NWT Open File 2017-03

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NORTHWEST TERRITORIES
GEOLOGICAL SURVEY

Government of
Northwest Territories

Cover Image:
The general surrounding landscape of the Yellowknife area.

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Abstract

The objective of this NWT Open File is to report the concentration of arsenic and other elements in 479 near-surface soil samples collected within 30 km of Yellowknife in 2015, 2016, and 2017. For this research, the soil samples were cored from locations that were undisturbed by buildings, roads, mining or other visible human activities to minimize the influence of recent post-mining activities and examine the effect of natural processes and the legacy of airborne emissions from former ore roasting. Sampling targeted four distinct terrain units: outcrop soils, forest canopy soils, forest canopy outcrop soils, and peatland soils. Most of the analyses have been done on the Public Health Layer, which is defined as the top 5 cm of material. The arsenic concentration in outcrop soils ranged from 3.5 mg/kg to 3000 mg/kg arsenic with a median of 165 mg/kg. The arsenic concentration in the forest canopy soils ranged from 1.0 mg/kg to 1300 mg/kg with a median value of 38 mg/kg. The arsenic concentration in the forest canopy outcrop soils ranged from 2.1 mg/kg to 4700 mg/kg with a median of 150 mg/kg. Finally, the arsenic concentration in peat soils ranged from 2.9 mg/kg to 3400 mg/kg of arsenic with a median of 95.5 mg/kg. Forty soil samples collected from below the Public Health Layer (approximately 10 cm to 40 cm below the surface) were analysed and are lower in total concentration of arsenic than the corresponding samples from the Public Health Layer, with the exception of two samples. Arsenic concentrations are highly variable at the local and regional scale, and even vary between field duplicates, likely due to the uneven distribution of arsenic-rich minerals in the soil samples, which were unsieved and unground. Statistical analysis indicate that the distance and direction from the former ore roasters, soil depth, elevation and terrain type influence total arsenic concentration.

Introduction

The Yellowknife Greenstone Belt was one of the most productive and profitable gold districts in Canadian history, with the Giant and Con mines producing approximately 13 million ounces of gold (Bullen and Robb 2006; Moir *et al*. 2006). The production of this gold resulted in the deposition of arsenic-bearing materials on the mine properties and beyond through tailings spills, the use of waste rock for construction, and aerial deposits from the roaster stack emissions.

The gold-bearing arsenopyrite (FeAsS) ore was roasted as a pretreatment for cyanidation, creating sulfur dioxide and arsenic vapour, some of which was released into the surrounding environment through roaster stack emissions and condensed to arsenic trioxide (As_2O_3) dust (Walker *et al*. 2005 and references therein; Jamieson 2014). It is estimated that more than 20 000 tonnes of arsenic trioxide dust were released into the surrounding area via roaster emissions at Giant Mine between 1948 and 1999 (Wrye 2008). Annual emissions exceeded 7500 tonnes of arsenic trioxide dust per year between 1949 and 1951 and approximately 86 % of total arsenic trioxide emissions were released prior to 1963 at Giant Mine (Wrye 2008). After the installation of more efficient technologies in 1951, arsenic trioxide emissions were gradually reduced and by the 1990s less than 4 tonnes per year were being released into the surrounding environment until roasting ceased in 1999 (Wrye 2008). Arsenical gold ores were also roasted at the nearby Con Mine (1938—2003), but this ceased in 1970 as ores extracted from deeper in the mine became more free-milling and less refractory (Walker *et al.* 2015). Pressure oxidation, which does not produce stack emissions, was used instead of roasting to process the refractory ore in the early 1990s (Wright 1992). From 1948—1970, approximately 2500 tonnes of arsenic trioxide were released to the atmosphere as stack emissions from the Con Mine roaster (Hocking *et al*. 1978). Despite high emissions in the early years of the two mines, most of the arsenic trioxide dust produced during the operation of both mines was captured. At Giant, 90 % of the dust is currently stored in underground chambers, whereas at Con, the arsenic trioxide was treated on site, integrated with tailings, or sold (Hauser *et al*. 2006).

This report focuses on the concentration of arsenic in soils beyond the mine properties and within 30 km of the City of Yellowknife. Arsenic in lake waters and lake sediments in the Yellowknife region has been examined by (Palmer *et al*. 2015) and (Galloway *et al*. 2015) respectively. The overall objective of this project is to determine, where present, the legacy contamination from aerial roaster stack emissions and the natural arsenic concentrations occurring as a result of bedrock weathering and glacial transport. Areas where arsenic may have been introduced to (or removed from) the soil via other activities such as transport of waste rock and application of fill materials were avoided, as were areas with obvious high public use. Based on evidence that arsenic concentration in soil affected by stack emissions declines rapidly with depth (Hocking *et al.* 1978; Bromstad *et al.* 2017), and since there is a concern regarding exposure of humans to these soils, most samples were collected by coring the Public Health Layer (0 cm to 5 cm; Rencz *et al.* 2011), with only larger pebbles and fragments of plant material removed. The effect of terrain type, including the presence of a forest canopy, distance and direction from the former ore roasters, and elevation have been examined. The distinction of anthropogenic (roaster-generated) arsenic-hosting solid phases from natural arsenic-hosting soil materials can be determined using advanced mineralogical methods, and has been reported for soils on the Giant Mine property (Bromstad *et al*. 2015, 2017). These methods will be applied to a subset of the soils described in this report and the results released in a future publication. This research is part of two Master of Science (M.Sc.) projects by Kirsten Maitland and Jonathan Oliver at Queen's University, supervised by Dr. Heather Jamieson. When completed, the M.Sc. theses will be published online through the Queen's University library.

Previous Research on Soils in the Yellowknife Region

Hocking *et al.* (1978) collected soil samples from 52 sites within 40 km of Giant and Con mines as part of a study instigated by the federal government in response to public concern, and demonstrated that arsenic concentrations were very high near the roasters (hundreds to thousands of mg/kg arsenic with one sample at more than 20 000 mg/kg arsenic) and decreased rapidly with soil depth and distance from the two mines. The authors indicated that the distribution pattern was consistent with attribution to the two gold roasters. Hutchinson *et al*. (1982) reported similar results within 15 km, and particularly high arsenic concentrations in the top 2 cm of soil near the roasters. Kerr (2006) documents a comprehensive study of till (from 10 cm to 70 cm depth), humus and leaf litter in the Yellowknife area, and discusses the influence of soil depth, bedrock type, mineralization and the legacy of roaster emissions. The arsenic concentrations for the clay and silt fraction of the buried tills sampled by Kerr (2006) are not directly comparable with the shallower soils collected by the other studies reviewed here, and nor are they directly comparable with this report. However, Kerr (2006) reports several soil profiles and compares arsenic concentrations in the tills with overlying humus and leaf litter, concluding that those profiles taken near Giant and Con mines show evidence of airborne contamination. St. Onge (2007) collected soil samples (0 cm to 30 cm depth) from 12 sites within 40 km of the former mines and found a decline in arsenic concentration with distance.

Bromstad *et al*. (2017) summarizes the results of two M.Sc. theses completed at Queen's University (Wrye 2008; Bromstad 2011) on the speciation (mineral form) of arsenic in soils on the Giant Mine property. The soil samples were taken in undisturbed areas away from mine infrastructure and activities. Given that the Giant Mine property is approximately 5 km long in a north-south direction and 1 km to 2 km wide, these results provide useful data on the proximal distribution of arsenic in soils in the region. The results of Bromstad *et al*. (2017) indicate that near-surface undisturbed soils on this mine property contain up to 7700 mg/kg arsenic, and the highest concentrations are in pockets of soil on bedrock outcrops. Examination of soil samples using scanning electron microscopy (SEM) and synchrotron-based microanalysis showed that the most common arsenic hosts in the soils are arsenic trioxide and roaster-generated iron oxide (maghemite and hematite). Bromstad *et al.* (2015) presents arsenic concentration analysis results of 360 soil samples collected from the Giant Mine property, which confirms earlier results based on 40 samples taken for the M.Sc. theses in 2007 and 2010 and summarized in Bromstad *et al.* (2017). Coring soil samples instead of using grab samples proved extremely useful to determine the variation of arsenic concentration with depth. Results indicate that the top soil horizons (usually 0 cm to 5 cm depth) at a given sample site typically had the most arsenic for that site compared to samples collected from depths up to 100 cm (highest value 17 000 mg/kg As – see Figure 1). These results have important implications for interpreting the results of previous soil sampling studies, of the area where soil samples were, in some cases, taken over larger depth intervals (the top 20 cm to 30 cm) and then combined. SEM with automated mineralogy software was used to find and determine the relative proportions of arsenic hosts in 50 samples (Bromstad *et al*. 2015). Arsenic trioxide was present at every sample site and is the dominant host in many of the samples. In the two studies (Bromstad *et al*. 2015, 2017), there appears to be a relation between the dominant wind direction and the highest arsenic soil concentrations, but this is difficult to quantify in the relatively small and elongated area of the mine property. Van Den Berghe *et al.* (2016) collected several near-surface soil samples on outcrops near lakes within 3 km of the western boundary of the Giant Mine property and identified the presence of arsenic trioxide in those samples.

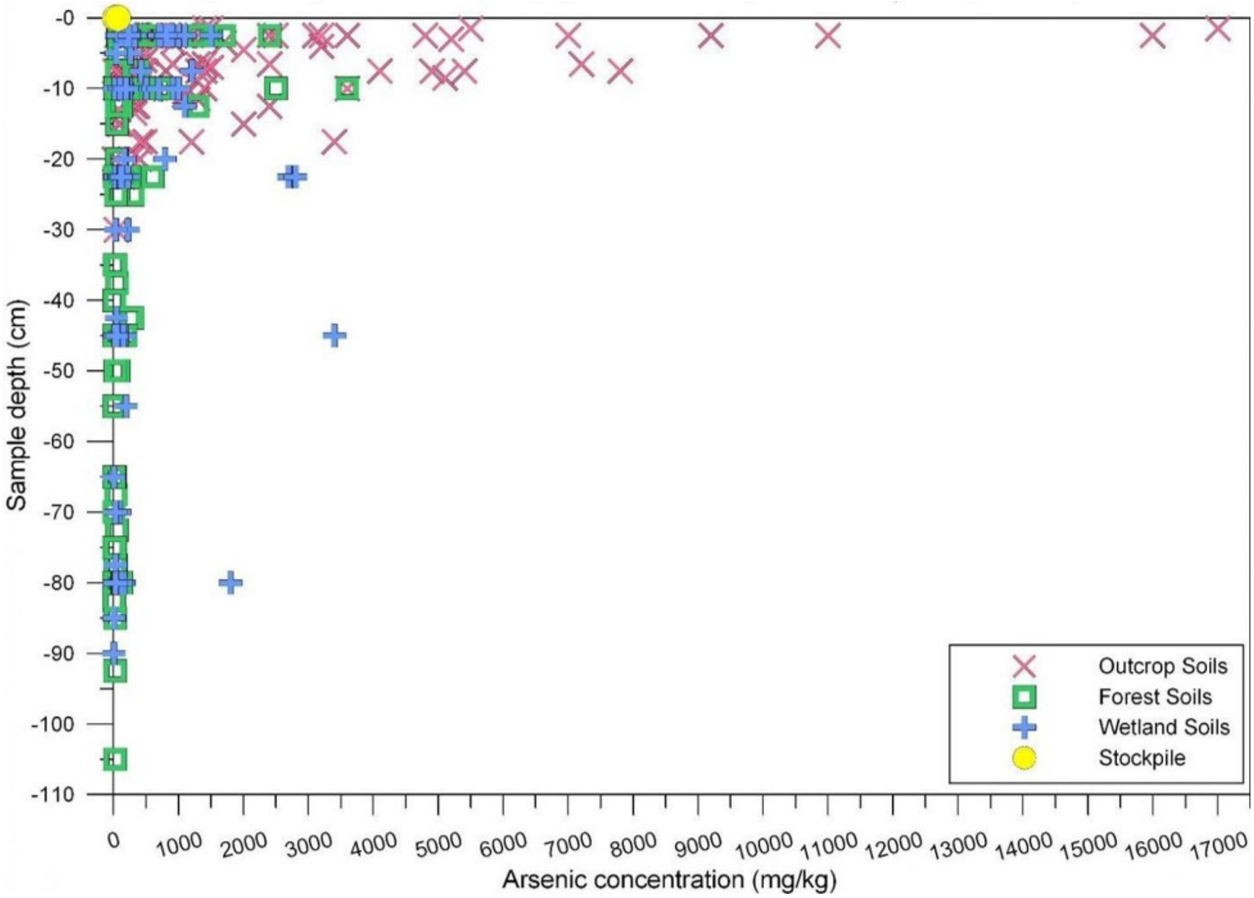

Figure 1. The variation of arsenic concentration in soil sample types (outcrop, forest, wetland, and stockpile) at sample mid-point depths on the Giant Mine property (from Bromstad *et al*. 2015).

Geology and Physiography of the Study Area

The study area, similar to that described by Palmer *et al.* (2015) and Galloway *et al*. (2015), is located along the southern edge of the Canadian Shield Slave Structural Province in the Northwest Territories. The Slave Structural Province hosts a number of gold deposits, with the largest deposits, including both Giant and Con, residing within the Yellowknife greenstone belt (YGB). The greenstone belt is a linear, north-south-trending formation located between the Western Plutonic Complex of the Defeat Plutonic Suite to the west and the Duncan Lake Group metaturbidite basin that conformably overlies the Yellowstone greenstone belt to the east (Siddorn *et al.* 2006). A map illustrating the occurrence of these simplified rock units throughout the study region can be found in Figure 2. The YGB consists of a northeast striking, southeast dipping and facing homocline, of mafic volcanic and intrusive rocks belonging to the Kam Group (2.72–2.70 Ga), intermediate metavolcanic and intrusive rocks of the Banting Group (2.66 Ga), as well as conglomerates and sandstones of the Jackson Lake Formation (2.60 Ga) tectonically emplaced between the two former groups. Archean deformation zones consisting of hydrothermally altered and deformed sericite- or chlorite-rich rocks with or without schistosity, crosscut the Kam Group, and host the Giant and Con gold deposits (Siddorn *et al.* 2006).

The study area lies in the Great Slave Uplands and Great Slave Lowlands of the Taiga Shield High Boreal Ecoregion (Ecosystem Classification Group 2009), which is characterized by elevated bedrock terrain interspersed with small areas of forest and peat. During the last period of glaciation (8000 to 12 000 years ago), Glacial Lake McConnell covered most of the study region and deposited silts and clays, which now infill many of the topographic depressions throughout the Great Slave Lowlands (Wolfe and Morse 2015).

Prevailing wind directions and wind speeds likely influence the distribution of roaster emissions throughout the study area (Bromstad *et al*. 2017). Wind measurements taken at the Yellowknife Airport indicate the dominant wind direction is from the east (34 %), while smaller wind components exist from the south-southeast (25 %), and from the northwest (23 %) (Pinard *et al.* 2008). Figure 2 shows the calculated mean wind directions at the Yellowknife Airport between 1953 and 1999, indicating wind energy is predominately generated from the east and southeast. East winds are dominate for most of the year, but not the summer. In June, July, and August, winds regularly originate from the South (Environment Canada 2017). Over a ten-year period, the mean annual wind speed measured at the Yellowknife Airport weather station was 3.28 m/s (Pinard *et al.* 2008).

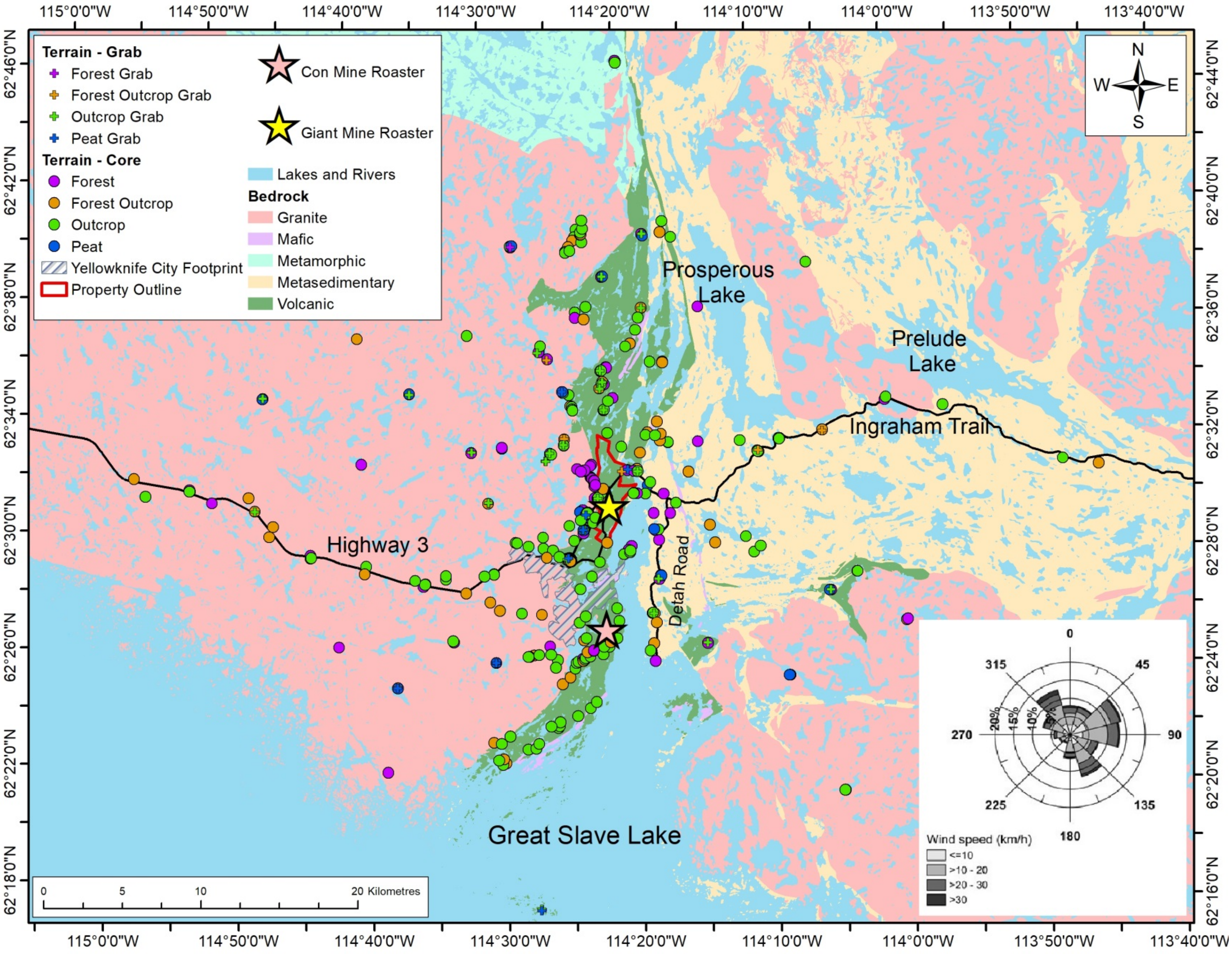

Figure 2. Soil sampling locations in the Yellowknife area. The inset shows the frequency of wind direction and corresponding wind speed at the Yellowknife airport (1953–1999). Data from Environment Canada (2017).

Soil Sampling and Analytical Methods

Sampling Protocol

During the summers of 2015, 2016, and 2017, 479 soil samples were collected from within a 30-km radius of the City of Yellowknife (sample locations are shown in Figure 2; coordinates provided in Appendix A—Table A1). The objective of the regional soil sampling survey was to collect a large quantity of samples from undisturbed sites throughout the study region from a variety of terrain units. Target areas for sampling were selected based on: (1) the distance from the former Giant and Con Mine roasters, (2) the direction from the roasters with respect to prevailing wind direction, and (3) the location of past or ongoing research.

Within each target area, soils were sampled from the four terrain units to observe how terrain impacts arsenic variation on a local scale. The four primary units sampled were: forested canopy outcrop soils, outcrop soils, forested canopy soils, and peatlands. It was recognized that accumulation of arsenic may differ within soils of different terrain types through differences in canopy interception, plant uptake, erosional transport, and soil properties (e.g., Bromstad *et al*. 2017). Forested canopy outcrop soils were collected from soil pockets located on exposed rock outcrops with significant tree canopy cover. These samples were often located at higher elevations and had numerous trees growing within the soil pockets. Outcrop soils were defined as soil pockets with little to no tree canopy cover. These sample locations were also often located at higher elevations, and contained vegetation in the form of small willow trees, juniper bushes, or long grass. Forested canopy soils were obtained from areas with dense tree canopy cover, often in low-lying areas. Peat samples were collected from peatlands and collapsed peatland areas also referred to as fens.

Bypass Road target area (BP, n=155)

Samples in this target area were retrieved from sites near the new Giant Mine Bypass Road, which was built in 2014 to re-route the Ingraham Trail. Areas with surface rock fragments that appeared to be sourced from blasting during construction of the road were avoided. These samples are close to the Giant Mine Property, often within less than 1 km to 2 km from the former roaster.

Detah Road target area (n=18)

Samples were collected along the Detah road. One sample taken in this target area was retrieved directly within the Detah community, amongst the willow trees next to the community gardens.

Ndilǫ target area (n=5)

Soil samples were retrieved throughout Ndilǫ with the help of community members. Areas that were likely to be the least disturbed by human activities were chosen.

TerraX Properties – Northbelt (n=42) and Southbelt (n=20) target areas

TerraX Minerals Inc. has led recent renewed interest in gold exploration within the YGB. Exploration efforts are occurring in the TerraX Northbelt and Southbelt properties (TerraX, 2017). The TerraX Northbelt is located directly north of the Giant Mine Property, and samples were collected at this site around Landing and Vital lakes, as well as at Berry Hill. The Southbelt property is located south of Yellowknife in the YGB along the shore of Great Slave Lake.

Ingraham Trail target area (n=22)

The Ingraham Trail, a public highway east of Yellowknife, was used to access sampling sites not located downwind from the predominant wind direction of the roasters, up to 30 km from the mining and processing operations.

Highway 3 target area (n=28)

Highway 3, west of Yellowknife, was used to access sampling sites downwind from the roasters and away from the mine properties.

City of Yellowknife target area (n=9)

Samples collected within the City of Yellowknife in areas not belonging to a specific target area are in this group. Samples were taken in undisturbed areas and, as much as possible, away from human activity.

Long Lake target area (n=8)

Long Lake is a recreational area for local community members and tourists visiting Fred Henne Territorial Park. The area is also subject to other current research projects focused on the fate, transport, and mobility of arsenic in lake sediments (Schuh *et al.* 2017). Samples were retrieved within 15 m to 175 m of the lake to assist in risk assessment studies, in addition to drawing connections between terrestrial and aquatic systems.

Martin Lake and nearby previous research sites target area (n=22)

Previous research completed by Van Den Berghe (2016) focused on the mobility and speciation of arsenic in lake sediments in three research lakes located to the west of the Bypass Road: Lower Martin Lake, BC-20 and Handle Lake. In an attempt to link previous lake sediment research to ongoing terrestrial investigations, soil samples were retrieved near these three lakes. Samples collected near Gar Lake are also included in this group.

Kam Lake target area (n=34)

Kam Lake is located south of Yellowknife, close to the former Con Mine, which also operated a roaster during the early years of mining. This is currently an area of residential, industrial and recreational use.

Hay Lake target area (n=6)

Hay Lake is accessed from the Ingraham Trail, approximately 2 km from the Detah Road. Hay Lake and the surrounding area are used for subsistence and recreational purposes by local community members.

Yellowknife River target area (n=12)

Soil sampling was completed along the Yellowknife River. The river is a popular recreational area and has been used by the local Dene people as a means of travel to remote fishing and trapping locations for centuries. The river extends to the north and east of the mine properties.

North of Giant Mine Property (n=32)

These samples were obtained using a helicopter to access areas around Duckfish Lake, Homer Lake, Chan Lake and Icing Lake. Some samples that were obtained north of the mine properties, but did not fall within the TerraX Northbelt were also included in this target area.

Distal East and Southeast Sites (n=11)

Helicopter access was required to sample distal locations east and southeast of the mine properties near Duck Lake and Mason Lake.

Distal Southwest and West Sites (n=11)

Samples sites greater than 15 km from Yellowknife in the westerly and southwesterly directions. Sites were accessed by helicopter.

Mirage Islands (n=3)

Three samples (one outcrop, one forest and one peat) were collected on the Mirage Islands, located at the mouth of Yellowknife Bay, south of the Giant and Con mine properties.

Sample Collection Methods

Soil cores, ranging from 4.9 cm to 40 cm in length, were collected so that they could be examined and subsampled at various depths. During the summer of 2015, multiple soil samples of similar terrain units were collected along the Bypass Road and from one outcrop soil pocket on Vital Lake, to test for variability at the local scale. Two grids of detailed sampling were also completed in the summer of 2016; one grid near Fred Henne Provincial Park and the second grid collected along the Giant Mine Bypass Road. Soil cores were retrieved with aluminum tubing (outside diameter of 5.08 cm) that was driven into the soil surface using a drive-head and a sledgehammer. To account for soil compression resulting from this method of sampling, the distance from the top of the core tube to the soil surface, and the top of the core to the sample surface was measured and corrections made accordingly. Peat cores were obtained by using a shovel to remove a section of material approximately 3000 cm^3 to 5000 cm^3 in size. A sharp knife or saw was then used to cut into the peat and extract a smaller core sample, which covered a surface area of approximately 10 cm^2, and extended the total depth of the previously removed material. In areas where core samples were not feasible due to a thin soil cover, and depth control was difficult to achieve due to local conditions, grab samples were retrieved at varying depths using a trowel. Outcrop, forest, and forest outcrop grab samples ranged between 3 cm and 14.5 cm in depth. Peat grab samples were taken down core in some areas where natural transitions within the peat occurred; these samples ranged between 10 cm and 40 cm in depth. All sampling equipment was cleaned between sample sites. Cores and grab samples were frozen for transport back to Queen's University and kept frozen prior to lab preparation.

Soil samples from 2016 and 2017 were collected with a lead weight to drive the aluminum tubes into the ground instead of a drive-head. This resulted in elevated lead and antimony values compared to the 2015 samples, indicating contamination from the weight, thus the lead and antimony values are removed from the table of results (Appendix A—Table A1).

Sample Preparation

Samples were prepared and sub-sampled for analysis at Queen's University. The aluminum core tubes were placed in a refrigerator kept at 4 °C to defrost, then cut lengthwise using a table saw and carefully separated using a ceramic blade. The tube cutting process resulted in minor contamination of the soil cores by aluminum fragments, and the analytical results for aluminum have been removed from the dataset. Chemical analysis of aluminum tube fragments indicated trace quantities of other elements (Cu, Mg, Fe, Zn, Mn, Ti, V, Ni, and Cr) and thus the results for these have also been removed from Appendix A—Table A1. (Soil compression was calculated prior to sub-sectioning the Public Health Layer (top 5 cm) (Health Canada, 2010). The Public Health Layers were homogenized and divided into sub-samples. Grab samples were homogenized and emptied into plastic trays where they could also be separated into sub-samples. Peat samples were kept frozen prior to being sub-sectioned. A hacksaw was used to remove a 5-cm section representing the Public Health Layer from the top of the peat cores. At 37 locations, samples from greater depths (10 cm to 40 cm) were sub-sectioned from cores of sufficient length to document changes in arsenic concentrations at depth.

Soil Sample Analysis

A portion of each sub-sectioned core and grab sample were submitted for near-total elemental analysis at the Analytical Services Unit (ASU) at Queen's University.

The majority of the soil samples were neither sieved nor ground before analysis, the intention being that they represent the surface material as it might be encountered in the field. Sieving is usually done to compare results to guidelines or other data sets (e.g., Parsons and Little 2015), but our previous work (Bromstad *et al.* 2017) had indicated that arsenic trioxide may be present in a range of grain sizes and it was a priority to capture this solid phase. Moreover, a split of many of the samples will be characterized by scanning electron microscope to identify arsenic-hosting minerals, determine their morphology and degree of liberation, and calculate the distribution of arsenic amongst solid phases, which precludes sieving. Some samples (n=19) were sieved to less than 2 mm and the results are included in Appendix A—Table A1.

A total of 46 grab samples and 433 core samples were submitted for analysis. Samples were digested using aqua regia solution containing hydrochloric acid and nitric acid, at a 3:1 volume ratio. Digestion time was 300 minutes at 90 °C. Inductively coupled plasma - mass spectrometry (ICP-MS) was used to determine gold concentrations, and for antimony at concentrations less than 10 mg/kg. The digestion procedure for gold used a rinse composed of hydrochloric acid/cysteine rinse and standard stabilization method. This method was developed by ASU based on stabilization methods provided in Wang and Brindle (2014) and Wang *et al.* (2014). The remaining elements were analyzed by inductively coupled plasma - optical emissions spectrometry (ICP-OES).

A random selection of 258 samples were air-dried and ground before being analyzed for total organic carbon content using the LECO SC444 method. This method calculates organic carbon by subtracting inorganic carbon, determined by ashing the samples at 475 °C, from the total carbon result. Carbon percentages are determined through the combustion and oxidation of carbon to form carbon dioxide by burning the sample at 1350 °C in a stream of purified oxygen. The amount of evolved carbon dioxide is measured using infrared detection and used to calculate the percentages of carbon in each sample (Nelson and Sommers 1982).

Statistical Analysis

All statistical analyses were performed in Minitab 17. A Kruskal-Wallis test and Dunn's post hoc analysis was performed to compare arsenic concentrations between terrain units. Regression analysis was used to investigate the relationship between arsenic concentration, distance and direction from the Giant Mine roaster, and elevation. All tests were performed at a 95 % confidence interval.

Quality Assurance and Quality Control

Reproducibility

Field duplicates, split samples (two samples taken from the same depth in the same core), and internal laboratory duplicates were all analysed to evaluate sample homogeneity and analytical reproducibility (Appendix B—Table B1). The relative percent difference (RPD), calculated between the original sample (denoted as the parent (P) sample) and the quality assurance/quality control (QAQC) sample, was defined by:

$$RPD = \frac{Absolute\ value(Parent - QAQC)}{Average\ (Parent, QAQC)}\ x\ 100$$

Parameters below the analytical detection limit were disregarded for RPD calculations.

Field duplicates (FD) were samples collected in the field as close as possible to the original sampling location and provide an indication of consistency of field methods and sample homogeneity. The results indicate a high degree a variability among the parent sample and the duplicate sample, with 72 % of the samples (n=29) having an RPD greater than 20 % for arsenic. Field observations and detailed soil descriptions in the laboratory suggest that despite samples being collected in close proximity, generally within less than 1 m, soil characteristics can vary widely and thus contributed to the high degree of variability. Examples of these characteristics include the presence or absence of moss, an organic layer, low-lying vegetation or other ground cover, and grain size distribution. Moreover, any components added to the soil from aerial deposition may not have been deposited homogeneously. Wash-down of contaminants into soil pockets surrounded by bare outcrops would also have contributed to an uneven distribution of elements associated with stack emissions. Given that several of the potential arsenic-hosting solid phases contain large amounts of arsenic (arsenopyrite contains 46 wt.% As and arsenic trioxide contains 76 wt.% As), an uneven distribution of particles would result in a relatively large difference in total arsenic concentration.

Split samples (SS) were prepared by dividing a single sample evenly into multiple samples, and submitting these with unique sample names to the laboratory. This provides an indication of variability within individual samples due to the natural properties of the soil varying at the scale of mm to cm. Although the reproducibility is higher than in the case of the field duplicates, with only 32 % of the samples (n=57) having an RPD for arsenic greater than 20 %, there is variability in the distribution of arsenic at this scale. Lab duplicates (LD) were prepared by ASU. Before analysis, samples for laboratory duplicates were selected at random. Two separate portions of the same sample were extracted, analyzed, and reported separately. Reproducibility was higher than split samples and field duplicates, with only 23 % of the samples (n=62) having an RPD for arsenic greater than 20 %. The generally low reproducibility of the duplicates is probably also due to the fact that the samples were neither mixed nor ground.

Accuracy

Accuracy of analytical results was evaluated by repeated measurements (n=44) of several certified reference materials—purchased soil sample standards (SS-1 and SS-2 from SCP Science, Quebec, and MESS-3 and MESS-4). MESS-3 and MESS-4 soil sample standards are based on the National Research Council Canada (2016) certified values for *Marine Sediment Reference Material for Trace Metals and other Constituents*. ASU's expected result of 18 mg/kg for MESS-3 and MESS-4 is based on an average of results obtained for partial digestion. One RPD for arsenic was 23 %, all others less than 20 % and most less than 10 % (Appendix B—Table B2).

Results and Discussion

Appendix A—Table A1 is a compilation of all the data for the 2015, 2016, and 2017 sampling period, including sample location, terrain type, distance to, and direction from, former ore roasters, elevation, and concentration of all elements reported. Appendix A—Table A2 explains the labels used for sampling.

Regional Variability in Arsenic Concentrations Within the Public Health Layer

Total arsenic concentrations within the Public Health Layer of soils are presented in Figure 3. Arsenic concentrations are indicated as lower than the residential remediation guideline of 160 mg/kg, between the residential and industrial remediation guideline of 340 mg/kg, or above both these concentrations in three higher concentration ranges (GNWT 2003). Concentrations are highest in the area close to the Giant Mine property, including the Bypass Road target area. Elevated concentrations are also observed near the Con Mine property. Arsenic concentrations at sites near the outer perimeter of the sampling area are considerably lower than near the two former mines. Based on the samples collected to date (n=18), 83 % of arsenic concentrations in samples collected along the Detah road were below the residential guideline of 160 mg/kg, ranging from 5.9 mg/kg to 270 mg/kg (median = 54 mg/kg). Arsenic concentrations in Ndilǫ (n=5) ranged from 130 mg/kg to 280 mg/kg (median = 180 mg/kg); one sample was below the residential guideline and the remaining four samples were between the residential and industrial guidelines. Arsenic concentrations near Long Lake and Fred Henne Territorial Park (n=70) range from 32 mg/kg to 3000 mg/kg, with 33 % of samples below the residential guideline (median = 240 mg/kg). Figure 4 shows the variation of arsenic concentrations in target areas in a box and whisker plot, as well as guidelines. The number of samples in each target area is highly variable and therefore comparisons between target areas should be made with caution. Figure 5 shows arsenic concentrations in the Public Health Layer in undisturbed sites near Yellowknife. In this figure, higher arsenic concentrations are indicated by larger symbols and darker colours.

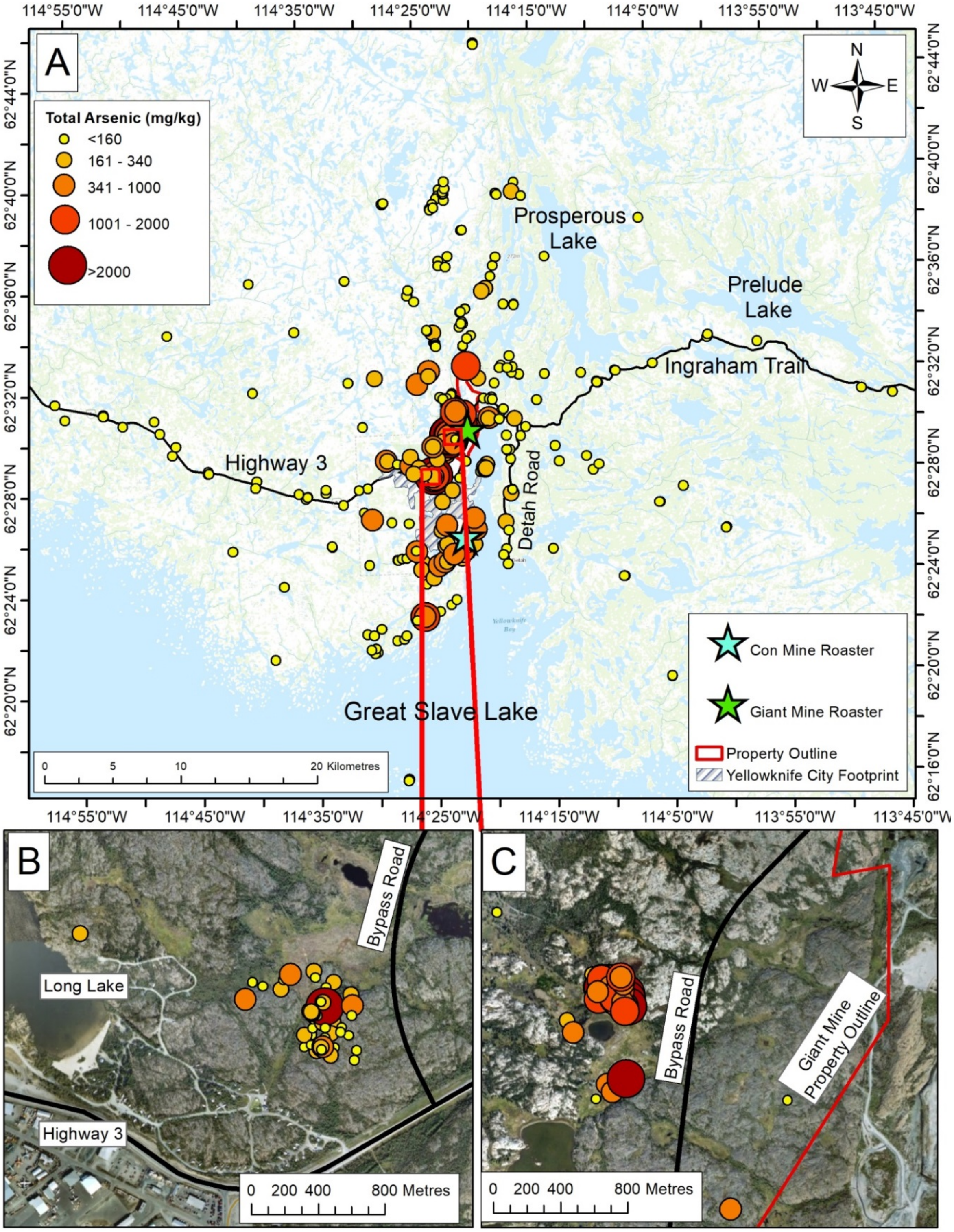

Figure 3. Total arsenic concentrations in the Public Health Layer at all core sample sites. The size and colour of the symbol indicates arsenic concentration. Figure A shows the results for the entire study area; Figure B shows the results in the detailed sampling grid located near Fred Henne Territorial Park; Figure C shows the results for the detailed sampling grid along the Giant Mine Bypass Road, west of Giant Mine.

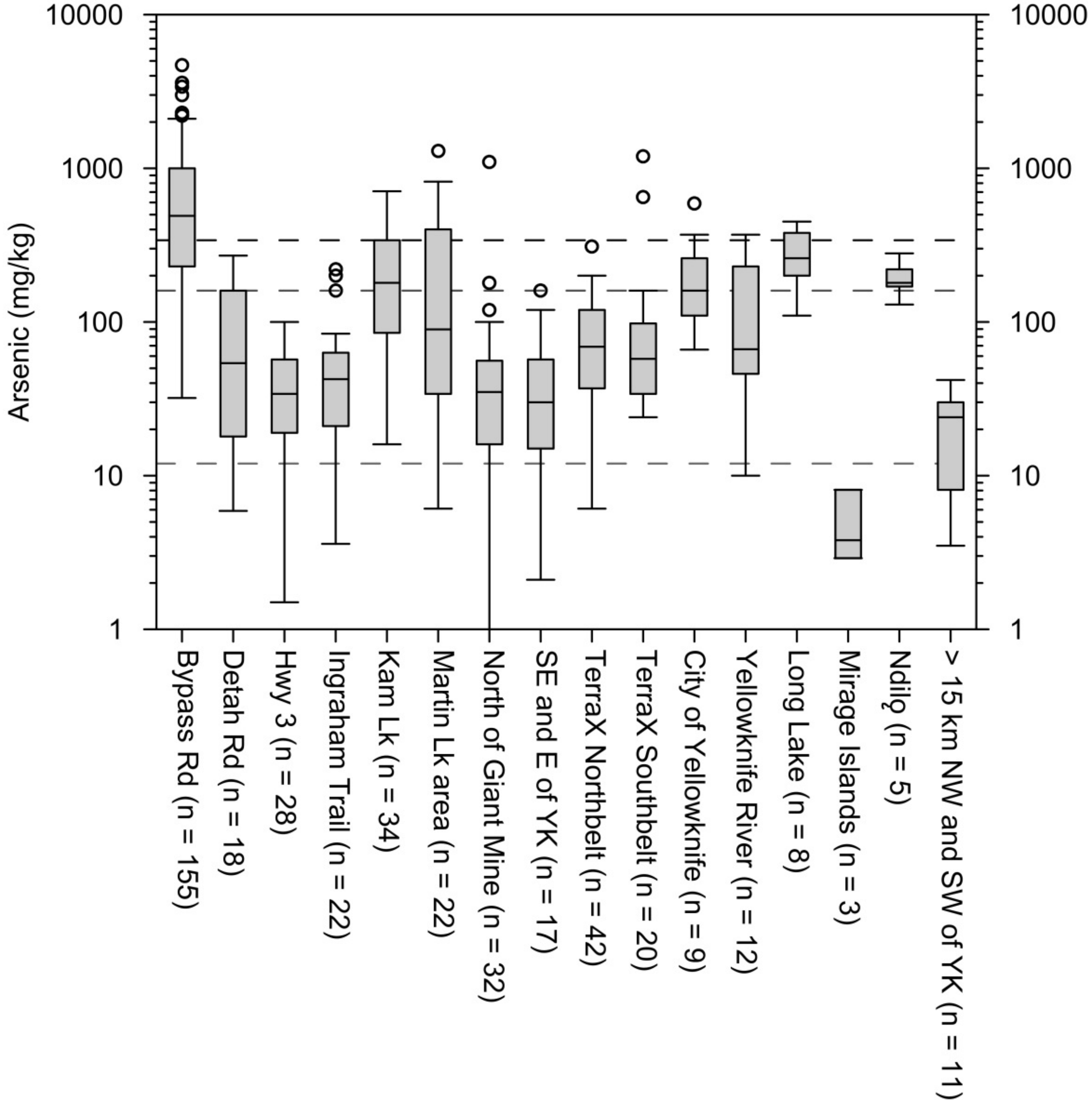

Figure 4. Box and whisker plots depicting the concentration of arsenic in the randomly selected target areas. The upper and middle dashed lines represent the Government of Northwest Territories site-specific health based soil quality remediation objectives of arsenic concentrations in Yellowknife soils (2003)—the upper dashed line is the Industrial Remediation Objective (340 mg/kg) and the middle dashed line is the Residential Remediation Objective (160 mg/kg). The lower dashed line represents Health Canada’s 2010 Canadian Soil Quality Guideline of arsenic in soil for agriculture, residential/parkland, commercial, and industrial (12 mg/kg) concentrations. The “Distal East and SE sites" and "Hay Lake" were combined as “SE and E of YK” due to small sample sizes.

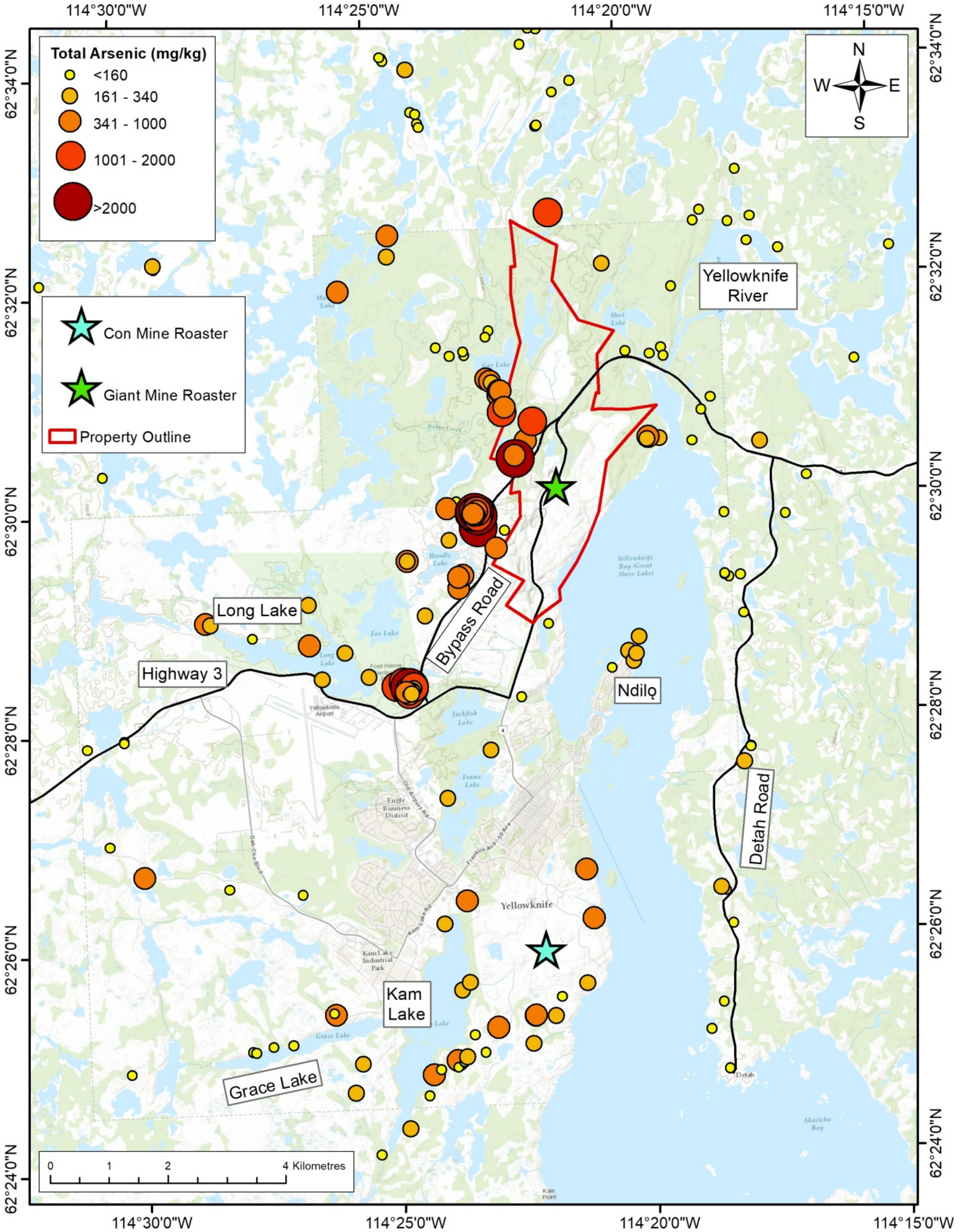

Figure 5. Total arsenic concentrations in the Public Health Layer at sites near Yellowknife. Higher arsenic concentrations are shown by larger symbols and darker colours.

Influence of Distance to Former Roasting Sites

Figure 6 shows the relationship between the concentration of arsenic in the Public Health Layer of soils and distance to the former Giant Mine roaster. There is a clear inverse relation, with arsenic concentrations decreasing with increased distance from the roaster. This observation is similar to previous studies in the area focused on soils (Hocking *et al*. 1978; Hutchinson *et al.* 1982; Kerr 2006; St. Onge 2007; Bromstad *et al*. 2017), lake waters (Palmer *et al.* 2015, Houben *et al*. 2016) and lake sediments (Galloway *et al*. 2015). Although the trendline does not reach a clear plateau, the concentrations are much lower at 25 km away from the roaster and are likely approaching background at this distance. The arsenic concentration in Public Health Layer soils from sites 20 km to 30 km from the Giant roaster ranges from 1.0 mg/kg to 63 mg/kg (n = 25; median = 40 mg/kg). This is generally consistent with the findings of Hocking *et al.* (1978), Hutchinson *et al.* (1982), Kerr (2006), and St. Onge (2007). Our range of background values is substantially lower than the background value of 150 mg/kg As indicated by previous studies (Risklogic 2002; GNWT 2003).

Soil samples taken within 3 km of the former Con roaster are highlighted in Figure 6, and are mostly distributed above the trendline, suggesting that arsenic concentrations in these soils may be affected by the Con mine, as concluded by Hocking *et al.* (1978). For all the data, there is considerable scatter about the trendline, suggesting that additional factors influence arsenic concentration in soils in the region.

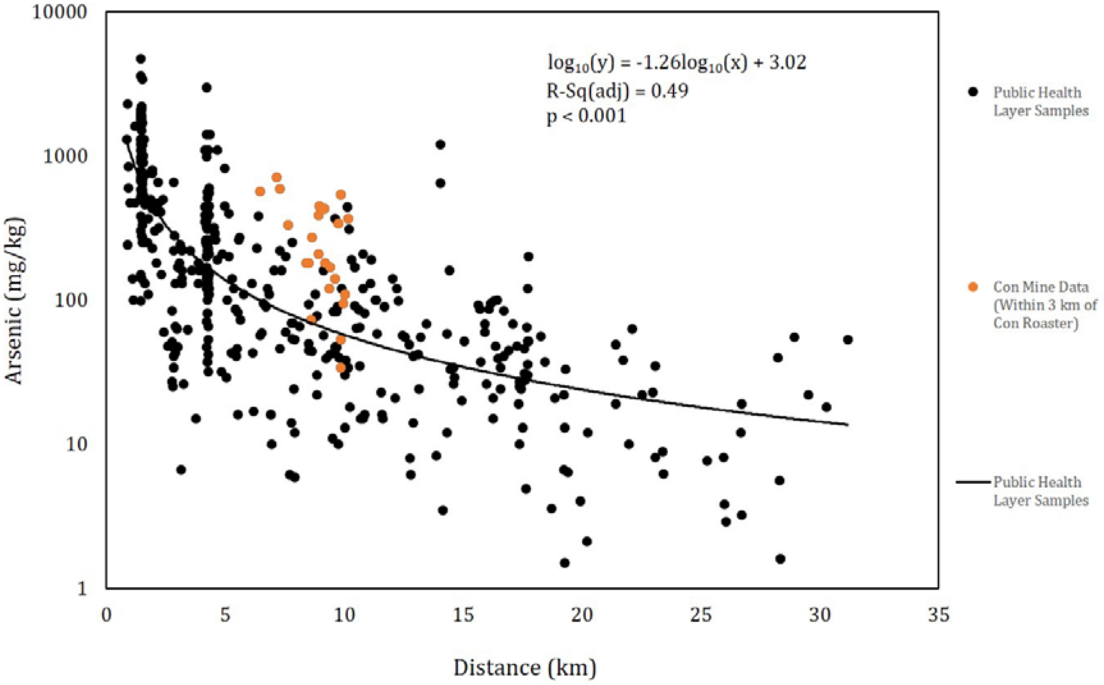

Figure 6. The variation of arsenic concentration in soil samples with distance from the former Giant Mine roaster. Samples collected from the Public Health Layer are in black and samples collected within 3 km of Con Mine are highlighted in orange and are included in the analysis.

The Influence of Wind Direction

The influence of direction from the former roasters was explored since previous studies have shown that the influence of stack emissions is greatest at sites downwind of predominant wind directions in the region (e.g., Hocking *et al*. 1982; St. Onge 2006; Palmer *et al*. 2015; Bromstad *et al*. 2017). The inset to Figure 2 shows that winds blow most frequently from the east and northeast in the region. Figure 7 demonstrates that median arsenic concentrations in near-surface soils were highest to the west of Giant Mine, consistent with the predominant wind direction in the region. Concentrations were lowest east of the historical roaster along the least frequent wind direction.

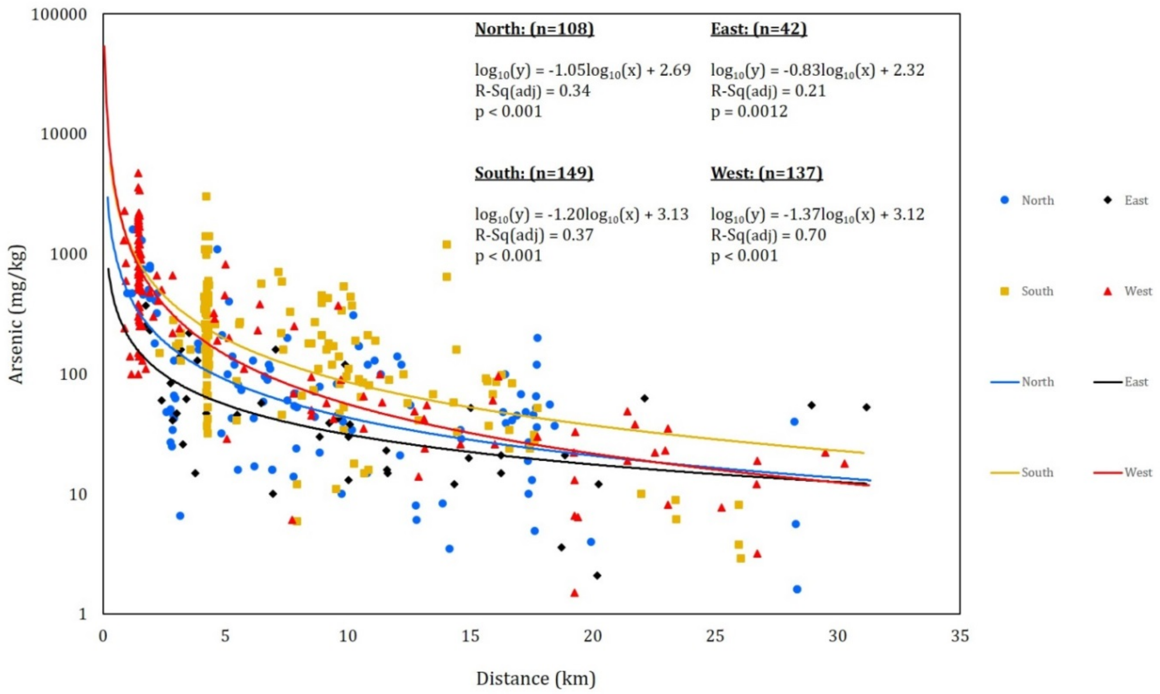

Figure 7. The variation of arsenic concentrations within 35 km of the former Giant Mine roaster with respect to direction. Direction classification is centred around the former Giant Mine roaster as East (45° to 135°), North (315° to 45°), West (225° to 315°), and South (135° to 225°). Coloured lines represent slope and coloured symbols represents the arsenic concentration in samples.

The Influence of Elevation

Figure 8 shows the variation of arsenic concentration in soils with elevation. This relation was explored since higher sites may have intercepted more stack emissions during the early years of roaster operations. There is a correlation between higher elevation and higher arsenic concentration, although it is not very strong and undoubtedly influenced by other factors such as distance and wind direction. The high elevation sites were found west of the Bypass road and north of Giant mine near Landing and Vital lakes (Appendix A—Table A1).

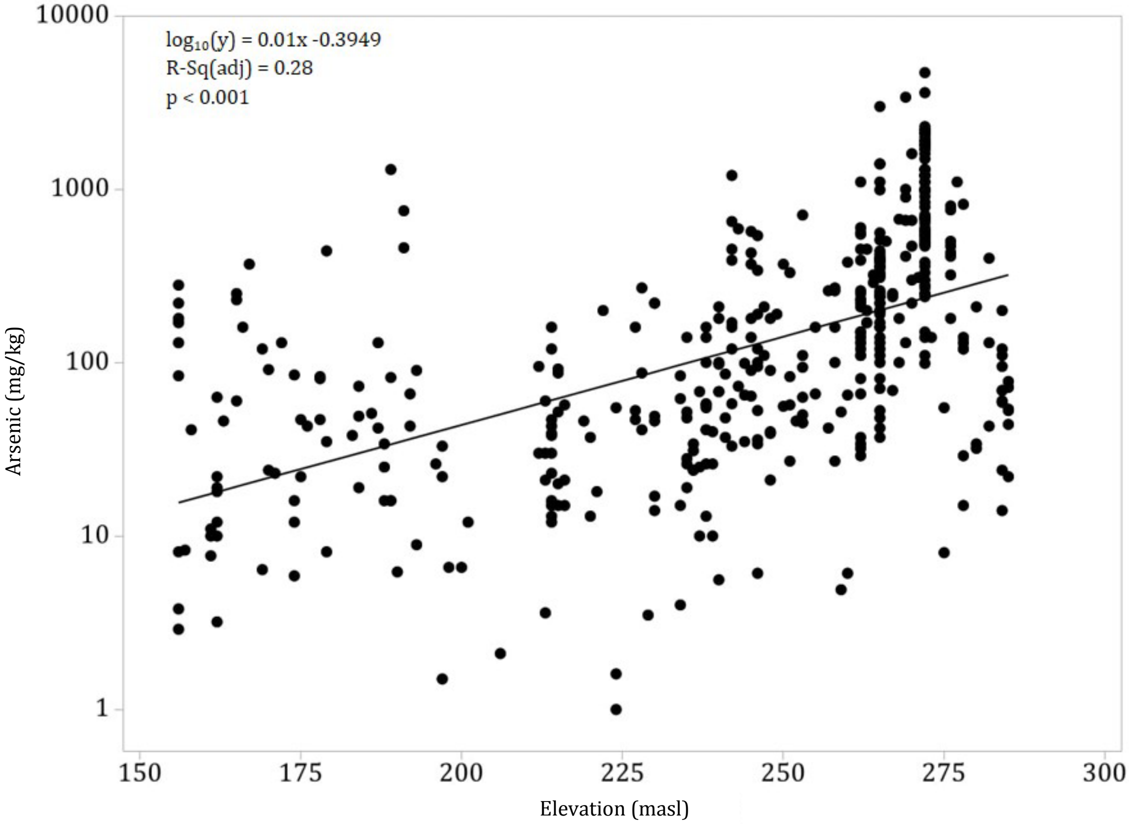

Figure 8. The relation between arsenic concentration in the Public Health Layer of soils and elevation (masl, metres above sea level) in the study area.

The Influence of Terrain Type

Figure 9 shows the relation of arsenic concentration in the Public Health Layer to terrain type, as defined above in Sampling Protocol, and documented in Appendix A—Table A1. Total arsenic in forest soil samples range from 1.0 mg/kg to 1300 mg/kg with a median of 38 mg/kg; forest outcrop samples have a range of 2.1 mg/kg to 4700 mg/kg with a median of 150 mg/kg; outcrop soil samples range from 3.5 mg/kg to 3000 mg/kg with a median of 165 mg/kg; finally, peatland samples range from 2.9 mg/kg to 3400 mg/kg with a median of 95.5 mg/kg. The distribution of arsenic concentrations in forest samples were significantly different than forest outcrop, outcrop, and peatland samples ($p < 0.001$). There were no statistically significant differences between the distributions of arsenic within the median outcrop, forest outcrop, and peatland terrain units ($p > 0.05$).

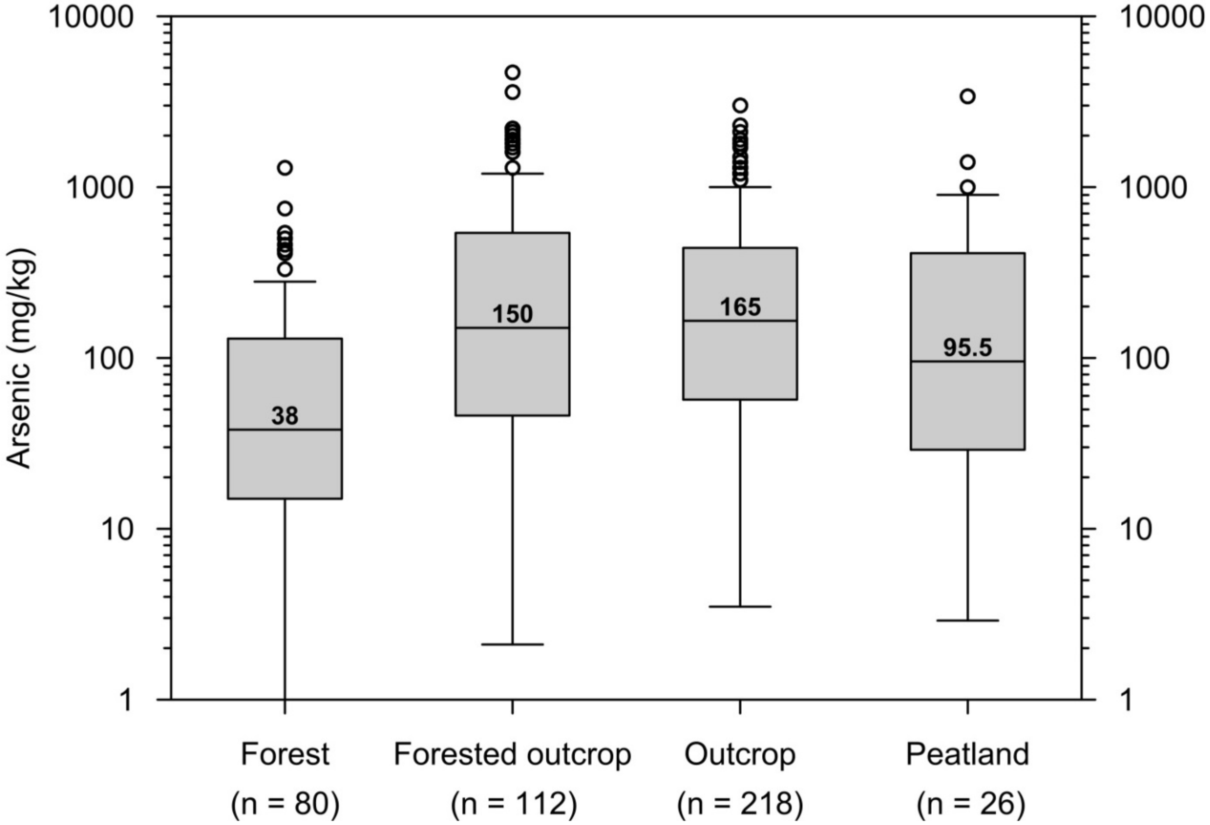

Figure 9. The relation of arsenic concentration in the Public Health Layer to terrain type. The distribution of arsenic concentrations in forest samples were significantly different than forest outcrop, outcrop, and peatland samples (Kruskal-Wallis Test, $p < 0.001$). There were no statistically significant differences between the distributions of arsenic within the median for outcrop, forest outcrop, and peatland terrain units (Kruskal-Wallis Test, $p > 0.05$). Mean values of terrain type are shown.

Considerable variability was observed between multiple samples collected in detailed sample plots (Figure 3B and Figure 3C), suggesting several factors could play a role in the distribution of arsenic and soil geochemistry at the local scale. Further research results, including identification of the arsenic-hosting solid phases, is expected to identify factors controlling local variation in arsenic concentration.

Effect of Soil Depth

Most of the results reported here are from the top 5 cm of soil (Public Health Layer) except for the 37 samples labelled DC (down core). Figure 10 shows that, with two exceptions, all of the deeper samples have much lower concentrations of arsenic than the corresponding sample from the Public Health Layer. The dashed lines connect the two samples from the same core, but are not meant to imply that arsenic decreases linearly with depth. Previous work (Hocking *et al.* 1978; Kerr 2006; Bromstad *et al.* 2015, 2017) describes a rapid drop of arsenic concentration with soil depth in the Yellowknife area.

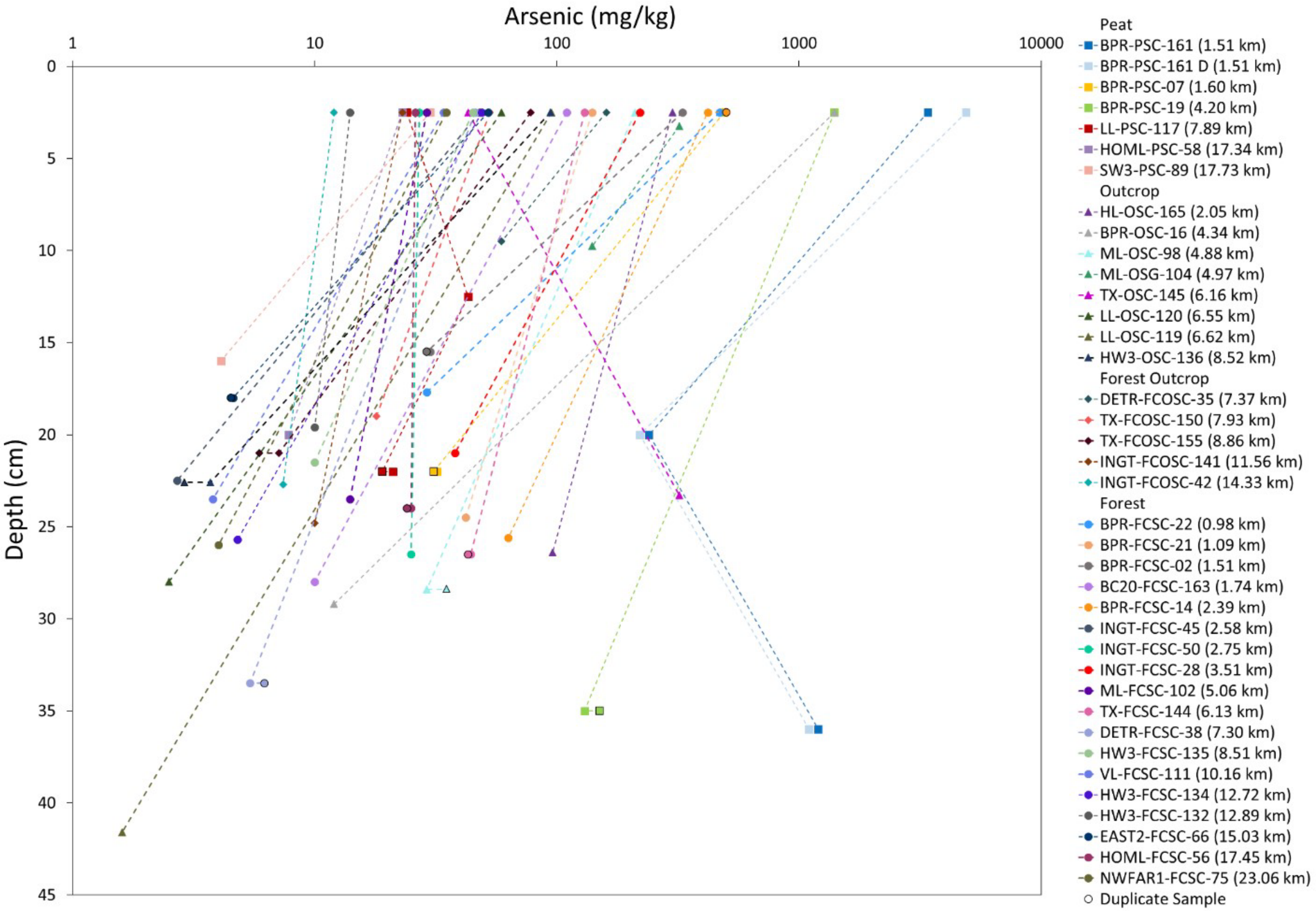

Figure 10: Arsenic concentrations from the Public Health Layer and at various depths for 17 soil cores from the Yellowknife region. Sample sites are organized by terrain type (Peat – square, Outcrop – triangle, Forest Outcrop – diamond, and Forest – circle); the distance from Giant Mine to the respective sample sites is indicated in parentheses.

Grab Samples

Grab samples, collected mainly in areas of outcrop terrain, ranged in arsenic concentration from 2.9 mg/kg to 930 mg/kg (n = 46; median = 120 mg/kg). Grab sample concentrations fall within the range of core sample concentrations collected in nearby areas. The samples with the highest arsenic concentrations were collected near the Bypass Road, near Giant Mine.

Conclusions

This report provides data from the top 0 cm to 5 cm of soil for 479 samples from undisturbed sites within 30 km of Yellowknife. The influence of distance from the former roasters, wind direction, elevation and terrain type have all been explored. Based on the relation between arsenic concentration and distance from the Giant roaster, soil concentrations of arsenic likely approach background concentrations at a distance of 20 km to 30 km. Arsenic concentrations range between 1 mg/kg and 63 mg/kg (n = 25; median = 40 mg/kg) at this distance, and can be considered a reasonable, albeit conservative, estimate of background conditions in the region. The results show that arsenic concentrations are also elevated near the former Con roaster. Higher arsenic concentrations are found downwind (i.e., west) of the Giant roaster, at higher elevations, and in outcrop and forest outcrop soils compared to forest soil. The presence of elevated concentrations of arsenic in soils in the Yellowknife area that are hundreds to thousands of times the Canadian Soil Quality Guideline has been known for at least forty years, and this enrichment is attributed to the operation of gold ore roasters based on the spatial distribution and wind direction (e.g., Hocking *et al.* 1978).

Further work on selected subsamples will include identification of the arsenic-hosting solid phases, using techniques described in Bromstad *et al*. (2015, 2017). These results will help distinguish natural from roaster-generated arsenic-bearing particles. In particular, the presence of arsenic trioxide and roaster-generated maghemite and hematite have been interpreted as clear evidence of stack emissions (Bromstad *et al*. 2015, 2017). It is not clear whether roaster products from Con can be distinguished from those from Giant.

The results documented in this report contribute to an understanding of the risk to human and ecosystem health associated with arsenic in soils in the Yellowknife region. However, evaluation of that risk requires additional information, particularly an assessment of exposure. Oral ingestion, inhalation and dermal contact are all pathways by which humans and other organisms can be exposed to arsenic in soil. In the case of arsenic, it is well known that particle size, degree of liberation, and the chemical form (mineralogy) of arsenic in the solid phase all influence bioaccessibility, or the solubility of arsenic in bodily fluids (Ruby *et al*. 1999; Meunier *et al*. 2010; Plumlee and Morman 2011). Arsenic trioxide, the dominant solid phase of arsenic released by roaster stack emissions, is considered to be the most toxic and bioaccessible form of solid-phase arsenic (Plumlee and Morman 2011;

Jamieson 2014). Exposure, in a general sense, also depends on physical access to contaminated soil. In the Yellowknife area, this may increase as a result of site remediation and construction of new infrastructure such as the Giant Mine Bypass Road.

An industrial site-specific remediation guideline of 340 mg/kg total As and residential guideline of 160 mg/kg As were established by the Government of the Northwest Territories in 2003 (GNWT 2003). These criteria are much higher than the Canadian Soil Quality Guideline of 12 mg/kg (CCME 2007), and were based on the expectation that exposure would be limited by the cold climate and that the "average natural background concentration of arsenic in and around Yellowknife was determined to be 150 ppm". The industrial guideline was also based on the expectation of "little or no public access". The results reported here show the natural background in near-surface soil concentrations of arsenic are much lower (~ 3 mg/kg to 63 mg/kg). This is consistent with previous studies (Hocking *et al.* 1978; Hutchinson *et al.* 1982; Kerr 2006; St. Onge 2006), which show elevated natural background arsenic is restricted to soils above mineralized veins and shear zones (Kerr 2006). The results of our ongoing research on the speciation of arsenic in soils with hundreds to thousands of mg/kg arsenic will provide insight into whether these high concentrations are the result of natural or anthropogenic sources, and will help identify the main solid-phase hosts of arsenic.

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Appendices

Appendix A — Table A1 – Compilation of all the data for the 2015, 2016, and 2017 sampling period

Appendix A — Table A2 – Definitions and abbreviations

Appendix B — Table B1 – Reproducibility of measured As concentration in Field Duplicates, Lab Duplicates and Split Samples

Appendix B — Table B2 – Comparison of measured and expected arsenic concentrations in Certified Reference Materials

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