

A HOLISTIC ASSESSMENT OF TRACE METAL MOBILITY NEAR
ABANDONED MINE SITES IN THE WHEATON RIVER WATERSHED,
YUKON, CANADA

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Thesis Abstract

The Wheaton River Watershed, located in southern Yukon, feeds Bennett Lake and ultimately the Yukon River, and has been heavily impacted by recreation and mining activities dating back to the Klondike Gold Rush (1896). This watershed is within the Traditional Territory of the Carcross/Tagish First Nation (C/TFN), who have stewarded these lands since time immemorial, and hold concerns over impacts of point source contamination on broader ecosystem health. Despite the common understanding of ecosystem connectivity among locals, assessments of trace metal mobility across multiple environmental media are overlooked when approaching research using perspectives exclusively rooted in Western Science. Despite this knowledge, no assessment of trace metal mobility has been conducted in the watershed, leaving questions surrounding the extent of contaminant point source impacts on the catchment as a whole largely unanswered.

Co-created with C/TFN staff and community members, this thesis took a holistic approach to assess the current distribution and potential for trace metal mobility on a watershed scale. Water, plant, scat and sediment samples collected at mine-impacted and downstream sites were analyzed for trace metals, focusing on those posing known risks to human and animal health (As, Cd, Hg, Pb, and Sb) to investigate the extent of metal mobility between media and throughout the catchment, supported by sequential extraction and stable isotope analyses. There were both region and metal-specific patterns of trace metal distribution and mobility, with most trace metals stored in sediments at mine-impacted sites. Metals in water samples followed similar trends to solid samples; however, lower concentrations, especially in the mainstem Wheaton River relative to mine-impacted samples, suggested either or both low mobility and dilution from other water sources as driving factors at this time. Flux analyses and yield calculations indicated that the cumulative Wheaton River trace metal exports into Bennett Lake were comparable or below western Arctic references. This thesis explores trends of multi-media metal mobility from mine-impacted sites on a watershed scale, informed by perspectives rooted in Western Science and Indigenous knowledge, highlighting the complex, connected, and dynamic nature of the watershed. Findings from this thesis will help inform land planning and stewardship initiatives led by C/TFN, remediation efforts, and guide future research in the region.

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Glossary

AA = Arctic Amplification

AGS = Arctic Gold and Silver Mine

ANOVA = Analysis of Variance

As = Arsenic

BNAA = British North America Act

C/TFN = Carcross/Tagish First Nation

Ca = Calcium

Cd = Cadmium

CO_3^{2-} = Carbonate Ion

Co = Cobalt

Cr = Chromium

Cu = Copper

DO = Dissolved Oxygen

DOC = Dissolved Organic Carbon

DOM = Dissolved Organic Matter

EF = Enrichment Factor

EPA = (US) Environmental Protection Agency

Fe = Iron

FWC = Flow-Weighted Concentrations

GP = Goddell Portal

HWW = How We Walk with Land and Water

ICP-MS = Inductively Coupled Plasma Mass Spectrometry

ICP-OES = Inductively Coupled Plasma Optical Emission Spectrometry

IJC = International Joint Commission

IKS = Indigenous Knowledge Systems

KDFN = Kwanlin Dün First Nation

LW = Lower Wheaton

MeHg = Mercury (methylated)

Mg = Magnesium

MMU = Moose Management Unit

Mn = Manganese

Ni = Nickel

OWS = Open Water Season

PCA = Principal Component Analysis

Pb = Lead

Sb = Antimony

SD = Standard Deviation

SE = Standard Error

SEP = Standard Error of Prediction

SM = Skukum Mountain

TEK = Traditional Ecological Knowledge

THg = Mercury (total)

TKC = Ta'an Kwäch'än Council

TRTFN = Taku River Tlingit First Nation

TTC = Teslin Tlingit Council

UFA = Umbrella Final Agreement

USGS = United States Geological Survey

UW = Upper Wheaton

WS = Western Science

YFN = Yukon First Nations

YG = Yukon Government

Zn = Zinc

YRITWC = Yukon River Inter-Tribal
Watershed Council

Statement of Authorship Contributions

Conceptualization of the study presented herein was conducted by Kate Rorabeck (KR), Kyra St. Pierre (KSP), Tyler Obediah (TO; C/TFN), Amaya Cherian-Hall (ACH; C/TFN), Ashley Dubnick (AD; Yukon University), and Colleen James (CJ; C/TFN). Data compilation, analysis, figure generation and writing were conducted by KR under the supervision of and with guidance from KSP, incorporating feedback from AD. Chapters 1 and 3 were written by KR with feedback and comments from KSP. Local context informing writing was provided by TO, ACH, CJ, and Laurence Patterson-Smith (LPS; C/TFN). Chapter 2 was prepared as a manuscript for *Environmental Science: Processes and Impacts*, and drafted while incorporating feedback from KSP and AD at the time of thesis submission with feedback from the remaining co-authors to follow. All co-authors were involved in study design and fieldwork. Sample analyses performed at BASL, JVSIL, LANSET, and UOGL service laboratories, where KR assisted with sample analysis and method development, with guidance from laboratory technicians and KSP. We declare no conflicts of interest relevant to this study. We declare no ethics approval was required. The research funding was supported by the University of Ottawa, the Natural Sciences and Research Council of Canada (NSERC), and Northern Contaminants Program (NCP).

Chapter 1: Thesis Introduction

Background and Literature Review

1.1 Stressors in the Western Canadian Sub-Arctic

1.1.1 Mining in the North

Pre-Gold Rush Mineral Extraction and Trade in Northwestern Canada

Prior to European contact, the mining and trading of extracted goods was conducted by Indigenous peoples across what is now referred to as southern and northern Canada. Tools created from copper mined in the Great Lakes region (Fowler, 1973; Mihalicz et al., 2022), southwestern Yukon (Cooper & Simonetti, 2021; Mihalicz et al., 2022), and along the Coppermine River in the Northwest Territories (Gilbert, 1931; Mihalicz et al., 2022) have been traced across North America and are attributed to trade activity among Indigenous peoples prior to European contact and influence. Additionally, products made with mined obsidian were used and traded within northern British Columbia, Yukon, and Alaska (Mihalicz et al., 2022). Following European contact and establishment of settlements, European ‘explorer’ Sir Martin Forbisher travelled across northern Canada in the quest for the Northwest Passage, taking interest in valuable minerals on Baffin Island in the late 1500s (Marsh & Panneton, 2008). Infrastructure development in the late 1800s by an influx of settlers triggered a series of mineral discoveries including nickel-copper ore in northern Ontario towns of Sudbury (1883) (Mihalicz et al., 2022) and silver near Cobalt Lake (1903) (Town of Cobalt, 2026), all while constructing the Canadian Pacific Railway. While these discoveries of minerals were happening in eastern Canada, prospectors from Canada and the United States flocked to northwestern Canada in the pursuit of gold in the late 1800s and early 1900s (C/TFN Government, 2014; Gates, 2009).

Klondike Gold Rush

The Klondike Gold Rush originated in the late 1800s, resulting in a large influx of people from southern Canada and United States to the Yukon (Gates, 2009). To reach the mining site, travelers usually hiked from Alaska over the Coast Range Mountains until they reached the Yukon River headwaters, eventually travelling north by boat (Gates, 2009). The high demand for travel along this route inspired the construction of the White Pass Trail Railway from Skagway, Alaska to

Lindeman, BC, and eventually to Carcross in the Yukon in the early 1900s (C/TFN Government, 2014). The initial construction and use of the railway had negative impacts on the citizens of the Carcross/ Tagish First Nation (C/TFN), impacting trading and trapping regimes (C/TFN Government, 2014). This large influx of people to Canada's North triggered widespread mining infrastructure development and mineral exploration of several regions (Gates, 2009).

20th Century Mining in Northwestern Canada

In the mid to late 1900s, advancement in technology, increased infrastructural capacity, and changing environmental conditions (e.g., glacial melt, thawing permafrost) have rendered some mineral deposits in Canada's North more accessible, driving increases in capacity for exploration and extraction (Jones et al., 2024; Palmer et al., 2025; Rodon et al., 2022). In Canada's subarctic, several largescale operations for extraction of minerals including gold, silver, uranium, copper, zinc, and diamonds were established in the mid to late 1900s (Jones et al., 2024; Rodon et al., 2022; Shigley et al., 2016) (Figure 1).

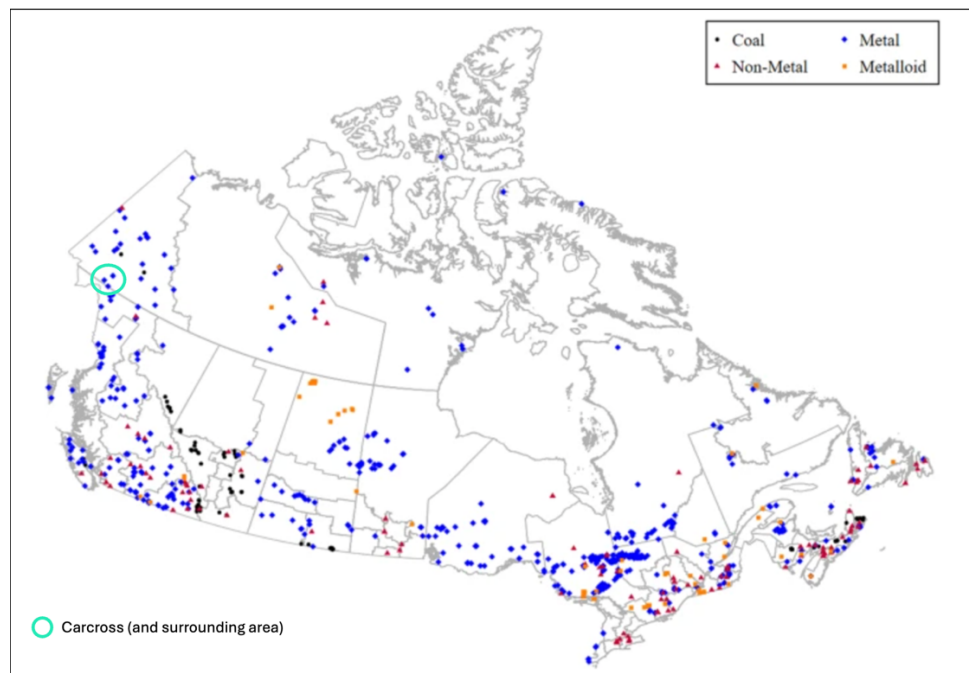


Figure 1. Map displaying spatial distribution of mines in MinCan dataset by mineral type (Dallaire-Fortier, 2024) and modified to note Carcross and surrounding area.

In Yukon, several mines have been active in the past and present, primarily mining gold, silver, lead, zinc, and copper ores (Fyles et al., 1960; Yukon Government, 2025d). Major mine licenses have been granted by the Yukon Government (YG) following a transfer of responsibility from the federal government in 2003 (Yukon Government, 2025d). Major mine licenses granted by YG include Carmacks Copper, Eagle Gold, Keno Hill Silver, Minto, and Wolverine Mines (Figure 2). Other major mining licenses granted by the federal government prior to 2003 are displayed in Figure 2.

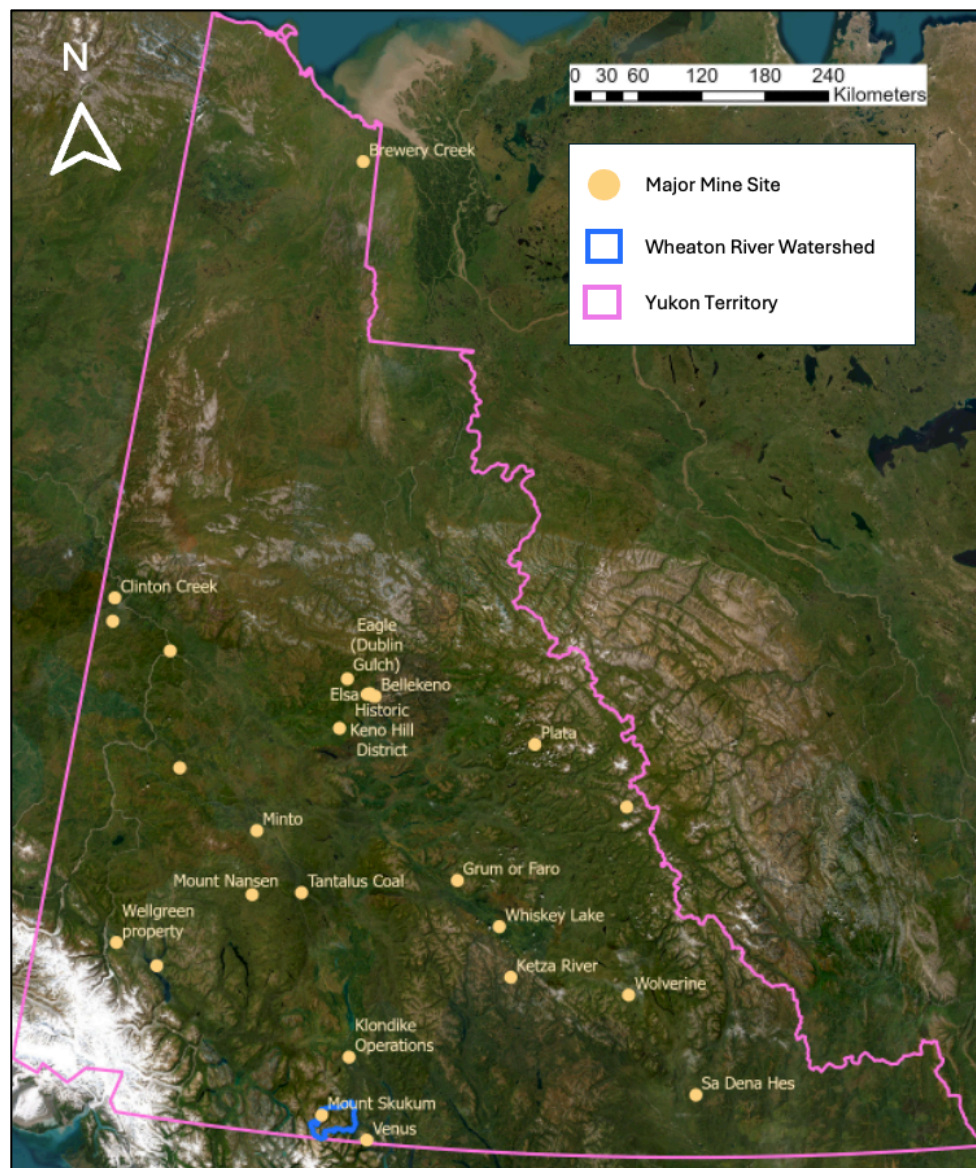


Figure 2. Major Mine Operations in the Yukon Territory (Dallaire-Fortier, 2023; MacNeil et al., 2025).

Social and Environmental Implications of Mining in the North

The social and environmental impacts of mining in the North on locals, including First Nation, Métis and Inuit communities cannot be understated. Given the ‘boom-and-bust’ nature of remote resource economies, many sites have been suddenly closed or abandoned, outcomes that were historically paired with minimal obligations for remediation (Sandlos & Keeling, 2016). During production and following closure, mines perpetuate a series of social and environmental issues including abandoned contaminated sites, disrupted access to traditional lands, social unease, and an economy reeling from the habitual ‘boom-and-bust’ nature of mining operations (Monosky & Keeling, 2021; Palmer et al., 2025). This wide array of negative implications are all known to impact environmental health through potentially widespread contamination of land, water, and all who rely on them (Jones et al., 2024; Palmer et al., 2025; Sandlos & Keeling, 2016). Extraction greatly increases surface concentrations of those metals, thereby influencing natural biogeochemical cycles by increasing bioavailability and potential for ecosystem transport (Yin et al., 2021). To date, much of mine remediation efforts are regarded as purely technical or scientific issues, often resulting in limited to no inclusion of Indigenous knowledge in decisions (Monosky & Keeling, 2021; Sandlos & Keeling, 2016). The omission of Indigenous perspectives has often resulted in inadequate remediation, which can broadly impact Indigenous wildlife management, subsistence hunting, and safe, sustainable interaction with all aspects of the surrounding environment (Monosky & Keeling, 2021; Sandlos & Keeling, 2016).

1.1.2 Climate Change

The Arctic is warming at three to four times the global average (Rantanen et al., 2022). This accelerated warming is a phenomenon called Arctic Amplification (AA), fed by changing oceanic current patterns, ice-albedo feedback and near surface temperature inversion (England et al., 2021). AA has resulted in drastic changes in the physical and chemical structure of arctic and subarctic landscapes, including through the deepening of the active layer, which itself resulting in increased erosion, metal mobility, and changes in vegetation composition across the landscape (Jin et al., 2021; O’Donnell et al., 2024; Skierszkan et al., 2024, 2025).

Mountainous regions are particularly sensitive to changes associated with anthropogenic climate change (Knight, 2022; McDowell et al., 2023; Rasouli et al., 2014). Warming weather and increased frequency and magnitude of precipitation in these regions have been connected to

changing landslide activity (Cloutier et al., 2017; Huggel et al., 2012; Mann et al., 2025; Owczarek et al., 2020). Although landslides are ubiquitous in mountainous regions under a stable climate, these events are likely to increase in frequency and magnitude with increased temperatures, precipitation, and magnitude of storm events (Cloutier et al., 2017; Huggel et al., 2012; Mann et al., 2025; Owczarek et al., 2020). Warming climate has driven increased rockfall activity within mountainous regions, particularly in the summer with permafrost thaw (Stoffel et al., 2024). Both rockfalls and landslides undoubtedly have great influence over the biophysical diversity of affected regions, altering soil, sediment and vegetation, and freshwater composition, paths and magnitude of sediment transport, and wildlife habitat and mobility (Cloutier et al., 2017; Geertsema & Pojar, 2007; McDowell et al., 2023). Further, the impacts on local, Indigenous communities, including through the reshaping of ancestral trails and impacting travel through mountainous areas cannot be understated (McDowell et al., 2023).

1.2 Biogeochemistry of Metals of Interest

1.2.1 Arsenic

Arsenic (As) is a metalloid that can assume several different oxidation states (-3, -1, 0, +3, and +5) (Couture et al., 2010; Rodie et al., 1995). A principal source of arsenic to the environment is the weathering of arsenopyrites (-1), where its microbial oxidation can enhance the release of As into its dissolved phase (Rodie et al., 1995). Arsenite (+3), which is highly toxic and mobile in the environment and arsenate (5+), known to bind with Fe oxide minerals, are the predominant oxidation states assumed in reducing and oxidizing environments, respectively (Rodie et al., 1995; Schlesinger et al., 2022; Smedley & Kinniburgh, 2002). Arsenic plays no known essential role in biochemistry and has toxic and carcinogenic effects at low concentrations, with sediment-bound arsenic posing risk to drinking water safety (Couture et al., 2010; Hong et al., 2014; Schlesinger et al., 2022). In humans, As exposure can lead to the development of various cancers, diabetes, neurological disorders, cardiovascular disorders, and damage to reproductive organs (Hong et al., 2014). In natural environments, As can be bound to organic or inorganic ligands, or as arsine gas, all with different properties and toxicological effects (Rodie et al., 1995).

Arsenic is naturally occurring in bedrock geology but can also be further propagated through ecosystems through anthropogenic activities including mining operations and release of other industrial effluents (Palmer et al., 2024; Rodie et al., 1995). Anthropogenic sources include mining,

phosphorous-containing fertilizer runoff and fuel combustion, which have dominated the global biogeochemical cycling of As (Schlesinger et al., 2022). The mining of As-bearing sulfides is common during the mining of gold, silver, copper, lead, zinc, nickel and iron (Jones et al., 2024; Rodie et al., 1995; Schlesinger et al., 2022; Sivarajah et al., 2024). As a result, ecosystems impacted by mining can experience widespread environmental As contamination in sediment, water, and vegetation (Palmer et al., 2025; Sivarajah et al., 2024).

Aquatic Geochemistry

The mobility and speciation of As throughout freshwater systems is influenced by redox potential, dissolved organic carbon (DOC), iron (Fe) and sulfur (S) concentrations (Couture et al., 2010; Palmer et al., 2024; Rodie et al., 1995). Like phosphorous, As forms a triprotic acid in aqueous solution, which is known to be an equilibrium of oxidized states in aqueous solutions (Schlesinger et al., 2022). In oxic waters, As (+5) dominates as triprotic arsenic acid, and in anoxic waters, As (+3) dominates as triprotic arsenous acid (Rodie et al., 1995; Schlesinger et al., 2022). Under oxic conditions, iron (+3) dominates and is favoured to form Fe (oxy)hydroxides that have a high affinity to arsenate (Couture et al., 2010; Palmer et al., 2024; Rodie et al., 1995).

Arsenic abundance and transformation in riverine systems is largely controlled by geological contributions and interactions within river sediment (Gorny et al., 2015; Palmer et al., 2024; Schlesinger et al., 2022). However, bedrock concentrations were seen to have a smaller influence on As concentrations in rivers with neutral pH and typical alkalinity due to oxidation of As, commonly followed by adsorption to fluvial sediments (Smedley & Kinniburgh, 2002). Further, river waters are often diluted by surface runoff, influencing the transport of As in the system (Smedley & Kinniburgh, 2002).

Terrestrial Biogeochemistry

Natural concentrations of As in sediments and soils are largely related to local geology, with higher concentrations typically associated with bog, peaty, and acid sulfate soils (Kossoff & Hudson, 2012). Like aquatic environments, speciation of As, and its uptake by vegetation, is largely controlled by Eh, pH, microbial activity, and DO: however, root physiology can also control uptake (Kossoff & Hudson, 2012; Vithanage et al., 2012). Uptake by plants of As is variable among species, with arsenate uptake considered more favourable under oxic conditions (Vithanage

et al., 2012). Chemical similarities of As and phosphorous species can play a major role in plant-soil interactions and uptake by vegetation, as arsenate is commonly taken up by vegetation using the same pathways as phosphate (Vithanage et al., 2012). Different trends related to conditions for As uptake by horsetail have been observed surrounding a mine in Cobalt, Ontario where plants were seen sequestering As through the formation of an oxidized iron-bearing plaque on plant roots (Turcotte et al., 2025). To date, widespread knowledge of the dynamics of vegetation uptake of As remains limited.

1.2.2 Cadmium

Cadmium (Cd) is a transition metal exclusively held within a 2+ oxidation state, with similar chemical properties to zinc and mercury (Ankush et al., 2024; Jha & Kumar, 2024). Cadmium is commonly found as an impurity in zinc, lead, and copper ores, and can enter the environment through natural and anthropogenically driven processes (Ankush et al., 2024; Jha & Kumar, 2024). Natural liberation of Cd occurs through weathering of local geology, eruption of Cd-containing volcanic ash, and from forest fires resulting in burning of Cd-containing vegetation and organic matter (Ankush et al., 2024; Jha & Kumar, 2024; Satarug et al., 2010). Anthropogenic processes associated with release of Cd into the environment include industrial activities such as smelting, refining and effluent emissions, mining operations, fossil fuel combustion, and the synthesis and use of Cd-containing fertilizers (Jha & Kumar, 2024; Verbruggen et al., 2013). Cadmium is a heavy metal with unique toxicological properties (Gamberg et al., 2005; Jha & Kumar, 2024; Satarug et al., 2010). Once ingested, Cd has a high affinity for absorption to kidneys, where it can accumulate and cause significant physiological damage (Jha & Kumar, 2024; Satarug et al., 2010). Cadmium has also been shown to contribute to chronic health issues within the gastrointestinal tract, lungs and liver of exposed animals, all influenced by speciation (Jha & Kumar, 2024; Satarug et al., 2010).

Terrestrial Biogeochemistry

When present in the terrestrial environment, Cd mobility in soils and sediment is highly influenced by the presence of carbonates, clays, organic matter, hydrous oxides, and a series of physicochemical parameters including redox potential, pH and DO (Jha & Kumar, 2024; Nungula et al., 2024). Cadmium has an affinity for exchangeable, carbonate and organic matter fractions within soil, which contributes to its increased potential for liberation from soils into the

surrounding ecosystem (Ankush et al., 2024; Jha & Kumar, 2024; Nungula et al., 2024). Factors such as high pH (often associated with the presence of carbonates), aerobic soils, and the formation of Cd complexes including clay and inorganic ligands, all promote the formation of low-solubility complexes, reducing potential for mobility in the environment (Jha & Kumar, 2024; Nungula et al., 2024; Wang et al., 2006). In contrast, factors such as high proportions of soluble Cd complexes bound to organic matter, low pH, and anaerobic soils are all contributors to a higher potential for Cd mobility within the ecosystem (Nungula et al., 2024; Tandy et al., 2009).

Plants display a wide range of variability in tolerance and tendency to accumulate Cd within their tissues, largely related to physicochemical conditions in soils and plant physiology (Gamberg et al., 2005; Nungula et al., 2024; Verbruggen et al., 2013). Further, hydrological and climate dynamics paired with soil dynamics can directly influence Cd mobility in soils (Jha & Kumar, 2024; Nungula et al., 2024). Factors such as high rainfall paired with poor soil drainage and low pH can create a favourable environment for Cd leaching from soil, resulting in increased potential for mobility into surface and groundwater and having possible implications on water quality (Jha & Kumar, 2024; Nungula et al., 2024).

Aquatic Biogeochemistry

Cadmium primarily enters aquatic systems by sediment deposition, where a multitude of factors such as pH, DO, hardness and salinity and Cd concentration are all controls on speciation (Ankush et al., 2024; Jha & Kumar, 2024). In freshwater systems, Cd is present as soluble forms Cd^{2+} , $\text{Cd}(\text{OH})_2$, and CdCO_3 , with water chemistry strongly influencing complex formation (Ankush et al., 2024; Jha & Kumar, 2024; Mok et al., 2015). Alternatively, Cd can remain undissolved in water adsorbed to sediment particles (Ankush et al., 2024).

Implications for Ecosystem Health

Cadmium mobility within the food web is highly variable, as different species of vegetation have different physiology and tolerances to Cd (Gamberg et al., 2005; Nungula et al., 2024; Verbruggen et al., 2013). Some plant species with a high affinity for, and tolerance of, Cd are ‘hyperaccumulators’, however, Cd uptake by plants is also highly influenced by physicochemical conditions within soil and sediment (Nungula et al., 2024; Verbruggen et al., 2013). Wildlife who feed regularly on Cd-hyperaccumulating vegetation, such as caribou and moose consuming large

quantities of willow, often present with elevated renal Cd concentrations, posing risks for chronic health issues (Gamberg et al., 2005; Jensen et al., 2009; Nungula et al., 2024; Verbruggen et al., 2013).

1.2.3 Lead

Lead (Pb) is a heavy metal that can exist naturally in +2 and +4 oxidation states, behaving most similarly to tin (Cullen & McAlister, 2017). This non-essential heavy metal is acutely toxic to living organisms, with high levels of exposure associated with kidney damage and neurological issues in humans, and even low levels leading to anemia, reproductive health issues, and developmental issues in children (Abadin et al., 2020; Cullen & McAlister, 2017; Fillion et al., 2014; Stalwick et al., 2023). Lead is naturally occurring and can be mobilized into the environment through natural and anthropogenic processes (Cullen & McAlister, 2017; Stalwick et al., 2023). Natural sources of Pb include volcanic eruptions, geological weathering, radioactive decay, and liberation from vegetation and organic matter by forest fire (Cullen & McAlister, 2017). Despite natural contributions, anthropogenic sources of Pb dominate inputs, altering global biogeochemical cycling of Pb (Cullen & McAlister, 2017; Stalwick et al., 2023). These sources include mining, ore smelting, combustion of leaded fossil fuels, and the use and disposal of lead-containing products such as lead-acid batteries, paints, pipes, and ammunition (Abadin et al., 2020; Cullen & McAlister, 2017). Despite the phase-out of Pb in gasoline in the 1970s and regulations resulting in a reduction of Pb emissions, Pb continues to persist and mobilize within the natural environment (Cullen & McAlister, 2017). Given its potential for long-range transport, Pb can be detected in air, snow, rain and glacial ice cores around the world, with disproportionately high deposition recorded with increasing distance from mid-latitudes (Cullen & McAlister, 2017; Stalwick et al., 2023; Zhang et al., 2015).

Lead Exposure in the Arctic and Subarctic

Despite reduction in Pb emissions in recent decades, average blood levels of Pb in residents of northern communities are significantly higher compared to the rest of the Canadian population (Laird et al., 2013; Liberda et al., 2018; Stalwick et al., 2023). A multitude of Pb sources have been identified including the use of lead ammunition for the harvesting of country food, mining, and long-range transport, household sources including lead pipes, dust and paint, all of which disproportionately impacting Indigenous and northern communities reliant on older homes and

country food harvesting (Fillion et al., 2014; Laird et al., 2013; Liberda et al., 2018; Pérez-Rodríguez et al., 2018; Tsuji et al., 2008).

Long-range transport and proximity to mining sites have been shown to influence Pb concentrations in plants and animals (Stalwick et al., 2023). Vegetation uptake of Pb was higher near mining activities at a Pb-Zn mine in Faro, Yukon, with labrador tea identified as a promising indicator of environmental Pb concentrations (Pugh et al., 2002). Another study conducted an assessment of Pb concentrations in vegetation at both pristine and industrial sites across a large area in Canada's North, highlighting the potential in using Pb/La ratios to determine dominant Pb sources (Shotyk et al., 2020). In the Athabasca oilsands region in Alberta, particulate Pb has been found to be the dominant source of Pb in vegetation on the surface of berries (Stachiw et al., 2019). Assessment of Pb concentrations in animals in relation to mining sites have yielded variable results (Bordeleau et al., 2016; Quakenbush & Citta, 2009; Stalwick et al., 2023; TetraTech, 2009). For example, elevated concentrations were reported near a lead-zinc mine in Alaska with Pb concentrations in animal blood and liver samples at mine impacted sites reported as 20 times higher than those from reference sites (TetraTech, 2009). In contrast, Pb concentrations in snowshoe hare and bearded seals near a copper smelter and lead-zinc mines, respectively, were below detection limits (Bordeleau et al., 2016; Quakenbush & Citta, 2009). These results highlight a large degree of regional and circumstantial variability, emphasizing the importance of regional monitoring to identify Pb concentrations and sources.

Terrestrial Biogeochemistry

The presence, mobility, and fate of Pb speciation in soil is largely controlled by its composition and physicochemical properties including redox potential, pH, presence of inorganic and organic ligands, soil composition, and other ions in solution (Bradl, 2004; Cullen & McAlister, 2017; Degryse et al., 2009). Lead's general tendency to form progressively less soluble compounds through adsorption, complexation reactions, and co-precipitation is slowed by its relatively large ionic radius (Bradl, 2004; Cullen & McAlister, 2017; Degryse et al., 2009). The formation of PbCO_3 is favoured in carbonate rich soil under oxidizing and mildly acidic to alkaline soil conditions (Cullen & McAlister, 2017). When carbonates are lacking, Pb favours adsorption to metal oxides, with a relatively stronger affinity for Mn-oxides (Bradl, 2004; Cullen & McAlister, 2017; Degryse et al., 2009). Soils rich in organic matter can greatly influence the complexation of

Pb, largely controlled by the composition of inorganic and organic ligands, however with drastically different behavior in experimental and natural systems (Bradl, 2004; Cullen & McAlister, 2017). Across different soil compositions, increased temperature and pH are thought to increase Pb adsorption (Cullen & McAlister, 2017).

Aquatic Biogeochemistry

The fate and uptake of Pb in freshwater systems is directly related to chemical speciation (Cullen & McAlister, 2017). Fractions of particulate and dissolved Pb in freshwaters are attributed to local geology and anthropogenic inputs; however, physicochemical conditions and water chemistry directly impact its fate in the environment (Cullen & McAlister, 2017). Under typical freshwater conditions, circumneutral pH, and in contact with the atmosphere, Pb tends to form complexes with sulfate and hydroxy ions, with OH more abundant at high pH and sulfate more abundant at low pH (Cullen & McAlister, 2017). Lead favours complexation with organic matter in oxygenated freshwater; however, complexation dynamics largely depend on composition of organic and inorganic ligands, pH and other cations present in the system (Cullen & McAlister, 2017; Morel et al., 2025). Alternatively, suspended particles can serve as a site for Pb adsorption, governed by ionic strength, pH, ligand composition, and other cations present (Cullen & McAlister, 2017; Morel et al., 2025).

1.2.4 Mercury

Mercury (Hg) is a transition block d-metal that can assume three different oxidation states +1, +2, and 0 (Lehnherr, 2014). Its occurrence in the environment is attributed to both natural and anthropogenic processes; however, anthropogenic sources such as fossil fuel combustion, oil, coal, and gold mining activities largely dominate inputs (Hammerschmidt & Fitzgerald, 2006; Hudelson et al., 2020; Lehnherr, 2014). Stability of Hg (0) and its characteristic high vapour pressure allow for easy volatilization and long-range transport of Hg far from its source, allowing it to reach high-latitude and remote locations, far from its source (Hammerschmidt & Fitzgerald, 2006; Lehnherr, 2014). Following transport, Hg can be deposited and either sequestered in soils and plants, aquatic sediments, or reduced by biotic and photochemical pathways (Hammerschmidt & Fitzgerald, 2006; Hudelson et al., 2020; Lehnherr, 2014). Once introduced in the natural environment, Hg can undergo methylation, forming a series of methylated complexes (MeHg) with a high potential for bioaccumulation and magnification throughout the ecosystem (Bernhoft, 2012; Lehnherr, 2014).

Methylmercury is the form of Hg most toxic to humans and other animals due to its bioaccumulative properties in tissue and its ability to cross the blood-brain barrier, posing severe risk as a neurotoxin among other negative health issues (Bernhoft, 2012). In northern and mountainous regions holding glaciers and permafrost, melt has caused release of sequestered Hg, resulting in increasing fluxes of mercury in the surrounding environment (McDowell et al., 2023; St. Pierre et al., 2019).

Terrestrial Biogeochemistry

In forested ecosystems, 60-90% of vegetation uptake of mercury is through uptake of elemental gaseous Hg within the atmosphere (Zhou et al., 2021). Although plants like lichens and mosses are known to show higher rates of Hg uptake relative to vascular plants, large knowledge gaps still exist in our understanding of environmental and physiological controls of Hg uptake (Zhou et al., 2021). Hg concentrations found within soils are thought to be introduced most dominantly through deposition and decomposition of local tree litter, with Hg entering leaves through stomatal uptake (Gworek et al., 2020). While atmospheric deposition dominates in environments unimpacted by mining and other anthropogenic activities, point source contamination is the dominant source of Hg deposition in its surrounding environment (Armstrong et al., 2021; Cheney et al., 2024; Sivarajah et al., 2024). When deposited onto the terrestrial environment from the atmosphere, Hg is primarily in its most oxidized state, Hg 2⁺; however, redox sensitivity paired with environmental conditions including soil moisture, light exposure, temperature, matrix chemistry, and microbial community composition can result in transformations of its state and subsequent speciation (Gworek et al., 2020). Given the high potential of bioaccumulation and magnification within the food web, trophic transfer of Hg has been well-studied in northern ecosystems. A series of studies done on various species across the Yukon have generated valuable information regarding concentrations of mercury in both fish and terrestrial wildlife (Gamberg et al., 2005, 2005; Gamberg & Braune, 1999; Hinck et al., 2006; Schuster et al., 2011). Tissue, liver, and kidney in Yukon caribou, moose, and Arctic wolf were analyzed for Hg concentrations in multiple assessments (Gamberg et al., 2005, 2005; Gamberg & Braune, 1999; Gamberg & Gamberg, 2000). Generally, concentrations appeared to be below those that would cause toxicological concern, with some renal concentrations found to increase with age in arctic wolves and caribou (Gamberg et al., 2005; Gamberg & Braune, 1999). Caribou had higher concentrations than other species, attributed

to their typical consumption of lichen, known for its atmospheric uptake of Hg (Gamberg et al., 2005; Gamberg & Gamberg, 2000).

Aquatic Biogeochemistry

Hg primarily enters aquatic environments through snowmelt, glacial melt, precipitation, and atmospheric deposition, with anthropogenic activities dominating Hg inputs in environments adjacent to point-source contamination (Branfireun et al., 2020; Gworek et al., 2020; St Louis et al., 2005; St. Louis et al., 2007). Once Hg has been deposited into the environment, it can undergo a variety of transformations, greatly influencing its availability throughout aquatic systems (Branfireun et al., 2020; Lehnherr, 2014). Redox potential is a driving factor in controlling Hg speciation and mobility in the environment, and more specifically, controlling amounts of Hg available for methylation (Branfireun et al., 2020; Gworek et al., 2020; Lehnherr, 2014). Methylation is favoured in anoxic environments with moderate of DOM, where Hg efficiently converts to its 2+ oxidation state (Branfireun et al., 2020; Chiasson-Gould et al., 2014; Wildman, 2016; Xin et al., 2023). Benthic environments in wetlands are commonly anoxic, therefore in wetland-fed catchments, they are found to be dominant sources of MeHg to boreal forest ecosystems (St. Louis et al., 1994). MeHg formation in rivers is highly controlled by flow, with MeHg formation occurring heterogeneously in rivers, related to flow and the resulting oxygen dynamics and microbial community composition (Branfireun et al., 2020; Wildman, 2016; Xin et al., 2023). Once methylation occurs in these benthic environments, MeHg diffuses from sediments to water (Hammerschmidt & Fitzgerald, 2006; Hudelson et al., 2020). In low flow conditions, rivers play an important role in Hg loading into downstream systems, directly impacting Hg methylation dynamics (Wildman, 2016; Xin et al., 2023). With a warming climate leading to more variable precipitation patterns paired with anthropogenic activities, discharge and floodplain influence will undoubtedly play an increasingly important role in methylation controls in these freshwater systems (Hall et al., 2005; St-Aubin et al., 2025).

Long-term Hg Monitoring Records in Yukon

The proven efficiency of elemental mercury in enhancing gold recovery from mined ores resulted in its widespread use in the Klondike region for gold mining operations in the 20th century (Clackett et al., 2021). Given this context, long-term assessment of elemental Hg concentrations have been conducted to assess connections to the Klondike Gold Rush and other mining activities,

and response to regulations in more recent years (Clackett et al., 2021; MacSween et al., 2022). Changes in atmospheric mercury in the region have been assessed through analysis of tree cores, sediment cores, and a long-term air quality monitoring program including a site in Little Fox Lake, southern Yukon (Cheney et al., 2024; Clackett et al., 2021; MacSween et al., 2022). Tree rings were analyzed, producing a 151-year record near the Bear Creek mining camp, with results showing a rapid increase in Hg (0) from 1923 to 1930 (Clackett et al., 2021). Results showed a correlation between Hg concentrations and operational dates of several mining camps in the area, followed by a decrease in concentrations from 1939 to 1966, coinciding with the closure of the mine camp (Clackett et al., 2021). A series of sediment cores collected surrounding Giant Mine, near Yellowknife, Northwest Territories, were collected, with cores collected within five kilometers of the mine proving to be affected by mine-related Hg emissions, also coinciding with dates of known mining activities (Cheney et al., 2024). Relative to other methods discussed, atmospheric Hg records are much smaller in temporal scale, with data collection starting in 1995, (MacSween et al., 2022). An assessment of atmospheric Hg trends across 10 sites from 1995-2018 found that Little Fox Lake was the only site displaying a positive trend (MacSween et al., 2022). Data from Little Fox Lake also shows the only significant positive annual trend across all seasons, with drivers contributing to this trend not fully understood; however, researchers suggest that air masses arriving from Asia, a place with increased Hg emissions, to this western subarctic monitoring station paired with increasing forest fire activities might contribute to observed trends (Fraser et al., 2018; Huang et al., 2017; MacSween et al., 2022).

1.3 Rivers on Watershed Scale

1.3.1 Discharge and Flux

Discharge

Water discharged from the landscape within an ecosystem is a fundamental control of river and stream ecosystems (Gore & Banning, 2017). Understanding changes and ranges of magnitude in flow provide valuable insight into sediment and organic matter transport, distribution of vegetation, and migration of wildlife within lotic ecosystems (Gore & Banning, 2017; Poff & Ward, 1989). Fluctuating discharge is significantly correlated with changing water chemistry, undoubtedly impacting nutrient dynamics in downstream ecosystems (Shogren et al., 2021). Discharge is the

volume of water passing through a cross-section of known area in a river channel (Gore & Banning, 2017).

$$Discharge \left(\frac{m^3}{s} \right) = Area \text{ of water in cross section } (m^2) \times Flow \text{ velocity } \left(\frac{m}{s} \right)$$

Flux

Local geology and anthropogenic activities can greatly influence the quantity and species of metals and nutrients released into lotic ecosystems (Hall et al., 2005; Lafrenière et al., 2025; St-Aubin et al., 2025). Seasonal and groundwater dynamic changes influence water chemistry, resulting in dynamic metal, nutrient and physicochemical parameters and concentrations in each system (Gore & Banning, 2017; Lafrenière et al., 2025; Poff & Ward, 1989). The collection of continuous discharge measurements paired with sporadic water chemistry measurements present an opportunity to calculate riverine metal concentration fluxes used to assess associations between discharge and pollutant transport to downstream ecosystems (Gore & Banning, 2017).

Fluxes are calculated as a mass of a given chemical constituent transported in a riverine system over time, calculated using the following equation (Gore & Banning, 2017; USGS, 2007):

$$Mass \text{ Flux } \left(\frac{kg}{d} \right) = Discharge \left(\frac{m^3}{s} \right) \times Concentration \left(\frac{mg}{L} \right) \times 86.4$$

1.3.2 Integrators of Signals Throughout the Watershed

Fluvial systems are key players in transport and continuity within an ecosystem and serve as the cornerstone to many cultural traditions and ecosystem services (Carone et al., 2009; Moore, 2015). Rivers serve as integrators of various ecosystems throughout a watershed, and their processes contribute significantly to the molding of landscapes and ecosystem composition (Moore, 2015). The role a river plays in connecting ecosystems within a watershed has been long understood by Indigenous peoples, who hold a deep connection and awareness of the land and its changing dynamics (Kimmerer, 2013; McDowell et al., 2023). The River Continuum Concept highlights the gradient of changing physical conditions and ecosystem composition along a river system to be a dynamic gradient of biotic change, sediment loading, transport, and storage along a river (Vannote et al., 1980). This concept emphasizes that rivers are highly dynamic ecosystems that are in constant response to the surrounding environment, including seasonal cycles, ecosystem dynamics,

and ever-changing tributary flow regimes (Vannote et al., 1980). This understanding of interconnectedness was applied in the creation of the *Integrated Catchment Management* (ICM) approach, used to align efforts across all stakeholders to protect land, water, and biodiversity and the *Ecosystem Approach*, focused on maintaining biodiversity and finding equitable management solutions (Stewardson et al., 2017). These principles have been applied at a watershed level in mountainous and boreal forest landscapes in Canada to assess point-source impact on contaminant mobility within watersheds impacted by forest fires (Emmerton et al., 2020), mining (Cooke et al., 2025), agriculture (Emmerton et al., 2023), and broader ecosystem stressors such as climate change (Emmerton et al., 2019). In an assessment of contaminant mobility downstream of supposed reclaimed mountain-top coal mines, Cooke et al. (2025) found that downstream selenium concentrations surpassed safe levels, highlighting needs for further remediation. Additionally, Emmerton et al. (2020) showed that rivers in burn-affected regions had much higher concentrations of metals, nutrients and suspended sediments at a large-watershed scale relative to unburned catchments following the 2016 Fort McMurray wildfires.

1.3.3 Hydrological Controls of Rivers in Canada's Subarctic

Rivers in Canada's subarctic cross a breadth of different landscapes, ranging from mountainous to barren tundra, which both influence and are influenced by different climatic patterns, resulting in variable accumulation of snow, melt regimes, ice cover, and permafrost dynamics (Brabets & Walvoord, 2009; Chesnokova et al., 2020). In subarctic mountainous watersheds, mountains serve as important water sources for both high elevation and downstream lowland regions (Immerzeel et al., 2020; McDowell et al., 2023; Woo & Thorne, 2006). In recent years, these characteristic features of subarctic watersheds are undergoing rapid change, with alpine watersheds proving to be particularly sensitive to warming (Immerzeel et al., 2020; McDowell et al., 2023; Woo & Thorne, 2006).

Rivers in Southwestern Yukon

Rivers in southwestern Yukon, a mountainous region, were studied for their sensitivity to changes in temperature and precipitation patterns, and glacial retreat, resulting from changes in climate (Chesnokova et al., 2020; Rasouli et al., 2014; Woo & Thorne, 2006). Across the Yukon River Basin, warmer temperatures and thawing permafrost have contributed to increased winter streamflow (Brabets & Walvoord, 2009). In the headwaters region located in southwestern Yukon,

many rivers carry strong glacial influence; however, a wide range of glacial influence in the region has resulted in characteristically different hydrological regimes (Chesnokova et al., 2020). Minimally glacierized watersheds were characterized by peak discharge in June, corresponding to freshet at low elevations (Chesnokova et al., 2020). These systems are also more sensitive to warming temperatures, causing changes in freshet timing and ice cover, impacting peak water flow as a result (Stewart et al., 2005).

1.4 Holistic approaches to Natural Sciences Research

1.4.1 Indigenous Knowledge Systems and Western Science

Anthropogenic activities including resource extraction and development have become a dominant force on earth (Henri et al., 2021; Vitousek et al., 1997), causing undeniable changes to natural processes and contributing to biodiversity loss. As a result, many researchers have identified that although Western Science (WS) has been effective at studying simpler systems and specific questions, it holds great limitations in its ability to tackle broader questions related to environmental degradation of complex ecosystems (Alexander et al., 2021; Henri et al., 2021; Sharma et al., 2024; Wilcox et al., 2023). Despite these longstanding challenges, there has only recently been a push for inclusion of Indigenous knowledge systems (IKS) into conversations surrounding environmental stewardship and sustainability (Berkes, 2017; Henri et al., 2021; Mazzocchi, 2020; Sharma et al., 2024; Tengö et al., 2014; Wong et al., 2020).

There is no one ‘accepted’ definition of IKS given the broad range of teachings and knowledge held by Indigenous peoples worldwide (Berkes, 2017). Although this heterogeneity can pose challenges in assigning a formal definition, IKS can be generally understood as a body of knowledge encompassing knowledge, practice and belief (Berkes, 2017). IKS is understood to be much more than simply a way of conducting science but is a way of life (Berkes, 2017; Kimmerer, 2013; Sharma et al., 2024). These teachings are passed along generations rooted in teachings from the land and spiritual health of all beings, all offering sustainable insights into harmonious living with the land (Berkes, 2017; Kimmerer, 2013; Sharma et al., 2024). Also sometimes referred to as traditional ecological knowledge (TEK), Indigenous scholars Battiste and Henderson argue “what is traditional about traditional ecological knowledge is not its antiquity, but the way it is acquired and used” (Battiste & Henderson, 2000). This perspective suggests that knowledge within IKS and

TEK systems is shaped based on lived experiences, taking a more process-based approach (Battiste & Henderson, 2000; Berkes, 2017).

In contrast, western scientific knowledge systems are rooted in Eurocentric perspectives, typically prioritizing quantitative data and linear thinking (Datta & Starlight, 2024). These perspectives typically result in scientists favouring the compartmentalization of problems into specific research questions over a broader holistic assessment (Datta & Starlight, 2024; Henri et al., 2021). Although aspects of knowledge from other countries and cultures have been accepted into this knowledge system, these findings are often modified to fit within Eurocentric perspectives (Aikenhead & Ogawa, 2007).

Despite their inherent differences, many scientists, scholars, and rights holders believe that these knowledge systems can work complementary to one another to enhance our understanding of natural systems (Henri et al., 2021; Kimmerer, 2013; Tengö et al., 2014; Wright et al., 2019). Despite this potential, these knowledge systems can only work in symbiosis if bridged meaningfully through authentic conversations, and with ongoing recognition of colonial structures at play (Kimmerer, 2013; Tengö et al., 2014; Wright et al., 2019). If done correctly, the inclusion of strengths from both IKS and WS will generate more meaningful, accurate findings and, more broadly, solutions to complex issues around climate change, biodiversity loss, and other questions of importance that honor values of all rights holders (Henri et al., 2021; Kimmerer, 2013; Wright et al., 2019). There are a number of frameworks that encourage the complementary use of IKS and WS, including Two-Eyed Seeing (*Etuaptmumk*), and Two Row Wampum (*Kaswentha*) (Abu et al., 2020; Reid et al., 2021; Reid, 2020).

Barriers to Implementation

For much of Canadian history, First Nation, Inuit and Métis people have faced a series of injustices, including their forced assimilation into settler culture through the establishment of residential schools and a series of racist, colonial policies (Hanson, 2025; McGregor, 2018). Despite recent efforts made by all levels of government to understand the extent of these injustices and begin walking down a path towards reconciliation, many barriers are ever-present in policies and Canadian culture, including research practices (Johnson et al., 2023; Martin et al., 2017; McGregor, 2018; Wong et al., 2020).

In many cases, WS-dominated research with direct impacts on Indigenous communities has not always been respectful, with much of the research characterized as ‘on’ rather than ‘with’ Indigenous peoples, and many communities not getting anything in return (Johnson et al., 2023; Reid, 2020). Given their exclusion from much of the research, Indigenous peoples and their knowledge are underrepresented in the scientific landscape (Council of Canadian Academies (CCA), 2023; McDowell et al., 2023; McGregor, 2018; Wilson et al., 2020). Disciplinary divides and structural challenges associated with conducting interdisciplinary research has also served as a large barrier for Indigenous participation in research, further emphasizing the presence of colonial structures in academia and funding agencies (Council of Canadian Academies (CCA), 2023). Additionally, colonial structures present a series of accessibility barriers including language, limited community capacity, and educational barriers largely resulting from widespread impact of residential schools (Council of Canadian Academies (CCA), 2023; Wilson et al., 2020; Wilson et al., 2024; Wong et al., 2020).

Barriers for Indigenous involvement and leadership in research within Canada cannot be ignored by researchers (Council of Canadian Academies (CCA), 2023; Wong et al., 2020). Thoughtful reflection on the perpetuation of colonial structures within research must be at the forefront of planning at all stages while also considering the vast variety of barriers faced by Indigenous groups across Canada, focused within a place-based lens to account for local context (Council of Canadian Academies (CCA), 2023; Wilson et al., 2024; Wong et al., 2020). In response to the growing demand for decolonization of natural science research in Canada, Wong et al. (2020) published calls to action for Canadian researchers, designed to promote meaningful inclusion of First Nation, Inuit and Métis voices in natural science (Wong et al., 2020). While continued awareness among researchers is imperative in removing barriers associated with Indigenous involvement in research, academics have called for meaningful implementation of IKS at a structural level to enact meaningful change (Council of Canadian Academies (CCA), 2023; Inuit Tapiriit Kanatami, 2018; Wilson et al., 2024; Wong et al., 2020).

1.4.2 Decolonizing Research Practices in the Yukon

Historically, research conducted across the North has been directed by the needs, priorities, voices, and questions of southerners (Council of Canadian Academies (CCA), 2023; Johnson et al., 2023; Wilson et al., 2020). This disconnect greatly limits the impact and effectiveness of research

findings in the implementation of Arctic governance and decision making (Council of Canadian Academies (CCA), 2023; Wilson et al., 2024). With a growing recognition that northern research disproportionately benefits non-Indigenous researchers, substantial efforts have been made by both academic institutions and Indigenous groups to work towards decolonizing research practices in the north (Wilson et al., 2020). The diversity of landscapes, governance structures and priorities across the North has resulted in a vast variety of procedures and guidelines put in place for community engagement in research (Perrin et al., 2021; Wilson, 2019; Wilson et al., 2024). In the Yukon, unique self-government agreements in their early stages of implementation have resulted in little practical guidance being provided to non-Indigenous on how they can meaningfully alter their practices to decolonize research (Gaudry & Lorenz, 2018; Morton Ninomiya & Pollock, 2017; Wilson et al., 2020; Wilson et al., 2024). Community members and academic partners continually emphasize variability in priorities and capacity by community, and that simply consulting prior to conducting research is not enough to meaningfully decolonize northern research, nor a one-size-fits-all approach (Wilson et al., 2020; Wilson et al., 2024; Wong et al., 2020). Further, no widespread obligations in place for engagement at all stages including result sharing, can limit knowledge mobilization on a local scale (Perrin et al., 2021). These circumstances have resulted in some academics to call for changes in federal regulations to facilitate more meaningful decolonization of northern research through improved permitting structures offered to southern researchers by the territorial government by providing guidance on how to meaningfully address regional policy priorities (Perrin et al., 2021; Wilson et al., 2024; Wong et al., 2020). Regardless of barriers in place, there are examples of meaningful community engagement in research within the Yukon related to assessing contaminants in wildlife (Gamberg, 1994; Gamberg & Braune, 1999; Gamberg & Gamberg, 2000; Schuster et al., 2011). Significant advances in decolonizing research practices have been made in ecological studies relative to the geosciences (Bretzlaff, 2022; Wilson, 2019, 2020; Wilson et al., 2024).

1.5 Yukon River Watershed

The Yukon River extends over 3,000 km from northern British Columbia to the Bering Sea (Arctic Ocean), serving as the third longest river in North America and the longest inland Pacific salmon run (Yukon River Inter-Tribal Watershed Council, 2025). The watershed (850,000 km²) is the fourth largest in North America by area, holding approximately 126,000 residents (2000) including

First Nations in BC and Yukon, and tribes throughout Alaska (Brabets et al., 2000; Yukon River Inter-Tribal Watershed Council, 2025). Throughout history, the Yukon River has connected communities that are otherwise inaccessible by road, serving as a means for connection among communities who have stewarded these waters and surrounding lands since time immemorial (Yukon River Inter-Tribal Watershed Council, 2025). Eventually, this river also became a route for trade and eventually used to access sites mined during the Klondike Gold Rush (C/TFN Government, 2014). The river and its headwaters have historically served as a critical salmon spawning habitat, which is currently inactive due to blockage by various anthropogenic activities including hydroelectric infrastructure. Today, many communities along the river hold concerns over water quality due to both current and legacy contamination in both Canada and the United States.

1.5.1 Current Governance Landscape

The transboundary nature of this watershed presents both significant challenges and opportunities related to water governance and land planning throughout the watershed. Currently, government led monitoring efforts are coordinated nationally, with the United States Geological Survey (USGS) collecting water quality and permafrost monitoring data within Alaska, and Environment and Climate Change Canada (ECCC) and YG carrying out water quality and monitoring efforts within Canada (IJC, 2026b; USGS, 2013; Yukon River Inter-Tribal Watershed Council, 2025). Recognizing that water knows no borders, the International Joint Commission (IJC) was created by both Canada and the United States as a framework for co-governance of transboundary watersheds between federal governments (IJC, 2026b). Guided by the *Boundary Waters Treaty* (1909), this commission is responsible for resolving disputes affecting water flow and levels across national borders and recommending solutions to conflicts including water quality, hydroelectric power generation, fishing, health, and industry (IJC, 2026b). Although this commission fills an important role around conflict resolution, there are no large-scale government-led efforts surrounding co-management and governance of this watershed.

Indigenous-Led Governance

Many of the Yukon First Nations (YFN) and Alaskan Tribes within the drainage basin are committed to the protection of the watershed, recognizing the need to maintain watershed health for maintenance of cultural practices, food security, and traditional way of life. Established in 1997,

the Yukon River Inter-Tribal Watershed Council (YRITWC) is an Indigenous-led grassroots organization dedicated to the protection and preservation of the Yukon River watershed through coordinating collective efforts including community-led water quality monitoring efforts among YFN and Alaskan Tribes (Yukon River Inter-Tribal Watershed Council, 2025). Within the US and Canada, YRITWC has successfully integrated the Indigenous Observation Network program, facilitating water sample collection that has been included in reports published by the USGS (USGS, 2013; Yukon River Inter-Tribal Watershed Council, 2025). The YRITWC fills an important role surrounding collaborative governance led by local First Nations and Tribes. However, differing government priorities, frameworks, and processes relating to environmental protection, resource development and Indigenous/ Tribal collaboration remains as a large barrier to effective co-governance.

1.5.2 Yukon River Headwaters

The Southern Lakes region of southern Yukon and northern BC are considered the headwaters of the Yukon River. Situated within the Coast Mountain range, this expansive region is home to a large diversity of wildlife and vegetation, mountainous landscapes, and a series of large deep lakes (Smith et al., 2004). Atlin, Tagish, and Bennett Lakes are the largest lakes in the region, fed by a series of tributaries, all with chemistry highly influenced by water sources, surrounding landscapes, and development (MacNeil et al., 2025). Whereas Atlin and Tagish Lakes receive major glacier meltwater contributions from the Juneau Ice Field (Smith et al., 2004), the sub watersheds feeding Bennett Lake contain only a small residual glacier area (ranging from 0 - 9.4 %) and it is much more strongly influenced by precipitation (snow and rain) in the high alpine zones (MacNeil et al., 2025).

The Southern Lakes region is located within the Traditional Territories of the Carcross/Tagish First Nation (C/TFN), Kwanlin Dün First Nation (KDFN), Taku River Tlingit First Nation (TRTFN), Ta'an Kwäch'än Council (TKC), and Teslin Tlingit Council (TTC), who have stewarded these lands and waters since time immemorial. Currently, both individual and coordinated efforts are in place for monitoring of water quality and permafrost, largely coordinated by YRITWC, and land planning initiatives (How we Walk with Land and Water, 2025; James & Wilson, 2024; Yukon River Inter-Tribal Watershed Council, 2025). How We Walk with the Land and Water (HWW) is an Indigenous-led coordinated monitoring effort between C/TFN, KDFN,

and TKC focused on working together on land planning strategies related to modern treaty agreements (How we Walk with Land and Water, 2025). Although it is widely recognized that the concept of land use and planning is highly colonial, these nations are working together to approach this concept in a holistic fashion, seeking solutions informed by both WS and IKS (How we Walk with Land and Water, 2025).

1.6 Carcross/Tagish First Nation

C/TFN Traditional Territory covers an expanse of glaciers, lakes, rivers, and small ‘desert’ holding glacial sediments (C/TFN Government, 2014). Although C/TFN considers their land to be without borders, what are commonly known as Yukon, British Columbia and Alaska now claim portions of their lands today (C/TFN Government, 2014). From nomadic and subsistence lifestyles, to trading, trapping, and traversing the Chilkoot Pass, the people of C/TFN have always placed immense value on their relationship to the land and its resources (C/TFN Government, 2014). The C/TFN Statutes recognize and emphasize their responsibility to protect the lands in a sustainable manner, stating that co-management of the land and resources is a traditional practice (C/TFN Government, 2014; James & Wilson, 2024).

1.6.1 Governance Landscape

While governed by the *British North America Act* (BNAA) and *Indian Act*, C/TFN citizens navigated a series of tensions and colonial influence in the region including the Klondike Gold Rush, various infrastructural developments including mining, railway, forestry, hydroelectric power, and recreation infrastructure, all while governed by policies limiting assertion of Indigenous self-determination (Hanson, 2025; Wilson, 2019, 2020).

Following 20 years of negotiations between the Council of Yukon First Nations, Yukon Territorial Government, and Government of Canada, all parties agreed to the *Umbrella Final Agreement* (UFA) framework (Bretzlaff, 2022; Wilson, 2020). The UFA provided a framework for each YFN to negotiate terms for their respective final agreements for self-governance (C/TFN Government et al., 2005; Wilson, 2020). In 2005, the C/TFN UFA was signed, acknowledging C/TFN’s asserted rights, titles and interests, agreeing to self-governance of their settlement of lands and waters (Bretzlaff, 2022; Wilson, 2020). Of the 1500 squared kilometers of C/TFN’s settlement lands, approximately two thirds are ‘Category A’, granting C/TFN surface and subsurface rights

to these lands and waters; and the remaining third is ‘Category B’ lands, granting only surface rights (Carcross/ Tagish First Nation, 2005). These different categories grant C/TFN different rights to govern lands and waters on their settlement lands; however much of their Traditional Territory and mine-impacted sites are not included in this allotment, presenting complexities for environmental stewardship (Figure 3) (Carcross/ Tagish First Nation, 2005; C/TFN Government, 2014; Wilson, 2020).

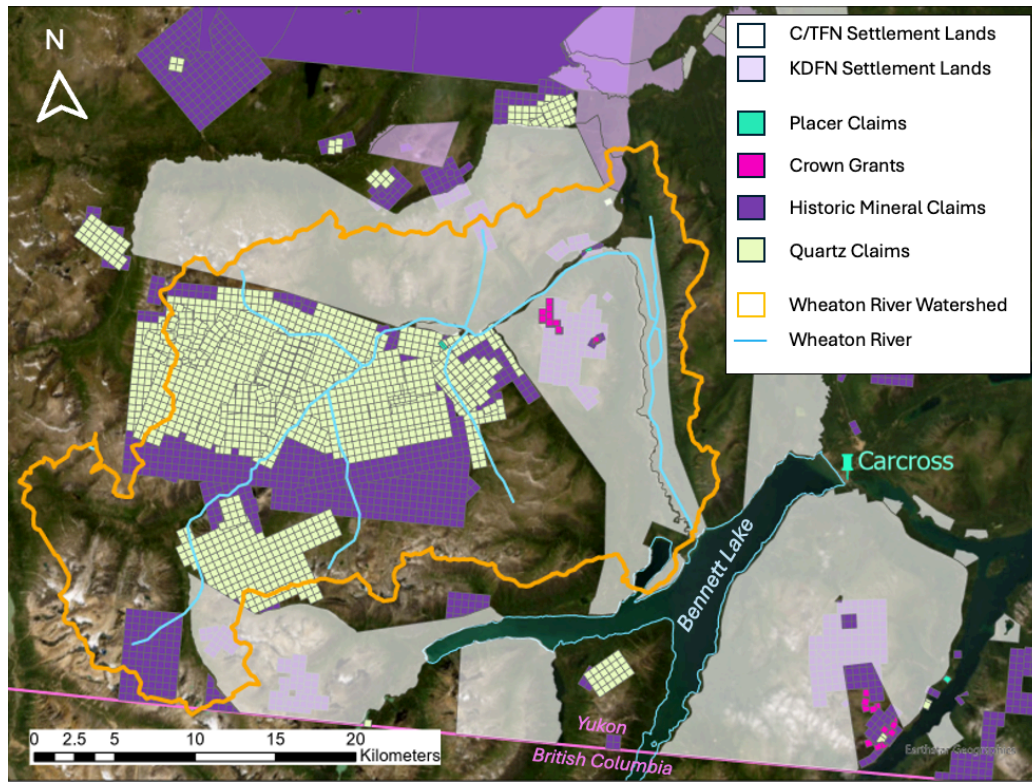


Figure 3. Mining claims and grants mapped alongside First Nation Settlement Lands within and surrounding the Wheaton River Watershed (Messenger et al., 2016; Yukon Government, 2021).

Water Governance

Chapter 14 of the UFA formally recognizes C/TFN’s rights to manage water flowing through, adjacent to, or on their settlement lands. Through this agreement, C/TFN can create laws to better assert their rights outlined in Chapter 14 which better positions them to protect waters (C/TFN Government et al., 2005; Wilson, 2019). Further, this agreement provides all signatory YFN to create legislation that supersedes colonial water legislation (Wilson, 2020). Although this agreement is progressive relative to others of its kind by installing safeguards to ensure C/TFN involvement in decisions pertaining to waters flowing through, on, or adjacent to their settlement

lands, it does not give them equal authority throughout the process (James & Wilson, 2024; Wilson, 2019). In the UFA, settler water laws are referred to as ‘general laws of application’ which apply to all lands regardless of settlement claim, and have not been updated since the implementation of the UFA (C/TFN Government et al., 2005; Wilson, 2020). More broadly, some C/TFN citizens have criticized the agreements for failure to respect water, centering legislation in a colonial worldview (James & Wilson, 2024; Wilson, 2020). In 2025, the C/TFN water committee released the *Heén/Tū (water) Strategy and Action Plan*, which includes plans to monitor, manage, and protect water using methods rooted in traditional values, supported by WS (James & Wilson, 2024).

Land Planning and Resource Management

Chapter 11 of the UFA outlines land planning initiatives, which details the co-management strategy of settlement lands between YFN and the Yukon Government, including the creation of the Land Use Planning Council, who holds authority over non-municipal lands in the Yukon (Staples et al., 2013; Wilson, 2019). Although created with the intention of creating a sustainable and consistent co-management strategy throughout the Yukon, shortcomings of this co-management strategy became evident during management of the Peel River Watershed, situated within the territory of the First Nation of Na-Cho Nyak Dun, Tr’ondëk Hwëch’in and Gwich’in Tribal Council (Staples et al., 2013; Wilson, 2020). This case showed that although this legislation facilitates co-management in theory, lack of collaboration by the Government and their clear desire to lead the land management process by deciding allocations of protected land showed that collaboration must be enforced in the spirit of reconciliation rather than simply just enforcing legislation (Staples et al., 2013; Wilson, 2020).

In Chapter 18, the agreement addresses the responsibilities of YFN and government in the operation and restoration of a quarry (C/TFN Government et al., 2005). Although this agreement makes progress in formally recognizing the requirement of YFN approval and consent to be received by the Yukon Government for any new quarries or mines, the process for remediation of legacy sites remains unclear (C/TFN Government et al., 2005; Wilson, 2019, 2020). The agreement states that the Yukon Government has an obligation to restore legacy quarries and mines to ‘accepted land use standards’ in a ‘reasonable time frame’ (C/TFN Government et al., 2005). Although binding, standards outlined in the agreement are highly vague and unclear. Further, this agreement further perpetuates First Nation dependence on government processes and timelines,

further ingraining colonial ideas into this co-management agreement (C/TFN Government et al., 2005; Wilson, 2020).

1.6.2 C/TFN Research Priorities

After several discussions including C/TFN staff and citizens, community members, and academic partners, a series of research questions were synthesized to best align with community concerns and priorities. Some of which, including those listed below, are considered research priorities for this region:

What contaminants are released into waterways from what sources, and what is their fate?

How does land use affect water? How do landscapes affect water?

Based on conversations with community and emerging questions, little is known in the area about the effects of point source contamination on both water quality and broader impacts on ecosystem health. Further, community members and representatives have expressed a strong interest in being involved in the research process, starting with the establishment of research questions, and continuing with methodologies, sample collection, and the interpretation and validation of research findings. Given a mutual desire for collaboration on this research, we have worked closely with C/TFN staff and community members towards answering these questions. All research partners are working together with the eventual goal of using research findings to inform reclamation efforts, habitat assessments, monitoring, land planning, and management plans for areas of interest for C/TFN. Further, it is a priority of the research team to conduct research supporting the implementation of C/TFN's *Héen/Tū (water) Strategy and Action Plan* (James & Wilson, 2024). The research conducted was designed by all parties to facilitate capacity sharing, co-learning, and co-production throughout all stages of the project.

1.6.3 Contaminated Sites Within the Bennett Lake Watershed

C/TFN Traditional Territory has been impacted by both natural and point source contaminants throughout various environmental media, posing a risk to drinking water quality, country food security, and broader ecosystem health (Figure 4). Until 1990, the community of Carcross sourced drinking water from groundwater wells; however, naturally elevated arsenic concentrations motivated a change in supply to Bennett Lake surface waters (C/TFN Government, 2014; EDI &

YUCAN Environmental Planning, 2010). Although historical data is limited, current available data shows that Bennett Lake surface waters fall within water quality guidelines (Baker, 1979; EDI & YUCAN Environmental Planning, 2010; Oliver, 2023). Within the literature, the last complete assessment of water quality of Bennett Lake was published in 1979, where water quality was assessed for mercury and other basic chemical and physicochemical parameters (Baker, 1979). Findings revealed that turbidity was the only parameter that fell outside of accepted limits at the time, though was also highly variable, often falling within accepted range (Baker, 1979).

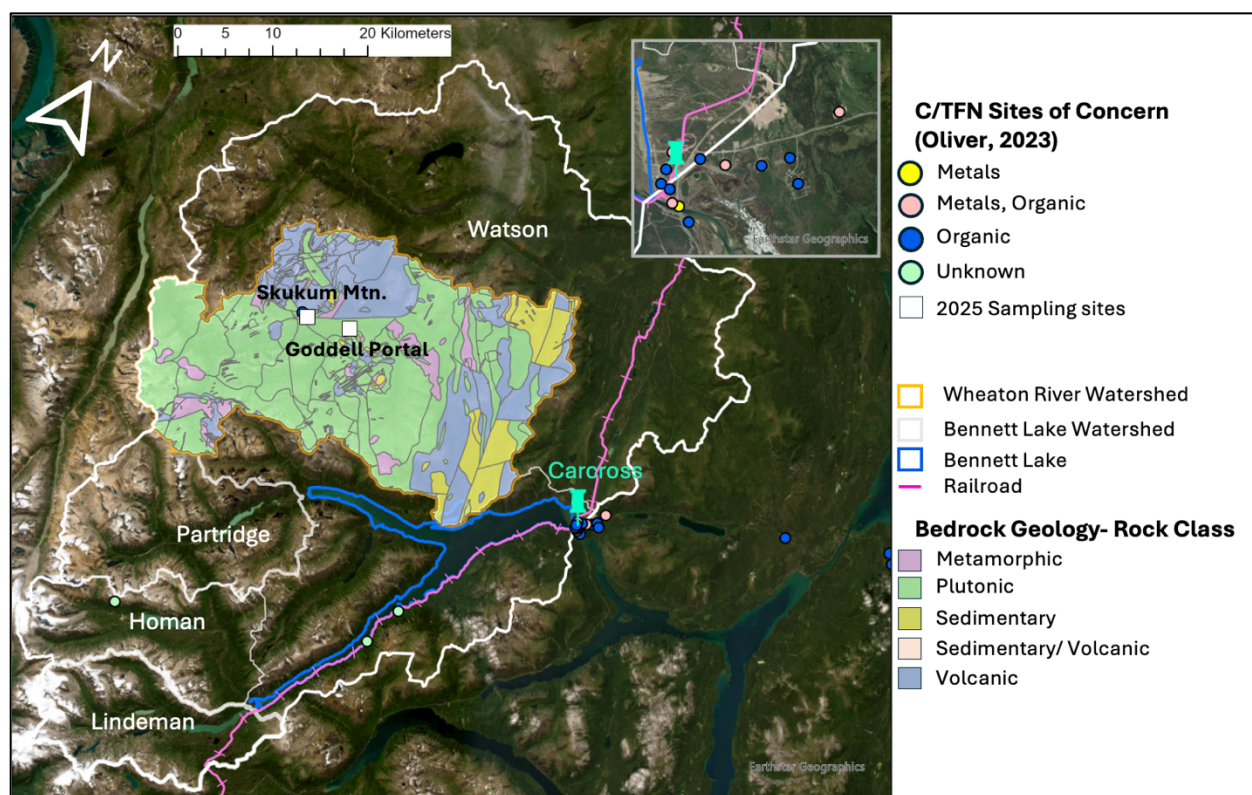


Figure 4. Map of sites of C/TFN sites of concern within and surrounding the Bennett Lake Watershed, plotted by contaminant group of primary concern (MacNeil et al., 2025; Messenger et al., 2016; Oliver, 2023). Wheaton basin underlying geology (rock class) is shown (Chikanda et al., 2023).

Within this region, mineral deposit mining is common to source specific metals such as antimony, cadmium, cobalt, copper, gold, mercury, silver, titanium and zinc (EDI & YUCAN Environmental Planning, 2010; Oliver, 2023). More presently, mineral placer mining is a growing area of economic activity in the region, which is expected to increase in demand in the coming years (van Loon, 2024). Much of this mining is concentrated within the Wheaton River Watershed; however, multiple mines have also operated on Montana Mountain, a sacred place for C/TFN citizens

located across Bennett Lake opposite Carcross (C/TFN Government, 2014). Located in both Bennett and Tagish Lake Watersheds, Montana Mountain mining operations conducted by both Venus and Arctic Gold and Silver (AGS), have sparked concerns among locals surrounding water quality and broader environmental impacts in the region (T. Obediah, Pers.Comms.). Tailings from the AGS mine drainage (1964-1969) flow into Bennett Lake through Tank Creek. Consultant and monitoring reports indicate elevated concentrations of various metals, specifically arsenic throughout environmental media near the tailings pond; however, do not report elevated concentrations within Tank Creek at the Bennett Lake inflow (INAC, 2010; Roach & Cunningham, 2000). Recently, the federal government has agreed to support a community-led remediation of both Venus and AGS mines and point-source contamination residing at the former site of Choutla Residential School, all within C/TFN Traditional Territory (Calissi, 2025).

1.7 Wheaton River Watershed

The catchment of the Wheaton River and its tributaries feed Bennett Lake in the Yukon River headwater region (Figure 4). Once nearly entirely covered by the Cordilleran Ice Sheet during the Late Wisconsinan McConnell Glaciation, the U-shaped valleys where the Wheaton and its tributaries flow are evidence of significant glacial coverage in the region (Church & Clague, 2008; Painter et al., 2023). During a relatively rapid Holocene deglaciation, the watershed filled with proglacial lakes, eventually thought to have drained into what is now known as the Wheaton River (Church & Clague, 2008; Painter et al., 2023). Today, the Wheaton basin holds two small alpine glaciers, the Wheaton and Radelet, both covering an area of less than 1 km² (Painter et al., 2023). Despite their close proximity to one another, these two glaciers have different melt dynamics, with the Wheaton Glacier retreating the most in the recent decade, and the Radelet Glacier retreating most between 2001 and 2011 (Painter et al., 2023). This minimal glacial coverage in the region amounts to less than 1% of the catchment; therefore, the Wheaton River is primarily fed by snowmelt, precipitation, and groundwater influences (MacNeil et al., 2025).

1.7.1 Geological Setting

The Wheaton River drainage basin is within Stikinia Terraine (Smith et al., 2004). This region is characterized by volcanic complexes near Skukum Mountain containing andesite, dacite flows and breccia (Hart & Radloff, 1990; Smith et al., 2004) (Figure 4). Volcanic activity has resulted in a strong presence of volcanic intrusions throughout the region (Hart & Radloff, 1990) (Figure 4).

Nearby these complexes, reports describe significant mineralization of metallic minerals such as gold, silver, and antimony within emerging veins, primarily composed of quartz (Hart & Radloff, 1990).

Tectonic activities within this area have resulted in the formation of faults and a shear zone trending northwest within the Wheaton River valley (Hart & Radloff, 1990; Smith et al., 2004). Exploration reports detail the emergence of Jurassic Laberge and Lewes River groups, primarily composed of sandstone, conglomerate, augite porphyritic basalt, and limestone, as a result of steep faulting (Smith et al., 2004). The Tally Ho shear zone within the river valley has also been described to contain significant mineralization (Hart & Radloff, 1990).

1.7.2 Mining

The abundance of mineralization of high-value minerals, and mining interests in the region following the Klondike gold rush resulted in mineral exploration in the Wheaton River valley starting in the early 1900s (Hart & Radloff, 1990). Over time, a variety of mining activities occurred within the watershed, with many concentrated around the Skukum Mountain volcanic complex (Smith et al., 2004). Given rich mineral resources, both large scale and hobby placer mining operations occur within the region (van Loon, 2024).

Skukum Mountain

The Skukum Mountain deposit is an area rich in gold mineralization, largely within quartz carbonate veins (MacDonald & Godwin, 1986; Yukon Government, 2025b). Further, the Mount Skukum Volcanic Complex holds a series of felsic rocks and andesitic flows holding veins with widely distributed gold and silver mineralization (Yukon Government, 2025b). Exploration and extraction was done by a series of companies, primarily interested in gold and silver, with the site originally staked in 1922 (Yukon Government, 2025b). Various veins within the region were drilled and assessed for geochemical properties. Even with inevitable variation between veins, reports estimate high concentrations of gold, silver, arsenic, lead, and sulfides (Yukon Government, 2025b). Other drilling operations to assess the presence of diamonds and more metals were done at the site in the early 2000s (Yukon Government, 2025b).

Nearby the access to the mine site at Skukum mountain is the convergence of two waterways, Skukum and Berney Creeks. The Skukum Creek Watershed is known to have hosted previous

mining operations; however, Berney Creek Watershed has not (T. Obediah, Pers. Comms.). Following extreme precipitation events in summer 2024, both Skukum and Berney Creek Watersheds were heavily impacted by landslides, undoubtedly altering water chemistry through altering flow paths, loading them with debris, and mobilizing mining infrastructure within Skukum Creek Watershed. This series of landslides has also rendered the Skukum Mountain Mine's 1000 m portal, a main portal for mineral exploration, inaccessible by road. The extent of changes in water chemistry resulting from mining activities and landslide events on nearby creeks, ecosystems, and downstream waterways are currently unknown.

Goddell Portal

Located southeast of Skukum Mountain, the Goddell shear zone first underwent mineral exploration in the early 1900s (Yukon Government, 2025a). The area is characterized by veins of quartz-stibnite, quartz-hematite, and gold-pyrite-arsenopyrite, and the Goddell shear zone containing gold-bearing pyrite and arsenopyrite (Yukon Government, 2025a). Following exploration confirming presence of gold, silver, and antimony- containing minerals, several companies have mined the area from the 1960s until recently (Yukon Government, 2025a). Despite several efforts to characterize the extent of mineralization within the Goddell shear zone, the Yukon Geological Survey believes that the limits of the deposit are not yet defined (Yukon Government, 2025a).

In 2024, an atmospheric river event triggered landslide activity around the Goddell Portal mining site, impeding road and portal access, and disturbing the tailings pond adjacent to the portal entrance and generating new streams. The landslide undoubtedly altered hydrological regimes within the region and facilitated transport of materials from the mine site and tailings pond within the watershed. This transport of contaminants and resulting implications for downstream water quality of the Wheaton River and Bennett Lake are currently unknown.

1.7.3 Land Management

Easily accessed by car via Annie Lake Road, the Wheaton River Watershed is a popular destination for camping and ATVing, and famous among paddlers for river travel by canoe and kayak. The tourism industry within this watershed has also become increasingly commercialized, with several options available for lodging in the area. Hunting and fishing are also popular recreational

activities within this watershed, with permits issued and regulations managed by the Yukon Government (Yukon Government, 2025c, 2025e).

In the 2025-2026 hunting season, hunting of large game including male moose, wood bison, deer, elk, and various bear and canine species were open for specific windows for harvest by permit (Yukon Government, 2025e). Male goats and sheep can be hunted with a permit in specific zones in the area, but hunting of male caribou is prohibited. Based on a moose population survey in the Mount Lorne Moose Management Unit (MMU) conducted in November 2021, the Yukon Government has increased the number of permits issued in this hunting season, doubling the number of permit hut authorizations from four to eight (Goorts & Czetwertynski, 2021; Yukon Government, 2025e). Findings from the survey reported an above average moose population density in the area, however found that the number of calves and yearlings per 100 adult cows was below average relative to other MMU within Yukon (Goorts & Czetwertynski, 2021). This increase in hunting permit allocations raises concerns among C/TFN citizens, who feel that ecosystems throughout the watershed are sick after withstanding significant stress from mining, recreation and hunting activities (A. Cherian- Hall, Pers. Comms). C/TFN staff and citizens are instead advocating to reduce the frequency of these activities by saying that the Wheaton River and broader watershed is showing signs of stress and must be given an opportunity to rest. The *'Let Wheaton Rest'* initiative is rooted in Traditional Knowledge and a comprehensive understanding of broader ecosystem health, suggesting the use of a quadrant system, where activities rotate among quadrants annually (A. Cherian-Hall, Pers. Comms).

1.8 Thesis Objectives and Outline

The overarching objective of this thesis is to assess the mobility of trace metals from mine-impacted sites within the Wheaton Watershed on downstream Bennett Lake using a holistic, co-developed approach through investigating 1) trace metal distribution and potential for mobility between different environmental media at a watershed-scale; and 2) quantifying concentrations downstream of mine-impacted sites to assess potential contributions of trace metals from the Wheaton River to Bennett Lake.

Following the quantification of trace metal concentrations in all sampled environmental media, concentrations will be relatively compared across all regions and media, to identify patterns and assess chemical similarities. Findings from these analyses will be supported by stable isotope

analyses and all validated by local knowledge. Further, current uptake and conditions for potential mobility of trace metals will be investigated through enrichment factor calculations, and a sequential extraction, respectively.

This watershed-scale assessment of trace metal mobility in the vicinity of mine-impacted sites will be followed by an analysis of water samples downstream of mine-impacted sites collected from the Wheaton River, near its inflow to Bennett Lake. Here, trace metal concentrations will be related to daily discharge values through flux calculations to estimate annual loads and mean annual flow weighted concentrations. This assessment of the relationship between discharge and contaminant concentrations will allude to potential trace metal contributions from the Wheaton River to Bennett Lake and compared to dynamics observed in other regional catchments.

Findings related to the objectives outlined above will be discussed in the context of the *Umbrella Final Agreement* and community-led initiatives in water governance, monitoring, and environmental stewardship.

Chapter 2: Manuscript

A Holistic Assessment of Trace Metal Mobility near Abandoned Mine Sites in the Wheaton River Watershed, Yukon, Canada

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This chapter is in preparation for submission to *Environmental Science: Processes and Impacts*.

Abstract

The Wheaton River Watershed, located in southern Yukon, feeds Bennett Lake and ultimately the Yukon River, and has been heavily impacted by recreation and mining activities dating back to the Klondike Gold Rush. This watershed is within the Traditional Territory of the Carcross/Tagish First Nation, who have stewarded these lands since time immemorial, and hold concerns over impacts of point source contamination on broader ecosystem health. Co-created with C/TFN staff and community members, this work took a holistic approach to 1) investigate current trace metal distribution and potential mobility between different environmental media on a watershed scale and 2) quantify concentrations downstream of mine-impacted sites to assess potential contributions of trace metals from the Wheaton River to Bennett Lake. Following collection of water, plant, scat, and sediment samples in 2025 from two mine-impacted regions and one downstream site, trace metal concentrations were quantified in all samples, focusing on those posing known risks to human and animal health (As, Cd, Pg, Pb, and Sb) and supported by sequential extraction of sediments and stable isotope analyses (C, N, O, H). Findings show that most metals are stored in sediments at mine-impacted sites with proportions of readily bioavailable metals varying regionally: Cd and Sb were more bioavailable at Skukum and Goddell, respectively, while other metals (As, Pb) were not preferentially taken up by biota at either site, and still others (THg, MeHg) showed no correlation between concentrations in sediment and concentrations in biota. Trace metal distribution followed the same regional trends in water samples; however, holding much lower concentrations, especially in the mainstem Wheaton River where concentrations were below water guidelines. Flux analyses and yield calculations suggest Wheaton River trace metal exports into Bennett Lake are comparable or below regional averages. This research provides an initial overview of contaminant mobility, highlighting the complex, connected, and dynamic nature of the watershed. Findings from this pilot study will help inform land planning initiatives, remediation efforts, and guide future research in the region.

Introduction

Rivers serve as integrators of various ecosystems throughout a watershed, acting as key players in shaping landscape composition and as the cornerstone to many cultural traditions and ecosystem services (Carone et al., 2009; Cooke et al., 2025; Emmerton et al., 2023; McDowell et al., 2023; Moore, 2015). Landscapes shaped by rivers are highly dynamic, undergoing constant co-evolution (Emmerton et al., 2019; McDowell et al., 2023; Moore, 2015; Vannote et al., 1980). In subarctic and arctic regions, watersheds are experiencing change at disproportionately high rates due to both climate change, and anthropogenic impacts resulting from both longstanding and growing interests in natural resources (Immerzeel et al., 2020; McDowell et al., 2023; Woo & Thorne, 2006). Alpine and Arctic watersheds are particularly sensitive to these changes, as their landscapes host a series of complex ecosystems, endangered species, rapidly changing glaciers and permafrost, and dynamic systems that present many hazards and downstream impacts (Immerzeel et al., 2020; McDowell et al., 2023).

Rivers are also key players in the transport, storage, and transformation of trace metals through both natural processes like natural seasonal fluctuations in discharge; and anthropogenic processes including dam construction, and disruptions caused by industry within their catchments. These principles have been applied at a watershed level in mountainous and boreal forest landscapes within Canada to assess contaminant mobility within watersheds impacted by a series of natural and anthropogenic stressors including forest fires (Emmerton et al., 2020), mining (Cooke et al., 2025), agriculture (Emmerton et al., 2023) and climate change (Emmerton et al., 2019). One way to assess the potential for mobilization of metals is through sequential extraction methods, which have been used to assess anthropogenic contributions to contamination of floodplain sediments and potential for mobility; however, little is known of the broader ecosystem impacts on biota and beyond (Clark et al., 2014; Kanianska et al., 2022).

The Wheaton River Watershed (824 km²) is the second largest watershed feeding Bennett Lake in southern Yukon Territory (Canada) and overlaps with the Traditional Territories of both Carcross/Tagish First Nation (C/TFN) and Kwanlin Dün First Nation (KDFN). The watershed has been heavily impacted by mining activities since the Klondike Gold Rush (1896), undergoing extensive exploration for gold, silver and antimony focused within the Skukum Mountain Complex and Goddell Portal regions of the catchment. Given known extractants of interest paired with

concerns for contamination, metals recognized as non-essential, with known negative impacts on biota such as As, Cd, Pb, Hg, and Sb, are of special interest. While studies in the natural sciences have traditionally evaluated the impacts of mining on water or sediments or biota (Clackett et al., 2021; Kanianska et al., 2022; Palmer et al., 2019; Stachiw et al., 2019; Verbruggen et al., 2013), few studies consider trace metal distributions across multiple environmental media (water, sediments, plants, animals) on a watershed-scale, a holistic approach that recognizes the interconnectedness between these watershed components, rooted in Indigenous knowledge systems.

In this present study, we assessed trace metal mobility from mine impacted sites within the Wheaton River Watershed using a holistic, co-developed approach. We integrated perspectives from Indigenous Knowledge and Western Science to 1) investigate current trace metal distribution, 2) assess potential mobility between different environmental media on a watershed scale, and 3) quantified concentrations downstream of mine-impacted sites to assess potential contributions of trace metals from the Wheaton River to Bennett Lake. While current multi-media watershed-scale assessment of contaminants that draws on the strengths and perspectives of both Indigenous and Western scientific knowledge systems is especially limited, they are necessary to generate meaningful findings that are effective in addressing community research questions and priorities.

Methods

Site Description

The Wheaton River Watershed (875 km²) is within the Boreal Cordillera (Yukon Stikine Highlands Ecoregion) in the Yukon Territory, Canada (Smith et al., 2004). It is the second largest tributary to Bennett Lake (Figure 5) and characterized by a cool, dry climate. The Wheaton Watershed is dominated by high alpine barren grounds (55% watershed coverage), and lowlands composed of temperate coniferous, shrubland, and grassland landscapes (22.3 %, 6% and 9,8% watershed coverage, respectively) (MacNeil et al., 2025). Once heavily glacierized, the Wheaton and Radelet glaciers now cover only 1% of the Wheaton River Watershed (Church & Clague, 2008; Painter et al., 2023). Discontinuous and sporadic permafrost underlie the watershed (Smith et al., 2004; USGS, 2013). The region is characterized by large gradients in biophysical composition as a function of elevation, with sparse, lichen-dominated landscapes in the high alpine, and subalpine regions commonly featuring shrub birch, willow, labrador tea, soapberry, balsam

poplar, fireweed, horsetail, and lodgepole pines. Wildlife are present within the region, including mountain goats, sheep, moose, black and grizzly bears, wolves, and the small Ibex caribou herd.

The Wheaton River is fed by snowmelt originating in the high alpine, with spring freshet typically occurring in early May (Environment and Climate Change Canada, 2026) (Figure A1). In summer months, the river continues to be fed by high alpine snowmelt and precipitation, with water levels typically reaching the annual maximum in early July (Environment and Climate Change Canada, 2026) (Figure A1, A2). The mean annual discharge of the Wheaton River is highly variable, measured as 9.41 m³/s in 2024, and 6.41 m³/s in 2025.

The watershed is underlain by Stikinia terrain, holding several volcanic formations composed of dacite, andesite, and breccia (Hart & Radloff, 1990) (Figure A3). Tectonic activity paired with favourable hydrothermal conditions have resulted in several mineralized faults, veins, and shear zones, hosting gold, silver, and other metals of economic value (Hart & Radloff, 1990; Smith et al., 2004). Given the market value of gold and silver, mining and industrial developments have been a long-standing anthropogenic pressure in this watershed, dating back to the Klondike Gold Rush in the late 1800s (C/TFN Government, 2014).

Sample Sites and Collection

This project was co-developed by engaging in conversations with the research team, composed of C/TFN staff and community members and academic partners, to ensure research questions aligned with local priorities both in the short- and long-term regarding understanding of environmental contaminants within C/TFN Traditional Territory. Sampling site selection was guided by C/TFN community concerns identified by the research team and access considerations. Of the four sites sampled, Skukum Mountain (SM) and Goddell Portal (GP) both represent landscapes that are directly mine-impacted; while Upper Wheaton (UW) and Lower Wheaton (LW) are both non-mine-impacted sites within the Wheaton River valley, downstream of mine-impacted sites (Figure 5). The Skukum Mountain mine site was originally staked in 1922 and actively mined for gold and silver (Yukon Government, 2025b). Since its establishment, Skukum Mountain has been the setting of a series of exploration and mining projects. Further downstream Goddell Portal mine site was actively mined for gold, silver, and antimony, originally staked in 1906, and has since undergone a series of exploration projects (Yukon Government, 2025a).

Water, plant, scat and sediment samples were collected from Skukum, Goddell, and LW in summer 2025 (Table A1). On July 9, an additional water sample was collected from UW. Both Goddell and Skukum required challenging back country site access with multiple river crossings and were sampled once each on June 24 and July 9, respectively. Both sites were heavily impacted by landslides in 2024, blocking direct access to the Skukum mine site. At the Skukum mine site, various landslide-impacted creeks and a pond suspected to hold tailings discharge were sampled in accordance with local knowledge of the mine footprint. Water samples from LW were collected on an approximately monthly basis ~20 km downstream of mine-impacted sites in both 2024 (May 16 to October 9) and 2025 (May 5 to November 10) to understand the magnitude and variability of chemical exports from the catchment into the lake. Plant samples from the LW site were collected on July 9, 2025.

Samples of all environmental media were collected, while prioritizing collection of plants with known traditional uses, and those known to be consumed by local fauna. Owing to the inevitable variability in plant community composition across sites, the same species were collected at each where possible.

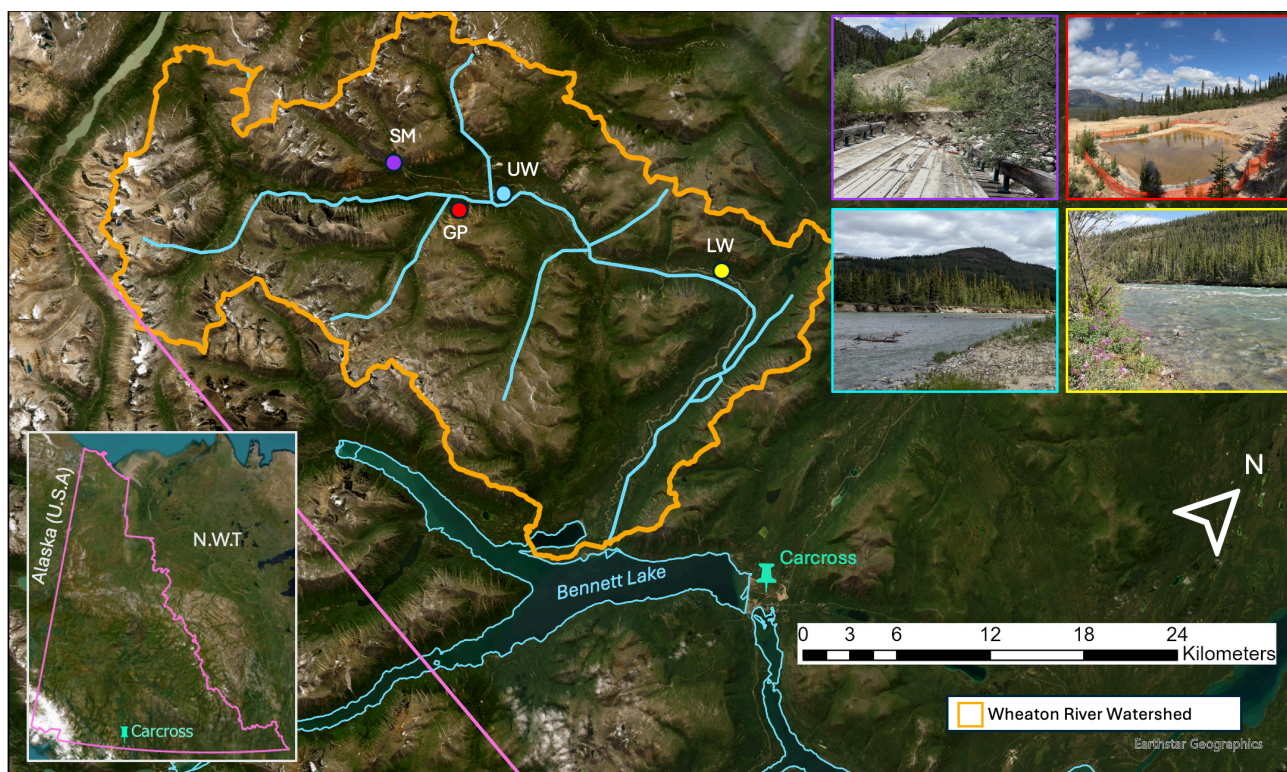


Figure 5. Regions for sample collection: Goddell Portal (GP), Lower Wheaton (LW), Upper Wheaton (UW) and Skukum Mountain (SM). SM region includes Skukum Creek and Berney Creek (unimpacted).

Water Samples

At each site, a multi-parameter YSI EXO1 sonde was used to measure pH, turbidity, dissolved oxygen, temperature, barometric pressure, and specific conductivity. Sample bottles for major cations (Ca, Mg, Na, K), anions (Cl^- , SO_4^{2-}), ammonium (NH_4^+), nitrate (NO_3^-), total dissolved phosphorus (TDP), phosphate (PO_4^{3-}), dissolved organic carbon and stable isotopes (DOC, $\delta^{13}\text{-DOC}$), and water stable isotopes ($\delta^2\text{H-}$ and $\delta^{18}\text{O-H}_2\text{O}$) were rinsed three times with filtrate (0.45- μm polyethylenesulfonate (PES) filter) using a sample-rinsed syringe. Bulk water samples were collected to measure total phosphorous (TP). Cation and DOC samples were preserved using 21 μL of 70% HNO_3 and 40 μL of 37% trace metal grade HCl , respectively. Cation, anion, TDP, TP, TDN, DOC and water isotope samples were kept refrigerated, while PO_4^{3-} , NH_4^+ , and NO_3^- samples were kept frozen prior to analysis. Bulk water collected at each sampling site was filtered through two muffled glass microfiber filters, acidifying one. Bulk water filters were folded and frozen in foil until preparation for analysis for particulate organic carbon ($\delta^{13}\text{C-POC}$) and nitrogen ($\delta^{15}\text{N-PN}$).

Both filtered and unfiltered trace metal samples were collected at all sample sites in certified amber HDPE bottles (Thermo Scientific, N3010125). Filtrate for filtered samples was passed through a 0.45- μm polyethylsulfonate (PES) filter using a sample-rinsed syringe. All trace metal samples were preserved with 70% trace metal grade HNO_3 . Unfiltered water was collected for mercury analysis in certified amber glass bottles (Thermo Scientific™ 1490125) using EPA method 1669 (USEPA, 1996). Samples were acidified to 0.2% HCl using 37% trace metal grade HCl. Samples were refrigerated prior to analysis.

Solid Samples

Solid Samples were collected into Whirlpaks and frozen immediately upon return from the field. All solid samples were freeze-dried and homogenized using an acid-washed stainless-steel grinder or mortar and pestle prior to laboratory analysis.

Laboratory Analysis

Water

Water samples were analyzed for the parameters described above using standard protocols at a series of laboratories including the University of Alberta Biogeochemical Analytical Service Laboratory (TP, TDP, TDN, PO_4^{3-} , NH_4^+ , and NO_3^-), and the University of Ottawa's Ján Veizer Stable Isotope Laboratory (DOC; filters; DOC, $\delta^{13}\text{-DOC}$; $\delta^2\text{H-}$ and $\delta^{18}\text{O-H}_2\text{O}$), the Laboratory for the Analysis of Natural and Synthetic Toxins (THg, MeHg, trace metals), and Geochemistry Laboratory (major cations, major anions, trace metals) (Table A2). Sample preparation and analysis of THg and MeHg was done following EPA methods 1631 and 1630, respectively (USEPA, 1998, 2002) (Section A1, A3).

Solid Samples

Plant, scat, and sediment samples were analyzed for trace metals (As, Ca, Cd, Co, Cr, Cu, Fe, Mg, Mn, Ni, Pb, Sb, Zn) by total acid digestion ($\text{HNO}_3/\text{H}_2\text{O}_2$) following EPA method 200.2 and 3050b for organic and sediment samples, respectively, followed by dilution (Section A2) (T. D. Martin et al., 1994; U.S Environmental Protection Agency, 1996). Digests were then analyzed by inductively-coupled plasma mass spectrometry (ICP-MS). Recoveries for all reference materials were calculated and reported (Table A4).

Solid samples for THg analysis were prepared using EPA method 7473 (USEPA, 2007), followed by analysis on the NIC MA-3000 Direct Mercury Analyzer (Section A3). Biota (scat and vegetation) and sediment samples were prepared for MeHg analysis using EPA method 1630 (USEPA, 1998), followed by analysis on a Tekran 2700 (Section A3). % C, %N and stable isotope analyses were performed following standard methods, running known and blind calibration standards and 5% of samples in duplicate (Section A4). Following lab analyses, recoveries for all reference materials were calculated and reported (Table A5, A6, A7).

Sequential Extraction of Sediment Samples

Sediment samples were analyzed in duplicate using a modified five-step sequential extraction procedure to constrain geochemical conditions for trace metal mobility from the sediments (Filgueiras et al., 2002; Rauret et al., 1999; Tessier et al., 1979). The five steps were: (1) water-soluble using milli-Q® water; (2) exchangeable using 1M NH₄Cl solution; (3) acid-soluble using 0.11M acetic acid solution; (4) reducible using 0.5M NH₂OH-HCl in 0.05M HNO₃; and (5) oxidizable using a 30% w/w H₂O₂ digestion followed by 1M NH₄OAc solution (Filgueiras et al., 2002; Rauret et al., 1999; Tessier et al., 1979) (Section A5, Table A8). Extractants were prepared for analysis by evaporation and a series of acid digestions and dilutions (Section A5). The residual fraction, accounting for metals bound to silicate minerals, was calculated as the difference between total digested and the sum of extractant concentrations from steps 1-5 (Filgueiras et al., 2002; Kanińska et al., 2022; Rauret et al., 1999; Tessier et al., 1979). All samples were analyzed for trace metal and major cation concentrations using ICP-MS and inductively-coupled plasma optical emission spectrometry (ICP-OES), respectively. Blanks were run in 10- sample intervals along with a series of eight reference samples and two digestion blanks.

Data Analysis

Following laboratory analysis, QA/QC on solid, water, and sequential extraction samples was completed and reported (Table A9, A10, A11). Resulting datasets were imported into RStudio (Posit Team, 2026) for visualization and subsequent statistical analyses using *tidyverse* (Wickham et al., 2019), *vegan* (Oksanen et al., 2026), *patchwork* (Pedersen, 2025), *rloadest* (Runkel et al., 2004), *ggrepel* (Slowikowski et al., 2026), *cluster* (Maechler et al., 2026), and *pheatmap* (Kolde, 2025).

Enrichment factors (EF) were calculated using Eq 1 to determine metal uptake by plants relative to the underlying substrate (Puckett & Finegan, 1980; St. Pierre et al., 2015).

$$\text{Enrichment Factor} = \frac{\left(\frac{\text{element x}}{\text{reference element (Fe)}} \text{ in } \mathbf{vegetation} \right)}{\left(\frac{\text{element x}}{\text{reference element (Fe)}} \text{ in } \mathbf{sediment} \right)} \quad (1)$$

Principal component analysis (PCA) for both water and solid samples were conducted using *vegan*. Analysis of variance (ANOVA) were used to compare metal concentrations, enrichment, and extractable fractions by site and matrix, followed by Tukey’s post hoc test where applicable. All sample types (water, plant, scat, sediment, tailings (water and sediment)) were then combined, z-scored, and log-transformed, followed by clustering analysis using ‘ward.D2’ clustering criterion (Ward, 1963).

Fluxes of metals from the Wheaton River to Bennett Lake were quantified by relating measured concentrations to mean daily discharge values (Environment and Climate Change Canada, 2026) using log-linear models within the *rloadest* package (Runkel et al., 2004). Models were constructed using data collected from 2024 and 2025 for the open water season (OWS) from April 1 to November 31. Daily load estimates for 2025 are missing between September 15 to 23 (inclusive) and following October 27th due to lack of published discharge data. In cases where constituent concentrations fell below detection limits, half the detection limit was used in its place (Analytical Methods Committee, 2001). *rloadest* models were initially chosen to minimize Akaike’s information criterion (AIC), followed by manual assessment for goodness of fit using bias percentage (Bp), partial load ratio (PLR), and the Nash Sutcliffe efficiency index (E) (Nash & Sutcliffe, 1970; Runkel et al., 2004; St. Pierre et al., 2021) (Table A12). Discharge values recorded during the atmospheric river event in July 2024 were well above the range of discharge values with known constituent concentrations, resulting in the prediction of negative fluxes for some chemical constituents (Co, Pb, Zn) during this period. Negative values in the affected models were replaced by the product of mean constituent concentration and daily discharge, with error calculated as the product of the standard deviation (SD) of the mean constituent concentration and daily discharge. Unimpacted constituent errors for annual flux were quantified by propagating the daily standard error of prediction (SEP), therefore incorporating both uncertainty of load estimate and model fit (St. Pierre et al., 2021). Using the same method selection and QA/QC criterion,

annual loads of As for Yukon and Mackenzie Rivers were calculated using available discharge and water quality monitoring records (ArcticGRO, 2024) to assess estimated annual loads from the Wheaton River in a regional context.

Mean annual flow-weighted concentrations (FWC) were calculated to account for large inter-annual variation in discharge using Eq 2 (St. Pierre et al., 2021).

$$\text{Mean annual FWC} = \frac{\sum \text{daily load estimate}}{\sum \text{daily water volume}} \quad (2)$$

Although all analytical decisions surrounding data handling and interpretation were performed by the authors, Chat GPT Language Model (OpenAI, 2025) was used to improve code efficiency, aesthetic presentation, and formatting of data handled in RStudio.

Results

Trace Metal Concentrations in Sediments, Plants, Scat and Water

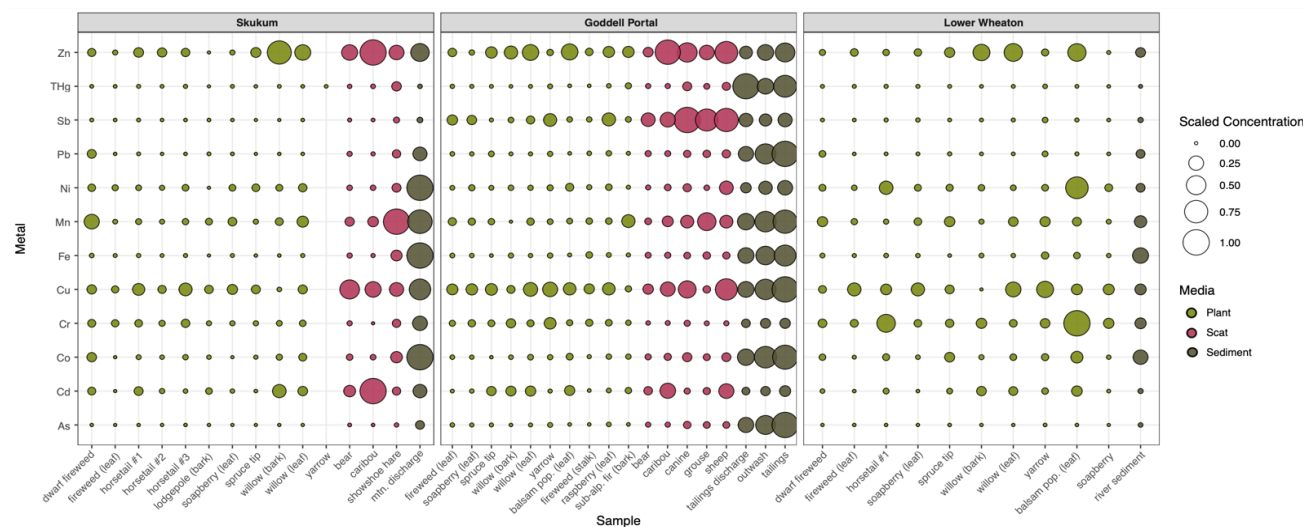


Figure 6. Relative metal concentrations within solid samples collected (plant, scat, sediment) scaled from 0-1 by metal. Samples are plotted by region and coloured by media. Mine-impacted regions (Skukum and Goddell Portal) and downstream site (Lower Wheaton) are featured.

Metals of concern will be discussed in more detail, however, remaining metals analyzed are presented in Table A9. As concentrations ranged between 0.020 to 4.60 mg kg⁻¹, 0.17 to 25.9 mg kg⁻¹, 3.27 to 780 mg kg⁻¹, and 0.108 to 52.5 µg L⁻¹ in plant, scat, sediment, and water, respectively (Table A9). Cd concentrations ranged from 0.05 to 3.47 mg kg⁻¹, 0.275 to 17.4 mg kg⁻¹, 0.130 to 3.65 mg kg⁻¹, and 0.002 to 0.232 µg L⁻¹ in plant, scat, sediment, and water, respectively (Table

A9). Pb concentrations ranged from 0.006 to 6.79 mg kg⁻¹, 0.228 to 5.47 mg kg⁻¹, 6.65 to 111 mg kg⁻¹, and 0.02 to 7.73 µg L⁻¹ in plant, scat, sediment, and water, respectively (Table A9). Sb concentrations ranged from 0.002 to 0.932 mg kg⁻¹, 0.005 to 4.84 mg kg⁻¹, 0.044 to 1.07 mg kg⁻¹, and 0.048 to 16.1 µg L⁻¹ in plant, scat, sediment, and water, respectively (Table A9). THg concentrations ranged from 0.070 to 23.0 ng g⁻¹, 4.50 to 113 ng g⁻¹, 1.51 to 1519 ng g⁻¹, and 0.122 to 62.7 ng L⁻¹ in plant, scat, sediment, and water, respectively (Table A9). MeHg concentrations ranged from 0.001 to 1.646, 0.028 to 11.587, and 0.047 to 0.841 ng g⁻¹ for plant, scat and sediment respectively (Table A9).

Metal concentrations differed between region and media (Figure 6 and 8). Concentrations of As, Pb, Co, THg, and Fe in mine-impacted sediments were relatively much higher than in all other samples (Figure 6, Table A9). Regional trends were also observed between mine-impacted sediments with Goddell sediments having higher concentrations of THg, As, Pb, and Sb, while Skukum sediments had higher concentrations of Cd and Zn. As, THg and Pb concentrations in Goddell sediments were significantly different than those in other samples from all media and regions, while Cd concentrations in scat were significantly higher from those in plant samples across all regions (ANOVA, $p < 0.05$). Cd concentrations were also relatively elevated in willow across all regions and in caribou scat collected at Skukum (Figure 6, Table A9).

Goddell water samples were chemically distinct from each other and other water samples, with significantly higher concentrations of As, Co, Cu, Mn, Ni, Pb, Sb and THg compared to samples collected from other regions (ANOVA, $p < 0.05$) (Figure 7, 8, and A4). Despite their proximity, the rockslide stream and tailings were isotopically and chemically distinct (Figure 7, 8 and A4). Cd and Zn concentrations in Skukum Creek were the highest in the watershed and unlike other samples collected in the Skukum region. Lower Wheaton River water showed a large range in Cr concentrations, with concentrations sometimes exceeding those in all other water samples (Figure 7, Table A13).

All mainstem Wheaton River (UW, LW) samples collected downstream of mine-impacted sites were below Canadian water quality guideline concentrations for the protection of aquatic life (Canadian Council of Ministers of the Environment (CCME), 1999) (Table A14), apart from Fe concentrations from the Lower Wheaton in samples from June 2025. Both rockslide stream and tailings samples exceeded guidelines for As, with the rockslide stream also exceeding guidelines

for Cd, Fe, and Pb, and the water overlying the tailings exceeding the THg guideline. Skukum Creek water exceeded guidelines for Cd and Zn and displayed distinct chemistry relative to other samples on both regional and watershed scales (Figure 8).

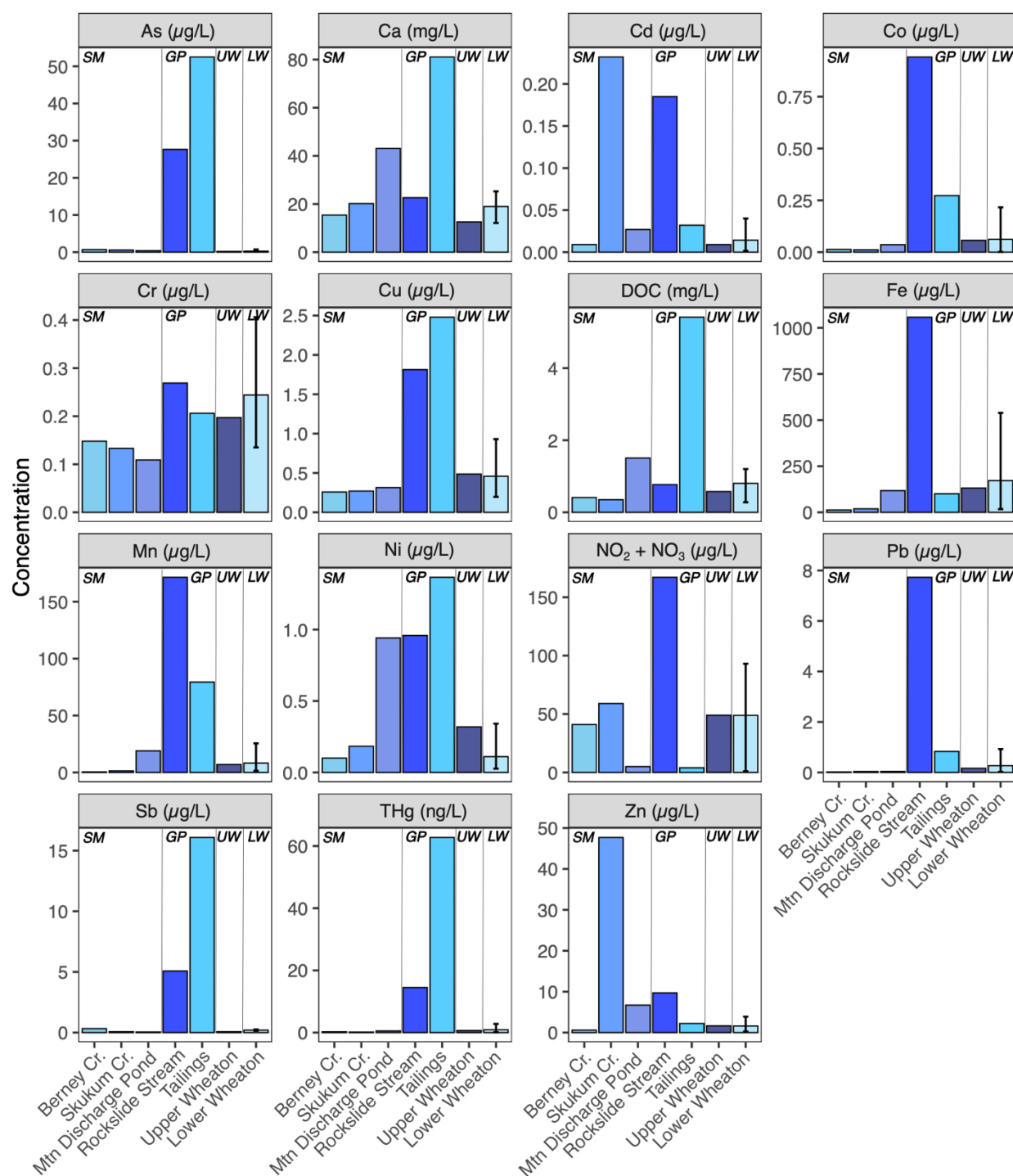


Figure 7. Bar graph of metal and nutrient concentrations in unfiltered water samples, plotted by constituent. Due to repeated sampling at Lower Wheaton, the mean concentration from 2024/25 and range (error bars) was plotted for each constituent. Please note that scale and units differ by constituent.

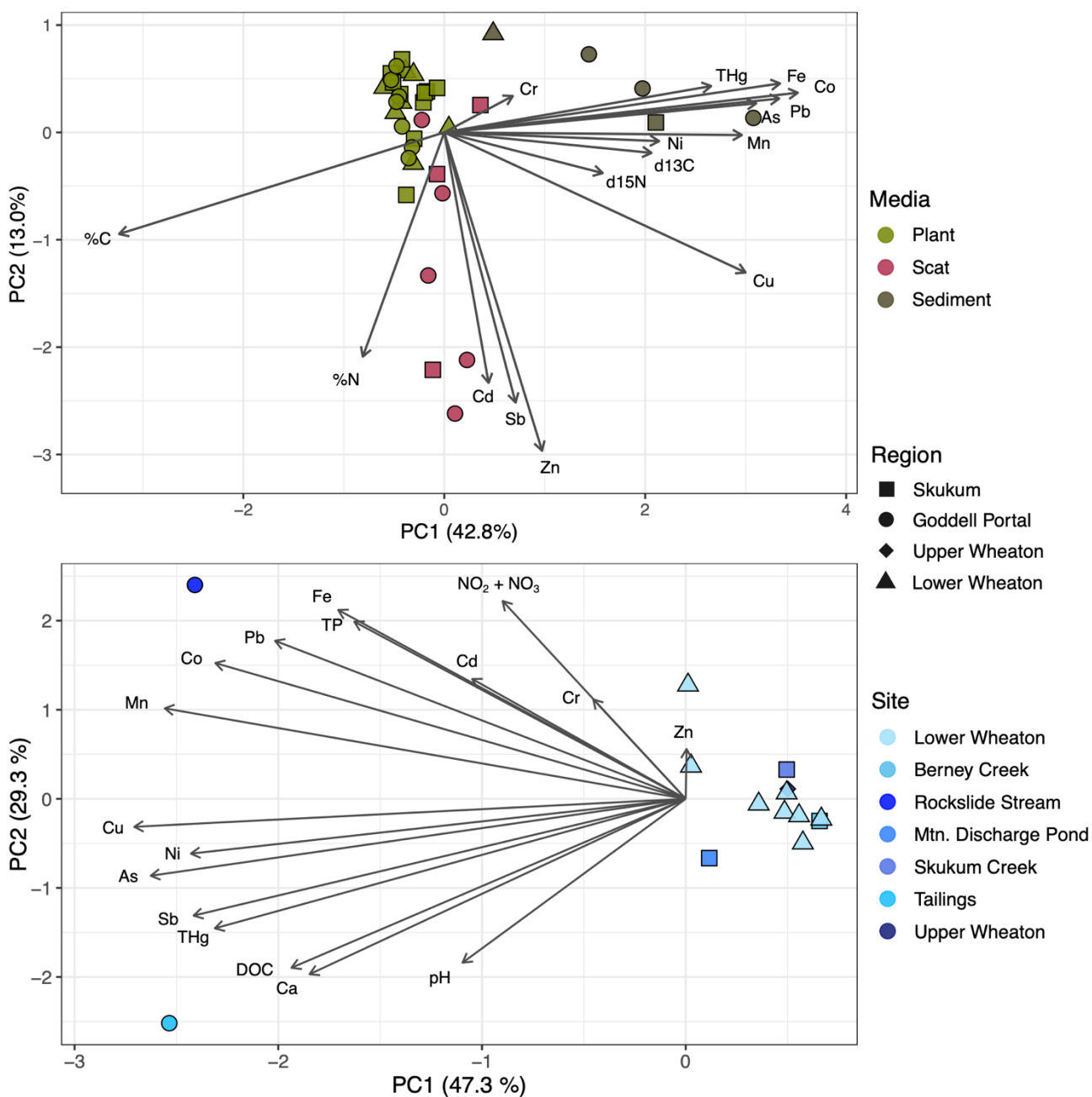


Figure 8. Principal component analysis of standardized sample chemical compositions showing a) trace metal concentrations, carbon and nitrogen content and stable isotope signatures ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$) for all solid samples coloured by media and shaped by region; b) trace metal, nutrient and dissolved organic carbon (DOC) concentrations in water samples coloured by site and shaped by region.

In total, 57.1% of the variance in solid sample chemical composition was explained by two principal components (Figure 8a), representing 47.1% and 14.8%, respectively. As, THg, Pb, Fe, Mn and Ni were the primary contributors to PC1, while Cd, Zn and Sb contributed predominantly to variability on PC2 (Figure 8a). Samples grouped by media, with the majority of variability in sediment and scat samples captured in PC1 and PC2, respectively. In total, 76.6% of the variance in water sample chemical composition was explained by two principal components (Figure 8b), accounting for 47.3% and 29.3%, respectively. As, THg, Sb, Pb, Fe, Mn, and Cu were the primary contributors to PC1 while Zn, pH, NO₂+NO₃, DOC, Ca, Fe, TP and Cd contributed predominantly to PC2 (Figure 8b). Samples collected at Goddell were both chemically distinct from other samples collected throughout the catchment, with variability in the region largely captured on PC2 (Figure 8b). Upper and Lower Wheaton samples were clustered together, with variability between them captured on both PC1 and PC2 (Figure 8b). Skukum water samples were not closely clustered together with PC1 and PC2 having contributed to variability among samples.

Potential for Trace Metal Mobility from Sediments

Across all regions, all metals except for As, Co, and Pb showed a median enrichment greater than 10, indicating significant enrichment in plant materials relative to the underlying sediment (Figure 9). A red horizontal line indicates the threshold for significant enrichment (EF>10). There was no enrichment (EF<1) of Pb or Co in plants from underlying soils across all regions, and for As at mine-impacted sites (GP and SM). Enrichment of THg, Sb, Cu, and Ni ratios in plants relative to regional underlying sediments differed significantly by region (ANOVA, $p < 0.05$). Sb and Cu EFs were significantly elevated in Skukum samples relative to Goddell, and Sb was significantly higher than those from the Lower Wheaton River (Tukey's post-hoc, $p < 0.05$). THg EFs were significantly lower (0.005 – 2.01) at Goddell, likely due to the higher THg concentrations (1518.951 ng g⁻¹) in the underlying sediments.

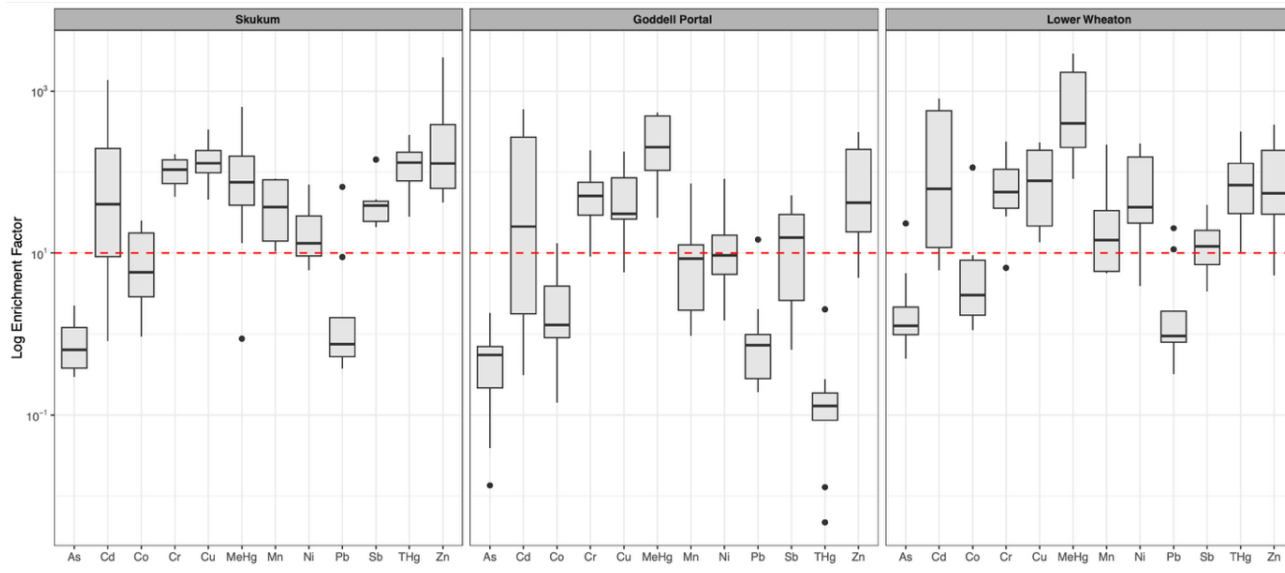


Figure 9. Local enrichment factors of trace metals in plants across all sampled regions in the Wheaton Watershed. Note that the y-axis is on a $\log_{10}$ scale.

The concentrations within each fraction for all non-Hg trace metals varied among mine-impacted and downstream sediments, and regionally (Figure 10). River sediments from Lower Wheaton showed significantly different distribution patterns of As compared to all other samples; however, all samples showed significantly higher concentrations of As leached in reducible conditions (13-23 % across all samples) and retained in the residual fraction (57 – 85%) relative to water and exchangeable fractions, which represented only 0.2-6 and 0.1-1 % of total concentrations, respectively. Like for As, Pb concentrations leached in the reducible fraction (45-60 %) was significantly higher than water, exchangeable, and acid-soluble fractions, with the rest primarily held within the residual fraction (31-53 %).

In contrast, significant differences were observed in Cd distribution in sediments between regions, and coherent for all samples from the same region. The fractions of exchangeable Cd and Zn were significantly higher at Lower Wheaton (30-32%), than at the Skukum (24%) and Goddell (9-13%). However, unlike Cd, larger concentrations of Zn were held in the residual fraction across all regions. Regional differences were less pronounced in Sb distribution; however, tailings discharge sediments were significantly different from all other Goddell samples. Further, Goddell samples had significantly more Sb in reducing fraction (31-87%) relative to samples from other regions (8-14%), where Sb was more abundant in the residual fraction.

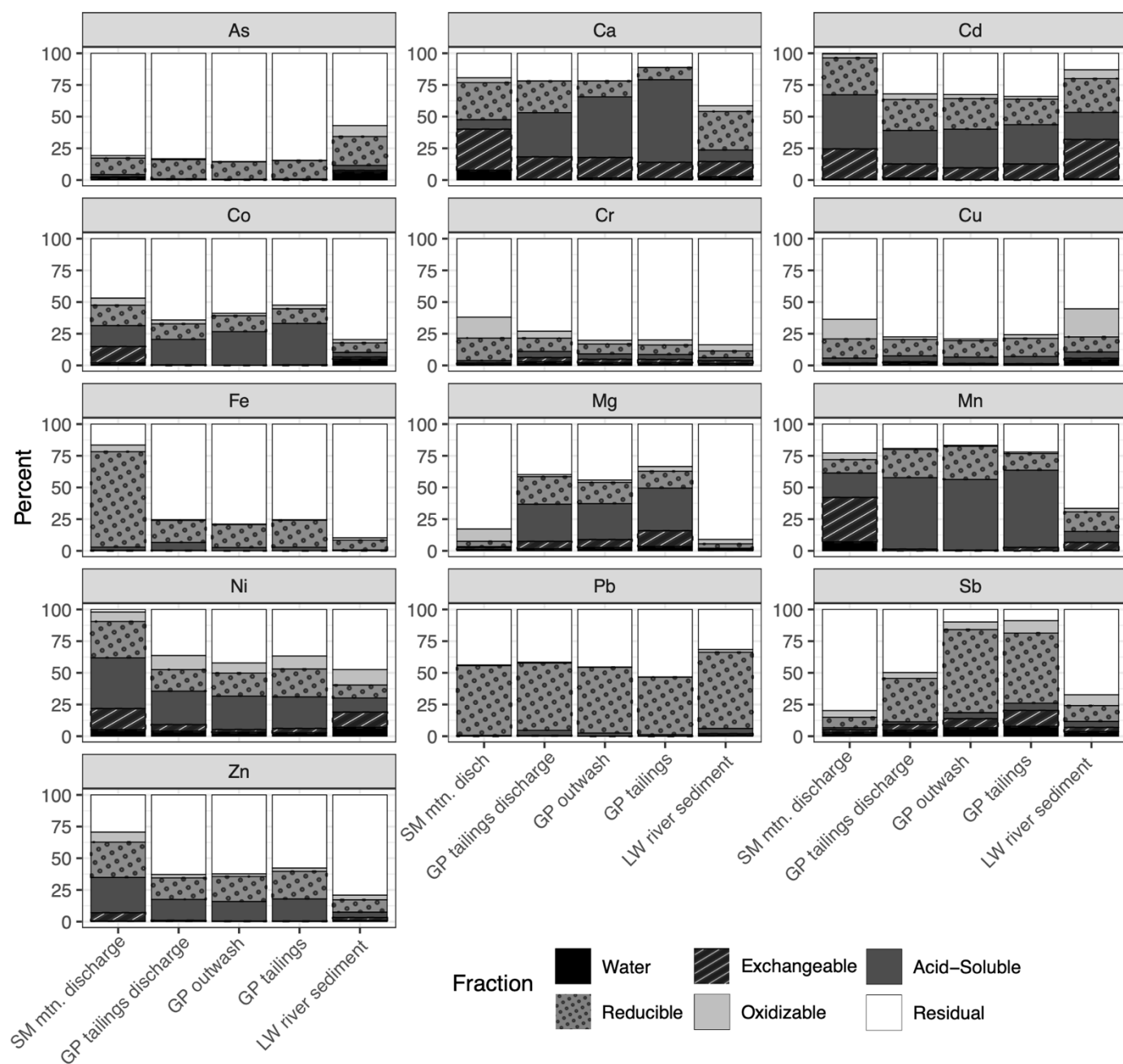


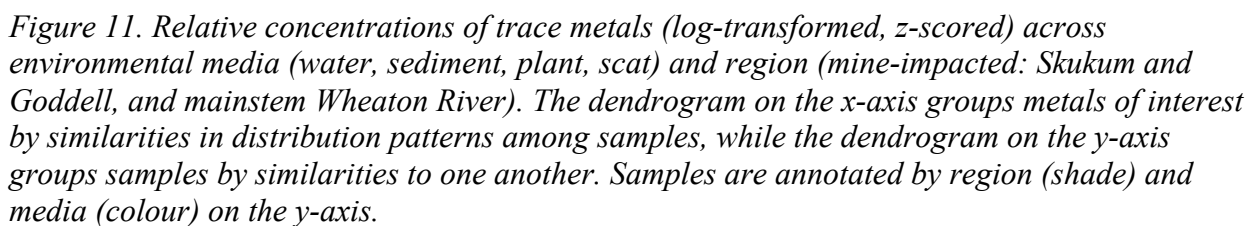
Figure 10. Percent of total non-Hg trace metal concentrations leached through sequential extractions in the water, exchangeable, acid-soluble, reducible, and oxidizable fractions from the sediment samples.

Metal Mobility within Environmental Media on a Watershed-Scale

We used clustering analysis to identify patterns in trace metal concentrations across environmental media and regions (Figure 11). THg distribution among samples was unique relative to other trace metals. Certain metals exhibited distinct pairings across sample types, namely As-Sb, Pb-Fe, Cd-Zn, Co-Mn, and Ni-Cr-Cu (Figure 11). Cd-Zn and As-Sb pairs behaved similarly with relatively elevated concentrations in willow (all sites), and Skukum and Goddell scat and plants, respectively (Figure 11). Mine-impacted sediments were clustered together, and clearly distinct from Lower Wheaton River sediments, which were clustered more closely with biota (Figure 11). Plant samples from Skukum and Lower Wheaton regions were clustered together, separate from other media, while most plants collected from Goddell were clustered with scat collected within the region. Relatively elevated Sb concentrations among Goddell plant and scat samples were a source of variability between samples collected at Skukum and Lower Wheaton.

Scat samples were generally clustered by region, with Skukum samples showing elevated concentrations of Cd relative to Goddell (Figure 11). Cd was consistently distributed among solid samples.

Water samples clustered distinctly from all other sample types, likely due to lower relative trace metal concentrations (Figure 11). All Skukum water samples were adjacent within the dendrogram; however, the water chemistry from the unimpacted Berney Creek was more similar to the Lower Wheaton River samples (Figure 11). Nearly all water samples from the Wheaton River mainstem (UW and LW) clustered together, apart from those collected in June 2025 (Figure 11).



Modelling Downstream Impacts: Chemical Exports into Bennett Lake

Annual estimated loads (SE) of As, Cd, Pb, and THg from the Wheaton River were 150.4 (49.5), 4.9 (0.7), 165.5 (253.0), and 0.5 (0.2) kg yr⁻¹ for the open water season (OWS) in 2024, respectively; and 42.7 (2.9), 2.3 (0.3), 46.3 (61.4), and 0.1 (0.01) kg yr⁻¹ in 2025. Estimated annual loads for all other chemical constituents discussed are reported in Table A15. Concentration-discharge relationships for water quality parameters were assessed with significant ($p < 0.05$) positive relationships observed for As and Cr (unfiltered), and negative relationships observed for Ca and Mg (Figure A5). Flux predictions for the OWS were higher across all constituents in 2024 compared to 2025. To account for large inter-annual variations in discharge, mean annual flow weighted concentrations (FWC) were calculated for 2024 and 2025 (Table A15). All constituents but Sb, Ca, Mg, and DOC had higher mean FWC in 2024.

Annual loads of As, Pb and THg flowing from the Wheaton River into Bennett Lake were compared to other subarctic and arctic rivers, including the Old Crow River (Pb and THg from (Staniszewska et al., 2022)), the Yukon River at the Bering Sea (As, THg) from and the Mackenzie River (ArcticGRO, 2024; Zolkos et al., 2020). To account for large differences in catchment area and magnitude, annual contaminant yields were calculated and then compared by dividing predicted annual loads by catchment area (Zolkos et al., 2020). Among the four rivers, the Wheaton River had the lowest annual yield of THg and Pb (Table 1). The Wheaton River's annual yield of As fell between estimates for the Mackenzie and Yukon (Table 1).

Table 1. Mean (Standard Deviation) of annual yields (g/km²/yr) calculated from corresponding annual flux predictions (Table A15) for As, Pb, and THg within specified catchments.

		Yield (g/km ² /yr)		
	Area (km ²)	As	Pb	THg
Wheaton (2024-2025)	823.5	117 (93)	129 (102)	0.35 (0.29)
Old Crow (2019)	13 900	-	885 (NA)	0.85 (NA)
Yukon (2012-2017)	830 000	184 (23)	-	4.0 (0.5)
Mackenzie (2012-2017)	1 780 000	55 (7)	-	1.6 (0.4)

Discussion

Trace Metal Mobility within Mine-Impacted Sites

Strong trends in trace metal distribution between and among regions and media were observed throughout the watershed. Across all samples, concentrations by weight in water were lower than those observed in sediment and biota, especially in the samples collected in the mainstem Wheaton River, suggesting either or both low mobility and significant dilution by other hydrological inputs (Figure 11). Sequential extraction from the sediments did, however, suggest that while sediments held the highest concentrations of metals among samples analyzed, most metals were not readily mobile in the environment at the time of sampling. Further analyses could be done including mineral characterization (Sposito, 2016), and tracer analyses (Turunen et al., 2020) to help distinguish between dilution and retention of metals in sediments.

Goddell sediments had higher As, Pb, THg and Sb concentrations, while Skukum sediments were higher in Cd and Zn (Figure 6, 8a). While no specific geological units can be attributed to these differences, it does highlight the highly heterogeneous geology within the region. Further, sediment matrix composition differed regionally with Goddell samples having higher carbonate and lower organic content compared to those from Skukum (Table A16). These regional differences in sediment composition translated to their distributions among biota within the region (Figure 6, 9-11). In scat samples, differing exposures to metals are suggested based on excretion patterns observed by region (Figure 6, 11). At Goddell, where sediments had relatively high Sb concentrations, scat and some plants also had relatively high Sb concentrations. Though Sb uptake tendencies are highly variable and dependent on flooding dynamics and resulting soil physiochemistry, Sb is commonly thought to be poorly mobile in plants and to have a low potential to bioaccumulate (Ngo et al., 2016; Zhu et al., 2020) and strongly bind to soils and minerals (Flynn et al., 2003; Zhu et al., 2020). EFs indicate that Sb was enriched in vegetation across all regions, though higher concentrations were found in vegetation at Goddell (Figure 9). While current studies of Sb uptake mechanisms are largely centered around uptake in flooded environments, these results highlight the importance of understanding physicochemical conditions within soils to better constrain drivers of uptake by biota (Zhu et al., 2020).

Among all metals, Cd was most preferentially mobilized into willow and scat samples in the Skukum Mountain region, showing higher concentrations in scat than sediments, significant EF,

significantly higher concentrations are available at Skukum relative to other regions (Figure 6, 9). Across all regions, Cd uptake was highly favoured by willow, well known hyperaccumulators of Cd (Figure 6, 11) (Gamberg et al., 2020; Jensen et al., 2009). Woodland caribou favour willow consumption in the summer months, which likely explains the high Cd in caribou scat within the region (Gamberg et al., 2020). While these results agree with studies of Cd levels in woodland caribou within the region, scat-only assessments prevent drawing conclusions around food safety and toxicity since they provide only snapshot insights into animal metal exposures and excretions, and no insights into metal retention (Gamberg et al., 2020). Zn follows similar trends to Cd in distribution throughout biota; however, their differing and potentially cumulative toxicological properties should be considered in studies surrounding animal health and food safety for subsistence hunters (Gamberg, 1994; Gamberg et al., 2020). Along with scat at both mine-impacted sites, hoofprints (goat or sheep) in the Goddell tailings pond were present, evidence that animals are interacting with the mine-impacted environment. While the collection of scat samples enabled a first assessment of animal interactions with the site, scat is an imperfect proxy for understanding whether and how animals are using the site (Reid & Koch, 2017; Varsha, 2013).

Conversely, high concentrations of As, Pb and THg measured in sediments were not reflected in biota. Median EFs for Pb and As indicated no enrichment across almost all regions, supported by sequential extraction results, indicate very limited bioavailability of both metals in the environment with median percent extracted in water and exchangeable fractions combined as 0.7% and 0.3% for As and Pb, respectively. Compared to the other metals of interest, THg uptake by plants is independent of underlying sediment concentrations, with stomatal respiration likely a key driver of THg uptake in plants (Wohlgemuth et al., 2023). This is supported by enrichment factor calculations, where low THg EFs at Goddell and significant enrichment at Skukum and Lower Wheaton suggest that uptake was not proportional to underlying sediments (Figure 9). While the amount of organic carbon (%OC) correlates to higher %MeHg (as a proportion of THg) in sediments across the watershed as expected (Gworek et al., 2020), no relationship was observed between %MeHg in sediments and overlying biota (Figure 9). No one driver of Hg methylation in biota sampled can be isolated in this study due to the highly complicated and multi-faceted nature of these reactions in natural environments (Gworek et al., 2020; St Louis et al., 2001).

Trace metal distributions in water collected at mine-impacted sites reflected regional trends in sediment and biota; however, mine-impact signals were not detected in waters from the mainstem Wheaton River, indicating strong local influence on water quality (Figure 7, 8b). In the Skukum region, although samples were collected from Skukum Creek and Berney Creek 10m upstream of their convergence, significantly elevated concentrations of Cd and Zn in Skukum Creek and the mountain tailings discharge pond suggest signals from legacy mining persisted within this mine-impacted tributary to the Wheaton (Figure 7, 8b). The distinct water chemistry of these two water bodies (despite their proximity) paired with their similar isotopic signatures highlight the influence of mine-impact on water quality within the Skukum Creek Catchment.

Like Skukum, water samples collected in the Goddell region (tailings, rockslide stream) had different water chemistry (Figure 8b); however, distinct isotopic signatures suggested that they did not share a common source, and suggesting potential evaporative enrichment of rockslide stream waters (Barnes, 2022; Turner et al., 2010) (Figure A4). Tailings water had high concentrations of Sb, THg, and As, while the rockslide stream had higher concentrations of Pb, Mn, Co, Fe, and $\text{NO}_2 + \text{NO}_3$. While the isotopic signatures of the tailings fell near the LMWL suggesting its source is precipitation, relative enrichment of $\delta^2\text{H}$ and $\delta^{18}\text{O}\text{-H}_2\text{O}$ in rockslide waters indicated post-precipitation modification, which could potentially reflect interactions with freshly exposed rockslide materials, groundwater origin and permafrost interactions, or evaporative effects (Aron et al., 2021; Barnes, 2022; Newman et al., 2023; Turner et al., 2010). While an unknown water source and lack of supporting data prevent the isolation of the dominant driver within the system, enrichment of both $\delta^2\text{H}$ and $\delta^{18}\text{O}$ suggest likely evaporative effects, while only $\delta^{18}\text{O}$ enrichment would suggest that rock-water interactions dominate fractionation processes. Evaporative effects can, however, point to a wide range of environmental processes (Aron et al., 2021; Barnes, 2022; Turner et al., 2010).

Conditions Required for Future Mobility of Trace Metals from Sediments at Mine-Impacted Sites

Sequential extraction data suggests that trends of metal mobility from sediments was highly metal and sample-specific; however, samples showed release of metals at very low concentrations in water, suggesting that mobility into waterways was limited when unsupported by changes in environmental conditions such as pH, weathering, and redox potential (Figure 10, 11) (Filgueiras

et al., 2002; Róžański, 2013). Arsenic was primarily within the residual fraction, suggesting that most was bound to silicate minerals or within stable mineral structures (Figure 10) (Filgueiras et al., 2002; Kanińska et al., 2022). Indeed, quartzes are known to be abundant at Goddell and throughout the watershed (Hart & Radloff, 1990; Yukon Government, 2025a). Pb was mostly within the residual and reducible fractions, suggesting that while some is not readily mobile, Pb adsorption to amorphous Fe and Mn-oxides (e.g. goethite, ferrihydrite) and the creation of anoxic conditions (e.g. flooding) could drive release into the environment (Figure 10) (Filgueiras et al., 2002). Conditions needed for Sb mobility into the environment would vary by region, with most leaching in the reducible fraction at Goddell, but largely stable (residual fraction) elsewhere (Figure 10).

While Cd was relatively bioavailable, large proportions are stored in sediments and are acid soluble or reducible, suggesting that Cd was largely adsorbed onto carbonates (e.g. calcite) or amorphous Fe and Mn-oxides, with its release thus conditional on decreasing pH and oxygen concentrations, respectively (Figure 10) (Filgueiras et al., 2002; Nungula et al., 2024). While Zn showed similar distributions, higher proportions were in the residual fraction, indicating comparatively less with the potential for future release (Figure 10). These findings suggest that oxygen depletion, which can be triggered by an array of environmental changes including flooding and changes in groundwater routing, could be accompanied by increasing mobility of several metals into the environment (Li et al., 2024).

While these data offer valuable insights into sediment geochemistry, limited sampling in the Skukum and Goddell regions combined with the known heterogeneity of the local surficial geology does limit our understanding of the variability of metals in soils and sediment deposits across the watershed. Further, the impacts of surficial geological processes in driving future metal mobility cannot be understated. Processes like landslides, which are widespread in the Wheaton Watershed, can trigger cascading effects on water quality and landscapes by reshaping waterways (Cloutier et al., 2017; Owczarek et al., 2020), vegetation composition and animal migration routes (Geertsema & Pojar, 2007), and altering floodplain dynamics (Cloutier et al., 2017; Owczarek et al., 2020).

Downstream Impacts: Contaminant Loading into Bennett Lake

Extreme hydrological events play a crucial role in determining the Wheaton River's interaction with its watershed (Li et al., 2024; Staniszevska et al., 2022; Zolkos et al., 2020). Both OWS

fluxes and FWC were higher in 2024 than 2025, reflecting the higher discharge resulting from an atmospheric river event in July 2024 that triggered several landslides in the watershed and likely contributed to increased metal fluxes into Bennett Lake (Table A15). As the frequency and severity of extreme events is expected to increase, continued monitoring is necessary to better understand the dynamics of the system at large in the context of a changing environment through consistent monitoring (Li et al., 2024; Staniszewska et al., 2022).

Significant increases in concentrations of As and Cr (unfiltered) with increasing discharge suggests that the presence of these constituents in water is transport limited (Figure A5) (Bieroza et al., 2018; Knapp et al., 2020). In contrast, Ca and Mg (filtered) showed significant decreases in concentration with increasing discharge, signaling that these constituents are likely source-limited and that dilution is a key driver in their dissolved concentrations within the river (Figure A5) (Bieroza et al., 2018; Knapp et al., 2020). While not showing significant relationships with a relatively small number of data points, other metals commonly bound to organic matter and sediment (Cd, Co, Pb, Fe, Cu, Mn, etc.) could follow either trend, however more data is necessary to indicate their transport dynamics (Knapp et al., 2020; Li et al., 2024).

Wheaton River yields were below regional averages, represented by the Yukon and Mackenzie Rivers, for all metals except As. Large differences in As yield estimates between the Mackenzie and Yukon Rivers estimates could be attributed to factors such as underlying geology, mining activity, human alteration, climate, permafrost coverage and erosion dynamics (Wohl et al., 2022; Zolkos et al., 2020). Notably, regional comparisons for Cd were limited by lack of available literature and datasets. Given consistent differences in the behaviour and distribution of Cd and other metals of interest, this gap highlights the need for increased Cd monitoring in subarctic and arctic watersheds.

Conclusions and Significance

This study was co-developed as a part of a collaborative partnership drawing on perspectives from both Western Science and Indigenous Knowledge to conduct a multi-media, watershed-scale assessment of trace metal mobility across the Wheaton River Watershed. Mine-impacted sites were sampled for plant, scat, sediment and water samples to assess variability by region and media and compared to a site downstream to understand propagation through the watershed. The results suggest both region and metal-specific patterns of distribution and mobility. Sediments collected

at mine-impacted sites held the highest concentrations of all sample types collected. Proportions of readily bioavailable metals were unique to each site: Cd and Sb were more bioavailable at Skukum and Goddell, respectively, while other metals (As, Pb) were not preferentially taken up by biota at either site, and still others (THg, MeHg) showed no correlation between concentrations in sediment and concentrations in biota (Figure 9-11). Water samples had higher concentrations of metals at mine-impacted sites rather than downstream but were much lower than they were in sediments. Trace metal distributions in water at mine-impacted sites were consistent with distribution among solid samples, with sequential extraction results further supporting minimal mobility into water from samples collected. Unfiltered water concentrations of trace metals of interest at LW fell below guidelines for protection of aquatic life. Despite the presence of historical mine activity in the watershed, yields of all trace metals were consistent with the Yukon and Mackenzie River basins, suggesting either or both of low mobility from mine-impacted sites and significant dilution from the larger watershed. Sub-Arctic and alpine watersheds are, however, dynamic, and landscape changes in response to land use and climate change could alter trace metal mobility dynamics in the future. These results emphasize the sensitivity, connectivity and complexity of Sub-Arctic watersheds, and the potential for trace metal mobility given current and potential environmental change. This assessment provides important context for implementation of local land conservation initiatives by getting a first glance at the extent of contaminants as stressors on watershed health.

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Conflicts of Interest

The authors declare no conflicts of interest relevant to this study.

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Chapter 3: Thesis Conclusions

Key Scientific Takeaways

This work contributes to filling critical knowledge gaps by taking a holistic approach to examine trace metal mobility dynamics at a watershed-scale, informed by 1) community concerns and priorities, 2) current mobility and 3) potential mobility of metals stored in mine-impacted sediments. By drawing on interdisciplinary techniques informed by Western Science and traditional knowledge, the study aimed to address the research question: *What is the extent of current and potential trace metal mobility from mine-impacted sites on a watershed-scale in the Wheaton River Watershed?*

The results discussed demonstrated that while metals were largely contained within mine-impacted sediments in the catchment, their current bioavailability and conditions for future mobility are highly metal- and region-specific. Cd and Sb were more bioavailable at Skukum and Goddell, respectively, while As and Pb largely remained in sediments, and Hg and MeHg showing no strong correlations between concentrations in sediment and biota (Hall et al., 2005; St Louis et al., 2001). Notably, Cd concentrations in scat and vegetation were related by known dietary habits in woodland caribou and willow and validated by traditional knowledge (Gamberg, 1994; Gamberg et al., 2020). Trace metal distribution followed the same regional trends in water samples; however, at much lower concentrations, especially in the mainstem Wheaton River where concentrations were below water guidelines (Canadian Council of Ministers of the Environment (CCME), 1999). Limited mobility of metals from sediments into water agrees with sequential extraction results presented (Filgueiras et al., 2002; Kanianska et al., 2022). Neither dilution nor low mobility can be isolated as the primary driver of the results presented in this study due to the lack of upstream reference site.

Flux analyses and yield calculations indicated that extreme hydrological events play a crucial role in determining the Wheaton River's interaction with its watershed (Li et al., 2024; Staniszewska et al., 2022; Zolkos et al., 2020). Further, annual yields from the Wheaton River were comparable or below regional averages, the sensitive and dynamic nature emphasizes the need for ongoing assessment of changes in the context of changing environmental conditions (Li et al., 2024; Staniszewska et al., 2022; Zolkos et al., 2020). Differences in water chemistry within sampling

regions compliment traditional knowledge, by further supporting the suspected strong local influence on water chemistry and emphasizing the complex, sensitive nature of the catchment.

Lessons and findings from this work are directly applicable towards the implementation of objectives declared by the C/TFN Water Strategy, which include but are not limited to increased water quality monitoring within their Traditional Territory, and eventual development of water quality objectives, for which the Wheaton Watershed is a candidate. This assessment provides important context for implementation of local land conservation initiatives by getting a first glance at the extent of contaminants as stressors on watershed health. Better understanding the impact of legacy mining projects on metal distribution provides valuable perspectives to help inform assessment of future mining proposals and implement C/TFN rights to govern the waters and lands within their Traditional Territory detailed in their Final Agreement (2005). Further, this initial multi-media assessment provides valuable insights for informing future study design, questions, and more specific community research priorities.

Limitations

While the results presented provided valuable insight into trace metal mobility on a watershed-scale, this study presents several limitations. The biota collected, with the exception of bark, represents only a snapshot of trace metal uptake dynamics, preventing a robust assessment of temporal variability in uptake dynamics. Scat samples represent only a snapshot of animal diet, not accounting for metals that were absorbed during digestion. Although scat analysis presents a non-invasive method to assess potential metal exposure of local fauna, lack of species-specific studies accounting for digestive enrichment and uptake greatly limits broader application of these results into isotopic and food safety studies. Further, although some species were collected across all regions enabling direct comparisons; lack of replicate plant samples of all species collected in each region limits our understanding of regional variability of trace metal uptake by a given species. While this is limiting, obtaining replicates of each species was not expected given the inherent contrasting of high alpine and river valley environments in this study. Understanding uptake dynamics in these systems is also limited by lack of corresponding physicochemical data for underlying sediments that limits our understanding of both current uptake conditions and preventing assessment of potential for future change.

As previously discussed, sites sampled for this study represented sites of concern (mine-impacted) for C/TFN and a downstream site. Although the samples collected provide valuable insights into the scale of trace metal mobility on a watershed scale, sampling upstream of mine-impacted sites was not possible in 2025 as the upstream area of the Wheaton watershed is extremely remote, with more ‘accessible’ sites still requiring travel on (abandoned) alpine mining roads and multiple river crossings. While this was logistically impossible for this study, lack of samples collected upstream of the mines does limit our understanding of their impact on water quality both regionally and downstream. Pending changes in access or circumstance, upstream sampling in future years could strengthen study design by provide insights into the extent of mining impact on water chemistry within the catchment through supporting identification of drivers of change.

Several of the limitations in sample collection discussed also presented conceptual and analytical challenges while conducting data analysis. Given the unique dataset structure, challenges associated with data analysis and statistical testing under a WS lens were encountered throughout this study. More specifically, challenges surrounding unit differences for water and solid samples presented difficulties in analyzing water as a connector of ecosystems in which they flow. While C/TFN are strong advocates for the importance of water to human and non-human life within this ecosystem, the statistical nature and conventions of WS commonly restricted us to comparing like with like rather than what we know is connected. Further, opportunistic sampling led to an ‘unbalanced’ dataset, limiting statistical significance testing and use of commonly used sophisticated western data analysis methods without careful planning. While initially perceived as challenges, these circumstances emerged as an opportunity to question current criteria for significance of results, and how this might be best broadened when approaching science through lenses of both WS and IKS. I now recognize that to include perspectives from WS and IKS in research, both perspectives must be considered at all stages of research to generate accurate, locally relevant results that meaningfully address local priorities. That said, institutional and political barriers to broadening these standards must be considered to ensure that data generated and results shared are best suited to support implementation of policies and agreements benefitting community members. To do so, ongoing conversations with local rights holders should be facilitated to assess the scope of the current results with data collection and analysis methods used to date to identify any necessary changes to best satisfy community research priorities in the context of working governance frameworks.

Future Directions

This work was the first known assessment of its kind, and has been regarded as a valuable, meaningful way to conduct natural sciences research within C/TFN Traditional Territory. Building on the limitations discussed, using similar frameworks within the region at other sites would support the construction of a robust dataset grounded in community priorities and concerns that can be applied to environmental assessments.

While doing this work, some questions to guide potential future research were raised, including (1) *How do trace metal mobility and uptake dynamics change seasonally and between years?* ; (2) *How do trace metal mobility dynamics in the Wheaton River watershed differ from other catchments within the Bennett Lake watershed and across the Yukon River Headwaters?* ; (3) *To what extent are these metals entering the food web, and what are the potential implications for consumption guidelines of country foods (e.g. tea leaves, animal tissue)?*; (4) *How might these mobility trends change with ongoing environmental change, landslide activities, and potential resumed mining activity within the catchment?*

Beyond the local scope of this study, these results emphasize the value of taking an interdisciplinary approach when addressing research questions in complex, natural systems. While studies in the natural sciences typically limit sample collection to one or two media, this work emphasizes the value of a multi-media assessment when conducting ecosystem-scale research. Beyond its obvious local relevance, this work presents a progressive framework for the co-creation of contaminants research in the north, whose results and lessons learned could be applied to adjacent, multidisciplinary work beyond the local context.

Personal Reflections on the Research Process

My experience with this research has shown me that like a river, research, especially in the environmental sciences, is highly dynamic and non-linear. I feel lucky to have been deeply engaged in this research at all stages. This experience enforced that meaningful collaboration and partnerships are necessary and at the core of any successful research.

When I first began this work as a Master's student, I planned to collect lake profiles, sediment cores, and tributary samples to create flux models of Bennett and Tagish Lake catchments; however, logistical complications during fieldwork (low bridges, high waters, high winds) quickly

changed the circumstances for data collection, and so my project naturally changed with it. While I was initially surprised by how quickly and drastically my plans had changed, the result greatly enriched my learning. I became exposed to a vast variety of new lab methods, and drawing on concepts from analytical and environmental chemistry, geochemistry, biology, and limnology to analyze the dataset under an interdisciplinary lens. Further, I learned a great deal about what meaningful community-led research looks like. Having been involved in the project at all stages, I've experienced firsthand the challenges and celebrated the successes that have resulted from this work. Working through logistical challenges in the field, literature review, riding the steep learning curve of using analytical tools including RStudio and ArcGIS Pro, the iterative nature of data analysis, and implementing feedback from peers and co-authors has only grown my respect for the research process and what it takes to produce high-quality, interdisciplinary work. Through these experiences I have learned how to be a better listener, and as a result, details about the surrounding landscape and how people engage with it. The Wheaton River Watershed is far more than just a study site; they are living lands with deep cultural significance. As a result, I have learned that findings from this research are far more than a scientific finding, as they have potential to carry implications both in policy and personally for locals who live with and love the land.

Appendix A: Supplementary Methodology, Figures, and Tables

Supplementary Methodology

Section A1.

Trace Metal Analysis- Water Samples

Acidified samples were subsampled into 15 mL vials for analysis by ICP-MS. Unfiltered samples were shaken and left to settle briefly prior to subsampling to allow for both analysis of a representative sample and avoid damage to the instrument by limiting its introduction to large particulate matter. Blanks and standards were run in 10-sample intervals, and samples were corrected for drift and detection limits, listed by analyte in Table A2 .

Total Mercury

Samples were prepared for analysis, following EPA method 1631 (USEPA, 2002). 700 μL of bromine chloride (BrCl) was added to acidified water samples with slightly variable volumes to ensure excess BrCl is available to bind to Hg. Samples were left at room temperature overnight. The following day, 30 μL of hydroxylamine hydrochloride was added before shaking the sample to remove its yellow tint. Then, 60 μL of SnCl_2 was added to reduce Hg (II) to Hg (0). To prevent loss of Hg (0), each sample was closed tightly immediately following reagent addition. Samples were run on a Tekran 2600 alongside a series of blanks and standards in 10-sample intervals.

Section A2. Laboratory methods for digestion of solid samples

Total Digestion

Biota Samples

A modified method for total digestion related to EPA method 200.2 revision 2.8 was followed for digestion of organic samples (Mahar & Dawydiak, 2000; T. D. Martin et al., 1994). Approximately 200-300 mg of plant and 500-1000 mg of scat and reference materials (SRM 1515, BCR 483, SRM 2977) were added to a 50 mL Digtube and saturated with 2-4 mL of Milli-A water (resistivity of $18.2 \text{ M}\Omega\cdot\text{cm}$) until sample was saturated. 4 mL of 70% HNO_3 was added to each sample and heated to 100°C for 3.5 hours until only about 1 mL of liquid remained in each sample. Once

cooled to room temperature overnight, 2 mL of H₂O₂ was added dropwise to each sample and heated to 95 °C for 1 hour and then cooled to room temperature. Following digestion, samples were diluted and filtered to <0.45 µm for analysis by ICP-MS. Analyte-specific detection limits (Table A3) and reference material recoveries (Table A4) were reported.

Sediment Samples

USEPA method 3050b was followed for total digestion of sediment samples (U.S Environmental Protection Agency, 1996). Approximately 1g of each sediment sample and reference material (PACS-3, SRM 8704) added into a 50-mL Digtube, followed by 5 mL of milli-Q® and 5 mL of 70% HNO₃. Samples were refluxed at 95 °C for 10 minutes and cooled to room temperature. Then, 5 mL of 70% HNO₃ was added to each sample and refluxed at 95 °C for 30 minutes. Once cooled to room temperature again, 2 mL of Milli-Q and 3 mL of H₂O₂ were added, and samples were heated to 95 °C for 180 minutes, loosely covered. Digested samples were diluted and vacuum-filtered using 0.45 µm DigiFILTER (catalogue: 010-500-070) for analysis by ICP-MS, where blanks and blind standards were run in 10-sample intervals and eventually corrected for DRIFT. Analyte-specific detection limits (Table A3) and reference material recoveries (Table A4) were reported.

QA/QC

Following analysis by ICP-MS, dry weight concentrations of each metal were calculated using sample-specific dilution factors calculated based on weight of diluted solutions. All sample concentrations were above detection limits. The mean, range, standard deviation (SD) and relative standard deviation (RSD, %) for each reference material for all metals discussed (As, Ca, Cd, Co, Cr, Cu, Fe, Mg, Mn, Ni, Pb, Sb, Zn) are listed by metal in Table A4.

Section A3. Laboratory analyses for mercury analysis in solid samples.

Total Mercury

Samples were prepared for analysis by following EPA method 7473 (USEPA, 2007). Freeze-dried and homogenized solid samples were weighed into cooled ceramic boats, with 25-50 mg and 50-100 mg of samples used for organic and sediment samples, respectively. Samples were run in a direct mercury analyzer (DMA) alongside a series of reference materials run in 10-sample intervals.

5% of samples were run in duplicate. Recovery ranges, mean, standard deviation (SD) and relative standard deviation (RSD) for reference materials are listed in Table A5.

Methylmercury

Organic Samples

Samples were prepared in accordance with EPA 1630 methods and suggested Tekran 2700 procedures (USEPA, 1998). 50-150 mg of dry, homogenized sample was added to pre-weighed 8 mL vials (60940A-4), followed by an addition of 3 mL of 25% KOH in methanol (MeOH) solution. Samples were gently swirled and loosely covered ahead of heating to 65 °C for 16h. After digestion was complete and samples were cooled, digestates are diluted to 8 mL using MeOH and stored in the freezer (-20 °C). Prior to analysis on the Tekran 2700, 29.5 mL of Milli-Q and 500 µL of sodium acetate buffer were added to a 40 mL glass vial with a septum lid. Following the addition of 50 µL of sample and 30 µL of NaBEt₄, samples were run on a Tekran 2700 accompanied by a series of blanks, standards, and reference materials. Summary statistics for reference material recoveries are reported in Table A6.

Sediment Samples

Samples were prepared in accordance with EPA method 1630 and 5A-7, and suggested Tekran 2700 procedures (USEPA, 2007). A 5:1 ratio of acidic KBr and 1M CuSO₄ was added to pre-weighed sediments, followed by approximately 10 mL of DCM. After resting at room temperature for 1 h, samples were shaken at 330 rpm for 1h, followed by centrifugation for 15 minutes at 3500 rpm. 3-5 mL DCM was extracted from each sample into 8 mL scintillation vials with aluminum-lined caps. 2-3 mL of 0.1 mM sodium thiosulfate was added to scintillation vials and shaken for 1h at 330 rpm, followed by centrifugation at 3500 rpm for 15 mins. Following the extraction of sodium thiosulfate solution from the vial, samples were stored in plastic centrifuge tubes at -80 °C. Prior to analysis on the Tekran 2700, 29.5 mL of Milli-Q and 500 µL of sodium acetate buffer were added to a 40 mL glass vial with a septum lid. Following the addition of 50 µL of sample and 30 µL of NaBEt₄, samples were run by cold-vapour atomic fluorescence spectroscopy (CVAFS) on a Tekran 2700 accompanied by a series of blanks, standards, and reference materials. Summary statistics for reference samples are reported in Table A7.

Section A4. Laboratory methods for analysis of stable isotopes

Sample Preparation

A subsample of each prepared sediment sample was acidified prior to analysis of %C and $\delta^{13}\text{C}$ -POC to eliminate carbonates that might otherwise skew $\delta^{13}\text{C}$ -POC results. 500-1000 mg of sample was added to a 20 mL beaker and then placed on a hot plate heated to 50 °C. 10% HCl was added slowly drop by drop and beakers were filled to ~2/3 with HCl, except for tailings and discharge channels which were nearly filled with HCl, accounting for weakly alkaline tailings and potential buffering reactions. Samples were left on the hot plate for 8 hours at 50 °C. Samples were cooled and rinsed 5 times using Milli-Q until rinse was a neutral pH. Following rinsing, samples were heated in an oven at 50 °C until dry. Acidified dry weights were recorded to calculate the proportion of carbonates in each sample.

Given the inherent high variability in %C and N values among different matrices and a paucity of local reference values, %CN was analyzed on a representative set of samples across all matrices prior to isotopic analysis. Following freeze-drying and homogenization of all samples, 2-4 mg of organic samples were measured using a microbalance and packed into a tin capsule. Tungstic Oxide (WO_3) was added to all capsules to act as a combustion catalyst and binder in case resilient solids are present that would hinder combustion. Standards and ‘blind’ samples were run to assist in instrument calibration. Samples were analyzed using the Elementar Cube, where they were combusted at 1800 °C using oxygen gas, and resulting gases were carried by helium to be quantified following a series of adsorption traps to isolate gases.

Stable Isotope Ratios of C and N

Following %CN analysis, C:N ratios were calculated to assess necessary dilutions and inform isotope analyses. Carbon and nitrogen were analyzed on the same sample using the isotope ratio mass spectrometer (IRMS) if the C:N ratio was below 50:1, and separately if exceeding it. In this case, all sediment, bark, and caribou scat samples exceeded the target ratio, and were therefore analyzed separately for C and N using individual elemental target weights. Target weights were calculated based on %CN using the following equations:

$$N_{tar} = \frac{10}{\%N}$$

$$C_{tar} = \frac{5}{\%C}$$

When C and N were run together, the N equation was used to calculate the target weight. Weights of analyzed samples were all within 5% of the target weight. A series of standards, bypass samples, duplicates, and blanks were prepared for analyses for all target weights. For sediment samples, C and N were analyzed separately on acidified and unacidified samples, respectively. Samples were analyzed on a Delta Q-IRMS (Thermo). All isotopic analyses were completed at the Ján Veizer Stable Isotope Laboratory at the University of Ottawa.

Section A5. Methods used for sequential extraction analyses of sediment samples

Procedure

A five-step modified sequential extraction procedure was completed on all sediment samples. Approximately 1 g of sediment was added to a 50-mL polypropylene centrifuge tube. Methods used are described in writing below and summarized in Table A8.

Matrix Preparation

Prior to analysis of all extractants by ICP-MS, all samples were prepared to obtain a consistent matrix of 1-2% HNO₃. Samples from each fraction were placed on a hot plate heated to 120 °C and evaporated until dry. Once completely dry, a few mL of concentrated trace metal grade HNO₃ were added to digest residual salts on the hot plate until dry. 0.5 mL of HNO₃ was then added to dissolve any residual materials, followed by up to 500 µL of H₂O₂ to assist with dissolution as necessary. 2 mL of milli-Q® was then added to each Teflon bottle and transferred to a pre-weighed polypropylene 15-mL centrifuge tube after slightly warming the solution. milli-Q® was added to the Teflon bottle and then to the pre-weighed centrifuge tube to dilute the extractant until approximately 10 g of solution was created, and the exact weight was recorded. Following further dilutions, solutions were then analyzed by ICP-MS and ICP-OES, and dry weight analyte concentrations leached from sediments during each fraction were calculated for interpretation of results.

Data Analysis

For samples with duplicates, the mean of the leached concentration from each fraction was used for plotting analysis of results. Mean standard deviation and mean relative standard deviation are reported by constituent in Table A9. If leached concentrations exceeded those from the total acid digestion, concentrations from each fraction were scaled accordingly relative to the total concentration obtained from the total acid digestion.

Supplementary Figures

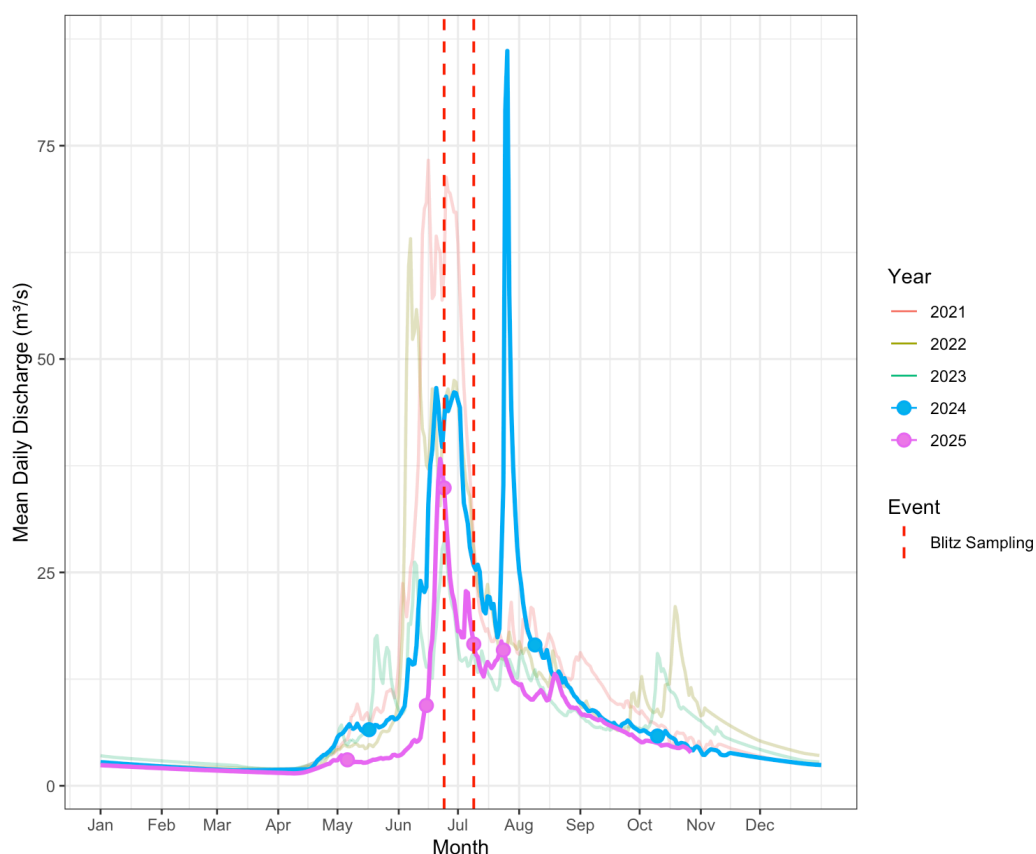


Figure A1. Wheaton River Discharge (Water Survey Canada, station ID 09AA012) from 2021-2025 with 2024/25 Lower Wheaton water sampling dates noted by points along the discharge record. Biota sampling dates (2025 only) are denoted by a vertical dashed red line.

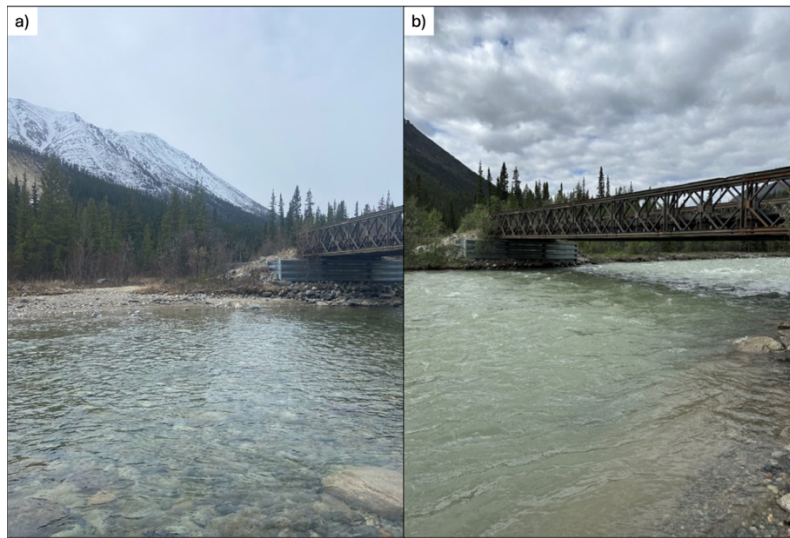


Figure A2. Images of the Lower Wheaton sampling site at the time of collection on a) May 5, 2025, and b) June 24, 2026.

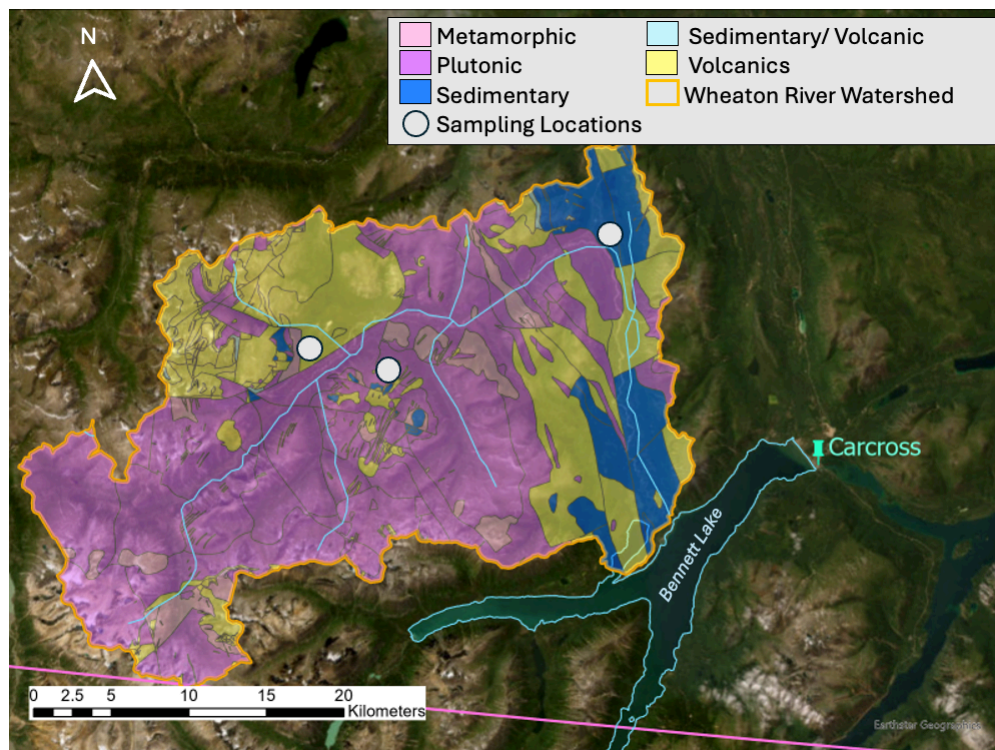


Figure A3. Bedrock geology of Wheaton River watershed, coloured by rock classification (Messenger et al., 2016; Yukon Government, 2023).

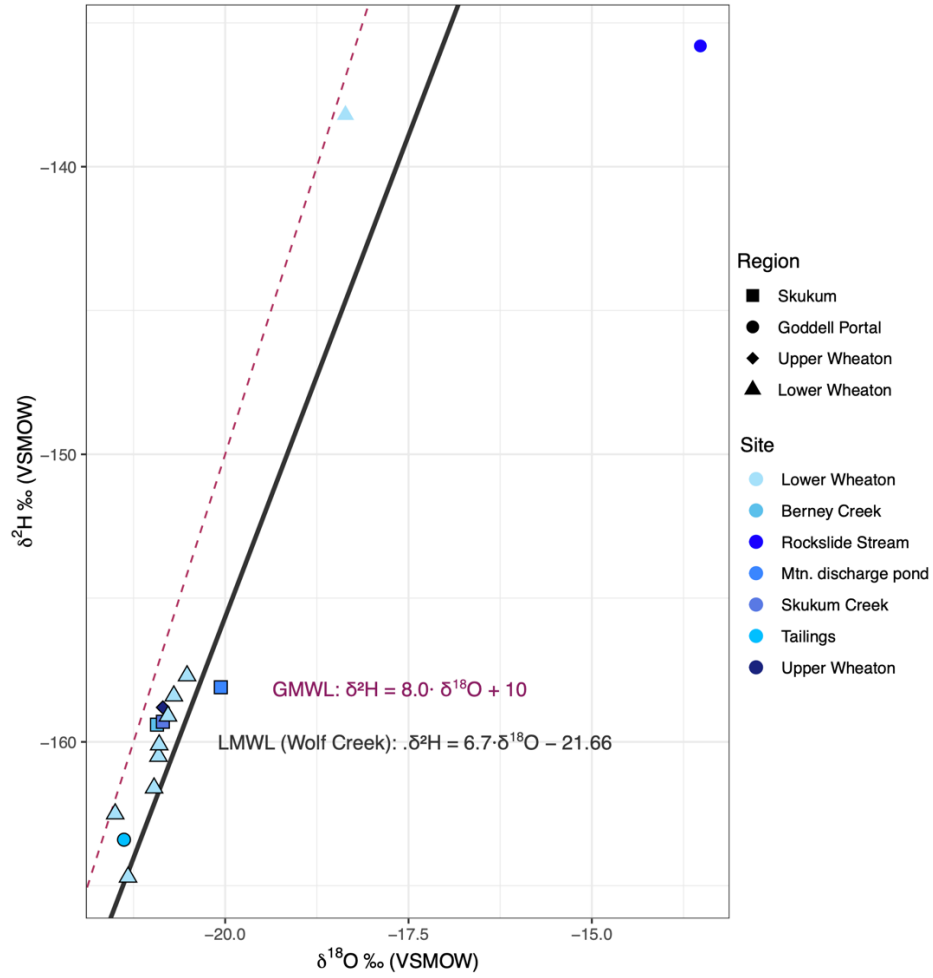


Figure A4. Isotopic compositions of water samples taken in 2024 ($n=3$) and 2025 ($n=12$) colored by site and shaped by region plotted in relation to the Global Meteoric Water Line (GMWL: $\delta^2\text{H} = 8 \delta^{18}\text{O} + 10$; (Craig, 1961)) and the Local Meteoric Water Line (LMWL: $\delta^2\text{H} = 6.7 \delta^{18}\text{O} - 21.66$; (Bourke, 2022)).

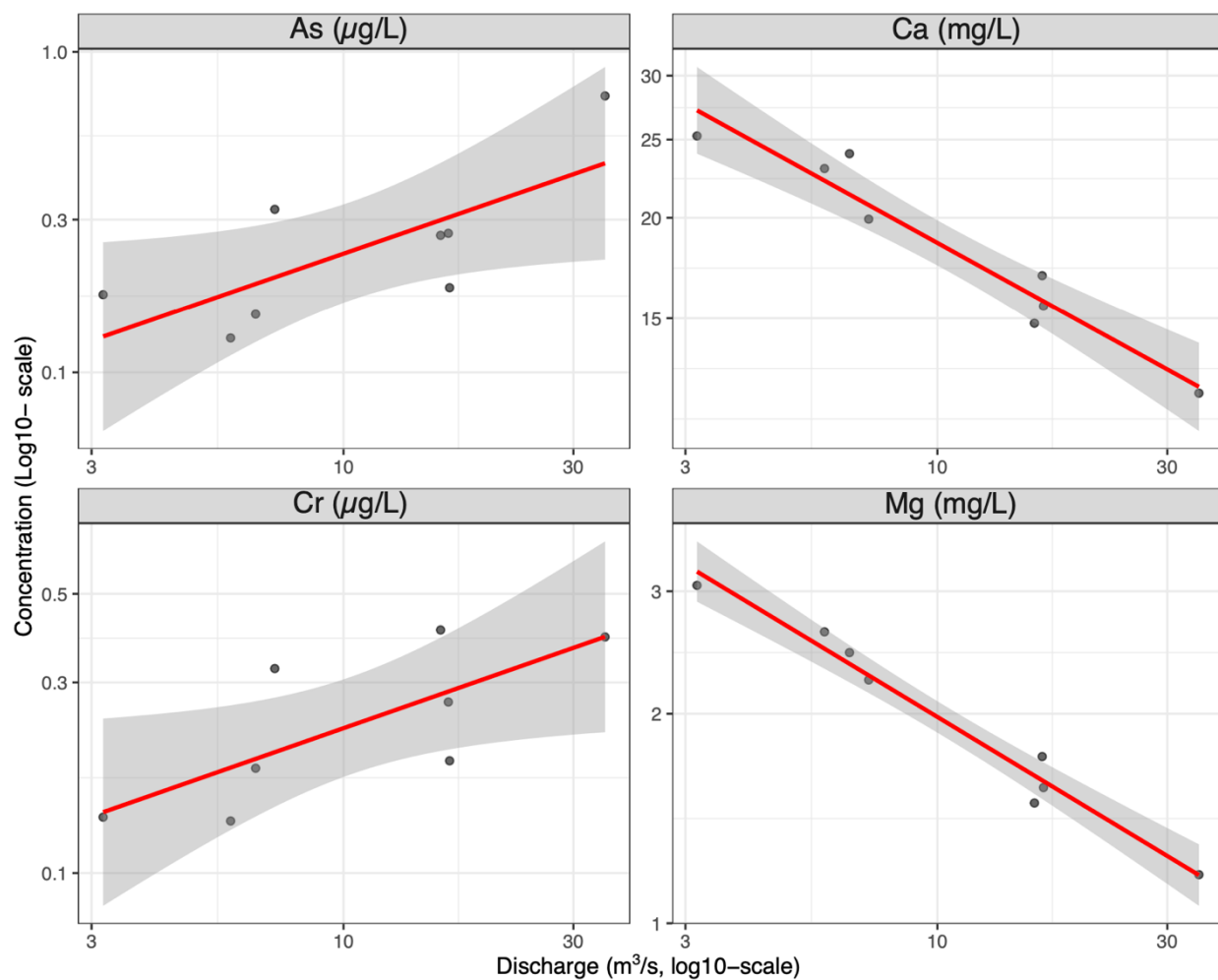


Figure A5. Log-concentration and Log-discharge for water quality constituents with significant relationships ($p < 0.05$). Please note concentration unit differences by constituent.

Supplementary Tables

Table A1. Frequency and media of samples collected at site of interest.

Site Abbrev.	Season			Water	Plant	Sediment	Scat
	Spring	Summer	Fall				
LW	X	X	X	X	X	X	
UW		X		X			
SM		X		X	X	X	X
GP		X		X	X	X	X

Table A2. Analytical methods and laboratories by chemical constituent in water samples, listing sample names, dates sampled, unit, instrument and labs where analysis was performed, instrument-specific detection limits and error.

Constituent	Name	Unit	Instrument/ Formula	Analytical Lab	Detection Limit
As	Arsenic	µg/L	Agilent Technologies 7700 series ICP-MS	LANSET	0.015
Ag	Silver	µg/L	Agilent Technologies 7700 series ICP-MS	LANSET	0.010
Ca	Calcium	mg/L	Agilent 5110 ICP-OES	UOGL	0.00015
Cd	Cadmium	µg/L	Agilent Technologies 7700 series ICP-MS	LANSET	0.003
Cl ⁻	Chlorine	mg/L	Dionex ICS-2100	UOGL	0.0177
Co	Cobalt	µg/L	Agilent Technologies 7700 series ICP-MS	LANSET	0.002
Cr	Chromium	µg/L	Agilent Technologies 7700 series ICP-MS	LANSET	0.016
Cu	Copper	µg/L	Agilent Technologies 7700 series ICP-MS	LANSET	0.009
DOC	Dissolved organic carbon	mg/L	OI Analytical TIC-TOC 1030 + DeltaPlus XP IRMS	JVSIL	0.1
$\delta^{13}\text{C}$ -DOC	Dissolved organic carbon isotopes	‰	OI Analytical TIC-TOC 1030 + DeltaPlus XP IRMS	JVSIL	-
$\delta^2\text{H}$ -H ₂ O	Hydrogen water isotopes	‰	Picarro Analyser L2130-i	JVSIL	-
$\delta^{18}\text{O}$ -H ₂ O	Oxygen water isotopes	‰	Picarro Analyser L2130-i	JVSIL	-
Fe	Iron	µg/L	Agilent Technologies 7700 series ICP-MS	LANSET	0.051
K	Potassium	mg/L	Agilent 5110 ICP-OES	UOGL	0.00524
Mn	Manganese	µg/L	Agilent Technologies 7700 series ICP-MS	LANSET	0.011
Mg	Magnesium	mg/L	Agilent 5110 ICP-OES	UOGL	0.00006
Na	Sodium	mg/L	Agilent 5110 ICP-OES	UOGL	0.00064
Ni	Nickel	µg/L	Agilent Technologies 7700 series ICP-MS	LANSET	0.052
NO ₃ + NO ₂	Nitrate and Nitrite	µg/L	Lachat QC8500 FIA Analyzer	BASL	2
Pb	Lead	µg/L	Agilent Technologies 7700 series ICP-MS	LANSET	0.005
PC	Particulate carbon	µg	Elementar Vario EL Cube Elemental Analyser	JVSIL	20
PIC	Particulate inorganic carbon	µg	PIC = PC - POC	-	-
POC	Particulate organic carbon	µg	Elementar Vario EL Cube Elemental Analyser	JVSIL	20

PN	Particulate nitrogen	µg	Elementar Vario EL Cube Elemental Analyser	JVSIL	25
Sb	Antimony	µg/L	Agilent Technologies 7700 series ICP-MS	LANSET	0.002
Se	Selenium	µg/L	Agilent Technologies 7700 series ICP-MS	LANSET	0.005
SO ₄ ²⁻	Sulfate	mg/L	Dionex ICS-2100	UOGL	0.016
THg	Total mercury	ng/L	Tekran 2600 CVAFS Sample Analysis System	LANSET	0.05
TP	Total phosphorous	µg/L	Lachat QC8500 FIA Analyzer	BASL	2
TDP	Total dissolved phosphorous	µg/L	Lachat QC8500 FIA Analyzer	BASL	2
TN	Total nitrogen	µg/L	TN = PN +TDN	-	-
TDN	Total dissolved nitrogen	µg/L	Lachat QC8500 FIA Analyzer	BASL	11
Zn	Zinc	µg/L	Agilent Technologies 7700 series ICP-MS	LANSET	0.065

BASL, Biogeochemical Analytical Service Laboratory in the Department of Biological Sciences at the University of Alberta (Edmonton, AB, Canada). JVSIL, Ján Veizer Stable Isotope Facility at the University of Ottawa (Ottawa, On, Canada). LANSET, Laboratory for the Analysis of Natural and Synthetic Environmental Toxins at the University of Ottawa (Ottawa, ON, Canada). UOGL, University of Ottawa Geochemistry Laboratory (Ottawa, ON, Canada).

Table A3. Analytical methods and laboratories by chemical constituent in solid samples, listing sample names, dates sampled, unit, instrument and labs where analysis was performed, instrument-specific detection limits and error.

Constituent	Name	Unit	Instrument	Analytical Lab	Detection Limit
As	Arsenic	µg/kg	Agilent 8800 QQQ	UOGL	0.041
Ca	Calcium	mg/kg	Agilent 5110 ICP-OES	UOGL	0.00015
Cd	Cadmium	µg/kg	Agilent 8800 QQQ	UOGL	0.001
Co	Cobalt	µg/kg	Agilent 8800 QQQ	UOGL	0.005
Cr	Chromium	µg/kg	Agilent 8800 QQQ	UOGL	0.012
Cu	Copper	µg/kg	Agilent 8800 QQQ	UOGL	0.005
d13C	Carbon isotopes	‰	Thermo Delta Q IRMS	JVSIL	-
d15N	Nitrogen Isotopes	‰	Thermo Delta Q IRMS	JVSIL	-
Fe	Iron	mg/kg	Agilent 5110 ICP-OES	UOGL	0.001
K	Potassium	mg/kg	Agilent 5110 ICP-OES	UOGL	0.006
MeHg	Methyl Mercury	ng/kg	Tekran 2700 Methyl Mercury Analyzer	LANSET	0.012
Mn	Manganese	µg/kg	Agilent 8800 QQQ	UOGL	0.001
Mg	Magnesium	mg/kg	Agilent 5110 ICP-OES	UOGL	0.001
Na	Sodium	mg/kg	Agilent 5110 ICP-OES	UOGL	0.001
Ni	Nickel	mg/kg	Agilent 8800 QQQ	UOGL	0.013
Pb	Lead	mg/kg	Agilent 8800 QQQ	UOGL	0.004
C	Mass of carbon	mg	Elementar Vario EL Cube Elementar Analyser	JVSIL	20
N	Mass of nitrogen	mg	Elementar Vario EL Cube Elementar Analyser	JVSIL	25
Sb	Antimony	µg/kg	Agilent 8800 QQQ	UOGL	0.002
THg	Total Mercury	ng/g	NIC MA-3000 Direct Mercury Analyzer	LANSET	0.002
Zn	Zinc	mg/kg	Agilent 5110 ICP-OES	UOGL	0.002

JVSIL, Ján Veizer Stable Isotope Facility at the University of Ottawa (Ottawa, On, Canada). LANSET, Laboratory for the Analysis of Natural and Synthetic Environmental Toxins at the University of Ottawa (Ottawa, ON, Canada). UOGL, University of Ottawa Geochemistry Laboratory (Ottawa, ON, Canada).

Table A4. Summary statistics of reference material recoveries following total acid digestion.

Reference Material	Mean % Recovery	Median % Recovery	Range % recovery	SD	RSD (%)
SRM 1515-apple (NIST)	96.9	93.2	69.5 – 129.5	16.1	16.7
BCR 483-lichen (JRC)	83.8	86.4	54.2 - 106.6	13.5	16.1
SRM 2977-mussel (NIST)	93.9	94.4	69.6 - 128.5	16.3	17.4
PACS-3 (NRC)	70.3	77.6	2.6 - 99.8	25.4	36.1
SRM 8704- Buffalo River Sed (NIST)	86.1	96.5	1.6 - 109.8	30.3	35.2

Table A5. Summary statistics of reference material recoveries for THg analysis.

Reference Material	Mean % Recovery	Range % recovery	SD	RSD (%)
BRC 483-lichen (JRC)	74.0	68.7 - 79.0	18.2	5.1
SRM 436-tuna (IAEA)	84.5	82.0 - 87.0	148.8	4.2
SRM 2704- Buffalo River Sediment	95.5	92.1 - 98.8	67.9	4.9

Table A6. Summary statistics for reference materials used in MeHg analysis of biota, including mean, range, standard deviation (SD) and relative standard deviation (RSD).

Reference Material	Mean % Recovery	Range % recovery	SD	RSD (%)
SRM 1566b-oyster (NIST)	91.4516	88.8505 – 94.0526	3.6784	4.0223
IAEA-476 (IAEA)	89.9070	86.3532 – 93.4608	5.0258	5.5900
IAEA-436 (IAEA)	98.3959	98.1899 – 98.6020	0.2914	0.2962

Table A7. Summary statistics for reference materials used in MeHg analysis in sediment, including mean, range, standard deviation (SD) and relative standard deviation (RSD).

Reference Material	Mean % Recovery	Range % recovery	SD	RSD (%)
SQC-1238	116.8770	102.0674 – 131.6866	20.9440	17.9197
ERM-CC580	110.2128	108.3459 – 112.0796	2.6402	2.3955

Table A8. Summarized methods for 5-step sequential extraction.

Fraction	Reagent Used	Procedure	Extractant Preparation
<i>Water-Soluble</i>	10 mL of Milli-Q water (resistivity of 18.2 M Ω ·cm)	Shake for 1h at room temperature (RT)	Consistent across all fractions. Following shaking, the extractant is decanted into an acid-washed Teflon bottle following centrifugation at 2150 rpm for 20 minutes. The residual is rinsed with 5-mL of Milli-Q to remove all extractant and decanted into sample specific bottles.
<i>Exchangeable</i>	10 mL of 1M NH ₄ Cl	Shake for 5h at RT	
<i>Acid-Soluble</i>	30 mL of 0.11 M Acetic Acid	Shake for 16h at RT	
<i>Reducible</i>	40 mL of 0.5 M NH ₂ OH HCl in a matrix of 0.05M HNO ₃	Shake for 16h at (RT)	
<i>Oxidizable</i>	10 mL of 30% w/w H ₂ O ₂ and 30 mL of 1M NH ₄ OAc	Add H ₂ O ₂ and, digest for 1h at RT, heat dry bath at 85 °C until the reaction is completed and until ~ 1 mL of sample remains. Once cooled, shake with NH ₄ OAc for 16 h at RT.	

Table A9. Summary statistics for samples by media and metal. Concentrations reported are in ug/g dry wt, except for THg which are in ng/g and marked by *.

Metal	Media	Mean	Median	Range
<i>As</i>	Plant	0.571	0.107	0.020 – 4.61
	Scat	9.76	6.04	0.157 – 25.9
	Sediment	290	224	3.27 - 780
<i>Cd</i>	Plant	0.629	0.227	0.005 – 3.47
	Scat	3.98	1.66	0.275 – 17.4
	Sediment	1.55	1.28	0.130 – 3.65
<i>Co</i>	Plant	0.303	0.138	0.024 – 2.16
	Scat	0.690	0.428	0.188 – 1.96
	Sediment	9.31	10.0	3.73 – 14.7
<i>Cr</i>	Plant	6.19	3.04	1.35 – 62.8
	Scat	1.68	1.34	0.950 – 4.02
	Sediment	8.46	6.89	4.38 – 16.5
<i>Cu</i>	Plant	6.71	6.47	2.29-15.3
	Scat	13.9	12.8	3.55 – 27.0
	Sediment	22.2	24.6	6.71 – 39.6
<i>Fe</i>	Plant	198	109	18.4 - 1189
	Scat	1136	727	88.0 - 4340
	Sediment	20 405	17 909	11 299 – 37 303
<i>Mn</i>	Plant	91.8	51.1	14.3 - 448
	Scat	442	254	52.2 - 1646
	Sediment	921	1010	264 - 1395
<i>Ni</i>	Plant	3.19	1.44	0.232 – 43.2
	Scat	3.09	1.22	0.629 – 12.9
	Sediment	20.0	12.2	3.96 – 61.4
<i>Pb</i>	Plant	0.559	0.104	0.006 – 6.79
	Scat	2.77	2.50	0.228 – 5.47
	Sediment	47.4	30.0	6.65 - 111
<i>THg*</i>	Plant	2.48	1.29	0.070 – 23.0
	Scat	41.2	15.8	4.50 - 113
	Sediment	624	496	1.51 - 1519
<i>Sb</i>	Plant	0.108	0.007	0.002 – 0.932
	Scat	1.83	1.17	0.005 – 4.84
	Sediment	0.589	0.776	0.044 – 1.07
<i>Zn</i>	Plant	61.5	34.4	13.0 - 293
	Scat	187	155	41.6 - 357
	Sediment	117	122	41.2 - 186
<i>MeHg</i>	Plant	0.283	0.179	0.001 - 1.646
	Scat	1.924	0.615	0.028 – 11.587
	Sediment	0.319	0.115	0.047 – 0.841

*Table A10. Summary statistics for water samples by metal. Concentrations are reported in µg/L, except for THg which are reported in ng/L and marked by *.*

Metal	Mean	Median	Range
<i>As</i>	5.62	0.272	0.108 – 52.5
<i>Cd</i>	0.042	0.012	0.002 – 0.232
<i>Co</i>	0.126	0.039	0.001 – 0.941
<i>Cr</i>	0.217	0.191	0.109 – 0.406
<i>Cu</i>	0.650	0.399	0.196 – 2.48
<i>Fe</i>	199	101	12.5 - 1058
<i>Mn</i>	23.5	6.44	0.502 - 171
<i>Ni</i>	0.324	0.18	0.026 – 1.37
<i>Pb</i>	0.748	0.122	0.02 – 7.73
<i>THg *</i>	6.14	0.655	0.122 – 62.7
<i>Sb</i>	1.57	0.207	0.048 – 16.1
<i>Zn</i>	5.54	1.75	0.249 – 47.7

Table A11. Mean SD and RSD of duplicate leachant concentrations for each fraction, reported by metal.

Metal	Mean SD	Mean RSD (%)
As	0.426	24.641
Cd	0.006	14.000
Pb	0.304	18.427
Zn	0.540	11.478
Mg	24.215	7.503
Fe	44.126	22.704
Mn	7.645	9.820
Sb	0.025	18.792
Cu	0.187	33.122
Co	0.061	21.240
Cr	0.023	9.085
Ni	0.125	11.658
Ca	208.558	15.054

Table A12. Summary statistics for flux models listed by constituent. Models were created in R and assessed for fitness using Akaike Information Criterion (AIC), R-squared (R²), percent flux bias (Bp), partial load ratio (PLR), and Nash Sutcliffe efficiency index (E).

Analyte	Number of observations	auto-selected model	model AIC	R ²	Bp	PLR	E
THg	8	6	10.54	97.39	-6.064	0.9394	0.9856
V	8	1	18.89	79.82	-9.842	0.9016	0.7948
Cr	8	1	8.409	93.01	1.526	1.015	0.9428
Mn	8	1	22.13	78.74	-6.113	0.9389	0.8614
Fe	8	4	22.07	89.42	-10.92	0.8908	0.7935
Co	8	6	21.69	95.24	-6.252	0.9375	0.9136
Ni	5	2	16.79	86.37	8.852	1.089	0.491
Cu	8	4	15.94	87.28	-3.735	0.9626	0.8619
Zn	8	6	17.09	94.26	-2.61	0.9739	0.9136
As	8	2	10.63	93.88	-2.961	0.9704	0.9774
Se	3	1	3.659	72.74	0.1066	1.001	0.8002
Cd	8	1	22.3	66.51	-14.99	0.8501	0.6638
Sb	8	6	-	98.58	-0.2061	0.9979	0.9853
			5.5991				
Ag	5	1	10.93	73.16	1.119	1.011	0.9059
Pb	8	6	24.46	90.27	-6.21	0.9379	0.8801
SO4	5	4	-23.54	99.9	0.002832	1	0.9988
Cl	5	4	-	98.54	-0.7219	0.9928	0.9888
			5.2973				
Ca	8	3	-33.52	99.86	0.04402	1	0.9983
K	8	7	-	99.99	-0.03337	0.9997	0.9999
			50.346				
Mg	8	3	-26.85	99.58	-0.5789	0.9994	0.9957
Na	8	5	15.6	85.19	7.113	1.071	0.9223
TP	8	1	24.9	80.09	-2.613	0.9739	0.8943
TDN	8	4	2.744	93.85	-0.5332	0.9947	0.9792
DOC	8	4	12.96	87.77	-0.5664	0.9943	0.772

Table A13. Summary statistics for water samples collected from the Annie Lake Rd. sampling site within the Lower Wheaton region. All concentrations are reported in µg/L, except for THg which is reported in ng/L and marked by *.

Metal	Mean	Std. Dev.	Mean % filtered
As	0.201	0.077	65.3
Cd	0.014	0.013	54.0
Co	0.062	0.070	27.1
Cr	0.244	0.107	69.9
Cu	0.458	0.253	78.7
Fe	171.745	175.379	12.9
Mn	8.221	8.194	32.5
Ni	0.110	0.117	60.9
Pb	0.268	0.299	22.8
Sb	0.201	0.033	95.9
THg *	0.901	0.864	-
Zn	1.609	1.167	63.3

Table A14. Water and sediment quality guidelines reported by CCME (Canadian Council of Ministers of the Environment (CCME), 1999). Water quality guidelines marked by * indicate that guideline concentrations are calculated relative to hardness (Pearson, 2026). Values are reported for a hardness of 55 mg/L (420 mg/L) to account for differences between all other water samples and tailings. ** is marked by values that are calculated as a function of pH (average), DOC (average) and hardness. Values were reported for average of samples (tailings) to account for physiochemical differences.

Metal	Long-Term guideline concentration in freshwater for the protection of aquatic life (CCME) (ug/L)	Interim Sediment Quality Guidelines for the Protection of Aquatic Life (ug/kg dry wt.)	Probable effect level concentrations in freshwater sediments (ug/kg dry wt)
As	5	5900	17 000
Cd	0.1 (0.37) *	600	3500
Cr	-	37 300	90 000
Co	-	-	-
Cu	2 (4) *	35 700	197 000
Fe	300	-	-
Mn	390**	-	-
Ni	25 (150) *	-	-
Pb	1 (7) *	35 000	91 300
THg	0.026	170	486
MeHg	0.004	-	-
Sb	-	-	-
Zn	10 (36) **	123 000	315 000

Table A15. Annual combined fluxes (kg or ton/year) of major ions, trace metals, contaminants, and nutrients from samples taken near gauged site along the Wheaton River for the open water season (OWS; April-November). Values modified to more accurately reflect load estimates during the atmospheric river event are marked by *. Note unit differences by constituent.

		OWS		Mean FWC	
		2024	2025	2024	2025
Water (km ³)		0.040	0.010	0.040	0.010
Major Ions		(ton/yr)		(mg/L)	
	Ca	46878.3 ± 11.5	2528.0 ± 5.5	17.1 ± 0.5	17.2 ± 0.5
	Cl	-	13155.9 ± 223	-	0.1 ± 0.02
	K	195.7 ± 0.2	964.0 ± 0.1	0.7 ± 0.008	0.7 ± 0.007
	Mg	468.0 ± 1.6	275.6 ± 0.9	1.7 ± 0.1	1.9 ± 0.1
	Na	1442.3 ± 4354.9	227.7 ± 28.4	5.3 ± 27.6	1.6 ± 1.6
	SO4	-	2269.4 ± 11.1	-	15.5 ± 0.9
Trace Metals		(kg/yr)		(µg/L) ** (ng/L)	
	As	150.4 ± 49.5	42.7 ± 2.9	0.5 ± 0.5	0.3 ± 0.1
	Cd	4.9 ± 0.7	2.3 ± 0.3	0.02 ± 0.02	0.02 ± 0.02
	Co	39.9 ± 26.3 *	10.6 ± 3.4	0.1 ± 0.3	0.07 ± 0.2
	Cr	91.3 ± 4.7	38.8 ± 1.7	0.3 ± 0.1	0.3 ± 0.1
	Cu	133.0 ± 11.9	67.5 ± 5.1	0.5 ± 0.4	0.5 ± 0.3
	Fe	62858.3 ± 12926.8	28546.1 ± 4379.6	230.1 ± 313.0	195.3 ± 211.6
	Mn	3897.0 ± 701.4	1413.2 ± 196.3	14.3 ± 16.6	1.0 ± 1.0
	Ni	-	29.7 ± 5.1	-	0.2 ± 0.3
	Pb	165.5 ± 253.0 *	46.3 ± 61.4	0.6 ± 2.2	0.3 ± 2.0
	Sb	52.8 ± 1.5	28.8 ± 0.5	0.2 ± 0.04	0.2 ± 0.04
	THg	0.5 ± 0.2	0.1 ± 0.01	**1.7 ± 2.1	**0.8 ± 0.5
	Zn	1134.7 ± 435.6 *	324.7 ± 62.0	4.2 ± 5.0	2.2 ± 3.4
	Ag	-	0.9 ± 0.05	-	0.006 ± 0.004
	Se	29.4 ± 1.6	-	0.1 ± 0.06	-
Nutrients		(ton/yr)		(mg/L)	
	DOC	173.7 ± 10.0	111.2 ± 5.7	0.6 ± 0.4	0.8 ± 0.4
	TP	4.4 ± 1.4	1.3 ± 0.3	0.02 ± 0.03	0.009 ± 0.01

TDN	26.6 ± 0.8	12.3 ± 0.3	0.1 ± 0.03	0.08 ± 0.02
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*Table A16. Approximate percentage of carbonate composition (by weight) within each sediment sample. Values were calculated using sample mass pre and post acidification with 10% HCl. Samples marked with * indicate potential errors associated with loss of sample during acidification procedures.*

Region	Sample	Approximate % carbonate by weight
Skukum	S_mtndischansed	7.5 *
Goddell Portal	GP_outwashsed	9.5
Goddell Portal	GP_dischansed	6.5
Goddell Portal	GP_tailings	12.2
Lower Wheaton	LW_riversed	3.8

Appendix B: Results Summary Handout for Community

See following pages for front and back of the handout for local community partners summarizing results from the study.

Metal Mobility in the Wheaton Watershed

What are trace metals?

- Occur in the environment naturally or from human activities
- Some are required in small amounts by living things, but higher concentrations can be toxic
- Some are toxic to living things in any amount (ex. arsenic, antimony, cadmium, lead, mercury)
- Metals do not degrade, but they can transform in different environmental conditions to move between water, sediment, and wildlife

When can metals enter the environment?

- Most metals in the environment are stored in sediment, but their ability to be taken up by plants and animals depends on what they are attached to (ex. geology, soil chemistry)
- In sediments, metals can be:

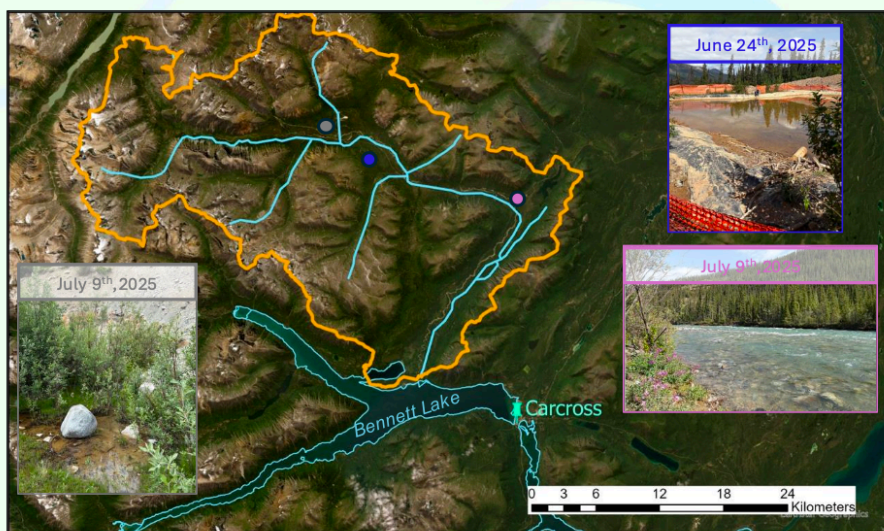
- 1** Ready to enter the environment
- 2** Currently stuck, but could be released later
- 3** Stuck in place

- Different sediments within the watershed hold different amounts of metals and have different ways of storing them.

Sampling

Sediment samples were collected from:

- 1. Skukum Mtn.** by tailings discharge pond
- 2. Goddell Portal** a) tailings; b) tailings discharge channel; c) other sediment nearby the portal.
- 3. Wheaton Riverbed** at Annie Lake Rd. Bridge



At these sites, nearby scat, plant, and water samples were collected to see how metals are moving throughout and beyond mine-impacted sites.

Samples collected include:

Scat

- ✓ Bear
- ✓ Caribou
- ✓ Sheep
- ✓ Grouse
- ✓ Snowshoe Hare
- ✓ Canine

Plant

- ✓ Soapberry (berry + leaf)
- ✓ Willow (bark + leaf)
- ✓ Yarrow
- ✓ Fireweed (stalk + leaf)
- ✓ Spruce tips
- ✓ And many more!

Water

- ✓ Skukum Mtn. tailings discharge pond
- ✓ Skukum Creek
- ✓ Berney Creek
- ✓ Tailings
- ✓ Wheaton River at Annie Lake Rd.

