for lead in the spring and fall of 2011. Concentrations ranged from below detection limits to 1,780 mg/kg, with a median of 64 mg/kg. The Spokane River at the Idaho border had the highest lead concentrations, followed by the Spokane River Nine Mile site. Lead concentrations were also elevated at three urban streams and the Upper Columbia River site.

Six out of 59 samples (10%) contained lead levels above Ecology's proposed Sediment Quality Standard (SQS) of 360 mg/kg. All samples collected from the Spokane River at the Idaho border were above this level, as well as both spring samples from the Spokane River at Nine Mile.

Four new sites were added to the project plan in 2011: three small urban streams and one small rural stream. Lead concentrations measured at the urban streams ranged from 117-237 mg/kg and were higher than 70-80% of all other data collected for this program from 2008 to 2011. Mean lead levels were significantly higher at the three urbanized streams than the reference stream Huge Creek.

In a qualitative temporal analysis of lead samples collected between 2008 and 2011, very few monitoring sites displayed consistently increasing or decreasing patterns. Yakima River data showed generally decreasing levels each year. The two mid-section sites on the Columbia River – McNary Dam and Rock Island Dam – showed lead levels generally increasing each year. At this time, the small sample size prohibits a quantitative statistical temporal analysis.

Acknowledgements

The authors of this report thank the following people for their contribution to this study:

- PGE Beaver Generating Plant for access to the lower Columbia River.
- Foster Golf Links for Duwamish River site access.
- Olympic National Park for Queets River research permission.
- Personnel at the following facilities for site access: McNary Dam, Nine Mile Dam, Rock Island Dam, and Wanawish Dam (Horn Rapids Dam).
- Washington State Department of Ecology staff:
    - Andrew Albrecht, Kristin Carmack, Casey Deligeannis, Chad Furl, Brian Gallagher, Mya Keyzers, Tanya Roberts, Jim Ross, Patti Sandvik, Keith Seiders, Tighe Stuart, and Scott Tarbutton for their help with sample collection.
    - Manchester Environmental Laboratory staff for analysis of the samples and assistance with this project: Dean Momohara, Aileen Richmond, Nancy Rosenbower, Leon Weiks, Daniel Baker, Meredith Jones, Sally Cull, and others.
    - Dale Norton for guidance and review of the project plan and report drafts.
    - Art Johnson for reviewing the draft report.
    - Jean Maust and Joan LeTourneau for formatting and editing the final report.

Introduction

Lead is a naturally occurring element that is toxic and persistent and has been widely redistributed in the environment by humans. It is classified as a metal of concern under the Washington State Department of Ecology (Ecology) Persistent Bioaccumulative Toxics (PBT) Rule (WAC 173-333-110).

Sources

The use of lead as an additive in gasoline in the 20th century resulted in widespread pollution until U.S. restrictions in the 1980s and 1990s phased out use (Ecology and WDOH, 2009). In Washington State, historical sources of lead to the environment also included smelting emissions and lead-arsenate insecticide applications. Currently, major anthropogenic sources of lead to the state's environment include releases from ammunition and hunting shot, fishing weights, and vehicle wheel weights (Ecology, 2011). Other significant sources include emissions from industrial, commercial, and institutional facilities, with the highest releases from military bases, paper mills, and steel manufacturers (Ecology, 2011).

Effects

Lead is a concern because it is highly toxic to humans and wildlife. It primarily affects developing nervous systems, but can also harm cardiovascular, gastrointestinal, immune, and reproductive systems, as well as kidneys and blood (Ecology and WDOH, 2009). Lead uptake occurs through ingestion and inhalation of lead-containing materials, such as dust, after which it accumulates in bones (ATSDR, 2009). There are many sources of lead exposure to humans from the indoor and outdoor environment, and even small exposures may cause some harm (Ecology and WDOH, 2009).

Actions in Washington State

Lead was the third PBT chemical to be addressed by Washington State through a Chemical Action Plan (Ecology and WDOH, 2009). The plan was developed by Ecology and the Washington State Department of Health (WDOH) to identify the toxic effects of lead, describe its occurrence in the environment, and recommend ways to reduce its harm.

In 2009 legislation was passed to require alternatives to lead wheel weights on vehicles 14,000 pounds or less (RCW 70.270). Effective January 1, 2011, lead weights must be replaced with environmentally preferred weights whenever tires are replaced or balanced.

Ecology began a monitoring program to assess temporal changes in lead levels of rivers and lakes in Washington State in 2008. SPM was the target medium because of lead's high affinity for the particulate phase. The primary goals of the lead monitoring program were to (1) establish baseline levels of SPM-associated lead in the aquatic environment, and (2) measure spatial and temporal trends in lead over time as Chemical Action Plan reduction strategies are implemented.

This report summarizes the fourth year of the long-term monitoring effort. The first three years of lead monitoring were conducted from 2008 - 2010. Data from the first three years are reported by Meredith and Furl (2009, 2010) and Meredith and Roberts (2011).

Study Design

Sixty SPM samples were collected via in-line filtration from 15 monitoring sites throughout Washington State for analysis of total lead. Samples were taken twice in the spring and twice in the fall at each site. Spring sample collections are timed to coincide with high flow from spring snowmelt and fall samples are collected to capture low-flow conditions. A more detailed project design description can be found in the Quality Assurance (QA) Project Plan (Meredith and Furl, 2008).

The 2011 monitoring locations shown in Figure 1 consisted of 11 established sites and 4 new sites (see Appendix A for location information). Newly added sites included three small urban streams (Leach Creek, Longfellow Creek, and Thornton Creek), and one small rural stream (Huge Creek). The small urban streams were added in an attempt to monitor changes in environmental lead levels due to Chemical Action Plan reduction strategies. Huge Creek has little development or paved roads in its drainage and was added as a reference site for the small urban streams. A QA Project Plan Addendum details the changes in monitoring sites (Meredith, 2011).

Selection criteria for the established monitoring sites covered a range of land uses and lead contamination potential. Three of the sites were included in the sampling design to represent areas with potential contamination: Hylebos Creek, Spokane River at the Idaho border, and Upper Columbia River. These sites are primarily impacted by historical smelting, mining in the Coeur D'Alene basin, and an upstream lead-zinc smelter, respectively, and are undergoing clean-up actions (Ecology, 2007; Butkus and Merrill, 1999; Ecology, 2009; EPA, 2008). The Queets River watershed lies in the Olympic National Park and represents reference conditions for mid-large sized rivers in this study.

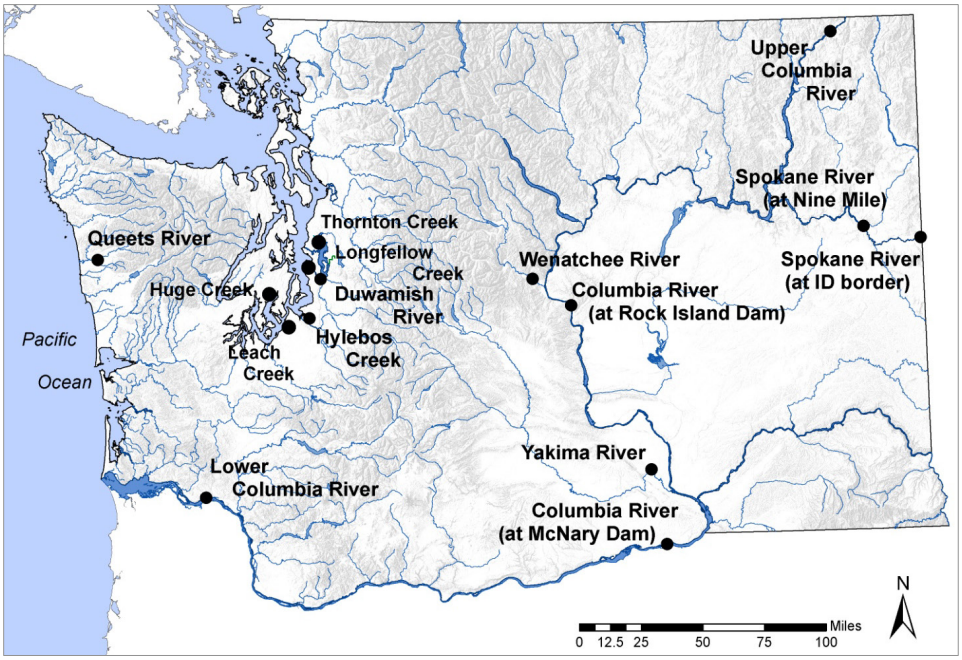

Figure 1. 2011 Lead Monitoring Site Locations.

Methods

Field Procedures

Ecology's Environmental Assessment (EA) Program staff collected SPM samples following *Standard Operating Procedure for Collecting Freshwater Suspended Particulate Matter Samples using In-line Filtration* (Meredith, 2008). River or stream water was pumped via a peristaltic pump and filtered through pre-weighed, 0.45 um pore-size (47 mm membrane) nitrocellulose filters using in-line Teflon filter holders. The intake of the tubing was placed 0.5-3 feet below the water surface, suspended above the river or lake bottom, for the duration of sampling. Water was pumped through the filtration apparatus until enough SPM had accumulated to restrict water flow. Filters were then carefully removed from the filter holder and placed in pre-acid-washed aluminum sample containers. Samples were bagged, labeled, and stored upright with blue ice for transport to Ecology headquarters.

Field blanks were sampled at rotating monitoring sites by pumping lab-prepared blank water through decontaminated tubing, filtration apparatus, and filter. Field replicates were collected following the same procedure used to collect the other SPM samples (Meredith, 2008).

The volume of water that passed through each filter was recorded after sampling. One filter was collected per sample. Samples were stored at Ecology headquarters at 4 °C until shipment to the Ecology Manchester Environmental Laboratory (MEL) for analysis. Chain of custody procedures were maintained throughout the project.

Ambient water temperature, pH, and conductivity were measured in situ at each site. pH was measured following *Standard Operating Procedures for the Collection and Analysis of pH Samples* (Ward, 2007).

Laboratory Procedures

MEL analyzed lead in SPM following EPA Method 200.8 (ICP-MS). Prior to analysis, filters were dried at 103-105 °C, weighed to determine dry SPM weight, and digested following EPA Method 3050B. Dry SPM weight was used to determine lead results in mg/kg. Results are presented in this report as lead by weight in mg/kg (parts per million) and as lead by volume in ug/L (parts per billion). Lead in ug/L was calculated by dividing the amount of lead measured on individual filters by volume of water passed through the filter.

Several lead samples were below the reporting limit, but above the method detection limit. The raw data (lead values below the reporting limit but greater than the method detection limit) were used in calculations presented in the Discussion section. Due to the level of uncertainty, only values above the reporting limit are included in the Results section.

Total suspended solids (TSS) values were estimated as the dried SPM weight on sample filters divided by filtrate volume. These are not standard TSS values due to the filter size used in the study (0.45 um pore-size). MEL's standard TSS analyses are conducted using 1.5 um pore-size filters. TSS calculations < 1 mg/L are reported as 1 U; however, for loading calculations and correlations the raw data was used.

Data Quality

MEL received all samples in good condition and performed all analyses within holding times. All instrument calibrations and internal standard recoveries were within laboratory acceptance limits. Copies of case narratives documenting instrument calibration, calibration checks, internal standards, method blanks, matrix spikes, and laboratory control samples are available upon request. All laboratory and field quality control (QC) tests are presented in Appendix B.

All laboratory analyses met measurement quality objectives (MQOs) for this project (Meredith and Furl, 2008). No analytically significant amount of lead was found in the method blanks. Laboratory control sample recoveries ranged from 98-106%. Recoveries of matrix spikes ranged from 99-104%. A laboratory error occurred where MEL spiked an incorrect sample resulting in no matrix spike for the first spring sampling period. A recovery could not be calculated due to lack of native sample material to subtract. This also resulted in no early spring sample for Hylebos Creek.

Field replicate relative percent differences (RPD) were generally good, with an average of 15%. One pair of samples, in late fall, had a high RPD (37%), but was within MQOs so no qualification was made to the data. The other pair of replicate samples taken during late fall had an RPD of 2.1%.

Four out of ten field blank samples contained lead concentrations at or above the reporting limit (0.05 ug/filter). These samples were collected at the Columbia River at McNary Dam, Upper Columbia River, Spokane River at the Idaho border, and Wenatchee River sites. The Columbia River at McNary Dam field sample had a lead result less than five times the concentration of the associated field blank. This sample was flagged "J" as an estimate. The Upper Columbia River and Spokane River sample results were greater than five times the field blank and no qualification was made. The field sample at Wenatchee River was below detection limits, so no qualification was made. See Appendix B for full field blank results.

Sampling Results

A total of 59 SPM samples were analyzed for lead in 2011. Table 1 displays a statistical summary of spring and fall lead data. Table 2 presents the individual lead and TSS results. Complete results including pH, temperature, and conductivity can be found in Appendix C.

Table 1. Statistical Summary of 2011 Spring and Fall Lead in SPM Results (mg/kg).

Season	n =	Det. Freq	Min.	Max.	Median	Mean	SD
spring	29	90%	ND	1780	53.3	225	412
fall	30	80%	ND	628	78.7	144	161

SD: standard deviation; ND: not detected above reporting limit.

Table 2. Lead and Total Suspended Solids Results, 2011.

Monitoring Site	Spring			Fall		
	Date	Pb (mg/kg)	TSS (mg/L)	Date	Pb (mg/kg)	TSS (mg/L)
Columbia River at McNary Dam	5/3/2011	64 J	6	8/31/2011	34	4
Columbia River at McNary Dam	5/31/2011	37	9	9/28/2011	50	1 U
Columbia River at Rock Island Dam	5/2/2011	43	4	8/30/2011	64	1 U
Columbia River at Rock Island Dam	6/1/2011	39	15	9/29/2011	64	1 U
Columbia River, Lower	5/4/2011	15	14	8/29/2011	16	10
Columbia River, Lower	6/1/2011	16	20	9/26/2011	17	6
Columbia River, Upper	5/4/2011	64	2	8/31/2011	93	2
Columbia River, Upper	6/1/2011	202	7	9/29/2011	168	2
Duwamish River	5/6/2011	11	9	8/29/2011	25 U	4
Duwamish River	6/3/2011	10	22	9/27/2011	28	10
Huge Creek	5/6/2011	38 U	2	8/29/2011	19 U	1
Huge Creek	6/3/2011	19 U	3	9/27/2011	21 U	2
Hylebos Creek	5/6/2011	n/a	5	8/29/2011	42	1
Hylebos Creek	6/4/2011	35	6	9/27/2011	63 U	2
Leach Creek	5/6/2011	151	2	8/29/2011	224	5
Leach Creek	6/3/2011	199	4	9/27/2011	225	5
Longfellow Creek	5/6/2011	145	2	8/29/2011	181	7
Longfellow Creek	6/3/2011	162	3	9/27/2011	149	5
Queets River	5/5/2011	17	12	9/1/2011	18	4
Queets River	6/2/2011	18	14	9/28/2011	18	30
Spokane at Idaho Border	5/3/2011	1010	1 U	8/30/2011	628	1 U
Spokane at Idaho Border	5/31/2011	1780	3	9/28/2011	558	1
Spokane at Nine Mile Dam	5/3/2011	662	1	8/30/2011	181	1
Spokane at Nine Mile Dam	5/31/2011	844	5	9/28/2011	243	1 U
Thornton Creek	5/6/2011	117	4	8/29/2011	237	13
Thornton Creek	6/3/2011	176	8	9/27/2011	185	10
Wenatchee River	5/2/2011	20 U	4	8/30/2011	22 U	2
Wenatchee River	6/1/2011	7.6	10	9/29/2011	31 U	2
Yakima River	5/3/2011	12	9	8/31/2011	13	7
Yakima River	5/31/2011	11	81	9/28/2011	18	4

n/a: sample not analyzed; U: not detected above reported limit; J: estimated value

Discussion

Ranking and Comparison to Guidelines

The 2011 monitoring sites are ranked in order of annual average lead concentration in Figure 2. The Spokane River at the Idaho border had the highest lead concentrations, followed by the Spokane River at Nine Mile. Lead concentrations were also elevated at the three urban streams and the Upper Columbia River. Out of all 2011 samples, 47% (28 of 59) had lead levels higher than 50 mg/kg. The remaining sites had low (<50 mg/kg) lead levels.

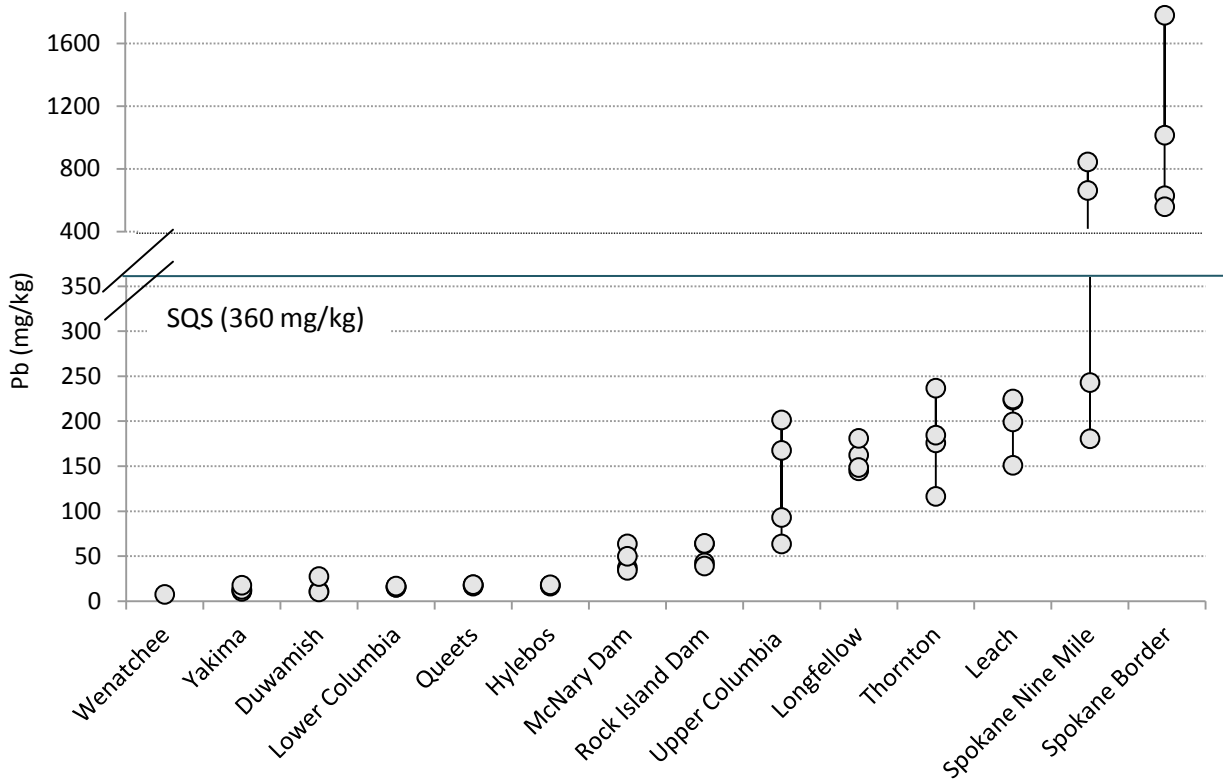

Figure 2. 2011 Monitoring Sites Ranked by Annual Average Lead Concentration (mg/kg).

No lead detections were made at Huge Creek. Note the broken y axis.
LAET = Lowest Apparent Effects Threshold.

Six samples (10%) contained lead concentrations above Ecology's proposed Sediment Quality Standard (SQS) of 360 mg/kg (Figure 2). All samples collected from the Spokane River at the Idaho border were above this level, as well as both spring samples from the Spokane River at Nine Mile. This value is based on new freshwater sediment quality values for Washington State (Michelsen, 2011) and replaces the guideline value used in previous reports (LAET, 335 mg/kg). Washington State does not currently have regulatory criteria for lead in freshwater sediments or SPM.

Seasonal Variation

Seasonal variation in the 2011 samples was assessed by comparing mean spring and fall lead values using Wilcoxon Signed Ranks tests. A Wilcoxon Signed Ranks test is a non-parametric paired samples test used when data is not normally distributed. Lead results (mg/kg) from the monitoring sites were compared seasonally and no significant seasonal difference was found ($p = 0.211$) [1].

The two Spokane River sites, Longfellow, Leach, and Thornton Creeks and the Upper Columbia River were separated from the sites with lower lead levels for further statistical analysis. When the sites with higher lead levels were evaluated for seasonal differences, no significant difference was detected ($p = 0.753$). Sites with lower lead levels were found to have a significant seasonal difference ($p = 0.038$), with fall samples having higher levels than spring (mean difference = 7.2 mg/kg). Lead levels are compared by season in Figure 3.

[1] A difference would be considered statistically significant if the p-value was less than 0.05.

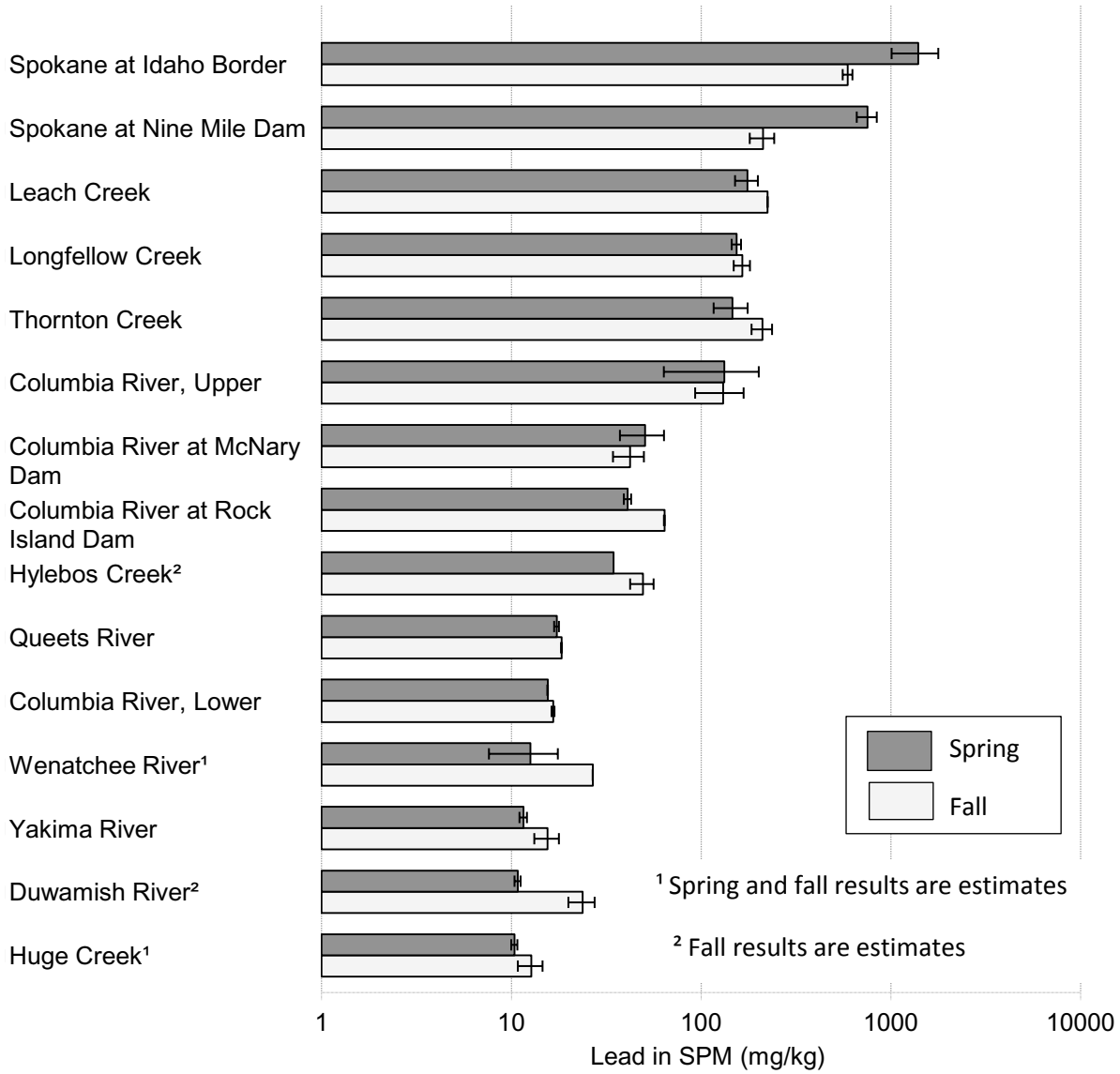

Figure 3. Lead Concentrations Measured in the Spring and Fall at the Monitoring Sites during 2008-2011.

Error bars represent one standard error.

Correlations

Non-parametric Spearman Rank correlations were calculated for each monitoring site using data from all sampling years (2008-2011) to evaluate relationships between lead levels, flow, and TSS. Lead concentrations by weight (mg/kg) were used, along with lead concentrations by volume (ug/L). Appendix D displays the correlation coefficients for each site.

Correlations between lead concentrations by weight (mg/kg) and flow and TSS were significant at six of the monitoring sites. Lead levels decreased as flow and TSS increased at all but Spokane Nine Mile and Thornton Creek (TSS only). As in past sampling years, a dilution effect may be an explanatory factor in the rivers without contaminated banks and streambeds. Higher TSS levels and larger particle sizes present in heavier flows contain lesser surface areas for metals binding (Benoit and Rozan, 1998; Dawson and Macklin, 1998).

Elevated lead concentrations at high flows in the Spokane River likely result from contaminated sediments being washed from the banks or streambed sediments being redistributed in the river at high flows. The small urban streams displayed higher concentrations of lead by weight (mg/kg) and by volume (ug/L) at higher TSS levels, but not flows. The three urban streams were the only sites showing inverse flow/TSS relationships.

Correlations between flow and TSS were significant (with the exception of Upper Columbia River) and positive at all sites except for the urban streams. Similarly, lead by volume correlated positively with TSS at all sites and positively with flow at all sites except for the urban streams.

Loading

Daily particulate lead loads were estimated for each sampling site (except Hylebos Creek) using daily mean flow (cfs) and lead concentrations by volume (ug/L). Figure 4 displays the mean load for spring and fall in increasing order.

Particulate lead loads were the highest in the Columbia River, ranging from 10-386 kg/day, followed by the Spokane River (1.7-193 kg/day). The small streams contained the lowest loads (0.001 – 0.018 kg/day), with the rural reference stream, Huge Creek, ranking the lowest.

Spring loads were higher than fall at all sites except for the three urban streams and Queets River. In the Columbia River, spring loads were around 90% higher in the spring. Spring loads at the Spokane River sites were 99% higher than fall loads. Sampling events captured high flow spring conditions and low flow fall conditions at all sites except for the small streams and Queets River, on the west side of the state.

The hydrological differences in the small streams likely explain the higher loading in fall compared to spring at those sites. High flows in the small streams are driven by rain events in the fall and winter instead of by snowmelt, like the other monitoring sites. Sampling at the small streams did not capture any storm events and little variation occurred in flow on the sampling date between seasons. Sampling at Queets River appeared to capture baseline flow levels during

spring and fall with little variation, except for a spike in the hydrograph in late fall corresponding to a rainfall event.

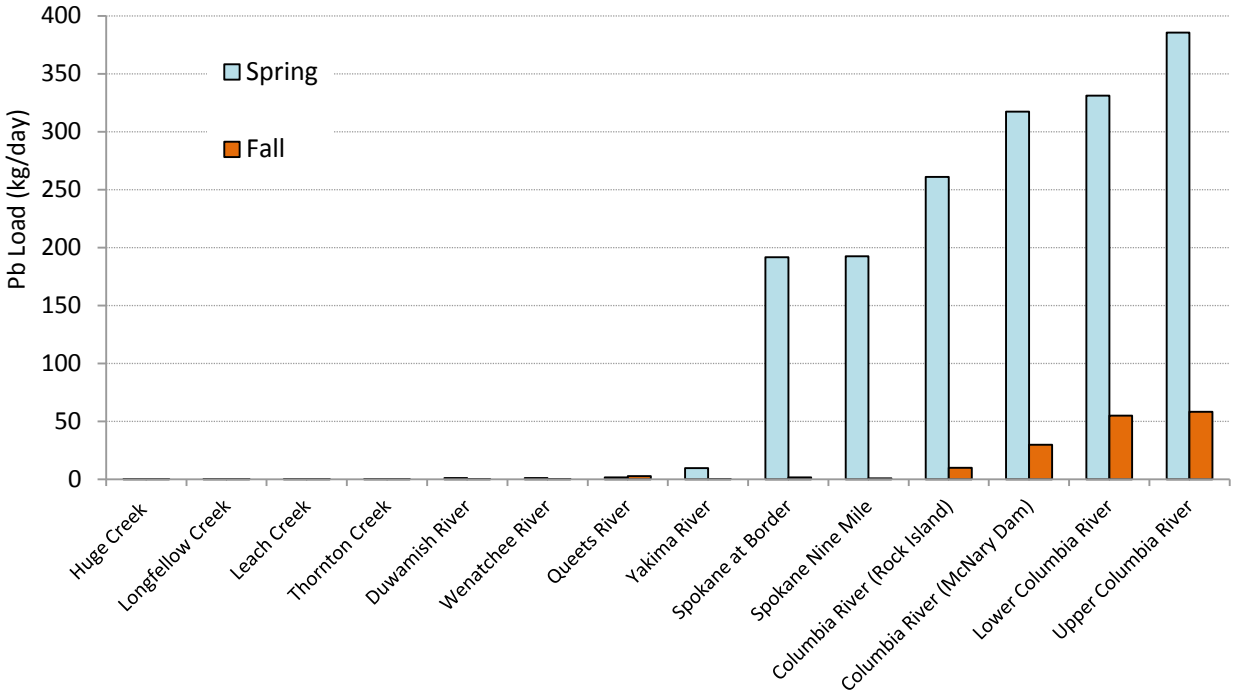

Figure 4. Mean Estimated Particulate-bound Lead Loadings (kg/day) for Spring and Fall Sampling Events, 2011.

Urban Streams

Four new sites were added to the sampling plan in 2011 to evaluate lead levels in small, urban watersheds. Leach, Longfellow, and Thornton Creeks were selected because of the high percentage of impervious surfaces and heavy traffic volumes in their watersheds. Huge Creek was chosen as a reference site for its rural and relatively undeveloped watershed. Here, baseline lead values are reported for the newly added sites, along with a comparison of levels between the sites.

Lead concentrations measured at the urban stream sites were consistently higher than most other monitoring sites, with the exception of the Spokane River. Spring concentrations ranged from 117-199 mg/kg and fall concentrations ranged from 149-237 mg/kg. Lead values from the urban sites were higher than 70-85% of all other lead data collected for this project between 2008 and 2011 (Figure 5). In contrast, data from the rural stream Huge Creek fell between 3 and 15% of the dataset values.

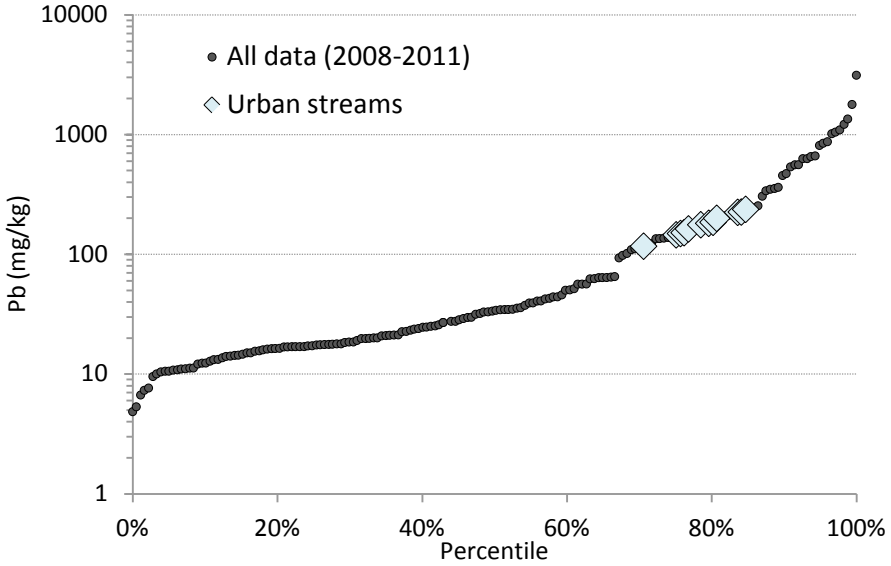

Figure 5. Frequency Distribution of All Lead Data Collected during 2008-2011.
Light blue diamonds represent urban stream samples.

A one-way ANOVA was conducted to test for differences in lead means among the urban and rural streams, with a Bonferroni post-hoc test for multiple comparisons. All distributions were normal (Shapiro Wilk > 0.05) and the homogeneity of variances assumption was met (Levene's Statistic = 1.87; $p = 0.172$).

Mean lead concentrations were significantly higher at the three urbanized streams than the reference stream Huge Creek (see Appendix E for statistical results). There was no significant difference between the three urban streams. Figure 6 shows the mean lead concentrations measured at the sites during 2011 sampling events. The four sites were similar in drainage area (4-6 square miles) and flow (2-12 cfs on sampling dates).

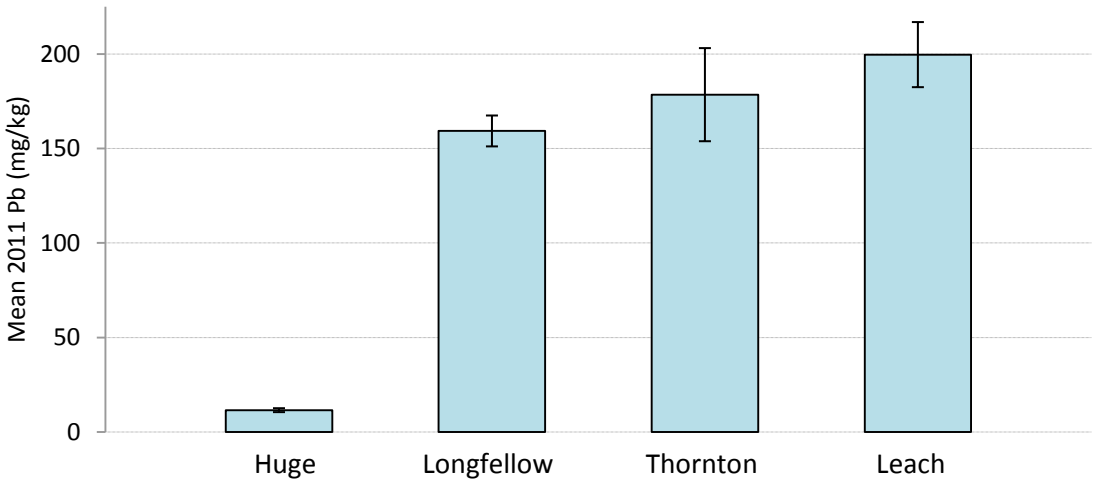

Figure 6. Mean 2011 Lead Concentrations (mg/kg) for Small Urban and Rural Streams.

n = 4; Error bars represent one standard error.

The higher lead concentrations observed at the urban streams are not surprising. Elevated lead levels in streams draining urban landscapes have been well-documented (Herrera, 2011; Tiefenthaler et al., 2008). There are many sources of lead in urban environments, including contaminated road dust from the wearing of vehicle parts, such as wheel bearings, tires, and brake pads (Carter, 2006; Ewen et al., 2009), as well as lead in runoff from house paints, traffic paints, and roofing materials (Ecology, 2011). Air emissions from nearby industry can also contribute to particulate lead levels in urban streams when contaminated dust is washed into stream systems during rain events. Lead has been found to associate with SPM more strongly in urban catchments than in rural, which may be attributed to a greater abundance of particulate forms of lead in urban streams (Bibby and Webster-Brown, 2005).

Temporal Analysis

As of 2011, four years of data have been collected at eleven of the monitoring sites. While the sample size is too small to detect statistical temporal trends at this point, the following sections qualitatively describe patterns in lead levels observed at the monitoring sites between 2008 and 2011. Non-detected data were excluded from the analysis below.

Spokane River

No monotonic (consistently decreasing or increasing) patterns emerged at either of the Spokane River sites. Maximum lead concentrations for both sites occurred in the spring of 2008 and 2011. These samples coincided with the highest flows recorded during the sampling period. Fall concentrations have remained fairly stable, ranging from 536-1,045 mg/kg and 135-362 mg/kg for the border and Ninemile sites, respectively. A separate Ecology study monitoring ambient total and dissolved lead concentrations in water samples from the Spokane River found that metals in the Spokane River have significantly decreased since 1994 (Hallock, 2010). Figure 7 shows lead concentrations of samples collected from 2008-2011.

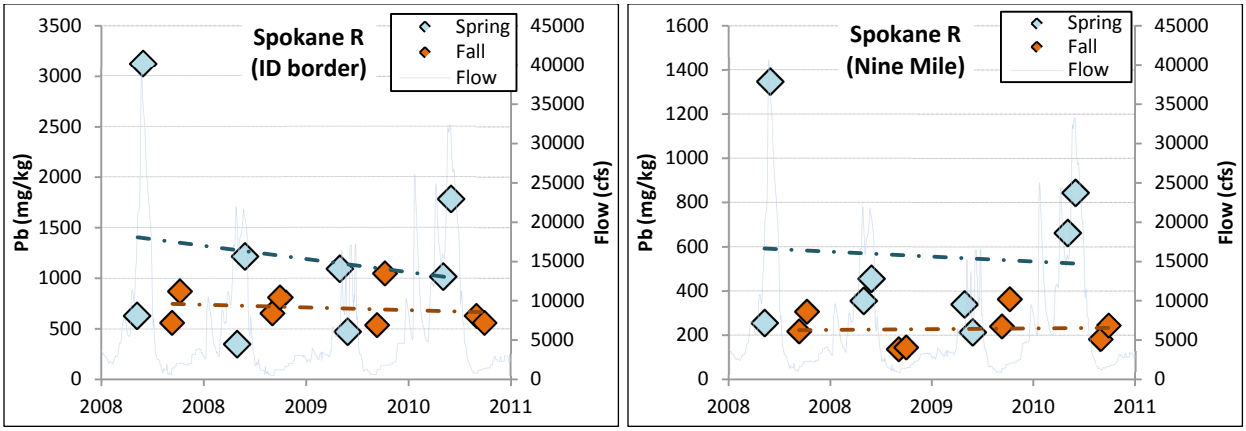

Figure 7. SPM-associated Lead Concentrations (mg/kg) at Spokane River Sites during the 2008-2011 Sampling Period.

Broken lines represent trendlines for spring (dark blue) and fall (orange) samples. Trendlines not statistically significant. Flow data is plotted along the secondary y-axis.

Columbia River

Lead concentrations of samples collected from the Columbia River showed mixed results. No pattern is apparent at the lower and upper stretches. The two mid-section sites – McNary Dam and Rock Island Dam – were the only sites with lead concentrations generally increasing each year. Lead concentrations were inversely correlated with flow at the Rock Island Dam site, driven by the higher levels during low flow fall periods. Figure 8 displays lead concentrations plotted against time and flow at the four Columbia River sites.

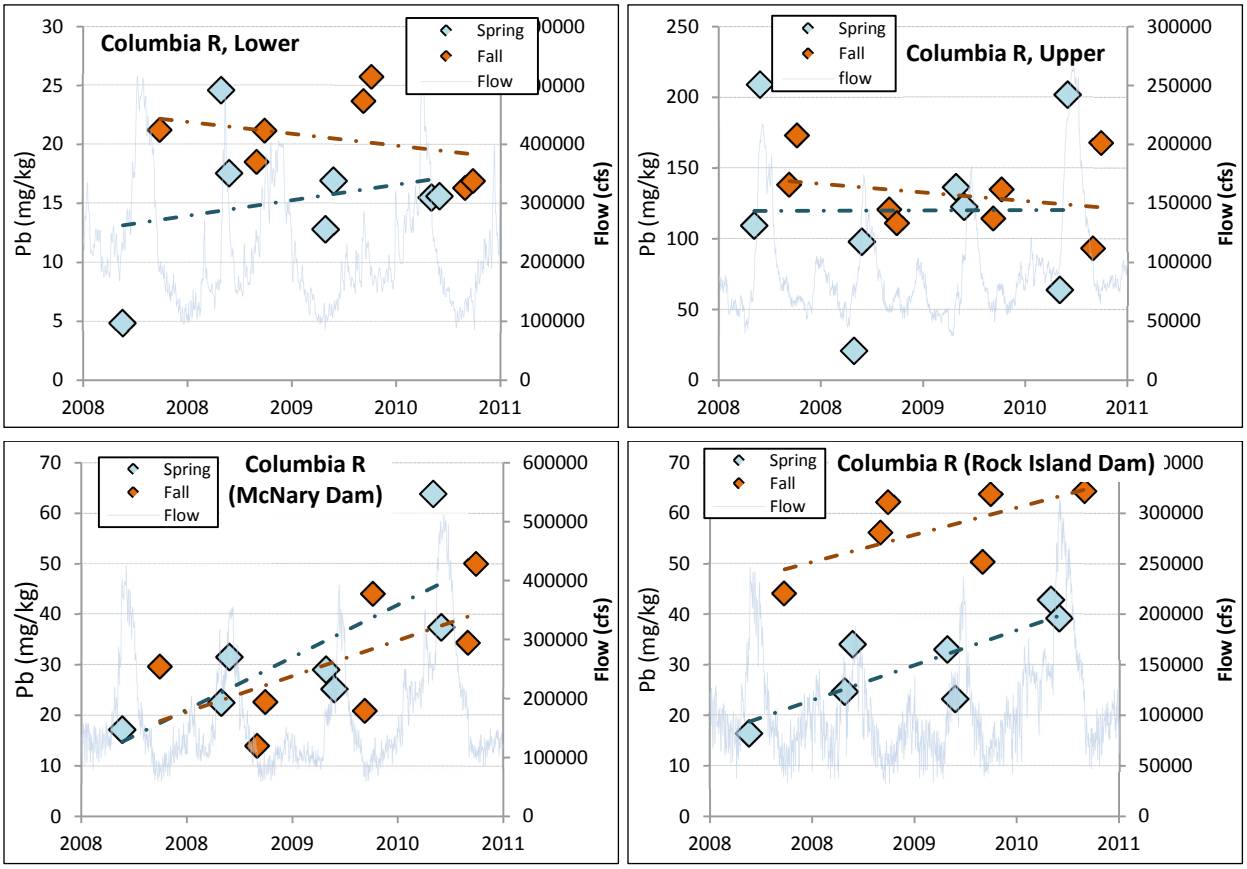

Figure 8. SPM-associated Lead Concentrations (mg/kg) at Columbia River Sites during the 2008-2011 Sampling Period.

Broken lines represent trendlines for spring (dark blue) and fall (orange) samples. Trendlines not statistically significant. Flow data is plotted along the secondary y-axis.

Mid-size Rivers

The four other rivers with lead data available for the years 2008-2011 were Duwamish, Queets, Wenatchee, and Yakima Rivers (Figure 9). Lead concentrations measured at these sites have generally been very low, reflecting baseline levels. Spring lead levels show a very stable pattern at Duwamish, Queets, and Wenatchee Rivers sites, whereas fall levels were more erratic without an apparent pattern. Yakima River data show generally decreasing levels each year. Lead levels and flow were not correlated at the Yakima River site.

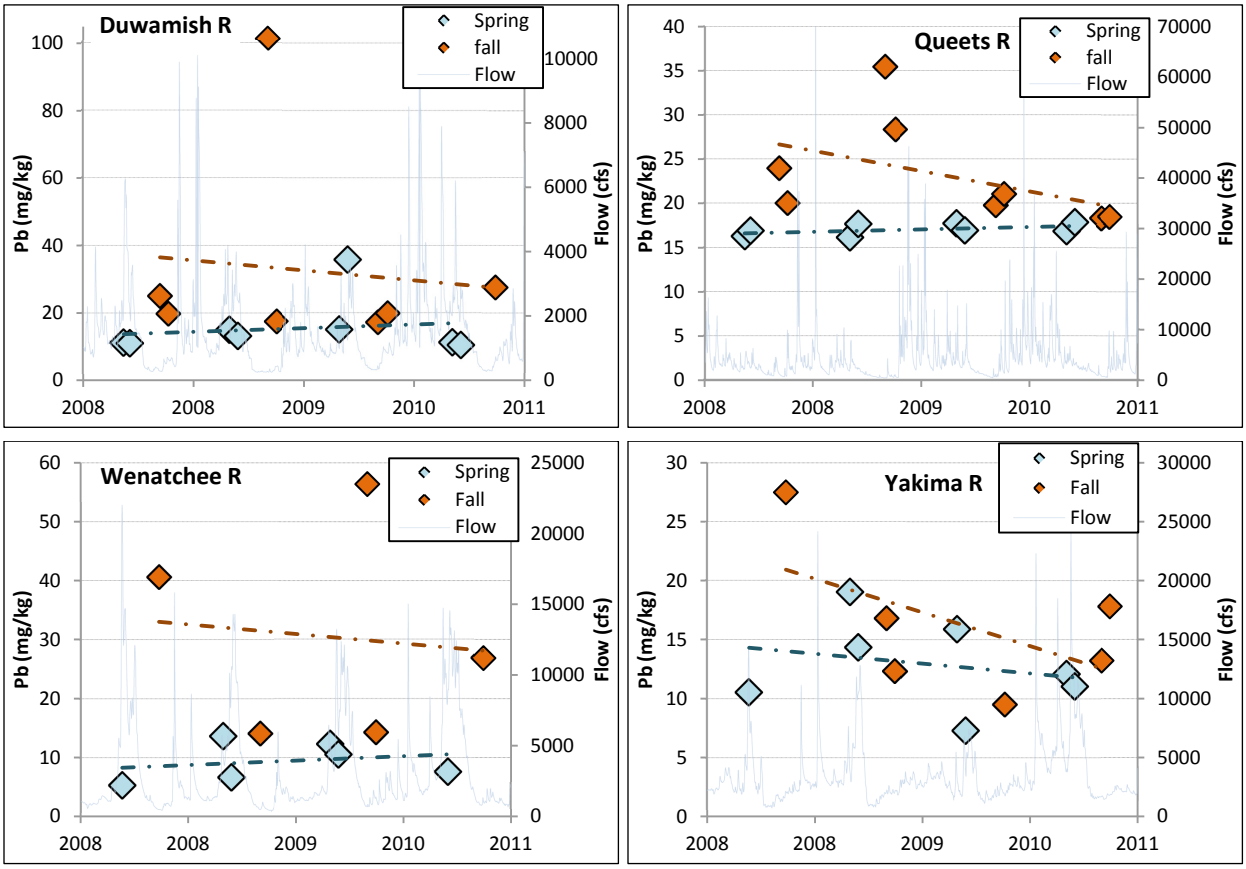

Figure 9. SPM-associated Lead Concentrations (mg/kg) at Mid-size River Sites during the 2008-2011 Sampling Period.

Broken lines represent trendlines for spring (dark blue) and fall (orange) samples. Trendlines not statistically significant. Flow data is plotted along the secondary y-axis.

Hylebos Creek

Lead concentrations measured at Hylebos Creek have varied year to year. Maximum concentrations for spring samples occurred in 2008 and have remained lower since then. Fall levels showed a similar pattern to the Spokane sites, with peak concentrations in 2008 and 2011, but no monotonic pattern. No flow data was obtained for Hylebos Creek, and therefore no correlations can be made between lead and flow. However, lead and TSS were not correlated at this site.

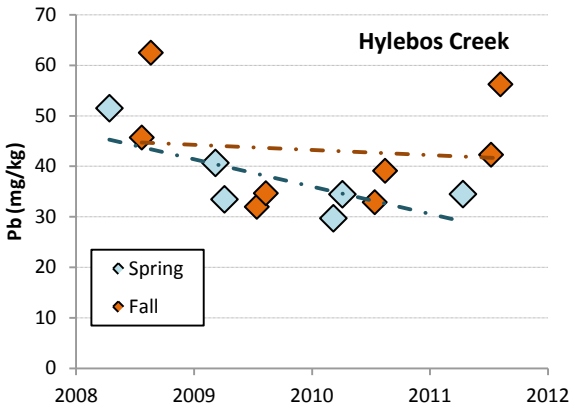

Figure 10. SPM-associated Lead Concentrations (mg/kg) at Hylebos Creek during the 2008-2011 Sampling Period.

Broken lines represent trendlines for spring (dark blue) and fall (orange) samples. Trendlines not statistically significant.

Conclusions

This report summarizes results from the fourth year of a long-term study to evaluate spatial and temporal trends in environmental levels of lead in Washington State waterbodies. A total of 59 SPM samples from 15 river and stream sites were analyzed for lead in 2011. Results of this study support the following conclusions:

- Samples from the Spokane River at the Idaho border contained the highest lead concentrations, followed by the Spokane River at Nine Mile. Lead concentrations were also elevated at the three urban streams and the Upper Columbia River.

- Six out of 59 samples (10%) contained lead levels above Ecology's proposed Sediment Quality Standard (SQS) of 360 mg/kg. All samples collected from the Spokane River at the Idaho border were above this level, as were both spring samples from the Spokane River at Nine Mile. The SQS is not a state criterion; currently no regulatory criteria exist for freshwater SPM.

- Estimated particulate lead loads were the highest in the Columbia River, ranging from 10-386 kg/day, followed by the Spokane River (1.7-193 kg/day). The small streams contained the lowest loads (0.001-0.018 kg/day), with the rural reference stream, Huge Creek, ranking the lowest.

- Lead concentrations measured at the urban stream sites were consistently higher than the other monitoring sites, with the exception of the Spokane River. Concentrations ranged from 117-237 mg/kg. Mean lead concentrations were significantly higher at the three urbanized streams than the reference stream of similar size, Huge Creek.

- In a qualitative temporal analysis of lead samples measured over 2008-2011, few sites showed consistently increasing or decreasing patterns. Yakima River data had generally decreasing levels each year. The two mid-section sites on the Columbia River – McNary Dam and Rock Island Dam – showed a generally increasing pattern.

Recommendations

Results of this (2011) study support the following recommendations:

- Continue lead sampling at the 15 monitoring sites in 2012. Statistical trends tests may be possible in the 2012 report, with five years of data collected. The long-term monitoring project plan and objectives should be reassessed in the 2012 report.
- Consider collecting more material at sites where lead was not detected above reporting limits. The lab may be able to analyze two filters as one sample, which would lower reporting limits.
- Continue to investigate sources of field blank contamination. Consider the use of a portable field screen for a "clean" environment to sample in, especially during windy conditions. Field audits with sampling staff should also be performed regularly.

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Appendices

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Appendix A. Monitoring Site Location Descriptions

Table A-1. Monitoring Site Descriptions, 2011.

Monitoring Site	County	WBID[1]	WRIA[2] Number	Latitude[3]	Longitude[3]	Description
Columbia River, Lower	Wahkiakum	WA-CR-1010	25	46.18490	-123.18760	Columbia River near Clatskanie, OR, RM 54.
Columbia River at McNary Dam	Benton	WA-CR-1026	31	45.93940	-119.29720	Columbia River at McNary Dam near Umatilla, OR, RM 292.0.
Columbia River at Rock Island Dam	Chelan-Douglas	WA-CR-1040	44	47.34390	-120.09390	Columbia River at Rock Island Dam, RM 453.5, 10 miles south of Wenatchee.
Columbia River, Upper	Stevens	WA-CR-1060	61	48.92161	-117.77445	Upper Columbia River at Northport, RM 735.
Duwamish River	King	WA-09-1010	9	47.48525	-122.26140	Duwamish River at Foster Golf Links in Tukwila, RM 10.
Hylebos Creek	Pierce	WA-10-1011	10	47.25335	-122.35013	Hylebos Creek in Fife, at 4th St. bridge.
Huge Creek	Pierce	n/a	15	47.38899	-122.69887	Huge Creek near Wauna, WA.
Leach Creek	Pierce	n/a	12	47.22092	-122.50925	Leach Creek at the Emerson St. crossing.
Queets River	Jefferson	WA-21-1030	21	47.55220	-124.19780	Queets River in Olympic National Forest, 2 miles up Queets River Rd, RM 11.5.
Longfellow Creek	King	WA-09-1000	9	47.55370	-122.36662	Longfellow Creek in West Seattle, at Brandon St.
Spokane River at ID Border	Spokane	WA-57-1010	57	47.69483	-117.05133	Spokane River near the Idaho border, RM 96.
Spokane River at Nine Mile	Spokane	WA-54-1020	54	47.77470	-117.54440	Upstream side of Spokane River's Nine Mile Dam, RM 58.1.
Thornton Creek	King	WA-08-1020	8	47.69477	-122.27250	Thornton Creek near mouth at Matthews Beach Park.
Wenatchee River	Chelan	WA-45-1010	45	47.50070	-120.42570	Wenatchee River, about 5 miles NW of Wenatchee, RM 7.1, near Old Monitor Rd. Bridge.
Yakima River	Benton	WA-37-1010	37	46.37830	-119.41810	Yakima River, 12 mi NW of Richland, RM 18.0. Diversion structure at Wanawish Dam.

[1]WBID - Waterbody Identification Number
[2]WRIA - Water Resource Inventory Area
[3]NAD83 HARN
RM - River Mile

Appendix B. Quality Assurance Data

Table B-1. Measurement Quality Objectives.

Analysis	Check Standards	Matrix Spikes (%recovery)	Field Replicates (RPD)	Lowest Concentration of Interest
Lead	± 15% LCS	70-130%	>50%	1 mg/Kg dw

dw - dry weight

Table B-2. Laboratory Blanks.

Sample Number	Analysis Date	Result (ug)
B11E120-BLK1	5/19/2011	0.05 U
B11F099-BLK1	6/16/2011	0.05 U
B11I149-BLK1	9/23/2011	0.05 U
B11J020-BLK1	10/7/2011	0.05 U
B11J027-BLK1	10/7/2011	0.05 U

Table B-3. Laboratory Control Samples.

Sample Number	Analysis Date	Recovery (%)
B11E120-BS1	5/19/2011	102
B11F099-BS1	6/16/2011	106
B11I149-BS1	9/23/2011	98
B11J020-BS1	10/7/2011	104
B11J027-BS1	10/7/2011	103
B11J027-BSD1	10/7/2011	106

Table B-4. Laboratory Matrix Spikes.

Sample Number	Analysis Date	Recovery (%)
1106034-19	6/16/2011	104
1109062-12	9/23/2011	99
1110028-03	10/7/2011	104

Table B-5. Field Blanks.

Sample Number	Collection Date	Site	Result (ug/filter)
1105048-06	5/3/2011	Columbia River (McNary)	**0.46**
1105048-16	5/6/2011	Longfellow Creek	0.05 U
1106034-04	6/1/2011	Upper Columbia River	**0.11**
1106034-08	6/1/2011	Lower Columbia River	0.05 U
1106034-10	6/1/2011	Columbia River (Rock Island)	0.05 U
1109062-03	8/30/2011	Spokane River at ID Border	**0.05**
1109062-16	8/30/2011	Wenatchee River	**0.10**
1110028-09	9/27/2011	Longfellow Creek	0.05 U
1110028-15	9/28/2011	Yakima River	0.05 U
1110028-20	9/28/2011	Queets River	0.05 U

U: Not detected at or above the reported result.

Table B-6. Field Replicates.

Sample Number	Collection Date	Result (mg/Kg)	RPD (%)
1105048-08	5/2/2011	20.0 U	n/a
1105048-09	5/2/2011	17.9 U	
1106034-17	6/3/2011	162	18
1106034-18	6/3/2011	196	
1109062-10	8/29/2011	224	4.6
1109062-11	8/29/2011	214	
1110028-01	9/28/2011	243	37
1110028-02	9/28/2011	168	
1110028-19	9/28/2011	18.5	2.1
1110028-25	9/28/2011	18.9	

Appendix C. 2011 Results

Table C-1. Spring Data.

Monitoring Site	Sample #	Date	Pb (mg/kg)	Pb (ug/L)	Temp. (°C)	pH	Cond. (us/cm)	TSS (mg/L)
Columbia River at McNary Dam	1105048-05	5/3/2011	63.8 J	0.382 J	12.0	8.5	161	6
Columbia River at McNary Dam	1106034-06	5/31/2011	37.4	0.345	18.2	7.9	124	9
Columbia River at Rock Island Dam	1105048-07	5/2/2011	42.8	0.179	10.3	7.4	82	4
Columbia River at Rock Island Dam	1106034-09	6/1/2011	39.2	0.591	14.5	8.1	194	15
Columbia River, Lower	1105048-18	5/4/2011	15.5	0.217	10.3	7.9	140	14
Columbia River, Lower	1106034-07	6/1/2011	15.6	0.319	12.6	7.7	65	20
Columbia River, Upper	1105048-03	5/4/2011	63.7	0.123	7.4	8.0	139	2
Columbia River, Upper	1106034-03	6/1/2011	202	1.365	9.8	7.9	131	7
Duwamish River	1105048-14	5/6/2011	11.2	0.097	11.0	7.4	99	9
Duwamish River	1106034-16	6/3/2011	10.4	0.224	10.8	7.6	63	22
Huge Creek	1105048-12	5/6/2011	38.5 U	0.064 U	10.0	7.6	69	2
Huge Creek	1106034-14	6/3/2011	19.0 U	0.053 U	11.0	7.5	71	3
Hylebos Creek	1106034-15	6/3/2011	34.5	0.195	12.2	7.5	228	6
Leach Creek	1105048-11	5/6/2011	151	0.378	10.8	7.4	305	2
Leach Creek	1106034-13	6/3/2011	199	0.883	12.4	7.4	316	4
Longfellow Creek	1105048-15	5/6/2011	145	0.341	11.4	8.0	294	2
Longfellow Creek	1106034-17	6/3/2011	162	0.429	13.6	7.9	262	3
Queets River	1105048-19	5/5/2011	16.8	0.206	8.3	7.0	21	12
Queets River	1106034-12	6/2/2011	17.8	0.258	9.4	7.6	11	14
Spokane River at Idaho Border	1105048-02	5/3/2011	1010	0.951	5.9	7.7	46	1 U
Spokane River at Idaho Border	1106034-02	5/31/2011	1780	4.778	10.7	7.5	40.9	3
Spokane River at Nine Mile Dam	1105048-01	5/3/2011	662	0.772	6.6	7.9	68	1
Spokane River at Nine Mile Dam	1106034-01	5/31/2011	844	4.576	11.0	7.5	55.4	5
Thornton Creek	1105048-17	5/6/2011	117	0.445	11.2	7.9	226	4
Thornton Creek	1106034-20	6/3/2011	176	1.385	14.7	8.0	198	8
Wenatchee River	1105048-08	5/2/2011	20.0 U	0.071 U	8.3	8.5	64	4
Wenatchee River	1106034-11	6/1/2011	7.63	0.077	8.1	8.2	95	10
Yakima River	1105048-04	5/3/2011	12.1	0.108	12.6	8.5	172	9
Yakima River	1106034-05	5/31/2011	11.0	0.898	16.2	8.0	131	81

Table C-2. Fall Data.

Monitoring Site	Sample #	Date	Pb (mg/kg)	Pb (ug/L)	Temp. (°C)	pH	Cond. (us/cm)	TSS (mg/L)
Columbia River at McNary Dam	1109062-18	8/31/2011	34.4	0.126	21.6	7.7	126	4
Columbia River at McNary Dam	1110028-16	9/28/2011	50.0	0.048	20.4	7.9	196	1 U
Columbia River at Rock Island Dam	1109062-14	8/30/2011	63.8	0.045	21.3	7.6	203	1 U
Columbia River at Rock Island Dam	1110028-17	9/29/2011	64.4	0.042	18.2	6.8	372	1 U
Columbia River, Lower	1109062-05	8/29/2011	16.3	0.157	21.4	n/a	133	10
Columbia River, Lower	1110028-13	9/26/2011	16.9	0.110	19.2	6.8	141	6
Columbia River, Upper	1109062-04	8/31/2011	93.1	0.216	18.6	8.2	118	2
Columbia River, Upper	1110028-05	9/29/2011	168	0.350	14.85	8.0	129	2
Duwamish River	1109062-07	8/29/2011	25.0 U	0.106 U	17.8	7.3	170	4
Duwamish River	1110028-07	9/27/2011	27.5	0.265	14.6	7.2	92	10
Huge Creek	1109062-13	8/29/2011	19.2 U	0.022 U	13.1	7.6	90	1
Huge Creek	1110028-12	9/27/2011	20.8 U	0.033 U	11.9	7.8	83	2
Hylebos Creek	1109062-06	8/29/2011	42.3	0.059	14.1	8.0	300	1
Hylebos Creek	1110028-06	9/27/2011	62.5 U	0.152 U	13.4	7.7	327	2
Leach Creek	1109062-10	8/29/2011	224	1.226	14.7	7.8	376	5
Leach Creek	1110028-11	9/27/2011	225	1.160	14.3	6.9	27	5
Longfellow Creek	1109062-08	8/29/2011	181	1.288	15.4	7.9	297	7
Longfellow Creek	1110028-08	9/27/2011	149	0.756	15.2	7.4	174	5
Queets River	1109062-19	9/1/2011	18.3	0.079	14.0	7.0	65	4
Queets River	1110028-19	9/28/2011	18.5	0.561	9.9	7.5	49	30
Spokane River at Idaho Border	1109062-02	8/30/2011	628	0.504	22.8	7.5	47.7	1 U
Spokane River at Idaho Border	1110028-04	9/28/2011	558	0.797	18.0	7.5	45.4	1
Spokane River at Nine Mile Dam	1109062-01	8/30/2011	181	0.221	15.7	8.0	261	1
Spokane River at Nine Mile Dam	1110028-01	9/28/2011	243	0.233	13.0	7.9	219	1 U
Thornton Creek	1109062-09	8/29/2011	237	2.961	16.3	8.1	235	13
Thornton Creek	1110028-10	9/27/2011	185	1.920	15.2	7.7	177	10
Wenatchee River	1109062-15	8/30/2011	21.7 U	0.045 U	16.4	7.6	35	2
Wenatchee River	1110028-18	9/29/2011	31.3 U	0.054 U	12.0	7.4	49	2
Yakima River	1109062-17	8/31/2011	13.2	0.090	20.1	8.1	239	7
Yakima River	1110028-14	9/28/2011	17.8	0.063	17.9	6.9	258	4

Appendix D. Correlation Coefficients

Table D-1. Spearman Rank Correlation Coefficients for Relationships between Lead by Weight (mg/kg), Lead by Volume (ug/L), TSS, and Flow.

Waterbody	n	Pb (mg/kg) correlated with:			Pb (ug/L) correlated with:	
		Pb (ug/L)	TSS (mg/L)	Flow (cfs)	TSS (mg/L)	Flow (cfs)
Columbia River at McNary Dam	14	0.31	-0.13	0.20	**0.85**	**0.73**
Columbia River at Rock Island Dam	14	**-0.71**	**-0.84**	**-0.67**	**0.97**	**0.78**
Columbia River, Lower	14	-0.26	**-0.57**	**-0.61**	**0.90**	**0.79**
Columbia River, Upper	16	0.36	-0.04	0.16	**0.87**	**0.54**
Duwamish River	15	-0.31	**-0.81**	**-0.79**	**0.75**	0.24
Huge Creek	1	---	---	---	---	---
Hylebos Creek	15	0.25	-0.08	---	**0.92**	**---**
Leach Creek	4	0.80	0.80	-0.40	**1.00**	-0.80
Longfellow Creek	4	0.80	0.80	-0.80	**1.00**	**-1.00**
Queets River	16	**-0.57**	**-0.62**	**-0.59**	**0.98**	**0.76**
Spokane River at Idaho Border	16	0.34	-0.02	0.46	**0.89**	**0.94**
Spokane River at Nine Mile Dam	16	**0.79**	0.39	**0.83**	**0.83**	**0.92**
Thornton Creek	4	**1.00**	**1.00**	**-1.00**	**1.00**	**-1.00**
Wenatchee River	12	-0.31	-0.54	-0.45	**0.91**	**0.87**
Yakima River	13	-0.11	-0.28	-0.24	**0.95**	**0.80**

Bolded, shaded values are significant at the 95% confidence level.
A Spearman Rank Correlation Coefficient indicates the strength and direction of a relationship between two variables. Values closer to 1 or -1 indicate the two are strongly correlated.

Appendix E. Statistical Results

Table E-1. Results of ANOVA with Bonferroni post-hoc test.

	Sum of Squares	df	Mean Square	F	p
Between Groups	87557	3	29186	30.0	0.00001
Within Groups	11660	12	972		
Total	99217	15			

(a) site	(b) site	Mean Difference (a-b)	Standard Error	p	95% Confidence Interval	
					Lower Bound	Upper Bound
Huge	Longfellow	-147.8	22.04	0.0001	-217.25	-78.27
Huge	Thornton	-167.0	22.04	0.00004	-236.45	-97.47
Huge	Leach	-188.2	22.04	0.00001	-257.64	-118.66
Longfellow	Thornton	-19.2	22.04	1.000	-88.69	50.29
Longfellow	Leach	-40.4	22.04	0.551	-109.88	29.10
Thornton	Leach	-21.2	22.04	1.000	-90.68	48.30

df: degrees of freedom
F: F-ratio
p: p-value

Appendix F. Glossary, Acronyms, and Abbreviations

Glossary

Ambient: Background (environmental). Away from point sources of contamination.

Anthropogenic: Human-caused.

Conductivity: A measure of water's ability to conduct an electrical current. Conductivity is related to the concentration and charge of dissolved ions in water.

Hydrograph: A graph showing the discharge of a river over a period of time.

Monotonic: Consistently increasing or consistently decreasing.

Persistent, bioaccumulative, toxic substance (PBT): A distinct group of chemicals that threaten the health of people and the environment. They (1) remain in the environment for a long time without breaking down (persist), (2) are accumulated by animals and humans and increase in concentration up the food chain (bioaccumulate), and (3) are linked to toxic effects in fish, wildlife, and humans (toxic).

pH: A measure of the acidity or alkalinity of water. A low pH value (0 to 7) indicates that an acidic condition is present, while a high pH (7 to 14) indicates a basic or alkaline condition. A pH of 7 is considered to be neutral. Since the pH scale is logarithmic, a water sample with a pH of 8 is ten times more basic than one with a pH of 7.

Suspended particulate matter (SPM): Solids suspended in freshwater captured by filtration through a 0.45 um filter membrane.

Total suspended solids (TSS): The suspended particulate matter in a water sample as retained by a filter.

Acronyms and Abbreviations

Ecology	Washington State Department of Ecology
EPA	U.S. Environmental Protection Agency
MEL	Manchester Environmental Laboratory
MQO	Measurement quality objective
PBT	Persistent, bioaccumulative, and toxic substance
RPD	Relative percent difference
SOP	Standard operating procedures
SPM	Suspended particulate matter
TSS	Total suspended solids
WAC	Washington Administrative Code
WDOH	Washington Department of Health

Units of Measurement

°C	degrees centigrade
cfs	cubic feet per second
dw	dry weight
mg	milligrams
mg/kg	milligrams per kilogram (parts per million)
mg/L	milligrams per liter (parts per million)
mm	millimeters
ug/L	micrograms per liter (parts per billion)
um	micrometer
uS/cm	microsiemens per centimeter, a unit of conductivity