

A diatom-based paleolimnological survey of environmental changes since ~ 1850 in 18 shallow lakes of the Athabasca Oil Sands Region, Canada

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Abstract Aerially transported contaminants from the industrial development of the bituminous sands in the Athabasca Oil Sands Region (AOSR) of western Canada may threaten the water quality and community structure of the region's lakes. This environmental threat is further compounded by the region's changing climate (i.e. increased temperatures and reduced moisture). Because environmental monitoring began ~ 30 years after the initiation of the region's bitumen-based industry (~ 1967), a paleolimnological approach is required to document pre-disturbance conditions and to determine how lakes have changed, if at all, in response to environmental stressors. Our

bottom–top (before and after) study of dated sediment records compared pre-disturbance (~ 1850) and modern subfossil diatom assemblages from 18 shallow, isolated lakes (which are typical of the region), located along a spatial gradient (up to ~ 110 km) relative to the main area of local industry. Despite the region's substantial environmental stressors, the changes in mostly benthic-dominated diatom communities were minor at 15 of the 18 lakes. We conclude that the muted biological responses may be a consequence of naturally high nutrient concentrations and/or the typically subtle nature of changes in assemblages dominated by benthic generalist taxa. At all sites, including three lakes with marked changes, we found no evidence of a diatom assemblage response to airborne inputs of contaminants (i.e. dibenzothio-phenes) and nutrients (i.e. bioavailable nitrogen) from the AOSR industry. Instead, it is likely that regional warming played a role in the modest diatom assemblage changes observed in these shallow lakes, with responses mediated by lake-specific characteristics.

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Introduction

The Athabasca Oil Sands Region (AOSR) is well-known for its enormous bitumen extraction and processing industry that began with the development of the first commercial upgrader in 1967 (Government of Alberta 2014). The Athabasca deposit is the largest of three bitumen deposits in Alberta and Saskatchewan, with current production rates of ~ 1.76 million barrels per day, and forecast rates of ~ 3.16 million barrels per day by 2030 (Alberta Energy Regulator 2015; Canadian Association of Petroleum Producers 2015). The environmental concerns that accompany this large industrial expansion are becoming increasingly apparent. For example, development of facilities, mines, and supporting infrastructure has resulted in habitat fragmentation (Rooney et al. 2012), ground deformation (Stancliffe and van der Kooij 2001), and ultimately the loss of ecosystem services (Rooney et al. 2012). Moreover, bitumen-based industrial processes require large volumes of freshwater, generate copious amounts of tailings (Sauchyn et al. 2015), and can contaminate the ground (e.g. flow-to-surface events; Korosi et al. 2016), water (Kelly et al. 2009, 2010), and atmosphere (Kurek et al. 2013a; Kirk et al. 2014; Manzano et al. 2017).

Industrial airborne contaminants from upgrader stacks and machinery, as well as fugitive dusts from mine sites, industrial waste piles, and exposed bitumen (Landis et al. 2012; Jautzy et al. 2013; Zhang et al. 2016; Manzano et al. 2017), are deposited across the AOSR. Paleolimnological data suggest that contaminants may reach aquatic environments at least 90 km from the main industrial area (Kurek et al. 2013a). The industrial airborne contaminants include a suite of potentially toxic compounds (Kelly et al. 2009, 2010; Kirk et al. 2014). For example, dibenzothiophenes (DBTs) are a group of polycyclic aromatic compounds (PACs) that are among the industrial contaminants released to the atmosphere by the oil sands operations (Jautzy et al. 2013; Kurek et al. 2013a) and are an established proxy for exposure of an AOSR aquatic system to industrial airborne contamination (Jautzy et al. 2013; Kurek et al. 2013a, b; Summers et al. 2016). Several studies that explored atmospheric contamination in the AOSR also detected elevated concentrations of crustal, anthropogenic, and priority pollutant elements (Guéguen et al. 2016; Kelly et al. 2010; Kirk et al. 2014; Landis et al. 2012).

Biologically available nitrogen, which can potentially enhance primary production and alter biotic communities, is known to be emitted in large quantities by oil sands activities. For example, 102 tonnes of dissolved inorganic nitrogen were deposited in 2014 springtime snowpack within 50 km of the main industrial oil sands development (Summers et al. 2016).

Consistent with Canada's prairie provinces, warming temperatures and a drying landscape characterize the AOSR (Schindler and Donahue 2006). During the past ~ 100 years, mean annual temperatures in the prairie provinces have increased by $\sim 1\text{--}4$ °C, while total annual precipitation has declined by $\sim 25\%$ (Schindler and Donahue 2006). Previous paleolimnological studies investigating the effects of industrial activity in the AOSR, and downwind, originally focused on the effects of acid deposition from emissions of nitrogen and sulphur oxides (Hazewinkel et al. 2008; Curtis et al. 2010; Laird et al. 2013). Although they produced no consistent evidence of widespread surface water acidification, these studies noted potential increases in primary production, which Summers et al. (2016) identified as a regional effect that was most closely linked to recent climate warming and not the deposition of bioavailable nitrogen. Other recent paleolimnological studies that investigated biological responses to environmental stressors in the AOSR found an overriding influence of climate warming, instead of industrial inputs of contaminants and nutrients, in structuring aquatic communities of lakes (Hazewinkel et al. 2008; Kurek et al. 2013a, b; Laird et al. 2017; Mushet et al. 2017; Summers et al. 2017).

Direct long-term monitoring data and baseline conditions are largely lacking for the AOSR (Dowdeswell et al. 2010). Thus, paleolimnological methods, which use physical, chemical, and biological proxies preserved in lake sediments (Smol 2008), are required to understand past environmental conditions of these aquatic systems. In this study, we used diatoms as the primary proxy to advance our understanding of the effects of industrial airborne pollution (i.e. contaminants and nutrients) and climate change on shallow lake ecosystems. Subfossil remains of diatoms are one of the most commonly used biological proxies in paleolimnological and biomonitoring studies and are used as indicators of past changes in pH, salinity, nutrients, and climate-mediated conditions, as

well as many other variables (Smol and Stoermer 2010).

Previous diatom studies tracked temporal changes in the AOSR in specific lakes of interest (Curtis et al. 2010; Hazewinkel et al. 2008; Laird et al. 2017; Summers et al. 2017). To explore the regional patterns of temporal change in diatom assemblage composition, however, we used a top–bottom (i.e. diatom assemblages in recent sediments compared to assemblages preserved in pre-disturbance sediments) paleolimnological approach (Smol 2008), including 18 shallow study lakes situated within ~ 110 km of the main industrial activities. Sites were selected to represent shallow, isolated lakes that receive different amounts of airborne pollution from the local bitumen industry. For all sites, pre-disturbance (bottom) sediment sections were dated and are known to represent ~ 1850. We addressed the following research questions: (1) Do modern diatom assemblages differ from pre-disturbance assemblages? (2) Has aerially transported industrial pollution (i.e. contaminants and bioavailable nitrogen) affected diatom assemblage composition? (3) What physical and chemical variables best explain the spatio-temporal patterns of change in diatom assemblages among shallow AOSR lakes?

Study area

The AOSR is situated primarily in the northeast corner of Alberta, Canada, and extends into northern Saskatchewan (Fig. 1). Decades of bitumen-based industry and development have altered this once-remote part of the Boreal Plains ecozone. The landscape is characterized by boreal forest, peatlands, uplands featuring jack pine (*Pinus banksiana*) and trembling aspen (*Populus tremuloides*), and many shallow lakes and wetland systems (Natural Regions Committee 2006). The region's surficial geology is composed mainly of coarse glacio-fluvial till, which overlies sand, shale, bituminous sands, and carbonate-dominated limestone and dolomite bedrock (Canadian Society of Petroleum Geologists 1973). The winters are long and cold (2014 mean winter air temperature = -19.7 °C), and the summers are short and warm (2014 mean summer air temperature = 17.2 °C; Environment and Climate Change Canada's Fort McMurray temperature station; ID: 3062696; 56°36'00.0"N, 111°13'12.0"W; [\[ahccd\]\(http://www.ec.gc.ca/dccha-ahccd\)\). Most precipitation falls as rain between June and October \(Kerkhoven and Gan 2011\), with an annual average of 400–500 mm \(Leong and Donner 2015\).](http://www.ec.gc.ca/dccha-</p></div><div data-bbox=)

Materials and methods

Site selection

The 18 lakes used in this top–bottom paleolimnological survey are a subset of shallow lakes that were strategically selected to represent a range of airborne contamination from local bitumen-based industry (Table 1; Fig. 1). Given that this project explores aerially transported industrial contamination and nutrient deposition, the selected study lakes are isolated basins that are not directly part of a larger hydrologic system, such as the Athabasca River, and have small catchments relatively undisturbed by industry at the time of coring. The sites are located at different distances (within ~ 110 km) in all directions from the main area of industrial bitumen-related activities (Fig. 1).

Sample collection

As reported in earlier AOSR studies, including Kurek et al. (2013a) and Summers et al. (2016), sediment cores and water samples were collected in early March 2011–2015 as part of the Canada-Alberta Joint Oil Sands Monitoring (JOSM) program (www.jointoilsandsmonitoring.ca). All lakes are located on provincially owned and publicly accessible lands that were accessed by helicopter. Three sediment cores were collected through the ice from the deepest part of each shallow lake using a Uwitec gravity coring system and were sectioned using a Uwitec vertical extruder immediately upon returning to the field base (www.uwitec.at). Given that the cores were sectioned contiguously every 0.5 cm down to a depth of 20 cm and contiguously every 1.0 cm thereafter, the top sections for each lake were 0.5 cm thick and the bottom sections were 1.0 cm thick. The core sections were frozen until analysed. The primary core from each site was used for radiometric dating (Flett Research Ltd., Winnipeg, Manitoba), PAC analyses (AXYS Analytical Services, Sidney, British Columbia), and bioindicator and sedimentary chlorophyll-

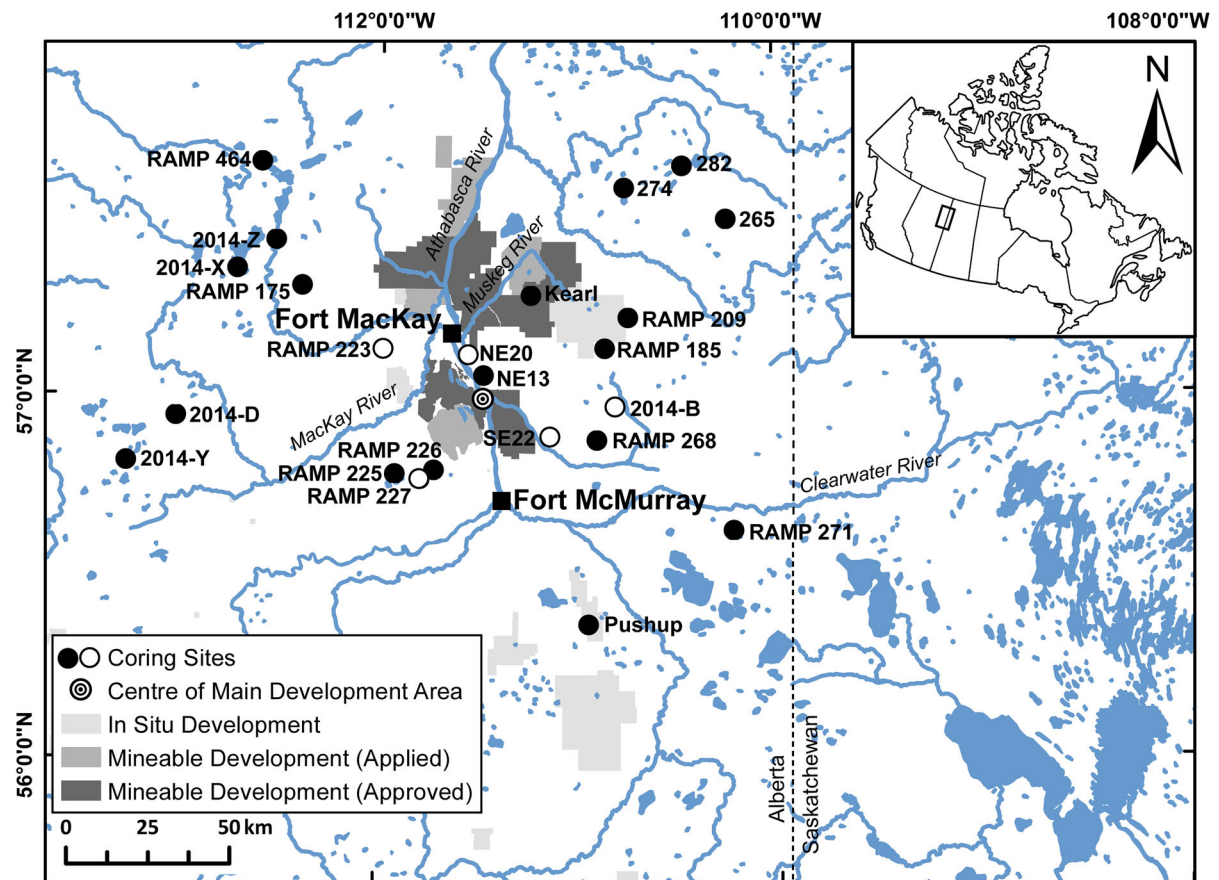


Fig. 1 Locations of the 23 cored study lakes, two local communities, and the footprint of industrial oil sands development. Black circles mark the 18 shallow sites included in the histogram of the changes in diatom assemblages from the top-bottom sediment analyses (Fig. 3) and statistical analyses.

White circles mark sites that were not included in the study because their chronologies do not extend to pre-disturbance times or there is a lack of diatoms in the pre-disturbance sediment sections (see text for details)

a analyses (Paleoecological Environmental Assessment and Research Laboratory (PEARL), Queen's University, Kingston, Ontario). Replicate cores were retained for additional analyses. At the time of coring, water samples were collected through the ice at each site and were processed following standard techniques at the National Laboratory for Environmental Testing (NLET; Burlington, Ontario). Measures of environmental variables used in numerical analyses were obtained from field measurements, map measurements, sediment samples, and water samples (Electronic Supplementary Material [ESM] Table S1).

Radioisotopic analyses

Radioisotope activity and age modelling were completed by Flett Research Ltd and were previously reported in Summers et al. (2016, 2017). Briefly, ^{210}Po and ^{226}Ra activities were measured using alpha spectrometry as proxies for total and background (supported) ^{210}Pb , respectively. Age-depth relations (± 2 SD) of the measured sediments in which total ^{210}Pb exceeded supported ^{210}Pb were calculated using the constant rate of supply (CRS) age model (Appleby 2001). Ages for the remaining sediment sections were estimated by fitting a polynomial of the lowest order with a reasonable fit to the cumulative dry mass of the measured sediment, and setting the intercept to the time of coring (Summers et al. 2016, 2017).

Table 1 Physical, chemical, and industry-related characteristics for each of the 18 lakes

Lake name	Lake code	Year cored (CE)	Surface area (ha)	Coring depth (m)	pH	DIC (mg/L)	DOC (mg/L)	TN (µg/L)	TP (µg/L)	TN:TP (mass)	NH3 (mg/L)	SiO2 (mg/L)
RAMP 271	271	2013	399.9	1.5	7.2	23.2	24	1290	33.1	39	0.134	5.2
2014-Y	Y	2014	6.0	1	6.8	20.6	94.5	3740	1290.0	3	0.959	23.9
282	282	2015	50.3	1.6	8.2	27.7	13.3	1660	2.6	638	0.823	1.9
2014-D	D	2014	4.4	0.9	6.6	15.6	92.9	2940	341.0	9	0.504	7.4
RAMP 464	464	2013	93.2	4.6	7.1	8.6	26	1460	90.0	16	0.027	3.9
NE13	NE13	2011	3.4	1.7	7.6	76.2	33.5	2950	9.8	301	1.67	21.0
RAMP 226	226	2011	18.1	1.5	6.8	2.7*	33.6	1162*	41*	28	0.013*	0.87**
2014-X	X	2014	7.9	0.75	6.7	15.5	39.8	2860	24.2	118	1.3	2.6
2014-Z	Z	2014	50.8	3.2	7.0	24.6	28.8	2300	75.5	30	0.455	1.4
RAMP 225	225	2012	23.3	1.2	9.3	27	62.2	3690	33.6	110	0.15	7.4
RAMP 175	175	2013	38.1	1.5	7.4	24.8	68.7	4720	61.2	77	1.83	8.7
Kearl	Kearl	2012	546.2	3.5	9.5	41.9	35.3	4380	26.4	166	0.059	5.8
274	274	2015	9.7	1.2	8.1	34	49	1550	6.4	242	0.173	6.5
RAMP 209	209	2013	10.9	1.2	6.3	5.6	51.7	773	37.0	21	0.09	5.9
265	265	2015	2.1	1.4	6.4	2.1	9.7	687	11.6	59	0.356	1.8
RAMP 268	268	2012	188.5	1.8	6.8	9.8	67.8	3130	44.9	70	0.609	8.8
RAMP 185	185	2012	10.5	2.0	6.4	3.5	55	2120	21.2	100	0.582	8.8
Pushup	Pushup	2014	16.9	5.1	6.8	12.6	28.7	2660	130.0	20	1.14	1.8
Mean	-	-	82.2	2.0	7.3	22.0	45.3	2524	131.7	19	0.6	7.2
Median	-	-	17.5	1.5	6.9	20.6	37.6	2660	33.6	79	0.5	5.9
Maximum	-	-	546.2	5.1	9.5	76.2	94.5	4720	1290.0	4	1.8	23.9
Minimum	-	-	2.1	0.8	6.3	2.1	9.7	687	2.6	264	0.0	1.4

Table 1 continued

Lake name	Chl-a ($\mu\text{g/L}$)	Specific conductance ($\mu\text{S/cm}$)	Cl-(mg/L)	Ca ⁺² (mg/L)	Mg ⁺² (mg/L)	K ⁺ (mg/L)	Na ⁺ (mg/L)	SO ₄ ⁻² (mg/L)	DBT EF	Top sed. [DBT] (ng/g dry wt)	Top sed. [DBT] depth (cm)/year (CE)
RAMP 271	37.5	191.0	0.46	25.9	7.19	1.41	6.21	0.4	0.7	61.2	0/2012
2014-Y	1.7	220.0	0.3	30.8	8.54	1.85	11.5	6.8	1.1	19.3	0/2014
282	<0.1	188.0	0.31	25.8	11.2	1.01	0.98	0.1	1.3	119.8	0.5/2014
2014-D	8.1	176.4	0.44	25.9	7.41	0.8	7.25	5.9	1.6	36.9	0/2014
RAMP 464	<0.1	86.0	0.17	10	3.26	1.14	4.06	8.5	1.9	65.6	0/2012
NE13	4.0	591.0	0.94	97.6	18.6	1.19	5.74	0.7	2.1	205.9	0/2011
RAMP 226	28.3	41.9*	0.08*	6.64*	2.08*	0.62*	1.53*	0.28*	2.3	121.4	0/2011
2014-X	10.7	142.0	0.38	15.8	4.75	3.6	1.62	1.6	2.4	34.4	0/2013
2014-Z	41.4	255.0	0.26	27	9.44	1.62	11.4	24.4	2.6	92.2	0/2014
RAMP 225	9.6	123.0	0.85	16.6	8.9	1.6	2.39	1.1	2.7	308.0	0/2012
RAMP 175	23.0	234.0	0.78	25.9	7.61	2.77	13.8	0.4	2.9	63.3	0/2013
Kearl	63.0	203.0	0.78	40.5	12.8	1.36	19	2.6	3.1	573.5	0/2012
274	0.6	240.0	0.4	38.6	19.2	0.46	1.24	0.4	4.4	118.6	0/2015
RAMP 209	1.4	48.0	0.27	7.43	2.04	0.18	0.77	0.2	6.2	96.2	0/2013
265	<0.1	11.3	0.09	0.4	0.32	0.25	0.41	0.4	7.0	7.3	0/2014
RAMP 268	9.3	108.0	0.63	14.3	3.25	0.83	5.61	0.2	7.1	361.9	0/2012
RAMP 185	1.4	62.0	0.44	9.06	2.51	0.52	1.24	2.0	8.6	538.0	0/2012
Pushup	97.1	125.0	0.41	13.9	3.38	3.03	2.27	0.8	37.4	220.6	0/2014
Mean	22.5	176.7	0.5	25.0	7.7	1.4	5.6	3.3	5.3	169.1	–
Median	9.6	176.4	0.4	25.8	7.4	1.2	4.1	0.8	2.7	107.4	–
Maximum	97.1	591.0	0.9	97.6	19.2	3.6	19.0	24.4	37.4	573.5	–
Minimum	0.6	11.3	0.1	0.4	0.3	0.2	0.4	0.1	0.7	7.3	–

Single asterisks (*) mark measures that were collected by the Regional Aquatics Monitoring Program (RAMP) in the same year that the lake was cored. These measures were used because no data were collected from the site when the lake was cored. Double asterisks (**) mark averages of RAMP-collected water chemistry data from 2003 to 2007, as no data were collected from the site when the lake was cored. Lakes with TN:TP mass ratio < 14 are considered nitrogen limited (Downing and McCauley 1992)

The top–bottom paleolimnological approach

A top–bottom paleolimnological approach (reviewed in Smol 2008) was used to compare two temporal snapshots (present and ~ 1850) of diatom communities from 18 shallow, isolated sites across the AOSR. Top–bottom analyses are time-efficient and useful for exploring environmental change across broad spatial scales; however, the approach does not identify the specific timing of historical changes (Smol 2008). The uppermost available sediment section from each core was used as the top, providing an integrated sample from the whole basin that represents the most recent (ca. 2010–2014) conditions (ESM Table S2). Sediments below 0–0.5 cm were used only if no sediments from the original top section were available (sites 2014-Z, RAMP 268, and 274). Given that chronologies for all of the sediment cores were previously established (Summers et al. 2016, 2017), ^{210}Pb -estimated dates of sediment deposition were used to select the section from each lake that was most representative of ~ 1850 (available sections ranged from ~ 1825 to ~ 1897). These sections were used as the bottom (pre-disturbance) sections for each lake and provided an integrated sample of conditions preceding industrial oil sands development and anthropogenic climate change in this region (ESM Table S2). In this study, the top sections were 0.5 cm thick and the bottom sediment sections were 1.0 cm thick, which, in addition to the effects of downcore sediment compaction, resulted in the bottom sections representing a longer period of sediment deposition than the top sections. In this study, however, the time intervals for each sediment section were known (based on ^{210}Pb dating) and the differences between tops and bottoms were investigated through the use of multiple sites.

Diatom sample preparation and analyses

Diatom samples were prepared from either wet or freeze-dried sediment generally following standard methods (Battarbee et al. 2001). Briefly, sediment samples were digested in a 1:1 molar solution of strong nitric and sulphuric acids and heated in a hot water bath at 80 °C for at least 2 h. The samples were allowed to settle for 24 h before the acid was removed and the remaining slurry was diluted with deionized water. The settling and dilution procedure was

repeated until neutral pH was achieved. At least four dilutions of the digested slurry were plated onto coverslips and mounted onto slides using Naphrax[®]. Enumeration and identification of subfossil diatom remains (minimum = 303 valves, average = 418 valves) was completed at 1000 \times magnification using a Leica DMR light microscope fitted with differential interference contrast optics and oil immersion lenses. Taxonomic references primarily included Krammer and Lange-Bertalot (1986–1991) and Camburn and Charles (2000). Identified diatom taxa were expressed as relative abundances (%) of the total number of valves counted in each sediment section.

Numerical analyses

Environmental variables

A principal components analysis (PCA) was performed on modern environmental data (including physical, chemical, and industrial data) collected from the 18 study lakes to examine the variation of sites with respect to modern environmental variables. Prior to inclusion in any ordination, the Vegan (Oksanen et al. 2012) and Analogue (Simpson and Oksanen 2016) packages in the R software environment (R Development Core Team 2015) were used to test the environmental variables for normality, and non-normal variables were transformed as necessary (using square-root, log, or $\log(x + 1)$ transformations; Supplemental Table 1). A Pearson correlation matrix with a Bonferroni correction was completed with R's Hmisc package (Harrell 2017) and was used to calculate correlations among the environmental variables. The PCA was performed using CANOCO version 5.0 (ter Braak and Šmilauer 2012). The analysis was completed on a correlation matrix with variables centred and standardized to account for the different units of measurement.

Diatom assemblages

For most statistical analyses using diatom assemblage data, unless stated otherwise, rare taxa (not exceeding 2% in at least one sediment section) were excluded. Additionally, relative abundances of diatoms were square-root transformed in most statistical analyses to reduce the influence of dominant taxa and increase the weight of taxa with lower abundances, thereby

equalizing the variance among taxa and preventing subtle trends from being obscured (Borcard et al. 2011).

Changes in the diatom communities were quantified in several ways. First, the trajectories of change between pre-disturbance times and the present day were determined using a detrended correspondence analysis (DCA). A DCA is an unconstrained ordination method that assesses compositional changes between species and between cases (Hill and Gauch 1980), which were bottom and top sediment sections in this scenario. Modern assemblage data were actively included in the DCA, whereas pre-disturbance assemblage data were included passively to compare the trajectories of assemblage change within the context of the modern samples. The results were similar when both modern and pre-disturbance assemblage data were run actively. The DCA was performed with CANOCO version 5.0 software (ter Braak and Šmilauer 2012) using the default settings of detrending by segments and no additional down-weighting of rare species. Trajectories of change in DCA space between pre-disturbance and present day (Δ DCA1 and Δ DCA2) were determined by subtracting bottom sediment section DCA axis 1 and 2 site scores from the corresponding top sediment section DCA axis 1 and 2 sites scores (Quinlan et al. 2003; Perren et al. 2008; Sweetman et al. 2008). DCAs are scaled in standard deviation (SD) units, which are units of species turnover along the first underlying gradient of compositional variation (Birks 2007). This metric of beta diversity quantifies assemblage change in a species-based framework (Birks 2007) and is thus potentially more ecologically relevant than other metrics commonly used to understand biological change (Birks 2007; Perren et al. 2008).

To explore trends down to the species level across the 18 study lakes, pre-disturbance (bottom) and modern (top) samples of DCA sample scores, species richness, species diversity, and relative abundances of select taxa were compared in 1:1 scatter plots. DCA sample scores (axes 1 and 2) for bottom sediments were plotted against DCA sample scores (axis 1 and 2) for top sediments with a 1:1 line to make comparisons between the two time periods. Differences between modern and pre-disturbance species richness and Hill's N2 diversity (alpha diversity), were included in plots in the same way. Species richness and Hill's N2 diversity were calculated using the Vegan

(for both analyses; Oksanen et al. 2012) and the Rioja (for Hill's N2 diversity; Juggins 2015) packages in R. Prior to richness and diversity calculations, raw count data, with rare taxa included, were rarified to a common sum to account for differences in the total number of diatoms counted in each sediment section (Birks 2012). Species richness is expressed as the number of taxa in a sample, whereas Hill's N2 diversity incorporates richness and evenness in a sample, and is relatively easy to interpret ecologically because it is measured in units of very abundant taxa within a sample (Birks 2012). Percent relative abundances of common taxa or groups of taxa were also included in scatter plots with a 1:1 line to assess and compare compositional changes between modern and pre-disturbance assemblages. Finally, temporal assemblage changes in DCA 1 and 2 axis scores (Δ DCA1 and Δ DCA2), richness, diversity, and the taxa included in the 1:1 scatter plots were displayed as histograms and arranged along gradients of environmental variables and PCA axis 1 and 2 scores.

Results

Core chronologies

Radioisotope activity profiles and age-depth models from all the lakes included in this study are shown in ESM Fig. S1. Most of the chronologies were published previously in Summers et al. (2016). ^{210}Po (a proxy for total ^{210}Pb) in the majority of cores exhibited a typical exponential decay curve with some irregularity, although some cores showed low ^{210}Po in the surface sediments. Sediment cores from sites 274 and RAMP 271 only extended to ~ 1897 and ~ 1880 , respectively. Thus, those sediment sections, although more than 30 years younger than ~ 1850 , were selected as the closest representations of ~ 1850 . Pre-disturbance sediment sections selected from the other lakes ranged from ~ 1825 to 1865.

Top–bottom sediment availability

In this study, 23 lakes were originally sampled; however, only 18 sites were ultimately included in the analyses. One sediment core (RAMP 227) only extended to ~ 1953 (41–42 cm; ESM Fig. S1) and was thus inappropriate for inclusion. Furthermore,

despite ample diatoms in modern sediments, four sites (SE22, RAMP 223, NE20, 2014-B) did not have a sufficient number of diatoms to count in the pre-disturbance sections and were therefore also excluded from the study. On average, based on our estimated ²¹⁰Pb dates, the bottom sediment sections in our study represent approximately 6 years whereas the top sections represent approximately 1 year of diatom accumulation. The pre-disturbance sections with no/low diatoms were high in siliciclastic materials, whereas diatoms and other autochthonous siliceous indicators (e.g. chrysophyte cysts and scales) were scarce but well preserved, ruling out dissolution as the cause for no/low diatoms. There were no apparent patterns or common features in the lakes lacking pre-disturbance diatoms. Although 18 study lakes is a relatively low number for a regional assessment, the sites cover a sufficient range of industry contamination to justify an exploratory survey of lakes

(dibenzothiophene enrichment factors (DBT EFs) 0.7–37.4; Summers et al. 2016).

Limnological trends across study lakes

All environmental variables actively included in the PCA, except for pH (which is already expressed on a log scale), were either normally distributed or displayed an acceptably normal distribution following transformation (ESM Table S1). The sources of the measurements for each environmental variable are included in ESM Table S1, and ranges are in Table 1. Sixteen environmental variables were actively included in the PCA, and six variables were included passively (ESM Table S1). The passive variables included specific conductance and five ions (chloride, calcium, magnesium, potassium, and sodium) that were positively correlated to and thus represented by dissolved inorganic carbon (DIC), which was actively included in the PCA. In addition to conventional water chemistry measures and physical lake characteristics, the PCA included representative variables of industrial impact such as DBT enrichment factors (DBT EFs), DBT concentrations from the uppermost sediment section (top [DBT]), and sulphate (SO₄²⁻) from the water column. DBT EFs were calculated in Summers et al. (2016) and compare DBT concentrations in modern sediment sections to concentrations in sediment sections immediately preceding industrial activity in the AOSR (~ 1955–1970). The EFs characterize the change in DBT concentrations at a site over time and are thus useful as metrics of industrial airborne contaminants (Summers et al. 2016). The top [DBT] is not a measure of industrial impact, but rather, is the combined natural and industrially sourced DBT concentration in the lake sediments deposited most recently. Sulphate is included independently because AOSR pet coke, a bitumen industry waste product, is rich in sulphur (Alberta Energy Regulator 2015; Manzano et al. 2017).

The PCA demonstrated that water chemistry and other limnological variables were generally dissimilar among the lakes, as no lakes clustered together in the ordination (Fig. 2). The amount of variance explained was similarly distributed between the two main axes, with PCA axis 1 explaining 25% and axis 2 explaining 23%. Although total nitrogen loaded the most strongly of all variables onto axis 1 (score = - 0.91), the axis

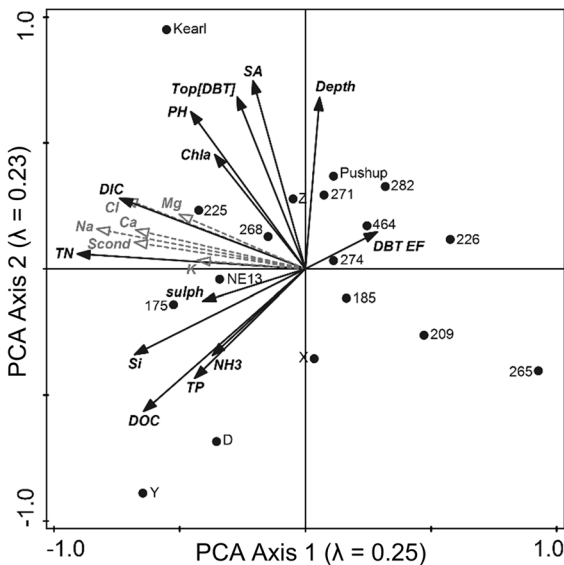


Fig. 2 Principal components analysis (PCA) plot including the first two ordination axes of measured environmental variables from the 18 lakes included in statistical analyses. Lake surface area (SA), coring depth (Depth), pH, dissolved inorganic carbon (DIC), dissolved organic carbon (DOC), total nitrogen (TN), total phosphorus (TP), ammonia (NH₃), silicon dioxide (Si), chlorophyll-*a* (Chla), dibenzothiophene enrichment factor (DBT EF), top sediment DBT concentrations (Top [DBT]), and sulphate (sulph) were included actively and are marked in solid black. Specific conductance (Scond), chloride (Cl), calcium (Ca), magnesium (Mg), potassium (K), and sodium (Na) were included passively and are marked in grey. Site names are abbreviated (Table 1)

represented a complex gradient of nutrients as well as ions, represented by DIC (score = -0.72). Axis 2 represented a gradient of multiple variables, but mostly represented physical characteristics, including surface area and depth, which loaded most strongly onto axis 2 (score = 0.74 and 0.68 , respectively; Fig. 2). Interestingly, sulphate was not positively correlated to DBT contaminant variables.

Trends in diatom assemblages

A total of 311 diatom taxa were identified in the 18 lakes. A total of 97 taxa remained when a cut-off criterion was applied retaining only taxa exceeding 2% in at least one sediment section. Fourteen taxa are shown in the top–bottom relative abundance histogram, including taxa exceeding 5% relative abundance in at least one sediment section and demonstrating notable change in abundance from bottom to top in at least one site (Fig. 3). Small, benthic fragilarioid taxa were common to most sediment sections and, in fact, were dominant in the majority of both top and bottom samples (Fig. 3). NE13 was an exception. NE13 is a marl lake characterized by many benthic taxa (e.g. *Brachysira neoexilis*, cymbelloid taxa, and long *Navicula* taxa including, for example, *Navicula cryptocephala*, *Navicula cryptotenella*; Fig. 3).

The trajectories of overall assemblage change through time (i.e. the differences between the pre-disturbance and the modern DCA sample scores from each lake) showed modest changes with no consistent directional change among sites (Fig. 4). It is evident, however, that pre-disturbance diatom assemblages shared more similarities among lakes than modern assemblages. The plot of DCA sample scores (Fig. 4) highlighted the differences among assemblages, with only modest separation of sites (except for the marl lake, NE13). DCA axis 1 explained 13.5% of the variance and axis 2 explained 9.9% (Fig. 4). Examining the diatom taxonomic loadings on the DCA axes (i.e. species scores), *Aulacoseira* taxa and pennate planktonic *Fragilaria* taxa were most important to DCA axis 1 (ESM Fig. S2, ESM Table S3). Three lakes demonstrated notable changes between pre-disturbance and modern assemblages (2014-D, 2014-Z, and Pushup; Fig. 3). Sites 2014-D, 2014-Z, and Pushup demonstrated clear compositional turnover from high relative abundances of small, benthic

fragilarioid taxa to high relative abundances of centric planktonic taxa (Fig. 3). Diatom assemblage changes in other sites were less pronounced (Fig. 4), but nevertheless exhibited changes among numerous benthic taxa, mostly transitioning from *Staurosira construens* and *Staurosirella pinnata* taxa to other benthic taxa with more specialized habitat preferences, or from benthic taxa to pennate planktonic taxa in one case (Fig. 4).

An examination of trends in the dataset's most common taxa highlighted distinct differences between pre-disturbance and modern relative abundances (Fig. 5). One of the clearest changes was a decrease in the relative abundances of taxa in the dominant, benthic fragilarioid complex (*Staurosira construens* and *Staurosirella pinnata*) in the modern assemblages of most study lakes (Fig. 5). Notable relative abundance increases in the modern samples are evident in plots of *Achnanthydium minutissimum*; the small, benthic naviculoid complex (*Eolimna minima*, *Sellaphora atomoides*, and *Sellaphora seminulum*); the long, benthic *Navicula* complex (*Navicula cryptocephala*, *Navicula cryptotenella*, *Navicula pseudolanceolata*, *Navicula radiosa*, *Navicula rhynchocephala*, and *Navicula vulpina*); and all *Nitzschia* taxa summed (Fig. 5 plots).

Scatter plots of pre-disturbance and modern species richness and Hill's N2 diversity (rarefied $n = 303$) showed no directional trend among the 18 lakes (Fig. 5). However, as is the case in all paleolimnological studies, changes in species diversity through time probably involve different time spans in sediment sections and thus should be interpreted with caution (Smol 1981). Plots of DCA Axis 1 scores demonstrated minimal change between the two time periods investigated, and mostly plotted along the 1:1 line, while several sites had noticeably larger DCA axis 2 site scores in modern diatom assemblages, indicating marked diatom assemblage changes at those sites (Fig. 5).

Environmental variables and their relation to diatom assemblages

Temporal assemblage changes in richness, diversity, DCA 1 and 2 axis scores (Δ DCA1 and Δ DCA2), and the taxa included in the scatter plots were displayed as histograms and arranged along gradients of environmental variables and PCA axis 1 and 2 scores. The

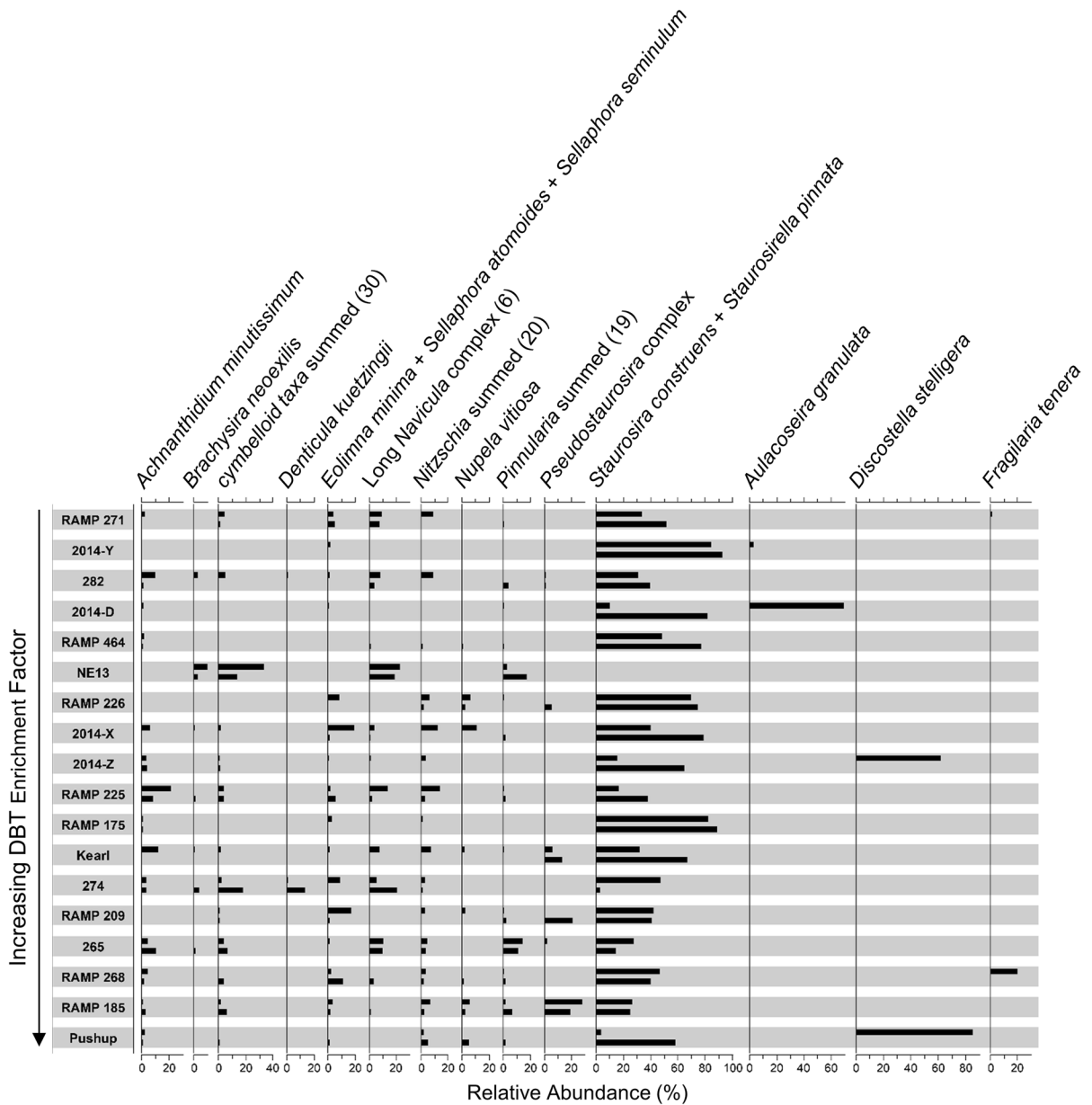


Fig. 3 Relative abundances of select diatom taxa (> 5% relative abundance in at least one interval and demonstrating notable abundance change from bottom-to-top in at least one site) in the modern (top of each shaded pair) and pre-disturbance (bottom of each shaded pair) sediment sections from the 18 study lakes. Where applicable, the number of taxa included in

the complexes and summed groups are indicated in parentheses following the group name. Taxa included in complexes and summed groups are listed in ESM Table S4. Lakes are arranged from top to bottom in order of increasing dibenzothiophene enrichment factor (DBT EF; Table 1)

ordered histograms did not show consistent, directional patterns of biological change (ESM Fig. S3). It is notable that these metrics of assemblage change, when plotted along gradients of DBT EF and distance

to nearest major industrial source (Landis et al. 2017), which are proxies for aerial industrial oil sands impact, showed no clear directional trends (ESM Fig. S3).

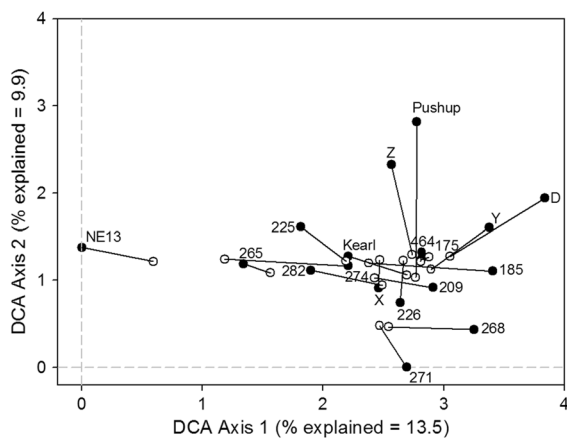


Fig. 4 Detrended correspondence analysis (DCA) plot including site scores from the first two ordination axes. Black circles mark the actively included modern diatom assemblages for each site. White circles mark the passively included pre-disturbance diatom assemblages for each site. The length and direction of the lines connecting pre-disturbance and modern assemblages indicates the magnitude and direction of assemblage change over time at each site. Site names are abbreviated as in Table 1

Discussion

Collectively, there is little change between pre-disturbance and modern diatom assemblages in our 18 study sites. When assemblage changes are examined at individual sites, however, lake-specific responses are observed. Furthermore, the absence of directional patterns in top–bottom metrics of assemblage change (i.e. species richness, Hill’s N2, DCA axis 1 and 2 sample scores, and the relative abundances of common taxonomic groups), when plotted along measured environmental gradients (e.g. pH, nutrients, DBT enrichment factor, etc.) indicate that there are no clear relationships between the degree of change and any measured environmental variables in these shallow lakes (ESM Fig. S3).

Importantly, the absence of consistency in diatom assemblage responses along industrial gradients (i.e. DBT EF, distance to nearest major industrial source) suggests that airborne pollution from local oil sands industry, including contaminants and bioavailable nitrogen, has not notably altered the assemblages of the mostly benthic algae of these lakes. The lack of a biological response to deposition of industrial airborne pollution is consistent with findings from other studies across the AOSR (Kurek et al. 2013a, b; Laird et al. 2017; Mushet et al. 2017; Summers et al. 2017) and

may be a consequence of naturally high nutrient concentrations. Given that our study lakes have high modern concentrations of nutrients (Table 1) and that there are lakes in Alberta with known nutrient-rich histories prior to settlement (Blais et al. 2000; Bayley et al. 2007), it is plausible that our study lakes were historically rich in nutrients. Furthermore, the majority of our study lakes are phosphorus-limited (Table 1; TN:TP column; $> 14 = \text{P-limited}$) and, as such, it is logical that the addition of growth-stimulating nitrogen (Summers et al. 2016) has not yielded marked changes in biological assemblages. The absence of biological changes corresponding to elevated levels of aerily deposited industrial contamination suggests that the diatom assemblages in our study lakes are resistant to the atmospheric pollution from the local activities.

Lake 2014-D is a unique case in this study. 2014-D is a shallow lake (0.9 m) that demonstrated a marked diatom compositional turnover from small, benthic fragilarioid taxa ($> 85\%$) in its pre-disturbance assemblage to over 60% abundance of *Aulacoseria granulata* in its modern assemblage. Given that 2014-D is very high in total phosphorus (TP; 2014-D = 341 $\mu\text{g/L}$) and that *A. granulata* is commonly found in meso-eutrophic to eutrophic conditions (Kilham and Kilham 1975), it is possible that the assemblage changes at this site were caused by increasing nutrient concentrations. 2014-D, however, is > 80 km away from the nearest major industrial source and its DBT EF is 1.6 (i.e. minimal “industrial impact”; Summers et al. 2016); thus, the community changes are not attributed to industrial inputs of biologically available nitrogen.

The muted diatom community changes in most of our shallow study sites are consistent with known responses to climate change in this region. In the AOSR, mean annual air temperatures have increased markedly since the beginning of the record in 1916 and, since the 1970s, have mostly exceeded the record-long average (Laird et al. 2013). Moisture in the region has declined with an increase in evaporation driven by warming that has outpaced the cyclical patterns of precipitation (Schindler and Donahue 2006). The shallow depths of our study lakes mostly preclude high abundances of planktonic taxa and, as such, the observed changes in the diatom communities are mainly, but not exclusively, among benthic taxa. In the majority of our shallow sites, we found changes from

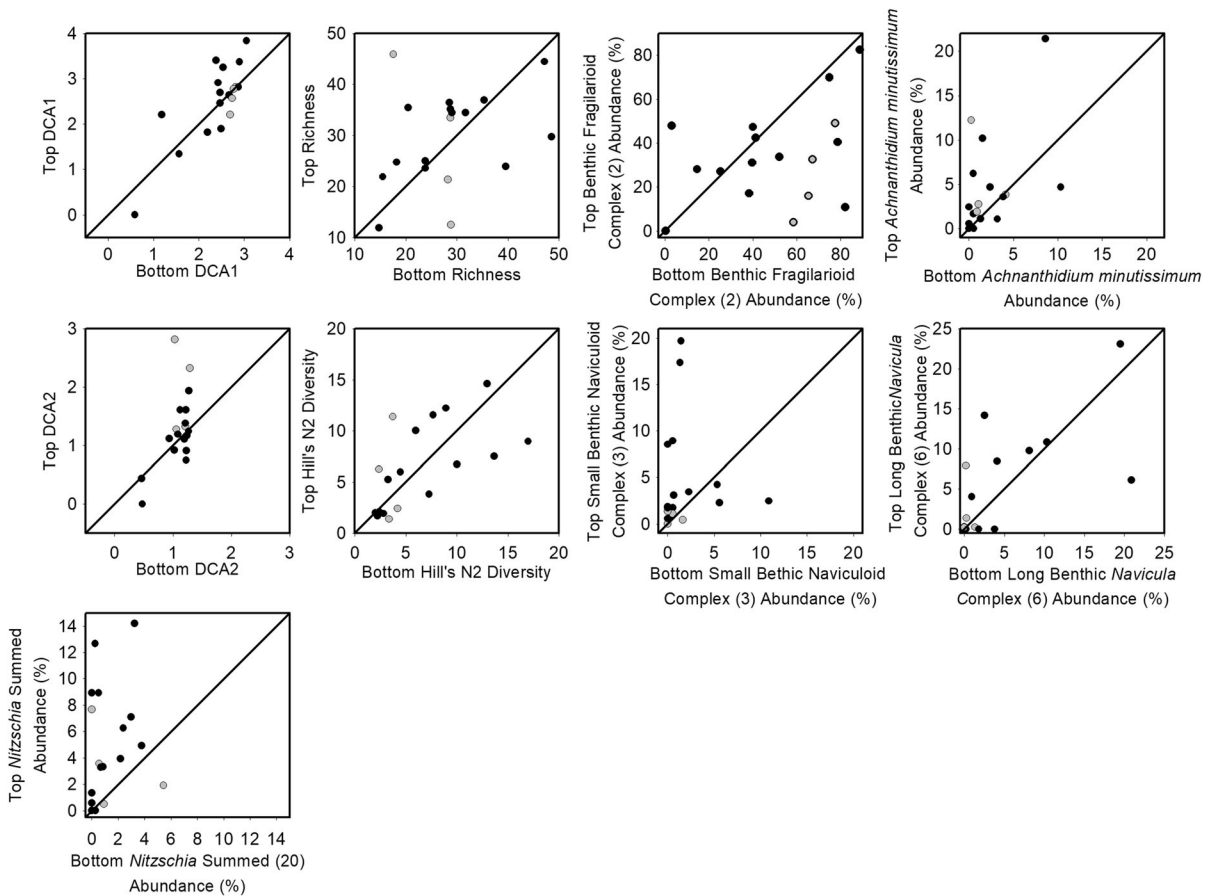


Fig. 5 Scatter plots of assemblage metrics and select common diatom taxa from the pre-disturbance (bottom; x axis) and modern (top; y axis) sediment sections of the 18 Athabasca Oil Sands Region (AOSR) study lakes. The benthic fragiliarioid complex includes *Staurosira construens* and *Staurosirella pinnata*. The small benthic naviculoid complex includes *Eolimna minima*, *Sellaphora atomoides*, and *Sellaphora seminulum*. The long benthic *Navicula* complex includes *Navicula cryptocephala*, *Navicula cryptotenella*, *Navicula*

pseudolanceolata, *Navicula radiosa*, *Navicula rhynchocephala*, and *Navicula vulpina*. Black circles denote sites with coring water depths ≤ 2 m. Grey circles denote sites with coring water depths > 2 m. Where applicable, the number of taxa included in the complexes and summed groups are indicated in parentheses following the group name. A 1:1 line is included. Samples that plot above the 1:1 line indicate greater values in the modern assemblages than in the pre-disturbance assemblages

pre-disturbance assemblages dominated by *Staurosira construens* and *Staurosirella pinnata* (small, epilithic, generalist, fragiliarioid taxa; Lotter and Bigler 2000) to modern assemblages that are still dominated by these fragiliarioid taxa, but with increased abundances of other benthic taxa with more complex environmental preferences (e.g. naviculoid taxa, *Nitzschia*; Figs. 3, 5). In many shallow lakes and ponds, warmer temperatures and extended growing seasons have been found to increase the availability and complexity of the benthic environment (e.g. increased abundance and diversity of macrophytes), favouring the

development of diatom assemblages with complex and diverse habitat requirements (e.g. periphytic taxa; Douglas et al. 1994; Rühland et al. 2014; Summers et al. 2017). The transitions can be subtle given the generalist nature of the small, benthic fragiliarioid taxa (Lotter and Bigler 2000). As such, the diatom assemblage transitions we recorded in the majority of our study lakes are subtle, but such changes are expected responses to a warming climate given the shallow depths of the sites (Rühland et al. 2014; Summers et al. 2017).

Although the majority of our study lakes demonstrated similar assemblage changes among benthic taxa (i.e. shifts from benthic, fragilarioid taxa to specialized benthic taxa), several sites showed increased abundances of planktonic taxa in modern assemblages. We found substantial assemblage shifts from a high dominance of benthic fragilarioid taxa to modern assemblages dominated by small, planktonic *Discostella stelligera* in some of the relatively deeper, but still shallow, sites in our set of lakes (2014-Z, 3.2 m; Pushup, 5.1 m; Fig. 3). We also observed changes in RAMP 268 (1.8 m) from a benthic fragilarioid-dominated (~ 40%) pre-disturbance assemblage to a modern assemblage comprised of > 20% *Fragilaria tenera*, which is a pennate, planktonic species. These assemblage changes are consistent with expected responses to changes in lake water properties and associated changes in resource availability that result from warming and longer ice-free periods in deeper lakes (Rühland et al. 2015). For example, top–bottom studies from across the Northern Hemisphere (Rühland et al. 2008), including lakes from the central Canadian subarctic (Rühland et al. 2003), northwestern Ontario (Enache et al. 2011), and New Brunswick (Harris et al. 2006), attributed high modern abundances of small, cyclotelloid taxa, including *D. stelligera*, to decreases in vertical mixing and increases in thermal stability and associated changes in light and internal nutrient distributions. Although our study sites are likely too shallow to thermally stratify, warmer conditions and a longer open-water period may have reduced the duration and strength of vertical mixing and increased water column stability, enabling planktonic diatoms to thrive in some of our sites.

The diatom assemblage changes in our study lakes are mainly subtle, but the nature of these changes are consistent with more substantial increases in whole-lake primary production that were found in all lakes of this study and are documented in Summers et al. (2016). The increasing primary production trends in the Summers et al. (2016) study were linked to climate warming and the associated effects on the region's lakes, while no clear link to a fertilization effect from local industry was detected (Summers et al. 2016). The clear increases in sediment-inferred whole-lake primary production corroborate the changes in the diatom assemblages (although they are subtle) in our 18 study sites, and support the conclusion that the region's

changing climate likely played an important role in structuring the biological communities in shallow AOSR lakes.

The differences in assemblage responses in our shallow lakes (i.e. most lakes transitioned to more complex benthic assemblages, whereas three lakes shifted to planktonic assemblages) are likely explained by site-specific conditions, and underscore the importance of the limnological context of a lake prior to a shift (Rühland et al. 2015). For example, 2014-Z (3.2 m) and Pushup (5.1 m), the two lakes that demonstrate a large assemblage turnover from benthic fragilarioid taxa to *Discostella stelligera*, are comparable in depth to RAMP 464 (4.6 m) and Kearl (3.5 m). Rather than shifting to assemblages dominated by planktonic taxa (as recorded in 2014-Z and Pushup), RAMP 464 and Kearl change to a more complex benthic assemblage, as is common in the majority of our study sites. Diatom assemblages from different lakes can have different responses to a widespread stressor such as climate change. Such different responses have been documented in the AOSR (Laird et al. 2017) and may be accounted for by differences in lake characteristics, such as morphologic and littoral characteristics (e.g. basin shape, shoreline substrate), that provide a variety of different benthic habitats for diatoms to exploit. Although we observed lake-specific responses, broad interpretation of the bottom-to-top changes in the assemblages of our 18 study lakes, spanning a range of industrial airborne contamination, provide a valuable assessment of the region's lakes and suggest relative resistance in the face of substantial environmental stress. Detailed, downcore analyses of the diatom assemblages in lakes that have high inputs of industrial airborne pollution and/or show large taxonomic changes between pre-disturbance and modern assemblages will further elucidate the effects of ongoing multiple stressors on the lakes in the AOSR.

Conclusions

Overall, the diatom assemblages in our shallow AOSR study lakes record only minor changes since ~ 1850. The muted responses may be a consequence of naturally high nutrient concentrations in the lakes buffering the response to contaminant and nutrient deposition from nearby industry. In addition, the

benthic diatom taxa that dominate the assemblages of these shallow lakes in most top and bottom samples are considered environmental generalists and would therefore tolerate stress and a wide range of environmental conditions. Although three study sites showed marked diatom community change between bottom and top sediment sections, no sites demonstrated changes clearly linked to deposition of industrial pollution. Instead, the majority of diatom assemblage responses are broadly consistent with indirect changes linked to climate warming. The differences in the nature and magnitude of the assemblage responses among seemingly comparable sites suggest that individual lake changes are modified by site-specific characteristics. Our research corroborates the findings of other paleolimnological studies in the AOSR, and farther downwind, that find aquatic community responses primarily determined by climate change, rather than industrial contaminant and nitrogen deposition (Hazewinkel et al. 2008; Curtis et al. 2010; Kurek et al. 2013a; Laird et al. 2013, 2017; Summers et al. 2016). The findings of this study highlight the relative biological stability of primary producers found in the region's lakes despite ongoing environmental stress.

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