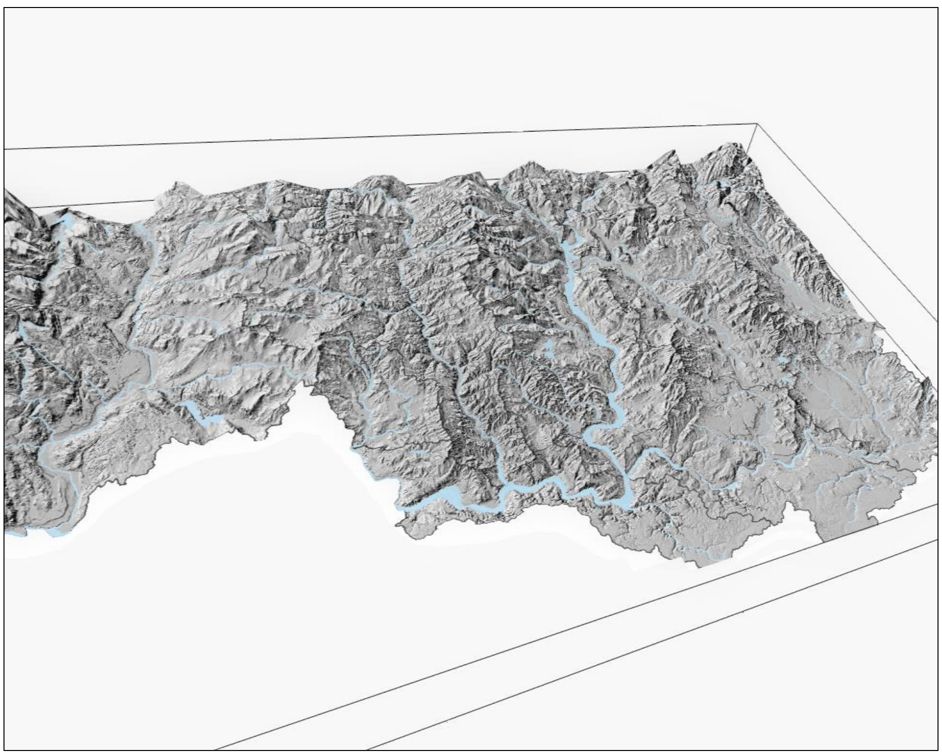
Upland Regional Soil Background Characterization for Select Metals in Northeast Washington Watersheds



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Water Resource Inventory Area (WRIA) numbers for the study area:

- 49 – Okanogan
- 51 – Nespelem
- 52 – Sanpoil
- 53 – Lower Lake Roosevelt
- 54 – Lower Spokane
- 55 – Little Spokane
- 58 – Middle Lake Roosevelt
- 59 - Colville
- 60 – Kettle
- 61 – Upper Lake Roosevelt
- 62 – Pend Oreille

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Cover photo: Rendering of the northeast Washington data-use area.

Upland Regional Soil Background Characterization for Select Metals in Northeast Washington Watersheds

by

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Acknowledgments

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- Dr. Nairanjana Dasgupta and the team at the Center for Interdisciplinary Statistics Education and Research at Washington State University in Pullman, WA.

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- Erika Bronson and Charles Gruenenfelder of the Toxics Cleanup Program at the Washington State Department of Ecology.

Executive Summary

Naturally occurring concentrations of metal and metalloid elements, or natural background, in upland soils is an important baseline environmental metric for assessing the effects and influence of human activities. This background soil metals analysis is intended to guide the application of environmental regulations in northeast Washington State and offers a framework for possible application in other areas.

The analysis establishes natural background metal values that represent upper-percentile thresholds in soils within 11 state-defined watersheds (Water Resource Inventory Areas) for 18 metals and metalloids. The geographic boundaries of this natural background analysis are selected to guide and inform cleanup decisions and other environmental work occurring in the greater Upper Columbia River region. In support of this analysis, the Washington State University Center for Interdisciplinary Statistics Education and Research performed detailed exploratory analyses, metal-specific evaluations, and provided recommendations for data pooling.

Table 1 summarizes the final 90th percentile concentration ranges for application across the study area. Median (50th percentile) and 75th percentile values from each reported range are also provided. The use of a concentration range accounts for the inherent diversity and variability within natural surfaces sampled across the 10,000-square-mile study area that encompasses different geologic settings, soil assessment study designs, analytical methods, and other factors. The statistical evaluation conducted establishes that wholesale pooling of the data sources may not be appropriate unless data availability is limited. The background concentration ranges selected for background applications are reported from watershed-specific data sources unless otherwise defined.

Investigator-detected soil concentrations above the 90th percentile range midpoint values (Table 1) are indicative of anthropogenic effects or influences from localized geologic ore mineralized anomalies. For environmental investigators utilizing only partial digestion analytic methods (total recoverable EPA digestion methods), the lower-end values within the Table 1 90th percentile ranges generally may serve as better representation under those applications.

The 90th percentile ranges and associated midpoint results from the uniquely large dataset compiled for this study serve as a resource to regulators, scientists, and others interested in the conditions of surface soils of northeast Washington. The 50th percentile ranges are relevant to those interested in central tendency.

Why Complete this Study?

The practice of combining multiple existing datasets for the collective application to establish natural background metal concentrations has previously been underutilized. An abundance of ground-surface analytic data exists over a broad area of northeast Washington. Many of these datasets were derived in conjunction with federal and state programs intended to evaluate earth resources or environmental conditions. This analysis achieves environmentally relevant upper-bound estimates of natural background metal concentrations by combining and comparing data from an array of soil assessment studies with different sample site selection criteria, sample collection and analysis methods, and sample sizes.

The Washington State Department of Ecology's Toxics Cleanup Program previously performed and published data-limited, soil sample collection results to estimate natural background concentrations statewide (Ecology, 1994). High-density regional sample collection and analysis can be cost prohibitive and high-resolution geographic coverage difficult to achieve. By comprehensively screening and compiling existing region-specific metal concentrations data from multiple studies and investigations, this report establishes estimates of upland natural background metals concentrations for environmental regulatory application for surface soils of northeastern Washington State. *Soil* is operationally defined as unconsolidated earth surface materials less than 2 millimeters in size, and a natural medium capable of supporting rooted plants and other upland flora and fauna. Careful examination of sampling and analysis protocols addresses comparability and compatibility of datasets used.

Table 1. Ninetieth percentile background metal concentrations in northeast Washington State.

Analyte	90th Percentile Natural Background Concentration Range[1]	Range Midpoint[1]
Aluminum[2]	24,870 to 81,320	53,095
Antimony	0.49 to 2.03	1.26
Arsenic	3 to 20	12
Barium	432 to 1,137	785
Beryllium	0.7 to 3.3	2
Bismuth	0.2 to 2.1	1.1
Cadmium[2]	0.4 to 0.84	0.44
Chromium	35 to 164	100
Copper	16 to 49	33
Indium	0.05 to 0.06	0.055
Iron	26,950 to 54,900	40,925
Lead[2]	19 to 43	31
Manganese	621 to 2,493	1,557
Mercury[2]	0.02 to 0.15	0.09
Nickel	12 to 50	31
Silver	0.4 to 1.4	0.9
Thallium	0.2 to 0.7	0.5
Zinc[2]	55 to 139	97

[1]All reported concentrations are reported in milligrams per kilogram (mg/kg) and represent 90th percentiles.

[2]Adjusted background concentrations as described in the Final Results and Natural Background Determinations section.

Introduction

An understanding of natural background is necessary for evaluating the extent, magnitude, and potential human or environmental risks caused by anthropogenic pollution. Large datasets are preferred for estimating natural background metals concentrations for watershed or regional scale pollution assessments. The statistical power of larger datasets can overcome inherent variability associated with locale and study objectives. Using existing data from multiple sources, this analysis establishes estimated background metal concentrations obtained from the analysis of upland soil and tributary data collected in 11 Upper Columbia River watersheds of northeast Washington State (Figure 1). Upper-bound background values are presented for 18 metals and metalloids. Several of the evaluated metals are recognized as common inorganic pollutants associated with mineral industry operations.

Model Toxics Control Act Considerations

The Washington State Department of Ecology (Ecology) implements the Model Toxics Control Act (MTCA). Determining natural background requires that samples be collected, to the degree possible, from areas that have the same basic characteristics as the media of concern at a particular site. MTCA and associated guidance also provide minimum sample size requirements and methods for defining natural background conditions (Ecology, 1992).

For state regulatory purposes, *natural background* refers to hazardous substance (metals) concentrations consistently present in the environment in areas that are not influenced by human activities. Estimating natural background concentrations in the study area requires carefully avoiding soil metals data collected in areas where anthropogenic impacts from smelting, mining, and other potential metal-liberating land-use practices. For soil investigations, determining natural background calls for the collection of a minimum of 10 samples (Ecology, 1992). For this report, the term *background* refers to *natural background*, unless otherwise clarified.

In 1994, Ecology collected soil samples and performed data analysis to determine background conditions across the state (Ecology, 1994). The calculated ranges issued in the report routinely inform remedial actions at cleanup sites across the state where site-specific background determinations are not mandated. Ecology (1994) describes a number of applications for background datasets that include estimating background conditions, evaluating waste streams, determining regulatory compliance, and designing remedial investigations and feasibility studies related to the cleanup of contaminated sites. The 1994 study involved the collection of soil and sediment materials from various depths and areas, often avoiding the upper-most surface soil profile and collecting samples no more than 36 inches below ground surface. The approach applied in the 1994 guidance did not emphasize the estimation of natural near-ground-surface upland conditions in northeast Washington, nor the diversity of analytic methods that can be applied by various investigators.

The current study estimates site-specific, near-surface natural metal concentrations in northeast Washington at a regional scale, integrating a broad suite of data, not inclusive of the 1994

Ecology work. Historical metals pollution from smelter emissions in the Upper Columbia River Valley occurred within the study area for much of the 20th century. This includes emissions from a large, active smelter complex in Trail, British Columbia, Canada, and emissions from a smelter facility that operated intermittently in Northport, Washington, from the late 1890s through portions of the 1920s. Historical smelter emissions in the Columbia River Valley resulted in adverse impacts to upland flora from sulfur dioxide (SO_2) exposure and aerial deposition of metal particulate (Scheffer and Hedgcock, 1955). Figure 1 depicts the total estimated area of SO_2 injury to upland vegetation. Smelter metal enrichment of shallow soils in the Upper Columbia River Valley has been previously documented within parts of the study area from the U.S.-Canadian border to the municipality of Northport, WA, and beyond. These emissions and associated potential soil contamination are a primary consideration to ensure local metal sources do not bias the data. Shallow, smelter-impacted soils often exhibit high metals concentrations with an analytical fingerprint that is consistent with the type of local smelter emissions (Hart Crowser, 2013a). These elevated concentrations can be a confounding factor for investigators evaluating background conditions at sites that are unrelated to historical smelter emissions in the region.

The data curated for this project and presented in this document have a variety of possible regulatory uses, but principally establish actionable thresholds for maximum natural background concentrations to guide decision making associated with risk, cleanup, and remedial actions in the Upper Columbia River Valley and northeast Washington watersheds. The background concentrations established in this document represent upper-percentile estimates of naturally occurring metals in soils of northeast Washington.

Study Design

Study and Area

Data from independent existing soil assessments performed within the region of study are compiled, evaluated, and presented to provide geographic coverage throughout northeast Washington, while avoiding suspect data collected in areas of local or potential anthropogenic influence.

Soil is operationally defined as unconsolidated earth surface materials less than 2 millimeters in size, and a natural medium capable of supporting rooted plants and other upland flora and fauna. Within the context of assessment, these surface soils reside within 24 inches (typically shallower) of the ground surface at the time of sample collection. The data used for estimating background were collected from a broad geographic area of variable terrain, elevation, and underlying geology to obtain a regional assessment of background conditions. The area of sample coverage is over 10,000 square miles to the north of Highway 2, between the Washington-Idaho border and the east slopes of the Cascade Mountains. Sample collection locations range in elevation from 800 to 7,000 feet above mean sea level. Three-quarters of the samples were collected at elevations below 3,900 feet.

The study area is comprised of eleven state-defined watersheds, or Water Resource Inventory Areas (WRIAs), that are associated with the Columbia River and its tributaries (Figure 1). The final project dataset includes a suite of 18 metals and metalloids for which a sufficient quantity of data were available to achieve a desired level of spatial representation and statistical relevance.

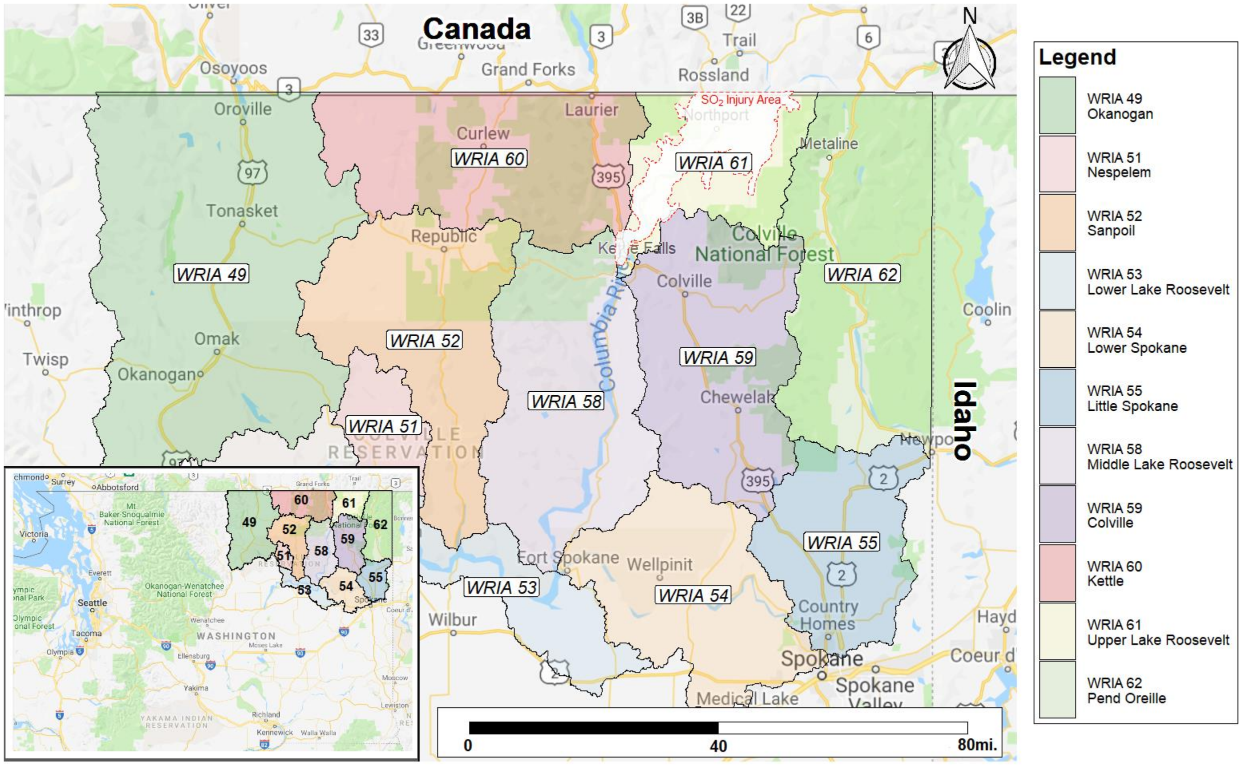

Figure 1. Data use area and associated 11 Washington Water Resource Inventory Areas (WRIAs) evaluated for this study.

Data Selection

The 18 metals of interest in this study are naturally occurring in rocks, soils, and sediments (aluminum, antimony, arsenic, barium, beryllium, bismuth, cadmium, chromium copper, indium, iron, lead, manganese, mercury, nickel, silver, thallium, and zinc). However, some of these metals have an established association with industrial operations that can affect measured concentrations over broad areas.

In brief, the studies and investigations from which data are used to perform this natural background metal/metalloid evaluation can be grouped within three broad categories:

1. Large-scale geochemical studies related to resource exploration.
2. State and federal environmental assessment and site-specific contamination studies.
3. Other geologic, agronomic, ecological, and supplemental research.

A summary of all data used to perform this evaluation as well as source references are included in Appendix A (Table A-1). The selected published soil assessments were performed between 1975 and 2015.

While most of the existing studies included the objective of characterizing soil conditions with minimal anthropogenic influence, additional screening of sample data was performed. Evaluations included:

- Quality assurance/quality control protocols.
- Sample preparation and analytical methods.
- Sample collection depth and location.
- Potential anthropogenic influence to identify and select appropriate data for the estimation of natural soil conditions.

Target Metals

Table 2 summarizes the final selection of metal analytes and the total number of results for each.

Table 2. Total number of records evaluated for background estimation by analyte and Water Resource Inventory Area (WRIA).

Analyte	WRIA 49	WRIA 51	WRIA 52	WRIA 53	WRIA 54	WRIA 55	WRIA 58	WRIA 59	WRIA 60	WRIA 61	WRIA 62	*Total No. Records by Analyte*
Aluminum	366	42	395	249	626	175	753	101	1,442	168	30	*4,347*
Antimony	10	0	4	2	2	2	11	3	2	69	6	*111*
Arsenic	272	32	165	74	185	80	192	14	170	76	24	*1,284*
Barium	29	2	12	8	52	6	65	15	18	49	25	*281*
Beryllium	361	42	174	80	188	80	219	28	189	79	19	*1,459*
Bismuth	10	0	4	2	44	2	4	2	2	0	6	*76*
Cadmium	10	0	4	2	44	2	24	6	8	74	12	*186*
Chromium	362	42	176	80	187	80	222	30	197	85	30	*1,491*
Copper	364	42	154	80	187	79	165	29	196	85	30	*1,411*
Indium	10	0	4	2	2	2	4	2	2	0	6	*34*
Iron	364	42	393	249	619	173	745	99	1,413	162	30	*4,289*
Lead	361	42	174	80	187	80	221	30	196	85	30	*1,486*
Manganese	364	42	395	246	625	175	753	101	1,442	168	30	*4,341*
Mercury	29	2	12	8	11	6	31	13	8	70	18	*208*
Nickel	364	42	175	80	186	79	220	30	195	85	30	*1,486*
Silver	335	40	163	72	135	73	165	14	173	74	6	*1,250*
Thallium	10	0	4	2	44	2	11	3	2	67	6	*151*
Zinc	364	42	175	80	187	79	221	29	196	85	30	*1,488*
Total No. Records by WRIA	*3,985*	*454*	*2,583*	*1,396*	*3,511*	*1,175*	*4,026*	*549*	*5,851*	*1,481*	*368*	*25,379*

NURE-HSSR and Reanalysis of Archived Samples

The National Uranium Resource Evaluation Hydrogeochemical and Stream Sediment Reconnaissance program (NURE-HSSR), initiated by the Atomic Energy Commission in the 1970s, is the largest singular data package in the study and provides extensive geographic coverage of the study area (Figure 2). This data set represents approximately 95 percent of the individual samples evaluated to estimate background, or 83 percent of the total number of records. Under NURE-HSSR, sample locations used in this study were systematically determined and distributed across a broad area of the Upper Columbia River region. During the NURE-HSSR program, focus areas in northeast Washington were selected for a high-density sample collection effort that included portions of the study area.

The NURE-HSSR sample archive maintained by the U.S. Geological Survey (USGS) is one of the largest repositories available. More recent soil and sediment studies have incorporated archived NURE-HSSR samples for analysis using other analytical methods. Key reanalysis studies for this project include Pre-mining Geochemical Background Downstream from Midnite Mine (Church et al., 2008) and a USGS NURE-HSSR Reanalysis Program (Grossman et al., 2004). NURE-HSSR and the USGS Reanalysis Program incorporated aggressive sample digestion methods.

The NURE-HSSR program employed national laboratories and field personnel (Church, 2010b). For this study, the national laboratories primarily involved in Washington, were Savannah River and Lawrence Livermore. Savannah River also sub-contracted supplemental high-quality analysis of NURE-HSSR samples by atomic absorption spectroscopy (AA). In some cases, NURE-HSSR analytical detection limits were not low or reliable enough to determine meaningful estimated background concentrations evaluated for the purposes of this study. In many cases, these analyses resulted in extensive record removals in datasets not well suited for background estimation. Specifically, the NURE-HSSR analytics for determining concentrations of barium, bismuth, indium, mercury, and thallium are of questionable quality or inadequate for quantitating background concentrations in soil and sediment samples. Table B-1c (Appendix B) describes data omitted from background estimation due to inadequate detection limits.

NURE-HSSR samples analyzed at Lawrence Livermore National Laboratory (LLL) resulted in significant analytical limitations (Church, 2010b) and data reporting issues. Although LLL was able to provide quality analyses for aluminum, iron, and manganese, a data reporting issue in the USGS NURE-HSSR database assigned the same sample type code for primary samples, duplicate samples, and blank samples. Because of various analytic and reporting concerns, LLL data are excluded from background estimation. LLL was responsible for the majority of NURE-HSSR samples in the Sandpoint 1x2 degree quadrangle, which covers large portions of Pend Oreille, Stevens, and Ferry counties in northeast Washington.

The introduction of inductively coupled plasma/atomic emission spectroscopy (ICP-AES) and the more recently adopted inductively coupled plasma/mass spectrometry (ICP-MS) methods provided analysis of archived NURE-HSSR samples with improved instrument detection limits and accuracy. Importantly, the USGS NURE-HSSR Reanalysis Program of archived NURE-HSSR samples using ICP-AES methods provides limited coverage of the Sandpoint quadrangle that was previously unfulfilled by shortcomings in the original LLL dataset.

When available, data from the USGS NURE-HSSR Reanalysis Program and the Midnite Mine study underwent a substitution process using a tiered approach based on improved analytical detection, precision, and accuracy. Data from the Midnite Mine study were the optimal source if they were available for reanalyzed archived NURE-HSSR samples at a given sample location, followed by USGS NURE reanalysis data. Silver was not part of the analytical suite during the Midnite Mine study, and the analytical detection limit for silver during the NURE reanalysis study was insufficient to determine background concentrations. Thus, all of the results for silver associated with NURE-HSSR samples are from the original NURE-HSSR study, which represent 93 percent of the silver results in the background dataset.

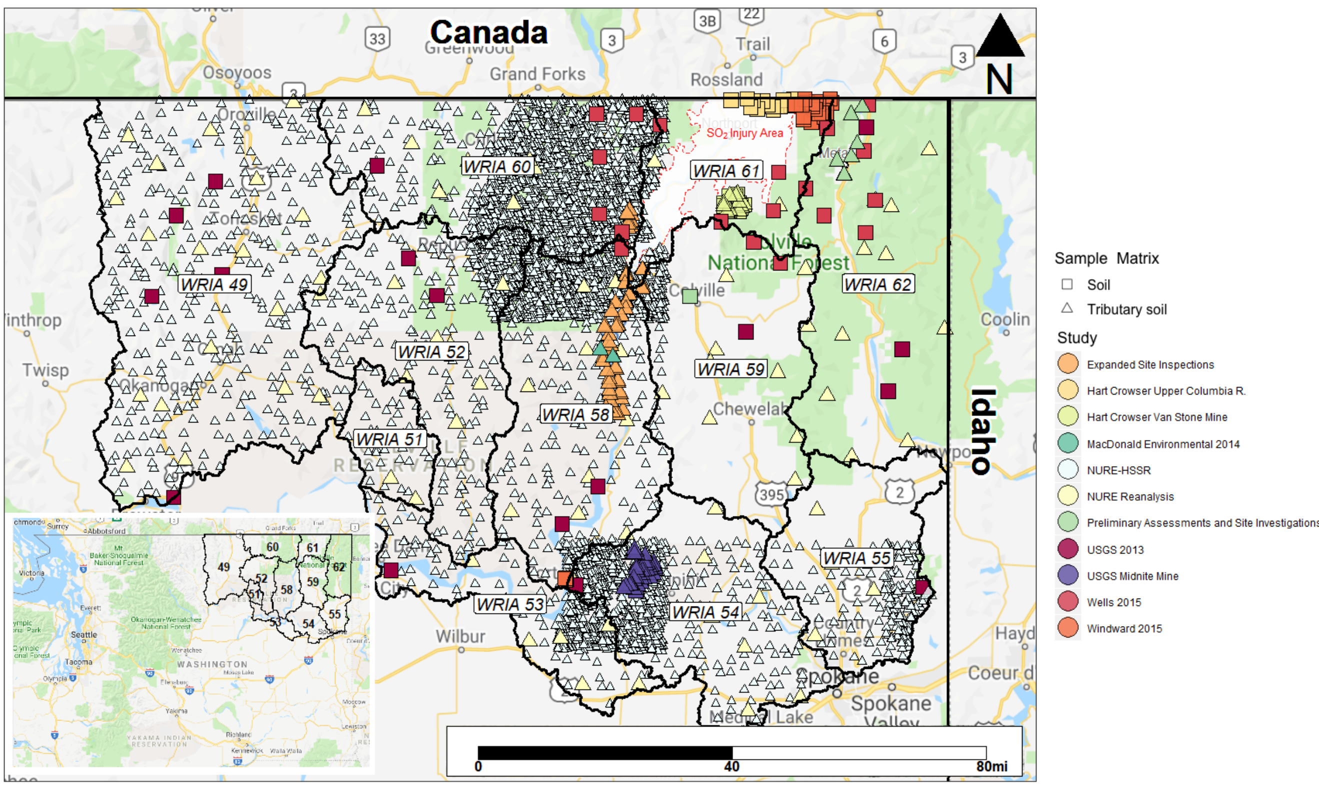

Figure 2. Geographic distribution of samples used for background estimation.

MESL – MacDonald Environmental Services Ltd.
NURE-HSSR – National Uranium Resource Evaluation Hydrogeochemical and Stream Sediment Reconnaissance Program
UCR – Upper Columbia River
USGS – U.S. Geological Survey

Final Sample Screening

After compiling data from existing studies with the appropriate metadata, a final review identified any remaining issues within the dataset to make final decisions about data usability.

As noted previously, soils and sediments within portions of the Upper Columbia River Valley exhibit metal enrichment caused by emissions from historical smelting operations (Ecology, 2013; Hart Crowser, 2013a). Complex air transport and dispersion mechanisms affected the distribution and magnitude of historical smelter emission metals deposition on surface soils in northeast Washington State, impacting terrestrial vegetation (Scheffer & Hedgcock, 1955). This is more recently affirmed by actions under federal authority that resulted in residential yard soil removal actions to protect human health (U.S. Environmental Protection Agency [EPA], 2005; CH2M Hill, 2015; Ramboll, 2017).

After an initial screening step, samples were excluded from the dataset in accordance with predefined geographic screening criteria for the portions of the study area known to be affected by pollution from historical smelting or localized mining activities. Initial screening steps, summarized in Appendix B, involved the removal of sampled materials within a documented historical smelter air pollution (SO_2) impact area, illustrated in Figure 1 (Scheffer & Hedgcock, 1955).

Geographic screening also excluded data from highly impacted areas including current and former mining sites and mineral claims using geographic information systems (GIS) map layers available from the USGS (2005). Ecology did not apply additional geographic exclusions in an effort to retain the utility of the dataset for other environmental applications; the majority of the published assessments previously included such exclusions within their study designs or in their initial sample screening.

Using ArcGIS™, 35 samples were excluded from the background evaluation because sampling locations were within the historical SO_2 injury footprint. The historical SO_2 impact area covers approximately 40 percent of WRIA 61. Samples collected in WRIA 61 that met the geographic screening criteria in Appendix B were advanced for background estimation as part of the final dataset. Twelve samples from the Upper Columbia River Upland Soil Assessment (Hart Crowser, 2013a) were advanced for background estimation despite collection within the SO_2 footprint. The Hart Crowser soil samples were collected at discreet depth intervals from 0–6, 6–12, 12–18, and 12–24 inches. Samples collected from the 0- to 6-inch interval exhibited elevated metals concentrations consistent with smelter impacts. These samples were removed from the final dataset, and samples from deeper intervals were independently retained.

For the 11 WRIAs, an additional 39 samples were removed from the dataset due to close proximity (< 500 meters) to mines, mineral claims, or related facilities. Mines and related facility locations were based on map layers available from the USGS (2005). Any available replicate sample results were averaged prior to analysis.

A final review of sample appropriateness and eligibility was performed once all samples meeting selection criteria were sorted and organized. Ecology reassessed proximity to mines, localized mineralization, and underlying bedrock geology for any remaining samples with anomalous results. While minimal, 15 additional samples were excluded from the background evaluation dataset. Those samples are listed in Appendix B (Table B-3).

Number and Type of Samples

Depending on the metal analyte of interest, the total number of data results per metal ranges from 34 to 4,347. The final dataset advanced to estimate background contains 25,379 results obtained from analyzing 4,374 individual samples collected in northeast Washington. Appendix A summarizes the background dataset. Of the 25,379 total records, NURE-HSSR represents 21,154 results (83 percent), which corresponds to 4,049 individual samples (93 percent), and does not include the archived samples analyzed during the USGS NURE-HSSR Reanalysis Program (Grossman et al., 2004) and USGS Midnite Mine (Church et al., 2008) studies.

NURE-HSSR sample collection protocols called for collecting samples over an approximate depth of 20 centimeters (Smith et al., 2012). The remaining samples in the background dataset were collected from the ground surface to a maximum depth of 24 inches below ground surface. Over 90 percent of the samples were collected from the top 8 inches.

The resulting data represent or are responsive to watershed-scale upland soil and soil erosion processes and are judged appropriate for statistical analysis of upland soil conditions.

Sample Digestion, Analysis, and Detection Limits

Instrument detection limitations were significant for certain analytes within particular soil assessments. Table A-5 (Appendix A) summarizes the analytical methods and typical reporting limits for the final analytical data used in the study. In some cases, complete analyte datasets were removed from the background dataset if the minimum detection limits produced data that were primarily below the detection limits. The proportion of non-detects per metal is summarized in Table A-6 (Appendix A). Analyte datasets removed because they lacked adequately low detection limits are detailed further in Appendix B.

Sample Preparation Considerations

The analytic preparation or digestion protocols used to determine metals from a sample will influence the reported concentrations. NURE-HSSR samples were sieved to 100 mesh (<149 micron [µm]) in size before undergoing crushing, followed by a near total multi-acid digestion, often referred to as a total sample digestion. The NURE-HSSR four-acid soil digestion leaches metals from the sample using a sequence of nitric, hydrochloric, perchloric, and hydrofluoric (HF) acids. The USGS will typically employ total sample digestion methods for geochemical analysis of rock and soil samples, as the digestion sequence will dissolve all but the most resistant matrices. Descriptions of the sequence of the four-acid digestion are widely available (Box et al., 2001). Aggressive USGS digestion typically results in higher measured metal concentrations for a given sample, relative to routine EPA environmental measurement methods.

Eighty-eight of the original NURE-HSSR samples collected in northeast Washington have additional results reported by the USGS (Grossman et al., 2004). Archived NURE-HSSR samples were reanalyzed using improved ICP-AES analytical methodology and detection limits, with similar sample preparation and four-acid digestion methods.

Alternatively, the total recoverable EPA digestion methods for soils (EPA Methods 3005 and 3052) involve dissolution using nitric and hydrochloric acids to analyze dissolved metals in water samples (EPA 3005A), with an optional microwave treatment (EPA 3052) for soil and other organic matrices. This total recoverable method is more common for analyzing metal concentrations in water and soil samples from contaminated cleanup sites. The four-acid USGS method will typically dissolve greater concentrations of metal constituents in samples than the total recoverable methods.

Of the total sample count, over 96 percent of the samples analyzed were composed of a prepared size less than 150 µm, and 95 percent of the samples were prepared using aggressive four-acid digestion methods, described by Box et al. (2001). All samples used in this study were analyzed by laboratories at a size fraction no greater than 2 mm.

Statistical Methods and Analysis

General Approach

The analysis of the background data curated in this study establishes upper-bound regional background thresholds for decision making associated with the smelter-impacted Upper Columbia River Valley and for environmental and toxics-based applications across the Upper Columbia River watersheds of northeast Washington.

The MTCA cleanup regulation typically utilizes specific methods for estimating 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.

This study embodies an exceptionally large data inventory to establish upper-bound background estimates at watershed and regional scales, or for site-specific applications.

Analysis of Background Dataset

The Washington State University (WSU) Center for Interdisciplinary Statistics Education and Research (CISER), performed independent statistical analyses to define, refine, and validate appropriate approaches for estimating background. Ecology provided the refined upland data sets to WSU, following screening steps previously discussed. During exploratory analysis of the dataset, three distinct data 'Source' categories were defined:

NURE: These are the original NURE-HSSR results for metals, as screened.

Non-NURE: Assessments that also provide tributary soil data include:

- USGS reanalysis of NURE archive samples (Grossman et al., 2004).
- Midnite Mine and Blue Creek Drainage Pre-mining Geochemical Background Assessment (Church et al., 2008).
- Preliminary Assessments and Site Investigations Report on Lower Pend Oreille River Mines and Mills (EPA, 2002a).
- Preliminary Assessments and Site Investigations Report on Upper Columbia River Mines and Mills (EPA, 2002b).

- Upper Columbia River Expanded Site Inspection Report Northeast Washington (EPA, 2003).
- Van Stone Mine Remedial Investigation report on background concentrations (Hart Crowser, 2013b).
- Confederated Tribes of the Colville Reservation background (MacDonald, 2014).

Soil: Concentration data were obtained from the following studies:

- Soil Study Data Summary Report (Windward, 2015).
- Geochemical and Mineralogical Data for Soils of the Conterminous United States (Smith et al., 2013).
- Van Stone Mine Remedial Investigation report on background concentrations (Hart Crowser, 2013b).
- Upper Columbia River Upland Soil Sampling Study (Hart Crowser, 2013a).
- Midnite Mine and Blue Creek Drainage Pre-mining Geochemical Background Assessment (Church et al., 2008).
- Preliminary Assessments and Site Investigations Report on Upper Columbia River Mines and Mills (EPA, 2002b).
- Heavy Metal Contamination in Soil and Lichen Tissue in the Colville National Forest (Wells, 2015).

Proportions of each of these unique data sources with respect to the total dataset are tabulated in Appendix A (Table A-3). Figure 3 illustrates the geographic distribution of samples that fall into each of the three data source categories.

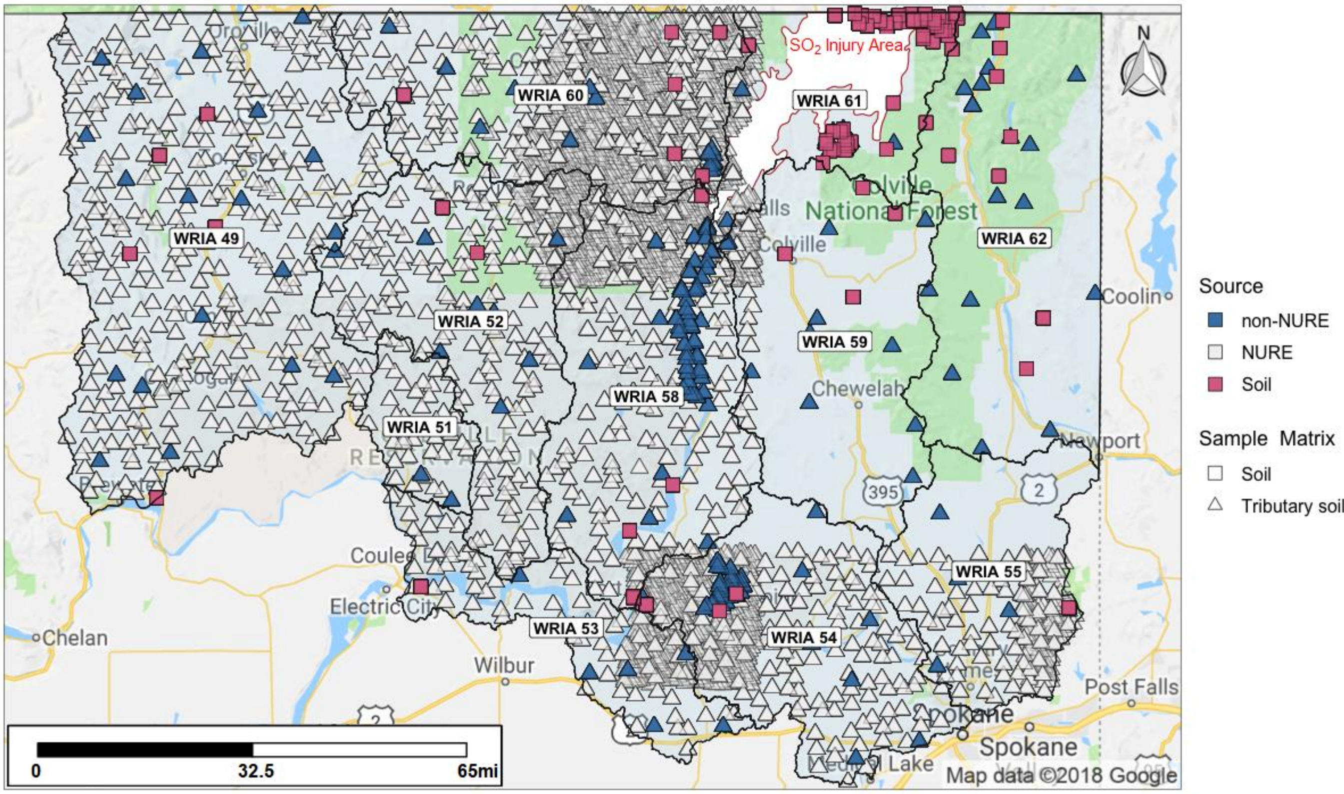

Figure 3. Geographic distribution of samples by data source.

NURE – National Uranium Resource Evaluation.

WSU performed an independent evaluation of the datasets for each metal using data-appropriate statistical approaches. A comprehensive summary of the three data sources WSU identified can be found in Appendix A. WSU's report to Ecology is presented as Appendix E. Briefly, that evaluation provided:

- Exploratory analysis that included graphical inspection of metal distributions and distribution fitting (11 distributions evaluated).
- One-way analysis of variance and non-parametric Kruskal-Wallis tests comparing primary data sources to each other to provide recommendations on pooling data from different data source categories (NURE, non-NURE, and soil) and geographic watersheds (WRIAs).
- Calculation of empirical 90th percentiles and bootstrapped confidence intervals ($CI_{.95}$) to estimate background concentrations over a range of groupings.

Figures 4 and 5 illustrate the distribution of each metal analyte between each of the three distinct data sources. Colored points outside the upper and lower fences on the boxplots represent potential outlier values that may have a pronounced effect on empirical 90th percentile values. WSU performed a final background estimation without outlier removal in part because of (1) the lack of a well-defined distribution makes inference about outlier effects unclear, and (2) the non-parametric bootstrapping methods used to estimate $CI_{.95}$ are not significantly influenced by outlying observations.

A sample count (n) greater than or equal to 10 ($n \geq 10$) is established by WSU as the minimum accepted sample size to report $CI_{.95}$ and 90th percentiles as estimates of background concentrations.

Tabulated results from the WSU analysis of upland data are presented in Tables 4.1–4.18 in Appendix E. Based on the source data, results are divided into four different data group categories for each metal, as available:

- All data pooled and presented as one data set [*All-Pooled*].
- Pooled results from all combined WRIAs assigned to each of the three data sources: NURE, non-NURE, and soil [*All-WRIA-Source*].
- Individual WRIA results presented by WRIA and calculated from combined data within each WRIA, with sufficient sample size ($n \geq 10$) [*WRIA-Specific*].
- WRIA results according to each of the three data sources having sufficient sample size ($n \geq 10$): NURE, non-NURE, and soil [*WRIA-Source*].

WSU findings concluded that none of the 11 distributions evaluated fit the data well enough to proceed using parametric analyses. Distributional differences persisted between and across WRIAs and the three data sources. WSU identified that disparity in the Kruskal-Wallis test results (significant interactions) between and across sources and individual WRIAs indicate that total data pooling in general for most metals data is less representative than using WRIA-grouped (*WRIA-Specific*) and/or source-grouped (*WRIA-Source*) data for estimating background concentrations. Where feasible, WSU advised applying the lower bound of the bootstrapped $CI_{.95}$ and the 90th percentile results as lower and upper threshold estimates of background concentration, respectively.

Finally, WSU explored anomalous thresholds observed in WRIA 61 results. All-Pooled and *All-WRIA-Source* 90th percentiles without WRIA 61 data were calculated for this comparison. Notable metals exhibiting apparent anomalous outcomes when compared to non-WRIA 61 data results include aluminum, cadmium, lead, nickel, and zinc. WRIA 61 anomalies suggest that for certain metals, particularly in soil results, depletions or enrichments may be influenced by metal particulate deposition from smelter emissions, and extend over a broader area than the initially applied exclusion area defined by the SO_2 impact area (Scheffer & Hedgcock, 1955).

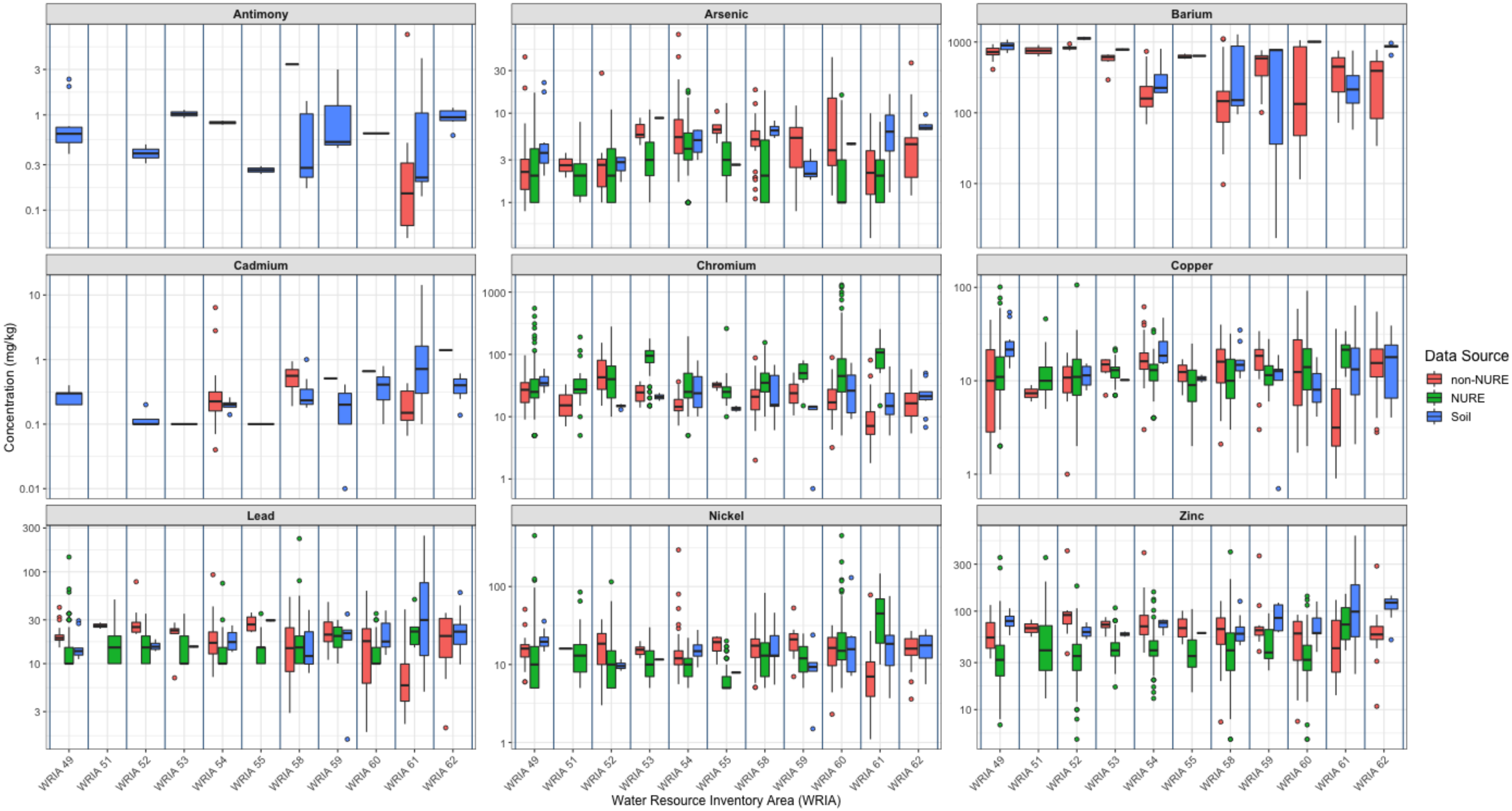

Figure 4. Distributions of metals concentrations by WRIA and data source.

Note: Logarithmic y-axis scale
NURE – National Uranium Resource Evaluation
WRIA – Water Resource Inventory Area

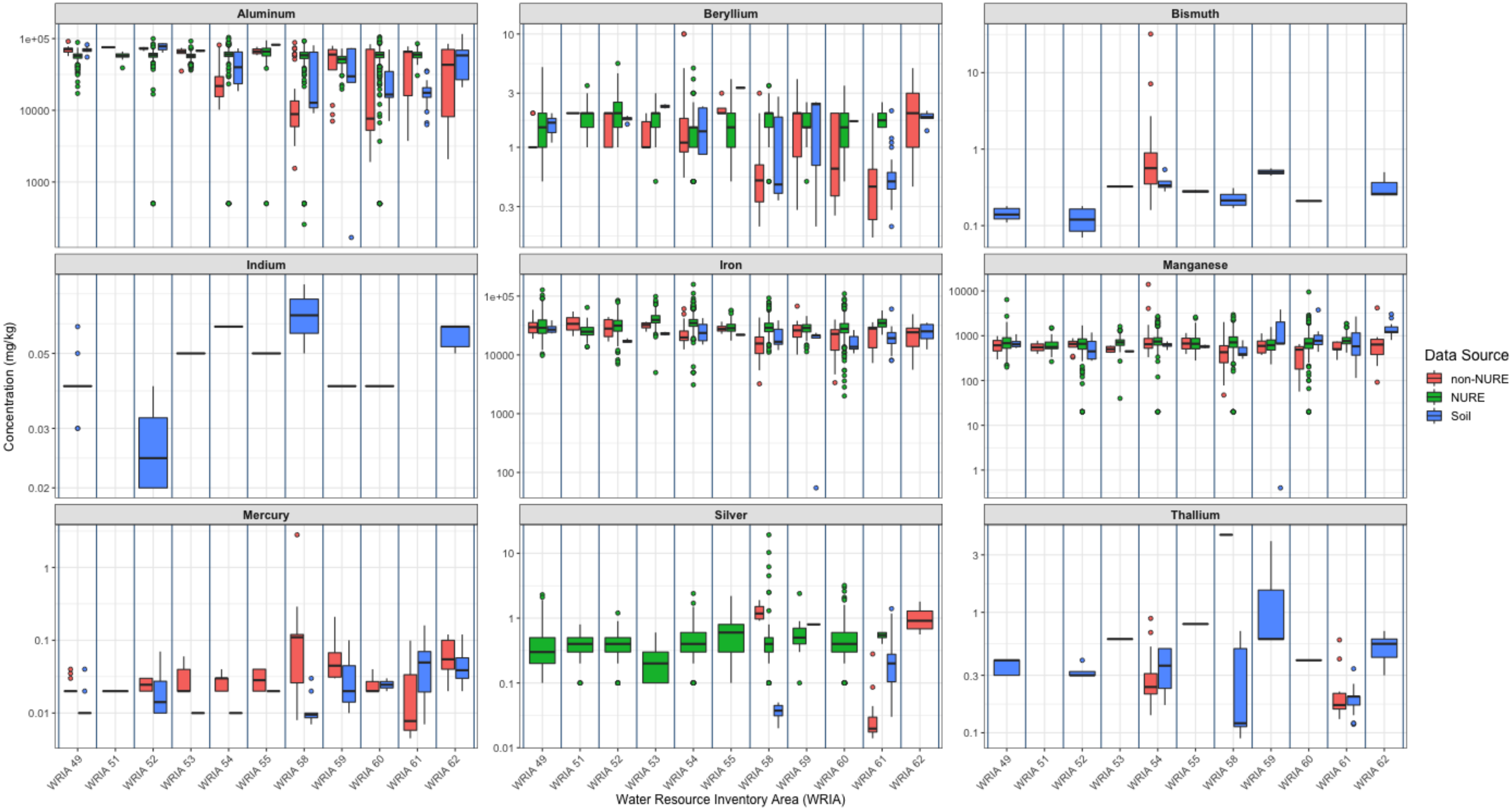

Figure 5. Distributions of metals concentrations by WRIA and data source.

Note: Logarithmic y-axis scale
NURE – National Uranium Resource Evaluation
WRIA – Water Resource Inventory Area

Results and Natural Background Determinations

Previous Data on Background

Others have reported estimations of natural background metal concentrations in the area encompassed by this study (Ames & Prych, 1995; Ecology, 1994; Church, 2010a; Ecology, 2011; MacDonald, 2014). Both Church and MacDonald utilized selected NURE data in their evaluations. Average crustal abundance concentrations are also widely published and guide assessments and reasonable concentration expectations (Hansford & Boerngen, 1984; University of Oklahoma, 1985; Gunter, 1998).

In general, upper-bound background concentrations reported in earlier studies are often more conservative (lower concentrations than the ranges presented here). Previous studies were not as comprehensive and may not have been fully inclusive of the cumulative variability between analytical methods, regional variability in geology, and other factors. The methods used in this study provide a comprehensive analytical framework for background estimation of northeast Washington soils, including the Upper Columbia River Valley, which includes considerations for multiple factors influencing soil measurement conditions. This is achieved by performing comprehensive exploratory data analysis and the application of statistical bootstrapping methods when appropriate. The results from the detailed exploratory dataset analysis and subsequent partitioning into the three data source categories demonstrate the value of exploratory data analysis when building a large background dataset.

The datasets applied in this study extend beyond previous background estimation efforts in terms of the geographic scale, the density of sample collection locations, and the incorporation of a broad suite of metals for evaluation. Additionally, this project applied screening to ensure data quality by identifying detection limit issues, replicate samples, and variability between analytical methods. The final dataset Ecology developed is a comprehensive tool for informing a broad range of natural background soil and sediment conditions across portions of northeast Washington on regional or watershed-specific scales.

90th Percentile Approach

The refinements discussed below yield final 90th percentile concentration ranges to be applied as inclusive and representative of the upper-bound natural background variability across all WRIAs, accounting for the diversity of geography, geology, study objectives, and analytical methods.

A tiered background-value selection process reports the ranges of maximum acceptable empirical 90th percentile results and further reduces the potential of selecting concentrations that might inappropriately underestimate soil metal background concentrations. Application of the WSU bootstrapped $CI_{.95}$ bounds for background determinations has statistical validity, but several high variability datasets result in ranges of $CI_{.95}$ estimates that do not readily integrate with regulatory applications.

As the WSU data analyses established, 90th percentile results are reported only for sample sets having 10 or more samples to calculate from ($n \geq 10$). While, theoretically in applying results, there may be an inherent simplicity and convenience to pooling data from the entire study area and data sources, the outcomes from exploratory statistics demonstrate the importance of considering variability inherent to the geography and geology between WRIAs and data sources when feasible.

Ecology's final determinations employ an inclusive range of 90th percentile thresholds of natural background data applicable to the Upper Columbia River Valley and Upper Columbia River watersheds of northeast Washington.

The outcome maximally assigns the range of 90th percentiles for each metal within the WRIA-Specific and *WRIA-Source* results. For indium, the available or appropriate data are limited, prohibiting robust *WRIA-Specific* or *WRIA-Source* 90th percentile calculations. In this case, the maximum 90th percentile range includes incorporation of the *All-Pooled* results.

Final Refinements and Determinations

To support final screening, and for comparative purposes between WRIAs, generalized comparative map figures (using *WRIA-Specific* 90th percentile concentration outcomes for each metal) included in Appendix C (Figures C-1 through C-3) illustrate watershed-specific variation. Consideration of watershed inter-comparisons and further examinations of underlying data and known area-scale influences assisted in final appropriate background range determinations. These considerations, other factors, and final value determinations are described below.

Certain *WRIA-Specific or WRIA-Source* 90th percentile outcomes that deviate by factors of 1.5 or greater compared to the other WRIAs underwent further review and adjustment to establish representative background thresholds. The 90^{th} percentile ranges for cadmium, lead, mercury, and zinc underwent additional selection steps within the WSU results, incorporating WRIA inter-comparisons and other factual considerations. The adjusted ranges ensure elimination of anthropogenic or other localized factors and data limitations. Additional review may be of particular importance when evaluating smaller, less robust data sets and those associated with smelter-influenced WRIA 61. Specifically, WRIA 61 is excluded from the final upper-bound background value ranges for cadmium, lead, and zinc due to smelter effects. WRIA 58 is excluded for mercury. Appendix D documents final 90^{th} percentile evaluation and determinations for each of the 18 metals.

In summary, Table 3 presents the established 50^{th}, 75^{th}, 90^{th} percentile ranges and the 90^{th} percentile midpoint of the range values of natural background for 18 metals across the 11 watersheds, with the 90^{th} percentile values assigned as the upper-bound background concentration range.

Table 3. Ranges of final 50th, 75th, 90th percentiles.

Analyte	50th Percentile Range	75th Percentile Range	90th Percentile Natural Background Concentration Range[1]	90th Percentile Range Midpoint
Aluminum[2]	17,550 to 65,900	20,675 to 73,850	24,870 to 81,320	53,095
Antimony	0.15 to 0.64	0.31 to 0.74	0.49 to 2.03	1.26
Arsenic	2 to 3	3 to 9	3 to 20	12
Barium	214 to 854	335 to 1,073	432 to 1,137	785
Beryllium	0.5 to 1.5	0.6 to 2	0.7 to 3.3	2
Bismuth	0.14 to 0.57	0.17 to 0.9	0.2 to 2.1	1.1
Cadmium[2]	0.3 to 0.56	0.3 to 0.7	0.4 to 0.84	0.44
Chromium	25 to 45	30 to 85	35 to 164	100
Copper	3 to 22	8 to 26	16 to 49	33
Indium	0.04 to 0.05	0.04 to 0.06	0.05 to 0.06	0.055
Iron	19,200 to 39,260	23,956 to 46,600	26,950 to 54,900	40,925
Lead[2]	15 to 22	15 to 27	19 to 43	31
Manganese	490 to 1,210	572 to 1,588	621 to 2,493	1,557
Mercury[2]	0.01 to 0.05	0.01 to 0.07	0.02 to 0.15	0.09
Nickel	5 to 15	7 to 26	12 to 50	31
Silver	0.2 to 0.2	0.4 to 1	0.4 to 1.4	0.9
Thallium	0.1 to 0.2	0.2 to 0.5	0.2 to 0.7	0.5
Zinc[2]	40 to 122	48 to 132	55 to 139	97

[1] All reported concentrations reported in milligrams per kilogram (mg/kg) and represent 90th percentile values. Ranges derived from WSU *WRIA-Specific* and *WRIA-Source* 90th percentile outcomes, unless otherwise specified.

[2] Adjusted background concentrations, as described in this section.

Conclusion

An extensive screening and assimilation of 25,379 data records, including the evaluation of geologic and anthropogenic factors were explored, and upper-percentile background concentration ranges calculated. Background concentration ranges provided in Table 3 represent the ranges of 90th percentile maximum background concentrations for the study area. Natural soils to be encountered or sampled across the 11 WRIA study areas can be expected to be below (less than) the range midpoints presented. Variability can be expected across watersheds as a result of variability in underlying bedrock or glacially influenced terrain.

Detected concentrations above the 90th percentile range midpoint values (Table 3) indicate probable anthropogenic effects or influence from localized geologic anomalies that warrant additional evaluation or sample collection. The reported values account for the diversity and variability inherent to natural surface materials across different settings, encompassing a broad regional area of northeast Washington, while incorporating significant variability in analytic preparations and laboratory methods, grain-size considerations, study designs, and other factors. For environmental investigators utilizing partial digestion analytic methods (total recoverable EPA digestion methods), the lower end range 90 percentile values generally may serve as better representative under those applications.

Upper-percentile concentrations of certain metals (cadmium, lead, zinc) in upland soil surfaces within the smelter-impacted WRIA 61 watershed that are above the Table 3 values, outside of potential localized ore mineralization, represent definitive metal enrichment resulting from historical anthropogenic activity, shown primarily as a result of mineral smelting operation emissions.

Recommendations

The established background ranges are generated from the analysis of a uniquely large dataset compiled during this study to provide a resource to regulators, scientists, or others interested in upper-bound natural background conditions of upland surface soils across watersheds of northeast Washington. Along with detailed exploratory analysis and the bootstrapping methods applied, the methods and approaches used offer a methodology for other area-wide or regional-scale background efforts. Use of the 50^{th}, 75^{th}, and 90^{th} percentile ranges in Table 3 for application to hazardous substance assessment and site cleanup is appropriate on a site-specific basis in direct consultation with Ecology's Toxics Cleanup Program.

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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 that are the result of human activities unrelated to releases from that site.

Confidence interval: A range of values with a defined probability that an observation or parameter lies within it.

Emissions: Production and discharge of gas-phase or particulate substances.

Model Toxics Control Act (MTCA): The environmental cleanup law in Washington State. It informs decisions of liability and defines what cleanup actions are necessary to protect Washington residents and the environment.

Natural background: The concentration of a constituent consistently present 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.

Sample digestion: The process of dissolving samples for chemical analysis, typically by treatment with acids and the addition of heat.

Sediment: Solid fragmented material (soil and organic matter) that is transported and deposited by water.

Smelter: A facility where metal is extracted from ore using heat and chemical-reducing agents.

Soil: For the purposes of this study, soil is operationally defined as unconsolidated earth surface materials less than 2 mm in size and a natural medium capable of supporting rooted plants and other upland flora and fauna. Within the context of assessment, these surface soils reside within 24 inches (typically shallower) of the ground surface at the time of sample collection.

Surficial sediment: Recently deposited sediment in watershed tributaries.

Acronyms and Abbreviations

$CI_{.95}$	95 percent confidence interval
CISER	Center for Interdisciplinary Statistics Education and Research
Ecology	Washington State Department of Ecology
EPA	U.S. Environmental Protection Agency
GIS	Geographic information system software
LLL	Lawerence Livermore National Laboratory
MTCA	Model Toxics Control Act
n	number of samples
NURE-HSSR	National Uranium Resource Evaluation Hydrogeochemical and Stream Sediment Reconnaissance
PASI	Preliminary Assessments and Site Investigations
SO2	sulfur dioxide
USGS	U.S. Geological Survey
WAC	Washington Administrative Code
WRIA	Water Resource Inventory Area
WSU	Washington State University

Units of Measurement

cm	centimeter
ft	feet
in	inch
kg	kilogram, a unit of mass equal to 1,000 grams
km	kilometer, a unit of distance equal to 1,000 meters
mg	milligram
mm	millimeter
mg/kg	milligrams per kilogram (parts per million)

Metals

Ag	silver
Al	aluminum
As	arsenic
Ba	barium
Be	beryllium
Bi	bismuth
Cd	cadmium
Cr	chromium
Cu	copper
In	indium
Fe	iron
Hg	mercury
Mn	manganese
Ni	nickel
Ag	silver
Pb	lead
Sb	antimony
Tl	thallium
Zn	zinc

Appendices

Appendix A. Summary of Background Dataset

Table A-1. Primary data sets summary for background soil estimation development.

Soil Assessment Name	Soil Assessment Reference	No. Samples	No. Records	Metal Analytes Advanced for Background Estimation
NURE-HSSR	Smith, S.M. 2006. "National Geochemical Database-Reformatted Data from the National Uranium Resource Evaluation (NURE) Hydrogeochemical and Stream Sediment Reconnaissance (HSSR) Program: Version 1.40." USGS Open-File Report 97-492.	4,049	21,154	Al As Be Cr Cu Fe Pb Mn Ni Ag Zn
Reanalysis of Archived NURE-HSSR Samples	Grossman, J.N., Grosz, A.E., Schweitzer, P.N., and Schruben, P.G. 2004. The National Geochemical Survey-Database and Documentation: USGS Open-File Report 2001-1001.	87	1,045	Al As Ba Be Cr Cu Fe Pb Mn Hg Ni Zn
USGS 2013	Smith, D.B., Cannon, W.F., Woodruff, L.G., Solano, F., Kilburn, J.E., and Fey, D.L. 2013. Geochemical and mineralogical data for soils of the conterminous United States: USGS Data Series 801.	34	578	Al Sb As Ba Be Bi Cd Cr Cu In Fe Pb Mn Hg Ni Tl Zn
USGS Midnite Mine and Blue Creek Drainage	Church, S.E., Kirschner, F.E., Choate, L.M., Lamothe, P.J., Budahn, J.R., and Brown, Z.A. 2008. Determination of Premining Geochemical Background and Delineation of Extent of Sediment Contamination in Blue Creek Downstream from Midnite Mine, Stevens County, Washington: USGS Scientific Investigations Report 2007-5262.	43	588	Al As Ba Be Bi Cd Cr Cu Fe Pb Mn Ni Tl Zn
Hart Crowser Van Stone Mine Background Assessment	Hart Crowser. 2013b. Van Stone Mine Remedial Investigation: "Appendix B-Results of Van Stone Mine Background Sampling and Natural Background Metal Concentrations." (Prepared for Ecology).	24	286	Sb As Be Cd Cr Cu Pb Hg Ni Ag Tl Zn
Hart Crowser Upper Columbia River	Hart Crowser. 2013a. Upper Columbia River Upland Soil Sampling Study, Stevens County ,Washington. (Prepared for Ecology).	25	400	Al Sb As Ba Be Cd Cr Cu Fe Pv Mn Hg Ni Ag Tl Zn
USEPA PASI: Upper Columbia River Mines and Mills	EPA [U.S. Environmental Protection Agency Region 10 Superfund Technical Assessment and Response Team]. 2002b. Preliminary Assessments and Site Investigations Report, Upper Columbia River Mines and Mills, Stevens County, Washington. TDD: 01-02-0028, Contract 68-S0-01-01.	3	36	Al As Ba Be Cd Cr Cu Fe Pb Mn Hg Ni Ag Tl Zn
USEPA PASI: Lower Pend Oreille Mines and Mills	EPA [U.S. Environmental Protection Agency Region 10 Superfund Technical Assessment and Response Team]. 2002a. Preliminary Assessments and Site Investigations Report, Lower Pend Oreille River Mines and Mills, Pend Oreille County, Washington. TDD: 01-08-0009, Contract 68-S0- 01-01.	6	68	Al As Ba Be Cd Cr Cu Fe Pb Mn Hg Ni Ag Zn
USEPA: Upper Columbia River Expanded Site Inspections	EPA [U.S. Environmental Protection Agency Region 10 Superfund Technical Assessment and Response Team]. 2003. Upper Columbia River Expanded Site Inspection Report Northeast Washington. TDD: 01-02-0028.	57	625	Al Sb As Ba Be Cd Cr Cu Fe Pb Mn Hg Ni Ag Tl Zn
Windward Soil Study Data Summary Report	Windward. 2015. Upper Columbia River Soil Study Data Summary Report. (Prepared for Teck American Incorporated in association and consultation with Exponent, Parametrix, Inc., and ENVIRON.)	25	400	Al Sb As Ba Be Cd Cr Cu Fe Pb Mn Hg Ni Ag Tl Zn
MacDonald Envrironmental Assessment for the Colville Confederated Tribes	MacDonald Environmental Sciences, Ltd. 2014. Determination of Regional Background Concentrations of Selected Metals in Sediments of the Upper Columbia River Drainage Basin. (Prepared for Confederated Tribes of the Colville Reservation).	2	28	Al As Ba Be Cd Cr Cu Fe Pb Mn Hg Ni Ag Zn
Kevin Wells M.Sc Dissertation: Metals Contamination in the Colville National Forest	Wells, K. 2015. Heavy Metal Contamination in Soil and Lichen Tissue in the Colville National Forest, Washington, USA. Dissertation submitted for the degree of Master of Science in Environmental Forestry, Bangor University, Wales, United Kingdom.	19	171	Al Cd Cr Cu Fe Pb Mn Ni Zn

Ag – silver, Al – aluminum, As – arsenic, Ba – barium, Be – beryllium, Bi – bismuth, Cd – cadmium, Cr – chromium, Cu – copper, Fe – iron, Hg – mercury, In – indium, MESL – MacDonald Engineering Services Ltd., Mn – manganese, MSc – Masters of Science degree, Ni – nickel, NURE-HSSR – National Uranium Resource Evaluation Hydrogeochemical and Stream Sediment Reconnaissance, PASI – Preliminary Assessments and Site Investigations, Pb – lead, Sb – antimony, Tl – thallium, USEPA – U.S. Environmental Protection Agency, USGS – U.S. Geological Survey, Zn – zinc

Table A-2: Proportion of samples and records represented by each study used for estimating background.

Study Name	No. Samples	% Total Samples	No. Records	% Total Records
NURE	4,049	92.6%	21,154	83.4%
NURE Reanalysis	87	2.0%	1,045	4.1%
USGS Midnite Mine	43	1.0%	588	2.3%
Hart Crowser UCR	25	0.6%	400	1.6%
Windward 2015	25	0.6%	400	1.6%
Hart Crowser Van Stone Mine	24	0.5%	286	1.1%
Weston ESI	57	1.3%	625	2.5%
Wells 2015	19	0.4%	171	0.7%
USGS 2013	34	0.8%	578	2.3%
PASI 0009: Pend Oreille County	6	0.1%	68	0.3%
PASI 0028: Stevens County	3	0.1%	36	0.1%
MESL 2014	2	0.0%	28	0.1%
Totals	*4,374*	*100.0%*	*25,379*	*100%*

ESI –EPA Expanded Site Inspections, MESL – MacDonald Environmental Services Ltd., NURE – National Uranium Resource Evaluation, PASI – EPA Preliminary Assessments and Site Investigations, UCR – Upper Columbia River, USGS – U.S. Geological Survey

Table A-3: Proportion of total records represented by each unique data source.

Data Source	NURE Sediment		Non-NURE Sediment[1]		Soil		*Total No. Records*
Analyte	**No. Records**	**% Total Records**	**No. Records**	**% Total Records**	**No. Records**	**% Total Records**	
Aluminum	4,046	93%	195	4%	106	2%	*4,347*
Antimony	0	0%	14	13%	97	87%	*111*
Arsenic	1,004	78%	181	14%	99	8%	*1,284*
Barium	0	0%	194	69%	87	31%	*281*
Beryllium	1,163	80%	197	14%	99	7%	*1,459*
Bismuth	0	0%	40	53%	36	47%	*76*
Cadmium	0	0%	68	37%	118	63%	*186*
Chromium	1,167	78%	206	14%	118	8%	*1,491*
Copper	1,087	77%	206	15%	118	8%	*1,411*
Indium	0	0%	0	0%	34	100%	*34*
Iron	3,989	93%	194	5%	106	2%	*4,289*
Lead	1,163	78%	205	14%	118	8%	*1,486*
Manganese	4,041	93%	194	4%	106	2%	*4,341*
Mercury	0	0%	111	53%	97	47%	*208*
Nickel	1,164	78%	204	14%	118	8%	*1,486*
Silver	1,165	93%	22	2%	63	5%	*1,250*
Thallium	0	0%	53	35%	98	65%	*151*
Zinc	1,165	78%	206	14%	117	8%	*1,488*

[1] Includes NURE reanalysis data
NURE – National Uranium Resource Evaluation

Table A-4a: Proportion of total records represented by NURE sediment samples.

Analyte	NURE Sediment											Total No. Records
	WRIA 49	WRIA 51	WRIA 52	WRIA 53	WRIA 54	WRIA 55	WRIA 58	WRIA 59	WRIA 60	WRIA 61	WRIA 62	
Aluminum	337 (8%)	40 (1%)	383 (9%)	241 (6%)	573 (13%)	169 (4%)	687 (16%)	84 (2%)	1,419 (33%)	113 (3%)	-	*4,046*
Antimony	-	-	-	-	-	-	-	-	-	-	-	*0*
Arsenic	243 (19%)	30 (2%)	153 (12%)	66 (5%)	133 (10%)	74 (6%)	144 (11%)	156 (12%)	5 (0%)	- (0%)	-	*1,004*
Barium	-	-	-	-	-	-	-	-	-	-	-	*0*
Beryllium	332 (23%)	40 (3%)	162 (11%)	72 (5%)	135 (9%)	74 (5%)	156 (11%)	13 (1%)	173 (12%)	6 (0%)	-	*1,163*
Bismuth	-	-	-	-	-	-	-	-	-	-	-	*0*
Cadmium	-	-	-	-	-	-	-	-	-	-	-	*0*
Chromium	333 (22%)	40 (3%)	164 (11%)	72 (5%)	135 (9%)	74 (5%)	156 (10%)	13 (1%)	174 (12%)	6 (0%)	-	*1,167*
Copper	335 (24%)	40 (3%)	142 (10%)	72 (5%)	135 (10%)	73 (5%)	99 (7%)	12 (1%)	173 (12%)	6 (0%)	-	*1,087*
Indium	-	-	-	-	-	-	-	-	-	-	-	*0*
Iron	335 (8%)	40 (1%)	381 (9%)	241 (6%)	567 (13%)	167 (4%)	679 (16%)	82 (2%)	1,390 (32%)	107 (2%)	-	*3,989*
Lead	332 (22%)	40 (3%)	162 (11%)	72 (5%)	135 (9%)	74 (5%)	156 (10%)	13 (1%)	173 (12%)	6 (0%)	-	*1,163*
Manganese	335 (8%)	40 (1%)	383 (9%)	238 (5%)	573 (13%)	169 (4%)	687 (16%)	84 (2%)	1,419 (33%)	113 (3%)	-	*4,041*
Mercury	-	-	-	-	-	-	-	-	-	-	-	*0*
Nickel	335 (23%)	40 (3%)	163 (11%)	72 (5%)	135 (9%)	73 (5%)	155 (10%)	13 (1%)	172 (12%)	6 (0%)	-	*1,164*
Silver	335 (27%)	40 (3%)	163 (13%)	72 (6%)	135 (11%)	73 (6%)	155 (12%)	13 (1%)	173 (14%)	6 (0%)	-	*1,165*
Thallium	-	-	-	-	-	-	-	-	-	-	-	*0*
Zinc	335 (23%)	40 (3%)	163 (11%)	72 (5%)	135 (9%)	73 (5%)	155 (10%)	13 (1%)	173 (12%)	6 (0%)	-	*1,165*
Total No. Records	*3,587*	*430*	*2,419*	*1,290*	*2,791*	*1,093*	*3,229*	*496*	*5,444*	*375*	*0*	***21,154***

NURE – National Uranium Resource Evaluation
WRIA – Water Resource Inventory Area

Table A-4b: Proportion of total records represented by non-NURE sediment samples.

Analyte	Non-NURE Sediment											Total No. Records
	WRIA 49	WRIA 51	WRIA 52	WRIA 53	WRIA 54	WRIA 55	WRIA 58	WRIA 59	WRIA 60	WRIA 61	WRIA 62	
Aluminum	19 (0%)	2 (0%)	8 (0%)	6 (0%)	49 (1%)	4 (0%)	55 (1%)	12 (0%)	16 (0%)	5 (0%)	19 (0%)	*195*
Antimony	-	-	-	-	-	-	1 (1%)	-	-	13 (12%)	-	*14*
Arsenic	19 (1%)	2 (0%)	8 (1%)	6 (0%)	48 (4%)	4 (0%)	38 (3%)	11 (1%)	12 (1%)	15 (1%)	18 (1%)	*181*
Barium	19 (7%)	2 (1%)	8 (3%)	6 (2%)	48 (17%)	4 (1%)	55 (20%)	12 (4%)	16 (6%)	5 (2%)	19 (7%)	*194*
Beryllium	19 (1%)	2 (0%)	8 (1%)	6 (0%)	49 (3%)	4 (0%)	53 (4%)	12 (1%)	14 (1%)	17 (1%)	13 (1%)	*197*
Bismuth	-	-	-	-	40 (53%)	-	-	-	-	-	-	*40*
Cadmium	-		-	-	40 (22%)	-	13 (7%)	1 (1%)	1 (1%)	12 (6%)	1 (1%)	*68*
Chromium	19 (1%)	2 (0%)	8 (1%)	6 (0%)	48 (3%)	4 (0%)	55 (4%)	12 (1%)	16 (1%)	17 (1%)	19 (1%)	*206*
Copper	19 (1%)	2 (0%)	8 (1%)	6 (0%)	48 (3%)	4 (0%)	55 (4%)	12 (1%)	16 (1%)	17 (1%)	19 (1%)	*206*
Indium	-	-	-	-	-	-	-	-	-	-	-	*0*
Iron	19 (0%)	2 (0%)	8 (0%)	6 (0%)	48 (1%)	4 (0%)	55 (1%)	12 (0%)	16 (0%)	5 (0%)	19 (0%)	*194*
Lead	19 (1%)	2 (0%)	8 (1%)	6 (0%)	48 (3%)	4 (0%)	54 (4%)	12 (1%)	16 (1%)	17 (1%)	19 (1%)	*205*
Manganese	19 (0%)	2 (0%)	8 (0%)	6 (0%)	48 (1%)	4 (0%)	55 (1%)	12 (0%)	16 (0%)	5 (0%)	19 (0%)	*194*
Mercury	19 (9%)	2 (1%)	8 (4%)	6 (3%)	9 (4%)	4 (2%)	21 (10%)	10 (5%)	6 (3%)	14 (7%)	12 (6%)	*111*
Nickel	19 (1%)	2 (0%)	8 (1%)	6 (0%)	47 (3%)	4 (0%)	54 (4%)	12 (1%)	16 (1%)	17 (1%)	19 (1%)	*204*
Silver	-	-	-	-	-	-	4 (0%)	-	-	12 (1%)	6 (0%)	*22*
Thallium	-	-	-	-	40 (26%)	-	1 (1%)	-	-	12 (8%)	-	*53*
Zinc	19 (1%)	2 (0%)	8 (1%)	6 (0%)	48 (3%)	4 (0%)	55 (4%)	12 (1%)	16 (1%)	17 (1%)	19 (1%)	*206*
Total No. Records	*228*	*24*	*96*	*72*	*658*	*48*	*624*	*142*	*177*	*200*	*221*	***2,490***

NURE – National Uranium Resource Evaluation, WRIA – Water Resource Inventory Area

Table A-4c: Proportion of total records represented by soil samples.

Analyte	Soil											Total No. Records
	WRIA 49	WRIA 51	WRIA 52	WRIA 53	WRIA 54	WRIA 55	WRIA 58	WRIA 59	WRIA 60	WRIA 61	WRIA 62	
Aluminum	10 (0%)	-	4 (0%)	2 (0%)	4 (0%)	2 (0%)	11 (0%)	5 (0%)	7 (0%)	50 (1%)	11 (0%)	*106*
Antimony	10 (9%)	-	4 (4%)	2 (2%)	2 (2%)	2 (2%)	10 (9%)	3 (3%)	2 (2%)	56 (50%)	6 (5%)	*97*
Arsenic	10 (1%)	-	4 (0%)	2 (0%)	4 (0%)	2 (0%)	10 (1%)	3 (0%)	2 (0%)	56 (4%)	6 (0%)	*99*
Barium	10 (4%)	-	4 (1%)	2 (1%)	4 (1%)	2 (1%)	10 (4%)	3 (1%)	2 (1%)	44 (16%)	6 (2%)	*87*
Beryllium	10 (1%)	-	4 (0%)	2 (0%)	4 (0%)	2 (0%)	10 (1%)	3 (0%)	2 (0%)	56 (4%)	6 (0%)	*99*
Bismuth	10 (13%)	-	4 (5%)	2 (3%)	4 (5%)	2 (3%)	4 (5%)	2 (3%)	2 (3%)	6 (8%)	11 (14%)	*47*
Cadmium	10 (5%)	-	4 (2%)	2 (1%)	4 (2%)	2 (1%)	11 (6%)	5 (3%)	7 (4%)	62 (33%)	11 (6%)	*118*
Chromium	10 (1%)	-	4 (0%)	2 (0%)	4 (0%)	2 (0%)	11 (1%)	5 (0%)	7 (0%)	62 (4%)	11 (1%)	*118*
Copper	10 (1%)	-	4 (0%)	2 (0%)	4 (0%)	2 (0%)	11 (1%)	5 (0%)	7 (0%)	62 (4%)	11 (1%)	*118*
Indium	10 (29%)	-	4 (12%)	-	2 (6%)	2 (6%)	2 (6%)	4 (12%)	2 (6%)	2 (6%)	6 (18%)	*34*
Iron	10 (0%)	-	4 (0%)	2 (0%)	4 (0%)	2 (0%)	11 (0%)	5 (0%)	7 (0%)	50 (1%)	11 (0%)	*106*
Lead	10 (1%)	-	4 (0%)	2 (0%)	4 (0%)	2 (0%)	11 (1%)	5 (0%)	7 (0%)	62 (4%)	11 (1%)	*118*
Manganese	10 (0%)	-	4 (0%)	2 (0%)	4 (0%)	2 (0%)	11 (0%)	5 (0%)	7 (0%)	50 (1%)	11 (0%)	*106*
Mercury	10 (5%)	-	4 (2%)	2 (1%)	2 (1%)	2 (1%)	10 (5%)	3 (1%)	2 (1%)	56 (27%)	6 (3%)	*97*
Nickel	10 (1%)	-	4 (0%)	2 (0%)	4 (0%)	2 (0%)	11 (1%)	5 (0%)	7 (0%)	62 (4%)	11 (1%)	*118*
Silver	-	-	-	-	-	-	6 (0%)	1 (0%)	-	56 (4%)	-	*63*
Thallium	10 (7%)	-	4 (3%)	2 (1%)	4 (3%)	2 (1%)	10 (7%)	3 (2%)	2 (1%)	55 (36%)	6 (4%)	*98*
Zinc	10 (1%)	-	4 (0%)	2 (0%)	4 (0%)	2 (0%)	11 (1%)	4 (0%)	7 (0%)	62 (4%)	11 (1%)	*117*
Total No. Records	*170*	*0*	*68*	*32*	*62*	*34*	*171*	*69*	*79*	*909*	*152*	***1,746***

WRIA – Water Resource Inventory Area

Table A-5: Summary of analytical methods and typical detection limit (actual results may vary).

Analytical Method	Analytes	Detection or Reporting Limit Range (mg/kg)
AA	Arsenic	0.6 - 1
	Beryllium	0.5
	Chromium	5
	Copper	2
	Lead	10
	Nickel	5
	Silver	0.1
	Zinc	Unknown
ICP-MS	Aluminum	50
	Antimony	0.009 - 0.2
	Arsenic	0.04 - 1
	Barium	0.02 - 0.2
	Beryllium	0.005 - 0.03
	Bismuth	0.06
	Cadmium	0.006 - 0.025
	Chromium	0.036 - 0.5
	Copper	0.034 - 2
	Iron	50
	Lead	0.02 - 0.4
	Manganese	0.7
	Nickel	0.03 - 0.3
	Silver	0.004 - 0.2
	Thallium	0.002 - 0.41
	Zinc	0.2 - 3

AA – atomic absorption spectroscopy
ICP-MS – inductively coupled plasma/mass spectrometry

Analytical Method	Analytes	Detection or Reporting Limit Range (mg/kg)
INAA	Aluminum	500
	Iron	5000
	Manganese	20
ICP-AES	Aluminum	0.4 - 168.1
	Antimony	0.05 - 60
	Arsenic	1 - 10
	Barium	0.059 - 20
	Beryllium	0.066 - 5
	Bismuth	0.04
	Cadmium	0.01 - 0.6
	Chromium	0.37 - 2
	Copper	0.5 - 2.5
	Indium	0.02
	Iron	0.73 - 200
	Lead	0.14 - 4
	Manganese	0.02 - 5
	Mercury	0.1 - 0.2
	Nickel	0.12 - 4
	Silver	0.5 - 10
	Thallium	0.1 - 10
	Zinc	0.5 - 6
CVAA	Mercury	0.0004 - 0.02
HGAAS	Arsenic	0.6

CVAA – cold vapor atomic absorption spectroscopy, HGAAS – hydride generation atomic absorption spectroscopy, ICP-AES – inductively coupled plasma/ atomic emission spectroscopy, INAA – instrumental neutron activation analysis, mg/kg – milligrams per kilogram

Table A-6: Summary of non-detect results in background dataset.

Number of Non-detect Results and Percentage of Non-detect Results by Analyte													
Analyte	**WRIA 49**	**WRIA 51**	**WRIA 52**	**WRIA 53**	**WRIA 54**	**WRIA 55**	**WRIA 58**	**WRIA 59**	**WRIA 60**	**WRIA 61**	**WRIA 62**	**Total No. Non-detect**	**% Total Records by Metal**
Aluminum	0 (0%)	0 (0%)	6 (12%)	0 (0%)	8 (16%)	3 (6%)	19 (38%)	1 (2%)	13 (26%)	0 (0%)	0 (0%)	*50*	*1%*
Antimony	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	1 (4%)	0 (0%)	25 (96%)	0 (0%)	*26*	*23%*
Arsenic	0 (0%)	0 (0%)	25 (27%)	0 (0%)	0 (0%)	0 (0%)	25 (27%)	1 (1%)	39 (43%)	0 (0%)	1 (1%)	*91*	*7%*
Barium	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	1 (100%)	0 (0%)	0 (0%)	0 (0%)	*1*	*0%*
Beryllium	10 (36%)	0 (0%)	0 (0%)	1 (4%)	1 (4%)	6 (21%)	4 (14%)	1 (4%)	4 (14%)	0 (0%)	1 (4%)	*28*	*2%*
Bismuth	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	*0*	*0%*
Cadmium	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	2 (40%)	1 (20%)	2 (40%)	0 (0%)	0 (0%)	0 (0%)	*5*	*3%*
Chromium	1 (50%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	1 (50%)	0 (0%)	0 (0%)	0 (0%)	*2*	*0%*
Copper	1 (33%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	1 (33%)	0 (0%)	1 (33%)	0 (0%)	*3*	*0%*
Indium	0 (0%)	0 (0%)	1 (100%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	*1*	*3%*
Iron	0 (0%)	0 (0%)	0 (0%)	1 (14%)	5 (71%)	0 (0%)	0 (0%)	1 (14%)	0 (0%)	0 (0%)	0 (0%)	*7*	*0%*
Lead	75 (28%)	7 (3%)	31 (11%)	25 (9%)	44 (16%)	12 (4%)	19 (7%)	1 (0%)	57 (21%)	0 (0%)	0 (0%)	*271*	*18%*
Manganese	0 (0%)	0 (0%)	13 (32%)	0 (0%)	6 (15%)	0 (0%)	11 (27%)	1 (2%)	10 (24%)	0 (0%)	0 (0%)	*41*	*1%*
Mercury	14 (33%)	2 (5%)	3 (7%)	4 (10%)	2 (5%)	1 (2%)	2 (5%)	1 (2%)	2 (5%)	10 (24%)	1 (2%)	*42*	*20%*
Nickel	41 (29%)	3 (2%)	33 (24%)	4 (3%)	10 (7%)	23 (17%)	21 (15%)	1 (1%)	3 (2%)	0 (0%)	0 (0%)	*139*	*9%*
Silver	11 (22%)	0 (0%)	1 (2%)	0 (0%)	0 (0%)	1 (2%)	6 (12%)	1 (2%)	13 (25%)	16 (31%)	2 (4%)	*51*	*4%*
Thallium	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	1 (3%)	0 (0%)	35 (97%)	0 (0%)	*36*	*24%*
Zinc[1]	-	-	-	-	-	-	-	-	-	-	-	-	-

[1]None of the zinc results used for background estimation were reported below the analytical detection limit.

WRIA – Water Resource Inventory Area

Appendix B. Data Usability Screening:

Table B-1a: Number of samples removed from background dataset due to proximity to mines.

Soil Assessment	No. Samples	Analyte List
PASI 0009: Pend Oreille County	2	As, Ba, Be, Cd, Cr, Cu, Fe, Pb, Mn, Hg, Ni, Ag, Zn
PASI 0028: Stevens County	4	Al, Sb, As, Ba, Be, Cd, Cr, Cu, Fe, Pb, Mn, Hg, Ni, Ag, Tl, Zn
Expanded Site Inspection	6	Al, Sb, As, Ba, Be, Cd, Cr, Cu, Fe, Pb, Mn, Hg, Ni, Ag, Tl, Zn
NURE Reanalysis	3	Al, As, Ba, Be, Cr, Cu, Fe, Pb, Mn, Hg, Ni, Zn
NURE-HSSR	19	Al, As, Be, Cr, Cu, Fe, Pb, Mn, Ni, Ag, Zn
USGS Midnite Mine	2	Al, As, Ba, Be, Bi, Cd, Cr, Cu, Fe, Pb, Mn, Ni, Tl, Zn
USGS 2013	1	Al, Sb, As, Ba, Be, Bi, Cd, Cr, Cu, Fe, Pb, Mn, Hg, Ni, Tl, Zn
Wells 2015	1	Al, Cd, Cr, Cu, Fe, Pb, Mn, Ni, Zn
Windward 2015	1	Al, Sb, As, Ba, Be, Cd, Cr, Cu, Fe, Pb, Mn, Hg, Ni, Ag, Tl, Zn

Table B-1b: Number of samples excluded from background dataset located within the SO_2 exclusion area.

Soil Assessment	No. Samples	Analyte List
PASI 0028: Stevens County	3	Al, Sb, As, Ba, Be, Cd, Cr, Cu, Fe, Pb, Mn, Hg, Ni, Ag, Tl, Zn
Expanded Site Inspection: UCR	3	Al, Sb, As, Ba, Be, Cd, Cr, Cu, Fe, Pb, Mn, Hg, Ni, Ag, Tl, Zn
Hart Crowser UCR: 6-12 inch interval	13	Al, Sb, As, Ba, Be, Cd, Cr, Cu, Fe, Pb, Mn, Hg, Ni, Ag, Tl, Zn
NURE-HSSR	9	Al, As, Be, Cr, Cu, Fe, Pb, Mn, Ni, Ag, Zn
NURE Reanalysis	3	Al, As, Ba, Be, Cr, Cu, Fe, Pb, Mn, Hg, Ni, Zn
Windward 2015	4	Al, Sb, As, Ba, Be, Cd, Cr, Cu, Fe, Pb, Mn, Hg, Ni, Ag, Tl, Zn

Table B-1c: Analyses excluded from the background dataset due to inadequate detection limits.

Soil Assessment	No. Records	Analyte
NURE-HSSR	498	Ba
NURE Reanalysis	67	Ag
NURE Reanalysis	95	Bi
NURE Reanalysis	87	Cd
MESL 2014	2	Tl
USGS 2013	50	Ag

Ag – silver
Al – aluminum
As – arsenic
Ba – barium
Be – beryllium
Cd – cadmium
Cr – chromium
Cu – copper
Fe – iron
Hg – mercury
Mn – manganese
Ni – nickel
Pb – lead
Sb – antimony
Tl – thallium
Zn – zinc

MESL – MacDonald Engineering Services Ltd.
NURE-HSSR – National Uranium Resource Evaluation Hydrogeochemical and Stream Sediment Reconnaissance
PASI – Preliminary Assessments and Site Investigations
USGS – U.S. Geological Survey

Note: Initial sample data sets used in this study, and further screened for usability as summarized, typically underwent earlier pre-selection data refinements as presented in Attachment A of the Upper Columbia River Site's: U.S. Environmental Protection Agency (site technical team) Level of Effort (LOE) for Assessment and Estimation of Upland Soils – Upper Columbia River Basin. 2016.

Table B-2: Replicate samples averaged prior to background analysis.

Soil Assessment	Duplicate	Triplicate	Triplicate + Split
Windward 2015	ADA-103-2mm	CBN-003-2mm	ADA-106-2mm
	ADA-171-2mm	-	ADA-169-2mm
	ADA-176-2mm	-	-
	ADA-180-2mm	-	-
	CBN-001-2mm	-	-
Wells 2015	4811881	Colville 7	-
	Colville 20	-	-
	Colville 3	-	-
	Colville 8	-	-
	Colville 9	-	-
	Deadman	-	-
USGS Midnite Mine	DSOR100S1	-	-
	DSOR129SI	-	-
	DSOR134S1	-	-
	DSOR313S1	-	-
NURE Reanalysis	24287	-	-
	DSOP089S1	-	-
	ONBB022S1	-	-
Hart Crowser Van Stone Mine	VS-BG-9-SD	-	-
	VS-BG-9-SS	-	-

NURE – National Uranium Resource Evaluation
USGS – U.S. Geological Survey.

Table B-3: Samples excluded from background analysis with additional rationale.

Station Name	Soil Assessment	Analyte List	Rationale for Exclusion
US005 US010 US012 TS029 TS031 TS038 TS043 TS061	Expanded Site Inspection (EPA, 2003)	Al, Sb, As, Ba, Be, Cd, Cr, Cu, Fe, Pb, Mn, Hg, Ni, Ag, Tl, Zn	Sample collection location is located within the 500-year floodplain of the Columbia River. Locations may be subject to influence from historical slag deposits in the river channel.
ONCE015S1	NURE-HSSR (Smith, 2006)	Al, As, Be, Cr, Cu, Fe, Pb, Mn, Ni, Ag, Zn	Sample collection location is immediately down drainage from Rammore and Hercules mines.
ONDH020S1			Sample collection location is immediately down drainage from Cleveland Mine.
RZAH023S1			Sample collection location is down drainage from Germania Mine/Mill.
24355 24361 24382 ONDE003S1	NURE Reanalysis (Grossman et al., 2004)	Al, As, Ba, Cr, Cu, Fe, Pb, Mn, Hg, Ni, Zn	Samples were excluded from background evaluation of the Reanalyzed NURE data by Church (2010a) due to mining influence.

Ag – silver, Al – aluminum, As – arsenic, Ba – barium, Be – beryllium, Cd – cadmium, Cr – chromium, Cu – copper, Fe – iron, Hg – mercury, Mn – manganese, Ni – nickel, NURE-HSSR – National Uranium Resource Evaluation Hydrogeochemical and Stream Sediment Reconnaissance, Pb – lead, Sb – antimony, Tl – thallium, Zn – zinc

Appendix C. Chloropleth Maps of Maximum 90th Percentile Background Concentrations.

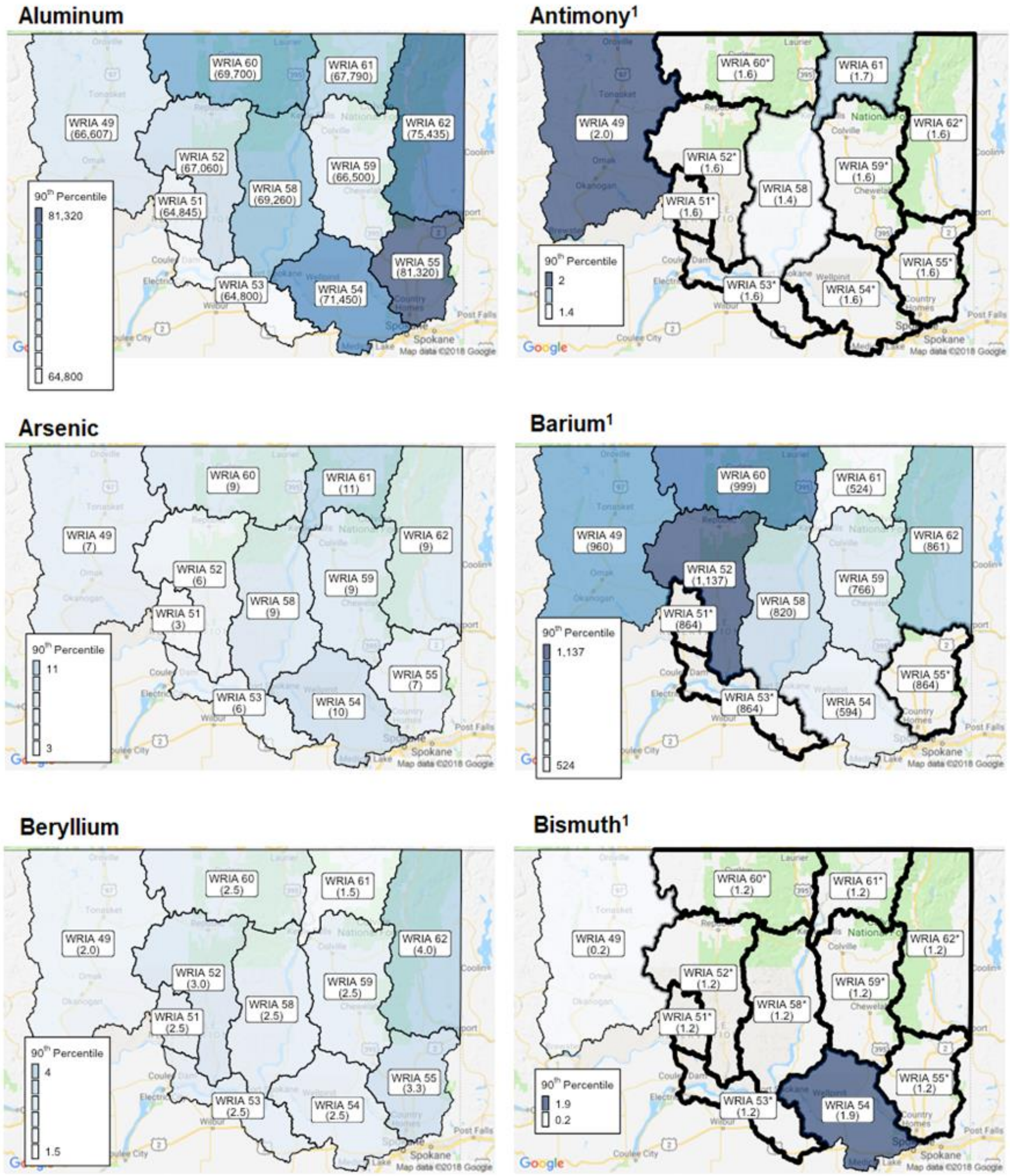

Figure C-1: Maximum 90th percentile *WRIA-Specific* background concentrations (mg/kg) of aluminum, antimony, arsenic, barium, beryllium, and bismuth in northeast Washington soils and sediments.

[1]Values for WRIAs identified with an asterisk (*) and bold WRIA outline are calculated from *All-Pooled* result due to an absence of WRIA-specific outcomes resulting from small sample size (n<10), and are intended for general comparison only

WRIA – Water Resource Inventory Area.

Figure C-2: Maximum 90th percentile *WRIA-Specific* background concentrations (mg/kg) of cadmium, chromium, copper, iron, indium, and lead in northeast Washington soils and sediments.

[1]Values for WRIAs identified with an asterisk (*) and bold WRIA outline are calculated from All-Pooled result due to an absence of WRIA-specific outcomes resulting from small sample size (n<10), and are intended for general comparison only.

WRIA – Water Resource Inventory Area

Figure C-3: Maximum 90th percentile *WRIA-Specific* background concentrations (mg/kg) of manganese, mercury, nickel, silver, thallium, and zinc in northeast Washington soils and sediments.

[1]Values for WRIAs identified with an asterisk (*) and bold WRIA outline are calculated from *All-Pooled* result due to an absence of WRIA-specific outcomes resulting from small sample size (n<10), and are intended for general comparison only.

WRIA – Water Resource Inventory Area

Appendix D. Data-use Considerations and Quallity Assurance.

The following considerations were taken while using this background dataset:

- In the USGS (Smith et al., 2013) and Hart Crowser Upper Columbia River (2013a) studies, samples collected at discrete depth intervals are assigned to the same sample ID.
- For Ecology's background analysis, replicate samples were averaged prior to use (Table B-2).
- Final quality assurance review of the metadata identified 132 duplicate records that are detailed in Table D-1.

Table D-1: Duplicate sample results in background dataset.

Duplicate Type	Duplicate Record Data Source	Primary Record Data Source	Sample ID	No. Stations	No. Records Duplicated	Analyte List
True Duplicate	NURE-Reanalysis	NURE-Reanalysis	SKAA037S1	1	1	Be
Substitution Error[1]	NURE-HSSR	USGS Midnite Mine	RZAH027S1, RZAH037S1	2	20	Al, As, Be, Cr, Cu, Fe, Mn, Ni, Pb, Zn
Substitution Error[1]	NURE-HSSR	USGS Midnite Mine	DSOR404S1, DSOR403S1, DSOR420S1, DSOR419S1, DSOR100S1, DSOR421S1, DSOR102S1, DSOR101S1, DSOR426S1, DSOR425S1, DSOR422S1, DSOR423S1, DSOR448S1, DSOR118S1, DSOR441S1, DSOR128S1, DSOR442S1, DSOR120S1, DSOR119S1, DSOR121S1, DSOR460S1, DSOR398S1, DSOR129S1, DSOR314S1, DSOR133S1, DSOR126S1, DSOR313S1, DSOR399S1, DSOR134S1, DSOR389S1, DSOR127S1, DSOR388S1, DSOR131S1, DSOR136S1, DSOR135S1, DSOR341S1, DSOR340S1	37	111	Al, Fe, Mn

[1]Duplication resulted from the augmentation of the background dataset with additional NURE-HSSR data retrieved from the USGS website (Smith, 2006).

Al – aluminum, As – arsenic, Be – beryllium, Cr – chromium, Cu – copper, Fe – iron, NURE-HSSR – National Uranium Resource Evaluation Hydrogeochemical and Stream Sediment Reconnaissance, Mn – manganese, Ni – nickel, Pb – lead, USGS – U.S. Geological Survey, Zn – zinc

Final Background Determinations with Additional Qualifying Rationale

Aluminum

For aluminum, the total available sample pool is n=4,347, composed of soil, NURE, and non-NURE samples. All 11 WRIAs have calculated 90th percentiles. A 90th percentile of 24,870 mg/kg from WRIA 61 (50 soil samples) represents a concentration approaching 2.5 times lower than the next lowest WRIA result (WRIA 59 at 60,770 mg/kg; n=84 NURE samples). Lower aluminum soil measurements from total recoverable analytic digestion methods in WRIA 61 within historical smelter-emission impact areas have been previously observed in soil results (Hart Crowser, 2013). While analytical partial digestion methodology could influence these lower values and WRIA 61 is not outside the potential range of carbonate crustal abundance (University of Oklahoma, 1985; Hansford & Boerngen, 1984; Gunter, 1998), the WRIA 61 soil 90th percentile is out of range in the context of watershed scale 90th percentile background applications. WRIA 61 smelter impact could also be a factor, but no clear anthropologic or geologic WRIA-specific modifying factors were established. Removal of the lower WRIA 61 value would result in a range of 60,770 to 81,320 mg/kg. The final background range includes WRIA 61 and is 24,870 to 81,320 mg/kg. This range considers all the tiered selection steps and analytics, and is selected as the upper-percentile natural background.

Antimony

Certain metals results, such as antimony, were notably limited in total sample pool counts. Because of the small sample counts, individual samples within specific WRIAs or sources resulted in susceptibility to significant movement of the 90th percentile outcomes. For antimony, the total available data pool is n=111, composed of soil and non-NURE samples. The WRIAs with n≥10 having reported 90th percentiles were WRIA 49, 58, and 61, and the concentration range 0.49 to 2.0 mg/kg. The soil and source samples from WRIAs 49 and 58 did not exceed n=11. Sixty-nine of the total sample count (62 percent) were located in WRIA 61. Within WRIA 49, a single soil sample of 2.4 mg/kg results in a 90th percentile of 2.03 mg/kg; without that sample, the 90th percentile becomes 1 mg/kg.

From WRIA 61, one non-NURE sample result of 7 mg/kg notably influences certain 90th percentile results, in particular the pooled non-NURE (n=14) result (2.53 [with] vs. 0.49 mg/kg [without]). This sample is suspect in light of the small data pool and locational considerations for this metal. Antimony is an established smelter emissions pollutant (SO_2 exclusion area) in WRIA 61. Ecology’s present-day natural background characterization of northeast Washington upland lakes established an antimony 90th percentile of 0.14 mg/kg (Ecology, 2011). While potentially too broad of an upper-percentile range given the above observations, in the final analysis an overall study 90th percentile range of 0.49 to 2.03 mg/kg is accepted for natural background.

Arsenic

For arsenic, the total available sample pool is n=1,284, composed of soil, NURE, and non-NURE samples. All 11 WRIAs have calculated 90th percentiles, the concentration range being 3 to 20 mg/kg. This range is the upper-percentile natural background.

Barium
For barium, the total available sample pool is n=281, composed of soil, NURE, and non-NURE samples. Eight WRIAs have calculated 90th percentiles, the concentration range being 432 to 1,137 mg/kg. This range is the upper-percentile natural background.

Beryllium
For beryllium, the total available sample pool is n=1,459, composed of soil, NURE, and non-NURE samples. All 11 WRIAs have calculated 90th percentiles, the concentration range being 0.7 to 3.3 mg/kg. This range is the upper-percentile natural background.

Bismuth
For bismuth, the total available sample pool is n=76 composed of soil and non-NURE samples. The WRIAs with $n \geq 10$, having calculated 90th percentiles were limited to WRIA 49 and 54, the concentration range being 0.2 to 2.1 mg/kg. This range is the upper-percentile natural background.

Cadmium
For cadmium, detection limitations reduced the available pool to n=186 soil and non-NURE samples. Five WRIAs (*WRIA-Specific)* have calculated 90th percentiles. Of the total pool of samples, 74 were located in WRIA 61. A 90th percentile of 3.62 mg/kg is calculated for cadmium from WRIA 61 (62 soil samples). Similarly, WRIA 61 also biases the total combined soil (*All-WRIA-Source)* outcome (2.42 with WRIA 61 soil vs. 0.50 mg/kg without) as well as significantly influencing the *All-Pooled* result (1.50 mg/kg with WRIA 61 soil vs. 0.60 without). The result of 3.62 mg/kg is more than 4 times greater than the next highest 90th percentile result. Additionally, cadmium data from WRIA 61 has a significant effect on the 90th percentile results for *All-Pooled* and *All-WRIA-Source* data. Removal of WRIA 61 soil data is appropriate for the estimation of representative background cadmium concentrations in northeast Washington.

Cadmium is an established historical metal smelter air emission pollutant affecting uplands of the Upper Columbia River Valley. The 90th percentile soil outcome from WRIA 61 indicates that the predefined historical SO_2 forest-injury exclusion footprint along the river valley is inadequate for cadmium enrichment impacts, being significantly above reasonable maximum natural background range limits. For comparison, Ecology’s present-day natural background characterization of northeast Washington upland lakes established a cadmium 80th percentile (normal distribution method) of 0.63 mg/kg (Ecology, 2011). Removal of the WRIA 61 soil results is appropriate for upland watershed-scale background value estimation. The revised range is 0.4 to 0.84 mg/kg. This is the upper-percentile natural background range.

Chromium
For chromium, the total available sample pool is n=1,491, composed of soil, NURE, and non-NURE samples. All 11 WRIAs have calculated 90th percentiles. The 90th percentile of 164 mg/kg for chromium covering WRIA 60 (driven by 174 NURE samples) represents the highest WRIA upper-most-bound concentration. A review of geologic bedrock identified certain tributaries in WRIA 60 with high reported concentrations in association with the

presence of chromium-bearing ultramafic mineralization. WRIA 60, and potentially others (WRIAs 52 and 53), exhibit background values considerably higher than most other watersheds. Caution is advised for chromium background range applications, as the maximum value in particular may be best reserved to WRIA 60 only. With WRIA 52, 53, and 60 removed—the highest upper-bound value would be 68 mg/kg (WRIA 49). The concentration range including WRIA 52, 53, and 60 is 35 to 164 mg/kg. This is the upper-percentile natural background range.

Copper

For copper, the total available sample pool is n=1,411, composed of soil, NURE, and non-NURE samples. All 11 WRIAs have calculated 90th percentiles, the concentration range being 16 to 49 mg/kg. This is the upper-percentile natural background range.

Indium

For indium, the total available sample pool is n=34, composed of soil samples. The data are distributed across nine WRIAs, with only a single WRIA (49) with n≥10, and having calculated 90th percentile. Data spanned detection limitations of 0.02 to 0.06 mg/kg (WRIAs 52, 53, 54, 55, and 62). The 90th percentile outcomes for both WRIA 49 (n=10) as well as the combined WRIA data total (n=34) for this small data set yields a concentration range being 0.05 to 0.06 mg/kg. While influenced by detection limitations, this is the upper-percentile natural background range.

Iron

For iron, the total available sample pool is n=4,289, composed of soil, NURE, and non-NURE samples. All 11 WRIAs have calculated 90th percentiles. The value of 26,500 mg/kg reported for WRIA 61 soil is low compared to most other results, though not reaching a factor difference of 1.5. By including this lowest value, the concentration range is 26,950 to 54,900 mg/kg. This is the upper-percentile natural background range.

Lead

For lead, the total available sample pool is n=1,486, composed of soil, NURE, and non-NURE samples. All 11 WRIAs (*WRIA-Specific* and *WRIA-Source*) have calculated 90th percentiles. The 90th percentile (*WRIA-Source*) of 106 mg/kg for lead soil covering WRIA 61 (driven by 62 soil samples) represents an upper-bound WRIA concentration approaching 2.5 times the next highest WRIA (WRIA 62 at 43 mg/kg; n=11 soil samples). WRIA 61 also notably influences the *WRIA-Specific* and *All-WRIA-Source* soil 90th percentile outcomes (99 and 86 mg/kg, respectively).

Lead is an established historical metal-smelter air-emission pollutant affecting uplands of the Upper Columbia River Valley. The 90th percentile outcomes from WRIA 61 indicate that the predefined historical SO_2 forest-injury exclusion footprint along the river valley is inadequate for lead enrichment in the area—remaining significantly above an appropriate maximum natural background range for lead. Regional underlying geologic bedrock terrain, frequently carbonates, does not support elevated concentrations, except in localized zones of ore mineralization (Gunter 1998). Removal of WRIA 61 for reporting the range of watershed-scale background values is appropriate, the revised concentration range being 19 to 43 mg/kg. This is the upper-percentile natural background range.

Manganese
For manganese, the total available sample pool is n=4,341, composed of soil, NURE, and non-NURE samples. All 11 WRIAs have calculated 90th percentiles. The 90th percentile of 2,493 mg/kg for manganese covering WRIA 62 (11 soil samples) represents a concentration approximately 1.6 times the next highest WRIA (WRIA 61 at 1,570 mg/kg; n=61 NURE samples). Re-evaluation of WRIA 62 did not identify specific anthropogenic or geologic exclusionary anomalous conditions. Caution is advised because the application of an upper-range value of 2,493 mg/kg for WRIA 62 appears appropriate, but the 90th percentile range for applications to other area concentrations (ignoring WRIA 62 result) for background is reported as 621 to 1,570 mg/kg. In summary, the concentration range including all outcomes is 621 to 2,493 mg/kg. This is the upper-percentile natural background range.

Mercury
For mercury, the total available sample pool is n=208, composed of soil and non-NURE samples. Seven WRIAs (*WRIA-Specific*) have calculated 90^{th} percentiles. The 90^{th} percentile *WRIA-Source* of 0.27 mg/kg for mercury from WRIA 58 (n=31; dominated by 21 non-NURE samples) represents a concentration 1.6 times the next highest WRIA (WRIA 59 at 0.15 mg/kg; n=10 non-NURE samples). WRIA 58 non-NURE samples also strongly influence the combined WRIA 58 outcome (0.24 mg/kg).

Previous Ecology inland lake studies observed potentially higher area mercury concentrations in WRIA 58 surface sediments (Ecology, 2011; Ecology, 2013). Local anthropogenic factors were not ruled out. The 90^{th} percentile outcomes from WRIA 58 indicate that undefined anthropogenic sources for mercury may be causing enrichment above reasonable maximum natural background range limits. Ecology's natural background characterization of northeast Washington upland lakes established a mercury 90^{th} percentile of 0.12 mg/kg (Ecology, 2011). Taken collectively, the removal of the WRIA 58 outcome for reporting the range of watershed-scale background values is appropriate. The corrected range of concentrations for natural background is 0.02 to 0.15 mg/kg. This is the upper-percentile natural background range.

Nickel
For nickel, the total available sample pool is n=1,486, composed of soil, NURE, and non-NURE samples. All 11 WRIAs have calculated 90th percentiles. The concentration range being 12 to 50 mg/kg. This is the upper-percentile natural background range.

Silver
For silver, the total available sample pool is n=1176, composed of soil, NURE, and non-NURE samples. Ten WRIAs have calculated 90th percentiles, the concentration range being 0.4 to 1.0 mg/kg. The 1.0 mg/kg result was from WRIA 55. In summary, the concentration range including all outcomes is 0.4 to 1.0 mg/kg. This is the upper-percentile natural background range.

Thallium

For thallium, the total available sample pool is n=151, composed of soil and non-NURE samples. Four WRIA 90th percentile results were reported. Other data (n<10) are distributed across other WRIAs. The WRIAs with n≥10, having calculated 90th percentiles are WRIAs 49, 54, 58, and 61. The upper-percentile WRIA 90th percentile concentration range being 0.2 to 0.7 mg/kg. Note that the 0.2 outcome (WRIA 61 soil) appears to be a detection-limiting result, thus the lower end of the background range in actuality may be lower. The 0.2 value approaches being two times less than the next lowest 90th percentile (0.4 mg/kg). The significance of the observed difference could not be established. The range carried forward is 0.2 to 0.7 mg/kg. This is the upper-percentile natural background range.

Zinc

For zinc, the total available sample pool is n=1,488, composed of soil, NURE, and non-NURE samples. All 11 WRIAs (*WRIA-Specific* and *WRIA-Source*) have calculated 90th percentiles. The 90th percentile *WRIA-Source* soil of 347 mg/kg for zinc for WRIA 61 (62 soil samples) represents a concentration approaching 2.5 times the next highest WRIA (WRIA 62 at 139 mg/kg; n=11 soil samples). WRIA 61 also strongly influences the *WRIA-Specific* and *All-WRIA-Source* soil outcomes (252 and 225 mg/kg).

Zinc is an established historical metal-smelter air-emission pollutant affecting uplands of the Upper Columbia River Valley. The 90th percentile outcomes from WRIA 61 indicate that the predefined historical SO_2 forest-injury exclusion footprint along the river valley is inadequate for zinc enrichment and WRIA 61 results remain significantly above a reasonable maximum natural background range. Removal of the WRIA for reporting the range of watershed-scale background values is appropriate. The range of concentrations for natural background after removal of the WRIA 61 90th percentile outcomes is 55 to 139 mg/kg. This is the upper-percentile natural background range.

Appendix E. WSU CISER Report Prepared For Ecology

Preface to Appendix E

The following supplemental information is provided to help the reader interrelate the Washington State University (WSU) statistical analyses report in this appendix to that of the data source and *data group categories* applied in the main report.

The WSU report refers to three different organized data sources that underwent statistical exploratory and percentile outcome presentations. These relate to the main report terminology as follows:

Identified WSU source analyses	**Main Report**[1]
Soil	Soil
Sediment (NURE); sedi_NURE; NURE	NURE
Sediment (non-NURE); Sedi_nonNURE; nonNURE	non-NURE

[1] Described in the Statistical Methods and Analysis section.

WSU tables 4.1–4.18 in Appendix E summarize statistical outcomes associated with grouping and combination variations of the sources: Soil, NURE, and non-NURE. The following list and WSU table for aluminum provide an example associating the four main report *data group categories* (see Statistical Methods and Analysis section for definitions) to the WSU tables.

- *All-Pooled:* All data are pooled and presented as one data set.
- *All-WRIA-Source:* Pooled results from all combined Water Resource Inventory Areas (WRIAs) assigned to each of the three data sources: NURE, non-NURE, and soil.
- *WRIA-Specific:* Individual WRIA results presented by WRIA and calculated from combined data within each WRIA, with sufficient sample size (n ≥ 10).
- *WRIA-Source:* WRIA results according to each of the three data sources having sufficient sample size (n ≥ 10): NURE, non-NURE, and soil.

In addition, the established minimally and maximally assigned range of 90th percentiles (and also 50th and 75th percentile range information) for each metal in Table 2 of the report is derived from the *WRIA-Specific* and *WRIA-Source* results. The two 90th percentile range values for aluminum (24870 and 81320) also are **bold** highlighted in the example table, below. For indium, available or appropriate data are limited, prohibiting robust *WRIA-Specific* or *WRIA-Source* 90th percentile calculations. In this case, the 90th percentile range included incorporation of the *All-Pooled* results.

Table E-1. Example WSU aluminum table.

Pooled	WRIA	Source	n	IQR	50th Percentile	75th Percentile	90th Percentile	Lower bound for 90th percentile	Upper bound for 90th percentile
All			4347	10500	58827	64100	69800	69200	70200
Source	49		366	8280	58130	62315	66607	64813	67700
	51		42	6077	58287	61490	64845	57149	68183
	52		395	7347	59100	62763	67060	65614	68472
	53		249	7082	57400	61282	64800	63675	66460
	54		626	9580	59800	64675	71450	69000	73355
	55		175	15697	65900	73850	**81320**	78940	84020
	58		753	12613	57800	64100	69260	68340	70440
	59		101	13600	52500	58200	66500	60700	72600
	60		1442	10775	59700	64775	69700	68910	70300
	61		168	40762	54550	61700	67790	66300	69880
	62		30	58980	43250	70580	75435	63040	80249
WRIA		Non-NURE	195	59690	23500	69840	74316	72182	75592
		NURE	4046	9800	59060	64000	69367	68983	69882
		Soil	106	49413	20700	65000	72950	65150	75650
None	49	Non-NURE	19	8425	71230	73315	76758	62046	80388
	49	NURE	337	7747	57613	61060	64853	63893	65814
	49	Soil	10	5675	68400	72175	77430	72660	85030
	51	NURE	40	5965	57677	61225	62325	58990	63203
	52	NURE	383	7273	58900	62397	65789	64379	66842
	53	NURE	241	6515	57307	60615	63800	62600	64900
	54	Non-NURE	49	14000	21800	29500	69774	64908	105622
	54	NURE	573	8700	60400	64900	71653	69220	73806
	55	NURE	169	16000	65893	73300	81107	78433	83993
	58	Non-NURE	55	7585	8840	13500	60876	49042	104152
	58	NURE	687	11704	58600	64437	69340	68420	70580
	58	Soil	11	53875	12732	64700	70200	59800	127200
	59	Non-NURE	12	27290	59923	70438	77510	75620	87900
	59	NURE	84	11125	51800	56475	60770	56840	62740
	60	Non-NURE	16	65685	7660	70965	74170	64850	83025
	60	NURE	1419	10600	59700	64700	69700	69054	70387
	61	NURE	113	10200	59700	64200	68680	64065	70260
	61	Soil	50	5496	17550	20675	**24870**	22630	29007
	62	Non-NURE	19	62420	43150	70570	75990	67280	82006
	62	Soil	11	42257	58300	68950	74600	33200	90200

IQR – interquartile range

The rest of Appendix E is available only on the internet, linked to this report at: https://fortress.wa.gov/ecy/publications/SummaryPages/1903014.html.