Geochemistry of lakes across ecozones in the Northwest Territories and implications for the distribution of arsenic in the Yellowknife region. Part 1: Sediments

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ABSTRACT

We obtained near total element geochemistry on 211 near-surface sediment samples from lakes along a transect across the Western Interior Platform to the central portion of the Slave Geological Province with a focus near the City of Yellowknife, Northwest Territories, to document regional concentrations of arsenic (As) and other elements in lake sediments. Concentrations of major and trace elements, including elements of potential human and ecological concern (As, Cd, Cr, Cu, Pb, Zn), were extracted from sediments using a modified *aqua regia* digestion. Concentrations of As exceed Canadian federal guidelines for the protection of aquatic life in most of the lakes sampled in the Slave Geological Province. Seventy one percent (n=149) of all sediment samples contain As concentrations higher than the Canadian Council of Ministers of the Environment (CCME) Interim Freshwater Sediment Quality Guideline of 5.9 mg/kg and 54% (n=114) of the samples contain As concentrations that exceed the CCME Probable Effect Level of 17 mg/kg. Sediments with the highest As concentrations are from lakes near the City of Yellowknife and likely reflect a combination of contamination associated with past industrial activity and geogenic input from mineralized bedrock and derived surficial materials (median As concentration 107.9 mg/kg, range 6.30->10,000+, n=95). Arsenic concentrations in lake sediments sampled elsewhere in the central Northwest Territories are lower (Ingraham Trail, median As concentration 10.6 mg/kg, range 1.9-101.6, n=27; Tibbitt to Contwoyto Winter Road, median As concentration 7.9 mg/kg, 0.3-101.4, n=52; Western Interior Platform, median As concentration 1.1 mg/kg, 0.1-7.1, n=37). Based on our data and a review of existing literature, background As concentration in lake sediments appear to ~25 mg/kg for the Yellowknife region, and lower for other regions in the central NWT. Other elements (Cr, Cu, Pb, Zn) are below sediment quality guidelines in the majority of lake sediments sampled.

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1. INTRODUCTION

1.1 Geochemical background and baseline

Geochemical background refers to natural variations in the concentration of elements in the environment, whereas "baseline" may include both geogenic and anthropogenic inputs (Salminen and Tarvainen, 1997; Salminen and Gregorauskiene, 2000; Reimann and Garrett, 2005). Careful determination of both geochemical background and baseline is necessary to assess chemical change in the environment and provide guidelines for regulation and policy. Results from local to continental-scale geochemical mapping projects demonstrate that variability in the spatial distribution of elements can be governed by both natural variation and anthropogenic inputs (Reimann et al., 2009). Geology provides a first-order control of geochemical concentrations at the Earth's surface, in all ecosystem components (e.g., sediment, soil, water, plants) (Yan-Chu, 1994). Variability in natural processes, such as climate and precipitation, also strongly influence chemical distribution in the environment, for example, by altering biogeochemical processes, although these factors are likely minimized at the local scale (Reimann and Garrett, 2005; Rencz et al., 2011). Concentrations of elements thus vary widely between and within regions, making country-wide average backgrounds of limited utility in addressing regional questions. To effectively identify and map geochemical background, baseline, and change in the environment, local scale (0.5 to 500 km^2) surveys are necessary and can provide information on the sources and transport mechanisms of elements of concern (Reimann and Garrett, 2005; Reimann et al., 2009).

In Archean greenstone belts hosting mesothermal gold deposits enrichment of arsenic (As) and other elements, including copper (Cu), lead (Pb), and zinc (Zn), is common. Arsenic can be released from ore minerals through oxidation and form new minerals in soils or be carried in solution depending on pH-Eh conditions. Arsenic released from bedrock can also be retained through adsorption on Fe-Mn oxyhydroxides present in altered rocks. When released to overlying soils, As concentrations generally increase by a factor of 2 to 3 due to the tendency of As to accumulate with Fe- and Mn-oxyhydroxides and organic matter in soil (Reimann et al., 2009). Thus, natural geochemical soil anomalies are common in areas of mineralization (Reimann et al., 1998; Ashley and Lottermoser, 1999). Soils are also receptors of atmospherically-derived elements (McMartin et al., 1999; Walker et al., 2005; Jamieson, 2014). Similarly, lake basins and their catchments are sinks for particles and metals. Sediments, particularly fine-grained portions, offer high adsorption capacity to elements derived from the catchment and thus act as reservoirs for pollutants that can eventually be liberated to surrounding ecosystems (Gurung et al., 2005). Substantial impacts of sediment contamination on ecosystem health have been reported in many previous studies, ranging from effects on benthic communities (e.g., Swartz, 1999) to upper trophic levels (e.g., Foley et al., 1988; Bishop et al., 1999). Geochemical characterization of lacustrine sediments is a useful tool for monitoring concentrations of elements of concern in the environment and assessing the potential for interaction with aquatic and terrestrial biota (Balachandran et al., 2006).

1.2 Arsenic

Arsenic is a naturally occurring metalloid that is relatively abundant in the Earth's crust, occurring with an average concentration of ~2.5 ppm (Wedepohl, 1995; Bowell et al., 2014). Arsenic is often enriched to 10's or 100's of ppm in hydrothermal alteration zones (frequently together with elements such as Au, Mo, and Sb) and coal deposits (Smedley and Kinniburgh, 2002). However, some of the highest reported concentrations of As in materials near the Earth's surface (1000's mg/kg) are associated with soils contaminated by mining and mineral processing

(Bowell et al., 2014, Jamieson, 2014). Of the more than 560 As minerals, the most common are As-bearing pyrite and arsenopyrite, both of which are associated with various ore deposits (Majzlan et al., 2014). Arsenic rarely substitutes in the mineral structures of silicates and carbonates, but it is found in many soils and sediments associated with Fe- and Mn-oxyhydroxides or organic material.

The mobility of As in the near-surface environment is governed by pH, redox, microbial activity, adsorption, and dissolution and precipitation reactions, with a variety of As oxidation states possible in both aqueous species and solid phases. When arsenopyrite and pyrite are exposed to aerobic conditions, they may oxidize to form Fe-oxides and other secondary minerals leading to the release of As, which may be attenuated by sorption to clay and hydrous (oxyhydr)oxides (Smedley and Kinniburgh, 2002). The adsorption of As(III) and As(V) species to Fe-oxides, and to a lesser extent Mn-oxides, is the most common means of As sequestration in soils and sediments under oxic conditions (DesChamps et al., 2002). However, if aqueous concentrations of dissolved As are high, such as in mine waste environments, secondary As minerals including scorodite and yukonite may form (Walker et al., 2009). Under reducing conditions that are common in bottom waters and sediments of lakes, As-bearing sulphide minerals will be relatively stable, but dissolution of detrital oxide minerals can cause strongly sorbed As-oxyanions to be released into pore water (Smedley and Kinninburg, 2002). Arsenic may be desorbed prior to dissolution of oxide minerals if As(V) is reduced to As(III) on the oxide surface. In some cases, As can also be leached from soils to the water table and transported large distances (e.g., Tarvainen and Paukola, 1998; Borba et al., 2000). Arsenic not retained in rocks or soils can also be carried by surficial drainage to watershed lakes, which also receive As bound to detrital sedimentary particles and from the atmosphere.

The chemical diversity and mobility of As make it one of the Earth’s most prevalent toxic species (Bowell et al., 2014). In water, inorganic As(III) and As(V) species are stable as oxyanions, which are negatively charged (arsenate) or neutral (arsenite). Thioarsenic species (negative or neutral) can be important in environments with abundant reduced sulphur. Organic As species are usually less abundant than inorganic species but may be present in wetlands and other organic-rich environments (Campbell and Nordstrom, 2014). Increasing evidence for chronic and acute toxicity of As, as well as widespread exposure of human populations to groundwater naturally enriched in As and subsequent negative health impacts, have led to a recent decrease in the World Health Organization drinking water limit from 50 to 10 ppb (Bowell et al. 2014). Inorganic aqueous species are considered more toxic than organic species, and As(III) species are more toxic than As(V) species, although as Leybourne et al. (2014) point out, dissolved As may be reduced on ingestion into the body. The health impact of ingestion or inhalation of As in solid materials such as soil, sediment, or dust depends on the bioaccessibility of the As-hosting minerals or compounds. Bioaccessibility refers the solubility of As in bodily fluids such as gastric, intestinal or pulmonary solutions, and is usually evaluated using animal experiments or simulated body-fluid experiments (Basta and Juhasz, 2014). If a mineral or solid compound with high bioaccessibility is orally ingested, a higher proportion of As is likely to be dissolved in the gastro-intestinal system and be available for uptake into other organs. Solids with low bioaccessibility tend to be excreted. Plumlee and Morman (2011) ranked As minerals on a scale of relative bioaccessibility with arsenopyrite and scorodite having the lowest bioaccessibility, and As trioxide having the highest. Relatively little information is available on the bioaccessibility of As sorbed on organic material or Fe-Mn oxides, or whether the data acquired for human systems can be extended to other organisms (Saunders et al., 2011). However, it is clear that understanding the risk associated with As-enriched soils and sediments depends on the identification of the solid species hosting As.

1.3 Arsenic in study region

Abundant gold mineralization in the Slave Geological Province of the Northwest Territories (NWT), Canada, led to the establishment of numerous mines from the late 1930s to the present day, such as (Giant Mine [1948-2002], Con Mine [1938-2003] and Tahera/Salmita [1964-1968 & 1983-1986], and the region remains highly prospective for additional gold development. Historical mineral processing and waste disposal practices at some mines (Giant Mine, Con Mine, Tundra/Salmita) that were employed prior to the development of more efficient technologies and stringent environmental regulations resulted in substantial contamination by As, some of which is in a highly toxic, soluble, and bioaccessible form (As trioxide, As_2O_3) (Jamieson, 2014; Stavinga, 2014). The predominant element of concern associated with gold projects in the region is arsenic, although the concentration of antimony, copper, and zinc are elevated in environmental media near many of these sites. Little geochemical background or baseline information was collected prior to development of these mines and local contamination now complicates the determination of background and baseline concentrations, which is necessary for development of site specific remediation objectives and identification of reference or control sites for environmental assessment (GNWT, 2003; Golder, 2013). This highlights the importance of regional geochemical surveys in understanding regional background and baseline geochemistry.

Arsenic is an element of community concern near the City of Yellowknife, NWT, because concentrations in lake sediments and soils are high relative to the rest of the country after ~75 years of gold mining (Ollson, 1999; Webster, 1999; Reimer et al., 2002). Abundant gold mineralization in the supracrustal rocks of the Yellowknife Supergroup of the Slave Geological Province led to the establishment of at least three major gold mines in the immediate vicinity of the city: Negus Mine/Con Mine, and Giant Mine, that collectively operated from 1938 until 2003. Shear zone hosted deposits at Con and Giant mines were the largest of these operations, collectively producing over 14 million ounces of gold. Mining and processing at Giant Mine in particular represents one of the longest continuous gold mining operations in Canadian history, where over 7 million ounces of gold was produced. Due to the refractory nature of ores in the Yellowknife Greenstone Belt, extraction of gold from arsenopyrite ore at Con and Giant mines involved roasting to volatilize As and Sb and transform sulphide minerals into porous Fe-oxides of maghemite and hematite amenable to cyanidation. Roasting was volumetrically a more significant portion of the milling process at Giant Mine than at Con Mine where a large percentage of gold was free milling (Moir et al., 2006). These Fe-oxides commonly incorporated As in their structure and are examples of primary anthropogenic As-bearing solids that have little similarity to naturally occurring minerals such as arsenopyrite (Walker et al., 2005). Roaster-generated Fe-oxides produced at Giant Mine contained as much as 6.85 wt.% As, including both As^{3+} and As^{5+} (Walker et al., 2005). Roasting of ore at Giant Mine released As-rich emissions, approximately 7.3 tonnes/day of arsenic (predominantly As_2O_3) and SO_x vapours directly to the atmosphere from 1949 until 1951 when a Cottrell Electrostatic Precipitator was installed and reduced aerial arsenic emissions to an estimated 3.3 tonnes/day between 1954 and 1958 (Table 1; Sandlos and Keeling, 2012). Collection rates of arsenic were further improved by the installation of a baghouse in 1958 (Table 1; MacDonald, 1997; SRK Consulting, 2002). The majority of the estimated 20,824 tonnes of As_2O_3 released from stack emissions during operation of the Giant Mine roaster from 1949 to 1999 was released during the first decade of ore processing at Giant Mine (MacDonald, 1997; SRK Consulting, 2002). The more stringent controls developed and implemented after 1958 continued to decrease emissions substantially and led to the underground storage of 237,176 tonnes of As_2O_3 until 1999 when roasting of ore ceased at Giant Mine (Table 1; Tait, 1961; CPHA, 1977; GNWT, 1993;

MacDonald, 1997; SRK Consulting, 2002; Environment Canada, 2007; INAC, 2007; Wyre, 2008; Sandlos and Keeling, 2012).

Initial ore processing at Con and Negus mines involved standard cyanidation to release gold from host rock but in 1940 the discovery of ore associated with arsenopyrite resulted in construction of a roasting facility that began operations in 1942 (Sandlos and Keeling, 2012). Con Mine roasted ore until November 1942 when wartime restrictions on gold production resulted in suspension of the facility. Ore roasting resumed at Con Mine in 1948, followed by commencement of roasting at Giant Mine in 1949. It is estimated that Con Mine released approximately 2 tonnes/day of arsenic trioxide between 1949-1953 because only about 20% of ore processed there was refractory (Sandlos and Keeling, 2012).

Table 1: Estimates of aerial emissions of As_2O_3 dust from the Giant Mine roaster

Year	As_2O_3 (tonnes/day)[a]	As_2O_3 (tonnes/year)[b]	Year	As_2O_3 (tonnes/day)[a]	As_2O_3 (tonnes/year)[b]
1949-1951[c]	7.3	7963.6	1968	0.2	83.0
1952[c]	5.5	1989.3	1969	0.3	109.5
1953[c]	5.5	1989.3	1970	0.2	80.5
1954	5.5	1990.9	1971	0.9	320.2
1955	2.9	1061.8	1972	0.4	145.2
1956	2.7	995.5	1973	0.4	147.7
1957	3.0	1078.4	1974	0.2	80.5
1958[c]	1.5	547.5	1975-1992[c]	0.3	1698.9
1959	0.1	19.1	1991-1993[c]	0.1	54.8
1960	0.1	27.4	1994	0.01	3.6
1961-1963[c]	0.2	163.3	1995	0.01	3.5
1964	0.3	114.5	1996	0.01	3.5
1965	0.0	0.0	1997	0.02	7.6
1966	0.2	88.8	1998	0.015	5.3
1967	0.1	47.3	1999	0.01	3.7

Total As_2O_3 emissions (tonnes) = 20,824

[a] As_2O_3 emissions estimates were measured from the top of the roasting stack and are reported in Tait, 1961; CPHA, 1977; GNWT, 1993; Environment Canada, 2007; INAC, 2007; summarized after Wyre (2008).

[b] As_2O_3 emissions for the year(s) are assuming that roasting occurred for 365 days. This will be an overestimate of emissions because it is not known when the roaster was not in operation and the majority of As_2O_3 generated was in the first 10 years of roasting.

[c] Emissions for this year or years are estimated based on information from above sources and known changes to roaster operations.

Other roaster products at Giant Mine, including As-bearing maghemite and hematite were deposited with mine tailings, some of which were transported into local creek and lake sediments. Under reducing conditions post-depositional re-mobilization of As associated with roaster-derived Fe-oxides has released As into sediment pore-water (Fawcett and Jamieson, 2011), while As(III) in maghemite and As_2O_3 persists in the oxidizing conditions of near-surface tailings and soils (Walker et al., 2005). Recent work by Howell (2014) on select lakes demonstrates that roaster-emitted arsenic oxide also persists in regional lakes within ~5 km NE of the Giant Mine roaster and within 6.5 km from the City of Yellowknife (airport).

Due to the long history of anthropogenic release of As and the presence of As-enriched mineralized bedrock in the area, numerous studies have measured the concentration of As and other metal(loid)s in a variety of matrices in the Yellowknife region, including surface and pore waters, ground water, lake sediments, soils, trees and other plants, fungi, garden produce, and animals, including food fish (Table 1).

Table 2: Literature summary of As concentrations in environmental materials in the Yellowknife region[a]

Environmental Media	**Reference[b]**
Mine waste	EBA Engineering Consultants Ltd., 1998; Koch, 1998; Mace, 1998; Environmental Sciences Group, Royal Military College of Canada, 2000a, 2001a; EBA Engineering Consultants Ltd., 2001; SRK Consulting, 2004; Walker et al., 2005; Fawcett et al., 2006; Wrye, 2008; Fawcett, 2009; Bromstad, 2011; Fawcett and Jamieson, 2011; Bromstad and Jamieson, 2012; Nash, 2012; Jamieson et al., 2013; Jamieson, 2014; Fawcett et al., 2015; Walker et al., 2015
Bedrock	Boyle, 1960; Yamashita and Creaser, 1999; Cousens, 2000; Cousens et al., 2002; Ootes, 2004; Ootes et al., 2006
Till	Kerr and Wilson, 2000; Kerr, 2001, 2006
Soil	Hutchinson et al., 1982; EBA Engineering Consultants Ltd., 1998; Dave, 1999; Dene Nation, 1998; Ollson, 1999; Ollson et al., 2000; Environmental Sciences Group, Royal Military College of Canada, 2000b; Stephens et al., 2001; Kerr and Wilson, 2000; YASRC, 2001; Risklogic, 2002a; Golder Associates Ltd., 2005; Kerr, 2006; Wrye, 2008; Bromstad, 2011
Lake and stream sediments	Falk et al., 1973a,b[b]; Nickerson, 1972; Allen et al. 1972[b]; Moore et al., 1978; Wagemann et al., 1978[b]; Allan, 1979; Sutherland, 1989; Mudroch et al., 1989[b]; HydroQual Consultants Inc. and Gore & Storrie Ltd., 1989; Dushenko et al., 1995; Bright et al., 1994[b], 1996[b]; Jackson et al., 1996, Jackson, 1998; Koch, 1998; Mace, 1998; Ollson, 1999; Lorax Environmental, 1999; YASRC, 2001; Dillon Consulting Ltd., 2002a,b, 2004; Andrade, 2006; Fawcett et al., 2006; Fawcett, 2009; Andrade et al., 2010[b]; Stephen, 2011; Galloway et al., 2012; Nash, 2012; Golder Associates Ltd., 2006, 2013; Howell, 2014; Nasser, 2014; Fawcett et al., 2015
Ground water	INTERA Consultants Ltd., 1997; Bursey et al., 1999; Clark, 2001; Clark and Raven, 2004
Surface and pore waters	Falk et al., 1973a,b[b]; Moore et al., 1978; Hamilton et al., 1989; HydroQual Consultants Inc. and Gore & Storrie Ltd., 1989; Dushenko et al., 1995; Bright et al., 1996[b]; Jackson, 1989; Jackson et al., 1996; Jackson, 1998; Mace, 1998; Koch, 1998; Ollson, 1999; EBA Engineering Consultants Ltd., 2001; Environmental Sciences Group, Royal Military College of Canada, 2001c; Dillon Consulting Ltd., 2002a,b; Connell, 2002, 2003, 2004; Andrade et al., 2010; Golder Associates Ltd., 2006; 2013; SENES Consulting Ltd., 2006; Fawcett et al., 2015
Snow	Environment Canada, 1977; Stanley Associates Engineering Ltd., 1986; Ferguson Simek Clark Engineers & Architects, 1994
Vegetation, including aquatic plants, terrestrial plants, leaf litter, humus, tree bark, berries, garden produce, and fungi	O'Toole et al., 1972; Wagemann et al., 1978; Soniassy, 1979; Hutchinson et al., 1982; Dushenko et al., 1995; Dene Nation, 1998; Koch, 1998; Dene Nation, 1998; Davey, 1999; Nickerson, 1999; Akaticho Study, 2000; Koch et al., 2000a,b; Stephens et al., 2001; Obst et al., 2001; Environmental Sciences Group, Royal Military College of Canada, 2001b; Dunn et al., 2001, 2002; Dillon et al., 2002a; Chan, 2003; Kerr, 2006; St-Onge, 2007; Ollson et al., 2009
Animals, including invertebrates and fish	Falk et al., 1973a,b*; Wagemann et al.,1978’ Moore et al. 1978; Jackson et al., 1996; Koch, 1998; Koch et al., 2000a,b; Evans et al., 2001; Dillon et al., 2002a,b, 2004; Koch et al., 2003; DIAND, 2004; Golder Associates Ltd., 2004b; de Rosemond and Liber, 2004; de Rosemond et al., 2008.; Ollson et al., 2009; Koch et al., 2013; Cott et al. (submitted)

[a] This literature review focuses on reports of arsenic in environmental media in off mine lease areas; there is numerous literature, in addition to what is shown here, on the concentration of arsenic in environmental media from on mine lease areas.

[*] Location shown in Figure 2

Some studies have suggested that natural geochemical background concentrations of As in soils in the Yellowknife area can be as high as 150 mg/kg (YASRC, 2001; Reimer et al., 2002; Risklogic, 2002b), well above the Canadian Council of the Ministers of the Environment (CCME) guideline of 12 mg/kg for soils for the protection of human and ecosystem health (CCME, 2002). Territorial site-specific remediation objectives for As in Yellowknife area soils are 160 ppm for residential areas and 340 mg/kg for industrial areas (GNWT, 2003). The GNWT has also set 150 mg/kg as a site specific guideline for lake sediments at the Yellowknife boat launch (GNWT, 2003). However, previous studies have been largely focused on areas of concern, such as mine lease areas, known discharge pathways, the Giant Mine townsite, the Con Mine trailer court, and other sites likely to be impacted (e.g., Environmental Sciences Group, Royal Military College of Canada, 2001c). The body of previous work demonstrates that a substantial contamination problem exists in the Yellowknife region (Hutchinson et al., 1982;

Kerr and Wilson, 2000). For example, a report from 2006 indicates that As concentration in soils from the Giant Mine Townsite range into the 1000s of mg/kg (dry weight) (mean 998 ± 4.4 mg/kg, min=19, max=16,600, n=226) while As concentrations in soils from Latham Island average 106 ± 3.6 mg/kg (min=7, max=780, n=24), from the City of Yellowknife average 42 ± 3.8 mg/kg (min=2.5, max=1190, n=282), and from the community of Dettah average 26 ± 3.1 mg/kg (min=7.2, max=144, n=8) (SENES Consulting Ltd., 2006). However, comparing mean chemistry of soil samples can be problematic as concentrations vary with depth (e.g., Kerr, 2006). Comparison can be further limited by differences in the size fraction tested (e.g., <2 mm vs. <63 µm) and digestion protocol used.

Limited attention has been placed on assessing As concentrations outside of specific areas of concern or in lake sediments that pre-date industrial activity despite the fact that atmospheric emissions of As from the Giant Mine ore roaster resulted in widespread dispersion to the surrounding region (CPHA, 1977; Hocking et al., 1978; Wagemann et al., 1978; Bright et al., 1996; Kerr and Wilson, 2000; Andrade et al., 2010; Galloway et al., 2012). The paucity of sediment geochemical data from lakes outside of known impact areas currently limits the understanding of geochemical background and the extent of As dispersion in the Yellowknife region. Sites used as reference areas or control sites (e.g., Akaitcho Bay, Grace Lake) for environmental assessment (e.g., Golder Associates, 2006) are located proximal to mine sites to minimize variation associated with bedrock geology and other factors. However, recent work by Andrade et al. (2010) show elevated As, Cu, and Zn concentrations coincident with the beginning of mining in the region in a dated sediment core from Akaitcho Bay, Great Slave Lake, demonstrating downstream impact from mining and thus the unsuitability of this site as a reference location. The limited work on lake sediments deposited before initiation of gold mining or from sites un-impacted by mining nonetheless provide some evidence for what background and baseline As concentrations may be in lake sediments of the Yellowknife area (e.g., Mudroch et al., 1989; Bright et al., 1996; Andrade et al., 2010; Galloway et al., 2012). The studies by Bright et al. (1996), Mudroch et al. (1989), and Andrade et al. (2010) suggest that background As concentrations in Grace Lake, the mouth of Baker Creek at Great Slave Lake, Back Bay, Yellowknife Bay, and Akaitcho Bay (Great Slave Lake) may be as low as ~10-35 mg/kg. SENES Consulting Ltd. (2006) used As concentrations in sediment cores from Yellowknife Bay collected by Mudroch et al. (1989) to suggest that background As concentration in lake sediments is ‘on the order of 15 µg/g [mg/kg]’. Mudroch et al. (1989) consider background concentrations of As in sediments of Back and Yellowknife bays to be in the range of 15 to 25 mg/kg based on analysis of sediments 25 to 28-cm below the sediment-water interface captured in sediment cores. Based on ^{210}Pb and ^{137}Cs analyses, 13 cm depth in the Back Bay sediment core is dated as 1942 AD and 6 cm depth in the Yellowknife Bay sediment core is dated at 1947 cm (Mudroch et al. 1989). Therefore, the 25 to 28-cm interval referenced above most certainly pre-dates release of arsenic and other elements of concern associated with mining in the region (e.g., based on the linear sedimentation rate calculated based on the ^{210}Pb chronology, 25 to 28-cm could date as ~1889 AD to 1872 AD in the Yellowknife Bay sediment core and to 1921 AD to 1913 AD, assuming core collection in 1989; Mudroch et al., 1989).

Surface sediments from Grace Lake, often used as a control lake and located proximal (<5 km) to Con Mine, contain sediment As concentrations ranging from 15 to 105 mg/kg, while in Likely and Chitty lakes, located farther away (23 km N and 35 km NE, respectively) from Con Mine and on volcanic (Likely) and metasedimentary (Chitty) bedrock, As concentrations in near-surface sediments ranges from 19-38 mg/kg (Wagemann et al., 1978; Bright et al., 1996). Grab samples from Grace Lake (Falk et al., 1973a,b) were obtained after mining had commenced in the region and a contamination problem was recognized (Wagemann et al., 1978). The anomalous result of 105 mg/kg As concentration in surface sediments from this lake may represent background sediment concentration since Grace Lake is located proximal to Con Mine where ore was roasted and as Andrade et al. (2010) demonstrate, As can be upwardly mobile in lake sediments. Galloway et al. (2012) show in a survey of near-surface sediments from 19 lakes from a 90 km E-W transect along the Ingraham Trail and through the City of Yellowknife that As concentrations are mostly below the CCME Probable Effects Level (PEL) of 17 mg/kg (CCME, 2002) in sediment of lakes east of the City on metasedimentary bedrock. In lakes located near Giant and Con Mines and west of the City of Yellowknife on granitoid bedrock, As concentrations in sediments range up to 150 mg/kg, probably reflecting contamination from roasting of gold ore. However, analyses from only these few sites do not provide the information necessary to develop robust sediment quality guidelines for As and other elements of concern in the Yellowknife region. Additional sampling and analyses are required to determine influences on the distribution of elements of potential concern in regional lake sediments.

In this study we present new sediment geochemical data from lakes surrounding the City of Yellowknife. Some of these lakes have been influenced by accumulation of elements of potential concern from the legacy of mining in the study area. We also present sediment geochemical data from lakes located north of Yellowknife along the Tibbitt to Contwotyo Winter Road in the central Slave Geological Province and lakes west and south of Yellowknife in the Western Interior Platform to provide data for comparison to sites not likely impacted by mining activities. These data may provide insight into background and baseline geochemistry of lake sediments for the Yellowknife area as well as the central Northwest Territories and contribute to a better understanding of the environmental impact of mining.

2. STUDY AREA

The City of Yellowknife and surrounding area is located in the southwestern Slave Geological Province, District of Mackenzie (Figure 1). Elevation in the region rises gradually from 157 m above sea level (MSAL) near Great Slave Lake to approximately 400 MASL north of 63° latitude. The Yellowknife River is the main drainage for the area. Its southern outlet flows into Yellowknife Bay, Great Slave Lake. Many lakes east of Yellowknife lie within the Cameron River-Prelude Lake watershed. Drainage in the region is influenced by bedrock structure; numerous small elongate lakes have formed along fault and joints in the bedrock.

Lakes northeast of the City of Yellowknife along the Tibbitt to Contwoyto Winter Road and associated waterbodies drain into four primary drainage basins: the Great Slave Lake and Mackenzie River Drainage Basin, the Coppermine River Drainage Basin, the Back River Drainage Basin and the Hood and Burnside rivers Drainage Basin. On the southern end, within the Great Slave Lake and Mackenzie River Drainage Basin, the winter road traverses the Cameron, Beaulieu and Lockhart river watersheds.

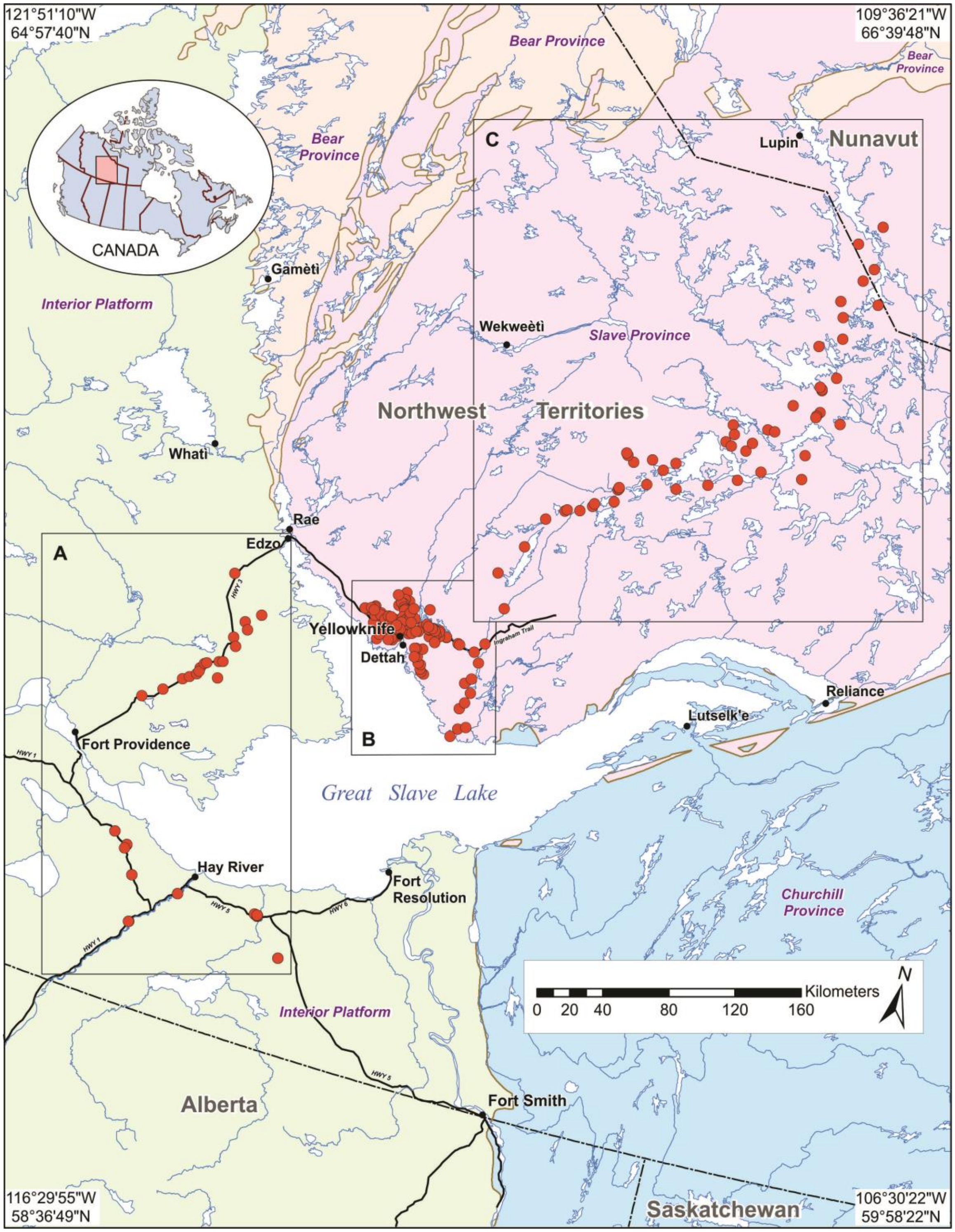

Figure 1: Map showing sampling locations and the regional study area with geological provinces of the central Northwest Territories shown by colour. Outlined areas shown in Figure 3 (Western Interior Platform; inset A), Figure 4 (Yellowknife area and Ingraham Trail; inset B), and Figure 5 (Tibbitt to Contwoyto Winter Road; inset C)

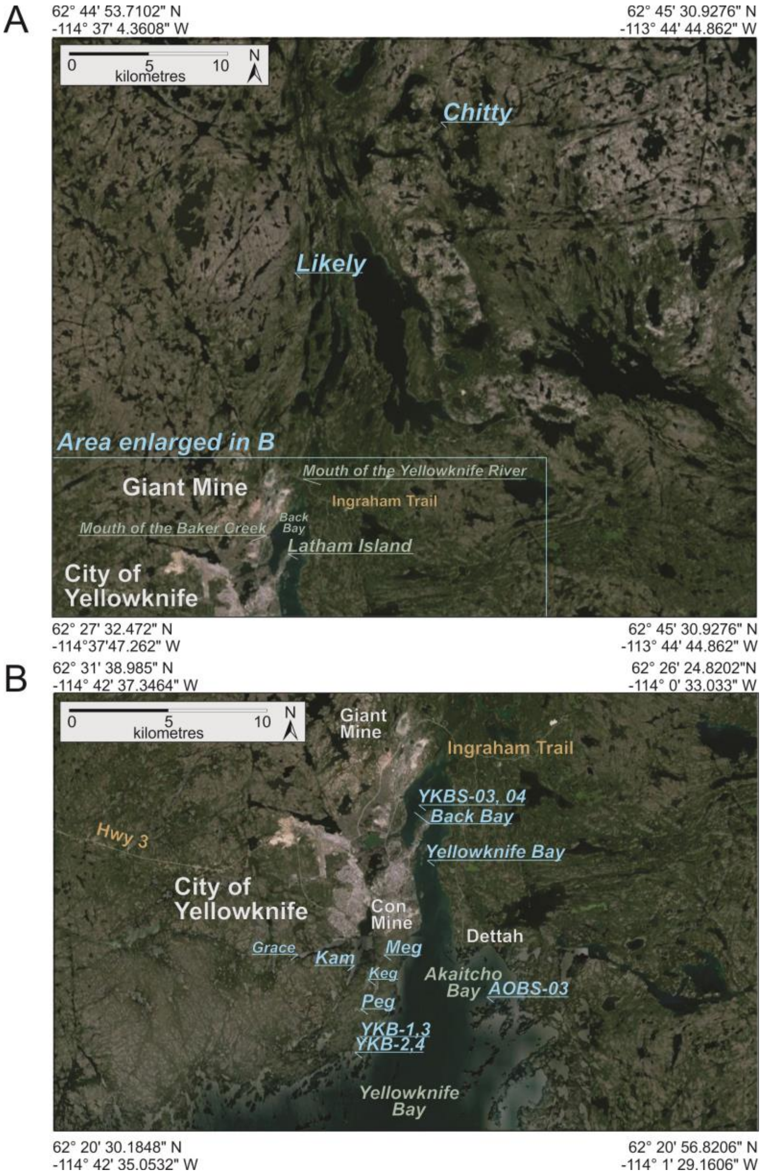

Figure 2: Satellite imagery showing the location of previous lake sediment sampling near the City of Yellowknife (source satellite imagery: Google Earth, 2015). A - Likely and Chitty lake sediment surface samples (0-20 cm) (Wagemann et al., 1978); B - Grace, Meg, Keg, Peg, Kam, Yellowknife Bay-3 (YKB-3) and Yellowknife Bay-4 (YKB-4) sediment surface samples (0-5 cm) (Bright et al., 1994, 1996); Grace, Meg, Keg, Peg, Kam, Yellowknife Bay-1 (YKB-1), Yellowknife Bay-2 (YKB-2) lake sediment cores (Bright et al., 1996); Back Bay and Yellowknife Bay sediment cores (Mudroch et al., 1987); Grace, Kam, Keg lake sediment surface samples (0-20 cm) (Wagemann et al. (1978); YKBS-03, 04 and AOBS-03 sediment cores (Andrade et al., 2010)

2.1 Climate and vegetation

The Hay River Lowland of the Taiga Plains Ecozone occurs west and south of Great Slave Lake (Ecosystem Classification Group, 2007). This broad lowland plain is characterized by a subhumid mid-boreal climate. The mean summer temperature is 13°C and mean winter temperature is -19°C. Mean annual precipitation ranges from 350 to 450 mm. Vegetation is composed of closed mixed stands of trembling aspen (*Populus tremuloides*), balsam poplar (*Populus balsamifera*), white spruce (*Picea glauca*), balsam fir (*Abies balsamea*), and black spruce (*Picea mariana*) on dry or boggy sites. Poorly drained fens and bogs cover 30% of the ecoregion and also support larch (*Larix laricina*).

West of the North Arm of Great Slave Lake lies the Great Slave Lake Plain of the Taiga Plains Ecozone (Ecosystem Classification Group, 2007). The ecoregion has a low subarctic climate and is underlain by discontinuous permafrost. Mean annual temperature is -6°C with a mean summer temperature of 11.5°C and mean winter temperature of -25°C. Mean annual precipitation ranges between 225 to 300 mm. The ecoregion is dominated by open stands of black spruce and groundcover or Labrador tea (*Ledum decumbens*), ericaceous shrubs, and mosses. Wetlands cover about half of the region.

The Yellowknife study area occurs south of the treeline and lies within the Great Slave Lake Lowland and Great Slave Lake Upland ecoregions of the Taiga Shield Ecozone (Ecosystem Classification Group, 2007). Climate is sub-humid, high-boreal, and characterized by a mean summer temperature of 11°C and a mean winter temperature of -21.5°C. Mean precipitation ranges between 200 and 375 mm. Vegetation is composed of closed stands of trembling aspen, balsam poplar, paper birch (*Betula papyrifera*), and jack pine (*Pinus banksiana),* with white spruce and black spruce dominating later successional stands. Poorly drained fens and bogs are covered with open stands of larch and black spruce.

Northeast of Yellowknife, the Tibbitt to Contwoyto Winter Road (TCWR) traverses the central Slave Geological Province. The southern end of the TCWR begins in the Tazin Lake Upland Ecoregion of the Taiga Shield Ecozone and travels north into the Coppermine River Upland Ecoregion of the Southern Arctic Ecozone near latitudinal treeline and then into the barrenlands of the Takijuq Lake Upland Region of the Southern Arctic Ecozone near Lac de Gras (Ecosystem Classification Group, 2007). The Coppermine River Upland Ecoregion is characterized by a high subarctic climate with a mean summer temperature of 9 °C and mean winter temperature of -24.5 °C. Annual precipitation ranges from 200 to 300 mm. Vegetation consists of open, stunted stands of black spruce and larch with lesser amounts of white spruce. Shrub or ground cover in these open communities consists of dwarf birch (*Betula glandulosa*), willow (*Salix spp.*), ericaceous shrubs, such as northern Labrador tea and red bearberry (*Arctostaphylos rubra*), cotton grass (*Eriophorum spp.*), and lichen. Poorly drained areas support tussocks of sedge (*Carex spp.*), cotton grass and sphagnum moss (*Sphagnum spp.*). Low shrub tundra consisting of dwarf birch and willow are also common in this ecoregion. Climate of the Takijuq Lake Upland Ecoregion of the Southern Arctic Ecozone is characterized as low arctic, with a mean summer temperature of 6°C and mean winter temperature of -26.5°C (Wiken, 1986). Annual precipitation ranges from 200 to 300 mm. Vegetation is composed of shrub tundra consisting of dwarf birch, willow, Labrador tea, and ericaceous plants. Low-lying sites support willow, sphagnum moss, and sedge. Scattered stands of spruce may occur along the southern boundary of the ecoregion.

2.2 Bedrock geology

The study area covers two distinct geological districts (Figure 1). The southern and western portion is underlain by the flat-lying Paleozoic sedimentary successions of the Western Interior Platform while the northern portion of the study area, including the Yellowknife region, rests of the Canadian Shield consisting of the Archean to Neo-Proterozoic rocks of the Slave Geological Province.

Bedrock of the Western Interior Platform geological province is composed of a succession of Phanerozoic sedimentary rocks that range in age from Cambro-Ordovician to Devonian and overly Archean and Proterozoic crystalline basement rocks that include Archean cratons (Slave and Rae provinces), accreted Proterozoic terranes, and magmatic arcs (Gebert, 2008). This sedimentary cover includes quartz-rich and carbonate sandstone, mudstone, dolomite, and limestone (e.g., Rhodes et al., 1984).

The main bedrock elements of the Slave Geological Province are founded on foliated granodioritic to tonalitic rocks and related granitoid gneiss, termed the Central Slave Basement Complex (Bleeker and Davis, 1999). A thin, ca. 2.8 Ga cover sequence, conformably overlies the basement complex and is structurally overlain by post 2.27-2.66 Ga volcanic and turbiditic sedimentary rocks belonging to the Yellowknife Supergroup (Villeneuve et al., 1997). The supracrustal rocks were intruded by several different generations of magma, including syn-volcanic felsic to mafic dykes and sills, three suites of Post-Yellowknife Supergroup granites, and Proterozoic intrusions (predominately diabase dykes) (Kjarsgaard et al., 2002). In the Yellowknife study area, the bedrock is dominated by mafic meta-volcanics of the Yellowknife Supergroup (2.71-2.65 Ga; Villeneuve and Relf, 1998) that include basalt, andesite, and pillowed flows that trend north-south and host the major gold deposits. East of Yellowknife, Archean meta-sedimentary rocks predominate and consist of greywacke, slate, schist, and phyllite. Yellowknife Supergroup meta-volcanics and meta-sedimentary rocks are intruded by younger granitoid rocks. West of Yellowknife, granitoid intrusions, consisting of granite, granodiorite, and tonalite, compose the majority of the bedrock. The region is crosscut by early Proterozoic diabase and gabbro dykes and several major fault lines, such as the Kam Lake Fault and the West Bay Fault that run through the city of Yellowknife, separating the volcanic rocks from younger granitoids (Yamashita and Creaser, 1999; Yamashita et al., 1999; Cousens, 2000; Cousens et al., 2002).

In the northern part of the study region, Yellowknife Supergroup rocks consist of a thinly bedded (1-10 cm), graded, psammite-pelite succession with minor volcanic rocks (Kjarsgaard and Wyllie, 1994). Plutonic rocks intruded the lithological units of the Central Slave Basement Complex, the Central Slave Basement Complex cover sequence, and the Yellowknife Supergroup between 2.71 and 2.58 Ga (Villeneuve et al., 1997). The region is transected by a variety of un-metamorphosed Proterozoic diabase dykes and over 150 Cretaceous to Eocene kimberlite pipes (Kjarsgaard and Heaman, 1995; Davis and Kjarsgaard, 1997; Kjarsgaard et al., 2002) that have resulted in a lucrative Canadian diamond industry. Numerous greenstone belts (e.g., Courageous Lake greenstone belt) in the Slave Geological Province have resulted in gold exploration and exploitation throughout the region. For detailed description of each component, the reader is referred to Hoffman and Hall (1993) and Kjaarsgaard et al. (2002).

2.3 Surficial Geology

Bedrock of the Western Interior Platform is overlain by blanks and veneers of fine-grained glaciolacustrine sediments associated with Glacial Lake McConnell that formed following deglaciation of the region between 10,000 and 9,600 ^{14}C yr BP (Smith, 1994; Dyke, 2004). Till deposits range from <1 to 3 m thick to >25 m thick, with thickness increases locally where karst collapse have created depressions in the bedrock surface. Historic drill hole data show that till can be up to 100 m thick in paces (Lemmen, 1990). Till in the area has a silt to fine sand matrix with very little clay (Lemmen, 1998). Till, moraine, and glaciofluvial deposits are extensively reworked into beach deposits composed of bedded sand and gravel (Lemmen, 1998; Gebert, 2008). In other places, fills were washed by wave action that produced a discontinuous coarse lag. Wind action following drainage of the glacial lake formed locally concentrated fine-grained glaciolacustrine sediments into eolian dunes and ridges. Recent deposits in this region include alluvial sediments and organic accumulations in bog and fen settings (Gebert, 2008).

The surficial geology of the Yellowknife region is dominated by a mosaic of Glacial Lake McConnell sediments and glacial tills that infill the topographic lows of the abundant bedrock outcrops. Till is primarily matrix-supported diamicton (Kerr and Wilson, 2000). Clasts consist of various locally derived lithologies and range in size from small pebbles to large boulders. Till in the Yellowknife area may be composed of up to 60% clasts, but most exposures contain approximately 20% to 40% (Kerr and Wilson, 2000). Till exposures have been generally eroded, are less than 2 m thick, and form discontinuous cover in topographic lows or on bedrock outcrops. Glaciofluvial sediments are relatively uncommon in the study region, and where present consist of fine sand to cobbles in the form of eskers, kames, and outwash (Kerr and Wilson, 2000). A number of surficial sedimentary deposits may be attributed to Glacial Lake McConnell, which infilled Great Slave Lake, Great Bear Lake, and Athabasca Lake basins during deglaciation between 11,800 and 8,300 years ago (Dyke and Prest, 1987; Smith, 1994; Kerr and Wilson, 2000; Wolfe et al., 2014). Sedimentary deposits of Glacial Lake McConnell consist of poorly to moderately sorted coarse to fine sand, silt, and clay that can be up to 20 m thick in some topographic lows (Kerr and Wilson, 2000). These sediments may overlie till, outwash, or bedrock and finer grained sediments deposited in deep water environments, and may be overlain by sand and gravel deposited in regressive fluvial or littoral successions. Accumulations of Holocene-aged peat also occur in the study area and can be 1 m thick or greater in bogs and other low-lying wetland types (Kerr and Wilson, 2000).

The Lac de Gras area (NTS map sheets 76C and 76D) was covered by the Laurentide Ice Sheet during the last glaciation (Dyke et al., 2002). A transition from southwesterly to northwesterly ice flow occurred during late glacial time (Dredge et al., 1995; Ward et al., 1997). The timing of deglaciation is constrained by a single terrestrial ^{14}C date from plant material contained within an esker of 8500 yr BP, suggesting the landscape was partially deglaciated at this time (Dredge et al., 1995). Sediment cover in this region is mostly composed of a single unstratified till unit. Dredge et al. (1995) and Ward et al. (1997) describe the till as having silty-sandy to sandy matrix and a clast content varying from 5 to 40%. Till can be divided based on surface morphology as well as thickness from till veneer, till blanket, to hummocky till. Till silt content is likely derived from local bedrock sources (granitoids or Yellowknife Supergroup metasediments; Stubley, 2005). Areas of hummocky till have a different geochemical signature to thinner till for some kimberlite pathfinder elements (e.g., Ba, Cr, Mg, Ni; Wilkinson et al., 2001). Variations are attributed to the different bedrock sources of till and transport distances as well as attrition and mixing. Kimberlite indicator minerals such as pyrope, almandine-spessartine garnet, grossular and andradite garnet, diopside, chromite, ilmenite, and olivine are associated with a specific source at transport distances exceeding 70 km (McClenaghan et al., 2002). Modified shaped gold grains with unidentified sources and unknown transport distances are also present (Kerr and Knight, 2007). Potential sources of indicator minerals and till matrix geochemical anomalies include kimberlites, ultramafic dykes, and known VMS showings as well as a variety of new base, precious, and rare metal showings such as porphyries, alkaline intrusions, and pegmatite. On a landscape scale, stagnant ice associated with the last glaciation created large areas of hummocky till and moraine ridges in the central Slave Geological Province (Kerr et al., 1997). Massive ground ice or icy sediment within till blankets may be present northeast of Contwoyto Lake, evidenced by stabilized thawslides (Wolfe, 2000). Glaciofluvial sediment in the region is commonly contained within tracts of thin or scoured till and exposed bedrock and consists of fine sand to cobbles in the form of eskers, kames, and proglacial outwash

(Wolfe, 2000). The tracts, or "corridors" are oriented towards the west of northwest. The relative influence of subglacial scouring, meltwater erosion, and proglacial stagnant water on the origin of these corridors is still debated. Eskers range from small ridges a few tens of metres long to large complexes up to 32 km long and 20-30 m high. Outwash plains with braided channels and kettle lakes are sometimes associated with eskers (Wolfe, 2000). A number of erosional and depositional shoreline landforms associated with a sequence of proglacial lakes that formed in the region surrounding Contwoyto Lake during the last deglaciation are present. These include perched deltas, raised beaches, terraces and spits, wave-cut beaches and washed boulder till surfaces that occur up to 45 m above present lake level (Kerr et al., 1997). Alluvial sediments consisting of gravel and silt sized sediment deposited by modern streams and rivers are also present (Wolfe, 2000). Accumulations of peat up to 1 m thick are present in topographic lows and poorly drained areas.

2.4 Bedrock and till geochemistry in the Yellowknife region

Due to the high mineral potential of bedrock in the Yellowknife area a large body of literature exists on bedrock geochemistry (Table 2). Arsenic concentrations in granitoid and gneissic rocks in the region are ~2 mg/kg (Boyle, 1960). However, in mineralized granitoid rocks As concentrations can range up to 90 mg/kg (Kerr and Wilson, 2006). Globally, As concentrations in acidic and intermediate igneous rocks are between <0.1-15 mg/kg (Turekian and Wedepohl, 1961; Koljonen, 1992; Smedley and Kinninburgh, 2002). Arsenic concentration in metasedimentary bedrock ranges from 10 ppm to 30 mg/kg (Boyle, 1960), within the range reported for global metamorphic rocks (<0.1-143 mg/kg; Smedley and Kinninburgh, 2002). Arsenic content in meta-volcanic bedrock in the Yellowknife region ranges from 1 mg/kg to 33 mg/kg (Yamashita and Creaser, 1999; Yamashita et al., 1999; Cousens, 2000; Cousens et al., 2002; Ootes, 2004; Ootes et al., 2006), comparable to the concentrations reported for basic and ultrabasic igneous rocks from around the globe (0.03-113 mg/kg; Turekian and Wedepohl, 1961; Smedley and Kinninburgh, 2002) but elevated relative to continental curst (Rudnick and Gao, 2004) (Table 2). Relative to average crustal composition, granitoid rocks of the Yellowknife area are enriched in As and Pb with lower concentrations of Cr, Cu, and Zn (no data for Cd or Hg) but concentrations are comparable to global granitoid bedrock values. Meta-sedimentary bedrock in the Yellowknife region is enriched in Cr and Zn (no data for Cd, Pb, or Hg) relative to global values for meta-sedimentary and sedimentary rocks (Table 2).

Tills in the Yellowknife region can contain As with concentrations up to 1560 mg/kg within *in situ* weathered material over mineralized zones, although typically As concentrations are between 5 to 30 ,g/kg in regional tills (Kerr, 2006). The As concentrations in glaciofluvial, glaciolacustrine, and peat deposits in the region are not known.

The geochemistry of bedrock and surficial materials in the Western Interior Platform and central Slave Geological Province are not known.

Table 3: Summary of bedrock geochemistry in the Yellowknife region and comparison to global values

Bedrock Type	**Median metal(loid) concentrations (mg/kg)[a] (min, max, n)[b]**						
	As	**Cd**	**Cr**	**Cu**	**Pb**	**Hg**	**Zn**
Till							
Kerr (2006)[c]	22.7 (14.2-1560, n=152) INAA clay+silt; 93 (28-702, n=14) ICP-ES clay	No data	82.5 (29-640, n=80) INAA clay+silt; 118 (90-226, n=14) ICP-ES clay	108 (54-295, n=14) ICP-ES clay	20 (10-58, n=14) ICP-ES clay	Below detection limit	50, 25-290, n=80) INAA; 71 (54-250, n=14) ICP-ES clay

Granitoid							
Boyle (1960); Yamashita et al. (1999); Cousens et al. (1999); Kerr (2006)	2 (no range reported; up to 90 near mineralized zones	No data	7 (0-72, n=22)	8 (3-40, n=22)	20 (11-37, n=22)	No data	44 (5-86, n=22)
Global (Turekian and Wedepohl, 1961; Koljonen, 1992; Smedley and Kinninburgh, 2002)	<0.1-15	0.13	2-22	5-30	12-19	0.08	39-130
Meta-sedimentary							
Boyle (1960); Jackson (1974) Yamashita and Creaser (1999); Kerr (2006)	2-64	No data	153 (101-265, n-38)	56.5 (4-106, n=38)	No data	No data	83 (33-152, n=38)
Global (Smedley and Kinninburgh, 2002; Turekian and Wedepohl, 1961)	<0.1-143	0.3 (shale)	35 (sst); 90 (shale)	45 (shale)	7 (sst)	0.03 (sst); 1.4 (shale)	16 (sst); 95 (shale)
Meta-volcanic							
Yamashita and Creaser (1999); Cousens (1999); Cousens et al. (2002); Ootes (2004); Ootes et al. (2006); Kerr (2006)	8 (1-33, n=15)	No data	136 (3-1061, n=144)	62 (0-230, n=60)	No data	No data	85 (1-172, n=144)
Global (Smedley and Kinninburgh, 2002; Ultrabasic and basaltic rocks - Turekian and Wedepohl, 1961)	0.03-113	0.22	170-1600	10-87	1-6	0.09	50-105
Composite bedrock in Yellowknife region	5 (1-33, n=15)	No data	128 (0-1061, n=205)	43 (0-230, n=120)	21 (11-37, n=21)	No data	80 (1-172, n=204)
Continental Crust) (Rudnick and Gao, 2004)	2.25 (0.2-5.1, n=18)	0.09 (0.06-0.2, n=18)	106 (32-490, n=34)	28 (8-90, n=28)	13 (2.2-22, n=33)	0.03 (0.01-0.10, n=15)	72 (50-102, n=28)

[a]1/2 method detection limit used where concentration was below method detection; [b]min, max, n when reported; [c]clay+silt <63 µm; clay <2 µ; sst – sandstone

3. METHODS

Two-hundred and eleven near-surface sediment samples from 196 lakes were collected during summer and fall 2009-2014 from canoe and helicopter (Figures 1, 3, 4, 5). The sediment samples were collected from four main regions: the Western Interior Platform, Yellowknife area, Ingraham Trail, and the Tibbitt to Contwoyto Winter Road (Figures 1, 3, 4, 5). Elemental analyses on a subset of near surface sediment samples from the Yellowknife region are reported in Galloway et al. (2012). Surface water chemistry of the lakes along the TCWR are reported in Macumber et al. (2012). Surface water chemistry in the Yellowknife area will be reported in Part 2 of this report. The locations of samples are tabulated in Appendix 1.

Figure 3: Map showing the location of lake sediment sample locations in the Western Interior Platform region

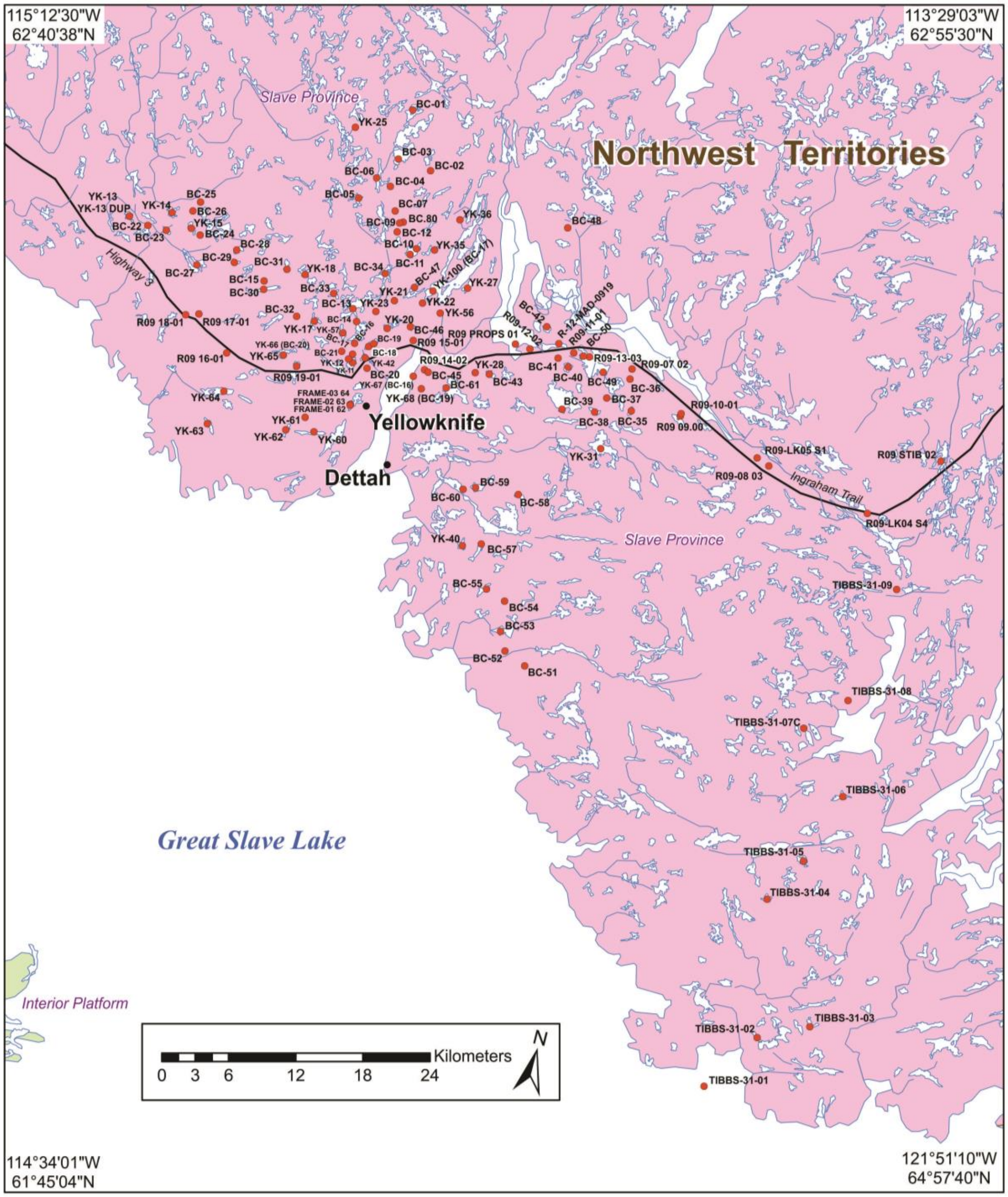

Figure 4: Map showing the location of lake sediment sample locations in the Yellowknife region and along the Ingraham Trail

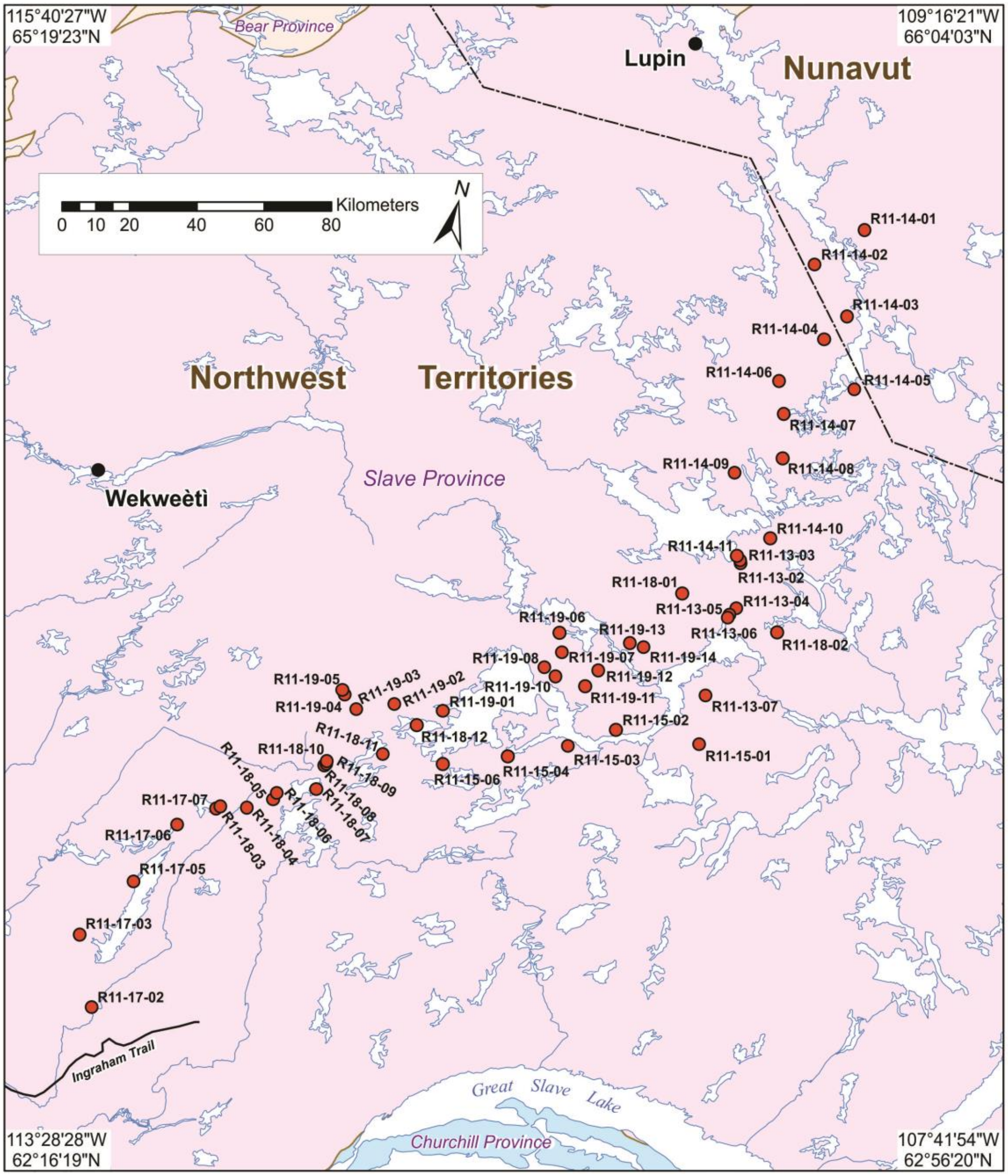

Figure 5: Map showing the location of lake sediment sample locations along the Tibbitt to Contwoyto Winter Road (TCWR)

3.1 Sampling

Many of the lakes located along Highway 3 and the Ingraham Trail were targeted based on accessibility by canoe (Galloway et al., 2012). Lakes along the TCWR were accessed by truck in winter and helicopter in summer. Lake bathymetry is not known for sample lakes. Metadata and water quality variables were measured at one metre intervals using a multi-metre probe at the time of collection (Galloway et al., 2012; Macumber et al., 2012). Near-surface sediment samples were collected using an Ekman Grab sampler. The top 2 to 5 cm of sediment retrieved was sub-sampled for element geochemical analyses. Samples were kept cool in the field and during shipping to Carleton University where they were kept cold at 4 °C until submission to the laboratory for geochemical analyses.

3.2 Geochemistry

Sediment sub-samples were submitted to Acme Laboratories (now Bureau Veritas), Vancouver, for geochemical analyses. Sub-samples were freeze dried and screened to -180 µm (-80 mesh ASTM) at the laboratory. Concentrations of elements in sediment samples were determined by inductively coupled plasma-mass spectrometer (ICP-MS) (ICP-MS 1F/AQ250 package). Samples were digested using modified *aqua regia* digestion with the exception of phosphorus, which was extracted using $NaHCO_3$. Near total digestion was used to extract metals that could become bioavailable and because complete digestion methods that involve high-temperature fuming can volatilize As and Sb, both contaminants of potential concern in this study (Parsons et al., 2012). Certified reference material was not submitted for laboratory quality control/quality assessment. Two pulp duplicates generated by the laboratory for sediment samples collected in the Yellowknife area show that Relative Percent Difference ranges from <0 to 66.7% for one pulp duplicate sample when RPD for two elements (Sn and B) are removed and from <0 to 48% (all elements included) for the other pulp duplicate sample.

4. RESULTS

Two-hundred and eleven near-surface sediment samples were collected from 196 lakes within three ecozones (Figures 2-5). Metadata and physical limnological characteristics are also reported in Galloway et al. (2010), Macumber et al. (2012), and Galloway et al. (2012) for a sub-set of the lakes sampled along the Ingraham Trail and the Tibbitt to Contwoyto Winter Road.

4.1 Element geochemistry

Element geochemistry results are reported in Appendix 1. The median As concentration for all lake sediment samples is 21.3 mg/kg (min=0.05, max=>10,000, n=211). Where element concentration was below method detection we used ½ of the method detection limit in statistical analyses. Where element concentration exceeded method detection, we used the upper method detection limit in statistical analyses. Exceedance of method detection limit occurred in a single sample for As. Significant figures reported by Acme Laboratories are retained in this report for each analyte.

Yellowknife area lake sediment samples contain the highest median As concentration (median 107.9 mg/kg, min=6.3, max=>10,000, n=95) (Table 3, Figure 4) compared to sediment samples from the lakes in the Western Interior Platform and along the Ingraham Trail and Tibbitt to Contwoyto Winter Road. The distribution of As concentration in lake sediment samples from the Yellowknife area is skewed (skewness=6.35) relative to the distribution of As for the other regions. Twenty-five percent of samples from the Yellowknife area contain As concentrations above 295 mg/kg. Median As concentrations in samples from all other regions sampled are below the CCME PEL of 17 mg/kg for this element. Element geochemistry results are compared to the Canadian Council of Ministers of the Environment Interim Sediment Quality Guidelines (ISQG) and Probable Effect Levels (Table 3).

Table 4: Descriptive statistics of metal(loid) concentrations (mg/kg) in lake sediment samples from the four regions sampled and comparison to Canadian Council of Ministers of Environment and Probable Effect Levels (PEL) for the Protection of Aquatic Life are included for comparison. Results from previous regional (Puznicki, 1997) and national (Friske and Hornbrook, 1991) sediment geochemistry surveys also included for reference.

Descriptive statistic	**Element concentration[a] (mg/kg)**						
	As	**Cd**	**Cr**	**Cu**	**Pb**	**Hg**	**Zn**
Method detection limit	*0.01*	*0.01*	*0.5*	*0.01*	*0.01*	*0.005*	*0.01*
CCME ISQG[c] (mg/kg)	5.9	0.6	37.3	35.7	35	0.17	123
CCME PEL[d] (mg/kg)	17	3.5	90	197	91.3	0.486	315
NGR[e] mean in lake sediments (Friske and Hornbrook, 1991) (mg/kg)	2.5	0.32	47	31	6	0.074	104
Slave Structural Province (Puznicki, 1997) (mg/kg)[f]	~83% samples <17	~70% samples <MDL[g] (0.2)	68% of samples < 37.3.	99% samples < 197, 81% of samples within 12-57	Mean 17, 98% samples <91	86% of samples <0.20	Mean 90, 86% of samples <120, 98% of samples <315
All samples (n=211)							
Minimum	0.05	0.03	1.1	0.84	0.60	0.025	9.80
Maximum	>10000.00[h]	0.95	74.2	141.32	48.52	0.369	344.60
Range	9999.95	0.92	73.1	140.48	47.92	0.366	334.80
Interquartile range	107.10	0.24	21.6	25.80	5.85	0.059	49.10
Median	21.30	0.30	21.2	26.10	5.88	0.071	77.60
Arithmetic Mean	199.08	0.32	23.3	32.26	7.30	0.079	82.25
Standard Deviation	823.21	0.18	14.8	23.95	6.45	0.0535	48.40
Skewness	9.30	0.74	0.7	1.32	3.43	0.0197	1.98
Western Interior Platform samples (n=37)							
Minimum	0.05	0.03	1.1	0.84	0.60	0.003	9.80
Maximum	7.10	0.52	23.5	28.72	13.68	0.107	133.50
Range	7.05	0.49	22.4	27.88	13.08	0.105	123.70
Interquartile range	1.33	0.22	4.15	6.78	2.40	0.031	32.10
Median	1.10	0.19	3.9	5.71	2.32	0.039	41.10
Arithmetic Mean	1.37	0.20	5.4	6.93	3.54	0.040	45.05
Standard Deviation	1.27	0.13	5.2	5.91	3.45	0.024	28.45
Skewness	2.74	0.54	2.29	1.83	1.82	0.578	1.23
Ingraham Trail samples (n=27)							
Minimum	1.90	0.04	4.9	3.87	0.71	0.003	11.60
Maximum	101.60	0.44	51.1	53.19	13.45	0.154	190.10
Range	99.70	0.40	46.2	49.32	12.74	0.152	178.50
Interquartile range	17.28	0.14	20.08	10.15	5.53	0.049	24.15
Median	10.60	0.25	24.3	24.49	6.34	0.070	67.10
Arithmetic Mean	21.04	0.24	26.6	24.72	6.78	0.073	71.99
Standard Deviation	26.28	0.11	13.2	10.28	3.46	0.043	32.57

Table 4 cont.

Descriptive statistic	Element concentration[a] (mg/kg)						
	As	Cd	Cr	Cu	Pb	Hg	Zn
Method detection limit	*0.01*	*0.01*	*0.5*	*0.01*	*0.01*	*0.005*	*0.01*
Skewness	2.20	-0.19	0.3	0.49	0.22	0.093	1.45
Yellowknife area samples (n=95)							
Minimum	6.30	0.06	2.8	11.39	1.66	0.019	19.30
Maximum	>10000.00	0.83	56.1	86.71	48.52	0.369	344.60
Range	9993.7	0.77	53.3	75.32	46.86	0.350	325.30
Interquartile range	295.48	0.27	14.53	14.12	5.38	0.066	42.50
Median	107.90	0.34	21.2	25.96	7.85	0.095	86.30
Arithmetic Mean	426.524	0.35	24.0	29.53	9.96	0.101	91.57
Standard Deviation	1190.93	0.16	11.0	14.21	8.23	0.061	52.41
Skewness	6.35	0.43	0.8	1.62	2.83	1.985	2.53
Tibbitt to Contwoyto Winter Road samples (n=52)							
Minimum	0.30	0.04	6.1	14.77	1.73	0.003	33.30
Maximum	101.40	0.95	74.2	141.32	11.91	0.207	238.50
Range	101.10	0.91	68.1	126.55	10.18	0.204	205.20
Interquartile range	14.25	0.25	16.55	31.40	2.58	0.042	57.25
Median	7.90	0.38	30.3	57.45	5.32	0.066	90.00
Arithmetic Mean	16.67	0.40	33.0	59.17	5.37	0.069	97.01
Standard Deviation	22.18	0.20	15.2	25.75	2.19	0.037	45.01
Skewness	2.51	0.76	0.8	0.58	0.78	1.164	1.12

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[a] – ½ of the method detection limit used where concentration was below method detection; [b]mg/kg – milligram per kilogram; [c]ISQG – Interim Sediment Quality Guideline; [d]PEL – Probable Effect Level; [e]NGR – National Geochemical Reconnaissance Programme; [f]estimated from figures in report because digital data was not available, cutoffs used for description of data were the Ontario Sediment Quality Guidelines (Jaagumagi and Persaud, 1996) and National Interim Guideline. Mean reported for some elements as reported by Puznicki (1997); [g]MDL – method detection limit; Results are compared to CCME (2002) guidelines where possible; [h]when concentration exceeded method detection the upper method detection limit was used for statistical analyses. ½ of the method detection limit was used for statistical analyses when element concentration was below detection.

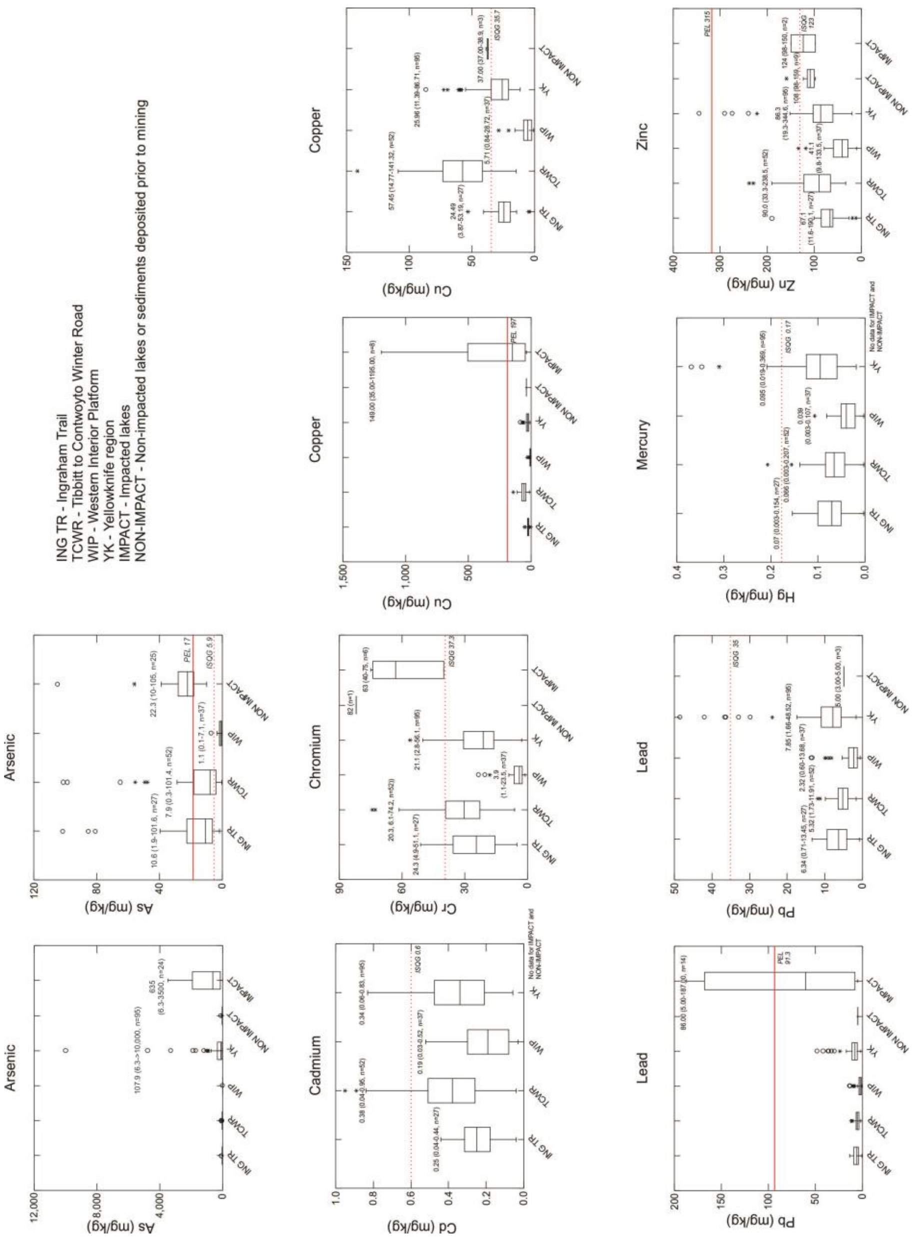

Figure 6: Box and Whisker plot of metal(loid) concentrations elements in lake sediment samples by region. Impact and non-impact samples summarized from literature shown in Table 4. Descriptive statistics shown in Tables 3 and 4. For As, Cu, and Pb, sample categories with high element concentrations are removed and a second plot is shown to visualize the distribution of element concentration in samples from remaining categories. Box represents interquartile range (IR), with bottom of box representing the 25th percentile and top of box representing the 75th percentile. Line in box presents median. Bottom whisker denotes data within 0.5*IR, top whisker denotes data within 1.5*IR, star outliers denote data within 3*IR, open circle denote extreme outlier data. ISQG – Interim Sediment Quality Guideline (CCME, 2002); PEL – Probable Effect Level (CCME, 2002).

5. DISCUSSION

5.1 Regional variation in sediment geochemistry

To provide insight on the provenance of As and other metal(loid)s in sediments of lakes near the City of Yellowknife, we compare element concentrations from sediments in the Yellowknife area with those from the Western Interior Platform, east of the city along the Ingraham Trail, and northeast through the central Slave Geological Province along the TCWR. We summarize data presented in the literature from known impacted sites in the Yellowknife area, and to probable non-impacted sites in the region (Figures 1, 2a-d; Table 4). It should be noted that differences in the grain size fraction of sediments analyzed, digestion protocols, and analytical method could limit the validity of comparison of our dataset to previous work.

Table 5: Metal(loid) concentrations in lake sediments of the Yellowknife area summarized from selected published literature* and compared to concentrations in lake sediments sampled as part of this study. Canadian Council of Ministers of the Environment (2002) guidelines and regional data are also shown.

Reference	Site	Sample Type	Concentration (mg/kg)[a]						
			As	Cd	Cr	Cu	Pb	Hg	Zn
NON-IMPACTED SAMPLES[b]									
Bright et al. 1996	Grace Lake	Core (0-5 cm)	22						
Bright et al. 1996		Core (5-10 cm)	56						
Bright et al. 1996		Core (10-15 cm)	15						
Bright et al. 1996		Core (15-18 cm)	23						
Bright et al. 1994		Grab (0-5 cm)	22.3		82	38.9	5		159
Falk et al. 1973a,b		Grab (0-20 cm)	105*						
Falk et al. 1973a,b		Grab (0-20 cm)	34						
Falk et al. 1973a,b		Grab (0-20 cm)	26						
Nickerson, 1972; Allen et al. 1972		Grab (0-20 cm)	20						
Bright et al. 1996	Yellowknife Bay, Great Slave Lake	Core 1 (15-18 cm)	34						
Bright et al. 1996		Core 2 (40-44 cm)	16						
Mudroch et al. 1987		Core 1 (27-28 cm)	16						102
Mudroch et al. 1987		Core 2 (27-28 cm)	18						108
Mudroch et al. 1987		Core 3 (27-28 cm)	15						123
Andrade et al. (2010)		YKBS-03 Core (34 cm)	10			37	5		100
Mudroch et al. 1987	Back Bay, Great Slave Lake	Core 1 (25-26 cm)	20						110
Mudroch et al. 1987		Core 2 (25-26 cm)	25						98
Mudroch et al. 1987		Core 3 (25-26 cm)	23						115
Andrade et al. (2010)	Akaitcho Bay, Great Slave Lake	Core (28 cm)	10			37	5		100
Bright et al. 1996	Kam Lake	Shallow core (25-37 cm)	39						
Wagemann et al. 1978	Likely Lake	Grab (0-20 cm)	20						
Wagemann et al. 1978		Grab (0-20 cm)	27						
Nickerson, 1972; Allen et al. 1972		Grab (0-20 cm)	19						
Nickerson, 1972; Allen et al. 1972		Grab (0-20 cm)	38						
Wagemann et al. 1978	Chitty Lake	Grab (0-20 cm)	28						
	Median		*22.3*		*82*	*37*	*5*		*108*
	Min		*10*		*82*	*37*	*5*		*98*
	Max		*105*		*82*	*38.9*	*5*		*159*
	n		*25*		*1*	*3*	*3*		*9*

Table 5 cont.

Reference	Site	Sample Type	Concentration (mg/kg)[a]						
			As	Cd	Cr	Cu	Pb	Hg	Zn
IMPACTED SAMPLES[c]									
Bright et al. 1994	Meg Lake	Grab (0-5 cm)	1160		74	781	72		
Bright et al. 1994		Grab (0-5 cm)	1043		74	151	<10		
Falk et al. 1973a,b	Keg Lake	Grab (0-20 cm)	3300						
Nickerson, 1972; Allen et al. 1972		Grab (0-20 cm)	6.3						
Bright et al. 1994	Peg Lake	Grab (0-5 cm)	3090		40	232	<10		
Bright et al. 1994	Kam Lake	Grab (0-5 cm)	2186		75	1195	49		
Nickerson, 1972; Allen et al. 1972		Grab (0-20 cm)	3500						
Falk et al. 1973a,b		Grab (0-20 cm)	1670						
Falk et al. 1973a,b		Grab (0-20 cm)	640						
Falk et al. 1973a,b		Grab (0-20 cm)	1700						
Falk et al. 1973a,b		Grab (0-20 cm)	120						
Nickerson, 1972; Allen et al. 1972		Grab (0-20 cm)	40						
Nickerson, 1972; Allen et al. 1972		Grab (0-20 cm)	63						
Wagemann et al. 1978		Grab (0-20 cm)	2700						
Mudroch et al. 1987	Back Bay, Great Slave Lake	Core 1 (0-1 cm)	1010				170		
Mudroch et al. 1987		Core 2 (0-1 cm)	630				168		
Mudroch et al. 1987		Core 3 (0-1 cm)	2800				168		
Mudroch et al. 1987	Yellowknife Bay, Great Slave Lake	Core 1 (0-1 cm)	550				187		
Mudroch et al. 1987		Core 2 (0-1 cm)	490				173		
Mudroch et al. 1987		Core 3 (0-1 cm)	280				157		
Bright et al. 1994		YKB3 Grab (0-5 cm)	115		52	38.6	<10		
Bright et al. 1994		YKB4 Grab (0-5 cm)	174		40	147	19		
Andrade et al. (2010)		YKBS-03 Core (0 cm)	600			50	8		150
Andrade et al. (2010)	Akaitcho Bay, Great Slave Lake	Core (0 cm)	47			35	18		98
	median		*635*		*63*	*149*	*157*		*124*
	min		*6.3*		*40*	*35*	*8*		*98*
	max		*3500*		*75*	*1195*	*187*		*150*
	n		*24*		*6*	*8*	*11*		*2*

[a] literature selected based on inclusion of lake sediment geochemical data from lakes other than Great Slave Lake and sediment core study design. This is not meant to be a complete compilation of of existing sediment quality in the region. For that, the reader is directed to SENES Consulting Ltd. (2006).

[b] Non-impacted samples refer to sediment-water interface samples from lakes not likely to have been impacted by mining and from depth in sediment cores that were collected from known impact sites

[c] Impacted samples refer to sediment-water interface samples or upper sediments of sediment cores collected in lakes likely to have been impacted by mining

n/a – not applicable; [a] - when concentration is below the method detection limit (MDL), ½ of the MDL is used

*sample may be impacted

Arsenic concentrations in sediments from lakes throughout the study area are highly variable, ranging from 6.3 ppm to above detection (>10,000 ppm) (Table 3). Three lakes, Lake

BC-19, Lake BC-17, and Lake YK-11, contain sedimentary As concentrations that are extreme outliers (Figure 3; Table 4). The median As concentration in Yellowknife area lake sediments (107.9 ppm, min 6.3, max > 10,000 ppm, n=95) is well below the median concentration in impacted sediments (median 635, min 6.3, max 3500, n=24 ppm). However, some of the impacted lakes (e.g., Meg, Keg, Peg) are known tailings discharge sites and are therefore not representative solely of contaminant input from aerial roaster stack emission. The high skewness of As concentrations in Yellowknife area lake sediments (skewness=6.4) relative to other sample regions (Table 3) most likely indicates that some lakes in the Yellowknife area have received anomalously high As loading from industrial activity and/or input from enriched mineralized zones. Howell (2014) demonstrates the presence of As-oxide mineral phases in the sediments of BC-13 and BC-17, suggesting anthropogenic input of As into these lakes.

The concentration of Pb is highest in Baker Creek Watershed region lakes compared to regional lakes, but this element is not particularly high in known impacted sites. Other elements, such as Cd, Cr, Cu, and Zn, are higher in sediments of lakes located along the Tibbitt to Contwoyto Winter Road than in sediments of lakes in the Baker Creek Watershed region. These results suggest that elevated As and Pb relative to regional lakes characterize lakes of the Baker Creek Watershed region.

5.2 Determination of geochemical background

Geochemical data from lake sediment cores in the Yellowknife area show that As concentrations in near-surface sediments are elevated compared to concentrations at depth, which represents sediments deposited prior to mining (Table 4). These published lake sediment core data suggest that background As concentrations in the region could be as low as 23 ppm and that As concentrations above this background level in Yellowknife area lakes reflect anthropogenic impact. Three of the sediment core sites are from Great Slave Lake, which is substantially larger than Grace and Kam lakes, yet sediment core As concentrations are comparable (Table 4).

Lakes located along the Ingraham Trail overlying metasedimentary bedrock have a median As concentration of 10.6 ppm (min 1.9, max 101.6, n=27), or about half of the regional background suggested from the Great Slave, Kam, and Grace lake sediment cores of 23 ppm. Similarly, lakes along the Tibbitt to Contwoyto Winter Road traversing the central Slave Geological Province have a median sediment As concentration of 7.9 ppm (min 0.3, max 101.4, n=52). Based on these data, regional geochemical background concentration for As in lake sediments is a range, reflecting differences in bedrock geology and other landscape factors, and appears to be between 7.9 to 23 mg/kg (Tables 4,5).

In the Puznicki (1997) dataset, 83% of lakes sampled throughout the Slave Geological Province contained As concentrations below the CCME PEL of 17 ppm, consistent with our findings of sedimentary As concentrations in lakes along the Tibbitt to Contwoyto Winter Road. Lake sediment As concentrations in other mineralized mafic volcanic-dominated greenstone belts in the NWT and Nunavut, such as the Hope Bay Greenstone Belt, also contain As concentrations that are comparably low (Roberts Lake, median 7 ppm, min 4, max 15 ppm, n=15; Sherlock, 2005).

6. CONCLUSIONS

We obtained near-total element geochemistry by ICP-MS on 211 sediment-water interface samples from lakes in the Yellowknife region, lakes in the Western Interior Platform, and lakes along the Tibbitt to Contwoyto Winter Road corridor in the central Northwest Territories. These analyses were used to evaluate regional occurrences of As and other elements of potential environmental concern in lake sediments, and to estimate geochemical background and baseline against which chemical change in the environment may be evaluated. Concentrations of As exceed Canadian federal guidelines in near-surface sediments from most of the lakes sampled. Seventy one percent (n= 149) of the sediment samples contain As concentrations higher than the Interim Freshwater Sediment Quality Guideline of 5.9 ppm and 54% (n=114) of the samples contain As concentrations that exceed the Probable Effect Level of 17 ppm. Samples with the highest As concentrations were collected near the City of Yellowknife and likely reflect a combination of contamination associated with past land-use and geogenic input from mineralized bedrock and derived surficial materials.

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