Trace elements in lake catchments surrounding Iqaluit, Baffin Island, NU

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Abstract

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In this study, twenty lake catchments surrounding Iqaluit, Baffin Island, were assessed for trace element concentrations (Al, As, Ba, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, THg, V, Zn) in lake water, catchment soil, lake sediment, and moss (*Hylocomium splendens*). Additionally, the sources of each trace element were determined through the positive matrix factorization (PMF) model and enrichment factors (only in moss). Geogenic trace elements had the highest median concentrations (Fe>Al>Mn) throughout the study media and across the study catchments. Anthropogenic trace elements had the lowest median concentrations (Pb>As>Cd>THg) across the lake catchments, which were generally several orders of magnitude lower compared to geogenic elements. The PMF model identified trace elements associated with geogenic or anthropogenic sources, i.e., THg (47%), Cd (40%), Zn (34%), and Al (27%) were identified as originating from industrial emission sources deposited onto lakes because they accounted for a large proportion within the model.

Keywords: Trace elements, Arctic lake catchments, water chemistry, biomonitoring, positive matrix factorization (PMF) model, enrichment factors

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List of Acronyms

Al	Aluminum
NH_3	Ammonia
Sb	Antimony
AMAP	Arctic Monitoring and Assessment Programme
As	Arsenic
Ba	Barium
BD	Bulk Density
Ca^{2+}	Calcium
CAPMoN	Canadian Air and Precipitation Monitoring Network
Cd	Cadmium
Cl^-	Chloride
Cr	Chromium
Cu	Copper
DOC	Dissolved Organic Carbon
DIC	Dissolved Inorganic Carbon
EF	Enrichment Factor
Fl^-	Fluoride
H_2O_2	Hydrogen Peroxide
HCl	Hydrochloric Acid
ICP	International Cooperative Programme on Effects of Air Pollution on Natural Vegetation Crops
Fe	Iron
Pb	Lead
LOI	Loss on Ignition
Mg^{2+}	Magnesium
Mn	Manganese
Hg	Mercury
NAPS	National Air Pollution Surveillance
Ni	Nickel
NO_3^-	Nitrate
HNO_3	Nitric Acid
NMAD	Normalized Median Absolute Deviation
NCP	Northern Contaminants Program
OM	Organic Matter
Pg	Paleoproterozoic Cumberland Batholith
PM	Particulate Matter
PSA	Particle Size Analysis
PO_4^{3-}	Phosphate

POPs	Persistent Organic Pollutants
K^+	Potassium
PCA	Principal Component Analysis
PC	Principial Component
PMF	Positive Matrix Factorization Model
RPD	Relative Percent Difference
Sd	Sediment
Se	Selenium
Ss	Soil
SOM	Soil Organic Matter
SiO_2	Silicon dioxide
SO_4^{2-}	Sulphate
TDN	Total Dissolved Nitrogen
THg	Total Mercury
TP	Total Phosphorus
Tb	Till Blanket
Tv	Till Veneer
V	Vanadium
Zn	Zinc

1 Introduction

1.1 Trace Metals in the Environment

Trace elements[1], or trace metals as they are commonly referred to, play a key role in many ecological and environmental processes. For example, some trace elements are micronutrients (e.g., zinc, copper, and iron), which are required in small quantities, playing an important role in maintaining the integrity of various chemical, physiological, and metabolic processes in humans and plants (Bhattacharya, 2016). In contrast, other elements, (e.g., mercury, cadmium, and lead) have no known biological function and are toxic in small concentrations (Peñarroya, 2011). Trace elements exist throughout the natural environment in aqueous (seawater and freshwater), geologic (minerals and rocks), and atmospheric (troposphere and stratosphere) compartments (Robbins et al., 2019). Naturally occurring sources include: geogenic (i.e., soil-derived dusts, volcanic emissions, mineral weathering, soil erosion and leaching, and sediment suspension), forest fires, sea salt aerosols, metal corrosion, and metal evaporation from water resources (Nriagu, 1989; Shallari et al., 1998; Fergusson, 1990; Bradl, 2002; He et al., 2005; Tchounwou et al., 2012; Halbach et al., 2017). The principal release pathway into the environment is through abiotic and biotic mineral weathering processes (breakdown of rock and soil) and anthropogenic activities including mining, fossil fuel combustion, and industrial emissions; Peñarroya, 2011).

[1]The term trace element refers to elements that exist in trace amounts (aqueous between 1 fM (femtometre) to 10 μM and geologic <0.1% by weight), in natural and disturbed environments, with excess bioavailability negatively impacting living organisms (Bhattacharya et al., 2016). Trace elements encompasses metals, metalloids, and radionuclides.

Trace elements are recognized as potential pollutants in the environment due to their toxicity, bioaccumulation potential, and persistence (Kabata-Pendias and Pendias, 2001; Peñarroya, 2011). Their bioavailability is influenced by physical, chemical, and biological factors. More specifically, physical factors, such as temperature, phase association (i.e., dissolved, non-dissolved (humic substances), particulate (clays, metal oxides)), adsorption, and sequestration contribute to their availability. Furthermore, chemical factors influencing bioavailability include speciation, complexation kinetics, lipid solubility, and water partition coefficients (hydrophobicity; Hamelink et al., 1994). Lastly, biological factors include species characteristics, trophic interactions, and biochemical and physiological adaptations (Verkleji, 1993; Tchounwou et al., 2012;).

Physical and chemical processes can transform elements from one species to another and can convert them from inorganic to organic forms, as well as soluble and insoluble forms (Peñarroya, 2011). The toxicity of a trace element depends on its concentration in the environment, where it's found (i.e., water, sediment, soil, air), the source (i.e., mining, industry, natural rock breakdown, and agriculture), pH, temperature, redox potential and whether the element exists by itself or as part of a larger chemical compound (Morel and Price, 2003; Gamberg et al., 2005;Loureiro and Hepp, 2020). Further, factors that affect element mobility (either increasing or decreasing) in the environment can vary over time and among micro-organisms, plants, and animals (Leventhal and Taylor, 1995). For example, development of acidic or oxidizing conditions can release large amounts of arsenic into waters from sediment, due to decreased sorption capacity (Léonard, 1991).

Although trace elements occur naturally throughout the environment, they have been transferred to environmental compartments at greater rates and distances due to human activities (Lau et al., 1998; AMAP, 2005; Aras et al., 2012), leading to elevated concentrations that can negatively impact humans and wildlife. Anthropogenic sources of trace elements include: mining and smelting operations, domestic and agricultural use of metals (i.e., lead, arsenic, cadmium, chromium, and copper) and metal-containing compounds in inorganic fertilizers, phosphate fertilizers, and fungicides (Shallari et al., 1998; He et al., 2005; Tchounwou et al., 2012; Srivastava et al., 2017), metal processing in refineries, coal burning, petroleum combustion, nuclear power plants and high tension lines, plastics, textiles, microelectronics, wood preservation, shipping emissions, transportation (automobiles and shipping), and paper processing plants (Pacyna et al., 1996; Arruti et al., 2010; Tchounwou et al., 2012).

The main pathways of trace elements into the environment include atmospheric emissions and transport (deposition and re-emission) and oceanic currents to remote regions, such as the Canadian Arctic. The atmosphere provides a vital link between southern and northern latitudes, by acting as a vector and carrying pollutants (i.e., particulate matter and trace elements) to remote northern regions. Specifically, atmospheric deposition can provide a constant pathway (by continuous transport) of anthropogenic sourced trace elements to northern environments, which could potentially cause deleterious effects. Local sources of pollution (e.g., shipping, domestic heating and power generation) can also affect the atmospheric composition at regional and Arctic-wide scales (Law et al., 2017; Schmale et al., 2018), with little research focusing on the proportion of long-range atmospheric transport of contaminants. However, studies

(Benson, 1969; Tran and Molders, 2011; Schmale et al., 2018;) in northern cities (e.g., Fairbanks, Prudhoe Bay, and Utqiagvik (Alaska), Norilsk and Murmansk (northern Russia)), have found that wintertime pollution is more prominent from local sources (compared to the summertime), as there are more heating and electricity requirements.

1.2 Positive Matrix Factorization (PMF) Model

Establishing element sources is useful in differentiating between geogenic and anthropogenic contributions within a given area (Wu et al., 2021). This information is helpful in developing management and mitigation plans, updating air monitoring programs and policies, and increasing knowledge on element loadings and deposition rates within the Arctic region. Receptor models have been widely used for source apportionment (Thurston et al., 2005; Watson et al., 2008). The most common receptor models can be categorized into univariate models (chemical mass balance) and multivariate models (principal component analysis (PCA) and positive matrix factorization (PMF), which incorporate non-negative (positive) results on the output, frequently resulting in a more physically interpretable result (Dutton et al., 2010). Each model has its advantages and disadvantages, but all rely on the basic principles of mass balance (Dutton et al., 2010). In comparison to the other models, PMF is well suited to multi-year datasets, dealing with both high and low concentration data (Bzdusek et al., 2006). PMF also incorporates individual measurement uncertainties into the model framework, making it a good choice for source apportionment datasets (Dutton et al., 2009a; Dutton et al., 2009b).

The PMF model has been widely used in source apportionment studies (Wu et al., 2021; Zhuang et al., 2019; Christensen et al., 2018) to interpret aerosol, water, soil, sediment, moss, and lichen elemental composition data (Paatero and Tapper, 1994;

Jaeckels et al., 2007; Brown et al., 2015; Guan et al., 2018; Isely et al., 2018; Landis et al., 2019; Isely and Tayolor, 2020). PMF is a type of factor analysis, whereby it reduces large datasets into smaller groups or factors that share commonalities (see section 2.5.4). The usefulness of the model is in how it interprets the data- it shows the amount of source contributions per element in each factor, and source types based on the data input into the model (EPA, 2005). The model is often used to characterize sources from populated regions, and little research has been used to understand sources within the Canadian Arctic. Nonetheless, it is important to differentiate sources of trace elements to better understand potential impacts to the environment and the health of wildlife and northern communities, as many rely on subsistence lifestyles.

1.3 Atmospheric Transport and Deposition

Long-range atmospheric transport of trace elements has been well documented (Arimoto et al., 1987; Dick, 1991; Lee and Lehmden, 1973; Rahn et al., 1981; Rahn and Lowenthal, 1984; Wolff et al., 1999), with studies showing the distribution patterns of trace elements (i.e., depending on particle size and metal solubility) to the Arctic and Antarctic and throughout the globe (AMAP, 2005). Trace metals that are on particles of anthropogenic or marine origin are more soluble than crustal particles (Desboeufs et al., 2005; Giusti et al., 2003; Laing et al., 2014). Fine particulate matter (PM <2.5 μm) from mining, industry, transportation, and agriculture contribute heavily to the transportation of trace elements, by creating metal-particulate complexes through the process of adsorption (Zhang et al., 2018). Once in the atmosphere, gases, particles, and elements can undergo physical and chemical transformation during transport and deposition (Pacyna, 2008). Depending on the region, different elements will be prominent in the atmosphere. For example, historically lead (Pb) was mostly concentrated in areas with

high traffic as a result of additives in fuel, similarly, vanadium (V) is emitted through fossil fuel combustion, particularly residual fuel oils (Wang et al., 2013; Gad, 2005), and arsenic (As) is found in the atmosphere near ceramic industries (Tan et al., 2014; Sahu and Basti, 2020). Moreover, atmospheric chromium (Cr) is common in areas with mining activities, copper (Cu), zinc (Zn) and cadmium (Cd) can be emitted from biomass burning (Fang et al., 2015), and mercury (Hg) is a product of coal burning (Feng and Qiu, 2008; Sahu and Basti, 2020).

The processes of wet and dry atmospheric deposition refer to the fallout of precipitation (i.e., rain, snow, fog), particles or aerosols, and gases to the earth's surface (i.e., land, water). Various meteorological (i.e., wind speed, clouds), hydrological (i.e., hydrodynamic interaction), physical (i.e., particle size), chemical (i.e., phase, oxidation state), and biological (i.e., transport of particles through vegetative canopies) processes are involved in the fallout of elements from the air to the earth's surface (Pacyna, 2008). The process of dry deposition involves particle size and gas exchange (Macdonald et al., 2000), while wet deposition depends on particle impaction by rain droplets, precipitation scavenging (through adsorption), solubility of vapor phase substances in water and adsorption to snow (Macdonald et al., 2000). In the Arctic, cold, dense air, and increased darkness during the winter lead to accumulation and increased possibility of atmospheric trace element deposition (Halbach et al., 2017) due to the atmosphere's ability to 'capture' air masses for months at a time (NPI, n.d.). After deposition, elements can be accumulated in the upper layer of soil and snow, on vegetation and in aquatic environments (Pacyna, 2008).

In Canada, atmospheric monitoring programs, such as the National Air Pollution Surveillance (NAPS) and the Canadian Air and Precipitation Monitoring Network (CAPMoN) have been established to assess air quality in urban and background locations, however stations are spatially intermittent (Cowden and Aherne, 2019), due to limited resources and infrastructure, especially in Arctic regions. The use of mosses to measure trace metal deposition has been practiced in Europe since the 1970s (Ruhling and Tyler, 1970; Cowden and Aherne, 2019), and since 1990, the International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops (ICP Vegetation) has been carrying out large-scale moss surveys every five years across Europe (Harmens et al., 2010; Cowden and Aherne, 2019). The use of mosses in the assessment of atmospheric deposition can provide information on the relative deposition of nutrients, trace elements, and contaminants (Pacyna, 2008) in regions where mosses proliferate.

1.4 The Canadian Arctic

The Canadian Arctic (area > 60°N) contains 40% of Canada's land mass with less than 1% of Canada's population, and includes the Northwest Territories (1,144,000 km^2), Nunavut (1,877,787 km^2) and Yukon (482,443 km^2). Surface waters in the three territories make up 36.8% of Canada's surface water extent (Reist et al., 2015). Baffin Island is Canada's largest island, located in the Canadian Eastern Arctic and hosting Nunavut's capital city Iqaluit (Peramaki, 2000). The Canadian Arctic was once considered to be a pristine environment because of its geographical location away from southern agricultural and industrial regions (Donaldson et al., 2010). Since the mid-1980s and early 1990s, studies have found environmental contaminants (i.e., heavy metals, organochlorines, DDT, and PCBs) in all compartments of the Arctic ecosystem (Wong,

1986; Barrie et al., 1992; Lockhart et al., 1992; Muir et al., 1992; Thomas et al., 1992). As populations continue to rise within the Arctic (populations are expected to increase 30%), more local emission sources are expected to be added to the atmosphere (Schmale et al., 2018). In recent years, Hg, Cd, and Pb have been identified as elements of concern, due to their ability to biomagnify and bioaccumulate up the food chain (NCP, 2012), and negatively impact northern communities and wildlife. Many Arctic inhabitants rely on the land for subsistence living, which is both nutritionally and culturally important to them (The Arctic Council, 2021). The Arctic Monitoring and Assessment Programme (AMAP) and Canada's Northern Contaminants Program (NCP) were created to carry out research on the status of elements of concern (e.g., Hg, Cd, and Pb), heavy metals, climate change, persistent organic pollutants (POPs), and more recently microplastic concentrations within the Arctic region.

1.5 Climate Change

The Canadian North is warming at twice the global average rate, which is affecting the land, biodiversity, and culture and traditions of northern communities (Government of Canada, 2018). Arctic ecosystems are particularly vulnerable to changes in temperature, snow, and ice-cover (Post et al., 2009). Some of the most rapid ecological changes associated with a warming climate have been observed in freshwater environments (Post et al., 2009).

Climate change may also result in the increased mobilization of trace elements through enhanced geochemical weathering, a reduction in snow cover, and a reduction in freezing days (Zaharescu et al., 2016). Thawing of permafrost and snowmelt can increase trace element concentrations (accumulated over the winter) in soil water, leading to higher uptake through vegetation, and enhance trace element transport in surface waters

and soil layers (Prowse et al., 2006; Muskett and Romanovsky, 2011; Perryman et al., 2020). Elevated concentrations of Hg, Fe, Mn, Ni, Zn, Al, Cr, and Pb in soil have all been observed in areas that have experienced rapid thaw because of the release of accumulated (from increased weathering and legacy sources) elements (Antcibor et al., 2014; Loiko et al., 2017; Schuster et al., 2018; St. Pierre et al., 2018). Furthermore, increases in soil organic matter (SOM) degradation during a period of thaw can impact the mobility of metals bound to SOM (Rajkumar et al., 2013; Perryman et al., 2020). Given the recent and projected rapid changes in climate, it is important to understand current concentrations and pools of trace elements to be able to predict the potential impacts of climate change on catchment transport rates and pathways (Miller et al., 2003; Middelkoop, 2008; Wigngaard et al., 2017).

Additionally, decreases in sea ice have made the Arctic easier to navigate. This has led to increases in shipping activities, and the potential use of new transit routes, such as the Northern Sea Route and the Transpolar Sea Route (Stephenson et al., 2015). As most of the shipping emissions occur within 400 km of coastlines, they primarily contribute to air pollution in coastal areas (Eyring et al., 2010; Aulinger et al., 2016). Moreover, emissions can be transported hundreds of kilometres downwind and impact a much broader region (Eyring et al., 2010; Aulinger et al., 2016). A secondary impact to loss of sea ice is a predicted increase in mining activities (exploration and development), and human populations (Gong et al., 2018; Schmale et al., 2018), which will further increase emissions to the atmosphere and degrade the landscape.

1.6 Lake Catchments as Integrators of Change

Lake catchments are an important component in the hydrologic cycle allowing for the determination of mass balances (i.e., fluxes of solutes into and out of a catchment),

and the assignment of solutes to specific sources and sinks such as atmospheric deposition, biomass change, or bedrock weathering (Drever, 1997; Hynes, 1975; Allan et al., 1997; Allan, 2004). Within catchments, lakes are effective indicators because they act as ‘sentinels of change’, reflecting process changes at the landscape scale, and providing spatial and temporal information on the impacts of anthropogenic and natural activities (Liang, 2018; Adrian et al., 2009). Lake catchments are broadly defined as a depression of land that receives and collects water, usually from subsurface (groundwater) and over surface flow and precipitation; water will then drain into an outlet (river, lake, reservoir, or other bodies of water).

Lakes and ponds are ubiquitous in the Arctic environment, playing a role in the ecology of fish and wildlife, providing a reliable source of freshwater to northern communities and mining camps (Woo et al., 1981), and reflecting internal and external catchment processes. High latitude Arctic lakes are generally oligotrophic and remain ice-covered for most of the year (Zaharescu et al., 2009); they are exposed to low temperatures, relatively small amounts of precipitation, and seasonally low inputs of solar radiation, which limits the development of vegetation and the chemical weathering of soils within their catchment (Liang, 2018). The hydrochemistry (i.e., nutrients, anions, trace elements) of these water bodies is predominantly influenced by atmospheric inputs, surface sediments and bedrock geochemistry of the catchment, i.e., the geological structure, the mineralogical/chemical composition of the rocks, the proportion of rock types and their weathering resistance (Hutchinson, 1957; Wetzel, 1983; Lewin and Macklin, 1987; Hamilton et al., 2001; Zaharescu et al., 2009; Liang, 2018). The biogeochemical cycling of trace elements in Arctic environments is generally governed

by a weathering-limited regime (Stallard and Edmond, 1983), leading to an overall low input into an ecosystem, i.e., aqueous concentrations are usually between 1–2 $\mu g \cdot L^{-1}$ (Markert et al., 1997; Zaharescu et al., 2009). Further, lake sediments can serve as both a sink and a source of trace elements, and act as the main pool of elements in the aquatic environment (Fernandes and Nayak, 2012; Ali and Khan, 2019;). The distribution of elements in lake sediments is affected by their chemical composition, grain size, and organic content (Zhao et al., 2014; Ali and Khan, 2019). The source of trace elements in high latitude lakes can be geogenic and anthropogenic (Zaharescu et al., 2009). Catchment soils act as a reservoir for trace elements to the surrounding environment by acting as a natural pathway through surface runoff. Factors affecting the presence and distribution of trace elements in soils are the composition of parent material, weathering, soil permeability, and surface roughness. Trace elements tend to accumulate in soils and sediments (Foster and Charlesworth, 1996) because of their affinity for sorption processes (particularly with organic matter), when introduced into the environment (Wijngaard et al., 2017). There have been several studies characterizing the chemical properties of lake water, lake sediment and surface soils in the Canadian Arctic (Stewart and Macdonald, 1981; Stewart and Bernier, 1983; McNeely and Gummer, 1984; Welch and Legault, 1986; Fee et al., 1988; Hamilton et al., 1994; AMAP, 2005 Halbach et al., 2017); however, most studies rely on historic data (Liang, 2018), and knowledge on the role of catchment characteristics (water, sediment, and soil) is limited.

The catchment of a waterbody is the area where water will travel overland or through subsurface movement to enter or drain into a waterbody (Davies et al., 2008). Lake catchments are typically divided into three groups: with inlets and outlets (visible

during periods of low flow), headwater lakes (no inlets but at least on outlet), and isolated lakes (no visible stream inlets or outlets; Yan et al., 2019). Lakes can be connected in a sequential lake stream network (tandem lakes), a lake chain network (mixed lakes), or single lakes (Figure 1.1; Sadro et al., 2012; Yan et al., 2019). Atmospheric inputs and trace element accumulation, transport, distribution, and fate in remote lake catchment systems show that once deposited, elements can be stored and redistributed throughout environmental compartments (i.e., soil, lake water, sediment, and terrestrial and aquatic biota; Peñarroya, 2011). Transfer processes such as deposition, storage, and redistribution of elements can be spatially different, as they may follow the latitudinal or longitudinal gradients throughout different regions (Peñarroya, 2011). In general, concentrations of solutes such as major cations and anions are expected to increase along downstream flow paths, whereas biogeochemically reactive materials (i.e., trace elements, nutrients, organic matter) can increase or decrease depending on reaction dynamics within lakes (Soranno et al., 1999; Sadro et al., 2012).

Biomonitoring using moss has become a useful tool in assessing air pollutants in Europe (ICP Vegetation, 2015). This approach has proven effective at providing temporal and spatial insight into air pollution deposition (Cowden and Aherne, 2019). There are few Canadian studies that use moss as a biomonitor (Cowden et al., 2015; Pott and Turpin, 1998; Shotyk et al., 2016). *Hylocomium splendens* and *Pleurozium schreberi*, are pleurocarpous moss that are commonly used in biomonitoring studies because they are easy to identify (Berg et al., 1994); furthermore, they are found in high abundance throughout the globe, including the Canadian Arctic (Iqaluit, Nunavut). They form on the ground and are an effective trap for trace elements in precipitation, airborne particles, and

windblown dust (ICP Vegetation, 2015). Additionally, enrichment factors (EFs) are often used to assess the variation of the deposition of pollutants over a given area, and to determine an elements origin (anthropogenic or geogenic; Klos et al., 2011; Macedo-Miranda et al., 2016). EFs do this by comparing the concentration of an analyte accumulated in mosses to that in soil with reference to a conservative element (Klos et al., 2011; Cowden, 2018).

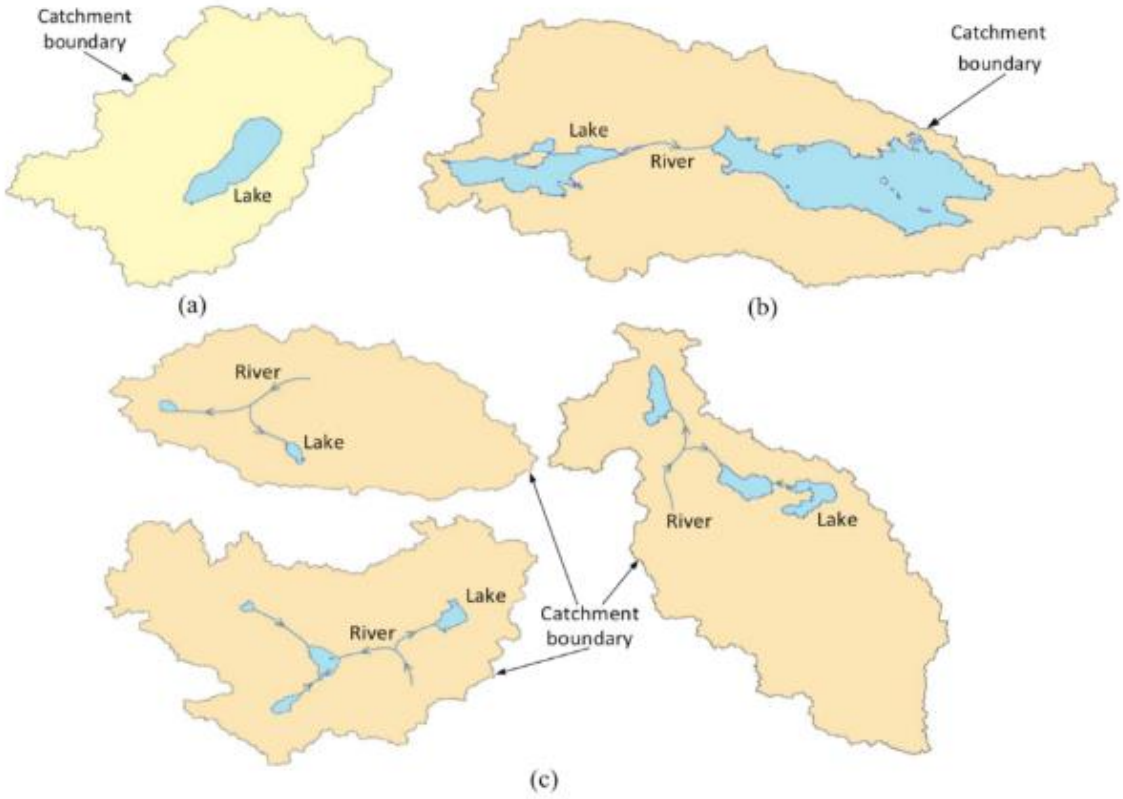


Figure 1.1: Diagram of different lake catchment types: single-lake catchment (a), tandem-lake catchment (b), and mixed-lake catchment (c). Note image derived from Yan et al. (2019).

1.7 Thesis Objectives

The objective of this study was to assess the variation and concentration of trace elements (Al, As, Ba, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, THg, V, and Zn[2]) in the low-Arctic, specifically focusing on concentrations in lake water, lake sediment, catchment soil, and moss (*Hylcomium splendens*) from 20 lake catchments surrounding Iqaluit, Baffin Island; and to investigate sources (geogenic versus anthropogenic) of trace elements in the lake catchments, and moss by using the PMF receptor model and enrichment factors (moss). In addition, Arctic lake catchments were characterized for chemistry of lake water (major ions [Cl^-, Fl^-, NO_3^-, SO_4^{2-}, Ca^{2+}, Mg^{2+}, Na^+, and K^+], nutrients [NH_3 and TP]), soil, and lake sediment composition.

Baffin Island represents a unique opportunity to study Arctic environments because it is easily accessible compared to other Arctic regions, where helicopter and charter flights are often required. Furthermore, the Nunavut Research Institute provided accommodations and the opportunity to engage the community through oral presentations and workshops. The 20 lake catchments represented replicate sampling, as the area surrounding Iqaluit is homogenous in nature, suggesting that the catchments experience the same geologic and pollution sources. The trace elements in this study had varying concentrations levels, e.g., Fe and Al were several orders of magnitude higher than all other elements. Nonetheless, for the purposes of this study every element was referred to as a trace element.

[2] These elements were selected because they provide a good representation of anthropogenic (associated with human and environmental health hazards) and geogenic (abundant within the earth's crust) sources.

The hypothesis of this study was that trace elements emitted from both local and distant anthropogenic sources are present in Arctic lake catchments. This hypothesis was further explored through research questions.

The following research questions were addressed: 1) Does soil and sediment composition (organic content, pH, and particle size) influence trace element concentrations in soil and sediment? 2) Are pH and (DOC) associated with element concentrations in lake water? 3) What elements are dominant in lake water, lake sediment, catchment soil and moss? 4) What trace elements show a high variability (spatial and interannual) across the catchments? 5) Using the PMF receptor model and enrichment factors (in moss), can anthropogenic and geogenic sources be distinguished in lake water, lake sediment, moss, and catchment soil?

This thesis is written in a traditional style, which includes six individual chapters as follows: Chapter 1: Introduction, Chapter 2: Methods, Chapter 3: Lake catchment characteristics (results), Chapter 4: Trace elements in Arctic lake catchments: concentrations and sources (results), Chapter 5: Discussion, and Chapter 6: Conclusions. The study region, sampling methods, and laboratory methods are the same throughout the entire thesis. Chapters 3 and 4 address the primary objectives for this thesis. Details on plant species diversity in the 20 Arctic study catchments is provide in Appendix A.

1.8 Significance of Research

This research integrates the use of lake water, sediment, soil, moss, and vegetation as monitors of ecosystem health at the catchment scale, and how they can be impacted by local factors and long-range transport. Trace element concentrations and the general chemistry of the catchment provide a more holistic understanding of baseline conditions in lake catchments in the Iqaluit region. Few studies (Permaki, 2000; Halbach et al.,

2017; Wilkie and La Farge, 2018) have integrated Arctic lake catchments into their results, leaving knowledge gaps and failing to capture the 'whole' picture throughout the region. In this work, twenty lake catchments were sampled to provide a broader understanding of how environmental compartments are linked and influenced by trace element concentrations. Furthermore, there is little information regarding the use of *Hylcomium splendens* as biomonitors (following ICP Vegetation protocol), and vegetation species composition (through the use of surveys), with poor spatial coverage and lack of overall available data in the eastern Canadian Arctic, specifically Iqaluit, Baffin Island, Nunavut. This research has the potential to link Canadian Arctic observations, with other northern regions. Further, the results support the mandate, strategy, and work plan of AMAP and NCP.

2 Materials and Methods

2.1 Study Region

Iqaluit (63.7467ºN, –68.5170ºW) is the capital city of Nunavut, covering a land area of 52 km^2. It is north of the treeline, and situated on southern Baffin Island, along Frobisher Bay. Iqaluit has a population of 7,740 residents, 83.8% of whom identify as Inuit (Statistics Canada, 2017; Masina et al., 2019). Long-term climate data for Iqaluit is available 1 km inland at 33.5 m a.s.l. (Canadian Climate Normals 1971–2000 Station Data; 63.7500ºN, –68.5500ºW). Annual mean daily air temperature is –9.5°C, ranging between a mean daily temperature of 7.7°C in July, and –26.8°C in February. Long-term annual precipitation is 424.1 mm (192.9 mm of rain; 256.8 cm of snow), 95% of rain occurs from June–September, and 90% of snow occurs from October–April. Iqaluit lies on the Precambrian Canadian Shield, which is composed of a variety of metamorphic rocks, such as granite, migmatite, and quartz-feldspar gneissic rocks (Andrews, 1989; Blackadar, 1967; Peramaki and Decker, 1999), giving low weathering rates, low base cations (Ca^{2+}, Mg^{2+}, Na^{+}, and K^{+}), and high silicon dioxide (SiO_2) content (Skjelkvåle et al., 2001), leading to lakes with neutral pH and low conductivity (Liang, 2018).

Iqaluit is situated within the low arctic tundra zone, which is an ecologically sensitive area (and therefore sensitive to climate warming and transboundary pollution) and is continuously underlain by permafrost (Peramaki and Decker, 1999). Soils in the region are broadly described as regosolic static (cryoturbation exists in regional pockets throughout Baffin Island) cryosols (i.e., no B horizon, and has permafrost at a depth of 1–2 m), developed on sandy morainal material, while frost boils and other micro-landforms are commonly found on till. Organic soils are considered rare as wetlands are generally limited to a few centimetres of peat (Tarnocai and Kroetsch, 1994; Hodgson, 2005).

Constant freezing and thawing have led to nutrient poor, silty, and shallow soils with little soil profile development (Peramki and Decker, 1999; Tarnocai, 2009), leading to small, stunted (<30 cm in height) vegetation. Mosses, lichens, willow (*Salix*), heathers (*Ericaceae*), and dwarf shrubs are found abundantly across the tundra, with extensive areas of sedges and grasses found in low, wet areas, and on moist (till or marine deposit) slopes (Hodgson, 2005; Short and Jacobs, 1982) (Figure 2.1 shows IQ 34 the study site). Study sites were dominated by moss spp., lichen spp., grasses, sedges, and small trees (see Figure 2.1 for example).

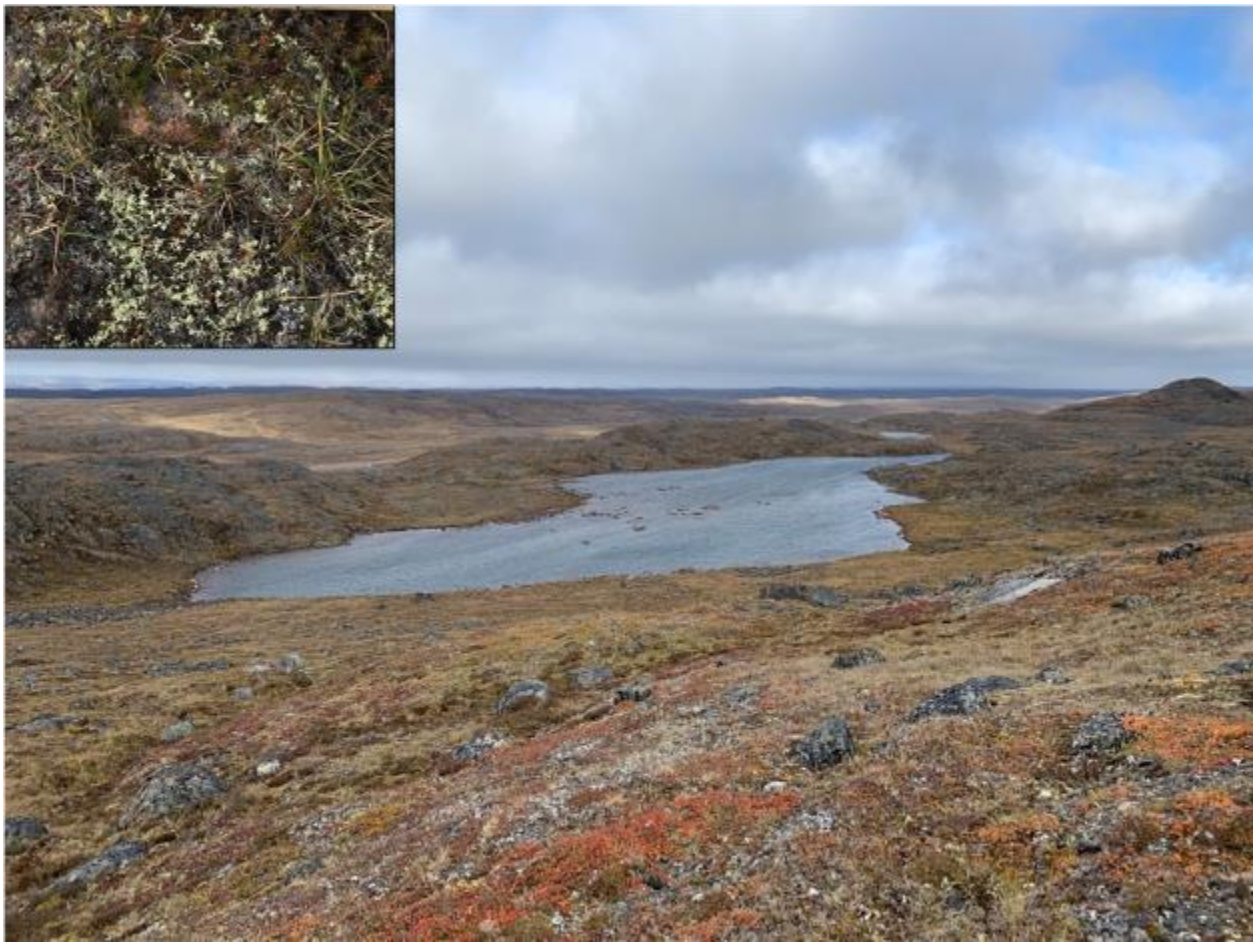


Figure 2.1: Sampling site near Iqaluit (IQ 34); tundra mostly comprised of mosses, lichens, heathers, dwarf shrubs, and grasses are found predominantly at the sites. Inset shows a typical tundra plant community (e.g., lichen spp., moss spp., grass spp., Labrador tea, and white heather) found throughout Iqaluit and surrounding area.

2.2 Site Description

The twenty study lake catchments were chosen from an existing lake survey, where 50 lakes were selected using stratified-random sampling (NTS Grid 25N 025N15/025N16; Jefferies et al., 2010; Liang and Aherne, 2018), with a greater weighting to lakes >1 ha in surface area and accessible (i.e., walking distance was no more than 10 km) from Iqaluit (Figure 2.2; Table 2.1). Catchments (*n*=20; Figure 2.2) were sampled during September 2018 for lake water, lake sediment, catchment soil, moss (*Hylocomium splendens*); Figure 2.2 and Figure 2.3), and vegetation diversity (Appendix A). During August 2019, lake water and moss (Figure 2.3) were sampled again to assess interannual variability between trace elements and major chemistry. Vegetation diversity was also sampled again in 2019 (details are provided in Appendix A). Moss, vegetation, soil, and sediment were sampled from the north, east and west facing slopes of each lake catchment, while lake water was collected only from the north facing slope. Chemical analysis associated with each environmental parameter is described in Table 2.1.

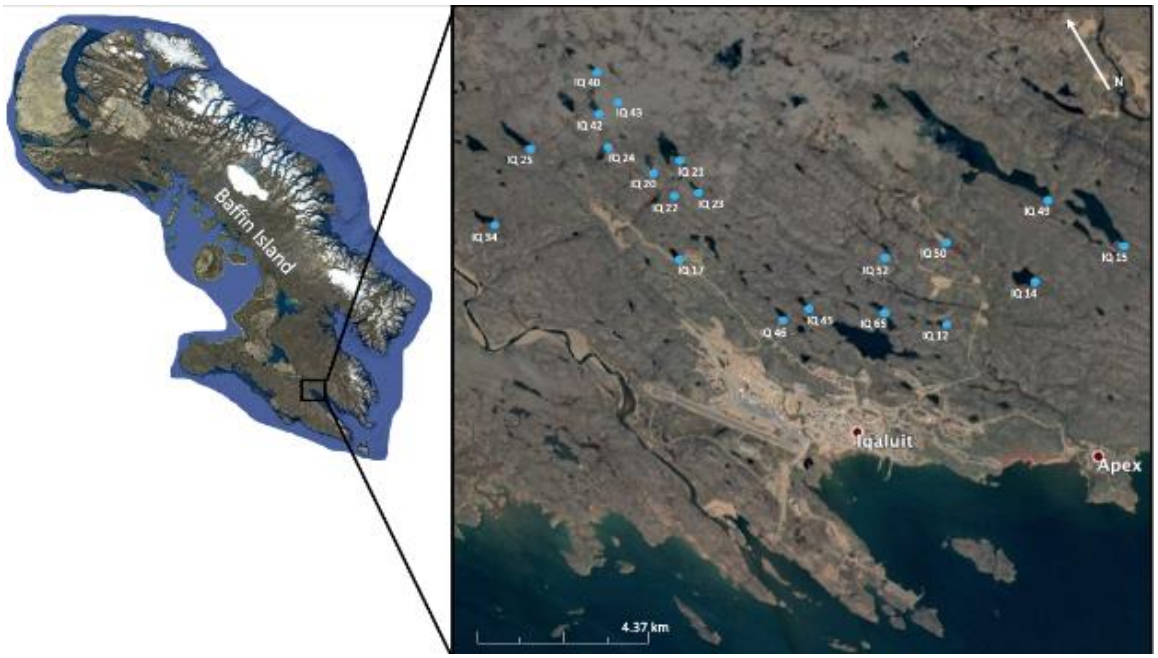


Figure 2.2: The location of the twenty lake catchment study sites surrounding Iqaluit southern Baffin Island.

Figure 2.3: *Hylocomium splendens* (mountain fern moss) sampled at each of the 20 study sites in the Iqaluit region.

Table 2.1: Chemical analyses on samples collected from environmental compartments across the study catchments (n = 20) in the Iqaluit region. Sediment and soil were only sampled in 2018.

Environmental compartments	Analyses
Lake (surface) water	• Trace elements including total mercury (THg); • Major ions; • Nutrients; • Alkalinity, pH, conductivity; and • Dissolved organic carbon (DOC), dissolved inorganic carbon (DIC), total dissolved nitrogen (TDN)
Lake (surface) sediment	• Trace elements including THg; • Particle size analysis (PSA); • Organic matter (OM); and • pH
Catchment (surface) soil	• Trace elements including THg; • Particle size analysis (PSA); • OM • pH; and • Bulk density
Hylocomium splendens (moss; Figure 2.3)	• Trace elements including THg • Enrichment factors
Vegetation (Appendix A)	• Community composition; • Abundance; • Species richness; and • Cover

2.3 Sample Collection

2.3.1 Lake water

All surface water samples (for trace elements, THg, major ions, and nutrients) were collected from the shoreline (lake edge), from an area free from emergent vegetation, and sampled at a depth of 15 cm below the surface in pre-cleaned containers. Samples for trace metals were collected in a 15 mL conical tube, THg was collected in two 40 mL I-Chem vials, nutrient samples were collected in a 125 mL high-density polyethylene (HDPE) bottle, and alkalinity and major ions were collected as a bulk water (unfiltered) sample in a 250 mL HDPE bottle. Trace metal (excluding THg) and nutrient samples were filtered in the field using a 0.45 µm nylon syringe filter; trace metal

samples were then acidified with HNO_3 2% v/v, and THg (unfiltered) samples with HCl 0.5% v/v within four hours of sample collection. At predetermined sites, field duplicates were collected to ensure sample consistency and accuracy (10% of the sites were selected). Samples were kept in the dark and stored at 4ºC until laboratory analysis. Supplementary water chemistry (pH, conductivity, temperature, and dissolved oxygen) data were collected on site during sampling using a YSI 650 MDS probe.

2.3.2 Lake sediment

A surface sediment (0–5 cm) sample (~100 g) was collected from the shoreline (~30 cm–1 m below water line) of the north, east and west facing slopes (i.e., each slope had one sample collected) at a distance of approximately 1.2 meters from the shore. Samples were collected away from emergent vegetation and debris, transferred to a large Ziploc bag, where all three samples were composited, homogenized, and transferred to a glass jar (~100 g). Samples were stored in the dark and refrigerated (4°C) until laboratory analysis.

2.3.3 Catchment soil

Vegetation plots (quadrats) were designed to capture species richness and composition at the study sites and provide a general picture for the surrounding area. Vegetation plots and transects were established on the north, east and west facing slopes. Two plots and one transect was setup per slope, with a total of six plots and three transects per study site (catchment). One quadrat was placed at the start and end of the 10 m transect, where species richness and composition were visually assessed (Appendix A). A bulk density soil core (2.5 cm internal radius, 5 cm in height, and 102.14 cm^3 volume) was collected from the top left outside corner of the vegetation quadrats. The first quadrat placed at the start of the transect (10 m from shoreline) was used for collecting soil

samples (bulk density and composite samples). In total, three bulk density cores were collected per catchment (e.g., one core was collected per slope) and transferred to individual Ziploc bags. Composite surface soil samples (0–5 cm; ~100 g) were collected using a hand trowel at the top left corner outside of the vegetation quadrat, and added to a Ziploc bag, where the three samples (one per quadrat) were homogenized. Samples were refrigerated (4°C) and stored in the dark until laboratory analysis.

2.3.4 Moss

Approximately 5 g (wet weight) of *Hylocomium splendens* was collected from a 50 m by 50 m area at each lake catchment, following the ICP Vegetation protocol (ICP Vegetation, 2015; Fernández et al., 2015). Typically, 5–10 sub-samples within each catchment were collected and combined in a brown paper bag. The moss colonies were usually sampled from the ground or growing on sheltered boulders. To avoid the possibility of sample disturbance, moss samples were collected away from trails or foot paths. The fresh (green) portion of the moss was collected, and cleaned in the field, i.e., brown moss parts, lichen, soil, and plant debris was removed (ICP Vegetation, 2015). Moss was stored in the dark and kept cool until laboratory analysis.

2.4 Laboratory Analysis

2.4.1 Lake water

Unfiltered bulk water samples were analysed for alkalinity, pH, and conductivity using a Mantech PC-titrator. Filtered (0.45 µm) water samples were analysed for major cations (Ca^{2+}, K^{+}, Mg^{2+}, and Na^{+}) using inductively coupled plasma optical emission spectrometry (ICP-OES). Major anions (filtered 0.45 µm; (Cl^{-}, Fl^{-}, NO_3^{-}, PO_4^{3-}, and SO_4^{2-})) were analysed through ion chromatography. Water samples were filtered in the laboratory using a 0.45 µm nylon syringe filter and analysed for dissolved organic carbon (DOC), dissolved inorganic carbon (DIC), and total dissolved nitrogen (TDN) using a

Shimadzu TOCV. Nutrients: ammonia (NH_3), and total phosphorus (TP) were analysed colorimetrically using EPA methods 350.1 (NH_3; EPA, 1993a), and 365.1 (TP; EPA, 1993b; analyses was completed within the Environmental GeoScience Research Groups laboratories). THg was analyzed through a Tekran 2600 mercury analyzer (EPA method 1631; EPA, 2002). Sixteen elements (Al, As, Ba, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, V, and Zn) were analyzed using a Triple Quad inductively coupled plasma mass spectrometry (ICP-MS) analyzer at the Trent University Water Quality Centre.

2.4.2 Catchment soil and lake sediment

Surface soil and sediment samples were oven dried at 40°C for 72 hours and sieved to fine (<2 mm) and coarse (>2 mm) fractions. Samples <2 mm were then analysed for organic matter, bulk density (soil only), pH ($CaCl_2$), and particle size (sand, silt, and clay). Percent (weight) organic matter (%OM) was estimated as loss-on-ignition (LOI; muffle furnace was set to 400^{o}C for 10 hours), which is widely used for soil and lake sediment (Hoogsteen et al., 2018). The weight losses observed at temperatures above 400°C can be mostly attributed to the destruction of SOM (Hoogsteen et al., 2015; Hoogsteen et al., 2018). Bulk density (g/cm^3) was calculated by dividing the dry soil weight (g) of the sieved fine <2mm fractions by the volume of the soil core (102.14 cm^3). The pH was measured using an Oakton multimeter in a 1:2.5 soil/sediment:solution ratio; 0.01M $CaCl_2$ and ultra-pure water (b-pure). Soil and sediment samples were analysed for percent sand, silt, and clay by laser diffraction using a Horiba Particle-size Distribution Analyser LA-950V2. Each sample was measured in triplicate in a solution of 40 g of Calgon per litre of reverse osmosis (RO) solution.

Catchment soil and lake sediment (fine fraction) were pulverized (to ensure homogenization of sample) using a mixing mill (Spex mixer model 6000D). Trace

element (Al, As, Ba, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, V, Zn) concentrations were then measured using a Triple Quad ICP-MS (within Trent University Water Quality Centre) analyzer following acid digestion using a Mars 6 microwave digester (1:1 HNO_3 and H_2O_2; EPA method 3052).

2.4.3 Moss

Moss samples were cleaned of dead material and debris (i.e., vegetation, soil particles) (ICP Vegetation, 2015), dried for two weeks in their paper bags at room temperature (20°C) and pulverized using a mixing mill (Spex mixer model 6000D), with an 8100 Spex adapter for use with plastic vials and agate balls. Trace element concentrations (Al, As, Ba, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, V, Zn) were determined on a Triple Quad ICP-MS analyzer following acid digestion using a Mars 6 microwave digester (1:1 HNO_3 and H_2O_2; EPA method 3052). The THg samples were analysed using a Milestone DMA-80 mercury analyzer (EPA Method 7473; EPA, 2007).

2.4.4 Data treatment: blank correction and transformations

Blank concentrations were used to determine potential laboratory contamination. Blanks were made for each environmental compartment and were used during individual analytical batch runs for each laboratory analysis (Appendix B). Blank concentrations were averaged and subtracted from the measured concentration of each sample. Some trace element concentrations (Co, Se, As) exhibited values that were below detection in lake water and moss (water 2018 sample: Co = 30%; water 2019 sample: Se = 80%; moss 2018 sample: Co = 20%; moss 2019 samples: As = 95% and Se = 90%). Values less than the detection limit can cause difficulties for statistical analysis, as such appropriate replacement values must be used (Cowden and Aherne, 2019). For this study, values below detection limit were set as half the detection limit to standardize across all datasets.

Values were set as half the detection limit because they made up a small portion of the results, and this has been shown that it minimally impacts the outcome of the analysis (Wendelberger and Campbell, 1994).

2.4.5 QA/QC analysis results

Replicates were analyzed for each environmental compartment for all laboratory analyses for this study. Replicates were used to ensure analytical and instrument precision, and to determine potential sample contamination. Ten percent of samples collected in the field had triplicates made in laboratory (i.e., one sample had 3 sub-samples analysed from it). Triplicate values for all environmental compartments were within range of the three sub-samples. The mean percent relative standard deviation (%RSD) amongst replicate analysis for surface water samples were Al, As, Ba, Cd, Fe, Mn, Sb, Se, and V < 30%, while Co, Cr, Cu, Ni, Pb, and Zn ranged from 35–68%. In moss, Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Se, As, Sb, Ba, THg, and Pb < 30%, and Cd had the largest CV value of 45%. Replicates < 30% for catchment soil were: Al, V, Cr, Zn, Se, As, Sb, THg, and Ba, and Pb, Cd, Cu, Ni, Co, Fe, and Mn ranged between 31–38%. Lastly, lake sediment values were THg, Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Se, As, Ba, Cd, Sb, and Pb < 30%.

2.5 Data Analysis

2.5.1 Delineation of lake catchments and elevation values

Based on visual assessment, most lake catchments were defined as being single lake catchments. Lake catchment boundaries (perimeter) were delineated using topographic lines, where the apex of the hill surrounding the catchment was considered the boundary. Catchment area and elevation was calculated in Google Earth Pro (7.3.3) using topography, water body/course, and linear flow data from CanVec obtained through the Government of Canada's National Hydrological Network (Appendix C).

Distance to coast (km) was calculated using the Grass GIS 7 plugin in QGIS and a coastal shapefile of Canada (Liang, 2018). Lake:catchment ratios were calculated from data provided by Google Earth. Elevation values that were missing (required) for lake catchment elevation (m) was determined using Google Maps Elevation Application Programming Interface (Appendix D).

2.5.2 Data analysis for lake catchment variables

All data used in this study for water, sediment, soil, and moss were tested for normality using the Shapiro-Wilks test. Variables were considered normal if the *p*-value was >0.05. Log transformations were attempted to normalize the data that did not meet this criterion, however many variables remained non-normal after the transformations, therefore non-parametric tests, and statistical summaries (e.g., median, Spearman rank correlation, etc.) were calculated for all lake catchment datasets. The median was presented for all data because it is less susceptible to outliers. Spearman rank correlations were carried out between catchment attributes, general water chemistry, physiochemical (soil and sediment), environmental variables, and trace elements, and only significant (p-value = <0.05) correlations are presented here. Spatial variability in general chemistry and trace element data for the lakes, sediment, soil, and moss was calculated using the normalized median absolute deviation (%NMAD). Interannual variability in general chemistry and trace element data for surface water and moss was calculated as relative percent difference (RPD). Spatial and interannual variability <35% were considered to be low (not variable), while values above were considered to be variable.

PCAs was carried out to identify relationships between individual datasets in water, sediment, soil, and moss. Additionally, a PCA was also used to evaluate the relationships between trace elements and environmental variables (i.e., pH, conductivity,

DOC in water, and pH and OM in soil and sediment), as they are known to influence trace element cycling (Lydersen et al., 2002).

Trace element soil pools were calculated to quantify total element presence and distribution throughout the catchments. Soil pools (mass per unit area; $mg \cdot m^{-2}$) were calculated using bulk density ($kg \cdot m^{-3}$) and soil chemistry from a depth of 5 cm.

Source apportionment is the practice of deriving information about pollution sources and the amount they contribute to pollution levels (Belis et al., 2014). This is usually accomplished using three main approaches: emission inventories, source models and receptor models (Belis et al., 2014). For this study the PMF model and enrichment factors (EFs) (receptor models) were used to categorize similar groupings of trace elements, and further determine source apportionment such as, geogenic and anthropogenic sources. The PMF model is able to produce similar groupings of elements, whereby source apportionment can be determined, such as geogenic (natural) or anthropogenic sources (see Section 2.5.4).

Statistical analysis was performed using R (Version 3.3.2; packages: nortest, Hmisc, ggplot2, stats), paleontological statistics (PAST Version 4.04), PMF model (Version 5.0.14), and Microsoft Excel (Version 16.35).

2.5.3 Enrichment factors in moss

Enrichment factors (EFs) have been used in moss biomonitoring studies to illustrate sources of natural or anthropogenic trace elements accumulated in moss tissue.

Enrichment factors (EFs) were used to assess sources of natural or anthropogenic trace elements accumulated in moss tissue. Enrichment factors compare the ratios of accumulated trace elements in moss tissue to that in soil collected from the same location (Klos et al., 2011), both normalized to a reference soil element, this allows for inferences

to be made regarding the source of that element (Cowden, 2018). The EF calculation uses the concentration of an examined element in moss and soil and divides it by the concentration of a reference element in moss and soil. The calculated enrichment factor value allows for a differentiation between natural (substrate, overhang leaching) or anthropogenic (atmospheric deposition, windblow dust) sources in moss tissue (Dragovic and Mihailovic, 2008; Klos et al., 2011; Macedo-Miranda et al., 2016; Cowden, 2018). Good reference elements for crustal sources are typically Al, Fe, Sc, and Ti because they rarely enter atmospheric aerosols from anthropogenic sources (Klos et al., 2011; Ford and Hasselback, 2001; Dragovic and Mihailovic, 2008; Biegalski et al., 2011). In this study, soil was used as a reference material instead of parent (crust) material. This is most commonly found in moss biomonitoring studies (Dragovic and Mihailovic, 2008; Klos et al., 2011; Biegalski et al., 2011. Macedo-Miranda et al., 2016; Cowden, 2018) as sampling soils at different locations within a catchment can represent local soil parent material (Puckett and Finegan, 1980; Nash and Gries, 1995). Additionally, using soil allows for the separation of recent deposition from legacy sources, and wind-blown soil from deposition[3].

By definition, EFs that are <3 are considered not enriched, resulting in an element from soil contributions (Klos et al., 2011). An EF that ranges from 3–9 is considered enriched, while EFs that are > 9 are considered highly enriched, suggesting an anthropogenic source (Dragovic and Mihailovic, 2008). In this study Al was used as the reference element and EFs were computed as per eq. 2.1 below, where *x* is the trace element concentration and Al is the reference element.

[3] Limitations exist as soil also receives recent deposition.

Eq. (2.1)

$$EFx = \frac{\left(\frac{x}{Al}\right) in\ moss}{\left(\frac{x}{Al}\right) in\ soil}$$

2.5.4 Positive matrix factorization (PMF) model

The PMF model (U.S. Environmental Protection Agency PMF 5.0; Norris and Duvall, 2014) has been widely used to distinguish between anthropogenic and geogenic sources of trace elements. The PMF model is a type of factor analysis, whereby it reduces large datasets into smaller groups (factors) that share commonalities (Norris and Duvall, 2014; Gholizadeh et al., 2016; Isley et al., 2018; Wu et al., 2021). The factors can then be related to emission or natural sources. Trace element concentrations (2018 data for moss, lakes, sediment, and soil) for Al, As, Ba, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, V, and Zn were input into the PMF model to determine their source contributions. The concentration data and measurement error for each trace element were required inputs for the model. The measurement error was calculated through percent relative standard deviation for each element replicate (i.e., the triplicate of the element), and then multiplied by the element concentration (Appendix E).

Once the required data has been inputted, the model analyzes it by conducting 20 base model runs, e.g., the model automatically runs 20 different simulations. Twenty runs are the optimal number for the model to evaluate the correlations and variation between the data (Norris and Duvall, 2014; Wu et al., 2021). Base model runs produce the primary PMF output of profiles and contributions (Norris and Duvall, 2014). Based on these data, the model creates factor fingerprints, which are stacked bar charts that display the concentration (in percent) of each species contributing to each factor[4]. For the purpose of

[4] The figures cannot be manipulated as they are a direct output of the model.

this study, factor fingerprints were focused on because of the visual and numerical data they provide.

The PMF model runs a correlation analysis, whereby the default R-value is 0.6 (Norris and Duvall, 2014; Wu et al., 2021). This model will only show positive correlations (hence positive matrix), allowing users to 'easily' interpret the data. Using the correlation analysis function in PMF, elements were considered usable (strong) based on their R-value (> 0.5); elements were not excluded from the analysis. Once the model has finished running, it chooses the best run or best representation of the data.

The PMF model outputs factors, which are groups of elements that occur repeatedly throughout the data set in specific ratios (Wu et al., 2021). For each environmental compartment, the R-values were used to determine which factor best suited the data. The 4-factor model was selected based on more significant (higher) R-values for all environmental compartments, and the higher >80% bootstrapping results (Norris and Duvall, 2014). The Residual analysis (an output from the model), displays a histogram showing the percent of all residuals in a given bin (each bin is equal to 0.5; Norris and Duvall, 2014). These plots are useful to determine how well the model fits each element. If an element has many large residuals or displays a non-normal curve, it may be an indication of a poor fit (Norris and Duvall, 2014). Elements that are modelled well, will have residuals between +3 and –3 and follow a normal distribution. In this study, all elements except Co were well modelled. However, Co still fell within the 'normal' range of +3 and –3 and was used in the analysis.

3 Lake Catchment Characteristics

3.1 Introduction

In the current study, each lake catchment was characterized for major ions (lakes), nutrients (lakes), and soil and sediment composition. In addition, catchment attributes, such as distance to coast, elevation, and lake:catchment ratios were evaluated as they may further influence the fate of trace elements (Halbach et al., 2017; Wieczorek et al., 2018). This chapter provides details on lake catchment characteristics (area and size), general water chemistry, and soil and lake sediment physiochemical variables for the 20 study sites (located north and east of Iqaluit, and within 10 km). Additionally, vegetation species (and diversity) were identified and described at each study site (Appendix A). The purpose of providing lake catchment characteristics is to build a database for the Iqaluit area.

3.2 Catchment Attributes

The lake catchments in this study were dominated by low arctic tundra, with several sites (25%) having some human disturbance (~50 m to disturbance) associated with them, such as: road activity (i.e., vehicle and foot traffic), construction, cell phone towers, and a sled dog team that lived year-round at one site. Based upon visual surveys of the sites, lake catchments differed slightly based on topography (steep hill gradients or low-lying hills) and landscape type (i.e., areas with glacial boulder deposits or organic soils (humus). A majority (>50%) of study sites had either glacial till or glacial boulder deposits present in the catchment. In the Iqaluit region, surficial geology is dominated by till blanket (Tb), till veneer (Tv), and monzogranite of Paleoproterozoic Cumberland batholith (Pg). A large proportion (Tv = 100%; Tb = 45%) of the sample sites are underlain by Tb (glacial till) and Tv (till that is <1.5 m thick) surficial (over bedrock) geology type, with few sites (20%) dominated by Pg (includes high-quartz plutonic,

alkali-feldspar granite, tonalite, mafic plutonic, and monzogranite rocks; Whalen et al., 2010).

Lake area ranged from 1.1–102.6 ha across the 20 study sites; median area was 3 ha, with 18 lakes <10 ha (Table 3.1). The two lakes with the largest surface area were IQ 15 (31.9 ha) and IQ 49 (102.6 ha; Table 3.1) and were located adjacent to each other east of Iqaluit. Terrestrial catchment area ranged between 2.7–497.4 ha (median of 15 ha). Lake surface area (NMAD = 50%) and catchment area (NMAD = 65%) were highly variable, while elevation (NMAD =10%) varied only slightly between the study sites (Table 3.1). Generally, larger lakes had larger catchments surrounding them (significant correlation $r = 0.69$; p-value < 0.05; Table 3).

Table 3.1: Coordinates, elevation (m a.s.l), lake area (ha), terrestrial catchment area (ha; minus the lake size), and lake:catchment ratios of 20 sample sites surrounding the Iqaluit region.

Site ID	Latitude	Longitude	Elevation (m a.s.l)	Distance to coast (km)	Lake area (ha)	Catchment area (ha)	Lake:catchment (ratio)
IQ 12	63.75639	–68.47514	128	2.4	1.3	14.5[a]	11.2:1
IQ 14	63.75787	–68.44772	156	3.3	7.9	76.8	9.7:1
IQ 15	63.75651	–68.40860	236	3.5	31.9	497.4[b]	15.6:1
IQ 17	63.78196	–68.55050	159	3.9	1.1	3.7	3.4:1
IQ 20	63.79485	–68.54530	182	5.3	2.9	12.5	4.3:1
IQ 21	63.79617	–68.53853	171	5.6	9.7	22.7[a]	2.3:1
IQ 22	63.79044	–68.54859	173	4.9	2.2	15.4	7:1
IQ 23	63.78978	–68.53420	182	5.1	1.3	6.0	4.6:1
IQ 24	63.80022	–68.55978	170	5.7	3.2	14.8	4.6:1
IQ 25	63.80573	–68.58057	129	5.7	6.4	56.0	8.8:1
IQ 34	63.79695	–68.60198	112	4.5	3.7	75.1	20.3:1
IQ 40	63.81269	–68.54939	182	7.2	1.2	9.2	7.7:1
IQ 42	63.81276	–68.54865	177	6.9	4.3	17.6	4.1:1
IQ 43	63.80871	–68.56215	163	6.7	5.4	27.6	5.1:1
IQ 45	63.76689	–68.51498	183	2.7	1.5	7.6	5.1:1
IQ 46	63.76851	–68.52488	135	2.7	1.5	8.0	5.3:1
IQ 49	63.76792	–68.42772	203	4.8	102.6	497.4[b]	4.8:1
IQ 50	63.76868	–68.46346	143	3.9	1.4	80.7	57.6:1
IQ 52	63.77085	–68.48226	122	3.6	1.8	49.0	27.2:1
IQ 65	63.76148	–68.49212	131	2.5	2.3	2.7	1.2:1
Average	—	—	162		10	29	—
Median	—	—	167		3	15	—
NMAD(%)	—	—	10		50	65	—

[a] More than one lake in the catchment that is not part of the study; [b] Lakes part of the same catchment.

3.3 Catchment Soil and Lake Sediment Characteristics

Surface soil and lake sediment in the Iqaluit region had low OM, with a median of 2.03% (soil; %w) and 0.81% (sediment; %w), with soil ranging from 1.1–17.2% and sediment ranging from 0.40–3.97% (Table 3.2 Figure 3.1 and Figure 3.2). Organic matter was significantly correlated with silt ($r_s = 0.65$; p-value <0.05) and sand ($r_s = –0.63$; p-value <0.05) in sediment, and weakly correlated with silt in soil. Organic matter also showed little variation across the study region; 65% of sites had <2.5% OM in soil, and in sediment (<1.4%). One site had an OM content of 17%, which was attributed to the

large deposits of natural organic soils in the area (Figure 3.2). Iqaluit is mostly overlain by nutrient poor soils, however organic ‘pockets’ of soils can be found throughout southeastern Baffin Island (Hodgson, 2005).

Soil weathering regimes, surface area, particle size, mineral content, and environmental factors play a role in the pH of soil (Cao et al., 2016). Median pH in soil (4.66) and sediment (5.56) were acidic to moderately acidic (Table 3.2 and Figure 3.2). Fifteen out of the 20 study sites had a soil pH <5; Figure 3.2). Soil texture, specifically clay and silt, displayed a weak positive correlation with soil pH, which suggests that particle size can influence soil pH. Bulk density (0.82–1.57 $g \cdot cm^{-3}$; median of 1.31 $g \cdot cm^{-3}$) across study sites was within the range of undisturbed soils (1.0–1.4 $g \cdot cm^{-3}$; MPCA, 2021; USDA, 2008). Overall, soil variables had low spatial variation across the study sites had a low spatial variability in soil variables (BD = 14%, OM = 25%; Table 3.2; Figure 3.3)). The highest spatial variability was found in silt (47% sediment) and clay (52% soil; Table 3.2; Figure 3.3). Soil and sediment had similar particle size composition properties, with the majority being sand (soil median: 80.8%; sediment median: 85.4%), followed by silt (soil median: 16.82%; sediment median: 13.14%), and then clay (soil median: 1.68%; sediment median: 1.51%; Figure 3.3). In general, it is worth noting that spatial variability between soil and sediment was consistent (Table 3.2).

Table 3.2: Summary statistics [5th and 95th percentile range, average, median, and normalized median absolute deviation (%NMAD)] for surface soil (Ss) and surface sediment (Sd) analytes at 20 lake catchment sites sampled in 2018 in the Iqaluit region.

Analytes		Median	Average	5th and 95th Percentile	Spatial variability (%)
pH[a] (H^+)	Ss	4.71	4.66	4.13–5.35	7
Organic matter (%)	Ss	2.03	3.00	1.36–6.45	25
Bulk density ($g \cdot cm^{-3}$)	Ss	1.31	1.19	0.82–1.57	14
Sand (%)	Ss	80.84	78.99	64.11–94.92	9
Silt (%)	Ss	16.82	18.30	4.54–31.39	35
Clay (%)	Ss	1.68	1.86	0.24–3.81	52
pH[a] (H^+)	Sd	5.45	5.56	5.04–6.24	4
Organic matter (%)	Sd	0.81	1.04	0.47–2.34	33
Sand (%)	Sd	85.39	81.94	54.62–96.63	9
Silt (%)	Sd	13.14	16.02	2.76–43.88	47
Clay (%)	Sd	1.51	1.65	0.45–2.89	35

Spatial variation was estimated as the normalized median absolute deviation (%). [a]pH is based on H^+ concentration.

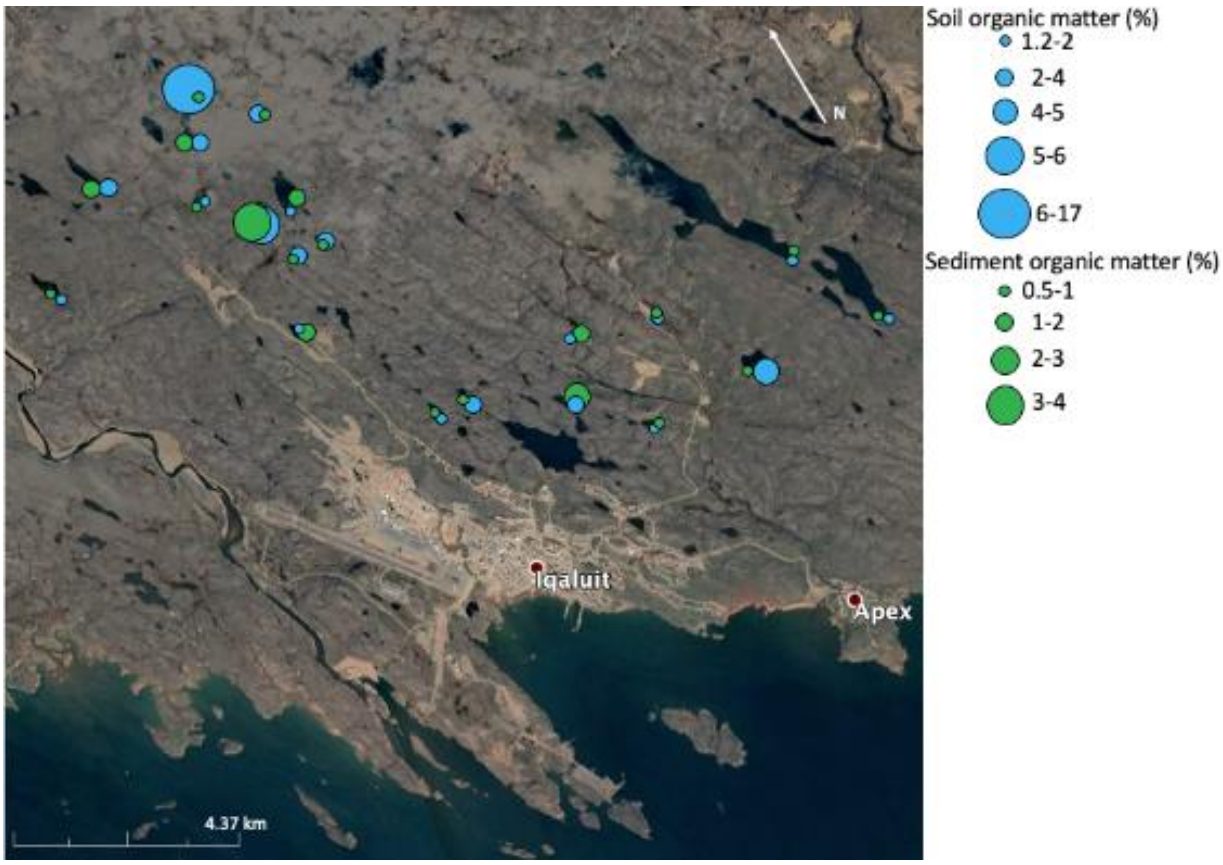


Figure 3.1: Map of soil (blue) and sediment (green) organic matter content (%) in Iqaluit across the 20 study sites (see Figure 3.2; Table 3.1).

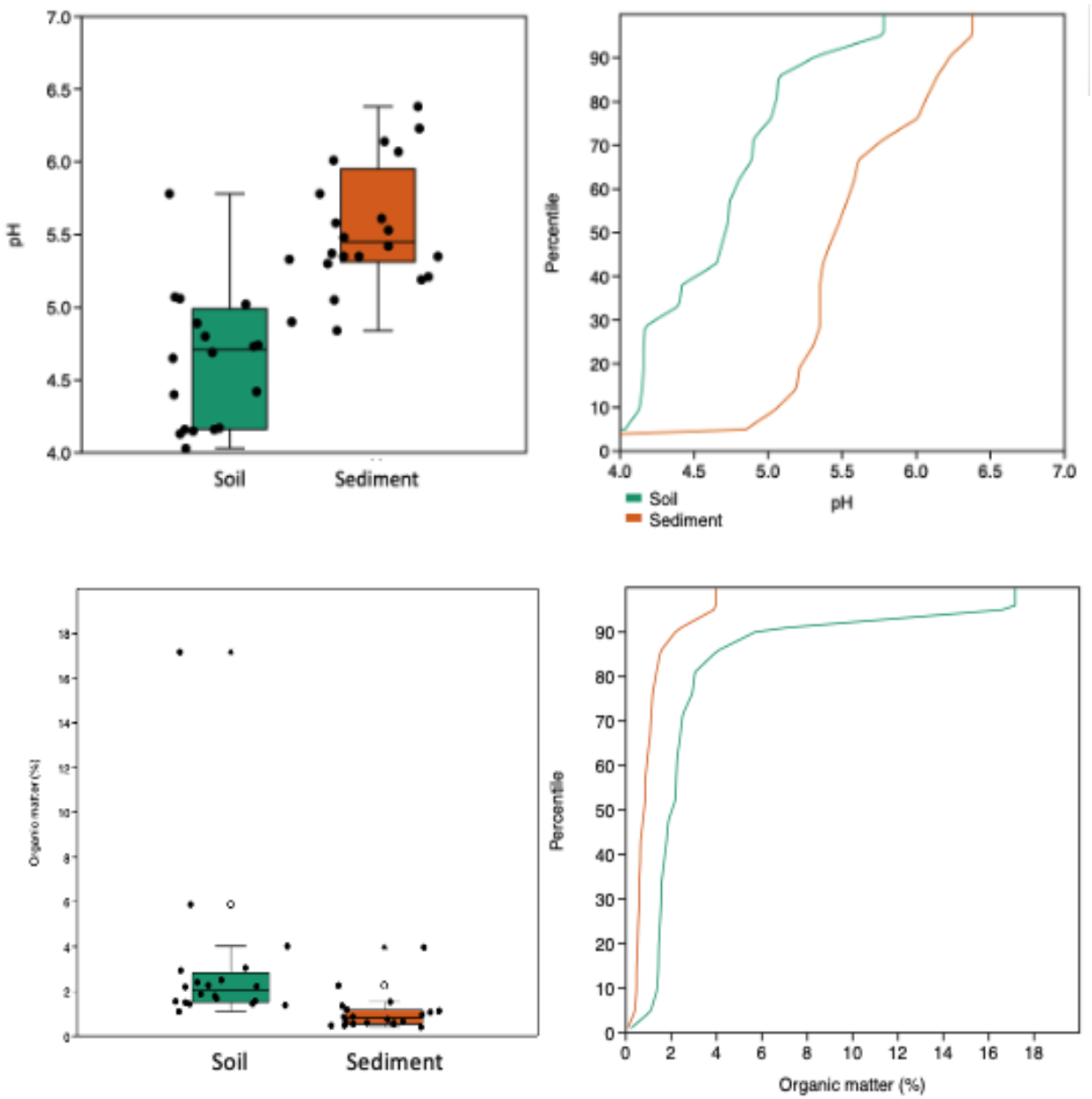


Figure 3.2: Box and jitter plot and cumulative frequency of pH and organic matter (%) in catchment soil and lake sediment for the 20 study sites located in the Iqaluit region. The purpose of the jitter is to displace the points to reduce the effect of overplotting a singular point[5].

[5] A jitter plot moves the data points horizontally so that they are not overlapping one another. This allows for better interpretation of the results.

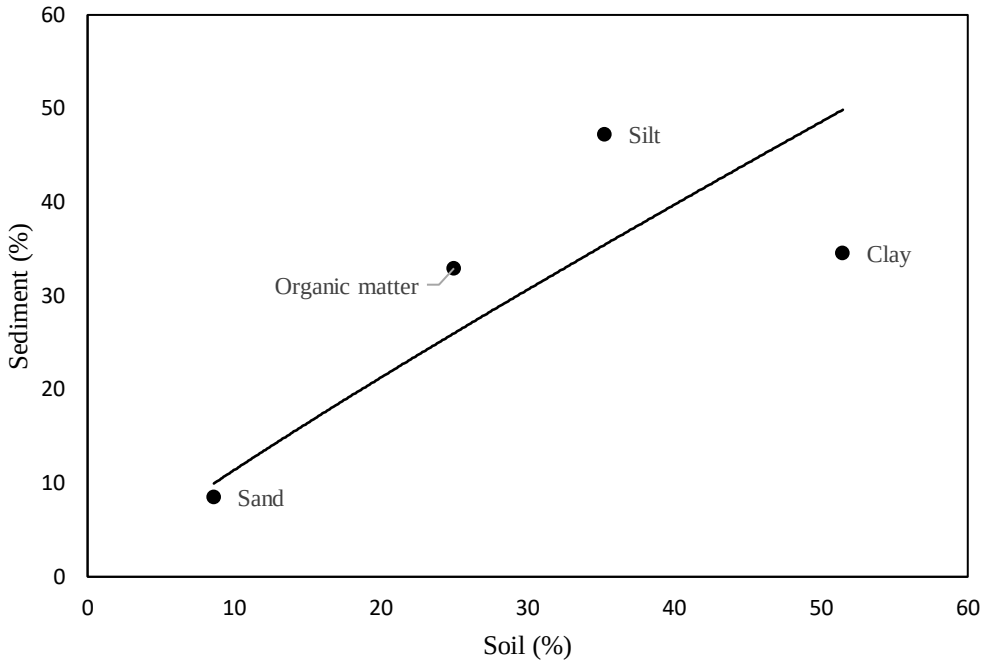


Figure 3.3: Spatial variability (%) of soil and sediment characteristics for the 20 study sites (see Table 3.2). The solid line represents the 1:1 line.

3.4 Lake Chemistry

Median lake pH was 7.34, with most sites (83%) having a pH >7. Iqaluit is underlain by acid sensitive geology, with belts of calcium running through the bedrock (Hodgson, 2005). The input of dissolved elements released from weathering directly affects pH, alkalinity and ion concentrations in lakes (Hamilton et al., 2001). In most lakes in the high Arctic, calcium carbonate is the major buffering ion, and lakes underlain by Precambrian metamorphic, intrusive and sedimentary rocks, ranged in pH from 5.8 to 8.1 on Baffin Island, Devon Island and Ellesmere Island (Hamilton et al., 2001). A significant positive correlation was found between pH and Ca^{2+} ($r_s = 0.91$), K^+ ($r_s = 0.73$), and DIC ($r_s = 0.85$), which suggests that pH is influenced by carbonate content and potassium feldspar in the bedrock geology of the catchment (Hodgson, 2005). Despite the lakes coastal location and high pH, they were dilute as evidenced by their low

conductivity (median = 46.2 $\mu S \cdot cm^{-1}$), which varied little between years (14%) and across the study lakes (23%; Table 3.3). Further, conductivity was more strongly significantly correlated with Ca^{2+} (r_s = 0.94), DIC (r_s = 0.87), K^+ (r_s = 0.80), Mg^{2+} (r_s = 0.60), and SO_4^{2-} (r_s = 0.59), compared to Na^+ (r_s = 0.18) and Cl^- (r_s = –0.15), suggesting that weathering of bedrock geology had more of an influence on conductivity than sea-salt aerosols. However, given the low conductivity, this suggests that the catchments are underlain by slow weathering bedrock (e.g., Ca^{2+}, K^+, Mg^{2+}, and SO_4^{2-} are released).

Ratios between Na^+ and Cl^- can be used to assess the presence of marine aerosols. A ratio of <0.85 (based on units of μeq/L; Möller, 1990) can be used to assess the inputs of marine aerosols. Out of the twenty lakes, only one (IQ 46) site had observations for Na^+ and Cl^-, where median Na^+:Cl^- ratios (0.74) were lower than 0.85, which suggests a marine aerosol influence (Möller, 1990). Distance to coast was significantly negatively correlated with Na^+ (r_s = –0.66) and Cl^- (r_s =–0.57), and study sites that were located farther from the coast had low concentrations of Na^+ and Cl^- (Figure 3.4). The prevailing wind direction for the region, originates from the northwest, so while the lakes are near a marine source, they do not necessarily receive high marine aerosol inputs. Generally, <30% of wind direction and windspeed (7−11 knots) originated from the southeast (coast) of Iqaluit between 2018 and 2019 (see WindRose Plot; Appendix F). DOC (median of 2.77 $mg \cdot L^{-1}$) varied equally between years (24.2%), and across the study sites (23.3%) and was significantly positively correlated with Cl^- (r_s = 0.42) and Na^+ (r_s = 0.40). DOC may be entering the catchment through surface runoff or windblow soil particles.

The median concentration of major ions from highest to lowest during 2018–2019 was: $Ca^{2+} > SO_4^{2-} > Mg^{2+} > Cl^- > Na^+ > K^+ > NO_3^- > PO_4^3$ (Table 3.3; Figure 3.5). Median cation and anion concentrations were within the lower range of inland freshwater systems in Canada and the Arctic (McNeely et al., 1979; Liang, 2018). In this study, major cations and Cl^- were below the concentrations found in background studies conducted in high Arctic Nunavut, Northwest Territories and Yukon (Liang, 2018), while SO_4^{2-} was comparable (median = 2.35 $mg \cdot L^{-1}$; Liang, 2018 median = 3.10 $mg \cdot L^{-1}$). Generally, major ion concentrations, and to a lesser extent DOC, increased with increasing lake area and catchment area (Appendix G). Potassium ($r_s = 0.41$) was associated with lake area, and SO_4^{2-} ($r_s = 0.42$), Ca^{2+} (0.61), and K^+ (0.67) were related to catchment area, suggesting terrestrial catchment (weathering) sources compared to within lake sources for K^+. Major anions (NO_3^-, PO_4^{3-}, SO_4^{2-}) showed the highest degree of spatial variability (> 48%), while all other lake chemistry analytes remained low (< 30%; Table 3.3). Low concentrations typically have higher variability as slight increases in a value can influence the variability by several orders of magnitude (Birch et al., 2001). This results in large variability between study sites and years (e.g., median PO_4^{3-} = 0.02 $\mu g \cdot L^{-1}$, resulting in a high spatial (88%) and interannual (190%) variability). Spatial variability and interannual variability were significantly positively correlated for major ions ($r_s = 0.74$; Figure 3.5), analytes that varied between sites also varied between years (Table 3.3). Overall, lake water chemistry varied more between years then between lakes.

Table 3.3: Summary statistics (minimum–maximum range, average and median) for water chemistry variables of the study sites (n=20) during 2018–2019.

Analytes	Median ($mg \cdot L^{-1}$)	Average ($mg \cdot L^{-1}$)	5th and 95th percentile	Spatial variability (%)	Interannual variability (%)
Alkalinity	16.96	18.15	10.44–26.35	29	10
pH[a]	7.34	7.28	7.00–7.51	31	28
Conductivity ($\mu S \cdot cm^{-1}$)	46.23	45.73	28.41–64.94	23	14
Ca^{2+}	7.13	7.15	3.91–11.61	31	4
K^{+}	0.17	0.17	0.11–0.24	14	29
Mg^{2+}	0.95	0.95	0.66–1.14	20	11
Na^{+}	0.74	0.74	0.60–0.86	11	4
Cl^{-}	0.91	0.90	0.71–1.16	10	23
NO_3^{-}	0.04	0.10	0.01–0.19	75	146
PO_4^{3-} ($\mu g \cdot L^{-1}$)	0.02	0.20	0.01–0.25	88	190
SO_4^{2-}	2.35	2.94	0.91–5.64	50	57
DOC	2.77	3.68	1.20–2.65	23	24
DIC	1.09	2.07	1.86–5.42	24	17
TDN	0.39	1.07	0.09–0.18	10	61
NH_3	0.03	0.05	0.02–0.12	77	65
TP[b] ($\mu g \cdot L^{-1}$)	11.29	12.14	8.67–15.62	16	–

Spatial variation was estimated as the normalized median absolute deviation (%). Interannual (2018–2019) variability was estimated as the average absolute relative percent difference. [a]Spatial and interannual variability is based on H^{+} concentration. [b]Based on one sampling period (2019).

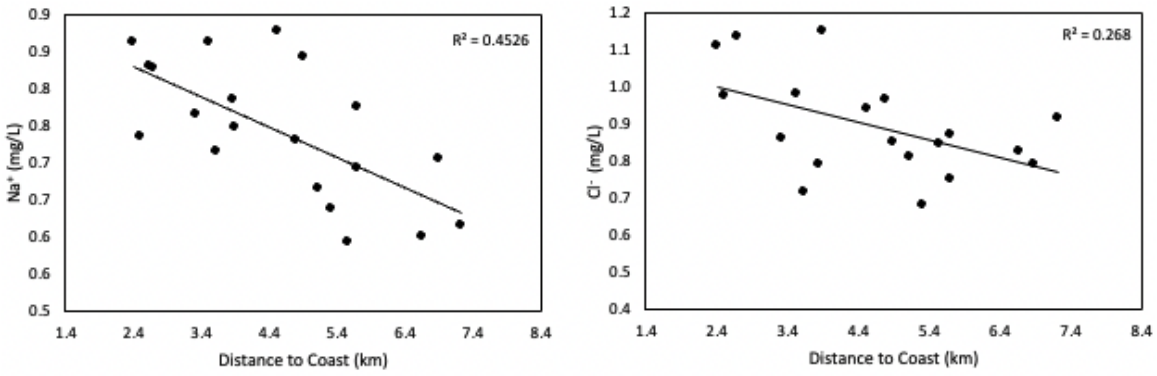


Figure 3.4: Scatterplots showing the relation between Na^{+} and Cl^{-} and distance to coast. Study sites located farther from the coast have a lower concentration.

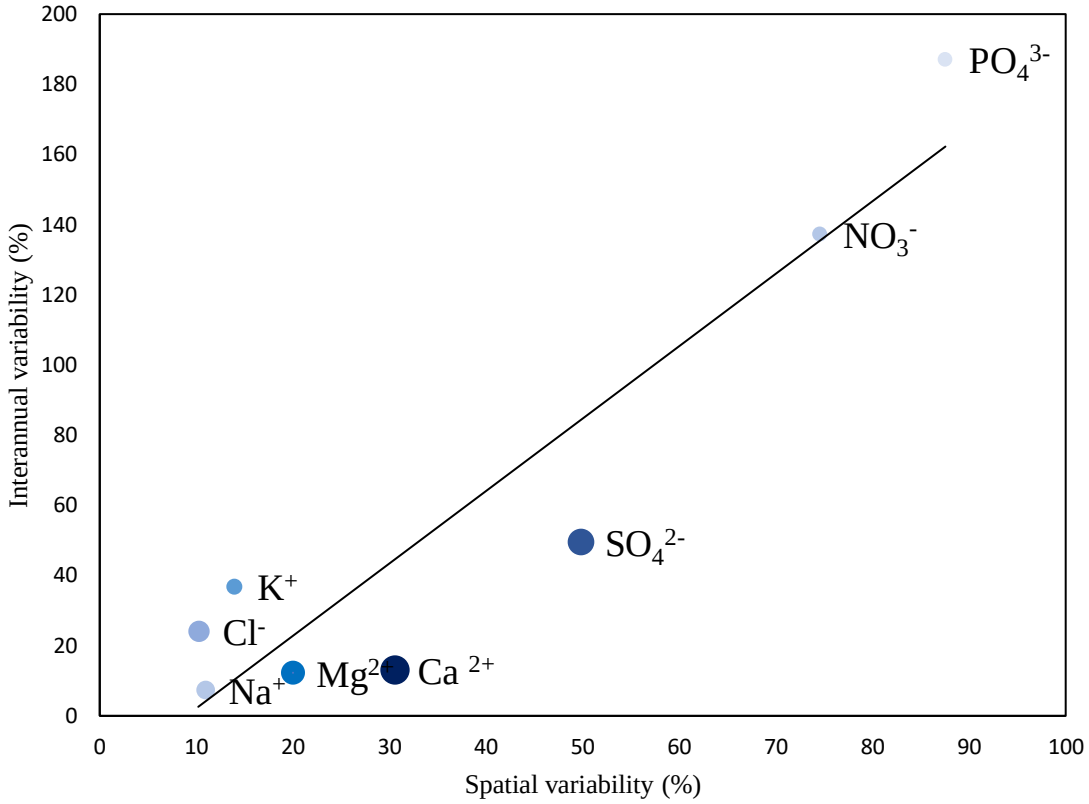


Figure 3.5: Relationship between spatial variability (%) and interannual variability (%) of the major ions (Spearman correlation rs = 0.74; p-value = <0.05). The 1:1 solid line is shown for reference.

3.5 Terrestrial and Aquatic Associations

A PCA was used to show which variables explain the variability in the data based on their similarity within water chemistry (pH, conductivity and DOC, major ions, and nutrients), catchment soil and lake sediment datasets. The first four principal components (PC; Appendix H) explained ~64% (PC1 = 26.1%; PC2 = 16.7%; PC3 = 11.0%; PC4 = 10.1%) of the variation amongst the catchment data (Figure 3.6). The variables that were associated with PC1 were pH, conductivity, DIC, Ca^{2+}, Mg^{2+}, and K^+, suggesting that weathering of geogenic (carbonate) material influences lake chemistry. The second axis PC2 was associated with Cl^-, Na^+, and sand, silt and clay particles in catchment soil,

reflecting marine inputs and potentially reflecting buried marine sediments. Baffin Island has been subjected to repeated glaciation episodes causing part of the land to sink into the ocean (Deering et al., 2021). Most recently (~11,000 years ago), the Laurentide Ice Sheet retreated from the southeast corner, resulting in isostatic uplift of the island, meaning as the ice sheet retreated the land rebounded to its former level (Miller et al., 1980; McCann et al., 1981; Jacobs et al., 1985; Figure 3.7). The third axis PC3 was mainly associated with sediment variables, such as OM, silt and sand, and TP and K^+ (lake), reflecting internal loading for nutrients. Lastly, the fourth axis PC4 was related to DOC, Na^+ and Mg^{2+}, lake area, catchment area, and elevation, suggesting that sea-salt aerosols (through wind) are a source (but not dominant) to the catchments. Nutrients (TP, NH_3 and TDN) had a stronger correlation with soil bulk density and lake sediment (silt, clay, and OM) particles; in contrast, major ions were correlated more with soil texture (Figure 3.6 and Figure 3.7). This is most likely due to nutrients binding to soil and sediment particles within the terrestrial catchments. Inputs of P into lakes is usually from weathering of bedrock and leaching from soil (Vonk et al., 2015).

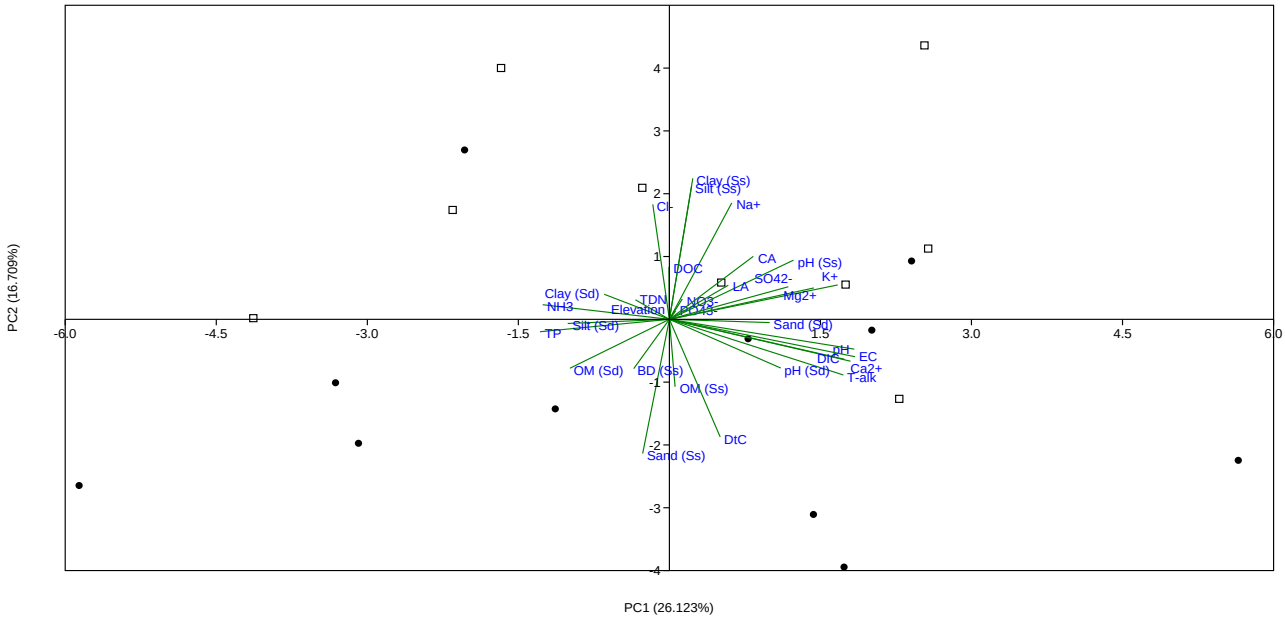


Figure 3.6: Biplot of the first two principal components (PC) of lake water (average between 2018 and 2019), surface soil (Ss) and surface sediment (Sd) data (listed in Table 3.1 and Table 3.2) physiochemical variables, and elevation (m a.s.l), lake area (ha), catchment area (ha) and distance to coast (km). Lake water was sampled in 2018–2019, while soil and sediment data were collected in 2018.

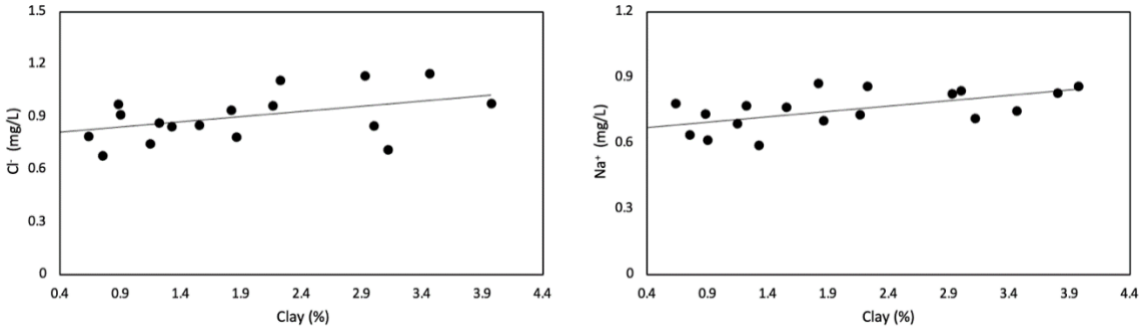


Figure 3.7: Scatterplots showing the relationship between Na^+ and Cl^- (mg/L) and clay soil particles.

4 Trace Elements in Arctic Lake Catchments: Concentrations and Sources

4.1 Introduction

The Canadian Arctic receives atmospheric and oceanic transport of trace elements from the northern hemisphere, potentially negatively impacting its ecosystems (Halbach et al., 2017; AMAP 2005). Aluminum and Fe are usually from geogenic sources and are often used as indicators of crustal material (Wu et al., 2021). Cadmium, Pb, Hg, As, Zn, Ba, Cr, V, Cu, Ni, Co, Se, and Sb are typically associated with anthropogenic sources (Wu et al., 2021), however some of these elements (As, V, Ba, Zn) occur naturally in bedrock.

Inland waters in remote regions are considered to be sentinels of terrestrial and atmospheric processes because they are linked through changes in the catchment (geologic weathering), transport, nutrients, elements and contaminants (storing and mobilizing; Host et al., 1997; Johnson et al., 2004; Wiley et al., 1997; McRae et al., 2004; Williamson et al., 2008). This study sampled twenty lake catchments in Iqaluit to determine trace element concentrations, their spatial distribution across study sites, and across several environmental compartments within each catchment. Further, relationships between environmental compartments and trace elements concentrations were investigated.

This chapter provides information on the concentration of trace elements (n = 16) in four different environmental compartments (lakes, sediment, soil, and moss) in each of the study catchments. Trace element concentrations were then evaluated for variability between study sites (using %NMAD) and years (to provide a larger dataset; using %RPD; used only for lakes and moss), and related to environmental variables such as,

DOC (OM), pH, and EC (water) using a PCA. These variables were chosen because they have been known to influence trace element cycling (Lydersen et al., 2002).

Enrichment factors (EF) in moss were calculated to determine elements from anthropogenic sources as trace element concentrations in moss reflect atmospheric sources (wind-blown soil particles and wet and dry deposition). As previously stated, the use of soils in assessing EFs in moss tissue has been widely used in moss biomonitoring studies (Dragovic and Mihailovic, 2008; Klos et al., 2011; Macedo-Miranda et al., 2016; Cowden, 2018). The potential sources for each trace element in lakes, soil, sediment, and moss was assessed using the PMF source apportionment model. Where previously noted, the PMF model is helpful in identifying potential natural (geogenic) and industrial sources of trace elements (Wu et al., 2021; Isley et al., 2018; Isley and Taylor, 2020).

4.2 Trace Element Concentrations in Water, Sediment, Soil, and Moss

Sixteen trace elements were analyzed in lake water (Sw = surface water), catchment soil (Ss = surface soil), lake sediment (Sd = surface sediment), and moss (Ms; *Hylocomium splendens*; Table 4.1–4.4). Trace elements in the Iqaluit region are primarily from current and legacy anthropogenic sources and geogenic sources (combustion of fossil fuels), which may be available (free species) or bound to organic matter. Geogenic sourced elements (Fe>Al) had the highest median concentrations throughout the media and across the study catchments (Table 4.1–4.4), e.g., Fe [22,401 $mg \cdot kg^{-1}$ (Sd); 15,254 $mg \cdot kg^{-1}$ (Ss); 536 $mg \cdot kg^{-1}$ (Ms); 23 $\mu g \cdot L^{-1}$ (Sw)], and Al [5678 $mg \cdot kg^{-1}$ (Sd); 4781 $mg \cdot kg^{-1}$ (Ss); 400 $mg \cdot kg^{-1}$ (Ms); 17 $\mu g \cdot L^{-1}$ (Sw)], suggesting that geologic input is a governing source (Appendix I). Additionally, soil pools were calculated for trace elements (Appendix I), with the largest pools being: for Fe (958.50 $g \cdot m^{-2}$), Al (297.81 $g \cdot m^{-2}$), and Ba (2.18 $g \cdot m^{-2}$), showing that geogenic elements are dominant in the

catchments. Soil and sediment followed the same median rank order (and were significantly positively correlated $r_s = 0.95$; Figure 4.1) for most trace elements (except for Cr, V and Cu). Overall, sediment had higher concentrations of all elements compared to soil. It is recommended that sediment should be continued to be sampled over soil since both compartments have similar concentration orders. Sediments act as a sinks and sources (through adsorption of materials in suspension) of metals and contaminants in aquatic systems (Rainey et al., 2003; Wardhani et al., 2022). Soil (2.03%) organic matter was higher at the study sites compared to sediment (0.81%), allowing for increased metal binding (uptake). Lake (surface) water and moss had the same median ranking order for Fe, Al, Zn, Ba, and Cu. These elements are added into the study sites through weathering of bedrock, leaching of surface soils, or atmospheric deposition.

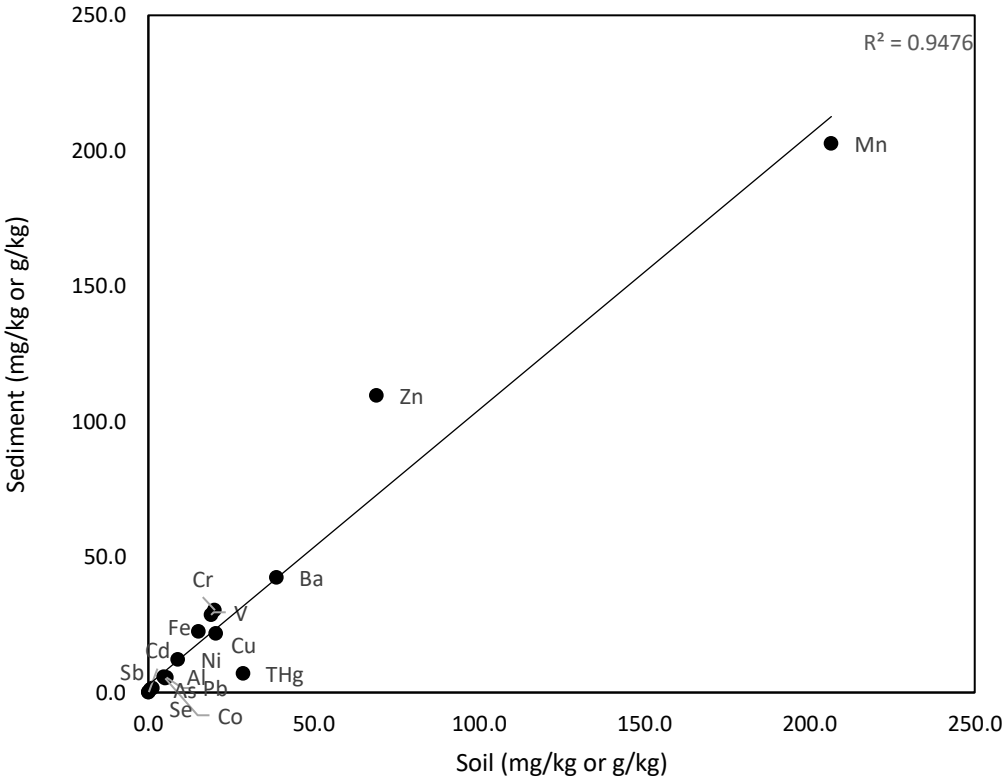


Figure 4.1: Scatterplot showing a significant regression analysis (R2 = 0.95) between trace element (mg·kg^{-1} or g·kg^{-1}) rank order in soil and sediment. Note Al and Fe (g·kg^{-1}) were rescaled (divided by 1000) to fit plot.

4.2.1 Trace elements in lake water

In lake water (n = 20), Fe had the highest median concentration (26.54 µg·L^{-1}), followed by Al (21.60 µg·L^{-1}), Zn (8.50 µg·L^{-1}), and Ba (4.02 µg·L^{-1}; Table 4.1). Compared to anthropogenic sourced elements (e.g., Cd, Pb, As, Hg, Zn, Sb, V), geogenic element concentrations were at least one order of magnitude higher in surface water samples (e.g., Fe (26.54 µg·L^{-1}) is one order of magnitude higher than Pb (0.31 µg·L^{-1}); Table 4.1). Trace elements that have already been associated with human activities, such as As (and local geology), Cd, Pb and THg (Wu et al., 2021) showed concentrations of 0.05, 0.02, 0.31 µg·L^{-1}, and 3.85 ng·L^{-1}, respectively. These concentrations were within range of other Arctic studies (Crane et al., 2001; Camarero et al., 2009; Lou et al., 2013).

Approximately half of the elements had a spatial variability <35%, with Al (48%), Co (59%), Fe (51%), Mn (49%), Sb (56%), and Zn (45%) >35%; geologic variation is attributed to the study sites high variability. Additionally, low variability within the lakes is attributed to the homogeneity across the region. In general, interannual variability (2018 versus 2019 across all lakes) was slightly higher with more elements >35% compared to spatial variability (Table 4.1).

Table 4.1: Summary statistics (5^{th} and 95^{th} percentile range, average, median and spatial and interannual variability) for sixteen trace elements in lake water sampled in 2018 and 2019.

Elements	Median ($\mu g \cdot L^{-1}$)	Average ($\mu g \cdot L^{-1}$)	5^{th} and 95^{th} percentile ($\mu g \cdot L^{-1}$)	Spatial variability (%)	Interannual variability (%)
Al	21.60	28.06	10.68–72.80	48	62
As	0.05	0.05	0.04–0.07	23	46
Ba	4.02	3.94	1.96–5.59	36	35
Cd	0.02	0.02	0.01–0.04	42	41
Co	0.04	0.22	0.02–0.63	59	22
Cr	0.14	0.16	0.10–0.25	25	46
Cu	2.46	3.22	1.13–9.44	36	24
Fe	26.54	33.64	5.99–94.20	51	35
Mn	1.37	1.71	0.64–3.28	49	57
Ni	0.28	0.29	0.19–0.43	32	70
Pb	0.31	0.36	0.20–0.57	35	52
Sb	0.03	0.04	0.02–0.06	56	67
Se	0.04	0.04	0.03–0.05	30	60
THg ($ng \cdot L^{-1}$)	3.85	4.82	2.28–12.57	28	—
V	0.03	0.04	0.02–0.08	26	35
Zn	8.50	11.90	5.15–28.08	45	39

Spatial variation was estimated as the normalized median absolute deviation (%). Interannual (2018–2019) variability was estimated as the average absolute relative percent difference.

4.2.2 Trace elements in lake sediment

As expected, trace elements with the highest concentration in surface sediment were also associated with geogenic catchment sources, e.g., median Fe (22,401 $mg \cdot kg^{-1}$), Al (5678 $mg \cdot kg^{-1}$), Mn (203 $mg \cdot kg^{-1}$), and Zn (110 $mg \cdot kg^{-1}$; Table 4.2). Sediment did not follow the same order of concentration as the lake water, however geogenic elements

dominated both environmental compartments, e.g., Fe, Al, Mn, and Zn in sediment (Table 4.2) compared to Fe (26.54 µg·L^{-1}), Al (21.60 µg·L^{-}1), Zn (8.50 µg·L^{-}1) and Ba in lakes (Table 4.1). The difference in the order of magnitude is likely because sediments act as a sink, collecting and storing elements over the long term, while lake chemistry changes seasonally owing to hydrology fluctuations, and their short retention time (Horowitz, 1985b). Generally, spatial variability was relatively low (<35%); THg had the highest variability of 40%, followed by Al, As, Ba and Ni (Table 4.2). Spatial variability was lower in sediment than in lake water. This is to be expected as sediment reflects long-term storage and accumulation, with little changes seasonally. Sediment also contains a historical record of chemical conditions (Horowitz, 1985b). Furthermore, smaller concentrations of trace elements will show a higher proportional variability.

Table 4.2: Summary statistics (5th and 95th percentile range, median, average, and spatial variability) for sixteen trace elements in sediment sampled in 2018.

Elements	Median ($mg \cdot kg^{-1}$)	Average ($mg \cdot kg^{-1}$)	5^{th} and 95^{th} percentile ($mg \cdot kg^{-1}$)	Spatial variability (%)
Al	5677.89	6348.63	3257–10,064	37
As	1.14	1.20	0.52–3.30	37
Ba	42.44	46.20	23.17–97.76	37
Cd	0.15	0.16	0.05–0.48	26
Co	5.21	6.78	2.94–11.41	29
Cr	30.39	29.34	11.93–49.25	21
Cu	21.63	23.05	10.50–53.84	31
Fe	22401.10	24781.78	10,349–37,832	29
Mn	202.60	249.00	93.93–498.67	34
Ni	12.19	13.61	5.90–25.89	37
Pb	5.54	6.05	2.62–11.27	31
Sb	0.02	0.04	0.01–0.34	33
Se	1.64	1.75	0.71–2.75	29
THg ($ng \cdot g^{-1}$)	6.90	8.98	1.40–20.84	40
V	28.64	29.58	11.89–51.84	18
Zn	109.60	106.33	69.66–147.15	20

Spatial variation was estimated as the normalized median absolute deviation (%). Sediment was sampled in 2018.

4.2.3 Trace elements in catchment soil

In catchment soil, Fe (15,253 $mg \cdot kg^{-1}$), Al (4781 $mg \cdot kg^{-1}$), Mn (207 $mg \cdot kg^{-1}$) and Zn (69 $mg \cdot kg^{-1}$) had the highest median concentrations (Table 4.3), which was consistent with sediment (Table 4.2). As previously mentioned, geogenic sources in the catchments are greater than external anthropogenic sourced elements. Organic matter in soil (2.03%) was slightly higher compared to sediment (0.81%), which likely influenced the concentration of organic bound trace elements (i.e., THg in soil = 28.7 $ng \cdot g^{-1}$ vs. THg in sediment = 6.9 $ng \cdot g^{-1}$) in the study sites (Table 4.2). Overall, spatial variability in trace elements was low in soil and sediment, as they reflect long term accumulation of trace elements from weathering and deposition. Similarly, soil and sediment follow the same concentration order, reflecting similar weathering and deposition rates. Most elements had a spatial variability <35% (Table 4.3). Antimony (50%), THg (47%), Mn (46%), V

(43%), Cr (41%), Ni (41%), and Cu (37%) had the highest spatial variability (Table 4.3) in soil. In general, trace element concentrations showed a consistent pattern between lake water, sediment and soils. Furthermore, there were no distinguishable directional gradient, i.e., trace element concentrations were not higher in study sites located to the north or east of Iqaluit.

Table 4.3: Summary statistics (5th and 9th percentile range, median, average, and spatial variability) for sixteen trace elements in catchment soil sampled in 2018.

Elements	Median ($mg \cdot kg^{-1}$)	Average ($mg \cdot kg^{-1}$)	5^{th} and 95^{th} percentile ($mg \cdot kg^{-1}$)	Spatial variability (%)
Al	4781.01	5935.76	2704–12,890	32
As	0.78	2.69	0.43–10.97	32
Ba	38.79	78.63	20.44–179.82	22
Cd	0.14	0.47	0.08–0.84	31
Co	5.12	6.37	2.41–14.34	34
Cr	20.15	26.08	8.81–62.91	41
Cu	20.54	50.62	7.60–84.42	37
Fe	15253.56	20836.68	8418–40,337	35
Mn	206.62	445.53	0.11–1.16	46
Ni	8.98	14.81	5.06–30.95	41
Pb	5.47	71.98	0.00–0.10	31
Sb	0.04	0.11	0.02–0.16	50
Se	1.24	1.34	0.71–2.19	27
THg ($ng \cdot g^{-1}$)	28.71	31.65	6.78–62.98	47
V	18.98	21.81	6.65–32.93	43
Zn	69.16	185.38	43.58–343.52	26

4.2.4 Trace elements in moss **(***Hylocomium splendens*)

The highest median concentrations in moss were Fe (536.62 $mg \cdot kg^{-1}$), Al (462.28 $mg \cdot kg^{-1}$), Zn (115.41 $mg \cdot kg^{-1}$), and Ba (27.39 $mg \cdot kg^{-1}$). Lakes also followed this concentration order (Table 4.1 and 4.4). Trace element concentrations in moss are representative of atmospheric (wet and dry deposition) sources, suggesting that lakes may also show the same patterns. Deposition is highly influenced by wind-blown dust and pollutants, which may further explain the similarity between moss and lakes. Generally, interannual variability was high for Co (70%), Ni (61%), Cu (49%) and Zn (44%), while

THg (19%) and Fe (19%) were low (Table 4.4). Low interannual variability in iron suggests consistent sources (e.g., wind-blow and weathering) each year, while THg has a relatively long atmospheric lifetime (several months; Halbach et al., 2017). All environmental compartments had low spatial variability (e.g., most trace elements were <35 % variability), with lake sediment having the lowest. Lake sediment integrates the heterogeneity of soils, while water, sediment, soil, and moss reflect homogenous sources (e.g., soil weathering, geology and atmospheric deposition) across the region. Cobalt (54%; driven by low concentrations) and Al (37%; reflecting geogenic variation) had the highest spatial variability, while THg (15%), As (20%), and Se (22%) had the lowest.

Table 4.4: Summary statistics (5th and 9th percentile range, median, average and spatial and interannual variability) for sixteen trace elements in moss (Hylocomium splendens) sampled in 2018 and 2019.

Elements	Median ($mg \cdot kg^{-1}$)	Average ($mg \cdot kg^{-1}$)	5^{th} and 95^{th} percentile ($mg \cdot kg^{-1}$)	Spatial variability (%)	Interannual variability (%)
Al	462.28	457.41	209–689	37	39
As[a]	0.11	0.12	0.08–0.18	20	—
Ba	27.39	30.09	15.32–61.13	25	30
Cd	0.24	0.24	0.15–0.33	26	38
Co	0.44	0.54	0.21–0.97	54	70
Cr[a]	0.85	1.12	0.45–2.87	35	—
Cu	16.93	19.38	9.68–33.85	32	49
Fe	536.62	682.93	297–1580	32	19
Mn	0.13	0.16	0.06–0.41	33	34
Ni	2.99	3.45	2.04–5.44	31	61
Pb	2.37	3.22	1.72–5.65	25	36
Sb[a]	0.11	0.20	0.07–0.62	28	—
Se[a]	0.24	0.26	0.13–0.49	22	—
THg ($ng \cdot g^{-1}$)	42.98	44.97	37.25–60.31	15	19
V	0.70	0.86	0.39–1.98	34	28
Zn	115.41	125.57	63.14–215.38	32	44

[a]Based on one sampling period.

4.3 Enrichment Factors in Moss

The median EFs for each element across the study sites ranged from 0–581 (Table 4.5). Generally, the higher the EF the more anthropogenic or enriched the element is,

while a lower EF value suggests a geogenic (natural) source. Barium, Cd, Cu, Pb, and Sb were the only elements that were highly enriched (EF >9; Figure 4.2 and Table 4.5), suggesting that they are of anthropogenic origin. Total mercury is mostly associated with atmospheric pollution although it wasn't identified as being enriched. Potential reasons include: low THg concentration was not recognized through EF equation or that THg is from legacy sources and comes from the redistribution of soil bound THg. Antimony (95%), Cd (85%) and Cu (50%) had the greatest percentage of study sites that were highly enriched compared to the other elements. Anthropogenic sources of Sb are fossil fuel combustion, including sources of heat generation in the winter and diesel generators as a power source). Nearly half of the trace elements showed a degree of enrichment (46%), while 56% were not enriched (EF value <3), suggesting that they are predominantly of soil origin (wind-blown dust containing geogenic and anthropogenic elements). Study sites located the farthest from the Iqaluit (north and east) had high EFs, compared to sites closer to the city (Figure 4.2). Prevailing winds (study sites to the north will receive the winds first) or elevation (study sites to the east are higher in elevation) of the study sites may 'intercept' the deposition.

Figure 4.2: The location of study sites and study sites with high enriched factors (>9) in moss tissue trace element concentrations (pink star) and their corresponding elements.

Table 4.5: Mean moss (2018–2019) enrichment factors and degrees of enrichment per element with Al as the common soil element.

	Enrichment factor		Percentage (%) of sites enriched (*n*=20)		
Element	Mean	Degree of enrichment	Not enriched (<3)	Enriched (3–9)	Highly enriched (>9)
Al	—	—	—	—	—
As[a]	2	not	85	10	5
Ba	9	slightly	5	70	25
Cd	20	highly	5	10	85
Co	1	not	100	0	0
Cr[a]	1	not	100	0	0
Cu	16	highly	10	40	50
Fe	0	not	100	0	0
Mn	0	not	100	0	0
Ni	4	slightly	10	90	0
Pb	6	slightly	10	80	10
Sb[a]	87	highly	0	5	95
Se[a]	3	not	55	45	0
THg	0	not	100	0	0
V	1	not	100	0	0
Zn	0	not	100	0	0

[a]Based on one year of sampling.

4.4 Trace Element Associations with Environmental Variables

Spearman's correlation was used to determine potential associations between trace elements, soil texture, and general water chemistry for lake water, sediment, and catchment soil. Clay, silt, and sand particles in soil showed little to no relationships with trace element concentrations. Most notably, As (r_s = 0.24; soil) was the only element to be weakly positively correlated with soil clay particles, while SOM was weakly correlated with Cd (r_s = 0.36) and THg (r_s = 0.34) in soil. In sediment, silt particles were positively significantly correlated with Cd (r_s = 0.64), Cu (r_s = 0.65), Se (r_s = 0.55), and THg (r_s = 0.69). As previously noted, sediment acts as both a sink and a source to trace elements in lake water. Fine and coarse grain particles readily adsorb elements onto their surface (Rowe et al., 2004). Cadmium, Cu and THg suggest anthropogenic sources,

whereas Se is representative of geochemical cycling through biological sources and of fossil fuel combustion (Winkel et al., 2015).

In lakes, most trace elements showed little correlations with general water chemistry. Aluminum, As, Ba, Cu, and V had the largest number of significant correlations with major ions. Total mercury (r_s = 0.35 and 0.37), Sb (r_s = 0.43 and r_s = 0.60) and Mn (r_s = 0.42 and r_s = 0.30) were correlated with nutrients (NH_3 and TP) and DOC respectively, suggesting that organic matter influences element and nutrient binding.

Environmental variables such as OM (DOC) and pH can influence trace element cycling, potentially influencing element concentrations as they change over time (Nelson and Aherne, 2020; Lydersen et al., 2002; Asmala et al., 2019) in environmental chemistry. Major ions such as Ca^{2+} are used as indicators of geologic weathering (Hamilton et al., 2001). Factor analyses are often used to help interpret relationships between variables in large datasets. PCAs are a type of factor analysis, which are often used to interpret trace elements and environmental variables. Multivariate associations were used to explore relationships between environmental variables (pH, Ca^{2+}, DOC, or OM) and trace element concentrations in lake water, lake sediment and catchment soil.

In lake water, the first four PCs explained 68% (PC1 = 33.30%, PC2 = 14.05%, PC3 = 11.12%, PC4 = 9.51%; Appendix J) of the variation (Figure 4.3). Variables that influenced PC1 were Al, V, Co, Pb, Ba, and Cr, suggesting that elements clustered with Al are of geogenic origin (runoff from the catchment). Variables that influenced PC2 were Cu, THg and Zn (Figure 4.4). As previously discussed, Cu had high EF values in moss tissue, suggesting that deposition from anthropogenic sources is the main pathway.

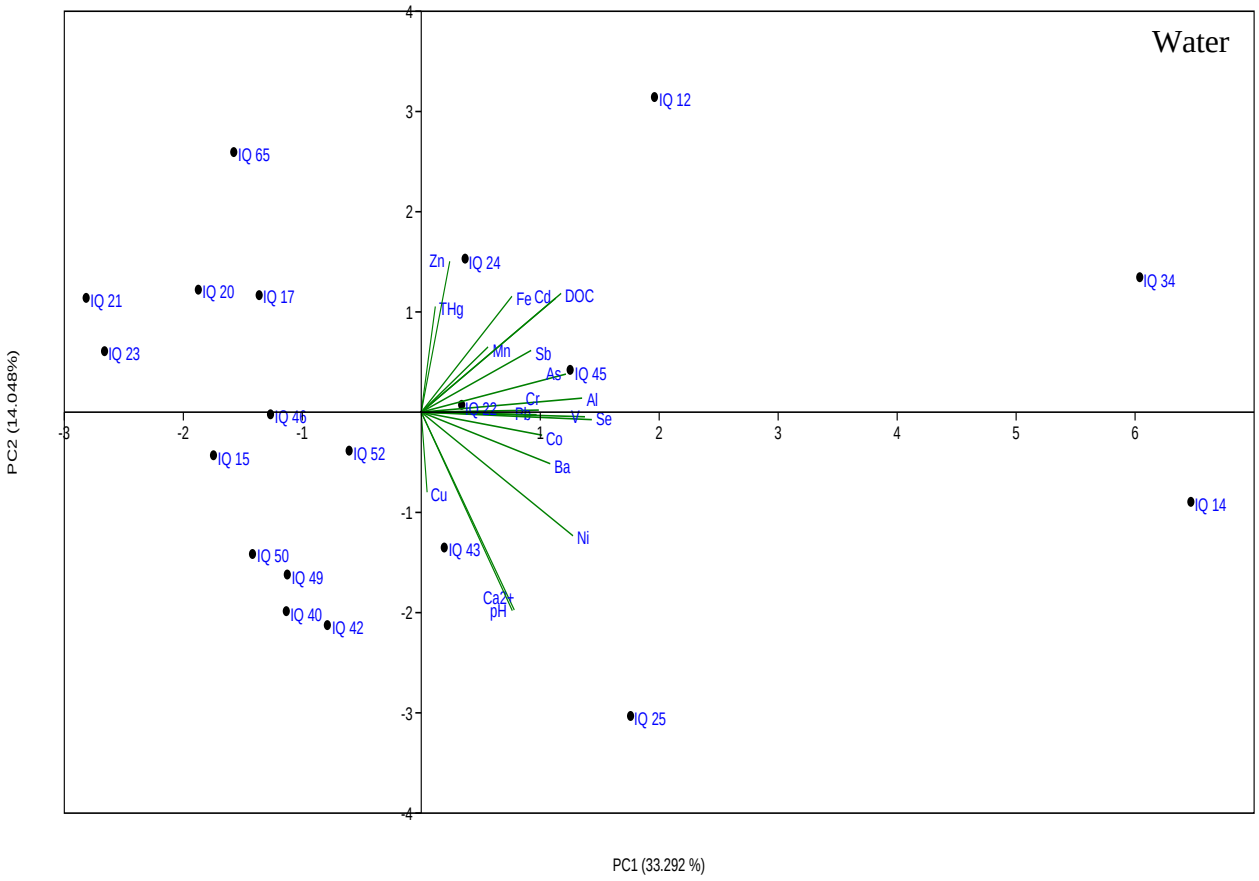
DOC had a high positive loading with Fe, Cd and Sb, and to a lesser extent Mn, indicating that a large fraction of total Fe, Cd, Sb, and Mn are organic bound. pH had low positive loadings for As, Ba, Se, Al, V, Co, Pb, Cr, and Ni, but was highly correlated with Ca^{2+}, suggesting that pH plays a minor role in the availability or speciation of elements to the surrounding catchments, and that pH could be influenced by geology through Ca^{2+} weathering.

The first two PCs in lake sediment explained 74% of the variation (PC1 = 46% and PC2 = 28%). Variables that influenced PC1 were Ba, As, and Ni, indicating an atmospheric influence. PC2 was primarily influenced by organic matter, THg, Se, V, Al, and pH (Figure 4.3). As previously stated, a large fraction of these elements are organic bound. In catchment soil, the first two PCs explained 73% (PC1 = 61.44% and PC2 = 11.18%; Appendix J). Like sediment, PC1 was dominated by As, Ni, Cu, and Fe, while axis 2 was dominated by organic bound elements: pH, Se, V, Mn, Al, OM, Pb, Zn, and Cd (Figure 4.3) in soil. pH had low positive loadings for Se, Al, Mn, and V, suggesting that pH plays a minor role in the availability of elements.

pH seems to be influenced by the weathering of Ca^{2+} from the bedrock, as trace elements are enriched directly from the weathering profile in soil, sediment and lake water. Moreover, high concentrations of Fe, Al, Mn and DOC indicate that bedrock weathering is the primary source of these elements into the lake catchments.

It was hypothesized that the lake catchments would be influenced by anthropogenic sources through atmospheric deposition. The PCAs showed there are anthropogenic elements in soil, sediment and water, however most trace elements were from leaching and weathering of bedrock. The PCAs for sediment and soil show similar

groupings of elements along PC1 (e.g., As, Ba, Cu, Pb, Zn, and Cd) and PC2 (e.g., Al, Fe, Mn, Co, Cr, and V; Figure 4.3). Trace elements that were clustered around Al and Fe were indicative of geogenic sources or have been bound by organic matter (or Fe-oxides) to Al and Fe. The PCA for lake water does not show a clear source of trace elements. Trace elements were evenly distributed along PC1 and PC2, suggesting vertical mixing in the water column and showing seasonal turnover in the lakes.



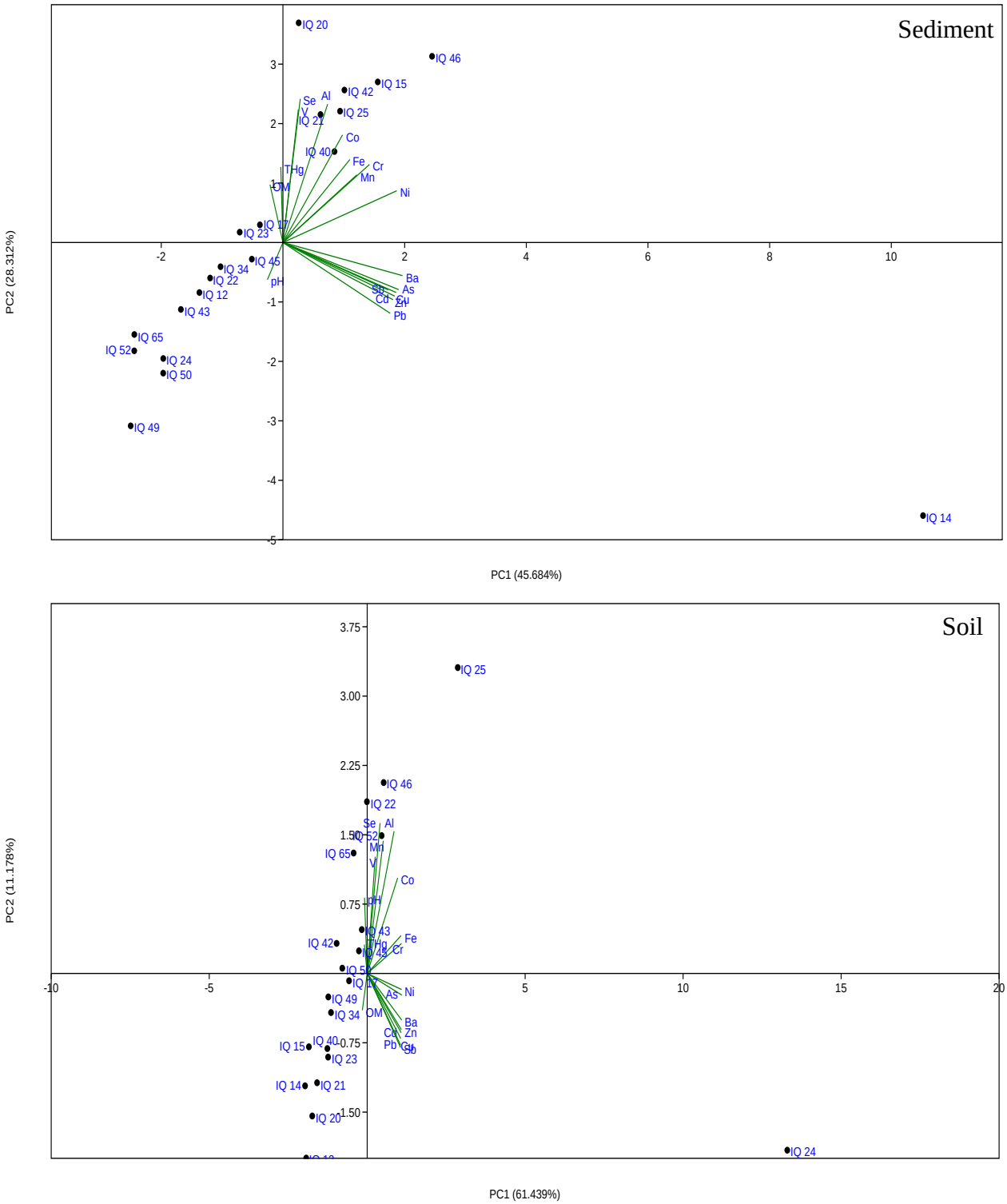


Figure 4.3: Biplots of the first two principal components of water, sediment, soil, and environmental variables (Ca^{2+}, DOC, pH, and OM). Note variables in lake water data were averaged between 2018 and 2019.

In moss, the first two PCs explained 67.65% (PC1 = 45.25%, and PC2 = 12.91%; Appendix J) of the variation. The major elements found along PC1 were geogenic and consisted of Se, Al, Fe, V, and Co, while PC2 was dominated by anthropogenic elements, Zn, Sb, Cd, Cu and THg (Figure 4.4). Similar to lake water, trace elements were not clearly defined into groupings like soil and sediment. This most likely reflects seasonal growth and an even distribution of trace elements within the moss tissue. Enrichment factor analysis also showed Cd, Sb and Cu as 'highly enriched', indicating anthropogenic activity from an atmospheric source, although it is unclear if it is local or long-range transport.

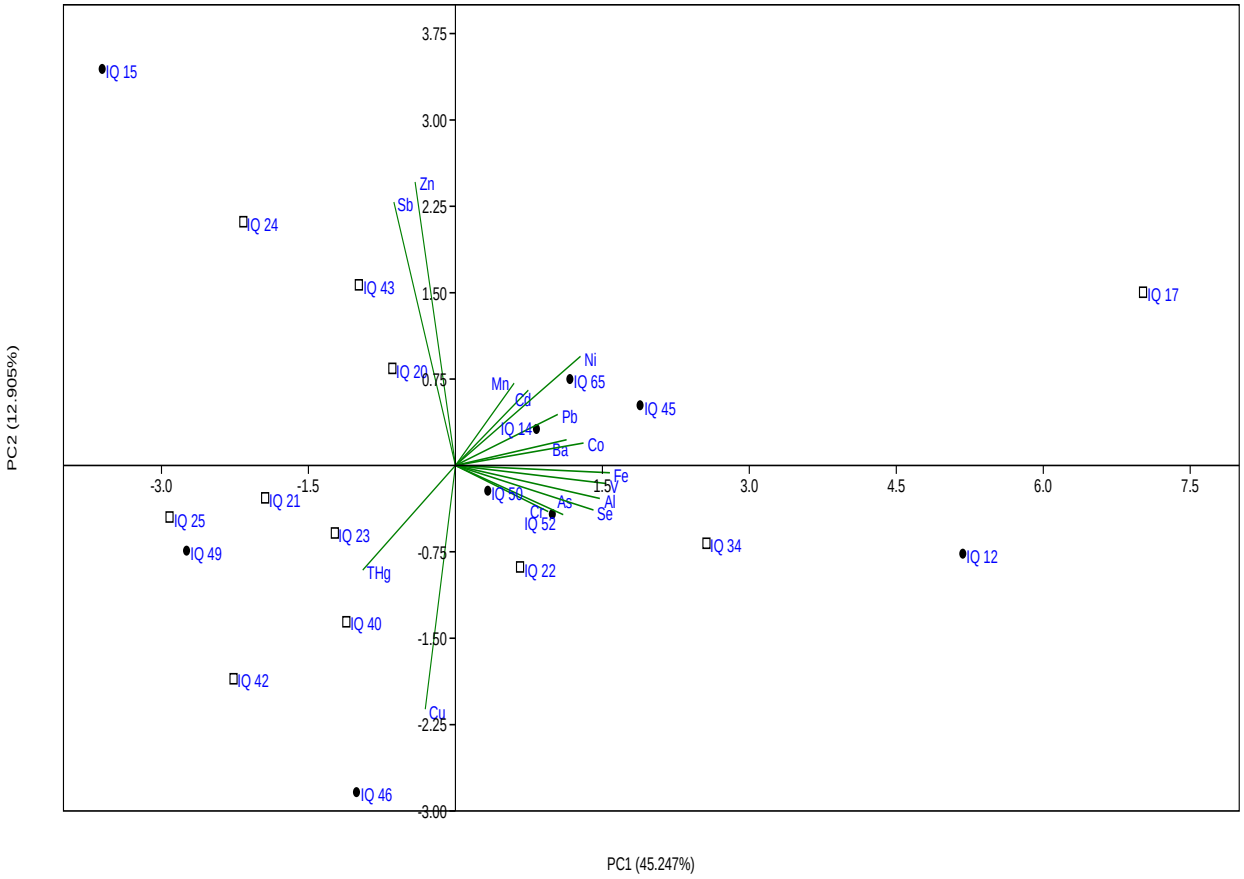


Figure 4.4: Biplots of principal components of trace elements analyzed in *Hylocomium splendens* (moss). Note variables in moss data were averaged between 2018 and 2019.

4.5 Source Apportionment using Positive Matrix Factorization (PMF) Model

The PMF model was based on 4 factors, comprising of geogenic and anthropogenic sources of trace elements. Four and 5 factor models were used to determine the best fit for the data. Using the PMF outputs from the correlation analysis and boot strapping results, the 4-factor model was selected over the 5-factor model, based off high correlation values and boot strapping of the modelled runs. In the correlation analysis, elements are compared to modelled values and concentration (actual) values of an element, the closer they fit or correlate to each other the higher the r^2 values reads. Boot strapping results >80% were achieved in the 4-factor model, while the 5-factor had lower values. Boot strapping results should read as close 100 as possible and show the same (or similar) values (numbers) for each factor. Using the correlation analysis built into the model, most elements were well predicted, with observed r^2 values >0.6 (Appendix E). Although each environmental compartment had four factors, the source contributions differentiated between individual compartments (i.e., each factor per environmental compartment had different trace elements associated with them). DOC and OM (soil and sediment) were used as a tracer for identifying factors representing organic bound elements.

The two anthropogenic sources selected for the PMF were industrial emissions and fossil fuel combustion since they represent a large portion of anthropogenic atmospheric emissions (UNEP, 2016; AMAP, 2005). These two sources were chosen based on the results of other PMF studies (Xia et al., 2020; Wu et al., 2021), along with their prevalence in the atmosphere (AMAP, 2005; Schmale et al., 2018). Emission sources in Iqaluit include transportation (vehicles, airplanes, shipping) wood stoves (for heat), and diesel generators (electric power generation). Long distance emissions include

industrial, fossil fuel combustion, agriculture, and biomass burning. Both local and long distance can be deposited onto the lake catchments surrounding Iqaluit.

Certain anthropogenic and geogenic elements were selected as indicators of these sources, e.g., Fe, Al, and Mn (geogenic), and THg, Cd, and Pb (anthropogenic) were key drivers in determining groupings of elements because of their known associations (Wu et al., 2021). Based on the EF results, Cd and Pb were highly enriched, suggesting that they likely originate from an anthropogenic source. Based on previous results in this study, Iqaluit still represents 'background' concentrations of trace elements.

The 4-factors were composed of two geogenic and two anthropogenic sources. Each element represents a percentage of the total mass within the factors (column in the figure). Elements that have a higher percentage have a larger contribution to that source. As described below, moss showed a greater weighting to atmospheric deposition from anthropogenic sources, while soil showed equal weighting to geogenic and anthropogenic sources. Finally, sediment and lakes were predominantly influenced by geogenic sources, with a small proportion of elements related to anthropogenic sources.

For lake water, factor 1 (industry emissions and fossil fuel combustion associated with organic matter) was characterized by the prominence of THg (47%), Cd (40%), Zn (34%), and Al (27%); Factor 2 (geogenic elements associated with organic matter) was characterized by Fe (78%), Mn (40%), and Al (37%); Factor 3 (geogenic) included Co (84%), V (62%), As (55%), and Ni (53%); and Factor 4 (fossil fuel combustion) was dominated by Zn (58%), Pb (56%), Cu (32%), and Sb (31%) (Figure 4.5; Table 4.6; Appendix E).

For lake sediment, factor 1 was characterized by organic bound elements associated with fossil fuel combustion; THg (75%), Cd (31%), and Cu (29%); Factor 2 and factor 3 were characterized by geogenic elements; Fe (96%), Al (80%), Mn (70%; associated with organic bound elements), and Zn (65%) and Mn (76%), Co (66%), Ba (62%), and As (58%); Factor 4 (industry) was characterized by Sb (57%), V (46%), Cr (43%) and Se (36%) (Figure 4.5; Table 4.6; Appendix E).

Similar to sediment, catchment soil in factor 1 was identified as fossil fuel combustion (organic bound elements) and was characterized by THg (86%), Cd (24%), and Se (22%); Factor 2 was characterized by industrial emission sources and consisted of Sb (80%), Cu (70%), Zn (68%), and As (60%); Factor 3 (organic bound) and Factor 4 were both geogenic in origin and dominated by, organic bound elements V (73%), Se (66%), Al (51%), and Fe (49%; Factor 3 represented organic bound elements); and Mn (72%), As (27%), Co (24%), and Al (22%) (Factor 4; Figure 4.5; Table 4.6; Appendix E).

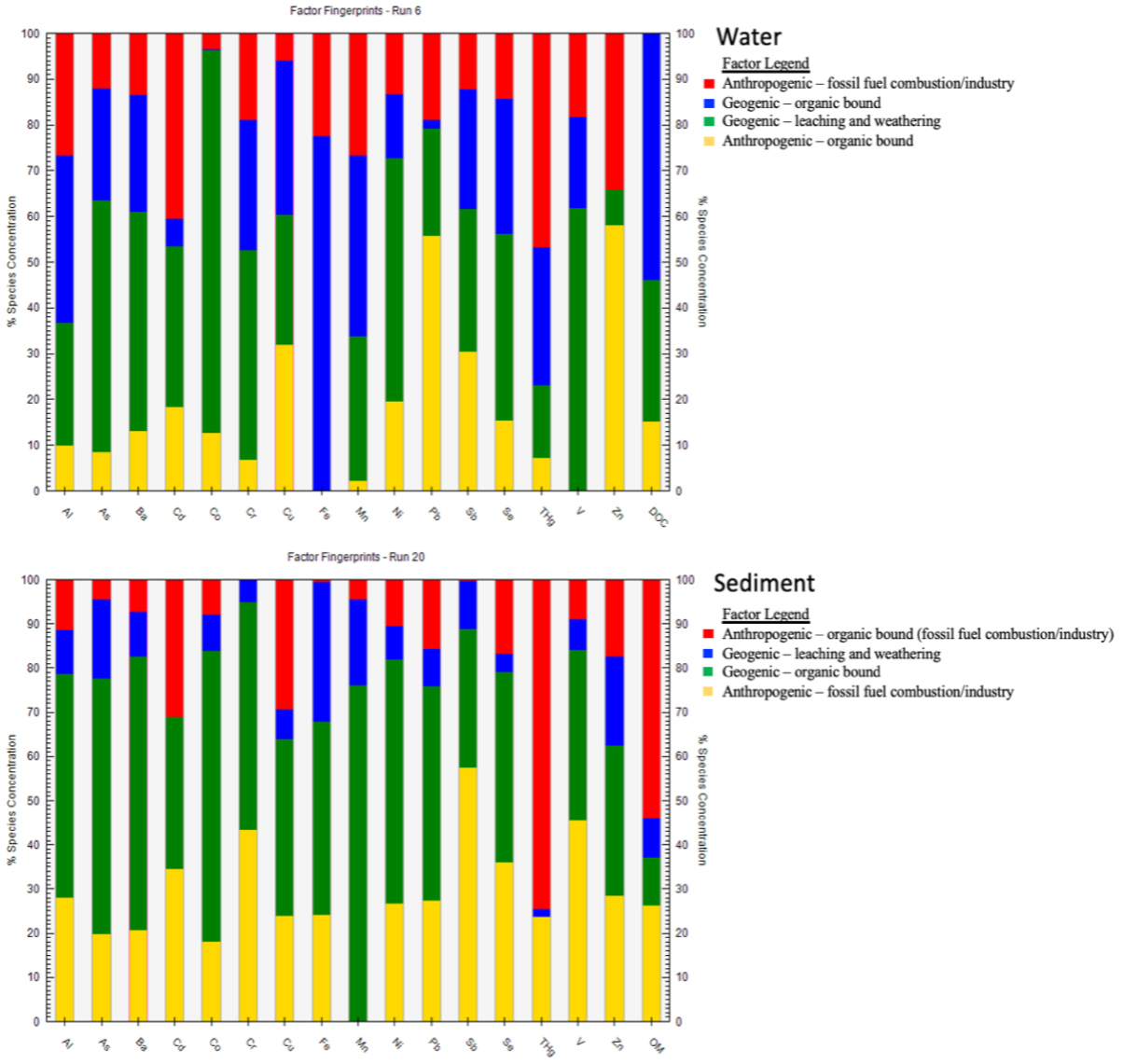
In moss, Factor 1 and 2 were identified as geogenic (wind-blown soil) and was characterized by Ba (51%), Cd (30%), Se (28%; Factor 1), and V (28%); and Fe (64%), Al (57%), V (57%), and Cr (53%; Factor 2 represented organic bound elements); Factor 3 (fossil fuel combustion) and Factor 4 (industrial emissions) and were characterized by Sb (78%), Zn (60%), Ni (25%), and THg (20%); and Co (64%), Mn (57%), Cu (55%) and THg (50%) (Figure 4.5; Table 4.6; Appendix E).

The PMF model is a useful tool in interpreting concentration data and differentiating the proportion of an element within each factor. Using tracer elements such as Fe, Al, Pb, THg, and OM and DOC helped to identify sources. Environmental compartments showed a greater weighting toward elements from geogenic sources

(leaching and weathering), compared to those from anthropogenic sources. Elements (e.g., Pb, Cd, Cu, Ba; Figure 4.3; Figure 4.5) identified as originating from an anthropogenic source were also identified by EFs and PCA receptor models. Similar results were also achieved with geogenic sources of trace elements.

Table 4.6: Summary of PMF factor fingerprint results for lake, sediment, soil, and moss data. Each factor represents either a geogenic or anthropogenic source. OM was used as a tracer for organic bound elements.

Lakes	Factors				
Anthropogenic	F1	THg	Cd	Zn	Al
Geogenic (organic bound)	F2	Fe	Mn	Al	Ba
Geogenic	F3	Co	V	As	Ni
Anthropogenic (organic bound)	F4	Zn	Pb	Cu	Sb
Sediment					
Anthropogenic (organic bound)	F1	THg	Cd	Cu	Zn
Geogenic	F2	Fe	Al	Mn	Zn
Geogenic (organic bound)	F3	Mn	Co	Ba	As
Anthropogenic	F3	Sb	V	Cr	Se
Soil					
Anthropogenic (organic bound)	F1	THg	Cd	Se	Zn
Anthropogenic	F2	Sb	Cu	Zn	As
Geogenic (organic bound)	F3	V	Se	Al	Fe
Geogenic	F4	Co	Mn	Cu	THg
Moss					
Geogenic	F1	Ba	Cd	Se	V
Geogenic (organic bound)	F2	Fe	Al	V	Cr
Anthropogenic	F3	Sb	Zn	Ni	THg
Anthropogenic (wind-blown soil)	F4	Co	Mn	Cu	THg


Factor Fingerprints - Run 6
% Species Concentration
Al
As
Ba
Cd
Co
Cr
Cu
Fe
Mn
Ni
Pb
Sb
Se
THg
V
Zn
DOC
Water
Factor Legend
Anthropogenic – fossil fuel combustion/industry
Geogenic – organic bound
Geogenic – leaching and weathering
Anthropogenic – organic bound
Factor Fingerprints - Run 20
% Species Concentration
Al
As
Ba
Cd
Co
Cr
Cu
Fe
Mn
Ni
Pb
Sb
Se
THg
V
Zn
OM
Sediment
Factor Legend
Anthropogenic – organic bound (fossil fuel combustion/industry)
Geogenic – leaching and weathering
Geogenic – organic bound
Anthropogenic – fossil fuel combustion/industry

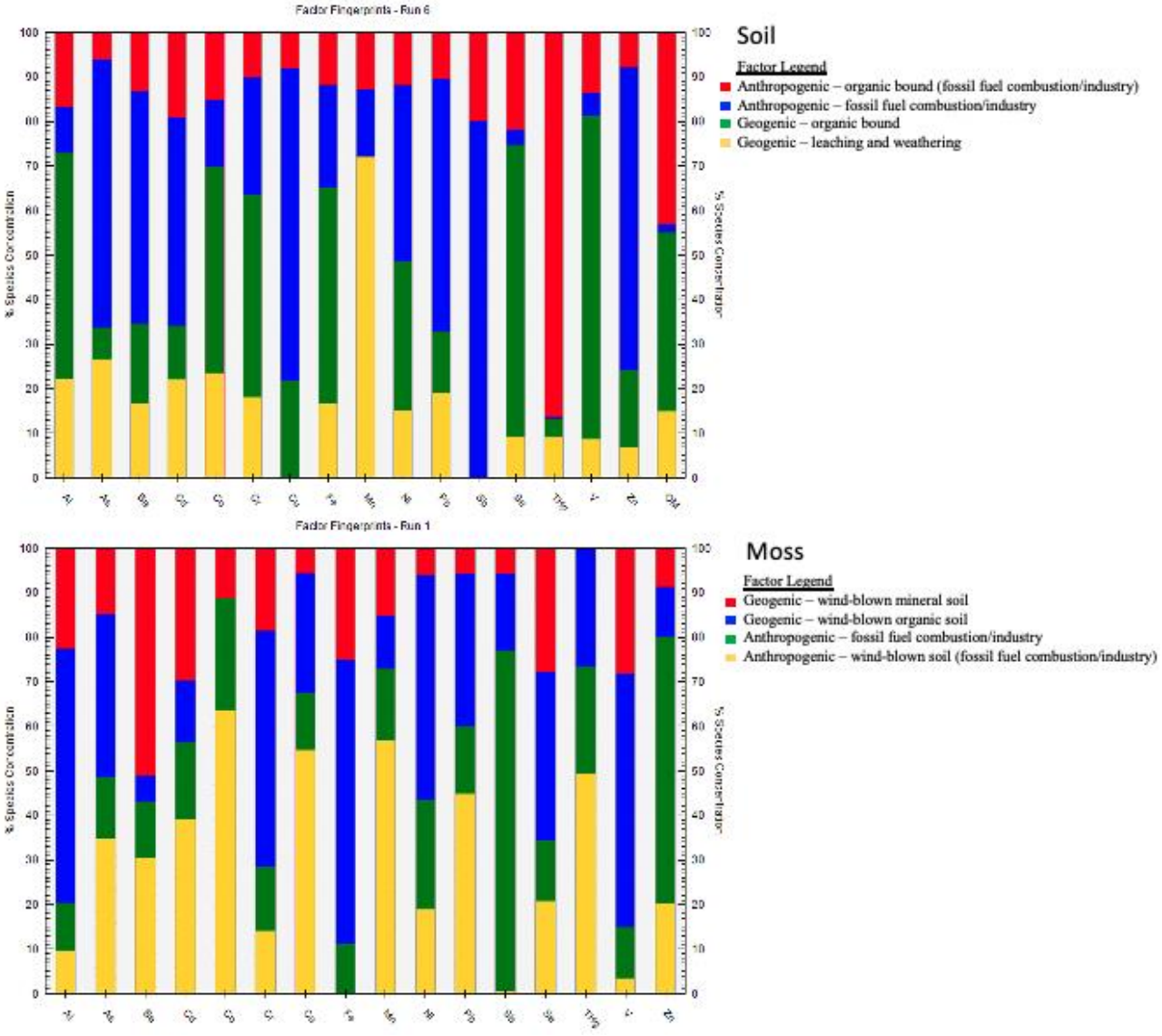


Figure 4.5: Estimated percent of measured species attributed to each PMF source factor in lake water, lake surface-sediment, catchment soil, and moss. Factors represented geogenic or anthropogenic sources. DOC and OM were used as tracers for organic bound elements. Geogenic sources were generalized to cover all mechanisms. Anthropogenic factors were simplified to fossil fuel combustion and industry sources.

5 Discussion

Lake catchments are integral to trace element biogeochemical cycling between terrestrial and aquatic phases in the environment (Adrian et al., 2016). Surface soils and sediments in the Arctic reflect atmospheric deposition and accumulation over relatively long periods of time (~10 years), compared to mosses (2-3 years) and lakes (1 year) which reflect shorter time periods (AMAP, 2005; Halbach et al., 2017). Lakes are influenced by catchment runoff from longer term soil pools, while mosses are impacted by wind-blown soil particles from legacy sources.

Previous studies have concluded that climate, geology, and vegetation are key factors affecting lake water chemistry across high-latitude regions (ACIA, 2005; Dranga et al., 2017), however, there have been few attempts to characterize lake catchment compartments (i.e., lake water, surface sediment, catchment soil, moss, and vegetation) within the Canadian Arctic, with most studies (Dushenko et al., 1996; Peramaki and Decker, 2000; Wilkie and La Farge, 2011; Halbach et al., 2017) focusing on one aspect of an environmental compartment (i.e., surface, mineral layer, sediment cores, depth profiles).

Although the Canadian Arctic is generally considered a ‘pristine’ environment, there is still a need for establishing monitoring programs (water, soil, sediment, vegetation, air) as data can be difficult to collect. Frequently studied regions of the Arctic include Devon Island, Bathurst Island, Cornwallis Island, Alert, and northern Baffin Island (Liang, 2018; Huser et al., 2020). Comparatively, the low Arctic, specifically southeastern Baffin Island has few studies or long-term monitoring programs established. This is important as climate change, shipping emissions, and other anthropogenic

activities are expected to increase in the future, potentially having negative implications on Arctic ecosystems (Adrian et al., 2016).

5.1 Water Chemistry and Trace Element Concentrations in Lakes

The Iqaluit region is mostly underlain with metamorphic (Precambrian) bedrock, with a low buffering capacity (Hamilton et al., 2001; Harrison et al., 2011), low ion, and low alkalinity levels (Huser et al., 2020). This is common in igneous geology (granite, and rhyolite >69% SiO_2, trachyte ~63% SiO_2, and gabbro and basalt 45–52% SiO_2; Hodgson, 2003; Harrison et al., 2011), which provides limited buffering capacity against inputs of acidity (Liang, 2018). Regional variation in pH, alkalinity, and ions can reflect the spatial distribution of different geology types and can represent natural gradients within a region (Huser et al., 2020). Further, the content of trace elements in lake waters is an indicator of local bedrock geology and ecological state of the catchments (Kakareka et al., 2019). In all media, geogenic trace elements were the highest in this study.

In general, cations and anions were within range of lakes sampled in the Siksik Creek Catchment, 45 km north of Inuvik, Northwest Territories. For example, mean concentrations of Ca^{2+} (7.15 mg L^{-1}) and mean SO_4^{2-} (2.35 mg L^{-1}) compared to mean Ca^{2+} (6.58 mg L^{-1}) and SO_4^{2-} (3.85 mg L^{-1}) concentrations in the Northwest Territories (Dean et al., 2016). Calcium is most likely derived from geochemical mineral weathering thawing permafrost, and glacial till deposits (Hodgson, 2005; Dean et al., 2016). Similarly, SO_4^{2-} can be attributed to iron sulphide minerals commonly found throughout the region (Hodgson, 2005).

Concentrations of major ions (e.g., Ca^{2+}, Mg^{2+}, and SO_4^{2-}) are predicted to increase as climatic changes occur. For example, permafrost degradation (e.g., deepening of the

active layer and exposing mineral soils) is expected to increase nutrients and major ions (Frey and McClelland, 2009; Prowse et al., 2006), and lowering of the water table, which can cause mobilization and release of accumulated elements in Arctic lakes (Dean et al., 2016).

In general, trace element concentrations in the study lakes were similar to other northern latitudes. For example, Ni concentrations in the study lakes (median = 0.28 µg L^{-1}) were similar to concentrations in Finland (0.37 µg L^{-1}), Norway (0.24 µg L^{-1}), Sweden (0.35 µg L^{-1}), and Russian Kola Islands (0.33 µg L^{-1}; Skjelkvale et al., 2001). Further, median concentrations of Ba (6.45 µg L^{-1}), Al (17 µg L^{-1}), and Mn (0.40 µg L^{-1}; Axel Heiberg Island; Johannesson and Lyons, 1995; Liang et al., 2018), THg (3.2 ng L^{-1}; Nunavik, QC); Chetelat et al., 2015) from other regions of the Canadian Arctic were within range of this study (median Ba = 4.02 µg L^{-1}; Al = 21.60 µg L^{-1}; Mn = 1.37 µg L^{-1}; THg = 3.85 ng L^{-1}). A mine impacted lake located in northwestern Ontario on Precambrian bedrock had Ni (29 µg L^{-1}) and Mn (20 µg L^{-1}) concentrations several orders of magnitude higher than non-impacted lakes (Martin and Pedersen, 2002).

Several environmental and chemical factors influence trace element mobilization in environmental matrices (pH, organic matter, SO_4^{2-}, Ca^{2+}, Mg^{+}). Despite the acid sensitive geology, surface waters in Iqaluit had a neutral pH. Lakes had low DOC (2.77 mg L^{-1}) and low (Ca^{2+} = 7.13 mg L^{-1}; SO_4^{2-} = 2.35 mg L^{-1}; Na^{+} = 0.74 mg L^{-1}) ion concentrations, compared to other Arctic lakes. For example, Ca^{2+} (25.39 mg L^{-1}), SO_4^{2-} (29.91 mg L^{-1}), Na^{+} (25.38 mg L^{-1}) concentrations were at least one order of magnitude higher in lakes sampled within the Arctic Archipelago (e.g., Baffin, Cornwallis, Devon, Ellesmere Islands; Hamilton et al., 2001). However, DOC (3.93 mg L^{-1}) in the Arctic

Archipelago lakes were comparable to this study. Calcium most likely comes from geochemical weathering, as there are several large deposits in the region (Hodgson, 2005). Results from the PCA analysis indicated that Ca^{2+} is associated with geochemical weathering as evidenced by the strong relationship with pH. Therefore, trace elements that are correlated with Ca^{2+} are most likely from geogenic sources. A large proportion of the trace elements were not correlated to organic matter, even though other studies have found correlations (Burton et al., 2011; Skjelkvale et al., 2001; Sedlak et al., 1997; Driscoll, 1985). This is most likely due to the low organic content and abundance of Ca^{2+} ions in the lakes. Metal concentrations can be reduced in the presence of cations (Ca^{2+} and Mg^{2+}) since metals compete with ions for binding (uptake) with organic matter. (Kozlova et al., 2009).

Trace elements can be controlled by bedrock geology and soil minerology throughout the region. In some areas, groups of lakes can show high concentrations of a specific element relative to other lakes in their vicinity. This is interpreted as a geochemical influence from the bedrock (Skjelkvale et al., 2001). Trace element concentrations in the study lakes had higher spatial variability (Co (59%), Fe (51%), and Sb (56%)) and interannual variability (Ni (70%), Sb (67%), and Al (62%)), when compared to all other compartments (e.g., spatial variability in soil, Sb (50%) and THg (47%), sediment THg (40%), and As (37%) and moss Co (54%) and Al (37%)). This is most likely due to the low concentrations found in surface waters. Low concentrations typically have higher variability as slight increases in a value can influence the variability by several orders of magnitude (Birch et al., 2001). However, elements such as Fe and Al

also had a high spatial variability, but this most likely reflect geologic variation between the study sites.

Iqaluit and other remote northern communities rely on lakes and rivers for their drinking water supply, which is often pumped directly from the system without further treatment. All study lakes were below the Health Canada's Drinking Water Quality Guideline (Appendix K).

5.2 Trace Element Concentrations in Lake Sediment

Trace element concentrations were comparable or lower to other studies for Cd, Cr, Cu, and Zn i.e., Cd concentration in the study catchments (median = 0.15 mg kg^{-1}) was similar to concentrations in Svalbard (0.19 mg kg^{-1}; Lou et al., 2013), Siberia (0.20 mg kg^{-1}; Crane et al., 2001), and Greenland (0.10 mg kg^{-1}; Camarero et al., 2009). Comparatively, this study reported low values for THg (6.90 ng g^{-1}), Co (5.21 mg kg^{-1}), Mn (202.60 mg kg^{-1}), Ni (12.19 mg kg^{-1}), and Pb (5.54 mg kg^{-1}), whereas in other northern regions they were considerably higher, i.e., in Svalbard (Hg = 22.07 ng g^{-1}; Co = 15.08 mg kg^{-1}; Mn = 440.03 mg kg^{-1}; Ni = 29.98 mg kg^{-1}; Pb = 22.59 mg kg^{-1}; Lou et al., 2013), and Bathurst Island (THg = 25 ng g^{-1}; Chetelat et al., 2015).

Generally, organic matter (median = 0.81%) was low in all catchment sediments. This is most likely due to the low soil cover and unproductive nature of the lakes, which effectively reduces scavenging of metals (Camarero et al., 2009; Davis, 1984). Similarly, low organic matter was reported in lake sediments in the Northwest Territories (mean = 1.3%; Vonk et al., 2016).

The spatial variability of trace element concentrations in sediment was low (<35%; V (18%), Zn (20%), and Cr (21%)). Sediment reflects the heterogeneity of soil and the low disturbance of the region.

5.3 Trace Element Concentrations in Catchment Soil

The determination of trace elements in surface soils is an important precondition for understanding their cycling in terrestrial ecosystems and interaction with other compartments (Halbach et al., 2017).

Iron, Al, Mn, and Zn had the highest concentrations in catchment soil, whereas the remainder of the trace element concentrations were low in comparison. Overall, soils had slightly higher trace element concentrations and spatial variability compared to sediment. This is most likely attributed to the higher organic matter in soils (soil = 2.03% compared to sediment = 0.81%) and mixing (metal transport, accumulation, retention, and loss; Buggy and Tobin, 2008) that sediment will undergo during seasonal and temperature changes within lakes resulting in reduced variability. Sources of trace element variation across the Arctic may be explained through varying soil organic matter content, i.e., the organic matter is the main factor driving the soil's ability to retain many trace elements (Halbach et al., 2017). However, this was not the main driver for this study. Typically trace elements that had low concentrations resulted in higher spatial variability. For example, THg concentrations in soil (47%) and sediment (40%) showed higher spatial variability, which is similar to other studies results (AMAP, 2005; Allen-Gil et al., 2013; Beldowski et al., 2015; Hao et al., 2013; St. Pierre et al., 2015).

Trace element concentrations are consistent to those in Svalbard and Norway. Median concentrations of As (0.78 mg kg^{-1}), THg (28.71 ng g^{-1}), Cd (0.14 mg kg^{-1}), Pb (5.47 mg kg^{-1}), and Ni (8.98 mg kg^{-1}) were lower than those reported for surface soils in mainland Norway and Svalbard (As = 2.96 mg kg^{-1}; THg = 107 ng g^{-1}; Cd = 0.38 mg kg^{-1}; Pb = 10.70 mg kg^{-1}; Ni = 11.90 mg kg^{-1}; Lag and Steinnes, 1978a; Nygard et al., 2012; Halbach et al., 2017), and coastal soils in Tuktoyaktik in the Northwest Territories,

Herschel Island, King Point, and Komakuk Beach along the Yukon coast (THg ranged from 26–303 ng g^{-1} and median values ranged from 61–114 ng g^{-1}; Leitch, 2006). High organic matter at study sites, and/or local pollution sources (i.e., smelters, mines located within 150 km) typically lead to higher element concentrations (AMAP, 2005; Loesto et al., 2014; St. Pierre et al., 2015; Halbach et al., 2017). Additionally, lithogenic elements (Al, Fe) that lack a correlation with THg, suggest that atmospheric deposition rather than mineral dust input is a source of Hg accumulation in soils (Givelet et al., 2004). Results from the PMF analysis showed Fe and Al were mostly attributed to geogenic factors, suggesting that sources of Hg are through deposition.

5.4 Trace Element Concentrations and Enrichment Factors in *Hylocomium splendens* (moss)

In this study, *Hylocomium splendens* was chosen as a biomonitor due to its abundance throughout the study catchments, and it is one of the species that is widely used for biomonitoring (ICP Vegetation, 2015). Geogenic sourced elements (Al 462.28 mg kg^{-1}; Fe 536.62 mg kg^{-1}; and Zn 115.41 mg kg^{-1}) had the highest concentrations in moss tissue. The lack of cover (shelter) from atmospheric influences, such as wind, increases the opportunity of wind-blown soil particles to be captured onto moss tissue (Steinnes, 1995; Gombert et al., 2004). Further, leaching from overhanging vascular plants (mosses can be found growing under <30 cm in height vegetation) is another potential pathway (Oakland et al., 1999; Steinnes, 1995; Gombert et al., 2004).

The trace element concentrations from this study were higher than concentrations reported from Serbia (Dragovic and Mihhailovic, 2008), Bosnia and Herzegovina, Bulgaria, and Finland (UNECE, 2003; Poikolanien et al., 2004). For example, median Ni concentrations are reported as 2.99 mg kg^{-1} (this study), 1.69 mg kg^{-1} (Bosnia and

Herzegovina), 1.49 mg kg^{-1} (Bulgaria), 1.96 mg kg^{-1} (Serbia), and 1.83 mg kg^{-1} (Finland).

Antimony, Cd, and Cu were the only elements that were highly enriched, while Pb, Ni, As, Ba, and Se were enriched, meaning they are most likely from anthropogenic deposition sources (Dragovic and Mihailovic, 2009; Macedo-Miranda, 2016), such as fossil fuels and industrial activities (mining, metal and ore processing, smelting; Wu et al., 2021). Trace elements (Ba = 27.39 mg kg^{-1}, Cu = 16.93 mg kg^{-1}, Ni = 2.99 mg kg^{-1}, and Pb = 2.37 mg kg^{-1}) were slightly elevated in concentration compared to other studied trace elements, suggesting that these elements could be of geogenic origin or long-range anthropogenic sources (Macedo-Miranda, 2016). In lower latitude regions, high concentrations of an element can often be reflected in high EF values due to their proximity to industrial activity (Wilkie and La Farge, 2018), whereas Arctic environments typically reflect enrichment, albeit at lower concentrations, for example Cd and Sb had the highest EF values (>20).

Spatial variation of trace elements in moss was low with a majority (88%) of sites < 35%. Variation between sampling years was slightly higher, with 44% of sites > 35%. Variation between study sites and years may relate to differences in growing conditions, microclimates, and varying contribution of wind-blow soil particles (Halleraker et al., 1998; Reimann et al., 2001; Poikolainen et al., 2004). These include conditions that affect growth rates, productivity, and adsorption. Variation between years may also indicate shorter time periods of element accumulation in moss tissue in which they typically reflect (Halbach et al., 2017).

The use of bryophytes for large scale, long-term biomonitoring of atmospheric elements in the Arctic provides an efficient, inexpensive, less labor-intensive compared to establishing air (precipitation and aerosol) monitoring programs. However, further monitoring is required to establish long-term trends within this study and the Canadian Arctic (Gamberg et al., 2005).

5.5 Source Apportionment

Iqaluit receives trace elements from geogenic and anthropogenic (long-range and local) sources. A PMF model was used to identify trace elements that share a common source. Based on the model, groupings (factors) of natural (mineral soil, parent material, geology), organic bound (natural and anthropogenic) and anthropogenic elements were identified as potential sources. All environmental compartments showed geogenic and anthropogenic sources of elements.

DOC and OM shared similar proportions of geogenic (Al, V, Fe, Mn) and anthropogenic (THg, Cd, Pb, Sb) trace elements in lakes, sediment, and soil. In particular, lakes had the highest percentage and proportion of elements that were geogenic and associated with DOC, which is consistent with previous studies noting significant correlations between Fe, Al, and DOC (Gjessing, 1964; Tipping, 1981; Skjelkvåle et al., 2001; Burton et al., 2013).

Anthropogenic sources of trace elements were determined based on the assumption that fossil fuels and mining (industry) comprise a large portion of pollutants to northern regions. Based on reported results from other studies, Pb, THg, Cd, Cr, Cu, Sb, Co, Ni, V, and As were most often associated with anthropogenic sources (Wu et al., 2021). For example, Hg, As, Cd, Pb, Cu, Cr, and V from other studies were associated with long-range atmospheric transport from fossil fuel combustion and industry, such as

ore and metal processing in the sub-arctic (Northern Finland; Kyllonen et al., 2020), and that Co, Cd, Cu, Cr, and Ni were most likely from metal and ore processing activities (Wu et al., 2021; Davis and Gulson, 2005; Khan et al., 2010; Cloquet et al., 2015; Sha et al., 2019).

In moss, all factors represent atmospheric deposition (e.g., wind-blown surface and mineral soil; Steinnes, 1995). Other sources of trace elements to moss include, leaching by vascular plants (Steinnes, 1995; Oakland et al., 1999; Gombert et al., 2004). Combining EFs and multivariate statistics helps to further apportion sources of trace elements; providing more information of potential sources. Results from the PMF model and EFs, identified Al (57%), Fe (64%), and V (28%) as originating from wind-blown soil particles. Similarly, EFs identified Cr, Fe, Mn, and Zn as representative of soil contributions (less enriched), while Pb, Sb and Cu most likely originate from anthropogenic sources. EFs reported other studies identified Fe, Al and Mn as originating from geogenic sources (Wu et al., 2021; Cohen et al., 2011; Wu et al., 2021; Zhuang et al., 2019). High EFs for Cd (EF = 22.5) and Pb (EF = 25.2; Chiarenzelli et al., 2001; Dragovic and Mihailovic, 2009) were also reported in mosses and lichens, compared to this study where Pb (EF = 6) was enriched, and Cd (EF = 20) was highly enriched. Pb deposition is most likely attributed to leaded gasoline (legacy sources), while phosphate fertilizers, waste incineration and fossil fuel combustion were identified as main sources of Cd (Dragovic and Mihailovic, 2009). Both elements are highly volatile, thus undergoing long-range transport (Puxbaum, 1991).

Although the study sites are not considered coastal, they are located within 8 km of the coast near major international shipping routes (Northwest Passage, Arctic Bridge

Gateway, and Fury and Hecla Strait) that are expected to see an increase in vessel traffic within the next several years (Loeng et al., 2005; Gavrilchuk and Lessage, 2014). This will result in increased shipping emissions throughout the region. Elements that are associated with shipping emissions include Zn, Sb, Cu, Cd, and Pb (Wu et al., 2021). The prevailing wind direction for the region originates from the northwest, where mainland Nunavut and the Northwest Passage are located. Prevailing winds can carry particulate matter, along with elements to other regions in the Arctic (Aulinger et al., 2016; Gong et al., 2018). During the sampling periods (August and September, the wind direction predominantly originated from the northwest (Appendix H), which is in relative proximity to Frobisher Bay and additional shipping routes, albeit less frequented. This suggests that marine influences (i.e., shipping routes, shipping port) could impact trace element concentrations. Local factors influencing trace element concentrations are emissions from diesel generators and a domestic airport. Regions that are in relative proximity (~400 km) of a pollution source, can reflect the pollution onto the landscape (Chen et al., 2008; Zhou et al., 2018; Wu et al., 2021).

6 Conclusions

The objectives of this study were to assess the variation and concentrations of trace elements in Iqaluit, investigate sources (geogenic or anthropogenic) of trace elements and characterize physiochemical characteristics of 20 lake catchments surrounding Iqaluit. It was hypothesized that trace elements emitted from local and long-range anthropogenic sources are present in Arctic lake catchments.

These objectives were addressed by asking the following research questions:

Research Question 1: Does soil and sediment composition (organic content, pH, and particle size) influence trace element concentrations in soil and sediment?

The studied lake catchments were underlain with Precambrian bedrock geology, which dictated trace element concentrations through processes of geochemical weathering. The thin layer of topsoil (<5 cm), low organic matter and large composition of sand particles likely inhibits trace element mobilization in the lake catchments. Understanding the degree that physiochemical characteristics have on trace element mobilization and transport requires a detailed survey of the catchments.

Research Question 2: Are pH and (DOC) associated with element concentrations in lake water?

Studying different environmental compartments can help to determine how geogenic sources influence environmental variables in lake water. The high Ca^{2+} content in the bedrock impacts lake water pH; most likely raising it from from a typical acid-sensitive lake (6.2–6.9) to a neutral pH, which reduces the impact of pH on trace element speciation and mobilization. DOC had strong associations with trace elements, and most likely influences trace element concentrations in the study lakes through adsorption and distribution within the water column.

Research Question 3: What elements are dominant in lake water, lake sediment, catchment soil and moss?

Iron and Al had the highest concentrations in all environmental compartments (and were not considered trace). High concentrations of these elements were from local bedrock weathering in the region. Trace elements from anthropogenic sources (and atmospheric deposition) were several orders of magnitude lower than geogenic sourced elements. Catchment soil and lake sediment are the by-product of geologic weathering and breakdown; therefore, resulting in both compartments having similar trace element concentration order, with Fe and Al having the highest concentration. Lakes and moss were less influenced by geogenic elements as they do not receive direct inputs.

Research Question 4: What trace elements show a high variability (spatial and interannual) across the catchments?

Overall, trace elements had a low spatial and interannual variability in all environmental compartments. Lake catchments were generally influenced by the same sources (underlying geology and regional deposition), owing to the lack of local point sources and the homogeneity of the region. Trace elements that had low concentrations at or below the detection limit often had high variability. Variability between years and study sites is indicative of gradual geology changes across the landscape.

Research Question 5: Using the PMF receptor model and enrichment factors (in moss), can anthropogenic and geogenic sources be distinguished in lake water, lake sediment, moss, and catchment soil?

Source receptor models used in this study were useful in separating and differentiating anthropogenic and geogenic sources. However, it can be difficult to interpret the results because sources are not always clearly defined. Additionally, the models cannot differentiate between local or long-range anthropogenic sources, as prevailing winds and elevation influence deposition onto the study sites. Future air

monitoring studies in and surrounding Iqaluit would be beneficial for the interpretation of source apportionment.

By combining results obtained through EF analysis and PMF model, this study showed that different sources of trace elements exist. Both receptor models showed similar results of trace element sources, e.g., EFs and PMF identified Fe as geogenic and Pb, Cd, As, Se, Ba, Cd, and Cu as originating from anthropogenic sources. The trace element concentrations in the Iqaluit area are most likely from long-range atmospheric transport of industrial emissions (and fossil fuel) and local sources of fossil fuel combustion.

6.1 Recommendations

Current Arctic lake catchment studies are limited in sample collection due to logistics, resulting in spatial and temporal knowledge gaps. More studies are needed on regional surveys of climatic data, lake water chemistry, lake sediment and catchment soil chemistry including lithologic and pedologic factors, trace element composition, and bathymetric surveys. Further, detailed surveys of lake water, lake sediment, catchment soil should be included, along with an assessment of vegetation community composition, to monitor vegetation community shifts. Detailed environmental surveys would provide more information on trace element cycling within the catchment, and potential physiochemical characteristics that can aid or inhibit mobilization, transport and leaching.

An additional moss survey (every 5 years) of the region would allow for insight into deposition trends following decreased global trace element emissions but increased anthropogenic influences (in the Arctic) and monitor potential element uptake of wildlife and the community. Further, analyzing vegetation tissue and conducting a detailed surface soil survey for trace elements and nutrients analysis, and soil pore water will

provide insight into the uptake and subsequent leaching of elements through their root system (Berg et al., 1994). Consumption of mosses by herbivores can facilitate the transfer of atmospherically derived elements into Arctic food webs (Ihl and Barboza, 2007; Wilkie and La Farge, 2018), as climatic shifts in vegetation communities have resulted in ungulate grazing changes. More research is required to monitor trace element concentration in mosses as Arctic ecosystems accumulate and biomagnify trace elements, resulting in high levels of dietary exposure for upper trophic level wildlife and the people who rely on the wildlife for subsistence (NCP, 2012).

Finally, data from this study can be used to represent background concentrations of the Iqaluit area and provide future studies with a baseline. These results can be used to monitor the impacts of climate change within the Iqaluit area and give direction on future policy development.

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Appendices

Appendix A: *Species Diversity in Arctic Tundra Catchments*

Species Diversity in Arctic Tundra Catchments

The Canadian Arctic tundra covers a landmass of 2,167,000 km^2 and is simply defined as the land beyond the northern limit of trees. The tundra is comprised of stunted vegetation, mosses, and lichens, that have adapted to surviving in harsh conditions (Kauffman and Pyke, 2008). The tundra is relatively homogenous and is part of the periglacial environment, affected by past ice ages, permafrost, and freeze-thaw cycles (Harmsen, 2008). Arctic tundra vegetation has adapted to endure harsh environmental conditions such as, low year-round temperatures, low precipitation, limited soil nutrient availability (low nitrogen and phosphorus ions; Billings and Mooney, 1968; Iturrate-Garcia et al., 2016), and shortened growing seasons (<3 months) with snow coverage for the remainder of the year (Iturrate-Garcia et al., 2016).

Arctic species have a distinct equatorial-polar transition in reference to diversity and productivity, whereby plant species that occur in the southern Arctic will decrease in diversity and productivity the further north they are (i.e., as species move farther from the equator there is an overall decrease in plant species diversity; Walker, 1995; Young, 1971). Furthermore, the overall flora is similar to other biomes, and there are few species endemic to northern regions, i.e., northern plants are primarily subsets of their southern counterparts (Young, 1971). Most habitats are dominated by flowering plants such as grasses, sedges, dwarf shrubs, prostrate shrub, and a variety of perennial herbaceous plants (Peterson, 2014). In general, vegetation distribution is based primarily on summer warmth, with the dominant vegetation species being *Salix* (willow) and *Ericaceae* (heather), with extensive areas of grasses, sedges, lichens, and mosses occurring in low,

wet, covered areas, and on moist (till or marine deposit) slopes (Short and Jacobs, 1982; Hodgson, 2005). Tundra vegetation are low in stature with growth buds near the soil surface, where the temperature is less variable (Kauffman and Pyke, 2001).

Determining community composition, species diversity and species richness is an important component of monitoring Arctic vegetation, as knowing these factors will provide insight into soil conditions, growing conditions, nutrient availability, and micro habitats. A basic feature of plant communities is the distribution of abundance among species (Smith and Wilson, 1996). There are many aspects of plant communities that can be measured, the most common ones include: species richness, abundance, and diversity (Pielou, 1977; Smith and Wilson, 1996; Sax, 2002). The Shannon-Wiener Diversity Index and the Simpson Index are statistical representations of diversity within a given area. New indices continue to be proposed, however there is no agreed upon usage for a specific index (Smith and Wilson, 1996).

Community composition, species richness, and vegetation cover

The objective of this appendix is to provide a general characterization of Arctic tundra vegetation (species diversity and species richness). During August 2018 and September 2019 vegetation surveys were carried out (i.e., 10 catchments were sampled in 2018 and the remaining 10 sites were sampled in 2019). A 10 m transect running perpendicular to the shoreline with one quadrat (50 cm x 50 cm) placed at the start and the end of the transect on each slope (i.e., two quadrat readings per slope, with a total of 3 slopes per catchment). The transect and two quadrats were placed at a distance of 10 m from the shoreline. In total, there were 6 vegetation quadrats surveyed per catchment. Quadrat and transect locations were selected based on similarity, i.e., soil, vegetation, slope, and aspect. Vegetation within each quadrat had percent (%) groundcover visually

estimated and assigned to two plant functional types (PFT): vascular (shrubs and herbaceous) and nonvascular (bryophytes and lichens) (Walker, 2000). The diversity for each quadrat and PFT was described using species richness (number of species of each PFT present in a quadrat), and the percent cover of individual species found within the quadrat. Community composition was defined as the list of species in each quadrat including their abundance (estimated as the number of quadrats where a species was present) (Iturrate-Garcia et al., 2016). The dependence of vegetation on edaphic factors, combined with small-scale heterogeneity in soil characteristics, promotes patchy distribution of communities (Walker, 2000; Lantz et al., 2010; Mod et al., 2014). These factors made identifying individual plants difficult. Instead, species that dominated a quadrat were assumed to be one plant, as they can reproduce with rhizomes and offshoots (i.e., clonal growth).

Information is sparse on the vegetation species and composition of the Iqaluit area. Further, there has been no attempt at quantifying the microhabitats that exist at different study sites. For example, sites dominated by glacial till and boulders have mosses and lichens, while less till (smaller particle size) shows grasses and vascular plants. Understanding community composition can help with monitoring vegetation shifts in Arctic landscapes, as climate change is expected to shift communities toward trees and shrubs. Shifts will influence soil temperature, moisture, surface runoff, and deepening of the active layer (Anderson and Bliss, 1998).

Assessing vegetation diversity through surveys

All analyses were based upon visual survey data. The number of individual plant species per quadrat could not be calculated if they dominated the sample area. For the purpose of comparative analysis and to avoid loss of information, vegetation diversity

was characterized by three measures: (1) species richness; (2) Shannon-Wiener Diversity Index; and (3) Simpson Index of Diversity. The principal measure of diversity used in this study was species richness, which refers to the number of species within a given sample. Species richness (which was not impacted by clonal growth) provides a robust and accurate measure of species diversity as long as sampling methods are standardized (Sax, 2002). Species richness and species abundance are key elements in describing and predicting biodiversity. Species richness was calculated by summing the total number of species per catchment, while species abundance was calculated by adding individual plants within their species. The Shannon-Wiener Diversity Index (H'c; Eq. A.1; Shannon, 1948; Iturrate-Garcia et al., 2016) and the Simpson Index of Diversity (D'c; Eq. A.2; Simpsons, 1949; Pielou, 1977), use a scale of 0–1, with 1 having the highest diversity. The diversity indices vary slightly, with Simpson's being weighted toward dominate species, and Shannon's showing species richness and evenness.

Eq. (A.1)

$$H'c = -\sum p_i \log p_i$$

Eq. (A.2)

$$D'c = \frac{\sum c_i \, (c_i - 1)}{C \, (C - 1)}$$

For H'c (Shannon Wiener Diversity Index), pi=ci/C, where C is the total species coverage and ci is the coverage of the ith species (Loya, 1972; Dodge et al., 1982; Tomascik and Sander, 1987). For D'c (Simpson Index of Diversity), ci is the coverage of the ith species and C is the total coverage of the species within the quadrat (Tomascik and Sander, 1987).

Vegetation species, diversity, richness, and abundance

A total of twenty vegetation species were identified across all lake catchment quadrats. Comparisons of D'c and H'c diversity indices showed similarities, both using a

0–1 scale, with 1 having a high diversity. Ruszkowsli (2020) reported values for both indices within the same range, however with this study the Simpson index resulted in higher values. Simpsons index is often impacted by dominant species (i.e., species that have a high coverage), which explains the consistently higher diversity index value, when compared to the Shannon–Wiener index (DeJong, 1975; Ruszkowski, 2020). Sites that had the highest diversity value for both indices were as follows: IQ 46>IQ 34>IQ 20>IQ 42. Sample sites IQ 34, IQ 20 and IQ 42 are the northern most, and are located <3 km to one another. Site IQ 17 had the lowest diversity and species richness, because it was mostly comprised of gravel patches and boulders, which potentially impacts the chemistry of soils and lakes. Species richness at the study sites ranged from 4–12, with 80% of the values above 7. Species abundance values were slightly higher (range 7–21), with over 95% of the sample sites registering a value above 7 (Table A1). Confidence intervals were used to assess the significance of vegetation diversity indices, which were defined by the upper and lower bounds that were calculated for vegetation survey data to determine their statistical significance. Upper bounds were calculated by adding the sample mean to the confidence interval value, while lower bounds were calculated by subtracting the sample mean from the confidence interval. Vegetation survey values found outside of the upper and lower range for each vegetation indices (i.e., Simpson and Shannon Index, and species richness and abundance) were considered significant. Sites IQ 17, 20, 24, 25, 34, 42, 43, 45, 46, and 49 had values outside of the upper or lower bound and were therefore considered to be statistically significant. Out of the twenty species, lichen, grass and moss were shown to be the dominant species, with at least one out of the three being present at each sample site.

Table A1: Species richness and abundance, and the median Shannon-Wiener Diversity Index (H'c; Eq.2) and the Simpson Index of Diversity (D'c; Eq.3) found within catchment sites (n=20). * indicates level of significance ($\propto = 0.05$).

Site ID	Richness	Abundance	D'c	H'c
IQ 12[a]	**9***	15	0.62	0.50
IQ 14[a]	8	13	0.59	0.41
IQ 15[a]	8	12	0.51	**0.34***
IQ 17[b]	**4***	**7***	**0.41***	**0.32***
IQ 20[a]	7	**17***	**0.69***	**0.62***
IQ 21[b]	**5***	12	0.63	0.45
IQ 22[b]	7	14	0.68	0.56
IQ 23[a]	8	15	0.65	0.47
IQ 24[a]	**6***	13	**0.44***	**0.37***
IQ 25[b]	**10***	13	**0.54***	**0.38***
IQ 34[a]	**12***	**21***	**0.74***	**0.66***
IQ 40[b]	**9***	**16***	**0.63***	**0.55***
IQ 42[b]	**10***	**18***	**0.71***	**0.59***
IQ 43[b]	**7***	**11***	**0.38***	**0.30***
IQ 45[a]	**9***	**18***	0.69	0.51
IQ 46[b]	**11***	**20***	**0.76***	**0.67***
IQ 49[a]	**5***	**8***	**0.46***	**0.39***
IQ 50[b]	7	12	0.63	0.50
IQ 52[b]	8	12	**0.57***	**0.41***
IQ 65[a]	7	**11***	**0.66***	**0.59***
Median	8	13	0.63	0.49
NMAD (%)	20	15	10	20

All moss, lichen and liverwort species were grouped together, as identification can be difficult without an expert. [a]Sampled in 2019. [b]Sampled in 2018.

Relationships between vegetation diversity and catchment soil

A PCA was used to determine relationships between vegetation diversity indices, species richness, abundance, and soil physiochemical variables, and catchment area (ha). Similarly, correlations were used to show close associations with vegetation data and catchment soil variables. The first two principal components explained 61% of the variance (PC1 = 33.07%; PC2 = 27.72%) (Figure A2). Variables that influenced axis 1 were Simpsons' diversity and Shannon's diversity indices, species richness and species abundance. Catchment area was negatively associated with axis 1, suggesting that as catchment area increases, species diversity decreases. Axis 2 was dominated by clay, silt, sand, and bulk density. Both axes show that soil physiochemical variables have little influence on how diverse the vegetation is in the catchments. Despite this, other studies have reported vegetation influencing soil chemistry, pH, moisture content and organic matter (Prowse et al., 2006).

Significant positive correlations existed between the Simpson diversity index and species richness ($r_s = 0.59$), species abundance ($r_s = 0.80$), and Shannon diversity index ($r_s = 0.95$). In general, correlations between vegetation and soil variables were relatively low or non-existent. The most notable correlations between diversity and catchment soil physiochemical variables were between the Simpson diversity index and pH ($r_s = -0.25$), organic matter ($r_s = 0.24$), and catchment area ($r_s = -0.43$), and the Shannon diversity index and pH ($r_s = -0.26$) and catchment area ($r_s = -0.44$). Soil moisture content and species abundance were significantly negatively correlated ($r_s = -0.66$) and soil porosity and species richness ($r_s = -0.48$) (Figure A3). Generally, Arctic vegetation have adapted to receive minimal precipitation throughout the year. They rely on soil texture, organic

content, nutrient content, water-holding capacity, and slow decomposition rates to support their growth (Chu and Grogan, 2010).

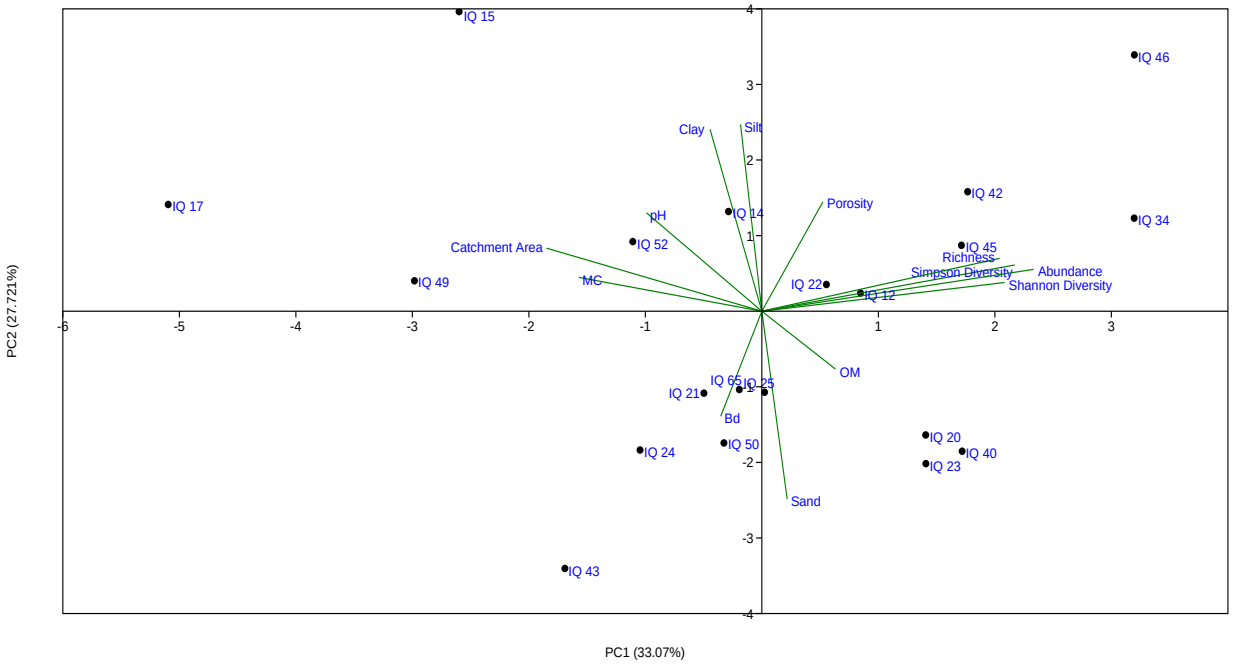


Figure A2: A biplot of the first two principal components of median vegetation and soil physiochemical data (Table 3.5 and Table 3.3). *Note* OM refers to organic matter, Bd refers bulk density, H'c refers to Shannon Diversity Index and D'c refers to the Simpson Diversity Index.

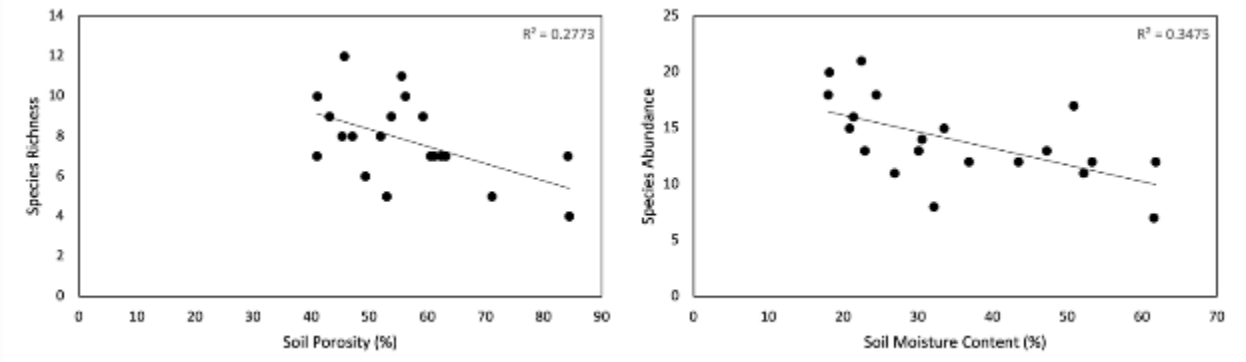


Figure A3: Scatterplot showing the relation between soil moisture content and soil porosity (%) and species abundance and richness.

Table A1: Principal components, eigenvalues and %variance of median vegetation (abundance, richness and Shannon and Simpson Diversity Indices) and soil physiochemical data.

PC	Eigenvalue	% variance
1	3.58	35.83
2	3.12	31.21
3	0.94	9.42
4	0.84	8.38
5	0.70	7.00
6	0.48	4.77
7	0.19	1.88
8	0.10	0.95
9	0.05	0.51
10	0.00	0.04

Relationships between vegetation diversity and trace elements

A Spearman's correlation was used to determine potential association between trace elements found in catchment soil and vegetation indices. Generally, most trace elements were not correlated with any vegetation index. This was to be expected as the vegetation tissue was not submitted for trace element analysis. A few elements showed weak positive and negative correlations with vegetation indices. Total mercury and species richness (0.30), Shannon (0.36) and Simpson (0.38) diversity indices were positively correlated. Higher species diversity in catchments can lead to increasing levels of mercury. As plants senesce the decomposing leaf litter can release Hg into the surrounding soil and surface waters (Obrist et al., 2012). Further, more organic matter can lead to increased plant diversity, influencing mercury levels (Orbrist et al., 2012).

Appendix B: *QA/QC trace element results for lake water, surface sediment, catchment soil, and moss.*

Table B1: QA/QC results for 2018 lake water and moss trace element samples. *Note* samples were run within the same batch.

	Lake water (ppb)	Moss (ppb)	
Element	Blank 1 (ppb)	Blank 1 (ppb)	Blank 2 (ppb)
Al	2.85	21.56	9.53
As	<0.0054	0.01	0.01
Ba	0.17	0.18	0.32
Cd	0.00	0.02	0.02
Co	<0.0220	<0.0057	<0.0057
Cr	0.19	0.04	0.09
Cu	0.66	0.53	0.50
Fe	4.75	1.91	15.66
Mn	0.07	0.04	0.04
Ni	<0.1306	0.08	0.17
Pb	0.04	0.06	0.07
Sb	<0.0040	0.00	0.01
Se	<0.0158	<0.0202	<0.0202
THg ($ng \cdot g^{-1}$ or $ng \cdot L^{-1}$)	–	–	–
V	<0.0164	0.01	0.01
Zn	6.69	3.98	18.89

Table B2: QA/QC results for 2019 lake water trace element samples. *Note* samples were run within the same batch.

Element	Blank 1 ($\mu g \cdot L^{-1}$)	Blank 2 ($\mu g \cdot L^{-1}$)	Blank 3 ($\mu g \cdot L^{-1}$)	Blank 4 ($\mu g \cdot L^{-1}$)	Blank 5 ($\mu g \cdot L^{-1}$)
Al	55.83	43.24	15.13	6.79	9.48
As	<0.0177	<0.0177	<0.0177	0.02	<0.0177
Ba	0.45	0.57	0.06	0.24	0.09
Cd	0.04	0.03	0.03	0.02	0.02
Co	0.03	0.02	0.01	0.01	0.02
Cr	0.12	<0.1184	<0.1184	<0.1184	<0.1184
Cu	2.43	0.89	0.47	0.38	1.48
Fe	11.75	6.84	28.91	5.25	9.98
Mn	0.10	0.08	0.14	<0.0364	0.06
Ni	0.05	0.06	0.05	<0.0463	0.12
Pb	0.18	0.11	0.07	0.06	0.09
Sb	0.06	0.05	0.04	0.03	0.01
Se	<0.0699	<0.0699	<0.0699	<0.0699	<0.0699
THg ($ng \cdot L^{-1}$)	–	–	–	–	–
V	0.03	0.02	<0.0177	<0.0177	<0.0177
Zn	10.75	8.05	10.83	6.17	11.12

Table B3: QA/QC results for 2019 moss trace element samples. *Note* samples were run within the same batch.

Element	Blank 1 (ppb)	Blank 2 (ppb)	Blank 3 (ppb)	Blank 4 (ppb)	Blank 5 (ppb)	Blank 6 (ppb)	Blank 7 (ppb)	Blank 8 (ppb)
Al	25.83	4.70	6.07	2.19	3.80	1.86	2.65	<0.1640
As	<0.0169	<0.0169	<0.0169	<0.0169	<0.0169	<0.0169	<0.0169	<0.0169
Ba	0.34	1.47	0.08	2.49	1.10	0.15	0.66	1.34
Cd	0.01	0.01	0.04	0.01	0.04	0.01	0.02	<0.0001
Co	<0.0089	<0.0089	<0.0089	0.01	0.06	0.04	0.05	<0.0089
Cr	–	–	–	–	–	–	–	–
Cu	0.68	0.41	1.28	0.53	3.00	0.39	5.42	1.589
Fe	7.18	6.34	10.84	2.71	3.05	3.19	13.87	8.81
Mn	0.63	1.04	<0.1386	<0.1386	<0.1386	<0.1386	<0.1386	<0.1386
Ni	0.10	0.186	<0.0216	<0.0216	0.07	0.10	0.05	<0.0216
Pb	0.41	0.41	0.41	0.41	0.41	0.41	0.41	0.41
Sb	–	–	–	–	–	–	–	–
Se	<0.0214	<0.0214	<0.0214	<0.0214	<0.0214	<0.0214	<0.0214	<0.0214
THg ($ng \cdot g^{-1}$)	–	–	–	–	–	–	–	–
V	<0.0008	<0.0008	<0.0008	<0.0008	0.04	0.02	0.03	<0.0008
Zn	25.17	14.07	3.91	1.02	8.19	4.07	6.43	1.38

Table B4: QA/QC results for 2018 surface sediment trace element samples. *Note* samples were run within the same batch.

Element	Blank 1 (ppb)	Blank 2 (ppb)	Blank 3 (ppb)	Blank 4 (ppb)
Al	2.29	5.46	4.47	11.99
As	<0.0062	<0.0062	<0.0062	0.01
Ba	1.51	0.35	0.12	1.40
Cd	0.00	0.005	0.01	0.003
Co	<0.0057	<0.0057	<0.0057	<0.0057
Cr	0.09	0.10	0.11	0.08
Cu	0.46	0.36	0.47	3.23
Fe	7.53	14.80	12.95	24.84
Mn	0.06	0.06	0.34	0.02
Ni	0.20	0.20	0.21	0.17
Pb	0.08	0.05	0.08	0.22
Sb	0.00	0.01	0.01	<0.0011
Se	<0.0202	<0.0202	<0.0202	<0.0202
THg ($ng \cdot g^{-1}$)	–	–	–	–
V	0.01	0.02	0.02	0.00
Zn	1.56	1.58	6.41	4.53

Table B5: QA/QC results for 2018 catchment soil trace element samples. *Note* samples were run within the same batch.

Element	Blank 1 (ppb)	Blank 2 (ppb)	Blank 3 (ppb)
Al	12.01	4.52	2.61
As	0.012	<0.0062	<0.0062
Ba	1.12	1.10	0.61
Cd	0.01	0.01	0.10
Co	<0.0057	<0.0057	<0.0057
Cr	0.11	0.05	0.06
Cu	1.95	0.54	1.09
Fe	33.62	4.50	5.07
Mn	0.35	<0.0056	<0.0056
Ni	0.28	0.18	0.18
Pb	0.46	0.08	0.10
Sb	0.01	0.01	0.10
Se	<0.0202	<0.0202	<0.0202
THg ($ng \cdot g^{-1}$)	–	–	–
V	0.02	0.00	0.01
Zn	16.52	3.10	2.43

Appendix C: *Lake catchment calculation.*

Using CanVec's topographic data series, layers (topography, water body and water course, and linear flow) were extracted using the Government of Canada's geospatial extraction tool and imported into Google Earth Pro (version 7.3.3; Figure C1). The water body dataset was used to collect the lake area (ha) for each sample site. The water course, and linear flow data were used to determine the flow direction (slope of the land), and the number of inflow and outflow points within each lake (catchment). The catchment boundaries were delineated by using elevation data contained within the topography layer (Figure C2). The apex of the first hill, or the highest elevation point closest to the lake was the catchment boundary. The topographic lines were used to trace the outline of each hillside to provide a more accurate catchment area (Figure C1). Once the catchment area had been established, the lake area within that catchment was subtracted to get the terrestrial catchment area (i.e., catchment area minus lake area).

Figure C1: Google Earth Pro images of the layers (waterbody, water course, linear flow and topography; left), and catchment area (right).

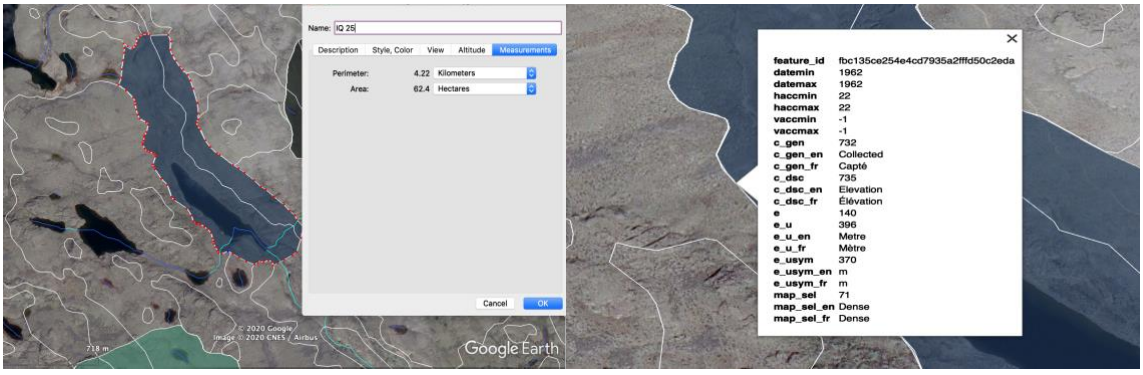


Figure C2: An example of mapping the perimeter of the catchment to determine the area (left) and obtaining elevation data from the contour lines (right).

Appendix D: *Google Maps Elevation Application Programming Interface.*

Missing values for elevation (m) were determined using Google Maps Elevation Application Programming Interface (Figure D1). Steps to retrieve unknown elevations for samples sites include:

- Inputting coordinates into the Elevation API
- The API outputs three different values
    - Location (latitude and longitude)
    - Elevation (m)
    - Resolution (indicates the maximum distance between data points)


```
{
   "results" : [
      {
         "elevation" : 128.2594451904297,
         "location" : {
            "lat" : 63.75639,
            "lng" : -68.47514
         },
         "resolution" : 610.8129272460938
      }
   ],
   "status" : "OK"
}
```


Figure D1: An example of the output received using sample site IQ 12 from Google Maps Elevation Application Programming Interface.

Appendix E: *Positive matrix factorization results*.

The concentration data and uncertainty data are required inputs for the PMF model. The measurement error (uncertainty) was calculated through the percent relative standard deviation calculation for each element replicate, and then multiplied to the element concentration. Concentration data for all trace elements were calculated for each element and study site.

For example: Al has a concentration of 756.75 mg kg^{-1} for study site IQ 12. 756.75 mg kg^{-1} is multiplied by the average triplicate (replicate; 0.41) of the samples, which is equal to the calculated uncertainty (311.43) for the model (Figure E1).

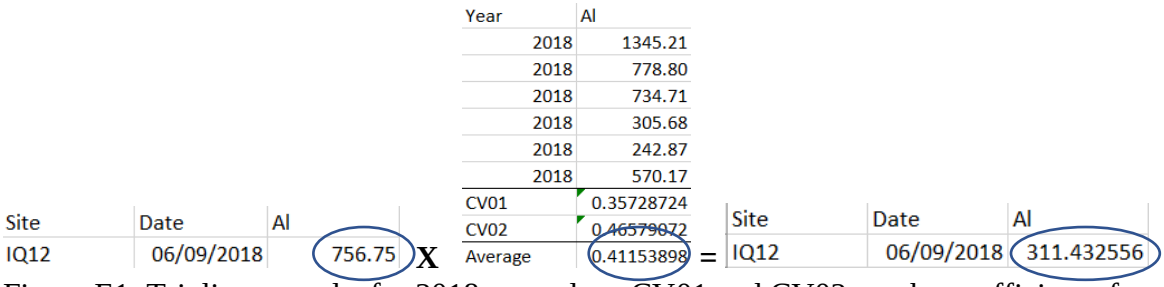


Figure E1: Triplicate results for 2018 moss data. CV01 and CV02 are the coefficient of variation of the triplicate.

Table E1: r^2 values of trace elements with their environmental compartments using PMF model. Values are results from the correlation analysis that the PMF model performs.

Element	r^2 value (water)	r^2 value (sediment)	r^2 value (moss)	r^2 value (soil)
Al	0.62	0.97	0.87	0.96
As	0.79	0.60	0.57	1.00
Ba	0.23	0.90	0.99	1.00
Cd	0.36	0.74	0.36	0.94
Co	0.01	0.94	0.26	0.93
Cr	0.33	0.83	0.46	0.97
Cu	0.25	0.86	0.37	1.00
Fe	0.19	0.96	0.96	0.93
Mn	0.15	0.95	0.06	1.00
Ni	0.82	0.96	0.51	0.95
Pb	0.68	0.47	0.72	0.956
Sb	0.17	0.27	0.97	1.00
Se	0.37	0.90	0.82	0.91
THg	0.90	1.00	0.11	1.00
V	0.21	0.91	0.92	0.34
Zn	1.00	0.80	0.63	1.00
OM or DOC	0.99	0.80	—	0.00

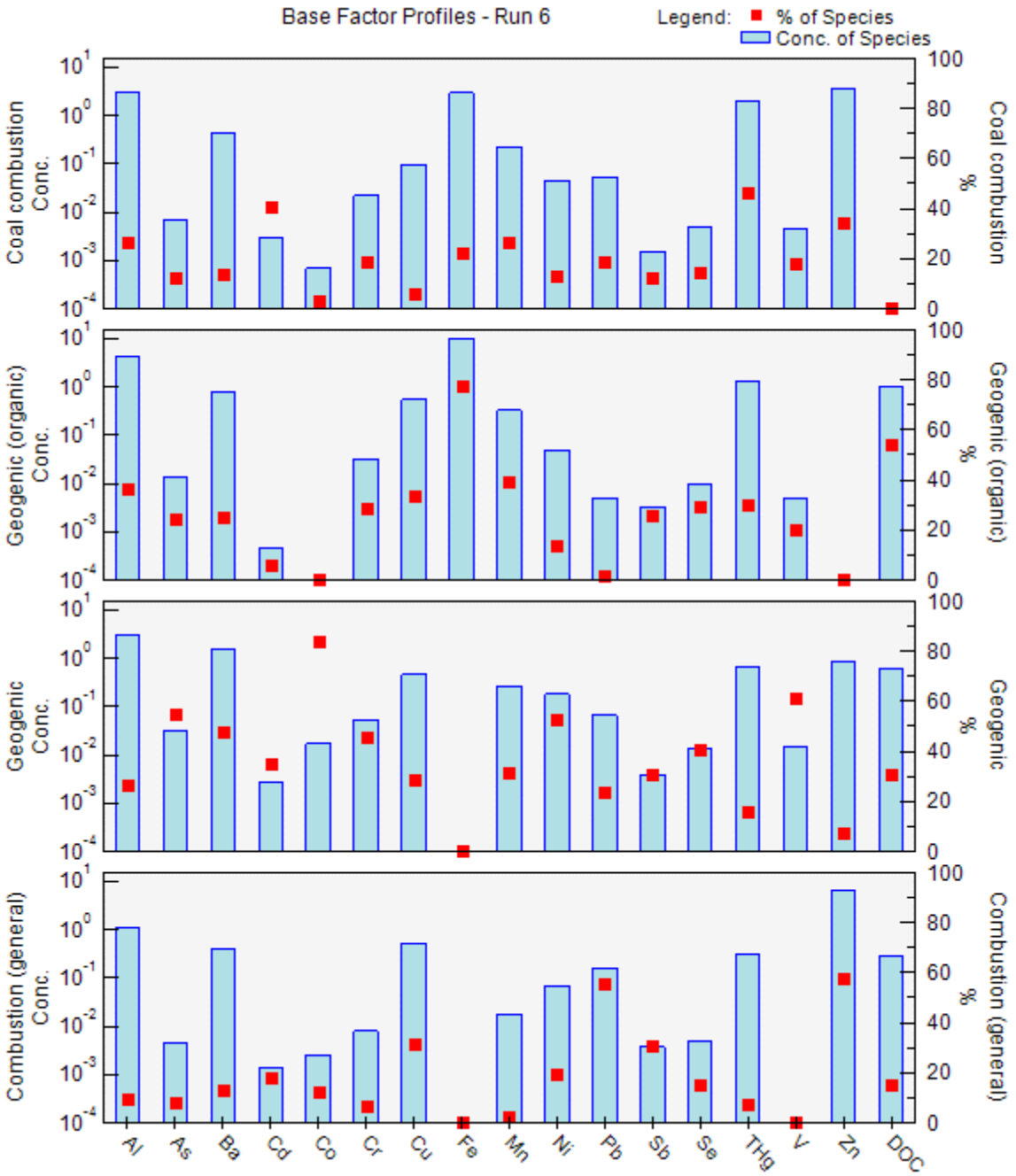


Figure E2: Profile contributions of 4-factor model using PMF for lake water (2018). *Note* the concentration of the species represents the concentration of an element within a factor (left side). The percent of a species represents the amount of a species within a factor (right side).

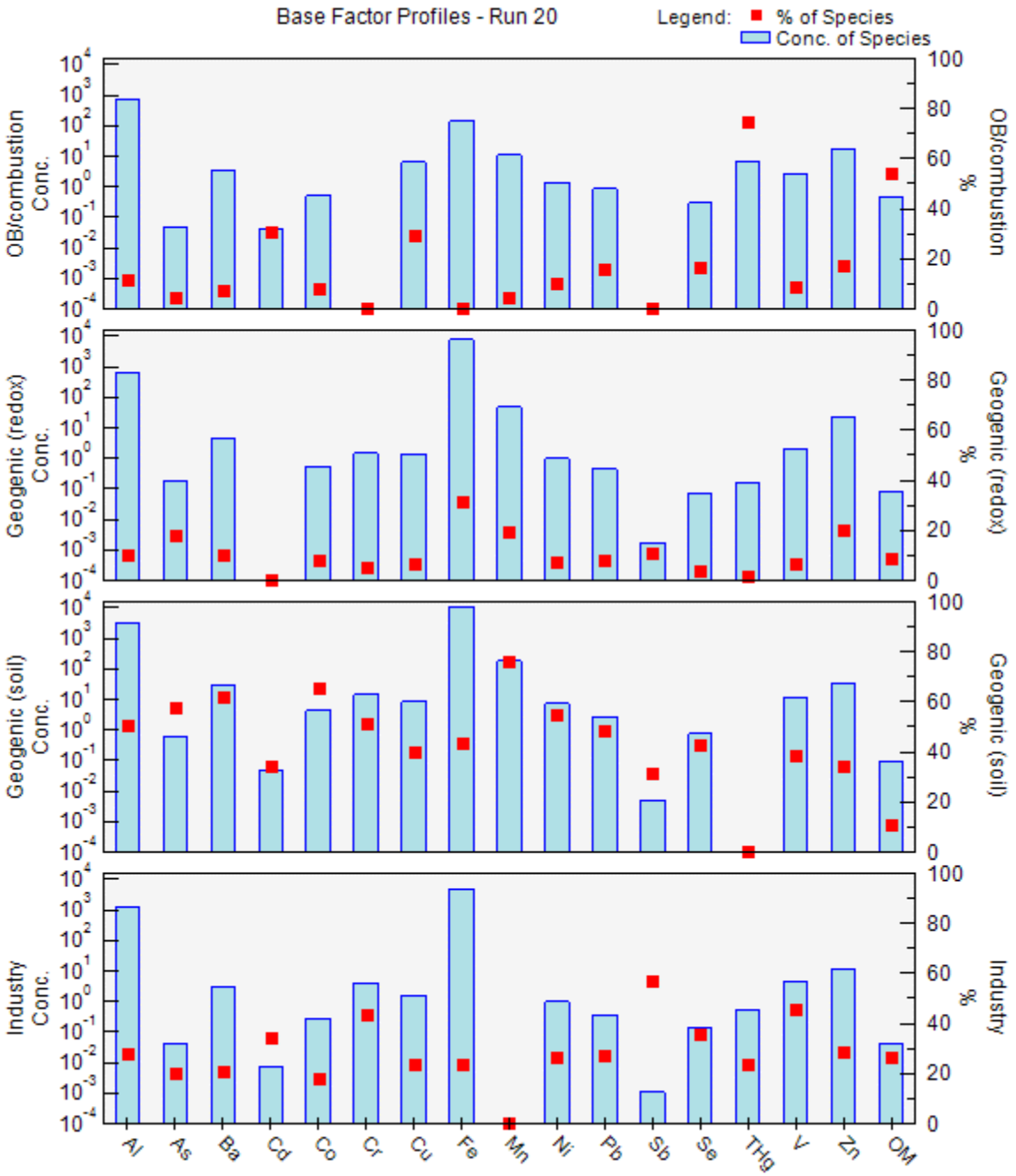


Figure E3: Profile contributions of 4-factor model using PMF for surface sediment (2018). *Note* the concentration of the species represents the concentration of an element within a factor (left side). The percent of a species represents the amount of a species within a factor (right side).

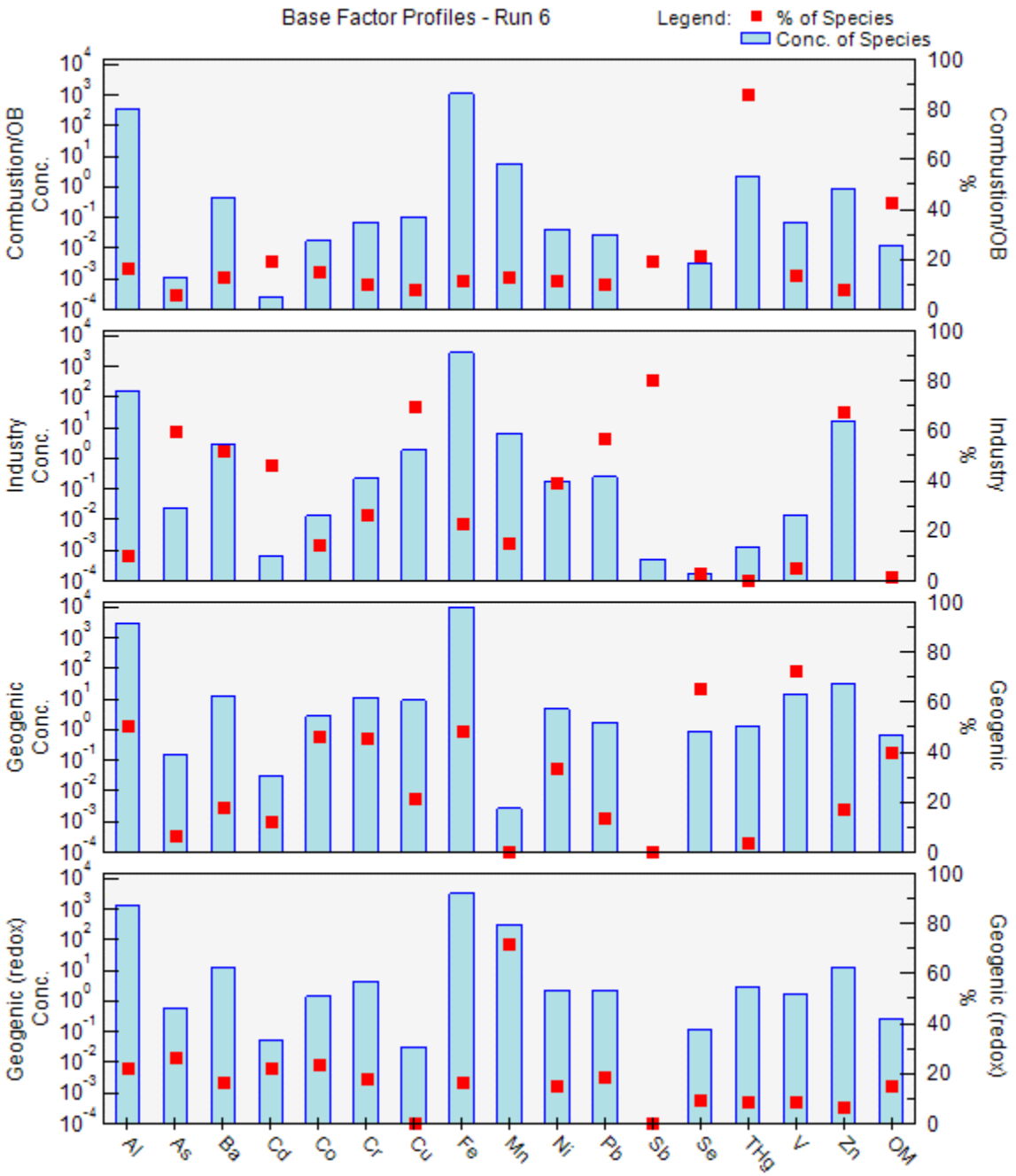


Figure E4: Profile contributions of 4-factor model using PMF for catchment soil (2018). *Note* the concentration of the species represents the concentration of an element within a factor (left side). The percent of a species represents the amount of a species within a factor (right side).

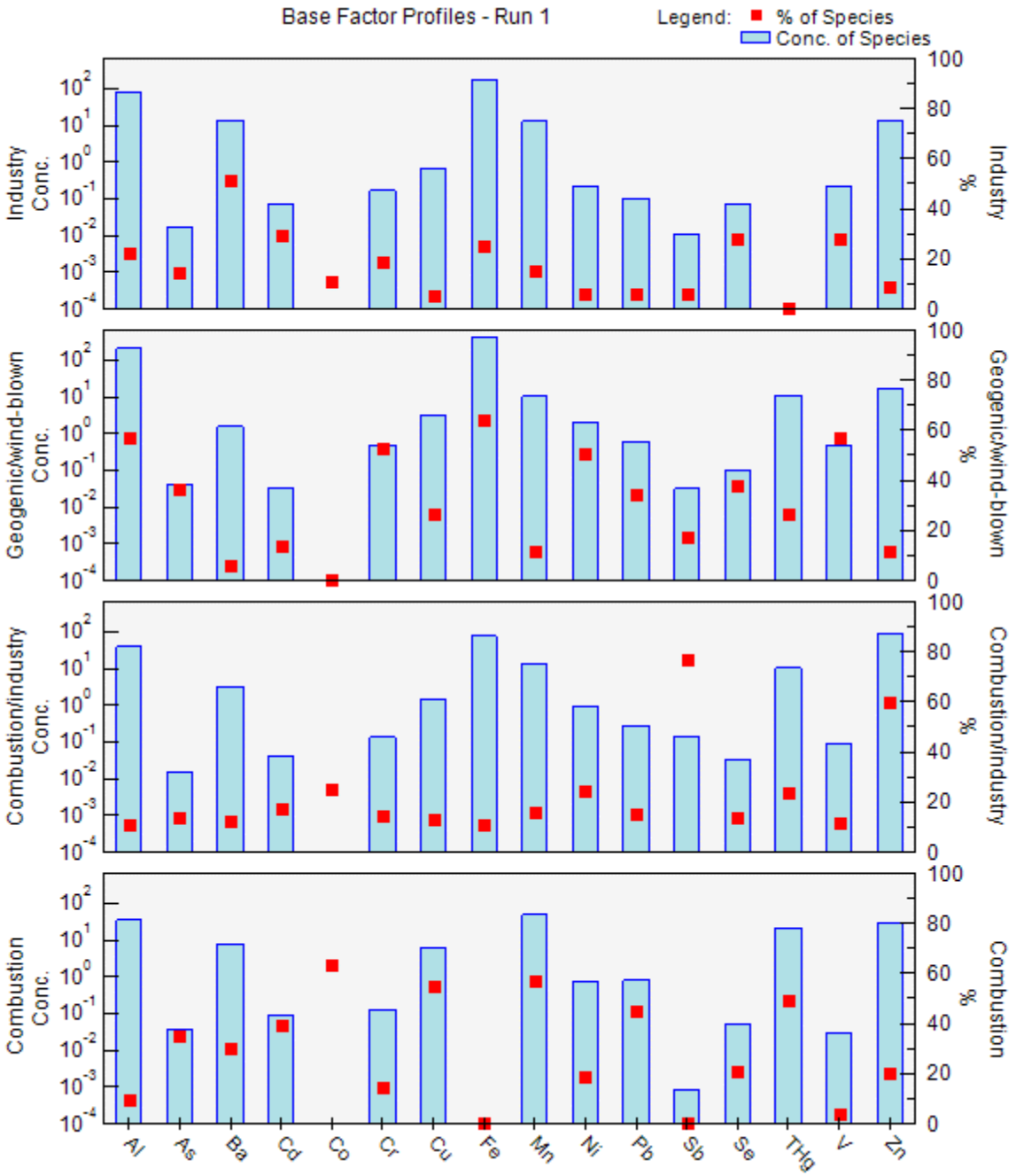


Figure E5: Profile contributions of 4-factor model using PMF for moss (2018). *Note* the concentration of the species represents the concentration of an element within a factor (left side). The percent of a species represents the amount of a species within a factor (right side).

Appendix F: *WindRose Plots of 2018 and 2019 wind speed and wind direction for the Iqaluit area.*

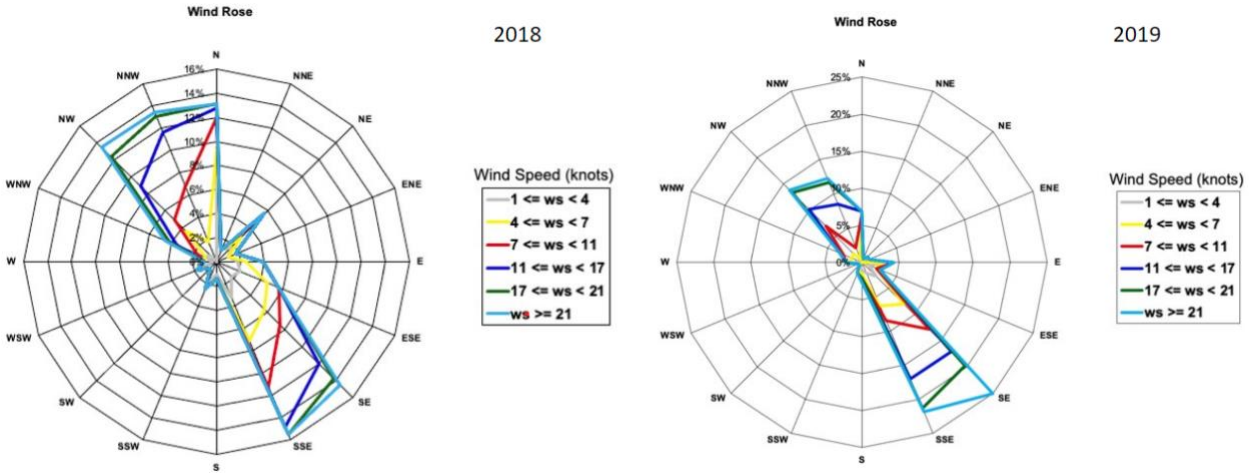


Figure F1: WindRose plots of wind speed and wind direction for the study sites.

Appendix G: *Correlation coefficients between lake chemistry and catchment attributes.*

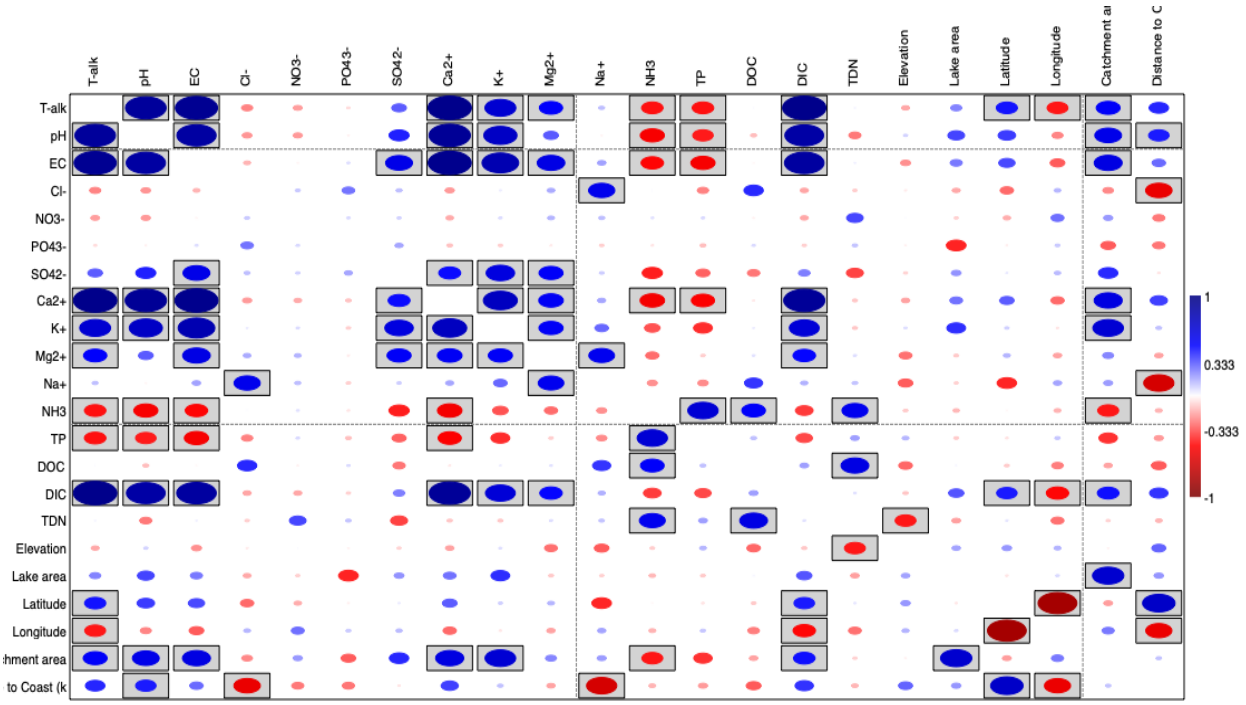


Figure G1: Correlation matrix of lake water chemistry variables (sampled 2018 and 2019) and catchment attributes. *Note* Larger circles represent stronger correlations, while smaller circles show less of a correlation. Correlations in a box represent a significant (p-value = <0.05) correlation.

Appendix H: *Principal components, eigenvalues and variance (%).*

Table H1: Principal components, eigenvalues and %variance of water chemistry and soil and sediment texture variables. *Note* water chemistry was sampled in 2018 and 2019, while soil and sediment were sampled in 2018.

PC	Eigenvalue	% variance
1	7.85	29.06
2	4.55	16.87
3	3.10	11.47
4	2.51	9.30
5	1.68	6.21
6	1.47	5.43
7	1.31	4.84
8	1.09	4.04
9	0.95	3.53
10	0.69	2.56
11	0.55	2.05
12	0.42	1.54
13	0.32	1.19
14	0.23	0.84
15	0.14	0.53
16	0.07	0.24
17	0.05	0.19
18	0.02	0.09
19	0.01	0.02

Appendix I: *Median Rank order of trace elements in lakes, sediment, soil, and moss, trace elements of concern, and study site distribution.*

Water: Fe > Al > Zn > Ba > Cu > Mn > Pb > Ni > Cr > As > Co > Se > V > Sb > Cd > THg
Sediment: Fe > Al > Mn > Zn > Ba > Cr > V > Cu > Ni > Pb > Co > Se > As > Cd > Sb > THg
Soil: Fe > Al > Mn > Zn > Ba > Cu > Cr > V > Ni > Pb > Co > Se > As > Cd > Sb > THg
Moss: Fe > Al > Zn > Ba > Cu > Ni > Pb > Cr > V > Co > Se > Cd >Mn> As > Sb > THg

Soils contain large terrestrial pools of trace elements that can potentially be taken up by terrestrial organisms, leached into groundwater, or transferred to surface waters (Richardson et al., 2015). Soil pools typically reflect the geology of the area, however soils near anthropogenic pollution sources will reflect greater quantities of elements (Pacyna et al., 2001).

Table I1: Trace element ($mg \cdot m^{-2}$) soil pool (mass per unit area) calculations.

Element	Median ($mg \cdot m^{-2}$)	Average ($mg \cdot m^{-2}$)	5^{th}–95^{th} percentile ($mg \cdot m^{-2}$)
Al	297,813	360,228	138,118–867,191
As	42.57	175.67	24.15–694.63
Ba	2175.06	5094.29	1102.54–11,503.39
Cd	8.88	30.50	3.89–54.80
Co	300.70	387.88	134.42–900.28
Cr	1206.87	1640.30	430.79–3964.91
Cu	1248.31	3383.12	364.48–6315.45
Fe	958,490	1,292,407	477,130–2,599,163
Mn	13.43	27.38	4.74–80.41
Ni	597.88	942.69	241.38–2300.21
Pb	310.74	4989.56	150.00–6547.35
Sb	2.24	7.07	0.89–11.47
Se	76.24	80.72	34.15–125.86
THg	1.74	1.74	0.38–3.03
V	1182.24	1331.19	405.52–2299.13
Zn	4.25	12.20	1.92–22.24

Trace elements of concern

Trace elements of concern were low compared to geogenic elements. Most notably, lakes, sediment and soil followed the same rank order. Pb was highest in lakes

(0.31 µg·kg^{-1}), sediment (5.54 mg·kg^{-1}) , soil (5.47 mg·kg^{-1}), and moss (2.17 mg·kg^{-1}; Figure 4.1). Arsenic was second highest in lakes (0.05 µg·kg^{-1}), sediment (1.14 mg·kg^{-1}) and soil (0.78 mg·kg^{-1}; Figure G1). Cadmium was second highest in moss tissue collected throughout the catchments (0.24 mg·kg^{-1}). This order of concentration was different compared to the other environmental compartments, whereby As was usually the second highest. Total mercury was the lowest in all environmental compartments and varied the most between sample sites.

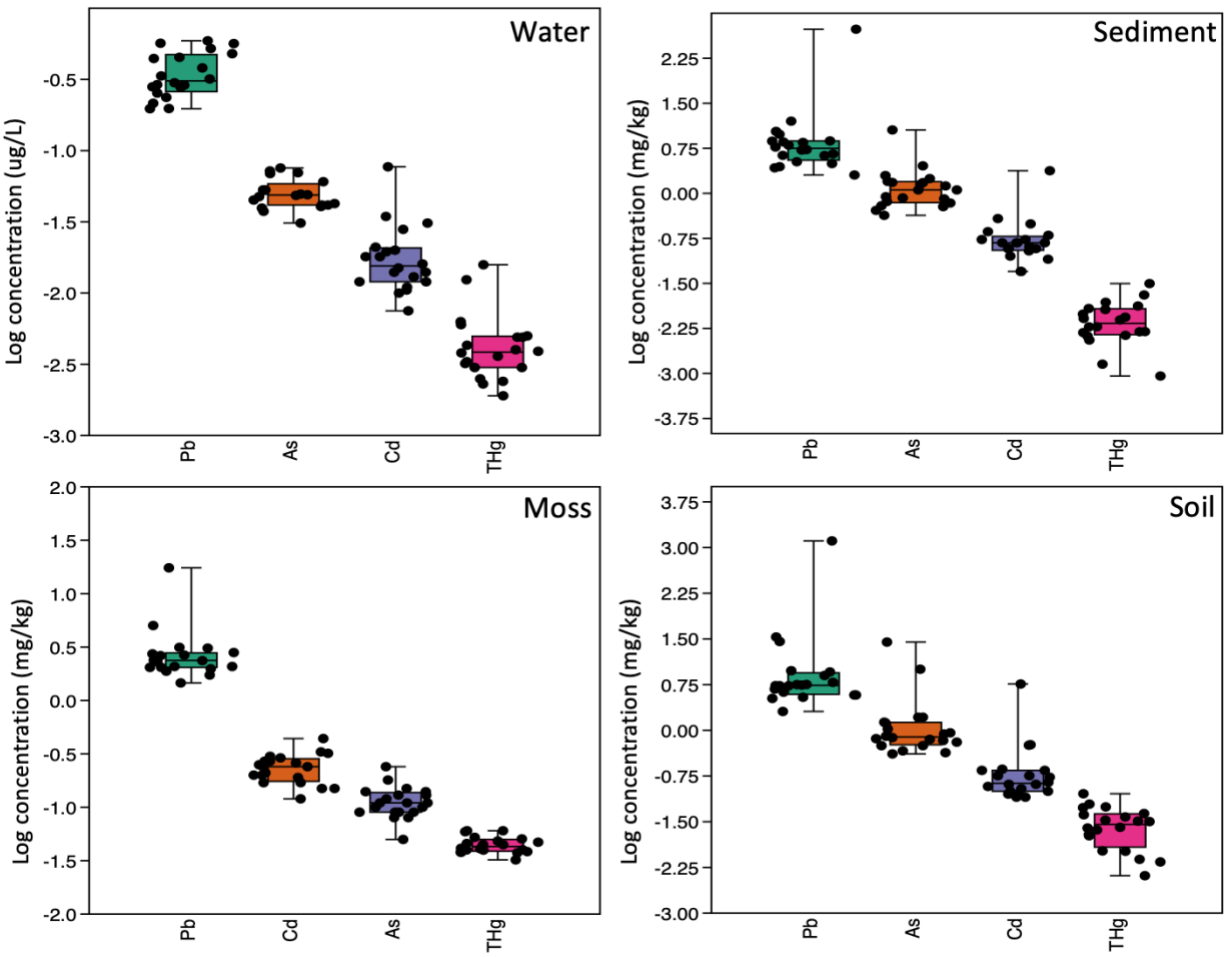
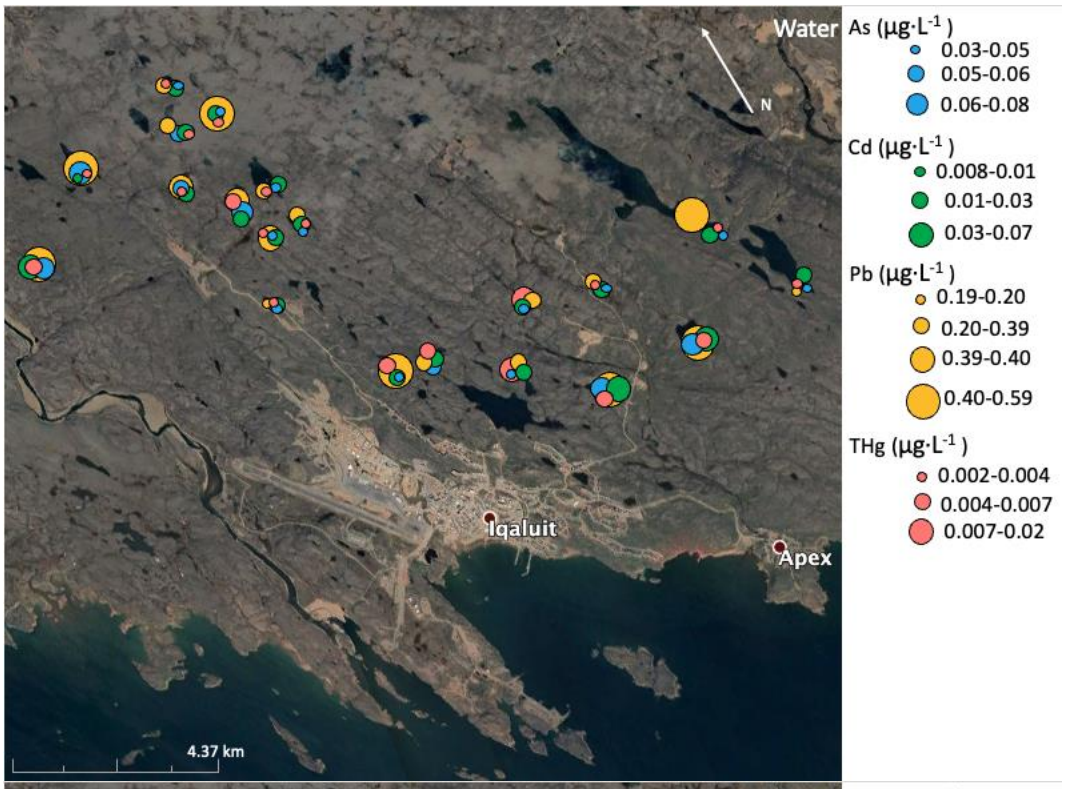
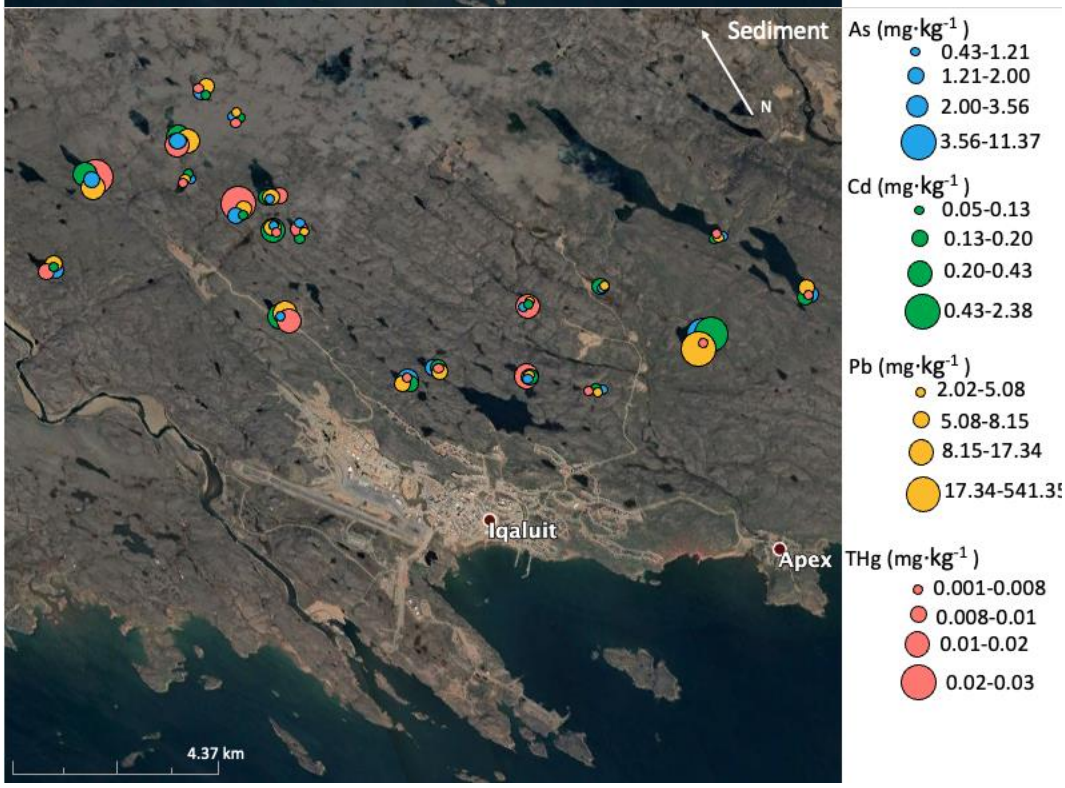


Figure I1: Box and jitter plot of log concentration of core trace elements in lake water, surface sediment, catchment soil, and moss.


Water
N
Iqaluit
Apex
4.37 km
As (µg·L-1)
0.03-0.05
0.05-0.06
0.06-0.08
Cd (µg·L-1)
0.008-0.01
0.01-0.03
0.03-0.07
Pb (µg·L-1)
0.19-0.20
0.20-0.39
0.39-0.40
0.40-0.59
THg (µg·L-1)
0.002-0.004
0.004-0.007
0.007-0.02


Sediment
N
Iqaluit
Apex
4.37 km
As (mg·kg-1)
0.43-1.21
1.21-2.00
2.00-3.56
3.56-11.37
Cd (mg·kg-1)
0.05-0.13
0.13-0.20
0.20-0.43
0.43-2.38
Pb (mg·kg-1)
2.02-5.08
5.08-8.15
8.15-17.34
17.34-541.35
THg (mg·kg-1)
0.001-0.008
0.008-0.01
0.01-0.02
0.02-0.03

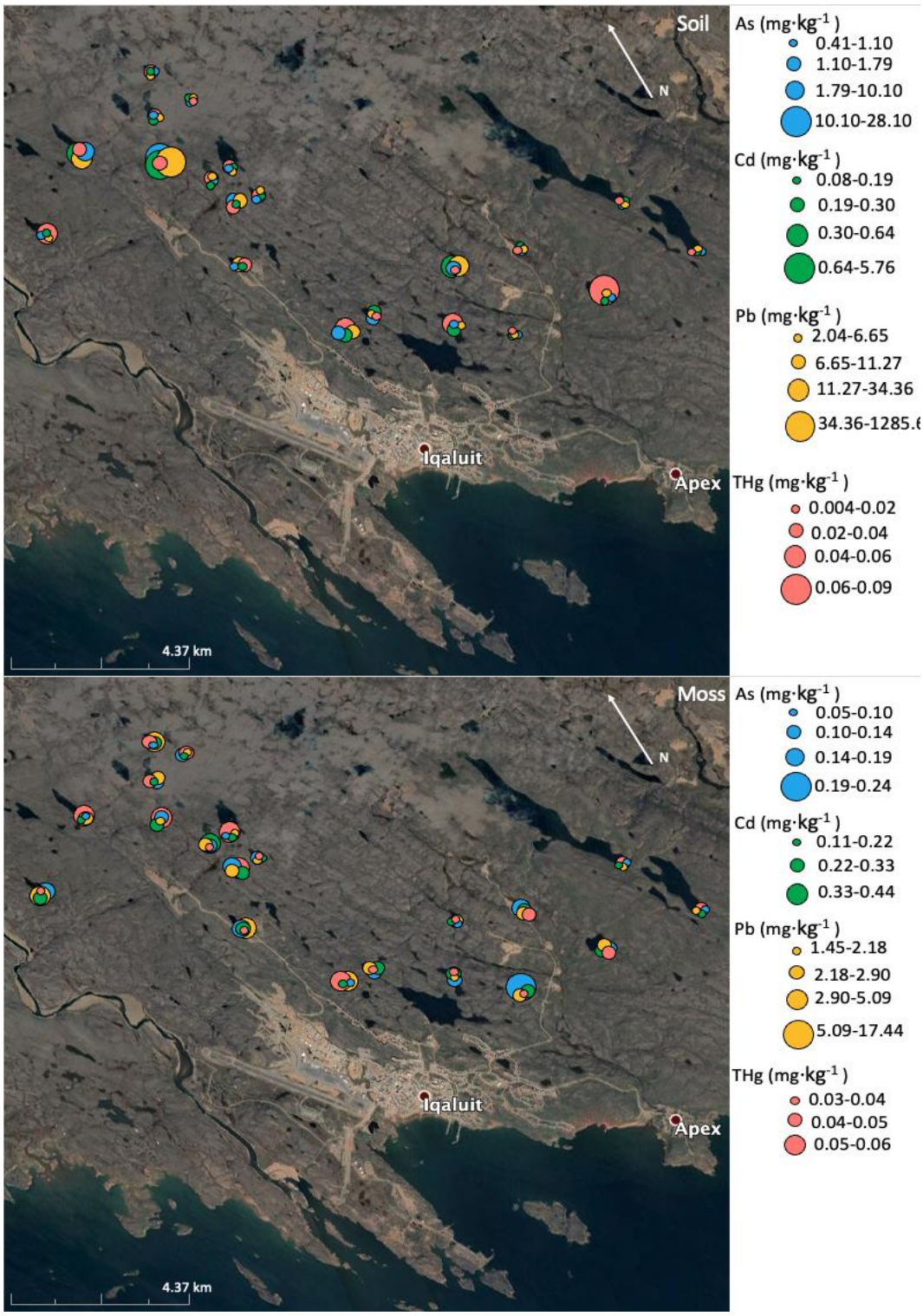


Figure I2: Spatial distribution of As, Cd, Pb and THg in lakes, sediment, soil, and moss. *Note* lake and moss were sampled during 2018 and 2019, whereas soil and sediment were only sampled in 2018.

Distribution of trace elements in the sample sites

Several trace elements in lakes (i.e., Ni in east = 0.30 $\mu g \cdot L^{-1}$ vs. Ni in north = 0.24 $\mu g \cdot L^{-1}$) and soil (i.e., Cd in east = 0.17 $mg \cdot kg^{-1}$ vs. Cd in north = 0.13 $mg \cdot kg^{-1}$) had slightly higher concentrations at sites east of Iqaluit, while moss (i.e., Cu in north = 17.45 $mg \cdot kg^{-1}$ vs. Cu in east = 16.56 $mg \cdot kg^{-1}$) and sediment (i.e., Cr in north = 30.60 $mg \cdot kg^{-1}$ vs. Cr in east = 24.60 $mg \cdot kg^{-1}$) showed higher concentrations in the north.

Lake catchment trace element sources were primarily of geogenic origin, but catchments also received inputs from anthropogenic sources. The geogenic elements, Al, Fe and Ba had the highest concentrations in lakes. Other elements with higher concentrations found at the sites were Cu and Pb, which are usually released from anthropogenic sources such as, automotive and fossil fuel (by-product) combustion (Viskup et al., 2020). In sediment and soil, Al, Ba, Fe, Mn, Cu, and Zn were present in high concentrations at all study sites, suggesting a large geogenic influence and a smaller anthropogenic influence, such as industry and mining. Similarly, moss had high concentrations of Al, Fe, Mn, Ba and Cu.

Appendix J: *Principal components, eigenvalues and variance (%).*

Table J1: Principal components, eigenvalues and %variance of surface water data sampled 2018-2019.

PC	Eigenvalue	% variance
1	6.19	30.97
2	3.32	16.58
3	2.07	10.35
4	1.79	8.97
5	1.45	7.25
6	1.37	6.86
7	0.89	4.39
8	0.71	3.53
9	0.58	2.88
10	0.44	2.21
11	0.38	1.88
12	0.23	1.15
13	0.21	1.03
14	0.15	0.75
15	0.10	0.51
16	0.07	0.35
17	0.05	0.25
18	0.016	0.08
19	0.01	0.04

Table J2: Principal components, eigenvalues and %variance of lake sediment data sampled 2018.

PC	Eigenvalue	% variance
1	9.94	55.21
2	3.20	17.79
3	1.40	7.90
4	0.94	5.22
5	0.79	4.41
6	0.48	2.65
7	0.43	2.37
8	0.23	1.27
9	0.21	1.16
10	0.13	0.74
11	0.08	0.46
12	0.07	0.38
13	0.05	0.25
14	0.03	0.15
15	0.02	0.09
16	0.01	0.06
17	0.00	0.01
18	0.002	0.00

Table J3: Principal components, eigenvalues and %variance of catchment soil data sampled 2018.

PC	Eigenvalue	% variance
1	11.06	61.44
2	2.01	11.18
3	1.36	7.57
4	1.13	6.29
5	0.99	5.47
6	0.87	4.84
7	0.41	2.30
8	0.07	0.37
9	0.05	0.26
10	0.03	0.17
11	0.02	0.09
12	0.00	0.01
13	0.00	0.01
14	0.00	0.01
15	0.00	0.00
16	0.00	0.00
17	3.31	0.00
18	1.48	8.22

Table J4: Principal components, eigenvalues and %variance of moss data sampled 2018-2019.

PC	Eigenvalue	% variance
1	7.23	45.20
2	2.09	13.06
3	1.51	9.44
4	1.46	9.09
5	1.06	6.64
6	0.712	4.49
7	0.48	3.03
8	0.39	2.41
9	0.38	2.37
10	0.21	1.33
11	0.19	1.17
12	0.12	0.76
13	0.08	0.51
14	0.06	0.37
15	0.01	0.10
16	0.00	0.02

Appendix K: *Comparison of Health Canada Water Quality Guidelines with this study.*

Table K1: Comparison of Health Canada Water Quality Guidelines with median results of this study.

Element	Median ($\mu g \cdot L^{-1}$)	Median:Health Canada Ratio (<1)	Health Canada ($\mu g \cdot L^{-1}$)
Al	21.60	0.01	2,900
As	0.05	0.01	10
Ba	4.02	0.00	2,000
Cd	0.02	0.00	7
Co	0.04	–	–
Cr	0.14	0.00	50
Cu	2.46	0.00	2,000
Fe	26.54	–	–
Mn	1.37	0.01	120
Ni	0.28	–	–
Pb	0.31	0.06	5
Sb	0.03	0.01	6
Se	0.04	0.00	50
THg ($ng \cdot L^{-1}$)	3.85	0.00	1,000
V	0.03	–	–
Zn	8.50	–	–