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Article

Assessing long-term changes in aquatic ecosystems near a small conventional oil and gas operation in the Cameron Hills, southern Northwest Territories, Canada

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With 7 figures and 3 tables

Abstract: The Cameron Hills is a freshwater-rich region located at the border of Alberta and the Northwest Territories and is the site of a small, remote oil and gas operation. Ecological monitoring data are scarce in the Cameron Hills, and absent prior to the onset of oil and gas development in the 1960s. Consequently, the potential impacts of industrial activities on freshwater ecosystems in the Cameron Hills are unknown. Identifying ecosystem responses to industrial activities is further confounded by the effects of climate change, as this region has undergone substantial warming since ~1900. To address this important knowledge gap, we used an integrated spatial and temporal approach to investigate how climate warming and industrial activities may have altered water quality in the region. Water samples and sediment cores were collected from lakes with varying degrees of catchment disturbance related to oil and gas activities. Comparison of catchment characteristics and modern water chemistry data suggest that catchment disturbance may be increasing dissolved organic carbon (DOC) export to lakes. Additionally, lakes in close proximity to the central battery exhibit lower pH than more distant lakes, which may be due to inputs of organic acids. Changes in diatom assemblages preserved in a dated sediment core from a lake with a disturbed catchment are consistent with modern water chemistry, indicating a trend toward increasing DOC and decreasing pH. Despite evidence of localized impacts related to oil and gas activities, changes in diatom assemblages suggest that regionally climate warming is currently the dominant driver of changes in lakes in the Cameron Hills.

Keywords: diatoms; paleolimnology; dissolved organic carbon; pH; subarctic; climate change; oil and gas development

Introduction

The Cameron Hills, situated in the southwestern Northwest Territories (NT, Canada), was the second largest actively producing oil and gas field in the NT until the suspension of operations in 2015, second only to the Imperial Oil operations in Norman Wells. Although relatively small compared to the large petroleum deposits in the Alberta oil sands regions ~500 km to the southeast, the Cameron Hills operation is of

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particular interest as it represents oil and gas exploration and development in a relatively pristine subarctic environment. Additionally, it is situated upstream of Tathlina Lake, a culturally and economically important area to local communities. The Ka'a'gee Tu First Nation use the Tathlina Lake area for hunting, fishing, and trapping activities, and Tathlina Lake also sustains a small commercial winter walleye fishery (Stewart et al. 2016). Despite the exploration of oil and gas beginning here as early as the late-1960s (Paramount Resources Ltd. 1991), little is known about the impacts the Cameron Hills operation has had on aquatic ecosystems in this region.

Many activities associated with oil and gas operations have the potential to alter aquatic ecosystems. Land disturbance related to seismic exploration, well installations, pipeline installations, and transportation infrastructure may lead to the acceleration of permafrost thaw and soil erosion (Williams et al. 2013), which can alter aquatic ecosystems by increasing solute and sediment loads from the catchment (Kokelj et al. 2009; Kokelj et al. 2013; Coleman et al. 2015). Flaring, which is used to burn gas produced during well testing and gas production, removes most of the hydrogen sulfide (H_2S) and methane (Paramount Resources Ltd. et al. 2003). However, by-products from gas emissions and flaring, such as sulfur dioxide (SO_2) and mono-nitrogen oxides (NO_x) (Paramount Resources Ltd et al. 2003), can cause lake acidification (e.g. Dixit et al. 1987; Haines 1981; Schindler 1988; Psenner 1994; Hazewinkel et al. 2008; Curtis et al. 2010). Another major environmental concern related to oil operations is the potential for leaks and spills, which can result in widespread contamination of the landscape. Possible sources include drilling fluid leaking from storage facilities or pipeline leaks or breaks (Paramount Resources Ltd. et al. 2003). A recent study examining the impacts of oil and gas exploration in the Mackenzie Delta region, NT, Canada, found evidence of increased conductivity, particularly chloride, in lakes affected by saline drilling fluids leaching from compromised drilling sumps excavated in permafrost (Thienpont et al. 2013). Although the operation in the Cameron Hills uses storage tanks rather than sumps to store drilling fluid waste, the potential for leaks or failures remains. These potential stressors can have serious impacts on local fisheries, as they can be disruptive to spawning habitats (e.g. Soulsby et al. 2001; Greig et al. 2005), disrupt food chains (e.g. Schindler 1988) and introduce potentially deleterious contaminants to aquatic ecosystems.

Examining the specific impacts of industrialization on aquatic ecosystems is complicated further by the added stress of a warming climate (Smol 2010). The southern NT has been warming rapidly, with an increase in mean annual air temperature of 3.6 °C since 1896 (Coleman et al. 2015). This warming has the potential to alter aquatic ecosystems within the Cameron Hills, particularly as this region is situated within the sporadic discontinuous permafrost zone, which is highly sensitive to climate warming (Kettles & Tarnocai 1999). Recent studies examining landscape changes in the lowlands north of the Cameron Hills found evidence of actively degrading permafrost, and corresponding changes in aquatic ecosystems such as increases in concentrations of dissolved organic carbon (DOC) and nutrients (Coleman et al. 2015), as well as increased allochthonous mercury transport (Korosi et al. 2015). Increasing DOC concentrations, and associated increases in water colour, can have significant consequences for aquatic ecosystems such as changes in lake stratification and mixing regimes (Seekell et al. 2015; Vonk et al. 2015), and reductions in light penetration resulting in decreases in whole-lake primary production and reduced dissolved oxygen levels (Couture et al. 2015; Deshpande et al. 2017). As the Cameron Hills is within this sensitive permafrost region, it is possible that aquatic ecosystems here are being impacted by ongoing permafrost thaw. Other possible limnological effects resulting from warming temperatures include changes in the length of the ice-free season, changes in water depth and lake size due to permafrost degradation, and increasing habitat complexity due to a lengthened growing season (Smol & Douglas 2007; Vincent 2009; Rühland et al. 2013; Rühland et al. 2015). As a result, separating the influence of these individual stressors, as well as identifying interacting effects of industrialization and regional climate change, is difficult, especially given the minimal environmental monitoring that has occurred in this remote northern region.

The lack of long-term monitoring data poses a challenge in determining if aquatic ecosystems in the Cameron Hills are being affected by oil and gas activities and/or climate warming. In the absence of monitoring data, indirect methods are required to determine if aquatic ecosystems are changing as a result of these stressors. Paleolimnological methods offer a means to reconstruct past environmental conditions using physical, chemical, and biological indicators stored in lake sediments (Smol 2008). Diatoms (siliceous algae of the class Bacillariophyceae) are particularly useful indicators as they remain well-preserved in lake sediments and are ubiquitous in aquatic environments (Smol & Stoermer 2010). Additionally, diatoms are abundant, ecologically diverse, and often have well-defined environmental optima and tolerances, which makes them ideal for tracking changes in lake environments (Smol 2008). Diatoms can be used to reconstruct lakewater chemistry (e.g. pH, nutrients, and dissolved organic carbon) (Dixit et al. 1987; Pienitz & Smol 1993; Curtis et al. 2010; Hyatt et al. 2011), as well as habitat characteristics (e.g. depth and extent of littoral vegetation) (Michelutti et al. 2003; Rühland et al. 2008; Laird et al. 2011), and have been useful in tracking ecosystem change as a result of industrial operations (Hazewinkel et al. 2008; Curtis et al. 2010; Laird et al. 2013)

Modern water chemistry and dated sediment cores were examined in order to investigate the potential of the Cameron Hills oil and gas fields to affect nearby aquatic ecosystems. To assess the present-day impact of industrial activities, the modern water chemistry of 27 lakes was examined, and compared to the level of disturbance in the catchment, as well as distance from the main flare stack.

Using the paleolimnological approach, we examined past limnological changes, both in the context of industrial development and with regional climate change. We analyzed the sediments of seven lakes, with varying levels of catchment disturbance in a before-after (i.e. top-bottom) paleolimnological study (Smol 2008) to give a snapshot of how environmental conditions in the Cameron Hills have changed during the recent past. Briefly, the uppermost sediment interval from each core was used to characterize the modern (post-industrial) diatom assemblage, and an interval selected down-core was used to characterize pre-industrial diatom assemblages. Additionally, we selected two sediment cores among these seven lakes (one from a lake with relatively high catchment disturbance, and one from a lake with little disturbance), representing a few hundred years of sediment accumulation, to analyze changes in diatom assemblages in detail over time. Additionally, organic carbon content (%OC) was analyzed in these two sediment cores to assess changes in organic matter. To estimate changes in primary production over time, we selected four sediment cores from lakes with a range of catchment disturbance to analyze visible reflectance spectroscopy-inferred chlorophyll-*a* (VRS chl-*a*, Michelutti et al. 2010; Michelutti & Smol 2016).

Site description and operational history

The Cameron Hills region is located within the Taiga Plains High Boreal Ecoregion in the southern NT (Canada), straddling the border with Alberta (Fig. 1), and sits 400–500 m above the surrounding lowland region. Several lakes and bogs dot the landscape. The Cameron River winds its way throughout the region and terminates in Tathlina Lake, which is situated in the lowland region (Fig. 1). Surface materials in the Cameron Hills consist mainly of glacial till deposits and postglacial sediment, but rock outcropping consisting of shale, sandstone, and siltstone are also present (Paramount Resources Ltd. 2003; Ecosystem Classification Group 2007). The most common forests in the region include Lodgepole Pine (*Pinus contorta*) forests, mixed-wood forests, riparian forests and black spruce (*Picea mariana*) forests, which are often underlain by permafrost (Paramount Resources Ltd. 2003; Ecosystem Classification Group 2007). Mean annual air temperature (MAAT) has been increasing in the region since at least the beginning of the instrumental climate record at the Hay River Climate Station, located ~120 km away from the Cameron Hills, with an increase of 3.6 °C since the late-1800s (Coleman et al. 2015).

Interest in the Cameron Hills region for oil and gas development by Hudson's Bay and Oil Ltd. began in the early-1960s and exploration drilling began in the late-1960s (Paramount Resources Ltd. 1991). The exploration permit was converted to petroleum and natural gas leases in 1971, and in 1978 Paramount Resources Ltd became a participant. By 1991, 22 wells had been drilled associated with the Cameron Hills Significant Discovery License. In 2013, Strategic Oil & Gas Ltd. acquired the Cameron Hills project from Paramount Resources Ltd. Strategic Oil and Gas Ltd recently ceased active operations in the Cameron Hills (January 2015) in response to reductions in oil prices.

At the time of sample collection, the operation in the Cameron Hills consisted of 55 gas and oil wells (active, suspended and abandoned), gas and oil gathering and measurement systems, borrow pits, and a central battery including a flare stack and a permanent camp (Strategic Oil and Gas Ltd. 2014). The gathering system consists of pipelines that run from wells to the central battery via a main pipeline. A pipeline constructed in 2002 also runs from the central battery to a processing facility in Bistcho, Alberta (Strategic Oil and Gas Ltd. 2014). The operation also includes winter roads and all-terrain vehicle trails. Location of the flare stack and well sites, including abandoned, suspended and active wells are indicated in Fig. 1.

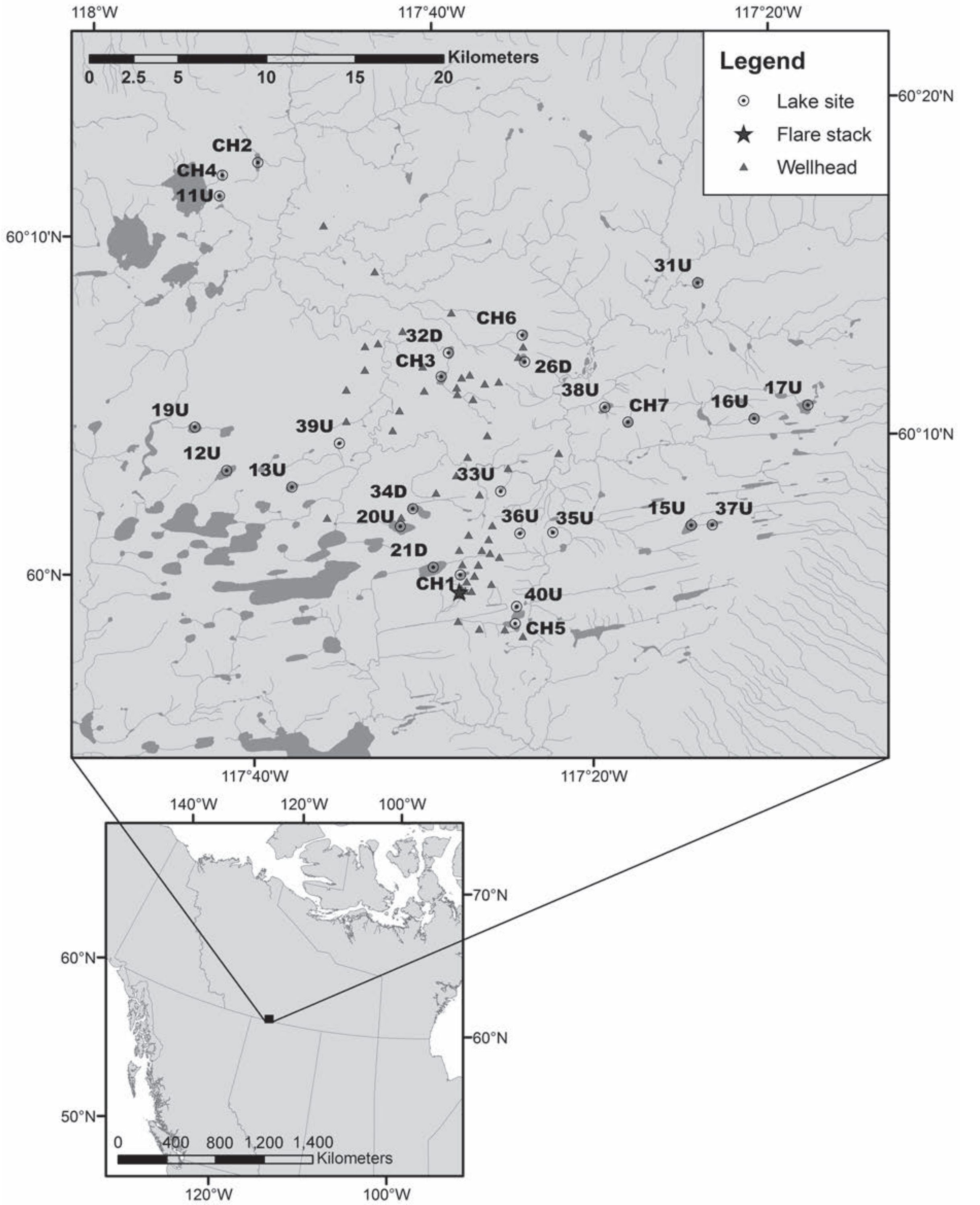

Fig. 1. Location of the study sites, abandoned, suspended, and active wells, and the flare stack within Cameron Hills (NT, Canada).

Methods

Field sampling

Twenty-seven lakes were selected for this study using 2010 IRS satellite imagery to capture a range of catchment disturbance. The catchment area for each of the study lakes was delineated using the Canadian Digital Elevation Data (CDED) digital elevation model (30 m resolution) by computing flow direction and using the watershed tool in ArcMap (v.10.1). Linear disturbance in the catchments, which includes pipeline, roads and seismic lines, was manually digitized in ArcMap from 2010 IRS satellite imagery. Only linear disturbance that could be identified with remote sensing was used in analyses. All lakes are shallow with maximum depths between 0.5–3 m and all but four study lakes (17U, CH6, 32A and CH3) are closed basins, having no visible water outlets. Sites were accessed by helicopter in September 2012, July 2013, and October 2014. Water samples were collected from a depth of approximately 30 cm in 1 L polyethylene bottles rinsed three times with lake water prior to collection. Following collection, sample bottles were placed in a cooler with ice packs and delivered to Taiga Environmental Laboratory in Yellowknife, NT (Canada) for analysis. Sediment cores were collected from seven of the study lakes (CH1 to CH7) (Fig. 1) in September 2012. Sediment cores were collected using a Glew (1989) gravity corer (internal diameter 7.6 cm), and sectioned using a Glew (1988) vertical extruder at 0.5 cm resolution until 10 cm, 1 cm resolution from 10–20 cm, and 2-cm resolution for the remainder of the core. Sediment samples were placed directly into individual WhirlPak® sample bags and frozen until analysis.

Water chemistry analysis

Water samples were analyzed for general chemistry (including major ions, dissolved organic carbon and nutrients) and 25 dissolved (filtered $< 0.45\ \mu m$) metal(loid)s following established methods (APHA 1992). Dissolved organic carbon was measured on a total carbon analyzer. Concentrations of cations and anions were determined by ion chromatography. Total (unfiltered) phosphorus was measured colorimetrically. Dissolved metal(loid) concentrations were determined by inductively coupled plasma mass spectrometry (ICP-MS) following EPA method 200.8 (Creed et al. 1994). Physical parameters of surface waters, including pH, specific conductivity and temperature, were measured *in situ* with a YSI 6920 multiprobe sonde (YSI Inc., Yellow Springs, Ohio). Water chemistry datasets were combined and for sites with more than one year of data the mean of all years of data was used in subsequent analyses. Water chemistry variables frequently ($> 50\,\%$) below detection limits were removed from all analyses. Normality was checked using the Shapiro-Wilk normality test and variables that were not found to be normally distributed were transformed either using a square-root transformation or log transformation if square-root transformation did not normalize the variable distribution. Pearson's correlation using the Bonferroni correction was calculated between the remaining water chemistry variables (total nitrogen (TN), DOC, total phosphorus (TP), alkalinity ($CaCO_3$), colour, conductivity (COND), pH, total dissolved solids (TDS), total suspended solids (TSS), calcium (Ca), magnesium (Mg), nitrate (NO_3), potassium (K), sodium (Na), sulphate (SO_4), aluminum (Al), arsenic (As), barium (Ba), copper (Cu), iron (Fe), lithium (Li), manganese (Mn), nickel (Ni), rubidium (Rb), strontium (Sr), titanium (Ti), and vanadium (V)), as well as site characteristics [extent of catchment disturbance (DSTRB), catchment area to lake area ratio (CA: LA), and distance from the main flare stack (DIST)]. The extent of catchment disturbance was calculated as the total kilometres of linear disturbance within a catchment divided by lake area to control for the influence of lake size. Generalized Additive Models were generated to determine whether catchment disturbance, the ratio of catchment area to lake area, or a combination of these site characteristics best predict water quality.

Principal components analysis (PCA) was used to assess variation between the lakes. Variables that were strongly correlated to each other were removed from the PCA (i.e. calcium, magnesium, strontium, potassium, iron, vanadium, and lithium are all strongly correlated to conductivity, and manganese was strongly correlated to colour and DOC). The PCA was conducted on the remaining 11 water chemistry variables, with skewed variables normalized using log or square root transformation. Catchment disturbance, catchment area to lake area ratio, and distance from the main flare stack were plotted passively on the PCA ordination plot. Correlations were conducted using the Hmisc package (Harrell & Dupont 2015) and the PCA was conducted using the vegan package (Oksanen et al. 2010) for R (R Development Core Team 2010).

Sedimentary analysis

^{210}Pb activity was analyzed on five of the seven sediment cores (CH1, CH2, CH3, CH5, and CH7) in order to establish chronologies (See Thienpont et al. 2017). This analysis was performed at the University of Ottawa using an Ortec high-purity germanium gamma spectrometer (Oak Ridge, TN, USA) and the constant rate of supply (CH1, CH2, CH3 and CH5) model and constant flux/constant sedimentation rate model (CH7)(Appleby & Oldfield 1978). Certified Reference Materials (IAEA 312 and IAEA 384) obtained from the International Atomic Energy Association (Vienna, Austria) were used for efficiency corrections. Results were analyzed using ScienTissiME (Barry's Bay, ON, Canada). Preindustrial sediments for the remaining two cores were selected using the chronologies and sedimentation rates of the five dated sediment cores as references. ^{137}Cs was also used as an independent dating verification method on the dated cores, as this radioisotope is associated with nuclear fallout which reached peak activity during 1963 when atmospheric nuclear testing was at its maximum (Appleby 2001).

From each of the seven cores, the uppermost sediment sample, and a sample from mid-core (to represent a pre-industrialization sample), were selected for diatom analysis. For CH2, CH4, CH5, CH6 and CH7 the midpoint samples were selected between 21 and 30 cm. Based on sedimentation rates estimated from ^{210}Pb dating these intervals were deposited before ~1850. The sediment cores for CH1 and CH3 were shorter than the other cores and as such interval selection was limited. For CH3, the interval at 17 cm was chosen as this was the bottom of the core, and for CH1, 7 cm was selected as diatoms concentrations were low below this interval. For both of these cores we are confident that these intervals occur prior to when oil and gas exploration began in the region, based on chronologies established from ^{210}Pb dating.

The cores obtained from lakes CH3 and CH7 were selected for detailed diatom and total organic carbon content analysis, as these cores were representative of a lake from a disturbed catchment (CH3) and a lake with little disturbance (CH7). The core obtained from CH1, arguably the most disturbed catchment, was 10 cm long, and deemed too short for detailed analysis, and diatom subfossils were sparse below 7 cm core depth.

Diatom slides were prepared according to standard methods outlined in Battarbee et al. (2001). To isolate diatoms, sediment subsamples were treated using a 1:1 (molecular weight) mixture of nitric and sulfuric acids. The mixture was placed in an 80 °C water bath for 3 hours in order to digest organic material. Samples were neutralized by being rinsed with deionized water daily for 7 days, with at least 22 hours between rinses to allow for diatom settling. Once samples reached a neutral pH (~7 rinses) they were plated on microscope coverslips to be mounted onto microscope slides using Naphrax®, a mounting medium with a high refractive index.

Diatom slides were analyzed using a Leica DMRB light microscope equipped with differential interference contrast (DIC) filters. Slides were viewed under an oil immersion lens at 1000× magnification. Diatom valves were identified to the species level according to numerous diatom references (Krammer & Lange-Bertalot 1997; Krammer & Lange-Bertalot 1999; Krammer & Lange-Bertalot 2000; Camburn & Charles 2000; Fallu et al. 2000). A minimum of 400 diatom valves were counted per interval up to a maximum of 700 valves, with counts generally between 450 and 600 valves per interval.

Total organic carbon content was determined by elemental analysis for selected intervals from cores CH3 and CH7. Freeze-dried samples were placed in a hydrochloric acid bath to remove inorganic carbon, rinsed and re-dried, and analyzed on a Vario EL Cube elemental analyzer at the G.G. Hatch (now Ján Veizer) Stable Isotope Laboratory at the University of Ottawa. Inferences of sedimentary chlorophyll-*a* (chl-*a*) were generated using visible reflectance spectroscopy (VRS) in order to reconstruct past primary production trends, following the method of Michelutti et al. (2010). This technique has been shown to be an accurate representation of trends in lake primary production through time, and incorporates estimates of both chlorophyll-*a* and its main diagenetic products (Michelutti et al. 2010; Michelutti & Smol 2016). For VRS-inferred chl-*a*, sediment intervals from 4 of the study lakes (CH1, CH2, CH3, and CH7) were sieved through 125 µm mesh and analyzed using a FOSS NIRSystem Model 6500 rapid content analyzer. VRS chl-*a* concentration was calculated using a linear regression following Michelutti et al. (2010).

Relative abundance was calculated for each diatom species, and the most abundant species were displayed in biostratigraphies using Tilia 1.7 (Grimm 2011). Some species were grouped together based on similar ecology or similar patterns of change. Species or ecological groups present at less than 2 % relative abundance were not included in the biostratigraphy. Biostratigraphic zones for CH3 and CH7 were identified using a constrained incremental sum of squares (CONISS) cluster analysis using the rioja package (Juggins 2014) for R and the number of significant zones was determined by comparison to a broken stick model in R (R Development Core Team 2010) using the vegan package (Oksanen et al. 2010). Species diversity was calculated using the Hill's N2 index (Hill 1973), performed using CANOWIN 4.51 (ter Braak & Šmilauer 2003). Both the Hill's N2 index and chl-*a* concentrations are displayed on the two full core diatom biostratigraphies, and for before-after samples the Hill's N2 values are displayed in a table.

Results

Water chemistry

Most lakes in the Cameron Hills region are mesotrophic to eutrophic (TP = 12–62 µg l^{-1}, with the exception of one lake (34D) that was highly eutrophic (TP = 94 µg l^{-1}). Lake water pH ranged from 5.9 to 8.0 with the majority of lakes circumneutral (6.5–7.5) (Table 1). Dissolved organic carbon concentrations ranged from 9.7–37.8 mg l^{-1} and are strongly correlated to lake water colour ($r = 0.86$, $p < 0.01$), which ranged from 9 to 404 TCU (Table 1).

The catchment characteristics examined are significantly correlated to many of the modern water chemistry variables. Catchment disturbance (calculated as the total kilometres of linear disturbance within a catchment divided by lake area, ranging from 0–0.87) is positively correlated to colour ($r = 0.59$, $p < 0.01$),

Table 1. Catchment characteristics and modern water chemistry statistics for 27 study lakes.

	Average	SD	Min	Max	Median
Lake area (ha)	32.6	24.8	5.8	92.5	20.9
Catchment area : Lake area	8.7	6.2	2.7	25.9	6.4
Disturbance (km disturbance/lake area)	0.2	0.2	0.00	0.8	0.08
Distance from Flare (km)	12.5	7.7	1.1	27.4	13.2
Nitrogen (unfiltered) (mg l^{-1})	0.9	0.3	0.5	1.7	0.9
DOC (mg l^{-1})	23.6	6.9	9.7	37.8	21.0
Phosphorus (unfiltered) (µg l^{-1})	35.9	19.0	12.	94.0	31.0
Alkalinity ($CaCO_3$) (mg l^{-1})	22.0	20.8	3.3	91.4	12.8
True Colour (TCU)	124	95	9	404	90
Conductivity (@25 °C) (µS cm^{-1})	59.7	39.0	24.9	184.0	44.9
pH	7.1	0.5	5.9	8.0	7.0
Total Dissolved Solids (mg l^{-1})	80.8	22.0	36.0	133.0	82.0
Total Suspended Solids (mg l^{-1})	5.3	3.4	3.0	16.0	4.0
Sodium (mg l^{-1})	0.8	0.4	0.2	1.6	0.8
Sulphate (mg l^{-1})	3.5	4.1	< 1.0	21.0	2.0
Aluminum (µg l^{-1})	37.4	34.5	0.6	132.7	32.6
Iron (µg l^{-1})	158.0	145.7	5.0	685.7	125.0
Titanium (µg l^{-1})	0.4	0.4	< 0.1	1.7	0.3

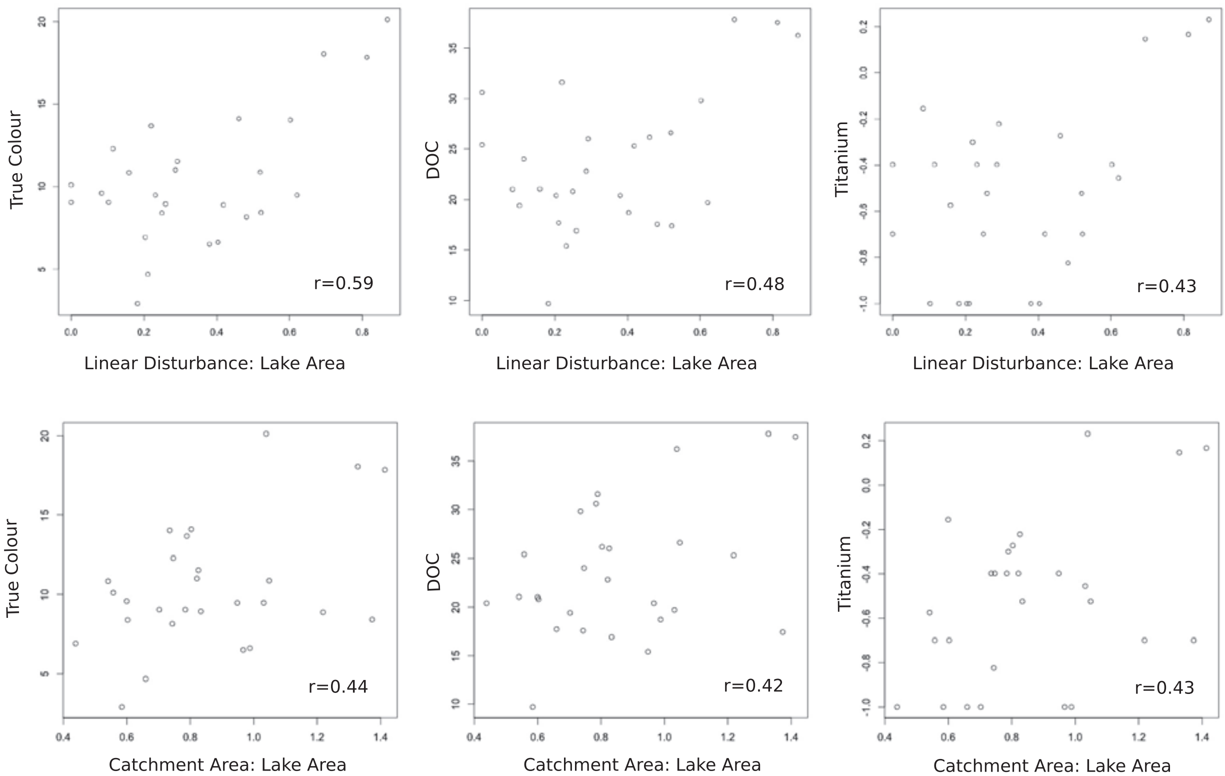

Fig. 2. Scatterplots of catchment characteristics and significantly correlated ($p < 0.05$) water chemistry variables. Colour and disturbance level have been square root transformed, titanium has been log transformed.

Table 2. Generalized additive model statistics.

Response	Predictor(s)	*p*-value	r^2 (adj)	AIC
DOC	Disturbance	0.01	0.205	178.8
DOC	CA:LA	0.03	0.144	180.85
DOC	Disturbance + CA:LA	0.16/0.58	0.182	180.54
Titanium	Disturbance	0.01	0.203	20.23
Titanium	CA:LA	0.03	0.151	21.94
Titanium	Disturbance + CA:LA	0.17/0.53	0.183	21.79

and concentrations of DOC (r = 0.48, $p < 0.01$), and titanium (r = 0.48, $p < 0.01$) (Fig. 2), as well as concentrations of aluminum (r = 0.50, $p < 0.01$) iron (r = 0.53, $p < 0.01$) and nickel (r = 0.49, $p < 0.01$). Catchment area to lake area ratio is positively correlated to colour (r = 0.44, $p < 0.01$), and concentrations of DOC (r = 0.42, $p < 0.01$), and titanium (r = 0.43, $p < 0.43$) (Fig. 2) as well as concentrations of iron (r = 0.41, $p < 0.05$) and nickel (r = 0.63, $p < 0.01$). Distance from the main flare stack is positively correlated to pH (r = 0.52, $p < 0.01$) as well as alkalinity (r = 0.44, $p < 0.05$) and negatively correlated to colour (r = –0.45, $p < 0.05$), and concentrations of DOC (r = –0.47, $p < 0.05$), sulphate (r = –0.60, $p < 0.01$), lithium (r = –0.46, $p < 0.05$), titanium (r = –0.38, $p < 0.05$), and vanadium (r = -0.55, $p < 0.01$). The Generalized Additive Models that best predict both DOC and titanium both have extent of catchment disturbance as the explanatory variable (Table 2).

The first PCA axis explains 55 % of the variation in the data set and is largely driven by pH and aluminum (Al) concentrations (Fig. 3). The second PCA axis primarily tracks nutrients (TN and TP) and explains 22 % of the variation (Fig. 3). In the PCA ordination space, lakes identified as having the highest disturbance level (CH1, CH3, 32D, 21D AND 39U) (disturbance level

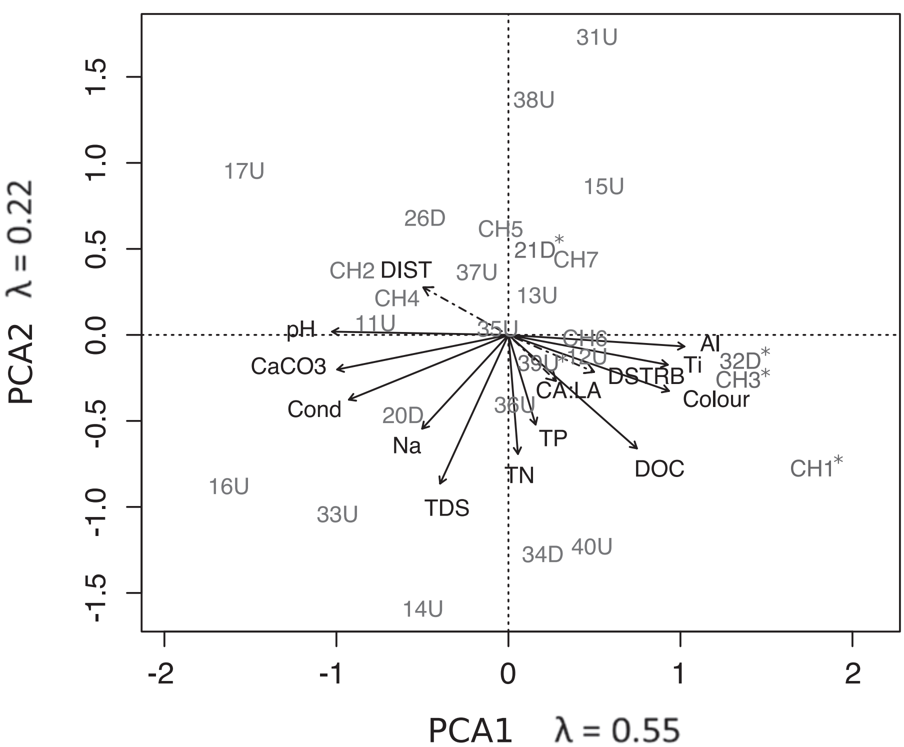

Fig. 3. Principal components analysis (PCA) of select chemical variables in the 27 lake data set. Dashed arrows represent variables that are plotted passively [distance from the flare stack (DIST), linear disturbance (DSTRB) and catchment area to lake area ratio (CA:LA)]. CaCO3, Cond, and Ti were normalized using log transformation, DSTRB, TDS, TP, Colour, and Al were normalized using square root transformation. *The 5 lakes with the most disturbed catchments are asterisked (Disturbance ratio >0.6).

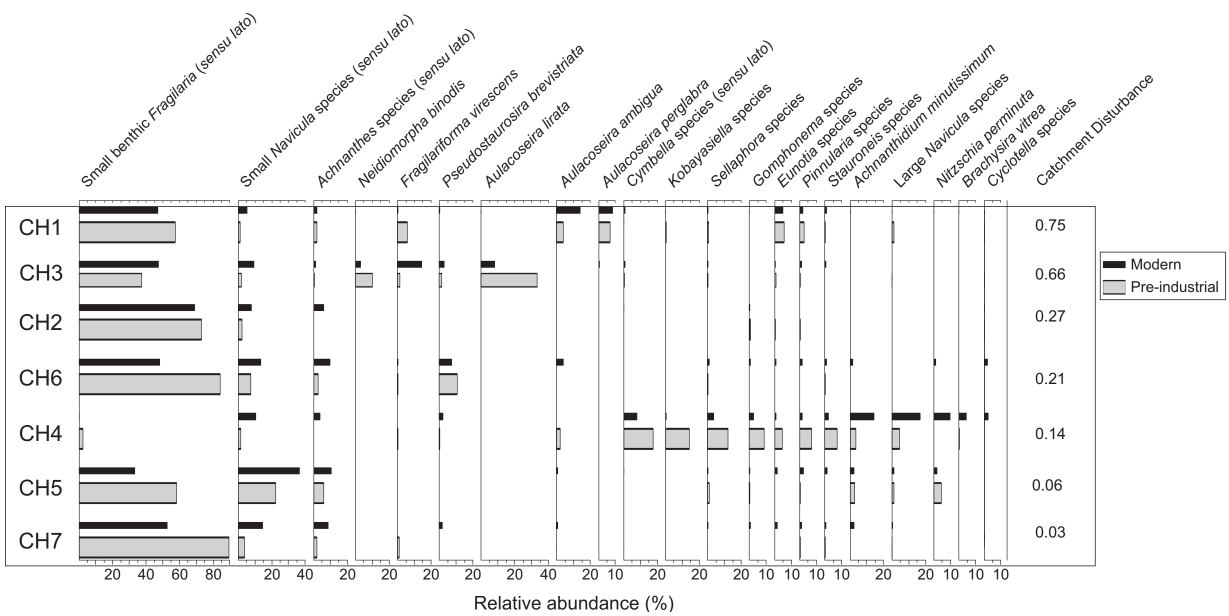

Fig. 4. Relative abundance diagram displaying the most common diatom taxa in the modern and pre-industrial sediments from seven of the study sites. Species or ecological groups present at less than 2 % relative abundance were removed.

between 0.60 and 0.87) cluster closely together, and are characterized by having higher concentrations of DOC, Al and titanium (Ti) as well as lower pH and alkalinity ($CaCO_3$) (Fig. 3). Additionally, CH1 has high total phosphorus concentrations (62.5 µg l^{-1}) compared to the mean of the data set (35.9 µg l^{-1}) (Fig. 3; Table

Table 3. Hill's N2 values for diatom species from top (after) and bottom (before) sediment samples from seven sediment cores.

	CH1	CH2	CH3	CH4	CH5	CH6	CH7
Top (After)	4.18	3.05	21.96	20.47	9.70	8.34	31.72
Bottom (Before)	2.86	4.25	12.20	21.03	3.51	2.38	7.25

1). Lakes identified as having the highest catchment area to lake area ratio (CH3, CH2, 32D, 33U, and 34D) are dispersed throughout the ordination space (Fig. 3).

Sedimentary analyses

Before-after analysis

Several trends in diatom assemblages from ~pre-1800s (pre-industrial) to present-day intervals were observed in the seven study lakes analyzed using the before-after (pre/post industrialization) approach (Fig. 4). All lakes except for CH3 show a decrease in small benthic *Fragilaria* species *sensu lato* (including *Staurosira construens*, *S. construens* var. *pumila*, *S. venter*, and *Staurosirella pinnata*). All seven lakes show an increase in small *Navicula* species *sensu lato* (*Eolimna submuralis*, *Fallacia indifferens*, *Microcostatus kuelbsii*, *Navicula minima*, *N. schadei*, *Naviculdicta pseudoventralis*, *Nupela vitiosa*, *Sellaphora disjuncta S. seminulum*, and *S. vitabunda*), and all lakes except for CH1 show an increase in *Achnanthes* species *sensu lato* (*Achnanthes acares*, *A. altaica*, *Achnanthidium subatomoides*, *Karayevia suchlandtii*, *Psammothidium curtissima*, and *Rossithidium pusillum*) (Fig. 4).

One lake, CH4, stands out as having a distinct diatom assemblage compared to the other study lakes (Fig. 4). In this lake, the pre-industrial assemblage is dominated by *Cymbella* species *sensu lato* (*Cymbopleura incerta*, *Encyonema gracile*, *E. hebridicum*, and *E. silesiacum)*, *Kobayasiella* species (*K. jaagii* and *K. subtilissima*), *Sellaphora* species (*S. laevissima* and *S. pupula)*, *Gomphonema* species (*G. acuminatum*, *G. angustatum*, *G. gracile* and *G. parvalum*), *Eunotia* species (*E. arcus*, *E. incisa*, and *E. praerupta*), *Pinnularia* species (*P. microstauron*, *P. gibba*, and *P. maior)* and *Stauroneis* species (*S. anceps*, and *S. phoenicenteron*). The modern assemblage is dominated by small *Navicula* species *sensu lato*, *Achnanthidium minutissimum*, large *Navicula* species (*N. cryptotenella*, *N. leptostriata*, *N. radiosa*, *N. rhynchocephala*, and *N. wildii*), and *Nitzschia perminuta.* For 5 lakes (CH1, CH3, CH5, CH6, and CH7) Hills N2 values are higher for the modern diatom assemblages compared to preindustrial assemblages (Table 3). For the remaining lakes (CH2 and CH4) Hills N2 values are slightly lower in the modern assemblage (Table 3).

Detailed analysis: CH7 – Low disturbance catchment

A significant shift in diatom species assemblage in CH7 occurred around the late-1800s/early-1900s (~12 cm) based on the CONISS cluster analysis (Fig. 5). Prior to this period (from 12–40 cm) the diatom assemblage remained relatively stable. This period was followed by a marked increase in *Navicula* species *sensu lato* (*Cavinula pseudoscutiformis*, *Navicula atomus*, *Naviculadicta absoluta*, *E. submuralis*, *F. indifferens*, *N. minima*, *N. vitiosa*, *S. disjuncta*, *S. pupula*, and *S. seminulum), Achnanthes* species *sensu lato* (*A. acares*, *A. altaica*, *A. ricula*, *A. minutissimum*, *A. subatomoides*, *K. suchlandtii*, *P. curtissima*, and *R. pusillum*), and *Nitzschia* species (mostly *Nitzschia fonticola*) and a decrease in small benthic *Fragilaria* species *sensu lato* (*S. pinnata*, *S. venter*, and *Pseudostaurosira brevistriata*) (Fig. 5). There was also an increase in species diversity around 12 cm, estimated using Hill's N2 number, and at about 6 cm there was an increase in estimated overall primary production, inferred using sedimentary chl-*a* (Fig. 5). *Fragilariforma exigua* was present throughout the period represented by the bottom part of the sediment core, but was not recovered above 4 cm (after ~1977).

Detailed analysis: CH3 – High disturbance catchment

Two significant shifts in diatom assemblage composition occurred throughout the length of the sediment core from CH3, based on the CONISS cluster analysis, and are marked by shifts between *Aulacoseira lirata*, and small benthic *Fragilaria* species *sensu lato* (*S. pinnata*, *S. venter*, and *P. brevistriata*) (Fig. 6). Specifically, an increase in *A. lirata* and a decrease in small benthic *Fragilaria* species began at approximately 10.5 cm and was followed by a reverse trend of decreasing *A. lirata* and increasing small benthic *Fragilaria* species *sensu lato* beginning in ~1970, near the time exploration began in the region (late-1960s). A slight increase in small *Navicula* species *sensu lato* (*Navicula subrotundata*, *F. indifferens*, *N. vitiosa*, *S. disjuncta*, and *S. seminulum)* and *F. exigua* occurred most notably from ~1974 to present (6–0 cm) (Fig. 6). Additionally, species diversity, estimated using Hill's

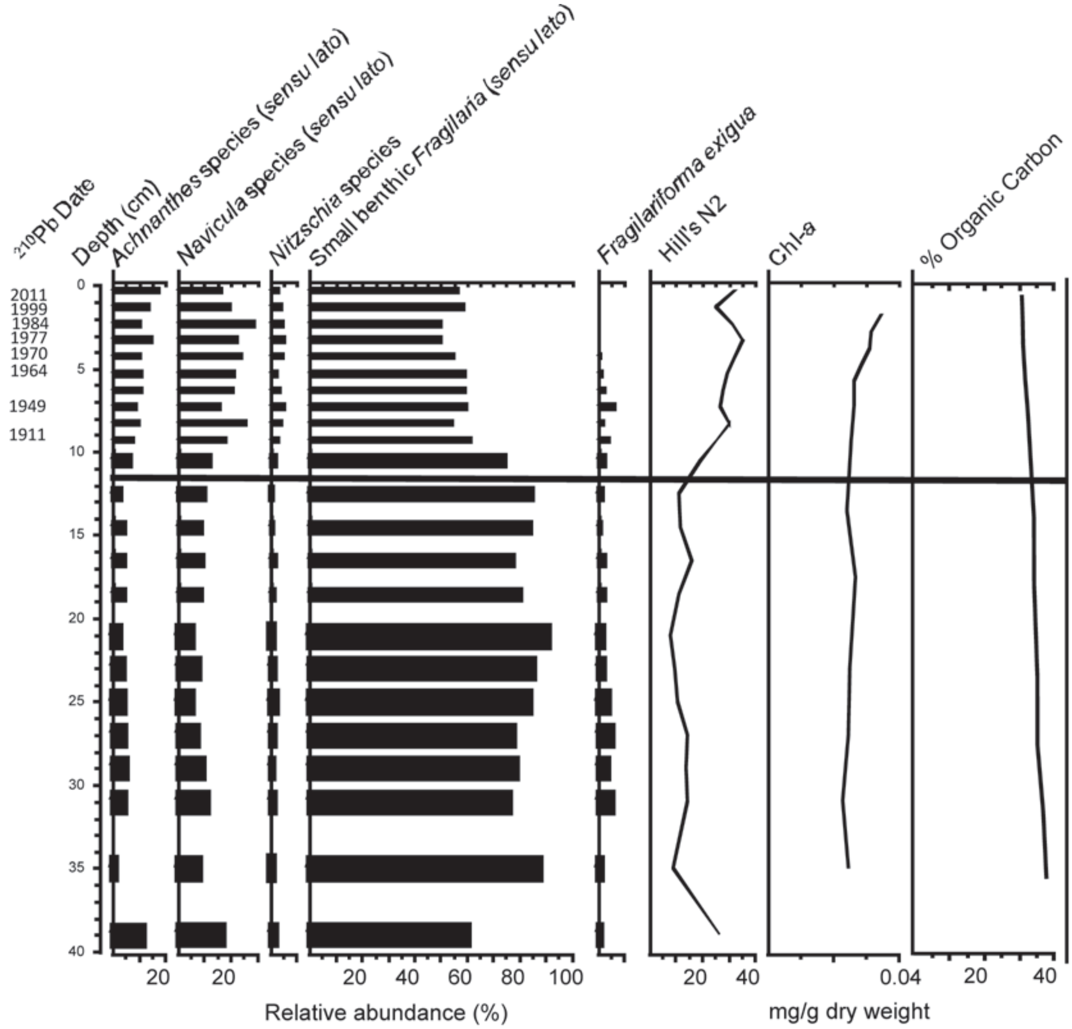

Fig. 5. Relative abundance diagram for the low-impact study lake, CH7, showing changes in the most common diatom taxa, as well as Hill's N2 diversity index, visible reflectance spectroscopy-inferred chlorophyll-*a* (VRS-chl-*a*), and percent organic carbon (%OC). Horizontal lines shows biostratigraphic zones based on constrained incremental sum of squares cluster analysis. Dates derived from ^{210}Pb activity using a constant rate of supply (CRS) model are shown on the left. Species or ecological groups present at less than 2 % relative abundance were removed.

N2 number, and overall primary production, estimated using VRS chl-*a,* also began to increase from ~1974 to present (6–0 cm) (Fig. 6).

Total organic carbon and chlorophyll-*a* reconstructions

A slight and gradual decrease in sedimentary total organic carbon content from the bottom towards the top of the core is observed in both CH7 (~39 % at 35 cm to 30 % at the surface) and CH3 (~22 % at 17 cm to 17 % at the surface). Chlorophyll-*a* and its main diagenetic products (referred to here as chl-*a*), which was inferred from sediment cores from four lakes (CH1, CH2, CH3 and CH7), shows an increasing trend in three lakes that began in the mid-1900s and an increase in one lake, CH1, beginning in the mid-1800s to early -1900s (Fig. 7).

Discussion

Impacts of catchment disturbance

A significant, positive correlation between catchment disturbance and several modern water chemistry variables (DOC, colour, Al, Fe, Ni and Ti) (Fig. 2) suggests that catchment disturbance may be impacting the water chemistry of these lakes, however it is important to note that these relationships are strongly driven by the

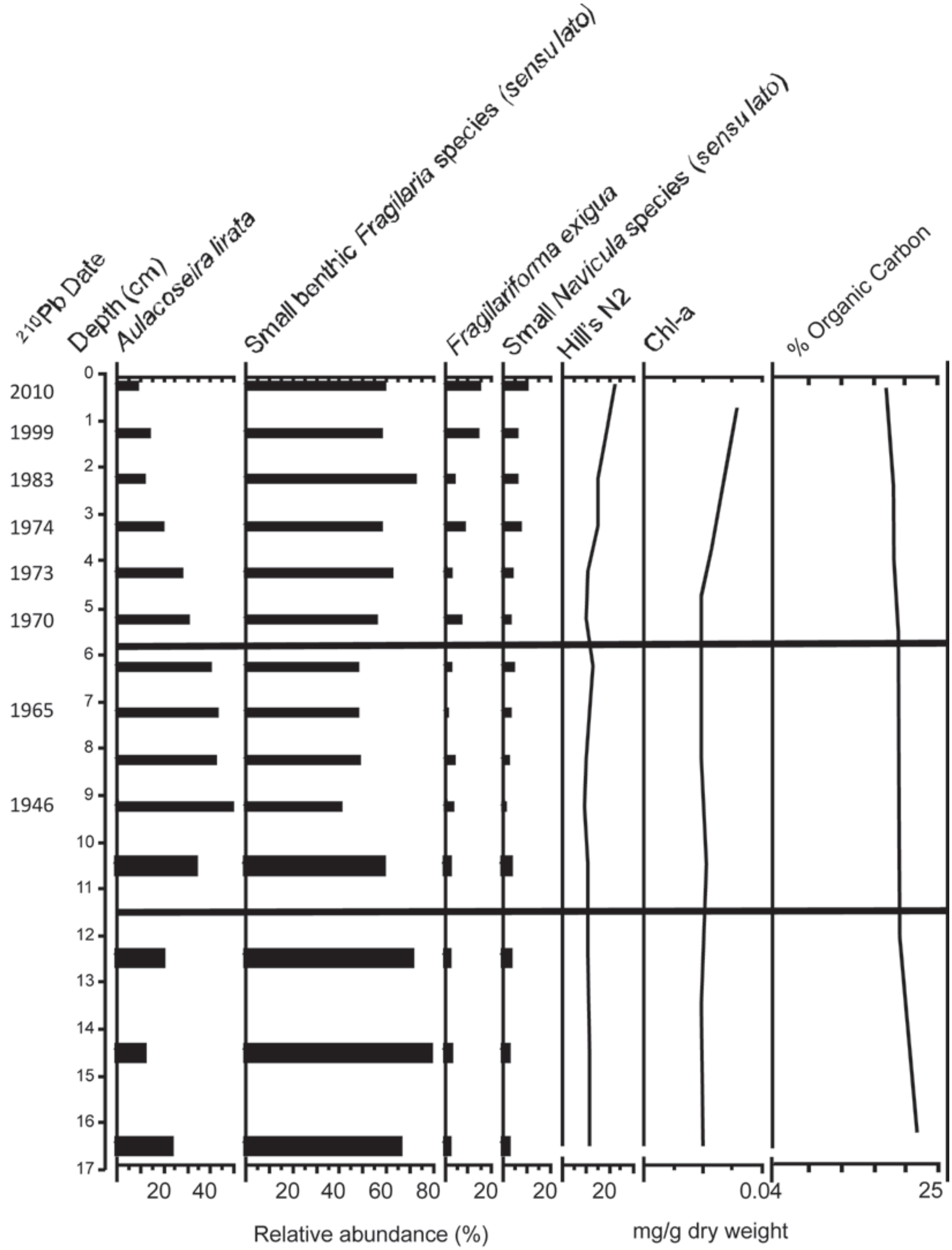

Fig. 6. Relative abundance diagram for the disturbed lake, CH3, showing changes in the most common diatom taxa, as well as Hill's N2 diversity index, visible reflectance spectroscopy-inferred chlorophyll-*a* (VRS-chl-*a*), and percent organic carbon (%OC). Horizontal lines shows biostratigraphic zones based on constrained incremental sum of squares cluster analysis. Dates derived from ^{210}Pb activity using a constant rate of supply (CRS) model are shown on the left. Species or ecological groups present at less than 2 % relative abundance are not included in the figure.

three lakes with the most disturbed catchments (CH1, CH3, and 32D). Increases in DOC and associated increases in colour have been linked to permafrost thaw in the lowland region surrounding the Cameron Hills (Coleman et al. 2015). Catchment disturbance related to seismic exploration, well installations, pipeline installations, and transportation infrastructure, may be accelerating permafrost thaw, particularly in CH1, CH3, and 32D, resulting in elevated DOC and colour. Additionally, increased concentrations of Ti have been

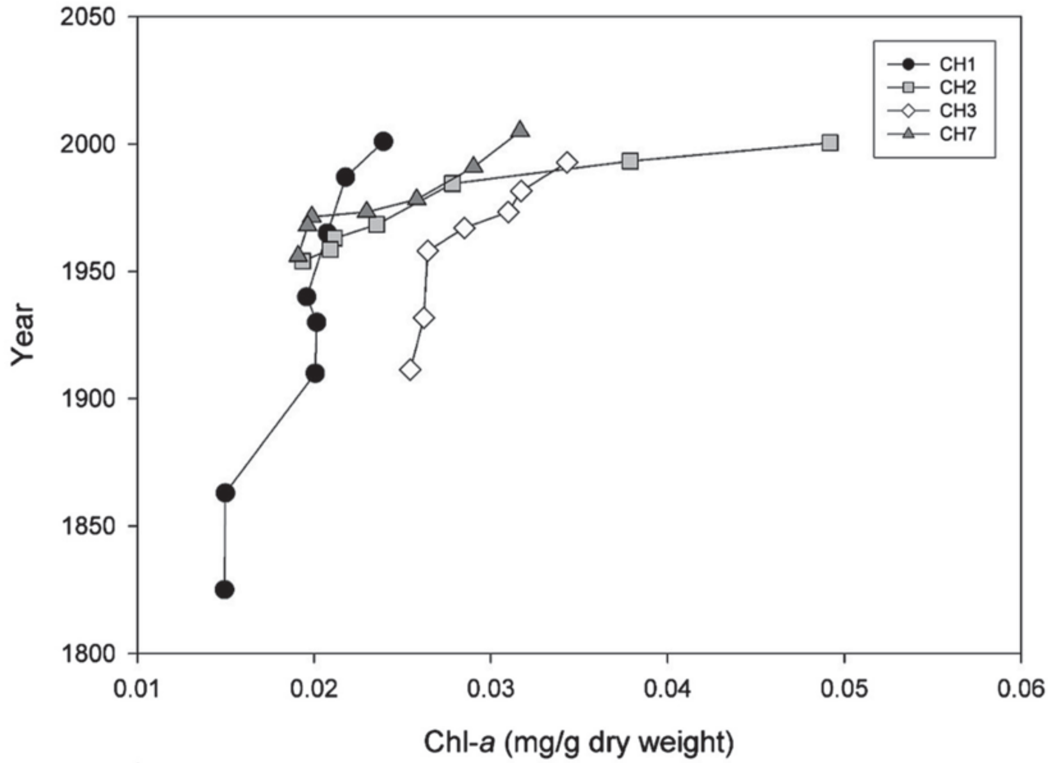

Fig. 7. Profile of visible reflectance spectroscopy-inferred chlorophyll-*a* (VRS-chl-*a*) trends, which includes the main diagenetic products (Michelutti et al. 2010) analyzed from four of the seven sediment cores that were obtained from the study sites.

linked to soil erosion (e.g. Massa et al. 2012). As both DOC and titanium are also significantly correlated to the catchment area to lake area ratio, generalized additive models were used and indicate that the level of catchment disturbance is a better predictor of both DOC and titanium, over catchment area to lake area ratio or a combination of the two variables (Table 2). Additionally, the five most disturbed lakes cluster together on the PCA ordination plot, and are characterized as having high DOC, colour, Al, Fe (not shown) and Ti and lower pH (highlighted in Fig. 3), while the lakes with the highest catchment area to lake area ratio are dispersed throughout the ordination. Together these analyses suggest that catchment disturbance is altering the water chemistry of these lakes. Research examining land use and DOC suggests that land disturbance can alter DOC export to aquatic ecosystems (Pagano et al. 2014).

The detailed downcore diatom analysis for CH3, a lake with a higher disturbance in the catchment, was challenging to interpret as a result of multiple potential stressors. The major trend recorded in diatom assemblages from CH3 is an apparent fluctuation between *Aulacoseira lirata* and small benthic *Fragilaria* species *sensu lato* (Fig. 6). The initial increase in *A. lirata* and decrease in small benthic *Fragilaria* species *sensu lato* (beginning around 12 cm) predates the beginning of oil and gas exploration in the region, and therefore this increase is unrelated to industrial activities. Tychoplanktonic *Aulacoseira* species are typically relatively heavy because of their heavily silicified valves, requiring turbulent mixing to remain in the water column, so decreases in relative abundances are often driven by enhanced lake stratification (Rautio et al. 2000; Rühland et al. 2008; Rühland et al. 2015). As this lake is very shallow, between 0.6-1 m, stratification is unlikely to occur, and unlike many other locations, *Aulacoseira* are increasing in CH3, thus the fluctuation in *A. lirata* is likely tracking another limnological change. This region has been warming since at least the late-1800s, coincident with the timing of the shift in relative abundance between *Fragilaria* and *Aulacoseira* taxa. *A. lirata* may be responding to changes related to warming temperatures such as a longer ice-free growing season impacting this shallow, polymictic lake.

Changes in diatom assemblages since the onset of industrial operations are consistent with an increase in

DOC and associated water colour, as observed in the water chemistry analyses outlined above. The increase in small benthic *Fragilaria* species *sensu lato* that begins around 1970 (Fig. 6) is atypical, although not entirely unexpected here given the high DOC and colour of CH3. These species are characteristic of harsh environments, such as low-light conditions found with extended ice cover and short growing seasons (e.g. Smol 1988; Lotter & Bigler 2000; Smol et al. 2005). With warming air temperatures and a corresponding increase in the ice-free period, previous studies have mainly recorded decreases in these species coinciding with an overall increase in species diversity (e.g. Smol et al. 2005). However, several studies have also found links between these species and high DOC and colour (Pienitz & Smol 1993; Pienitz et al. 1999; Bouchard et al. 2013; Coleman et al. 2015). These small benthic *Fragilaria* species were present in high abundance at the bottom of the CH3 sediment core (Fig. 6), and began to decrease slightly in relative abundance, perhaps as other species became more competitive in response to warming temperatures, only to increase again around the time oil and gas exploration began in the region. This increase may be a result of increasing DOC, resulting in high colour and low light penetration in the lake. A study examining 77 lakes across the subarctic Canadian treeline that *A. lirata* is more often found in lakes with low DOC (Rühland et al. 2003), so its decrease here may be a result of increasing DOC over the last 40 years.

Impacts of industrial emissions

The flaring of natural gas has the potential to impact aquatic ecosystems through the emission of SO_2 and NO_x, which can result in acidification as these chemicals are removed from of the atmosphere by precipitation and are deposited on the landscape. However, studies of limnological impacts of the much larger operations to the south in the Alberta oilsands region have found little evidence of acidification (Hazewinkel et al. 2008; Curtis et al. 2010; Laird et al. 2013). In the Cameron Hills, a significant relationship between pH and distance from the flare stack was detected ($r = 0.52$, $p < 0.01$), which would suggest that the operation may be acidifying nearby lakes, however the mechanism is not straightforward. There is no correlation between pH and sulphate ($r = 0.10$, $p > 0.05$) and CH1, the lake closest to the flare stack (Fig. 1) and which has the lowest pH (5.9) in the data set, has sulphate concentrations below detection (<1 mg l^{-1}). However, there is a strong, negative relationship between DOC and pH ($r = -0.66$, $p < 0.01$). The high DOC in lakes near the flare stack may therefore be contributing to lower pH due to the presence of natural organic acids, potentially from increased catchment disturbances.

In CH3, an increase in the relative abundance of the acidophilic diatom species *Fragilariforma exigua* (basionym *Fragilaria virecens* var. *exigua*), began in ~1974, reaching its highest abundance from ~1999 to the present (Fig. 6). CH3 is also one of the lakes identified as having lower pH (6.5) compared to the regional average (7.1), and this may therefore be indicative of decreasing pH in this lake. As discussed above, decreasing pH may be due to emissions (although this lake also has sulphate levels below detection) or it may also be associated with increasing DOC concentrations in the lake.

Interestingly, in CH1 (the lake closest to the main flare stack) there are no shifts in diatom relative abundances since pre-industrialization that are consistent with acidification based on our before-after comparison (Fig. 4). Instead there is indication of nutrient enrichment, identified by the increase in *Aulacoseira ambigua* between preindustrial and modern assemblages, and is supported by the modern water chemistry which shows this lake to be high in TP (62.5 µg l^{-1}), especially compared to the other lakes in this region (Table 1). Nutrient enrichment may be the overriding stressor in this lake and changes in diatom assemblages related to acidification may be too subtle to detect. Studies of lake impacts from larger developments have identified a trend toward greater productivity in lake systems near oilsands operations (Hazewinkel et al. 2008; Curtis et al. 2010), corroborating the results observed in CH1. Although we can only speculate as to the cause of nutrient increases in CH1, internal nutrient loading, atmospheric deposition, and increased export of nutrients from the catchment have been suggested in other similar environments (Hazewinkel et al. 2008; Curtis et al. 2010).

Impacts of climate change

The before-after analysis comparing changes between preindustrial and modern diatom assemblages in lakes with varying levels of catchment disturbance suggest that the aquatic ecosystems in the Cameron Hills are following similar trajectories, despite differences in disturbance from industrial activities (Fig. 4). The seven lakes show some consistent trends: all but one lake (CH3) exhibited a decrease in small benthic *Fragilaria* species *sensu lato*, all lakes showed an increase in small *Navicula* species *sensu lato*, and all but two lakes (CH1 and CH3) showed an increase in *Achnanthes* species (Fig. 4). As stated above, decreases in small benthic *Fragilaria* species have been associated with increases in the ice-free period with warming air temperatures (e.g. Rühland et al. 2015). The corresponding increases in *Navicula* species *sensu lato* and *Achnanthes* species s*ensu lato* are most likely a result of a more complex habitat that is also associated with a warming environment. In these lakes, there was a high diversity of both of these diatom groups , and this is reflected in the increasing Hill's N2 species diversity estimates seen in most lakes (Table 3). The detailed core analysis of CH7 is consistent with what is seen in the before-after analysis of the seven lakes and show a trend consistent with a warming environment. Most notably we see a decrease in small benthic *Fragilaria* species *sensu lato* and a corresponding increase in *Achnanthes* species *sensu lato* and small benthic *Navicula* species *sensu lato* beginning between 10 and 12 cm (dates could not be established for this period) (Fig. 5). An increase in primary production, inferred by sedimentary chl-*a* concentrations, is observed in all four cores analyzed in detail (Fig. 7) and is also indicative of a warming environment. A slight decrease in %OC is observed in both the CH3 and CH7 sediment cores (Figs 5 and 6) over the time period represented by these records. As primary production appears to be increasing based on VRS-chl-*a* inferences, the decline in %OC is unlikely due to decreased algal production, but may instead be related to increased mineral inputs from the surrounding catchment. Since %OC is already declining prior to oil exploration in the region it is unlikely to be related to industrial activities, instead representing a long-term, gradual trend.

Conclusions

The Cameron Hills region has been impacted by multiple environmental stressors, and untangling the relative impacts of these stressors can be challenging. The comparison of modern water chemistry and environmental disturbance due to oil and gas activities suggests that these activities can result in localized impacts to these pristine, remote environments. Specifically, the positive relationship between catchment disturbance, Ti (an indicator of soil erosion), and DOC indicate that land disturbance may be increasing export of DOC from catchments to aquatic ecosystems in the Cameron Hills. The modern water chemistry results also show lower pH in lakes closer to the main central battery. This relationship may be due to emissions from the flare stack, however the strong relationship to DOC concentrations suggest it may be more likely that increasing DOC concentrations are driving pH in these lakes.

Environmental inferences using proxies preserved in lake sediments are consistent with water chemistry interpretations. Changes in diatom assemblages in a sediment core from a lake with a disturbed catchment (CH3) suggest increasing DOC and associated colour, which is consistent with the timing of oil and gas development, as well as a more recent acidification signal, possibly related to the increasing DOC. Additionally, the sediment core from the currently eutrophic lake that is nearest to the central battery (CH1) shows a shift in diatom assemblages from pre-industrialization to present that is indicative of higher nutrient conditions.

Despite indications that oil and gas activities are driving changes in nearby aquatic ecosystems through landscape disturbance, environmental changes inferred from proxies stored in lake sediments indicate that, regionally, climate warming appears to be the dominant factor driving aquatic ecosystem change. The dominant shift in diatom species identified from sediment cores collected from seven lakes is from an assemblage of species that are typical under harsher environments (often associated with colder temperatures) to a more diverse assemblage often associated with warming temperatures and a more complex ecosystem. Similarly, an increase in inferred chlorophyll-*a* identified in the sediment records is indicative of increasing primary production, consistent with warming temperatures and decreased ice cover.

As oil and gas continues to be a valuable Canadian export, interest in oil and gas development is expected to continue. Increases in DOC concentrations in aquatic ecosystems has been observed across the circumpolar north, and although different driving mechanisms have been implicated (Pagano et al. 2014), permafrost regions appear to be particularly susceptible (Coleman et al. 2015; Vonk et al. 2015; Wauthy et al. 2018). Results from this study suggest that the activities of this small operation may have increased DOC loading to nearby aquatic ecosystems. This highlights the importance of establishing environmental monitoring activities for these small oil and gas operations since they may be an important, yet underappreciated, mechanism in altering terrestrial loading to aquatic systems.

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Authors' contributions

J.P.S., J.M.B., M.J.P., J.B.K., J.R.T. and K.A.C. conceived of and designed this study. K.A.C. performed experiments and analysed data. M.J.P. analysed remote sensing data. J.P.S., J.M.B., M.J.P., J.B.K., J.R.T contributed materials and analytical expertise. K.A.C. wrote the paper with significant contributions from all authors.

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