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Impacts on aquatic biota from salinization and metalloid contamination by gold mine tailings in sub-Arctic lakes☆

Madi Perrett [a, 1], Branaavan Sivarajah [a, *, 1], Cynthia L. Cheney [b], Jennifer B. Korosi [c], Linda Kimpe [b], Jules M. Blais [b], John P. Smol [a]

[a] Paleoecological Environmental Assessment and Research Laboratory, Department of Biology, Queen's University, Kingston, K7L 3N6, Ontario, Canada
[b] Laboratory for the Analysis of Natural and Synthetic Environmental Toxicants, Department of Biology, University of Ottawa, Ottawa, K1N 6N5, Ontario, Canada
[c] Department of Geography, York University, Toronto, M3J 1P3, Ontario, Canada

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ABSTRACT

Precious metal mining activities have left complex environmental legacies in lakes around the world, including some sites in climatically sensitive regions of the Canadian sub-Arctic. Here, we examined the long-term impacts of past regional gold mining activities on sub-Arctic lakes near Con Mine (Yellowknife, Northwest Territories) based on sediment core analysis (paleolimnology). In addition to receiving metal(loid)s from roaster stack emissions, the study lakes were also influenced by salt-rich mine drainage from Con Mine tailings. Water samples from these lakes had some of the highest concentrations for salinity-related variables (e.g. Ca^{2+}, Cl^{-}, Na^{+}) and metal(loid)s (e.g. As, Cu, Ni, Sb) in the Yellowknife area. Furthermore, the presence of halophilic diatom (Bacillariophyceae) taxa (*Achnanthes thermalis* and *Navicula incertata*) in the recent sediments of Keg and Peg lakes suggest that the extreme saline conditions are strongly influencing the present biota, more than 10 years after the cessation of gold mining activities at Con Mine. The sedimentary metal(loid) profiles (e.g. As, Cu, Ni) of Kam Lake tracked the influence of regional gold mining activities, particularly those at Con Mine, while the algal assemblages recorded the biological responses to salinization and metal(loid) pollution (e.g. marked decreases in diatom species richness, Hill's N2 diversity, and chrysophyte cyst:diatom valve ratio). At Kam Lake, the algal assemblage changes in the post-mining era were indicative of climate-mediated changes to lake thermal properties (e.g. rise in planktonic diatoms), nutrient enrichment related to urbanization (e.g. increase in eutrophic *Stephanodisusc* taxa), and/or a combination of both stressors. The lack of biological recovery (i.e. return to pre-mining assemblages) is consistent with investigations of mine-impacted lakes in temperate regions where elevated contaminant levels and emerging stressors (e.g. climate warming, land-use changes) are influencing lake recovery.

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1. Introduction

Metal mining activities often pose a serious threat to the integrity and functioning of aquatic ecosystems by altering chemical and biological conditions (Little et al., 2020; Pelletier et al., 2020; Pociecha et al., 2020; Tenkouano et al., 2019). While much of the previous research on the ecological legacies of mining activities have focused on temperate freshwater systems, a growing number of studies are examining the long-term limnological consequences of mining activities in northern environments (Cheney et al., 2020; Chételat et al., 2019; Leppänen et al., 2019; Sivarajah et al., 2021; Thienpont et al., 2016). The lakes in circumpolar regions are also sensitive to additional environmental stressors, including those related to accelerated recent climate warming and urbanization (Gavel et al., 2018; Solovieva et al., 2005). Hence, understanding the long-term effects of these stressors is necessary to determine how high-latitude lake ecosystems may respond to future environmental perturbations (e.g. potential exploration and extraction of natural resources; Cott et al., 2015). Long-term monitoring data, however, are scarce for almost all lake regions

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* Corresponding author. Present Address: Department of Geography and Institute of Environmental Studies Carleton University, Ottawa, K1S 5B6, Canada.
E-mail address: branaavan.sivarajah@queensu.ca (B. Sivarajah).
[1] Both authors contributed equally to this work.

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(Smol, 2019) and are especially sparse for northern ecosystems. Therefore, we used paleolimnological techniques to examine the long-term biological impacts of salt-rich mine tailings and metal(loid) pollution on sub-Arctic lakes around Con Mine, Yellowknife, Northwest Territories, Canada.

Gold mining operations around Yellowknife were key to the development of this sub-Arctic city (Silke, 2009; Wolfe, 1998), but it also resulted in the metal(loid) contamination of surrounding aquatic and terrestrial environments (Galloway et al., 2015; Jamieson et al., 2017; Palmer et al., 2019). For example, recent studies of lakes around the City of Yellowknife have reported some of the highest lake-water and sediment arsenic (As) concentrations in Canada (Cheney et al., 2020; Nasser et al., 2016; Palmer et al., 2019). Most of the environmental impact assessments near Yellowknife have focused on the ecological impacts of roaster stack emissions from Giant Mine (Giant Yellowknife Gold Mines Limited; 1948–2004; Fig. 1) as it released 20,824 tonnes of toxic arsenic trioxide into the environment (Cheney et al., 2020; Jamieson, 2014; Palmer et al., 2019; Thienpont et al., 2016). However, gold mining operations in Yellowknife began at Consolidated (Con) Mine (Consolidated Mining and Smelting Company of Canada; 1938–2003), located on the northern shores of Great Slave Lake and bordering the southern limits of the city (Fig. 1), a decade before Giant Mine operations opened.

The abandoned Con Mine is underlain by the Yellowknife greenstone belt, where the gold-bearing ores were extracted from the Con, Rycon-Negus, and Campbell shear zones (Duke et al., 1991; Silke, 2009). Relative to nearby Giant Mine, the roasting of arsenopyrite ores at Con released less arsenic through roaster stack emissions (Hocking et al., 1978). For example, between 1948 and 1970, Giant released 15,785 long tons (16,038 tonnes) of arsenic, while Con emitted only 2484 long tons (2524 tonnes; Hocking et al., 1978). Furthermore, ore roasting at Con ended in 1971 because, by this time, most of the ores processed there were recovered from the Campbell shear zone that were free-milled and did not require thermal treatments such as roasting (Hocking et al., 1978; Silke, 2009). Importantly, however, Con Mine has left an additional environmental legacy, because some of the salt-rich mill effluents from Con were routed to Pud Lake. This resulted in the contamination of a chain of small shallow lakes (Meg-Keg-Peg) as well as Kam Lake, as they were all connected to Pud (Bright et al., 1994; Falk et al., 1973; Koch et al., 2000). Presently, these lakes have exceptionally high concentrations for salinity-related variables (e.g. Cl^{-1}; Table 1). In addition to the impacts of historic gold mining activities, the lakes around Yellowknife were also affected by local land-use changes (e.g. urbanization) and regional climate warming (Gavel

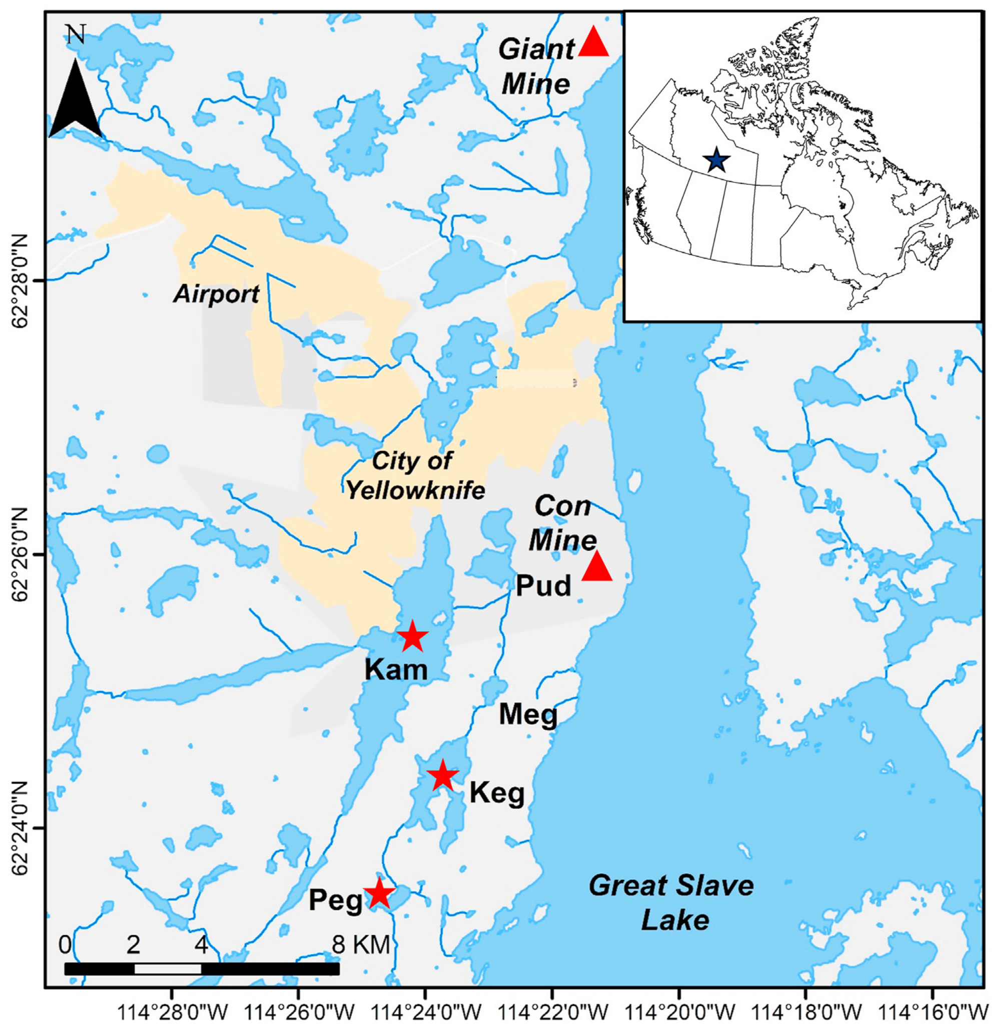

Fig. 1. Location of the study sites. Kam, Keg, and Peg lakes in Yellowknife, Northwest Territories, are indicated with red stars. The inset map identifies the location of Yellowknife on a map of Canada with a blue star.

Table 1
Selected limnological variables for the three lakes sampled around Con Mine (Yellowknife, Northwest Territories, Canada). The data presented here were point measurements taken when the sediment cores were retrieved in April 2014 and the lakes were still ice-covered. Hence, it is likely that the water chemistry values reported may be higher relative to the open-water period as a result of solute exclusion during ice formation. The complete data for the 33 lakes are provided in Sivarajah et al. (2019) and the median values for most variables are presented therein, except for Cu, Fe, Ni, and Sr.

	Latitude (decimal degrees)	Longitude (decimal degrees)	Distance to Giant Mine (km)	Distance to Con Mine (km)	Coring depth (m)	Surface area (km^2)	pH	Total nitrogen (mg L^{-1})	Dissolved organic carbon (mg L^{-1})
Kam	62.42242	−114.401	9.25	1.97	7.5	2.16	7.55	0.88	17
Keg	62.40597	−114.398	11.09	3.22	1	0.41	6.94	3.77	45.2
Peg	62.3904	−114.41	12.86	5.08	1	0.11	7.08	2.68	41.9
	Ca^{2+} (mg L^{-1})	**Cl^{-}(mg L^{-1})**	**Mg^{2+} (mg L^{-1})**	**K^{+} (mg L^{-1})**	**Na^{+} (mg L^{-1})**	**Sulphate (mg L^{-1})**	**Specific conductance (μS cm^{-1})**	**Dissolved solids (mg L^{-1})**	**Alkalinity (mg L^{-1})**
Kam	46.8	33	12.5	3.9	15.2	41	400	246	107
Keg	3690	7780	257	56	1820	2920	22,900	17,400	168
Peg	1850	4150	137	29.1	973	1190	13,200	9040	179
Median of 33 lakes	33.8	5.2	13.7	3.6	9.2	11	285	246	110
	Al (μg L^{-1})	**As (μg L^{-1})**	**Ba (μg L^{-1})**	**Cu (μg L^{-1})**	**Fe (μg L^{-1})**	**Li (μg L^{-1})**	**Ni (μg L^{-1})**	**Sb (μg L^{-1})**	**Sr (μg L^{-1})**
Kam	28.2	238	33.6	11.2	56	6.6	5.2	22.6	272
Keg	468	300	269	19	1120	73.7	98.1	31.9	44,300
Peg	435	2780	257	4.1	12,900	46.5	44.4	7.2	23,000
Median of 33 lakes	16.4	41.7	35.7	0.7	51	8.1	0.4	1.1	125

et al., 2018; Sivarajah et al., 2020; Stewart et al., 2018).

Salinization and metal(loid) pollution can substantially alter the chemical and biological conditions in freshwater ecosystems. For instance, disposal of mine wastes into a Finnish lake led to higher levels of contaminants in the water column and induced biological changes across multiple trophic levels (Leppänen et al., 2019). In Yellowknife, the biological consequences of metal(loid) pollution from historic gold mining activities have been documented across several lakes (e.g. Nasser et al., 2016; Persaud et al., 2021; Sivarajah et al., 2019; Thienpont et al., 2016). However, the cumulative long-term impacts of both salinization and metal(loid) pollution on aquatic biota has not been extensively investigated. Hence, the lakes around Con Mine provide an important opportunity to assess the long-term biological impacts of these dual stressors on sub-Arctic lakes.

The objectives of this study were to: 1) determine the cumulative biological impacts of metal(loid) contamination and salinization; and 2) assess potential biological recovery patterns in the lakes near Con Mine. We reconstructed the pollution history of Kam Lake by measuring the sedimentary metal(loid) concentrations and determined the biological responses throughout the 20th and early-21st centuries by assessing diatom (Bacillariophyceae; siliceous unicellular algae) assemblages in a dated sediment core, with additional data on shifts on chrysophyte cysts and past estimates of whole lake primary production. We specifically chose a diatom-based approach because diatoms respond rapidly to changes in salinity (Fritz et al., 2010) and metal(loid) pollution (Salonen et al., 2006; Pociecha et al., 2020). This makes them excellent bio-indicators to track the biological impacts of mining operations at Con Mine. We also identified extremophilic diatom taxa, specifically halophiles, from the surficial sediments of Keg and Peg lakes, where salt concentrations are still extremely high.

2. Materials and methods

2.1. Study area

Con Mine and the study lakes (Fig. 1) are located within the Great Slave Uplands and Lowlands of Taiga Shield High Boreal ecoregion (Ecosystem Classification Group, 2008) and underlain by the Slave Structural Province of the Canadian Shield (Wolfe, 1998). The Yellowknife region experiences a continental sub-Arctic climate with relatively long winters and short summers (Wolfe, 1998), where mean annual air temperatures were below 0 °C between 1942 and 2016 (range: - 1 to - 7 °C; data retrieved in April 2017 from http://www.ec.gc.ca/dccha-ahccd/). Due to the cold conditions, the lakes in the region are ice-covered for more than half the year. Although we do not have ice-cover information for our study lakes, the data collected at nearby Long Lake between 1956 and 1994 indicates that the ice-on duration ranged from 196 to 233 days (Benson et al., 2013).

Kam, Keg, and Peg lakes are less than 6 km from Con Mine (Fig. 1) and were affected by tailing spills from Pud Lake (Bright et al., 1994; Koch et al., 2000; Wagemann et al., 1978). Kam Lake is connected to Pud by a small stream on the northwestern shore, while Keg and Peg are part of a chain of lakes that are connected to Pud via streams to the south. Kam is a large (surface area = 2.16 km^2), relatively deep (maximum depth = ~12 m) lake (Healey and Woodall, 1973) and the surrounding area is well developed with residences and local businesses in the northwestern part of the lake (Fig. 1). In the late-1970s, sled dog kennels were established at the shores of Kam Lake (Yellowknife Dog Trotters Association) and untreated raw sewage was also directed into the lake briefly (Wagemann et al., 1978). Unlike Kam, both Keg and Peg lakes are small (<0.5 km^2) and shallow (<2 m maximum depth; Bright et al., 1994) with no catchment development (Fig. 1).

2.2. Field methods

Kam, Keg, and Peg lakes were sampled in the early spring of 2014 (April) when the lakes were ice-covered. The water and sediment samples were retrieved from the middle of all three lakes. Water samples were collected after triple rinsing the bottles with lake water. The water samples were analyzed at the Taiga Environmental Laboratory (Canadian Association for Laboratory Accreditation Incorporated (CALA Inc.) certified facility for the analysis of water chemistry parameters presented herein) in Yellowknife, following analytical protocols based on Standard US Environmental Protection Agency methods.

Sediment cores were retrieved using a Uwitec gravity corer, and vertically extruded at 0.5 cm contiguous intervals using a modified Glew (1988) extruder for the Kam Lake core. The coring depth for Kam, Keg and Peg lakes were 7.5 m, 1 m, and 1 m, respectively. For Keg and Peg lakes, only the surface 0.5 cm sediment increments, reflecting recent diatom assemblage composition, were analyzed.

2.3. Sediment core dating

The ^{210}Pb and ^{214}Pb activities for selected sedimentary intervals from the Kam Lake sediment core were measured using gamma spectroscopy at the University of Ottawa. Briefly, freeze-dried sediments were placed in a plastic vial and sealed with clear epoxy prior to analysis by an EG&G Ortec high purity germanium gamma spectrometer (Oak Ridge, Tennessee, USA). Efficiency corrections was made using certified reference materials obtained from the International Atomic Energy Association (Vienna, Austria). The supported activities of ^{210}Pb were determined by measuring the activities of ^{214}Pb and the unsupported activities of ^{210}Pb were calculated by subtracting the supported activities of ^{210}Pb from the total activities of ^{210}Pb as recommended by Appleby (2001). The unsupported activities of ^{210}Pb and the constant rate of supply (CRS) model described by Appleby (2001) were used to establish the chronology for the sediment core from Kam Lake using the computer software program ScienTissiME (Barry's Bay, Ontario, Canada). The results of core dating are presented in Supplement 1.

2.4. Sedimentary metal(loid) analyses

The sedimentary metal(loid) analyses was conducted at the SGS Laboratory in Lakefield, Ontario, Canada (CALA Inc. accredited facility for all elements reported herein) and the details of the laboratory procedures are presented in Cheney et al. (2020). Briefly, ~0.5 g of freeze-dried sediments from select intervals of the Kam Lake core were analyzed using aqua regia extraction, followed by Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS). The Method Detection Limits (MDL) for the analyzed metal(loids) are presented in Supplement 2. All sedimentary metal(loid) concentrations presented herein are reported as total concentrations and plotted stratigraphically using the computer program C2 version 1.5 (Juggins, 2007).

2.5. Algal analysis

Approximately 0.02 g of freeze-dried sediment was processed with concentrated acid mixture (50:50 M ratio of nitric (HNO_3) and sulfuric (H_2SO_4) acids) to digest the organic material in the sediment, and isolate the siliceous components (diatom valves, chrysophyte scales) of the sediments. The samples were then placed in an 80 °C hot water bath for ~2 h and the samples settled for ~24 h. The supernatant from the subsequent slurries was extracted from each sample, and deionized water was used to rinse the remaining sample until circumneutral pH was attained. Using a standardized mixing regime, aliquots of the mixed slurries from each sample interval were plated onto individual glass cover slips using a pipette. The samples were air dried and Naphrax® was used as a mounting medium to mount the glass coverslips onto microscope slides permanently. A minimum of 350 diatom valves were enumerated and identified from each sample. The diatom counts were presented as percent relative abundances to the total valves counted for each sample and the most common taxa presented stratigraphically using the computer program C2 version 1.5 (Juggins, 2007).

In addition to the down-core diatom assemblage data from Kam Lake, we examined the ratio of chrysophyte cysts to diatom valves (C:D) and this ratio (Smol, 1985) has been used to assess limnological changes associated with salinity, nutrients, and pH (reviewed in Zeeb and Smol, 2001). However, its utility has not been explored for mining contamination in sub-Arctic regions. The C:D was calculated for each sample from the Kam Lake sediment core and expressed as a percentage, according to Eq. (1) (*sensu* Cumming et al., 1993). Here, C:D is a ratio of the number of cysts (N_c) as a function of the total number of cysts (N_c) and diatom valves (N_{dv}).

$$C : D = \left(\frac{N_c}{N_c + N_{dv}}\right) \times 100 \qquad \text{Eq. (1)}$$

Visible reflectance spectroscopy of chlorophyll *a* (VRS Chl-*a*) was used to reconstruct the overall trends in primary production of Kam Lake over the past ~ 200 years. This is a cost effective and non-destructive method of analyzing trends in past production, which includes the isomers and main diagenetic products of chlorophyll *a* (Michelutti and Smol, 2016). Freeze-dried sediment samples from various intervals were sieved through a 125 μm mesh sieve to avoid the influence of particle size and were transferred into glass cuvettes (Michelutti et al., 2010). The glass cuvettes were then placed in a FOSS NIR System model 6500 rapid content analyzer, and spectral signatures between 650 and 700 nm wavelengths were analyzed to reconstruct VRS Chl-*a* in the samples (Michelutti et al., 2010).

2.6. Data analysis

Ordination analyses were completed for the full diatom data set (i.e. no groupings of species) from Kam Lake, using the program CANOCO version 5.0 (ter Braak and Šmilauer 2012). The relative abundances of the diatom assemblage data were square-root transformed before ordination analyses to equalize the variance among the diatom taxa. Detrended correspondence analysis of the down-core diatom data of Kam Lake revealed a gradient length of less than 2 standard deviation units. Therefore, the linear ordination technique of principal component analysis (PCA) was used to summarize the variation in the diatom data. The PCA axis 1 and 2 sample scores were plotted in the diatom stratigraphy of Kam Lake along with the assemblage data. The rarified species richness and Hill's N2 diversity (Hill, 1973) of the diatom data from Kam Lake were calculated using the 'vegan' (Oksanen et al., 2019) and 'rioja" (Juggins, 2017) statistical packages developed for R software environment (R Core Team, 2018).

2.7. Temperature data

The closest climate station to Kam Lake is located at the Yellowknife Airport (Climate Station ID: 2204101), which is approximately 5 km northwest of Kam Lake. The long-term mean air temperature data for this site was retrieved from the Environment Canada Historical Climate Record (http://climate.weather.gc.ca/). The mean annual and summer air temperature records from the Yellowknife Airport contained data from 1943 to 2016 and it was mostly complete with missing annual temperature data from 1945. We applied a linear regression to the air temperature data and then used the slope of the trend line to calculate the change in temperature over the entire record.

3. Results

3.1. Present limnology and surface diatom assemblages of lakes around Con Mine

The concentrations for ions (e.g. Ca^{2+}, Cl^-, K^+, Mg^{2+}, Na^+, SO_4^{2-}) and metal(loid)s (e.g. Al, As, Ba, Cu, Fe, Li, Ni, Sb, Sr) in the water columns of lakes around Con Mine were higher relative to other lakes in the Yellowknife region (Table 1). The arsenic levels exceeded the guidelines for the protection of aquatic life (5 μg L^{-1}; Canadian Council of Ministers of the Environment (CCME), 2001) and drinking water (10 μg L^{-1}; Health Canada, 2006) by several fold in all three lakes (Table 1). The surface sediment diatom assemblages of all three study lakes were dominated by benthic fragilarioid taxa (*Staurosira construens* var. *venter* (Ehrenberg) Hamilton and *Staurosirella pinnata* (Ehrenberg) D. M. Williams & Round, *Pseudostaurosira brevistriata* (Grunow) Williams & Round, *P. brevistriata* type) (Table 2). Taxa tolerant of high ionic concentrations such as *Achnanthes thermalis* (Rabenhorst) Schoenfeld, and *Navicula incertata* Lange-Bertalot were preset in greater than 5% relative abundances at Keg and Peg (Table 2). Additionally, pollution tolerant *Navicula libonensis* Schoeman was present in 6% abundance at Keg Lake. Planktonic diatoms were only observed at Kam Lake, while the diatom assemblages at the shallower Keg and Peg lakes were composed of benthic diatom taxa exclusively (Table 2).

3.2. Sedimentary metal(loid) concentrations at Kam Lake

The sedimentary concentrations of cadmium (Cd), copper (Cu), nickel (Ni), lead (Pb), and zinc (Zn) were lowest prior to the onset of mining operations and increased to varying degrees during the mining era (Fig. 2). Specifically, the concentrations of Cu, Ni, Pb, and Zn increased by 19, 2.3, 2.6, and 1.7-fold, respectively, above baseline during the mining period (~15 cm—~4 cm; Fig. 2). As and Sb, which are associated with roaster stack emissions and mine tailings, were elevated in the sediments from the mining and pre-mining eras (Fig. 2). The first increase in the concentrations of As and Sb occurred around 1897 (~18 cm, pre-mining period) and the second increase was recorded during the mining era (~1971, ~12 cm; Fig. 2).

3.3. Algal assemblage changes at Kam Lake

At Kam Lake, benthic *S. pinnata* dominated the assemblages throughout the entire record (Fig. 3). Prior to the onset of gold mining, notable abundances (~7%) of *Achnanthes sensu lato* and naviculoid taxa were observed in the sediments (Fig. 3). Although these groups were primarily composed of *Achnanthes acares* Hohn & Hellerman and *Achnanthes lanceolata* (Brébisson ex Kützing) Grunow, and *Navicula minima* Grunow, *Navicula jarnefeltii* Hustdet and *Navicula submuralis* Hustedt, several other taxa from these genera that occurred in low abundances (<0.5%) in the pre-industrial sediments were also included in these groups (21 taxa in each group; Fig. 3). Furthermore, during the pre-mining period, the diatom species richness and the ratio of chrysophyte cysts to diatom valves (C:D) were relatively high, whereas diatom species diversity and whole lake primary production were stable (Fig. 3). When gold mining began at Con Mine around the late-1930s, *Achnanthes sensu lato* and naviculoid taxa declined to negligible abundances while *S. pinnata* increased from ~70% to >90% (Fig. 3). The declines in *Achnanthes sensu lato* and naviculoid taxa were clearly tracked by the PCA axis 1 samples scores. Additionally, the diatom species richness, diversity, and C:D declined substantially when gold mining operations began in the region (Fig. 3). For instance, the pre-mining averages for richness, diversity, and C:D were ~32, ~2.1, and ~9.4, respectively. However, during the mining era (late-1930s to early-2000s), the average richness, diversity, and C:D decreased to ~ 14, ~1.2, ~3.1, respectively. In the most recent sediments (~post-2000), the relative abundances of certain pennate (*Asterionella formosa* Hassall, *Fragilaria tenera* (W. Smith) Lange-Bertalot, *Fragilaria nanana* Lange-Bertalot) and centric (*Discostella stelligera* (Cleve & Grunow) Houk & Klee) planktonic diatoms have increased in abundance and this taxonomic shift was captured by the PCA axis 2 sample scores (Fig. 3). The diatom species richness and diversity in the most recent sediments (~20 and ~1.9, respectively) were higher relative to the mining era; however, the C:D did not show notable signs of recovery (~3; Fig. 3). The whole lake primary production (inferred from trends in VRS Chl-*a*) at Kam Lake has been increasing gradually since the mid-to late- 20th century and the highest values were recorded in the most recent sediments (Fig. 3).

3.4. Yellowknife air temperature record

The mean annual and summer air temperatures have increased notably over the past ~70 years in Yellowknife (Supplement 3). The mean annual air temperature in Yellowknife has increased by 2.63 °C while the mean summer air temperature has increased by 1.78 °C between 1940 and 2016 (Supplement 3). Furthermore, the mean annual and summer air temperatures during the most recent decades (post-1990s) were generally higher than the previous decades (Stewart et al., 2018).

4. Discussion

4.1. Limnology and recent diatom assemblages of lakes around Con Mine

The present chemical conditions and biological assemblages in lakes around Con Mine tracked the ecological legacies of 20th century gold mining operations in the region. Specifically, our three study lakes had some of the highest concentrations for surface water ionic compounds (e.g. specific conductance, total dissolved solids, Ca^{2+}, Cl^-, K^+, Mg^{2+}, Na^+) and metal(loid)s (e.g. Al, As, Ba, Cu, Fe, Li, Ni, Sb, Sr) in the Yellowknife area (Table 1). While the high concentrations of arsenic and antimony can be attributed to the roaster stack emissions at Giant and Con mines (Cheney et al., 2020; Palmer et al., 2019), the elevated levels of other metal(loid)s and salinity-related variables (e.g. specific conductance, Cl^-, Na^+) are a legacy impact of salt-rich tailings from Con Mine (Falk et al., 1973). These lakes were sampled during the ice-covered period, which

Table 2
The most common diatom taxa in the surface sediments (i.e. taxa in greater than 5% abundance) of Kam, Keg, and Peg lakes .

Lake	*Achnanthes thermalis*	Benthic fragilarioid taxa	*Navicula incertata*	*Navicula libonensis*	*Nitzschia* taxa	Planktonic taxa	*Stauroneis tackei*
Kam	0	75	0	0	0	16	0
Keg	10	40	5	6	19	0	15
Peg	19	71	5	0	0	0	0

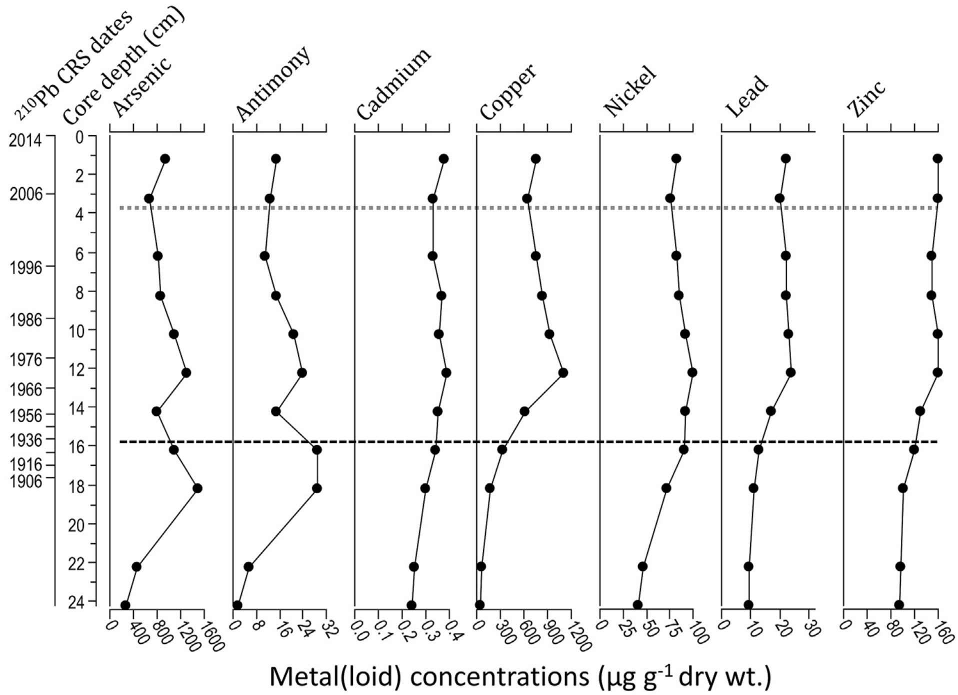

Fig. 2. Sedimentary metal(loid) concentrations (weight per gram dry weight) scaled by core depth (with CRS estimated ^{210}Pb dates plotted secondarily) from Kam Lake. The horizontal black-dashed and grey-dashed lines indicate the onset and end of mining activities at Con Mine, respectively.

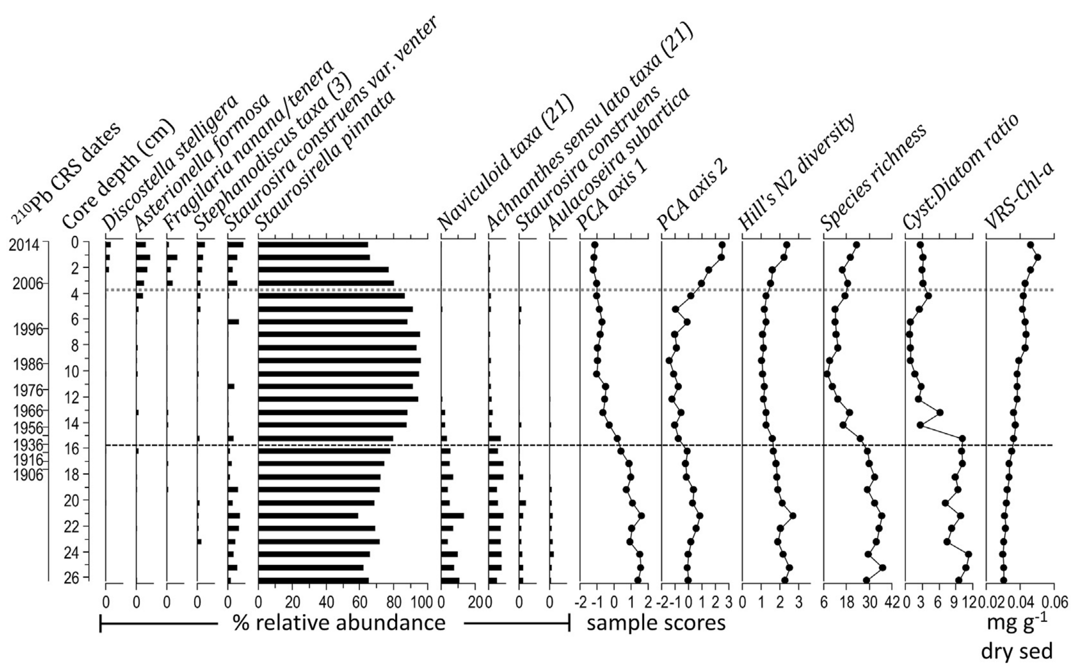

Fig. 3. Diatom assemblage profiles scaled by core depth (with CRS estimated ^{210}Pb dates plotted secondarily) showing relative abundances of the most common taxa from Kam Lake. For clarity of display, species belonging to *Achnanthes sensu lato* and naviculoid taxa that displayed similar trends through time were grouped together. The sedimentary PCA axis 1 and 2 sample scores, Hill's N2 species diversity, species richness, chrysophyte cyst to diatom valve ratios, and trends in VRS Chl-*a* (mg g^{-1} dry weight) are also included in the stratigraphy. The horizontal black-dashed and grey-dotted lines indicate the onset and end of mining activities at Con Mine, respectively.

would result in elevated salinity levels compared to the open-water season due to solute-exclusion from lake-ice formation (Palmer et al., 2019). However, field observations during sampling noted that the ice was slushy on Keg and Peg lakes, an observation unique to these two lakes in a sampling campaign of more than 20 lakes from the region. This suggests that the anomalously high concentrations of ionic compounds from mine tailings may have affected lake-ice formation.

At the time of sampling (2014), a decade had passed since mining activities ceased at Con Mine. Nonetheless, the concentrations for many contaminants were still high. This provides some indication that the chemical recovery of these lakes has not occurred and the local drainage, particularly via Pud Lake, may potentially still be acting as an important source of mine tailings via connecting streams to these lakes. Additionally, the concentrations of conservative ions (i.e. Mg^{2+}, Na^{+}, K^{+}, Cl^{-}) were extremely high at Keg and Peg lakes, which would be expected as these ions are not readily sequestered into the sediments and are minimally influenced by microbial metabolism (Wetzel 2001). Hence, in these highly saline environments, certain extremophiles can thrive while others succumb to the effects of extreme salinity. For instance, halophilic *A. thermalis* (Last et al., 1998) and *N. incertata* (Gell et al., 2002) are rarely reported from freshwater sub-Arctic lakes; however, at Keg and Peg lakes they occurred in greater than 5% relative abundance, highlighting the strong influence of salt-rich mine tailings on the biota of these lakes. Similarly, *N. libonensis*, a taxon tolerant of high copper concentrations and conductivity (Laing et al., 1999), also occurred in greater than 5% abundance at Keg Lake, providing further evidence of long-lasting biological responses to salinization and metal pollution of lakes around Con Mine. These bioindicators and the water chemistry data suggest that the mine tailings from Con Mine have substantially altered these ecosystems.

4.2. Long-term impacts of mining pollution on Kam Lake

Metal(loid) concentrations in lake sediments are important tools to track the impacts of mining activities on aquatic ecosystems; however, sedimentary trends in redox-sensitive elements suspected of post-depositional mobility should be viewed with caution and, when possible, compared to elements that are less mobile in sediments (e.g. lead, zinc; Cheney et al., 2020; Outridge and Wang, 2015). In Yellowknife, for example, gold mining activities resulted in the emissions of arsenic and antimony (Cheney et al., 2020; Palmer et al., 2019); however, both of these elements are redox-sensitive and may become unbound from organic matter and other minerals in the sediments under anoxic conditions, consequently becoming mobile within the sediment column (Bose and Sharma, 2002; Chen et al., 2003). Indeed, previous paleolimnological assessments from Yellowknife have noted that redox-sensitive elements have been mobile in lake sediments (Van Den Berghe et al., 2018) as we observed in the sediments of Kam. Unlike arsenic and antimony, the concentrations of less mobile element lead and other metals associated with mine tailings from Con Mine (i.e. Cu, Ni, and Zn; Falk et al., 1973) began increasing around 15 cm depth in the core when gold mining operations started in Yellowknife. This suggests that the sediments of Kam are indeed tracking the known inputs of mine tailings from Con Mine. Therefore, the earlier increase in redox-sensitive arsenic and antimony around 18 cm were likely a result of post-deposition mobility in the sediments. However, sedimentary metal(loid) analyses of a longer sediment core, along with data on hypolimnetic and sediment redox conditions, coupled with pore-water chemistry analyses, would be necessary to determine the dynamics of arsenic and antimony in the sediments of Kam Lake.

Staurosirella pinnata, a benthic fragilarioid taxon ubiquitously present in circumpolar lakes around the world (Jones and Birks, 2004; Laing et al., 1999; Solovieva et al., 2005; Smol and Douglas 2007), dominated the diatom assemblages at Kam Lake throughout the entire sedimentary record. However, its increase during the mining era was consistent with spatial surveys of present diatom assemblages in arsenic-contaminated lakes around Yellowknife and Cobalt (Ontario), where benthic fragilarioid taxa were dominant (Sivarajah et al., 2019; Little et al., 2020). Furthermore, *S. pinnata* increased relative to benthic *Achnanthes sensu lato* and naviculoid taxa that are known to tolerate metal(loid) polluted waters (Hirst et al., 2002; Salonen et al., 2006; Morin et al., 2012). While this was surprising, it provides some indication that benthic diatom responses to the combined effects of salinization and metal(loid) pollution may differ from metal(loid) contamination alone. Hence, the increase in the abundance of *S. pinnata* during the mining era at Kam and the general dominance of benthic fragilaroids in the recent sediments of Keg and Peg lakes suggest that these pioneering taxa in northern lakes may also tolerate relatively high salinity and metal(loid) pollution.

Diatom species diversity and richness are useful indicators of environmental pollution; however, these indices should be used with caution when interpreting biological responses to metal(loid) contamination as they are often influenced by multiple factors (De Laender et al., 2012). In this study we used these biological indices as complementing lines of evidence to the taxon-specific shifts discussed previously. For instance, a variety of *Achnanthes sensu lato* and naviculoid taxa occurred in low abundances in the pre-mining sediments and then disappeared during the mining era, which was tracked by the sharp declines in species richness. Unlike species richness, only modest declines in Hill's N2 diversity were observed during the mining period. This is because Hill's N2 diversity considers both the number of species and evenness (i.e. how abundant taxa are in a sample), and it was generally lower throughout the sedimentary record of Kam Lake as *S. pinnata* dominated the assemblages. Although the ratio of chrysophyte cysts to diatom valves (C:D) has not often been explored as a tool to assess the impacts of mining pollution, in a spatial assessment of saline temperate lakes in British Columbia (Canada) Cumming et al. (1995) suggested that chrysophyte cysts are rare or not observed in mesosaline and hypersaline lakes. Hence, the sharp declines of cysts during the mining era was likely a response to the salinization of Kam Lake from salt-rich mine tailings. The general synchrony in the timing of the decreases in diatom richness, diversity and C:D, and changes in taxonomic composition suggests that salinization and metal(loid) contamination have substantially altered the algal assemblages during the 20th century.

4.3. Multiple stressors influence recent biological assemblages at Kam Lake

Metal(loid) concentrations continue to be elevated at Kam Lake; however, the diatom assemblage shifts observed around the turn of the 21st century were indicative of climate-mediated changes to lake thermal properties and nutrient enrichment from development in the catchment. In particular, the increase in the relative abundance of certain planktonic diatoms (e.g. *A. formosa*, *F. tenera*, *F. nanana,* and *D. stelligera*) in the most recent (~post-2000) sediments was likely a response to regional climate warming, as these diatom shifts occurred during the warmest recorded years in Yellowknife (Supplement 3) and because similar increases in planktonic taxa have been observed in other lakes from Yellowknife (Sivarajah et al., 2020) and elsewhere (Winder et al., 2009; Rühland et al., 2015). Specifically, shorter-ice cover, longer and stronger thermal stratification, and weaker water column mixing often favor certain pennate and centric planktonic diatom taxa (Rühland et al., 2015). This is because, amongst other characteristics, these taxa have relatively high surface area to volume ratios that allow them to remain longer in the photic zone and maximize nutrient uptake and reproduction during periods of longer thermal stratification and/or longer ice-free periods (Ptacnik et al., 2003; Winder et al., 2009).

Increases in eutrophic diatom taxa were not observed during the 1970s, when raw sewage was routed into the lake, albeit only briefly. However, eutrophic *Stephanodiscus* taxa increased in the most recent sediments, which may be a response to additional urbanization (e.g. development of businesses, dog kennels) around

Kam Lake. Furthermore, the gradual increase in whole lake primary production (inferred by trends in sedimentary chlorophyll *a*) is likely a response to both warming mediated lengthening of growing season and urbanization. Similar observations have been made in urban lakes around Yellowknife (Sivarajah et al., 2020) and other high latitude lakes where longer growing seasons have resulted in higher overall primary production (Michelutti et al., 2005; Griffiths et al., 2017).

Although diatom species richness and diversity have increased in the most recent sediments of Kam Lake, the species composition is markedly different from the pre-mining era. Increases in diversity indices may not always represent biological recovery to pre-impact conditions (Gray and Arnott 2009), and this is particularly important to consider in systems where multiple environmental stressors may be influencing biological assemblages in the post-mining era. In fact, our observations are consistent with a growing number of studies from Yellowknife and other Northern regions where mining activities have substantially altered limnological characteristics, impacting aquatic biota across multiple trophic levels (e.g. Thienpont et al., 2016; Gavel et al., 2018; Leppänen et al., 2019; Sivarajah et al., 2020; Persaud et al., 2021). Collectively, these investigations demonstrate that the biota of mine-impacted Northern freshwaters are now vulnerable to the impacts of multiple environmental stressors, particularly those related to urbanization and climatic changes.

5. Conclusions

The anomalously high ionic and metal(loid) lakewater concentrations in our study sites highlight the complex long-term limnological legacies of gold mining operations in this region. Furthermore, the taxonomic composition of the present-day diatom flora, where taxa indicative of saline and metal(loid) polluted waters are present, suggest that the biota of freshwaters around Con Mine were severely impacted by both roaster stack emissions of metalloids and tailings drainage via Pud Lake. At Kam Lake, the increases in the relative abundance of benthic *S. pinnata* and concomitant decreases in diatom species richness, diversity and C:D during the mining era were key biological responses to salinization and metal(loid) contamination from gold mining activities. The algal assemblages at Kam Lake have not recovered to pre-mining conditions and biological recovery trajectories at this lake are further influenced by urbanization and regional climatic warming as has been observed in the freshwaters of other mine-impacted regions.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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Author statement

Madi Perrett: Investigation, Formal analysis, Writing – original draft; Branaavan Sivarajah: Conceptualization, Formal analysis, Visualization, Writing – original draft; Cynthia L Cheney: Investigation, Writing – review & editing; Jennifer B Korosi: Conceptualization, Writing – review & editing; Linda E. Kimpe: Investigation, Writing – review & editing; Jules M. Blais: Funding acquisition, Resources, Supervision, Writing – review & editing; John P. Smol: Funding acquisition, Resources, Supervision, Writing – review & editing.

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