A paleolimnological approach for interpreting Aquatic Effects Monitoring at the Diavik Diamond Mine (Lac de Gras, Northwest Territories, Canada)

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Abstract

A paleolimnological assessment of Lac de Gras (Northwest Territories, Canada) showed pronounced aquatic ecological and biogeochemical changes occurring since at least ~1950, well before diamond mining operations began in 2000. These changes are likely a response to regional climate warming, which is confounding the interpretation of an Aquatic Effects Monitoring Program (AEMP) intended to identify early-warning indicators of diamond mining impacts to water quality. In the latest AEMP report, action level exceedances based on three years of monitoring in Lac de Gras were reported for chlorophyll *a* and strontium, yet sediment cores collected from three different sites in the lake exhibited notable increasing trends in these parameters since pre-1950. Increases in the small, centric diatom *Discostella pseudostelligera* have also been occurring since pre-1950, which we infer to be a response to climate warming. Recent (post-1996) *D. pseudostelligera* increases observed from aquatic effects monitoring of nearby small lakes have previously been linked to nitrogen fertilization from diamond mining. Thus, our paleolimnological results clearly indicate that parameters predicted to respond to mining impacts are also responding similarly to regional climate warming. Based on this, AEMP adaptive management strategies need to consider the potential additive or synergistic effects of mining and climate change when establishing action level exceedances for water quality and ecological indicators. Using our paleolimnological data, we calculated background (pre-mining) rates of change in key geochemical parameters that can provide a benchmark for evaluating ongoing changes in the current mining period, and for establishing AEMP significance thresholds.

Key words: Barren Lands tundra; climate change; diamond mining; diatoms; metals; nitrogen; subarctic

Diamond-bearing kimberlite pipes were first discovered in northern Canada in 1991, in a vast and remote tundra landscape known colloquially as the Barren Lands (Northwest Territories). The Ekati Diamond Mine opened in 1998 under BHP Billiton Canada Inc. as the first commercial diamond mine in Canada, and is one of three diamond mines operating in the Barren Lands today. To obtain a Water License to operate in the Northwest Territories (a jurisdiction with its own government within Canada), the mines are required to implement an Aquatic Effects Monitoring Program (AEMP) to determine the short- and long-term impacts of mining activities on surface water quality, intended to guide the implementation of mitigation strategies (MVLB/GNWT 2019). AEMPs include the routine monitoring of water and sediment chemistry, plankton, benthos, and fish over the duration of the project, in impacted surface waters and reference sites. AEMPs also typically include 1-2 years of baseline monitoring conducted prior to the onset of operations.

Of particular concern is the potential for increases in total suspended and dissolved constituents (Rollo and Jamieson 2006) resulting from the release of diamond mine discharge waters, including some ions and trace elements (e.g. chloride, copper, aluminum) that can have toxic effects on aquatic biota (ERM 2014). As part of mining operations, explosives are used to fragment rock, and waste rock is commonly stockpiled on site. Dust, as well as leaching and groundwater seepage from waste rock constituents and blasting residuals, can contribute phosphates, nitrate, and ammonia to nearby surface waters (Bailey et al. 2012, Vandenberg et al. 2016; Golder Associates 2018). The potential ecological impacts of nitrate and phosphorus inputs to surface waters are a central water quality concern for diamond mining activities in the Barren Lands, including Lac de Gras (Bailey et al. 2013, Golder Associates 2018). Phosphorus, typically the limiting nutrient in freshwater ecosystems, has been reported to be increasing in lakes receiving mine discharge waters (ERM 2014). Consequently, nutrients and measures of lake productivity (such as chlorophyll *a* and phytoplankton community composition) are also carefully monitored. In the 2017 AEMP report, based on the previous three years of monitoring in Lac de Gras, documented increases in chlorophyll *a* triggered Action Level 2, defined as trending towards a pre-established significance threshold that indicates departure from baseline conditions (Golder Associates 2018).

Climate warming is an important, independent driver of Arctic limnological change that may confound the interpretation of AEMP data (Vincent et al. 2013). Although AEMPs include 1-2 years of baseline monitoring, this is likely not sufficient for characterizing the limnological responses to climate warming, as the instrumental record shows air temperatures in the Barren Lands have been steadily increasing since ~1960 (Mullen et al. 2017), and paleolimnological records show striking lake ecosystem changes beginning in ~1850 (Rühland et al. 2005). For example, several small lakes receiving discharge waters from the Ekati Diamond Mine exhibited increases in *Cyclotella* (small, centric diatom taxa) over the course of 19 years of AEMP data, which has been interpreted as a response to nitrogen discharge from the mine (St. Gelais et al. 2017). However, widespread increases in *Cyclotella* have also been reported in lakes across the northern hemisphere as a response to climate warming (Rühland et al. 2015), including in Slipper Lake, the most distal in the chain of small lakes receiving discharge waters from Ekati (Rühland et al. 2005). Based on Ekati AEMP data, *Cyclotella* increased in Slipper Lake after 2005 (St. Gelais et al. 2017), while a paleolimnological analysis of Slipper Lake conducted on a sediment core collected in 1997 (prior to the mine opening in 1998) recorded an abrupt increase in *Cyclotella* beginning in the 19th century (Rühland et al. 2005).

The discrepancy between the monitoring and paleolimnological records of Slipper Lake clearly demonstrates the importance of timescales in understanding drivers of limnological change and the challenges associated with attempting to understand mining impacts on northern aquatic ecosystems that are being transformed as a result of climate warming. Thus, paleolimnological studies have much to contribute to our understanding of the impacts of diamond mining activities on the tundra lakes of the Barren Lands because they provide an indirect mechanism for identifying trajectories of environmental change in response to climate signals that began before the onset of mining impacts. In this study, we collected sediment cores from three locations in Lac de Gras, a large, cold monomictic lake where the Diavik Diamond Mine is located (Figure 1). We performed sedimentary inferences of geochemical and ecological conditions prior to the onset of mining, in order to interpret water quality changes reported by the Diavik AEMP in the context of long-term limnological trends occurring in Lac de Gras in response to climate warming. We measured trace elements (e.g. aluminum, copper, strontium, lead, and zinc) because mine discharge waters and dust from waste rock piles are potential sources of these elements to nearby surface waters, and AEMP excedences have been reported for aluminum, copper, silicon, antimony, and strontium, among others. We also analyzed temporal trends in chlorophyll and nitrogen isotopes to identify a possible eutrophication signal related to nitrogen pollution. Finally, subfossil diatoms were also analyzed, as diatom species responses to climate warming and nutrient pollution are well established based on previous studies (Malik et al. 2018; Rühland et al. 2015; Saros et al. 2014).

Study Site

The Lac de Gras watershed is located ~50 km north of treeline and ~300 km northeast of the town of Yellowknife, within the Wek’èezhìi area (Figure 1). The area is part of an historic land claim agreement with the Government of Canada, known as the Tłı̨chǫ Agreement (2005), which recognizes the land, resources, and self-government rights of the Tłı̨chǫ peoples. It is in the Slave Geological Province and includes Precambrian granitic, gneissic, metasedimentary, and metavolcanic rocks (Geological Survey of Canada 2014). The Lac de Gras kimberlite field is of Late Cretaceous to Eocene age (Sarkar et al. 2015), and supports the Dominion Diamond Corporation’s (DDC’s) Ekati Diamond Mine (operational since 1998) and the DDC-RioTinto Diavik Diamond Mine (operational since 2000). The area is characterized by low topographical relief, thin surficial tills, glaciofluvial sediments (Dredge et al. 1999; Hu et al. 2003), continuous permafrost (Heginbottom et al. 1995), and an abundance of lakes and streams (Basar et al. 2012). Vegetation consists primarily of dwarf birch (*Betula glandulosa*), willow (*Salix* spp.), and northern Labrador tea (*Rhododendron tomentosum;* Ritchie 1993). Climate is continental, with short cool summers and long cold winters (Hu et al. 2003). Mean annual air temperature recorded at the Ekati Diamond Mine (Figure 1) from October 2016 to September 2017 was -7.9 C (15 C in July; -26.1 C in February), and total annual precipitation is 236.5 mm, with 34% falling as snow (ERM 2018).

Lac de Gras is the headwater of the Coppermine River, which flows north and discharges into the Arctic Ocean. It is a large lake, with a surface area of 569 km^2, an average depth of 12 m, a maximum depth of 56 m, and is typically ice-covered from approximately late-October to late-June (Deton’ Cho Stantec 2015). Limnological depth-profile data for temperature was collected monthly from June to September between 1996-2009 (albeit from different sampling sites on the lake, all roughly 20 m in depth) show that the maximum surface water temperatures in summer are 10°C and the lake does not exhibit thermal stratification at least in the top 20 m during the open-water season (Deton' Cho Stantec 2015). Lac de Gras is an ultraoligotrophic, slightly acidic to circumneutral, dilute, and clear lake (Table 1). The Diavik Diamond Mine operates on the East Island (Figure 1), and effluent flows into Lac de Gras through two diffusers at the North Inlet Water Treatment Plant. Annual estimates of discharge volumes from the Diavik Diamond Mine have been estimated to range from approximately 4,000,000,000 to 12,000,000,000 L from 2002 to 2013 (Deton' Cho Stantec 2015). The Ekati Mine is located ~15 km north of Lac de Gras, and mine waste waters discharge into Lac de Gras at the Slipper Lake and Lac du Sauvage outlets. Modeled annual effluent discharge volumes at the Slipper Lake outlet is estimated at 12,000,000,000 to 51,000,000,000 L annually from 2000 to 2013, and flows through a chain of seven lakes (including Slipper Lake) before it discharges into the northwest arm of Lac de Gras (Deton' Cho Stantec 2015). The total residence time in the series of lakes between the Ekati containment facility and its discharge into Lac de Gras is estimated to be 324 days (Rescan 2012). Data are unavailable for effluent discharge from Ekati's Misery pit into the Lac de Sauvage outlet at the eastern arm of Lac de Gras. Dust deposition rates are lower than the British Columbia dustfall objective for the mining industry, and ranged from an average of 354 mg dm^{-2} yr^{-1} for sites within 100 m of the mine to 139 mg dm^{-2} yr^{-1} for sites located 250-1000 m from the mine (Golder Associates 2018). Water chemistry, plankton, and benthos have been monitored at several sites in Lac de Gras regularly since 2001.

Monitoring of chlorophyll *a*, phytoplankton and zooplankton biomass, total phosphorus, and total nitrogen indicate some nutrient enrichment has occurred since the opening of the mine, as spatial gradients evident in these parameters are evident with distance from mine effluent discharge sites and the spatial extent of dust deposition (Golder Associates 2018).The magnitude of the effect in chlorophyll *a* triggered an Action Level 2 designation, which requires the establishment of an Effects Benchmark (Golder Associates 2018). Water hardness, conductivity, chloride, sulphate, and strontium showed significant increasing trends lake-wide over the period of water quality monitoring (Deton' Cho Stantec 2015). Concentrations of ammonia, lead, and tin were greater at mid-field sampling stations potentially affected by dust deposition compared to reference conditions (Golder Associates 2018).

Table 1 – Lac de Gras sediment coring sites. Lake parameters and water chemistry data from Lac de Gras sampling sites collected by the Dominion Diamond Corporation Diavik Aquatic Effects Monitoring Program. Data were collected in August, 2014, with the exception of pH and conductivity, which were collected in August, 2011. Data are accessible from the Wek'èezhìi Land and Water Board public registry.

Site	**Latitude (°N)**	**Longitude (°W)**	**Depth (m)**	**pH**	**Specific Cond. (μS cm^{-1})**	**TN (mg L^{-1})**	**TP (mg L^{-1})**	**TDS (mg L^{-1})**	**Alkalinity (mg L^{-1} $CaCO_3$)**
NF3	64°30.586'	110°14.741'	19.1	6.6	27.3	0.177	0.003	20.0	8.2
MF 2-3	64°31.562'	110°08.688'	20.7	7.0	26.2	0.128	0.005	17.3	8.8
MF 3-4	64°26.837'	110°19.857'	18.2	7.4	10.6	0.143	0.002	13.3	6.4

TN = total dissolved nitrogen
TP = total dissolved phosphorus
TDS = total dissolved solids

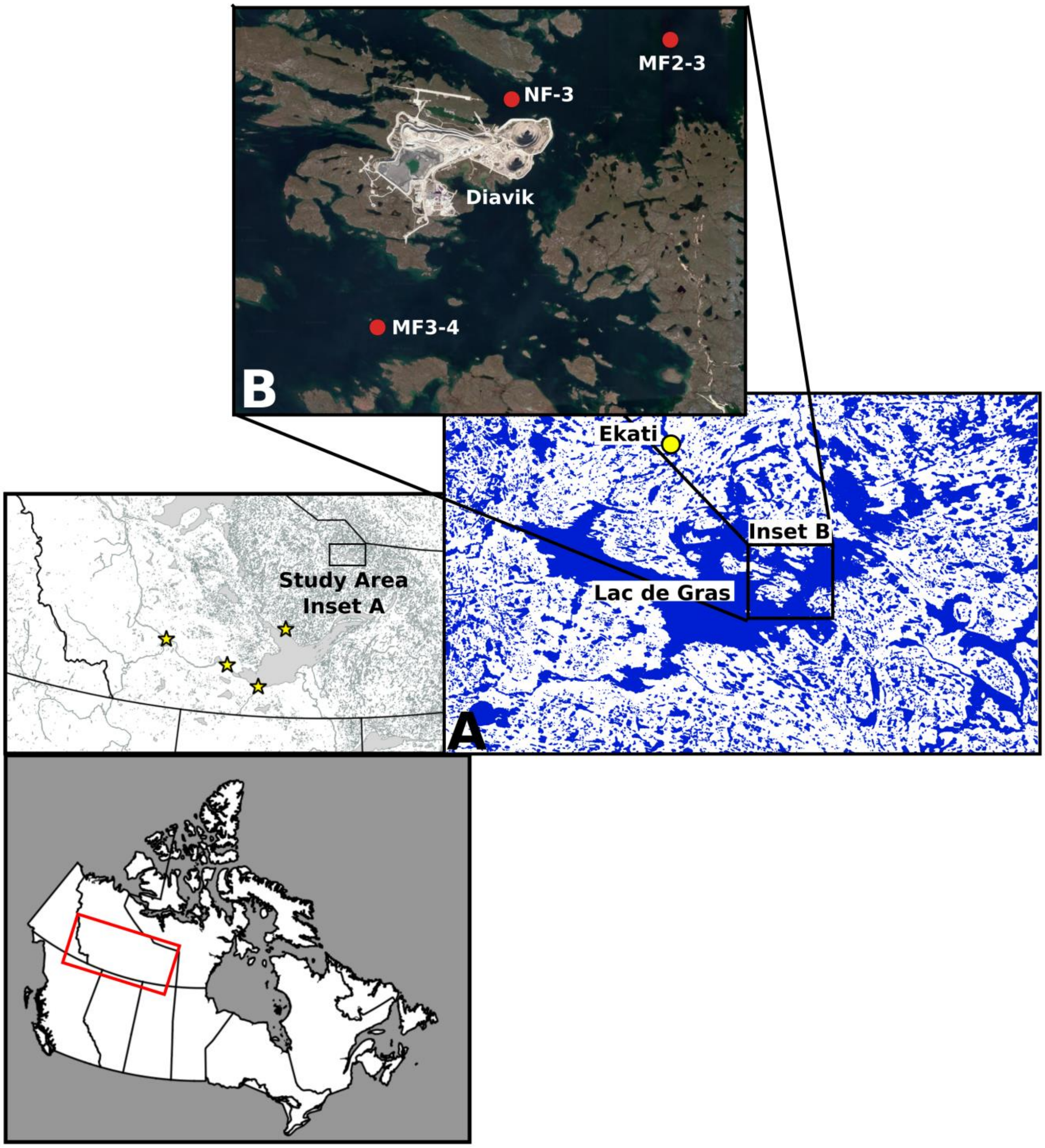


Figure 1 – **Study site locations**. Map showing the location of Lac de Gras within the Northwest Territories of Canada (Inset A), and the locations of the Diavik Diamond Mine and the three sediment coring locations within Lac de Gras (Inset B). Yellow stars represent the locations of major population centres in the Northwest Territories, including the town of Yellowknife (northernmost star).

Methods

Field Methods

Three coring locations were selected from among the sites routinely monitored for aquatic biota, water, and sediment chemistry as part of Diavik's AEMP (Golder Associates, 2018). Site Near Field 3 (NF3; depth=19.1 m) is located nearest to the effluent diffuser, and thus considered to have the greatest exposure to mine impacts. Sites Mid Field 2-3 (MF2-3; depth=20.7 m) and Mid Field 3-4 (MF3-4; depth=18.2 m) are both located mid-field to the effluent diffuser, surrounding the East Island where Diavik Diamond Mine is located, and considered to have intermediate exposure to the mine. One sediment core (18.5-23.5 cm in length) was collected from each of three locations in Lac de Gras (Figure 1; Table 1) in August 2014 using a gravity corer (UWITEC, Austria). Sediment cores were sectioned into 0.25 cm intervals on site using a modified Glew vertical extruder (Glew 1988). Sediment cores were shipped frozen to the University of Ottawa and freeze-dried prior to further analyses.

Laboratory Methods

Cores were ^{210}Pb dated using an Ortec high purity germanium Gamma Spectrometer (Oak Ridge, TN, USA) following methods in Appleby (2001). Certified Reference Materials obtained from the International Atomic Energy Association (Vienna, Austria) were used to calibrate and perform efficiency corrections on the gamma spectrometer. Results were analyzed using ScienTissiME, a Matlab-based program (Barry's Bay, ON, Canada). A chronology was established using the Constant Flux Constant Sedimentation Rate (CFCS) model (Appleby 2001), which gave near identical results to constant rate of supply model (Appleby and Oldfield 1978). Approximately 0.5 g of freeze-dried sediments was shipped to SGS Minerals Services in

Lakefield, Ontario, a Canadian Association for Laboratory Accreditation (CALA) accredited facility, for analysis of total metals. Briefly, samples were digested using microwave-assisted aqua regia and analyzed using inductively-coupled plasma mass spectrometry. In order to reconstruct trends in algal production, visual reflectance spectroscopy (VRS) was used to infer sedimentary chlorophyll *a* following methods in Michelutti et al. (2005, 2016), a procedure that incorporates estimates of chlorophyll *a* and its diagenetic products. Rates of percent change through time were calculated for geochemical parameters also included in Lac de Gras AEMP monitoring. Rates of percent change were calculated first by calculating the % change between adjacent sediment intervals, then dividing that by the difference in the age of the intervals.

For analysis of % organic carbon and % nitrogen, freeze-dried sediment was acid-fumigated with concentrated HCl for 48 hrs in an acid desiccator to remove inorganic carbon (Harris et al. 2001). Sample vials were then filled with deionized water, centrifuged at 3000 rpm for 10 min and the supernatant discarded. This step was repeated twice more, and then samples were freeze-dried again. Approximately 5-10 mg of sample were weighed into tin capsules containing 20 µg tungsten trioxide and analyzed using a Vario EL III Elemental Analyzer (Elementar, Germany) at the Ján Veizer Stable Isotope Laboratory (University of Ottawa), following methods described in Brazeau et al. (2013). For $\delta^{15}N$ analysis, 15-100 mg of untreated sediment was weighed into tin capsules (weight dependent on %N content of sediment samples) and loaded into the Elemental Analyzer interfaced to the DeltaPlus XP Isotope Ratio Mass Spectrometer (ThermoFinnigan, Germany) at the Ján Veizer Isotope Laboratory. Calibrated internal standards were used for normalization of the data, and precision was better than 0.20‰ (Pella 1990).

Subfossil diatoms were analyzed in the sediment cores collected from a near-field site (NF3) and one of the mid-field sites (MF2-3). Diatoms were isolated from the sediment matrix following methods outlined in Battarbee et al. (2001). Briefly, sediments were digested with a 1:1 mixture of nitric and sulfuric acids that was placed in a water bath at 80 C for 3 hrs. Samples were then neutralized by adding fresh deionized water daily for 7 days until a neutral pH was reached. Samples were then plated onto microscope coverslips to dry, and then mounted onto microscope slides using Naphrax®. Diatoms were identified to species-level on an AmScope T690C-PL light microscope at 1000x magnification using Krammer & Lange-Bertalot (1991, 1997, 1999, 2000) and Fallu et al. (2000) as taxonomic guides. A minimum of 400 diatom valves were counted per interval. A principal components analysis was conducted on the diatom stratigraphy data (a separate PCA for each sediment core) in order to visualize trends in overall species assemblage through time.

Results

^{210}Pb Dating

The three Lac de Gras cores exhibited an exponential decay in total ^{210}Pb with depth, which reached background ^{210}Pb (i.e. no more unsupported ^{210}Pb was present) at ~4-5 cm (Figure 2), with the entire current mining period included in the top 0.5-0.75 cm. In each core, a slight increase in ^{137}Cs was observed in the top 3 cm, with no distinct ^{137}Cs peak (Figure 2). Sedimentation rates were estimated at 0.0138 ± 0.0017 g cm^{-3} yr^{-1} in MF2-3 and NF3, and 0.00138 ± 0.0001 g cm^{-3} yr^{-1} in MF3-4. The basal sediment dates based on the CFCS age model were as follows: NF3=1395±77 CE, MF2-3=1050±43 CE, MF3-4=415±43 CE.

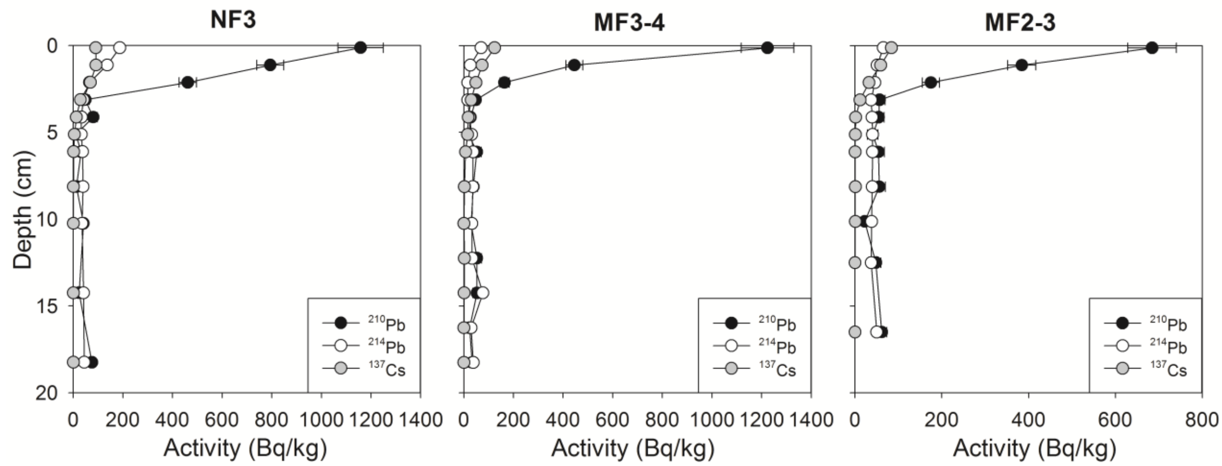


Figure 2 – Results of 210Pb gamma dating of Lac de Gras cores. Radioisotopic activities for 210Pb, 214Pb, and 137Cs in sediment cores collected from three sites (NF3, MF2-3, MF3-4) in Lac de Gras. Chronology and inferred sedimentation rates were determined using the Constant Flux Constant Sedimentation Model (Appleby and Oldfield 1978).

Geochemical Analysis

NF3 (Near-Field)

Concentrations of arsenic (As), copper (Cu), aluminum (Al), and iron (Fe) were relatively stable throughout the sediment core, while manganese (Mn), strontium (Sr), and zinc (Zn) increased above a core depth of 3.0 cm to the surface interval, increasing from 1500 to 7100 µg g^{-1}, 7.8 to 18 µg g^{-1}, and 53 to 89 µg g^{-1}, respectively (Figure 3). Rates of change in Mn in the pre-mining period of increase (3.0 to 0.5 cm) ranged from 0.85 to 4.1 % yr^{-1} (mean=2.25, SD=1.64), compared to 0 to 8.55 % yr^{-1} (mean=3.95, SD=3.51) in the current mining period (top 0.5 cm). Rates of change in Sr in the pre-mining period of increase (4.0 to 0.5 cm) ranged from 0.67 to 2.15 % yr^{-1} (mean=1.20, SD=0.58), compared to 0 to 4.91% yr^{-1} (mean=1.67, SD=2.30) in the current mining period (top 0.5 cm). Rates of change in Zn in the pre-mining period of increase (4.0 to 0.5 cm) ranged from -0.25 to 2.01 % yr^{-1} (mean=0.62, SD=1.01), compared to 0 to 2.81 % yr^{-1} (mean=1.52, SD=1.23) in the current mining period (top 0.5 cm). Lead (Pb) concentrations also increased between 3.0 and 0.5 cm, from 4.9 to ~15 µg g^{-1}, and stabilized during the current mining period (Figure 3). Rates of change in Pb in the pre-mining period of increase (4.0 to 0.5 cm) ranged from 0.3 to 4.55 % yr^{-1} (mean=2.24, SD=1.45), compared to -1.5 to 1.75% yr^{-1} (mean=0.46, SD=1.56) in the current mining period (top 0.5 cm). Sedimentary nitrogen (%N) and organic carbon (%TOC) increased above 12 cm to the surface interval, from 0.1 to 0.5% and 1.2 to 4.8%, respectively (Figure 4). The C/N ratio, a commonly used indicator of the relative fraction of terrestrial versus algal organic matter sources (Meyers and Teranes 2002), declined slightly above a core depth of 2.0 cm, from 13.2 to 11.5 (Figure 4). Sedimentary sulfur (%S) increased slightly above background in the current mining period (Figure 4). $\delta^{15}N$ exhibited a steady decrease from the bottom of the core to 0.75 cm (10.7 to 5.0‰), and increased again above 0.75 cm (the current mining period) to 6.3‰ in the surface interval (Figure 4). VRS-inferred chlorophyll *a* increased beginning at core depth of 2.0 cm (Figure 4). Rates of change in chlorophyll *a* in the pre-mining period of increase (2.0 to 0.75 cm) ranged from -4.5 to 44.78 % yr^{-1} (mean=11.35, SD=20.75), compared to 6.48 to 41.93 % yr^{-1} (mean=17.8, SD = 16.4) in the current mining period (top 0.75 cm).

MF3-4 (Mid-Field, Downstream)

Increases in Pb, Sr, Mn, Zn were observed above a core depth of 3.0 to 4.0 cm, from ~4-5 to ~9.0 µg g^{-1}, ~5-6 to 16 µg g^{-1}, 2300 to 6200 µg g^{-1}, and ~50 to ~70 µg g^{-1}, respectively (Figure 3). Rates of change in Pb in the pre-mining period of increase (4.0 to 0.5 cm) ranged from -0.1 to 0.9 % yr^{-1} (mean=0.4, SD=0.34), compared to -0.8 to 0.5% yr^{-1} (mean =-0.12, SD=0.94) in the current mining period (top 0.5 cm). Rates of change in Sr in the pre-mining period of increase (4.0 to 0.5 cm) ranged from 0.27 to 1.06 % yr^{-1} (mean=0.65, SD=0.28), compared to 0 to 1.82%

yr^{-1} (mean=0.91, SD=1.28) in the current mining period (top 0.5 cm). Rates of change in Mn in the pre-mining period of increase (3.0 to 0.5 cm) ranged from 0.18 to 2.3 % yr^{-1} (mean=0.95, SD=0.89), compared to 0 to 0.9% yr^{-1} (mean=0.44, SD=0.61) in the current mining period (top 0.5 cm). Rates of change in Zn in the pre-mining period of increase (4.0 to 0.5 cm) ranged from 0.05 to 0.58 % yr^{-1} (mean=0.22, SD=0.16), compared to -0.8 to 1.66 % yr^{-1} (mean=0.42, SD=1.75) in the current mining period (top 0.5 cm). The %TOC and %N increased steadily from 12 cm to the surface of the core, from 0.13 to 0.58% and 2.4 to 6.4%, respectively, while no trend was observed for %S (Figure 4). The C/N ratio decreased from 14 to 12 cm, and again from 6 cm towards the surface of the core, a total decrease from 21.4 to 12.8 (Figure 4). The $\delta^{15}N$ exhibited a steady decrease above a core depth of 4.0 cm, from 8.2 to 5.7‰ (Figure 4). VRS-inferred chlorophyll *a* increased above 2.0 cm, increasing above the lower limit of detection in the current mining period (Figure 4). Rates of change in chlorophyll *a* in the pre-mining period of increase (2.0 to 0.5 cm) ranged from -2.1 to 12.2 % yr^{-1} (mean=3.98, SD=5.68), compared to 0 to 15.5% yr^{-1} (mean=7.72, SD=11.00) in the current mining period (top 0.5 cm).

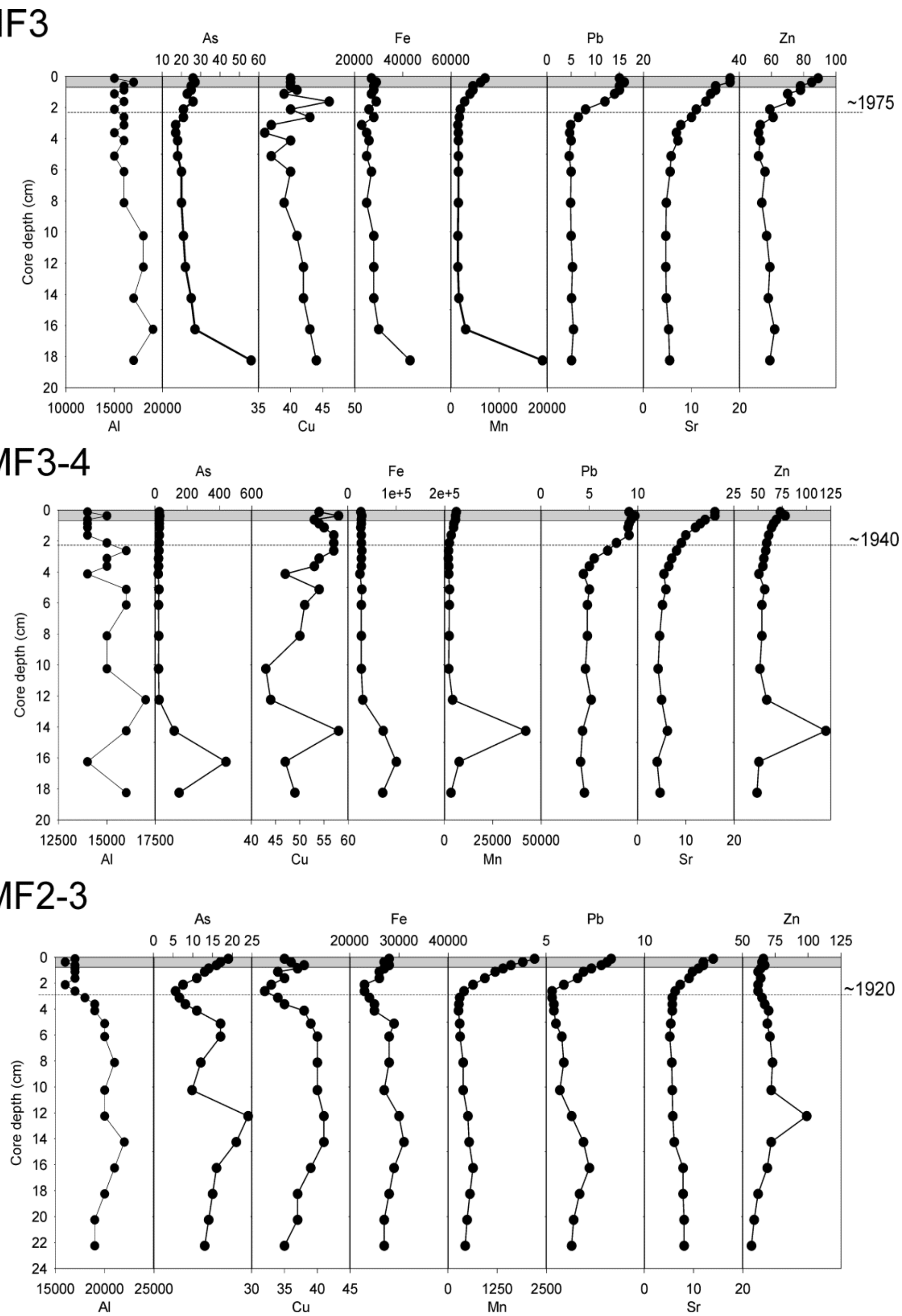
MF2-3 (Mid-Field, Upstream)

A slight decrease in Al concentrations was observed above a core depth of 8.0 cm, decreasing from 21000 μg g^{-1} to 17000 μg g^{-1} in the upper 2.0 cm, approximately 1950 (Figure 3). Arsenic concentrations were variable, and increased from 5.6 to 19 μg g^{-1} above 2.5 cm, which was still within the range of background for the sediment core (Figure 3). Concentrations of Pb and Sr increased above 2.0 cm, from 5.3 to 8.3 μg g^{-1}, and 7.3 to 14 μg g^{-1}, respectively (Figure 3). Rates of change in Pb in the pre-mining period of increase (2.0 to 0.75 cm) ranged from 0.3 to 1.1 % yr^{-1} (mean=0.60, SD=0.34), compared to 0.5 to 1.3 % yr^{-1} (mean=0.80, SD=0.41) in the current mining period (top 0.75 cm). Rates of change in Sr in the pre-mining period of increase (2.0 to 0.75 cm) ranged from 0.48 to 2.26 % yr^{-1} (mean=1.30, SD=0.76), compared to 0 to 3.08 % yr^{-1} (mean=1.6, SD=1.5) in the current mining period (top 0.75 cm). The %TOC and %N increased steadily above 6 cm towards the surface of the core, from 0.06 to 0.41% and 0.6 to 4.1%, respectively, and C/N decreased slightly between 14 to 12 cm, and again between 6 and 0 cm, for a total decrease from 13.8 to 11.5 (Figure 4). The %S increased slightly in the current mining period, from 0.12 to 0.15% (Figure 4). $\delta^{15}N$ exhibited a steady decrease from 14.2‰ at core depth 20 cm, to ~6-7‰ in the surface intervals (Figure 4). VRS-inferred chlorophyll *a* exhibited an increasing trend above 3.0 cm, increasing above the lower limit of detection in the current mining period (Figure 4). Rates of change in chlorophyll *a* in the pre-mining period of increase (3.0 to 0.75 cm) ranged from -7.6 to 51 % yr^{-1} (mean=11.7, SD=2.79), compared to -3.3 to 10.2 % yr^{-1} (mean=3.5, SD=9.64) in the current mining period (top 0.75 cm).

Diatoms

NF3

Distinct shifts in diatom assemblage structure occurred at NF3 prior to the opening of the Diavik Diamond Mine in the year 2000 (Figure 5). Diatom assemblages in the pre-industrial period were dominated by *Aulacoseira lirata* (30-40%), *Aulacoseira perglabra* (15-20%), and *Cyclotella ocellata* (15-20%), with *Cyclotella bodanica* var *lemanica* and *Encyonema herbridicum* present at 5-10% abundance. Above a core depth of 8 cm, *A. lirata, C. bodanica,* and *E. herbridicum* decreased in relative abundance, from ~30-40 to ~5-10%, 15-20% to 1-2%,



364 **Figure 3 –Temporal trends in sedimentary metal concentrations in Lac de Gras sediment**
365 **cores.** Stratigraphy showing downcore changes in several metals at three sites in Lac de Gras
366 along a gradient of impact from diamond mining operations. Concentrations are in µg g^{-1} dry
367 weight. The grey box represents the current mining period.

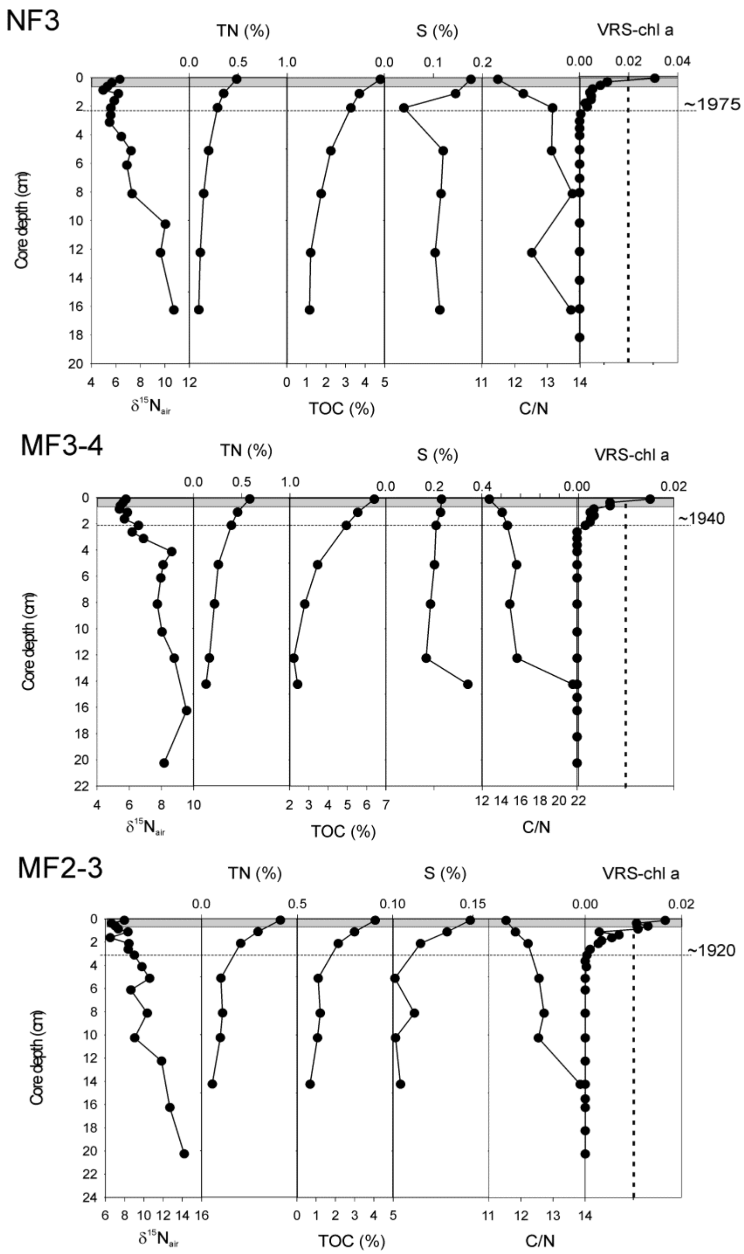


Figure 4 - Temporal changes in organic matter composition in Lac de Gras cores. Stratigraphy showing downcore changes in nitrogen stable isotope composition, % total nitrogen (TN), % total organic carbon (TOC), % sulfur (S), carbon: nitrogen ratio (C/N), and chlorophyll *a* inferred using visible reflectance spectroscopy (mg g^{-1} dry weight) in sediment cores from Lac de Gras along a gradient of impact from diamond mining operations. The grey box represents the current mining period. The dashed line represents the estimated lower method detection limit for VRS-chlorophyll *a* (Michelutti et al. 2005).

and 5-10% to 1-2%, respectively. *Discostella pseudostelligera*, *Microcostatus kuelbsii*, *Humidophila schmassmannii*, *Sellaphora* spp., and *Encyonema minutum* first appeared and increased in relative abundances above 8.0 cm. Above a core depth of 2.0 cm (~1975), further increases in *D. pseudostelligera* occurred, as well as increases in *Achnanthes acares*, small benthic *Fragilaria* spp., and *Psammothidium microscopicum*. In the current mining period (above core depth 0.75 cm), *D. pseudostelligera* further increased to become the dominant taxon at ~40% relative abundance, *M. kuelbsii* and *H. schmassmannii* decreased to <1%, and the ratio of chrysophyte scales:diatoms increased. Post-1950 diatom assemblage changes were most evident from PCA axis 2 scores.

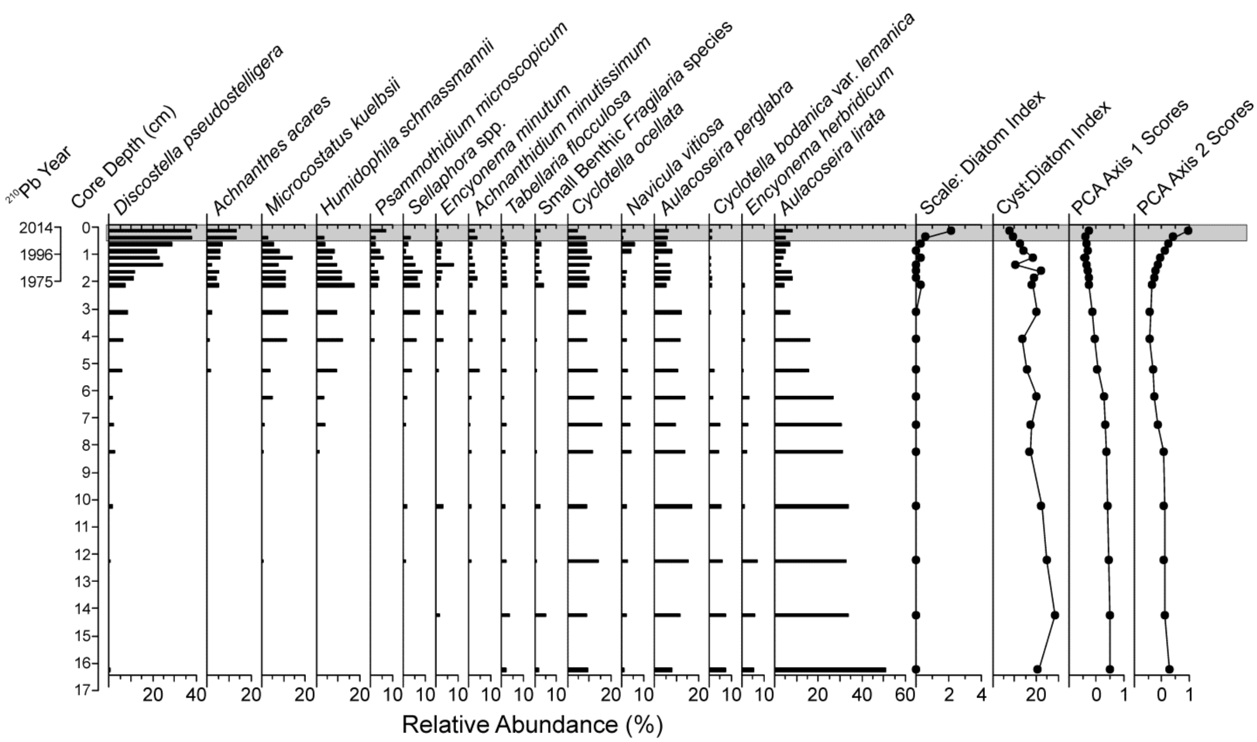


Figure 5 – **Temporal changes in diatoms at site NF3.** Stratigraphic profile of the most abundant diatom taxa (as relative abundances) in a sediment core collected from site NF3 in Lac de Gras, located near the effluent discharge site at the Diavik Diamond Mine. ^{210}Pb-inferred dates are shown as a secondary y-axis. Principal Components Analysis Axis 1 and 2 scores for diatom assemblages are also shown, as well as the ratio of chrysophyte scales to diatoms, and chrysophyte cysts to diatoms. The grey box represents the current mining period.

MF2-3

A shift in diatom assemblage occurred from 4.0 to 2.0 cm (Figure 6). Below core depth 4.0 cm, the assemblage was dominated by *A. lirata*, at relative abundances of 40-50%. *C. ocellate* (10-20%), *Encyonema* spp. (~10%), *Stauroneis anceps* (5-10%), *Frustulia rhomboides* (~5%), and *C. bodanica* (~5%) were also prevalent. At core depth 2.0 cm (~1950), decreases in relative abundance occurred for *A. lirata* (to ~10% relative abundance), *C. bodanica* (to <2%), *F. rhomboides* (to <2%), *S. anceps* (to <2%), and *Encyonema* spp. (to <5%). Increases in relative abundance were observed for *D. pseudostelligera*, *A. acares*, *P. microscopicum*, H. *schmassmannii*, *Sellaphora seminulum*, *M. kuelbsii*, *A. perglabra*, and *Nitzscia* spp., species which were mostly absent below core depth 4.0 cm. A further increase in *D. pseudostelligera* (from ~25 to 40%), and decrease in *A. lirata* (from ~10 to 2%) occurred in the post-mine period. The chrysophyte scale:diatom index increased above core depth 1.0 cm (~1990), and no further increases occurred in the post-mine period. PCA axis 1 scores decreased from ~5 to 2 cm (~1960), and were stable in the uppermost 2.0 cm. PCA axis 2 scores increased from ~5 to 2 cm, and then decreased again between 1.5 and 1.0 cm.

Discussion

Long-term changes in eutrophication indicators

Our paleolimnological findings for organic carbon, nitrogen, and chlorophyll *a* indicate that lake productivity has been increasing since pre-1950, well before the opening of the Diavik and Ekati diamond mines in the late 1990s and early 2000s. Only nitrogen stable isotope composition showed a clear deviation from the pre-mining trajectory of change. Sedimentary organic carbon and nitrogen content exhibited gradual increases since ~1700 in all three

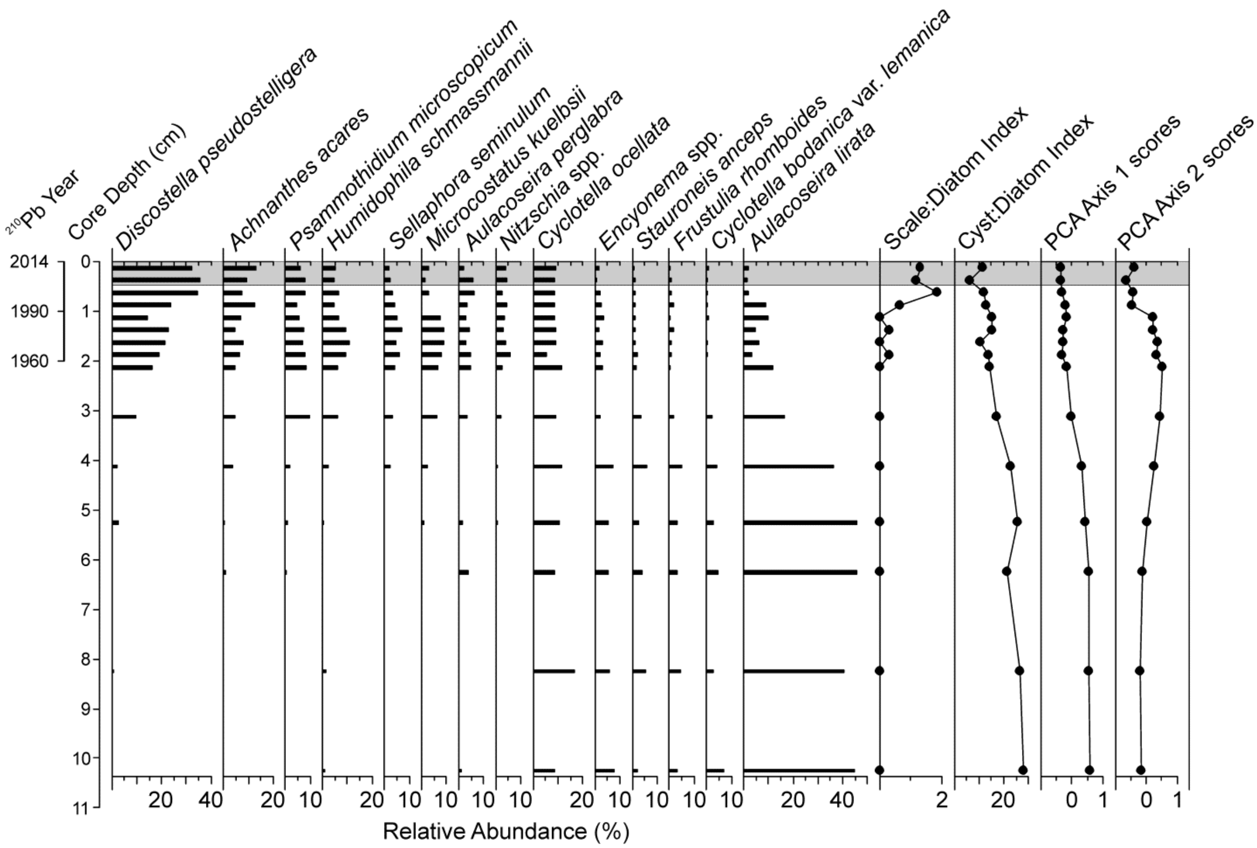


Figure 6 – **Temporal changes in diatoms at site MF2-3.** Stratigraphic profile of the most abundant diatom taxa (as relative abundances) in a sediment core collected from site MF2-3 in Lac de Gras, located mid-field and upstream from the Diavik Diamond Mine. ^{210}Pb-inferred dates are shown as a secondary y-axis. Principal Components Analysis Axis 1 and 2 scores for diatom assemblages are also shown, as well as the ratio of chrysophyte scales to diatoms, and chrysophyte cysts to diatoms. The grey box represents the current mining period.

sediment cores collected from Lac de Gras, although absolute percentages remained low (~5-7% for TOC and ~0.5% for TN), indicating a long-term gradual increase in aquatic productivity. C/N ratios, which provide an estimate of the relative contributions of algal versus terrestrial sources of organic matter to lake sediments, decreased after ~1950. Fresh organic matter derived from phytoplankton has C/N values in the range of ~4-10, whereas organic matter originating from vascular land plants typically has C/N values of >20 (Meyers and Teranes 2001). In Lac de Gras, C/N decreased from ~14 to ~11, which is indicative of an increased contribution of organic matter originating from in-lake primary productivity. Similarly, post-1950 increasing trends in VRS-inferred chlorophyll *a* were also apparent in all three Lac de Gras sediment cores, although we caution that VRS-chl *a* values that are below the estimated method detection limit should be interpreted with caution. Widespread increases in lake primary productivity have also been reported for lakes across the circumpolar Arctic and subarctic, a trend that has been inferred as a response to a longer open-water growing season due to climate warming (Michelutti et al. 2005, Griffiths et al. 2017, Hadley et al. 2019). For cores NF3 and MF3-4, the current mining period showed, on average, higher rates of percent change in VRS-chl *a*, which may indicate an additive or synergistic effect between climate warming and nutrient release from mining activities. Similarly, the only intervals with VRS-chl *a* above method detection limits for all three Lac de Gras cores are in sediments deposited in the current mining period, which may provide supporting evidence for at least a partial eutrophication effect of mining activities.

Long-term decreases in $\delta^{15}N$ (from >10‰ to ~4-6‰) were observed in all three sediment cores beginning ~1900, indicating that changes in nitrogen cycling have occurred in Lac de Gras. The limnological processes contributing to changes in bulk $\delta^{15}N$ are complex, making it challenging to interpret the underlying mechanisms driving the long-term shift in $\delta^{15}N$ in lake sediment records (Meyers 2003). However, the temporal changes in $\delta^{15}N$ values in the Lac de Gras sediment cores are consistent with seasonal changes in $\delta^{15}N$ in Lake Ontario (northeastern United States and southern Canada), where $\delta^{15}N$ values were low (~6-8‰) during the spring and summer phytoplankton bloom, and high (10-12‰) during the late fall and winter, when organic matter originated from heterotrophic sources (Hodell and Schleske 1998). Based on this, we suggest that the post-industrial decrease in $\delta^{15}N$ in Lac de Gras is likely tracking a shift from predominantly heterotrophic processes and sources of organic matter to more isotopicallydepleted phytoplankton detrital sources as the duration of the ice-free season has lengthened. In the current mining period, $\delta^{15}N$ increased in NF3 and to a lesser extent MF3-4, but not in MF2-3. Large inputs of nitrates from blast residue would be expected to alter the isotopic nitrogen signature, and thus the increase in $\delta^{15}N$ (a reversal of the long-term trend) can be plausibly linked to diamond mining activities. Inorganic nitrogen inputs, for example from agricultural fertilizers, have been shown to alter bulk sedimentary nitrogen isotope signatures in previous studies (Wang et al. 2019, Woodward et al. 2010). Furthermore, the current mining trend of increased $\delta^{15}N$ is most pronounced in NF3, the most impacted site. This is what would be expected if mining activities were responsible for the change in $\delta^{15}N$.

Striking diatom assemblage changes are evident from the Lac de Gras subfossil record since pre-1950, well before mining operations began, likely reflecting changes in lake ice phenology and associated limnological responses to regional climate warming. Beginning pre-1950, decreases in the heavily-silicified, tychoplanktonic taxon *Aulacoseira lirata*, and large-celled *Cyclotella bodanica* var *lemanica* occurred coincident with increases in the relative abundances of a range of benthic and periphytic species of the *Achnanthes sensu lato* (*acares*, *microscopicum*) and small *Navicula sensu lato* (*kuelbsii*, *schmassmani*). A longer ice-free season, and consequently a longer growing season, in response to regional warming would result in increases in littoral habitat growth (e.g. aquatic mosses, macrophytes), supporting the population growth of associated periphytic diatom taxa. Similar diatom assemblage changes have been documented across the circumpolar Arctic (Smol et al. 2005, Rühland et al. 2015).

Since the ~1950s, *Discostella pseudostelligera* has proliferated to become the dominant diatom taxon in Lac de Gras (at least at sites NF3 and MF2-3). Scaled chrysophytes, a holoplanktonic group that requires appropriate pelagic habitat to flourish, increased at the same time as *D. pseudostelligera*. *D. pseudostelligera* is a small, fast-growing, centric diatom that is often indicative of climate-driven physical changes in lakes (Winder et al. 2009, Wang et al. 2012). Circumpolar increases in *D. pseudostelligera* in the post-industrial period have been reported in lakes across the northern hemisphere, often corresponding with a longer ice-free period and greater water column stability (reviewed in Rühland et al. 2015). Synergistic interactions between nutrients (particularly nitrogen), light, and water column stability have also been postulated as potential drivers of increases in *D. pseudostelligera* (Saros et al. 2012, Saros et al. 2014, Malik et al. 2018). Based on long-term limnological monitoring conducted as part of the Diavik AEMP, Lac de Gras does not exhibit thermal stratification over the ice-free period, although short-lived periods of stratification and weakened mixing events are possible and may impact diatom assemblages (Rühland et al. 2013, Paterson et al. 2014). In the Lac de Gras watershed, permafrost thaw occurring in response to regional warming likely enhanced the flux of nutrients into the oligotrophic Lac de Gras (Petrone et al. 2006, Reyes and Lougheed 2015). Thus, the proliferation of *D. pseudostelligera* beginning after ~1950 likely reflects climate-driven changes in lake physical and biogeochemical processes, resulting in the crossing of ecological thresholds for phytoplankton communities. In small waterbodies north of Lac de Gras that receive effluent from the Ekati Diamond Mine, increases in the small centric diatom taxa *Cyclotella* have also been observed over the period of aquatic effects monitoring; however, this was interpreted as a response to nitrogen fertilization (St. Gelais et al. 2017). Importantly, diatom changes in Lac de Gras occurring in the current mining period are a continuation of trends that began in ~1950, and thus cannot be solely interpreted as a response to nitrogen and/or phosphorus fertilization.

Long-term changes in sedimentary metal concentrations

A paleolimnological approach can be used to establish long-term trajectories of change in trace metals in Lac de Gras, to place current monitoring observations in the context of ongoing climate warming. Based on the sediment cores collected from the 3 AEMP monitoring sites in Lac de Gras, strontium, lead, zinc, and manganese concentrations have been increasing over the post-industrial period. The % sulfur has also been increasing in NF3 and MF2-3, but not MF3-4. Increases in lead are likely a result of long-range transport of airborne pollutants from leaded fuels, and similar increases have been noted in sediment cores from across Arctic and subarctic regions (e.g. Bindler et al. 2001, Liu et al. 2012). Strontium is notably elevated in western Canadian Arctic lakes impacted by retrogressive thaw slumping (Houben et al. 2016, Mesquita et al. 2010), and its long-term increase in Lac de Gras may similarly provide a signature for thawing permafrost processes in the watershed. The thawing of permafrost and subsequent deepening of the seasonally-thawed active layer can expose previously frozen soils to decomposition and mineral weathering processes, enhancing the flux of trace elements into hydrologically connected waterbodies. For example, seasonal geochemical signatures of trace metals in surface waters of two Alaskan watersheds were linked to the extent of active layer thaw, indicating that surface water trace metal composition may provide a watershed-scale proxy for thawing permafrost soils (Barker et al. 2014). Thawing of permafrost may also result in reducing conditions in watershed soils if melted water stagnates, enhancing the mobility of redox-sensitive transitional metals such as manganese, iron, and zinc (Davidson 1993). Increases in the concentration of manganese in Lac de Gras sediments, decoupled from iron, may reflect an increased input of organic matter from the catchment (Kaff 2001), and/or the development of mildly reducing conditions in catchment soils, as manganese is more readily soluble than iron (Davidson 1993).

Among the trace elements that triggered AEMP Action Level 1 or 2, strontium and sulfur showed clear increasing trends prior to the opening of the Diavik and Ekati Diamond Mines, indicating that the exceedances may be unrelated or only partially related to mining operations. In contrast, aluminum and copper, which had AEMP Action Level 2 exceedances, have been stable or decreasing over the last several decades based on the paleolimnological record. As aluminum and copper are potentially toxic to aquatic biota (Brix et al. 2017, DeForest et al. 2018), these elements should continue to be carefully monitored for potentially mining-related impacts in Lac de Gras.

Management implications

An Aquatic Effects Monitoring Program (AEMP) is intended to guide adaptive management strategies to minimize and mitigate any potential adverse effects of project activities (MVLB/GNWT 2019). The design and implementation of an AEMP requires the determination of initial predictions of potential adverse effects, as well as the establishment of acceptable limits of impact (e.g. at what point is action required), in order to determine what data should be collected both before and during project operations (MVLB/GNWT 2019). For remote aquatic ecosystems like Lac de Gras, pre-determined “significance thresholds” or “action levels” are often defined as changes that are outside the range of natural variability, or departure from baseline conditions. Baseline conditions are typically established based on only a few years of pre-development monitoring, and often only as snapshots from one or a few sampling times, and thus do not provide a holistic understanding of ecosystem functioning, particularly for large northern lakes like Lac de Gras that are historically understudied systems (Cott et al. 2016),

An obvious advantage of a paleolimnological approach is the ability to characterize the range of natural variability or baseline conditions prior to monitoring. In situations where AEMP or current mining paleolimnological data show clear deviation from natural variability or baseline conditions, this would provide a compelling and early-warning indication of a potential impact of mining to trigger adaptive management strategies. For example, in Lac de Gras, nitrogen isotopic composition clearly deviates from a long-term decreasing trend in sediments deposited in the current mining period, providing compelling evidence that diamond mining activities have altered nitrogen cycling. Our paleolimnological study of Lac de Gras also supports AEMP Action Level 2 exceedences for aluminum and copper. In contrast, where anticipated mining impacts are similar to longer-term trajectories of limnological change, establishing significance thresholds is more challenging. Based on our paleolimnological data, we recommend the Diavik AEMP review its current significance thresholds and Action Levels for chlorophyll *a* and select metals (particularly Sr, Mn, Zn) that we show to be changing since at least 1950 due to climate warming. As an initial step to assist in this process, we calculated background rates of percent change in these geochemical indicators that can be used as a benchmark for evaluating changes that have occurred (or will occur) over the period of aquatic effects monitoring. We recommend that future efforts focus on developing statistical and conceptual approaches that will strengthen our capacity to extrapolate paleolimnological inferences of natural variability to aquatic effects monitoring, both within the context of ongoing AEMP design review in the Northwest Territories, and the application of paleolimnology to lake management more generally.

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