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ORIGINAL PAPER

Regional gold mining activities and recent climate warming alter diatom assemblages in deep sub-Arctic lakes

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Abstract

Previous paleolimnological investigations have examined the effects of gold mining operations, local land-use changes, and regional climate warming on the aquatic biota from shallow lakes in Yellowknife, Northwest Territories, Canada. However, the long-term impacts of these multiple environmental stressors on the biota of deeper lakes that support large-bodied fish species have not been investigated. In this study, we examined multiple sedimentary proxies from two deep lakes around Yellowknife to assess the long-term effects of metalloid contamination, development of the city, and recent warming over the past ~ 200 years. The sedimentary metalloid profiles tracked the influence of mining operations and local land-use changes in the Yellowknife area and there were some similarities in the diatom responses to multiple stressors across the two lakes. However, the increases in sedimentary metalloid concentrations, eutrophic diatom taxa, and whole-lake primary production were more pronounced at Grace Lake relative to Alexie, likely because Grace is located nearer to the gold mines, as well as local city development. The overall lake primary production and the relative abundance of the planktonic diatom *Discostella stelligera* increased at both sites suggesting that some of the biological changes may be influenced by changes in thermal stratification, as has been documented in a wide spectrum of lakes across the Northern Hemisphere. Furthermore, the diatom assemblage changes in these deep lakes differed from those observed from shallow lakes in the region, suggesting that site-specific limnological characteristics will influence the biological responses to multiple environmental stressors through time.

Keywords Paleolimnology · Contaminants · Arsenic · Land-use changes · Algae · Yellowknife · Northwest Territories

Introduction

The acceleration of mining operations to meet the global demand for metals after the Industrial Revolution and the two World Wars has led to the contamination of many aquatic ecosystems globally (e.g. Wong et al. 1999). For example, in Canada the exploration and extraction of precious metals such as gold and silver increased rapidly during the twentieth century and eventually led to the metalloid contamination of surrounding ecosystems (e.g. Sprague et al. 2016). The consequences of metalloid contamination on aquatic ecosystems can be catastrophic as biota from multiple trophic levels may be affected (e.g. Chen et al. 2015; Leppänen et al. 2017). A notable example of twentieth century metal mining operations and subsequent environmental damage occurred around the City of Yellowknife in the Northwest Territories, Canada, where gold mining operations led to the arsenic (As), antimony (Sb), and sulphate (SO_4^{2-}) contamination of numerous sub-Arctic lakes (Palmer et al. 2015; Houben et al. 2016). In addition to mining pollution, lakes within the City of Yellowknife were also affected by land clearance, establishment and growth of the city, and other municipal activities (e.g. Dirszowsky and Wilson 2016; Gavel et al. 2018). Furthermore, similar to many northern regions, Yellowknife has also undergone marked increases in air temperature (e.g. Mullan et al. 2017). The aquatic biological responses to the interactive effects of anthropogenic

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activities in watersheds and climate can be complex (Smol 2010) and have led to limnological surprises in this area. For instance, at Yellowknife's first sewage lagoon, Niven Lake, the legacy of eutrophication was exasperated by warmer temperatures in recent decades (Stewart et al. 2018). Therefore, understanding the cumulative impacts on aquatic organisms, within the context of multiple stressors, is necessary to determine appropriate remediation plans and postulate realistic recovery targets for lakes around Yellowknife and other Northern regions.

Previous and on-going paleolimnological research around Yellowknife have provided the much-needed long-term perspectives on the cumulative effects of multiple stressors (e.g. Dirszowsky and Wilson 2016; Thienpont et al. 2016; Gavel et al. 2018; Stewart et al. 2018). However, most of these studies have focused on shallow lakes (< 10-m maximum depth) as these are the dominant water bodies in this region (Palmer et al. 2015). While these paleolimnological studies have been informative, the cumulative long-term effects of multiple stressors on the biota of deeper (> 10-m maximum depth) sub-Arctic lakes, which often support important fisheries, have not been extensively investigated in this region. Indeed, the deep sub-Arctic lakes around Yellowknife contain suitable habitat to support large-bodied and cold-water fish populations (e.g. lake trout—*Salvelinus namaycush*) that have cultural significance to local indigenous peoples and play an important role in regional economic activities (e.g. sports fisheries, tourism) (Healey and Woodall 1973; Stewart 1997). Meanwhile, shallow lakes in the region do not support similar fish species as they do not have suitable thermal habitats and/or were affected by anthropogenic activities, or simply freeze to the bottom in winter (e.g. Stewart 1997; Gavel et al. 2018). Recently, land-use changes associated with industrial development has been identified as a potential threat to the habitat of large-bodied and late-maturing fish species in Canada's North (Cott et al. 2015). For example, increased human activities, such as highway constructions, logging, contamination, and fishing pressure, can negatively impact the habitat quality and population sizes of fish species (Cott et al. 2015). Furthermore, investigations of temperate lakes have indicated that the depletion of hypolimnetic dissolved oxygen, often induced by anthropogenic activities and subsequent eutrophication, is an important environmental stressor affecting aquatic habitats that support large-bodied fish species (Evans et al. 1996). Recent climate warming has also been suggested as an emerging stressor altering the habitats and phenology of cold-water fish species in temperate lakes (Guzzo and Blanchfield 2017). Whilst previous paleolimnological studies have assessed the cumulative impacts of land-use changes and climate warming on deep lakes that support large-bodied, cold-water fish species in temperate regions (e.g. Nelligan et al. 2016), relatively little is known about the long-term impacts of multiple stressors on their sub-Arctic counterparts.

In this paleolimnological investigation, we examined dated sediment records from two deep sub-Arctic lakes (> 10 m) in the Yellowknife area that support large-bodied fish species. Since we were interested in comparing the effects of multiple environmental stressors, we selected one lake near the City of Yellowknife and the mines (Grace Lake) and another lake that is ~ 20 km away from these activities (Alexie Lake). We used sedimentary metalloid analysis to track the history of metal pollution, while subfossil diatom (Bacillariophyceae) assemblages and visible reflectance of chlorophyll-*a* from the sediments were examined to infer the effects of multiple environmental stressors. In this study, we specifically selected algal-based paleolimnological techniques to track the history of eutrophication and impacts of recent warming on the limnology of these sub-Arctic lakes as these proxies have been used extensively to investigate the consequences of nutrient enrichment and climatic changes on temperate lakes that support large-bodied fish species (e.g. Nelligan et al. 2016). However, future investigations of zooplankton-based paleolimnological studies can provide more insights on the long-term changes in the fish communities of these lakes.

Study area

The gold mining activities in Yellowknife commenced around the late-1930s but were briefly interrupted by World War II and resumed soon after the war ended when an increased labour force became available. The gold mining operations lasted for over five decades at Giant (Giant Yellowknife Gold Mines Limited; 1948–2004) and Con (Consolidated Mining and Smelting Company; 1938–2003) mines (Wolfe 1998; Silke 2009). When the gold-bearing arsenopyrite and pyrite ores around Yellowknife were roasted, large quantities of As-bearing iron oxides and sulfur dioxide were released into the environment via roaster stacks (Coleman 1957; Walker et al. 2005; INAC 2007; Palmer et al. 2015). The As vapour precipitated into solid-phase arsenic trioxide (As_2O_3) in the atmosphere and was deposited on the terrestrial and aquatic ecosystems around Yellowknife (Walker et al. 2005; Schuh et al. 2018; Van Den Berghe et al. 2018). It has been estimated that ~ 20,000 tonnes of As_2O_3 were introduced into the surrounding environment from gold mining activities at Giant between 1948 and 1999 with greater amounts being released during the early years (1948–1951) (summarized by Jamieson 2014). As a mitigation procedure, an electrostatic precipitator was installed in 1951 that dramatically reduced the amount of As_2O_3 entering the environment through roaster stacks (~ 5.5 vs. 1.5 tonnes per day in 1952 and 1958, respectively) (INAC 2007; Jamieson 2014). Further reductions in As_2O_3

emissions became possible by the construction of a baghouse and underground storage facilities in 1951 (INAC 2007; Jamieson 2014). Cumulatively, these emission controls substantially reduced the amount of As_2O_3 that entered the environment between 1959 and 1999 (varied between 0.01 and 0.9 tonnes per day). Nonetheless, recent surveys of many lakes in the region recorded As concentrations that exceeded the suggested guidelines for the protection of aquatic life (< 5 μg L^{-1}) and drinking water (< 10 μg L^{-1}) quality (Palmer et al. 2015).

Grace and Alexie lakes are located within the Slave Structural Province of the Canadian Shield. Grace Lake is underlain by granitoid bedrock, while Alexie lies in the transition zone of granitoid and metasedimentary rock formations (Henderson 1985). Both lakes are relatively deep (Alexie—21 m; Grace—16 m) and have very similar pH and alkalinity, however Alexie is substantially larger than Grace (surface area of Alexie is ~ 4.56-km^2, whilst Grace is ~ 0.63 km^2) (Table 1), hence there are likely differences in the volume of these two lakes. Alexie has an inlet on the northwest corner that connects it to Chitty Lake and has an outflow on the southeast corner. Grace Lake has an inlet on the eastern part of the lake and an outflow on the west that connects it to Kam Lake. The predominant wind direction in the Yellowknife area is from east to west and Alexie is located to the northeast of Giant and Con mines, while Grace is located to the south of the mines (Fig. 1). The present-day contaminant concentrations in the water are higher at Grace, as this site is closer to the mining activities and the city (Table 1 and Fig. 1). For example, the present As concentration in the water column at Grace (16 μg L^{-1}) is ~ 10 times higher than Alexie (1.5 μg L^{-1}), and exceeds the guidelines for the protection of aquatic life and drinking water quality.

Table 1 Limnological and environmental information collected for Grace and Alexie lakes

Limnological parameter	Grace	Alexie
Latitude	62° 40′ 35.32″ N	62° 25′ 14.44″ N
Longitude	114° 06′ 05.77″ W	114° 25′ 51.83″ W
Coring depth (m)	16	21
Surface area (km^2)	0.63	4.56
Distance from Giant Mine (km)	10.23	23.52
Distance from Con Mine (km)	3.66	29.97
Total As (μg L^{-1})	16.1	1.5
Total Sb (μg L^{-1})	0.6	< 0.1
Sulphate (mg L^{-1})	15	4
pH	7.42	7.69
Alkalinity (mg L^{-1})	59.4	59.2
Total nitrogen (mg L^{-1})	0.75	0.51

Total arsenic (Total As), total antimony (Total Sb), sulphate, pH, alkalinity, and total nitrogen were single point measurements from April 2014

Grace Lake experienced substantial anthropogenic disturbances during the twentieth and twenty-first centuries as it is located within the City of Yellowknife and near Con Mine. Specifically, development around Grace Lake occurred as early as 1937 with the construction of the Con Mine (~ 3.7 km away), and a road was built over the outflow from Grace Lake to Kam Lake between 1937 and 1950 (Fig. 2). Furthermore, the population of Yellowknife increased substantially around the mid-twentieth century, and so to meet the growing demand of wood for construction, logging became an important form of land clearance activity in the area (St-Onge 2007). Very recently (post-2010), some houses have been built along the shores of Grace Lake, and the lake is used for recreational purposes by residents and tourists visiting the city. Grace Lake supports a diverse fish population that includes burbot (*Lota lota*), northern pike (*Esox lucius*), lake whitefish (*Coregonus clupeaformis*), lake cisco (*Coregonus artedi*), ninespine stickleback (*Pungitius pungitius*), and walleye (*Sander vitreus*) (Stewart 1997). Unlike Grace, Alexie Lake is located ~ 20 km from major anthropogenic activities and is considered relatively pristine in terms of shoreline development. Importantly, Alexie also supports a diverse large-bodied fish population that includes species such as burbot, lake trout (*Salvelinus namaycush*), northern pike, and lake whitefish (e.g. Cott et al. 2011; Callaghan et al. 2016; Guzzo et al. 2016).

Materials and methods

Sediment coring and chronology

The high-resolution sediment cores were retrieved through the ice from Grace and Alexie lakes in April 2014 using a Uwitec gravity corer from the middle of the lakes to ensure the sediments are integrative and representative of past environmental information from multiple habitats in the lake. A modified Glew (1988) extruder was used to section the samples at 0.5-cm intervals to the base of the core. The samples were frozen for transportation to the Laboratory for the Analysis of Natural and Synthetic Environmental Toxicants at the University of Ottawa. Select sediment intervals were freeze-dried and analyzed for ^{210}Pb isotopes using an Ortec high-purity Germanium gamma spectrometer (Oak Ridge, Tennessee) at the University of Ottawa to radiometrically date the cores. Certified reference materials obtained from International Atomic Energy Association (Vienna, Austria) were used for efficiency corrections, and results were analyzed using ScienTissiME (Barry's Bay, ON, Canada). The sediment core chronologies were developed following

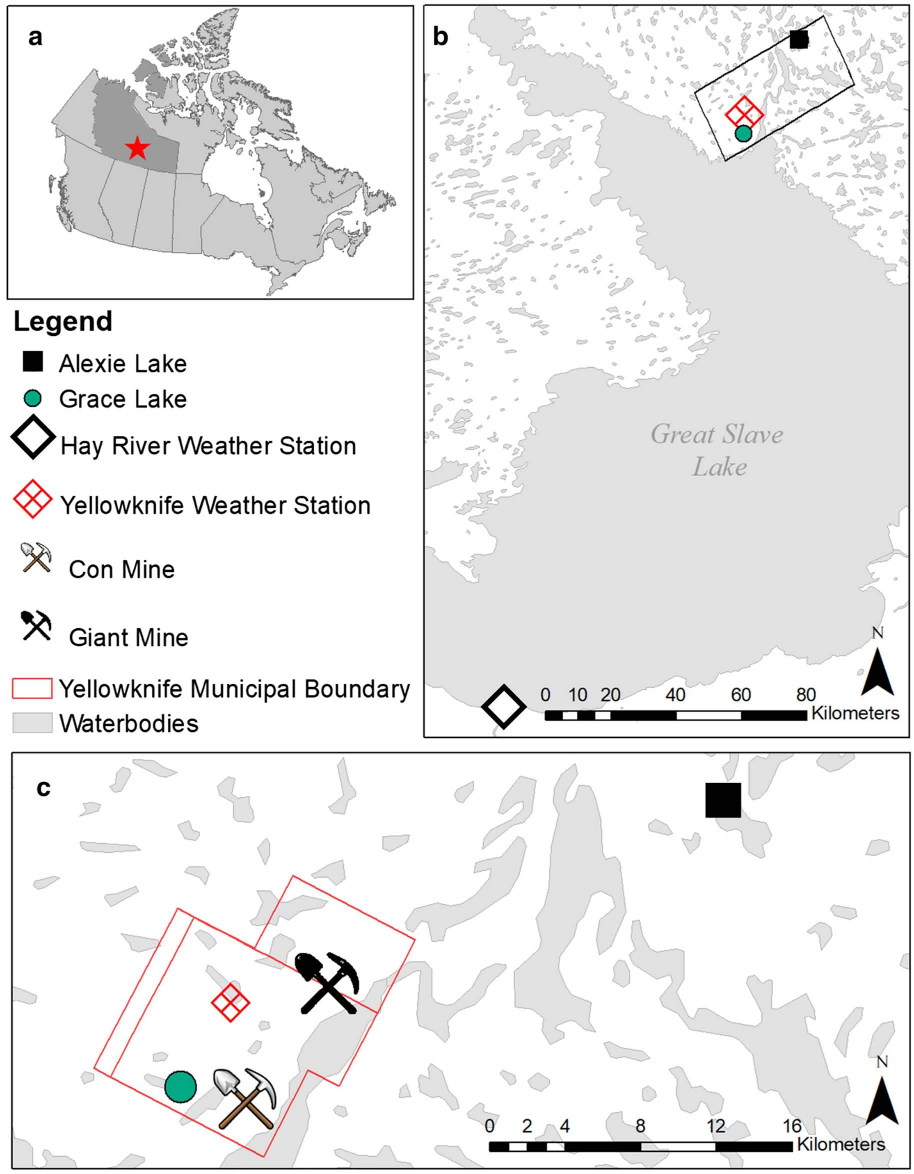

Fig. 1 Location of the study sites. **a** The red star indicates the location of Yellowknife, Northwest Territories on a map of Canada. **b** Map with the locations of meteorological stations and the two lakes in relation to Great Slave Lake. **c** Map of the Yellowknife area with the municipal boundary (indicated by red line), mines and the two study lakes

standard protocols described by Appleby (2001). Briefly, the supported activities of ^{210}Pb were determined by measuring the concentrations of ^{214}Pb in the sediments, and then the unsupported activities of the ^{210}Pb and the constant rate of supply model (CRS) were used to determine the ^{210}Pb dates.

Element analysis

Approximately, 0.5 g of freeze-dried sediments from select intervals were analyzed at SGS Laboratory (Lakefield, Ontario; Canadian Association for Laboratory Accreditation Inc. (CALA) accredited facility) for total metalloid concentrations using USEPA method 305B Aqua Regia Hot Plate Digest, followed by Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS) analysis. This method releases all elements that are not bound in silicate structures and are, therefore, potentially available to be mobile. All arsenic (As), antimony (Sb), and lead (Pb) concentrations presented herein are reported as total metalloid concentrations.

Diatom analysis

Subfossil diatom analyses were performed following procedures described in Sivarajah et al. (2019). A minimum of 350 diatom valves were identified and enumerated from each sample using a Leica DMR light microscope fitted with differential interference contrast optics under × 1000 magnification. We used the photomicrographs presented in Krammer and Lange-Bertalot (1986–1991), Håkansson (2002) and Rühland et al. (2003a) to identify the diatom taxa, and then verified the taxonomy with recent publications and online databases, such as Diatoms of North America (https

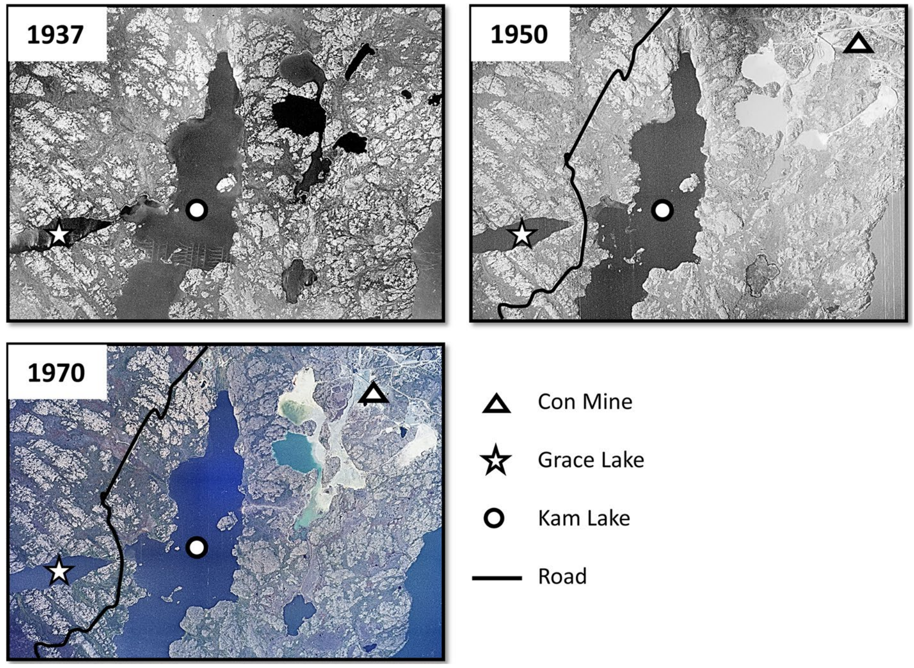

Fig. 2 Aerial photographs of Grace Lake and the surrounding area from 1937 (29 June 1937), 1950 (18 August 1950), and 1970 (2 September 1970). The original photographs were obtained from National Air Photo Library-Natural Resources Canada. A star and a circle were used to identify Grace and Kam lakes in all three photographs. The path of the road was traced with a black line and a triangle was used to identify Con Mine in photographs from 1950 and 1970

://diatoms.org/) to ensure the most updated names of species were used. The diatom taxa are expressed as percent abundance relative to the total number of diatom valves enumerated in a sample.

Sedimentary chlorophyll-*a*

Visible reflectance spectroscopy of chlorophyll-*a* (VRS Chl-*a*) is a rapid and non-destructive method to examine past trends in overall lake primary production (Michelutti et al. 2010) and has been used to assess past changes in trophic status in lakes across Canada (Michelutti and Smol 2016). Since chlorophyll-*a*, isomers of chlorophyll-*a*, and the main degradation products of chlorophyll-*a* (i.e., pheophytin-*a* and pheophorbide-*a*) have similar visible spectroscopic properties, the VRS Chl-*a* method tracks both primary chlorophyll-*a* and its main diagenetic products in sediments (Michelutti et al. 2010). Freeze-dried sediments were sieved through a 125-µm mesh to remove the effects of particle size and transferred to a glass vial. The samples were then scanned on a FOSS NIR System model 6500 rapid content analyzer, and the spectra in the range of 650–700-nm were analyzed to infer past trends in whole-lake primary production (Michelutti et al. 2010).

Temperature data

The long-term mean annual air temperature record for Yellowknife, NT (Climate Station ID: 2204101) was retrieved from the Adjusted and Homogenized Canadian Climate Database. The Yellowknife air temperature record was mostly continuous and contained data between 1944 and 2016, with 1 year (1945) of missing data. In addition, we used a longer record from Hay River, NT (Climate Station ID: 2202401) where air temperature data are available to the late-nineteenth century (1896–2016), but with 14 years of data missing (1897, 1899, 1902, 1908, 1909, 1917, 1918, 1922, 1927, 1943–1946, 1963). The Hay River climate station is located on the southern shores of Great Slave Lake, ~200 km south of Yellowknife (Fig. 1). The temperature records from both sites had very similar trends through time, however, the absolute values were generally lower at Yellowknife as it is more northerly.

Data analysis

Stratigraphic zones of the diatom assemblages were determined using constrained incremental sum of squares (CONISS) (Grimm 1987), and the broken stick model (Bennett 1996) was used to identify the important zones for each core using the R software environment (R Core Team 2018) using 'rioja' (Juggins 2017) and 'vegan' (Oksanen et al. 2018) software packages. Since the diatom species data had gradient lengths of < 2 standard deviation units (based on detrended correspondence analysis), linear ordination methods such as principal component analyses (PCA) were deemed appropriate to assess diatom assemblage changes through time (Birks 2010). Specifically, we examined the sedimentary diatom data from both lakes within the same ordination space using PCA (e.g. Sivarajah et al. 2017). This approach allowed us to determine if there were differences in diatom assemblages across both lakes and track the trajectories of changes in composition through time.

Results

Radiometric dating

Both Alexie and Grace lakes approached background ^{210}Pb activities around 12 cm (Online Resource 1). The CRS sedimentation rates were similar at both lakes around the early twentieth century (Grace Lake: 0.0031 g cm^{-2} $year^{-1}\pm0.0022$; Alexie Lake: 0.0021 g cm^{-2} $year^{-1}\pm0.0017$). However, the CRS sedimentation rates increased and fluctuated substantially during the mid- to late twentieth century at Grace Lake, while rates remained low and only subtly increased at Alexie (Online Resource 1). For example, between the 1940s and 2014, the average sedimentation rates at Grace and Alexie were 0.0137 and 0.0041 g cm^{-2} $year^{-1}$, respectively.

Element concentrations in sediments

There were notable differences in the elemental profiles of both study lakes (Fig. 3) with sedimentary metalloid concentrations generally higher at Grace than Alexie, even before the mining operations began. Coincident with the onset of regional gold mining operations, the concentration of mining-related metalloids, particularly arsenic and antimony, increased markedly around the 1930s–1940s at Grace Lake and reached maximum concentrations around the 1960s. However, the concentrations of As and Sb declined sharply after the 1960s and the present-day concentrations are only slightly higher than the pre-industrial period. Although lead concentrations of Grace Lake followed similar trajectories as arsenic and antimony until about the 1960s, the decline after the 1960s was more gradual. The metalloid concentrations at Alexie Lake also increased during the period of mining, however the changes were subtler compared to Grace Lake. In fact, when plotted on the same graph, the changes in metalloid concentrations at Alexie were not clearly distinguishable from background concentrations (Fig. 3). We therefore provided the metalloid profiles of Alexie Lake separately in Online Resource 2 to highlight the trends through time. We also explored the concentrations of metalloids in relation to titanium to determine watershed influence (Online Resource 3) and the trends remained similar to those presented in Fig. 3, indicating that the watershed inputs had little impact on the higher sedimentary metalloid concentrations during the mining era.

Diatom assemblage changes at Grace and Alexie lakes

The diatom assemblages of both study lakes were dominated by planktonic taxa throughout the last ~200 years. Specifically, *Discostella stelligera* (Cleve and Grunow) Houk and Klee was a common taxon in both lakes and increased in the most recent sediments (Figs. 4 and 5). The CONISS analysis, in conjunction with the broken stick model, identified two important zones in the diatom profiles of both lakes (Figs. 4 and 5) and the biggest split in assemblage composition occurred around the mid-1950s and early-1960s at Grace and Alexie lakes, respectively. However, the diatom assemblage changes were subtle at Alexie relative to the

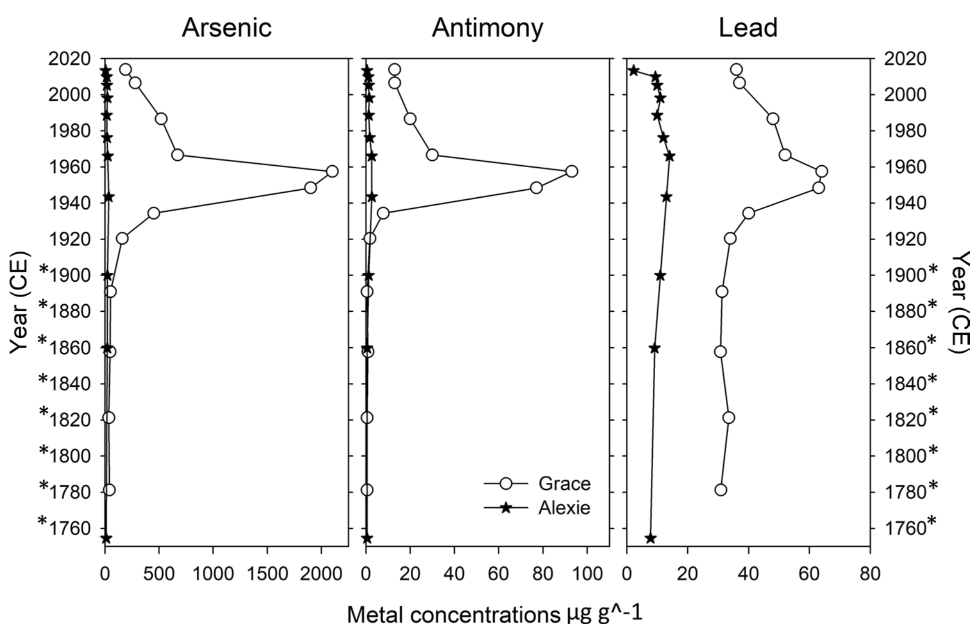

Fig. 3 Metal concentrations (weight per gram dry weight) in the sediments from Grace and Alexie lakes scaled by ^{210}Pb estimated years. The dates beyond background ^{210}Pb levels were extrapolated using a second-order polynomial. These dates have been identified with an asterisk (*) and should be interpreted with caution

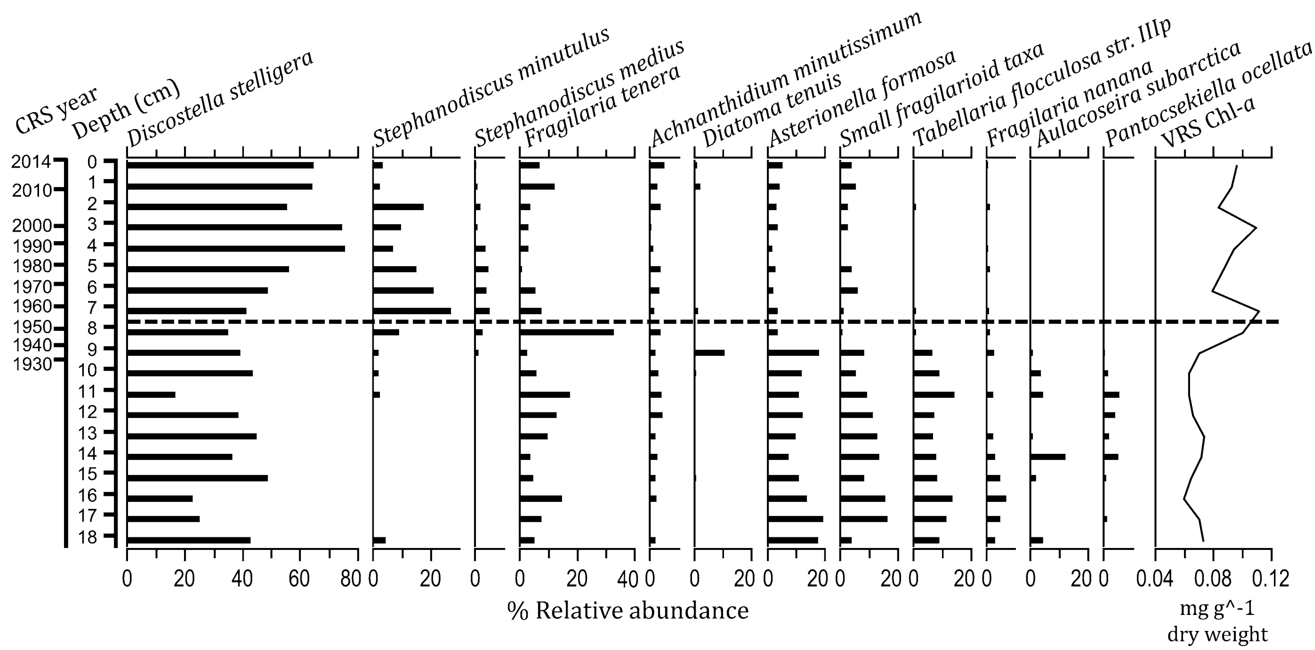

Fig. 4 Diatom assemblage profiles scaled by core depth (with ^{210}Pb estimated years plotted secondarily) showing relative abundances of the most common diatom taxa from Grace Lake, Yellowknife, Northwest Territories. For clarity of display, species from the same genera that exhibited similar trends through time were grouped together. CONISS zones are delineated with a horizontal broken line. The sedimentary VRS Chl-*a* (mg g^{-1} dry weight) are also included in the stratigraphy

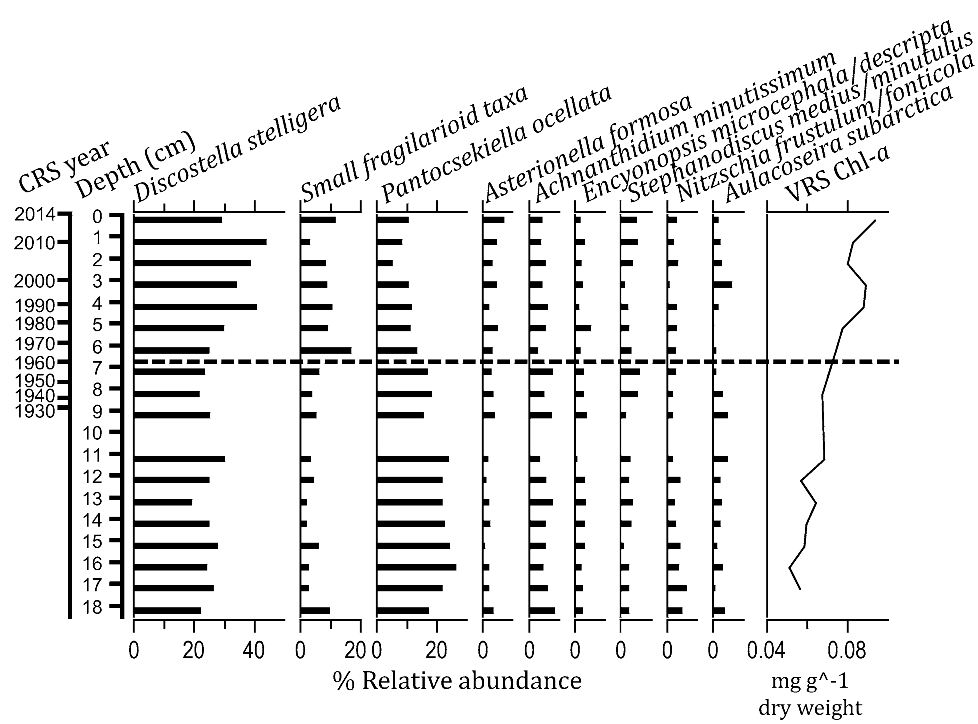

Fig. 5 Diatom assemblage profiles scaled by core depth (with ^{210}Pb estimated years plotted secondarily) showing relative abundances of the most common diatom taxa from Alexie Lake, Yellowknife, Northwest Territories. For clarity of display, species from the same genera that exhibited similar trends through time were grouped together. CONISS zones are delineated with a horizontal broken line. The sedimentary VRS Chl-*a* (mg g^{-1} dry weight) are also included in the stratigraphy

sharp changes that occurred at Grace Lake (Figs. 4, 5, and 6).

The diatom assemblages of Grace Lake were dominated by *D. stelligera* throughout the entire sedimentary record (Fig. 4). Although CONISS identified the major assemblage change around the mid-1950s (~ 7–8 cm), where the relative abundances of *D. stelligera* and *Stephanodiscus* taxa (*S. medius* Håkansson and *S. minutulus* (Kützing) Round) increased sharply, the changes in diatom assemblages were evident as early as the mid-to-late

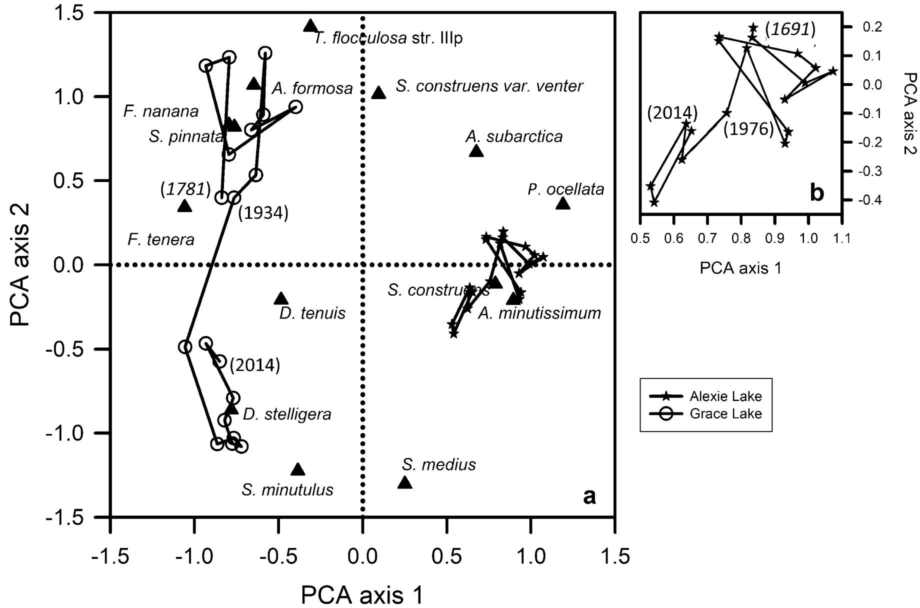

Fig. 6 Principal component analysis (PCA) ordination of axis 1 ($\lambda = 0.41$) and axis 2 ($\lambda = 0.17$) sample scores for the species and sites from Grace and Alexie lakes plotted within the same ordination space (**a**). The PCA axis scores for the most abundant species (i.e., > 5% in at least one sample from the two sites) are presented. The ^{210}Pb dates for select samples are included to highlight the major shifts in diatom composition through time. The dates beyond background ^{210}Pb levels have been italicized and these dates should be interpreted with caution. The PCA axis 1 and 2 samples scores for Alexie Lake has been plotted separately (**b**) to present the trends through time clearly

1940s. These assemblage changes included a decrease in the relative abundances of many planktonic taxa (*Asterionella formosa* Hassall, *Aulacoseira subarctica* (Müller) Haworth, *Pantocsekiella ocellata* (Pantocsek) Kiss & Ács, *Fragilaria nanana* Lange-Bertalot, *Fragilaria tenera* (W Smith) Lange-Bertalot, *Tabellaria flocculosa* str. IIIp Koppen) and small fragilarioid species (mostly *Staurosira construens* var. *venter* (Ehrenberg) Hamilton, *Staurosirella pinnata* (Ehrenberg) Williams and Round), with increases in planktonic *Stephanodiscus* taxa around the 1940s at Grace Lake.

The pre-disturbance diatom assemblages of Alexie Lake were co-dominated by *D. stelligera* and *P. ocellata* (Fig. 5). As further identified by CONISS, after the 1960s (~ 6–7 cm) the relative abundances of *D. stelligera* and small fragilarioid taxa (*S. construens* var. *venter*, *S. pinnata*, *Staurosira construens* Ehrenberg, *Pseudostaurosira brevistriata* (Grunow) Williams and Round) increased, while *P. ocellata* decreased. In contrast to Grace Lake, the relative abundances of *A. subarctica*, and *Stephanodiscus* taxa (*S. medius* and *S. minutulus*) did not change substantially through time. Similarly, taxa such as *Achnanthidium minutissimum* (Kützing) Czarnecki, *Nitzschia* taxa (*N. frustulum* (Kützing) Grunow and *N. fonticola* Grunow), and *Encyonopsis* taxa (*E. microcephala* (Grunow) Krammer and *E. descripta* (Hustedt) Krammer) occurred in < 10% relative abundances and did not display a directional trend throughout the sedimentary record of Alexie Lake.

The general direction of trajectories of the diatom assemblages through time, tracked by PCA axis 2 (Fig. 6a, b), were similar across both lakes. In the PCA ordination space, the pre-1940s samples from Grace Lake clustered together in the top-left quadrant while samples from post-1940s sediments plotted in the bottom-left quadrant (Fig. 6a). However, consistent with the diatom assemblage changes observed in the profiles, the changes in diatom composition at Alexie were subtler and the major shift occurred after the 1970s (Fig. 6b). Although both study lakes shared similar diatom taxa, the samples from the two lakes plotted separately along the PCA axis 1, highlighting the differences in the relative importance of each taxon in these lakes through time (Fig. 6a). For example, taxa that were present in higher abundances in the early sediments plotted in the top quadrant (e.g. *A. subarctica*, *P. ocellata*, *T. flocculosa* str. IIIp) while taxa that were present in higher abundances in the recent sediments plotted in the bottom quadrant (e.g. *D. stelligera*, *Stephanodiscus* taxa).

Past trends in whole-lake primary production

Trends in whole-lake primary production (inferred through VRS Chl-*a*) increased in both Grace and Alexie lakes (Figs. 4 and 5). The increase at Alexie was gradual through time with higher concentrations recorded in the post-1960s sediments (Fig. 5). The whole-lake primary production at Grace began to increase after the 1940s and fluctuated during the second half of the twentieth century. After the initial increase in VRS Chl-*a* between the 1940s and 1960s, it decreased briefly from ~1960 to ~1970, after which it increased until ~2000. Afterwards, the whole-lake primary production at Grace decreased subtly between 2000 and 2005 and then increased to the surface of the core.

Temperature data

Similar to previous investigations from Arctic and sub-Arctic environments, the annual air temperature records showed marked increases in Yellowknife and Hay River (Fig. 7). Specifically, the longer record from Hay River displayed a continuous increase in annual air temperature throughout the observational period (1896–2016). The two-segmented piecewise linear breakpoint analysis identified notable shifts in the air temperature records around 1956 and 1966 at Yellowknife and Hay River, respectively (Fig. 7). To examine if the differences in breakpoints were influenced by the length of the air temperature records, we applied the two-segmented piecewise linear breakpoint analysis on a section of the record from Hay River (1943 to 2016 where data were missing from 1943 to 1946 and 1963). This analysis identified a breakpoint around 1961 for the shorter annual air temperature record from Hay River.

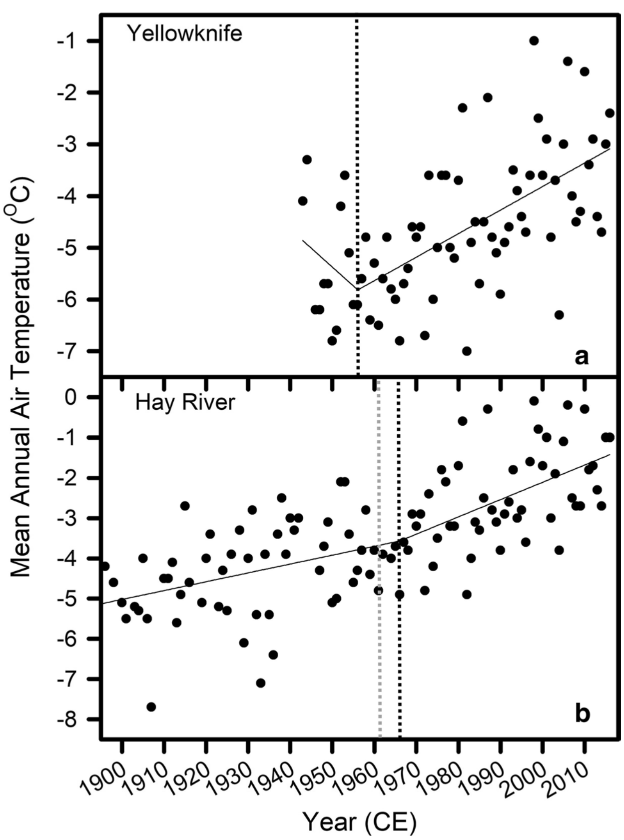

Fig. 7 The mean annual air temperature data from Yellowknife (**a**) and Hay River (**b**). A two-segmented piecewise linear regression of the mean annual air temperate data identified major breakpoints in the records around 1956 and 1966 at Yellowknife and Hay River, respectively (shown with black dotted lines in the figure). The grey-dotted line indicates a breakpoint around 1961 when the Hay River record was shortened to represent the same time frame as the Yellowknife record

Discussion

Geochemical signals of mining operations and land-use changes

The sedimentary biological and geochemical proxies at both lakes were generally stable prior to the onset of mining activities around Yellowknife (i.e. pre-1940s). The higher background concentrations of As at Grace relative to Alexie is likely linked to the underlying bedrock in the region, as well as Grace Lake's close proximity to the vein of arsenic-enriched arsenopyrite ore that made this area a prosperous location for gold mining operations (Cousens et al. 2006). The sedimentary metalloid profiles from Grace Lake (Fig. 3) clearly tracked the influence of mining in the region, as the As and Sb concentrations increased sharply during the early phase of gold mining (~1930s–~1970s), consistent with previous geochemical analyses from other lakes that are closer to the mine and the city (e.g. Dirszowsky and Wilson 2016; Thienpont et al. 2016). Unlike Grace, the increase in metalloid concentrations at Alexie were not pronounced and the differences in the magnitude of changes across the two sites highlight the influence of distance from the mines. The introduction of emission controls and the construction of the baghouse at Giant Mine to store the As_2O_3 in 1958 was also tracked by the notable and sharp declines in the concentrations of As and Sb after the 1960s.

The sedimentary Pb concentrations followed similar increasing patterns as As and Sb, with Pb concentrations declining gradually after the 1960s. Previous paleolimnological investigations, including one from the City of Yellowknife (Frame Lake; Dirszowsky and Wilson 2016), have noted that the sedimentary Pb concentrations tracked multiple anthropogenic activities but especially the use of leaded gasoline (Siver and Wozniak 2001). Hence, the gradual decline after the 1960s is tracking both the reduced atmospheric deposition of Pb from mining activities and use of leaded fuel. Furthermore, the CRS sedimentation rates at both lakes increased during the mid- to late-twentieth century, with pronounced changes and fluctuations occurring at Grace Lake relative to Alexie Lake (where the sedimentation rates remained low). The increases in average CRS sedimentation rates at Grace between the 1940s and 2014, relative to early twentieth century, suggests that local land-use changes (e.g. construction of the road adjacent to the lake and increased traffic, logging in the region, housing development on the shores) may have influenced the sedimentation rates of lakes in the City of Yellowknife. Meanwhile, the lack of catchment disturbances around Alexie led to minimal changes in sedimentation rates over the same time frame.

Biological responses to multiple environmental stressors at Alexie and Grace lakes

Disentangling the effects of multiple environmental stressors on aquatic biota can be challenging as stressors may interact, thus leading to complex biological responses (Smol 2010). In the case of Yellowknife, three major environmental stressors impacted the region after the late-1930s: 1) metalloid contaminants from gold mining operations (late-1930s to early-2000s); 2) land-use changes related to the development of the city (late-1930s to the present); and 3) marked increases in regional air temperature (~ post-1960s to the present). Although the onset of these stressors occurred as early as the 1930s, the major shift in diatom assemblages at Grace and Alexie occurred after the mid-twentieth century (Figs. 4 and 5). Similar to the trends in sedimentary metalloid profiles, larger diatom assemblage changes were observed in Grace Lake (Fig. 6) as it was closer to the mines and the city, thus receiving higher metalloid contaminants and nutrients from land-use changes in the area. The lake water As concentrations at Grace was 16.1 μg L^{-1} in 2014; however, metalloid concentrations in the water column were likely higher in the past, as we observed clear increases in the concentrations of As and Sb in the sediments during the mining era (Fig. 3). Hence, the higher concentrations of metalloids may have had direct impacts on the algal assemblages. For instance, the loss or decline in the abundance of several planktonic diatom taxa (*A. formosa*, *F. nanana*, *F. tenera*, *T. flocculosa* str. IIIp, *P. ocellata*) at Grace after the 1950s were likely responses to mining activities, as similar changes have been observed in other metal-contaminated lakes (e.g. Austin et al. 1985; Cattaneo et al. 2004; Leppänen et al. 2019). Since Alexie Lake was not contaminated to the same extent as Grace, the diatom assemblage changes in response to mining were subtle.

Interestingly, metal-sensitive planktonic taxa and some small fragilarioid taxa at Grace Lake were replaced by *Stephanodiscus* taxa and *D. stelligera* during the second half of the twentieth century (Fig. 4). Limnological and paleolimnological assessments from temperate and Arctic regions have shown that *Stephanodiscus* taxa often indicate eutrophication (e.g. Lotter 1998; Hadley et al. 2010; Nelligan et al. 2016). For example, in a paleolimnological assessment of a macrophyte-dominated shallow lake (Niven Lake, Yellowknife), Stewart et al. (2018) suggested that the subtle increase in the relative abundance of *Stephanodiscus* taxa was a response to raw sewage input from 1948 to 1981. While there was no documented evidence of sewage disposal into Grace Lake during the twentieth century, the land-use changes around Grace began as early as the 1930s with the construction of Con Mine less than 4 km away. Then, a road was constructed sometime between 1937 and 1950 near the outflow (Fig. 2) that likely increased human activity around the lake. Additionally, the population of the city also grew rapidly as a direct response to the economic prosperity with Yellowknife becoming the capital of the Northwest Territories in 1967. Hence, urbanization during the mid-to-late twentieth century likely resulted in higher nutrient inputs to lakes within the city as observed in previous studies (e.g. Dirszowsky and Wilson 2016; Gavel et al. 2018). Although we are not aware of any monitoring data on the historic nutrient inputs to Grace, we can infer that the increased sedimentation rates after the 1940s likely brought in additional nutrients from the landscape to Grace Lake which promoted algal production. Furthermore, the concomitant increases in eutrophic *Stephanodiscus* taxa and VRS-Chl-*a* at Grace after the 1940s suggests that the higher nutrient inputs from the local land-use changes may have contributed to the rise in whole-lake primary production. Meanwhile, the relative abundances of *Stephanodiscus* taxa at Alexie did not change substantially over the past ~ 200 years, as there was no catchment development and anthropogenic sources of nutrients to the lake. Despite the lack of changes in *Stephanodiscus* taxa, the whole-lake primary production increased at Alexie Lake too, suggesting that factors other than nutrient additions may also be influencing algal production in the lakes around Yellowknife. Unlike Grace, the increase in sedimentary chlorophyll-*a* (and its main diagenetic products) at Alexie was gradual throughout the record with greater increases occurring after the 1960s when temperatures became notably warmer. The rising temperatures likely led to declining ice-cover, and longer and warmer growing seasons that provided ideal conditions for increases in whole-lake primary production, as has been noted in many other Arctic (e.g. Griffiths et al. 2017) and temperate lakes (e.g. Nelligan et al. 2016; Summers et al. 2016; Paterson et al. 2017; Sivarajah et al. 2017).

The relative abundance of *D. stelligera*, a small-celled centric planktonic taxon that dominates the diatom assemblages in many deep lakes around the world, increased at both of our study lakes. Investigations of acidified and metal-contaminated lakes have identified *D. stelligera* as a taxon sensitive to pollution (Battarbee and Charles 1987; Cattaneo et al. 2008), including a severely As-contaminated shallow lake close to Giant Mine where *D. stelligera* complex decreased to negligible abundances while benthic naviculoid and *Denticula* taxa increased in response to severe metalloid contamination (Thienpont et al. 2016). However, its increase across both of our deep sites during the warmest years was consistent with trends observed in the paleolimnological records of lakes across the Northern Hemisphere where planktonic diatoms (mostly *D. stelligera*) have increased in response to recent warming mediated by changes to lake thermal properties (Sorvari et al. 2002; Rühland et al. 2008, 2015). Specifically, warmer air-temperatures often lead to longer ice-free periods with stronger thermal stratification which provides competitive advantages to small-celled centric diatoms, such as *D. stelligera*, that have high surface area to volume ratios and low sinking velocities (Ptacnik et al. 2003) and are able to remain in the photic zone longer relative to larger and heavier diatoms (Rühland et al. 2015). Furthermore, the increase in *D. stelligera* in response to warming was more pronounced in stratified deep lakes in the Canadian Arctic (Rühland et al. 2003b). Therefore, we conclude that climate-mediated changes to lake thermal properties may be stronger drivers of *D. stelligera* abundance relative to metalloid pollution from mining activities in these deep sub-Arctic lakes.

Changes in the limnology of deep sub-Arctic lakes around Yellowknife

The sedimentary geochemical and biological proxies from Alexie and Grace suggest that the limnology of these lakes that support large-bodied fish species have changed markedly since the mid-twentieth century in response to multiple environmental stressors. The degree of metal contamination and eutrophication varied between the two lakes as there were notable differences in distance to anthropogenic activities. Although both lakes received metalloid contaminants in the past (Alexie to a lesser extent), the recent decreases in sedimentary metalloid concentrations provide some indications of chemical recovery from gold mining operations. The recent biological assemblages at both lakes were substantially different relative to the ones from pre-mining and mining eras. This is consistent with some of the earlier work done on shallow lakes in the Yellowknife area (e.g. Thienpont et al. 2016; Stewart et al. 2018) and other industrially impacted lakes (e.g. Summers et al. 2016; Sivarajah et al. 2017) where recent warming has likely altered limnological conditions sufficiently that returning to pre-industrial conditions is no longer tenable.

Although speculative, some of the paleolimnological changes observed in this study may provide insights into the potential long-term impacts of multiple environmental stressors on the habitat of large-bodied fish species. For instance, the increased sediment inputs due to anthropogenic activities, such as road construction and housing development, may lead to changes in water clarity and impede visual abilities of fish during foraging and mate selection (Cott et al. 2015). Furthermore, the increases in primary production across both lakes may have indirect consequences on the fish habitat as the decomposition of large quantities of algal biomass consume oxygen in the cold-hypolimnion where many large-bodied fish species take refuge. The concomitant increase in *D. stelligera* at our study sites suggest that the lake thermal properties (i.e. less ice-cover, longer periods of thermal stratification) have been changing in these lakes and could eventually limit the optimal oxy-thermal habitat for cold-water fish species such as lake trout (Guzzo and Blanchfield 2017), especially in lakes that may be subjected to eutrophication from increasing urbanization. Hence, special consideration should be given to the management of sub-Arctic lakes that support large-bodied fish populations as they have important cultural and economic significance to local communities.

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Compliance with ethical standards

Conflict of interest The authors have no conflicts of interest to declare.

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