LONG-TERM LIMNOLOGICAL DYNAMICS IN MULTIPLE-STRESSOR SYSTEMS IN THE ATHABASCA OIL SANDS REGION, CANADA

by

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Abstract

Lakes in the Athabasca Oil Sands Region (AOSR) are threatened by multiple environmental stressors linked to industrial development of the local hydrocarbon deposits and climate change. Waterbodies are vulnerable to changes in water quality and quantity, as well as shifts in ecosystem structure and function. Due to insufficient monitoring prior to industrial development, little is known about the long-term changes in AOSR lakes and the effects of multiple stressors in the region. This thesis uses paleolimnological approaches to determine baseline conditions, track biotic changes through time, and investigate the relative effects of industrial pollution (i.e., contaminants and nutrients) and climate change on the environmental trajectories of the region’s shallow lakes. A multiproxy study of a shallow, isolated lake receiving substantial industrial contamination assessed how multiple trophic levels of a typical AOSR lake have changed over the past ~75 years. While there was no evidence of a threshold-type response to industrial pollution, biotic assemblages from multiple trophic levels suggest the benthic environment increased in complexity, consistent with the warming climate. Spectrally-inferred sedimentary chlorophyll-α profiles from 23 lakes and bioavailable nutrient deposition maps were used to investigate the extent, timing, and causes of increased primary production. Widespread, asynchronous increases in primary production and correlations to observed air temperatures suggest climate change as the main driver of elevated primary production, rather than bioavailable nutrients from industry. Finally, a “top-bottom” paleolimnological analysis comparing pre-disturbance (~1850) and present-day subfossil diatom assemblages from 18 shallow lakes was completed to survey the region’s biotic changes and investigate which environmental variables structured the diatom communities. While diatom assemblages in some lakes changed markedly, most AOSR lakes demonstrated resiliency to the region’s stressors. There were no apparent biological responses to industrial airborne pollution, including deposition of bioavailable nutrients. Instead, most diatom community shifts were consistent with known responses to climate change, and were likely mediated by lake-specific characteristics. Collectively, this research concludes that anthropogenic climate change is the main driver of overall muted community change and
marked primary production increases in the shallow lakes of the AOSR. Yet, future changes are possible given the expected continuation of the region’s industrial development.
Co-Authorship

Chapter 2 was co-authored with Joshua Kurek, Kathleen M. Rühland, Erin E. Neville, and John P. Smol. The chapter represents original work as part of my PhD thesis. Field work was competed as part of the Canada-Alberta Joint Oil Sands Monitoring (JOSM) program. I co-designed the project with Joshua Kurek, analysed the diatom samples, conducted all statistical analyses, and I am the primary author on the paper. Joshua Kurek analysed the cladoceran samples and Erin Neville analysed the chironomid samples as part of her Honours thesis project in Biology at Queen’s University. Kathleen Rühland assisted with diatom identification, statistical analyses, and development of the manuscript. The chapter has been published separately and is slightly modified in the thesis.


Chapter 3 was co-authored with Joshua Kurek, Jane L. Kirk, Derek C. G. Muir, Xiaowa Wang, Johan Wiklund, Colin A. Cooke, Marlene S. Evans, and John P. Smol. The chapter represents original work as part of my PhD thesis. Field work was competed as part of the Canada-Alberta Joint Oil Sands Monitoring (JOSM) program. I designed the project, analysed a majority of the spectrally-inferred sedimentary chlorophyll-α samples, conducted all statistical analyses, and am the primary author on the paper. The chapter has been published separately.

Chapter 4 was co-authored with Kathleen M. Rühland, Joshua Kurek, and John P. Smol. The chapter represents original work as part of my PhD thesis. Field work was completed as part of the Canada-Alberta Joint Oil Sands Monitoring (JOSM) program. I participated in the 2015 core sampling campaign. I designed the project, analysed the diatom samples, conducted all statistical analyses, and I am the primary author on the paper.
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Dedication

This thesis is dedicated to my parents – life-long learners, dreamers, and lovers of lakes
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List of Abbreviations

ANOSIM – analysis of similarities
AOSR – Athabasca Oil Sands Region
CCA - canonical correspondence analysis
CRS – constant rate of supply
DBT – dibenzothiophene
DCA – detrended correspondence analysis
DIC – dissolved inorganic carbon
DIN – dissolved inorganic nitrogen
DOC – dissolved organic carbon
ECCC – Environment and Climate Change Canada
EF – enrichment factor
IndVal – indicator value
JOSM – Joint Oil Sands Monitoring
PAC – polycyclic aromatic hydrocarbon
PCA – principal components analysis
PPE – priority pollutant element
RAMP – Regional Aquatics Monitoring Program
SRP – soluble reactive phosphorus
TDP – total dissolved phosphorus
TN – total nitrogen, unfiltered
TP - total phosphorus, unfiltered
VRS-chla – visible reflectance spectroscopy chlorophyll-a
Chapter 1

Introduction and literature review

1.1 Multiple stressors on lakes

Freshwaters are indispensable resources of market (e.g., drinking water, transportation, electricity generation, fisheries, and tourism) and nonmarket (e.g., habitat for plant and animal life, support for other ecosystems, and intrinsic natural value) goods and services (Wilson and Carpenter 1999). As the global environment changes due to expanding and intensifying anthropogenic impacts, the demands and stresses on many freshwaters increase and ecosystems may become degraded. Often, freshwaters are simultaneously affected by multiple stressors including, for example, natural processes and cycles, pollution, and over-use (e.g., overfishing). Further, climatic changes associated with global warming are now common worldwide and interact with stressors in a region, often multiplying the threats to lakes (Smol 2010). Multiple stressors may interact synergistically or antagonistically to yield amplified or diminished additive ecological responses, respectively (Folt et al. 1999; Piggott et al. 2015). Additionally, stressors can interact non-additively to yield complex and relatively unpredictable environmental consequences, which can be complicated and difficult to predict (Christensen et al. 2006; Smol 2010). The complex and sometimes confounding effects of several stressors occurring simultaneously are not only difficult to understand, but also challenging to manage (Smol 2010). Yet, understanding the effects of multiple stressors on freshwaters, particularly across ecologically-relevant time scales, is critical in supporting environmental management and stewardship.
1.2 Shallow lakes as sentinels of change in the Athabasca Oil Sands Region

Lakes are important sentinels, integrators, and regulators of environmental change (Williamson et al. 2009). Given their low-lying landscape position and relatively long water residence times, lakes archive and integrate physical, chemical, and biological information from the airshed and catchment (Adrian et al. 2009; Schindler 2009). Measurable indicators of change are recorded in lakes and provide direct and indirect information about lake and catchment responses to environmental stressors (Adrian et al. 2009; Williamson et al. 2009).

Shallow lakes, which are numerous, especially across temperate and high latitude regions (Bennion et al. 2010), can be particularly useful sentinels of change (Smol 2016). The relatively small volumes and high surface area-to-depth ratios of shallow systems may amplify and accelerate the responses to some environmental stressors (Smol and Douglas 2007a; Smol 2016). For example, small lakes with a low heat capacity will likely respond more rapidly to changes in solar irradiance (Gerten and Adrian 2001). Conversely, the small areas, depths, and volumes, and the related mechanisms, including ecological resistance to threshold-type responses (Smol 2016) and alternate states (Scheffer 1998), may dampen the consequences of environmental stressors in shallow lakes until ecological thresholds are surpassed (Smol 2016).

The Athabasca Oil Sands Region (AOSR; ~93,000 km², approximately centred around 57°01’12.0” N, 111°39’00.0” W) is located in the northeast corner of Alberta (Canada’s Boreal Plains ecozone), near the city of Fort McMurray (Figure 1.1). The area features extensive boreal forest, muskeg peatland, jack pine- and trembling aspen-dominated uplands, and a multitude of shallow, small lakes (Natural Regions Committee 2006). These lakes, which lie atop glaciofluvial till, bituminous sands, and limestone and dolomite bedrock (Canadian Society of Petroleum Geologists 1973), are sentinels and integrators of the multiple stressors in the region.
1.3 Climate change in the Athabasca Oil Sands Region

Currently, the AOSR is primarily stressed by climate change and industrial activities. The region’s continental climate is seasonally variable with winters that are long and cold, and growing seasons that are mild and wet. The Athabasca River Basin, in which the majority of the AOSR lies, has a mean annual temperature of 2 °C (Burn et al. 2004). Annual precipitation averages 400-500 mm (Leong and Donner 2015), with up to 75 % of the precipitation falling as rain from June to October (Burn et al. 2004; Kerkhoven and Gan 2011). Meteorological observations from Environment and Climate Change Canada’s (ECCC) Fort McMurray weather station (ECCC station ID: 3062696; 56°36’00.0” N, 111°13’12.0” W; www.ec.gc.ca/dccha-ahccd), the most representative ECCC station for the AOSR, show significantly rising annual temperatures since the beginning of the record in 1916 (mean of -1.1 °C between 1916 and 1940, mean of 1.3 °C from 1987 to 2011; Laird et al., 2013). Temperatures have increased ~1.65 °C since 1960 (Kurek et al. 2013a), and mean annual temperatures have mostly exceeded the record-long average since the 1970s (Laird et al. 2013). Observations of precipitation (ECCC station ID: 3062693; 56°36’00.0” N, 111°13’12.0” W; www.ec.gc.ca/dccha-ahccd) demonstrate periods of distinct below-average precipitation in the 1940s, 1950s, and 1990s (Laird et al. 2013). Further, the AOSR’s overall moisture has declined with regional warming and the associated effects on evaporation, evapotranspiration, and winter snowpack (Schindler and Donahue 2006). Despite increased contributions from glacial melting, summer flows in the lower reaches of the Athabasca River have decreased ~30 % since 1970 (Schindler and Donahue 2006). These trends are consistent with regional observations from Canada’s western prairie provinces (including the Canadian prairies and boreal transition zones) that identify increases in mean annual temperature (1-4 °C), declines in total annual precipitation (up to 24 % over the past ~100 years), and reductions in river flows (summer flows 20-84% lower than in the early 20th century; Schindler and Donahue 2006).
Models forecast intensification of the recent changes recorded in the AOSR’s climate. The 2013 projections for Northwest Canada from the Intergovernmental Panel on Climate Change (IPCC) predict a 2.7 °C increase in mean annual air temperature by the middle of this century and a 3.5 °C increase by the century’s end (Christensen et al. 2013). Annual precipitation is expected to increase 10 % by mid-century and 14 % by the century’s end (Christensen et al. 2013); however, the projected increases are not forecast to counteract the anticipated increases in evaporation (Schindler and Donahue 2006; Kerkhoven and Gan 2011; Leong and Donner 2015). Further, snowpacks in the western prairie provinces that have historically fed high river flows in May and June are expected to be increasingly diminished by winter rain, periodic melts, and an earlier spring freshet, thereby providing reduced water supply to the landscape (Zhang et al. 2001; Lapp et al. 2005). In summary, the western prairie provinces of Canada, in which the AOSR is situated, are likely to be warmer and drier in the coming decades (Schindler and Donahue 2006; Leong and Donner 2015).

1.4 Effects of climate change on lakes

Climate is a major abiotic driver of lake ecosystem structure and function. Shallow lakes are especially vulnerable to the effects of climate change given their small volumes and large surface area-to-depth ratios (Smol and Douglas 2007a). Although responses to climate change may be indirect and/or complex (Adrian et al. 2009), long-term monitoring records of physical (e.g., O’ Reilly et al., 2015), chemical (e.g., Keller et al., 2008), and biological (e.g., Winder et al., 2009) indicators, as well as paleolimnological records (e.g., Rühland et al., 2003, 2015), have identified changes in lakes due to climate warming. For example, increases in air temperature can alter surface water temperatures (De Stasio et al. 1996; Keller 2007; Adrian et al. 2009) and ice phenology (Magnuson et al. 2000; Latifovic and Pouliot 2007). These changes can affect the depth of light penetration and UV exposure (Pienitz and Vincent 2000), and thermal structure of a lake, for example (Schindler et al. 1996; Adrian et al. 2009). Warmer temperatures causing
increased evaporation-to-precipitation ratios have been shown to affect water levels and specific conductivity in isolated northern lakes (Smol and Douglas 2007a). These physical changes can yield alterations in chemical functioning. For example, in extreme cases, enhanced thermal stratification can cause hypolimnetic hypoxia or anoxia and subsequent changes in internal nutrient dynamics (Wilhelm and Adrian 2008). Chemical conditions more directly attributable to climate changes include, for example, reduced inputs of dissolved organic carbon (DOC), base cations, and nutrients from a catchment due to declines in precipitation (Schindler et al. 1996). Changes in DOC inputs can affect the physical structure of a lake by altering the photic zone and thermocline depth (Schindler et al. 1996). Additionally, physical changes, such as the aforementioned increases in temperature and decreases in ice cover, can alter lake seasonality and extend growing seasons, increase production, alter biological assemblage composition, and facilitate development of more complex habitats, especially in Arctic and subarctic lakes (Rühland et al. 2015).

1.5 Bitumen extraction and processing in the Athabasca Oil Sands Region

In addition to climate change, the AOSR is exposed to intense industrial activity, founded on its extensive deposits of bituminous sands. The Athabasca Oil Sands are one of three major hydrocarbon deposits in northern Alberta and Saskatchewan (Canada). Combined, they underlie ~142,000 km² and are estimated to contain almost 2 trillion barrels of in-place bitumen, ~170 billion barrels of which are recoverable with current technologies (Alberta Energy Regulator 2015). These hydrocarbon deposits were mostly formed from marine plankton and other organic material weathered in ancient sand nearly 144 million years ago (Alberta Energy Regulator 2015). They represent the world’s third-largest proven hydrocarbon deposits, after reserves in Saudi Arabia and Venezuela (Alberta Energy Regulator 2015). The Athabasca reserve is the largest of Alberta’s oil deposits, accounting for ~66 % of Alberta’s total oil sands area and ~80 % of oil sands production in 2014 (Alberta Energy Regulator 2015).
As well as being the largest, the Athabasca deposit is the only reserve in Alberta with extractable bitumen deposits less than 70 m below the surface, making it the only suitable deposit for surface mining. All other extraction is completed via *in-situ* methods, involving pressurized steam injection to heat and mobilize bitumen to the surface (Jiang et al. 2010). Once the oil sands are extracted, the bitumen is isolated and upgraded to marketable oil, and waste products are stored on-site (Zhang et al. 2016). The bituminous sands in the Athabasca region have been commercially exploited since the development of the first bitumen upgrader in the late 1960s (Government of Alberta 2014). By 1980, extraction and upgrading operations exceeded production of 100,000 barrels per day (Alberta Energy Regulator 2015). Today’s production rates are ~1.76 million barrels per day and are forecast to increase to ~3.16 million barrels per day by 2030 (Alberta Energy Regulator 2015; Canadian Association of Petroleum Producers 2015a). Future growth is expected to stem from ongoing expansion of *in-situ* recovery methods, which began outpacing surface mining recovery in 2012 (Korosi et al. 2016).

Environmental concerns accompany the region’s extensive industrial expansion. Development of industrial facilities and supporting infrastructure, and physical land disturbance from mining and exploration, result in habitat fragmentation and the loss of essential ecosystem services. For example, ~99 % of Alberta’s ~4,800 km² mineable footprint (excluding supporting infrastructure) is leased (Government of Alberta 2013) and ~904 km² were developed (realized active mining footprint) by 2015 (Canadian Association of Petroleum Producers 2015b). As of 2015, the total surface area of tailings ponds and associated structures was 220 km² (Canadian Association of Petroleum Producers 2015b). Despite mandated land restoration commitments, ~295 km² of peatlands will be permanently lost (reclaimed to upland forest and tailings storage lakes), and similar landscape changes from approved mines are estimated to markedly reduce carbon sequestration potential (Rooney et al. 2012). Surface mining activities generate large volumes of tailings and require massive amounts of freshwater, often removed from the...
Athabasca River (Sauchyn et al. 2015). In-situ activities can also cause ground deformation (Stancliffe and van der Kooij 2001) and bitumen flow-to-surface incidents have been observed (Korosi et al. 2016).

In addition to physical disturbances caused by local oil sands development, industry-related contamination of and through the AOSR’s surface waters (Kelly et al. 2009, 2010), groundwater (Frank et al. 2014), and atmosphere (Kurek et al. 2013a; Kirk et al. 2014; Manzano et al. 2017) has been identified and is an ongoing environmental concern. Atmospheric pathways of contamination can be far-reaching and can affect ecosystems, including lakes and their catchments, physically undisturbed by industry. Although one study (using Sphagnum moss as a proxy; Shotyk et al., 2014) found vanadium, a metal enriched in bitumen, as the only elevated trace metal in the AOSR environment, other studies exploring atmospheric contamination in the AOSR find evidence of elevated pollutants across the landscape. Studies find elevated levels of crustal and anthropogenic elements (including priority pollutant elements; Guéguen et al., 2016; Kelly et al., 2010; Kirk et al., 2014; Landis et al., 2012), acidifying agents (inorganic sulphur and nitrogen compounds; Simpson et al., 2010; Watmough et al., 2014), volatile organic compounds (Simpson et al. 2010), secondary organic aerosols (Liggio et al. 2016), nutrients (Kirk et al. 2014; Fenn et al. 2015), heterocyclic aromatics (Manzano et al. 2017), and polycyclic aromatic compounds (PACs; Kelly et al., 2009; Schuster et al., 2016; Zhang et al., 2016) surrounding the local oil sands developments, indicating industrial sources of these contaminants. Industrial sources of aerial contamination in the AOSR include upgrader stack emissions, diesel exhaust, volatization from tailings ponds (although this pathway is not fully understood), and dust from surface mines, mine reclamation sites, petcoke (a carbonaceous residual from the upgrading of crude oil) stores, haul roads, overburden components, and exposed bitumen (Landis et al. 2012; Jautzy et al. 2013; Parajulee and Wania 2014; Zhang et al. 2016; Manzano et al. 2017).
PACs are particularly well-studied industrial contaminants due to their recognition as priority pollutants by the US Environmental Protection Agency (2012). In addition to naturally occurring in all hydrocarbon deposits, volcanic eruptions, and wild fires, PACs are also emitted from the combustion of fossil fuels (Menzie et al. 1992). They are known mutagens and carcinogens (World Health Organization 2010), and are classified as toxic substances under Schedule 1 of the Canadian Environmental Protection Act (1999). Groups of PACs, namely C1-C4-alkylated PACs and dibenzothiophenes (DBTs), are relatively enriched in bitumen and generally accepted as suitable proxies for airborne industrial oil sands contamination (Kurek et al. 2013a; Schuster et al. 2016). They are measured in ongoing AOSR monitoring programs (e.g., Canada-Alberta Joint Oil Sands Monitoring Program; jointoilsandsmonitoring.ca). These petrogenic PACs (C1-C4-alkylated PACs and DBTs) are considered suitable industrial proxies because: 1) levels increase in regional lake (Jautzy et al. 2013; Kurek et al. 2013a) and peat (Zhang et al. 2016) cores concomitant with industrial AOSR development; 2) stable isotopic carbon signatures of these PACs in lake sediment cores shift to match the signatures of unprocessed AOSR bitumen at the pace and scale of local industrial activities (Jautzy et al. 2013); 3) deposition decreases with increasing distance from industrial sites (Kelly et al. 2009; Schuster et al. 2016; Zhang et al. 2016); and 4) levels were not elevated during a known forest fire event, while levels of other PACs known to originate from natural combustion were elevated (Schuster et al. 2016). Collectively, this evidence supports an association of C1-C4-alkylated PACs and dibenzothiophenes (DBTs) with industrial sources from the AOSR rather than a more distant signature of incomplete combustion processes.

1.6 Effects of the bitumen-based industry on lakes

Deposition of industrial contaminants can impact lakes directly and through complex mechanisms. Chemical contamination from industry, including deposition of metals and acid precursors, for example, can alter lake dynamics and exert toxic effects on organisms, which can
change how a lake functions and the structure of the aquatic community within the lake (e.g., Labaj et al., 2015; Thienpont et al., 2016). There are many North American and European examples of lake ecosystem reorganization in response to acid deposition (e.g., Cumming et al., 1994). However, there is little evidence of surface water acidification across the AOSR and farther downwind (Hazewinkel et al. 2008; Curtis et al. 2010; Laird et al. 2013). The limited response is likely due to a buffering effect provided by elevated deposition of base cations (Watmough et al. 2014).

Despite limited acidification of AOSR waters, the potential for community reorganization and/or toxic effects from industrial contamination persists, but is difficult to characterize fully given the occurrence of multiple stressors in the AOSR. Toxicological lab studies of oil sands related compounds, including PACs, identify significant negative effects on aquatic life, including greater mortality and malformations in juvenile fish (Colavecchia et al. 2006, 2007). However, the bioavailability and toxicity of these contaminants are likely modified in the natural environment by other contaminants (e.g., naphthenic acids, hydrocarbon degradation products, and metals) and the variability of physiochemical factors (e.g., water flow, volatization, evaporation, and ultraviolet radiation; Colavecchia et al., 2006). For example, PAC toxicity to fish increases with exposure to ultraviolet radiation via increased production of reactive oxygen species and lipid peroxidation (Choi and Oris 2000).

Although the relative roles of nitrogen and phosphorus in regulating primary production in lakes are actively debated (Lewis and Wurtsbaugh 2008; Schindler et al. 2016), inputs of these nutrients may stimulate elevated primary production in some lakes and potentially yield pronounced ecological community changes. Investigation of the biological response to reactive nitrogen deposition in lakes up to ~300 km downwind of the AOSR’s main industrial facilities found no demonstrable link to deposition of reactive nitrogen and changes in diatom and
chrysophyte assemblages (Laird et al. 2017; Mushet et al. 2017). The responses of lakes closer to the bitumen industry have not been fully explored.

1.7 Paleolimnology and its recent applications in the Athabasca Oil Sands Region

Changes in lake biota can integrate and track the effects of multiple stressors on lakes and the broader environment over long and ecologically-relevant time scales (Williamson et al. 2009). Assessments of past environmental conditions can be completed using historical measurements, space-for-time substitutions, hindcasting models, and/or paleoenvironmental reconstructions (Smol 2010). Given that monitoring datasets are often limited to no longer than a few years (Smol 2008), and the data required to track long-term environmental change in lakes are commonly lacking, a paleolimnological approach to tracking historical environmental conditions and disentangling the roles of multiple stressors is often instructive. In the AOSR, long-term environmental monitoring of aquatic systems only began in 1997, ~30 years after the establishment of industrial oil sands operations (Dowdeswell et al. 2010). A 2010 report by a formal advisory panel to the federal Minister of the Environment determined the monitoring programs in existence were ineffective in distinguishing industrial oil sands impacts because they were lacking rigorous sampling program designs, hypothesis-driven sampling regimes, adequate analytical capabilities, and sufficient understanding of baseline conditions (Dowdeswell et al. 2010). The report recommended paleolimnological analyses of the AOSR’s many lakes and ponds as an effective and timely course of action for characterizing baseline conditions and post-impact trajectories of change in the region (Dowdeswell et al. 2010).

Paleolimnology uses physical, chemical, and biological information archived in lake sediments to reconstruct past environments on timescales that often capture pre-disturbance conditions and extend prior to long-term monitoring (Smol 2008). Generally, the deepest point of a lake basin accumulates, integrates, and preserves proxies from many regions of the water body in a relatively undisturbed stratigraphic sequence, thereby providing a record of past conditions.
and processes in the lake and the catchment (Smol 2008). However, the plethora of interacting stressors often present in a modern environment and the overriding effects of climate change are challenging to disentangle (Smol 2010). Paleolimnological approaches that examine sites along gradients of interest, compare inferences from sediment records to historical data, and/or couple sediment records with experiments providing mechanistic information are especially useful in understanding the effects of multiple stressors on a system (Leavitt et al. 2009; Williamson et al. 2009). The relative abundance data used in paleolimnological studies generally has high reproducibility and effectively records changes in past environments (Smol 2008).

Paleolimnological studies on lakes in the broader AOSR have reconstructed historical deposition of airborne contaminants (Hall et al. 2012; Wiklund et al. 2012; Jautzy et al. 2013; Kurek et al. 2013a), assessed the extent of lake acidification (Hazewinkel et al. 2008; Curtis et al. 2010; Laird et al. 2013), identified potentially high levels of primary production in the region (Hazewinkel et al. 2008; Curtis et al. 2010; Kurek et al. 2013a; Laird et al. 2013), and explored potential causes of increased primary production and community changes in lakes within ~300 km of Fort McMurray (Laird et al. 2017; Mushet et al. 2017).

1.8 Paleoenviromental indicators

Diatoms (Bacillariophyceae) are microscopic algae that are diverse and abundant in almost all freshwater systems. Their subfossil remains are the most commonly used biological proxy in paleolimnological studies (Julius and Theriot 2010; Smol and Stoermer 2010), and are the primary indicator used in this thesis. There are currently > 24,000 officially recognized diatom taxa, which are distinguished by the morphology of their siliceous cell walls (frustules; Julius and Theriot, 2010). The durable and taxonomically-distinct frustules, quick turnover of generations, ability to rapidly and preferentially colonize new habitats, and relatively well-understood optima and tolerances of specific environmental variables (e.g., pH, salinity, and nutrient levels) allow the calibration of diatom taxa to environmental conditions and their use in
reconstructing and interpreting past environments (Smol 2008; Smol and Stoermer 2010). To date, diatom-based paleolimnological studies from the AOSR have focused on assessing the effects of acidification (Hazewinkel et al. 2008; Curtis et al. 2010; Laird et al. 2013) and nitrogen deposition (Laird et al. 2017; Mushet et al. 2017) from industry.

Groups of aquatic biota often respond differently to environmental stressors (e.g., Rühland et al., 2014) and, as such, assessments of multiple proxies typically provide a more holistic and improved understanding of past environmental change (Birks and Birks 2006). Other paleoindicators that are used in this thesis include chironomids (Chironomidae), cladocerans (Cladocera), and spectrally-inferred sedimentary chlorophyll-a (VRS-chla). Like diatoms, chironomids (Walker 2001) and cladocerans (Jeppesen et al. 2011) are useful environmental proxies because of their rapid reproduction rates, their diverse assemblages, and their specific optima and tolerances to environmental variables. Chironomids (Walker 2001; Smol 2008) and cladocerans (Korhola and Rautio 2001) are useful indicators of past ecological changes in lakes and catchments because components of their chitinous exoskeletons persist in the lake sediments and can be identified to the genus and often species level. Chironomids are established proxies of, for example, climatic changes and deep-water oxygen concentrations (Walker 2001; Smol 2008), while Cladocera are useful in inferring, for example, historical acidification, metal contamination, salinization, changes in water levels, and invasive species populations (Smol 2008). Sedimentary concentrations of chlorophyll-a are a biochemical proxy for whole lake primary production (Wolfe et al. 2006; Michelutti and Smol 2016). Given that the spectral measures incorporate the diagenetic products of chlorophyll-a in the sediments, the spectrally-derived inferences of the pigment can be used in paleo-reconstructions of nutrients and primary production (Wolfe et al. 2006; Michelutti and Smol 2016).
1.9 Thesis objectives

The main objective of this thesis is to assess the long-term effects of two major stressors, climate change and bitumen-based industrial development, on the biota of lake ecosystems in the AOSR. These stressors are expected to persist and intensify with time; thus, investigation of their cumulative impacts is important and timely. There are five chapters in this thesis. This outline of the objectives concludes the first, introductory chapter. The second, third, and fourth chapters are data chapters. The fifth and final chapter is a general discussion with conclusions and a synopsis of future directions.

The primary aim of the second chapter is to assess the biological community change over time and across multiple trophic levels in one shallow lake exposed to climate change and industrial oil sands contamination over recent decades. This study uses subfossil remains of multiple bioindicators, including primary producers, consumers, and detritivores, concentrations of petrogenic contaminants, and spectrally-inferred chlorophyll-α concentrations contained in the past ~75 years of deposited lake sediments. These proxies are used to infer historical conditions and examine the roles of climate change and industry in structuring aquatic communities. Together, the relatively detailed temporal resolution of this study and the use of multiple bioindicators enables a holistic investigation and, to my knowledge, the most comprehensive understanding of the biological response in a typical AOSR lake to the modern environmental stressors in the region.

Chapter three explores temporal changes in inferred aquatic primary production across the AOSR and the relative contributions of climate change and industrial oil sands development to the primary production trends. Although not explicitly exploring aquatic primary production, recent paleolimnological studies on lakes up to ~350 km away from the main area of AOSR industrial development found evidence of increased production over recent decades (Hazewinkel et al. 2008; Curtis et al. 2010; Kurek et al. 2013a; Laird et al. 2013). While the spatial extent of
the inferred increases in aquatic primary production suggests an important regional driver, such as climate change, marked deposition of nutrients within ~50 km of the main industrial upgrading facilities (Kirk et al. 2014) underscores industrial activity within the region as a potentially important driver. This chapter uses spectrally-inferred chlorophyll-a profiles from 23 lakes across the AOSR to characterize the spatial pattern and timing of primary production increases. The temporal and spatial trends are then compared to modern nutrient deposition maps and historical records of climate metrics to elucidate the regional driver(s) of aquatic primary production. This research provides important information for the effective management of the regional stressors and the stewardship of the region’s freshwaters.

Chapter four is a “top-bottom” paleolimnological study that compares subfossil diatom assemblages from the surface sediments (representing recent time periods) to assemblages in sediments deposited prior to industrial oil sands development and recorded climate change in the region. Top-bottom studies compare two discrete time intervals (one before and one after a disturbance) to efficiently investigate regional change in lakes over time (Smol 2008). Eighteen lakes are included in the study. The main objective of the chapter is to characterize how modern diatom assemblages differ from pre-disturbance assemblages and identify what environmental variables (including aerial deposition of industrial contaminants and nutrients) explain the patterns of diatom assemblage change and distribution among AOSR shallow lakes. This study examines the regional response of important primary producers (diatoms) to climate change and airborne industrial contamination, and demonstrates the value of a top-bottom approach as a useful tool for detecting and managing at-risk lakes.

Overall, this thesis explores and interprets the biological responses of AOSR aquatic communities to climate change and intense industrial activity. As such, understanding of the stressors’ relative effects on lakes, assessments of potential vulnerabilities, and the foundation for effective management are improved. Given the ubiquity of climate change and the relative
prevalence of the fossil fuel industry in other regions, lessons learned from this project are likely useful beyond the geographic scope of the AOSR. Further, our findings contribute insights into the biological functioning and responses of shallow lakes, in particular, to long-term environmental change and the cumulative effects of multiple stressors. Finally, this work demonstrates the value of paleolimnological methods, including multi-proxy and top-bottom approaches, for tracking ecosystem changes in the absence of long-term monitoring data and providing a critical basis for effective environmental management.
1.10 References


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**Figure 1.1.** The location of the Athabasca Oil Sands Region
Chapter 2

Assessment of multi-trophic changes in a shallow boreal lake simultaneously exposed to climate change and aerial deposition of contaminants from the Athabasca Oil Sands Region, Canada

2.1 Abstract

The Athabasca Oil Sands Region (AOSR) has been intensively developed for industrial bitumen extraction and upgrading since the 1980s. A paucity of environmental monitoring prior to development raises questions about baseline conditions in freshwater systems in the region and ecological responses to industrial activities. Further, climatic changes prompt questions about the relative roles of climate and industry in shaping aquatic ecosystems through time. We use aquatic bioindicators from multiple trophic levels, concentrations of petrogenic contaminants (dibenzothiophenes), and spectrally-inferred chlorophyll-\(\alpha\) preserved in well-dated sediments of a closed-basin, shallow lake ~50 km away from the main area of industry, in conjunction with climate observations, to assess how the biotic assemblages of a typical AOSR lake have changed during the past ~75 years. We examine the contributions of the area’s stressors in structuring aquatic communities. Increases in sedimentary measures of petrogenic contaminants provide clear evidence of aerial contaminant deposition from local industry since its establishment, while climate records demonstrate consistent warming and a recent period of reduced precipitation. Quantitative comparisons of biological assemblages from before and after the establishment of regional industry show significant (\(p < 0.05\)) differences; however, the magnitude and overall timing of the changes are not consistent with a threshold-type shift in response to the onset of regional industry. Rather, biotic assemblages from multiple trophic levels suggest transitions to an increasingly complex benthic environment and relatively warmer waters, which, like the increasing trends in inferred primary production, are consistent with a changing climate. These
findings highlight the important role of climate conditions in regulating primary production and structuring aquatic communities in these shallow systems.

2.2 Introduction

The Athabasca Oil Sands Region (AOSR) overlies massive deposits of bitumen, which have been commercially exploited since the development of the first upgrader in the region in the late-1960s (Government of Alberta 2014). By 1980, bitumen extraction and upgrading operations yielded production levels exceeding 100,000 barrels per day (Alberta Energy Regulator 2015), marking the establishment of the region’s oil sands industry. Today, Athabasca bitumen production rates are ~1.76 million barrels per day and are forecast to increase to ~3.16 million barrels per day by 2030 (Alberta Energy Regulator 2015; Canadian Association of Petroleum Producers 2015a). This intense development since ~1980, combined with forecasts of expanded production, raises environmental concerns among stakeholders, including industry, regulators, First Nations communities, and the public.

Industry-related environmental threats to aquatic ecosystems include widespread aerial deposition of contaminants (Kelly et al. 2009, 2010; Kirk et al. 2014), deposition of potentially growth-stimulating nutrients (Summers et al. 2016), and land disturbance from mining activities and related industrial development (e.g., seismic lines, roads, work sites, work camps, and aircraft facilities; Rooney et al., 2012; Timoney and Lee, 2009). Although toxicological lab studies have identified negative effects on aquatic life, such as greater mortality and malformations in fish associated with AOSR bitumen-contaminated sediments (Colavecchia et al. 2006, 2007), the ultimate ecological effects of industrial oil sands contaminant deposition in the region’s lakes are not well understood.

Coincident with AOSR development, the climate of northern Alberta has become warmer and drier since the beginning of the available meteorological record in the early 20th century, and models forecast intensification of these trends (Schindler and Donahue, 2006). For example,
temperatures and precipitation in northwestern Canada are expected to rise throughout the century (Christensen et al. 2013), while the ongoing declines in snowpack, river flows, and glaciers, due to elevated temperatures and rates of evaporation, are expected to reduce the region’s overall moisture (Schindler and Donahue 2006; Kerkhoven and Gan 2011; Leong and Donner 2015).

The long-term monitoring data required to more fully understand the effects of the multiple stressors in the AOSR are largely absent because strategic monitoring programs in the region were established decades after the beginning of industrial activities (Dowdeswell et al. 2010). Fortunately, paleoenvironmental studies can be used to extend and augment monitoring records, thereby establishing baseline environmental conditions and providing insights into aquatic responses to industrial activities and regional climate change (Smol 2008; Williamson et al. 2009). Some paleolimnological studies have explored the impacts of these multiple stressors on lakes in the AOSR and have demonstrated that aquatic ecosystems in the region have recently shifted to new ecological states, with linkages to both climate change (Hazewinkel et al. 2008; Curtis et al. 2010; Kurek et al. 2013a, b; Laird et al. 2013; Summers et al. 2016) and the effects of industrial oil sands activities (Kurek et al. 2013a). However, questions still remain regarding baseline environmental conditions, the timing and nature of ecological transitions, and the relative effects of climate change and industry across the region’s freshwaters.

As is common in multiple-stressor systems, disentangling the roles of individual stressors is challenging (Smol 2010). Here, we use bioindicators across a food web (i.e., primary producers, consumers, and detritivores), measures of industrial contaminants, and proxies of aquatic primary production from a strategically selected AOSR lake, in conjunction with historical climate observations, to assess changes in the biotic assemblages of an industrially contaminated, typical AOSR lake over the past ~75 years. We investigate the roles of regional industry and climate change in structuring aquatic communities. Our paleolimnological approach, spanning multiple trophic levels, provides a holistic assessment of aquatic ecosystem response to
oil sands activities overlapping with climate change, thereby establishing the trajectory of an aquatic ecosystem relative to baseline conditions.

2.3 Study area

2.3.1 The Athabasca Oil Sands Region

Alberta’s AOSR is located in the northeast corner of this western Canadian province (Figure 2.1). The landscape is dominated by boreal forest, wetlands, freshwater lakes and rivers, and is underlain by a bitumen-rich geology. The continental climate is seasonally variable with long, cold winters (2014 mean winter air temperature = -19.7 °C) and short, warm summers (2014 mean summer air temperature = 17.2 °C; Environment and Climate Change Canada’s Fort McMurray temperature station, ID: 3062696, www.ec.gc.ca/dccha-ahccd). Most precipitation falls between June and October (Kerkhoven and Gan 2011). Decades of industrial activities, including surface mining, in-situ bitumen recovery, and the development of related infrastructure, have industrialized this once-remote boreal landscape. For example, ~99 % of the 470,000 ha covering mineable oil sands deposits are leased (Government of Alberta 2013) and even after considering industry’s land restoration commitments, ~29,555 ha of peatlands will be lost, resulting in an estimated reduction in related carbon sequestration potential by ~5,700-7,200 tons C/year (Rooney et al. 2012).

2.3.2 Study lake

The Regional Aquatics Monitoring Program’s (RAMP) site 209 (hereafter RAMP 209; 57°13’ N, 110°44’ W) is a small (surface area = 10.9 ha, catchment = 82 ha), shallow (Z_{max} = 1.2 m), closed-basin lake, typical of the AOSR (Natural Regions Committee 2006). It is located ~50 km northeast of the region’s main area of oil sands industry (measured from AR6, which is a snow sampling site adjacent to two major upgraders commonly referenced in the scientific oil sands literature), and ~5 km east of nearby in-situ operations (Figure 2.1), which began
production in 2003 (Suncor Energy 2006). Importantly, the lake’s catchment is relatively undisturbed by land use, and ice cover in winter does not extend to the bottom of the lake. Based on measurements from March 2013, which were taken through ice cover, RAMP 209 is slightly acidic (pH ~6.3), eutrophic (unfiltered total phosphorus = ~37 µg/L), anoxic at least in winter (concentration of dissolved oxygen under 75 cm of ice < 0.1 mg/L), and has high concentrations of dissolved organic carbon (DOC; 51.7 mg/L). End-of-summer water chemistry measures collected annually by RAMP beginning in 2002 generally support the water quality measures we collected, indicating that RAMP 209 is slightly acidic, meso-eutrophic to eutrophic, and has a high concentration of DOC (http://www.ramp-alberta.org/data/AcidSensitiveLakes/AcidSensitiveLakeQuery.aspx).

RAMP 209’s closed-basin hydrology and relatively undisturbed catchment enable assessment of changes due to aerial pollution inputs, while its limnological characteristics that are similar to other shallow lakes in the region allow comparisons with other sites. RAMP 209 was strategically selected for this detailed downcore, multi-proxy study because its relatively high dibenzothiophene (DBT) enrichment factor (calculated from sediment core data as (mean [DBT] 2000-year of coring) / (mean [DBT] 1955-1970); Summers et al. 2016) indicates that the lake has received substantial inputs of airborne pollution from the region’s industrial oil sands development. DBTs are a group of petrogenic, sulphur-containing, alkylated polycyclic aromatic compounds (PACs) that were recognized as a suitable proxy of the aerial inputs from the region’s industrial oil sands activities (Jautzy et al. 2013; Kurek et al. 2013a, b). Although the average concentration of sedimentary DBTs in post-2000 RAMP 209 sediments is relatively low compared to other nearby lakes (162 ng/g dry weight; Summers et al. 2016), the large DBT enrichment factor of 6.2 reflects the change in DBT concentration since the earliest large-scale industrial oil sands operation. Summers et al. (2016) also identified a substantial increase in aquatic primary production at RAMP 209 starting in the mid-1980s and accelerating in the 2000s.
These data indicate that RAMP 209 is a suitable site to examine the potential ecological response to decades of industrial airborne pollution as well as climate change.

2.4 Methods

2.4.1 Sediment core collection

Samples were collected as part of the Canada-Alberta Joint Oil Sands Monitoring (JOSM) program (http://jointoilsandsmonitoring.ca), and sampling methodology has been previously reported in Summers et al. (2016). Three sediment cores were collected through the ice in March 2013 at the deepest part of RAMP 209 using a gravitational Uwitec coring system (www.uwitec.at). Cores were transported to the field base and a Uwitec vertical extruder was used to section the cores at 0.5-cm intervals for the upper 20 cm and 1-cm intervals for the remainder of the core. Sediments were frozen in Whirlpak® bags and polypropylene containers, and transported to the National Laboratory for Environmental Testing (NLET; Burlington, Ontario) for further processing. Sediments from one primary core (length = 40 cm) were transported to Flett Research Ltd. (Winnipeg, Manitoba) for radiometric dating, AXYS Analytical Services (Sidney, British Columbia) for PAC analyses, and the Paleoecological Environmental Assessment and Research Laboratory (PEARL) at Queen’s University (Kingston, Ontario) for sedimentary chlorophyll-α and bioindicator analyses.

2.4.2 Radiometric dating of sediments

As reported in Summers et al. (2016), dating of the full, primary sediment core (40 cm) was completed by Flett Research Ltd. using alpha spectroscopy, which measures the alpha radiation (±1 SD) of 210Po as a proxy for total 210Pb. Sixteen sediment intervals were measured for 210Po, and alpha radiation of 226Ra was also measured at 3 intervals (9.5-10 cm, 21-22 cm, and 33-34 cm) and used as a proxy for background (supported) 210Pb levels. The chronology of the measured sediment intervals was calculated using the Constant Rate of Supply (CRS) model.
(Appleby 2001). Ages for the remaining sediment intervals were estimated from a second-order polynomial curve with an intercept set to the time of coring (Summers et al. 2016).

2.4.3 Bioindicator processing and analyses

Diatoms, chironomids, and cladocerans were analyzed from the same 21 sediment intervals ranging from 0-21 cm and spanning the past ~75 years (~1938 to date of coring in 2013). The majority of the sediment intervals used for bioindicator analyses were subsampled at 0.5-cm thickness. Only the bottom-most 20-21-cm interval was subsampled at 1.0-cm thickness. To isolate diatom remains, ~0.02-0.03 g of freeze-dried sediment was digested in a 50:50 molar ratio of concentrated nitric acid (HNO₃) and sulphuric acid (H₂SO₄) and heated to 80 °C in a hot water bath for 2 hours. The digested samples were allowed to settle for 24 hours, the supernatant was removed, and the samples were rinsed with deionized water. The rinsing procedure was repeated until a neutral pH was attained. Diatom slurries were mixed and pipetted onto coverslips in four to eight dilutions, air-dried, and mounted onto microscope slides using Naphrax®. A minimum of 300 diatom valves was counted for every processed sediment interval (minimum = 302, maximum = 639, mean = 390 valves) using a Leica DMR microscope outfitted with differential interference contrast optics at 1000× magnification under oil immersion. Diatom valves were identified using taxonomic guides including Fallu et al. (2000) and Krammer and Lange-Bertalot (1986-1991).

To isolate chironomid remains, ~0.05 g of freeze-dried sediment was treated with a 5 % potassium hydroxide (KOH) solution. The samples were warmed to ~80 °C for 20 min, sieved through a 100-µm sieve, and rinsed with deionized water. The samples were rinsed into a beaker and aliquots were poured into a Bogorov tray and viewed with a dissecting microscope at 30× magnification. All chironomid head capsules were handpicked with fine forceps and distributed evenly across coverslips. Coverslips were mounted onto slides using Entellan®. A minimum of 50 chironomid head capsules was targeted from each sample (Quinlan and Smol 2001). Twenty of
the 21 intervals exceeded the targeted minimum count of 50 whole chironomids (minimum = 49, maximum = 115, and mean = 88 individuals). Chironomids were identified at 200 or 400× magnification under bright-field illumination. Taxa were identified to subtribe level or lower (Brooks et al. 2007).

Cladoceran remains were processed from ~0.1 g of freeze-dried sediment using 80 mL of 10 % KOH solution and heating at ~80 °C for 20 min. Samples were rinsed using deionized water and a 38-μm sieve and transferred to a glass vial. Three drops of 95 % ethanol were added as a preservative and three drops of safranin solution were added as a stain. No more than 50-μL aliquots were placed on each slide with glycerin jelly as a mounting medium. Cladoceran remains were identified under 200 or 400× magnification with bright-field illumination. Entire coverslips were scanned and individuals were identified to genus or species level using Korosi and Smol (2012a, b) as taxonomic references and Kurek et al. (2010) as a counting guideline.

2.4.4 Downcore numerical analyses of bioindicator assemblages

R Version 3.2.2 was used for all statistical tests (R Development Core Team 2015). Counts were converted to relative abundances (%) of the total assemblage, and rare chironomid and cladoceran taxa were eliminated using a cut-off criterion of at least 1 % relative abundance in at least 2 intervals. Due to the many diatom taxa present in low abundances, the rare cut-off criterion was increased to at least 2 % relative abundance in at least 1 interval for diatom indicator value (IndVal) analysis (explained below).

The statistical framework used in this study was designed a priori to compare recent environmental conditions with the conditions before the establishment of substantial AOSR oil sands industry ~1980 (Alberta Energy Regulator 2015). To summarize the assemblage data, downcore principal components analyses (PCAs) were completed (“vegan” R package; Oksanen et al., 2012) on square root-transformed relative abundances with rare taxa removed from each bioindicator group. Data were square root transformed to reduce the importance of common taxa.
and increase the importance of taxa with lower abundances (Borcard et al. 2011). PCA axes 1 sample scores were plotted stratigraphically to display the main patterns of variation for each proxy (Birks et al., 2012).

For each of the three bioindicators, an analysis of similarities (ANOSIM) test (Clarke 1993) was used to compare assemblage composition between groups before and after ~1980, which marks the establishment of the regional industry. These time periods encompass approximately similar lengths of time (~40 years pre-industry, ~30 years post-industry), similar numbers of sediment intervals (10 intervals pre-industry, 11 intervals post-industry), and similar ranges of sediment depth (10 cm of sediment pre-industry, 11 cm of sediment post-industry). The ANOSIM test statistic, R, compares the mean pair-wise similarity within the time period to the mean pair-wise similarity between the time periods selected a priori. ANOSIM R values can technically range from -1 to 1, although they are generally between 0 and 1. R values approaching 1 indicate greater differences in assemblage compositions between time periods, while R values ~0 indicate that similarities within and between time periods are the same on average (i.e., little to no differences in assemblage composition between time periods). R value significance (p < 0.05) was determined using 999 random permutations (Clarke 1993). ANOSIM tests were performed using the “vegan” package (Oksanen et al. 2012) in R on square root-transformed relative abundance data with rare taxa removed and Bray-Curtis dissimilarity coefficients (Bray and Curtis 1957).

IndVal analyses (Dufrêne and Legendre 1997), which consider the mean abundance of a taxon within a time period (specificity) and the taxon’s presence in most intervals of that time period (fidelity; Borcard et al., 2011; Dufrêne and Legendre, 1997), were used to identify the taxa that were deemed characteristic of the pre- or the post-1980 time periods for each group of bioindicators. IndVal uses randomizations (n = 999) to calculate the statistical significance of a taxon’s association with the time period. Taxa with p ≤ 0.05 were stratigraphically displayed. R’s
“labdsv” package (Roberts 2016) was used to run the analyses on untransformed relative abundance data with rare taxa removed. As previously stated, the rare criterion for diatoms was increased.

2.4.5 Historical climate observations and downcore trends in primary production and dibenzothiophene concentrations

Climate observations (temperature and precipitation), downcore DBT concentrations, and spectrally-inferred sedimentary chlorophyll-a (VRS-chla) concentrations were compared to the timing of biological assemblage changes to investigate the roles of industrial oil sands activities and climate change in structuring the ecological communities at RAMP 209. Climate data are publically accessible from Environment and Climate Change Canada. The DBT enrichment factor and VRS-chla data were previously published in Summers et al. (2016).

2.4.5.1 Historical climate observations

Adjusted and homogenized mean annual and seasonal temperature data from the beginning of the bioindicator records (~1938) to 2013 were obtained from Environment and Climate Change Canada’s Fort McMurray temperature station (ID: 3062696, www.ec.gc.ca/dccha-ahccd, ~70 km southwest of RAMP 209). Annual and winter temperatures were not available for 2013. Annual, spring, and fall measures were not available for 2012. Annual and spring temperatures were not available for 1942 and 1943. Thus, data for these time periods were absent in the analyses. Adjusted and homogenized total annual and seasonal precipitation data were available from the beginning of the bioindicator records (~1938) to 2006, and were obtained from Environment and Climate Change Canada’s Fort McMurray precipitation station (ID: 3062693, ~70 km southwest of RAMP 209, www.ec.gc.ca/dccha-ahccd). Annual total precipitation measures for 2007 to 2013 were calculated from the Regional Aquatics Monitoring Program’s (RAMP’s) Aurora climate station (C1-RAMP, 57°14’ N, 111°24’W), which is the closest RAMP weather station to Fort McMurray (~55 km northeast) with

Seasonal total precipitation for 2007-2013 was calculated from RAMP’s daily data. To account for missing days of RAMP data, the summed total seasonal precipitation was divided by the number of days with data (total precipitation (mm)/day) and then multiplied by the number of days in the season.

2.4.5.2 Polycyclic aromatic compounds including dibenzothiophenes

DBTs are a group of alkylated PACs, which are ubiquitous in the environment, produced during natural or anthropogenic combustion of fossil fuels and biomass (Menzie et al. 1992; Zhang et al. 2016). PACs have been linked to adverse effects on aquatic invertebrate communities in the AOSR (Gerner et al. 2017). Total DBTs (DBT + C1-C4-alkylated DBTs) are a suitable indication of industrial oil sands impact in the AOSR because they are a natural component of the region’s oil sands deposits and their atmospheric transport is relatively local (Kelly et al. 2009; Kurek et al. 2013a). As reported in Summers et al. (2016) and further detailed in Kurek et al. (2013a), PAC concentrations, including DBT concentrations, were determined from dried sediments using a standardized method for PACs (MLA-021), which is an AXYS method based on the United States Environmental Protection Agency methods 1625B and 8270C/D. The measured PAC record did not extend as far back in time as the bioindicator or VRS-chl\(a\) data because DBT analyses were only completed on sediments between 0 and 16.5 cm; however, the record was considered useful given that it extended prior to industry (~1955) and across more recent times.

2.4.6 Spectrally-inferred sedimentary chlorophyll-\(a\)

Visible reflectance spectroscopy (VRS) was used to estimate past changes in sedimentary chlorophyll-\(a\) (chl\(a\)) concentrations, as well as their main diagenetic products, which represent trends in whole lake primary production (Michelutti et al. 2005, 2010; Wolfe et al. 2006;
Michelutti and Smol 2016). VRS is an indirect measurement method that captures the sediments’ spectral signatures and uses the area under the reflectance trough between 650 and 700 nm. The 650-700-nm trough varies in magnitude with the concentration of chla and the main by-products of chla degradation (Wolfe et al. 2006); thus, the VRS method accounts for chla diagenesis (Michelutti et al. 2010).

To obtain a spectral signature encompassing the entire timeframe included in the bioindicator data, sediments from intervals dating back to ~1933 (21 cm) were freeze-dried, individually sieved through a 125-µm mesh, transferred to a 19 × 65-mm glass vial, and analyzed in a FOSS NIRSystem Model 6500 rapid content analyzer (detection limit ~0.01 mg/g dry weight).

2.5 Results

2.5.1 Chronology

Measured with alpha spectroscopy, total $^{210}\text{Pb}$ (using $^{210}\text{Po}$ proxy) activity exhibited an exponential decay with depth (Figure 2.2). The CRS model was calibrated against a linear regression model and was used to calculate the age estimates of the core. Overall, the radioisotope data and estimated dates are judged reliable for high-resolution paleolimnological sampling. The activity profiles and age-depth model have been previously published in Summers et al. (2016).

2.5.2 Downcore trends in bioindicator assemblages

A total of 155 diatom taxa, 47 chironomid taxa, and 22 cladoceran taxa were identified from the sediment core. After rare taxa were removed, 28 diatom taxa, 34 chironomid taxa, and 21 cladoceran taxa remained.

ANOSIM tests identified that the composition of all bioindicator assemblages significantly differed from pre- to post-1980 (establishment of regional oil sands industry). The
greatest difference identified by ANOSIM occurred for the diatoms, followed by cladocerans and chironomids (diatoms: $R = 0.60, p = 0.001$; chironomids: $R = 0.22, p = 0.004$; cladocerans: $R = 0.39, p = 0.002$). IndVal analyses on pre- and post-1980 time periods identified 13 diatom taxa, 4 chironomid taxa, and 6 cladoceran taxa as significantly ($p < 0.05$) affiliated with one of the pre-defined time periods (pre- or post-1980; Figure 2.3). All of the taxa identified by IndVal with $p \leq 0.05$ are presented in the stratigraphy (Figure 2.3) and were considered in the ecological interpretation. However, some taxa that were only present at very low abundances are not a focus of the discussion.

Stratigraphical display of the relative abundances (Figure 2.3) and downcore PCA axis 1 sample scores (Figure 2.4) showed gradual change in all biological proxies during the ~75-year record. PCA axis 1 sample scores explained 32.6% of the variance for diatoms, 14.8% for the chironomids, and 26.7% for the cladocerans (Figure 2.4). Downcore PCA axis 1 sample scores for all bioindicators showed directional changes in assemblage composition throughout the RAMP 209 sediment record (Figure 2.4).

Many diatom taxa were present at low abundances and/or were only present during specific periods of the record. IndVal identified the *Staurosira construens* complex (which includes *S. construens* var. *pumila*, *S. construens* var. *subsalina*, and *S. construens* var. *venter*, combined due to identification challenges) as characteristic of the pre-1980 time period. This species complex was dominant in the RAMP 209 sediment core, and was the only group that exceeded 20% relative abundance at any point in the diatom record. *Pseudostaurosira brevistriata* and *Staurosirella pinnata* are morphologically similar to taxa in the *S. construens* complex and, although these taxa were not truly dominant, they were present in relatively high abundances throughout the record (up to ~14 and 17%, respectively). Other taxa identified by IndVal as being significantly characteristic of the pre-1980 time period included *Aulacoseira nygaardii*, *Aulacoseira valida*, *Eolimna ütermoehlii*, *Eunotia sudetica*, and *Stauroneis kriegerii*.
which were all present in low relative abundances and did not exceed 9\% throughout the record. *A. nygaardii* and *A. valida* were nearly absent from the record after the late-1980s (Figure 2.3).

Diatom taxa identified by IndVal as associated with the post-1980 time period were low in relative abundances throughout the core (Figure 2.3). *Eolimna minima* and *Sellaphora seminulum* were present throughout the core and increased to relatively moderate abundances (~12\% and ~15\%, respectively) in the post-1980 sediments. *Nupela deiformis, Psammothidium curtissimum, P. subatamoides, N. vitiosa*, and the *Achnanthidium minutissimum* complex (which includes *Achnanthidium minutissimum* var. *affinis, A. minutissimum* var. *macrocephala*, and *A. minutissimum* var. *saprophila*, combined due to identification uncertainties), were only present in low relative abundances (< 5\%; Figure 2.3).

Unlike the diatoms, the chironomid assemblages were not dominated by a single taxon (Figure 2.3). *Dicrotendipes nervosus, Microtendipes pedullus, Paratanytarsus*, and *Tanytarsus mendax* were present throughout the core at relatively stable abundances, averaging ~6\%, 15\%, 5\%, and 7\%, respectively. *Psectrocladius barbatipes* was the sole IndVal-identified chironomid taxon characteristic of the pre-1980 time period. *P. barbatipes* was sparsely dispersed throughout the record in extremely low abundances (not exceeding ~3\%). Post-1980 IndVal-identified taxa included *Cladotanytarsus mancus, Stempellinella/Zavrelia, and Heterotanytarsus*, which were all present in low abundances (< 8\%). *Stempellinella/Zavrelia* and *Heterotanytarsus* were especially low in abundance and were rarely present in the record until approximately the mid-1990s (Figure 2.3).

Cladoceran assemblages were consistently dominated by a single taxon throughout the core (Figure 2.3). *Chydorus brevilabris* was dominant with abundances of ~17-40\%, and was identified as significantly associated with the pre-1980 time period (pre-1980 average abundance of ~32\%). *Alonella excisa* and *Alonella nana* were also identified as taxa characteristic of the pre-1980 assemblages because, although they were present throughout the core, their relative
abundances declined slightly after ~1980 (pre-1980 average abundances of ~5 % and 4 %, respectively and post-1980 averages of ~3 % and 2 %, respectively). Post-1980 IndVal-identified taxa included *Alona intermedia* (max abundance ~7 %), *Alona affinis* (max abundance ~27 %), and *Acroperus harpae* (max abundance ~11 %; Figure 2.3).

### 2.5.3 Historical climate observations and downcore trends in DBTs and VRS-chla

Given that ~1980 marked the beginning of AOSR industrial oil sands activity on a regional scale, overlapping stressors of climate change and local industrial activity were present from ~1980 onward. Mean annual air temperatures from Fort McMurray recorded increases since the beginning of the record, with recent decades, including those in the period of overlapping stressors, demonstrating some of the highest temperatures (Figure 2.5). Trends in seasonal mean air temperatures from Fort McMurray were similar, with the exception of fall temperatures that demonstrated no clear increasing trend (Supplemental Figure 2.1). Annual precipitation from Fort McMurray shows general declines and lower levels of precipitation recorded during the period of overlapping stressors (Figure 2.5). Trends in total precipitation from most seasons in Fort McMurray were similar, except spring records, which demonstrated no clear decreasing trend in recent decades (Supplemental Figure 2.1). As reported in Kurek et al. (2013a) and Laird et al. (2013), increasingly warmer and drier climate in recent decades were consistent with historical observations from other comparable regions of the Canadian prairies and boreal transition zones (Schindler and Donahue 2006).

Downcore concentrations of DBTs were used to characterize deposition of airborne pollution from the local industrial oil sands operations to RAMP 209 (Figure 2.3). The DBT profiles were relatively stable until the ~1980s, gradually increased until the early-2000s, and then abruptly increased to at least ~3 times higher than pre-1980 levels thereafter. As shown originally in Summers et al. (2016) and similar to the DBT concentrations, the downcore VRS-chla profile from RAMP 209 was stable until the early-1980s, increased gradually until ~2000s,
and increased markedly for the remainder of the core to the record’s highest levels in the most-recent intervals (Figure 2.3).

2.6 Discussion

2.6.1 Changes in biological assemblages

Assemblage shifts during the past ~75 years in the diatoms, chironomids, and cladocerans from RAMP 209 sediments represent the biological response of a typical, shallow AOSR lake to the multiple stressors in the region. Although ANOSIM tests identify significant (p < 0.05) differences between pre- and post-1980 assemblages in all examined bioindicator groups from RAMP 209, the assemblages (Figure 2.3) and trends in PCA axis 1 sample scores (Figure 2.4) do not reflect marked, abrupt changes. Rather, the biotic assemblages exhibit subtle, gradual responses (Figures 2.3-2.4) to long-term environmental stressors, such as climate change, which precedes the beginning of industrial development. Given that RAMP 209 is a typical AOSR lake (Natural Regions Committee 2006), the significant yet gradual multi-trophic changes identified in each trophic level may also be present in the many other comparable shallow lakes across the landscape.

2.6.2 Changes in diatom assemblages indicative of increased benthic habitat complexity

As expected, the diatom assemblages of this shallow lake are mainly composed of benthic taxa. Despite significant differences between assemblages from before and after the establishment of regional industry (identified by an ANOSIM test), the transitions in the community are subtle and gradual. There is no evidence of an abrupt, marked assemblage shift at any point in the record (Figures 2.3-2.4). Small, colonial, epipelic taxa (e.g., Staurosira construens complex, Pseudostaurosira brevistiata, and Staurosirella pinnata) were dominant throughout the diatom record, with slight declines in recent sediments (Figure 2.3). These fragilarioid taxa have been reported to be strong competitors across wide ranges of environmental
conditions, including broad gradients in temperature (Karst-Riddoch et al. 2009), total phosphorus (Bennion et al. 2001), and habitat preferences (Lotter and Bigler 2000; Bennion et al. 2010). They have also been observed in high abundances when they have the opportunity to outcompete other diatoms by tolerating relatively harsh environmental conditions that can constrain the growth of most other diatom taxa (Smol 1988; Lotter and Bigler 2000). For example, the high relative abundance of these taxa recorded over the entire sedimentary sequence in shallow RAMP 209 may be indicative of the persistence of low light levels over time due to ice cover and/or high concentrations of DOC.

Despite imperfect understanding of periphytic diatom habitat preferences (Bennion et al. 2010), the recent occurrence and increased relative abundance of taxa known to be associated with macrophytes (e.g., *Psammothidium curtissimum*, *P. subatamoides*, *Eolimna minima*, and *Sellaphora seminulum*), as well as other substrates (Figure 2.3; Blindow, 1987; Cattaneo and Kalff, 1978; Vermaire et al., 2011; Wetzel et al., 2015), indicate a potential transition to a more complex benthic habitat with increased abundance and/or diversity of substrates (e.g., macrophytes) available for colonization. Although IndVal identifies more diatom indicators (e.g., *Aulacoseira valida*, *A. nygaardii*, *Eunotia sudetica*, *Nupela vitiosa*, and *Achnanthidium minutissimum*; Figure 2.3), caution should be exercised in qualitative interpretation because, at low relative abundances, the presence of taxa may not necessarily be indicative of their ideal environmental conditions. Overall, the dominance of benthic fragilarioid taxa throughout the diatom record, with small and gradual transitions to more specialized periphytic species, suggests subtle, yet ecologically important increases in benthic habitat complexity (i.e., substrate/macrophyte availability and diversity) at RAMP 209.

### 2.6.3 Changes in chironomid assemblages indicative of warming water temperatures

Although the chironomid assemblages of RAMP 209 from before and after the establishment of regional industry are significantly different, the record-long dominance and
limited changes in abundance of several taxa (Figure 2.3) suggest that historical shifts in RAMP 209’s littoral environment have been minor. Three of the four most abundant taxa present throughout the core (Dicrotendipes nervosus, Microtendipes pedullus, and Paratanytarsus) are commonly associated with aquatic vegetation (Brodersen et al. 2001), indicating that RAMP 209’s littoral environment has likely included at least some macrophytes since the beginning of the record, and is relatively similar to today. The other most persistently abundant taxon, Tanytarsus mendax, is associated with more turbid lakes (Langdon et al. 2010).

The subtle changes that are evident in RAMP 209’s chironomid record are likely indicative of modestly increasing water temperatures and are reflected in several indicator taxa present at low abundances (Figure 2.3). Indicator taxa that are only present later in the chironomid record may indicate warming water temperatures, which agree with meteorological observations. Heterotanytarsus are thermophilic (Walker et al. 1991), and Zavrelia, although indistinguishable from Stempellinella when head capsules are lacking premandibles (Brooks et al. 2007), are typically present in temperate lakes (Walker et al. 1991). Further, Heterotanytarsus prefer low pH and high DOC waters (Walker et al. 1991), which are reflected in RAMP 209’s current water chemistry. The increased abundances later in the record of Cladotanytarsus mancus (Figure 2.3), a taxon with a known preference for warm waters (Barley et al. 2006), more clearly imply rising water temperatures. However, C. mancus appears to replace Psectrocladius barbatipes (Figure 2.3), which shares similar ecological preferences (Brooks et al. 2007). P. barbatipes is littoral (Lindegaard, 1992), commonly associated with macrophytes (Brodersen et al. 2001), and has a preference for relatively warm waters (Brooks et al. 2007). Thus, the qualitatively inferred changes in water temperatures are likely modest. As is common with the interpretation of chironomid assemblages, understanding the interactions among environmental factors, including climate variables, trophic dynamics, and micro-habitat variation, is difficult (Brodersen and Quinlan 2006). Although the early habitat of RAMP 209 may not have been
markedly different than it is today, the transitions in the chironomid assemblages are likely reflective of recent increases in water temperature and, like the diatoms, the changes are subtle and do not support a threshold-type response.

2.6.4 Changes in cladoceran assemblages indicative of increased benthic habitat complexity

Similar to the diatoms and chironomids, the cladoceran assemblages suggest subtle, gradual shifts (as summarized in the downcore PCA axis 1 sample scores; Figure 2.4). Also, similar to the diatoms, post-industry cladoceran assemblages suggest a more complex benthic habitat with increased availability and/or diversity of substrates for colonization. Again, the transitions are modest and not representative of a threshold-type response (Figure 2.3).

Broadly, declines in generalists coincident with increases in taxa with more specialized habitats and feeding strategies may indicate shifts to a more complex littoral environment as specific habitats become more important to the assemblage (Mosscrop et al. 2015; Thienpont et al. 2015). Although the cladoceran record shows dominance of a generalist taxon (*Chydorus brevilabris*; Kurek et al., 2011; Labaj et al., 2015; Mosscrop et al., 2015), its subtle decline in recent sediments, paired with the declines in taxa that have been found associated with mosses (e.g., *Alonella excisa* and *Alonella nana*; Duigan and Birks, 2000), suggests a simple littoral habitat early in the record. These taxa are replaced by taxa with more specialized habitats and feeding strategies (Figure 2.3), although *Alona affinis* is known to be tolerant of a broad range of conditions (Duigan and Birks 2000; Kurek et al. 2011; Mosscrop et al. 2015). For example, *Acroperus harpae* is a macrophyte-associated scraper (Dole-Olivier et al. 2000), whereas *Alona intermedia* is a detritivore (Chengalath 1982) that is often abundant in structurally-diverse habitats with mud and partial vegetation (Tremel et al. 2000). The subtle transition from generalists and taxa with simple habitat preferences to taxa with more complex life strategies and specialized physical habitat preferences supports the diatom-based inferences that RAMP 209’s
benthic habitat availability has become more diverse and complex with increased availability and
diversity of substrates for colonization (e.g., macrophytes).

2.6.5 Potential drivers of biotic change

Climate observations and downcore measures of airborne oil sands contaminant
deposition (DBT concentrations) indicate that RAMP 209 is simultaneously exposed to warming
annual (Figure 2.5) and seasonal (except fall; Supplemental Figure 2.1) temperatures, a general
trend toward reduced annual and seasonal (except spring; Supplemental Figure 2.1) moisture, and
elevated amounts of aerial pollution from local industrial oil sands developments (Figure 2.3).
Thus, recent decades encompass a period of co-occurring stressors (climate changes and regional
industrial activities; Figure 2.5). Further characterizing the DBT enrichment factor of 6.2
calculated in Summers et al. (2016), the relatively stable trend in the downcore DBT
concentrations, until the gradual increase following the 1980s, suggests that RAMP 209 has been
receiving elevated industrial contaminants since the establishment of the commercial bitumen
industry ~1980. The abrupt increase in DBT concentrations in the early-2000s occurs in concert
with the onset of nearby in-situ extraction, suggesting a localized contribution of industry
contaminants, although the pathways of industrial in-situ PAC contamination are not entirely
resolved (Korosi et al. 2013, 2016). A timeline of both stressors in the AOSR would demonstrate
a period of only climate change followed by a period of overlapping stressors (~1980 onward) in
which climate conditions were generally warmer than any other time in the record and relatively
dry, and industrial airborne contamination levels increased persistently. Further, all of these
stressors are forecast to continue and intensify (Schindler and Donahue 2006; Canadian
Association of Petroleum Producers 2015a).

2.6.6 No evidence of biological response to airborne industrial contaminants

Qualitative assessments of the taxa changes in all examined assemblages find no clear
evidence of a biological response to airborne industrial contamination (i.e., DBTs) despite RAMP
209’s sedimentary record of DBT concentrations tracking a signal of aerial industrial contamination, and ANOSIM tests identifying significant differences in diatom, chironomid, and cladoceran assemblages comparing before and after the establishment of regional industry. We find subtle, gradual changes in the examined biological assemblages and do not detect evidence of a change consistent in magnitude or timing with a threshold-type response attributable to airborne industrial contamination at the onset of regional industry (~1980) or the development of nearby in-situ extraction operations (early 2000s). Presumably, water-column and/or sedimentary concentrations of petrogenic contaminants have not yet surpassed levels that would cause severe biological responses at RAMP 209.

2.6.7 Biological transitions consistent with climate change

Interpretations of the biological assemblages from RAMP 209 and the downcore PCA axis 1 sample scores suggest subtle, gradual transitions in community composition rather than abrupt changes at the beginning of regional industry ~1980 or the development of nearby in-situ operations in the early 2000s. The nature of the assemblage changes from multiple trophic levels, which are inconsistent with the magnitude and timing of a marked biological response to airborne industrial contamination, suggests transitions to an increasingly complex benthic environment (interpreted through algal and invertebrate bioindicators) and warmer conditions (interpreted through indicator chironomids and climate observations). These assemblage changes are likely linked to a warming and drying regional climate. Although we do not find evidence of a threshold-type response in the biological assemblages, the ongoing deposition of industrial contaminants since ~1980 may be contributing to the subtle biotic transitions that we find to be primarily driven by climate change.

Warmer temperatures, which are direct consequences of the climate changes already present in the AOSR (Figure 2.5), can improve growing conditions for many aquatic organisms, including macrophytes, algae, and invertebrates, through reduced ice-cover duration and ice-
cover thickness, if other resources are not limiting (Adrian et al. 2009). In the northern region of Alberta, which hosts the AOSR, it is likely that ice conditions in the winter and spring are critically important controls on the biotic structure in shallow lakes and wetlands (Bayley et al. 2007), and it is likely that these mechanisms, as demonstrated in other scenarios (Thienpont et al. 2015), are important drivers of the subtle, biologically-inferred transitions in RAMP 209’s benthic environment. The warmer conditions qualitatively inferred by changes in the chironomid assemblages, and supported by climate observations, characterize a direct change in RAMP 209’s environment. Warmer conditions, through their effects on ice cover, for example, are also a potential mechanism driving the changes toward an increasingly complex and macrophyte-rich littoral habitat, which was inferred through diatoms and cladocerans. As such, the assemblage changes and inferred transitions in environmental conditions across all bioindicator groups are likely related.

In theory, habitat transitions in a lake could also be due to nutrient inputs from external contributors (e.g., industry), changing climate conditions (warmer, drier conditions yielding more nutrient-rich dust and/or concentrating nutrients in the closed-basin lake; Gibson et al., 2016; Smol and Douglas, 2007), and/or internal loading of phosphorus caused by anoxia. However, there is no evidence of marked changes in historical nutrient levels despite recent, clear increases in trends of inferred primary production at RAMP 209. The VRS-chla trend at RAMP 209 broadly matches inferred primary production increases across the AOSR that have been linked to warming temperatures (Kurek et al. 2013a; Summers et al. 2016). Further, Summers et al. (2016) found no evidence of substantial, industry-sourced deposition of biologically available phosphorus to stimulate primary production in AOSR lakes, including RAMP 209, which is phosphorus-limited (average TN:TP from 2002 to 2013 = 31.7, max = 65.6, min = 10.4). The climate change mechanisms (i.e., increasing temperatures and reduced moisture causing longer growing seasons and improved conditions for autotrophic growth) that Summers et al. (2016)
identified as the predominant drivers of the regional increases in inferred primary production in lakes across the AOSR, including RAMP 209, are further supported by water chemistry data and the subtle, long-term transitions in the examined biological assemblages. For example, end-of-summer water chemistry measurements from RAMP 209 extending back to 2002 indicate that the lake has been relatively high in nutrients for more than the last decade. Moreover, the nature and modest magnitude of changes in the diatom assemblages, which are established proxies for nutrient conditions, suggest that despite increases in inferred primary production, nutrient levels in RAMP 209 have remained relatively stable over at least the past ~75 years.

2.7 Conclusions

Our findings provide insights across multiple trophic levels that would not be possible through a conventional monitoring program. Although the statistical framework used in this study compares biological assemblages from before and after the establishment of the region’s industry, and significant differences are recorded across this time period for all biological proxies, the gradual, subtle changes across all three groups of bioindicators provide no clear evidence of an abrupt response to the deposition of airborne contaminants from industry or nutrient increases. Rather, the biotic shifts suggest climate change and associated mechanisms are likely the predominant drivers of transitions in all studied trophic levels from shallow RAMP 209. Despite the lack of threshold-type biological responses thus far, marked change in ecological structure and function cannot be ruled out of the lake’s future, given the forecast continuation and intensification of the multiple stressors acting on RAMP 209. Ongoing studies of bioindicators in multiple AOSR lakes with similar limnological and morphometric characteristics but differing industrial oil sands impacts should further elucidate the historical trends in the region’s lakes and continue to tease out the effects and mechanisms of the region’s environmental stressors on aquatic biota. Given that RAMP 209 represents a typical AOSR lake, the findings of this study
demonstrate the vulnerability of the region’s freshwaters to the ongoing industrial activity and climate change in the area.

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2.9 References


Figure 2.1. Map of study region and study site

Location of RAMP 209, the main development area (established ~1980), the in-situ operation near the study site (production beginning in 2003), two local communities, and the footprint of industrial oil sands development.
Figure 2.2. Radioisotope decay curves for sediment dating

Total $^{210}$Po activity (measured using alpha spectroscopy) is indicated by white circles and the associated errors (± 1 SD), and used as a proxy for total $^{210}$Pb activity. Background activity, measured using alpha spectroscopy on $^{226}$Ra, is denoted by the dashed vertical line. Black “Xs” denote the $^{226}$Ra activity in the three measured intervals (plotted on same scale as Total $^{210}$Po Activity). Constant rate of supply (CRS)-inferred dates are represented by black circles and the associated errors (± 2 SD).
Figure 2.3. Stratigraphic display of select bioindicator taxa, DBT (industrial oil sands contaminant) concentrations, and sedimentary-inferred primary production

Percent relative abundances of dominant diatom, chironomid, and cladoceran taxa and significant (p < 0.05) indicator taxa (identified by IndVal analyses) displayed stratigraphically. Taxa are arranged by the IndVal statistic (in parentheses after taxa names) within categories of pre- and post-establishment of regional oil sands industry (~1980) for each group of bioindicator. Taxa with no specified indicator time period or IndVal statistic are not indicators, but are included because they are the most abundant taxa in the records. Measured downcore DBTs and spectrally-inferred downcore chlorophyll-a (VRS-chla) concentrations are on the far right. The dashed horizontal line denotes 1980 (establishment of regional industry ~50 km away from RAMP 209).
Figure 2.4. Principal components analyses (PCAs)

Downcore PCA axis 1 (PC1) sample scores for diatom, chironomid, and cladoceran assemblages. Dashed horizontal line denotes 1980 (approximate establishment of regional industry ~50 km away from RAMP 209).
Figure 2.5. Historical annual climate records

Historical annual mean air temperature and total precipitation representative of the Athabasca Oil Sands Region (AOSR). Shaded grey areas represent 1980 onward, a period of overlapping stressors in the region (i.e., climate change and local industrial activity). Temperature data from Environment and Climate Change Canada station ID: 3062696. Precipitation data from Environment and Climate Change Canada Station ID: 3062693 and the Regional Aquatics Monitoring Program’s (RAMP’s) Aurora climate station.
Supplemental Figure 2.1. Historical seasonal climate records

Historical seasonal mean air temperature and total precipitation representative of the Athabasca Oil Sands Region (AOSR). Shaded grey areas represent 1980 onward, a period of overlapping stressors in the region (i.e., climate change and local industrial activity). Temperature data from Environment and Climate Change Canada station ID: 3062696. Precipitation data from Environment and Climate Change Canada Station ID: 3062693 and the Regional Aquatics Monitoring Program’s (RAMP’s) Aurora climate station.
Chapter 3

Recent warming, rather than industrial emissions of bioavailable nutrients, is the dominant driver of lake primary production shifts across the Athabasca Oil Sands Region

3.1 Abstract

Freshwaters in the Athabasca Oil Sands Region (AOSR) are vulnerable to the atmospheric emissions and land disturbances caused by the local oil sands industry; however, they are also affected by climate change. Recent observations of increases in aquatic primary production near the main development area have prompted questions about the principal drivers of these limnological changes. Is the enhanced primary production due to deposition of nutrients (nitrogen and phosphorus) from local industry or from recent climatic changes? Here, we use downcore, spectrally-inferred chlorophyll-\(a\) (VRS-chla) profiles (including diagenetic products) from 23 limnologically-diverse lakes with undisturbed catchments to characterize the pattern of primary production increases in the AOSR. Our aim is to better understand the relative roles of the local oil sands industry versus climate change in driving aquatic primary production trends. Nutrient deposition maps, generated using geostatistical interpolations of spring-time snowpack measurements from a grid pattern across the AOSR, demonstrate patterns of elevated total phosphorus, total nitrogen, and bioavailable nitrogen deposition around the main area of industrial activity. However, this pattern is not observed for bioavailable phosphorus. Our paleolimnological findings demonstrate consistently greater VRS-chla concentrations compared to pre-oil sands development levels, regardless of morphological and limnological characteristics, landscape position, bioavailable nutrient deposition, and dibenzothiophene (DBT)-inferred industrial deposition. Furthermore, breakpoint analyses on VRS-chla concentrations across a gradient of DBT-inferred industrial deposition show limited evidence of a contemporaneous change among lakes. Despite the contribution of bioavailable nitrogen to the landscape from
industrial activities, we find no consistency in the spatial pattern and timing of VRS-chlα shifts with an industrial fertilizing signal. Instead, significant positive correlations were observed between VRS-chlα and annual and seasonal temperatures. Our findings suggest warmer air temperatures and likely decreased ice covers are important drivers of enhanced aquatic primary production across the AOSR.

3.2 Introduction

Deposits of bitumen in northeastern Alberta are immense. Estimates indicate ~167 billion barrels of oil underlie ~140,000 km² of boreal forest (Canadian Association of Petroleum Producers 2015c). Of Alberta’s three major bitumen deposits, the Athabasca deposit is the largest, accounting for ~66 % of Alberta’s total oil sands area and ~80 % of current oil production (Government of Alberta 2013; Alberta Energy Regulator 2015). The Athbasca deposit hosts the only significant Canadian bitumen deposit accessible via surface mining as well as in situ techniques.

Since the late-1960s, the development of Alberta’s oil sands industry has expanded rapidly (Canadian Association of Petroleum Producers 2015d). As continued industrial growth is forecast (Environment Canada 2011; Canadian Association of Petroleum Producers 2015b), freshwaters in the Athabasca Oil Sands Region (AOSR) are vulnerable to pollutant emissions, wind-blown dust, land disturbance, and declines in water quality and quantity associated with local industrial activities (Schindler 2010; Rooney et al. 2012). However, regional freshwaters are also sensitive to the shifts in air temperature (~1.65 °C increase in average air temperature since 1960) and precipitation patterns (increased evaporation and glacial melt, and decreased snowpack and river flows) that have occurred in the region during the 20th century (Schindler and Donahue 2006). It is expected that climate change will have substantial effects on the ecology and biological production of the shallow lakes in the western Boreal Plain through both abiotic and biotic controls (Schindler 1998; Sass et al. 2008).
Paleolimnological studies can effectively compare conditions before and after disturbances, such as industrial activity, and are thus useful in the absence of, and in combination with, monitoring data (Smol 2008). Paleolimnological studies have played an integral role in assessing recent environmental changes in the AOSR. For example, studies have noted that aquatic systems in the AOSR and those nearby have shifted in ecological structure and function (Hazewinkel et al. 2008; Curtis et al. 2010; Kurek et al. 2013a, b; Laird et al. 2013). A recent study used dated lake sediment cores to demonstrate that sedimentary chlorophyll a and its diagenetic products (hereafter chla), a proxy for whole lake primary production, have substantially increased since the 1970s in five shallow lakes located up to ~50 km away from the main development area of the AOSR (Kurek et al. 2013a). Shifts in lake structure and function were also reflected by higher trophic levels as algal-grazing zooplankton such as Daphnia increased in both occurrence and relative abundance, despite increased contaminant deposition (Kurek et al. 2013a). Changes in diatom assemblage composition (Hazewinkel et al. 2008), diatom valve flux (Laird et al. 2013), stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotopes, and nutrient ratios (C:N; Curtis et al., 2010) in lakes across the broader region (up to ~350 km from the main development area) also support the findings of enhanced primary production in many of these systems during recent decades.

While many lakes in the AOSR are currently moderately to highly productive (Hazewinkel et al. 2008; Kurek et al. 2013a), increases in primary production may reflect the impacts from expanding industrialization. Bioavailable sources of reactive nitrogen from industrial upgrader emissions and wind-blown dust rich in cations and phosphorus from disturbed landscapes could also stimulate lake primary production (Landis et al. 2012; Watmough et al. 2014). Analyses of snowpack (Kelly et al. 2009, 2010; Kirk et al. 2014), lichen (Addison and Puckett 1980), moss (Zhang et al. 2016), and soil (Watmough et al. 2014) from the AOSR identified atmospheric deposition of metals and other pollutants from industrial oil sands.
activities. Kirk et al. (2014) estimated 28.6 tonnes of total phosphorus and 463 tonnes of total nitrogen, among other particulates, were deposited in the spring snowpack (winter 2011-2012) within a ~50-km radius of the main industrial upgrading facilities, suggesting that increased loading of bioavailable nutrients may also be occurring across the region.

Alternatively, increases in primary production in shallow lakes of the AOSR may reflect a regional stressor, such as climate change, as the key driver of lake shifts (Kurek et al. 2013a). Climate directly affects the physical properties of lakes and indirectly influences the availability of key resources for primary production, including light, nutrients, and habitat (Kraemer et al. 2015; Rühland et al. 2015). Temperature-mediated processes can affect primary production and subsequent concentrations of chl$\alpha$ in aquatic ecosystems. Increasing temperatures can, for example, reduce ice cover and lengthen the growing season, altering light availability, nutrient dynamics, and turbulence (Weyhenmeyer 2001; Smol and Douglas 2007b). Warmer temperatures can also alter the evaporation-to-precipitation ratio, and concentrate nutrients as well as ions and pollutants, with shallow lakes being particularly sensitive given their high surface area-to-volume ratio (Smol and Douglas 2007a). Precipitation-mediated processes affect transport of allochthonous materials into lakes, and influence water levels, water colour, and nutrient availability (Schindler et al. 1996; Sass et al. 2008; Cobbaert et al. 2015). Reduced water levels can concentrate nutrients (Markensten 2006) and, depending on turbidity, promote light penetration to greater depths, thereby facilitating an expanded zone of aquatic primary production (Beklioglu et al. 2006). Additionally, windy conditions may promote resuspension of nutrients and subsequent growth of phytoplankton, although the effect is dependent on the size and depth of a lake (Carrick et al. 1993; Bayley et al. 2007).

Shallow lakes and wetlands comprise a large portion of the AOSR landscape. Although alternate equilibria in shallow lakes can dampen or delay ecological responses, the relatively small volumes and lower dilution potential in shallow systems can predictably accelerate and/or
amplify their response to environmental stressors such as contaminant deposition and climate change (Smol 2016). Increases in primary production in AOSR lakes could thus indicate water quality shifts across the region.

This study uses dated cores from 23 limnologically-diverse lakes (Supplemental Tables 3.1-3.2) in catchments relatively undisturbed by oil sands industry located ~10 to 200 km away from the main development area (Figure 3.1) to characterize spatial patterns in primary production across the AOSR. Maps of nutrient deposition across the AOSR, developed from snowpack samples, are used to compare spatial patterns of nutrient deposition with the changes in aquatic primary production. We reason that an industrial fertilizing effect will diminish with declining aerial deposition and increased distance from industrial oil sands developments, while the effect of a climatic driver will persist across the region. With forecasts of continued climatic changes and intensifying industrial development in the AOSR, it is plausible that further ecosystem shifts will occur. Understanding the long-term dynamics and key drivers of this primary production increase is crucial in shaping the management strategies of freshwater resources in the AOSR.

3.3 Materials and methods

3.3.1 Sample collection and $^{210}$Pb dating

Sample collection and dating methods followed the procedure reported in Kurek et al. (2013a). Sediment cores from 23 limnologically-diverse lakes (Supplemental Table 3.2) within a ~200-km radius of the main development area in the AOSR were collected through augered ice holes at the deepest part of the lakes in March 2011, 2012, 2013, and 2014. Samples were collected as part of the Canada-Alberta Joint Oil Sands Monitoring (JOSM) program (http://jointoilsandsmonitoring.ca) and were taken from both phosphorus and nitrogen-limited lakes (Supplemental Table 3.2) with catchments that were relatively undisturbed by industrial development. Lakes were accessed by helicopter and were located on provincially-owned,
publically-accessible lands. No private lands or reserve areas were accessed, no protected species were sampled, and no permits or permissions were required.

Cores were collected using a Uwitec gravity corer, specially designed for high-resolution work on recent sediments, and the sediment profiles were sectioned at 0.5-cm intervals for the upper 20 cm, and 1-cm intervals thereafter. Sediment intervals were sampled into Nalgene jars or Whirlpak® bags and immediately frozen. Determination of total $^{210}\text{Pb}$, $^{226}\text{Ra}$ and $^{137}\text{Cs}$ activity ($\pm 1$ SD) was completed by Flett Research Ltd. (Winnipeg, Manitoba, Canada). $^{210}\text{Po}$ and $^{226}\text{Ra}$ alpha radiation were measured as proxies for total $^{210}\text{Pb}$ and supported$^{210}\text{Pb}$ activity, respectively (Appleby 2001). $^{137}\text{Cs}$ activity ($\pm 1$ SD) was measured via gamma spectrometry and was used as an independent chronological marker of the radioactive fallout peak following the 1963 moratorium on nuclear weapons testing (Appleby 2001). The constant rate of supply (CRS) age model (Appleby 2001) was used to estimate sediment core age-depth relations ($\pm 2$ SD) for all analyzed intervals (where total $^{210}\text{Pb}$ exceeded supported $^{210}\text{Pb}$) and the dry mass accumulation rate for each core was used to extrapolate the sediment ages below background depth. Lastly, to assign a date to every interval, a polynomial regression was fitted to the previously-determined dates using the lowest order polynomial that provided a reasonable fit (Supplemental Figure 3.1).

### 3.3.2 Chlorophyll-α inferences using visible reflectance spectroscopy (VRS-chla)

Visible reflectance spectroscopy (VRS) was used to estimate concentrations of sedimentary chlα, which is a proxy for whole lake primary production (Wolfe et al. 2006; Michelutti et al. 2010). VRS methods measure the entire suite of chlα degradation products; therefore, the spectral inferences are not affected by post-depositional diagenetic processes (Wolfe et al. 2006; Michelutti et al. 2010). The use of VRS-chlα determinations has now been assessed in a variety of limnological settings and has been shown to be a reliable method for estimating trends in whole-lake primary production (Michelutti et al. 2010). For example, it has faithfully tracked recent increases in production with known fertilization events (Labaj et al.
2014), recent declines in primary production as a result of reduced upwelling of hypolimnetic nutrients due to enhanced thermal stability (Michelutti et al. 2015), and the effects of past algaeicide treatments on production (Korosi and Smol 2012c).

In preparation for spectral analysis, sediments were freeze-dried, sieved through a 125-µm mesh, and the spectral reflectance of ~2-3 mm of sediment in a 19 x 65 mm glass vial was analyzed in a FOSS NIRSystem Model 6500 rapid content analyzer. A simple reflectance metric (the area under the absorbance peak from 650-700 nm) was used to infer chla concentrations using a linear equation derived from an earlier calibration study (Michelutti et al. 2010). The estimated detection limit of this technique is ~0.01 mg/g dry weight.

3.3.3 Dibenzothiophenes (DBTs)

Dibenzothiophenes (DBTs) are a group of alkylated polycyclic aromatic compounds (PACs) that are characteristic of AOSR bitumen (Akre et al. 2004; Wang et al. 2014). DBT deposition in snow in the AOSR declines as a proportion of total PACs with increased distance from industrial development, while unsubstituted PACs show no geographical trend. Thus, DBTs are considered indicative of industrial activities in the region (Akre et al. 2004; Kurek et al. 2013a). Downcore DBTs in dried sediments were analyzed by AXYS Analytical Services (Sidney, British Columbia, Canada) using a standardized method for PACs (MLA-021) based on the United States Environmental Protection Agency (US EPA) methods 1625B and 8270C/D. Concentrations of DBTs were calculated using the isotope dilution method of quantification. Five DBT analytes (Total DBTs = DBT + C1-C4-alkylated DBTs) were isolated. Additional details on PAC analysis in the sediment cores are given in Kurek et al. (2013a). A DBT enrichment factor, which is explained below, was used to characterize industrial impact at each site.

3.3.4 Analysis and mapping of nutrients in snow

To assess aerial inputs of nutrients across the landscape, loadings of total nitrogen (TN), dissolved inorganic nitrogen (DIN), total phosphorus (TP), total dissolved phosphorus (TDP), and
soluble reactive phosphorus (SRP) were measured in melted snow samples collected in March 2014. Snow was collected from ~135 sites located in a grid pattern up to 200 km from the major area of development (Supplemental Figure 3.2) using methods described in Kirk et al. (2014). TN and TP include both bioavailable and unavailable forms of nitrogen and phosphorus, respectively. DIN includes ammonia, nitrate, and nitrite, and thus represents bioavailable forms. Phytoplankton use orthophosphate, which is measured in SRP, and are also able to assimilate dissolved fractions of phosphorus, which is measured as TDP (Cortner and Wetzel 1992); therefore, SRP and TDP represent biologically-available forms. All samples were collected within 6 days in early March to ensure maximum snowpack depth and minimize snow variability over the course of sampling. Timing of sampling was selected based on historical snow accumulation data for the Fort McMurray region (Environment Canada, www.ec.gc.ca/dccha-ahccd). Composite snowpack profiles were collected using a pre-cleaned stainless steel corer into pre-cleaned high density polypropylene buckets using clean techniques as in Kirk et al. (2014). Analyses were completed at the National Laboratory for Environmental Testing (NLET), which is certified by the Canadian Association for Environmental Analytical Laboratories (CAEAL) using method 200.8 from the US EPA. Ten snow cores were collected at each site to determine average snow-water equivalence (SWE), which was then multiplied by nutrient concentrations at each site to calculate loadings, as described previously in Kirk et al. (2014) and Kelly et al. (2009). DIN snowpack loadings for 1978 were also available for 60 sites (Supplemental Figure 3.2) located varying distances from the main area of industry (Barrie and Kovalick 1980). Numerous site locations were similar between 1978 and 2014 and methods for sample collection were comparable.

ArcGIS10 Geostatistical Analyst software (Esri, Redlands, California) was used to interpolate 2014 spring-time loadings of all nutrients (TN, DIN, TP, TDP, SRP) for an area ~42,800 km², and 1978 DIN spring-time loading for an area ~23,800 km². Kriging settings and comparisons for
each interpolated parameter were similar to methods described in Kirk et al. (2014) and are provided in Supplemental Tables 3.3-3.5.

### 3.3.5 Numerical analysis

To facilitate comparisons of inferred primary production among lakes, standardized Z scores ((VRS-chla concentration – mean VRS-chla concentration)/standard deviation) were calculated for the VRS-chla concentrations. Enrichment factors were calculated to compare VRS-chla concentrations from the most recent sediment intervals (post-2000; n = 3-12 intervals per core) to concentrations ~15 years prior to industrial development in the region (1955-1970; n = 1-5 intervals per core). The ~15-year pre-industry time period was used because all sites had sediments dating back to at least 1955. To validate the appropriateness of the time intervals selected, enrichment factors were also calculated using the 2 and 3 most-recent intervals and the 2 and 3 intervals immediately preceding 1970. The various methods of calculating the enrichment factors yielded similar results, suggesting that the first method outlined was representative of the changes in VRS-chla concentrations occurring since industrial oil sands development.

In addition to measures of proximity to industry and landscape position, DBT enrichment factors were used to characterize the deposition of industrial oil sands contaminants at different sites. Although DBT enrichment factors represent the changing sedimentary concentrations of only one industrial contaminant, DBTs are the most representative indicators of industrial oil sands activity in the AOSR (Akre et al. 2004; Kurek et al. 2013a). Enrichment factors of DBTs are calculated from measured DBT concentrations in the sediments at each site, and are thus able to acknowledge the AOSR as a region of industry rather than a single facility. Additionally, DBT enrichment factors incorporate upwind/downwind landscape position and variable wind conditions in assessing the extent of airborne industrial pollution at each site. RAMP 227 was excluded because the DBT data did not extend to pre-industrial times (no measures for 1955-1970).
Sites were assigned to minimally DBT-enriched (DBT enrichment < 2) or highly DBT-enriched groups (DBT enrichment > 2). Given that there are no sedimentary guidelines for DBTs (Canadian Council of Ministers of the Environment 1999), we set the point of division for the categories at 2. The point of division was selected based on evidence that DBTs are generated locally (Kurek et al. 2013a), but to also account for possible long-range transport of DBTs from outside the AOSR. An independent two-sample t-test was used to identify any significant difference between the average VRS-chla enrichments of the highly and minimally DBT-enriched groups.

To characterize the timing of VRS-chla increases, piecewise linear regression models were applied to the VRS-chla concentration data (SigmaPlot Version 10). A linear relationship with a single breakpoint was assumed, and a two-segmented model was used. An ANOVA table and corresponding F test statistic from a null model was used to evaluate the statistical significance for each regression model. Breakpoint analyses were not completed on lakes where no stable baseline was captured (RAMP 175, RAMP 226, and 2014-B), where outliers would drive the timing of the breakpoint (Big Peter Pond), and where no DBT enrichment factor could be calculated (RAMP 227).

“Vegan” (Oksanen et al. 2012), “analogue” (Simpson and Oksanen 2016), and “Hmisc” (Harrell 2017) R packages (R Development Core Team 2015) were used to calculate Pearson correlations between VRS-chla enrichment factors, DBT-inferred industrial impact, and proximity to industry. Proximity to industry was defined as the distance to snow sampling site AR6, which is adjacent the Athabasca River and two major bitumen upgraders, and has been commonly used to represent the centre of industrial activities within the AOSR (e.g., Kelly et al., 2010, 2009; Kirk et al., 2014). Further Pearson correlation analyses were carried out on VRS-chla Z scores from minimally and highly DBT-enriched sites versus mean annual and seasonal air temperature (Fort McMurray, station no. 3062696) and precipitation (Fort McMurray, station no.
3062693) data from Environment Canada’s Adjusted and Homogenized Canadian Climate Data website (www.ec.gc.ca/dccha-ahccd). Given downcore sediment compaction and variation in sediment accumulation rates, temporal resolution of samples were reduced down core. To ensure each interval represented comparable amounts of time, the VRS-chla Z scores from cores in each group of DBT enrichment, and climate data were averaged across 5-year intervals (Supplemental Figures 3.3-3.4). Based on the $^{210}$Pb-estimated ages, most sediment intervals from highly DBT-enriched and minimally DBT-enriched cores represented less than 5 years. Therefore, resolution for VRS-chla Z scores and climate data averages was set to 5 years. Data from intervals representing more than 5 years were retained in the analyses because they represent data across the estimated time frames.

3.4 Results

Modern primary production, inferred from sedimentary chla, is greater than background values at all sites, regardless of proximity to industry (Figure 3.2, Supplemental Figure 3.5). Most of the 23 sites record relatively stable background (generally pre-1970s) VRS-chla concentrations followed by abrupt increases in surface sediments. Unlike the other sites, RAMP 175 shows a decreasing VRS-chla trend beginning ~2005. Similarly, RAMP 226 shows a decreasing trend from the late-1990s to the late-2000s with a subsequent return to early-1990s concentrations by 2011. Pushup Lake also shows a slight decrease in VRS-chla from the late-1990s to the mid-2000s (Supplemental Figure 3.5). Although VRS-chla profiles from these lakes differ from the regional patterns of consistent primary production increases, modern VRS-chla values are still higher than the pre-oil sands development values at these three sites, resulting in enrichment factors > 1. VRS-chla enrichment factors were > 1 at all 23 sites, averaging 1.8 (range 1.1 to 5.3; Table 3.1).

DBT enrichment factors exceeded 1 at all sites except RAMP 271 (0.7), averaging 4.4 (range 0.7 to 37.4; Table 3.1). Generally, DBT-enrichment factors are greater east of the main
development area and within the 50-km zone of high contaminant deposition identified in the study by Kelly et al. (2009); however, highly DBT-enriched lakes are not necessarily closer to the main area of industrial activity (Pearson correlation results for DBT enrichment factor versus distance to the main area of development: \( r = -0.4, p = 0.06 \)). Pushup Lake demonstrates the largest DBT enrichment factor due to a rapid increase to moderately high DBT concentrations from low pre-oil sands industry levels. The low pre-oil sands industry DBT concentrations at Pushup Lake contribute to the inflated DBT enrichment factor at this relatively distant site. NE20, which is close to the main area of industrial development and demonstrates the second-highest DBT enrichment factor, has modern DBT concentrations over an order of magnitude greater than Pushup Lake.

The average VRS-chla enrichment factors (Table 3.1) are not significantly different among the minimally and highly DBT-enriched groups (independent two-sample \( t(5) = 0.35, p = 0.74 \)). Further, no significant correlation was found between chla enrichment factors and proximity to the main area of industrial oil sands development \( (r = -0.23, p = 0.29) \) or DBT-inferred industrial impact \( (r = -0.23, p = 0.31) \). Breakpoint analyses on VRS-chla concentrations from each lake (except RAMP 175, RAMP 226, 2014-B, Big Peter Pond, and RAMP 227 for reasons described in Materials and Methods) identified abrupt changes ranging from \( \sim1919 \) to \( \sim2006 \) (Figure 3.2, Supplemental Figure 3.5), with 83% (15 out of 18) of the lakes experiencing an abrupt change at \( \sim1970 \) or later (Figure 3.2, Supplemental Figure 3.5).

Pearson correlation analyses identified significant \( (p < 0.05) \) positive correlations between mean annual and seasonal air temperatures, and VRS-chla Z scores in the highly DBT-enriched group and all sites combined (Table 3.2). Positive, significant correlations were also identified between mean annual, winter, summer, and fall air temperatures, and VRS-chla Z scores in the minimally DBT-enriched group. No significant correlations were identified between VRS-chla Z scores and mean annual and seasonal precipitation (Supplemental Table 3.6).
Maps of interpolated 2014 TN and DIN snowpack loadings show deposition of both total (TN) and biologically-available (DIN) nitrogen, with elevated deposition around the main area of local industrial activity (Figures 3.3-3.4). This deposition pattern is consistent with that observed for total PACs, unsubstituted PAHs, alkylated PAHs, DBTs, mercury and methyl mercury, several metals known to be emitted in large quantities from the oil sands upgrading facilities (e.g., Ni, V, and Zn), crustal elements (Al and La), and total suspended solids (Kirk et al. 2014). Using interpolated loadings, we calculated the quantity of nutrients deposited to the area within a 50-km radius of the major oil sands developments. In 2014, deposition of TN and DIN within 50 km of the main area of industrial oil sands development was 172 and 102 tonnes, respectively. In 1978, interpolated loadings of DIN (measures from Barrie and Kovalick, 1980) also showed elevated deposition around the main industrial area (Supplemental Figure 3.6), only ~15% lower than the 2014 value at 88 tonnes (corrected to account for differences in days of snow accumulation between winter 1977-1978 and winter 2013-2014, respectively 85.5 and 117.5 days). Using these interpolated loadings, we can estimate that at least 3,265 tonnes of DIN has been deposited on the landscape within 50 km of the main industrial oil sands developments.

Like TN and DIN, maps of interpolated 2014 spring-time TP across the same area show an industry-centred area of elevated deposition (Figure 3.5). Conversely, the bioavailable forms of deposited phosphorus (TDP and SRP) are not elevated around the main area of industrial activity (Figures 3.6-3.7). Total deposition concentrations of TP, TDP, and SRP within 50 km of the main area of oil sands industry in 2014 are 12.0, 2.9, and 1.0 tonnes, respectively.

3.5 Discussion

3.5.1 Is there a regional increase of primary production in the Athabasca Oil Sands Region?

We show modern primary production greater than background levels at all sites, including those located ~200 km away from the AOSR’s main oil sands developments. Further, there is no relationship between VRS-chla increases and proximity to the main area of
development or DBT-inferred industrial oil sands impact. There is also no consistent pattern of VRS-chla increases dependent on landscape position or bioavailable nutrient loading (Figures 3.4, 3.6-3.7). Finally, there is no significant difference between average VRS-chla enrichments from minimally and highly DBT-enriched groups. The pervasive, similar increases in VRS-chla concentrations across the AOSR, independent of proximity to industrial development or industrial emissions, suggests a region-wide driver of the enhanced primary production.

3.5.2 Is there a fertilization effect from upgrading and mining activities?

Our study finds no consistent evidence of nutrient deposition from oil sands industry stimulating increased primary production in industry-impacted lakes. We reason that industry-driven aerial fertilization and associated enhanced primary production would mimic the spatial patterns of bioavailable nutrient deposition (Figures 3.4, 3.6-3.7), and/or be most evident within the 50-km zone of high contaminant deposition surrounding the main area of industry (Kelly et al. 2009, 2010; Kirk et al. 2014) and diminish with increasing distance (Landis et al. 2012; Watmough et al. 2014). Enrichment factors summarize a change over time and enable us to identify an industrial fertilization effect as higher VRS-chla enrichment factors at sites with higher DBT enrichment factors. Our findings suggest no relationship with distance or direction from the centre of industry or DBT enrichment factor, which is independent of distance from the major developments. Nutrient deposition maps based on late winter snow loadings indicate industrial oil sands activities are currently sources of biologically-available nitrogen, but not phosphorus. Although a majority of the study sites are phosphorus-limited lakes, there is no pattern of increased VRS-chla in nitrogen- or phosphorus-limited lakes to match deposition patterns of bioavailable nutrients (Figures 3.4, 3.6-3.7). As such, we provide no spatial evidence of industrially-mediated production enrichments in AOSR lakes.

Further, the substantial range in VRS-chla breakpoints from lakes across a DBT-inferred industrial impact gradient (Figure 3.2, Supplemental Figure 3.5) suggests that primary production
is changing on a timeline specific to each lake. Despite this, the majority of lakes (83\%) on which breakpoint analyses were applied change after \~1970, suggesting most sites are responding to driver(s) in recent decades. However, the individualistic timing of breakpoints persists in recent decades and the contributions of co-occurring stressors cannot be disentangled using timing of change alone.

Although our findings do not provide consistent spatial or temporal evidence of an industrial fertilizing effect on primary production in lakes across the AOSR, they cannot rule out the threat of continued industrial activities as a potential driver of shifts in lake structure and function.

3.5.3 What is driving enhanced primary production across the Athabasca Oil Sands Region?

Our findings suggest climate warming as a likely driver of increased aquatic primary production in the AOSR. Significant, positive correlations between average annual and seasonal air temperatures (Table 3.2), which are increasing in the AOSR (Supplemental Figure 3.3), and standardized VRS-chla concentrations (Z scores) from both groups of DBT-enriched sites (Supplemental Figure 3.4) and all sites combined support the reasoning that a warming climate may be facilitating favourable conditions for primary producers. Specifically, the correlations between standardized VRS-chla concentrations and increased temperatures in the winter, spring, and fall suggest a longer and/or warmer growing season for primary producers as one likely mechanism. Lake ice phenology and stability of the water column are some of the most important controls on primary production and aquatic biological communities in ice-covered lakes (Smol and Douglas 2007b; Rühland et al. 2013, 2015), and favourable shifts in light conditions and nutrient availability that accompany an earlier ice-off period are known to stimulate primary production (Weyhenmeyer 2001). An increase in the ice-free season of these lakes may be
stimulating primary production. Further, warming temperatures, in scenarios where there are no other limiting resources, typically yield increased primary production (Adrian et al. 2009).

Our findings indicate that the variable, decadal-scale precipitation patterns in the AOSR are not playing a significant role in driving increased aquatic primary production. Although increases in primary production from our 23 lakes are not significantly correlated to precipitation (Supplemental Table 3.6), reduced moisture due to periods of low precipitation, reduced snowpack and ice-cover, and increased evaporation caused by warmer temperatures (Schindler and Donahue 2006) can concentrate nutrients and cations in waterbodies (Markensten 2006; Smol and Douglas 2007a, b) and could exacerbate production trends, especially in shallow lakes. The larger, though still not significant, correlation coefficients (r) between VRS-chla and winter precipitation and VRS-chla and fall precipitation suggest that snow may be an important component of the relationship between total precipitation and primary production in the AOSR. Further, shallow lakes with small catchments, which form the majority of our study sites, may not be reflecting changes in precipitation inflow to the same degree as would larger, deeper lakes with substantial catchments and inflows.

Our findings of significant correlations between standardized VRS-chla concentrations and warming temperatures are congruous with findings from biological species assemblages demonstrating changes consistent with recent regional climate warming in lakes near (within 50 km; Kurek et al., 2013a) AOSR industry and farther downwind (up to 300 km NE; Laird et al., 2013). The consistent findings of recent AOSR studies, including our study presented here, provide strong evidence of complex multiple-stressor systems in which climate warming plays an important role.

**3.5.4 Summary and future directions**

Our study’s inclusion of many diverse lakes located up to ~200 km away from the centre of industrial oil sands activities facilitates assessment of broad-scale patterns and development in
our understanding of the AOSR in a dual scenario of climate change and intense industrial activity. Our findings confirm a regional increase in primary production and indicate that warming temperatures are an important driver in AOSR aquatic primary production. Improved understanding of the roles and relative importance of industrial development and climatic change on foundational ecosystem processes in the AOSR likely lies in the assessment of downcore biotic and abiotic (e.g., stable nitrogen and carbon isotope) trends. These analyses should further elucidate the production histories and nutrient sources of regional lakes, and provide crucial information regarding the vulnerability of these aquatic systems.

3.6 Acknowledgments

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3.7 References


Table 3.1. Enrichment factors from minimally and highly DBT-enriched sites

VRS-chlα enrichment factors, corresponding DBT enrichment factors, and average post-2000 DBT concentrations, organized by DBT enrichment factor. Asterisks (*) denote sites for which breakpoint analysis was not considered appropriate and was thus not applied. RAMP 227 is excluded because the DBT record does not extend to pre-oil sands industry.

<table>
<thead>
<tr>
<th>Site</th>
<th>Chlα Enrichment Factor</th>
<th>DBT Enrichment Factor</th>
<th>Average [DBT] Post 2000 (ng/g dry weight)</th>
<th>DBT Enrichment Group</th>
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<td>RAMP 271</td>
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<td>1.6</td>
<td>29.8</td>
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<td>1.8</td>
<td>34.2</td>
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Table 3.2. Correlations between five-year averaged VRS-chla Z scores and five-year averaged temperature data

Results from Pearson correlations between VRS-chla Z scores (averaged over five-year intervals) from highly and minimally DBT-enriched sites and all sites combined and annual and seasonal AOSR temperature (averaged over the same five-year intervals).

<table>
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<th>Temperature</th>
<th>Annual</th>
<th>Winter</th>
<th>Spring</th>
<th>Summer</th>
<th>Fall</th>
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<td>r</td>
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<td>0.50</td>
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<td>p</td>
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<td>0.005</td>
<td>0.03</td>
<td>0.001</td>
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<td><strong>Minimally DBT-Enriched Sites</strong></td>
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<td>0.62</td>
<td>0.44</td>
<td>0.68</td>
<td>0.62</td>
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<td><strong>All Sites</strong></td>
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<td>0.047</td>
<td>0.001</td>
<td>0.01</td>
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Figure 3.1. Map of study area

Locations of 23 study lakes and two local communities (Fort McMurray and Fort McKay), and the footprint of industrial oil sands development. Lakes were cored in March 2011, 2012, 2013, or 2014. Many of the lakes have multiple names (Supplemental Table 3.1).
Figure 3.2. Changes in VRS-chla from each study site

Downcore profiles of VRS-chla concentrations from (A) minimally DBT-enriched sites and (B) highly DBT-enriched sites. Individual profiles are provided in Supplemental Figure 3.5. Timing of abrupt change in VRS-chla concentrations for all sites where breakpoint analysis was applicable, ordered by DBT-enrichment factor are shown in (C). Black rectangles with white midline denote timing of breakpoint and standard error. Sites (n = 5) left of dashed horizontal line are minimally DBT-enriched sites. Sites (n = 13) right of dashed horizontal line are highly DBT-enriched sites.
Figure 3.3. Deposition map of total nitrogen in 2014 snowpack

Interpolated loads of total nitrogen (TN; mg/m²) to the Athabasca Oil Sands Region in March 2014. Sedimentary VRS-chla enrichment factors and DBT enrichment factors from each study lake are overlain.
Figure 3.4. Deposition map of dissolved inorganic nitrogen in 2014 snowpack

Interpolated loads of dissolved inorganic nitrogen (DIN; mg/m²) to the Athabasca Oil Sands Region in March 2014. Sedimentary VRS-chla enrichment factors and DBT enrichment factors from each study lake are overlain.
Figure 3.5. Deposition map of total phosphorus in 2014 snowpack

Interpolated loads of total phosphorus (TP; mg/m²) to the Athabasca Oil Sands Region in March 2014. Sedimentary VRS-chla enrichment factors and DBT enrichment factors from each study lake are overlain.
Figure 3.6. Deposition map of total dissolved phosphorus in 2014 snowpack

Interpolated loads of total dissolved phosphorus (TDP; mg/m²) to the Athabasca Oil Sands Region in March 2014. Sedimentary VRS-chla enrichment factors and DBT enrichment factors from each study lake are overlain.
Figure 3.7. Deposition map of soluble reactive phosphorus in 2014 snowpack

Interpolated loads of soluble reactive phosphorus (SRP; mg/m$^2$) to the Athabasca Oil Sands Region in March 2014. Sedimentary VRS-chla enrichment factors and DBT enrichment factors from each study lake are overlain.
**Supplemental Table 3.1. Lake names**

Names and alternate names for study sites. Asterisks (*) denote names that have been previously used in academic literature.

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<th>Alternate Name</th>
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</tr>
<tr>
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</tr>
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## Supplemental Table 3.3. Summary of kriging settings


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**Supplemental Table 3.4. Estimated spatial extent of nutrient deposition**

Estimated spatial extent (km²) of interpolated spring-time snowpack loadings (mg/g²) of dissolved inorganic nitrogen (DIN) 1978, DIN 2014, total nitrogen (TN) 2014, soluble reactive phosphorus (SRP) 2014, total dissolved phosphorus (TDP) 2014, and total phosphorus (TP) 2014 obtained by geostatistical interpolation of measured spring-time snowpack loadings using ArcGIS Geostatistical Analyst software.

<table>
<thead>
<tr>
<th>Net Loading (mg/m²)</th>
<th>Area (km²)</th>
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<tbody>
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<td>DIN 1978</td>
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</tr>
<tr>
<td>0 – 3.4</td>
<td>25.88</td>
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<td>13.6 – 17.0</td>
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<td>17.0 – 20.4</td>
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<td><strong>Total:</strong></td>
<td><strong>23759.05</strong></td>
</tr>
</tbody>
</table>

| DIN 2014            |            |
| 0 – 3.4             | 0          |
| 3.4 – 6.8           | 0          |
| 6.8 – 10.2          | 2470.22    |
| 10.2 – 13.6         | 37274.39   |
| 13.6 – 17.0         | 3050.2     |
| 17.0 – 20.4         | 3.66       |
| **Total:**          | **42798.47**|

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<th>Net Loading (mg/m²)</th>
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<td><strong>Total:</strong></td>
<td><strong>42798.47</strong></td>
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</table>

| TDP 2014            |            |
| 0.0 – 0.2           | 0          |
| 0.2 – 0.4           | 38168.28   |
| 0.4 – 0.6           | 4218.9     |
| 0.6 – 0.8           | 323.21     |
| 0.8 – 1.0           | 77.79      |
| 1.0 – 1.2           | 10.29      |
| **Total:**          | **42798.47**|

| TP 2014             |            |
| 0.0 – 0.2           | 0          |
| 0.2 – 0.4           | 38168.28   |
| 0.4 – 0.6           | 4218.9     |
| 0.6 – 0.8           | 323.21     |
| 0.8 – 1.0           | 77.79      |
| 1.0 – 1.2           | 10.29      |
| **Total:**          | **42798.47**|
Supplemental Table 3.5. Comparisons of kriging range means and measured means


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<th>Kriging Range (mg/m²)</th>
<th>Kriging Range Mean (mg/m²)</th>
<th>Sampled Mean (mg/m²)</th>
<th>Difference from Sampled Mean</th>
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Supplemental Table 3.6. Correlations between five-year averaged VRS-chla Z scores and five-year averaged precipitation data

Results from Pearson correlations between VRS-chla Z scores (averaged over five-year intervals) from highly and minimally DBT-enriched lakes and annual and seasonal AOSR precipitation (averaged over the same five-year intervals).

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Supplemental Figure 3.1. Sediment core dating

Downcore profiles of supported ($^{226}$Ra activity; blue dashed line) and total $^{210}$Pb activity (red circles; ± 1SD). Downcore profiles of $^{137}$Cs activity (yellow triangles; ± 1 SD), and age-depth models (black and grey circles) for 23 sediment cores (A-W). Black circles represent constant rate of supply (CRS)-inferred dates (± 2 SD). Grey circles represent extrapolated dates. Age-depth models were developed using the depth midpoint of sediment intervals, the CRS-inferred age, and polynomial regression (second, third, or fourth-order) with intercept set to the time of coring. The yellow star overlain on the CRS dates denotes the depth of the 1963 $^{137}$Cs peak. Profiles are ordered by year of core collection.
Supplemental Figure 3.2. Snowpack sample sites

Maps of sites in the Athabasca Oil Sands Region where snowpack samples were collected in January 1978 and March 2014.
Supplemental Figure 3.3. Five-year averages of mean annual and seasonal temperature and precipitation data

Historical temperature (station no. 3062696) and precipitation (station no. 3062693) data for Fort McMurray obtained from Environment Canada’s Adjusted and Homogenized Canadian Climate Data website (www.ec.gc.ca/dccha-ahccd) dating back to 1916 and 1920, respectively.
Supplemental Figure 3.4. Five-year averages of VRS-chla Z scores

Five-year average VRS-chla Z scores from (A) minimally and (B) highly DBT-enriched sites.
Supplemental Figure 3.5. VRS-chla profiles

Downcore VRS-chla concentrations for each site calibrated to include diagenetic processes arranged by DBT enrichment factor. (A) Profiles and breakpoints for highly DBT-enriched lakes suitable for breakpoint analysis (n = 13), (B) profiles and breakpoints for minimally DBT-enriched lakes suitable for breakpoint analysis (n = 5), and (C) profiles for minimally (Big Peter Pond), highly (RAMP 175, RAMP 226, and 2014-B), and undetermined (RAMP 227) DBT-enriched lakes where breakpoint analysis is not applicable. RAMP 175, RAMP 226, and 2014-B do not demonstrate stable baselines, Big Peter Pond has one extreme point in recent sediments that erroneously impacts timing of a breakpoint, and RAMP 227 does not have a long enough DBT dataset to calculate a DBT-enrichment factor.
Supplemental Figure 3.6. Deposition map of dissolved inorganic nitrogen in 1978 snowpack

Interpolated loads of dissolved inorganic nitrogen (DIN; mg/m²) to the Athabasca Oil Sands Region in January 1978.
Chapter 4

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

4.1 Abstract

Aerially transported pollution (i.e., industrial contaminants and biologically available nitrogen) from the industrial development of the bituminous sands in the Athabasca Oil Sands Region (AOSR) may threaten the water quality and community structure of the region’s lakes. These environmental threats are further compounded by the changing climate (i.e., increased temperatures and reduced moisture). Since environmental monitoring only began ~30 years after the initiation of the region’s bitumen-based industry (~1967), a paleolimnological approach is required to better document pre-disturbance conditions and to determine how the lakes have changed, if at all, in response to environmental stressors. Our “top-bottom” study of dated sediment records compares pre-disturbance (~1850) and present-day 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 collective changes in diatom communities are minor. Given the shallow nature of our study lakes, most sites unsurprisingly demonstrate assemblage transitions among benthic taxa. At all sites, including lakes with marked changes, we find no evidence of a biological response to airborne inputs from industry. Instead, the diatom assemblages are mainly affected by climate warming, with responses mediated by lake-specific characteristics. We infer that the collectively muted biological responses may be due to naturally high nutrient levels and/or a strong resistance to change that is a common phenomenon in many shallow lakes. Despite their observed resiliency, the algal communities in the region’s lakes likely
remain vulnerable to the multiple, ongoing stressors in the AOSR, with potential future threshold-type responses as recorded in other shallow lake regions.

4.2 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 bitumen deposit is the largest of three 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 2015a). The environmental concerns that accompany this large industry 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).

Airborne industrial contamination from upgrader stacks and fossil-fueled 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, reaching aquatic environments at least 90 km from the main industrial sites (Kurek et al. 2013a). The industrial airborne contamination includes a suite of potentially toxic compounds (e.g., Kelly et al., 2010, 2009; Kirk et al., 2014). For example, dibenzothiophenes (DBTs) are a group of polycyclic aromatic compounds (PACs), which 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 airborne industrial contamination.
(Jautzy et al. 2013; Kurek et al. 2013a; Summers et al. 2016). Several studies exploring atmospheric contamination in the AOSR find evidence of 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 fertilize ecosystems and alter biotic communities, is known to be emitted in large quantities by industrial development of the oil sands (102 tonnes of dissolved inorganic nitrogen 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 (as evaporative processes outpace precipitation; Schindler and Donahue, 2006) characterize the AOSR. For example, over the past ~100 years, mean annual temperatures in the prairie provinces have increased by ~1-4 °C while total annual precipitation has declined by ~25 % (Schindler and Donahue 2006). Previous paleolimnological studies investigating the effects of local industrial activity in the AOSR and downwind originally focused on the effects of acid deposition from industrial emissions of nitrogen and sulphur oxides (Hazewinkel et al. 2008; Curtis et al. 2010; Laird et al. 2013). While finding no consistent evidence of widespread surface water acidification, the 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. Other recent paleolimnological studies investigating biological responses to environmental stressors in the AOSR have found an overriding influence of climate warming, instead of industrial inputs, in structuring aquatic communities (Kurek et al. 2013a; Laird et al. 2017; Mushet et al. 2017; Summers et al. 2017).

Direct long-term monitoring data are largely lacking for the AOSR given that environmental monitoring focusing on the effects of oil sands industry only began in 1997, ~30 years after the beginning of the commercial bitumen industry in the region (Dowdeswell et al.
Thus, paleolimnological methods, which use proxies naturally integrated and preserved in lake sediments (Smol 2008), are required to understand the historical environmental conditions of these aquatic systems. In this study, we use diatoms as the primary proxy to advance our understanding of the effects of industrial airborne pollution (i.e., contaminants and nutrients) and accelerated climate change on aquatic biota within ~110 km of the main area of industrial activity. Subfossil remains of diatoms are the most commonly used biological proxy in paleolimnological studies, and have been widely and effectively used as indicators of historical pH, salinity, nutrient levels, and climate mediated changes, for example (Smol and Stoermer 2010).

Previous studies have used diatoms to track changes in the AOSR in specific lakes of interest (e.g., Curtis et al., 2010; Hazewinkel et al., 2008; Laird et al., 2017; Summers et al., 2017). However, to explore the regional trends in diatom assemblage change across a broad spatial scale, we use a “top-bottom” paleolimnological approach (i.e., diatom assemblages in recent sediments compared to assemblages preserved in pre-impact sediments; Smol 2008) including 18 shallow study lakes situated within ~110 km of the main industrial activities. Sites have been selected to represent shallow, isolated lakes typical of the region (Natural Regions Committee 2006). Our research questions include: (1) Do modern diatom assemblages differ from pre-disturbance assemblages? (2) Has aerially transported industrial pollution (i.e., contaminants and bioavailable nutrients) affected diatom assemblage compositions? (3) What environmental variables best explain the patterns of diatom assemblage change and distribution among shallow AOSR lakes?

4.3 Methods

4.3.1 Study area

The AOSR is situated primarily in the northeast corner of the Canadian province of Alberta and extends into northern Saskatchewan (Figure 4.1). Decades of bitumen-based industry
and development have industrialized 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 (Natural Regions Committee 2006). The region’s surficial geology is composed mainly of coarse glaciofluvial 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; 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).

### 4.3.2 Site selection

The 18 lakes used in this top-bottom paleolimnological survey are a subset of typical lakes (i.e., shallow, closed basins; Natural Regions Committee, 2006) in the AOSR (Table 4.1; Figure 4.1). Given that this project explores aerially transported industrial contamination and nutrient deposition, the lakes selected for this study are isolated basins that are not directly part of a larger river system, such as the Athabasca River, and have catchments that were relatively physically undisturbed by industry at the time of coring. The sites are located at varying distances (within ~110 km) and in all directions from the main area of industrial bitumen-related activities (Figure 4.1).

### 4.3.3 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 land and were accessed by
helicopter. Three sediment cores were collected through the ice from the deepest part of each lake using a Uwitec gravitational coring system (www.uwitec.at) and were sectioned using a Uwitec vertical extruder immediately upon returning to the field base. Given that cores were sectioned at 0.5-cm intervals until 20 cm and at 1.0-cm intervals thereafter, the “top” intervals for each lake were 0.5 cm thick and the “bottom” intervals were 1.0 cm thick. Cores were frozen until they were analyzed. 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-α analyses (Paleoecological Environmental Assessment and Research Laboratory (PEARL), Queen’s University, Kingston, Ontario). Replicate cores were retained for additional analyses. Water samples were collected through the ice at each site and were processed 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 (Supplemental Table 4.2).

4.3.4 Radioisotopic analyses

As reported in Summers et al. (2016, 2017), measurements of radioisotope activity and age modelling were completed by Flett Research Ltd. $^{210}$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 intervals were estimated by fitting a polynomial of the lowest order with a reasonable fit to the measured sediment, and setting the intercept to the time of coring (Summers et al. 2016, 2017).

4.3.5 The “top-bottom” paleolimnological approach
A top-bottom paleolimnological approach (reviewed in Smol, 2008) was used to compare two temporal “snap shots” (~1850 and present) of diatom communities to infer environmental conditions 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 provide information about the specific timing of any identified historical changes (Smol 2008). The uppermost available sediment interval 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 (Supplemental Table 4.1). Sediments below 0-0.5 cm were only used if no sediments from the original top interval were available. Given that chronologies for all of the sediment cores were previously established, $^{210}\text{Pb}$-estimated dates of sediment deposition were used to select the interval from each lake that was most representative of ~1850 (available intervals ranged from ~1825 to ~1897). These intervals were used as the “bottom” or “pre-disturbance” intervals for each lake, and provided an integrated sample of conditions preceding industrial oil sands development (~1970) and anthropogenic climate change in this region (Supplemental Table 4.1).

4.3.6 Diatom sample preparation and analyses

Diatom samples were prepared from either wet or freeze-dried sediment generally following the standard methods outlined in 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 hours. The samples were allowed to settle for 24 hours 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× magnification using a Leica DMR light microscope fitted with differential
interference contrast and using oil immersion techniques. 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 interval.

4.3.7 Numerical analyses

4.3.7.1 Trends in environmental variables

A principal components analysis (PCA) was performed on modern environmental data (including physical, chemical, and industrial data) collected from the 18 study sites to examine the variation of sites with respect to environmental conditions. 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 transform any non-normally distributed variables as necessary (using square root, log, or log+1 transformations). 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. Like all ordinations in this study, 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.

4.3.7.2 Trends in diatom assemblages

For most statistical analyses using diatom assemblage data, unless stated otherwise, rare taxa (not exceeding 2% in one interval) were excluded. Additionally, relative abundances of diatoms were square root transformed in most statistical analyses to reduce the dominance of abundant 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 from pre-disturbance times to 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 sedimentary intervals in this scenario. Modern assemblage data were actively included in the DCA ordination while pre-disturbance assemblage data were included passively (i.e., as supplementary data). The DCA was performed using 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 from the pre-disturbance to the present-day (ΔDCA1 and ΔDCA2) were determined by subtracting bottom interval DCA axis 1 and 2 site scores from the corresponding top interval 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 determine if there was a significant, global (across all study lakes) difference in diatom assemblages from pre-disturbance times to present-day, a one-way analysis of similarity (ANOSIM) was completed on species data using the Vegan package (Oksanen et al. 2012) in R. An ANOSIM is a nonparametric analysis that tests the null hypothesis of no difference between species assemblages in pre-defined groups, which were bottom and top sediment intervals in this case. The ANOSIM ranked bottom and top species data in a matrix of Bray-Curtis similarity values and computed a test statistic (R), which quantified whether the pair-wise similarity was
greater within or between groups. Significance was calculated via 999 permutations (Clarke 1993).

To explore trends at 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 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 of diatom compositions between the two discrete 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 were rarified to a common sum to account for variation in the total number of diatoms counted in each interval (Birks 2012a). Species richness is expressed as the number of taxa in a sample, while Hill’s N2 diversity incorporates richness and evenness in a sample, and is relatively easy to interpret ecologically (Birks 2012a). Finally, percent relative abundances of common taxa or groups of taxa were included in scatter plots with a 1:1 line to assess and compare specific compositional changes between modern and pre-disturbance assemblages.

4.3.7.3 The relationship between environmental factors and diatom communities

A constrained ordination was performed (using CANOCO version 5.0 software) to investigate the relationships between the measured environmental variables and the distribution of modern diatoms from the 18 study lakes. The initial DCA performed in this study identified a gradient length of 3.9 SD; therefore, a unimodal ordination method (a canonical correspondence analysis; CCA) was deemed acceptable for additional analysis (Birks 2012a). The species data from top intervals were included actively in the ordination with modern environmental data. The species data from bottom intervals were included passively because associated pre-disturbance
environmental conditions were unknown. As in the PCA, environmental variables were tested for normality and transformed as necessary to attain an approximately normal distribution, and correlations among the variables were calculated in a Pearson correlation matrix with a Bonferroni correction (see PCA method details above; Supplemental Table 4.2). To obtain the most parsimonious set of potential explanatory variables for inclusion in the constrained ordination, variables with correlation coefficients (r) exceeding 0.6 were grouped and one representative variable from each group was used (Supplemental Table 4.2). A global permutation test was run on all candidate environmental variables to prevent the inflation of Type I errors (Blanchet et al. 2008). After the global permutation test was performed and deemed significant, forward selection was applied to sequentially test each environmental variable as a predictor of the diatom response, and determine which environmental variables should constrain the ordination (Birks 2012b). The significance of environmental variables was determined via 999 Monte-Carlo permutation tests. Bonferroni corrections were applied and an alpha (α) significance level of 0.05 (p_B < 0.05) was required for environmental variables to be considered significant predictors.

4.4 Results

4.4.1 Core chronologies

Radioisotopic activity profiles and age-depth models from all the lakes included in this study are shown in Supplemental Figure 4.1. Most of the chronologies have been previously published in Summers et al. (Summers et al. 2016). ²¹⁰Po (a proxy for total ²¹⁰Pb) in the majority of cores exhibited a typical exponential decay curve with some irregularity, although some cores showed depletions of ²¹⁰Po in the surface sediments. Sites 274 and RAMP 271 sediment cores only extended to ~1897 and ~1880, respectively. Thus, those intervals, although more than 30 years younger than ~1850, were selected as the closest representations of ~1850. Pre-disturbance sediment intervals selected from the other lakes ranged from ~1825-1865.
4.4.2 Top-bottom sediment interval 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; Supplemental Figure 4.1) and was thus inappropriate for inclusion. Further, 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 intervals and were therefore also excluded from the study. On average, based on our estimated $^{210}$Pb dates, the bottom sediment intervals in our study represent approximately six years while the top intervals represent approximately one year of diatom accumulation. The pre-disturbance intervals with no/low diatoms were high in siliciclastic materials while some 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 to make a regional assessment, the sites cover a sufficient range of industry contamination to justify an exploratory survey of lakes (DBT EFs 0.7 – 37.4).

4.4.3 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 (Supplemental Table 4.2). The sources of the measurements for each environmental variable are included in Supplemental Table 4.2 and ranges are included in Table 4.1. Sixteen environmental variables were actively included in the PCA while six variables were included passively (Supplemental Table 4.2). The passive variables included specific conductivity and five ions (chloride, calcium, magnesium, potassium, and sodium) that were 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 interval (top [DBT]), and sulphate (SO\textsubscript{4}\textsuperscript{2-}) from the water column. DBT EFs were calculated in Summers et al. (2016) and compare DBT concentrations in modern sediment intervals to concentrations in sediment intervals 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 used as metrics of industrial pollution from airborne contaminants (Summers et al. 2016). The top [DBT] is not a measure of industrial impact but is, rather, the combined natural and industrially-sourced DBT concentration in the lake sediments most recently deposited. SO\textsubscript{4}\textsuperscript{2-} 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 (Figure 4.2). The amount of variance explained was similarly distributed between the two main axes, with PCA axis 1 explaining 25 % and axis 2 explaining an additional 23 %. Although total nitrogen loaded the most strongly of all variables onto axis 1 (score = -0.91), the axis 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, which loaded most strongly onto axis 2 (score = 0.74; Figure 4.2).

4.4.4 Trends in diatom assemblages

A total of 311 diatom taxa were identified in the 18 lakes (Appendix A). When a cut-off criterion was applied retaining only taxa exceeding 2 % in at least one interval, 97 taxa remained. The top-bottom relative abundance histogram of these diatom assemblages showed that small, benthic fragilarioid taxa were common to most sedimentary intervals, and in fact were dominant in the majority of both top and bottom samples (Figure 4.3). NE13 was an exception. NE13 is a
marl lake characterized by many benthic taxa (e.g., *Anomoeoneis* + *Brachysira* summed, cymbelloid taxa, and long *Navicula* taxa (e.g., *Navicula cryptocephala*, *Navicula cryptotenella*; Figure 4.3).

Overall, diatom communities in the study lakes changed very little. A plot of the DCA sample scores (Figure 4.4) highlighted the differences among assemblages with modest separation of sites (except for the marl lake, NE13). DCA axis 1 explained 13.5 % of the variance and axis 2 explained an additional 9.9 % (Figure 4.4). Examining the diatom taxonomic loadings on the DCA axes (i.e., species scores), *Aulacoseira* taxa and pennate planktonic *Fragilaria* were most important to DCA axis 1 (Supplemental Figure 4.2, Supplemental Table 4.2). The trajectories of overall assemblage change through time (i.e., differences between the modern and the pre-disturbance sample scores from each lake) were relatively small and recorded no consistent directional changes. Diatom assemblages among the lakes were more similar in the past and diverged to their current states (Figure 4.4). Some individual lakes demonstrated notable changes between pre-disturbance and present-day assemblages (e.g., 2014-D, 2014-Z, and Pushup; Figure 4.3). Sites 2014-D, 2014-Z, and Pushup demonstrated clear taxa turnover from high relative abundances of small, benthic fragilarioid taxa to high relative abundances of centric planktonic taxa (Figure 4.3). Diatom assemblage changes in other sites were less pronounced (Figure 4.4), 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 (Figure 4.4). A global ANOSIM of the 18 shallow sites detected differences between pre-disturbance and modern diatom assemblages that were significant but not pronounced (one-way ANOSIM $R = 0.078$, $p = 0.01$, $n = 999$ permutations). The three lakes with the largest trajectories of change (i.e., 2014-D, 2014-Z, and Pushup) substantially affect the ANOSIM results, which are
not significant (one-way ANOSIM $R = 0.052$, $p = 0.10$, $n = 999$ permutation) when the three lakes are excluded.

An examination of trends at the species level highlighted distinct differences between pre-disturbance and present-day relative abundances of the dataset’s most common diatom taxa (Figure 4.5). 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 (Figure 4.5). Notable relative abundance increases in the modern samples are evident in plots of *Achnanthidium 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 (Figure 4.5 plots).

Scatter plots of pre-disturbance and modern species richness and Hill’s N2 diversity (rarified $n = 303$) showed no directional trend among the 18 lakes (Figure 4.5). 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 (Figure 4.5).

### 4.4.5 Environmental variables and their relation to diatom assemblages

Data screening procedures for the CCA reduced the number of environmental variables to 11 variables, namely depth, surface area, DIC, total nitrogen, total phosphorus, chlorophyll-$a$, ammonia, sulphate, silicon dioxide, DBT enrichment factor, and DBT concentration in the top sediment interval. A trial CCA identified top interval DBT concentration as an outlier variable (leverage exceeding 5×; Hall and Smol, 1992); thus, the top interval DBT variable was removed from the final CCA. The final CCA with a global permutation test and interactive forward
selection (Blanchet et al. 2008) identified DIC as the only variable explaining a significant ($p_B = 0.02$) amount of variation in the diatom assemblage (data not shown).

Temporal assemblage changes in richness, diversity, DCA 1 and 2 axis scores ($\Delta$DCA1 and $\Delta$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 ordered histograms did not show consistent, directional patterns of biological change (data not shown). It is notable that these metrics of assemblage change plotted along gradients of DBT EF and distance to nearest major industrial source (Landis et al. 2017), proxies for aerial industrial oil sands impact, showed no clear directional trends (data not shown).

4.5 Discussion

When diatom communities in the study lakes are examined as a group, there is little change from pre-disturbance times to the present day. However, when temporal changes are examined in individual sites, some diatom communities change markedly while others change minimally. The absence of directional patterns in metrics of temporal assemblage change (i.e., top-bottom comparisons of species richness, Hill’s N2 diversity, DCA axis 1 and 2 scores, and the relative abundances of the most common taxonomic groups) along gradients of measured environmental variables (data not shown) indicates that the measured variables may not have had a substantial effect on the diatom assemblage compositions through time in most lakes. As discussed in Rühland et al. (2014), the absence of clear trends through time along the environmental gradients may be due to the relatively small number of lakes sampled, insufficient gradient lengths (i.e., ranges) of measured environmental variables, little temporal change in the environmental variables, and/or diatom responses to factors not measured in this study (e.g., climate-induced changes, which could be occurring given that the ASOR climate is changing). Further, water chemistry measures represent conditions at the time of coring while sediment samples are an integrated representation of conditions throughout the basin over a longer period.
of time. These different temporal scales may explain the absence of clear trends in metrics of assemblage change along environmental gradients.

Importantly, the absence of patterns in diatom response along industrial gradients (i.e., DBT EF, distance to nearest major industrial source) suggests that airborne pollution, including contaminants and bioavailable nitrogen, from local oil sands industry does not consistently or predominantly affect the mostly benthic alga of these lakes. The lack of a biological response to deposition of industrial pollution is consistent with findings from other studies across the AOSR (e.g., Kurek et al., 2013; Laird et al., 2017; Mushet et al., 2017; Summers et al., 2017) and may be due to naturally high nutrient levels and/or the shallow depths and available habitats in the lakes. If our study lakes were historically rich in nutrients, which is plausible given the current high nutrient levels and phosphorus limitation in the majority of our sites (Table 4.1) and the known nutrient-rich histories of some lakes in Alberta (Blais et al. 2000; Bayley et al. 2007), the addition of growth-stimulating nitrogen would not be expected to yield marked changes in biological assemblages. Additionally, in some scenarios, biotic communities in shallow lakes are known to resist change and display stability in response to environmental stressors, including nutrient inputs (Scheffer 1998). The absence of consistent, marked biological changes in our study lakes may suggest that ecological thresholds of industrial pollution, for example, have not been surpassed.

In this study, a CCA indicated that diatom distribution was best explained by DIC. However, the absence of a directional pattern in the metrics of temporal assemblage change arranged along a gradient of DIC (data not shown) indicates that the magnitude of change in DIC concentrations since the pre-disturbance period was likely not sufficient to shape the diatom assemblages through time. Given that modern DIC concentrations span a relatively large gradient across our 18 study lakes (2.1-76.2 mg/L), and that lakes with the lowest DIC concentrations (265 DIC =2.1 mg/L; RAMP 226 DIC = 2.7 mg/L) as well as the lakes with the highest DIC
concentrations (NE13 DIC = 76.2 mg/L; Kearl = 41.9 mg/L) did not experience large assemblage changes between the two time periods (ΔDCA1 for 256 = -0.2; RAMP 226 = -0.03; NE13 = -0.6; Kearl = -0.5), the CCA’s identification of DIC is likely due to differences in DIC concentrations across space rather than the changes in DIC concentrations over time.

Instead of responding to industrial pollution, the muted biological community changes in most of our study sites are consistent with known responses to climate change (discussed below). Observations from Environment and Climate Change Canada’s (ECCC) Fort McMurray weather station (temperature station ID: 3062696; precipitation station ID: 3062693; 56°36’00.0” N, 111°13’12.0” W; www.ec.gc.ca/dccha-ahccd), the most representative ECCC station for the AOSR, show that 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). Given evidence of climate change, no directional patterns in metrics of assemblage change along gradients of physical and chemical lake characteristics, the nature of the changes consistent (although subtle) with climate-mediated changes, and several studies pointing to regional climate change as the principal driver affecting the biota of lakes in the AOSR (e.g., Kurek et al., 2013; Laird et al., 2017; Mushet et al., 2017; Summers et al., 2016), it is plausible that biological responses in our study lakes are influenced by climate change.

The shallow depths of our study lakes mostly preclude high abundances of planktonic taxa and thus, the observed changes are mainly, but not exclusively, among benthic diatoms. In the majority of our shallow sites, we find changes from pre-disturbance assemblages dominated by *Staurosira construens* and *Staurosirella pinnata* (small, epilithic, r-strategist, fragilarioid) taxa (Lotter and Bigler 2000) to modern assemblages that are still dominated by these fragilarioid taxa but with increased abundances of other benthic taxa with more complex environmental
preferences (e.g., naviculoid taxa, *Nitzschia*; Figures 4.3, 4.5). 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). As such, the diatom assemblage transitions we record in the majority of our study lakes are expected responses to a warming climate given the shallow depths of the sites (Rühland et al. 2014; Summers et al. 2017).

While the majority of our study lakes demonstrate similar assemblage changes among benthic taxa (i.e., shifts from benthic, r-strategist fragilarioioid taxa to specialized benthic taxa), several sites increase in abundances of planktonic taxa in modern assemblages. We find striking assemblage shifts from a high dominance of benthic fragilarioioid 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; Figure 4.3). We also observe changes in RAMP 268 (1.8 m) from a benthic fragilarioioid-dominated (~ 40 %) pre-disturbance assemblage to a present-day assemblage comprised of over 20 % *Fragilaria tenera*, which is a pennate planktonic species. These assemblage changes are consistent with expected responses to warming and longer ice-free periods in deeper lakes (Rühland et al. 2015). 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), for example, attribute high modern abundances of small, cyclotelloid taxa, including *D. stelligera*, to warming-induced decreases in vertical mixing and increases in thermal stability. However, in our relatively shallow sites, strong and prolonged thermal stratification would likely be difficult to establish. As such, although some heightened thermal stability may contribute to the assemblage shifts, it is likely that the increased abundances
of both centric and pennate planktonic taxa are primarily due to warming-induced extensions in the open water seasons and related changes that enable planktonic diatoms to thrive in these relatively shallow lakes.

The differences in assemblage responses to climate change in our shallow lakes (i.e., some lakes shift to more complex benthic assemblages while other lakes shift 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 fragilariod 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 was 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. These dissimilar assemblage responses may, for example, be due to differences in lake morphology driving varied availability of benthic habitat; however, the causes are currently unknown with only a top-bottom approach.

Although the majority of diatom responses observed in this study are qualitatively linked to climate change, 2014-D is an exception. 2014-D is a shallow lake (0.9 m) that demonstrates a marked turnover from small, benthic fragilariod taxa (> 85 %) in its pre-disturbance assemblage to over 60 % abundance of Aulacoseria granulata in its modern assemblage. The nature of this large change is inconsistent with known responses of shallow lakes to climate change (e.g. declines in the relative abundances of heavily silicified Aulacoseira taxa with weakened vertical mixing). Given that 2014-D is very high in total phosphorus (TP; 2014-D = 341 μg/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 are due to increasing nutrient levels. However, 2014-D has no known sources of nutrient inputs and historical nutrient levels are unknown. Despite the unknown causes, which cannot be determined with only a top-bottom approach.
paleolimnological approach, a potential nutrient-driven community change at 2014-D is not attributed to industrial inputs of biologically available nitrogen because the site is over 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). More detailed analyses are required to better understand the nature and timing of the diatom shifts in this shallow lake.

4.6 Summary and conclusions

Overall, the diatom assemblages in our shallow AOSR study lakes record minor changes since ~1850, despite substantial industrial pollution and a warming and drying climate. The collectively muted biological response may be due to naturally high nutrient levels and/or the commonly observed resilience of shallow lakes. Although some study sites track marked community change between bottom and top sedimentary intervals, the changes are clearly not linked to deposition of industrial contaminants or biologically available nitrogen. Instead, the majority of diatom responses, although variable, are broadly consistent with assemblage changes linked to climate warming. The differences in the nature and magnitude of the assemblage responses among seemingly comparable sites suggests the individual lake changes are modified by site-specific physical, chemical, and geographic lake characteristics. Our research corroborates the findings of other paleolimnological studies in the AOSR and farther downwind that find biological community responses primarily affected by climate change, rather than industrial contaminant and nutrient deposition (Hazewinkel et al. 2008; Curtis et al. 2010; Kurek et al. 2013a; Laird et al. 2013, 2017; Summers et al. 2016; Mushet et al. 2017). Downcore analyses of the diatom assemblages in lakes with high inputs of airborne industrial pollution and large taxonomic changes between pre-disturbance and modern assemblages will further elucidate the effects of ongoing multiple stressors on the lakes in the AOSR. The findings of this study contrast the relative biological stability found in the region’s lakes with ongoing vulnerability of the lakes as the region’s stressors are forecast to persist.
4.7 Acknowledgements

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4.8 References


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Oxford, United Kingdom


### Table 4.1. Summary of physical, chemical, and industry-related characteristics

Physical, chemical, and industry-related characteristics for each of the 18 lakes. 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 2003-2007, as no data were collected from the site when the lake was cored. Lakes with TN:TP mass ratio < 14 are nitrogen limited (Downing and McCauley 1992).
Figure 4.1. Map of study area

Locations of the 23 cored study lakes, two 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 (see Figure 4.3) 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 intervals (see text for details).
Figure 4.2. Principal components analysis (PCA)

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 interval DBT concentrations (Top [DBT]), and sulphate (sulph) were included actively and are marked in solid black. Specific conductivity (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 4.1).
Figure 4.3. Relative abundances of diatom taxa in modern and pre-disturbance sediments

Relative abundances of select diatom taxa in the present-day (top of each shaded pair) and pre-disturbance (bottom of each shaded pair) sediment intervals from the 18 study lakes. Where applicable, the number of taxa included in the complexes and summed groups are indicated in parenthesis following the group name. Taxa included in complexes and summed groups are listed in Supplemental Table 4.4. Lakes are arranged from top to bottom in order of increasing dibenzothiophene enrichment factor (DBT EF; Table 4.1).
Figure 4.4. Detrended correspondence analysis (DCA) site scores

Detrended correspondence analysis (DCA) plot including site scores from the first two ordination axes. Black circles mark the actively included present-day 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 present-day assemblages indicates the magnitude and direction of assemblage change over time at each site. Site names are abbreviated as in Table 4.1.
Figure 4.5. Scatter plots of assemblage metrics and relative abundances of select diatom taxa

Scatter plots of assemblage metrics and select common diatom taxa from the pre-disturbance (x axis) and modern (y axis) sediment intervals of the 18 Athabasca Oil Sands Region (AOSR) study lakes. The benthic fragilarioid 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 radios*, *Navicula rhynchocephala*, and *Navicula vulpina*. Black circles denote sites with coring depths ≤ 2 m. Grey circles denote sites with coring depths > 2 m. Where applicable, the number of taxa included in the complexes and summed groups are indicated in parenthesis following the group name. A 1:1 line is included. Samples that plot above the 1:1 line indicate greater values in the present-day assemblages than in the pre-disturbance assemblages.
### Supplemental Table 4.1. Summary of geographical and core information

Geographical lake characteristics and core information for each of the 18 lakes.

<table>
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<th>Lake Name</th>
<th>Latitude (DD)</th>
<th>Longitude (DD)</th>
<th>Distance to Nearest Industrial Source (km)</th>
<th>Top Interval Mid Year (CE)</th>
<th>Top Interval Mid Depth (cm)</th>
<th>Bottom Interval Mid Year (CE)</th>
<th>Bottom Interval Mid Depth (cm)</th>
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### Supplemental Table 4.2. Summary of environmental variables and their transformation for inclusion in the ordinations

Environmental variables included in the ordinations, the sources of the measurements, and the transformations applied to achieve approximately normal distributions. Variables in parentheses in the “Inclusion in CCA” column are the representative variables.

<table>
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<th>Variable</th>
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<th>Transformation</th>
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<th>Inclusion in CCA</th>
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<td>Map</td>
<td>Log10</td>
<td>Active</td>
<td>Included</td>
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<td>DBT EF</td>
<td>Sediment</td>
<td>Log10</td>
<td>Active</td>
<td>Included</td>
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<tr>
<td>Top interval [DBT] (ng/g dry wt.)</td>
<td>Sediment</td>
<td>Log10+1</td>
<td>Active</td>
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<tr>
<td>pH</td>
<td>Water Sample</td>
<td>None - already log10-transformed</td>
<td>Active</td>
<td>Not Included (DIC)</td>
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<td>TN (µg/L)</td>
<td>Water Sample</td>
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<td>Active</td>
<td>Included</td>
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<td>TP (µg/L)</td>
<td>Water Sample</td>
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<td>Active</td>
<td>Included</td>
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<td>NH₃ (mg/L)</td>
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<td>Included</td>
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<td>DOC (mg/L)</td>
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<td>Specific Conductivity (µg/L)</td>
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<td>Cl⁻ (mg/L)</td>
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<td>K⁺ (mg/L)</td>
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<td>Na⁺ (mg/L)</td>
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<td>Log10</td>
<td>Passive</td>
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Supplemental Table 4.3. Taxa represented by numbers in the DCA

Taxa represented by numbers in the detrended correspondence analysis (DCA) plot including species scores from the first two ordination axes.

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Supplemental Table 4.4. Taxa included in histogram groups

Taxa included in the complexes and summed groups shown in the histogram of diatom relative abundances (Figure 4.3).

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Supplemental Figure 4.1. Sediment chronologies

Downcore dating profiles of the 23 lakes that were included and/or considered for inclusion in the study. Profiles are arranged by the year the sites were cored (indicated in parentheses after the site name). Decay curves of total $^{210}$Po activity, which was used as proxy for total $^{210}$Pb activity, are shown by black circles and the associated error bars ($\pm$ 1 SD). Background activities, also known as supported $^{210}$Pb, were measured using $^{226}$Ra as a proxy and are plotted on the same scale as total $^{210}$Po activity (black dashed line). Constant rate of supply (CRS)-inferred dates are shown by grey circles and the associated error bars ($\pm$ 2 SD).
Supplemental Figure 4.2. Detrended correspondence analysis (DCA)

Detrended correspondence analysis (DCA) plot including species scores from the first two ordination axes. Taxa are indicated by numbers that are listed in Supplemental Table 4.3. Assemblage tops were included actively in the DCA while bottoms were included passively.
Chapter 5

General discussion and conclusions

5.1 General discussion and conclusions

As the AOSR becomes warmer, drier (Schindler and Donahue 2006), and more industrialized (Alberta Energy Regulator 2015), the region’s lakes and ecosystems are increasingly vulnerable to stressors and potential degradation. Identifying the cumulative effects and the relative contributions of stressors is essential in establishing realistic and effective management strategies (Smol 2010). Without monitoring to recognize environmental conditions preceding industrial development and anthropogenic climate change (Dowdeswell et al. 2010), a paleolimnological approach is necessary for determining baseline conditions, tracking ecosystem change over time, and deciphering drivers and their roles (Smol 2008). At the outset of this thesis, paleolimnological studies in the region characterized the deposition of contaminants from industrial oil sands activities (Hall et al. 2012; Wiklund et al. 2012; Jautzy et al. 2013; Kurek et al. 2013a), assessed surface water acidification (Hazewinkel et al. 2008; Curtis et al. 2010; Laird et al. 2013), identified potential increases in primary production (Hazewinkel et al. 2008; Curtis et al. 2010; Kurek et al. 2013a; Laird et al. 2013), and attributed biological change in some lakes as to an overriding influence of climate change (Kurek et al. 2013a; Laird et al. 2013). Further analyses of aquatic biota proximal to the main area of industrial activity were required to investigate the relative roles of aerially transported industrial pollution (i.e., contaminants and bioavailable nutrients) and climate change in structuring the biological communities in the region’s shallow lakes. Additionally, the extent, timing, and drivers of increased primary production had not been explicitly investigated. Addressing these knowledge gaps was the aim of this thesis. My research aids in understanding if and how climate change and industrial pollution, together and/or individually, have affected the aquatic biota in the AOSR’s many shallow lakes.
This knowledge is timely in providing information for effective management of the region’s multiple, ongoing stressors.

This thesis includes three studies. Chapter 2 uses a multi-proxy downcore approach, including diatom, chironomid, and cladoceran communities, to assess temporal changes since ~1940 in a typical AOSR lake receiving substantial inputs of airborne pollution from the regional bitumen-based industry. The gradual and relatively muted transitions in assemblages of bioindicator groups provide no evidence of a clear response to aerially transported industrial pollution. Rather, the assemblage changes are attributed to warmer conditions (inferred by chironomids) and related increases in habitat complexity (i.e., more abundant and diverse substrates for colonization; inferred by diatoms and cladocerans). The findings of this study indicate that changes in climate, potentially through mechanisms such as decreased ice cover, are important regulators of aquatic habitat availability and biotic structure across multiple trophic levels in the study lake, and potentially in other shallow lakes across the AOSR. Despite marked changes in environmental conditions and substantial pollutant deposition, the biological communities in a typical AOSR lake downwind of development appear to be relatively resilient to stressors associated with aerial contaminants from industry.

The third chapter of this thesis explores the relative contributions of regional climate change and industrial airborne contamination to trends in aquatic primary production across the landscape. Unlike the subtle and gradual biological changes in Chapter 2, aquatic primary production, tracked using spectrally-inferred sedimentary chlorophyll-\(a\) profiles, increased markedly at all sites, indicating an increase at a regional scale. The timings of increased production were asynchronous across sites. This study also used springtime snowpack data to investigate industrial contributions of nutrients to the AOSR landscape. Elevated levels of biologically available nitrogen surrounding the region’s main industrial facilities (shown with an interpolated nutrient deposition map), indicate industrial sources of potential growth-stimulating
nutrients. Spatial comparisons between the primary production trends in the study lakes and the patterns of nutrient deposition suggest whole lake primary production is not dominantly affected by nutrient deposition from industry. Correlations between production trends and historical climate records suggest warming temperatures are likely the predominant driver of the increases. Like biological community changes found in Chapter 2, the substantial changes in primary production are attributed to climate-driven mechanisms extending and/or improving the growing season. The relatively abrupt and marked changes from stable baselines in inferred primary production trends suggest conditions at the majority of study sites may have surpassed an ecological threshold for stability, and continued increases in aquatic production may be plausible.

Chapter 4 returns to investigating biological responses to stressors in the AOSR with subfossil diatoms. Using a “top-bottom” paleolimnological approach, the chapter characterizes how present-day diatom assemblages are different from assemblages preceding marked anthropogenic climate change and industrial oil sands development (sediments from ~1850), and identifies the environmental variables shaping the temporal changes and spatial differences. The mostly muted assemblage transitions (similar to the diatom community changes identified in Chapter 2), as well as the marked shifts in three sites, are not attributed to deposition of industrial contaminants or nutrients. Instead, as in chapters 2 and 3, the changes are broadly consistent with responses to climate change expected in shallow lakes. Differences in responses among seemingly comparable sites are likely modified by site-specific characteristics (e.g., benthic habitat availability), demonstrating that the pre-disturbance state of a lake importantly affects the type and magnitude of biological response (Rühland et al. 2015). In our study sites, it is possible that the observed biological resiliency to change may be due to naturally high nutrient levels and/or a strong resistance to change that is a common phenomenon in shallow lakes (Scheffer 1998).
Consistent with the results of recent paleolimnological studies in the AOSR and farther downwind (Kurek et al. 2013a; Laird et al. 2013, 2017; Mushet et al. 2017), the three approaches in this thesis consistently find little evidence of clear biological responses to the deposition of industrial contaminants and biologically available nutrients. Instead, the chapters suggest the more dominant role of climate in regulating both primary production and biotic community structure in AOSR lakes. Specifically, the nature of the biological transitions in Chapters 2 and 4, and the correlations to temperatures in Chapter 3, infer that longer and warmer growing seasons, which improve conditions for aquatic organism growth (Adrian et al. 2009), may be the main climate-driven mechanisms causing primary production increases, habitat diversification, and ultimately, changing biological assemblages in AOSR lakes. The gradual and overall muted assemblage transitions in Chapters 2 and 4, compared to the abrupt and marked inferred aquatic primary production changes in Chapter 3, demonstrate that the composition of the biological communities have been more stable and resistant than whole lake primary production levels to the effects of the region’s multiple stressors. Further, the findings of this research show how lake-specific physical, chemical, and landscape conditions can modify responses to an overriding stressor. Simply, the asynchronous increases in inferred primary production, identified in Chapter 3, point to lake-specific characteristics mediating the timing of production increases, while the climate-driven assemblage changes in Chapters 2 and 4 differ in their nature and magnitude among comparable shallow lakes. The differences in the individual lake responses demonstrate that the pre-disturbance state of a lake as well as the individual and combined effects of environmental stressors are important in understanding how aquatic ecosystems change over time.

5.2 Future research

Broadly, this thesis provides evidence of increased primary production and biological community changes in AOSR lakes that are primarily in response to climate change rather than
industrial airborne pollution. However, interesting and important research questions remain. The diatom assemblage changes, identified in Chapter 4, and the DBT enrichment factors, calculated in Chapter 3, provide the foundation for strategic selection of sites for downcore assessments. Detailed, downcore diatom analyses of lakes demonstrating large and small DBT enrichment factors and VRS-chla enrichment factors (as identified in Chapter 3), and large and small taxonomic turnover (as identified in Chapter 4) are underway and are expected to further characterize the timing and nature of the primary producers’ response to the region’s stressors. Ongoing top-bottom analysis is recommended as an efficient approach to identify lakes for further downcore assessment and monitoring. This work will likely identify any biological response to industrial airborne contamination and aid in identifying at-risk lakes and potentially important levels of contamination.

Given that stressors in the AOSR are expected to persist, marked biological change in the region’s shallow lakes is plausible in the future. The findings of this thesis provide a historical record of change that can be continued with strategic monitoring or repeated paleolimnological sampling of the study sites at longer intervals (e.g., 5-year intervals). Tracking the biological communities in the region’s lakes will not only provide information about the responses of shallow lakes as they become more stressed with climate change and industrial pollution, but also shed light on the various theories of alternate stable (Scheffer 1998) and unstable (Bayley et al. 2007) states in shallow lakes.

Finally, this thesis encourages careful and creative visual representation of the footprint and effects of the multiple stressors in the AOSR. Given the widespread and often polarized interest in the region and its industrial activity, it is imperative that the scientific findings are clearly communicated across research disciplines, to regulators, to industrial stakeholders, and to the public. A paleo-atlas will illustrate the histories of freshwaters across the AOSR, showing spatial and temporal changes in climate, land disturbance, biological communities, primary
production, isotope ratios, contaminant distribution, and metal concentrations. The broad-scale mapping will develop a foundation for further hypothesis testing (e.g., impact modelling) and effective monitoring in the region and other locations experiencing similar stressors. In general, continued paleolimnological research using multiple proxies and increased downcore resolution will advance our understanding of the complex interactions between the changing climate and the ongoing local industry. Visual representation of the data in this region where development priorities and environmental ethics often conflict should aid in widespread education and optimal management of this important and rapidly changing environment.
References


Summary

1) Gradual, muted biological assemblage changes, mainly driven by warmer conditions (reflected in chironomid assemblages) and associated increases in habitat complexity (reflected in diatoms and cladocerans), were evident in the downcore study investigating the effects of climate change and industrial contaminant and nutrient deposition on multiple biological proxies in a typical AOSR lake (RAMP 209). No evidence of a clear biological response to industrial airborne pollution was detected across the studied trophic levels. Given that RAMP 209 nutrient levels were likely relatively stable and not limiting to growth over the studied sedimentary record, light-controlling mechanisms (e.g., changes in ice cover) that are affected by warmer conditions are likely important in regulating the habitat availability and, ultimately, the biological community assemblage in the shallow lakes of the AOSR.

2) Increases were evident in all 23 lakes included in the investigation of primary production levels across the AOSR, indicating elevated production at a regional scale. Despite evidence of industrial activities depositing biologically available nutrients across the landscape, there was no evidence of the nutrients from local industry driving the production increases (i.e., no spatial pattern linking spectrally-inferred sedimentary chlorophyll-\(\alpha\) enrichment factors and interpolated deposition of biologically available nutrients). Rather, correlations to warming temperatures, which influence the growing season, are likely the main driver of the elevated aquatic primary production across the region.

3) Despite the region’s substantial environmental stressors, the collective changes in diatom communities identified in a “top-bottom” paleolimnological study of 18 shallow AOSR lakes were minor. There was no evidence of a discernable diatom response to airborne inputs from industry. Instead, the diatom community changes were mostly consistent with known responses to climate warming, which were mediated by lake-specific characteristics.
observed resiliency to change may be due to naturally high nutrient levels and/or a strong resistance to change that is a common phenomenon in many shallow lakes.
### Appendix A

#### A.1. Raw diatom counts for RAMP 209 diatom study (Chapter 2)

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- Neidium spp.
- Neidium affine v. longiceps
- Neidium ampliatum
- Neidium bisulcatum
- Neidium dubium
- Neidium hitchcockii
- Neidium testa
- Nitzschia spp.
- Nitzschia peisonis
- Nupela silvahercynia
- Nupela vitiosa
- Pinnularia alpina
- Pinnularia
- Pinnularia anglica
- Pinnularia anglica/interrupta
- Pinnularia cf. bacilliformis
- Pinnularia cf. bradelli
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### A.2. Raw diatom counts for “top-bottom” diatom study (Chapter 4)

<p>| Site       | Depth at Top of Interval (cm) | Year (CE) | Achnanthes spp. | Achnanthes spp. /Fragilaria spp. | Achnanthes acutissima | Achnanthes carinata | Achnanthes carinata | Achnanthes carinata | Achnanthes carissima | Achnanthes conspica | Achnanthes exigua | Achnanthes flexella | Achnanthes grana | Achnanthes lacus | Achnanthes lacus | Achnanthes lacus vulcani |
|------------|-------------------------------|-----------|-----------------|-----------------------------------|-----------------------|---------------------|---------------------|---------------------|---------------------|-------------------|----------------|----------------|----------------|----------------|
| RAMP 271 (T)| 0.5                           | 2012      | 0                | 9                                 | 37                    | 0                   | 0                   | 0                   | 0                   | 26                | 0              | 0              | 0              | 0              |
| RAMP 271 (B)| 43                            | 1880      | 0                | 3                                 | 8                     | 0                   | 0                   | 0                   | 0                   | 29                | 0              | 0              | 0              | 0              |
| 2014-Y (T)   | 0                             | 2014      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| 2014-Y (B)   | 31                            | 1849      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| 282 (T)      | 0                             | 2014      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 16                | 0              | 0              | 0              | 0              |
| 282 (B)      | 26                            | 1858      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 39                | 0              | 0              | 0              | 0              |
| 2014-D (T)   | 0                             | 2014      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| 2014-D (B)   | 22                            | 1850      | 5                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 2                 | 2              | 0              | 0              | 0              |
| RAMP 464 (T) | 0                             | 2012      | 0                | 0                                 | 8                     | 0                   | 1                   | 0                   | 0                   | 0                 | 1              | 0              | 0              | 0              |
| RAMP 464 (B) | 30                            | 1850      | 1                | 1                                 | 1                    | 6                   | 0                   | 0                   | 0                   | 0                 | 0              | 2              | 0              | 0              |
| NE13 (T)     | 0                             | 2011      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 7              | 0              | 0              | 0              |
| NE13 (B)     | 21                            | 1842      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| RAMP 226 (T) | 0.5                           | 2010      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| RAMP 226 (B) | 38                            | 1845      | 8                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| 2014-X (T)   | 0                             | 2013      | 0                | 0                                 | 2                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| 2014-X (B)   | 43                            | 1851      | 3                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| 2014-Z (T)   | 1                             | 2011      | 4                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| 2014-Z (B)   | 54                            | 1856      | 6                | 0                                 | 7                     | 0                   | 2                   | 15                  | 0                   | 0                 | 0              | 0              | 0              | 0              |
| RAMP 225 (T) | 0.5                           | 2012      | 1                | 0                                 | 11                   | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 10             |
| RAMP 225 (B) | 40                            | 1827      | 12               | 0                                | 7                    | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| RAMP 175 (T) | 0                             | 2013      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| RAMP 175 (B) | 45                            | 1860      | 1                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| Kearl (T)    | 0.5                           | 2012      | 6                | 0                                 | 0                     | 14                  | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| Kearl (B)    | 24                            | 1847      | 0                | 0                                 | 0                     | 0                   | 2                   | 0                   | 0                   | 0                 | 12             | 0              | 0              | 0              |
| 274 (T)      | 0.5                           | 2014      | 5                | 0                                 | 0                     | 0                   | 0                   | 0                   | 2                   | 16              | 0              | 0              | 0              | 0              |
| 274 (B)      | 43                            | 1897      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 2                 | 0              | 0              | 0              | 0              |
| RAMP 209 (T) | 0                             | 2013      | 6                | 0                                 | 0                     | 0                   | 0                   | 2                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| RAMP 209 (B) | 35                            | 1857      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| 265 (T)      | 0                             | 2015      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| 265 (B)      | 50                            | 1863      | 1                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| RAMP 268 (T) | 1.5                           | 2013      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| RAMP 268 (B) | 43                            | 1848      | 0                | 0                                 | 17                   | 0                   | 0                   | 0                   | 21                  | 0                 | 0              | 0              | 0              | 0              |
| RAMP 185 (T) | 0                             | 2012      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| RAMP 185 (B) | 30                            | 1850      | 1                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| Pushup (T)   | 0                             | 2014      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 0              | 0              | 0              | 0              |
| Pushup (B)   | 37                            | 1848      | 0                | 0                                 | 0                     | 0                   | 0                   | 0                   | 0                   | 0                 | 2              | 0              | 0              | 0              |</p>
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<th>Achnanthes nodosa</th>
<th>Achnanthes oestrupii</th>
<th>Achnanthes peragalli</th>
<th>Achnanthes petersenii</th>
<th>Achnanthes rechtersis</th>
<th>Achnanthes riula</th>
<th>Achnanthes cf. petersenii</th>
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**Note:** The table represents the abundance of different phytoplankton species at various sites, with each cell indicating the presence or absence of a species. The species names are listed in the header, and the values correspond to the presence (1) or absence (0) of each species at a specific site.
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<th>Tabellaria spp.</th>
<th>Tabellaria flocculosa</th>
<th>Ulnaria cl. ulna</th>
<th>Sum of diatom valves</th>
<th>cysts</th>
<th>scales</th>
<th>sponge spicule tips</th>
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Appendix B

B.1. Lake characteristics, core information, and diatom assemblage data from Big Peter Pond and La Loche (two larger, deeper lakes in the ASOR)

Geographical, physical, chemical, and industry-related characteristics, and core information for Big Peter Pond and La Loche.

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Relative abundances of select diatom taxa in the present-day (top of each shaded pair) and pre-disturbance (bottom of each shaded pair) sediment intervals from Big Peter Pond and La Loche. *A. granulata* (Kilham and Kilham 1975), *A. ambigua* (Clerk et al. 2000), and *Stephanodiscus* (Smol and Stoermer 2010) taxa are commonly found in meso-eutrophic to eutrophic conditions.
C.1. Scatter plots comparing select limnological measures from samples collected at the time of sediment coring (under ice cover; x axis) versus measures from samples collected during August, September, or October from the same year (no ice cover; y axis)

Water samples from under the ice at the time of coring were collected by the Environment and Climate Change Canada (ECCC) field crew. Water samples from August, September, or October from the same year that each lake was cored were collected by the Regional Aquatics Monitoring Program (RAMP). RAMP water chemistry was available to enable a comparison for 11 lakes (RAMP 175, RAMP 185, RAMP 209, RAMP 223, RAMP 225, RAMP 226, RAMP 227, RAMP 268, RAMP 271, Kearl (RAMP 418), and RAMP 464). A 1:1 line is included. Samples that plot above the 1:1 line indicate greater values measured by the RAMP.