Background Characterization for Metals and Organic Compounds in Northeast Washington Lakes

Part 1: Bottom Sediments

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Cover photo: Swan Lake, Ferry County (Michael Friese)

Background Characterization for Metals and Organic Compounds in Northeast Washington Lakes

Part 1: Bottom Sediments

by

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Waterbody Number or WRIA:

Swan Lake	WRIA 52
Davis Lake	WRIA 60
Ellen Lake	WA-58-9015
South Twin Lake	WA-58-9040
Summit Lake	WRIA 60
Pierre Lake	WA-60-9040
Cedar Lake	WA-61-9010
Pepoon Lake	WRIA 60
Williams Lake	WA-61-9080
Bayley Lake	WRIA 59
Sullivan Lake	WA-62-9190
Leo Lake	WA-62-9085
Browns Lake	WA-62-9030
Bead Lake	WA-62-9010
Upper Priest Lake	Idaho
St. Joe River	Idaho

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Abstract

A range of potentially toxic metals and organic compounds were analyzed in sediments and fish collected in 2010 from 14 northeast Washington lakes thought to be minimally impacted by local human activities. One lake and one river were also sampled in northern Idaho. The goal of the study was to provide regional-scale sediment and fish-tissue data that will support contaminant studies and cleanup activities associated with northeastern Washington waterbodies. This report presents the results on sediment samples only.

The following chemicals were analyzed in sediment: antimony, arsenic, barium, cadmium, chromium, copper, iron, lead, manganese, mercury, zinc, polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and polybrominated diphenylethers (PBDEs). Total organic carbon and grain size were also determined. Sensitive analytical methods were used to achieve low detection limits for the target chemicals.

Data presentation includes statistical summaries and figures showing spatial patterns, data distributions, and waterbodies ranked by concentration levels. Results are compared to previous sediment quality investigations in northeast Washington, including an indication of local *natural background* as deduced from an independently collected lake sediment core from Black Lake. The lake sediments are screened for potential sediment toxicity against sediment quality guidelines for protection of aquatic life. Approximate sediment concentration values representative of the 15 lakes, based on the objectives of sample collection methods employed, are calculated to support cleanup efforts at contaminated sites or for other uses.

A major study finding of concern is elevated concentrations of antimony, arsenic, cadmium, lead, mercury, and zinc in lake sediments from the western part of the study area along the Upper Columbia River. The probable source of contamination is historical transboundary air pollution from the Trail smelter in British Columbia and potentially to a lesser extent limited contribution from an historic smelter in Northport, Washington. Antimony, lead, and cadmium substantially exceeded sediment quality guidelines in several of these lakes. Dioxin (2,3,7,8-TCDD) was also higher in the western lakes, for reasons not determined.

The lakes in the eastern part of the study area exhibited lower levels of these and other contaminants, not much above estimated *natural background* as indicated by the Black Lake sediment core. With certain exceptions, the vast majority of the waterbodies had sediment concentrations of barium, chromium, copper, manganese, iron, PCBs, TCDD TEQs (toxic equivalents), and PBDEs that were broadly similar across the study area.

Acknowledgements

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

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    - Keith Seiders helped with study design.
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Introduction

An understanding of current background conditions provides an objective and consistent point of reference for assessing contaminated aquatic environments. Sediment chemistry, bioaccumulation, benthic community structure, and bioassay response are common measures where reference values are often established. San Juan (1994) lists a variety of potential uses for this type of information including defining background, screening contaminant data, risk assessments, regulatory compliance, evaluating waste streams, designing investigative studies, and research.

The goal of the present 2010 study is to provide data on the current status of freshwater sediments and fish that could be used as a reference for assessing and cleaning up various northeast Washington waterbodies potentially contaminated with toxic metals and halogenated[1] organic compounds. The fish tissue data are being reported separately in a second document (Part 2). The present report (Part 1) covers the sampling design, methods, results, and data interpretation for sediment samples.

The guidance for cleanup targets in the Model Toxics Control Act (MTCA, WAC 173-340-200) draws a distinction between two types of background:

- **Natural background:** For the purposes of hazardous substance cleanup under MTCA, natural background refers to the concentration of a constituent that occurs naturally in the environment and has not been influenced by localized human activities. Metals that occur naturally in bedrock and soils are cited as an example. Man-made chemicals such as PCBs are included by MTCA as part of the natural background when their presence is due to widespread use and global atmospheric transport.

- **Area background:** MTCA defines area background as the concentrations of substances that are consistently present in the environment in the vicinity of a site and which are the result of human activities unrelated to releases from that site. Blakley et al. (1992) gives the example of different lead levels in Seattle soils compared to Tacoma; area background for lead would therefore be different for the two cities. Area background is a site-specific determination.

Determination of background levels has also been an aspect or focus of numerous water quality investigations in Washington. Two in particular provided an impetus for the present study:

In 2007-2008, the Washington State Department of Ecology (Ecology) conducted *An Assessment of the PCB and Dioxin Background in Washington Freshwater Fish* (Johnson et al., 2010). Ecology needed this information to help prioritize the state's resources for cleaning up waterbodies that did not meet U.S. Environmental Protection Agency (EPA) Federal Clean Water Act human health criteria for fish consumption. The study showed that levels of these chemicals were often lower in the far eastern counties.

[1] In this study, compounds containing chlorine or bromine.

Another statewide effort by Ecology, *Baseline Characterization of Nine Proposed Freshwater Sediment Reference Sites* (Sloan and Blakley, 2009), was designed to screen for reference areas for freshwater sediment investigations. The study revealed a general lack of information on chemical and biological conditions for aquatic sediments in eastern Washington.

In the context of these two studies, the terms *background* and *reference* are essentially synonymous, meaning waterbodies that were thought to exhibit relatively low impact from human activities. Given the extent of urban, industrial, and agricultural development in the Pacific Northwest and world-wide, all Washington waterbodies have been affected to at least some degree by human activities.

The present report generally uses the term *background* location when referring to northeastern Washington waterbodies that were judged to be minimally impacted by local human activities. These waterbodies are further affected to varying degrees by watershed-scale atmospheric influences.

The regional variability and gaps in the background data on sediments and fish came to the attention of Ecology's Eastern Regional Office (ERO). ERO expressed concern that the use of statewide-based reference values for decision-making purposes in eastern and northeastern Washington would tend to inappropriately bias outcomes, particularly for cleanup actions. They saw a need for chemical data specific to northeast Washington. Sediment and fish tissue background assumptions affect the ability to differentiate between point-source impacts and appropriate background designations. In view of these concerns, Ecology initiated a project to survey a range of metallic and organic contaminants in sediments and fish from potential background eligible lakes in northeast Washington.

Project Summary

Current understanding of area or regional-scale conditions for chemical contaminants in aquatic environments in northeast Washington is limited. Therefore, a field study was conducted to achieve enhanced testing of selected waterbodies in Ferry, Stevens, and Pend Oreille counties, as well as two representative waterbodies in northern Idaho. Fourteen lakes were sampled in Washington. One lake and one river were sampled in Idaho.

The objective of the study was to characterize the levels of selected potentially toxic metals and halogenated organic compounds in bottom sediments and fish tissues from waterbodies that exhibit relatively low direct impact from human activities. Factors considered in waterbody selection included land-use development, proximity to historical mining, known industry and agriculture, general local watershed conditions, and known lake management history. The study focused principally on lakes whose quality was not believed to be influenced by notable human-oriented activities that are known to jeopardize environmental quality. Lakes dominated the study group since larger rivers and streams in the study area often could not be included due to a variety of known or potential anthropogenic influences.

The initial round of field work took place during the late summer and fall of 2010. An additional set of fish samples was collected in the spring of 2011 in an attempt to fill data gaps for certain waterbodies and species. Because of a delay in obtaining the final fish tissue data for the project, the present report is limited to the results on sediments.

Sediment samples were analyzed for antimony, arsenic, barium, cadmium, chromium, copper, iron, lead, mercury, manganese, zinc, polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and polybrominated diphenylethers (PBDEs). Also analyzed were grain size and total organic carbon (TOC). The same metals and organic chemicals were analyzed in fish, with the exception of barium, chromium, manganese, and iron. Sensitive analytical methods were used to achieve low detection limits for the target chemicals.

The study was designed and conducted by Ecology's Environmental Assessment Program with the assistance of ERO. Samples were analyzed primarily by Ecology's Manchester Environmental Laboratory (MEL) and Pacific Rim Laboratories, Inc. The study followed a Quality Assurance Project Plan (Johnson, 2010) developed in accordance with the Ecology guidance in Lombard and Kirchmer (2004).

Study Design

Study Area

The study area for this project encompasses Ferry, Stevens, and Pend Oreille Counties. These counties include or are adjacent to the majority of cleanup and hazardous waste sites in northeast Washington. The study area provides a number of lakes exhibiting relatively low impacts from development, compared to adjacent, more populated counties such as Spokane County.

ERO requested that Upper Priest Lake and the upper St. Joe River in northern Idaho be included in the study, in view of their remote locations in areas largely surrounded by wilderness and their proximity to eastern Washington. Upper Priest Lake lies within the Pend Oreille basin. The St. Joe River flows into Lake Coeur d'Alene, which drains to the Spokane River.

Target Chemicals

Sediment samples were analyzed for the metals, organic compounds, and other parameters listed in Table 1. These were identified by ERO as being of primary concern. The term *congener* means one of many variants or configurations of a common chemical structure. For example, the PCBs analysis included 209 individual compounds or congeners.

Table 1. Target Chemicals and Conventional Parameters for Sediment and Fish Samples.

Chemical	Sediments (present report)	Fish Tissues	
		Fillet	Whole Body
Antimony	**X**		X
Arsenic	**X**		X
Cadmium	**X**		X
Copper	**X**		X
Lead	**X**		X
Mercury	**X**	X	X
Zinc	**X**		X
Barium	**X**		
Chromium	**X**		
Manganese	**X**		
Iron	**X**		
PCBs (209 congeners)	**X**	X	X
PCDDs (7 congeners)	**X**	X	X
PCDFs (10 congeners)	**X**	X	X
PBDEs (36 congeners)	**X**	X	X
Grain size	**X**		
Total Organic Carbon	**X**		
% Lipids		X	X

Mercury, cadmium, lead, PCBs, PCDDs, PCDFs, and PBDEs are persistent, bioaccumulative toxics (PBTs) that are a hazard for fish and other aquatic life, wildlife, and human health (www.ecy.wa.gov/programs/swfa/pbt). The other metals analyzed also have toxic properties but seldom bioaccumulate in aquatic environments so are not classed as PBTs. Detailed profiles on the target chemicals for this study – describing health effects, physical/chemical properties, production and use, environmental occurrence, regulations, and analysis methods – have been prepared by the Agency for Toxic Substances & Disease Registry (www.atsdr.cdc.gov/toxprofiles/index.asp). This website profiles hazardous substances found at National Priorities List (Superfund) sites.

The metals analyzed in this study are naturally occurring at crustal concentrations in rocks and soils, and have a long history of use in industry and domestic products. Mining and ore processing in particular are known in some cases to locally affect water quality and sediment chemistry in close proximity to historic operations within certain northeast Washington tributaries by mobilizing or releasing mercury, cadmium, lead, and other metals (USGS, 2010; Pelletier and Merrill, 1998; Raforth et al., 2004). On a watershed scale, the Spokane River drainage exhibits metals impacts due to historic world-class mining and milling operations in Idaho. And the upper Columbia River exhibits metals impacts caused by metals smelting operations in Trail, British Columbia.

PCBs came into use in 1929. Commercial PCBs were manufactured as mixtures with varying chlorine content. PCBs were used as insulators in electrical transformers and capacitors, in plasticizers, lubricants, and hydraulic fluids, as well as in inks and sealers for gaskets and furnaces. Manufacture and use of PCBs was banned by EPA in the 1970s and 1980s due to ecological concerns. Historically, the Spokane River had some of the highest PCB levels in Washington freshwater fish (Serdar et al., 2011).

PCDDs and PCDFs, commonly referred to as dioxins and furans, are unintended byproducts found in association with certain industrial sites, waste incinerators, and combustion, especially of chlorinated material. 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) and 16 related compounds with similar structure are particularly toxic. Pulp and paper mills that used chlorine in their bleaching process were a major historical source of dioxins and furans in the Pacific Northwest, and mills operating in British Columbia discharged these wastes to the upper Columbia River, (EPA, 1991). This practice was discontinued in the 1990s. Nationwide and in Canada, reductions in dioxin and furan releases have occurred from a combination of regulatory activities, improved emission controls, voluntary actions on the part of industry, and the closing of a number of facilities.

PBDEs are brominated flame retardants added to a wide variety of household products such as upholstered furniture, computers, cable insulation, and textile coatings. PBDEs have been used extensively for the last 30 years, with the U.S. and Canada being the largest consumers of products treated with PBDEs (Ecology and WDOH, 2006). Studies have shown that toxic PBDEs leach from these products and accumulate in the environment. Here again, the upper Columbia and Spokane Rivers have figured prominently among the waterbodies with elevated PBDE levels in fish tissues (Rayne et al., 2003; Seiders et al., 2007).

Three main types of PBDEs are used in consumer products: Penta-BDE, Octa-BDE, and Deca-BDE. Manufacturers of Penta-BDE and Octa-BDE agreed to voluntarily stop producing these two forms by the end of 2004. In 2009, three major producers of Deca-BDE arrived at an agreement with EPA to stop producing, importing, and selling Deca-BDE by the end of 2012. A Washington State ban on the manufacture, sale, and distribution of televisions, computers, and residential upholstered furniture containing Deca-BDE took effect in January 2011.

Other parameters measured in the sediment samples from northeast Washington included TOC and grain size. These parameters can be useful for normalizing chemical data to compare contaminant levels among waterbodies. Metals tend to be associated with the finer particle sizes of aquatic sediments, while PCBs and the other target compounds have an affinity for the organic carbon fraction.

Waterbody Selection

ERO and Ecology's Environmental Assessment Program developed a preliminary list of potential minimally impacted lakes by examining Washington State maps and GIS coverages showing population density, agricultural land use, industrial and municipal outfalls, surface mines, and public lands. Recommendations were also provided by Bill Baker of the Washington Department of Fish and Wildlife (WDFW) and Sheri Sears, Resident Division Fish Manager for the Colville Confederated Tribes. This effort identified waterbodies that are believed to exhibit relatively low direct impact from human activities and have a low probability of local sources of contamination.

Each candidate lake was then checked against Ecology's Facility Site Identification System (www.ecy.wa.gov/fs/index.html) to identify potential activities that could affect their inclusion in the study. Facility Site identifies sites known to Ecology as having an active or potential impact on the local environment. Facility Site showed several mines or mining related sites were located in the Cedar Lake watershed (Lucky Four Mine, Redtop Mine, Northport Minerals). After further inspections it was decided to retain Cedar Lake in the study due to its favorable location in the uppermost part of the Columbia River drainage and a determination of the low potential for actual impact from mining and milling in the area.

Ecology and WDFW staff were contacted to determine if any of the lakes considered for study had been chemically treated to control aquatic plants, algae, or undesirable fish species. Records showed several of these lakes had been treated with rotenone in the past as part of a WDFW program to eliminate spiny-rayed fish and rehabilitate the trout fishery. The historic use of rotenone, a natural product derived from derris root, was not viewed as compromising a lake's usefulness for this study. Pepoon Lake was treated with toxaphene in 1962, also for fisheries enhancement. Toxaphene, a chlorinated pesticide, is not a regional chemical of interest in this study.

Based on the above evaluations, 26 lakes were preliminarily selected. After identifying location and condition of boat ramps or other means of access, contacting regional biologists to determine what fish species are present and fish stocking history, and field reconnaissance, 16 waterbodies were ultimately sampled (Table 2, Figure 1).

Table 2. Lakes and Rivers Sampled for the Northeast Washington Background Study in 2010.

Waterbody	County	WRIA No.	Elevation (feet)	Surface Area (acres)	Maximum Depth (feet)	Latitude	Longitude
Washington							
Swan Lake	Ferry	52	3,641	52	95	48.512	118.839
Davis Lake		60	4,550	17	45	48.739	118.231
Ellen Lake		58	2,300	78	34	48.501	118.256
South Twin Lake		58	2,572	973	57	48.264	118.387
Summit Lake	Stevens	60	2,600	7	35	48.959	118.127
Pierre Lake		60	2,012	106	75	48.905	118.139
Cedar Lake		61	2,135	52	28	48.943	117.594
Pepoon Lake		60	2,450	11	32	48.901	117.893
Williams Lake		61	1,980	38	47	48.755	117.968
Bayley Lake		59	2,400	17	12	48.420	117.664
Sullivan Lake	Pend Oreille	62	1,380	1,290	330	48.816	117.292
Leo Lake		62	2,588	39	37	48.648	117.495
Browns Lake		62	3,450	88	23	48.439	117.191
Bead Lake		62	2,850	720	170	48.299	117.116
Idaho							
Upper Priest Lake	Bonner	(Idaho)	2,441	1,338	100	48.786	116.889
St. Joe River	Clearwater	(Idaho)	3,198	na	na	47.202	115.516

WRIA: Water Resource Inventory Area.
na: not applicable.

An attempt was made to distribute the sampling effort more or less evenly across the study area, although this was not always practical. Most of the selected lakes lie in a north-south gradient along the Columbia River or Pend Oreille River drainages. In addition, an emphasis was placed on selecting representative upland lakes near the Columbia River and international border due to documented transboundary pollution issues. Historically, the vicinity has been subject to significant air emissions from industries in British Columbia, as described later in this report.

Potential pollutant source risks and geographic location distribution were defined as important selection factors; size was not. Larger lakes tend to have longer food chains, which may result in some species attaining higher levels of bioaccumulative chemicals in their tissues. High mountain lakes are subject to enhanced atmospheric deposition of synthetic organic compounds due to colder temperatures and larger amounts of precipitation (Wania and Mackay, 1993; Gillian and Wania, 2005). High lakes also typically have a low diversity of fish species. High mountain lakes were thus avoided for this study.

The study sampled a diverse range of lake sizes and elevations to obtain a regional assessment of the chemical background. The lakes selected for study range in size from less than 10 to over 1,000 acres, with maximum depths of 12 to 330 feet. Elevations are between about 2,000 and 4,500 feet; most lakes were below 3,000 feet.

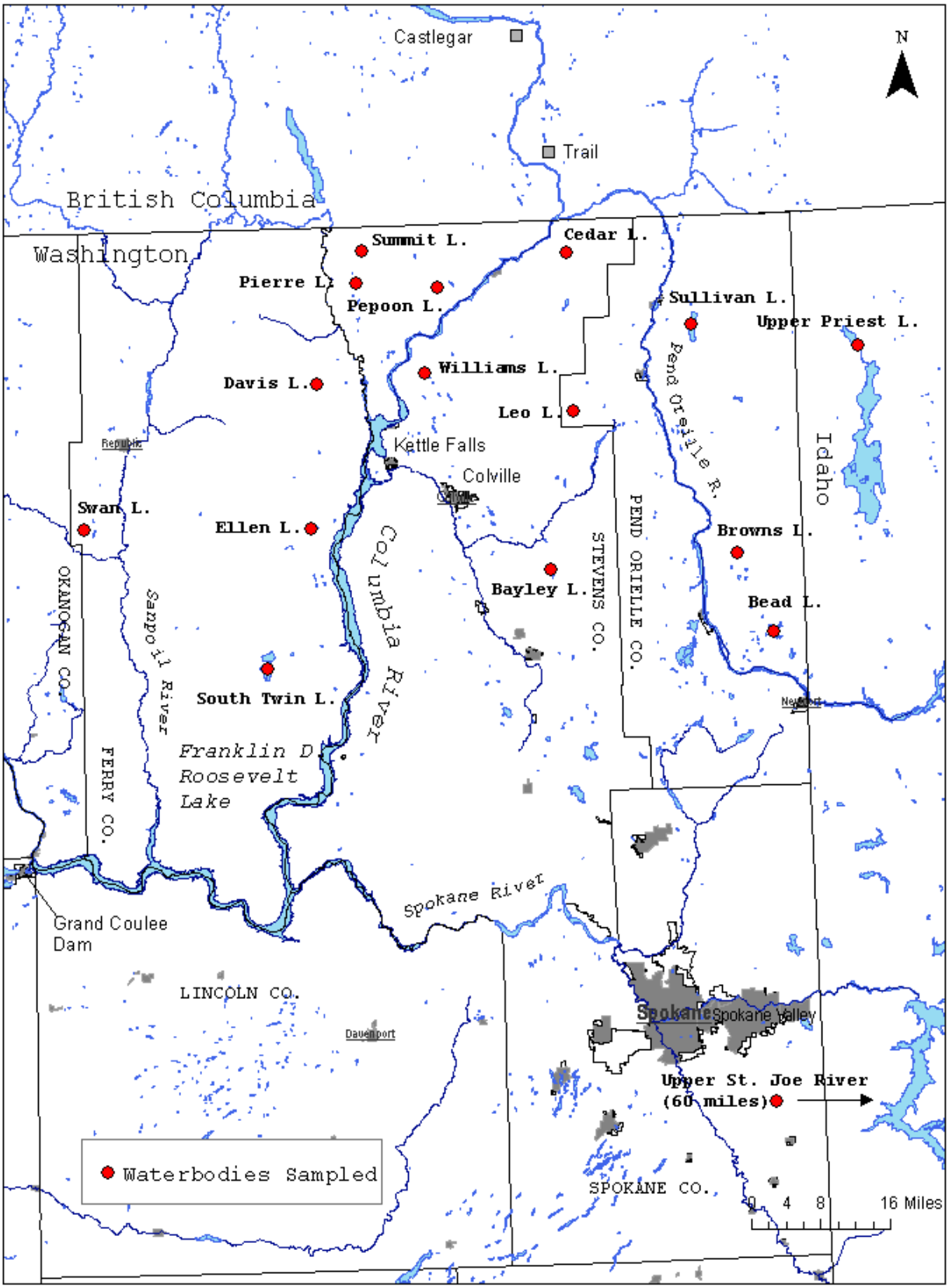


Figure 1. Waterbodies Sampled for the Northeast Washington Background Study in 2010.

Appropriate lakes could not be readily identified in the southern parts of Ferry and Stevens Counties or in northern Spokane County, which are more highly developed. Northwest Ferry County is lacking in lakes meeting the selection criteria.

Number and Type of Samples

MTCA has sample size requirements for defining background conditions (173-340-709 WAC). For soil investigations, determination of *natural background* requires collection of at least 10 samples, while determination of *area background* requires at least 20 samples. The minimum number of samples required for aquatic sediments or other media is not defined in the regulation and must be determined on a case-by-case basis. Sample size for the northeast Washington study was 16: one sediment sample per lake plus a single riverine sediment sample.

The project's regional orientation and budget considerations did not allow more than one sediment sample to be analyzed for each waterbody. This study assumes that the degree of spatial variability in contaminant concentrations in surficial sediments from each lake is reasonably low. Thus, a single composite sample was assumed to be representative of sediment quality for this initial regional assessment.

To enhance representativeness, each sediment sample was a composite of three separate grabs. The grabs were composited from three relative lake depths: shallow, mid-depth, and maximum depth. The grabs were placed along a longitudinal transect, with the shallow and mid-depth samples taken at approximately 1/3 and 2/3 of the maximum depth. For some of the lakes, heavy growths of aquatic plants prevented collection of a shallow sample. The samples were collected away from lake inflows, boat launches, cabins, camp sites, and other such disturbances. Appendix A has detailed information on the location of sediment samples in each lake and also on the types of fish tissue samples currently under analysis.

Sediment samples were from the top-10 cm layer. This increment is generally considered to include the biologically active zone (Ecology, 2008; Blakley, 2008). For these types of lakes, the 10-cm depth increment typically covers a depositional history spanning tens of years, as illustrated later in this report.

Sedimentation rates can be low in many upland lakes, depending on watershed characteristics and history. A top-10 cm sample may blend many decades of depositional history. Soft sediments make it difficult to representatively sample this layer using standard techniques. The Ponar sampling approach applied in this study was not intended to isolate distinct sediment time horizons, resulting in an anticipated depositional averaging and pollution history at any given waterbody.

Sediment samples from the upper St. Joe River likewise consisted of a composite from three separate locations collected within a 200-foot length of stream near the shoreline. These were taken from areas where deposits of fine material occurred, selected in such a way as to avoid bank sloughed material and local tributary inputs. Because bed sediments in a river of this nature are continually mixed and remobilized, it was judged not critical to adhere to a 10-cm depth increment.

Analytical Methods and Detection Limits

Low-level methods were used to minimize the number of non-detects in the data. Metals were analyzed by MEL using inductively-coupled plasma/mass spectrometry (ICP/MS) and cold vapor atomic absorbance techniques (CVAA, mercury only). Reporting limits for sediments were in the range of 0.005 mg/Kg for mercury and 0.l -0.5 mg/Kg for other metals, except 5 mg/Kg for zinc and 250 mg/Kg for iron (parts per million).

Organic compounds were analyzed by Pacific Rim Laboratories in Surrey, B.C. using high resolution gas chromatography/mass spectrometry (HR-GC/MS). Method detection limits in sediment were as low as 0.1 ng/Kg for PCBs and PBDEs, and 0.03 ng/Kg for PCDDs/PCDFs, depending on the congener in question (parts per trillion). These are the lowest detection limits currently available through laboratories accredited by Ecology for these methods.

Methods

Field

Sediment collection and handling followed the Ecology Environmental Assessment Program's standard operating procedure for freshwater sediment samples (Blakley, 2008). The lake samples consisted of composites from two to three grabs taken with a 0.05 m^2 Ponar sampler. A grab was considered acceptable if not over-filled with sediment, overlying water was not excessively turbid, the sediment surface was relatively flat, and the desired depth penetration was achieved. Stainless steel scoops were used to sample sediments from the upper St. Joe River.

For Ponar-collected samples, overlying water was siphoned off and the top-10 cm of sediment removed with a stainless steel scoop, placed in a stainless steel bowl, and homogenized by stirring. Material touching the side walls of the grab was not taken. The scoop samples from the upper St. Joe were homogenized in the same way.

Subsamples of the homogenized sediment were transferred to glass jars that had been cleaned to EPA (1990) quality assurance/quality control specifications and placed on ice immediately upon collection. The samples were returned to Ecology headquarters and held frozen until transported with chain-of-custody record to MEL. MEL shipped the organics and grain size samples to the contract laboratories. Sample containers and holding times for the project are listed in Table 3.

Table 3. Sample Containers, Preservation, and Holding Times.

Parameter	Container	Field Preservation	Holding Time
Metals	4 oz. glass w/ Teflon lid liner	Cool to 4°C	2 years (frozen); mercury 28 days
Organic Compounds	4 oz. glass w/ Teflon lid liner	Cool to 4°C	1 year (frozen)
Total Organic Carbon	2 oz. glass w/ Teflon lid liner	Cool to 4°C	6 months (frozen)
Grain size	8 oz. polyethylene	Cool to 4°C	6 months

Stainless steel implements used to collect and manipulate the sediments were cleaned by washing with Liquinox detergent, followed by sequential rinses with tap water, dilute nitric acid, deionized water, and pesticide-grade acetone. The equipment was then air dried and wrapped in aluminum foil for transport into the field. Cleaning of the Ponar between lakes consisted of thorough brushing and rinsing using on-site lake water.

Laboratory

Table 4 shows the methods and laboratories used to analyze the sediment samples. Bulk sediments were analyzed without additional sorting, sieving, or other preparation.

Excess sample was archived frozen at Ecology headquarters.

Table 4. Analytical Methods and Laboratories.

Analysis	Method	Reference	Laboratory
Antimony	ICP/MS	EPA 3050B / 200.8	MEL
Arsenic			
Barium			
Cadmium			
Chromium			
Copper			
Iron			
Lead			
Manganese			
Zinc			
Mercury	CVAA	EPA 245.5	
Total Organic Carbon	NDIR	PSEP-TOC	
Grain Size	Sieve and pipette	PSEP (1986)	Columbia Analytical
PCBs	HRGC/HRMS	EPA 1668A	Pacific Rim
PCDDs/PCDFs		EPA 1613B	
PBDEs		EPA 1614	

ICP/MS: Inductively coupled plasma - mass spectrometry.
CVAA: Cold vapor atomic absorbance.
NDIR: Non-dispersive infrared detector.
PSEP: Puget Sound Estuary Program.
HRGC/HRMS: High resolution gas chromatography/high resolution mass spectrometry.
MEL: Manchester Environmental Laboratory.

Data Quality

Data Review and Verification

MEL reviewed and verified all the chemical data for this project. The unit supervisor or an analyst experienced with the method performed the final review for the metals and TOC results generated by MEL. Quality assurance and quality control at MEL are described in MEL (2006, 2008).

MEL's quality assurance coordinator reviewed the organics analyses sub-contracted to Pacific Rim Laboratories and the grain size data produced by Columbia Analytical. The organics review followed National Functional Guidelines for Superfund Organic Methods Data Review (EPA, 2005a).

MEL prepared written case narratives assessing the quality of these data. The reviews include a description of analytical methods and an assessment of holding times, calibration, internal standard recoveries, ion abundance ratios, method blanks, on-going precision and recovery, labeled compound recoveries, matrix spike recoveries, laboratory control samples, and laboratory duplicates, as appropriate. With few exceptions, the results met acceptance criteria for these analyses, and the data are usable as qualified. The reviews and the complete MEL data reports are available from the author on request.

Method Blanks

Laboratory method blanks were included with each sample batch. EPA methods and guidance give wide latitude on how the method blank should be interpreted.

Low levels of some target compounds were detected in blanks for the organics analyses. In cases where the concentration measured in a sample was at least five times greater than the blank, the blank result was considered insignificant relative to the native concentration in the sample, and the data were used without further qualification (EPA, 2005a).

In a limited number of instances, results were accepted when only three times greater than the method blank. It was judged that a better estimate of the true concentration would be obtained by using, rather than ignoring, the detected values, as explained below.

- For one of the two PCDD/PCDF method blanks analyzed, the laboratory reported an estimated concentration between the practical quantitation limit (PQL) and estimated method detection limit (EMDL) for 1,2,3,7,8-PeCDD, 2,3,4,7,8-PeCDF, and 1,2,3,4,7,8-HxCDF. Eight affected results (2% of the data) were accepted and qualified as estimates when greater than three times this blank (EPA, 2005b). These congeners were not detected in the second blank.

- The laboratory reported concentrations of 15 PCB congeners (PCB-015, 107, 020/033, 022, 031, 047/048, 049, 056, 060, 105, 136, 141, 170, and 180) at or near the PQL (within a factor of 2) in one or both method blanks. Thirty-nine affected results (1% of the data) were accepted and qualified as estimates if greater than three times the blank.

- PBDE-049 was detected in both laboratory blanks at the EMDL. Two results greater than three times the blank were accepted and qualified as estimates. PBDE-209 was found at or slightly below the PQL in both laboratory blanks. Because similar levels were reported in both blanks, and because of the obvious presence of PBDE-209 in the study area, four results greater than three times the blank were accepted and qualified as estimates. Overall, these modifications affected less than 1% of the PBDE data.

Method blanks were not a data quality issue for metals or TOC. Method blanks are not analyzed for grain size.

Variability of the Data

The field variability inherent in chemical residue data was reduced by using composite samples. As discussed under Methods, the composite Ponar sampling method targeted a 10-cm top interval of sediments. Variability in sedimentation rates, depositional histories, and other characteristics may exist from waterbody to waterbody. For broad regional assessment purposes the methods applied are acceptable.

Estimates of analytical precision were obtained by analyzing laboratory duplicates (one homogenized sample split into two duplicate subsamples). The results are summarized for selected target chemicals in terms of relative percent difference (RPD) in Table 5. RPD is the difference between duplicates expressed as a percent of the mean value. Duplicates were not requested for the Upper Priest sample, but were analyzed for PCBs and PCDDs/PCDF at the discretion of the laboratory.

Most of the duplicate analyses for metals, TOC, and percent fines agreed within 12% or better, except for one mercury split at 22% RPD and one antimony split at 25% RPD. The lower level analyses (parts per trillion) for organic compounds agreed within a factor of 2 or better. The laboratory encountered co-extracting organic interferences or elevated method blanks for PBDE-209 in the South Twin and Bead samples which affected the precision of the PBDE duplicates.

The average of duplicate results is used in the remainder of this report. In the few instances where one sample of a duplicate pair was non-detect, the detected value was used.

Table 5. Precision on Duplicate (split) Sediment Samples for Selected Target Chemicals.
Metals in mg/Kg; organics in ng/Kg; dry weight.

Lake	Dup. #1	Dup. #2	RPD	Dup. #1	Dup. #2	RPD	Dup .#1	Dup. #2	RPD
	Antimony			Lead			Cadmium		
South Twin	0.69	0.64	8%	58	53	9%	1.2	1.1	9%
Bead	0.024 J	0.031 J	25%	24	24	0%	0.44	0.44	0%
	Arsenic			Mercury			Zinc		
South Twin	11	11	4%	0.14	0.11	22%	74	68	8%
Bead	6.1	6.2	2%	0.042	0.041	2%	76	81	6%
	Total PCBs*			TCDD			TCDD TEQs*		
South Twin	4,402	4,376	1%	0.45 NJ	0.20 NJ	78%	5.0	4.6	8%
Bead	1,877	2,642	34%	0.14 NJ	0.056 UJ	>86%	2.5	2.6	4%
Upper Priest	340	231	38%	.042 UJ	.042 UJ	0%	0.58	0.51	13%
	Total PBDEs*			Total Organic Carbon			Percent Fines		
South Twin	594	240	85%	14	15	7%	45	40	12%
Bead	399	210	62%	6.5	7.0	7%	75	72	5%

RPD: relative percent difference.
J: The analyte was positively identified. The associated numerical result is an estimate.
U: Not detected at or above the reported sample quantitation limit.
NJ: The analyte has been "tentatively identified"; the associated numerical value is the approximate concentration.
UJ: The analyte was not detected above the reported estimated quantitation limit.
*non-detects set at zero.

Results

General Physical Characteristics

The lakes sampled in this study tended to have soft, fine-grained sediments with a high organic carbon content. The single river sediment sample from the Upper St. Joe was mostly sand.

The silt and clay fractions (referred to as percent fines) typically comprised 40-80% of the lake sediments, except for the Cedar Lake deposits which were devoid of coarser material (Figure 2, Table 6). The median value was 58%.

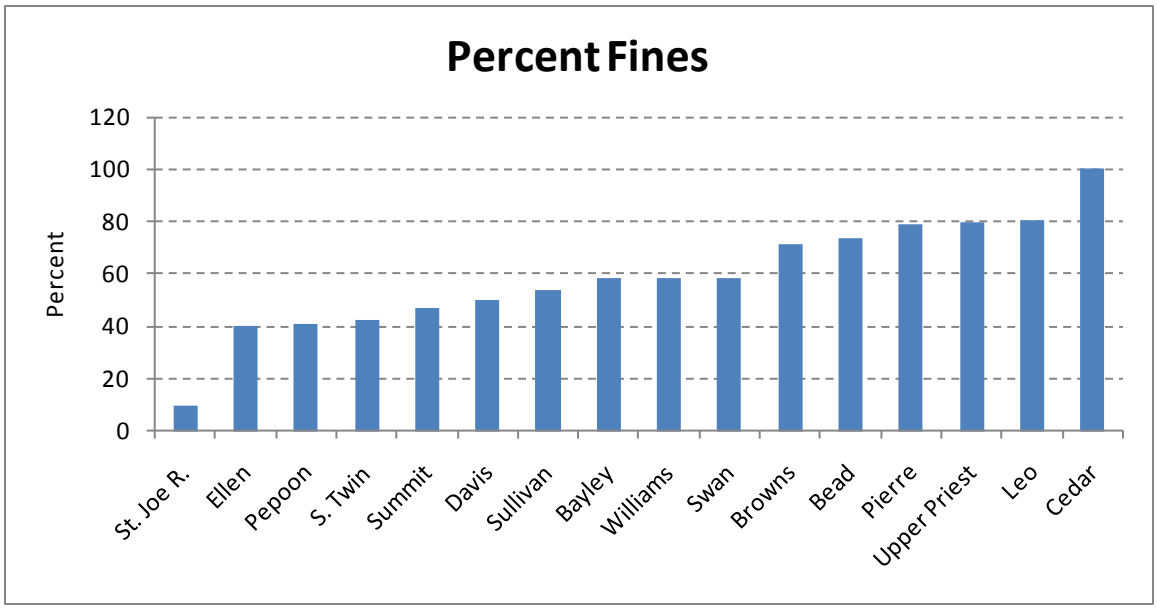


Figure 2. Percent Fines in Sediment Samples (silt+clay fractions; <0.0039-.0625 mm).

Table 6. Summary Statistics for Grain Size (Percent Fines*) in Sediment Samples.

Waterbodies:	Lakes	St. Joe River
N=	15	1
Median	58	9.3
Mean	62	- -
Minimum	40	- -
Maximum	100	- -
90th percentile	88	- -

*silt+clay fractions.

TOC in the lake sediments ranged from 3.5 – 24 mg/Kg, with a median of 15 mg/Kg (parts per million). The sediment sample from the Upper St. Joe River was relatively low in TOC (1.1 mg/Kg). (Figure 3, Table 7)

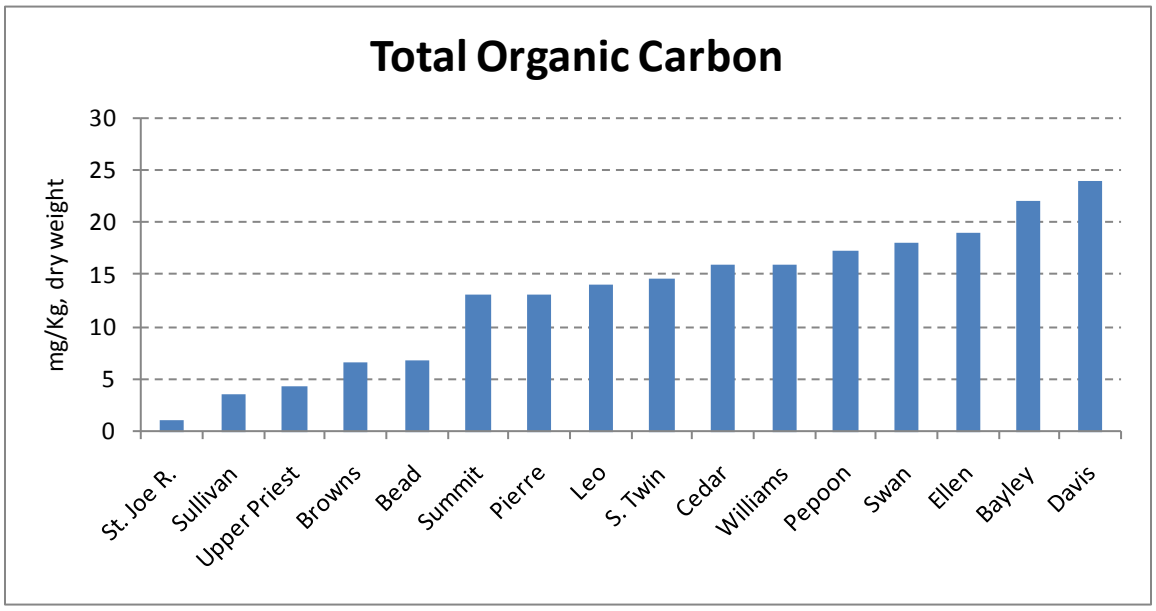


Figure 3. Total Organic Carbon in Sediment Samples (mg/Kg, dry).

Table 7. Summary Statistics for Total Organic Carbon in Sediment Samples (mg/Kg, dry weight).

Waterbodies:	Lakes	St. Joe River
N=	15	1
Median	15	1.1
Mean	14	- -
Minimum	3.5	- -
Maximum	24	- -
90th percentile	22	- -

Spatial patterns across the study area were not evident with respect to either grain size or TOC.

Metals Concentrations and Spatial Patterns

The metals concentrations measured in the sediments are shown in Table 8. All metals were present at quantifiable levels except for mercury and cadmium in the upper St. Joe River. The plots and data analysis that follow use the reporting limit for these non-detects. High concentrations of several metals in the Williams Lake sample caused these results to be qualified as estimates. Antimony concentrations less than 0.1 mg/Kg were between the reporting limit and method detection limit and were thus also qualified as estimates.

A matrix of correlation coefficients (R) was constructed to identify relationships among the metals analyzed (Table 9). R values that approach +1 indicate a strong positive relationship, while negative values suggest an inverse relationship. Statistically significant correlations are highlighted.

Antimony, lead, cadmium, arsenic, and zinc were significantly correlated. The relationship between antimony, lead, and cadmium was particularly strong (R=0.94-0.97). There was evidence of a moderate positive relationship with all five of these metals and mercury (R=0.32-0.51), although the correlations were not statistically significant. Additional metals pairs showing significant correlation were chromium/copper, barium/manganese, and manganese/iron.

Spatial patterns were observed that were consistent with these correlations. Lakes in the western part of the study area within the Columbia, Kettle, and Sanpoil river valleys – Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, South Twin, and Swan – had much higher concentrations of antimony, lead, cadmium, arsenic, and mercury than the eastern lakes in or around the upper Colville River and Pend Oreille River drainages. Zinc was also moderately elevated in the western lakes.

Concentrations of the five other metals analyzed – barium, chromium, copper, manganese, and iron – were either similar among lakes or slightly higher in the east. In most cases, the lowest metals concentrations were found in St. Joe River sediments.

Metals often concentrate in the finer size range of aquatic sediments. The influence of grain size on spatial patterns can be accounted for by normalizing dry weight-based metals concentrations to percent fines (Horowitz, 1985). However, sediment samples from the present study showed little or no evidence of a relationship between metals and fines (Table 9), indicating that normalizing was not appropriate for these fines-dominated samples.

Results for individual metals are discussed below. For each metal there is: (1) a statistical summary of the data, (2) a figure showing spatial patterns and data distribution, and (3) a graph ranking each waterbody by concentration. The same format is followed for organic compounds later in this report.

Table 8. Summary of Results for Metals Analyzed in Sediment Samples (mg/Kg, dry weight; parts per million).

Waterbody	Sb		Pb		Cd		As		Hg		Zn		Cu		Cr		Ba		Mn	Fe	% Fines
Washington																					
Swan Lake	0.68		53		1.1		8.1		0.197		78		24		8.9		115		410	15,900	58
Cedar Lake	3.4		141		3.0		12		0.075		151		13		3.8		182		270	6,120	100
Pepoon Lake	1.6	J	59	J	1.5		7.1		0.052		46		11		5.8		90		73	2,780	41
Summit Lake	0.70		27		0.77		8.0		0.099		20		8.1		19		32		228	2,940	47
Pierre Lake	0.73		41		1.4		14		0.133		76		77		53		83		69	19,300	79
Williams Lake	4.2	J	190	J	6.2		28	J	0.157		215	J	26	J	15		109	J	180	11,600	58
Davis Lake	1.6		63		1.5		10		0.208		62		11		7.5	J	79		225	19,900	50
Ellen Lake	1.2		89		2.3		11		0.202		104		23		8.7	J	96		289	16,500	40
S. Twin Lake	0.67		56		1.2		11		0.126		71		20		13		112		885	35,700	43
Sullivan Lake	0.03	J	9.9	J	0.24		4.5		0.031		63		23		15		111		243	23,500	54
Leo Lake	0.09	J	18		0.54		3.2		0.079		59		16		15		84		324	16,300	81
Browns Lake	0.07	J	21		0.51		4.6		0.060		62		17		12		83		103	11,300	71
Bayley Lake	0.13		14		0.58		3.7		0.103		50		22		9.5		140		475	14,000	58
Bead Lake	0.03	J	24		0.44		6.1		0.042		79		19		13		98		208	17,100	74
Idaho																					
Upper Priest Lake	0.02	J	27		0.65		6.1		0.082		117		27		21		333		3,010	43,500	80
St. Joe River	0.03	J	2.3		0.05	U	3.2		0.007	U	19		14		8.6		42		146	16,700	9

See Appendix D for metals abbreviations.
U: The analyte was not detected at or above the reported result.
J: The analyte was positively identified. The associated numerical result is an estimate.

Table 9. Pearson Correlation Coefficients (R) for Metals and Percent Fines in Sediment Samples

Significant correlations highlighted; >95% probability with Bonferroni correction for multiple comparisons.

	Sb	Pb	Cd	As	Zn	Hg	Cr	Cu	Ba	Mn	Fe
Sb											
Pb	0.97										
Cd	0.94	0.97									
As	0.84	0.88	0.94								
Zn	0.77	0.86	0.86	0.80							
Hg	0.35	0.45	0.44	0.51	0.32						
Cr	-0.18	-0.16	-0.05	0.21	0.00	0.06					
Cu	-0.06	0.00	0.09	0.30	0.17	0.20	0.90				
Ba	0.05	0.12	0.06	0.00	0.46	-0.02	0.01	0.09			
Mn	-0.22	-0.13	-0.15	-0.13	0.19	-0.02	0.09	0.04	0.88		
Fe	-0.37	-0.25	-0.24	-0.10	0.09	0.07	0.24	0.26	0.60	0.77	
Fines	0.17	0.20	0.14	0.10	0.45	-0.01	0.28	0.29	0.50	0.23	0.05

See Appendix D for metals abbreviations.

Antimony, Lead, Cadmium, Arsenic, Mercury, and Zinc

Antimony

The overall median and maximum concentrations of antimony in the lake sediments were 0.68 and 4.2 mg/Kg, respectively (Table 10). The 90th percentile[2] for the lakes was 3.7 mg/Kg. Antimony was estimated at 0.03 mg/Kg in the upper St. Joe River sediments.

Table 10. Summary Statistics for Antimony in Sediment Samples (mg/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Median	0.68	1.2	0.05	0.03
Mean	1.0	1.6	0.06	- -
Minimum	0.02	0.67	0.02	- -
Maximum	4.2	4.2	0.13	- -
90th percentile	3.7	3.5	0.11	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.

Of the 11 metals analyzed, antimony showed the greatest elevation in the western lakes compared to the eastern lakes. Concentrations in all western lakes were higher by approximately one order of magnitude (Figure 4). The western lakes ranged from 0.67 to 4.2 mg/Kg, while the eastern lakes had 0.02 to 0.13 mg/Kg. The quantile plot of the data in Figure 4 clearly shows two groups of lakes with discrete antimony levels. The differences between western and eastern lakes were statistically significant for antimony (Mann-Whitney test, $p<0.05$[3], St. Joe River excluded).

Lakes along the Columbia River, from South Twin to Cedar, showed evidence of increasing antimony levels moving northward toward the international border. The highest concentrations were recorded for the northern-most lakes in this area: Williams (4.2 mg/Kg), Cedar (3.4 mg/Kg), and Pepoon/Davis (1.6 mg/Kg). The lowest antimony levels were found in Sullivan, Leo, Brown, and Upper Priest Lakes to the east (<0.10 mg/Kg). As described in subsequent parts of this report, the level of chemical contamination in Williams Lake sediments stands out with respect to several additional metals (lead, cadmium, arsenic, zinc) as well as for PCBs.

[2] Percentiles describe a location in the distribution of data. For the 90th percentile, 10% of the data lie above the value and 90% lie below.

[3] p represents the probability of error in accepting a result as valid and representative of the population being sampled. For example, at p= 0.05 (i.e., 1/20) there is a 5% probability that the difference between a variable measured in set of samples is due to chance (a fluke). This report uses the 5% level as the cutoff for ascribing significant differences between results (i.e., 95% confidence).

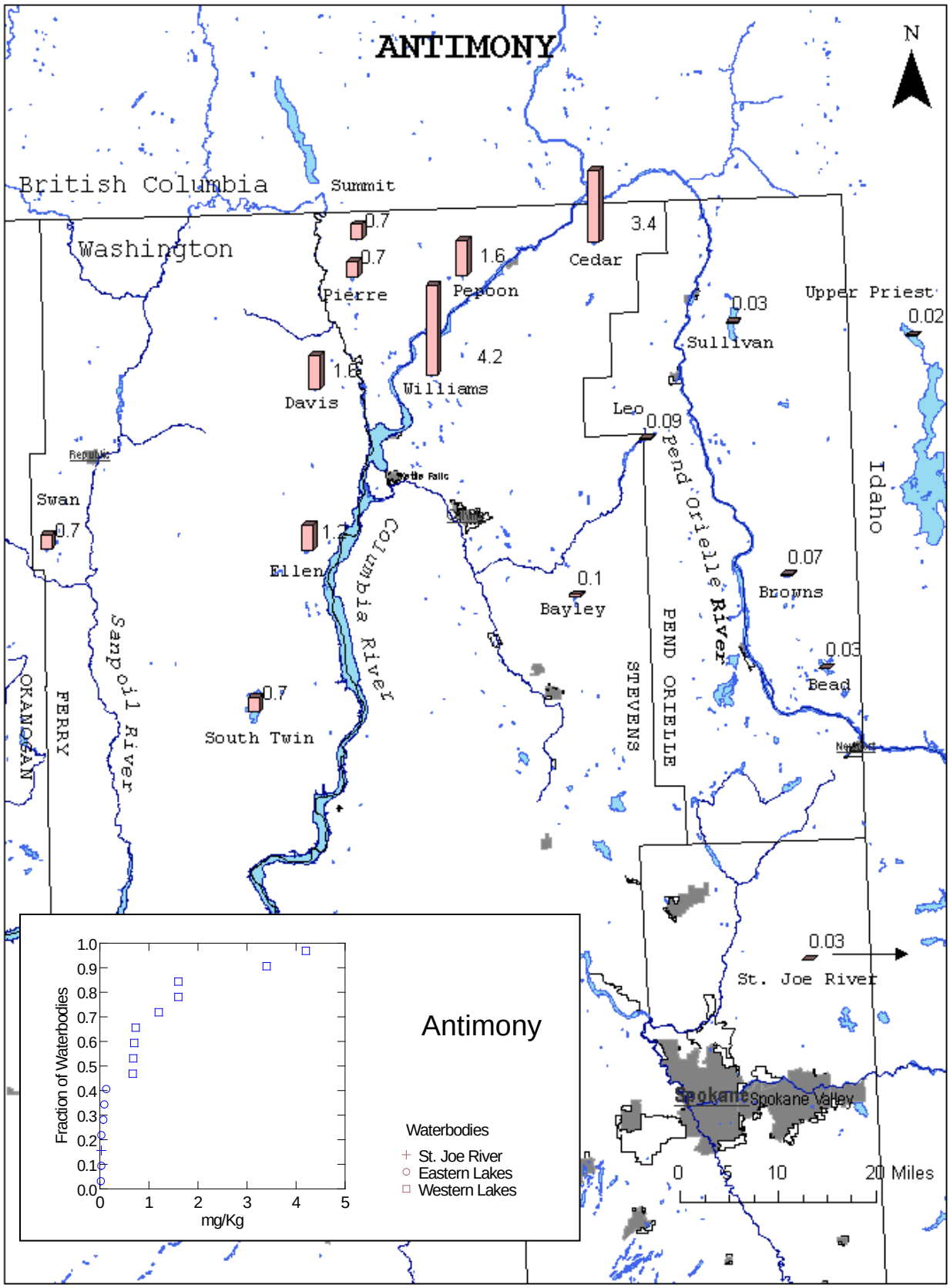


Figure 4. Spatial Patterns and Quantile Plot for Antimony in Sediment Samples.

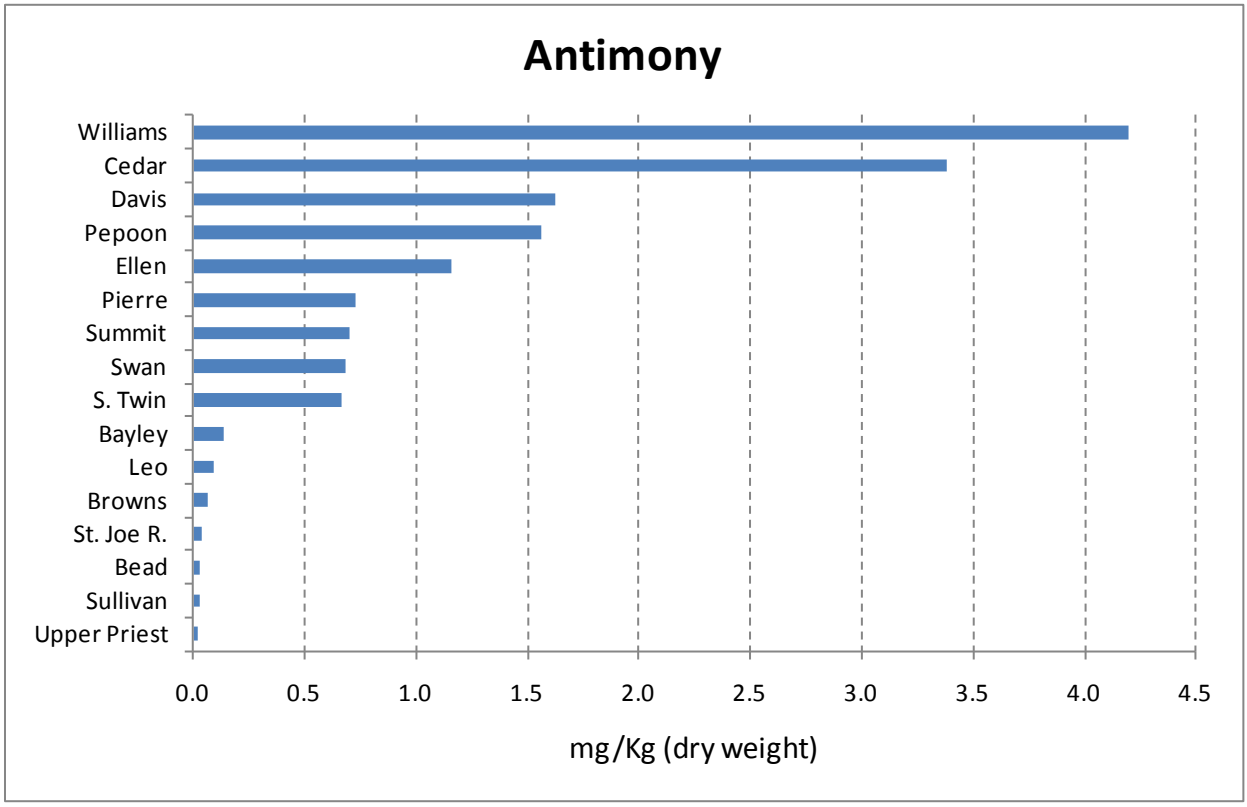


Figure 5. Waterbodies Ranked by Antimony Concentrations in Sediment Samples.

Lead

The overall median and maximum concentrations of lead in the lake sediments were 41 and 190 mg/Kg, respectively (Table 11). The lake 90th percentile was 120 mg/Kg. The lowest lead concentration, 2.3 mg/Kg, was recorded for the St. Joe River.

Table 11. Summary Statistics for Lead in Sediment Samples (mg/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Median	41	59	20	2.3
Mean	55	80	19	- -
Minimum	9.9	27	9.9	- -
Maximum	190	190	27	- -
90th percentile	120	151	25	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.

Like antimony, lead was consistently elevated in the western lakes (Figure 6), with median concentrations higher than the eastern lakes by about a factor of 3 (59 vs. 20 mg/Kg). A similar trend toward increasing concentrations moving north along the Columbia River was evident. Lakes that had the highest antimony concentration were also highest in lead. The lowest concentration was recorded for Sullivan Lake (9.9 mg/Kg), followed by Bayley, Leo, and Browns Lakes (14-21 mg/Kg), in that order. Here again, the distribution of the data suggests two separate groups of lakes with respect to lead levels (Mann-Whitney test, $p<0.05$).

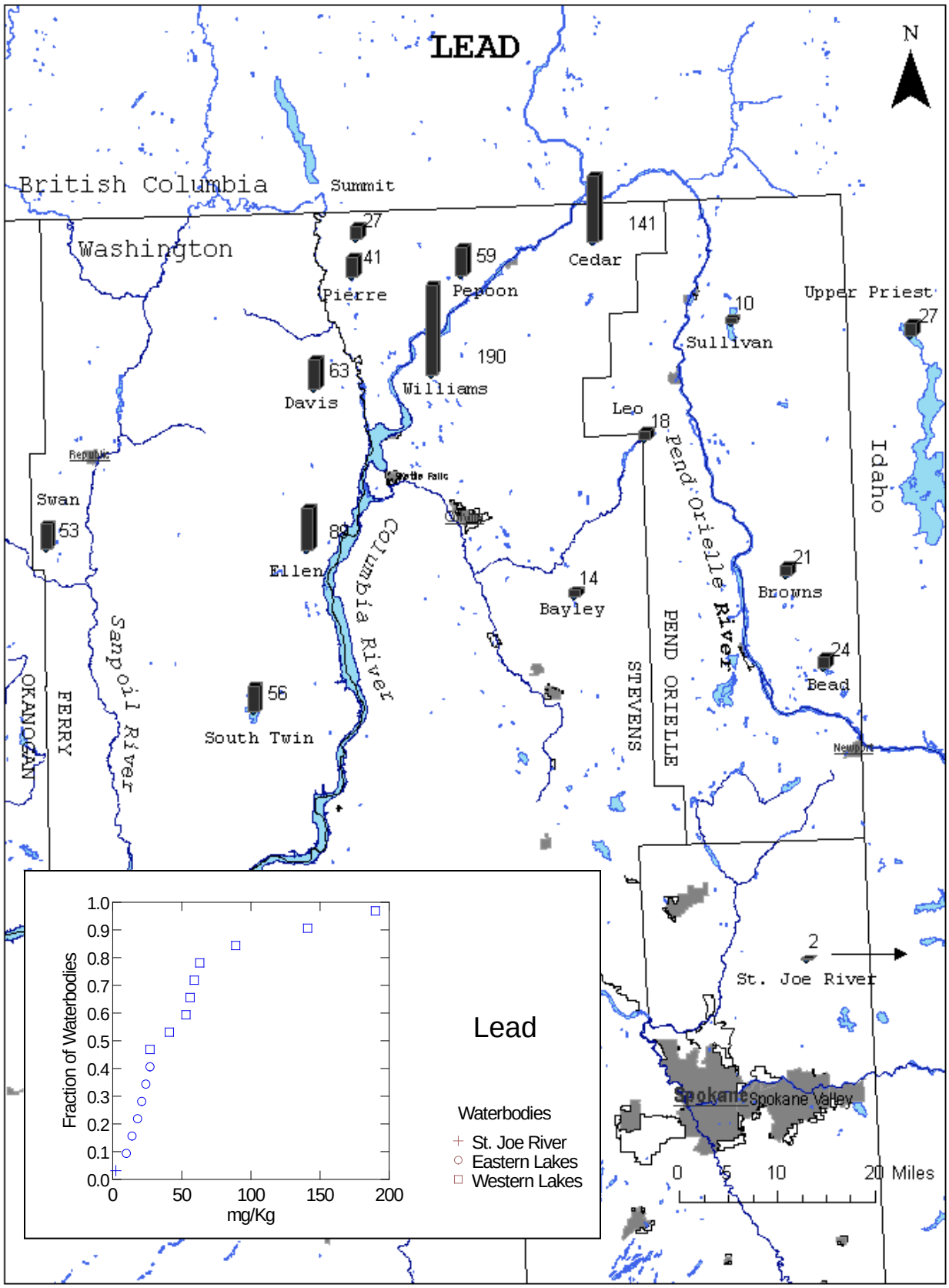


Figure 6. Spatial Patterns and Quantile Plot for Lead in Sediment Samples.

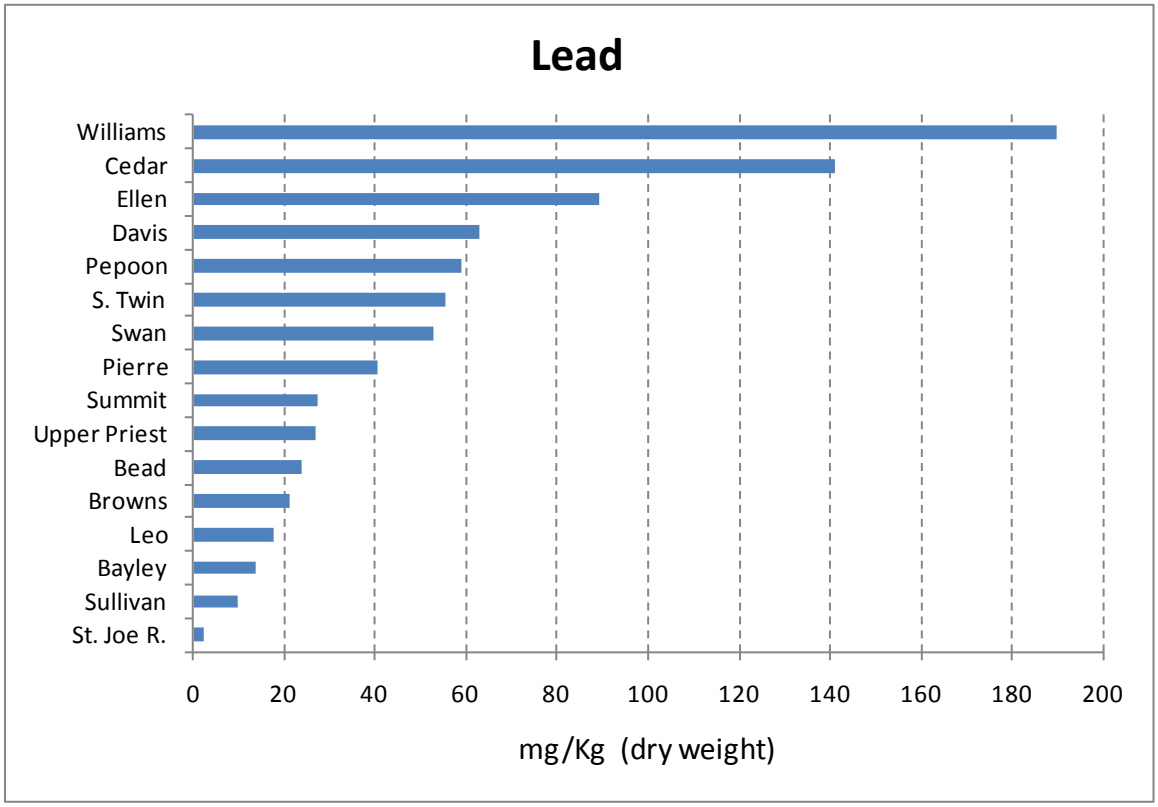


Figure 7. Waterbodies Ranked by Lead Concentrations in Sediment Samples.

Cadmium

For the lake sediments, cadmium had a median and maximum concentration of 1.1 and 6.2 mg/Kg, with 90th percentile of 2.8 mg/Kg (Table 12). Cadmium was not detectable in the St. Joe River sediments (<0.05 mg/Kg).

Table 12. Summary Statistics for Cadmium in Sediment Samples (mg/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Median	1.1	1.5	0.52	0.05 U
Mean	1.5	2.1	0.49	- -
Minimum	0.24	0.77	0.24	- -
Maximum	6.2	6.2	0.65	- -
90th percentile	2.8	3.7	0.61	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.
U: The analyte was not detected at or above the reported result.

As anticipated from their statistical correlation, spatial patterns for cadmium closely resembled antimony and lead (Figure 8). The western lakes ranged from 0.77-6.2 mg/Kg and the eastern lakes from 0.24-0.65 mg/Kg. Similar to lead, median cadmium concentrations were about three times higher in the western lakes.

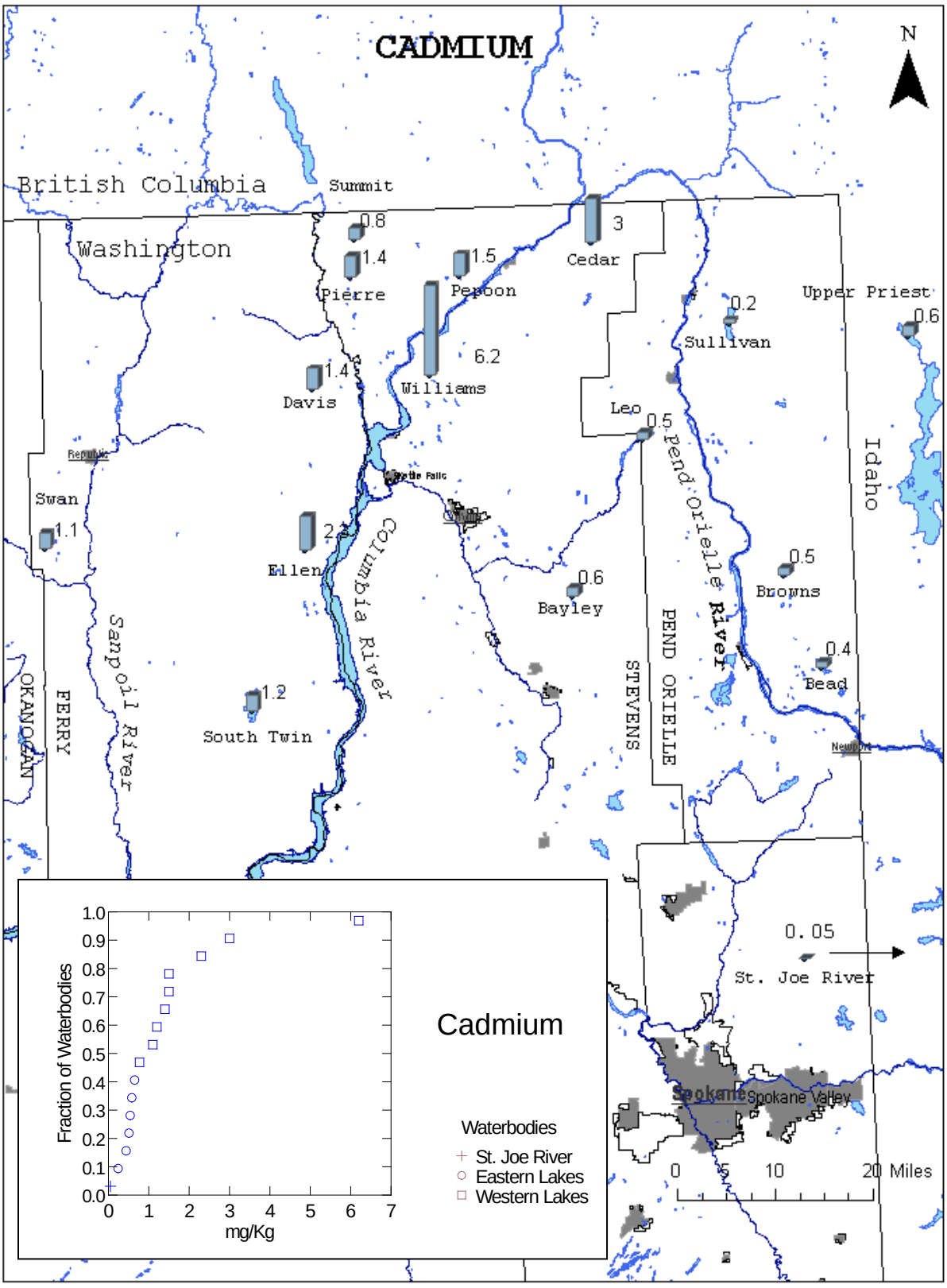


Figure 8. Spatial Patterns and Quantile Plot for Cadmium in Sediment Samples.
St. Joe River plotted at the reporting limit (0.05 mg/Kg).

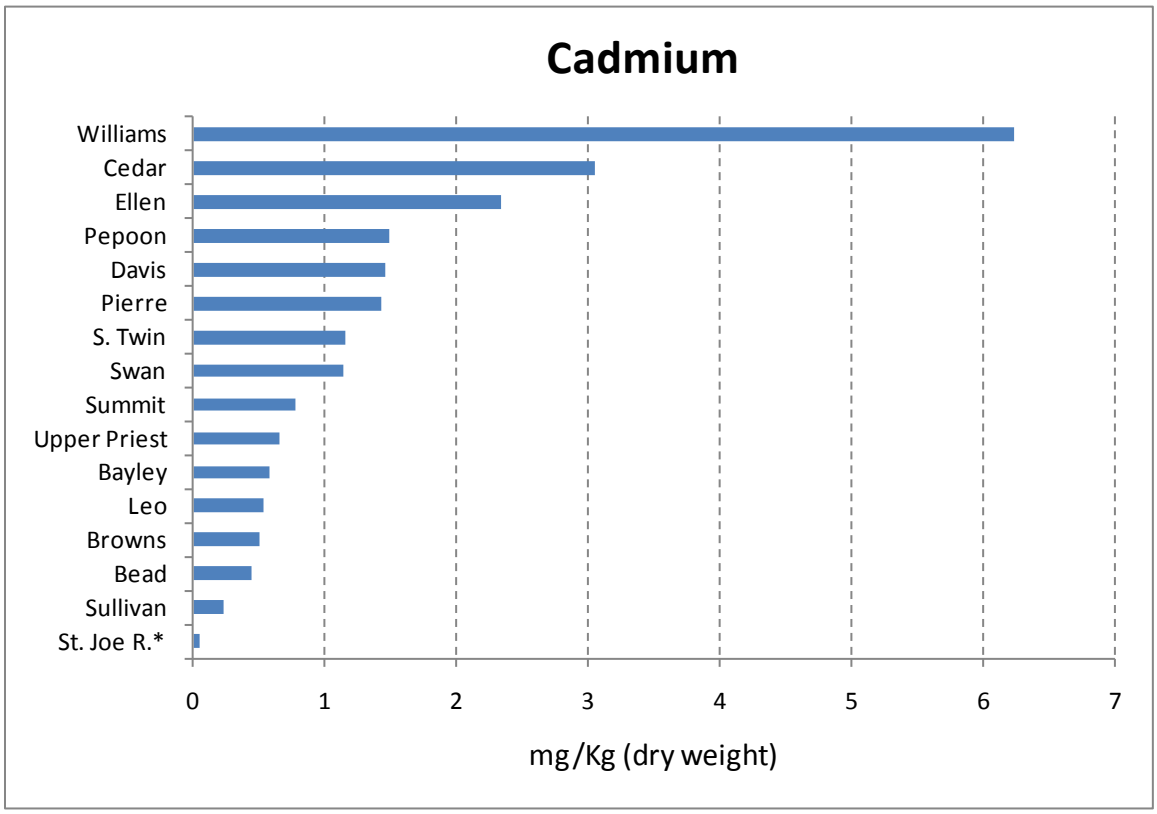


Figure 9. Waterbodies Ranked by Cadmium Concentrations in Sediment Samples.
**Not detected; plotted at the reporting limit (0.05 mg/Kg).*

Arsenic

The overall median and maximum concentrations of arsenic in the lake sediments were 8.0 and 28 mg/Kg, respectively (Table 13). The arsenic 90th percentile for the lakes was 13 mg/Kg. An arsenic concentration of 3.2 mg/Kg was measured for St. Joe River sediments.

Table 13. Summary Statistics for Arsenic in Sediment Samples (mg/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Median	8.0	11	4.5	3.2
Mean	9.2	12	4.7	- -
Minimum	3.2	7.1	3.2	- -
Maximum	28	28	6.1	- -
90th percentile	13	17	6.1	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.

Spatial patterns for arsenic (Figure 10) resembled antimony, lead, and cadmium, although western lakes were elevated to a somewhat lesser extent for arsenic. The median arsenic concentration in the western lakes (11 mg/Kg) was slightly more than twice the median in the eastern lakes (4.5 mg/Kg). Concentrations among eastern lakes were similar and uniformly low (3.2-6.1 mg/Kg), not much above sediments from the St. Joe River (3.2 mg/Kg). As with antimony, lead, and cadmium, the lakes fall into two distinct groups based on arsenic levels (Mann-Whitney test, $p<0.05$).

Although the highest arsenic concentrations were recorded for several of the northern-most lakes near the Columbia River – Williams (28 mg/Kg), Pierre (14 mg/Kg), and Cedar (12 mg/Kg) – a north-south gradient in concentrations was less clear. There was a broad area of elevated concentrations in the 8-14 mg/Kg range that included lakes from Cedar Lake near the international border to Swan Lake in the Sanpoil drainage. The lakes with the lowest arsenic concentrations were Leo, Bayley, and Sullivan at 3-4 mg/Kg.

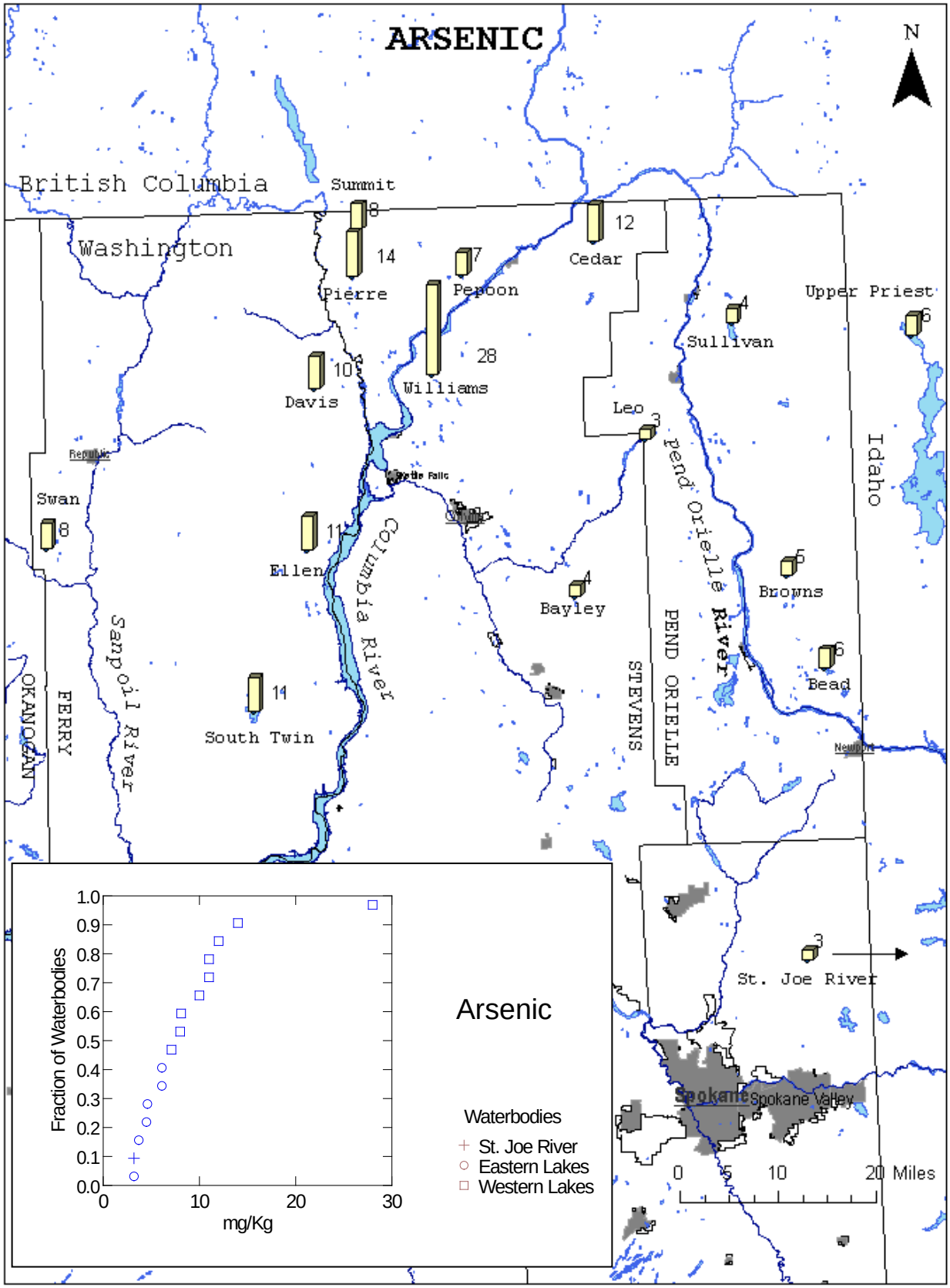


Figure 10. Spatial Patterns and Quantile Plot for Arsenic in Sediment Samples.

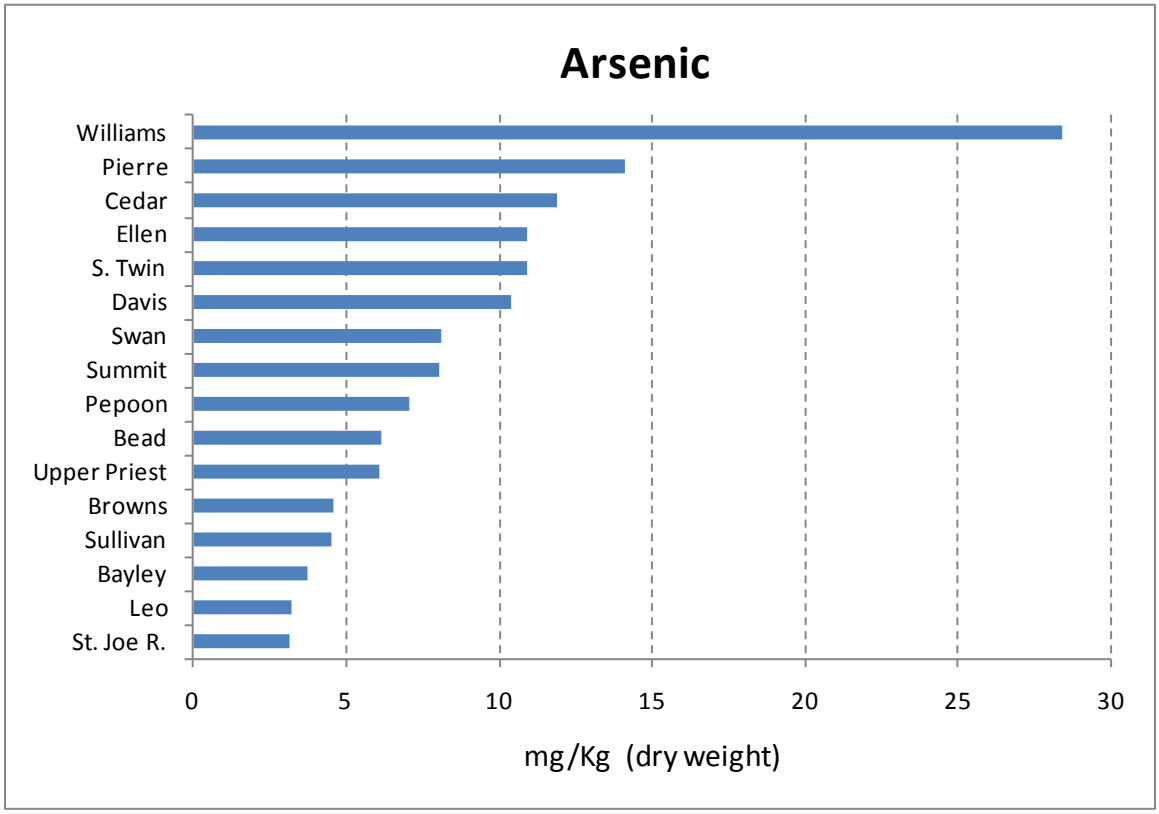


Figure 11. Waterbodies Ranked by Arsenic Concentrations in Sediment Samples.

Mercury

The overall median and maximum concentrations of mercury in the lake sediments were 0.099 and 0.208 mg/Kg, respectively (Table 14). The 90th percentile for the lakes was 0.200 mg/Kg. Mercury was below detection limits (< 0.007 mg/Kg) in the St. Joe River sediments.

Table 14. Summary Statistics for Mercury in Sediment Samples (mg/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Median	0.099	0.133	0.069	0.007 U
Mean	0.110	0.139	0.066	- -
Minimum	0.031	0.052	0.031	- -
Maximum	0.208	0.208	0.103	- -
90th percentile	0.200	0.203	0.092	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.
U: The analyte was not detected at or above the reported result.

Visually, the region of elevated mercury levels appears shifted further west than what was observed for antimony, lead, cadmium, or arsenic (Figure 12). The highest concentrations were found in Davis, Ellen, and Swan Lakes, west of the Columbia River. There was some overlap in concentrations among western and eastern lakes, although six out of nine western lakes still had the highest concentrations. Sullivan, Bead, Brown were the lowest eastern lakes. As with antimony, lead, cadmium, and arsenic, mercury concentrations were significantly higher in the western lakes (Mann-Whitney test, $p<0.05$).

Mercury differs from the other metals analyzed in being subject to complex biogeochemical cycling that can strongly affect its behavior in a lake. Logging and rainfall can greatly increase the net flux of mercury to a lake by increasing sedimentation rates (Furl et al., 2009). These types of factors may be at play in the apparent western shift of elevated mercury levels.

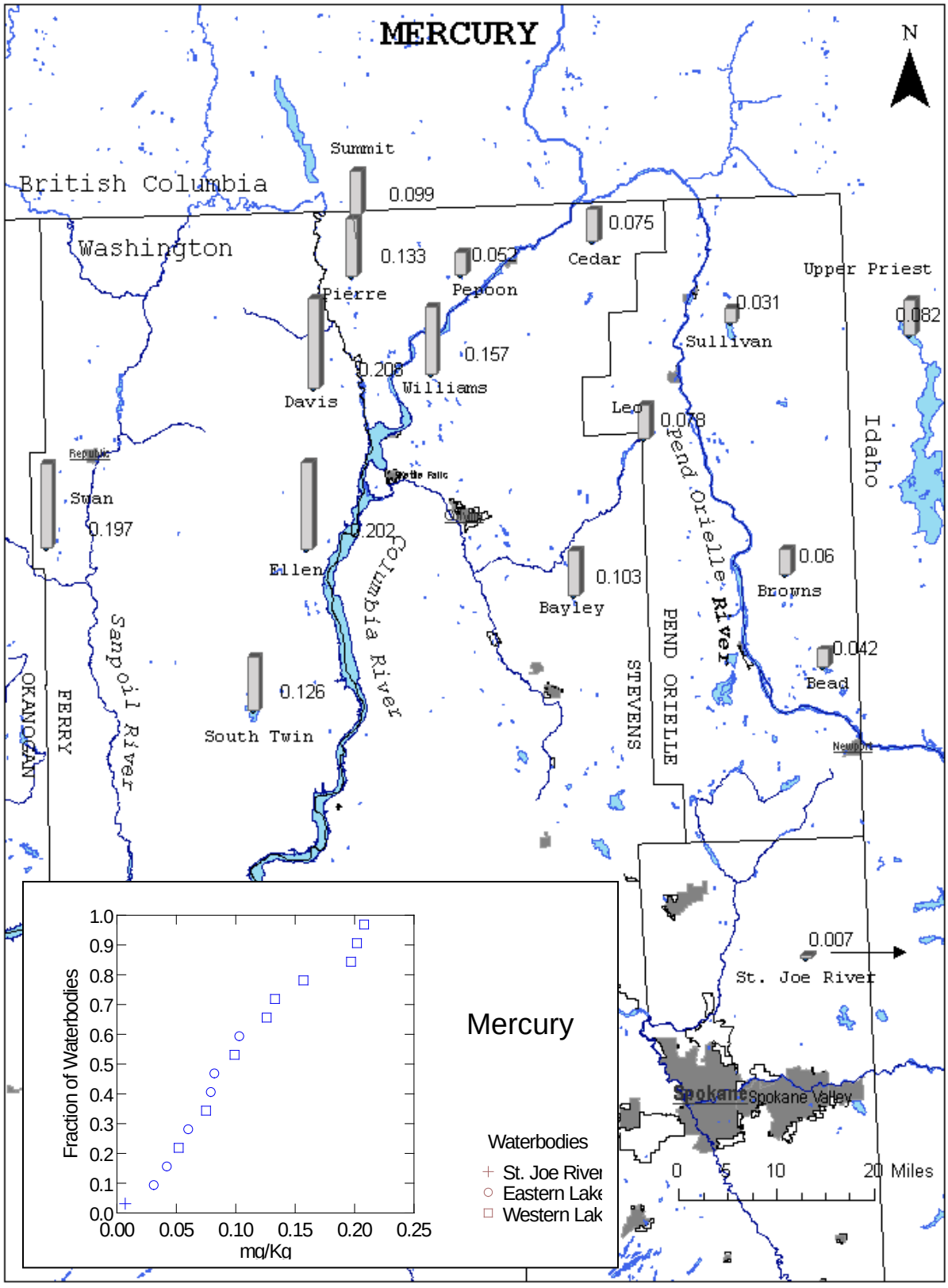


Figure 12. Spatial Patterns and Quantile Plot for Mercury in Sediment Samples.
St. Joe River plotted at the reporting limit; 0.007 mg/Kg.

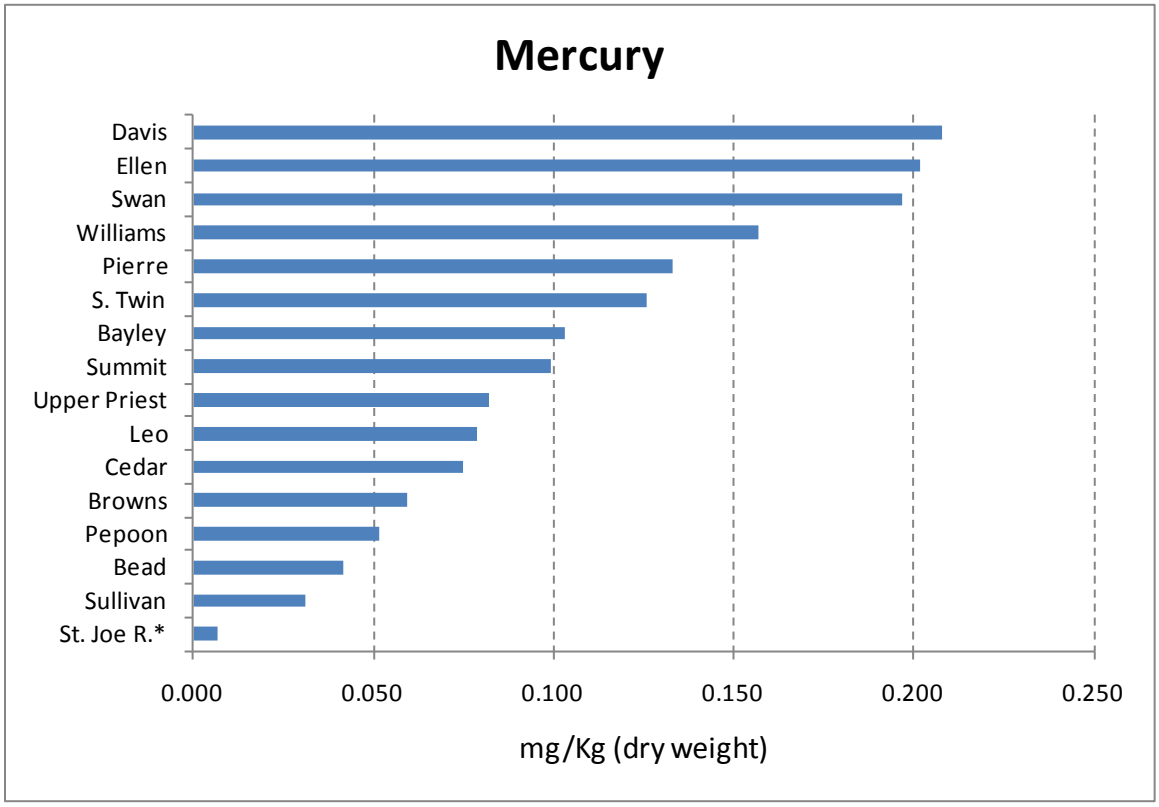


Figure 13. Waterbodies Ranked by Mercury Concentrations in Sediment Samples.
**Not detected; plotted at the reporting limit (0.007 mg/Kg).*

Zinc

The lakes had median and maximum zinc concentrations of 71 and 215 mg/Kg, respectively, with a 90th percentile of 137 mg/Kg (Table 15).

Table 15. Summary Statistics for Zinc in Sediment Samples (mg/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Median	71	76	63	19
Mean	84	91	72	
Minimum	20	20	50	- -
Maximum	215	215	117	- -
90th percentile	137	164	98	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.

Six of the eight highest zinc concentrations were recorded for the western lakes, with the highest concentrations in Williams and Cedar Lakes, 215 and 151 mg/Kg, respectively (Figure 14). As a group, however, the difference in zinc concentrations between western and eastern lakes was not statistically significant (Mann-Whitney test, p=0.60). Summit Lake and the St. Joe River had much lower zinc concentrations than the other waterbodies (19-20 vs. 46-215 mg/Kg).

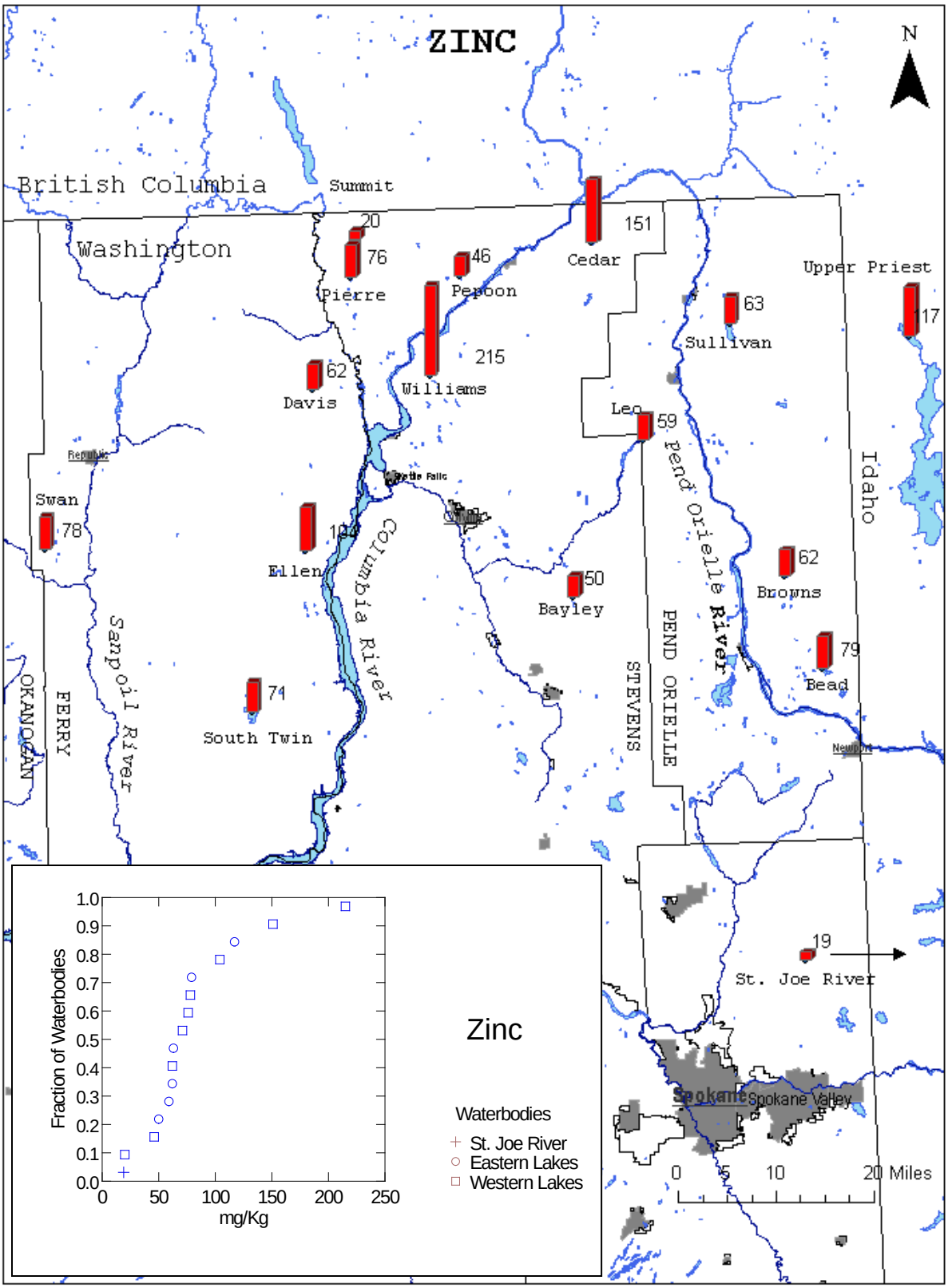


Figure 14. Spatial Patterns and Quantile Plot for Zinc in Sediment Samples.

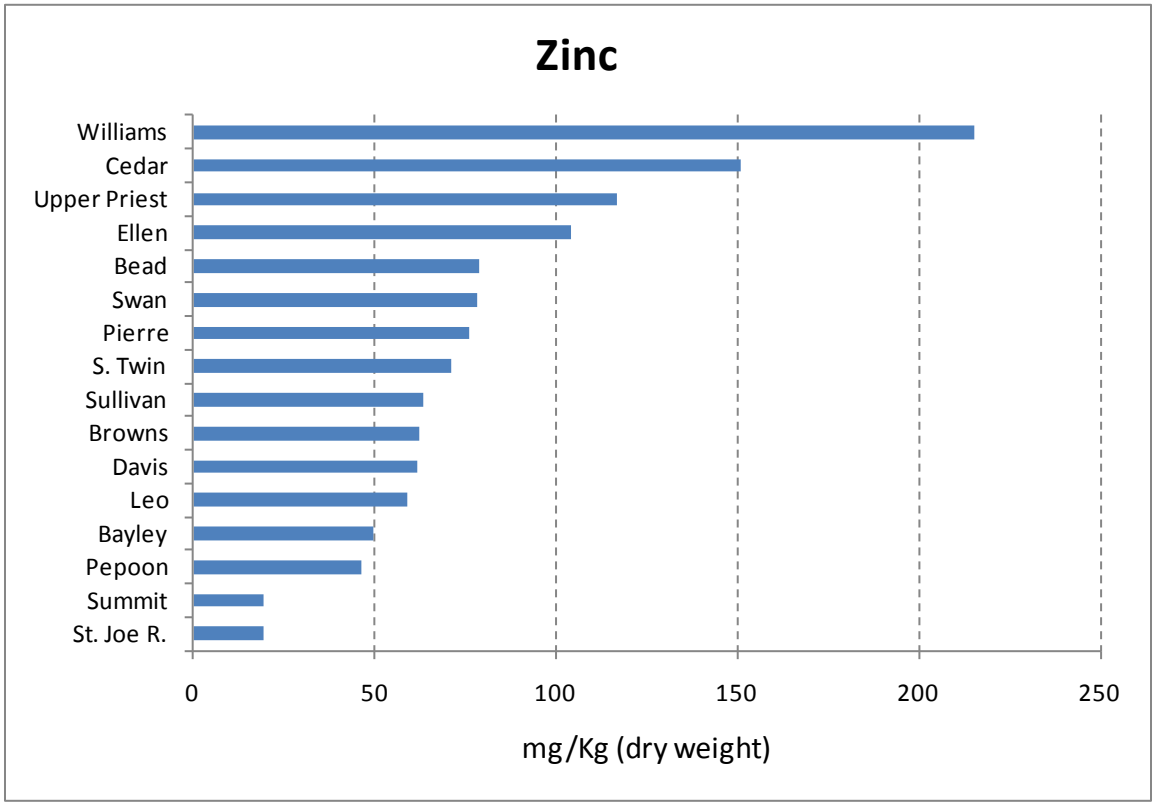


Figure 15. Waterbodies Ranked by Zinc Concentrations in Sediment Samples.

Barium, Chromium, Copper, Manganese, and Iron

Summary statistics and figures showing spatial patterns, data distribution, and waterbody rankings for barium, chromium, copper, manganese, and iron follow. These metals generally showed only modest variation across the study area. The data distributions indicate the western and eastern lakes form a common population with respect to these metals.

A few relatively elevated concentrations were observed, namely chromium and copper in Pierre Lake, and barium and manganese in Upper Priest Lake. Otherwise the results were unremarkable.

Barium

Table 16. Summary Statistics for Barium in Sediment Samples (mg/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Median	98	96	104	42
Mean	116	100	141	- -
Minimum	32	32	83	- -
Maximum	333	182	333	- -
90th percentile	165	128	237	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.

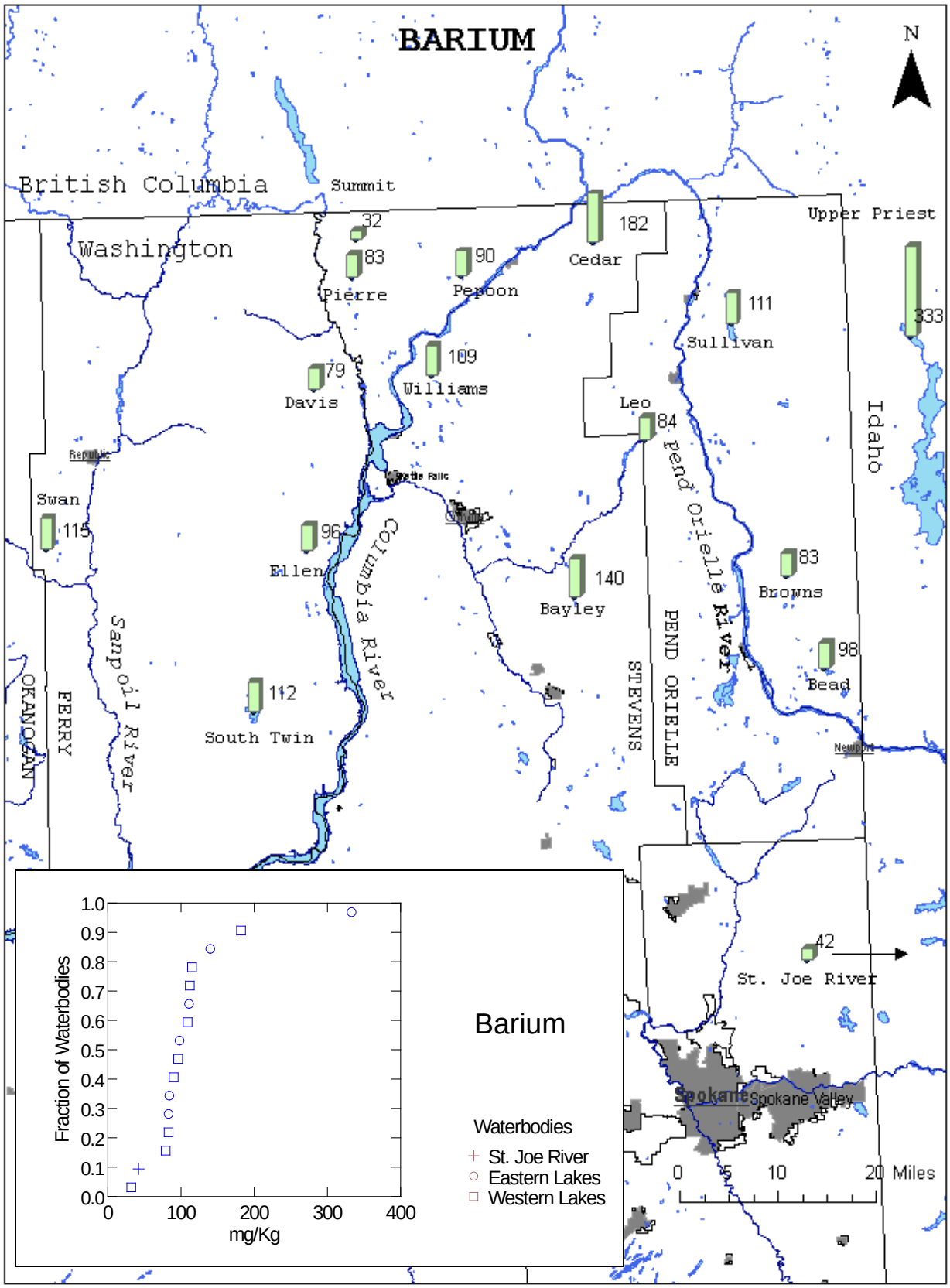


Figure 16. Spatial Patterns and Quantile Plot for Barium in Sediment Samples.

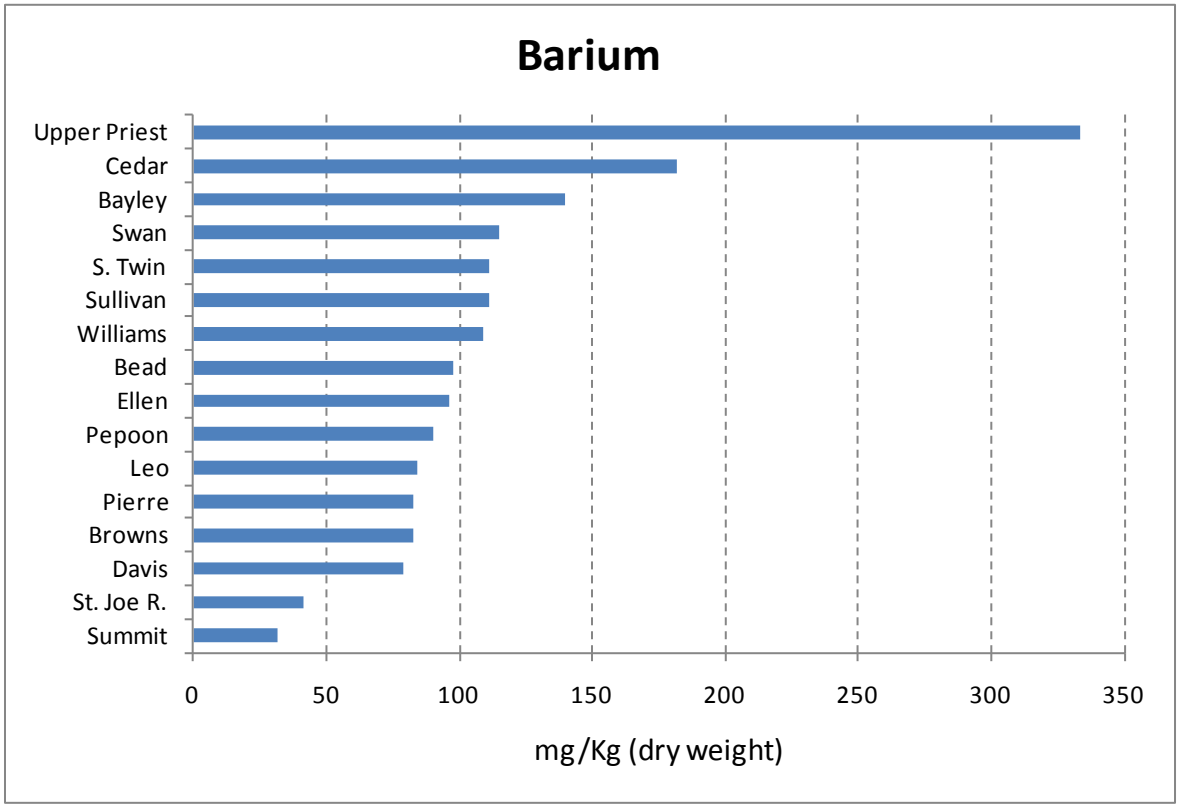


Figure 17. Waterbodies Ranked by Barium Concentrations in Sediment Samples.

Chromium

Table 17. Summary Statistics for Chromium in Sediment Samples (mg/Kg, dry weight)

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Median	13	8.9	14	8.6
Mean	15	15	14	- -
Minimum	3.8	3.8	9.5	- -
Maximum	53	53	21	- -
90th percentile	20	26	18	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.

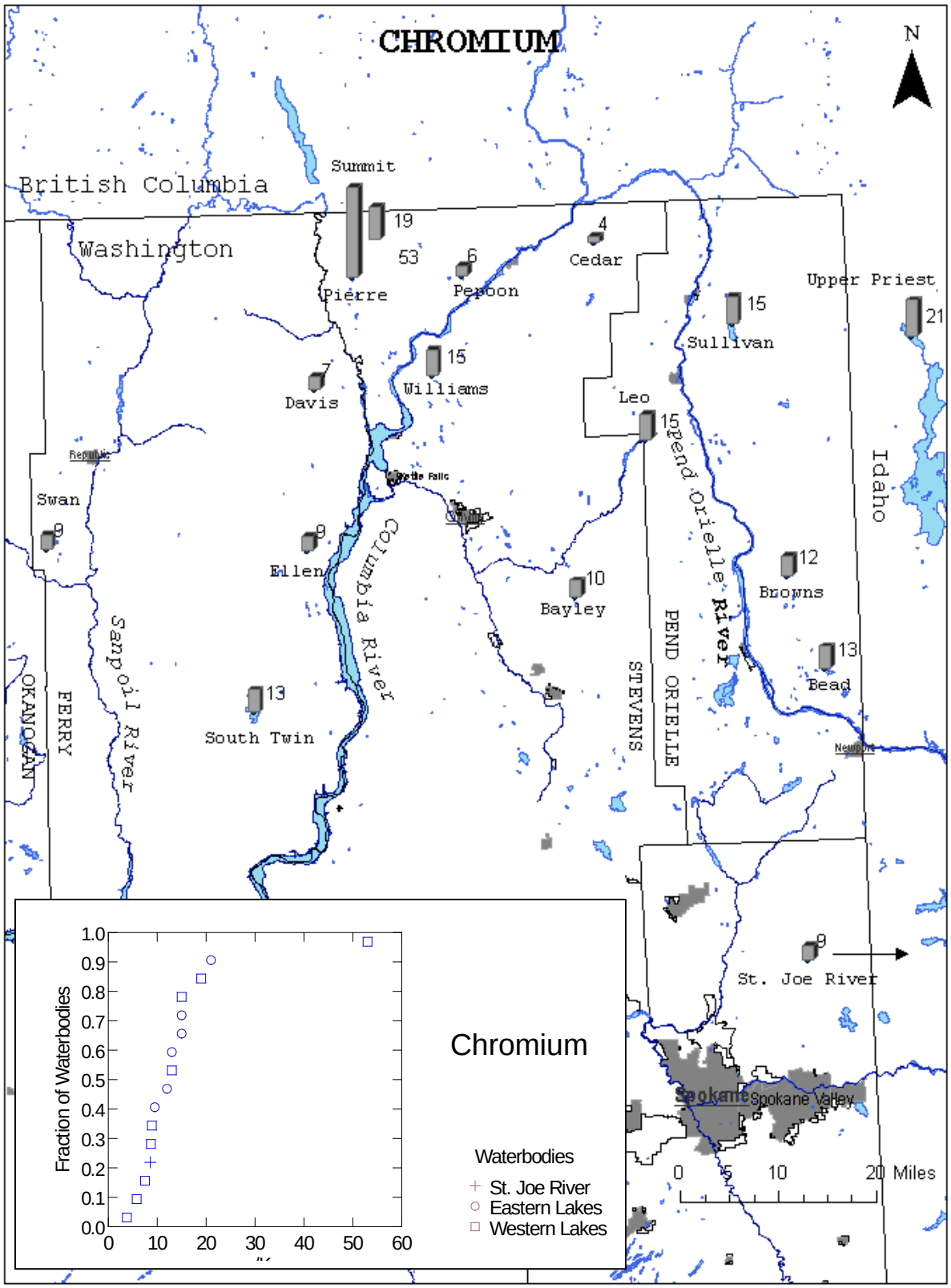


Figure 18. Spatial Patterns and Quantile Plot for Chromium in Sediment Samples.

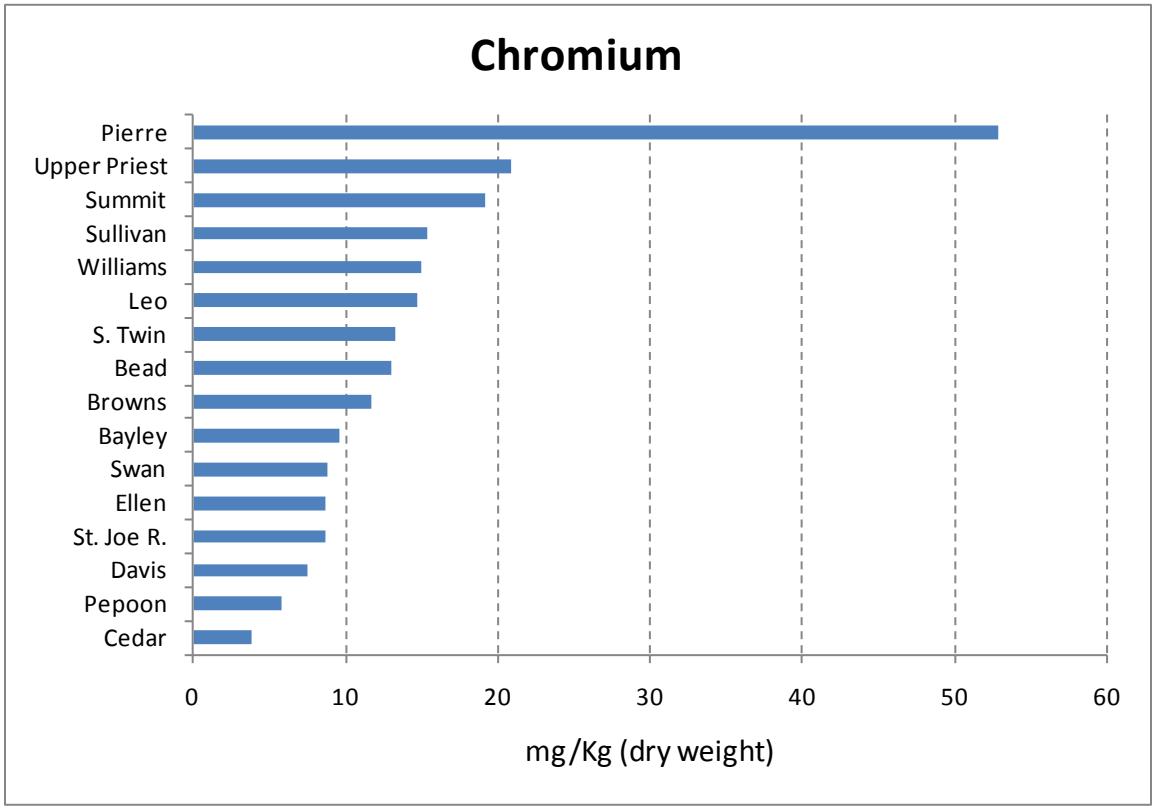


Figure 19. Waterbodies Ranked by Chromium Concentrations in Sediment Samples.

Copper

Table 18. Summary Statistics for Copper in Sediment Samples (mg/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Median	20	20	20	14
Mean	22	23	20	- -
Minimum	8.1	8.1	16	- -
Maximum	77	77	27	- -
90th percentile	26	36	25	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.

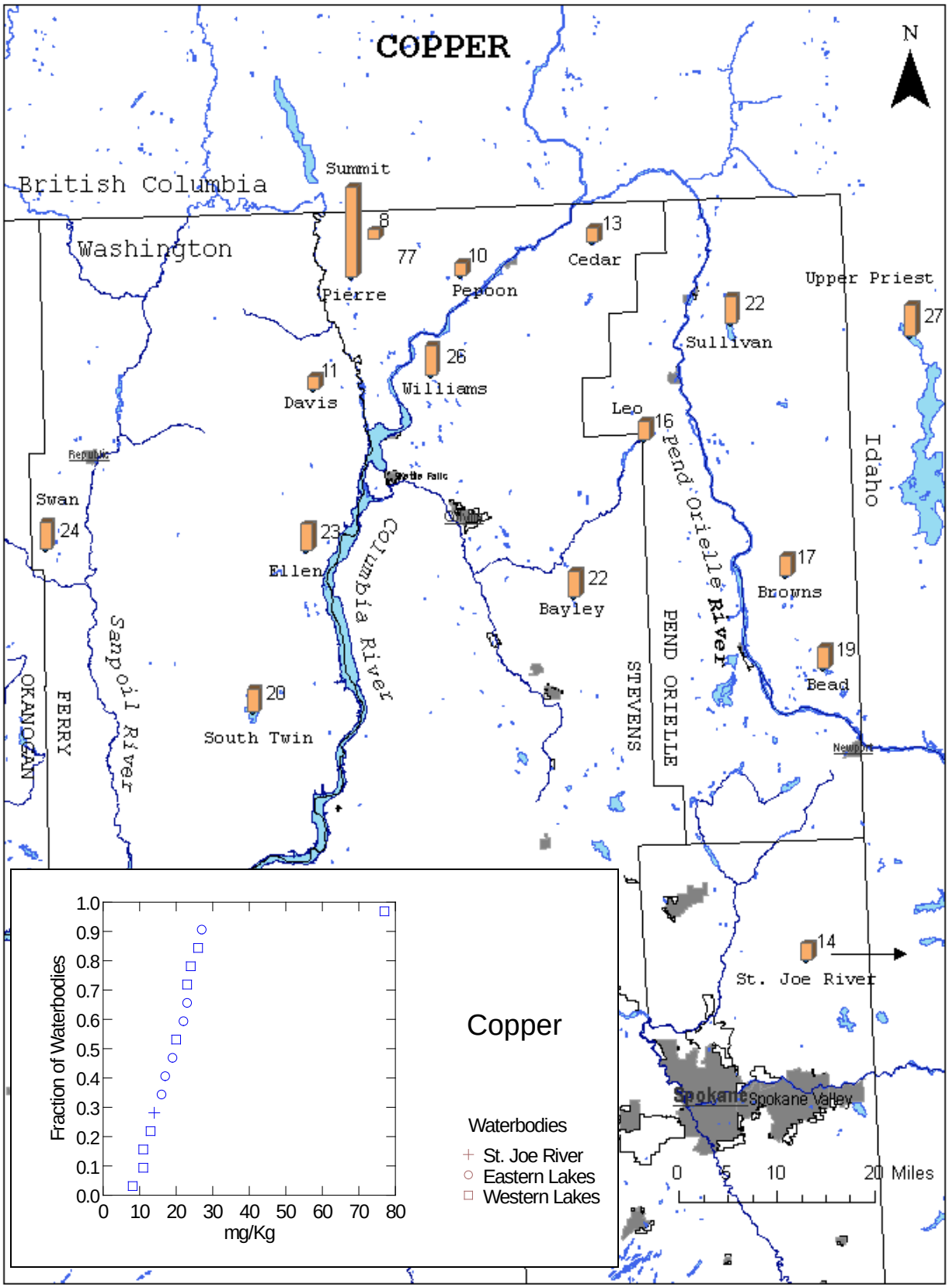


Figure 20. Spatial Patterns and Quantile Plot for Copper in Sediment Samples.

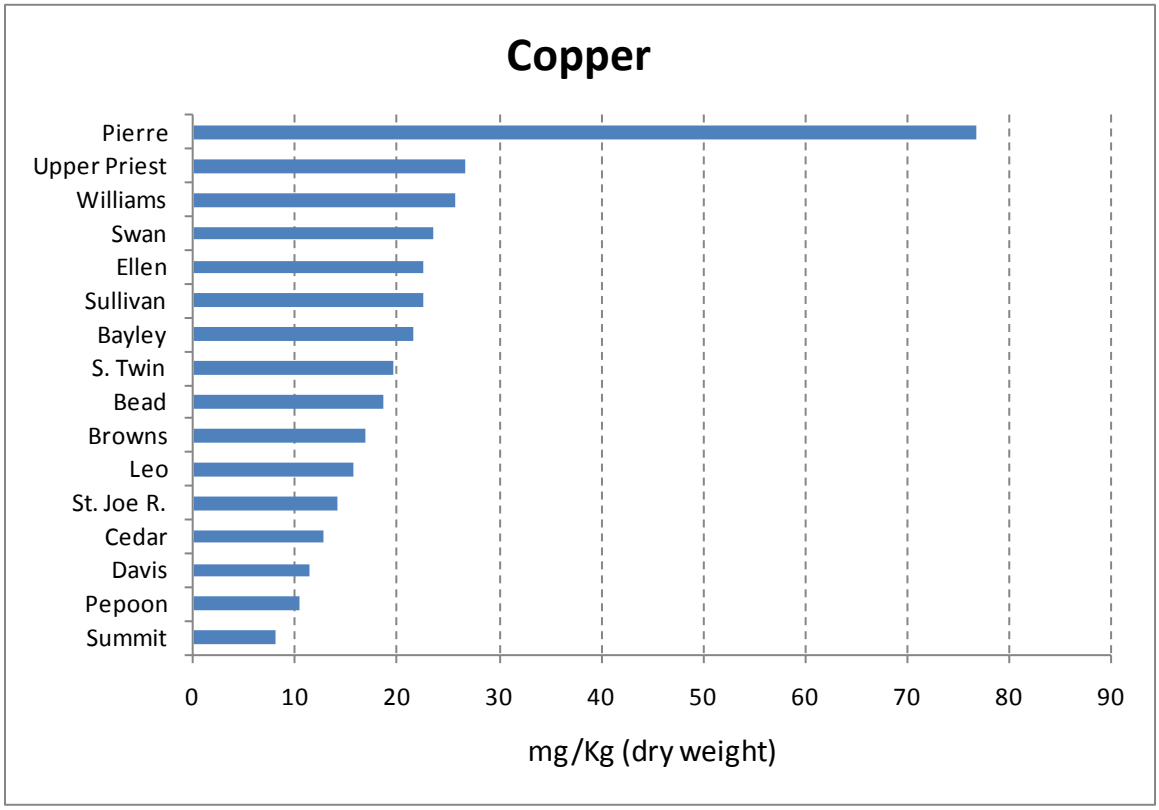


Figure 21. Waterbodies Ranked by Copper Concentrations in Sediment Samples.

Manganese

Table 19. Summary Statistics for Manganese in Sediment Samples (mg/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Median	243	228	284	146
Mean	466	292	727	- -
Minimum	69	69	103	- -
Maximum	3,010	885	3,010	- -
90th percentile	721	505	1,743	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.

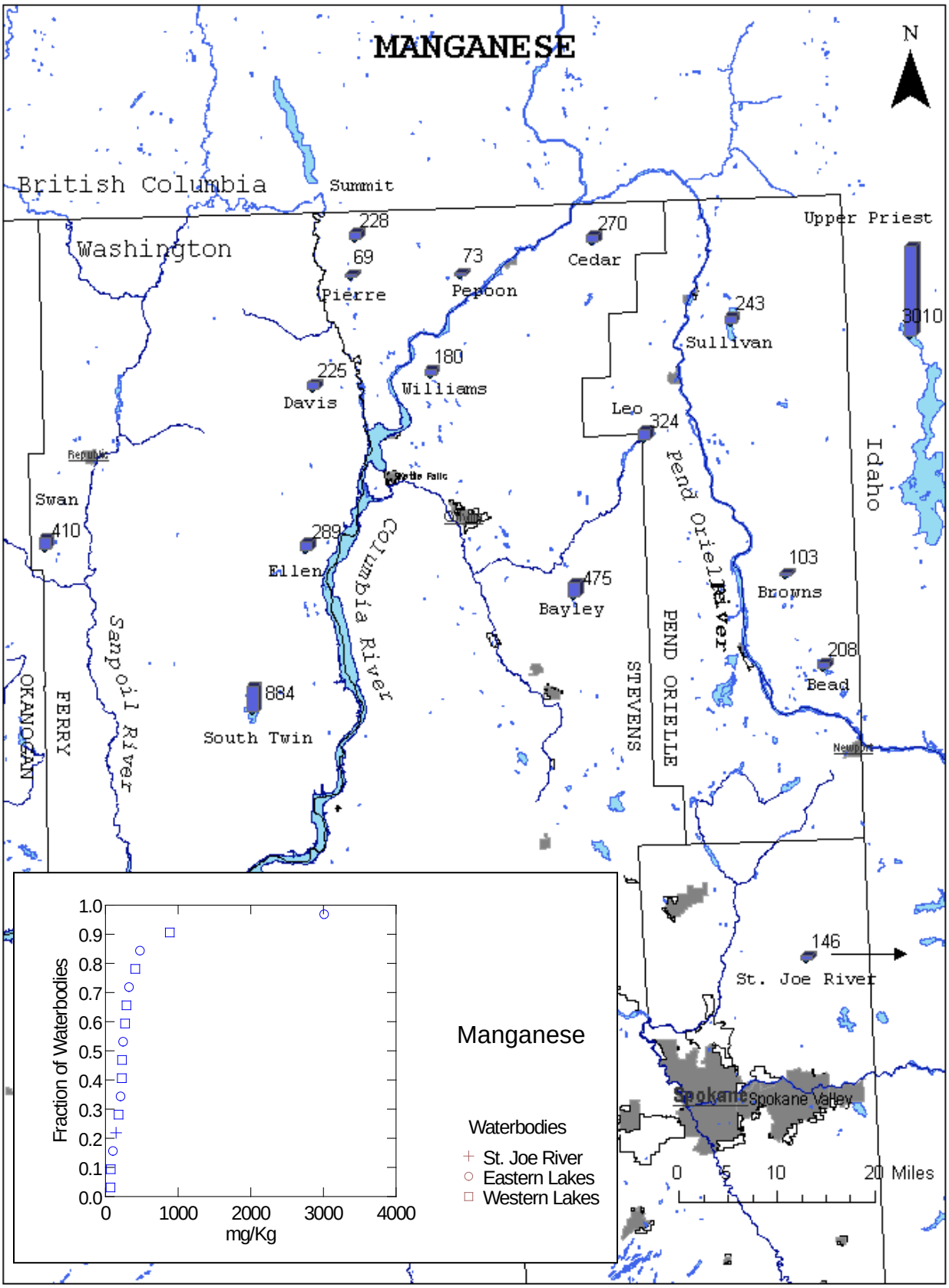


Figure 22. Spatial Patterns and Quantile Plot for Manganese in Sediment Samples.

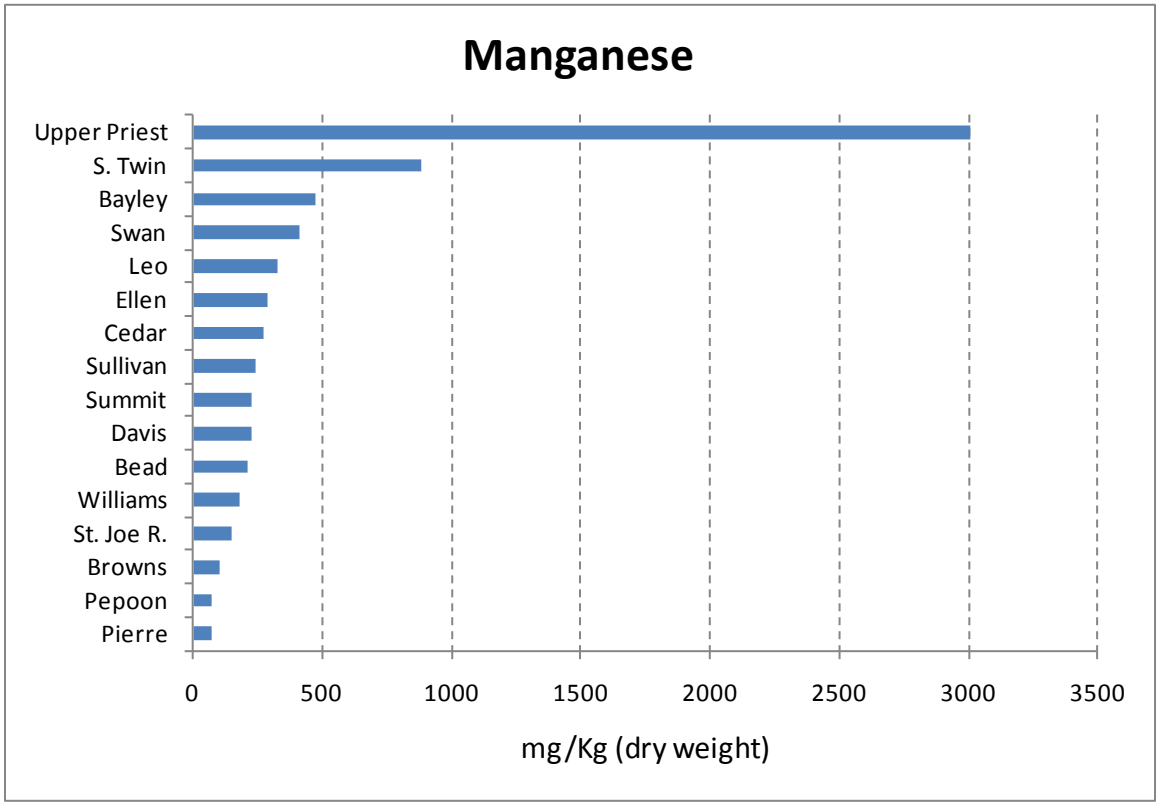


Figure 23. Waterbodies Ranked by Manganese Concentrations in Sediment Samples.

Iron

Table 20. Summary Statistics for Iron in Sediment Samples (mg/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Median	16,300	15,900	16,700	16,700
Mean	17,096	14,527	20,950	- -
Minimum	2,780	2,780	11,300	- -
Maximum	43,500	35,700	43,500	- -
90th percentile	30,820	23,060	33,500	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.

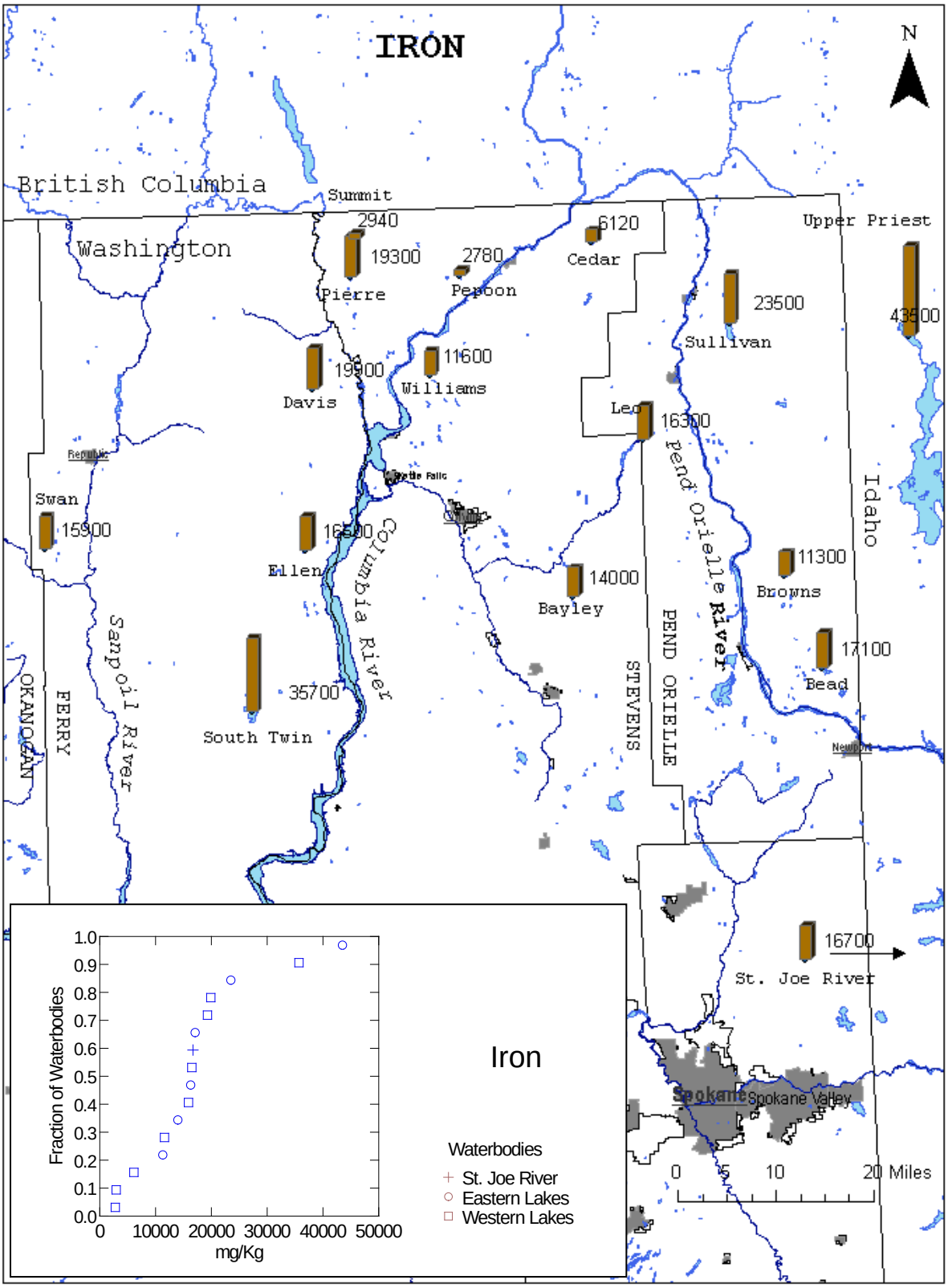


Figure 24. Spatial Patterns and Quantile Plot for Iron in Sediment Samples.

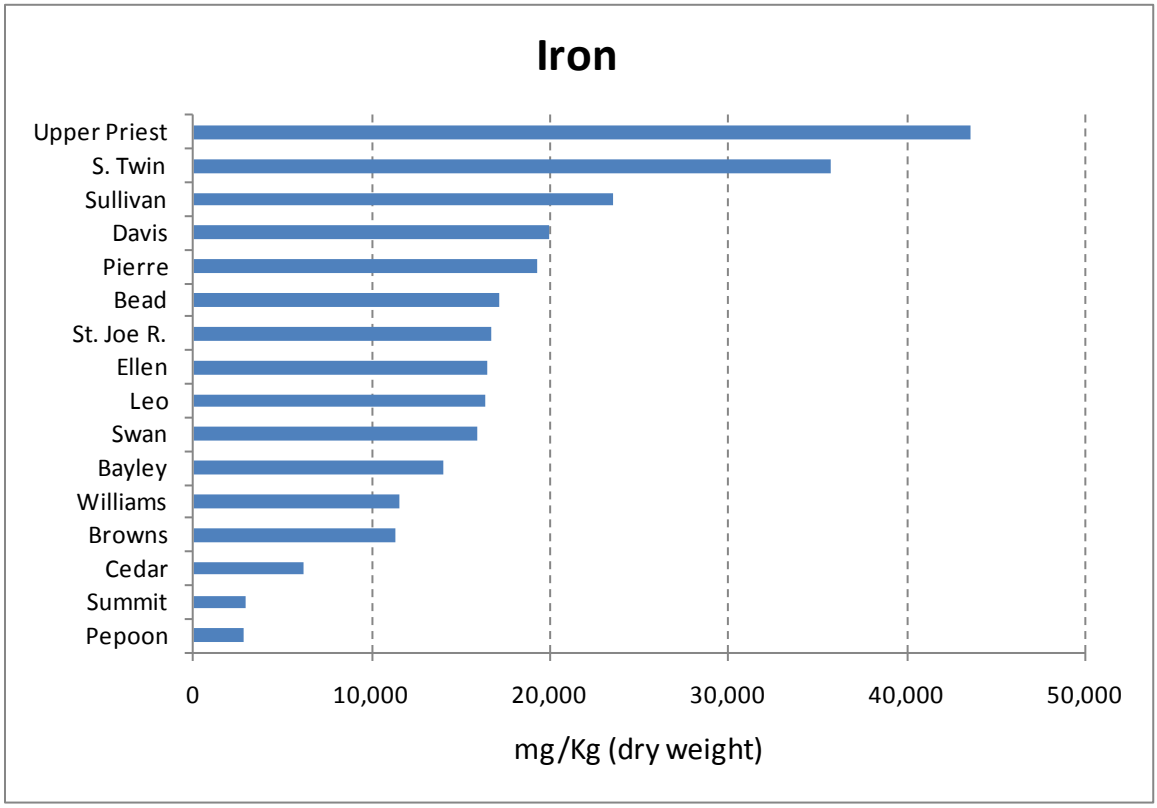


Figure 25. Waterbodies Ranked by Iron Concentrations in Sediment Samples.

Organic Compound Concentrations and Spatial Patterns

A range of halogenated organic compounds was also analyzed in the sediment samples. The analyses included 209 PCBs, 7 PCDDs, 10 PCDFs, and 36 PBDEs. The results are summarized in Table 21.

Definitions of previously unspecified chemical names in Table 21 are as follows:

- Total PCBs is the summed concentrations of the individual PCB compounds (congeners) detected.
- TEQ is the Toxicity Equivalent of a mixture of polychlorinated dioxins and furans or dioxin-like PCBs relative to TCDD, the most toxic of these compounds. TEQs were calculated using the human and mammalian Toxic Equivalency Factors (TEFs) which have been established for the seventeen 2,3,7,8-substituted dioxins and furans, and the dioxin-like PCBs[4] (Van den Berg et al., 2006; see Appendix B in the present report). The TEQ of a sample equals the sum of each congener concentration multiplied by its TEF. Non-detects were set at zero for Table 21.
- Total PBDEs is the summed concentration of the individual PBDE congeners detected. TEQs have not been established for PBDEs.

PCBs and PBDEs were detected in sediments from all 16 waterbodies. 2,3,7,8-TCDD was detected at about half the sites, with at least a subset of other 2,3,7,8-substituted PCDDs/PCDFs being found at all sites. Results for 2,3,7,8-substituted PCDDs/PCDFs and dioxin-like PCBs are summarized in terms of TEQs. PCB TEQs were insignificant compared to TCDD TEQs, being lower by several orders of magnitude.

The data were examined for correlations among the compounds analyzed and with the TOC content of the sediments. Hydrophobic compounds like these preferentially sorb to organic matter, potentially influencing their distribution patterns. Significant correlations, however, were not found.

A discussion of findings for each chemical or chemical group follows.

[4] Thirteen of the 209 PCB congeners with four or more lateral chlorines and with one or no substitution at the ortho (inner) position have dioxin-like properties.

Table 21. Summary of Results for Organic Compounds Analyzed in Sediment Samples (ng/Kg, dry weight; parts per trillion).

Waterbody	Total PCBs		PCB TEQs		TCDD		TCDD TEQs		Total PBDEs	% TOC
Washington										
Swan Lake	3,009		0.007		0.22		2.0		853	18
Cedar Lake	311		0.001		0.11		0.55		541	16
Pepoon Lake	298		0.001		0.063	U	0.21		101	17
Summit Lake	1,766		0.005		0.23	NJ	1.9		795	13
Pierre Lake	2,591		0.008		0.095	U	3.0		526	13
Williams Lake	8,335		0.43		0.10	U	3.9		685	16
Davis Lake	2,687		0.005		0.56	NJ	2.7		1,946	24
Ellen Lake	4,825		0.013		0.53	NJ	3.8		1,338	19
S. Twin Lake	4,389		0.013		0.32	NJ	4.8		417	14
Sullivan Lake	112		0.000		0.053	NJ	0.20		126	3.5
Leo Lake	3,807		0.016		0.057	UJ	5.3		612	14
Browns Lake	2,855		0.012		0.038	U	0.78		1,190	6.6
Bayley Lake	503		0.002		0.051	U	1.5		1,288	22
Bead Lake	2,260		0.008		0.14	NJ	2.6		304	6.8
Idaho										
Upper Priest Lake	286		0		0.042	UJ	0.55		273	4.2
St. Joe River	7		0		0.032	NJ	0.11		149	1.1

U: The analyte was not detected above the reported quantitation limit.
J: The analyte was positively identified; the associated numerical value is the approximate concentration.
NJ: The analyte has been “tentatively identified”; the associated numerical value is the approximate concentration.
UJ: The analyte was not detected above the reported estimated quantitation limit.

PCBs

Total PCBs in the lake sediments ranged from 112 ng/Kg (parts per trillion) in Sullivan Lake to 8,335 ng/Kg in Williams Lake (Table 22). The overall median for the lakes was 2,591 ng/Kg, with a 90th percentile of 4,651 ng/Kg. Total PCB in the St. Joe River sediments was only 6.8 ng/Kg.

Table 22. Summary Statistics for Total PCBs in Sediment Samples (ng/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Detection Freq.	100%	100%	100%	100%
Median	2,591	2,687	1,381	6.8
Mean	2,536	3,135	1,637	- -
Minimum	112	298	112	- -
Maximum	8,335	8,335	3,807	- -
90th percentile	4,651	5,527	3,331	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.

Spatial patterns and data distribution for PCBs are shown in Figure 26. Although the three highest total PCB concentrations were recorded in Williams, Ellen, and South Twin (8,335, 4,825, and 4,389 ng/Kg, respectively), the western lakes as a group did not exhibit consistently elevated concentrations and were not significantly different from the eastern lakes (Mann-Whitney test, $p>0.05$). PCB concentrations in Williams Lake were two or more times higher than in all other lakes.

A majority of lakes (ten) had total PCB concentrations in the approximate range of 1,800 – 4,800 ng/Kg (Figure 27.) These lakes lie along the Columbia and Pend Oreille Rivers suggesting a relationship with more highly populated areas. A second group of five lakes – Sullivan, Upper Priest, Cedar, Pepoon, and Bayley – had much lower PCB concentrations of approximately 100 - 500 ng/Kg. These lakes are located in more remote parts of the study area, four of the five being in the far northeast corner.

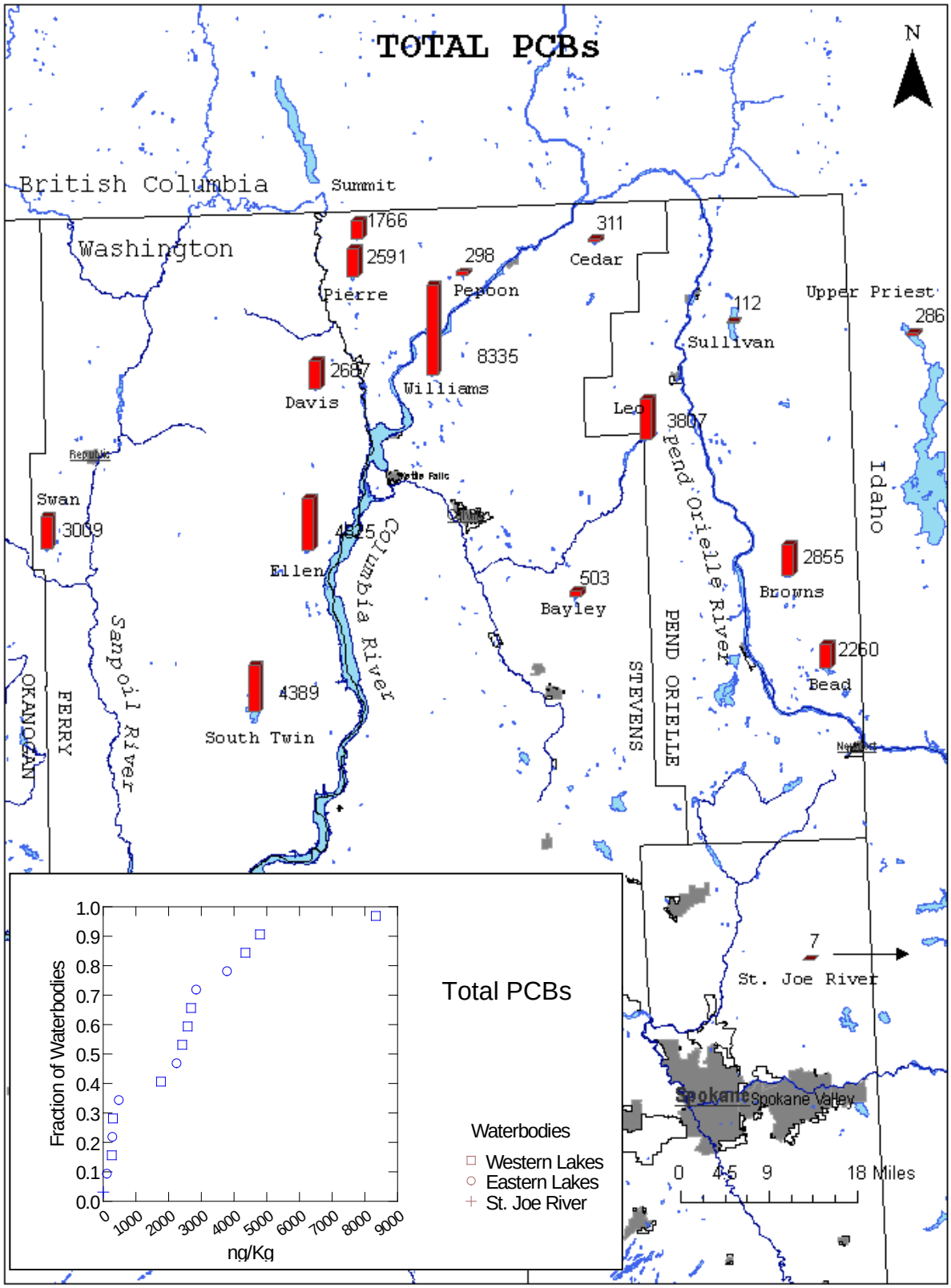


Figure 26. Spatial Patterns and Quantile Plot for Total PCBs in Sediment Samples.

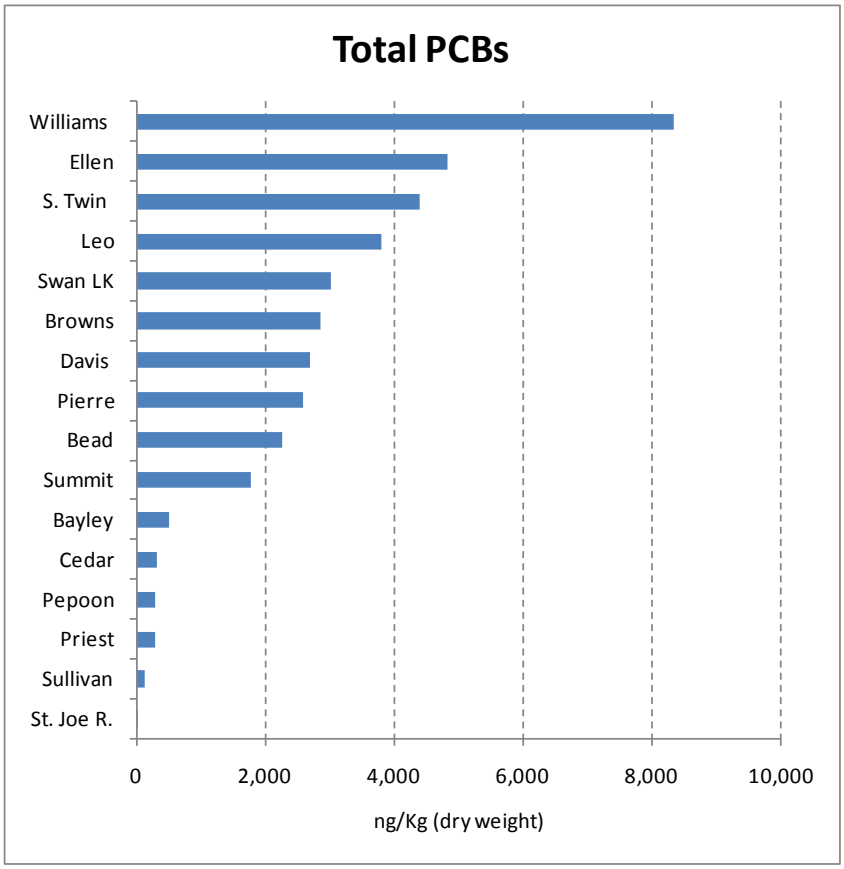


Figure 27. Waterbodies Ranked by Total PCB Concentration in Sediment Samples.

PCB TEQs

As already noted, PCB TEQs were low in most sediment samples (Table 23). All but one lake, Williams, were at or below about 0.02 ng/Kg (Figures 28 and 29). The relatively high PCB TEQ in Williams Lake, 0.43 ng/Kg, was due to a single congener, PCB-126, with a concentration of 4 ng/Kg. PCB-126 has a TEF of 0.1 (one tenth the toxicity of TCDD). PCB-126 was not detected in the other lakes.

Table 23. Summary Statistics for PCB TEQs in Sediment Samples (ng/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Median	0.0071	0.0071	0.0049	0
Mean	0.035	0.054	0.0063	- -
Minimum	0	0.0013	0	- -
Maximum	0.43	0.43	0.016	- -
90th percentile	0.015	0.096	0.014	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.

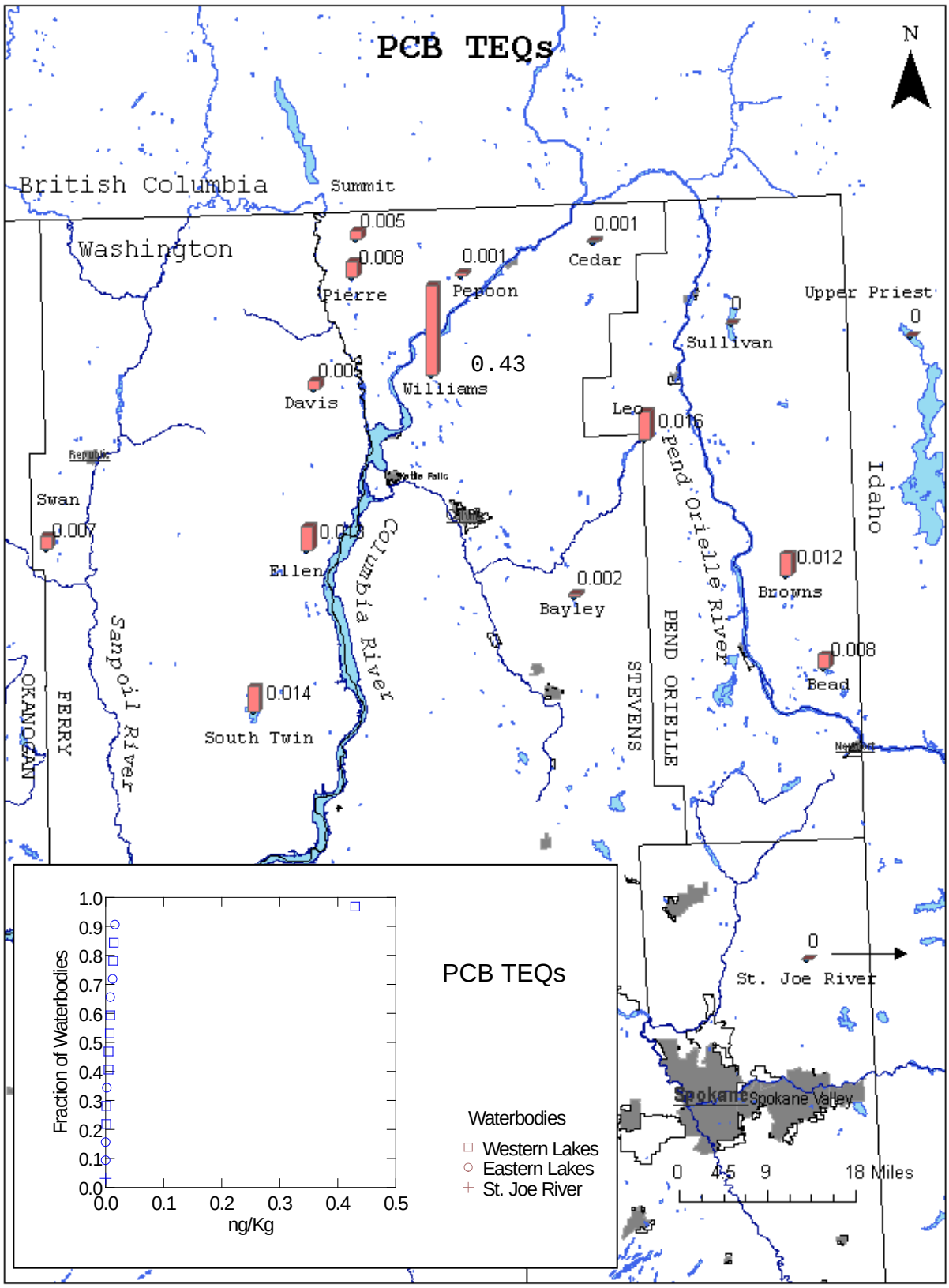


Figure 28. Spatial Patterns and Quantile Plot for PCB TEQs in Sediment Samples.

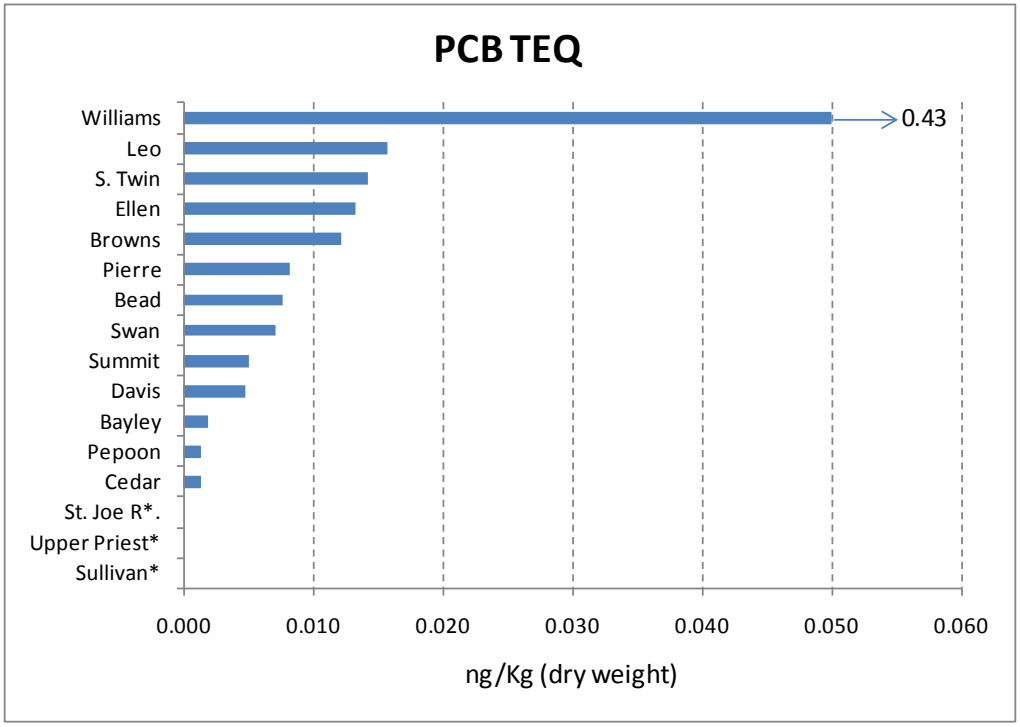


Figure 29. Waterbodies Ranked by PCB TEQ Concentrations in Sediment Samples.
**PCB TEQ = 0.*

TCDD

TCDD was detected in about half the waterbodies (Table 24). Detection was more frequent in the western lakes than the eastern lakes (67% vs. 33%).

The overall median and 90th percentile concentrations for the lakes were 0.098 and 0.50 ng/Kg, respectively. The western lakes median was 0.22 ng/Kg vs. less than 0.052 ng/Kg (non-detect) for the eastern lakes. TCDD was found at a low concentration of 0.032 ng/Kg in the St. Joe River.

Table 24. Summary Statistics for TCDD in Sediment Samples (ng/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Detection Freq.	53%	67%	33%	100%
Median	0.098	0.22	0.052 U	0.032
Mean	0.17	0.25	0.057	- -
Minimum	.021 U	0.063	0.038 U	- -
Maximum	0.56	0.56	0.098	- -
90th percentile	0.50	0.54	0.078	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.
U: Not detected at or above the reported sample quantitation limit.

As shown in Figure 30, TCDD was significantly higher in the western lakes (Mann-Whitney test, $p<0.05$; quantitation limit used for non-detects; St. Joe River excluded.) The highest concentrations tended to occur in lakes west of the Columbia River. There was no evidence of increasing TCDD concentrations moving up the Columbia River toward the Canadian border, as was observed for certain metals. The lakes with the highest TCDD concentrations were Davis, Ellen, and South Twin, 0.56, 0.53, and 0.32 ng/Kg, respectively.

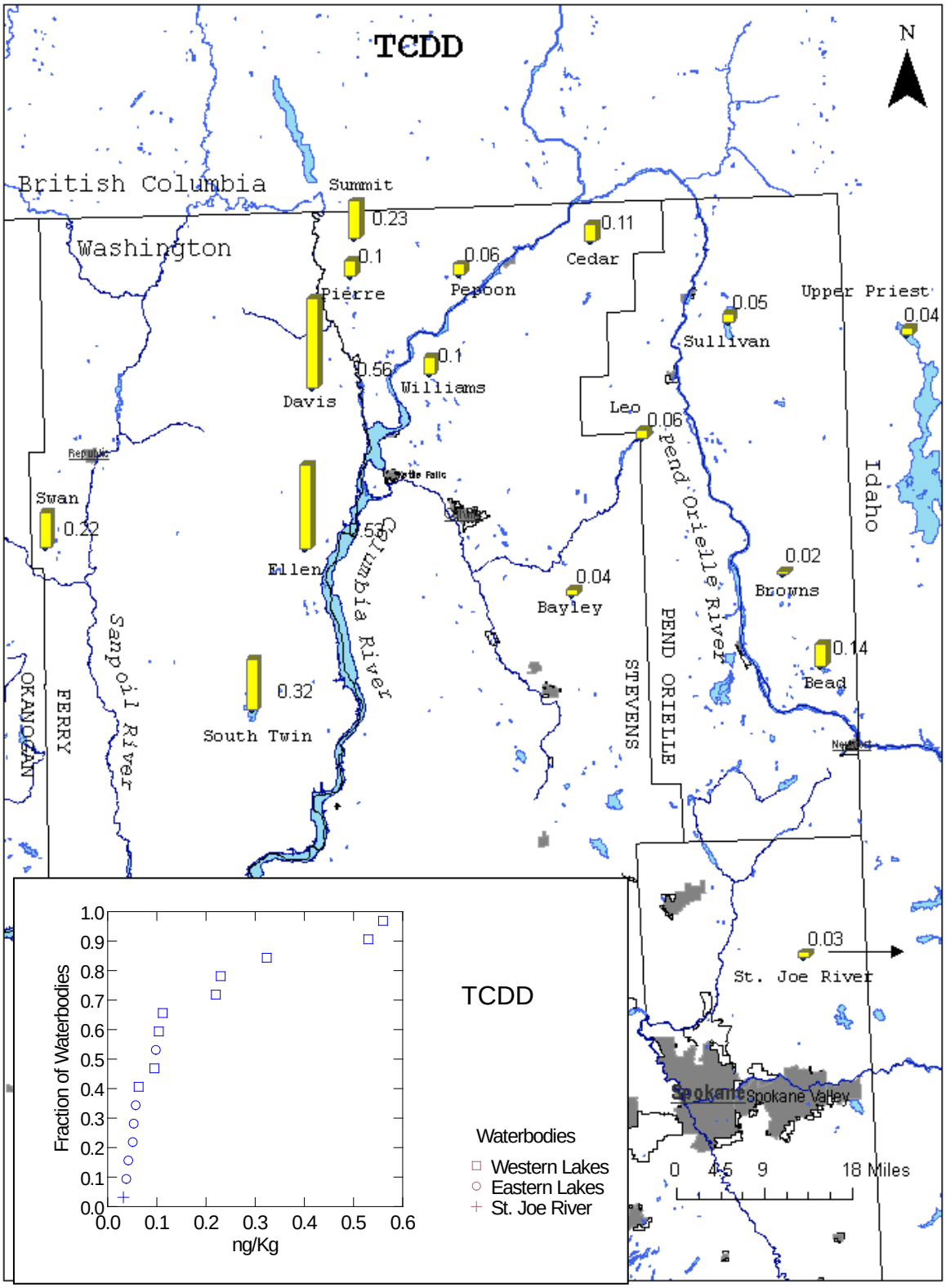


Figure 30. Spatial Patterns and Quantile Plot for TCDD in Sediment Samples.

Pepoon, Pierre, Williams, Leo, Browns, Bayley, and Upper Priest lakes plotted at the quantitation limit, 0.10-0.038 ng/Kg.

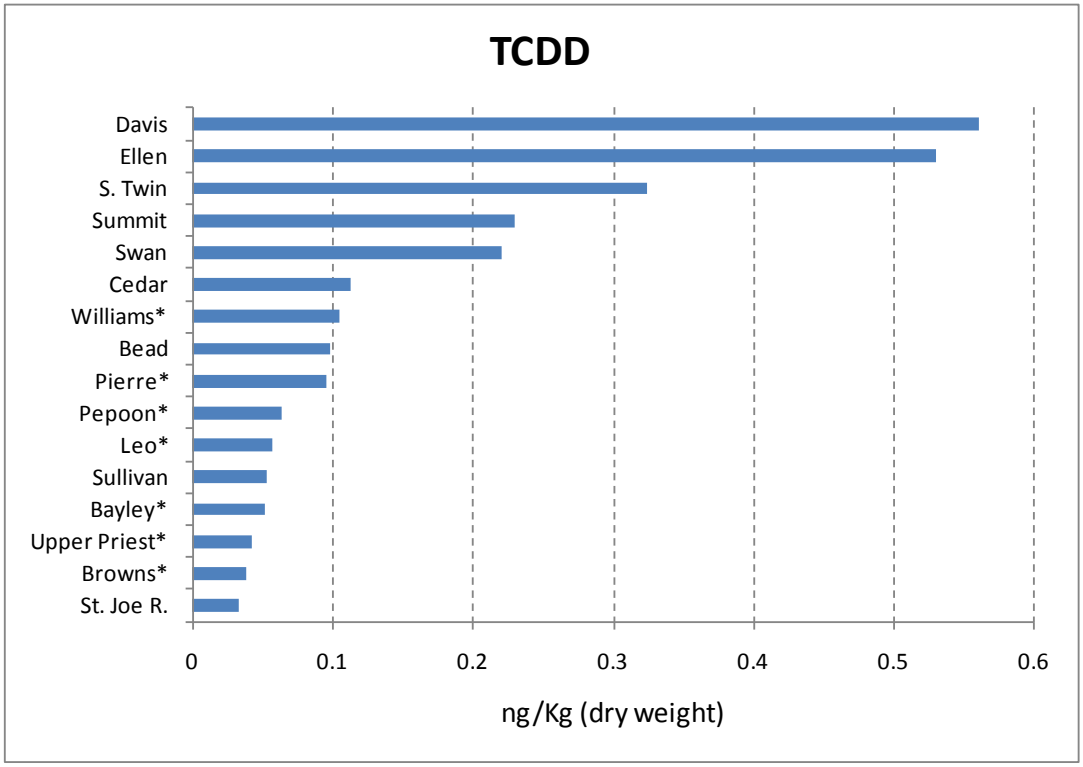


Figure 31. Waterbodies Ranked by TCDD Concentrations in Sediment Samples.
**Not detected; plotted at the quantitation limit (0.03 – 0.10 ng/Kg).*

TCDD TEQs

The TCDD TEQ ranged from 0.20 to 5.3 ng/Kg in the lakes and was at 0.11 ng/Kg in the St. Joe River (Table 25). Concentrations ranges were similar between western and eastern lakes (Figure 32).

Table 25. Summary Statistics for TCDD TEQs in Sediment Samples (ng/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Median	2.0	2.7	1.1	0.11
Mean	2.3	2.5	1.8	- -
Minimum	0.20	0.21	0.20	- -
Maximum	5.3	4.8	5.3	- -
90th percentile	4.6	4.1	3.9	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.

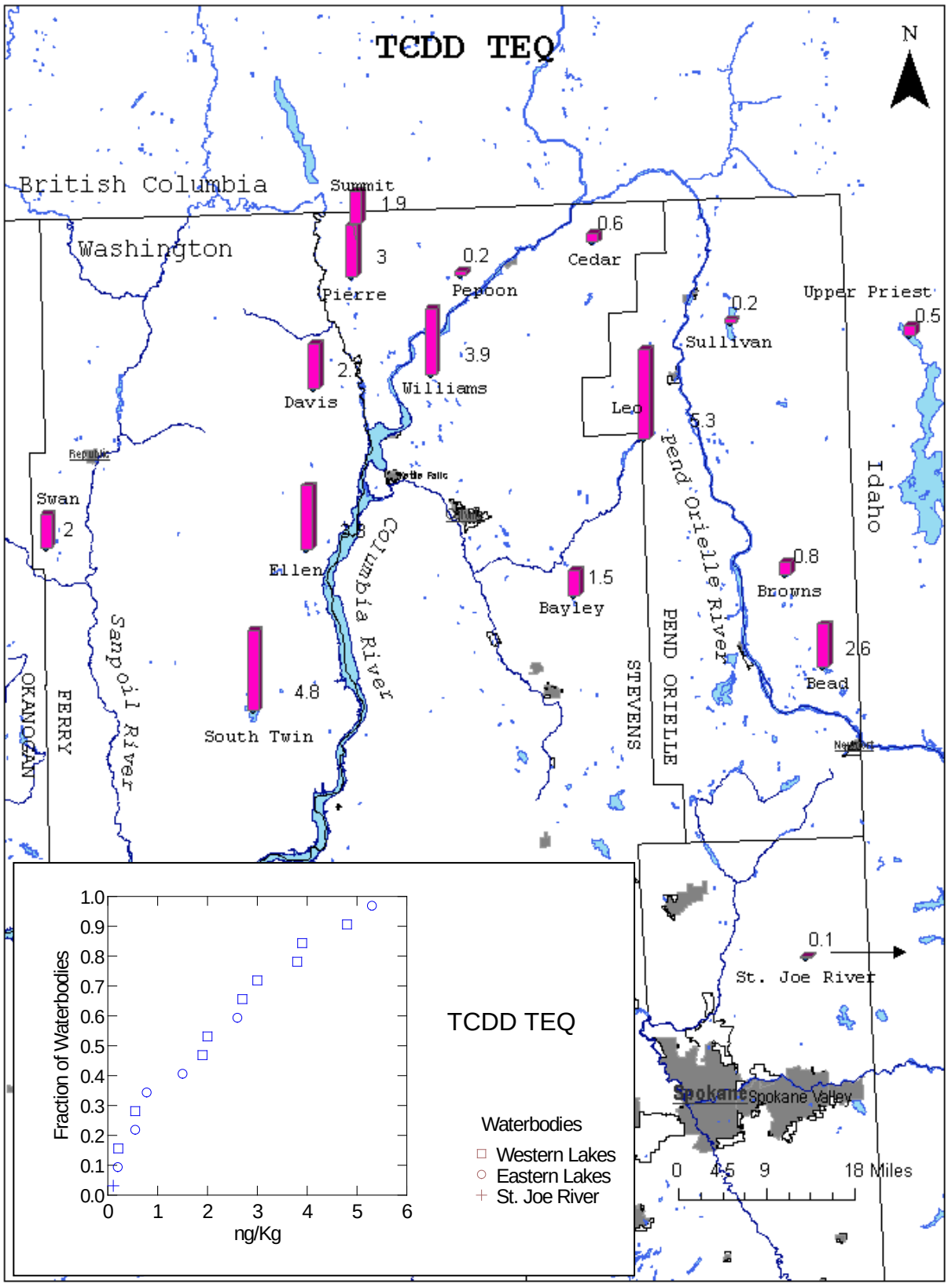


Figure 32. Spatial Patterns and Quantile Plot for TCDD TEQs in Sediment Samples.

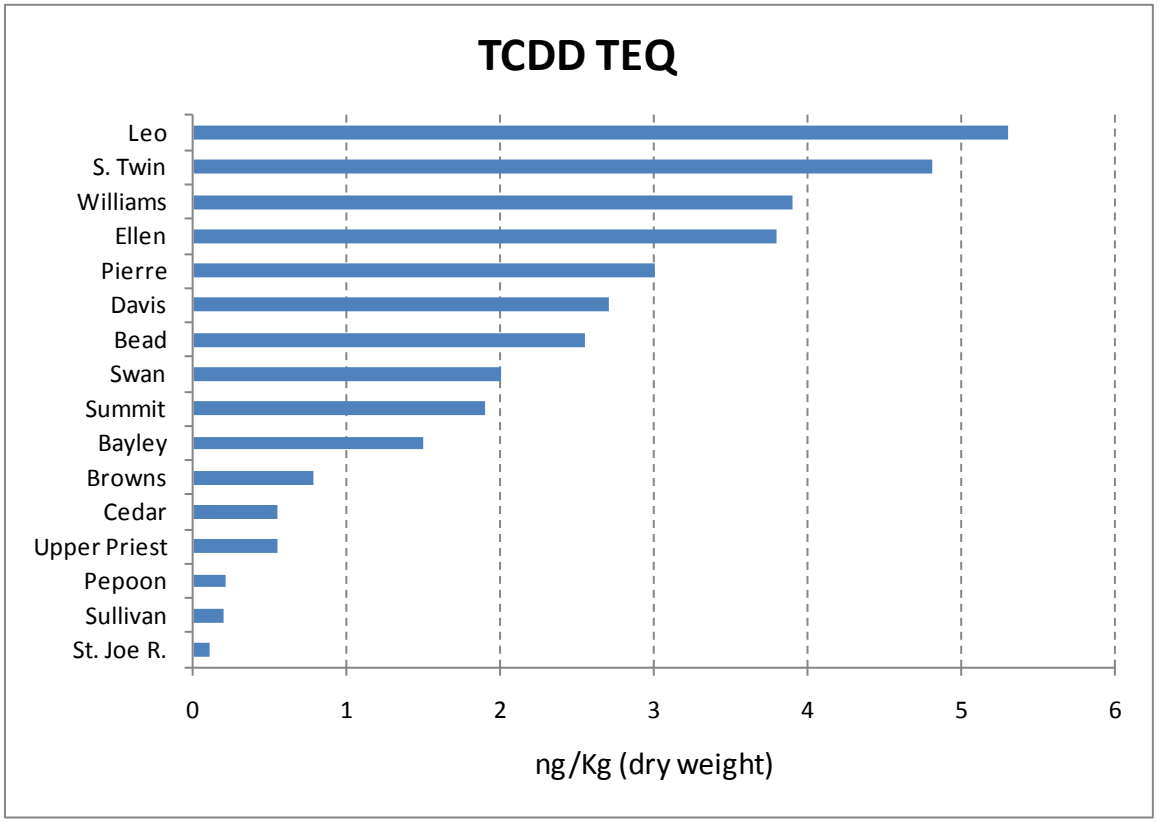


Figure 33. Waterbodies Ranked by TCDD TEQ Concentrations in Sediment Samples.

In those instances where the TEQ was greater than 1.0 ng/Kg, most of the TEQ (30-58%, 40% on average) was due to 1,2,3,7,8-PeCDD and less than 20% (6% on average) due to TCDD. These results are consistent with national findings that exposure of the U.S. population comes primarily from 1,2,3,7,8-PeCDD (EPA, 2003).

Figures 34 and 35 show how 1,2,3,7,8-PeCDD concentrations compared across the study area. No new insights on geographic patterns emerge.

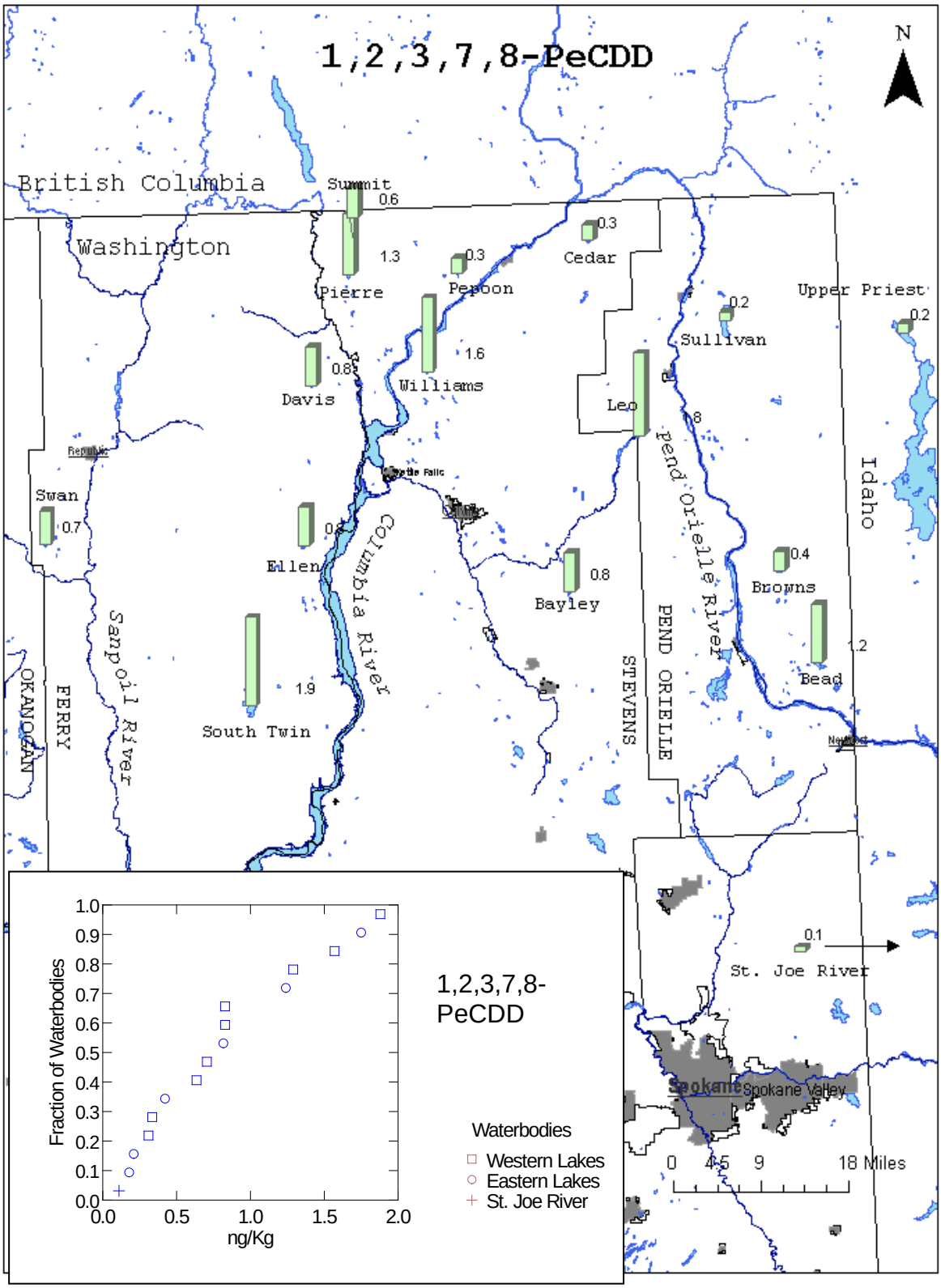


Figure 34. Spatial Patterns and Quantile Plot for 1,2,3,7,8-PeCDD in Sediment Samples.

Browns, Cedar, Pepoon, Sullivan, Upper Priest Lakes and St. Joe River plotted at the quantitation limit, 0.11-0.42 ng/Kg.

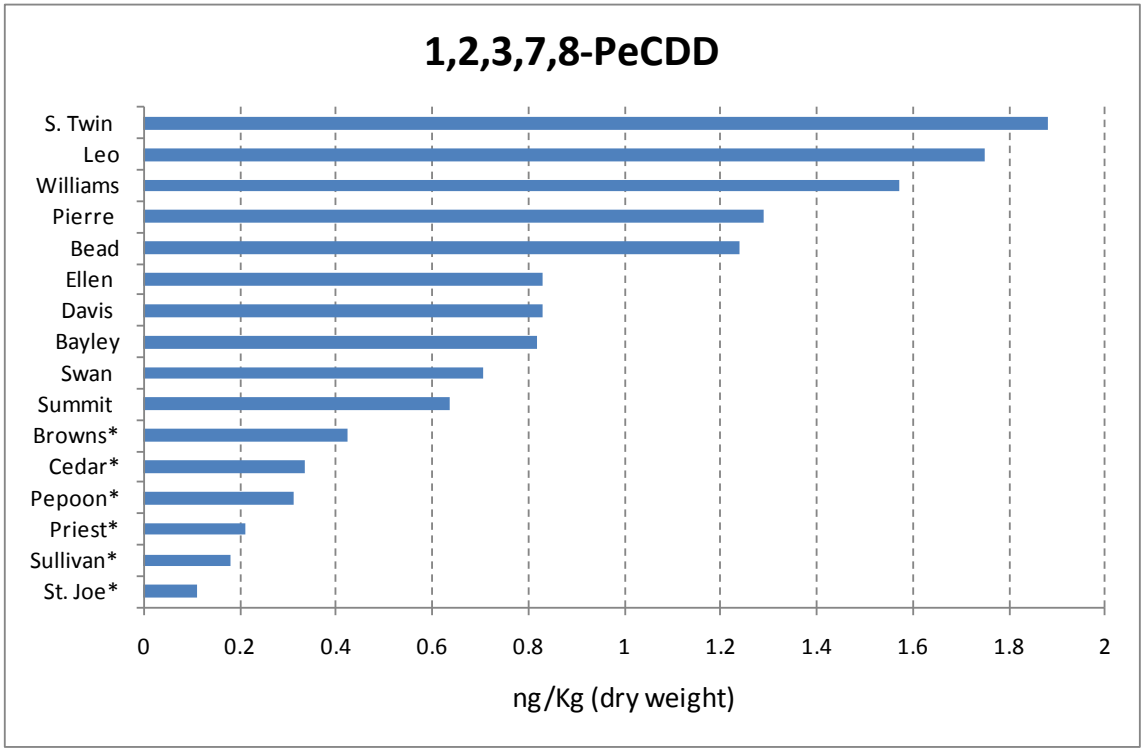


Figure 35. Waterbodies Ranked by 1,2,3,7,8-PeCDD Concentrations in Sediment Samples.
**Not detected; plotted at the quantitation limit (0.11 – 0.42 ng/Kg).*

PBDEs

PBDEs were detected in the sediments of all waterbodies at concentrations ranging from 101 – 1,946 ng/Kg (Table 26). Concentrations tended to be higher in the western lakes, but the differences were not statistically significant (Mann-Whitney test, $p>0.05$). The 90^{th} percentile total PBDE concentration for the lakes was 1,318 ng/Kg. The St. Joe River sediment sample had one of the lower total PBDE concentrations at 149 ng/Kg.

Table 26. Summary Statistics for Total PBDEs in Sediment Samples (ng/Kg, dry weight).

Waterbodies:	All Lakes	Western Lakes*	Eastern Lakes†	St. Joe River
N=	15	9	6	1
Detection Freq.	100%	100%	100%	100%
Median	612	685	458	149
Mean	733	800	632	- -
Minimum	101	101	126	- -
Maximum	1,946	1,946	1,288	- -
90th percentile	1,318	1,460	1,239	- -

*Cedar, Pepoon, Summit, Pierre, Williams, Davis, Ellen, S. Twin, and Swan.
†Sullivan, Upper Priest, Leo, Browns, Bayley, Bead.

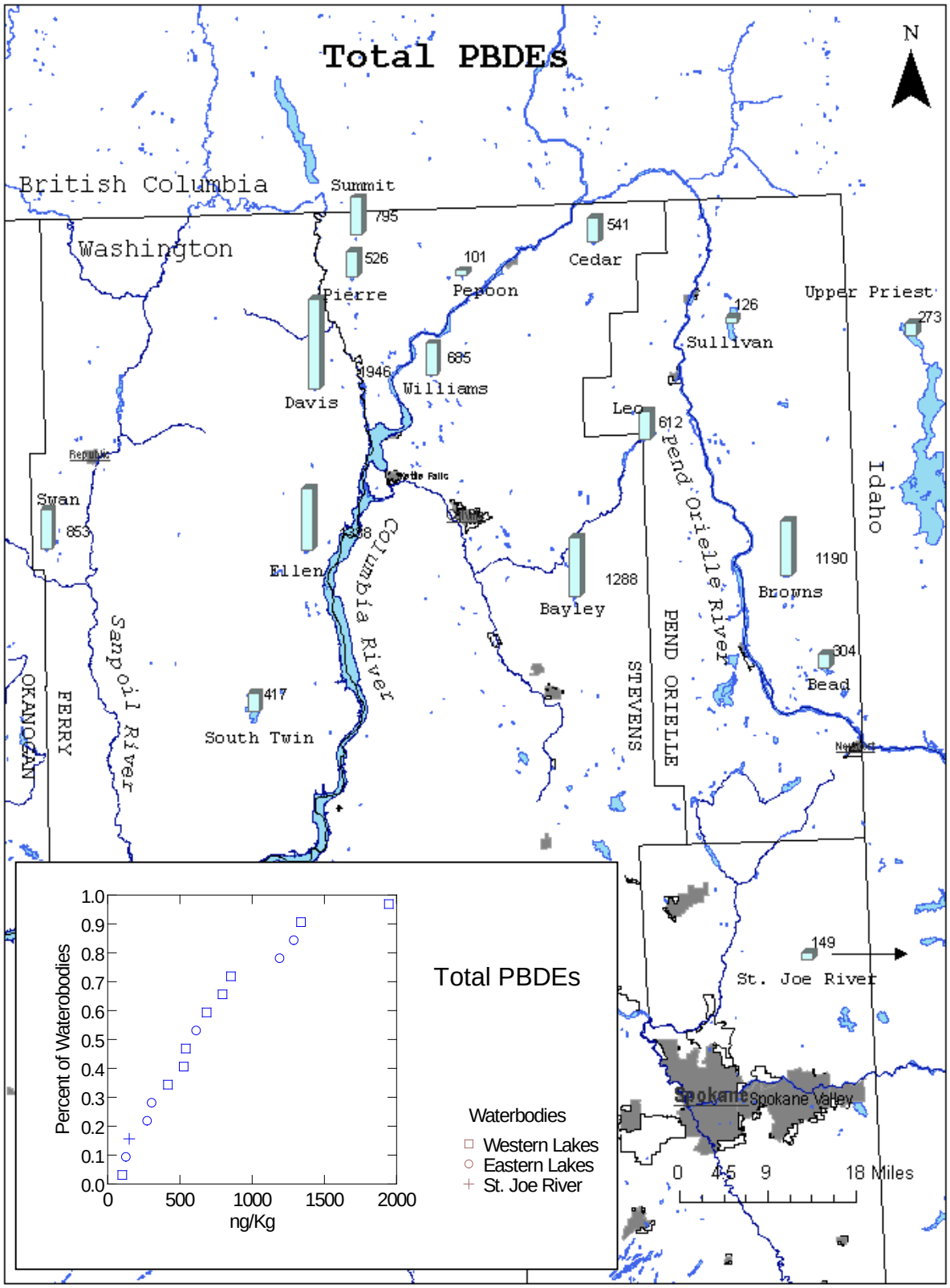


Figure 36. Spatial Patterns and Quantile Plot for Total PBDEs in Sediment Samples.

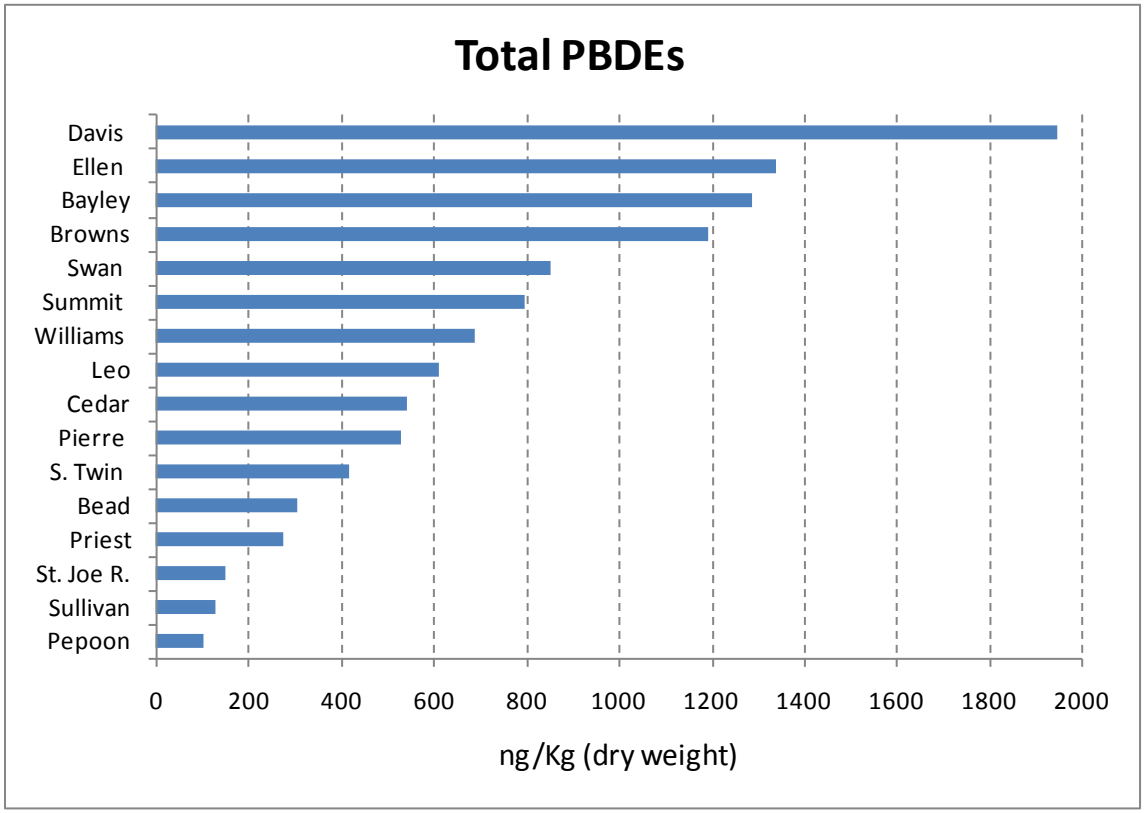


Figure 37. Waterbodies Ranked by Total PBDE Concentrations in Sediment Samples.

The most commonly occurring PBDEs were PBDE-209, -47, and -99, being detected in 50-63% of the samples. In waterbodies where these congeners were detected, total PBDEs averaged 82% PBDE-209, 21% PBDE-47, and 12% PBDE-99. Hites (2004) reviewed the environmental distribution of PBDEs and found roughly similar percentages for freshwater and marine sediments worldwide: 82% PBDE-209, 32% PBDE-47, and 23% PBDE-99. PBDE-47 and -99 were the major components of penta-PBDE flame retardants. Deca products, only recently banned in Washington State (January 2011), are almost entirely composed of PBDE-209.

Interferences or method blank issues for PBDE-209 in Pierre, South Twin, Leo, and Cedar lakes reduced the detection frequency of this important congener. Shortcomings in quantitating PBDE-209 adversely affected the results for total PBDEs, potentially obscuring patterns of interest.

Discussion

Previous Data on Background Study Lakes

Sediments from three of the lakes in the northeast Washington background study – Pierre, Leo, and Bead – have been sampled previously. Ecology's Washington State Toxics Monitoring Program (WSTMP) analyzed metals, PCBs, TOC, and grain size in sediments from Pierre and Leo Lakes, collected in 2009 in response to a request from Ecology's Eastern Regional Office (ERO) (Seiders, unpublished). This was part of ERO's initial effort to obtain expanded sediment geochemistry data on local waterbodies and served as a precursor to the present effort. Johnson and Norton (2001) report concentrations for a range of metals and organic compounds in Bead Lake sediments collected in 2000 in connection with toxicity tests on the Spokane River. As in the present 2010 study, all of these samples were composites of the top 10 cm from several grabs, one composite per lake. The samples were analyzed by MEL.

Results from the three investigations are compared in Table 27. Some differences are to be expected due to sampling locations, variations in subsampling to 10 cm, sensitivity of the analytical method (notice non-detects and estimated values), and laboratory variability. Despite these potentially confounding influences, metals concentrations, TOC, and grain size agree within a factor of 2 and often better, in most cases. The two earlier studies analyzed PCBs as Aroclor equivalents using a relatively insensitive gas chromatography/electron capture detection (GC/ECD) method. As a result, no PCBs were detected.

The Bead Lake sediments previously obtained were of substantially different character than those recently collected, having lower TOC and percent fines. The lower metals concentrations found are consistent with the coarser material analyzed. All of the earlier Bead Lake grabs were from a single nearshore location, rather than a transect across depths as in 2010. These factors are likely the predominant reasons for differences seen in the results.

The Pierre and Leo Lakes data demonstrate that sediment quality in these waterbodies can be consistently characterized at this level of resolution based on results from a single composite sample. This, in turn, implies that the sampling design for the 2010 northeast Washington regional study provides representative results for individual lakes within approximately a factor of 2.

Table 27. Comparison of Sediment Quality Data for Pierre, Leo, and Bead Lakes.
Metals and TOC in mg/Kg; PCBs in ug/Kg; dry weight basis; N=1.

Waterbody	Previous Results*	Present Study	RPD	Previous Results	Present Study	RPD	Previous Results	Present Study	RPD
	Antimony			Lead			Cadmium		
Pierre Lake	0.23 J	0.73	104%	22	41	59%	0.97	1.4	36%
Leo Lake	0.20 U	.09 J	- -	21	18	17%	0.48	0.54	12%
Bead Lake	5 UJ	0.03	- -	5 U	24	- -	0.50	0.44	13%
	Arsenic			Mercury			Zinc		
Pierre Lake	11	14	20%	0.084	0.13	45%	75	76	1%
Leo Lake	3.9	3.2	21%	0.12	0.079	43%	73	59	21%
Bead Lake	2.1	6.1	98%	0.030	0.042	33%	56	79	34%
	Chromium			Copper			Iron		
Pierre Lake	66	77	15%	86	53	47%	NA	19,300	- -
Leo Lake	15	15	2%	20	16	23%	NA	16,300	- -
Bead Lake	4.4 J	13	99%	7.0	19	92%	14,500	17,100	16%
	Total PCBs			Total Organic Carbon			Percent Fines		
Pierre Lake	11 U	2.6	- -	12	13	6%	63	79	23%
Leo Lake	25 U	3.8	- -	13	14	7%	60	81	29%
Bead Lake	13 U	2.2	- -	3.8	6.8	57%	18	74	121%

*Pierre Lake and Leo Lake (Seiders, unpublished); Bead Lake (Johnson and Norton, 2001).
RPD: relative percent difference.
U: not detected.
J: estimated concentration.
NA: not analyzed.

Reference Lakes from Other Investigations

Past sediment quality investigations in northeast Washington have selected other lakes to support an estimation of background or reference conditions (Figure 38). Separate studies by USGS and Ecology used sediments from Lower Arrow Lake in British Columbia as a reference for Lake Roosevelt sediments (Majewski et al., 2003; Era and Serdar, 2001). Lower Arrow Lake is upstream of the Trail lead-zinc smelter and Zellstoff Celgar pulp mill in B.C., major historical sources of transboundary pollution to the upper Columbia River basin. The same USGS study sampled bank sediments above the inferred high water mark in Lake Roosevelt as a second point of reference.

Era-Miller (2004) compared sediment chemistry and bioassay response in Spokane River sediments with sediments from Buffalo Lake in southeast Okanogan County. The WSTMP background samples from 2009, previously mentioned in connection with Pierre and Leo Lakes, included samples from Black, Amber, and Upper Twin Lakes (Stevens, Spokane, and Lincoln Counties, respectively). Sloan and Blakley (2009) investigated McDowell and South Skookum Lakes (Spokane and Pend Oreille Counties) as potential reference lakes for northeast Washington.

These previous sediment quality investigations included analysis for metals and, in a few cases, PCBs. Selected results are compared to medians from the present study in Figure 39. Except for PCBs, the few non-detects were plotted at the detection limit. The complete data are in Appendix C.

Median concentrations of antimony, lead, cadmium, arsenic, and mercury in sediments from the subset of western lakes used in this present study are approximately two to five times higher than corresponding median concentrations in sediments from the other past investigations. The western lakes also showed consistently elevated median concentrations for these same five metals as compared to the corresponding subset of eastern lakes in this study. Otherwise, considering the inherent variability of different studies and sediment types, the concentrations of other metals and PCBs in the lakes from the present study are generally comparable with the levels quantified by the earlier investigations. As such, the results from this present study are judged to provide representative data to define *area* and *natural* background sediment quality conditions in northeast Washington lakes.

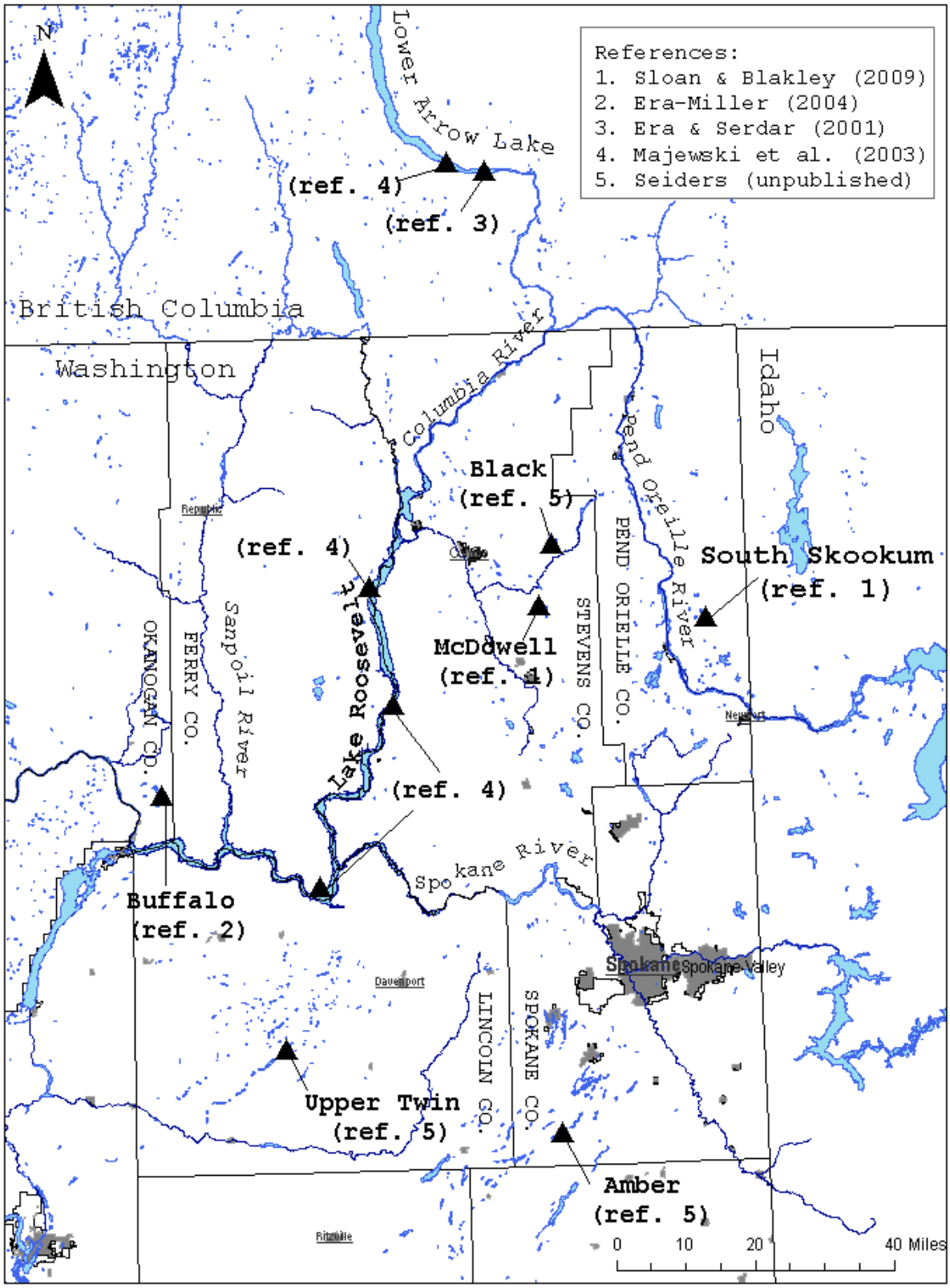
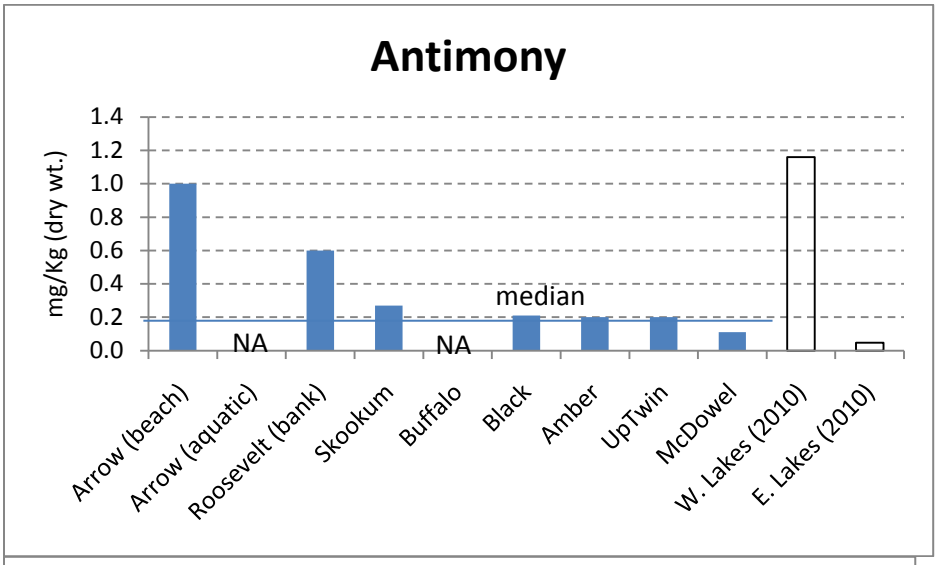
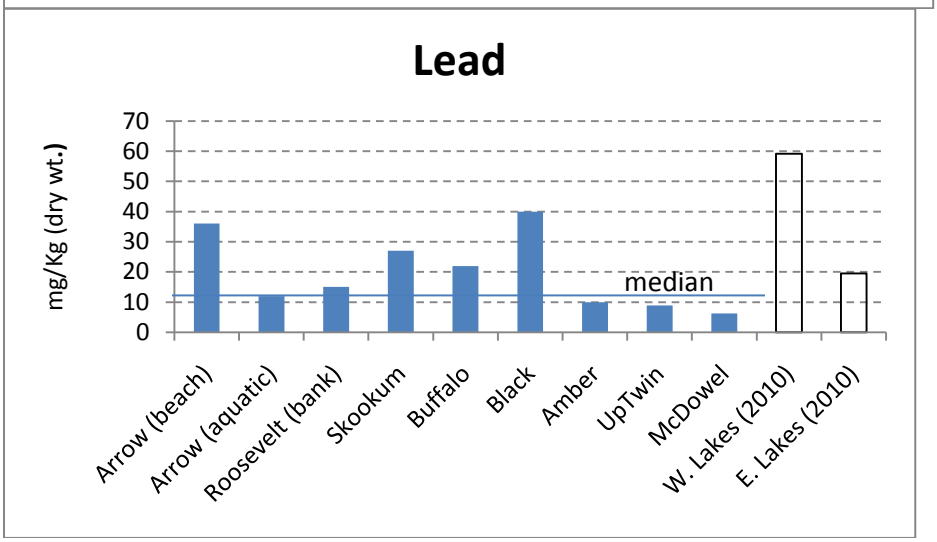
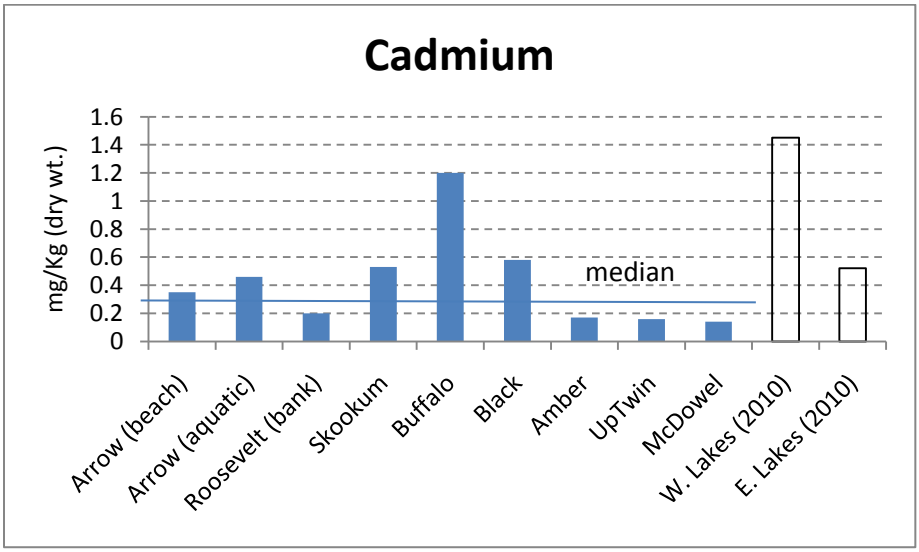
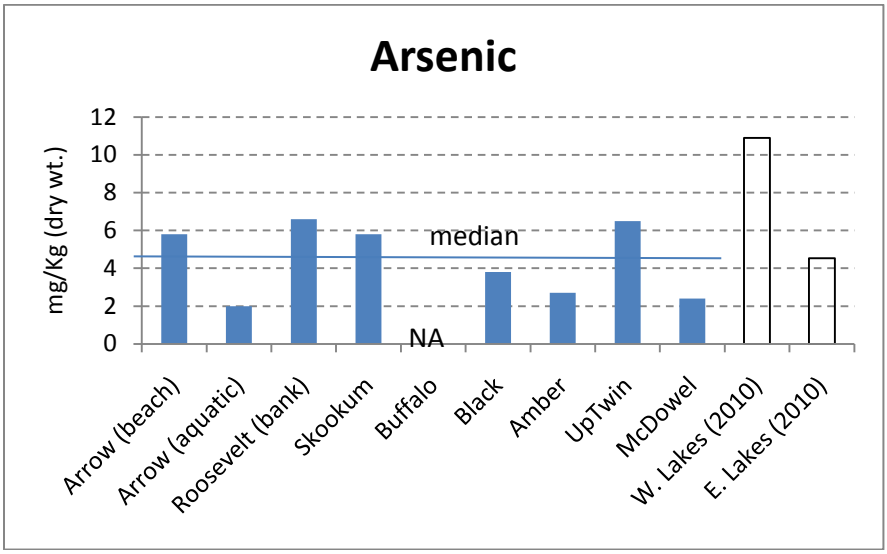


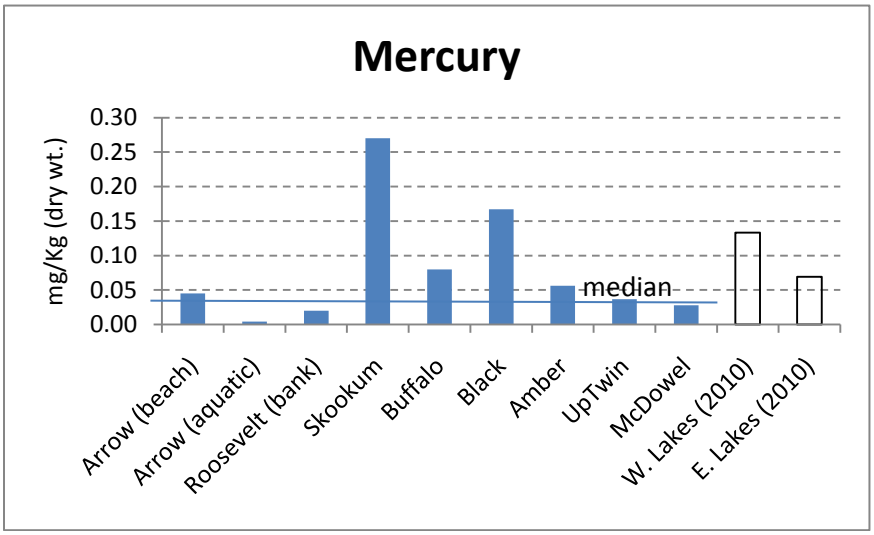
Figure 38. Reference Lakes Sampled in Other Sediment Quality Investigations in Northeast Washington.

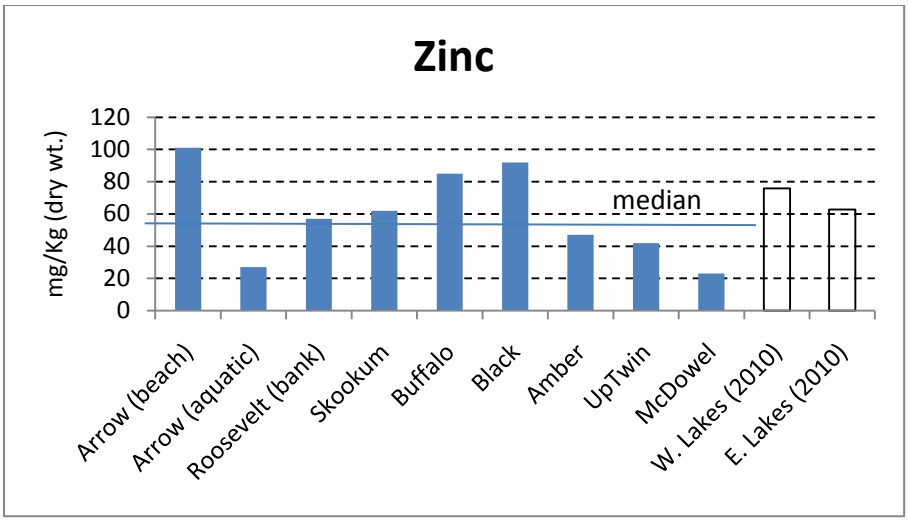

Antimony
mg/Kg (dry wt.)
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
median
NA
NA
Arrow (beach)
Arrow (aquatic)
Roosevelt (bank)
Skookum
Buffalo
Black
Amber
UpTwin
McDowel
W. Lakes (2010)
E. Lakes (2010)


Lead
mg/Kg (dry wt.)
70
60
50
40
30
20
10
0
median
Arrow (beach)
Arrow (aquatic)
Roosevelt (bank)
Skookum
Buffalo
Black
Amber
UpTwin
McDowel
W. Lakes (2010)
E. Lakes (2010)


Cadmium
mg/Kg (dry wt.)
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
median
Arrow (beach)
Arrow (aquatic)
Roosevelt (bank)
Skookum
Buffalo
Black
Amber
UpTwin
McDowel
W. Lakes (2010)
E. Lakes (2010)


Arsenic
mg/Kg (dry wt.)
12
10
8
6
4
2
0
median
NA
Arrow (beach)
Arrow (aquatic)
Roosevelt (bank)
Skookum
Buffalo
Black
Amber
UpTwin
McDowel
W. Lakes (2010)
E. Lakes (2010)


Mercury
mg/Kg (dry wt.)
0.30
0.25
0.20
0.15
0.10
0.05
0.00
median
Arrow (beach)
Arrow (aquatic)
Roosevelt (bank)
Skookum
Buffalo
Black
Amber
UpTwin
McDowel
W. Lakes (2010)
E. Lakes (2010)


Zinc
mg/Kg (dry wt.)
120
100
80
60
40
20
0
median
Arrow (beach)
Arrow (aquatic)
Roosevelt (bank)
Skookum
Buffalo
Black
Amber
UpTwin
McDowel
W. Lakes (2010)
E. Lakes (2010)

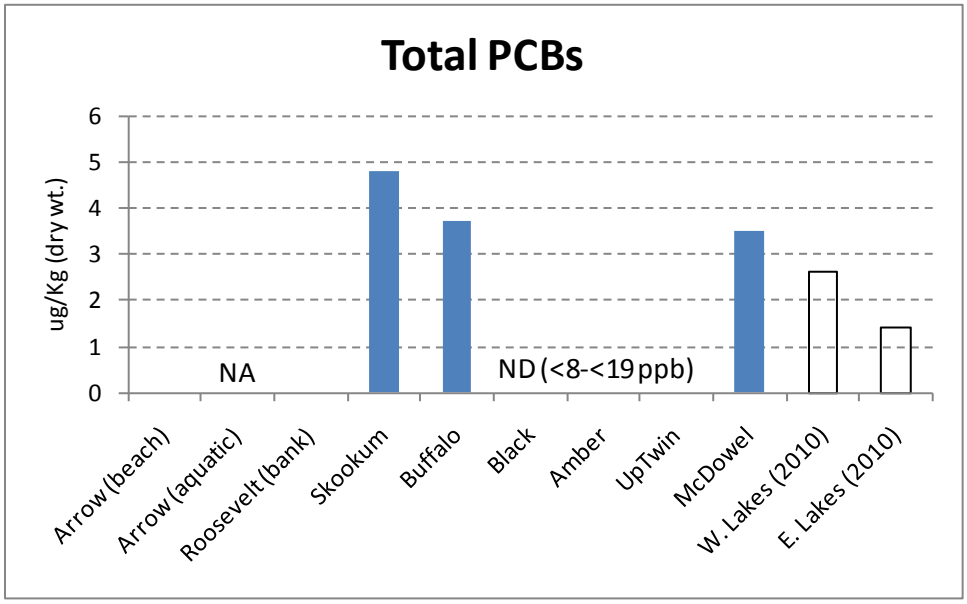


Figure 39. Metals and PCB Concentrations Reported in Sediments from Northeast Washington and British Columbia Reference Lakes (N=1-3) Compared to Medians from Present Study. *N=9 and 6, respectively, for "W. Lakes" and "E. Lakes."*

Historical Background

Metals

An Ecology program to measure trends in persistent, bioaccumulative toxics (PBTs) recently analyzed lead and mercury at a high level of resolution in an age-dated sediment core from Black Lake (Furl, unpublished). Black Lake is located in the approximate center of the study area for the present project, midway between Leo and Bayley Lakes (see Figure 38). Analysis and comparison of concentration variations between shallower and deeper layers from this core provides insights into the pollution history of lead and mercury within northeast Washington lakes over the past 100 to 200 years.

The lead and mercury profiles from the Black Lake core are shown in Figure 40. Dates were assigned based on Pb-210 activity, as described in Furl (2007). An ash layer from a 1920 forest fire in this part of the Colville National Forest provides a convenient marker to corroborate the Pb-210 dates.

As can be seen in these figures, lead and mercury remain relatively unchanged from the late 1700s to the early 1900s (i.e., the 20-40 cm layer). Concentrations steadily increase for both metals into the mid to late 1900s, followed by a lowering to present-day levels.

Thicker core increments of 0-10, 10-20, and 20-30 cm were analyzed for a wider range of metals and for PCBs. The results for the 20-30 cm layer (early 1900s to mid-1800s) provide a benchmark for concentrations prior to largescale urbanization and industrial development.

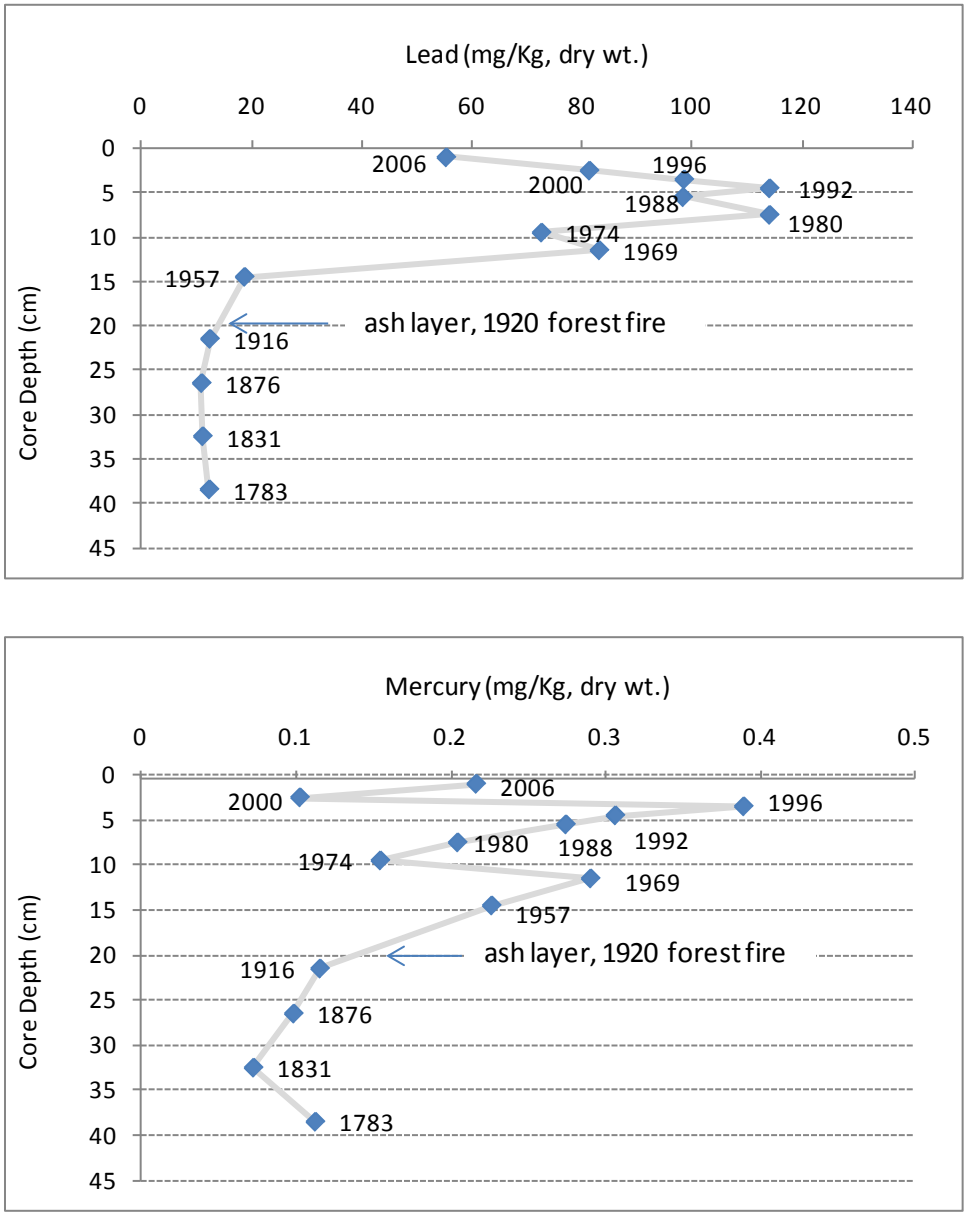


Figure 40. Changes in Lead and Mercury Concentrations Over Time in the Northeast Washington Study Area, as shown in a Sediment Core from Black Lake, Pend Oreille County. *Chad Furl, unpublished data.*

The metals concentrations in lake sediments from this present study were compared to the 20-30 cm layer in Black Lake to assess the extent to which they exceed historical, benchmark background (Figure 41). The data are plotted as a ratio of the northeast lakes medians and 90th percentiles to the benchmark background concentrations measured in the Black Lake core. Ratios greater than 1 exceed the benchmark background concentration.

Sediment concentrations of cadmium, antimony, lead, and arsenic in western study area lakes are consistently higher than the Black Lake benchmark; specifically, the median concentrations range from 7.5 times (cadmium) to 4.4 times (lead) higher than the Black Lake levels. The

calculated 90th percentile concentrations of cadmium, antimony, and lead in these same western lakes exceed the Black Lake background concentrations by a factor of 10 or more. For these same metals, the eastern lakes are elevated only slightly, by factors of 1.5-2.6.

Median concentrations of zinc, mercury, chromium, and copper from the eastern lakes are at or close to the Black Lake background, and elevated by factors of about 2 or less for the western lakes. As compared with the core study, the lower detection limits associated with the present study give the appearance of a reduction in the concentration of selected metals – most notably antimony.

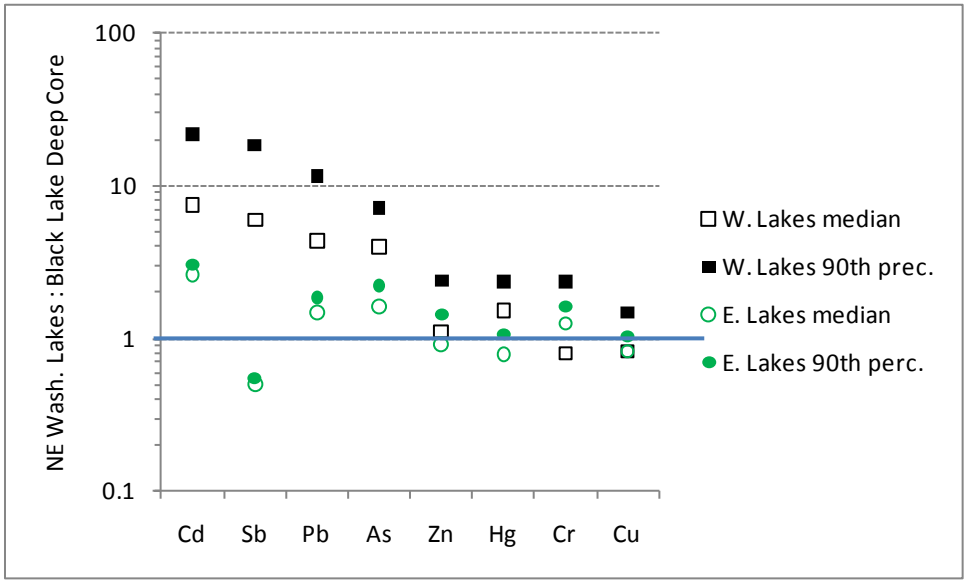


Figure 41. Metals Concentrations in Sediments from the Northeast Washington Background Study as Ratios with Historical Concentrations Measured at 20-30 cm in a Black Lake Sediment Core (log scale).

The Black Lake core results also offer a demonstration of the influences sediment sampling methods can have on observed results. The present study obtained composite grab samples collected from the top 10 cm (approximate) of the sediment column. In contrast, the Black Lake core was subdivided into discrete depth-specific sub-samples, resulting in one to four separate concentration values for a given 5-cm interval. The smaller depth interval represented by the core sub-samples provides a more detailed reporting of pre-industrial background and post-industrial anthropogenic impacts on lake sediment chemistry.

In contrast, results from the present study effectively represent a composited snapshot of sediment conditions, typically over decades and potentially over the last century. This is exemplified by comparison of the Black Lake profile to the current results from nearby Leo and Bailey Lakes. At these two similar and nearby lakes, lead measured less than 20 mg/Kg and mercury concentrations were at or below 0.10 mg/Kg. These results are more closely in line with the discrete historical 20-30 cm depth concentrations observed at Black Lake and demonstrate how variations in lake deposition rates and sample collection methodology potentially influence sediment concentration results.

Organic Compounds

PCBs were analyzed in the Black Lake core, but none were detected due to the relatively insensitive GC/ECD method used. However, the historical background for synthetic compounds like PCBs would be zero.

Sediment core data could not be located for the northeast Washington area for PCDDs, PCDFs, or PBDEs. Like PCBs, PBDEs are synthetic compounds and therefore have no historical background in aquatic sediments.

Forest fires are a natural source of PCDDs and PCDFs, although minor in comparison to anthropogenic sources (EPA, 2003). An EPA study of sediment cores from 11 geographically distributed lakes across the United States showed that PCDD/PCDF concentrations generally begin to rise in the 1930s and 1940s, declining in some lakes in the 1960s and 1970s (Cleverly et al., 1996). This study included two Washington lakes – Ozette and Beaver – both located on the Olympic Peninsula and subject to the influence of forest fires, as are the lakes of northeast Washington.

Unpublished data[5] (MRI, 1995) on the deepest layers from these two cores are compared to findings for the northeast lakes study in Table 28. The age assigned to individual core increments was not available. However, the period of record was established at 1720–1989 for Lake Ozette and 1884–1985 for Beaver Lake (Cleverly et al., 1996).

Table 28. Historical TCDD and TCDD-TEQ Levels in Deep Layers of EPA Sediment Cores from Two Olympic Peninsula Lakes Compared to Present-Day Northeast Washington Lake Surface Sediments (ng/Kg, dry weight).

	Ozette and Beaver Lakes* (range)	N.E. Washington Lakes	
		Western Lakes (median)	Eastern Lakes (median)
TCDD	0.025 U-0.044 U	0.22	0.052 U
TCDD-TEQs	0.020 - 0.062	2.7	1.1

*data source: Midwest Research Institute (1995).
U: not detected at or above reported sample quantitation limit.

TCDD was not detected in the deep sediments from Ozette or Beaver Lakes at or above 0.044 ng/Kg. Similar or potentially slightly higher levels were observed for TCDD in the eastern lakes from the present study, with a median of <0.052 ng/Kg (maximum of 0.098 ng/Kg). TCDD was frequently detected (67% of samples) in the western lakes; the median concentration was at least five times greater than historical background in the Ozette and Beaver cores. Both the western and eastern lake sediments show evidence of substantial elevations in present-day TCDD-TEQs compared to this benchmark for pre-industrial background.

[5] Obtained through the courtesy of Matthew Lorber, EPA, Washington D.C.

Evidence for Transboundary Pollution Effects

As discussed in previous sections of this report, industrial practices within British Columbia are known to have contributed to transboundary pollution in northeast Washington. These cross-border sources of pollution need to be considered when evaluating the findings from the present study.

Water, sediment, and fish contamination of the mainstem upper Columbia River (including Lake Roosevelt) by metals from the Teck[6] lead-zinc smelter in Trail, B.C. (Trail smelter) and dioxins from the Zellstoff Celgar (Celgar) pulp mill in Castlegar, B.C. has been documented by the U.S. Fish & Wildlife Service, USGS, EPA, and Ecology (e.g., Schmitt and Brumbaugh, 1990; Bortleson et al., 1994; Johnson et al., 1990, 1991; EPA, 2006). Studies show a gradient of increasing levels of these contaminants moving upstream toward the international border and attribute it primarily to wastes discharged into the Columbia River by the Trail smelter and Celgar pulp mill operations. Contamination of the upper Columbia River and Lake Roosevelt in the state of Washington is currently being investigated by EPA consistent with the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA, commonly known as Superfund).

Trail Smelter

The Trail smelter has been operating since 1906. It is one of the largest facilities of its type in the world. Throughout its history, the smelter has released sulfur dioxide (SO_2) and metals into the atmosphere. By 1925, SO_2 emissions from the smelter were estimated at 10,000 tons per month (EPA, 2008). Air emissions from the Trail smelter are known to have dispersed southward within the Columbia River Valley and associated airshed. These emissions have created measurable impacts to air quality on the U.S. side of the border.

Severe air pollution downwind of the smelter led to a famous arbitration in 1931 by the International Joint Commission which recommended compensation to Washington farmers for crop damages and changes in the emission of selected smelter-related air pollutants. Although pollution control devices were installed, emissions continued to the extent that further arbitration followed. A 1941 decision stipulated that the Trail smelter should avoid air emissions that could adversely impact Washington State, enhanced pollution control measures should be implemented, and Canada should pay for harm to the United States from future smelter emissions (Hess, 2005).

The Upper Columbia River Natural Resources Trustee Council (2009) has concluded that "*Historic smelter emissions have potentially impacted upland soils and other resources. Goodarzi (2002a, 2002b) documented aerial deposition originating from the smelter and measured elevated As, Cd, Cu, Pb, Hg and Zn concentrations in soil, as far as the US-Canada border. However, a data gap exists on soil contamination and resulting impacts to the natural floristic structure within the UCRS (Upper Columbia River Site) potentially caused by stack emission contaminants.*"

[6] Previously Teck Cominco, or Cominco

ICF International has done air quality modeling to examine the impacts of airborne SO_2 and deposition of metals to land and water surfaces of the upper Columbia Basin (ICF International, 2011). ICF concluded that "Emissions from Teck-Cominco (based on 1990 emissions levels) contribute to deposition of mercury and other metals (especially lead and zinc) over a broad area including the Upper Columbia River Basin and parts of the Colville Indian Reservation." It should be noted that air emission of metals from the Trail smelter continue to the present day although much reduced over historical levels (Environment Canada, National Pollutant Release Inventory www.ec.gc.ca/inrp-npri).

In recognition of this nearby, large-scale emission source, and documented air quality impacts attributed to its historical operations, the Trail smelter is likely the primary source for several of the metals whose concentrations are well above historical sediment background levels (see Figure 41). In particular, these past and ongoing smelter emissions are believed to have strongly contributed to the pattern of elevated metals in sediments measured in several of the western lakes included in this present study. The Black Lake core also suggests smelter impacts in the area of the eastern lakes, but to a lesser degree. In contrast, the metals concentrations observed in deeper portions of the sediment column within these upland lakes (e.g., at depths greater than 20 cm in the Black Lake core sample) are likely reflective of the natural, pre-industrial local soils within each lake's watershed area.

Metals concentrations from deeper lake sediment horizons typically provide a more representative indication of natural background conditions prior to industrial development. Conversely, metals concentrations approaching the top of the sediment column are expected to reflect a greater degree of anthropogenic impacts. This expected fine-scale vertical variability in metals concentrations must be properly considered when collecting and averaging sampled concentration values.

A perspective on the Trail smelter air plume is provided in Figure 42 which shows the area of injury to the forests of northern Stevens County in 1930 (ICF International, 2011). At the time of this survey by the U.S. Department of Agriculture, the damage extended as far south as Kettle Falls. The area of greatest forest damage encompasses a subset of lakes which exhibit some of the highest concentrations of antimony, lead, cadmium, and arsenic in the sediments, namely Cedar, Pepoon, Williams, and Davis Lakes (see Figures 4 through 10).

The somewhat lower concentrations of antimony, lead, and cadmium found in nearby Summit and Pierre Lakes may partly reflect their more westerly location outside the main SO_2 documented path of the plume. Elevated concentrations of copper and chromium were observed in the Pierre Lake sediment. No documented evidence (e.g., chemical treatment for fish enhancement or weed control) was discovered to account for these specific enrichments. The overall sediment concentration pattern from the present northeast Washington lakes study suggests that the elevated copper and chromium concentrations at Pierre Lake may be unrelated to the smelter emissions and documented SO_2-related impacts.

Lake elevation and local geography are also factors to consider in exposure to the smelter plume, as can be inferred from Figure 42. Elevation effects may partly explain the relatively lower metals concentrations in Summit Lake vs. nearby Pierre Lake (2,600 ft. vs. 2,012 ft.) and Pepoon

Lake vs. nearby Cedar and Williams lakes (2,450 ft. vs. 2,135 and 1,980 ft.). Elevations of the study lakes were given in Table 2.

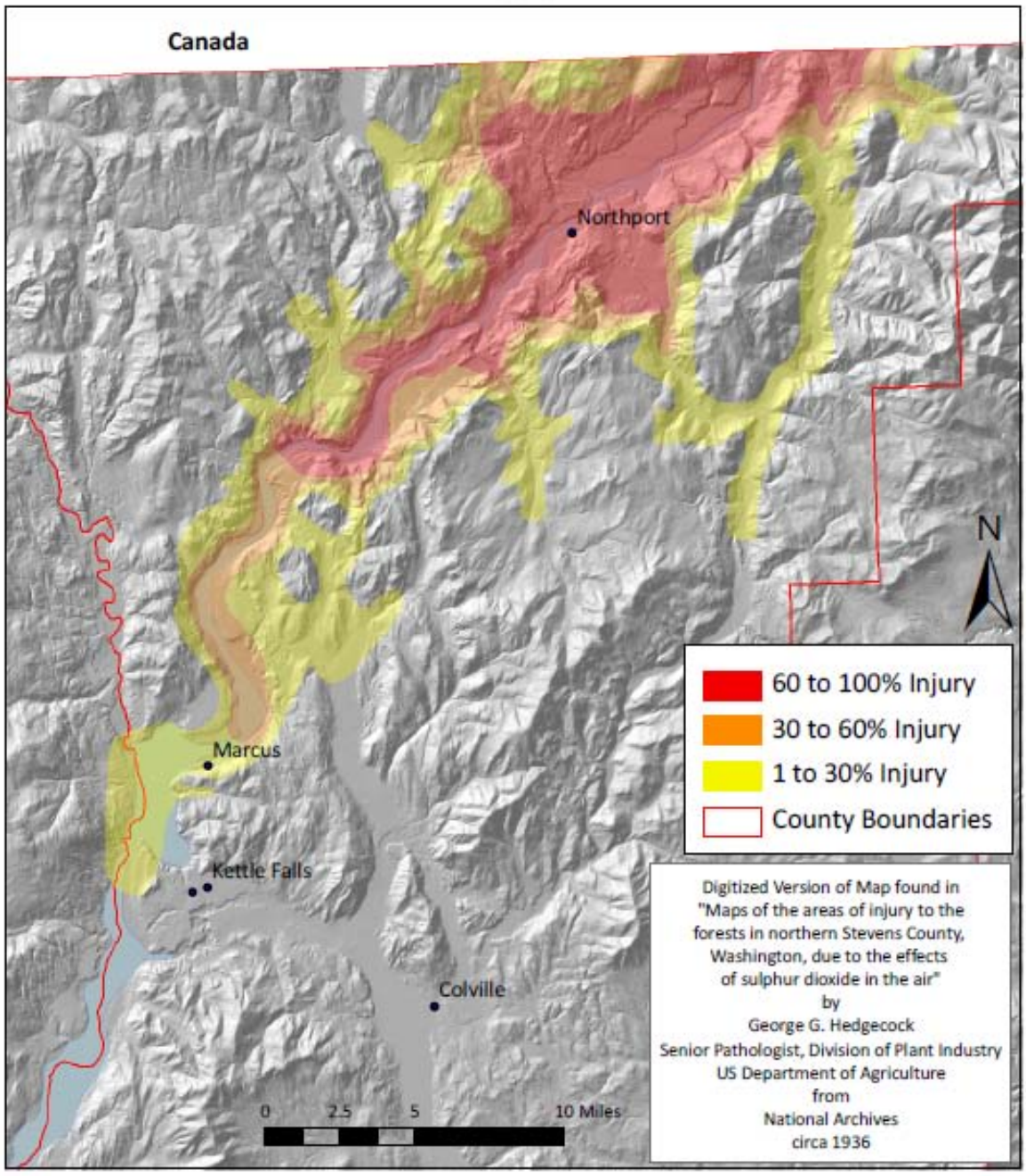


Figure 42. Forest Damage in Stevens County, Washington from Trail Smelter Sulfur Dioxide Air Emissions, 1930.

From ICF International, 2011.

A second, smaller smelter operation having a shorter operations history, known as the Le Roi Smelter, operated on and off in Northport, Washington until the 1920s. This facility also is expected to have locally contributed to emission pollution in the Northport vicinity (EPA, 2005c; Teck Cominco American Inc., 2008).

Recent data (unpublished) further supporting a link between Trail air pollution and contaminated lake sediments have been obtained by the Pacific Northwest Region of the U.S. Forest Service (USFS) Air Resource Management Program. Geiser (2011) has produced maps of air deposition patterns for metals in Washington and Oregon, based on analyzing epiphytic lichens growing on trees. According to Goodarzi et al., (2001) "[t]he deposition of particles and associated elements from a stationary source such as a smelter can be monitored using various biological media (e.g., transplanted lichen, moss) and/or passive collection methods (e.g., deposit gauges, sticky tray, dust jar/bucket or impactor)." As such, lichen can serve as effective biological media to track and monitor downwind aerial deposition of air pollutants such as metals that originate from industrial sources like the Trail smelter.

The USFS maps show that monitoring stations along the upper Columbia River near the U.S.-Canadian border continue to stand out as having higher metals levels than most of the rest of the region. Geiser (2011) estimates the average age of the USFS lichen specimens to be in the range of 5-20 years, thus representing metals deposition averaged over that period.

A draft map of the USFS cadmium data is shown as an example (Figure 43). Elevated cadmium levels extend down into the lower parts of the Lake Roosevelt area, far below Kettle Falls. The lichen samples show a similar concentration enrichment pattern for lead and zinc. USFS did not analyze antimony, and their mercury data do not include the Upper Columbia area. The USFS lichen data can be found at http://gis.nacse.org/lichenair.

The USFS lichen data and findings from other studies referenced in this report provide compelling lines of evidence to indicate a causal relationship exists between smelter-related air pollution and associated enrichment of selected metals in a number of upland lakes included in this present study. These findings and observations do not support local geologic mineral content of watershed soils as an explanation for the metal enrichment pattern which is most notable in the western study area lakes.

Celgar Pulp Mill

During the late 1980s effluent wastes from the Celgar bleach pulp mill were shown to contain high concentrations of PCDDs and PCDFs, an artifact of bleaching with elemental chlorine. In response to concerns about fisheries resources in the Columbia River, Celgar installed a chlorine dioxide bleach plant and added secondary treatment of its effluent. This ultimately led to an order of magnitude decrease in PCDD/PCDF levels in Columbia River fish (Serdar et al., 1994).

Dioxin (TCDD) levels were significantly higher in the sediments of western lakes compared to eastern lakes in the present study. However, unlike metals, a gradient of increasing concentrations moving north toward the international border was not apparent. Also, similar differences between western and eastern lakes were not observed for other dioxins or furans or for TCDD TEQs.

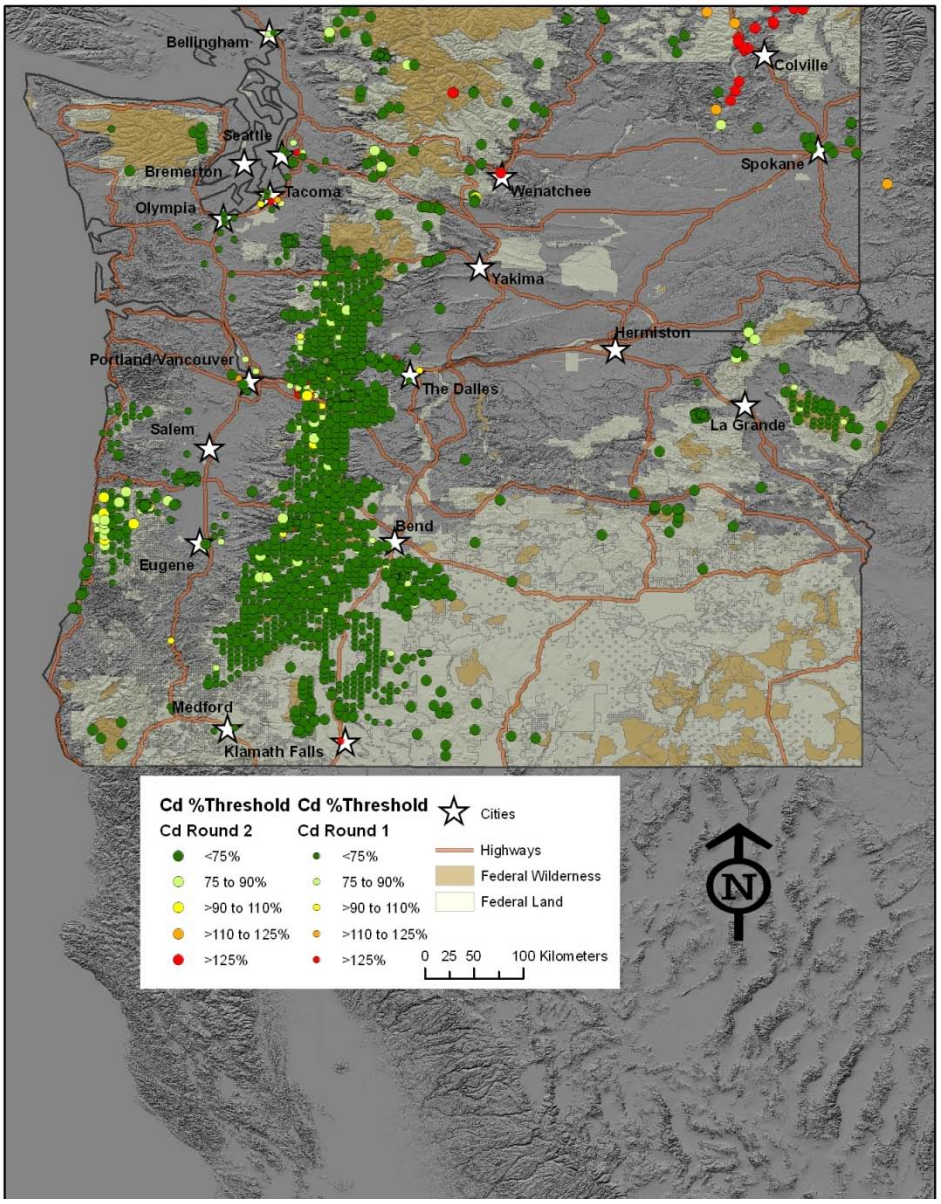


Figure 43. Cadmium Concentrations in Pacific Northwest Lichens as Percent of Clean-Site Thresholds.

1993 – 2008 samples.
Provided by Linda Geiser, USFS, Corvallis, OR.

While Celgar's impact on dioxin and furan levels within the waters and sediments of the upper Columbia River and Lake Roosevelt is well established, the effect on air emissions (and associated upland dispersion), if any, on northeast Washington land and water surfaces is unknown. Octachlorodibenzo-p-dioxin was the dominant congener in sediments from all of the lakes sampled in the present northeast Washington study, indicating that the major source is combustion generated emissions (EPA, 2003). Other potential sources of dioxin to the study area include burning domestic refuse in backyard burn barrels, diesel exhaust, and other incomplete combustion of fossil fuels and wood.

PCBs and PBDEs

The spatial distribution of PCBs and PBDEs in the lake sediments does not point to the existence of large localized sources within or adjacent to the northeast Washington area. Diffuse local inputs and global atmospheric transport are likely the most important sources of these compounds (Cleverly et al., 2007).

Sediment Quality Guidelines

An important consideration when using background values for setting cleanup targets for aquatic environments is the extent to which these values are protective of aquatic life and human health. Washington State currently has regulatory sediment quality criteria for marine waters only. Ecology determines appropriate criteria to evaluate potential chemical toxicity of freshwater sediments on a case-by-case basis (WAC 173-204-340).

MacDonald et al. (2000) have developed a set of consensus-based sediment quality guidelines for protecting freshwater ecosystems, based on their analysis of guidelines proposed by six federal, state, or provincial agencies in North America. MacDonald's Threshold Effect Concentrations (TECs) are intended to identify the concentrations of sediment-associated contaminants below which adverse effects on sediment-dwelling organisms are not expected to occur. Probable Effect Concentrations (PECs) are intended to define the concentrations of sediment-associated contaminants above which adverse effects on sediment-dwelling organisms are likely to be observed. Ecology's Eastern Regional Office uses the MacDonald TECs and PECs to screen sediment chemistry data for potential to adversely affect aquatic life.

Table 29 compares the MacDonald threshold and probable effects concentrations to the range of concentrations measured in the northeast Washington study area. MacDonald et al. does not provide guidelines for antimony. The antimony values in Table 29 are proposed sediment quality standards (SQS) and cleanup screening levels (CSL) from Betts (2003), an Ecology effort to establish freshwater sediment quality guidelines using local data.

The U.S. Army Corps of Engineers is currently using the Betts study and other sources to finalize sediment quality values as guidance for assessing freshwater sediments for dredging and disposal in the Pacific Northwest (USACE, 2009). The afore-mentioned sources do not have sediment quality guidelines for the other chemicals analyzed in the northeast Washington background study.

Table 29. Freshwater Sediment Quality Guidelines Compared to Concentrations Measured in Northeast Washington Sediments during Present Study.

Chemical	Threshold Effect Concentrations*	Probably Effect Concentrations*	Concentration Range in N.E. Washington Study Area
Metals (mg/Kg, dry wt.)			
Antimony	0.4†	0.6†	0.02 - 4.2
Lead	36	128	2.3 - 190
Cadmium	0.99	5.0	<0.05 - 6.2
Arsenic	9.8	33	3.2 - 28
Mercury	0.18	1.1	<0.007 - 0.21
Zinc	121	459	19 - 215
Copper	32	149	14 - 77
Chromium	43	111	8.6 - 53
Organics (ng/Kg, dry wt.)			
Total PCBs	60,000	676,000	7 - 8,335

*MacDonald et al. (2000).
†Sediment quality standard and cleanup screening level, respectively, proposed by Betts (2003).

As shown in Table 29, the higher of the antimony, lead, cadmium, arsenic, mercury, zinc, copper, and cadmium concentrations in the northeast Washington lake sediments exceed (do not meet) sediment quality guidelines. A detailed comparison by individual waterbody for antimony, lead, cadmium, arsenic, mercury, and zinc is provided in Figure 44, which plots the ratio of metals concentration in the sediments to the guideline value. Ratios greater than 1 exceed guidelines.

All or most of the western lakes exceed the SQS for antimony and the TEC for lead and cadmium, with several of these same lakes also exceeding TECs for arsenic, mercury, and zinc. Exceedance factors ranged from 1.1 to 10. Pierre Lake also exceeded TECs for copper and chromium (not shown in Figure 44). None of the eastern lakes exceed sediment quality guidelines.

Table 30 summarizes and ranks those lakes that exceed sediment quality guidelines. Sediments in these lakes are potentially toxic to bottom-dwelling organisms due to elevated metals concentrations. Williams and Cedar Lakes stand out as exceeding guidelines to the greatest extent. Antimony, lead, and cadmium appear to be a particular concern, exceeding the proposed CSL for antimony in all western lakes, the PEC for lead and cadmium in Williams Lake, and the PEC for cadmium in Cedar Lake.

Concentrations of bioaccumulative chemicals in freshwater sediments that may ultimately be harmful to human health through fish consumption have not been fully evaluated or finalized for screening purposes. Fish tissue data for the northeast Washington study area are being reported in a separate document, currently in preparation, and can be used to assess human health risk directly.

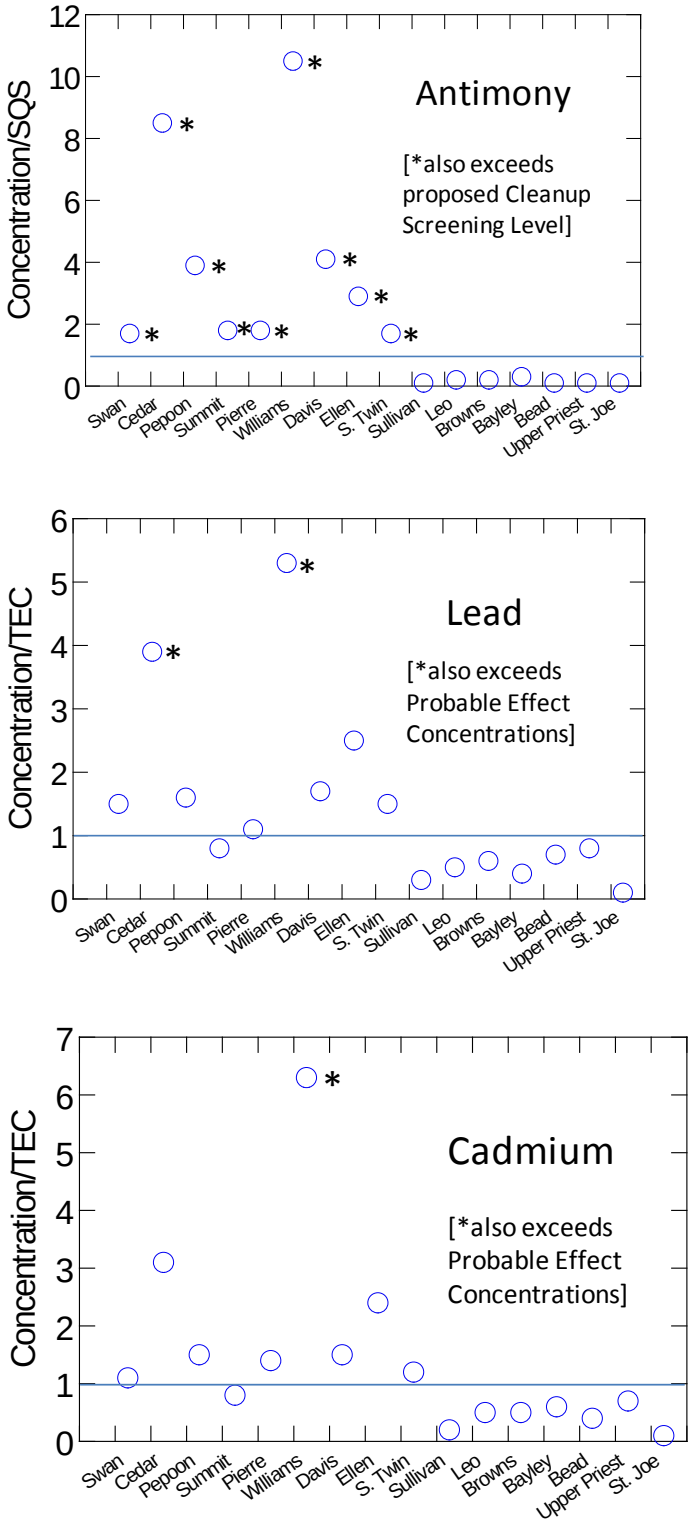


Figure 44. Metals Concentrations in Sediments from the Northeast Washington Study Area Compared to MacDonald et al. (2000) Threshold Effect Concentrations and Betts (2003) Proposed Sediment Quality Standard (Antimony).

Ratios > 1 exceed effects threshold or sediment standard.

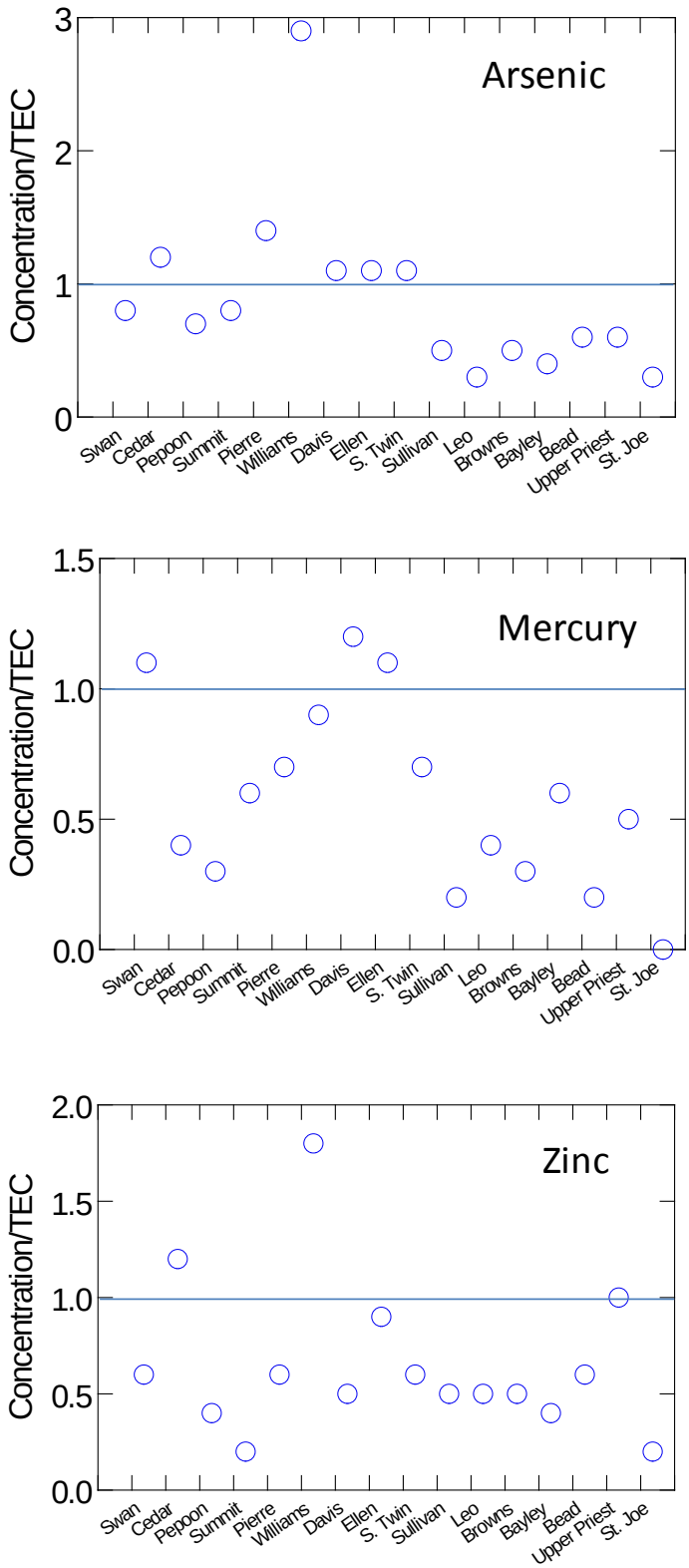


Figure 44. (continued)

Table 30. Summary of Northeast Washington Study Area Lakes that Exceed Sediment Quality Guidelines (exceedance factors).

Metal: Guideline:	Antimony SQS/CSL	Lead TEC/PEC	Cadmium TEC/PEC	Arsenic TEC	Mercury TEC	Zinc TEC	Copper TEC	Chromium TEC
Williams Lake	10/7.0	5.3/1.5	6.3/1.2	2.9	- -	1.8	- -	- -
Cedar Lake	8.4/5.6	3.9/1.1	3.1	1.2	- -	1.2	- -	- -
Davis Lake	4.1/2.7	1.7	1.5	1.1	1.2	- -	- -	- -
Ellen Lake	2.9/1.9	2.5	2.4	1.1	1.1	- -	- -	- -
Pepoon Lake	3.9/2.6	1.6	1.5	- -	- -	- -	- -	- -
Pierre Lake	1.8/1.2	1.1	1.4	1.4	- -	- -	2.4	1.2
S. Twin Lake	1.7/1.1	1.5	1.2	1.1	- -	- -	- -	- -
Swan Lake	1.7/1.1	1.5	1.1	- -	1.1	- -	- -	- -
Summit Lake	1.8/1.2	- -	- -	- -	- -	- -	- -	- -

SQS/CSL: Sediment Quality Standard and Cleanup Screening Level proposed by Betts (2003).
TEC/PEC: Threshold Effect Concentrations and Probable Effect Concentrations from MacDonald et al. (2000).

MTCA-based Background Values for Northeast Washington Aquatic Sediments

Although the data obtained through this project have a variety of possible uses, they were primarily collected in support of water quality-based and toxics-based cleanup studies and actions for east and northeast Washington waterbodies.

The Model Toxics Control Act (MTCA) Cleanup Regulation (WAC 173-340) uses specific methods for defining natural and area background concentrations. Several statistical approaches and considerations for determining a representative background concentration are described in WAC 173-340-709(3)(b) through (d):

(b) Background sampling data shall be assumed to be lognormally distributed unless it can be demonstrated that another distribution is more appropriate.

(c) For lognormally distributed data sets, background shall be defined as the true upper 90th percentile or four times the true 50th percentile, whichever is lower.

(d) For normally distributed data sets, background shall be defined as the true upper 80th percentile or four times the true 50th percentile, whichever is lower.

In the present study, lakes data were used to calculate background concentrations (90^{th} or 80^{th} percentiles) for metals and organic compounds in aquatic sediments of northeast Washington, following MTCA procedures. The MTCA*Stat*97 Background Module (BCKGD97.xlt) was used to calculate the statistics (www.ecy.wa.gov/programs/tcp/tools/toolmain.html), except where the number of samples or data distribution were such that the module could not calculate percentiles. In those instances, the standard Excel formula was used (Microsoft Office, 2007). Results of the calculations are shown in Tables 31 and 32.

Table 31. Background Values (bold font) for Aquatic Sediments in Northeast Washington Lakes: Metals and Percent Fines.

Calculated with MTCA Stat97 Background Module (BCKG97.xlt) or standard Excel formula (*); mg/Kg dry weight.

Lake Group and Parameter	Data Distribution	90th Perc.	50th Perc. (Median)	4x50th	Mean	Min.	Max.
Western Lakes (N=9)							
Antimony*	nonparametric	**3.5**	1.2	4.6	1.6	0.67	4.2
Lead	lognormal	**157**	67	270	80	27	190
Cadmium	lognormal	**4.3**	1.7	6.9	2.1	0.77	6.2
Arsenic*	nonparametric	**17**	11	43	12	7.1	28
Mercury	lognormal	**0.251**	0.127	0.506	0.139	0.052	0.208
Zinc	lognormal	**199**	75	302	91	20	215
Eastern Lakes (N=6)							
Antimony	lognormal	**0.14**	0.05	0.19	0.06	0.02	0.13
Lead	lognormal	**31**	18	71	19	9.9	27
Cadmium	normal	**0.63†**	0.49	2.0	0.49	0.24	0.65
Arsenic	lognormal	**6.7**	4.6	18	4.7	3.2	6.1
Mercury	lognormal	**0.117**	0.061	0.244	0.066	0.031	0.103
Zinc*	nonparametric	**98**	63	251	72	50	117
All Lakes (N=15)							
Zinc	lognormal	**157**	73	291	84	20	215
Copper	nonparametric	**47**	20	79	22	8.1	77
Chromium	lognormal	**28**	12	48	15	3.8	53
Barium	nonparametric	**242**	98	390	116	32	333
Manganese	lognormal	**1,032**	271	1,085	466	69	3,010
Iron	lognormal	**40,989**	13,538	54,154	17,096	2,780	43,500
% Fines	lognormal	**88**	60	240	62	40	99

†80th percentile for normally distributed data.

Table 32. Background Values (bold font) for Aquatic Sediments in Northeast Washington Lakes: Organic Compounds and Total Organic Carbon.

Calculated with MTCA Stat97 Background Module (BCKG97.xlt); ng/Kg dry weight.

Lake Group and Parameter	Data Distribution	90th Perc.	50th Perc. (Median)	4x50th	Mean	Min.	Max.
Western Lakes (N=9)							
TCDD (nd = 0)	normal	**4.0†**	2.6	10	2.5	0.21	4.8
TCDD (nd = 1/2 DL)	normal	**4.0†**	2.6	10	2.6	0.24	4.8
Eastern Lakes (N=6)							
TCDD (nd = 0)	lognormal	5.6	1.1	**4.4**	1.8	0.20	5.3
TCDD (nd = 1/2 DL)	lognormal	5.6	1.2	**4.6**	1.8	0.2	5.29
All Lakes (N=15)							
Total PCBs (nd = 0)	nonparametric	**6,229**	2,591	10,365	2,536	112	8,335
Total PCBs (nd = 1/2 DL)	lognormal	8,151	1,594	**6,375**	2,573	131	8,420
PCB TEQs (nd = 0)	nonparametric	0.18	0.01	**0.03**	0.035	0	0.429
PCB TEQs (nd = 1/2 DL)	lognormal	**0.33**	0.09	0.35	0.13	0.01	0.48
TCDD TEQs (nd=0)	lognormal	6.7	1.5	**5.9**	2.2	0.20	5.3
TCDD TEQs (nd = 1/2 DL)	lognormal	6.5	1.5	**6.1**	2.2	2.1	5.3
Total PBDEs (nd = 0)	lognormal	**1,774**	554	2,216	733	101	1,946
Total PBDEs (nd = 1/2 DL)	normal	**1,304†**	850	3,400	850	132	1,953
TOC	normal	**19†**	14	55	14	3.5	24

†80th percentile for normally distributed data.

The MTCA background module first assesses the distribution of values in the data set (lognormal, normal, or nonparametric) and then calculates percentiles based on that distribution. Some of the percentiles in Tables 31 and 32 therefore differ slightly from those presented earlier in this report, which are based on simple ranks rather than data distribution.

As previously discussed (see Results), antimony, lead, cadmium, arsenic, mercury, and TCDD differed significantly between the western and eastern lakes. In view of this finding, along with the presence of known or suspected sources of contamination, summary statistics were calculated separately for these chemicals. The western and eastern lakes data were pooled for the other chemicals. Because zinc contamination is a particular issue for the Lake Roosevelt vicinity, zinc statistics are provided for all three waterbody groupings, even though the concentration differences between western and eastern lake sediments were not statistically significant. The St. Joe River sample was excluded from all calculations.

Non-detects and outliers were handled as follows:

- Percentiles and other statistics for organic compounds were calculated in two ways:
  - Using half the method detection limit for non-detects, following MTCA and recent practice of Ecology's Toxics Cleanup Program (Bradley, 2010).

- Setting non-detects equal to zero, a common convention followed in earlier parts of this report.

- Although a few far outside values were observed for certain chemicals, these were included in the calculations since MTCA makes no provisions for excluding outliers that cannot be shown to be errors.

The values presented in Tables 31 and 32 are intended to aide in site evaluations and to improve current understanding of the *area* and *natural* chemical background in northeast Washington lake environments. In general, several of the western lakes show metal values that are representative of *area background* conditions; specifically, sediments from these lakes indicate influence from industrial pollution sources. Within the precision of the sampling methods applied for this study, the eastern lakes tend to return samples more consistent with *natural background* lake sediment conditions for northeast Washington. These findings are based on field collection methods that were consistently applied to all lakes included in this study.

One sediment sample from a free-flowing river (St. Joe River) also was collected as part of this study. This sample is intended for general comparison purposes to help benchmark background from this undeveloped, forested headwater watershed of the Spokane River basin. The St. Joe River sediment sample is believed to provide a representative indication of *natural background* concentrations for this type of moderate-sized riverine environment. The geochemical, biological, contaminant cycling, and hydraulic conditions and processes within river environments differ from those encountered in the upland lakes.

General sediment characterization and sediment toxicity studies conducted in accordance with the provisions of the MTCA (WAC 173-340) often focus on the top 10 cm of the sediment column as the primary target zone for sample collection. This depth interval is often associated with the most biologically active portion of the sediment column, both in terms of benthic organism abundance and as foraging habitat for certain fish species.

Ecology believes that the sediment collection methods used in this study have provided a representative background data set for this portion of northeast Washington. Resource managers and decision makers who are involved with sediment quality assessments at other upland sites or waterbodies in eastern and northeastern Washington may find these background data to be useful for a variety of project-specific needs. The data for this project are available through Ecology's Environmental Information Management System (EIM) for those wanting to analyze it in other ways (www.ecy.wa.gov/science/data.html).

Conclusions

Present-day, regional-scale background concentrations of potentially toxic metals and organic compounds have been characterized for aquatic sediments in northeast Washington, based on sampling at 14 upland lakes in northeast Washington, one lake in northwest Idaho, and one moderate-sized river in north Idaho. The waterbodies selected for sampling were judged to be minimally impacted by local human activities. Comparison with other studies suggests several of these waterbodies sampled are representative of *natural background*, except as noted below.

Elevated concentrations of antimony, cadmium, lead, arsenic, mercury, and zinc were observed in the sediments of lakes from the western part of the study area along the upper Columbia River. The probable source of contamination is historical transboundary air pollution from the Teck Cominco smelter in British Columbia and potentially, to a lesser extent, localized contribution from a smaller historic smelter in Northport, Washington. Antimony and cadmium substantially exceed sediment quality guidelines in several of these western lakes. TCDD (dioxin) was also higher in the western lakes, possibly related to combustion sources.

The eastern lakes had comparatively lower levels of these and other contaminants, not much above historical benchmark background data observed in the Black Lake age-dated sediment core samples.

Sediment concentrations of barium, chromium, copper, manganese, iron, PCBs, TCDD TEQs (toxic equivalents), and PBDEs were broadly similar across the study area.

Recommendations

As a result of this 2010 study, the following recommendations are made:

1. *Area* and *natural background* calculated values for metal and organic contaminants in northeast Washington lake sediments (Tables 31 and 32) should be referred to for possible use at MTCA cleanup sites, screening contaminant data, risk assessments, and other applications.

2. Further investigation of lakes within the western part of the study area should be considered to better assess potential impacts caused by selected contaminants. Concentrations of certain metals are shown to exceed various sediment quality guidelines by a factor of 2 or more in several of the western lakes sampled within this impacted area. Sediments from several western lakes sampled exceed *natural background* and should be considered for further investigation to better evaluate the potential for toxicity to benthic organisms and other aquatic life. Williams, Cedar, Davis, Ellen, and Pepoon lakes are recommended for additional testing.

3. Sediments from other northeast Washington lakes located within the historic sulfur dioxide footprint of the Trail smelter plume should be considered for investigation to better determine the spatial extent and severity of metals emission influences. Age-dated sediment cores would assist in establishing the history of pollution, sediment profiles, and the metals deposited as they relate to sediment aquatic life.

4. The chemical background for freshwater sediments in other regions of Washington State is a current data gap that should be addressed, to the extent that results of the present study prove useful.

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Appendices

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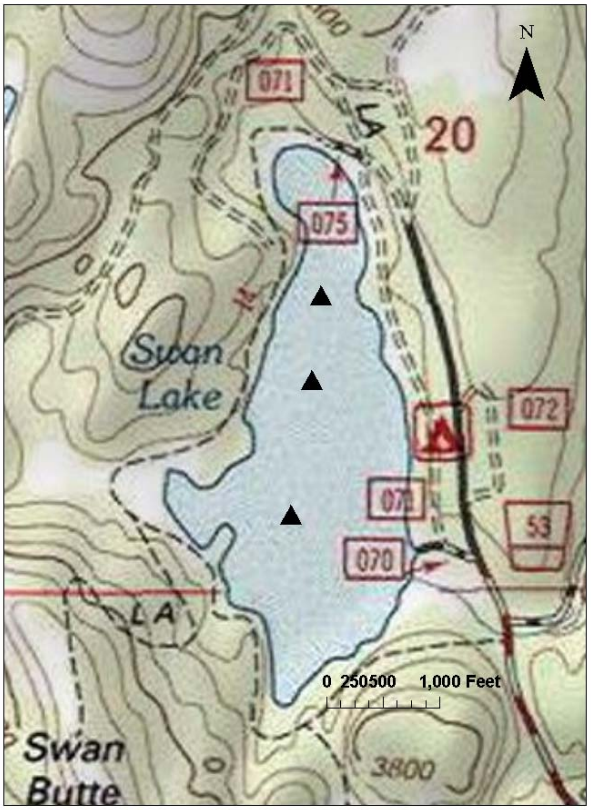
Appendix A. Location of Sediment and Fish Samples for the Northeast Washington Lakes Background Studies

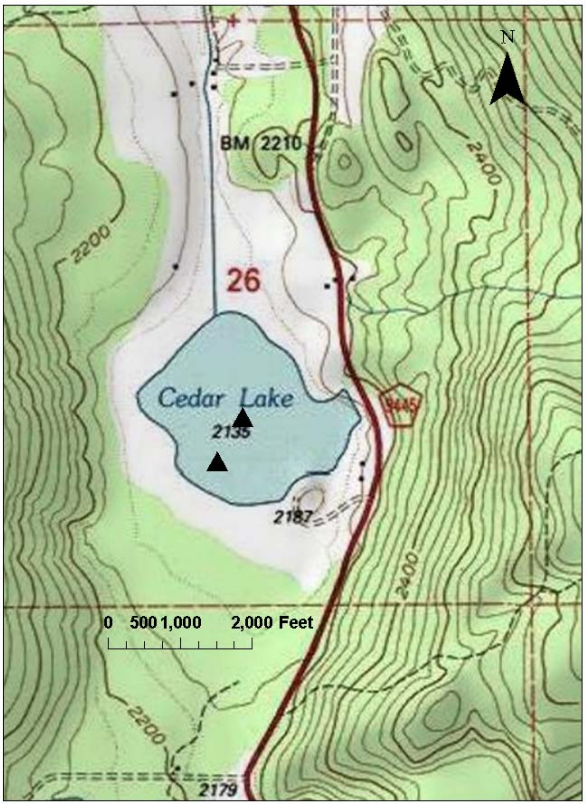
Table A-1. Sediment Samples (center of sample transect, NAD83).

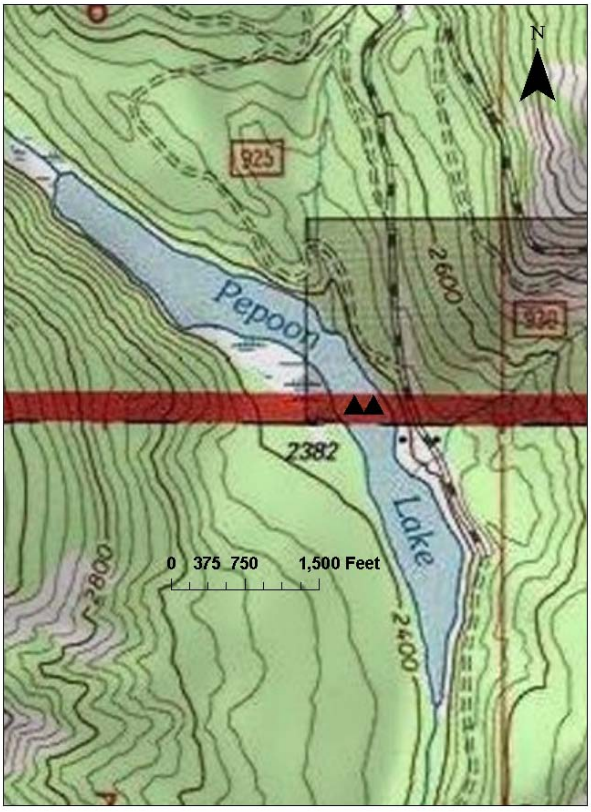
Waterbody	Date	Sample No.	Latitude	Longitude	Depths of Composite Sample (ft.)		
Williams	20-Sep-10	1009070-01	48.7568	-117.9674	41	26	16
Summit	20-Sep-10	1009070-02	48.9596	-118.1274	35	24	- -
Pierre	20-Sep-10	1009070-03	48.9010	-118.1389	75	50	25
Davis	20-Sep-10	1009070-05	48.7376	-118.2308	45	35	20
S. Twin	21-Sep-10	1009070-04	48.2676	-118.3934	55	38	19
Ellen	21-Sep-10	1009070-06	48.5026	-118.2529	34	23	- -
Swan	21-Sep-10	1009070-08	48.5141	-118.8380	99	63	31
St. Joe	28-Sep-10	1010041-01	47.202	-115.5160	1	- -	- -
Bayley	5-Oct-10	1010042-04	48.4187	-117.6625	17	12	- -
Browns	6-Oct-10	1010042-03	48.4384	-117.1909	21	17	8
Bead	7-Oct-10	1010042-01	48.3125	-117.1011	75	50	25
Cedar	18-Oct-10	1010043-03	48.9417	-117.5949	23	17	- -
Leo	19-Oct-10	1010043-01	48.6472	-117.4971	34	24	12
Sullivan	20-Oct-10	1010043-02	48.7974	-117.2921	75	51	24
Upper Priest	21-Oct-10	1010043-04	48.7742	-116.8750	74	50	24
Pepoon	18-Nov-10	1011041-01	48.9008	-117.8924	29	20	- -

Figure A-1. Maps of Sediment Grab Locations for the Northeast Washington Lakes Background Study, 2010.
Base map credit: ESRI ArcGIS Online and data partners including USGS and © 2007 National Geographic Society.

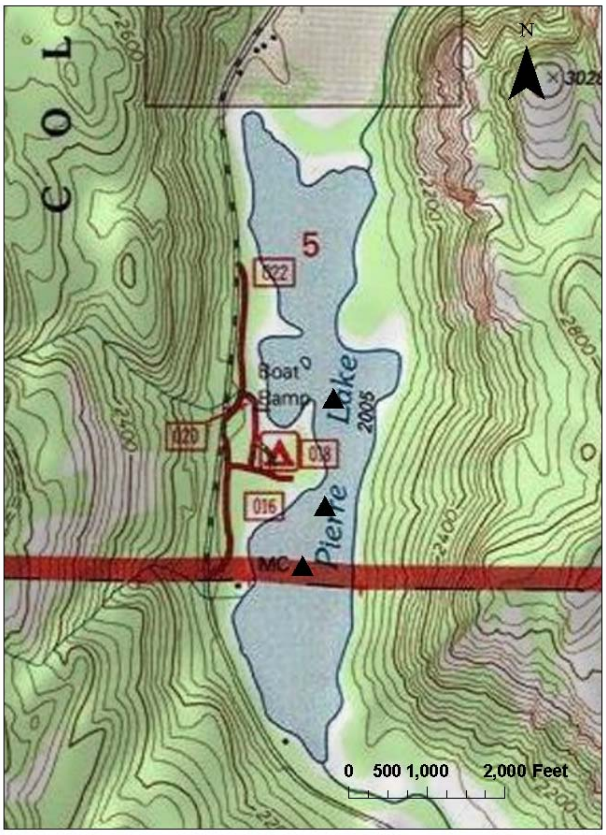
(See the following four pages.)

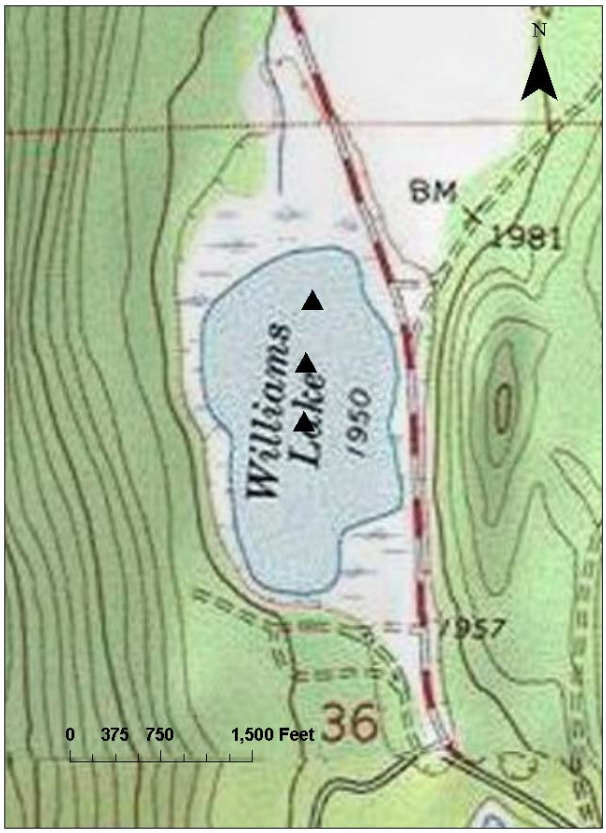

N
071
LA
20
075
Swan
Lake
072
073
53
070
LA
0 250500 1,000 Feet
Swan
Butte
3800

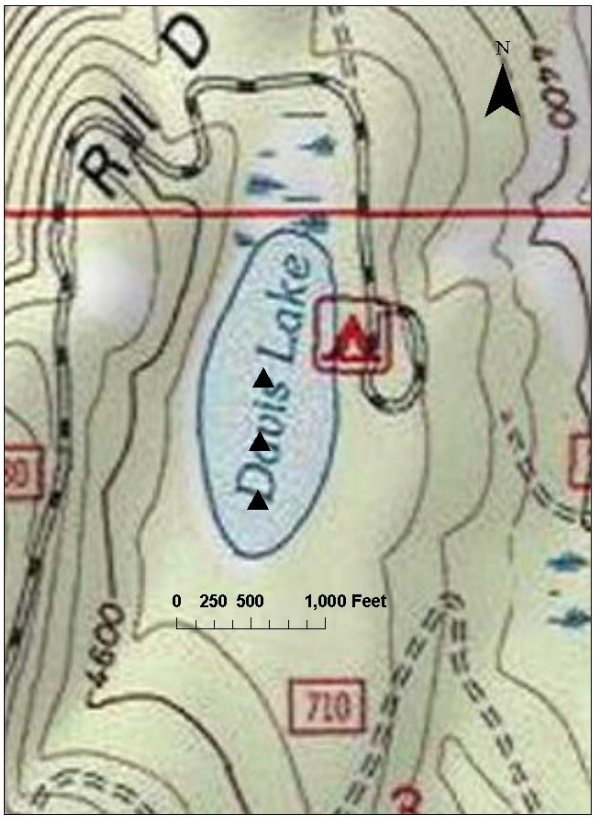

N
BM 2210
2400
2200
26
Cedar Lake
2135
2187
9445
2400
0 500 1,000 2,000 Feet
2200
2179

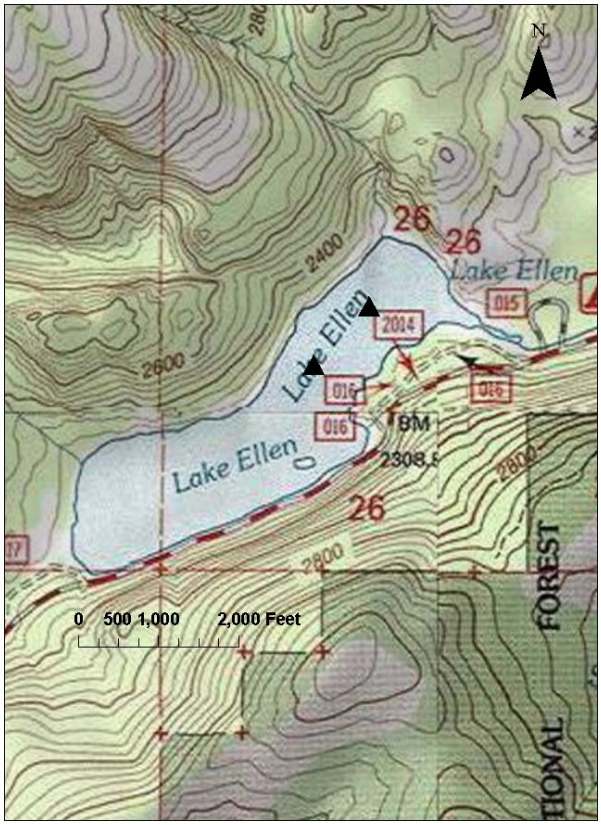

N
925
2600
Pepoon
926
2382
Lake
0 375 750 1,500 Feet
2800
2400

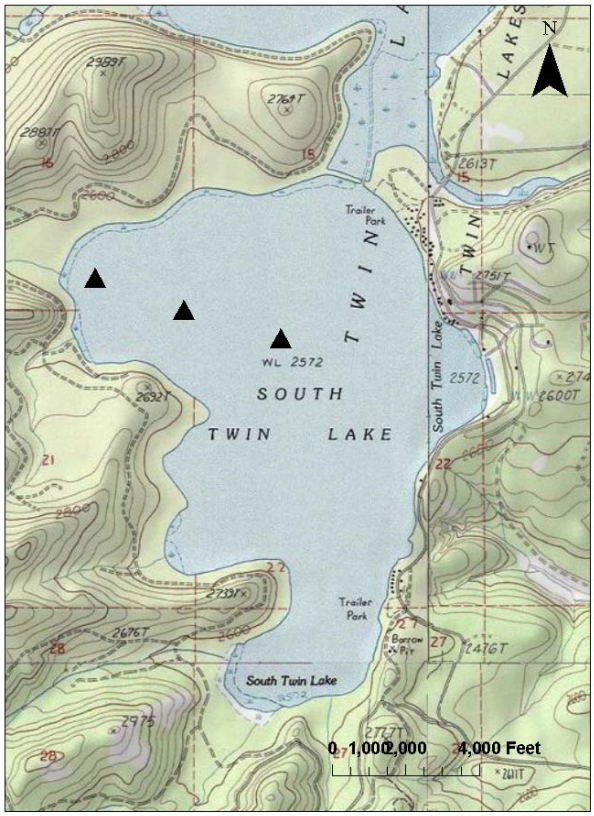

N
930
2600
WC
WC
WC
Summit
Lake
WC
036
042
0 250 500 1,000 Feet

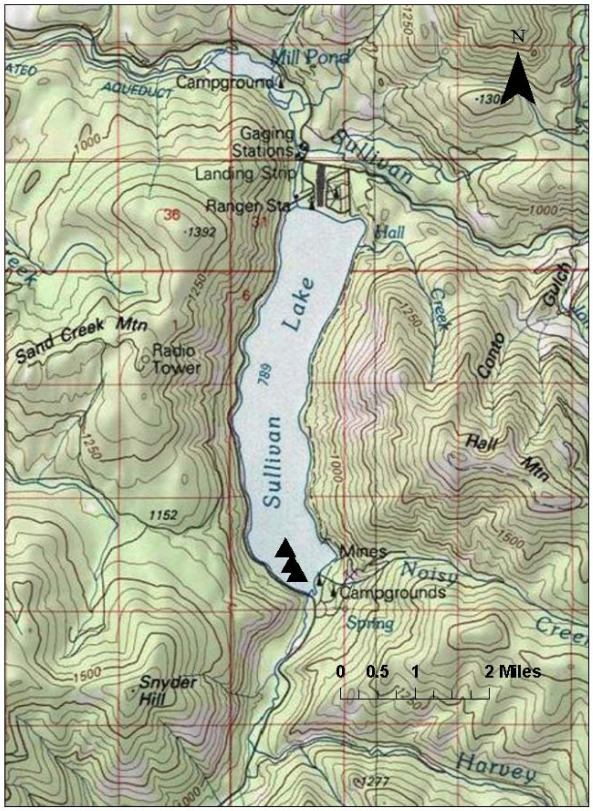

N
COLO
5
822
Boat Ramp
Lake
2005
820
018
016
Pierre
MC
2600
2400
2200
0 500 1,000 2,000 Feet


N
BM
1981
Williams Lake
1950
1957
36
0 375 750 1,500 Feet

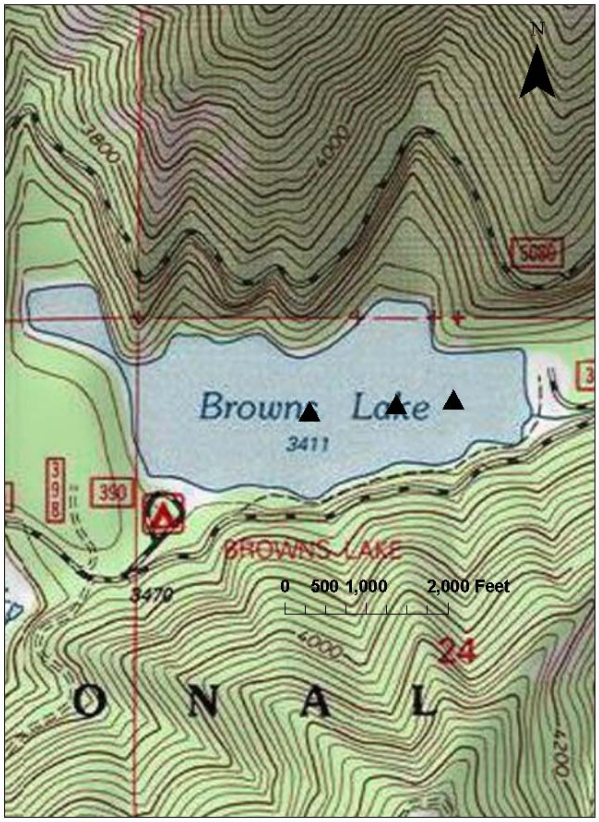

N
RIO
4600
Devils Lake
0 250 500 1,000 Feet
710

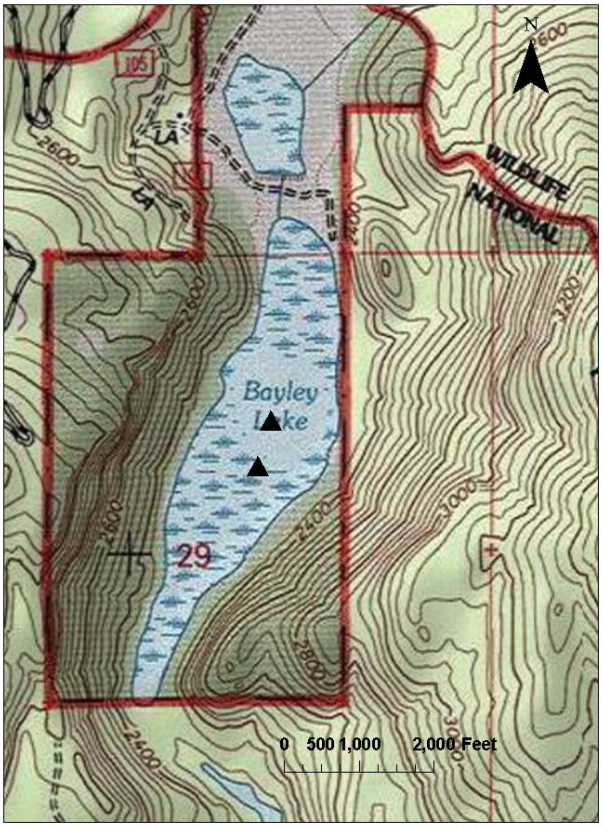

N
26
26
Lake Ellen
2400
2600
2014
015
016
016
016
Lake Ellen
BM
2308.5
Lake Ellen
2800
26
2800
0 500 1,000 2,000 Feet
FOREST

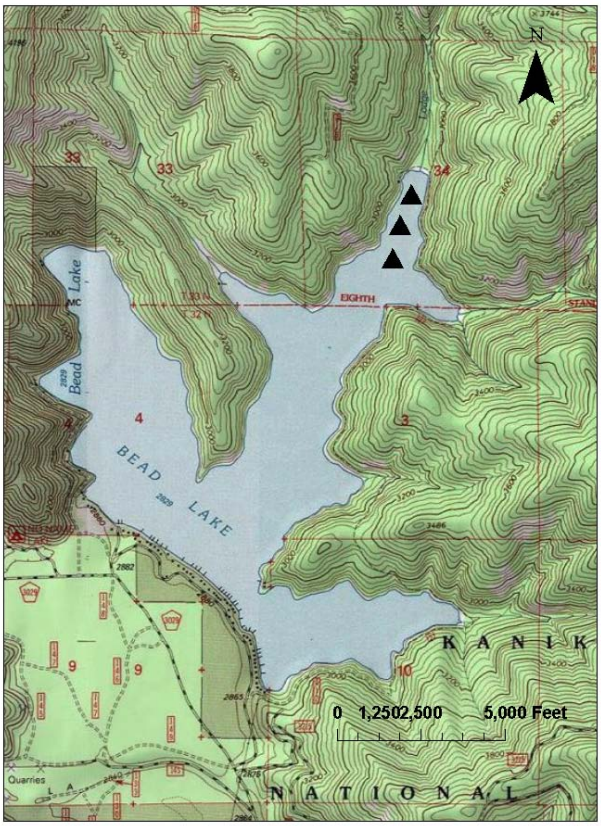

N
SOUTH
TWIN LAKE
WL 2572
Trailer Park
South Twin Lake
0 1,000 2,000 4,000 Feet

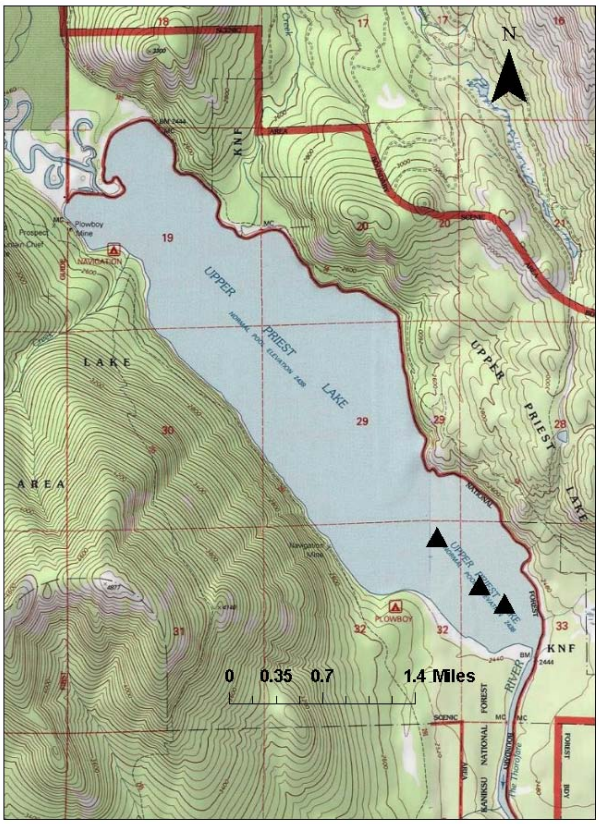

N
Mill Pond
Campground
Gaging Stations
Landing Strip
Ranger Sta
Sullivan Lake
Sand Creek Mtn
Radio Tower
Hall Mtn
Mines
Campgrounds
Noisy Creek
Spring
Snyder Hill
Harvey
0 0.5 1 2 Miles


N
505
506
Boat Ramps
Lake Leo
Little Pend Oreille Lakes
0 375 750 1,500 Feet


N
Brown Lake
3411
BROWNS LAKE
3470
0 500 1,000 2,000 Feet


N
WILDLIFE
NATIONAL
Bayley
Lake
29
0 500 1,000 2,000 Feet


N
Lake
BEAD LAKE
EIGHTH
K A N I K
N A T I O N A L
0 1,250 2,500 5,000 Feet


N
UPPER PRIEST LAKE
LAKE
AREA
KNF
0 0.35 0.7 1.4 Miles

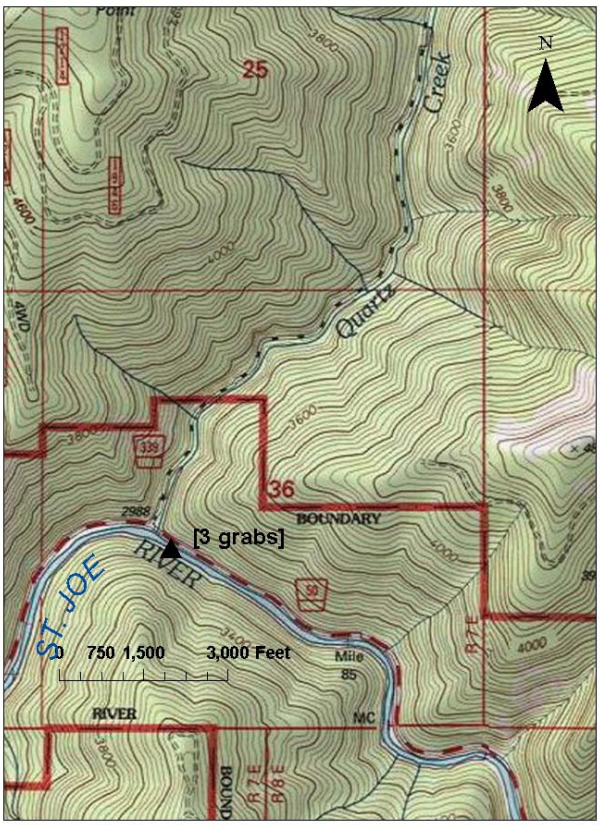

N
Creek
Quartz
25
36
BOUNDARY
[3 grabs]
ST. JOE
RIVER
0 750 1,500 3,000 Feet
Mile 85
MC

Table A-2. Fish Samples.

Waterbody	Species	Date	Collector	Sample No. 1102018-	Number Individuals	Tissue Analyzed
Washington						
Swan Lake	Rainbow trout	11-Oct-10	Colvilles	1	4	Fillet
Pierre Lake	Kokanee			3	1	Fillet
	Bluegill			4	2	Fillet
	Largemouth bass			5	3	Fillet
Ellen Lake	Rainbow trout			8	2	Fillet
	Bluegill			10	2	Fillet
South Twin Lake	Rainbow trout	21-Oct-10		11	2	Fillet
	Eastern brook trout			12	5	Fillet
	Largemouth bass			13/36/38	7	Fillet
Cedar Lake	Rainbow trout	18-Oct-10	Ecology	14/37	5	Fillet
Sullivan Lake	Kokanee	20-Oct-10		15	5	Fillet
	Largescale sucker			16	5	Whole body
	Tiger trout			39	5	Fillet
	Burbot			17	3	Fillet
	Brown trout			18	2	Fillet
Leo Lake	Black crappie	19-Oct-10		19	6	Fillet
	Rainbow trout			20	2	Fillet
	Yellow perch			21/33	10	Fillet
Bayley Lake	Rainbow trout	5-Oct-10		22/34/35	8	Fillet
Browns Lake	Cutthroat	6-Oct-10		23	5	Fillet
Bead Lake	Largescale sucker	7-Oct-10		24	3	Whole body
	Kokanee			25	1	Fillet
Idaho						
Upper St. Joe River	Cutthroat	28-Aug-10	Ecology	27	5	Fillet
	Mountain whitefish			28	4	Fillet
Upper Priest Lake	Lake trout	1-Oct-10	Idaho F&G	29	5	Fillet
	Smallmouth bass			30	5	Fillet
	Largescale sucker			31	5	Whole body

Colvilles: Colville Confederated Tribes.
Ecology: Washington State Department of Ecology.
Idaho F&G: Idaho Fish & Game.

Table A-2 continued.

Waterbody	Species	Date	Collector	Sample No. 1106039-	Number Individuals	Tissue Analyzed
	Largemouth bass -sm			1	3	fillet
	Largemouth bass -lg	23-May-11		2	3	fillet
	Largemouth bass			3	6	fillet
Colville River	Rainbow trout	10-May-11		4	4	fillet
	Largescale sucker		Ecology	5	2	whole
Bead Lake	Kokanee	11-May-11		6	5	fillet
	Yellow perch			7	6	fillet
Jumpoff Joe Lake	Brown trout	10-May-11		8	5	fillet
	Largemouth bass			9	2	fillet
	Smallmouth bass			10	4	fillet
	Brown trout			11	5	fillet
	Largescale sucker			12	5	whole
Pend Oreille River	Northern pike - sm	17-May-11	Kalispels	13	5	fillet
	Northern pike - med			14	5	fillet
	Northern pike - lg			15	2	fillet

Kalispels: Kalispel Tribe of Indians.

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Appendix B. Human and Mammalian Toxic Equivalency Factors for Dioxins, Furans, and PCBs

Table B-1. Human and Mammalian TEFs for Dioxins, Furans, and PCBs.

Compound	WHO 2005 TEF*
Chlorinated Dibenzo-p-dioxins	
2,3,7,8-TCDD	1
1,2,3,7,8-PeCDD	1
1,2,3,4,7,8-HxCDD	0.1
1,2,3,6,7,8-HxCDD	0.1
1,2,3,7,8,9-HxCDD	0.1
1,2,3,4,6,7,8-HpCDD	0.01
OCDD	0.0003
Chlorinated Dibenzofurans	
2,3,7,8-TCDF	0.1
1,2,3,7,8-PeCDF	0.03
2,3,4,7,8-PeCDF	0.3
1,2,3,4,6,7,8-HpCDF	0.1
1,2,3,4,7,8,9-HpCDF	0.1
1,2,3,4,7,8-HxCDF	0.1
1,2,3,6,7,8-HxCDF	0.1
1,2,3,7,8,9-HxCDF	0.01
2,3,4,6,7,8-HxCDF	0.01
OCDF	0.0003
Non-ortho substituted PCBs	
PCB-77	0.0001
PCB-81	0.0003
PCB-126	0.1
PCB-169	0.03
Mono-ortho substituted PCBs	
PCB-105	0.00003
PCB-114	0.00003
PCB-118	0.00003
PCB-123	0.00003
PCB-156	0.00003
PCB-157	0.00003
PCB-167	0.00003
PCB-189	0.00003

*Van den Berg, M., L.S. Birnbaum, M. Denison et al., 2006. The 2005 World Health Organization Re-Evaluation of Human and Mammalian Toxic Equivalency Factors for Dioxins and Dioxin-like Compounds. Toxicological Sciences 93: 223 - 241.

Appendix C. Sediment Chemistry Data for Reference Lakes in Northeast Washington and British Columbia

Table C-1. Sediment Chemistry Data for Reference Lakes in NE Washington and British Columbia (means show for N>1).

Waterbody	Location	Date	N=	Sb		Pb	Cd	As		Hg		Zn	T-PCBs		Reference
McDowell Lake	Spokane Co.	2008	3	0.11		6.2	0.14	2.4		0.028	U	23	3.5	J	1
S. Skookum Lake	Pend Oreille Co.	2008	3	0.27		27	0.53	5.8		0.27		62	4.8		1
Buffalo Lake	Okanogan Co.	2008	1	NA		22	1.2	NA		0.080		85	3.7		2
Lower Arrow Lake	British Columbia	2001	1	NA		12	0.46	2	U	0.004	U	27	NA		3
Lower Arrow Lake (beach)	British Columbia	2001	2	1.0		36	0.35	5.8		0.045		101	NA		4
Lake Roosevelt (bank)*	Ferry/Stevens Co.	2001	3	0.60		15	0.20	6.6		0.020		57	NA		4
Black Lake	Stevens Co.	2009	1	0.21		40	0.58	3.8		0.17		92	19	U	5
Upper Twin Lake	Lincoln Co.	2009	1	0.20	U	8.9	0.16	6.5		0.04		42	8.3	U	5
Amber Lake	Spokane Co.	2009	1	0.20	U	10	0.17	2.7		0.06		47	12	U	5
Western Lakes	Ferry/ Stevens Co.	2010	9	1.2		59	1.5	11		0.13		76	2.6		present study
Eastern Lakes	Stevens/ Pend Oreille Co.	2010	6	0.05	J	20	0.52	4.5		0.069		63	1.4		present study

*glacial sediments above high-water elevation on upper banks
U: not detected
J: estimated value
NA: not analyzed

References

1. Sloan and Blakley (2009)
2. Era-Miller (2004)
3. Era and Serdar (2001)
4. Majewski et al. (2003)
5. Seiders (2009)

Appendix D. Glossary, Acronyms, and Abbreviations

Glossary

Anthropogenic: Human-caused.

Area background: Concentrations of substances that are consistently present in the environment in the vicinity of a site and which are the result of human activities unrelated to releases from that site.

Background site: A waterbody thought to exhibit relatively low impact from human activities. Reference site.

Benthic: Bottom-dwelling organisms.

Bioaccumulative pollutants: Pollutants that build up in the food chain.

Congener: One of many variants or configurations of a common chemical structure. For example, the PCBs analysis includes 209 individual compounds or congeners.

Fingerprint: A multi-parameter chemical signature (distinctive chemical pattern) used to characterize the source of contaminants in an environmental sample or to differentiate the sample from contaminants present in samples representing background conditions.

Grab sample: A discrete sample from a single point in the water column or sediment surface.

Natural background: The concentration of a constituent that occurs naturally in the environment and has not been influenced by localized human activities.

Outlier: A number that deviates markedly from other numbers in a sample population.

Percent fines: Sediment texture.

Point source: Source of pollution that discharge at a specific location from pipes, outfalls, and conveyance channels to a surface water. Examples of point source discharges include municipal wastewater treatment plants, municipal stormwater systems, industrial waste treatment facilities, and construction sites that clear more than 5 acres of land.

Sediment: Solid fragmented material (soil and organic matter) that is transported and deposited by water and covered with water (example, river or lake bottom).

Sediment core: Vertical sediment sample.

Surficial sediment: Recently deposited sediment.

Acronyms and Abbreviations

B.C.	British Columbia, Canada
CSL	cleanup screening levels
Ecology	Washington State Department of Ecology
EIM	Environmental Information Management database

EPA	U.S. Environmental Protection Agency
ERO	Eastern Regional Office (Department of Ecology)
GIS	Geographic Information System software
MDL	method detection limit
MEL	Manchester Environmental Laboratory
MTCA	Model Toxics Control Act
N	number of samples
PBDEs	polybrominated diphenyl ethers
PBT	persistent, bioaccumulative, and toxic substance
PCBs	polychlorinated biphenyls
PCDDs	polychlorinated dibenzodioxins
PCDFs	polychlorinated dibenzofurans
PEC	probable effect concentration
PQL	practical quantitation limit
RM	river mile
RPD	relative percent difference
SO_2	sulfur dioxide
SOP	standard operating procedures
SQS	sediment quality standard
TCDD	2,3,7,8-tetrachlorodibenzo-p-dioxin
TEC	threshold effect concentration
TEQ	toxic equivalent
TOC	total organic carbon
USFS	U.S. Forest Service
USGS	U.S. Geological Survey
WAC	Washington Administrative Code
WDFW	Washington Department of Fish & Wildlife
WRIA	Water Resource Inventory Area
WSPMP	Washington State Pesticide Monitoring Program

Metals

As	arsenic
Ba	barium
Cd	cadmium
Cr	chromium
Cu	copper
Fe	iron
Hg	mercury
Mn	manganese
Pb	lead
Sb	antimony
Zn	zinc

Units of Measurement

cm	centimeter
ft	feet

kg	kilograms, a unit of mass equal to 1,000 grams.
mg	milligrams
mg/Kg	milligrams per kilogram (parts per million)
mm	millimeters
ng/Kg	nanograms per kilogram (parts per trillion)