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PRIMARY RESEARCH PAPER

Multiple environmental variables influence diatom assemblages across an arsenic gradient in 33 subarctic lakes near abandoned gold mines

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Abstract We examined surface sediment diatom assemblages from 33 subarctic lakes around the historic gold mines in Yellowknife, Northwest Territories (Canada), where lake-water As concentrations ([As]) still range between 1.5 and 2,780 μg/l, even though the roasting of the arsenopyrite-bearing gold ores ceased in 1999. Water chemistry variables related to gold mining pollution (arsenic, antimony, and sulfate) declined with increasing distance from the mines. The diatom assemblages varied substantially in species composition across our study lakes with the planktonic *Discostella stelligera/pseudostelligera* complex dominating in deep lakes where [As] were less than 5 μg/l. However, in lakes that exceeded the guideline for the protection of aquatic life ([As] > 5 μg/l), opportunistic benthic diatoms (small fragilarioid taxa and *Achnanthidium minutissimum*) were generally present in higher abundances. The subtle differences in diatom species richness and diversity among lakes with varying [As] suggest these indices may not be affected strongly by long-term arsenic pollution. Furthermore, canonical correspondence analysis of the diatom assemblage and environmental data indicated that nutrient and ionic compounds explained most of the variation in the assemblages, while mining-related variables played a limited role. Our results indicate that multiple limnological variables (nutrients, ions, and contaminants) and subarctic climatic conditions are important factors shaping the diatom assemblage composition in lakes impacted by mining activities and land-use changes near Yellowknife.

Keywords Contaminants · Arctic · Limnology · Algae · Indicators · Yellowknife · Northwest Territories · Metals

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Introduction

Diatoms (Bacillariophyceae) are excellent indicators of water quality as they rapidly respond to changes in chemical (nutrients, pH, metal concentrations) and physical (thermal structure, habitat availability) conditions in freshwater ecosystems (Smol & Stoermer, 2010). Furthermore, their siliceous cell valves are typically well preserved in most lake sediments making them ideal paleolimnological indicators to study a variety of environmental stressors, such as acidification, heavy metal pollution, and eutrophication (e.g., Dixit et al., 1992; Hall & Smol, 1996; Cattaneo et al., 2008; Fernández et al., 2018). Previous studies have examined the relationships between varying metal concentrations (e.g., aluminum, nickel) and diatom assemblages in acidified temperate lakes (e.g., Dixit et al., 1991; Cumming et al., 1992) and other lakes that have been affected by metal mining operations (Ruggiu et al., 1998; Salonen et al., 2006; Leppänen et al., 2017). However, their sensitivity and response to arsenic (As) contamination has not been extensively documented in lakes from climatically sensitive subarctic regions where mining is considered to be a major growth industry (Northwest Territories Industry, Tourism and Investment, 2014).

The lakes around Yellowknife, Northwest Territories, present an important opportunity to assess the influence of As on diatom assemblages, as these lakes have some of the highest As concentrations ([As]) in Canada (e.g., Palmer et al., 2015; Houben et al., 2016; Fig. 1), with many exceeding the federal water quality guidelines for the protection of aquatic life (5 µg/l; Canadian Council of Ministers of the Environment, CCME, 2001) and drinking water (10 µg/l; Health Canada, 2006). The gold mining operations around the City of Yellowknife, beginning in the mid-twentieth century, resulted in the emission of more than 20,000 tonnes of arsenic trioxide into the surrounding environment (Silke, 2009; Jamieson, 2014) and led to the atmospheric deposition of mining-related contaminants into nearby lakes (Palmer et al., 2015). Much of the arsenic trioxide was released from Giant Mine (Fig. 1) where the arsenopyrite-bearing gold ores were roasted between 1949 and 1999 and during this period it was the largest producer of gold in the region. Recent limnological surveys and geochemical analyses of lake sediments demonstrate the exceptionally high [As] are a legacy of the twentieth-century gold mining operations around Yellowknife, and not a result of the underlying geology (Palmer et al., 2015; Houben et al., 2016; Thienpont et al., 2016; Galloway et al., 2018; Schuh et al., 2018; Van den Berghe et al., 2018). Furthermore, the concentrations of As, antimony (Sb), and sulfate in the lake waters were inversely correlated to distance from the mine roaster stacks and corroborate that the increased concentrations of contaminants are a result of roasting arsenopyrite ores to extract gold (Houben et al., 2016). In addition to the aerial deposition of contaminants, lakes within the City of Yellowknife were also impacted by local land-use changes, such as urbanization (Dirszowsky & Wilson, 2016), raw sewage input (Stewart et al., 2018), and accidental spills from mine tailings (Bright et al., 1994). Although mining operations ceased in the early-2000s, the [As] in many lakes is still high but the effects of prolonged exposure to elevated [As] on key aquatic primary producers, such as diatoms, are poorly understood at a regional scale.

Previous studies in laboratory settings have reported that the acute toxicity of arsenic may lead to a decline in the relative abundance of green to blue-green algae and reduce the photosynthetic efficiency in diatoms (Tuulaikhuu et al., 2015). Furthermore, short-term field experiments have shown that the total abundance of diatoms on biofilms decreases when exposed to high arsenic concentrations (Barral-Fraga et al., 2018). A reduction in cell size and differences in community composition of biofilms have also been noted as a response to high arsenic concentrations (Rodriguez Castro et al., 2015; Barral-Farga et al., 2016). Here we examined diatom distributions along a gradient of lake-water arsenic concentrations ([As]) by analyzing surface sediment assemblages and measured environmental variables from 33 subarctic lakes around Yellowknife where algal communities have been exposed to high [As] for more than 50 years. The lakes were strategically chosen to cover a large gradient of [As] ($\sim$ 1.5–2,780 µg/l) along a $\sim$ 40-km radius from the city (Fig. 1). The goals of this exploratory study were to assess: (1) the potential influence of lake-water [As] on diatom assemblage composition, and (2) if other measured environmental variables were important in structuring the assemblages.

Study area

Yellowknife (Fig. 1) is situated on the northern shores of Great Slave Lake, within the Slave Structural Province of the Canadian Shield (Wolfe, 1998; Kerr &

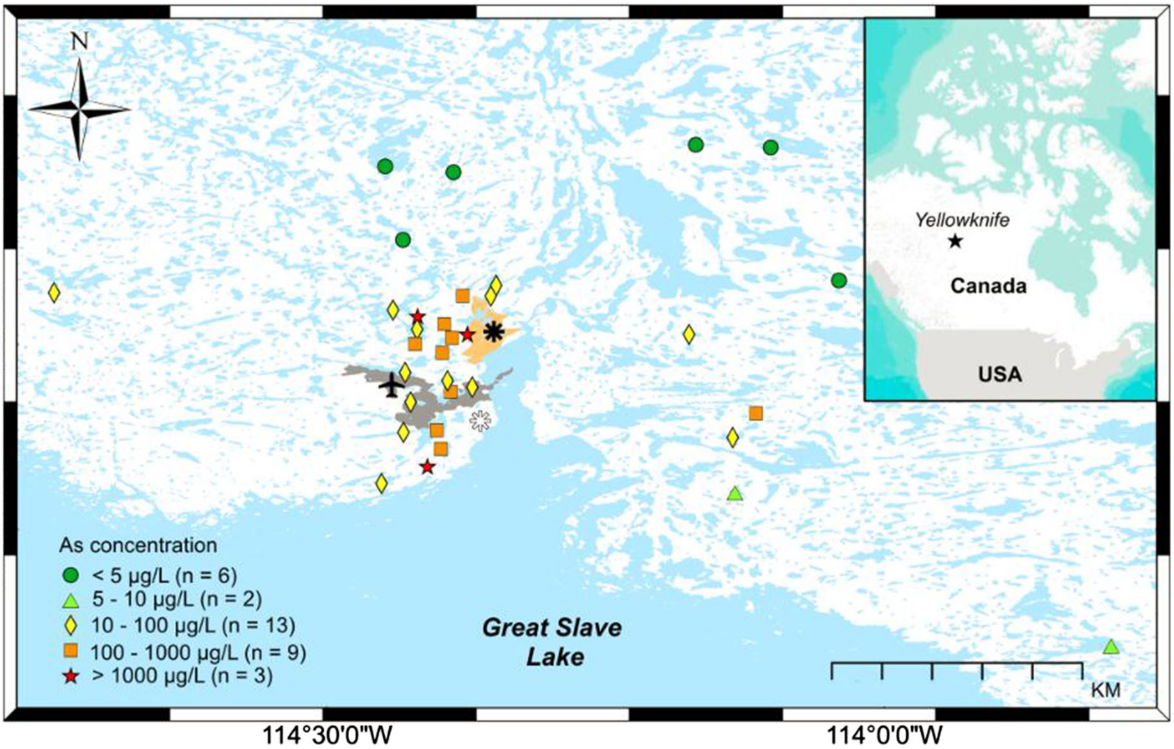


Fig. 1 A map of the study area of lakes around Yellowknife, Northwest Territories, Canada. The shaded asterisk indicates Giant Mine while the clear asterisk denotes Con Mine. The Yellowknife Airport is indicated by an air plane. The shapes of the symbols represent the concentration of arsenic in the water column and the number of lakes in each category are included. The inset map shows the location of Yellowknife within Canada

Wilson, 2000). The Yellowknife area contains pockets of discontinuous permafrost and experiences a continental subarctic climate (Wolfe, 1998). Based on the meteorological data collected at the Yellowknife airport from 1942 to 2016, the mean annual air temperature ranged between − 1 and − 7°C, while the mean annual precipitation varied between 195 and 507 mm (data retrieved in April 2017 from—http://www.ec.gc.ca/dccha-ahccd/). Yellowknife and the surrounding area are located within the Great Slave Uplands and Lowlands of Taiga Shield High Boreal ecoregion (Ecosystem Classification Group, 2008). The regional vegetation is characterized by the presence of black spruce [*Picea mariana* (Mill.) Britton, Sterns & Poggenburg], jack pine (*Pinus banksiana* Lamb.), paper birch (*Betula papyrifera* Marshall), trembling aspen (*Populus tremuloides* Michx), and white spruce [*Picea glauca* (Moench) Voss] with rock lichen occurring on exposed bedrock (Ecosystem Classification Group, 2008). The Weledeh Yellowknives Dene First Nations have used lakes around the Yellowknife area to travel, hunt, and fish for many centuries, as this area has traditionally supported excellent fisheries (Weledeh Yellowknives Dene, 1997). However, recent biological surveys from the freshwaters near the mines have recorded elevated [As] in the tissues of fish species and this may be of concern to human health (e.g., Cott et al., 2016). Regardless of the high arsenic concentrations in many lakes around this area, they continue to be used for recreational purposes.

The study lakes were generally small (mean 0.95 km^2, median 0.36 km^2, range 0.01–6.28 km^2), shallow (mean 4.6 m, median 3.5 m, range 0.5–21 m), and underlain by Precambrian granitoids, meta sedimentary bedrocks, or volcanic rocks (Kerr & Wilson, 2000; Houben et al., 2016). Ice-cover data were not available for all lakes in this study; however, the long-term ice phenological data available from 1956 to 1994 for one of the study lakes (Long Lake) and the Back Bay of the Great Slave Lake suggest that these lakes are ice-covered for most of the year (range

196–237 days; Benson et al., 2013). Detailed limnological characteristics are provided in Supplement 1 and discussed in subsequent sections. Most of the study lakes were circumneutral to alkaline and the concentrations of total arsenic in the lake water declined with increasing distance from Giant and Con mines. The lakes were divided into five groups based on varying total [As] (i.e., [As] > 1000 μg/l, 1000 > [As] > 100 μg/l, 100 > [As] > 10 μg/l, 10 > [As] > 5 μg/l, [As] < 5 μg/l) to better visualize the As gradient in our study lakes (Fig. 1). Two of the categories were based on the guidelines for the protection aquatic life and drinking water quality in Canada (i.e., 10 > [As] > 5 μg/l, [As] < 5 μg/l), and the other three categories were delineated to ensure the rest of the 25 lakes with > 10 μg/l of total [As] were not clumped into one large group as it may lead to the loss of valuable ecological information. However, this coding method was not used in the statistical analyses as there were notable differences in the number of samples within each category.

Methods

Sample collection

Surface sediments and water chemistry variables were collected from our study lakes (Fig. 1) between March 2014 and July 2016. Due to logistical challenges, some lakes in this study were sampled during the ice-on season while others were sampled after ice melt, with the latter the more common approach when working in subarctic regions. The water and sediment samples were usually obtained from the middle of the lake as we did not have detailed bathymetry for many of the remote study lakes. Hence, the depth of lakes reported in this study were measured at the coring site. Sediment cores were retrieved using an Uwitec gravity corer and sectioned using a modified Glew (1988) vertical extruder at 0.5 cm increments. The water-sampling bottles were triple-rinsed with lake water and then filled completely right before retrieving the sediment cores. Water chemistry variables were measured at the Taiga Environmental Laboratory in Yellowknife, Northwest Territories, following analytical protocols based on Standard US Environmental Protection Agency methods (Taiga Environmental Laboratory is a Canadian Association for Laboratory Accreditation Incorporated certified facility).

Diatom preparation and identification

The top 0.5 cm of the sediments were analyzed for siliceous microfossils following standard protocols described by Battarbee et al. (2001). However, for 10 lakes, surface sediments were no longer available from the top 0.5 cm (as they were used for other analyses); therefore, subsequent intervals (0.5–1.0 cm or 1.0–1.5 cm) had to be used. Approximately 0.2 g of wet sediments or 0.02 g of dry sediments from each sample was treated with strong acids (50:50 molar ratio of concentrated nitric and sulfuric acids) and placed in a hot-water bath for ∼ 2 h to accelerate the breakdown of organic material. Samples were then allowed to settle for ∼ 24 h prior to removing the supernatant, followed by rinsing with de-ionized water. This rinsing procedure was repeated about six times to dilute the strong acids. Once the samples reached circumneutral pH, aliquots of the slurries were plated onto glass cover slips and allowed to air-dry before mounting them on microscope slides using Naphrax®. A minimum of 500 diatom valves were enumerated and identified for each sample at × 1000 magnification using a Leica DMR light microscope fitted with differential interference contrast optics. Diatom taxonomy was based on an assortment of texts, including Krammer & Lange-Bertalot (1986, 1988, 1991a, b), Lange-Bertalot & Moser (1994), Camburn & Charles (2000), Fallu et al. (2000), Håkansson (2002), and Rühland et al. (2003a). *Staurosira construens* var. *venter* (Ehrenberg) Hamilton and *Staurosirella pinnata* (Ehrenberg) D. M. Williams & Round were both observed in our dataset. However, due to challenges in differentiating valves when present in girdle view, we grouped these taxa together for statistical analyses as they have similar optima for critical limnological variables such as dissolved inorganic carbon, dissolved organic carbon (DOC), total nitrogen (TN), pH, total phosphorus, and maximum depth (Hall & Smol, 1996; Rühland & Smol, 2002; Schmidt et al., 2004). The diatom assemblage composition was presented as percent abundance relative to the total number of valves counted in a sample, and all species were included in statistical analyses.

We examined the relative abundances of planktonic and benthic taxa in the diatom assemblages from the 33 lakes by grouping taxa with similar habitat preferences. As mentioned in Philibert & Prairie (2002), the classification of diatom taxa as benthic and planktonic species is a difficult task because different diatom surveys have reported the same taxa from diverse habitats. Therefore, we used a variety of sources including published literature (e.g., Round et al., 1990; Philibert & Prairie, 2002), online resources (e.g., Diatoms of North America), and discussions with researchers who have worked with diatom flora from the Canadian Arctic extensively to best determine the habitat preference for various taxa. In summary, we classified species from the following genera as benthic: *Achnanthes* sensu lato, *Amphora*, *Amphipleura*, *Brachysira*, cymbelloid taxa, *Caloneis*, *Cocconeis*, *Denticula*, *Diploneis*, *Diatoma*, *Eunotia*, *Epithemia*, *Frustulia*, benthic fragilarioid taxa, *Gomphonema*, *Gyrosigma*, naviculoid taxa, *Neidium*, *Hantzschia*, *Nitzschia*, *Rhopalodia*, *Pinnularia*, *Stauroneis*, and *Surirella*. The following genera were classified as planktonic: *Aulacoseira*, *Asterionella*, *Cyclotella* sensu lato, pennate fragilarioid taxa, *Melosira*, *Stephanodiscus*, and *Tabellaria*.

Data screening for ordination analyses

A total of 47 environmental variables were measured as part of this limnological survey. However, many water chemistry variables contained concentrations that were below the detection limits (BDLs). Therefore, we only used environmental variables that had measurements above the detection limits in at least 90% of the lakes ($\sim$ 30 lakes) (sensu Karst-Riddoch et al., 2009). Furthermore, if an environmental variable had concentrations BDL in 3 lakes or less ($< 10\%$), then it was replaced with the value of the detection limit (BDL at 1 site—chloride, total antimony, total iron, BDL at 2 sites—uranium, BDL at 3 sites—sulfate). Values for total phosphorus and dissolved phosphorus were not included in the analysis because measurements of phosphate are often interfered with by arsenate (AsO_4^{3-}) in lakes with high [As] (e.g., Stewart et al., 2018).

The chemical and physical variables from the 33 lakes were examined for normality using the Shapiro–Wilk test and appropriate transformations were applied, using the vegan (Oksanen et al., 2018) and analogue (Simpson & Oksanen, 2018) packages in the R Studio statistical software environment (R Core team, 2018). While assessing the environmental variables for normality, we noted that the two lakes (Keg and Peg) that received tailings from Pud Lake near the abandoned Con Mine had exceptionally high values for certain ions and specific conductivity. The anomalous values from Keg and Peg lakes prevented these variables to be normally distributed, even after transformations of the data. Therefore, Keg and Peg lakes were removed from further statistical analysis related to ordinations as these sites may have excess leverage on normalized ordination techniques.

Latitude (in decimal degrees) was the only environmental parameter that was normally distributed, and so all other environmental variables had to be transformed to meet the assumption of normality in the 31 lakes dataset. TN, DOC, total alkalinity, specific conductivity, total dissolved solids, calcium (Ca^{2+}), chloride (Cl^{-}), magnesium (Mg^{2+}), potassium (K^{+}), sodium (Na^{+}), sulfate (SO_4^{2-}), total metals [aluminum (Al), antimony (Sb), arsenic (As), barium (Ba), lithium (Li), manganese (Mn), rubidium (Rb), and uranium (U)], distance from Giant Mine, distance from Con Mine, coring depth, and surface area were all log (base 10)-transformed and used for subsequent ordination analyses. Lake-water pH was not normally distributed in our dataset. Therefore, we back transformed the pH [$pH = -\log(H^{+})$] to the concentration of hydrogen ions (i.e., 10^{-pH}). Then the resulting values were square-root transformed to ensure the concentration of H^{+} was normally distributed (hereafter SQRT[H^{+}]). After applying the appropriate transformations for the 25 environmental variables, a Pearson correlation matrix was generated in the R Studio software environment using the Hmisc package (Harrell, 2018) to assess the covariance among the chemical and physical parameters.

Ordination analysis

First, we performed a principal component analysis (PCA) with the 25 environmental variables to assess the distribution of the 31 sites along the environmental gradients (Fig. 2) using the statistical program CANOCO version 5.0 (ter Braak & Šmilauer, 2012). The detrended correspondence analysis of the full

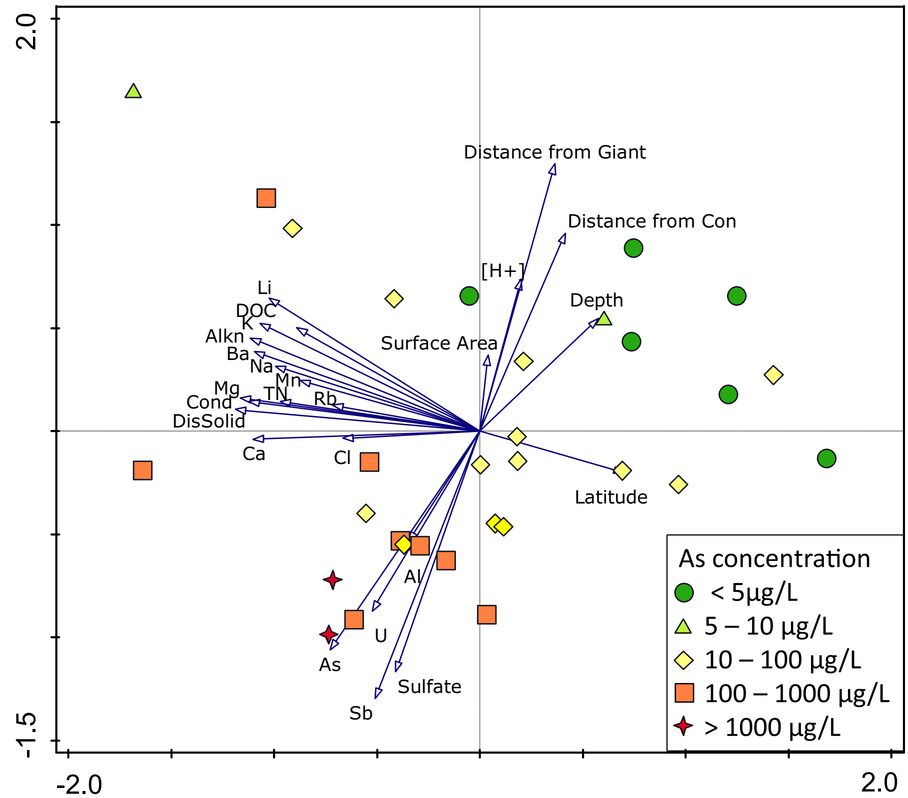


Fig. 2 The distribution of the 31 lakes along the gradients of the measured environmental variables using principal component analysis. The shapes of the symbols represent the concentration of arsenic in the water column

diatom species data revealed a gradient length > 2 standard deviation units, suggesting that a unimodal technique, such as canonical correspondence analysis (CCA), was a suitable method to explore the relationship between diatom species and environmental data (Birks, 2010).

A series of CCAs was run constrained to each environmental variable to assess if the selected variable explained a significant portion of the species data by examining the significance ($P < 0.05$) of the primary axes using 999 Monte Carlo permutation tests. Then we used the Pearson correlation matrix of the environmental variables in conjunction with the results from the 25 CCAs to reduce the number of explanatory environmental parameters by identifying groups of variables that were highly correlated (correlation coefficients > 0.6) and/or did not explain a significant portion of the variation in the diatom assemblages ($P > 0.05$). After removing the environmental variables that did not explain a significant portion of the species data, two groups of highly correlated variables were identified. Representative variables that explained the most variation in the diatom assemblage from each group were objectively selected using the forward selection option in CANOCO, following methods described by Blanchet et al. (2008). We elected to forward select the representative variables from each group to ensure the most parsimonious set of explanatory variables that were not correlated with each other were chosen objectively and included in the final CCA. However, these selected representatives were highly correlated to many variables within their groups and therefore they represent a group of explanatory variables in the CCA and the interpretations in the subsequent sections will be made within this context.

Additional statistical analysis

Analysis of similarities (ANOSIM; Clarke, 1993) was used to determine if the diatom assemblages differed significantly ($P < 0.05$) among three a priori defined groups ([As] > 100 µg/l, $n = 12$; 100 > [As] > 10 µg/l, $n = 13$; [As] < 10 µg/l, $n = 8$). The ANOSIM was based on the Bray–Curtis similarity matrix and the diatom assemblages were square-root transformed to stabilize the variance in the data using the statistical program PRIMER V6 (Clarke & Gorley, 2006). In ANOSIM, an R statistic of 0 suggests that the assemblages are not different among the two groups, while $R > 0$ (and closer to 1) suggests that there are differences in diatom assemblages among the two groups. However, R statistic can be less than 0 when there is higher variation in diatom assemblages within a group. Additionally, diatom species richness and Hill's N2 diversity for all 33 sites were calculated using the vegan (Oksanen et al., 2018) and rioja (Juggins, 2017) packages available for the R software environment (R Core Team, 2018). Prior to calculating richness and diversity, the diatom data from each of the sites were rarefied to a common sum of 500 diatom valves per sample.

Results

Limnological characteristics of lakes around Yellowknife

The study sites spanned relatively large physical and chemical limnological gradients (Supplement 1). For example, the surface areas and coring depths of the lakes ranged from 0.01 to 6.28 km^2 and 0.5 to 21 m, respectively (Supplement 1). Specifically, environmental parameters associated with gold mining operations, such as arsenic (range 1.5–2,780 µg/l, median 41.7 µg/l), antimony (range 0.1–49.8 µg/l, median 1.1 µg/l), and sulfate (range 1–2,920 mg/l, median 11 mg/l), had large gradients in this dataset (Supplement 1). The apparent large variation in the ionic strength of the water (e.g., calcium, chloride, sodium, specific conductivity) was influenced by the high values recorded at two lakes that were impacted by mine tailings (Keg and Peg lakes; Supplement 1). As mentioned previously, it was not possible to meet the assumption of normality of environmental variables related to ions due to the high values from Keg and Peg; therefore, these lakes were not included in subsequent ordination analyses.

Axes 1 and 2 of the PCA captured 62% of the total variation in the environmental data from the 31 lakes (Fig. 2). Axis 1 was primarily influenced by TN and DOC, as well as ionic composition (total alkalinity, specific conductivity, total dissolved solids, Ca^{2+}, Cl^-, Mg^{2+}, K^+, Na^+), while Axis 2 mainly reflected a gradient of physical properties (surface area and depth) and gold mining-related variables (distance from the mines, As, Sb, SO_4^{2-}) (Fig. 2). Many environmental variables in our 31-lake dataset were strongly correlated (correlation coefficients > 0.6) to each other, as identified by the Pearson correlation matrix (Supplement 2). Surface area of lakes and SQRT[H^+] were the only two variables that were not correlated to any other environmental parameters in this dataset. The PCA highlighted the strong relationships among the 25 environmental variables (Fig. 2) because highly correlated variables were grouped together (positively correlated) or distributed in opposite directions if negatively correlated. Chemical parameters, including nutrients, ionic composition, and some metals not related to mining activities in this area (e.g., Ba, Li, Mn, Rb), were positively correlated and plotted in the top left quadrant of the biplot (Fig. 2). The arrows representing environmental variables along Axis 1 and Axis 2 were perpendicular to each other suggesting minimal correlations among these parameters.

Generally, the concentrations of mining-related water chemistry parameters (As, Sb, SO_4^{2-}) declined with increasing distance from the mines (Figs. 1, 2) and were strongly negatively correlated (correlation coefficients > 0.6; Supplement 2). Furthermore, the PCA also captured the large [As] gradient of this dataset, as lakes with very low [As] were plotted in the top right quadrant, while high [As] lakes were plotted in the bottom left quadrant, and lakes with intermediate [As] were plotted between the two areas (Fig. 2). Although not strong, a negative correlation ($R = 0.54$) was observed between As and depth in this dataset. Due to logistical restraints, the water samples used in this study were collected from both ice-on and ice-free seasons and we acknowledge using samples from the same time window would have been preferable. Nonetheless, the patterns we observed in the water chemistry variables were consistent with previously published studies from the Yellowknife area (e.g., Palmer et al., 2015; Houben et al., 2016), which have also shown that the concentrations of contaminants associated with arsenopyrite ore processing (As, Sb, SO_4^{2-}) were higher in lakes closer to the mines.

Relationships between diatom assemblages and water chemistry parameters from arsenic-contaminated lakes around Yellowknife

Dominant diatom taxa in this 33-lake dataset included the planktonic *Discostella stelligera* (Cleve & Grunow) Houk and Klee/*Discostella pseudostelligera* (Hustedt) Houk & Klee complex, *Achnanthidium minutissimum* (Kützing) Czarnecki, and benthic fragilarioid species (*S. construens* var. *venter*, *S. pinnata*, *Staurosira construens* Ehrenberg, *Pseudostaurosira brevistriata* (Grunow) Williams & Round, *P. brevistriata* type) (Fig. 3). However, there were a few lakes where the diatom assemblages were dominated or co-dominated by *Stephanodiscus* taxa (Jackfish Lake), *Navicula cryptotenella* Lange-Bertalot (BC-18), *Denticula kuetzingii* Grunow (Range Lake), *Cocconeis placentula* Ehrenberg (Moose Lake), and *Fragilaria mesolepta* Rabenhorst (Niven Lake) (Fig. 3). Due to the high number of taxa in this dataset, we are only discussing some of the dominant species in subsequent sections.

Generally, in very shallow (< 1 m) and highly arsenic-contaminated lakes, the diatom assemblages were dominated by *A. minutissimum* while benthic fragilarioid taxa were dominant in shallow and deeper As-contaminated lakes (Fig. 4). In deep lakes where arsenic concentrations were lower than the guideline for the protection of aquatic life (< 5 µg/l), the assemblages were dominated by *the D. stelligera/ pseudostelligera* complex (Fig. 4). Furthermore, benthic diatom taxa were more common in arsenic-contaminated shallow and deep lakes, while planktonic taxa were dominant in deep lakes with very low arsenic concentrations in this dataset (Fig. 4). Clearly, there are some exceptions to these generalizations, because, as discussed later, the diatom assemblages in lakes around Yellowknife were also influenced by

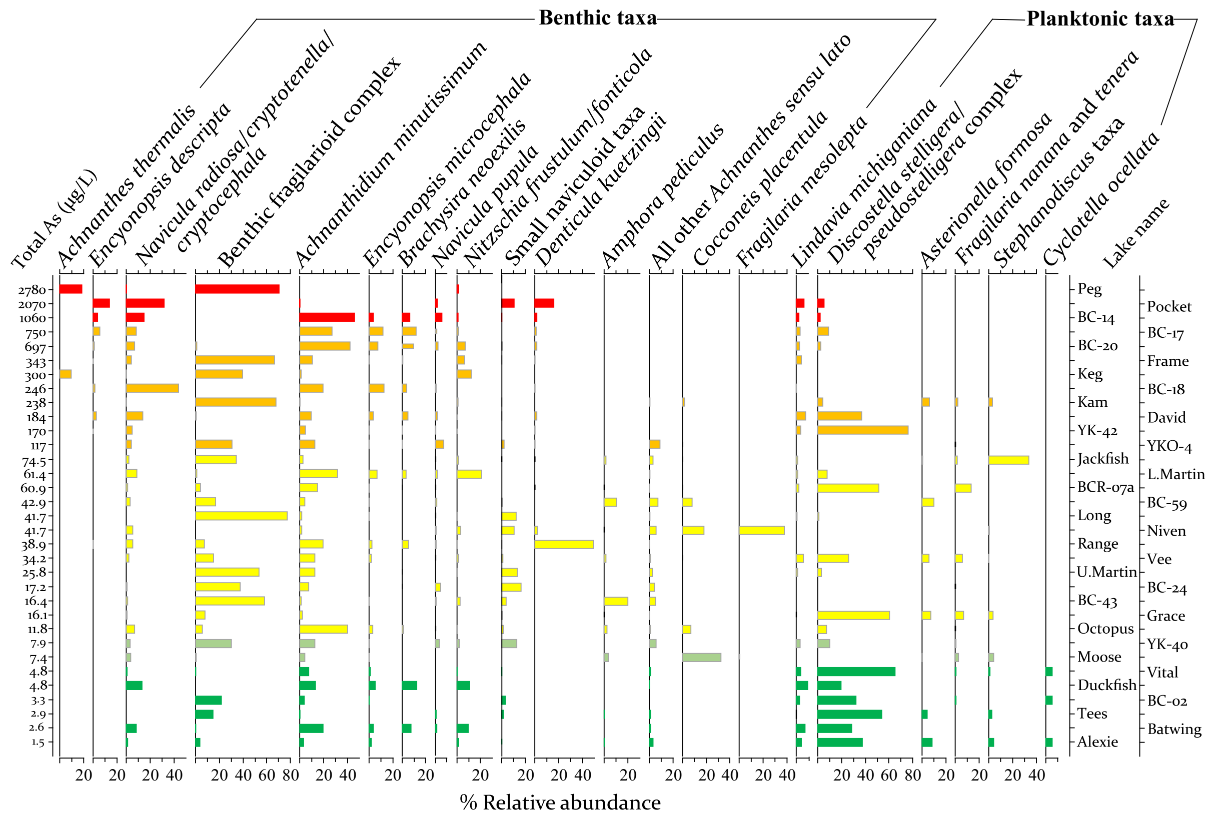


Fig. 3 Histograms of the dominant diatom taxa from the Yellowknife area. The sites are arranged according to As concentrations. The colors of the individual bars represent the varying arsenic concentrations at the site (red > 1000 µg/l, orange 100–1000 µg/l, yellow 10–100 µg/l, light green 5–10 µg/l, dark green < 5 µg/l)

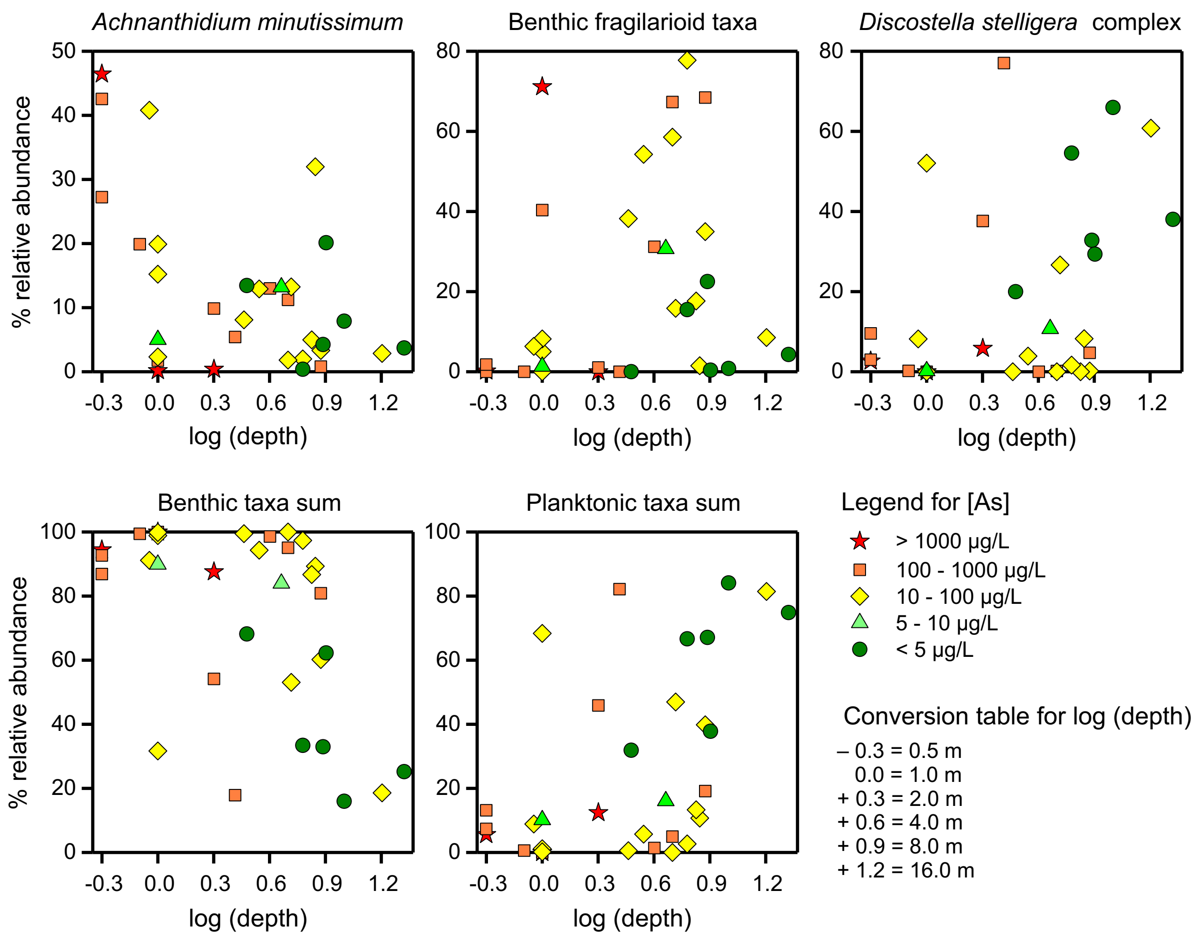


Fig. 4 Relationships between the relative abundances of *Achnanthidium minutissimum*, benthic fragilarioid taxa, the *Discostella stelligera* complex (*D. stelligera* and *D. pseudostelligera*), and sum of all planktonic and benthic taxa with log-transformed depth from lakes with varying arsenic concentrations. The shapes of the symbols represent the concentration of arsenic in the water column and a conversion table for the log depth is provided in the figure

other limnological variables related to nutrients and ions that may play an important role.

The diatom assemblage composition was diverse in this dataset (Fig. 3); however, the a priori divided groups (i.e., [As] > 100 µg/l, 100 > [As] > 10 µg/l, [As] < 10 µg/l) were not always significantly different from each other with respect to diatom distributions (Table 1). There were statistical differences in the diatom assemblages between lakes with [As] > 100 µg/l and 100 > [As] > 10 µg/l; however, these were not substantial (R = 0.159). The diatom species richness and diversity (Hill's N2) ranged from ~ 10 to ~ 47 and ~ 1.6 to ~ 13.4, respectively, and varied substantially across the 33 study lakes (Table 2). Additionally, the differences in species richness and diversity among lakes with varying [As] were subtle (Table 2). For example, the mean diatom species richness of lakes with > 100 µg/l of [As] was ~ 24, while the lakes with < 100 µg/l of [As] was ~ 30. Species diversity (Hill's N2) was marginally higher in lakes with < 10 µg/l of [As] (Table 2).

Most environmental variables were highly correlated ($R > 0.6$) in this dataset as identified by the Pearson correlation matrix (Supplement 2). A series of CCAs, constrained to each environmental variable individually, identified that the following 21 environmental variables explained a significant portion of the variations in the diatom assemblages (by examining the significance ($P < 0.05$) of the primary axes of CCAs), TN (9.3%), depth (8.2%), DOC (7.4%), Na^+ (7.1%), latitude (6.7%), Li (6.7%), Sb (6.6%), distance from Giant Mine (6.6%), Mg^{2+} (6.6%), As (6.4%), Rb

Table 1 Results of the analysis of similarity (ANOSIM) showing differences in diatom assemblages from the a priori groupings of lakes with varying arsenic concentrations (µg/l)

	[As] > 100 (n = 12)	100 > [As] > 10 (n = 13)	[As] < 10 (n = 8)
[As] > 100			
100 > [As] > 10	**0.159**		
[As] < 10	0.111	0.058	
Global R = **0.12**			

Bold values represent significant differences

Table 2 Results of the diatom species richness and diversity (Hill's N2) from the a priori groupings of lakes with varying arsenic concentrations (µg/l)

	Species richness				Species diversity			
	Min	Max	Mean	Median	Min	Max	Mean	Median
[As] > 100 (n = 12)	10.4	43	23.5	21.9	1.6	9.1	5.1	4.8
100 > [As] > 10 (n = 13)	16.9	47	30.5	29.8	2.1	13.4	5.5	5.0
[As] < 10 (n = 8)	22	41.8	30.1	29.3	2.2	9.8	5.9	6.3

(6.4%), total dissolved solids (6.3%), Ba (6.2%), total alkalinity (6.1%), Cl^- (6.1%), U (6.1%), specific conductivity (6%), K^+ (6%), Mn (5.3%), Ca^{2+} (5%), and SO_4^{2-} (4.9%). Only four environmental variables (i.e., Al, distance from Con Mine, surface area, SQRT[H^+]) did not explain a significant portion of the species data when they were the sole explanatory variable and therefore removed from subsequent analyses.

The environmental variables that explained significant amounts of variation in the diatom assemblages clustered into two groups of correlated variables (i.e., each variable was correlated with one or more variables within the group). Generally, variables related to ions (Ca^{2+}, Mg^{2+}, K^+, Na^+, Cl^-, specific conductivity, total alkalinity, total dissolved solids), TN, and DOC were correlated with some metals (Ba, Li, Mn, Rb), latitude, and depth. Furthermore, environmental variables related to mining (As, Sb, SO_4^{2-}, distance from Giant Mine) were correlated with each other and with U. The forward selection procedure in CANOCO identified three environmental variables to represent the two groups. TN and Na^+ were chosen from the nutrient and ion group to represent a suite of variables mentioned above and Sb was chosen from the mining-related variables. We note that the selected variables (TN, Na^+, Sb) represent a group of variables and subsequent interpretations should be viewed in this context. For example, when we use Sb as a representative variable to track industrial pollution, it allows us to track the potential influence of all the correlated mining parameters, including As.

The three representative environmental variables (TN, Na^+, and Sb) were then used to perform the final CCA that explained $\sim$ 22.9% of the variation in the diatom assemblages when constrained to the first three axes of the CCA. Furthermore, the variance inflation factors for these three variables were very low and ranged between 1.1 and 1.4, suggesting that correlations among these variables were near negligible. Conventionally, diatom–environment relationships are presented in two-dimensional figures by examining CCA Axis 1 versus Axis 2 (Fig. 5a) scores or Axis 1 versus Axis 3 (Fig. 5b). However, here we also display the results of the CCA by plotting Axis 1, Axis 2, and Axis 3 samples scores within the same ordination space (Fig. 5c) to examine if the underlying patterns in the distribution of sites could be presented more clearly. In Fig. 5a (Axis 1 vs. Axis 2), two sites with very low [As] plotted with sites where relatively high [As] were present (in the right quadrant along Axis 1). However, when Axis 1 and Axis 3 were

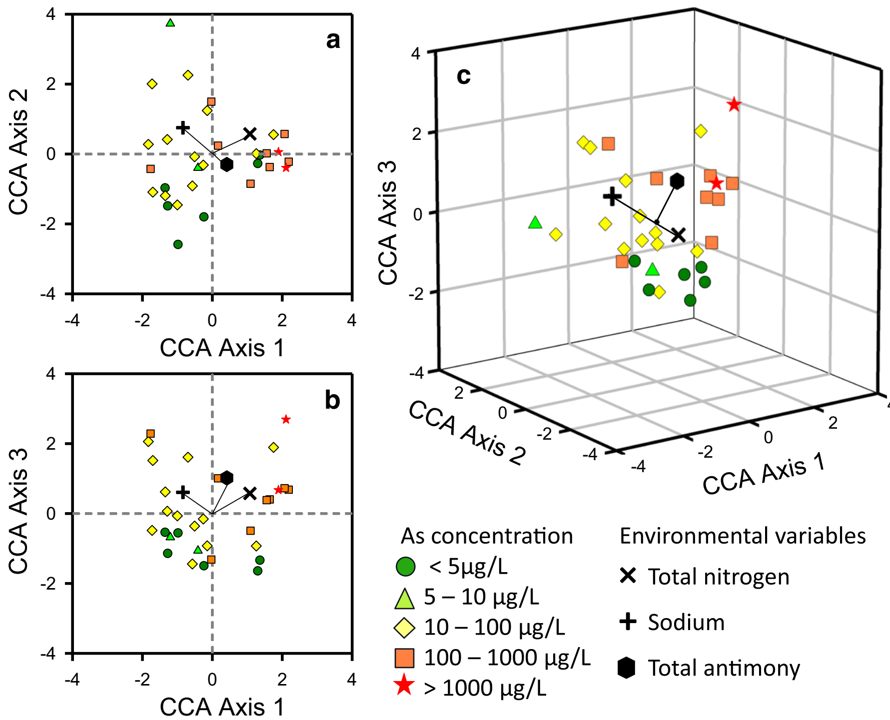


Fig. 5 Canonical correspondence analysis (CCA) showing the distribution of sites with total nitrogen (TN), sodium (Na^+), and antimony (Sb) in the ordination space. CCA Axis 1 $\lambda = 0.33$, CCA Axis 2 $\lambda = 0.20$, CCA Axis 3 $\lambda = 0.11$

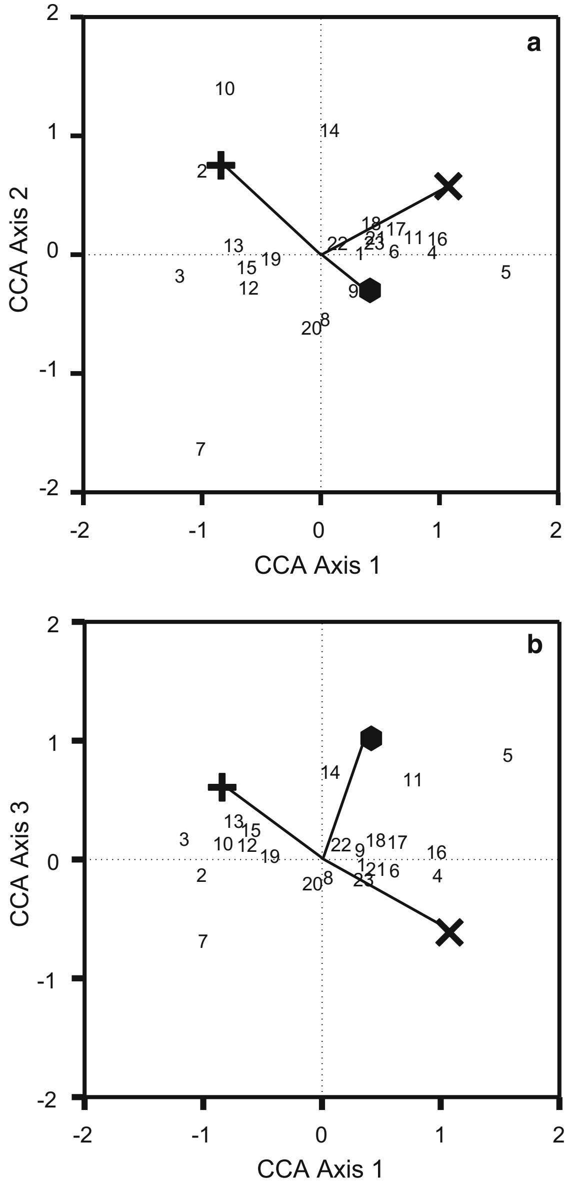
plotted within the same ordination space (Fig. 5b), sites with similar [As] generally clustered together. Similarly, in our three-dimensional CCA plot (Fig. 5c), sites with similar [As] grouped around each other. Therefore, Axis 3 is likely tracking the gradient of mining pollution, while Axes 1 and 2 are primarily tracking the influences of variables related to nutrients and ions. In addition to the separation of sites along Axis 3 of the CCA, lakes with relatively high [As] plotted in the same direction as Sb (representative variable for mining pollution) in Fig. 5b, c, suggesting that the inclusion of the third axis was necessary to appropriately present the distribution of sites in this dataset.

Due to the high number of diatom taxa present in this dataset, we only plotted the sample scores of the 23 most common diatom taxa (present in at least two samples in $> 5\%$ abundance). Similar to the CCA biplot of the site scores (Fig. 5), the plot with Axis 1 versus Axis 3 was the most informative in exploring the distribution of diatom taxa along a gradient of mining pollution (Fig. 6b). Benthic *D. kuetzingii* (11), *Encyonopsis descripta* (Hustedt) Krammer (5), and *P. brevistriata* type (14) plotted in the upper right quadrant, while *A. minutissimum* (1), *Brachysira neoexilis* Lange-Bertalot (4), *Encyonopsis microcephala* (Grunow) Krammer (6), *Navicula radiosa* Kützing (16), *N. cryptotenella* (17), *Navicula cryptocephala* Kützing (18), *Navicula pupula* Kützing (21), *Nitzschia frustulum* (Kützing) Grunow (22), and *Nitzschia fonticola* Grunow (23) plotted in the right quadrant and around the center of the biplot (Fig. 6b).

Discussion

Limnology of Yellowknife lakes

The range of arsenic concentrations reported in this study [range 1.5–2,780 μg/l (mean = 287 μg/l, median = 41.7 μg/l)] as well as from previous limnological assessments from the Yellowknife area (e.g., Palmer et al., 2015; Houben et al., 2016) represents one of the largest gradients of lake-water arsenic concentrations in Canada and likely around the world. The strong negative correlations between distance from Giant Mine and arsenic ($R = -0.82$), and antimony ($R = -0.86$) are consistent with the observations by Palmer et al. (2015) and Houben et al. (2016), who have shown that the exceptionally high



◀**Fig. 6** Canonical correspondence analysis (CCA) showing the distribution of the most common species (> 5% in at least two samples) with total nitrogen (×), sodium (plus), and antimony (hexagon) in the ordination space. CCA Axis 1 $\lambda = 0.33$, CCA Axis 2 $\lambda = 0.20$, CCA Axis 3 $\lambda = 0.11$. The diatom codes are as follows: (1) *Achnanthidium minutissimum*, (2) *Amphora pediculus*, (3) *Asterionella formosa*, (4) *Brachysira neoexilis*, (5) *Encyonopsis descripta*, (6) *Encyonopsis microcephala*, (7) *Cyclotella ocellata*, (8) *Discostella stelligera/pseudostelligera* complex (9) *Lindavia michiganiana*, (10) *Cocconeis placentula*, (11) *Denticula kuetzingii*, (12) *Fragilaria nanana*, (13) *Pseudostaurosira brevistriata*, (14) *Pseudostaurosira brevistriata* type, (15) *S. pinnata–S. construens* var. *venter* complex, (16) *Navicula radiosa*, (17) *Navicula cryptotenella*, (18) *Navicula cryptocephala*, (19) *Navicula minima*, (20) *Navicula seminulum*, (21) *Navicula pupula*, (22) *Nitzschia frustulum*, (23) *Nitzschia fonticola*

concentrations of these contaminants in the lakes around Yellowknife were a result of arsenopyrite ore-processing activities at Giant Mine.

Other chemical variables also had large environmental gradients; however, this was often influenced by two lakes (Keg and Peg). Keg and Peg lakes are part of a chain of lakes that begin with Pud Lake near Con Mine. Therefore, when Pud Lake received mine tailings from Con Mine, the mine effluent migrated to the downstream lakes (Pud–Meg–Keg–Peg) via connecting streams (Bright et al., 1994). Mining activities ceased at Con Mine in 2004 (Silke, 2009); however, the concentrations of ionic compounds associated with tailings (specific conductivity, total dissolved solids, Ca^{2+}, Cl^{-}, Mg^{2+}, Na^{+}, SO_4^{2-}) were exceptionally high when these lakes were sampled in 2014 (Supplement 1). Even though mining activities ended around the turn of the twenty-first century in the Yellowknife area, the high concentrations of mining-related contaminants (e.g., As and Sb) in many lakes in the region and ionic compounds at Keg and Peg suggest that these subarctic lakes are far from reaching any state of chemical recovery.

Diatom assemblages from arsenic-contaminated lakes around Yellowknife

The diatom assemblage composition was diverse in our dataset, and the lack of a clear separation in assemblage composition (Table 1) nor species richness and diversity among the a priori groups of lakes is likely a result of the high variability within each group. The mean Hill's N2 diversity of 5.4 in this study was lower than the means (range 6.72–13.88) reported from other diatom surveys from the Canadian Arctic (e.g., Michelutti et al., 2003; Antoniades et al., 2005; Keatley et al., 2009). Recently, Barral-Farga et al. (2016) noted low diatom species richness in biofilms exposed to As in a short-term (13 days) laboratory-based study. Similarly, multi-year limnological surveys of lakes from Canada (Austin et al., 1985) and a paleolimnological analysis of a lake in Finland (Leppänen et al., 2017) have also shown that diatom species richness and diversity generally decreased with metal input. However, no changes in the diversity and richness were observed in a diatom survey of streams impacted by metal pollution in Cornwall and Wales, UK (Hirst et al., 2002). Furthermore, diatom species diversity did not significantly differ between biofilms exposed to different As concentrations in Anllóns River, a system impacted by historical mining activities (Barral-Fraga et al., 2018). Based on our data, diatom species richness and diversity may not always clearly respond to metal contamination, and this is likely due to site specific limnological factors such as top-down biotic interactions and other abiotic factors including physio-chemical parameters. Furthermore, paleolimnological investigations from northern latitudes have provided evidence that diatom species diversity is partly determined by changes in ice-cover and length of the growing season (e.g., Griffiths et al., 2017). Therefore, subarctic lakes in the Yellowknife area, where the lakes are ice-covered for most of the year (196–237 days), changes in ice phenology may be influencing the algal diversity strongly and the response to metal pollution could be muted.

Although diatom teratology and size reduction have previously been reported from freshwaters affected by metal pollution (e.g., Ruggiu et al., 1998; Cattaneo et al., 2004; Morin et al., 2008; Tolotti et al., 2019), we only observed teratological forms in one lake (Pocket Lake) and in very low abundances. The general absence of teratological forms has been recorded in other aquatic ecosystems affected by industrial activities (e.g., Hamilton et al., 2015; Thienpont et al., 2016). The key drivers of teratology are not completely known. However, in contrast to morphological deformities of diatoms, a more consistent measure of metal contamination includes shifts in the community structure of diatom assemblages (Morin et al., 2012). For example, paleolimnological assessments of metal-contaminated systems have reported a shift from assemblages dominated by planktonic diatoms to benthic forms when metal load increased (e.g., Salonen et al., 2006; Cattaneo et al., 2008; Thienpont et al., 2016). Similarly, in our study, the diatom assemblages were generally dominated by benthic fragilarioid taxa and *A. minutissimum* in the shallow and deeper lakes that exceeded the concentration of As for the protection of aquatic life (i.e., [As] > 5 μg/l; Figs. 3, 4). Coincidentally, the lakes where As concentrations were below the levels set for the protection of aquatic life were also lakes that were generally deeper, and the planktonic *D. stelligera* complex often dominated these assemblages (Figs. 3, 4). However, there were exceptions to this generalization (i.e., *D. stelligera* was dominant in David, YK-42, BCR-07A, Vee, and Grace lakes, where [As] were greater than 5 μg/l) and these will be discussed more fully below.

The presence and/or dominance of a variety of benthic taxa in our dataset (*A. minutissimum*, small fragilarioid taxa, *N. radiosa*, *N. cryptotenella*, *N. cryptocephala*, *E. descripta*, *D. kuetzingii*, *Amphora pediculus* Grunow, and small naviculoid taxa) is consistent with previous diatom surveys of subarctic lakes from Canada (e.g., Pienitz & Smol, 1993; Rühland & Smol, 2002) and elsewhere (e.g., Weckström et al., 1997; Karst-Riddoch et al., 2009). However, some of these benthic taxa have also been reported from metal-contaminated systems around the world. For example, *A. minutissimum*, an opportunistic taxon, has been observed in many freshwaters with exceptionally high metalloid concentrations (Ruggiu et al., 1998; Ivorra et al., 2000; Szabó et al., 2005; Salonen et al., 2006), including a heavily As-contaminated lake in China (Chen et al., 2015). Furthermore, laboratory-based experiments have also shown that *A. minutissimum* can tolerate high [As] (e.g., Rodriguez Castro et al., 2015; Barral-Farga et al., 2016). In this dataset, *D. kuetzingii* and *E. descripta* were only present in lakes where As concentrations were well above the guideline for the protection of aquatic life (i.e., 5 μg/l) suggesting these taxa may also tolerate elevated As concentrations. A previous, detailed paleolimnological investigation of one of the most As-contaminated lake in our study (Pocket Lake: [As] = 2,070 μg/l) recorded an increase in benthic naviculoid taxa (*N. radiosa*, *N. cryptotenella*, and *N. cryptocephala*) and *D. kuetzingii* at the expense of planktonic *D. stelligera* when Giant Mine was in operation (Thienpont et al., 2016). Furthermore, benthic fragilarioid taxa were very common in many lakes where [As] were greater than 5 μg/l. Although benthic fragilarioid taxa have not been commonly associated with metal pollution, our study provides some indication that these opportunistic taxa, which are ubiquitously present in many High Arctic and subarctic environments, may tolerate high arsenic concentrations in lakes around Yellowknife.

Previous diatom surveys of metal-contaminated lakes have reported that planktonic cyclotelloid taxa (including the *D. stelligera/pseudostelligera* complex) and other planktonic forms are sensitive to metal pollution (e.g., Ruggiu et al., 1998; Cattaneo et al., 2008). In our dataset, *Cyclotella ocellata* Pantocsek, an oligotrophic planktonic diatom, was exclusively observed in deeper lakes (range 7.7–21 m) with [As] < 5 µg/l, suggesting that this taxon may be sensitive to As contamination in this region. Furthermore, as mentioned previously, at Pocket Lake, the *D. stelligera/pseudostelligera* complex was replaced by benthic taxa in response to severe As contamination (Thienpont et al., 2016). In our survey, the *D. stelligera/pseudostelligera* complex was dominant in deep lakes with [As] < 5 µg/l; however, its dominance in 5 lakes with relatively high [As] (David 184 µg/l, YK-42 170 µg/l, BCR-07A 61 µg/l, Vee 34 µg/l, Grace 16 µg/l) may suggest that multiple environmental factors may be influencing its distribution in this region. Generally, the *D. stelligera/pseudostelligera* complex has been associated with deeper lakes in the Canadian subarctic (Rühland et al., 2003a) and the Siberian Arctic (Laing et al., 1999); similarly, in our study, this species complex and a few other planktonic taxa occurred primarily in deep lakes (Fig. 4). However, these planktonic taxa were also present in shallow lakes in this survey, including some of the highly As-contaminated lakes (e.g., 77% relative abundance in YK-42: depth 2.6 m, [As] 170 µg/l; Fig. 4). Furthermore, the relative abundances of the *D. stelligera/pseudostelligera* complex have increased in subarctic lakes as a result of recent warming (e.g., Rühland et al., 2003b, 2008, 2015; Rühland & Smol, 2005); therefore, the high relative abundances of this taxon in some of the As-contaminated lakes may also be a recent response to changing climatic conditions such as longer growing season and stronger thermal stratification. Detailed diatom-based paleolimnological analyses of lakes in the region are necessary to better understand the underlying mechanisms that are leading to the high abundances of *D. stelligera/pseudostelligera* complex in lakes where [As] > 5 µg/l.

At moderately As-contaminated Jackfish and Niven lakes ([As] at Jackfish = $\sim$ 75 µg/l and Niven = 42 µg/l), *Stephanodiscus* taxa (mostly *Stephanodiscus medius-minutulus* type) and *F. mesolepta* dominated the assemblages, respectively. These taxa have been associated with nutrient enrichment in many previous diatom assessments from the Arctic (e.g., Moser et al., 2002) and elsewhere (e.g., Tropea et al., 2011; Nelligan et al., 2016). In addition to receiving As via atmospheric emissions, these lakes have also been impacted by local land-use changes. For example, Niven Lake was Yellowknife's first sewage lagoon from 1948 to 1981, and in a detailed paleolimnological assessment of this lake, Stewart et al. (2018) noted that eutrophic *Stephanodiscus* taxa increased subtly at the time of raw sewage input. However, the eutrophic *F. mesolepta* increased to dominance after $\sim$ 1990 (i.e., well after the cessation of sewage inputs), suggesting that Niven Lake may be subjected to internal nutrient loading, as this shallow lake currently undergoes hypolimnetic anoxia during summer months.

Similar to the diatom assemblages from many other As-contaminated lakes in this study, the assemblage composition of the two lakes that received mine tailings from the abandoned Con Mine (Keg and Peg lakes) were also dominated by benthic fragilarioid taxa (specifically *P. brevistriata* type). However, notable abundances of *Achnanthes thermalis* (Rabenhorst) Schoenfeld and *Navicula incerta* Lange-Bertalot were also observed in these lakes. Although rare, these taxa have been noted in saline environments (e.g., Gell et al., 2002) or during periods of high salinity (e.g., Last et al., 1998). Therefore, it is not surprising that these two taxa were present, exclusively in the two lakes that had the highest concentrations for total dissolved solids, specific conductivity, sulfate, aluminum, and other ionic compounds (Ca^{2+}, Cl^{-}, Mg^{2+}, K^{+}, Na^{+}) in our dataset.

The influence of multiple environmental variables on diatom assemblages in Yellowknife lakes

The proportion of variation explained by the environmental variables (TN, Na^{+}, and Sb) was relatively low ($\sim$ 22.9%), but similar to those reported from other Arctic and subarctic regions (e.g., Michelutti et al., 2006; Antoniades et al., 2009). The relatively low explained variation suggests that other environmental variables not measured (e.g., top-down biotic interactions, ice phenology) may also be important determinants of diatom assemblages in lakes around Yellowknife. TN and Na^{+} explained the highest amount of variation in the diatom assemblages in the final CCA (Fig. 5) and it is consistent with previous diatom-based limnological surveys from high latitudes (e.g., Douglas & Smol, 1995; Weckström & Korhola, 2001; Rühland & Smol, 2002; Tammelin et al., 2017). However, following TN and Na^+, Sb also explained a significant portion of the variation, suggesting that environmental variables related to mining pollution also influenced the diatom assemblages of lakes around Yellowknife.

Benthic taxa that were exclusively present in lakes with > 10 µg/l of As (i.e., *D. kuetzingii*, *E. descripta*, and *P. brevistriata* type) plotted in the upper right quadrant where Sb was also high (Fig. 6b), suggesting that these taxa are able to tolerate high metal pollution. Meanwhile, planktonic *C. ocellata* plotted in the lower left quadrant, furthest away from Sb (Fig. 6b), indicating that this taxon is very sensitive to metal contamination. Furthermore, benthic taxa such as *A. minutissimum*, *B. neoexilis*, *E. microcephala*, *N. radiosa*, *N. cryptotenella*, *N. cryptocephala*, *N. pupula*, *N. frustulum*, and *N. fonticola* have been observed in other subarctic lakes where TN and DOC concentrations were generally high and they have relatively high optima for these parameters (Pienitz & Smol, 1993; Rühland & Smol, 2002). In the Yellowknife area, these taxa may be influenced by environmental variables related to nutrients and mining pollution as they plotted along Axis 1 in the right quadrant and in between TN and Sb (Fig. 6b). Small benthic taxa such as *P. brevistriata*, *S. pinnata–S. construens* var. *venter* complex, and *Navicula minima* Grunow plotted along the first axis in the left quadrant and near Na^+ (Fig. 6b), indicating these taxa may be strongly influenced by ionic composition. This is consistent with a previous survey of diatoms from subarctic lakes in forested catchments where small benthic fragilarioid and naviculoid taxa were generally present in alkaline lakes where ionic concentrations were higher (Pienitz & Smol, 1993). The *D. stelligera/pseudostelligera* complex plotted close to the center of the biplot (Fig. 6b) suggesting that this taxon complex has a broad distribution in this dataset and occurs across a wide range of lakes around Yellowknife.

The CCA highlights that the diatom assemblages in the subarctic lakes around Yellowknife are structured by multiple limnological parameters, with mining pollutants playing a limited role. This is in contrast to the findings of surface sediment diatom surveys from metal-contaminated lakes in temperate regions, such as Sudbury (Ontario), where relationships were identified between metal concentrations in the water column and surface sediment diatom assemblages that led to the development and application of inference models to reconstruct past metal concentrations (Dixit et al., 1991, 1992). However, in the Sudbury region, heavy metal contamination was often accompanied by sharp decreases in pH (Dixit et al., 1992), whereas lakes around Yellowknife were not acidified due to the presence of carbonates that provided the necessary buffering capacity (Jamieson, 2014). Our results are similar to those reported from Arctic lakes near Norilsk smelters in Russia, where diatom assemblages were dominated by opportunistic benthic fragilarioid taxa and were not severely affected or altered by high metal contamination (Laing et al., 1999; Michelutti et al., 2001). The almost identical response of algal assemblages from lakes around Yellowknife and Norilsk is likely due to two reasons: first, these regions experience similar climatic conditions; and second, the underlying geology prevented acidification of these lakes. The role of local climatic conditions on diatom assemblages of lakes around the circumpolar Arctic has been investigated extensively, and many studies have concluded that the cold conditions and relatively long ice-cover lead to assemblages dominated by benthic diatoms, specifically higher abundances of opportunistic taxa, such as small benthic fragilarioids (Smol, 1988). Therefore, the responses of the diatom flora to metal contamination are likely partly influenced by local climatic conditions and catchment characteristics in northern latitudes.

Although some overall patterns are apparent, our diatom survey highlights some of the complexities of the biological responses to stressors linked to mining activities in climatically sensitive subarctic lakes. Since mining activities continue to increase in subarctic and Arctic regions (Northwest Territories Industry, Tourism and Investment, 2014), a better understanding of how key ecological indicators, such as diatoms, respond to mining-related stressors will be required. Furthermore, spatial surveys, such as this study, will be necessary to place paleolimnological assessments into appropriate environmental and historical contexts.

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