



NWT Open File 2015-06
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Yellowknife area**



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Cover photo: Photo taken from the air looking east showing Baker Creek on right flowing out of Lower Martin Lake (left hand foreground). Photo courtesy of Paul Vecsei.

ABSTRACT

Ninety-eight lakes were sampled within a 30 km radius of the City of Yellowknife to document elemental concentrations in surface waters in an area exposed to 50 years of emissions from gold ore processing. Concentrations of As, Sb, and SO₄ are elevated in lakes within 17.5 km of Giant Mine relative to lakes beyond this distance. Arsenic concentrations were highest in small lakes (< 100 ha) that were downwind and proximal to the historic stacks, suggesting a gradient in impact from historic roaster operations at Giant Mine consistent with the predominant wind direction in the region. Concentrations of As exceeded the federal drinking water guideline of 10 µg/L for many of the lakes sampled within 12 km of the roaster stacks, and in some lakes were more than 60 times this limit. This study provides an extensive survey of elemental concentrations in regional lakes surrounding the City of Yellowknife and should be supported by future work to investigate drivers of variation in As concentration in surface waters, interannual variability in water chemistry, and the long-term fate of As and other elements of potential concern in these lakes.

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Appendix 2 – KMZ file of sample locations for viewing in Google Earth.

INTRODUCTION

Terrestrial and aquatic environments of the Yellowknife area received large amounts of arsenic (As) from local mining and ore roasting activities during a 50 year period between 1948 and 1999 (Hocking et al., 1978; Wagemann et al., 1978; Bromstad and Jamieson, 2012; Houben et al., 2016). During this period, the Yellowknife Greenstone Belt was the center of one of the most productive and profitable gold districts in Canadian history, and led to the operation of two long-running gold mines and the establishment of the City of Yellowknife (Moir et al., 2006). The production of gold in the region resulted in the generation of large amounts of As-bearing waste, including waste rock, tailings, and arsenic trioxide produced as a byproduct of the ore roasting process. The influence of widespread distribution of As-bearing emissions across the landscape on the water quality of local lakes is not well understood.

Subarctic lakes on the Canadian Shield typically have low dissolved solids concentrations since most terrestrial runoff travels through a shallow seasonally frozen active layer or over bedrock and there is little contribution from groundwater sources (Pienitz et al., 1997; Rühland et al., 2003; Spence et al., 2014). This results in greater influence of landscape conditions on lake water chemistry, including bedrock geochemistry, catchment surficial materials and catchment vegetation (Kling et al., 2000; McClelland et al., 2006; Kokelj et al., 2009), but also suggests that the buffering capacity of subarctic lakes is limited, which may magnify anthropogenic and climate change impacts. In this report, we present chemical analyses of surface water samples for 98 lakes within 30 km of the City of Yellowknife to examine lake water conditions in an area that has been influenced by mining and municipal activities since the late 1930's.

The Environmental Legacy of Mining and Ore Processing in the Yellowknife Area

Mining History

The growth and prosperity of Yellowknife and the Northwest Territories (NWT) has largely been driven by the mining and exploration industries over the past 75 years (Bullen and Robb, 2006). Over 13.5 million ounces of gold were produced from the Yellowknife Mining District between 1938 and 2004, predominately from the three largest gold mines in the region: Giant, Con, and Discovery (Moir et al., 2006). Giant Mine (1948-2004) was the largest of the three mines, generating more than 7 million ounces of gold in total, making it one of the most productive gold mines in Canadian history (Moir et al., 2006). Gold production in the region ended in 2004 with the closure of Giant Mine, but recently there has been renewed gold exploration interest leading to the initiation of several new exploration projects in the Yellowknife Greenstone Belt (e.g. TerraX Minerals Inc., 2014).

Ore Processing

The majority of mining in the Yellowknife region occurred within the Giant and Con gold deposits of the Yellowknife Greenstone Belt. Gold in these deposits is predominately hosted in arsenopyrite (FeAsS) in quartz-carbonate veins and also occurs as free milling gold (Canam, 2006; Siddorn et al., 2006). At most modern gold mines, gold is extracted from ore using either gravity separation or cyanide leaching, but when gold occurs in the refractory phase, additional processing steps are required prior to cyanidation (Canam, 2006; Bromstad and Jamieson, 2012;

Walker et al., 2015). The Giant deposit contains mainly refractory gold. Consequently, throughout the mining process at Giant, and in the early years of production at Con (1948-1970), ore was roasted to make it more amenable to cyanidation (Hazra et al., 1977; Canam, 2006).

The roasting of arsenopyrite ore at Giant was a pre-leaching oxidation process that created porous iron oxides from which gold was extracted using cyanide leaching (Canam, 2006; Walker et al., 2015). Roasting produced As vapour, As-bearing iron oxides and sulfur dioxide (SO₂), which were emitted to the atmosphere via a roaster stack. In the atmosphere, As vapour precipitated to form the solid phase arsenic trioxide (As₂O₃). During the first three years of operations at Giant (1948-1951), As₂O₃ was vented freely to the atmosphere (SRK, 2002; Walker et al., 2005). In 1951, the first of many emission control technologies were implemented to reduce As emissions, but also to reduce the loss of gold in the roasting process (Bromstad, 2011). Arsenic emissions were gradually reduced over time, but As₂O₃ and As-bearing iron oxides continued to be released to the surrounding environment from the roaster stacks until 1999. An estimated 237,000 tonnes of As₂O₃, representing 90% of all As₂O₃ produced, were captured by emission control technologies at Giant Mine and are stored onsite (Indian and Northern Affairs Canada, 2010). This represents one of the largest deposits of As₂O₃ in the world (Bromstad and Jamieson, 2012). It is estimated that more than 20,000 tonnes of As₂O₃ were not captured and were instead released to the surrounding area via roaster emissions at Giant Mine between 1948 and 1999 (Wrye, 2008). Annual emissions exceeded 7500 tonnes per year between 1949 and 1951 and approximately 86% of total As₂O₃ emissions were released prior to 1963 at Giant Mine (Wrye, 2008). After the installation of more efficient technologies in 1951, emissions were gradually reduced and by the 1990's less than 4 tonnes per year were being released to the surrounding environment (Wrye, 2008). Roasting of ore was less common at Con Mine, and much of the As₂O₃ generated was captured, treated onsite and integrated with the tailings or sold and shipped to the south (Hauser et al., 2006). An estimated 2500 tonnes of As₂O₃ was not captured by emission control technologies and was released to the surrounding area between 1948 and 1970 (Hocking et al., 1978).

Arsenic and human health

Arsenic is of public concern because of its toxic effects on humans and other organisms (Nriagu, 1994). The toxicity, mobility and fate of As are dependent on chemical speciation, biological processes, and, in solid materials like soils and sediments, mineralogy (Bowell et al., 2014). The bioavailability of As is higher in aqueous solution relative to solid materials, as dissolved species can be more readily absorbed via the digestive system in wildlife and humans. In solution, inorganic As species are generally more toxic than organic species, with trivalent As [As (III)] typically more toxic than the pentavalent species [As (V)] to humans (Hughes, 2002). In general, arsenic (V) dominates in oxidizing environments, such as well-oxygenated surface waters, whereas As (III) is more common under reducing conditions, such as oxygen-poor bottom waters and porewater within lake sediments (Bowell et al., 2014). Organic forms of As may also be present in surface waters, but usually occur in smaller quantities than inorganic species. The long-term intake of small doses of inorganic As has been linked to several diseases in humans including liver, bladder, kidney and skin cancers (Chatterjee et al., 1995). As a result, Health Canada adopted the World Health Organization recommendation of 10 µg/L as the maximum allowable concentration of total As for potable water in the Canadian Drinking Water Guideline (Health Canada, 2014). It is important to recognize that ambient As concentrations in

surface waters vary widely across Canada, and in many cases exceed the drinking water guideline (Wang & Mulligan, 2006).

Previous Research

The potential ecological and human health risks associated with the large volumes of As-bearing waste at Giant and Con Mines has resulted in detailed research on the nature (e.g. Walker et al., 2005; Fawcett and Jamieson, 2011; Jamieson et al., 2011; Fawcett et al., 2015; Walker et al., 2015), extent (e.g. Hocking et al., 1978; Wagemann et al., 1978; Houben et al., 2016), and behavior of As in the aquatic environment on-site and in the immediate vicinity of Giant Mine, Con Mine and the City of Yellowknife (e.g. Mudroch et al., 1989; Bright et al., 1994, 1996; Dushenko et al., 1995; Andrade et al., 2010; Galloway et al., 2012, 2015). This work has generally been confined to the mine property, or in the downstream receiving environments near Giant Mine (e.g. Mudroch et al., 1989; Andrade et al., 2010) and Con Mine (e.g. Coedy, 1994; Bright et al., 1994, 1996). A more complete list of previous research on As in lake sediments, soils and surface waters in the Yellowknife area is available in Galloway et al. (2015).

Comparatively, relatively few investigations have addressed the regional influence of roaster emissions in the area. Hocking et al. (1978), St-Onge (2007), Galloway et al. (2012, 2015), and Houben et al. (2016) present regional data on the concentrations of As in soils, vegetation, lake sediments, and surface waters, respectively. These studies demonstrate that As concentrations decrease as a function of distance from the historic roaster stacks at Giant Mine and/or Con Mine, regardless of the material sampled. Hocking et al. (1978) investigated As concentrations in surface soils at 50 sites within a 40 km radius of Yellowknife and demonstrated that concentrations of As decreased with increasing distance from the roaster stacks at Giant and Con Mines. Galloway et al. (2012, 2015) reported near-surface lake sediment geochemistry from the Yellowknife region and showed that anomalously high levels of As occur in lakes on granitoid bedrock west of the City of Yellowknife, consistent with regional wind direction and aerial deposition of As from a point source. Houben et al. (2016) provided information from 25 small lakes in the region and estimated that concentrations of total As in lake surface water are elevated above federal guidelines for the protection of aquatic life (5 µg/L) within 17 km of the historic roaster stacks. Lake waters within 4 km of the stacks contained between 27 and 136 µg/L total As, while concentrations were as low as 2 µg/L beyond 18 km from the stacks. Houben et al. (2016) use backward multiple stepwise regression to suggest that the underlying bedrock geology has limited influence on the distribution of As in surface waters of their study lakes.

Published data from the 1970's provide an opportunity to compare historical and contemporary water chemistry information. Wagemann et al. (1978) reported As concentrations in surface waters of five lakes in the region. Extremely high dissolved As concentrations (700-5500 µg/L) were measured in two lakes directly impacted by mine effluent from Con Mine (Kam and Keg lakes). Total dissolved As concentrations were also higher in one of the three "reference" lakes (Grace Lake: 50-70 µg/L) that did not directly receive effluent from the tailings waste stream at Con, compared to two reference lakes (Likely and Chitty lakes) that are farther than 15 km from Giant and Con mines (< 10 µg/L). While these data provide important historical context for As concentrations in these specific lakes, an overall understanding of regional water quality conditions is lacking.

The objectives of this study were to undertake a detailed survey of lake water chemistry to better understand regional baseline surface water quality in the vicinity of the City of Yellowknife, and to assess the extent of influence of historic mining activities 75 years after the inception of mining and 15 years after roaster operations ended. A secondary objective was to determine pre-industrial background water quality conditions of local lakes. The differentiation between *baseline* and *natural background* is important in this context, as these terms are often used interchangeably yet have distinct definitions. *Geochemical background* of surface waters varies according to changes in hydrology and basic bedrock geology, whereas *geochemical baseline* refers to a specific period in time and includes geogenic (or background) variation in addition to influences from local mineralization and anthropogenic impacts (Salminen and Gregorauskiene, 2000; Albanese et al., 2007).

The results of this study are important to land managers, regulators and industry responsible for developing remediation and water license criteria that reflect current baseline or background conditions in the region. These results will also be useful for the municipality of Yellowknife and community members in future land-use planning initiatives, where an understanding of historical land-use impacts is required.

STUDY AREA

The study area is located within the Canadian Shield Slave Structural Province and lies within the Great Slave Uplands and Great Slave Lowlands of the Taiga Shield High Boreal Ecoregion (Ecosystem Classification Group, 2009). In both ecoregions, exposed bedrock plains and undulating to hilly bedrock uplands are the dominant landscape features. The Yellowknife Greenstone Belt, where both the Giant and Con deposits are located, transects the study area north to south and consists of Archean metavolcanic rocks (Figure 1) (Cousens, 2000). The area to the west was intruded by younger granitoids and the area to the east consists of Archean metasedimentary rocks (Figure 1). Extensive faulting divides granitoid and volcanic rock units and multiple gabbro and diabase dikes crosscut the area (Cousens, 2000).

Glacial Lake McConnell covered much of the present-day study area below 205 meters above sea level (MASL) during the last deglaciation 8000 to 12,000 years ago and marks the boundary between the Great Slave Lowlands and Great Slave Uplands Ecoregions (Figure 1) (Wolfe and Morse, 2015). Lacustrine sediments from glacial Lake McConnell and ancestral Great Slave Lake fill many of the topographic depressions in the southern part of the study area within the Great Slave Lowlands Ecoregion (Wolfe et al., 2014; Wolfe et al., 2015). Glacial tills commonly infill topographic lows of the abundant bedrock outcrops above 205 MASL in the Great Slave Uplands Ecoregion (Wolfe et al., 2014). 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). Permafrost is discontinuous in the region and is restricted to peatlands and areas with thick veneers of mineral soils (Wolfe et al., 2015).

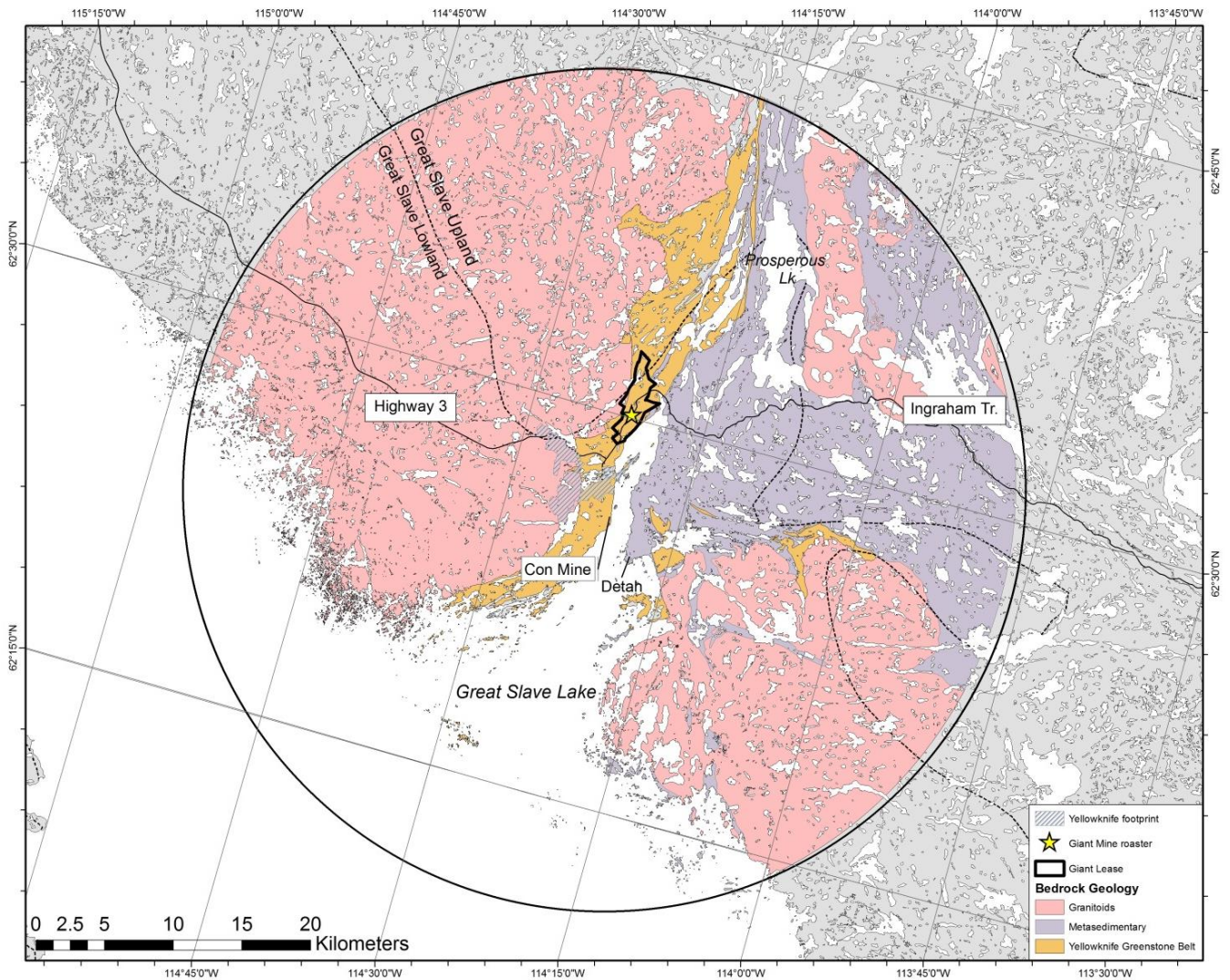


Figure 1. Map of the study area with communities, local mines, and underlying bedrock geology. The study area is delineated by the black circle and includes a 30-km radius around the city of Yellowknife. Bedrock geology is based on simplified geological unit classification presented in Wright et al. (2006).

The Yellowknife Greenstone Belt is one of the most well-studied greenstone belts in the world because of its high mineral potential and relative proximity to a major population center (Siddorn et al., 2006). In general, As concentrations in bedrock in the Yellowknife region are less than 30 mg/kg, but can be higher than 90 mg/kg in mineralized granites (Boyle, 1960; Kerr, 2006). Galloway et al. (2015) provide a thorough summary of local and global concentrations of As in bedrock. Tills overlying mineralization in the Yellowknife Greenstone Belt may contain As concentrations up to 1900 mg/kg (Kerr, 2006); however, As concentrations are generally between 5 to 30 mg/kg in regional tills (Kerr, 2006).

The distribution of roaster emissions across the landscape was likely controlled by prevailing wind directions and wind speeds. Predominant wind direction in the region is from the east in all months except June, July and August when winds are mostly from the south (Environment Canada, 2015). Mean wind energy calculated over a 6-year period (2001-2007) for Yellowknife (Pinard et al., 2008) demonstrates wind energy is predominately generated from the east and southeast (Figure 2).

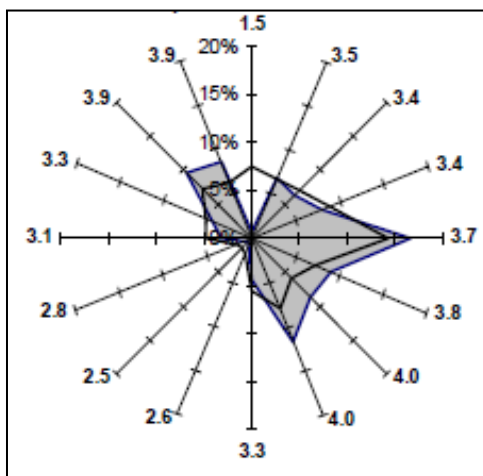


Figure 2. Wind rose depicting wind energy, frequency of direction and mean wind speed at the Yellowknife airport using data from Environment Canada (2001-2007). The shaded rose shows the relative wind energy by direction, and the outlined rose is the wind frequency of occurrence by direction. The mean wind speed by direction sector is indicated at the end of each axis. From Pinard et al. (2008).

METHODS

Study Lakes

Ninety-eight lakes were selected for sampling within a 30 km radius of Yellowknife to investigate variations in surface water chemistry in the region (Figure 3). Lakes up to 30 km away from the City of Yellowknife were targeted to ensure sampling extended beyond the probable influence of roaster emissions based on estimations of the spatial extent of impact

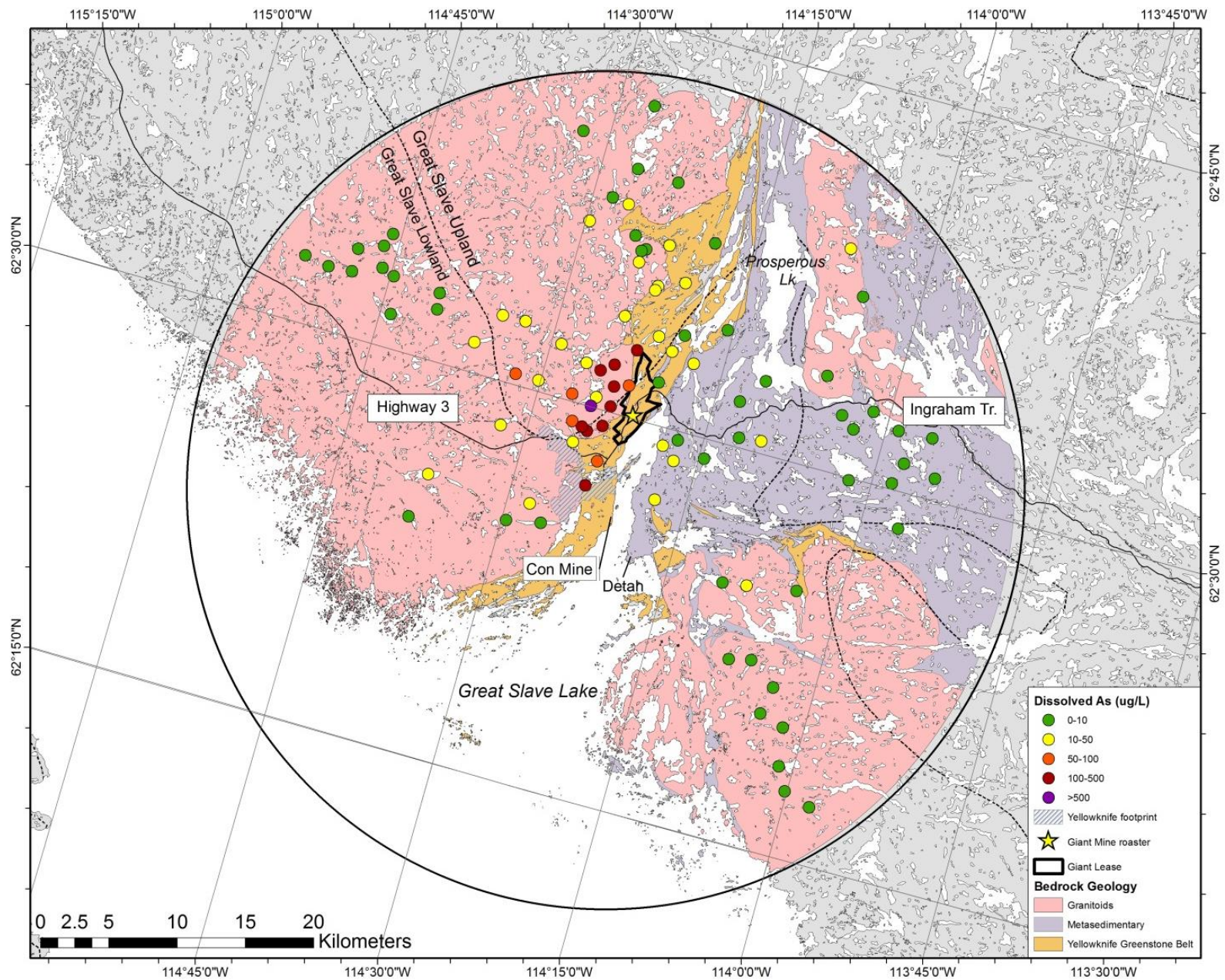


Figure 3. Concentrations of dissolved arsenic in surface water of lakes within a 30 km radius of Yellowknife.

reported by Hocking et al. (1978), Wagemann et al. (1978), and Hutchinson et al. (1982). The number and surface area of lakes within the study area were assessed using the digital 1:50 000 National Topographic Database in ArcMap (v. 10). To examine the influence of bedrock geology on lake water chemistry, representative lakes were selected from the simplified geological unit classification presented in Wright et al. (2006). Study lakes were divided into categories according to the size of the lake and its bearing from the historic roaster at Giant Mine to explore the influence of these characteristics on lake water chemistry. Lakes were considered small if the surface area was less than 100 ha. Lakes were described as downwind of the prevailing wind direction, based on data from Pinard et al. (2008), if the orientation from the roaster stack was between 270° (West) and 338° (Northwest).

Field Sampling

Surface water samples were collected for a variety of chemical and physical analyses from all lakes in either September 2012 or September 2014 from a helicopter equipped with pontoons. Sixteen of the lakes sampled in 2012 were resampled in 2014 to assess interannual differences in water chemistry. Best efforts were made to consistently sample in the center of lakes. In-situ measurements of dissolved oxygen, specific conductivity, total dissolved solids, temperature and pH were made with a YSI 6920 multiprobe sonde (Yellow Springs Inc., Yellow Springs Ohio). Water samples were collected from 30 cm below the surface in 250 mL polyethylene containers that had been rinsed three times with lake water. Following collection, water samples were stored out of direct sunlight in a cooler with ice packs and immediately delivered to a laboratory accredited by the Canadian Association for Laboratory Accreditation (CALA) for analysis. Water samples were analyzed for 15 physical and chemical variables, including: Ca, Cl, F, Mg, K, Na, SO₄, dissolved organic carbon, dissolved inorganic carbon, NO₃-N, NO₂-N, dissolved and total nitrogen, and dissolved and total phosphorous following standard methods (Clesceri et al., 1998). Water samples were also analyzed for 15 dissolved metal(loid)s by inductively coupled plasma - mass spectrometry (ICP-MS) following EPA method 200.8 (Creed et al., 1994). These samples were filtered immediately on arrival at the laboratory using a 0.45 µm filter and acidified with high purity nitric acid (HNO₃). Although we refer to these results as the “dissolved” fraction of the metal(loid)s in the sample, it should be recognized that colloids may pass through the 0.45 µm filter and may be included in the “dissolved” ICP-MS results. This is an important consideration as these colloids may play an important role in the downstream transport of metal(loid)s in watersheds.

Interannual variability in measured lake water parameters for sites with two years of data were assessed using multiple pairwise comparisons in SPSS (v. 13). No statistically significant difference existed between the two years of water chemistry data for any of the 16 lakes that were sampled in both years ($p > 0.05$); therefore, it was deemed appropriate to combine the water chemistry data from the 2012 and 2014 sampling surveys to maximize the number of samples available for analyses. All analyses, tables and figures presented here are based on the merged dataset. Where two years of data were available for a lake, the mean of the two years was calculated and used in further analyses. Both merged and unmerged datasets are presented in Appendix 1.

RESULTS

The 98 lakes sampled as part of this study represent 2% of the 4044 lakes in the entire 30 km radial study area (Table 1). Lakes of different sizes (0.7 - 3561 ha) were sampled, including several large lakes greater than 750 ha in area. The median lake size of sampled lakes was 35.7 ha, as compared to 1.07 ha for all lakes in the study area (Table 1). Proportionally, the distribution of lakes sampled in this study was skewed to larger lakes than the size distribution for the entire study area (Figure 4). Depths at the location of sampling were typically shallow (median = 1.75 m), with 70% of the sampling sites less than 5 meters deep (Table 1).

Table 1. Physical characteristics of study lakes and lakes within the entire study area.

	All lakes within study area	Lakes sampled in this study		
	Lake area (ha)	Lake area (ha)	Depth at sample location (m)	Distance to roaster (km)
Mean	8.49	122	3.19	12.9
Median	1.07	35.7	1.75	12.2
Min	0.01	0.73	0.26	1.77
Max	3561	3561	31.6	31.3
Stdev	66.51	384	4.03	7.56
N	4044	98	94	98

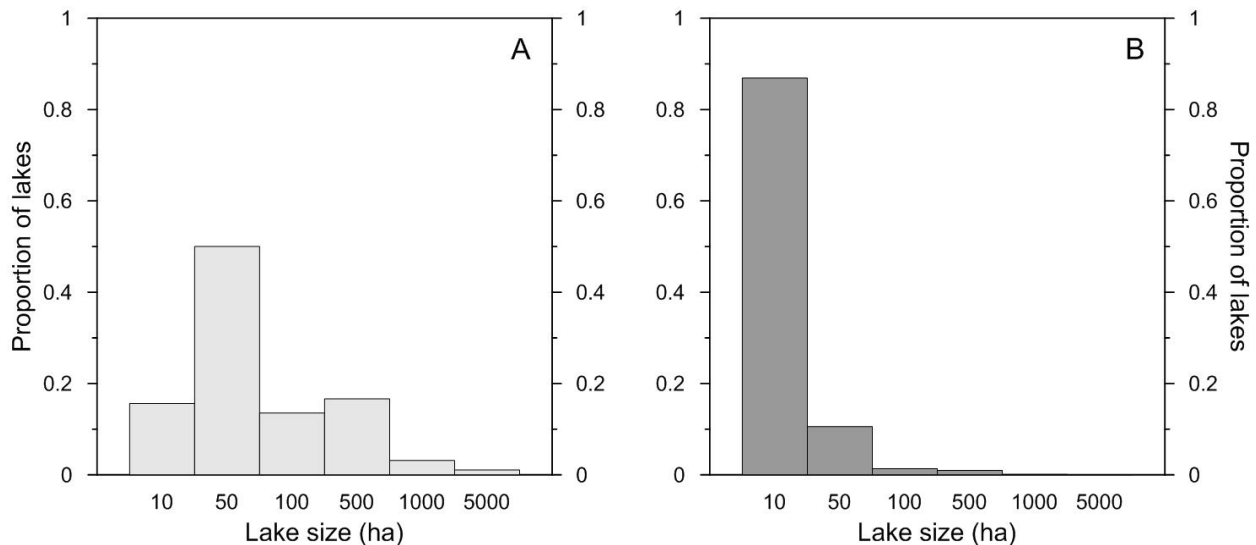


Figure 4. Histogram showing lake size distribution for: A) lakes sampled in this study (N=98); and B) all lakes within the study area (N=4044).

The trophic status of the lakes, as estimated by total phosphorous concentration (Canadian Council of Ministers of the Environment, 2004), ranged from ultraoligotrophic (TP < 4 µg/L) to hypereutrophic (TP > 100 µg/L), but approximately half the lakes sampled were either eutrophic or hypereutrophic (TP > 35 µg/L) (Appendix 1). Lakes were typically alkaline and Ca and Mg were the dominant cations (Table 2). Ionic concentrations of the sampled lakes were higher than reported in other studies north of the study region (Pienitz et al., 1997; Rühland et al., 2003). Concentrations of dissolved organic carbon were comparable to values reported for boreal lakes in the central NWT (Rühland et al., 2003), and were much higher than observed in more northerly studies from the high subarctic (Pienitz et al., 1997; Rühland and Smol, 1998) and the Mackenzie Delta area (Kokelj et al., 2009).

Arsenic concentrations varied considerably between lakes within the study area (0.5 – 646 µg/L) and exceeded the federal drinking water standard (10 µg/L) for 45% of the lakes sampled (Table 3). Arsenic concentrations in surface water declined steeply as a function of distance from the historic roaster stack at Giant and the relation was highly significant ($p < 0.01$) (Figures 5 and 6). Beyond 17.5 km from the historic roaster stacks As concentrations were typically below federal drinking water guidelines and the concentration of As no longer decreased with distance from the stack (Figure 6). The distribution of arsenic as a function of distance from the mine was most obvious for small lakes with a surface area less than 100 ha, and particularly for lakes downwind of Giant Mine (Figure 7). Arsenic concentrations were highest in small lakes proximal (<5 km) and to the west and northwest of the Giant Mine site, between the newly constructed section of Highway 4 that bypasses the Giant Mine property and Martin Lake (Figure 8).

Lake size is an important control on the concentration of As in study lakes, as concentrations were typically below 10 µg/L in lakes greater than 200 ha in size, irrespective of distance and direction to the mine (Figure 9). No pattern was observed between underlying bedrock geology and concentration of As in any of the lake waters (Figure 6).

Antimony (Sb) and sulfur dioxide (SO₂) were also associated with roaster emissions at Giant Mine (Bromstad, 2011) and, similar to As, concentrations of Sb and SO₄ (the atmospheric derivative of SO₂) in lakes decreased with distance from the mine (Figure 5). Concentrations of Sb exceeded federal drinking water guidelines in five lakes (Table 3). Spearman rank order correlation indicated a significant positive correlation between concentrations of As and Sb ($r = 0.796$, $p < 0.01$), and As and SO₄ ($r = 0.429$, $p < 0.01$; Table 4). The only other metal to exceed federal guidelines in the study lakes was iron (Fe), which exceeded the CCME guideline for the protection of aquatic life of 300 mg/kg at two sites, but was not significantly correlated ($\alpha = 0.01$) with As ($r = -0.197$, $p = 0.052$) (Table 4).

DISCUSSION

Arsenic, Sb and SO₂(g) were associated with roaster emissions in the Yellowknife region (Bromstad and Jamieson, 2012) and demonstrate a pattern of decreasing concentrations in lake waters as a function of distance from Giant Mine (Figures 5 and 6). Small lakes downwind (W-NW) and proximal (<5 km) to the roaster exhibited the highest concentrations of As (Figure 7).

Table 2. Summary statistics for selected physical and chemical properties of surface waters from the 98 study lakes.

	Major ions (mg/L)					Spec Cond (µs/cm)	pH	TDS (mg/L)	Alkalinity (mg/L)	Sulfate (mg/L)	Dissolved Organic Carbon (mg/L)	Nutrients			
	Ca	Cl	Mg	K	Na							Dissolved Nitrogen (mg/L)	Total Nitrogen (mg/L)	Dissolved Phosphorous (µg/L)	Total Phosphorous (µg/L)
Mean	21.0	6.40	10.5	3.80	8.91	163.6	7.84	142	100.8	5.74	29.0	1.68	2.09	39.2	97.3
Median	17.8	2.65	8.30	2.30	6.38	135.4	7.78	122	88.3	3.00	25.7	1.51	1.82	5.0	38.0
Min	5.70	0.35	2.00	0.40	1.40	31.3	7.12	28.0	20.6	0.50	5.00	0.26	0.27	1.0	4.00
Max	49.7	57.5	79.2	43.6	79.7	767.1	9.07	604	411	61.5	125	3.65	5.90	1780	2190
Stdev	10.1	10.0	10.1	5.50	10.5	121.5	0.37	96.84	63.2	9.38	17.8	0.90	1.25	184	299
N	98	98	98	98	98	94	62	61	53	98	97	50	53	95	53

Table 3. Dissolved metal(loid) concentrations in filtered (<0.45 µm) surface waters from the 98 study lakes. Summary statistics are only presented for metal(loid)s if concentrations exceeded the MDL for more than half of the sample sites. Guideline limits for the protection of aquatic life (CCME, 2004) and Maximum Allowable Concentration (MAC) as per Health Canada Guidelines for Canadian Drinking Water Quality (Health Canada, 2014) are indicated for reference. MDL refers to the method detection limit. Numbers in italics indicate below MDL. For data at or below MDL, 0.5 MDL was used in the generation of summary statistics. Numbers in bold indicate exceedance of one or more of the guideline values.

	Dissolved Metal(loid)s (µg/L)											
	Al	Sb	As	Ba	Cu	Fe	Pb	Li	Mn	Rb	Sr	Sulphate (mg/L)
MDL	0.6	0.1	0.2	0.1	0.2	5	0.1	0.2	0.1	0.1	0.1	1
CCME Freshwater^a	-	-	5	-	<i>equation</i>	300	<i>equation</i>	-	-	-	-	-
HC Drinking Water^b	-	6	10	1000	-	-	10	-	-	-	-	-
ALL LAKES												
Mean	11.1	1.18	40.2	23.3	0.37	25.0	0.09	7.51	1.27	4.00	83.3	5.74
Median	5.90	0.45	7.50	17.5	<i>0.10</i>	5.00	<i>0.05</i>	5.53	0.20	2.55	66.0	3.00
Min	0.80	0.10	0.50	4.40	<i>0.10</i>	<i>2.50</i>	<i>0.05</i>	1.50	<i>0.05</i>	0.80	20.3	<i>0.50</i>
Max	76.7	11.4	646	70.5	6.65	510	1.93	45.3	38.6	44.0	436	61.5
Stdev	13.1	1.86	92.6	15.8	0.76	64.4	0.20	6.84	4.29	4.89	62.2	9.38
N	98	98	98	98	98	98	98	98	98	98	98	98
LAKES DOWNWIND (270-338° from roaster)												
Mean	14.4	2.37	92.1	21.7	0.44	41.3	0.11	5.79	0.69	4.00	67.7	6.08
Median	10.6	1.15	25.4	19.2	<i>0.10</i>	3.75	<i>0.05</i>	5.45	0.30	3.20	60.9	3.25
Min	1.20	0.10	2.70	4.4	<i>0.10</i>	<i>2.50</i>	<i>0.05</i>	1.80	<i>0.05</i>	0.80	20.3	<i>0.50</i>
Max	56.3	11.4	646	53.6	6.65	510.0	1.93	13.3	2.80	10.1	176	61.5
Stdev	12.3	2.76	145	12.4	1.17	106.2	0.33	3.04	0.80	2.61	32.7	10.9
N	32	32	32	32	32	32	32	32	32	32	32	32
LAKES NOT DOWNWIND												
Mean	9.52	0.59	14.6	24.3	0.34	17.2	0.07	8.43	1.57	4.02	91.5	5.62
Median	4.20	0.40	6.20	17.1	<i>0.10</i>	6.00	<i>0.05</i>	5.70	0.20	2.40	69.6	3.00
Min	0.80	0.10	0.50	5.20	<i>0.10</i>	<i>2.50</i>	<i>0.05</i>	1.50	<i>0.05</i>	0.80	23.0	<i>0.50</i>
Max	76.7	4.20	187	70.5	1.90	133	0.58	45.3	38.6	44.0	436	57.0
Stdev	13.4	0.70	28.4	17.3	0.44	24.9	0.09	7.98	5.23	5.74	71.6	8.67
N	65	65	65	65	65	65	65	65	65	65	65	65

^aCanadian Council of Ministers of the Environment (CCME) Water Quality Guidelines for the Protection of Aquatic Life

^bMaximum allowable concentration (MAC) as per Health Canada Guidelines for Canadian Drinking Water Quality

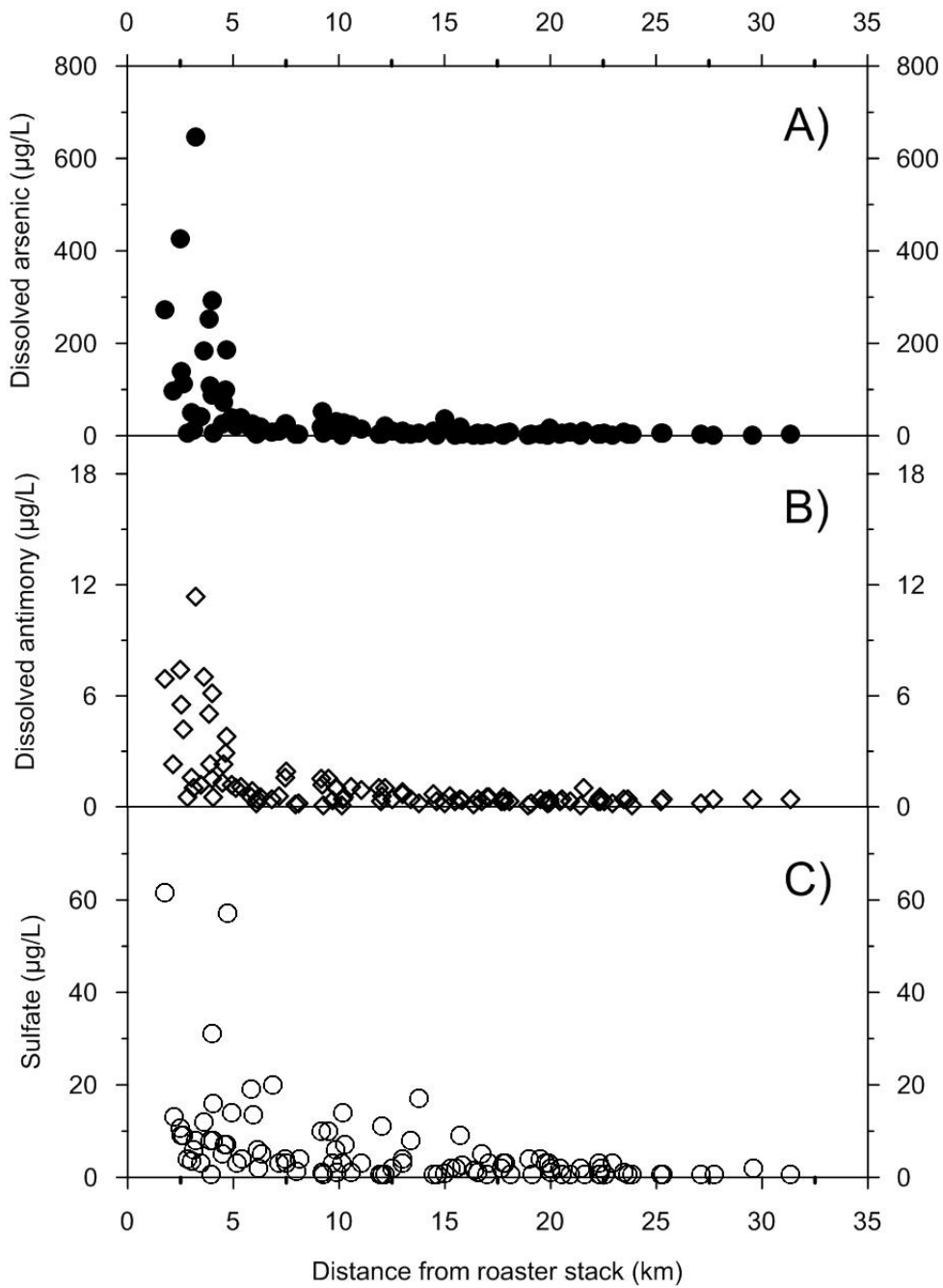


Figure 5. Element concentrations in filtered (<0.45 µm) surface waters from 98 lakes in the Yellowknife area as a function of distance from the historic roaster stack at Giant Mine, including: A) dissolved arsenic; B) dissolved antimony; and C) sulfate. Samples at or below the method detection limit (MDL) are included in the figures as ½ MDL.

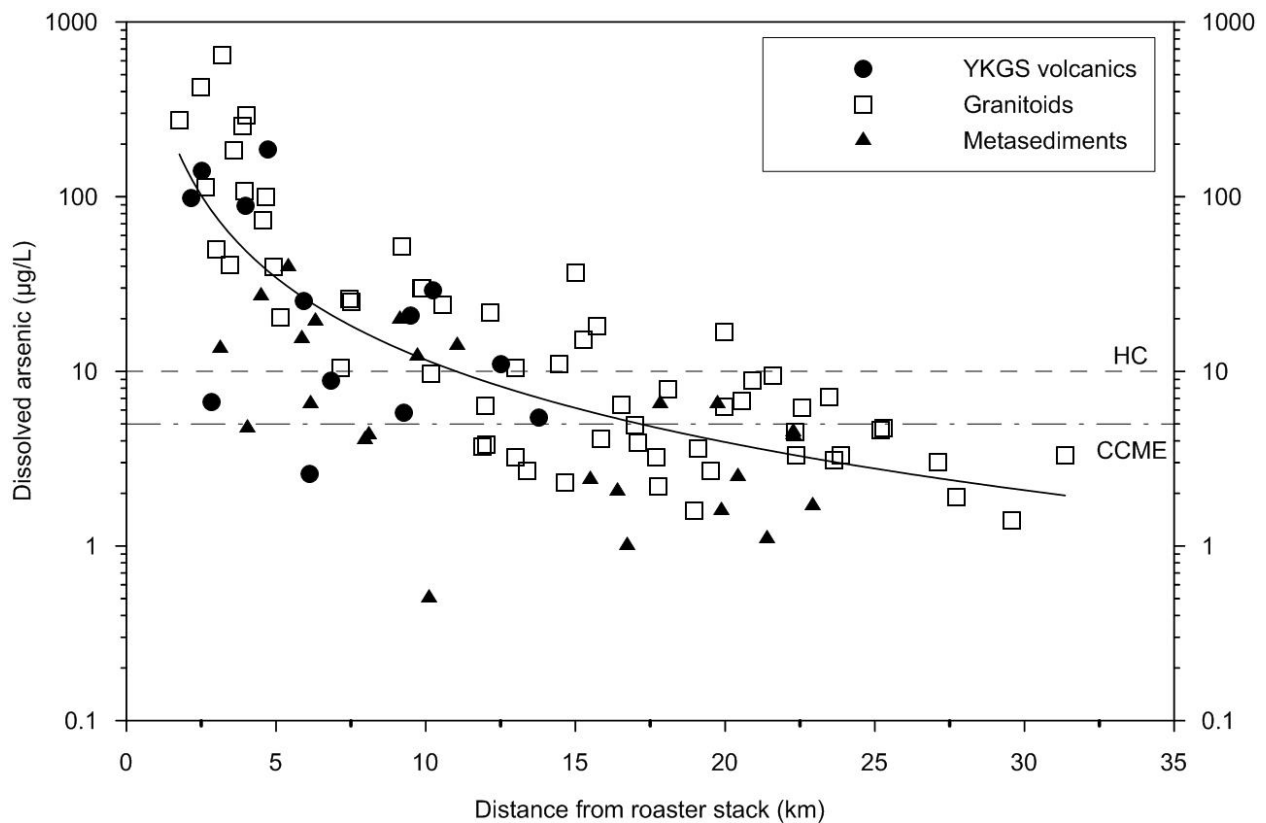


Figure 6. Concentrations of dissolved arsenic in lake waters from 98 lakes in the Yellowknife area with distance from the former roaster stack at Giant Mine. Individual lakes are symbolized according to underlying bedrock geology according to Wright et al. (2006). The dashed line at 10 µg/L represents the Health Canada Drinking Water Guideline for arsenic (Health Canada, 2014). The dashed and dotted line at 5 µg/L represents the CCME Guideline for the Protection of Aquatic Life for arsenic (CCME, 2004). The relation between dissolved As and distance from the roaster stack is shown by a solid line on the plot and is represented by the following equation: $y = 437.28x^{-1.57}$; $r^2 = 0.58$; $N = 98$; $p < 0.01$; $F_{1,97} = 128.87$.

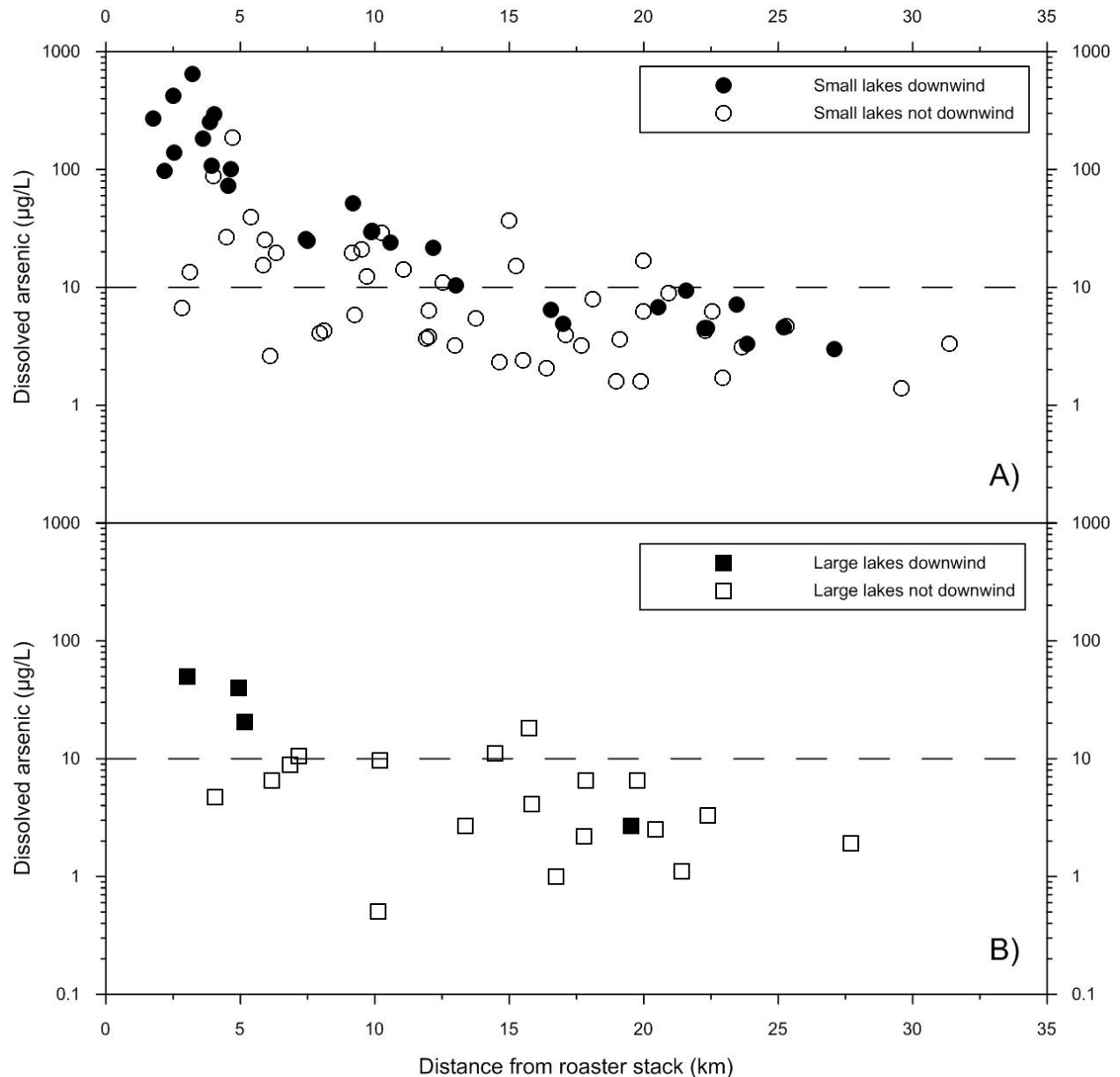


Figure 7. Concentrations of dissolved arsenic in lake waters from 98 lakes in the Yellowknife area with distance from the former Giant Mine roaster stack for: A) small lakes (<100 ha); and B) large lakes (>100 ha). Individual lakes are symbolized according to lake size and direction from the roaster. Lakes are considered downwind of the roaster if the bearing from the roaster to the sample lake is between 270 and 337 degrees. The dashed line at 10 µg/L represents the Health Canada Drinking Water Guideline for arsenic (Health Canada, 2014). Note: logarithmic scale on y-axis.

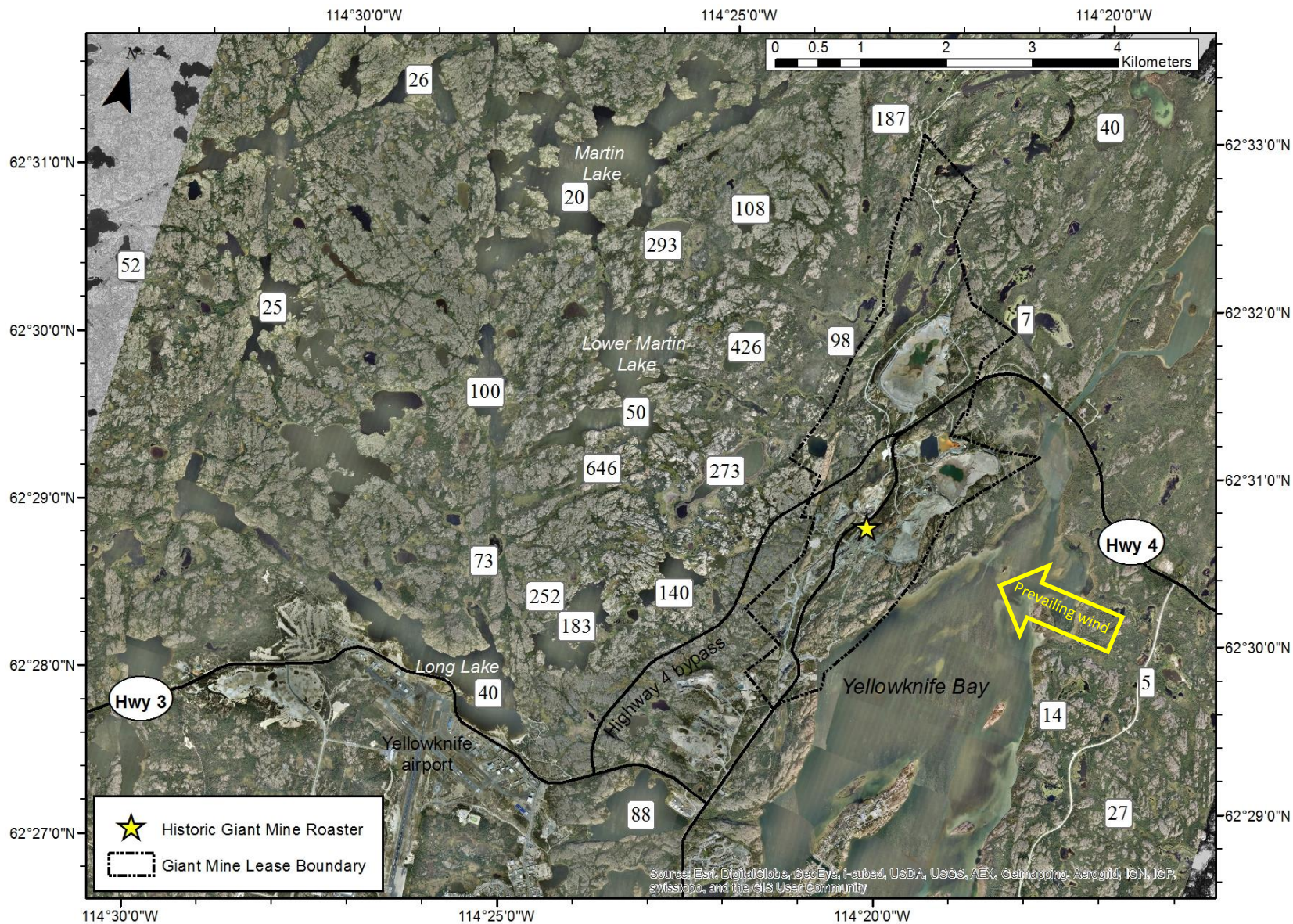


Figure 8. Concentrations of arsenic in lakes immediately downwind of the historic roaster stack at Giant Mine. The bordered numbers positioned over the lakes represent the concentration of arsenic ($\mu\text{g/L}$) in filtered ($<0.45 \mu\text{m}$) lake water samples measured during the two field surveys in September 2012 and 2014.

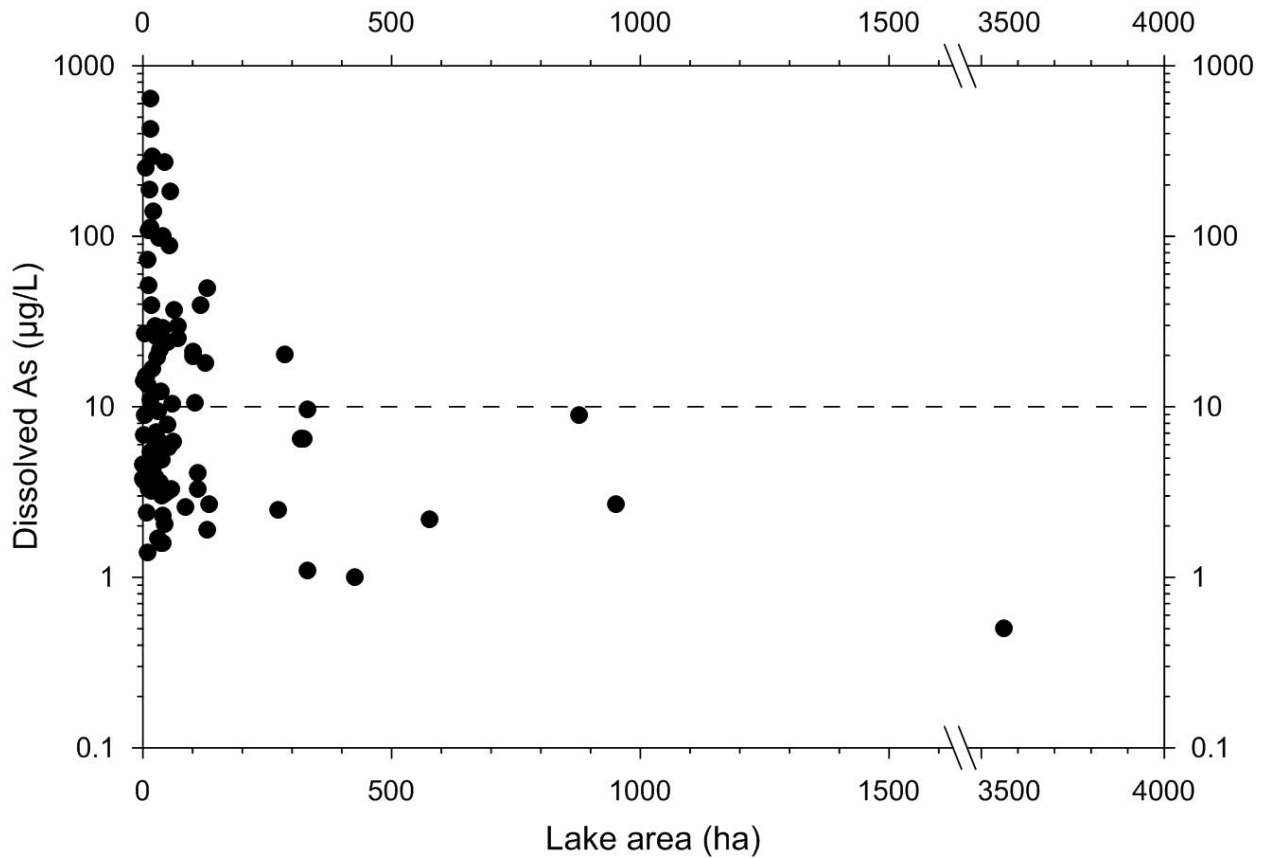


Figure 9. Concentrations of arsenic in filtered (<0.45 µm) lake waters from the 98 sampled lakes as a function of lake area. The dashed line at 10 µg/L represents the Health Canada Drinking Water Guideline for arsenic (Health Canada, 2014).

Table 4. Correlation matrix representing the results for Spearman rank order correlations between dissolved concentrations of arsenic, antimony, iron and sulfate in the 98 study lakes. The coefficient of correlation (*r*) is presented in the table as well as the *p* value associated with the correlation test. Significant correlations at *p*<0.01 are indicated in bold.

	As	Sb	Fe	SO ₄
As	X	0.796 (<0.01)	-0.197 (0.052)	0.429 (<0.01)
Sb	X	X	-0.201 (0.047)	0.435 (<0.01)
Fe	X	X	X	-0.246 (0.15)
SO ₄	X	X	X	X

This finding supports the work of Houben et al. (2016) that suggests stack emissions from the Giant and Con mines influenced lakes within 17 km of Yellowknife. The steep decline in As concentrations with increasing distance from the historic roaster stacks observed in this study and in several other studies, including those investigating soils, suggests that fallout from the roasters occurred predominately as particulate and not as gaseous dispersion (Hocking et al., 1978; Houben et al., 2016).

Our research explores additional physical and geographic influences on local lake water quality and shows that while regionally there is a general pattern of decreasing concentrations of As in lake water as a function of distance from Giant Mine (Figure 6), there is substantial variation in As concentration at the local scale (Figure 8), likely dependent on factors such as lake size and direction from the roaster stack point source. Several small lakes adjacent to the new bypass road around Giant Mine (Figure 8) had concentrations of As more than 10 times the federal drinking water guideline of 10 µg/L, including one lake (BC-17), where the concentration of As was more than 60 times (646 µg/L) the federal drinking water guideline (Figure 8). While the area to the west-northwest of the mine may be considered a “hotspot” relative to the rest of the region there are substantial differences in As concentrations even among the lakes in this “hotspot” area. For example, the concentration of As measured in Lower Martin Lake, less than 500 m north of BC-17, was 50 µg/L. The factors influencing metalloid concentrations in these lake waters are not well understood, but likely include lake size, hydrologic position of the lake on the landscape, catchment characteristics, lake water residence time, and other within-lake biogeochemical processes. Further research is required to determine the specific hydrological, landscape and within-lake biogeochemical processes that may affect elemental concentrations in lake waters within this “hotspot”.

Concentrations of As in surface waters of lakes proximal to historic mining activity in the Yellowknife area remain elevated 15 years after ore processing operations ended at Giant Mine and 60 years since peak stack emissions. Unfortunately, our understanding of the limnological recovery of lakes in the region is limited by the paucity of historical long-term water quality data in the area. Improvements in lake water quality have been reported in other areas of Canada after stack emissions were reduced, including the Sudbury area where lakes have exhibited substantial ecological and limnological recovery from the effects of metal loading and acidification (e.g. Keller et al., 1992, 2004). Considering the lack of long-term water quality information for lakes in the region, we suggest new research in the area should include paleolimnological and paleoecotoxicological approaches to better understand long-term trajectories of change in lakes impacted by legacy mining activities.

The presence of elevated sulfate in lakes close to Giant Mine may have indirect biogeochemical consequences in these lakes, including the enhanced methylation of mercury (Hg) (Houben et al., 2016), as Hg methylation is enhanced in the presence of sulfate reducing bacteria in aquatic systems (Jeremiason et al., 2006). Methylation rates of up to 44% were estimated by Houben et al. (2016) and our data highlights an area to the west-northwest of the mine where Hg pathways in the aquatic food web should be further explored.

This study provides the most extensive survey of water quality conditions in lakes of the Yellowknife area to date. These data will be useful to land managers responsible for determining

baseline geochemical conditions of local lakes and to inform the establishment of remediation criteria that reflect natural background conditions. Future work should be directed at investigating the specific drivers of geochemical variation in impacted lakes, including a better understanding of geochemical cycling in lakes, the interannual variability of water quality in impacted lakes, and the long-term fate of As and other elements of concern in these lakes.

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This report is part of a two part series on the chemistry of lake sediments and surface waters in the NWT. Part I is presented in Galloway et al. (2015) and describes the geochemistry of near-surface lake sediments across three ecozones in the NWT, including the 98 lakes that are discussed in this report.

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