

**Atmospheric Nitrogen Deposition and Nitrogen Dynamics of a Reclaimed
Fen-upland in the Athabasca Oil Sands Region, Alberta**

by
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Author's Declarations

I hereby declare that I am the sole author of this thesis. This is a true copy of the thesis, including any required final revisions, as accepted by my examiners.

I understand that my thesis may be made electronically available to the public.

Abstract

Surface mining in the Athabasca Oil Sands Region (AOSR) has resulted in the alteration of over 895 km² of wetland and forest ecosystems in the Western Boreal Plain (WBP) of northeastern Alberta. After surface mining ceases, the Alberta Government requires all disturbed land be reclaimed to an equivalent pre-mined land capability that consists of native boreal plant and wildlife species. The Nikanotee Fen Watershed is a reclaimed fen peatland – forest upland system constructed as a pilot attempt to reclaim the post-mined landscape back to a self-sustaining, carbon accumulating and biodiverse fen-upland ecosystem. During reclamation, ongoing mining operations continuously emit nitrogen (N) emissions that subsequently deposit onto the surrounding environment. This N deposition is a biogeochemical and ecological disturbance to the reclaimed system that may impact the N-status of the typically low N environment, which could threaten reclamation objectives. Quantifying N deposition and assessing the N-status of the reclaimed system will further our understanding of the impacts of N deposition on young post-mined landscapes. This study assesses N deposition both from a large- and small-scale perspective in the AOSR. The first objective quantifies N deposition loads to the reclaimed site (situated directly within the industrial centre) and three reference sites located between 10-65 km south from the industrial centre, to determine whether the reclaimed site receives greatest N deposition loads due to its industrial location (large-scale perspective). The following objectives focus only on the constructed fen-upland to analyze N availability and N dynamics on the reclaimed landscape, and determine the N-status of the fen-upland and assess the potential implications of N deposition on the development of the system (small-scale perspective).

Nitrogen deposition was collected using a network of passive atmospheric deposition collectors across all study sites and a variety of environmental samples were collected specifically at the reclaimed fen-upland to assess N availability, dynamics, and status of the soil-water-vegetation continuum of both reclaimed landscape units. Results indicate that N deposition loads of nitrate (NO₃-N), ammonium (NH₄-N) and total inorganic N (TIN) were greatest at the reclaimed site, decreasing with distance southwards from the industrial centre, suggesting that the reclaimed site is the most susceptible to the impacts of N deposition (particularly ecosystem N enrichment). Although the reclaimed site is situated in a high N deposition zone in the AOSR, N deposition is not currently increasing N availability or dynamics of either landscape unit. Ecosystem indicators within the soil-water-vegetation continuum suggest that the fen-upland is a low N environment. Findings from this study improve our understanding of the recovery of N nutrient pools ~7 years post-reclamation. Furthermore, findings from this study provide a foundation for understanding the N-status of a reclaimed landscape receiving greater-than-background N deposition loads and what this may mean for the development of the system.

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I would like to acknowledge that the land from which field data was collected for this thesis is within the boundaries of Treaty 8, traditional lands of the Dene and Cree, as well as the traditional lands of the Métis of northeastern Alberta.

I also acknowledge that my graduate courses, lab analyses and writing of this thesis took place on the traditional territory of the Neutral, Anishnaabeg, and Haudenosaunee Peoples. The University of Waterloo is situated on the Haldimand Tract, land promised to Six Nations, which includes six miles on both sides of the Grand River from its mouth to its source.

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List of Abbreviations

AOSR	Athabasca Oil Sands Region
BC	Base Cation
C:N	Carbon to Nitrogen Ratio
CRF	Controlled Release Fertilizer
Ext-N	Extractable Nitrogen
Ext-NH ₄ -N	Extractable Ammonium
Ext-NO ₃ -N	Extractable Nitrate
FFM	Forest Floor Mineral Mix
GW	Groundwater
IEC	Ion-Exchange Collector
LAI	Leaf-Area Index
LFH	Litter Fermentation Humus
LOI	Loss on Ignition
MET	Meteorology
N	Nitrogen
NH _x	Reduced Nitrogen
NH ₃	Ammonia
NH ₄ -N	Ammonium Nitrogen
NH ₄ -N:NO ₃ -N	Ammonium to Nitrate Ratio
NO _x	Nitrogen Oxide
NO ₃ -N	Nitrate Nitrogen
OM	Organic Matter
PM	Particulate Matter
PMM	Peat-Mineral Mix
PW	Porewater
RH	Relative Humidity
SAGD	Steam-Assisted Gravity Drainage
SF	Stemflow
SW	Surface water
TC	Total Carbon
TIN	Total Inorganic Nitrogen
TN	Total Nitrogen
VWC	Volumetric Water Content
WBP	Western Boreal Plain
WT	Water Table

1.0 Introduction

1.1 Background

The Athabasca Oil Sands Region (AOSR) lies within the sub-humid boreal region of northeastern Alberta, spanning an area of roughly 142,200 km² (Government of Alberta, 2021). The AOSR, situated in the Western Boreal Plain (WBP), is the largest of three Canadian oil deposits (Athabasca, Peace River and Cold Lake) and is the region that exhibits the most intense mining operations, associated with landscape disturbance and elevated atmospheric emissions from bitumen upgrading processes (Percy, 2013). In the AOSR, 20% of the oil sands deposit is located near the surface (~75 m) and as such is mined using large shovel and trucking machinery to create open pit mines (CAPP, 2019; Government of Alberta, 2021). The remaining 80% is deep below ground and can only be recovered using steam-assisted gravity drainage (SAGD) (in-situ method), which pumps bitumen to the surface using steam (CAPP, 2019; Government of Alberta, 2021). Both methods have direct impacts on the natural boreal forest environment in the AOSR, with the most concerning being the open-pit mining (Rooney et al., 2012). Open-pit mining results in complete stripping of the natural boreal landscape that was originally comprised of 64% wetlands, ~90% of which were fen peatlands, and 23% forested uplands (Rooney et al., 2012; Daly et al., 2012). To access the oil sand, this method creates pits that are several kilometers wide and up to 100 m deep (Rowland et al., 2009). The pre-disturbed wetland-upland mosaic is removed, and the vital ecosystem services that these ecologically significant environments provide, including water storage, ecological biodiversity, carbon storage and nutrient cycling, are lost (Nwaishi et al., 2015). To mitigate the disturbance of mining on the boreal environment, reclamation efforts are currently being explored to re-construct fen peatlands and forested uplands in the AOSR (Daly et al., 2012; Ketcheson et al., 2016). However, even during reclamation, atmospheric emissions, and regional

deposition from ongoing mining development, continue to occur and must be monitored due to the ecological concern of deposition on the environment (Vitt et al., 2003; Wieder et al., 2010).

Concerns regarding atmospheric deposition of reactive elements, particularly nitrogen (N) compounds, have been raised in the AOSR, since N emissions are expected to increase with expanding mining operations (Vitt et al., 2003; Laxton et al., 2010). In 2018, oil production hit 2.9 million barrels per day, and this is expected to increase to 4.25 million barrels per day by 2035 (CAPP, 2021). The bituminous oil sand that is recovered from both open-pit and in-situ mining is unprocessed, heavy crude oil that must be transported to crushers for processing and upgrading into synthetic crude oil (Landis et al., 2012). The techniques, petrochemicals and mechanical processes used in oil upgrading, refining and overall power generation create byproducts that are eventually emitted into the atmosphere via facility smokestacks. These smokestacks emit inorganic anthropogenic pollutants including reactive N compounds, sulfur oxides (SO_x), heavy metals and particulate matter (PM) (Landis et al., 2012; Hsu et al., 2016). Additionally, both extraction methods result in atmospheric emissions from truck and fleet operations that expel diesel engine exhaust and fugitive PM from haul road dust kick-up (Landis et al., 2012).

Although oil sands operations and fossil fuel combustion are the main source of industrial N emissions in the AOSR (Edgerton et al., 2020), there are other emission sources that are found within the region including urban, natural, and agricultural sources. Within urban areas, mobile source emissions from vehicles and residential heating are both sources of N (Landis et al., 2012). A natural emitter such as forest fires, which are common in the summertime, can also be a source of N. Long-range transport of N emissions (specifically ammonia, NH₃) from agricultural areas located to the south can travel northwards (aided by the SW winds) and deposit in the southern

portions of the AOSR (Hsu et al., 2016). These N sources in addition to industrial oil sands emissions, can distort source apportionment of N emissions within the region.

Atmospheric N Deposition

The two main N forms emitted into the atmosphere in the AOSR are reduced N (NH_x) and oxidized N (NO_x). After N is released into the atmosphere, it undergoes dilution followed by photochemical reactions and transformations as it moves away from the source (Hsu et al., 2016; Percy, 2013) (Figure 1.1). Atmospheric reactions transform the primary N pollutants (original N form) into secondary pollutants (altered N form) that contribute to both dry and wet deposition (Hsu et al., 2016). Ammonia (NH_3), nitric oxide (NO), and nitrogen dioxide (NO_2) are the most common reactive N compounds emitted into the atmosphere (Galloway et al., 2004; Li et al., 2016) and are the key drivers of N deposition in the AOSR (Hanson and Lindberg, 1991; Bytnerowicz and Fenn, 1996; Hsu et al., 2016). Ammonia is thought to contribute the greatest portion of N deposition in comparison to all other N species emitted in the AOSR (Whaley et al., 2018).

Upon being transformed, NH_3 is deposited as ammonium ($\text{NH}_4^+\text{-N}$) and NO_x is deposited as nitrate ($\text{NO}_3^-\text{-N}$) through dry and wet processes (Bytnerowicz and Fenn, 1996; Li et al., 2016). Dry deposition occurs when N from gases, aerosols and particles drop from suspension in the atmosphere and deposit onto the landscape in particulate or gas form (Hanson and Lindberg, 1991). Total N deposition is largely composed of dry deposition rather than wet deposition (Bytnerowicz and Fenn, 1996). Wet deposition occurs when N gases, aerosols and particles are scavenged by clouds and precipitation and are dissolved into water droplets and deposited onto the landscape in liquid form (Li et al., 2016). Dry and wet deposition that enter the environment are controlled by atmospheric turbulence, surface resistance, physical/chemical properties of the N compound, chemical potential gradient between the atmosphere and receptors, and the nature of the vegetative

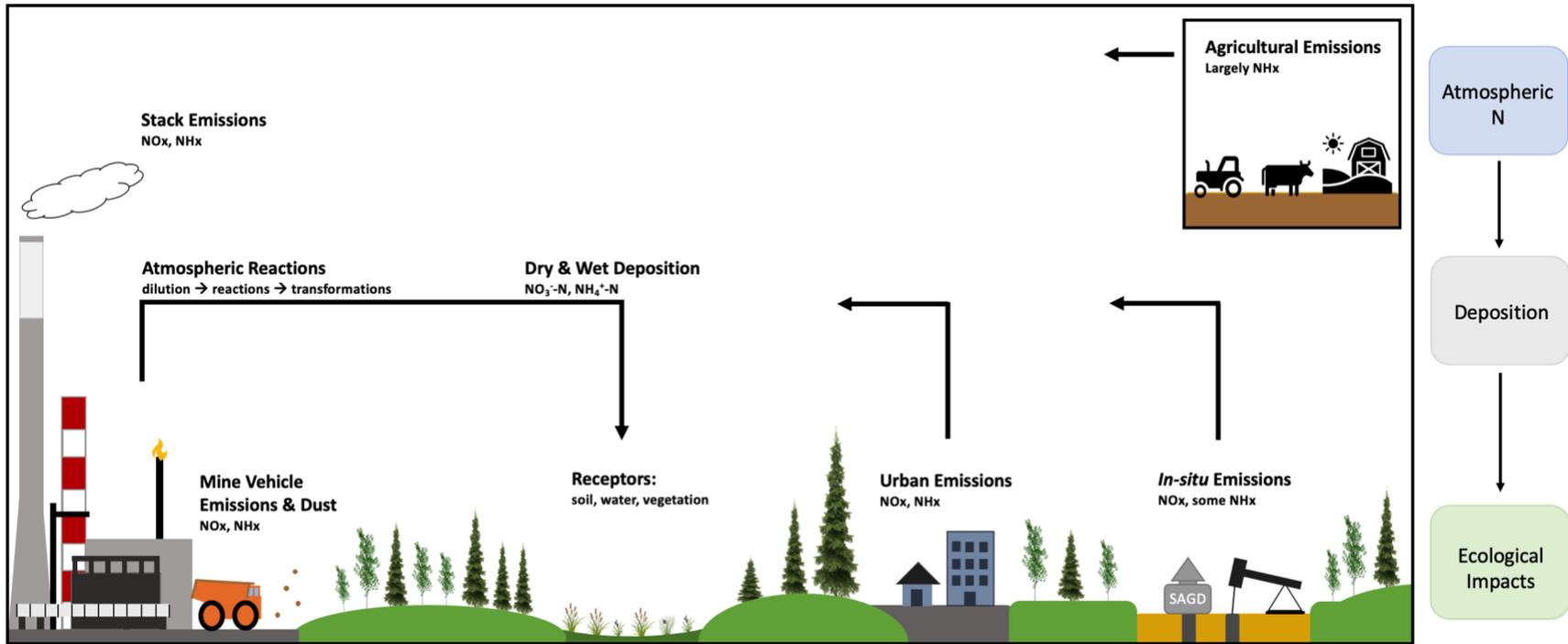


Figure 1.1: Diagram of the exogenous N sources in the AOSR and deposition receptors in the undulating wetland-upland environment. Agriculture is not a major N source to the AOSR but can occur. Figure modified from Foster et al. (2019) and Greaver et al. (2012).

surfaces (Hanson and Lindberg, 1991).

Environmental Impacts of N Deposition

Emissions of NH_x and NO_x are of concern as these two compounds act in combination with one another to alter the nutrient balance of ecosystems (Greaver et al., 2012). There are concerns that oil sands N emissions could potentially result in higher-than-normal N inputs to surrounding aquatic and terrestrial ecosystems, which could ultimately result in increased N concentrations eventually leading to increased N-status and saturation of ecosystems in the AOSR (Laxton et al., 2012; Hsu et al., 2016). Boreal ecosystems are typically N-limited (adapted to low N conditions); however, N deposition can accumulate and increase available N within the system (Laxton et al., 2012). This can stimulate plant growth and boost primary productivity, until a saturation point is reached, whereby nutrient imbalances can occur and ultimately result in community composition shift or plant decline and mortality (Fenn & Poth, 1998; Aber, 1992). Even low chronic N deposition loads can alter the N-cycle and overall N-status of the boreal environment (Aber et al., 1989; Laxton et al., 2010). Moreover, there have previously been concerns of the acidifying effects of N (and sulfur, S) deposition to acid sensitive ecosystems in the AOSR, particularly jack pine forest stands. It has been determined that this is only of minimal concern due to high base cation (BC) emissions in the region that generally match or slightly exceed the combined inputs of N and S deposition (Watmough et al., 2014). The zone of BC deposition that matches N (and S) deposition, however, it is likely limited to a radius of 20-30 km from the main industrial area (Fenn et al., 2015). In general, for ecosystems that are located in close proximity to the industrial centre, the risk of soil acidification is mitigated largely by high BC deposition and thus, the nutrient effects of N deposition are of more concern than the acidifying effects. Multiple studies have been conducted outside the active mining area to assess the impacts of N deposition on natural boreal

landscapes (Vitt et al., 2003; Wieder et al., 2010), and some studies have quantified N deposition loads to both natural and industrial sites in the AOSR (Hsu et al., 2016; Fenn et al., 2015; Proemse et al., 2013; Edgerton et al., 2020; MacKenzie & Dietrich, 2020). However, very few have assessed the biogeochemical implications of N deposition on reclaimed landscapes in the AOSR (Hemsley et al., 2019), that are built with the goal of recreating a low N environment typical of the region.

Fen and Upland Reclamation

Once oil sands mining ceases, the Alberta Government requires oil companies to reclaim the land back to an equivalent pre-mined capacity by means of complete physical engineering of landforms (Government of Alberta, 2019; Hemstock et al., 2010). Specifically, this means that the land previously mined for oil sands extraction must be reconstructed to a naturally comparable pre-mined functionality, which includes returning the land to an equivalent hydrological, ecological, and biogeochemical state (Daly et al., 2012). The land does not need to be reclaimed to its ‘original’ state, but rather to a self-sustaining landscape of equivalent capability that consists of native boreal plant and wildlife species (Rooney et al., 2012; CAPP, 2019). Although peatlands are the most common wetland type in the AOSR, they are highly complex systems to reconstruct (Daly et al., 2012). However, peatlands are a major feature in the pre-disturbed landscape, covering an estimated 29% of the oil sands administrative area with fens being the dominant peatland type (Lee & Cheng, 2009; Wieder et al., 2016b). Fens are minerotrophic wetlands that rely on their hydrologically connected mineral upland landscapes as a groundwater source (surface runoff is limited) (Devito et al., 2005a; Daly et al., 2012). It is because of this connectivity between fens and uplands that these ecosystems have been resilient in the sub-humid WBP environment. Fens rely on uplands as a water source, and during dry periods fens act as a reservoir to supply uplands (Devito et al., 2005a; Price et al., 2010). Since fens dominate the pre-mined landscape, recent

efforts have focussed on fen reclamation (Daly et al., 2012; Price et al., 2010). To reclaim fen peatlands, upland reclamation must also occur to provide an adequate water supply and ensure a self-regulating system in the sub-humid climate (Daly et al., 2012).

To test whether fen reconstruction can be a successful reclamation technique, the Nikanotee Fen Watershed was constructed within the Millennium mine lease at Suncor Energy Inc. The site consists of a connected fen-upland system situated in the centre of a larger constructed watershed (Price et al., 2010). The fen-upland was engineered to adequately supply the lower-lying fen with water via a tailings sand aquifer in the upland. The goal of this fen reclamation project is to construct a self-sustaining, carbon accumulating and biodiverse boreal ecosystem (Daly et al., 2012). Furthermore, construction and extensive research of this site will generate knowledge and techniques that can be implemented into future reclamation projects in the AOSR (Daly et al., 2012; Ketcheson et al., 2016).

Nikanotee fen-upland consists of multiple engineered layers that all lie on top of an overburden dump (Daly et al., 2012; Ketcheson et al., 2016). To build the 2.9 ha fen, peat from a nearby mining lease (a donor fen) was stripped, drained, and stockpiled (Rooney et al., 2012). The salvaged peat was then transferred to the reclaimed fen to serve as a substrate (~2 m deep) for vegetation establishment (Nwaishi et al., 2015). A structured experimental design was used as the revegetation strategy in the fen, which took place in 2013. Experimental planting in the fen followed a randomized factorial design that included bare peat plots and the planting of mosses (e.g., *Tomenthypnum nitens* and *Sphagnum warnstorffii*) and seedlings (e.g., *Juncus balticus*, *Triglochin maritima*, and *Carex aquatilis*) (Scarlett et al., 2017). Fertilizer was not applied to the fen during or after construction. Any nutrients within the donor peat were introduced to the fen at the time of construction. The 7.7 ha upland was built with 3 m of re-purposed tailings sand, which

was then capped by a thin (~0.3-0.5 m) layer of salvaged organic amendments. Organic amendments in the upland include a forest floor mineral mix (FFM) and some peat-mineral mix (PMM). The layer of salvaged organic amendments in the upland creates a soil-like substrate for microbial communities and vegetation establishment (Rowland et al., 2009; Hemstock et al., 2010). The organic cap increases the organic carbon, nutrient content, and water-holding capacity of the reconstructed upland (Rowland et al., 2009). Planting in the upland took place in 2013 and 2015. This involved the planting of a tree canopy consisting of jack pine (*Pinus banksiana*) and black spruce (*Picea mariana*), as well as an understory of various shrub and graminoid species (Gingras-Hill et al., 2018). During planting in 2015, each sapling was planted with a packet of controlled release fertilizer (CRF). Since the initial planting, however, the upland consists of a patchy distribution of scattered black spruce, jack pine, trembling aspen (*Populus tremuloides*) and balsam poplar (*Populus balsamifera*). Aspen and poplar may have been transplanted with the organic amendments or entered the reclaimed site by airborne seed dispersal.

Since the reclaimed fen-upland is situated directly within the zone of high N deposition in the AOSR (Proemse et al., 2013), it is unknown if N deposition may have an impact on the N-status and community composition of the site. Ecosystems in the AOSR have persisted in a region with naturally very low atmospheric N deposition inputs (Wieder et al., 2016b). However, due to the development of industrial oil sands activities and their associated N emissions, N deposition has become a threat to surrounding ecosystems in the AOSR (Vitt et al., 2003; Wieder et al., 2010), especially those situated in close proximity to the oil sands centre. As most N deposition studies have specifically assessed N deposition impacts on natural ecosystems in the AOSR, there is the need to understand the impacts of N deposition on reclaimed ecosystems and how N deposition may impact their development.

1.2 Research Objectives

Ecosystems in the AOSR are at risk of increasing N availability from atmospheric N deposition due to mining activities. The Nikanotee Fen Watershed is a young post-mined landscape situated directly within industrial activities. The site is ~7 years old and has had to develop within a highly disturbed environment. The amount of N deposition received by the site is currently unknown. It is anticipated that due to its location, deposition will be substantial. Hence, the overall goal of this research is to determine the current N-status of a reclaimed fen-upland in the AOSR and whether N deposition is impacting the N-status of the soil-water-vegetation continuum and community composition. A comparison of N deposition loads between the reclaimed site in relation to reference sites will show whether there is a gradient in deposition loads with distance southwards from the industrial oil sands centre, and thus, how N deposition in the reclaimed fen compares to wetlands that are less heavily impacted by mining activities. This research is important to our understanding of how external factors control boreal nutrient dynamics and vegetation communities and will also enhance our understanding of the recovery of N nutrient pools ~7 years post-construction. The primary research objectives of this study are to:

- 1) Quantify N deposition loads to a reclaimed fen-upland and reference sites in the AOSR;
- 2) Characterize N availability and N dynamics in a reclaimed fen and upland;
- 3) Determine the N-status of the reclaimed fen and upland, and assess the potential implications of N deposition on the development of the reclaimed system.

These three objectives are addressed in two chapters that form the basis of two manuscripts to be submitted to journals upon completion of this thesis.

2.0 Manuscript Chapter 1. Atmospheric nitrogen deposition loads at reclaimed and natural fens in the Athabasca Oil Sands Region, Alberta

2.1 Introduction

The Canadian oil sands deposit in northern Alberta and Saskatchewan is the third largest oil reserve in the world, following Venezuela and Saudi Arabia (Government of Alberta, 2020). 1.7 trillion barrels of oil are estimated to be found within the Canadian deposit, with 169 billion barrels estimated to be recoverable using current technology (Percy, 2013). These oil sands are found in three deposits that span across approximately 142,200 km² of boreal forest, of which 50% are wetlands (Government of Alberta, 2021; Vitt et al., 1996). The three deposits include the Peace River, Cold Lake and Athabasca, with the largest of these being the Athabasca deposit in northeastern Alberta (Government of Alberta, 2021). The Athabasca deposit is termed the Athabasca Oil Sands Region (AOSR) and the city of Fort McMurray is the largest municipality that falls within the region. In the AOSR, 20% of the oil reserve is located near the surface and as such is mined using large shovel and trucking machinery, including fleets of 400-ton heavy haulers (CAPP, 2019; Lynam et al., 2015). The remaining 80% is deep below ground and can only be recovered using steam-assisted gravity drainage (SAGD) (in-situ methods) of drilling and pumping bitumen to the surface using steam (CAPP, 2019). Both mining methods and their associated upgrading and processing activities result in significant industrial air emissions in the region (Proemse et al., 2013; Wieder et al., 2016a).

Atmospheric emissions from large-scale oil extraction and processing in the AOSR have raised concerns for the surrounding environment. In particular, nitrogen (N) emissions of N oxides (NO_x) and reduced N (NH_x) that are a byproduct from oil sands activities originating from point, mobile and fugitive sources (including smokestacks, mine fleets and dusts, respectively) (Hsu et

al., 2016), are a concern to the naturally low N boreal ecozone (Laxton et al., 2012). These N emissions originate largely from mining sources situated directly within the main industrial oil sands centre (where open-pit mining occurs), but can also originate from urban, agricultural, and natural sources (i.e., wildfires) (Lynam et al., 2015; Hsu et al., 2016). It is predicted that total industrial NO_x emissions will increase from 140 Mg/day in 2000 to 360 Mg/day in 2032 from mining expansion (Doram and Rawlings, 2006). The most common N compounds emitted by industrial activities are ammonia (NH₃), nitric oxide (NO) and nitrogen dioxide (NO₂) (Galloway et al., 2004; Li et al., 2016).

After being emitted into the atmosphere, NO_x and NH_x undergo dilution, followed by photochemical reactions and transformations as they disperse and are transported away from the source (Hsu et al., 2016; Percy, 2013). Upon being transformed and transported, NH₃ is deposited as ammonium (NH₄⁺-N) and NO_x is deposited as nitrate (NO₃⁻-N) via both dry and wet pathways (Bytnerowicz and Fenn, 1996; Li et al., 2016). Dry N deposition consists of N in gases, aerosols and particles that fall from suspension in the atmosphere and wet N deposition consists of N in gases, aerosols and particles that are dissolved in precipitation and deposit as water droplets (Hanson and Lindberg, 1991; Li et al., 2016). Deposition is typically collected as ‘bulk’ deposition using passive samplers, which collect both dry and wet deposition together.

Nitrogen deposition decreases exponentially within a distance of ~25 km from the main oil sands industrial centre (Fenn et al., 2015). However urban areas and remote SAGD sites are also sources of N emissions (Proemse et al., 2013; Moorhouse et al., 2010), which create N deposition hotspots in the AOSR separate from the main industrial operations. Atmospheric NH₄⁺-N and NO₃⁻-N ultimately deposit onto the surrounding aquatic and terrestrial ecosystems in the AOSR, consisting largely of bogs, fens and black spruce and jack pine forests (Wieder et al., 2016a;

Wieder et al., 2016b; Hsu et al., 2016; Edgerton et al., 2020). Nitrogen deposition may result in enhanced growth and productivity in these ecosystems; however, once an ecosystem's critical load is met or surpassed, this may result in plant mortality and a shift in community composition due to intensified decomposition and mineralization rates (Bytnerowicz and Fenn, 1996; Wieder et al., 2019). Excessive N deposition ultimately results in acidification and/or eutrophication of these sensitive boreal ecosystems (Lovett, 2013).

This research adds N deposition data to the already large network of atmospheric sampling sites across the AOSR. However, this paper focuses on N deposition moving southwards from the industrial centre, specifically over the summertime from May to August. Research for this study was conducted at one reclaimed fen-upland system located directly within the oil sands mining footprint (denoted as 0 km for this paper), and three reference fens situated southwards outside of the mining footprint, situated between 10 - 65 km away from the reclaimed site. Each study site included in this research has been extensively monitored for various hydrological, biological, and geochemical processes. However atmospheric N deposition inputs to each of these sites has not been quantified. Quantifying N deposition at each site will assist in the development of an integrative nutrient framework to better understand the biogeochemical functioning of disturbed and natural peatlands in the AOSR (Nwaishi et al., Under review). Therefore, the primary research objectives of this study are to: 1) quantify atmospheric N deposition loads at each study site; 2) determine whether the spatial pattern of decreasing N deposition with distance away from the main oil sands operations holds true at these specific sites in a southern direction; and 3) assess the potential ecological effects of N deposition at each site. Consistent with established literature in the spatial distribution of N deposition in the AOSR (Fenn et al., 2015; Proemse et al., 2013; Hsu et al., 2016; Bytnerowicz et al., 2010), we hypothesized that N deposition loads would be highest

at the reclaimed site, situated directly within industrial activities, decreasing at each reference site with distance southwards. A southerly transect from the industrial oil sands centre was selected because existing reclamation reference sites were used in this study.

2.2 Study Sites

Field research for this study was carried out between May and August 2018 at a reclaimed fen watershed and three reference fens located in the Athabasca Oil Sands Region (AOSR) in Fort McMurray, Alberta (Figure 2.1). The reclaimed fen is situated within the mine lease area of Suncor Energy Inc. and the remaining reference sites are situated outside of the mine lease area. Nikanotee Fen Watershed, hereafter referred to as Nikanotee (reclaimed site) and Poplar Creek Fen, hereafter referred to as Poplar (reference site) are both located approximately 30 km north of the city of Fort McMurray. Saline fen (reference site) and Pauciflora fen (reference site) are located approximately 15 km and 40 km south of the city of Fort McMurray, respectively. For purposes of this study, each fen will be denoted as distance (in km) away from Nikanotee, which is located directly within industrial oil sands activity, and thus, Nikanotee is 0 km, Poplar is 10 km, Saline is 40 km, and Pauciflora is 65 km (Figure 2.2). All sites are located within the sub-humid climate of the Western Boreal Plain (WBP), where mean annual air temperature (1981-2010) is 1 °C, with summer and winter averages fluctuating between 10 and 17°C and -17 and -6 °C, respectively (Environment and Climate Change Canada, 2017). Average annual precipitation of the region is 418.6 mm, where ~70% falls in the form of rainfall particularly during the summer months (Environment and Climate Change Canada, 2017).

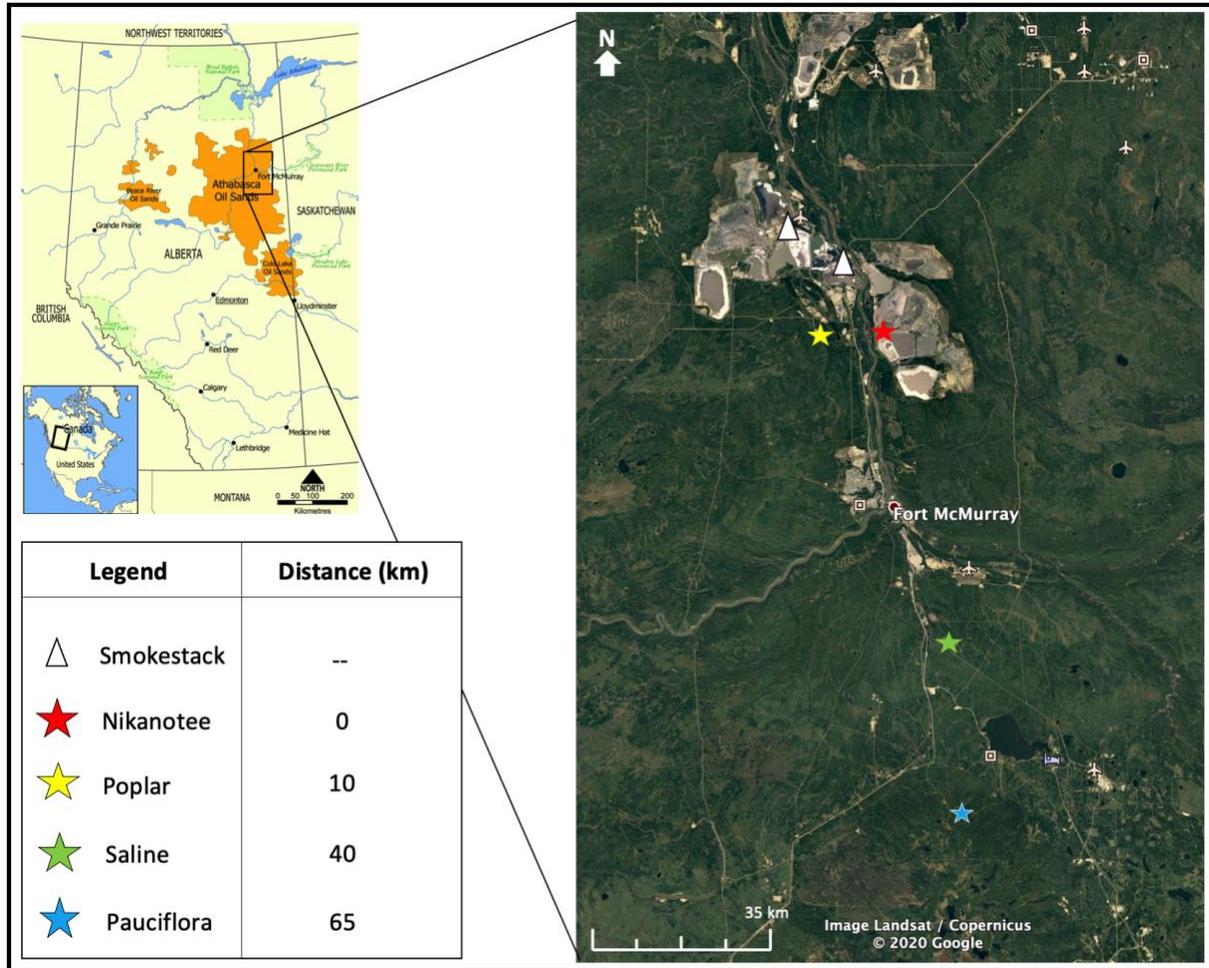


Figure 2.1: Map showing the location of the two main smokestacks (Syncrude left, Suncor right) and each research site in the Athabasca Oil Sands Region, Alberta.

Nikanotee Site

Nikanotee (56°55'54.3"N, 111°25'01.9"W; ~278 mASL) is a young, post-mined reclaimed fen-upland system (3 ha fen; 7 ha upland) located on Suncor's Millennium mine lease, situated directly within industrial oil sands activity (Figure 2.1). The fen-upland is surrounded by three previously reclaimed slopes (west, east, and southeast) and one natural slope to the south, which together form the watershed. The entire watershed is bordered by industrial dirt roads, including an active mine haul-road located to the east. Dominant vegetation cover in the fen includes salt-tolerant juncus (*Juncus balticus*), freshwater carex (*Carex aquatilis*) and invasive typha (*Typha latifolia*).

Similarly, the dominant vegetation cover in the upland includes black spruce (*Picea mariana*), jack pine (*Pinus banksiana*), trembling aspen (*Populus tremuloides*) and balsam poplar (*Populus balsamifera*). Average leaf-area index (LAI) for the reclaimed fen-upland in 2018 was 0.68.

Poplar Site

Poplar (56°56'14.8"N, 111°32'43.3"W; ~320 mASL) is a natural, 11 ha, moderate-rich treed fen with patches of upland forest located within Poplar Creek, ~10 km west of Nikanotee (Figure 2.1). Similar to Nikanotee, the site is bordered by industrial dirt roads, however, it is situated further away from active mining activities. Dominant fen vegetation cover consists of tamarack (*Larix laricina*), stunted black spruce, *Carex spp.* and mosses (*Sphagnum spp.*) (Borkenhagen, 2013). In the upland, dominant vegetation consists of trembling aspen, black spruce, jack pine, Labrador tea (*Ledum groenlandicum*), wood horsetail (*Equisetum sylvaticum*) and brown moss (*Tomenthypnum nitens*) (Borkenhagen, 2013). Average LAI in 2018 was 0.68. In May 2016, Poplar was almost entirely burned during the Horse River wildfire (MWF-009) and a large swath of tree cover and low-lying vegetation was destroyed, most notably in the drier upland. Since the fire, new growth has started to re-appear and re-establish on site (van Beest et al., 2019).

Saline Site

Saline (56°34'19.1"N, 111°16'27.5"W; ~400 mASL) is a natural, ~27 ha, open saline spring fen located within the Salt Creek Watershed (Wells and Price, 2015), approximately 40 km south of Nikanotee and 10 km south of the city of Fort McMurray (Figure 2.1). Unlike Nikanotee and Poplar, Saline is not surrounded by any industrial dirt roads or major industrial activities and instead, vegetated cutlines and undeveloped well pads. Salt-tolerant species make up the site including (but not limited to) juncus, sweetgrass (*Hierochloa hirta ssp.*) and reed grass (*Calamagrostis inexoansa*) (Phillips, et al., 2015; Wells and Price, 2015) with an average LAI of

0.96. The main fen complex consists entirely of low-lying vegetation (i.e., graminoids) and no tree canopy. However, the fen is surrounded by a mix of living and burned tamarack and black spruce trees, dwarf birch and other shrub species.

Pauciflora Site

Pauciflora (56°22'34.5"N, 111°14'10.3"W; ~740 mASL) is a natural, 8 ha, poor treed fen located on a regional topographic high in the Stony Mountain Wildland, approximately 65 km south of Nikanotee (Figure 2.1). Due to the site's topographic position (Figure 2.2), it experiences cooler temperatures and higher precipitation than the other study sites. An unpaved dirt road bisects the site at the very north edge of the fen and the rest of the fen is bordered by forested uplands. Dominant vegetation cover in the fen consists primarily of *Sphagnum spp.*, *Carex spp.*, some shrub species, dwarf birch, and stunted black spruce with an average LAI of 0.57. Dominant vegetation in the surrounding upland consists of a dense canopy of primarily black spruce, Labrador tea and a variety of feather moss and lichen species.

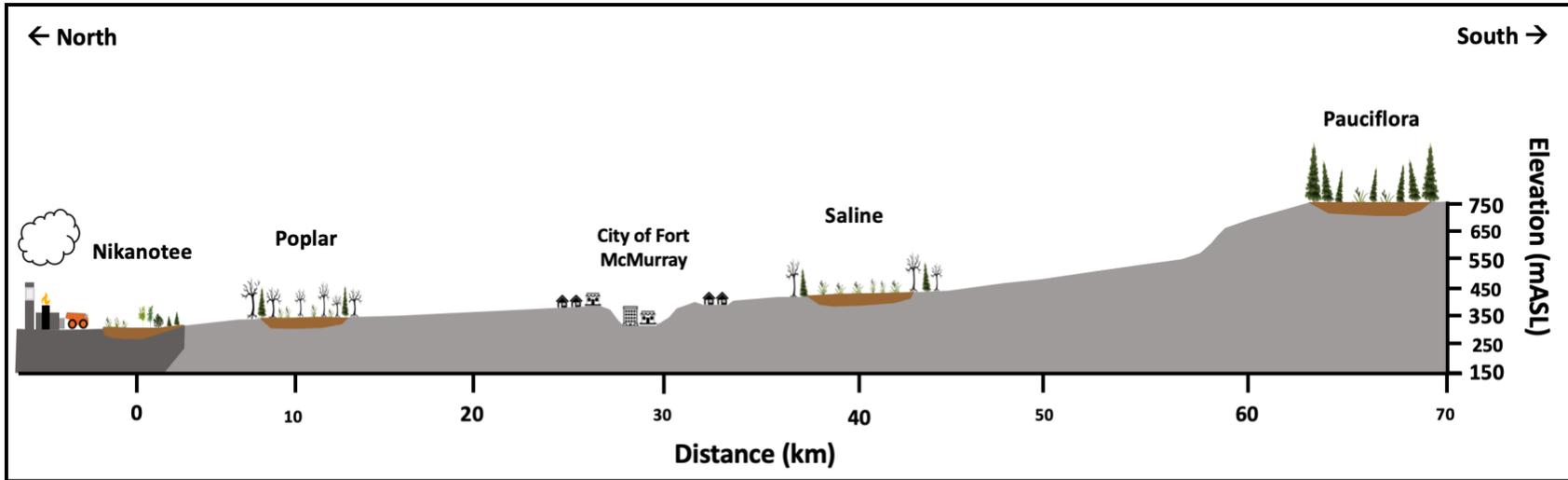


Figure 2.2: Illustration of the locations of all research sites with distance away from the reclaimed Nikanotee site (located on Suncor Energy Inc., dark grey area with smokestack to denote the open-pit mining footprint, 0 km). Each site is displayed at their respective elevation in meters above sea level (mASL). Study areas and vegetation are not drawn to scale. Note: Poplar is oriented 10 km west of Nikanotee and not directly south.

2.3 Materials & Methods

Atmospheric Nitrogen Deposition Collection, Analyses and Load Calculations

Passive atmospheric deposition collectors, known as ion-exchange collectors (IEC), were set up at each study site during the summer of 2018 from 7 May to 19 August (DOY 127 to 231; 105 days) (Nikanotee (n = 7), Poplar (n = 3), Saline (n = 3) and Pauciflora (n = 3)). All IECs were assembled following the methods of Brumbaugh et al. (2016). Briefly, each IEC was composed of polyvinyl chloride (PVC) and polypropylene components, including two pipes (1.27 cm x 15.24 cm) to create the main dual-stage resin column, and a plastic funnel (29.96 cm diameter). The upper PVC pipe (cation pipe) contained 17 ml cation exchange resin (*DOWEX™ 650C UPW*) and the bottom PVC pipe (anion pipe) contained 20 ml anion exchange resin (*DOWEX™ 550A UPW*). Clean poly-fil packing fibre was inserted in both ends of each pipe to keep the resin beads within their respective pipe. Both cation and anion pipes were then attached together with threaded PVC couplings/fittings. The IEC columns used for field sampling were open at both ends (not capped) and had a plastic funnel (29.96 cm diam. avg.) attached to the top of the column. This created an open column that permitted water flow via percolation through the entire column and out onto the ground. Water slowly moved through the column and ions in solution retained on the cation and anion exchange resin beads that were later extracted after the sampling period. A completely closed (capped) field blank IEC was installed at every site. All IECs were rinsed with ultra-pure deionized water prior to field installation.

During transportation, all IECs were kept upright and capped to avoid contamination. During installation, caps were removed, funnels were attached and sealed to the top of the cation pipe, nylon netting (1.2 cm mesh size) was placed over top of the funnel (to prevent leaves from entering) and metal stakes were taped around the outer rim of the funnel to keep birds away. All

IECs and field blanks were clamped and taped to wooden fence posts at an average height of 2.26 m. All IECs were installed in open areas at each site to avoid interception of deposition from trees.

At the end of the sampling period (end of August) columns were capped, placed upright in a cooler, and transported back to the Hydrometeorology Lab at University of Waterloo, where they were then stored in a refrigerator at 4°C until extraction. Cation exchange resin (ammonium, NH₄⁺ analysis) was extracted with four rinses of 50 ml 1M KCl. Similarly, anion exchange resin (nitrate, NO₃⁻ analysis) was extracted with four rinses of 50 ml 1M KI. The extracted 200 ml eluent was filtered and analyzed for NH₄⁺-N and NO₃⁻-N using colorimetric analysis at the Biogeochemistry Lab at University of Waterloo (Bran Luebbe AA3, Seal Analytical, Seattle, U.S.A., Methods G-102-93 (NH₄⁺-N) and G-109-94 (NO₃⁻-N)). Total inorganic N (TIN) was estimated as the sum of NO₃⁻-N and NH₄⁺-N. N deposition loads were estimated using,

$$N \text{ load} = \left(\frac{C * V}{A} \right) * 15.5 \quad (1)$$

where C is the measured NH₄⁺-N or NO₃⁻-N concentration (mg L⁻¹), V is the total extract volume (L) of KCl or KI eluent, A is the area of the funnel (in²) and 15.5 is the factor to convert from mg in⁻² to kg ha⁻¹ (Brumbaugh et al., 2016).

Meteorological Variables

Ambient air temperature and relative humidity (RH) sensors attached to meteorological (MET) towers (HC2S3 temperature and relative humidity sensor, Campbell Scientific, Logan, UT; ~3 m height) at each study site were sampled at 10 s intervals, averaged over 30-minute intervals, and recorded on dataloggers (CR1000, Campbell Scientific, Logan, UT) from May to September 2018. Daily precipitation (P) was recorded using tipping bucket rain gauges (Nikanotee and Pauciflora: Texas Instruments TR-525M; Poplar and Saline: Rg3-M, Onset, USA) located in open areas at each study site. P data for Saline was gap filled for a rain event from June 1-3rd 2018 using

Environment and Climate Change Canada (ECCC) Data for P from the Fort McMurray airport, situated 9 km north of Saline (Environment and Climate Change Canada, 2017). The RH sensor at Saline malfunctioned for most of the summer, therefore ECCC Data for RH from the Fort McMurray airport was used as a substitute for the study period. Wind data was obtained from the ECCC Data for the Fort McMurray airport (Environment and Climate Change Canada, 2017). Airport wind data was used as this provided a more accurate representation of wind speed and direction at the regional scale.

Statistical Analysis

All statistical tests and analyses were performed using R (Rstudio Team, 2016). Data was first tested for normality using the Shapiro-Wilkinson distribution test ($p > 0.05$). Nitrate was found to be normally distributed and as such was analyzed using a one-way parametric ANOVA test to determine differences among group means ($p < 0.05$ significance). This was followed by a post-hoc pairwise t-test test to examine significant differences between groups. Ammonium and TIN were found to be normally distributed (after being log-transformed) and were then also analyzed using a one-way parametric ANOVA test ($p < 0.05$ significance), followed by a post-hoc pairwise t-test.

2.4 Results

Climate Conditions

Mean daily air temperatures (for May 7 to August 19, 2018) were approximately 2 °C warmer at Nikanotee and Poplar than at Saline and Pauciflora, although all sites were warmer than climate normal for the same period (14.3 °C; Environment and Climate Change Canada, 2017) (Table 2.1). Mean RH across all study sites over the same time period was 61.4%, with the lowest RH at Saline (57.8%) and highest at Pauciflora (66.8%). Similarly, total P over the study period was lowest at

Saline (175.8 mm) and highest at Pauciflora (352.4 mm). Although Saline and Pauciflora are situated close together, Pauciflora received twice as much P due to the higher elevation of the site (Figure 2.2). Total P at Nikanotee and Poplar fell within the range of highest and lowest recorded values for all sites and received P amounts (Table 2.1) typical of climate normal (244.6 mm; Environment and Climate Change Canada, 2017).

Wind direction was primarily from a SW direction throughout the study period at Fort McMurray airport, indicating that this is the dominant regional flow direction (Figure 2.3). Frequency distributions of wind direction at Fort McMurray airport show that the predominant winds coming from the SW, W and NW occurred ~48% of the time. The highest wind speed at Fort McMurray airport was recorded as coming predominantly from the NW and W directions with an average wind speed of 3.2 ms⁻¹ over the summer 2018 study period.

Table 2.1: Climate conditions over the study period (May 7 to August 19, 2018) at all sites in the Athabasca Oil Sands Region, Alberta.

Site	Mean Air Temperature (°C)	Max Air Temperature (°C)	Min Air Temperature (°C)	Mean RH (%)	Total Precipitation (mm)
Nikanotee	18.6	29.5	7.3	59.4	236.1
Poplar	18.5	30.7	7.0	62.0	270.0
Saline	16.7	27.3	6.1	57.8	175.8
Pauciflora	15.9	26.8	3.9	66.8	352.4

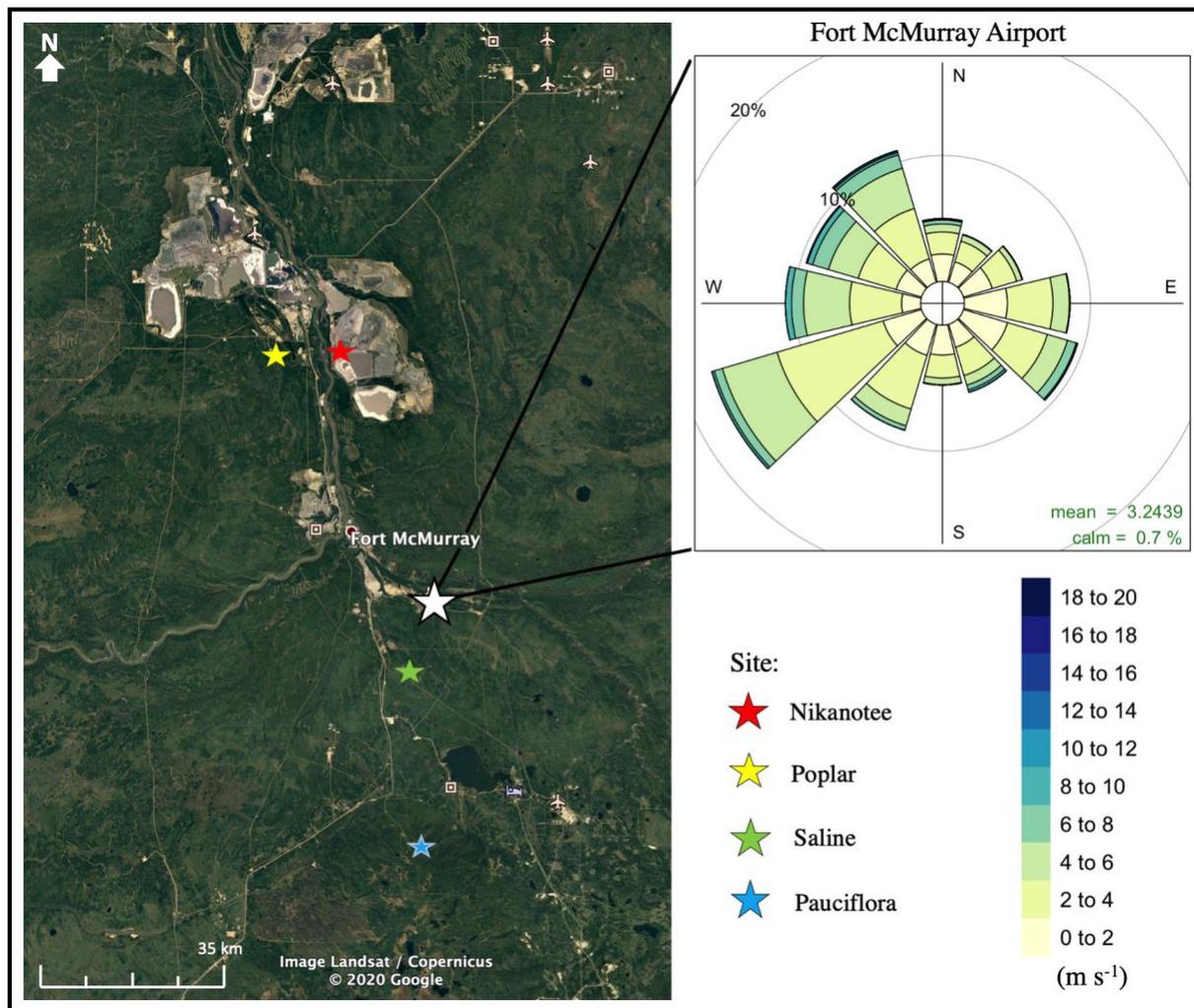


Figure 2.3: Half-hourly wind speed (m s^{-1}) and direction ($^{\circ}$) for the Fort McMurray airport (369 mASL) over the May-August 2018 monitoring period with associated frequency counts (%), Athabasca Oil Sands Region, Alberta. Airport wind data was used as it provides a more accurate representation of wind speed and direction at the regional scale.

Nitrogen Deposition

The N deposition loads of $\text{NO}_3\text{-N}$, $\text{NH}_4\text{-N}$ and TIN were greatest at Nikanotee (reclaimed site) relative to the three reference sites. Total inorganic N was largely dominated by $\text{NH}_4\text{-N}$ (>90% for Nikanotee, Poplar and Saline, and ~79% for Pauciflora). Both $\text{NH}_4\text{-N}$ and TIN deposition generally decreased with distance from 0-65 km (Figure 2.4B & C); however, significant differences were only observed between the industrial centre (Nikanotee, 0 km) and the farthest site (Pauciflora, 65 km) ($p = 0.02$). Between 0-65 km, average $\text{NH}_4\text{-N}$ deposition loads ranged

from 0.92 kg N ha⁻¹ (± 0.37) to 3.79 kg N ha⁻¹ (± 2.37). Similarly, average TIN deposition loads ranged from 1.16 kg N ha⁻¹ (± 0.38) to 4.07 kg N ha⁻¹ (± 2.40).

Nitrate did not display the same spatial pattern with distance from the industrial centre. Nitrate decreased from Nikanotee to Saline (40 km away), at which point NO₃-N increased again at Pauciflora (65 km away) (Figure 2.4A). Although NO₃-N was statistically higher at Nikanotee ($p < 0.05$), no differences were found between the three natural sites, regardless of distance ($p > 0.05$). Average NO₃-N deposition loads ranged from 0.22 kg N ha⁻¹ (± 0.016) to 0.33 kg N ha⁻¹ (± 0.045).

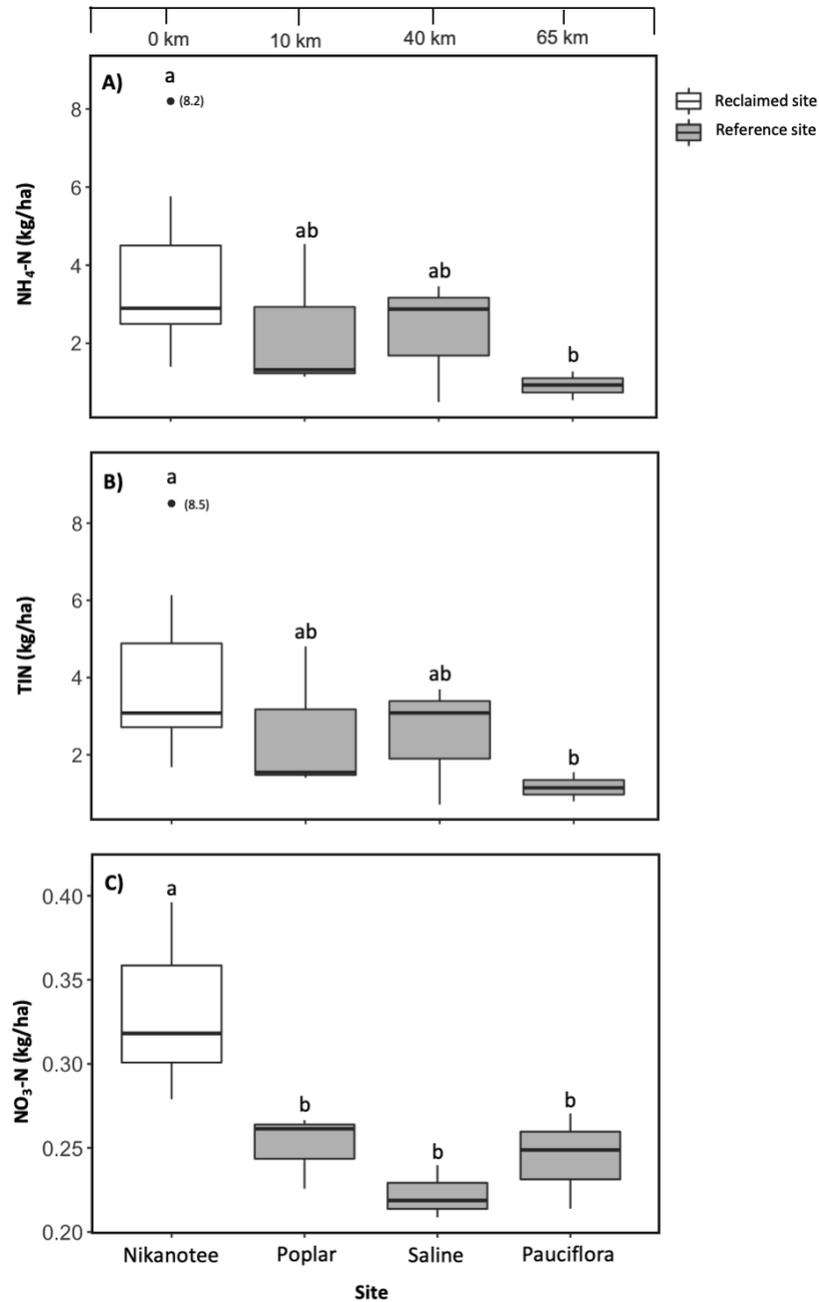


Figure 2.4: Box plots of (A) ammonium (NH₄⁺), (B) total inorganic N (TIN) and (C) nitrate (NO₃⁻) deposition loads at each study site in the Athabasca Oil Sands Region, Alberta. Means with different letters indicates a significant difference (p<0.05).

Larger Scale Spatial Patterns

Nitrogen deposition loads from the current study, along with additional values from the literature were compiled (Figure 2.5) to explore spatial patterns in N deposition loads relative to the

Syncrude Smokestack (57°2'52.75"N, 111°36'55.81"W; left-hand smokestack in Figure 2.1). All distances for sites in this study were corrected to reflect the Syncrude stack as distance zero. Therefore, Nikanotee, Poplar, Saline and Pauciflora are located 17, 13, 57 and 78 km away from the Syncrude smokestack, respectively. To compare with literature values, only 'summer' N deposition data (i.e., no 'winter') were used for comparison of spatial patterns for consistency. As the timeframes between this study (four-month sampling period) and that of the literature (six-month period) (Fenn et al., 2015; Proemse et al., 2013) were slightly different, only the relative spatial distributions of N deposition with distance south from the Syncrude smokestack were compared. It should also be noted that N deposition from the literature is from summers 2007 and 2008, ten years before this study.

A decreasing trend in NH₄-N, TIN and NO₃-N deposition loads was observed moving southwards from the stack. Sites situated near the stack (<17 km) are especially high, particularly for industrial sites. Although sites near the stack are quite high, there is variability and reduction in N species with distance south does not appear to be linear for both site types (i.e., industrial and natural). Overall, at a given distance from the stack, the industrial sites appear to have a greater N deposition load than the natural sites (Figure 2.5).

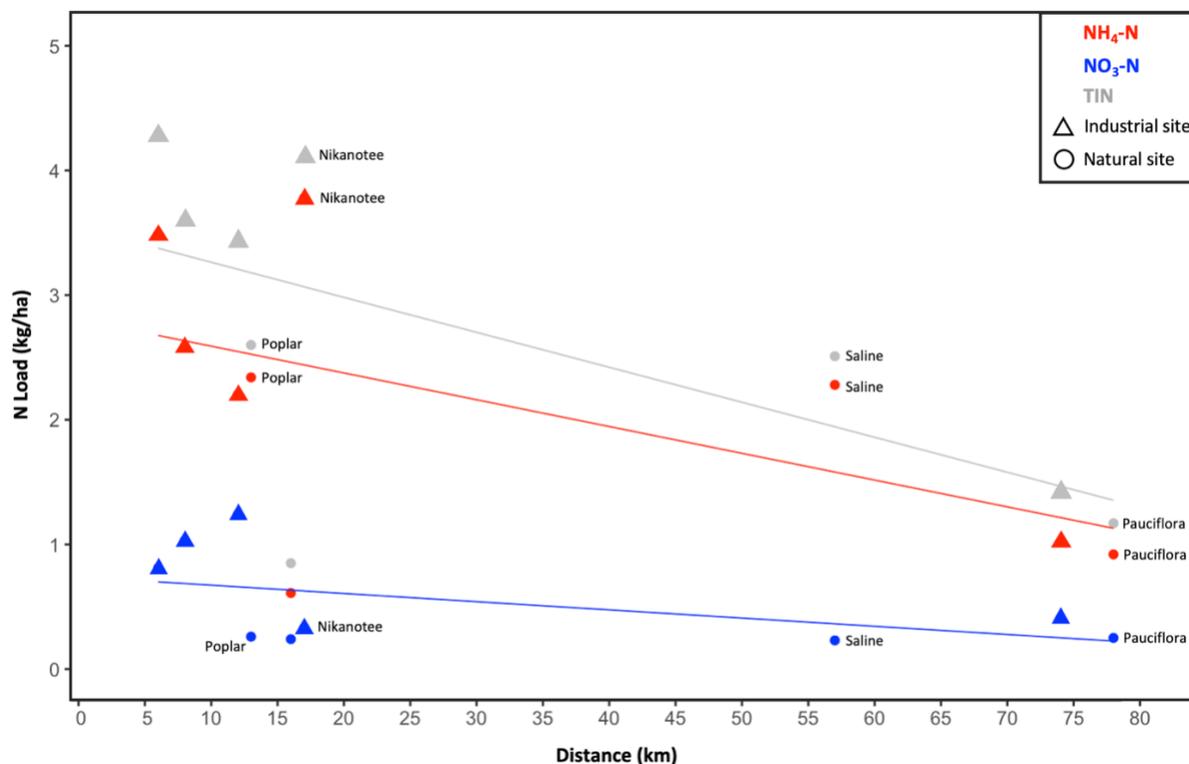


Figure 2.5: Spatial comparison of published N deposition (unlabeled shapes) versus results from this study (labeled shapes) of $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$ and TIN (kg ha^{-1}) as a function of distance from the Syncrude Canada Ltd. emissions stack towards the south of the industrial area in the Athabasca Oil Sands Region, Alberta. Industrial sites (including the reclaimed site) are denoted by the triangles and natural sites (including the reference sites) are denoted by circles. Note that only N deposition data from the literature at sites south of the industrial area were included to keep the spatial direction the same as this study. Only the spatial distribution of N is compared.

2.5 Discussion

Nitrogen Deposition Loading

The Nikanotee site, which was located directly within the primary oil sands source area, received the greatest N deposition loads over the 2018 summer season, compared to the three reference sites. This finding is consistent with previous N deposition studies conducted in the region that similarly reported the highest N deposition loads at sites located directly within (or closest to) the oil sands mining area (Fenn et al., 2015; Proemse et al., 2013; Hsu et al., 2016; Bytnerowicz et al., 2010). A major contributor to elevated N deposition at study sites located within the vicinity of oil sands activities is dry deposition from dust kick-up and particulate matter emissions (Proemse et

al., 2013). Dry deposition can freely deposit onto open ecosystems but can also accumulate on vegetative surfaces and wash off and deposit in concentrated quantities on the soil surface during rain events (Proemse et al., 2013). Nikanotee is bordered by major haul roads and often experiences dusty conditions from neighbouring mining activities. These roads are likely sources of dry N deposition to the reclaimed site. These high dry N loadings contribute to its overall increased N deposition, especially in terms of $\text{NH}_4\text{-N}$ deposition.

Ammonia (NH_3) emissions and their transformation into secondary $\text{NH}_4\text{-N}$ are concerning due to potential biological effects on surrounding ecosystems (i.e., increased N-status, growth, and productivity) and the greater overall contribution of $\text{NH}_4\text{-N}$ to N deposition (Bytnerowicz et al., 2010; Fenn et al., 2015). A considerable amount of NH_3 is readily deposited <5 km from its source. However, during dry summer conditions, NH_3 can be transported far away from its source, resulting in long-range $\text{NH}_4\text{-N}$ deposition (Whaley et al., 2018; Hsu and Bytnerowicz, 2015). Nikanotee received the highest $\text{NH}_4\text{-N}$ deposition due to its close proximity to stack and diesel emissions. Both Poplar and Saline received the next highest $\text{NH}_4\text{-N}$ deposition loads that were approximately equal for these two sites, even though Poplar is situated right next to the mining area and Saline is 40 km south. Sites located within 20-30 km distance to the main oil sands mining and upgrading area are significantly affected by industrial contributions of both $\text{NH}_4\text{-N}$ and $\text{NO}_3\text{-N}$ deposition (Lynam et al., 2015; Landis et al., 2012; Proemse et al., 2013). Poplar likely received moderate dry N deposition loads as it is surrounded by dirt roads and situated less than 20 km from the operations area, where the plume of dusts and particulates (dry N) often reach (Watmough et al., 2014; Proemse et al., 2013). However, despite the close proximity of Poplar to the main mining operations, the regional SW wind direction may have assisted in reducing overall N deposition to the site. Saline received comparable $\text{NH}_4\text{-N}$ loads to Poplar even though it is located further away

from the main mining area. The similarity in $\text{NH}_4\text{-N}$ loads is likely a result of the close proximity of Saline to the city of Fort McMurray, which is rapidly developing and expanding, creating a separate source of urban N emissions in the region (Proemse et al., 2013), especially dry N deposition. This is likely the main contributor of $\text{NH}_4\text{-N}$ deposition at Saline, since dry deposition is largely composed of $\text{NH}_4\text{-N}$ rather than $\text{NO}_3\text{-N}$ (Bytnerowicz et al., 2010). As expected, Pauciflora received minimal $\text{NH}_4\text{-N}$ loads since it is located much further away from the main mining source area, on a significant topographical high, and isolated in the Stony Mountain forested area.

Similar to $\text{NH}_4\text{-N}$ deposition, $\text{NO}_3\text{-N}$ deposition was highest at Nikanotee, which is consistent with previous studies that found sites located within industrial activities to be subjected to higher $\text{NO}_3\text{-N}$ loads than more distant sites (Proemse et al., 2013; Fenn et al., 2015). Nitrate has a short atmospheric lifetime of about one day, resulting in $\text{NO}_3\text{-N}$ deposition concentrating close to emission sources (Hsu et al., 2016). Moving southwards from Nikanotee, $\text{NO}_3\text{-N}$ deposition decreased by 33% up to a distance of 40 km (Saline), at which point $\text{NO}_3\text{-N}$ deposition increased 65 km away at Pauciflora. There are multiple factors that may have contributed to the observed increase in $\text{NO}_3\text{-N}$ deposition at Pauciflora, located furthest away from the industrial centre in this study. Wood et al. (2016) found that extractable $\text{NO}_3\text{-N}$ (ext- $\text{NO}_3\text{-N}$) concentrations in peat were highest at an industrial site located adjacent to a dirt road at the Japan Canada Oil Sands (JACOS) site. This increased ext- $\text{NO}_3\text{-N}$ in peat was attributed to an exogenous N source, likely due to proximity to a dirt road regularly used by vehicles. Similarly, Pauciflora is partially bisected by a dirt road at the northern part of the fen. During summer 2018, vehicles frequently used the road for recreational activities, research, and construction. Thus, Pauciflora may have also received an increased load in $\text{NO}_3\text{-N}$ deposition due to dirt road activities and resulting dust “kick-up”.

Conversely, increased $\text{NO}_3\text{-N}$ deposition may have been due to increased P at the study site, which was double that at Saline and the highest compared to Poplar and Nikanotee, located next to the industrial area. High total rainfall may also have helped scavenge $\text{NO}_3\text{-N}$ particles within the atmosphere, however, this would likely have also been reflected in $\text{NH}_4\text{-N}$ deposition (Lynam et al., 2015; Whaley et al., 2018). An additional exogenous source that should be considered is $\text{NO}_3\text{-N}$ emissions from wildfires. Fenn et al. (2015) suggested that an increase in $\text{NO}_3\text{-N}$ deposition at their study sites was possibly due to wood burning fires that burned in close proximity, which resulted in $\text{NO}_3\text{-N}$ particles from aged smoke plumes depositing on their sites. Summer 2018 experienced wildfires and smoky atmospheric conditions, particularly at the beginning and end of the season. Nitrate emissions from wildfires that burned south of Stony Mountain near the hamlet of Janvier, may have contributed to $\text{NO}_3\text{-N}$ deposition loads at this site. This demonstrates that not only are the activities from industrial oil sands an exogenous N source to the surrounding environment, but there are several other sources that need to be considered (i.e., wildfires, urban centres) when conducting depositional studies.

Pauciflora may have also exhibited higher $\text{NO}_3\text{-N}$ loads due to the site's close proximity to in-situ SAGD operations located around the Stony Mountain area. Wood et al. (2016) suggests the increase in ext- $\text{NO}_3\text{-N}$ in peat could also be due to the location of the site on a SAGD area composed of decommissioned well pads. SAGD operations are known to emit considerable NO_x emissions that are comparable to NO_x emissions from surface mining operations due to the amount of energy needed for steam generation (Moorhouse et al., 2010; McWhinney, 2014). The predominant SW and often NW winds that averaged just over 3 ms^{-1} may have transported NO_x emissions towards Pauciflora. A final reason for higher $\text{NO}_3\text{-N}$ deposition at Pauciflora may be because it is the most southern site in this study, located closest to the agricultural areas of

Edmonton, Athabasca and surrounding regions (Hsu et al., 2016). This is unlikely, however, as this was not reflected in $\text{NH}_4\text{-N}$ deposition. Although, long-range transport of NH_3 from agricultural activities does occur (particularly in low RH conditions, which are found in this region), considerably higher $\text{NH}_4\text{-N}$ deposition should also be observed at Pauciflora, since agriculture is a major emitter of NH_3 (Hsu et al., 2016).

Spatial Distributions of N Deposition

Results from this study show a decreasing trend in deposition for all inorganic N species with distance from the oil sands industrial centre, with $\text{NH}_4\text{-N}$ being several times greater than $\text{NO}_3\text{-N}$ (>3.7 times) at all sites. These findings are similar to previous depositional studies conducted in the AOSR that not only show a decreasing spatial pattern in N deposition, but also indicate a similar decreasing spatial pattern (typically exponential) for a number of other pollutants including sulfur (S), heavy metals and base cations (BC) with distance from the primary oil sands operations (Fenn et al., 2015; Wieder et al., 2016a; Hsu et al., 2016; Proemse et al., 2013).

Previous studies have found that $\text{NH}_4\text{-N}$ deposition is consistently greater than $\text{NO}_3\text{-N}$ deposition and has been shown to be up to 7.6 times greater over the summer at industrial sites located 6-8 km from the industrial centre (Proemse et al., 2013; Fenn et al., 2015). This study shows the greatest $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio to be 11.5 at Nikanotee, which is likely showing an increase in NH_3 emissions (from expanding mining developments) and thus, $\text{NH}_4\text{-N}$ deposition over the years. Conversely, this higher ratio could be reflective of a reduction in NO_x emissions (and thus reduced $\text{NO}_3\text{-N}$ deposition) from the implementation of emission control technologies such as NO_x reduction catalyst systems (Puckett, 2015; M.J. Bradley & Associates, 2008).

Average N deposition over the summer season (from 2008-2011) at an open industrial site located 3 km from the Syncrude stack (i.e., within the industrial zone) received N deposition loads

of 0.99, 1.8 and 2.8 kg ha⁻¹ for NO₃-N, NH₄-N and TIN respectively (Fenn et al., 2015). Although quantitative comparison of deposition loads between studies is not feasible, percent decrease is comparable. Fenn et al. (2015) found that NO₃-N, NH₄-N and TIN were reduced by 39, 24 and 30%, respectively at 20 km from the industrial centre, demonstrating a gradual decrease in N deposition at just 20 km from the main source area. It should be noted that deposition levels at 20 km in Fenn et al. (2015) are predicted from curve fitting equations that use empirical deposition data. Between 3 km (industrial levels) and >113 km (background levels) Fenn et al. (2015) found that NO₃-N, NH₄-N and TIN deposition were reduced by 74, 67 and 70%, respectively, over the same summer period. Similarly, this study found the largest percent decreases of NH₄-N and TIN deposition to be 76 and 71%, respectively, between Nikanotee (industrial) and Pauciflora (background). Between Nikanotee and Pauciflora, NO₃-N decreased by just 26%. However, as discussed above, there are multiple factors that likely contribute to this relatively elevated NO₃-N load at Pauciflora. Although there is a difference in distance (>30 km) and approximately 8 years between Pauciflora and the furthest sites in Fenn et al. (2015), both studies found a decrease of >65% in NH₄-N and TIN deposition at sites located within industrial activity and the distant, non-industrial locations. This confirms that sharp decreases in NH₄-N and TIN deposition occur over relatively short distances from the main emissions source.

Potential Ecological Effects of N Deposition

Nitrogen deposition affects ecosystems by acting as: (1) a nutrient enriching agent; and/or (2) an acidifying agent (Lovett, 2013). Nikanotee is the most susceptible ecosystem to N enrichment for all sites in this study as it received the highest N deposition loads and is situated directly within major oil sands activities, where atmospheric N is elevated (Fenn et al., 2015). As Nikanotee is a young, reclaimed site (~6 years old), N deposition may be acting as a fertilizing agent by

stimulating plant growth and productivity on the reclaimed ecosystem (Greaver et al., 2012). However, at the same time, N deposition inputs may also be skewing vegetation cover to a more N-tolerable community, which would result in a shift to a more vascular plant cover and a reduction in species richness (Bobbink et al., 1998; Macdonald, 2015; Bubier et al., 2007). With time, as N accumulates within the system, N deposition may become a chronic input to the site, increasing the N-status of ecosystem receptors, which could result in N saturation and shifts in population dynamics, species composition and overall community structure (Greaver et al., 2012). In terms of acidification, there is little concern at Nikanotee as the site is likely receiving equivalent amounts (or more) of buffering BC deposition from fugitive dust, however, this is sensitive to temporal change as industrial activities evolve (Watmough et al., 2014; Landis et al., 2012; Watmough, 2015).

Poplar is likely the most susceptible reference site to N deposition, however, only in terms of the N enriching effects of N deposition as Poplar is located within the high BC deposition zone (Watmough et al., 2014). Nitrogen deposition may also be acting as a fertilizing agent and may be increasing the N-status of soil, water, and vegetation (Greaver et al., 2012). A study conducted at Poplar (pre-2016 wildfire) found that peat and groundwater were dominated by N, but mosses and vascular vegetation were N-limited (Nwaishi et al., Under Review). High N content in peat and groundwater could be a result of long-term exogenous N deposition inputs, however, it is expected that this would also be reflected in foliar N concentrations (Aber et al., 1989). As such, N deposition might not currently be a chronic input to Poplar. Focus should be towards foliar N concentrations of vascular vegetation since moss appears not to be influenced by N deposition and is thus a poor indicator (Wieder et al., 2016b).

Saline is the next most susceptible reference site to N deposition, located outside of the previously determined high BC deposition zone (Watmough et al., 2014). As such it likely receives far less BC deposition than Nikanotee and Poplar, making the site more susceptible to the acidifying impacts of N deposition. Saline may be experiencing an increasing fertilization effect with the expansion of the city of Fort McMurray just north of the study site as well as a potential increase in soil acidification due to less BC buffering sources. This would result in a direct increase in porewater acidification and H⁺ uptake by vegetation (Wieder et al., 2016b). However, due to the underlying geology, the geochemistry of surface water is relatively neutral to base-rich (Volik et al., 2018). Thus, Saline may have an inherent ability to buffer the acidic effects of N deposition, making the nutrient enriching impacts of N deposition more pertinent.

Pauciflora is likely the least susceptible reference site to N deposition both in terms of enrichment and acidification. Although Pauciflora did not receive the lowest NO₃-N deposition load, total N deposition load was far less than all other sites indicating that it is the site the least impacted by N deposition in this study. Proximity to a dirt-road situated at the northern edge of the site, however, may introduce BC deposition loads that would buffer the potential acidic inputs of N depositing on site (Landis et al., 2012). Vegetation cover at Pauciflora includes some lichen species, which are highly sensitive and good indicator species of N deposition both in terms of N enrichment and acidification (Wieder et al., 2016b; Fenn et al., 2011). Future studies should include lichen sampling to examine just how susceptible Pauciflora is to N deposition.

In general, the nutrient enrichment (fertilization) effects of ecosystems in the AOSR align well with the N deposition gradient from the oil sands industrial centre. However, other sources of N deposition in the region (i.e., SAGD) may create concentrated N patches in other areas. Increased N deposition may result in a shift from a diverse ecosystem with lichen and mosses, to

a more simplified ecosystem of vascular vegetation and a reduction in peatland diversity (Bobbink et al., 1998; Macdonald, 2015; Bubier et al., 2007; Lamers et al., 2000). Increased vascular vegetation due to high N deposition will increase the productivity and evapotranspiration rates of peatlands in the AOSR and may lead to reduced rates of carbon accumulation (weakened carbon-sink function) as a result of changes to peat litter quality (i.e., increased vascular litter) and species composition (Bubier et al., 2007; Bragazza et al., 2006; Lamers et al., 2000). In general, acidifying effects of ecosystems in the AOSR will be of little concern to ecosystems located in close proximity to fugitive dust sources near oil sands mining (BC buffering) and will increase moving further away from the industrial centre (Watmough et al., 2014). However, impacts of acid deposition (i.e., decreased species diversity) to ecosystems will likely vary depending on close proximity to dirt roads (source of BC) and site-specific characteristics such as buffering capacity.

2.6 Conclusion

Ammonium and TIN deposition were greatest near the industrial centre and decreased with distance southwards. Results from this study indicate that even at 40 km south of the main industrial area, N loads are comparable to more industry affected sites. This study looked at sites in the AOSR that were relatively close in proximity (<70 km) compared to the literature (>100 km) (Fenn et al., 2015; Hsu et al., 2016; Proemse et al., 2013; Wieder et al., 2016a; Edgerton et al., 2020). In general, the spatial pattern of decreasing N deposition with distance southwards holds true for this study, with TIN decreasing 71% from 0-65 km. Susceptibility of nutrient enriching impacts from N deposition are expected to align with the spatial pattern of N deposition, however acidifying impacts likely vary due to different sources of buffering BC deposition.

This study is the first to examine exogenous N inputs to a reclaimed site relative to reference sites in the AOSR. Findings from this study provide a better understanding of N deposition loadings to each site and how this may contribute to greater nutrient cycling in young systems. Exogenous N sources are important to continually examine with expanding industrial (and urban) activities due to their potential ecological effects on the surrounding environment. Quantification of N deposition loads will assist in the creation of an integrative nutrient framework that will allow a better understanding of the biogeochemical functioning of reclaimed and natural peatlands in the AOSR. Furthermore, this knowledge will assist understanding how reclaimed and natural peatlands are impacted by exogenous N inputs and will aid in predicting the future of these peatland ecosystems in the AOSR.

3.0 Manuscript Chapter 2. Nitrogen deposition influence on the nitrogen-status and development of a reclaimed fen-upland in the Athabasca Oil Sands Region, Alberta

3.1 Introduction

Surface mining in the Athabasca Oil Sands Region (AOSR) situated in the sub-humid region of northeastern Alberta has resulted in the alteration of over 895 km² of boreal forest (Government of Alberta, 2021). The pre-mined AOSR landscape was comprised of 64% wetlands (~90% of which were fen peatlands) and 23% forest uplands (Rooney et al., 2012; Daly et al., 2012). To reach bituminous oil sand, the wetland-upland mosaic is removed, and vital ecosystem services that these ecologically significant environments provided, including water storage, ecological biodiversity, carbon storage and nutrient cycling, are lost (Nwaishi et al., 2015). Once surface mining ceases, the Alberta Government requires oil companies to reclaim the disturbed land to an equivalent pre-mined state that consists of native boreal plant and wildlife species (Natural Resources Canada, 2016). The construction of a fen peatland – forest upland system at the Nikanotee Fen Watershed is a pilot attempt to reclaim the post-mined landscape back to a self-sustaining fen-upland mosaic (Daly et al., 2012). However, during reclamation, nearby disturbances, specifically air pollution emissions and deposition, as a consequence of ongoing mining development, pose a biogeochemical and ecological concern to the reclaimed system.

Environmental concerns regarding air pollution and atmospheric deposition of nitrogen (N) compounds have been raised in the AOSR, since N emissions and subsequent deposition are expected to increase with expanding mining operations and related development (Vitt et al., 2003; Laxton et al., 2010). Nitrogen emissions originate from point, mobile and fugitive sources, which include smokestacks, mine fleets and dusts, respectively (Hsu et al., 2016). These N deposition loads have historically remained quite low, ~1 kg N ha⁻¹ yr⁻¹ (Vitt et al., 2003) across most of the

AOSR. However, mining development has led to increasing loads of $17 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ in the centre of operations (Wieder et al., 2019) and up to $24.3 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ on post-mined uplands (Hemsley et al., 2019). Inorganic N deposition, particularly as ammonium (NH_4^+) and nitrate (NO_3^-), are a concern to boreal ecosystems that have historically persisted under low N conditions (Laxton et al., 2012). These inorganic N forms are created through atmospheric transformations of ammonia (NH_3), nitric oxide (NO) and nitrogen dioxide (NO_2) (Galloway et al., 2004; Li et al., 2016). Greater NH_4^+ and NO_3^- availability may increase the N-status of these sensitive aquatic and terrestrial ecosystems, which could lead to N saturation and shifts in plant community composition (Laxton et al., 2012; Wieder et al., 2019).

Ecosystems in which N retention capacity is exceeded, and bioavailable N overwhelms biotic and abiotic demand, are considered N saturated (excess N availability) (Aber et al., 1989; Fenn & Poth, 1998; Stoddard, 1994). Nitrogen saturation occurs as a cascade of events where an ecosystem progresses from being strongly N deficient to strongly N sufficient; starting with satisfying vegetation N demand, followed by soil microbial N demand (Stoddard, 1994; Aber et al., 1989). Thus, a large soil-N pool, typically indicates increased N-status and fulfilment of vegetation N demand (Stoddard, 1994). Fulfilling vegetative and microbial demand is then followed by increased N availability and the inability of an ecosystem to process and retain increased N inputs, resulting in excess N being leached or emitted back into the environment in gaseous forms (i.e., net source of N) (Aber et al., 1989; Laxton et al., 2010). This may ultimately lead to reduced plant productivity and mortality (Aber et al., 1989), increased litter decomposition rates and acceleration of N dynamics (Wieder et al., 2019).

Aquatic and terrestrial ecosystems located within industrial activity may experience increased productivity due to high N availability, which is low in the boreal ecozone (Nwaishi et

al., 2016; Laxton et al., 2012). In general, most boreal ecosystems are efficient at retaining natural N inputs and thus are typically described as low N environments. However, chronic N inputs (even at low levels) represent a unique form of potential stress to these N-sensitive ecosystems (Aber et al., 1989; Wieder et al., 2019). Therefore, atmospheric N deposition has the potential to influence the N availability, cycling and overall status of a reclaimed ecosystem situated directly within the hub of industrial N emissions in the AOSR.

Due to the constructed nature of the reclaimed fen-upland, analyzing the N-status of the system is likely to be complex, as it is only ~7 years post-construction and is still establishing its vegetation community and overall N-cycling regime. As such, it may be difficult to draw generalizations on the N-status of the site and its potential response to ongoing N deposition processes. The overall objective of this study is to investigate the contribution of N deposition to the N dynamics of the reclaimed Nikanotee fen-upland. Specifically, the research objectives of this study are to: 1) quantify bulk N deposition to the fen and upland; 2) analyze N dynamics in the soil-water-vegetation continuum; and 3) assess the potential implications of atmospheric N deposition on the fen-upland by investigating N-status based on N indicators of different ecosystem receptors. Several indicators of N-status were investigated in the fen and upland, including $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio, N solution chemistry profile, soil and foliar N concentrations, C:N ratio and soil N-cycling processes. It can be challenging to evaluate the N-status of any ecosystem (Laxton et al., 2010; Fenn & Poth, 1998), therefore the use of multiple complimentary indicators within the soil-water-vegetation continuum is necessary to effectively analyze N content, N dynamics and the potential risk of N saturation (Fenn & Poth, 1998). No single parameter can be relied upon for indicating N enrichment, and rather accumulated evidence from multiple parameters increases our understanding of ecosystem N-status and could help indicate N saturation

due to chronic N deposition (Fenn & Poth, 1998). It is hypothesized that due to the reclaimed site being situated directly within industrial activities, it would receive an N deposition load that influenced the N-status of the site, resulting in increased N content in the soil-water-vegetation continuum. This work will enhance our understanding of the recovery of N nutrient pools ~7 years post-construction.

3.2 Study Site

Field research for this study was conducted between May and August 2019 on the constructed Nikanotee Fen Watershed approximately 30 km north of Fort McMurray, Alberta (56°55'54.3"N, 111°25'01.9"W). The Nikanotee Fen Watershed is a young, post-mined reclaimed landscape that is located within the boreal mixed wood ecological region (Beckingham & Archibald, 1996). The reclaimed landscape consists of a main fen peatland – forested upland system that is surrounded by three previously reclaimed slopes, and one remnant natural slope (South Slope). The previously reclaimed slopes include the East Slope (built in 2007), and the Southeast Slope and West Slope (both built in 2011). The final two landscape units to be built on the constructed watershed were the fen peatland and forested upland (built between 2010-2013). The entire constructed Nikanotee Fen Watershed covers a total area of 32.1 ha and is bordered by industrial dirt roads and an active mine haul-road located ~300 m upslope to the east.

The focus of this research is on both the reclaimed fen and upland landscape units. The reclaimed system was built with an upland:fen ratio of 3:1 that is located over top of a series of hydrologically engineered layers, creating a connected system that is able to feed water from the upland downslope into the lower lying fen (Daly et al., 2012). The reclaimed fen-upland landscape was built to maintain a near-surface fen water table that is capable of withstanding water stress,

which would ultimately benefit both the fen and the upland during dry periods (Daly, et al., 2012; Ketcheson et al., 2016). While the hydrological design of the reclaimed fen-upland is crucial for its overall functioning, it's design must also be able to sustain biogeochemical and ecological processes and functions typical of Western Boreal Plain (WBP) landscapes (Daly et al., 2012).

The reclaimed fen is a 2.9 ha reclaimed fen peatland located at the base of the upland. The fen is comprised of a 2 m deep layer of moderately decomposed peat, transplanted from a nearby natural peatland site that had opened for development (Daly et al., 2012; Ketcheson et al., 2017). The 2 m of peat overlies a 0.5 m petroleum coke layer, which lies on top of a geosynthetic clay liner that extends below the entire fen-upland. The liner creates a hydrological disconnect with the overburden materials located below the constructed system (Ketcheson et al., 2017). The reclaimed upland is a 7.7 ha reclaimed forestland that was built on a 3% gradient to provide a water source for the connecting fen (Daly et al., 2012). The upland area includes a ~100 m section called the 'transition zone' located at the toe of the upland slope (Figure 3.1; Figure 3.2). The entire upland is comprised of 3 m of re-purposed tailings sand discarded from oil sands processing and capped by a thin (~0.3-0.5 m) layer of organic cover soil, called LFH-mineral mix (herein referred to as LFH). The LFH, which typically contains propagules (i.e., seeds) from its original location to aid in natural recovery (Mackenzie & Naeth, 2010), was salvaged from a nearby upland lease and stored prior to construction. The LFH of most of the upland is a forest floor mineral mix (FFM) amendment, however, the transition zone also includes some peat substrate (peat-mineral mix, PMM) incorporated into its cover soil (Gingras-Hill et al., 2018). Topographical features including hummocks (~1 m height) and open recharge basins are located at the mid- and high-slope of the upland. These features were included in upland construction to promote infiltration and groundwater recharge (Daly et al., 2012).

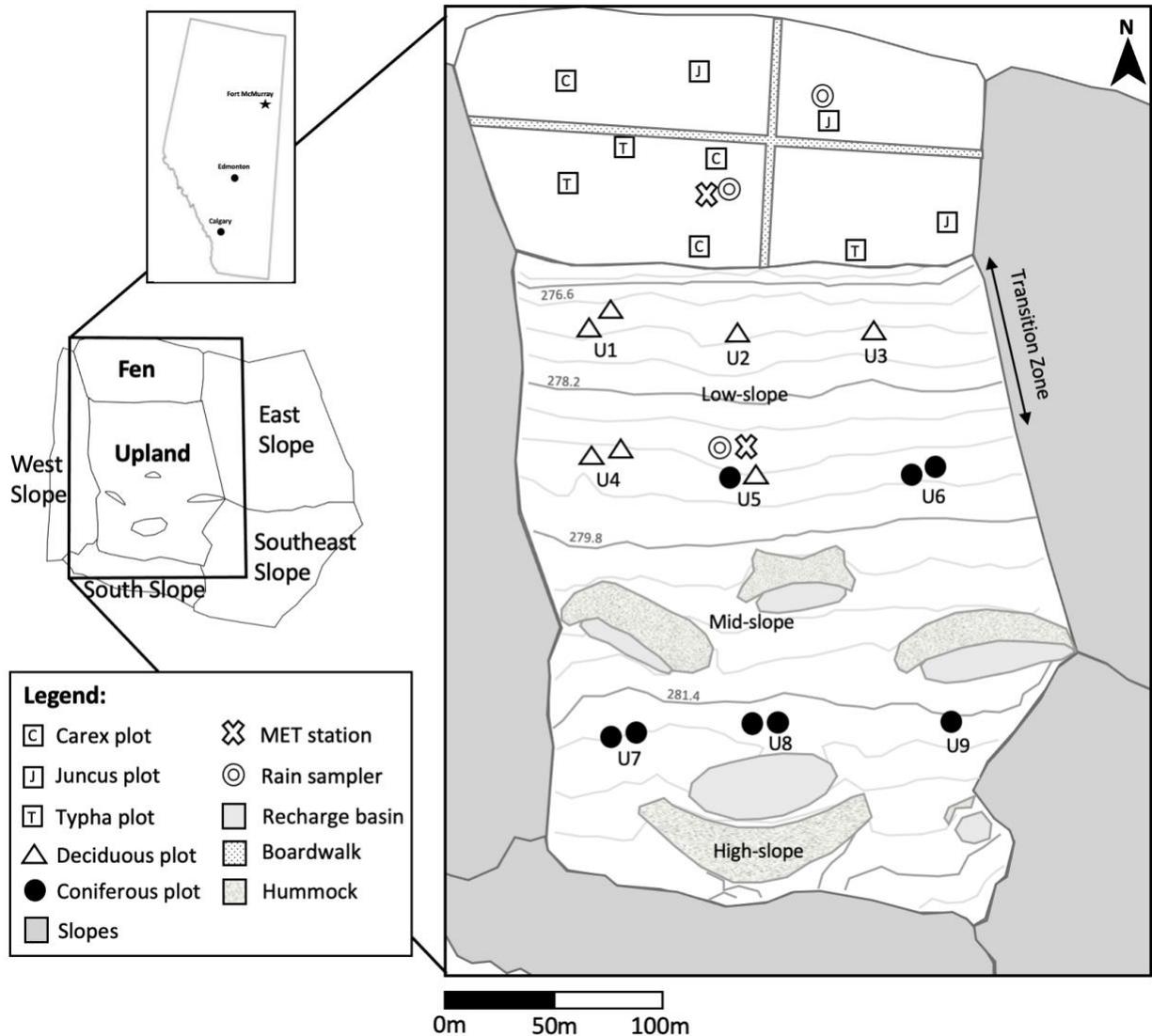


Figure 3.1: Map of the reclaimed fen-upland with monitoring plot locations. Nine fen plots covered the dominant vegetation species (N = 9 plots; carex n = 3, juncus n = 3, typha n = 3). Nine upland plots (N = 9 plots; U1-U9). Each upland plot included one or two of the dominant tree types (deciduous n = 7, coniferous n = 8).

Re-vegetation of the fen took place in June 2013. Experimental planting in the fen followed a randomized factorial design that included bare peat plots and the planting of mosses (e.g., *Tomenthypnum nitens* and *Sphagnum warnstorffii*) and seedlings (e.g., *Juncus balticus*, *Triglochin maritima*, and *Carex aquatilis*) (Scarlett et al., 2017). Fertilizer was not applied to the fen during or after construction. Details of the initial fen planting techniques are discussed in Borkenhagen & Cooper (2019). As of 2019, however, the fen is primarily dominated by three

vascular plant species including salt-tolerant juncus (*Juncus balticus*), freshwater carex (*Carex aquatilis*) and invasive typha (*Typha latifolia*).

Re-vegetation in the upland took place in 2013 and again in 2015. Re-vegetation involved the planting of a tree canopy consisting of jack pine (*Pinus banksiana*) and black spruce (*Picea mariana*), as well as an understory of various shrub and graminoid species (Gingras-Hill et al., 2018). In June 2015, saplings (>10,000 saplings total) were planted alongside a 10-g Continuum RT™ (18:9:9:9[S]) controlled release fertilizer (CRF) paper packet at a release rate of ~1,756 kg ha⁻¹. As of 2019 (six years post re-vegetation), the upland is dominated by a patchy distribution of scattered black spruce, jack pine, trembling aspen (*Populus tremuloides*) and balsam poplar (*Populus balsamifera*). Black spruce and jack pine are concentrated at the mid- and high-slope regions of the upland, whereas aspen and poplar are concentrated at the low-slope region. Between the low- and mid-slope region, there is a mix of all tree species. Aspen and poplar were not initially planted in the upland and were likely transplanted with the LFH or entered the reclaimed site by airborne seed dispersal. An understory vegetation survey conducted in the peak 2019 growing season determined that the understory is approximately 45% dominated by graminoid species (e.g., *Agropyron tracycaulum* and *Hordeum jubatum*), 26% bare ground, 13% forbs (e.g., *Sonchus arvensis* and *Taraxacum officiale*) and <4% shrub and alfalfa.



Figure 3.2: Photo of the constructed Nikanotee Fen Watershed from summer 2019. Photo taken on the East Slope

looking across the fen-upland towards the West Slope. Fen area is outlined in green, while the upland area is outlined in yellow (only half of the upland area is captured in this photo). A plume of dust over the constructed site from neighbouring industrial activities is apparent in this photo.

3.3 Materials & Methods

Sampling Design

Nine study plots were located in the fen (N = 9) and nine plots in the upland (N = 9) (Figure 3.1).

Within the fen, the nine monitoring plots included three each of carex, juncus and typha plots (n = 3 per species). Each plot was instrumented with one passive ion-exchange collector (IEC) (Brumbaugh et al., 2016), one lysimeter, one porewater sampler and a mineralization incubation. A groundwater well was situated in close proximity to each plot.

Six of the nine upland plots included two trees and three plots included one tree (Figure 3.1). In total, seven deciduous trees (n = 7) and eight coniferous trees (n = 8) were examined. Each plot was instrumented with one IEC, stemflow collector (per tree) and a mineralization incubation. A groundwater well was also situated in close proximity to each plot. Nutrient availability and dynamics were examined at each of these plots in the fen and upland between end of May to August 2019.

Atmospheric Nitrogen Deposition Collection, Analyses and Load Calculations

Ion-exchange collectors (IEC) were installed from 27 May to 19 August 2019 (DOY 147 to 231; 84 days) (fen (n = 9) and upland (n = 8)). The IECs were assembled following Brumbaugh et al. (2016). Briefly, each IEC was made of PVC and polypropylene components, including two pipes (each 1.27 cm x 15.24 cm) which formed the main dual-stage resin column, and a plastic funnel (29.96 cm diameter). The upper PVC (cation) pipe contained 17 ml cation exchange resin (*DOWEX™ 650C UPW*) and the bottom (anion) PVC pipe contained 20 ml anion exchange resin (*DOWEX™ 550A UPW*). Clean poly-fil packing fibre was inserted into both ends of each PVC

pipe to keep the resin beads from falling out. Both cation and anion pipes were then attached together with threaded PVC couplings/fittings. The IEC columns used for field sampling were open at both ends (not capped) and a plastic funnel was attached to the top. This created an open column that permitted water flow via percolation through the entire column and out onto the ground. Water moved through the column and ions in solution retained on the cation and anion resin beads that were later extracted in the laboratory. Completely closed (capped) field blank IECs were also installed. All IECs were rinsed with ultra-pure deionized (DI) water prior to field installation.

During transportation, all IECs were kept upright and capped to avoid contamination. During installation, caps were removed, funnels were attached and sealed to the top of the cation pipe, nylon netting (1.2 cm mesh) was placed over top of the funnel (to prevent leaves from entering) and metal stakes were taped around the outer rim to discourage birds from perching. All IECs and field blanks were attached to wooden and metal fence posts at an average height of 1.6 m in the fen and 2.1 m in the upland. Each IEC was installed in an open area above the canopy and collected bulk deposition, which is both wet and dry deposition.

At the end of the sampling period (end of August) columns were capped, placed upright in a cooler and transported back to the Hydrometeorology Lab at University of Waterloo, where they were stored in a refrigerator at 4 °C until extraction. Cation exchange resin (ammonium, NH_4^+ analysis) was extracted with four rinses of 50 ml 1M KCl. Similarly, anion exchange resin (nitrate, NO_3^- analysis) was extracted with four rinses of 50 ml 1M KI. The extracted 200 ml eluent was filtered and analyzed for $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ using colorimetric analysis at the Biogeochemistry Lab at University of Waterloo (Bran Luebbe AA3, Seal Analytical, Seattle, U.S.A., Methods G-102-93 ($\text{NH}_4^+\text{-N}$) and G-109-94 ($\text{NO}_3^-\text{-N}$)). Total inorganic N (TIN) was estimated as the sum of $\text{NO}_3^-\text{-N}$ and $\text{NH}_4^+\text{-N}$. N deposition loads were estimated using equation 1 in Cameron et al. (2021).

Water Sampling of Bioavailable N

Rainwater was collected in open areas (to ensure minimal effect of vegetation) in the fen and upland using water bottle rain gauges attached to 1.5 m wooden stakes ($n = 3$). A plastic funnel (10 cm diam.) was sealed to the neck of the bottle, nylon netting was placed over top of the funnel and metal stakes were taped around the outer rim of the funnel to ward off birds. Rain samples were collected within 24 hrs after a rainfall event and poured into clean, acid washed polyethylene bottles for N analysis. Due to destruction and contamination of samplers by wildlife, rainwater N was determined for the entire fen-upland, rather than both landscape units separately, a spatial distance over which precipitation should not vary.

Surface water infiltration was collected in the fen by installing plastic lysimeters ~5 cm below the peat surface. Mesh screening was attached to the top of the lysimeter to only allow water (shallow rainwater leachate) to be collected. Samples were collected within 24 hrs of rainfall events in a clean, acid washed polyethylene bottle for N analysis. Mid-way through the summer, lysimeters had to be raised to avoid constant pooling of surface water as the water table rose. This caused some drying of the upper layer of peat and may have altered N values for the second half of the sampling period.

Porewater was collected at each fen plot using 30 cm tensiometers retrofitted into suction lysimeters. The tensiometer cap was removed and rubber tubing (1.6 mm diam.) was threaded inside to the ceramic cup. 24 hrs prior to sampling, samplers were purged, and tubing was pinched at the top of the tensiometer to maintain a vacuum suction. A 60 ml syringe was used for sampling the following day and samples were poured into a clean, acid-washed polyethylene bottle for N analysis. Due to installation delay, porewater was only sampled between July and August.

Porewater and surface water N was not collected in the upland due to the hard soil surface and difficulty installing the samplers.

Stemflow (tree leachate) was collected at each monitored tree location in the upland. Trees were instrumented with a plastic funnel (~10 cm diam.) that was cut from the top of the funnel to the nozzle so it could be wrapped around the tree trunk. Once wrapped around the tree, the funnel was sealed shut with silicone, and tubing was inserted in the bottom of the funnel and secured into place using silicone and tape. The other end of the tubing was then inserted into a polyethylene bottle and secured to the base of the tree. Mesh screening and tape were placed around the top of the funnel (leaving a ~0.6 cm gap to not inhibit stemflow) to avoid capturing direct throughfall and prevent the entry of leaves. Samples were collected within 24 hrs after rainfall events and poured into clean, acid washed polyethylene bottles for N analysis.

Groundwater sampling was conducted in the fen and upland at wells located within the vicinity of each monitoring plot. Wells were purged 24 hrs prior to sampling and were sampled twice over the study period in June and July.

Before all bioavailable N sample collection, sampling bottles were thoroughly acid washed, and triple rinsed with deionized water. During time of sampling, bottles were rinsed again with deionized water and rinsed with some sample to coat the inside of the bottle. After samples were taken, they were placed on ice in a cooler and processed for storage within 24 hrs of collection. Samples were filtered using ashless 0.45 μm porosity filter paper (Whatman cellulose membrane filter) and frozen. All samples were analyzed for bioavailable $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ using colorimetric analysis at the Biogeochemistry Lab at University of Waterloo (Bran Luebbe AA3, Seal Analytical, Seattle, U.S.A., Methods G-102-93 ($\text{NH}_4^+\text{-N}$) and G-109-94 ($\text{NO}_3^-\text{-N}$) (Total inorganic N (TIN) = $\text{NH}_4^+\text{-N}$ + $\text{NO}_3^-\text{-N}$).

Soil Nutrients and Physical Properties

Extractable soil N pools and net rates of soil N-cycling (nitrification, ammonification, and mineralization) were determined with the in-situ buried-bag incubation method (Hart et al. 1994; Macrae et al. 2013). A pair of adjacent field moist soil cores were sampled at all monitoring plots in both the fen (n = 9) and upland (n = 9) using a Dutch auger and placed into polyethylene bags. One bag (time zero) was immediately placed on ice in a cooler and processed within 12-24 hrs for determination of extractable soil N concentrations of nitrate (NO_3^- -N) and ammonium (NH_4^+ -N). Peat and mineral soil extractable-N (ext-N) from this first bag (time zero) was used for the determination of soil bioavailable N. The second bag was flagged, placed back into the hole from which it was sampled and left to incubate in-situ over a three-week period. Two incubations both lasting three-weeks were performed in both the fen and upland during summer 2019: from 19 June to 19 July (DOY 170-200); and from 19 July to 17 August (DOY 200-229). Data were averaged over both incubation periods to obtain an overall summer average.

During sample processing, bags were mixed by hand until homogenized. An ~10 g sub-sample was shaken (to dissociate the soil) for one hour in 50 ml 2M KCl for the extraction of NO_3^- and NH_4^+ pools. All samples (including blanks) were then gravity filtered through ashless filter paper (Whatman no. 42) and stored in the freezer until colorimetric analysis for NH_4^+ -N and NO_3^- -N at the Biogeochemistry Lab at University of Waterloo (Bran Luebbe AA3, Seal Analytical, Seattle, U.S.A., Methods G-102-93 (NH_4^+ -N) and G-109-94 (NO_3^- -N)).

A second sub-sample from each bag was used for the determination of gravimetric moisture content. An 8-10 g sample was weighed out on an aluminum dish and placed in a drying oven at 80 °C for ~ 24 hrs (until all moisture had been removed). Oven dried samples were then re-weighed. Both wet and dry weights of each sample were incorporated into the mineralization

calculations and allowed for soil mineralization rates to be presented in this study in micrograms per gram of dry weight ($\mu\text{g g}^{-1} \text{dwt}^{-1}$). Net nitrification and ammonification rates were calculated by subtracting the initial NO_3^- and NH_4^+ concentration (time zero) from that of the incubated bag. Net N-mineralization rates were calculated as the sum of net nitrification and ammonification rates. Negative values for net NO_3^- and NH_4^+ mineralization indicates net N immobilization and conversely, positive values indicate net release of N into porewater.

Intact soil cores were sampled at each study plot in both the fen and upland for the analysis of soil physical properties and macronutrient concentrations. In the fen, 15 cm peat cores were extracted from each plot using PVC pipe (10 cm diam. x 15 cm ht.) and a knife. Similarly, in the upland, 10 cm soil cores were extracted from each plot using PVC pipe (10 cm diam. x 10 cm ht.) and a knife and hammer (very hard soil). Samples were transported back to the Hydrometeorology Lab where they were analyzed for bulk density and porosity (Freeze and Cherry, 1979). A sub-sample of soil was used to determine soil organic matter content (OM) by loss on ignition (LOI) at 550 °C for 3 hrs. A second sub-sample was used to determine macronutrients of total carbon (TC), total nitrogen (TN) and C:N ratio. Total C and TN were analyzed using a Thermo-Finnigan-Delta elemental analyzer-isotope ratio mass spectrometer (EA-IRMS) (Environmental Isotope Laboratory, University of Waterloo).

Vegetation Sampling and Nutrient Analysis

Vegetation samples were collected at peak 2019 growing season at all monitoring plots in both the fen and upland. To not interfere with stemflow in the upland, a “reference” tree was used for vegetation sampling. Each stemflow tree had a reference tree that was the same species, approximately the same height and in close proximity. Samples were frozen and transported back to University of Waterloo for the analysis of TC, TN and C:N ratio. In the lab, samples were

thawed, dried at 80 °C for 24 hrs and subsequently ground before analysis for TC and TN using EA-IRMS (Thermo Scientific, Waltham, MA) (Environmental Isotope Laboratory, University of Waterloo). Litter samples were collected and extracted for N in summer 2018. A sub-sample was extracted in 50 ml DI water for NO_3^- , while a second sub-sample was extracted in 50 ml KCl for NH_4^+ . All filtered extractions (including blanks) were analyzed using colorimetric analysis at the Biogeochemistry Lab at University of Waterloo (Bran Luebke AA3, Seal Analytical, Seattle, U.S.A., Methods G-102-93 (NH_4^+ -N) and G-109-94 (NO_3^- -N)).

Leaf area index (LAI) was recorded monthly at all plots in the fen and upland using the LAI-2200 plant canopy analyzer (LI-COR Biosciences, Lincoln, NE). Although a sparsely treed landscape unit, upland LAI was collected from individual trees at each plot, making upland LAI > fen LAI. All LAI data were processed, and scattering errors were corrected using the FV2200 version 2.1 program (LI-COR Biosciences, Lincoln, NE).

Micrometeorological Variables

Meteorological (MET) stations were set up in both the fen and upland, where ambient air temperatures (HC2S3 temperature and RH sensor, Campbell Scientific, Logan, UT) were sampled at 10 second intervals and averaged at 30-minute intervals on dataloggers (CR1000, Campbell Scientific, Logan, UT) over the study period. Daily precipitation was recorded using a tipping bucket rain gauge (TR-525M, Texas Electronics, Dallas, TX) (located in an open area in upland).

Fen soil temperature (copper constantan thermocouples) and volumetric water content (VWC, fraction) (Stevens Hydra Probe II) at 2 (surface), 5, 10 and 30 cm depths were recorded at 30-minute intervals on dataloggers (CR1000, Campbell Scientific, Logan, UT). Fen VWC was averaged from 2-30 cm to capture depth at which most water and soil sampling was conducted. Fen water table (WT) depth was recorded every 30 minutes with water level loggers (Dataflow

systems environmental monitoring Odyssey Capacitance Water Level Logger, Schlumberger Limited Mini Diver) in a well located next to the MET station.

Upland soil temperature (copper constantan thermocouples) and VWC (Stevens Hydra Probe II) at 5, 10, 15 cm depths, averaged from three locations along a north-south transect across the upland (SAF 130T (low slope), 220U (mid slope) and 360U (high slope)) (Figure 3.1) for spatial representativeness, were recorded at 30-minute intervals on dataloggers (CR1000, Campbell Scientific, Logan, UT). Upland VWC was averaged from 5-15 cm to capture depth at which most sampling was conducted. Upland WT depth was automatically recorded at 30-minute intervals on water level loggers (Dataflow systems environmental monitoring Odyssey Capacitance Water Level Logger, Schlumberger Limited Mini Diver). Only continuous WT data from SAF 220U (mid slope) was useable, as instrumentation issues led to extensive data gaps from the wells at SAF 130T (low slope) and 360U (high slope).

Statistical Analyses

All statistical analysis was performed using R (Rstudio Team, 2016). Data was first tested for normality using the Shapiro-Wilkinson distribution test ($p > 0.05$ significance). All bulk deposition data were found to be normally distributed and therefore were analyzed using a pairwise t-test to determine differences between the fen and upland ($p < 0.05$).

Fen N macronutrient concentrations in both soil and vegetation were found to be normally distributed and as such were analyzed using a one-way parametric ANOVA to determine differences among group means ($p < 0.05$). This was followed by a post-hoc pairwise t-test to examine significant differences between species. Upland N macronutrient concentrations in both soil and vegetation were found to be normally distributed and were therefore analyzed using a pairwise t-test to determine differences between tree types ($p < 0.05$).

All fen net N-cycling processes were found to be normally distributed and thus were analyzed using a one-way parametric ANOVA, followed by a post-hoc pairwise t-test to examine significant differences between species. Fen and upland extractable $\text{NO}_3\text{-N}$ and TIN were found to be non-normally distributed and were therefore analyzed using a non-parametric Kruskal-Wallis test ($p < 0.05$). Upland net N-cycling processes were found to be normally distributed and were analyzed using pairwise t-test to determine differences between tree types.

3.4 Results

Within each sub-section, the results for Nikanotee fen will be presented first, followed by the results for Nikanotee upland as they were analyzed as separate landscape units. When both landscape units are being compared, this will be indicated within the specific table or figure.

Climatic and Environmental Conditions

The summer 2019 study season, (27 May to 19 August, DOY 147 to 231), was overall wet and warm in comparison to the thirty-year climate normal (1981-2010) for the Fort McMurray region (Environment and Climate Change Canada, 2017). Total precipitation over the study period for the fen-upland was 231.8 mm, slightly higher than the thirty-year normal of 211.1 mm (Figure 3.3A and 3.4A). Mean daily air temperature for the fen and upland were 17.5 and 16.4 °C, respectively, both higher than the regional thirty-year climate normal (1981-2010) of 15.7 °C (Environment and Climate Change Canada, 2017) (Figure 3.3B and 3.4B).

Soil temperature in the fen gradually increased over the study period at each depth, averaging 11.1, 9.5, 8.3, and 6.1 °C, respectively (Figure 3.3B). On average between 2-30 cm, soil temperature in the fen was 8.8 °C. Fen WT depth averaged -2.8 cm bgs over the study period. Fen WT depth gradually increased over the study period, with a sharp increase coinciding with the first

large rainfall event of 28.8 mm on 28 June (DOY 179), where WT depth increased from -5 cm bgs to just over 0 cm bgs (Figure 3.3C). Fen VWC didn't respond significantly to precipitation events. There was a sharp increase in VWC at the beginning of the study period and then VWC gradually decreased over time (Figure 3.3D). Over the study period, VWC between 0-30 cm depth averaged 0.77 (fraction).

Soil temperature in the upland followed a similar pattern to ambient air temperature (Figure 3.4B). Specifically, at 5, 10 and 15 cm depths, average soil temperature over the study period was 15.1, 16.7 and 16.5 °C, respectively. Average soil temperature for 5-15 cm was 16.1 °C, almost equal to average ambient air temperature. Upland WT depth increased steadily over time (Figure 3.4C) averaging 319.9 cm bgs across the upland over the entire period. Upland VWC between 5-15 cm depth was relatively low (~0.09) at the beginning of the study period, but increased markedly following the first large rainfall event on 28 June (DOY 179; 28.8 mm) (Figure 3.4D). Upland VWC decreased to its lowest (~0.06) on 19 July (DOY 200) but increased again following large rainfall events near the end of the study period. On average, upland VWC was 0.11 over the entire study period.

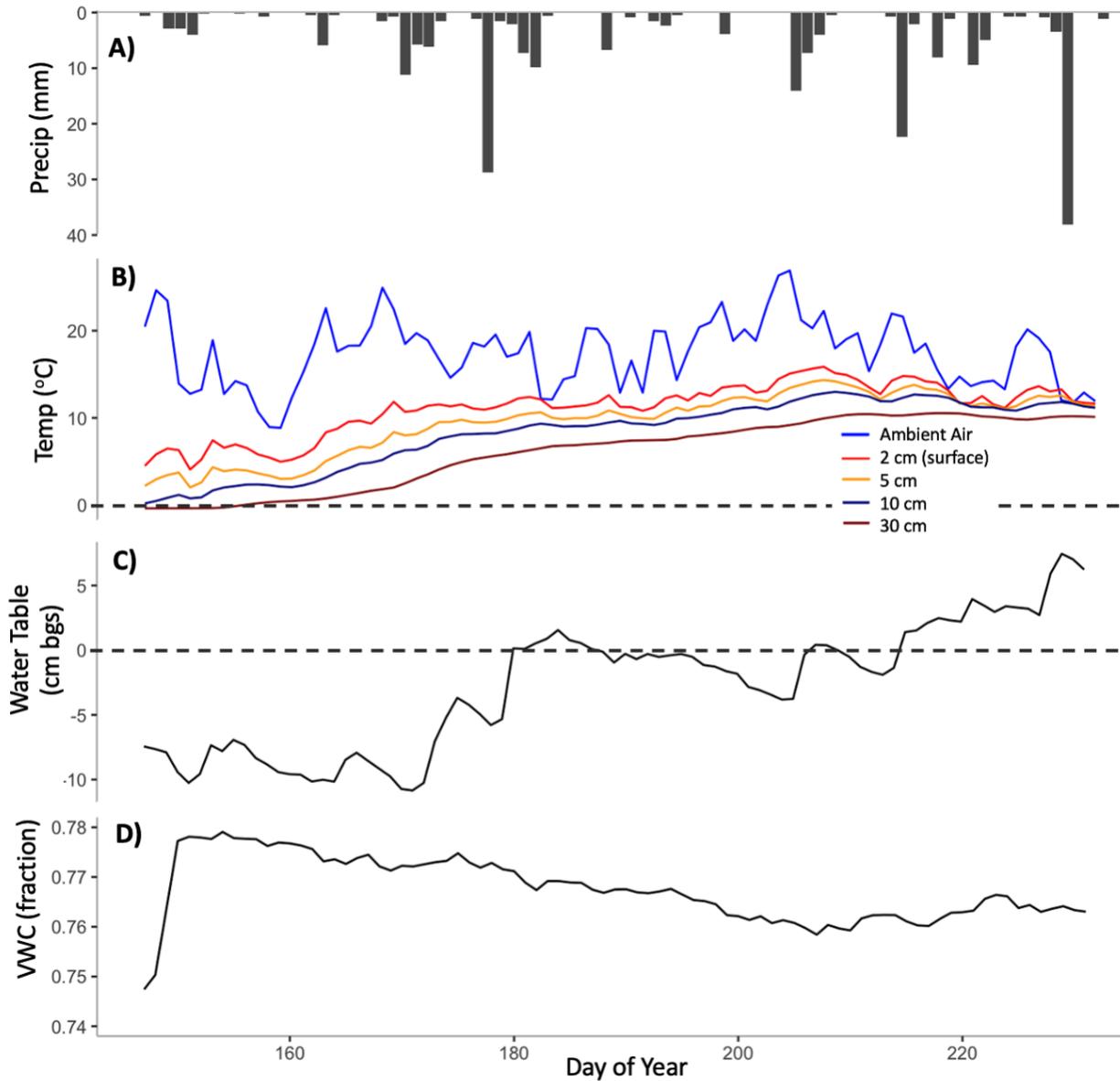


Figure 3.3: Hydrometeorological conditions during the 2019 growing season at Nikanotee fen, Fort McMurray, Alberta. A) total daily precipitation, B) average daily ambient air temperature and soil temperature at 2, 5, 10 and 30 cm depths, C) water table depth, and D) soil VWC averaged between depths of 2-30 cm.

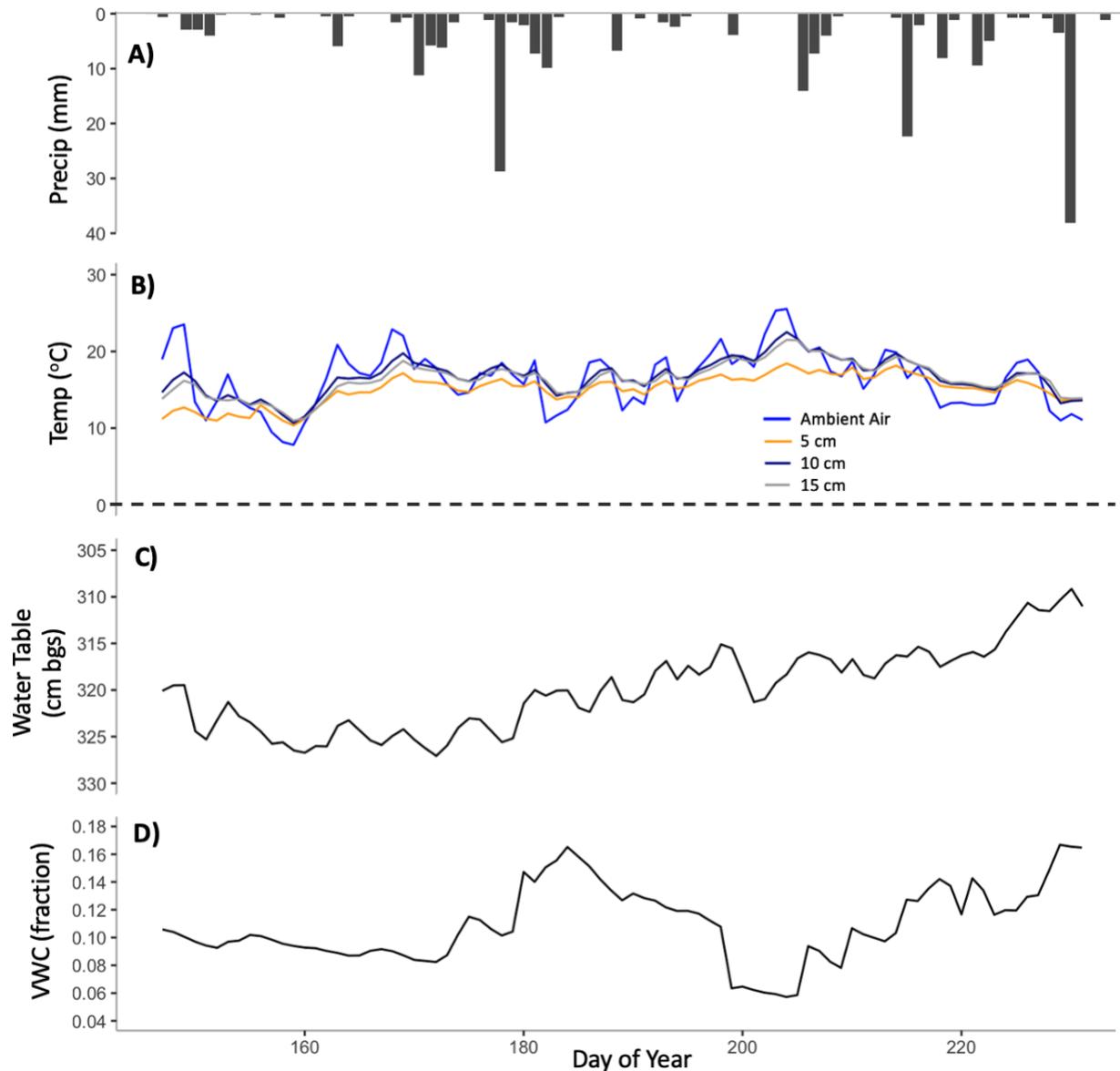


Figure 3.4: Hydrometeorological conditions during the 2019 growing season at Nikanotee upland, Fort McMurray, Alberta. A) total daily precipitation, B) average daily ambient air temperature and soil temperature at 5, 10 and 15 cm depths, C) water table depth, and D) soil VWC averaged between depths of 5-15 cm.

Nitrogen Deposition

Bulk N deposition in the fen was statistically greater than the upland for all N forms ($p < 0.01$) (Table 3.1). The fen received bulk $\text{NH}_4\text{-N}$ and $\text{NO}_3\text{-N}$ loads of $1.8 \text{ kg ha}^{-1} (\pm 0.4)$ (62.2 mg L^{-1} equiv.) and $0.3 \text{ kg ha}^{-1} (\pm 0.03)$ (12.2 mg L^{-1} equiv.), respectively, while the upland received $1.3 \text{ kg ha}^{-1} (\pm 0.2)$ (46.1 mg L^{-1} equiv.) and $0.2 \text{ kg ha}^{-1} (\pm 0.07)$ (8.8 mg L^{-1} equiv.), respectively. In total

(TIN), the fen received a bulk atmospheric N load of 2.1 kg N ha⁻¹ (±0.4) (74.4 mg L⁻¹ equiv.) and the upland received 1.5 kg N ha⁻¹ (±0.3) (55 mg L⁻¹ equiv.). Ammonium deposition was much greater than NO₃-N deposition, and consequently, TIN deposition rates reflected NH₄-N deposition rates. In both the fen and upland, >85% of TIN deposition was composed of NH₄-N and the remaining <15% was NO₃-N.

To compare and discuss N deposition with previous studies in the AOSR, inorganic N loads deposited on the fen and upland were converted to annual loads in kg ha⁻¹ yr⁻¹ (3-month N load x 4) (Table 3.1). In total (TIN) the fen and upland received approximately 8.5 kg N ha⁻¹ yr⁻¹ (±1.5) and 6.2 kg N ha⁻¹ yr⁻¹ (±1.1), respectively. These are a slight overestimation since N deposition loads tend to decrease over winter (Fenn et al., 2015; Hsu et al., 2016). However, considering that open bulk deposition collectors often underestimate total deposition (Lovett & Lindberg, 1993), these are good estimations with which to compare.

Table 3.1: Average bulk deposition for the fen and upland, Nikanotee Fen Watershed, Fort McMurray, Alberta. Summer 2019 bulk deposition (3-month collection) and estimated annual N deposition loads for nitrate (NO₃-N), ammonium (NH₄-N) and total inorganic nitrogen (TIN) are shown. Standard deviation provided in parentheses.

Site		NH ₄ -N	NO ₃ -N	TIN
Fen	kg ha ⁻¹	1.8 (0.4)	0.3 (0.03)	2.1 (0.4)
	kg ha ⁻¹ yr ⁻¹	7.1 (1.4)	1.4 (0.1)	8.5 (1.5)
Upland	kg ha ⁻¹	1.3 (0.2)	0.2 (0.07)	1.5 (0.3)
	kg ha ⁻¹ yr ⁻¹	5.2 (1)	1 (0.3)	6.2 (1.1)

Nitrogen Cascades

Total inorganic N overall was dominated by NH₄-N across all fen cascade levels (i.e., atmosphere, water, soil, vegetation), except for groundwater and Typha litter, in which NH₄-N and NO₃-N were equal (Table 3.2). Moving through the cascade beginning with atmosphere (N input), NH₄-N:NO₃-N ratios of both bulk and wet deposition were low but similar, with bulk and wet deposition exhibiting ratios of 5 and 2, respectively.

Water (N pools) showed a higher overall TIN concentration for surface water, compared to porewater and groundwater, with porewater TIN being lowest. $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio was 1 for both surface water and groundwater, however, porewater had a larger ratio of 10. Groundwater $\text{NO}_3\text{-N}$ increased to 0.09 mg L^{-1} below porewater (0.01 mg L^{-1}), indicating some leaching of NO_3^- at deeper depths.

Soil (N reservoir) showed the highest extractable TIN for Carex plots (18.9 ug g^{-1}), followed by Juncus plots (9.6 ug g^{-1}) and Typha (6.6 ug g^{-1}). $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratios were highest within this cascade level, demonstrating the dynamic processes and transformations occurring by soil microbes. Juncus plots had a higher $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio, compared to Carex and Typha plots. If we assume that the peat in the constructed fen has an approximate bulk density of 200 kg m^{-3} , then we have 30 kg m^{-2} in the top 15 cm of peat (refer to bulk density in Table 3.4). If the average peat extractable N in the fen is approximately 12 ug g^{-1} (equal to 12 mg kg^{-1}) (refer to peat extractable N in Table 3.2), then we have an average of 360 mg m^{-2} of N in the top 15 cm of peat. This translates roughly to 3.6 kg ha^{-1} of total inorganic extractable N in the top 15 cm of the fen, which is greater than the average bulk deposition that the fen received over the summer period.

Litter (N reservoir) showed almost identical extractable TIN for Carex and Typha litter with 17.1 and 16.9 ug g^{-1} , respectively. Juncus litter had highest extractable TIN of 24.9 ug g^{-1} . $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratios were lower in this cascade level reflecting accumulation of N in litter. If we assume that the litter in the constructed fen has an approximate biomass of 1000 g m^{-2} (equal to 1 kg m^{-2}) (Nwaishi, personal communication), and an average litter extractable N of approximately 20 ug g^{-1} (equal to 20 mg kg^{-1}) (refer to litter extractable N in Table 3.2), then we have an average of 20 mg m^{-2} of N in the fen litter. This translates roughly to 0.2 kg ha^{-1} of total inorganic extractable N in the fen litter.

Similar to the fen cascade, TIN was overall dominated by $\text{NH}_4\text{-N}$ across all upland cascade levels, except for stemflow, where $\text{NO}_3\text{-N}$ concentrations were higher than $\text{NH}_4\text{-N}$ (Table 3.3). Beginning with atmosphere (N input), $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratios of both bulk and wet deposition were the same as the fen cascade with ratios of 5 and 2 for bulk and wet deposition, respectively.

For water (N pools), stemflow TIN was dominated by $\text{NO}_3\text{-N}$ for both tree types, resulting in $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratios <1 . Since wet atmospheric input was dominated by $\text{NH}_4\text{-N}$, this indicates a tree process taking up $\text{NH}_4\text{-N}$ from wet deposition, resulting in more $\text{NO}_3\text{-N}$ than $\text{NH}_4\text{-N}$ flowing down the trunks. Groundwater TIN was much higher than stemflow and showed a larger $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio of 20, indicating leaching of NH_4^+ .

Soil (N reservoir) showed similar extractable TIN for both tree types, with 2 and 1.7 ug g^{-1} , for deciduous and coniferous soils, respectively. $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratios were identical (17) for both coniferous and deciduous soil. If we assume that the soil in the constructed upland has an approximate bulk density of 1200 kg m^{-3} , then we have 120 kg m^{-2} in the top 10 cm of soil (refer to bulk density in Table 3.4). If the average soil extractable N in the upland is approximately 2 ug g^{-1} (equal to 2 mg kg^{-1}) (refer to soil extractable N in Table 3.3), then we have an average of 240 mg m^{-2} of N in the top 10 cm of soil. This translates roughly to 2.4 kg ha^{-1} of total inorganic extractable N in the top 10 cm of the upland, which is greater than the average bulk deposition that the upland received over the summer period.

Litter (N reservoir) showed higher extractable TIN for coniferous litter (15.3 ug g^{-1}) compared to deciduous litter (9.5 ug g^{-1}). Once again, litter $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio decreased in this cascade level to 6 and 7 for deciduous and coniferous trees, respectively, reflecting plant uptake and storage of both inorganic N compounds. If we assume that the litter in the constructed upland has an approximate biomass of 500 g m^{-2} (equal to 0.5 kg m^{-2}) (Nwaishi, personal communication),

and an average litter extractable N of approximately 12 ug g^{-1} (equal to 12 mg kg^{-1}) (refer to litter extractable N Table 3.3), then we have an average of 6 mg m^{-2} of N in the upland litter. This translates roughly to 0.06 kg ha^{-1} of total inorganic extractable N in the upland litter.

Table 3.2: Bioavailable N cascade (N solution chemistry profile) of atmosphere (input), water (bioavailable pools), vegetation (reservoir) and soil (reservoir) N in Nikanotee fen over the growing season. Ammonium (NH₄-N), nitrate (NO₃-N), total inorganic N (TIN) and ammonium:nitrate ratio (NH₄-N:NO₃-N) are displayed. Litter data is from summer 2018, while all other variables are from summer 2019. Bulk deposition and wet deposition (total) show summed N values and all other variables including wet deposition (average) show mean N values over the study period. Standard deviation provided in parentheses.

Cascade Level	Species	Depth (cm)	Units	NH ₄ -N	NO ₃ -N	TIN	NH ₄ -N: NO ₃ -N ratio	
Atmosphere	Bulk deposition	--	--	mg m ⁻²	176.3 (35.5)	34.5 (3.2)	210.7 (37.2)	5
	Wet deposition (tot.)	--	--	mg m ⁻²	53.5 (2.4)	24.3 (1.1)	77.8 (3)	2
	Wet deposition (avg.)	--	--	mg m ⁻²	3.8 (2.4)	1.7 (1.1)	5.6 (3)	2
Water	Surface water	--	5	mg L ⁻¹	0.17 (0.2)	0.14 (0.2)	0.31 (0.3)	1
	Porewater	--	30	mg L ⁻¹	0.1 (0.09)	0.01 (0)	0.11 (0.09)	10
	Groundwater	--	150	mg L ⁻¹	0.09 (0.06)	0.09 (0.12)	0.18 (0.1)	1
Soil	Peat	Juncus	10	ug g ⁻¹	9.4 (9.7)	0.22 (0.2)	9.6 (9.7)	43
		Carex	10	ug g ⁻¹	18.3 (6)	0.64 (1)	18.9 (6.7)	29
		Typha	10	ug g ⁻¹	6.3 (7)	0.3 (0.2)	6.6 (7)	21
Vegetation	Litter	Juncus	0	ug g ⁻¹	22.3 (45.6)	2.7 (6.3)	24.9 (14.4)	8
		Carex	0	ug g ⁻¹	14.5 (23.5)	2.6 (5.2)	17.1 (26)	6
		Typha	0	ug g ⁻¹	8.4 (24.5)	8.4 (19.3)	16.9 (21.9)	1

*Wet deposition total is the summed value of wet N over the study period

*Wet deposition average is the average wet N per rain event

Table 3.3: Bioavailable N cascade (N solution chemistry profile) of atmosphere (input), water (bioavailable pools), vegetation (reservoir) and soil (reservoir) N in Nikanotee upland over the growing season. Ammonium (NH₄-N), nitrate (NO₃-N), total inorganic N (TIN) and ammonium:nitrate ratio (NH₄-N:NO₃-N) are displayed. Litter data is from summer 2018, while all other variables are from summer 2019. Bulk deposition and wet deposition (total) show summed N values and all other variables, including wet deposition (average), show mean N values over the study period. Standard deviation provided in parentheses.

Cascade Level		Tree Type	Depth (cm)	Units	NH ₄ -N	NO ₃ -N	TIN	NH ₄ -N: NO ₃ -N
								ratio
Atmosphere	Bulk deposition	--	--	mg m ⁻²	130.8 (24.1)	24.9 (6.7)	155.8 (27.9)	5
	Wet deposition (tot.)	--	--	mg m ⁻²	53.5 (2.4)	24.3 (1.1)	77.8 (3)	2
	Wet deposition (avg.)	--	--	mg m ⁻²	3.8 (2.4)	1.7 (1.1)	5.6 (3)	2
Water	Stemflow	Deciduous	--	mg L ⁻¹	0.09 (0.09)	0.2 (0.4)	0.29 (0.4)	0.5
		Coniferous	--	mg L ⁻¹	0.08 (0.08)	0.7 (1.1)	0.78 (1.1)	0.1
	Groundwater	--	250	mg L ⁻¹	1.19 (0.74)	0.06 (0.05)	1.25 (0.4)	20
Soil	Mineral	Deciduous	10	ug g ⁻¹	1.9 (1.7)	0.11 (0.1)	2 (1.7)	17
		Coniferous	10	ug g ⁻¹	1.7 (0.9)	0.1 (0.1)	1.7 (0.8)	17
Vegetation	Litter	Deciduous	0	ug g ⁻¹	8.1 (6)	1.4 (1.1)	9.5 (3.5)	6
		Coniferous	0	ug g ⁻¹	13.4 (11.7)	1.8 (1.9)	15.3 (6.8)	7

*Wet deposition total is the summed value of wet N over the study period

*Wet deposition average is the average wet N per rain event

Soil and Vegetation Properties & Macronutrients

Typha was the tallest species in the fen (>80 cm on average) but had the lowest LAI, while Juncus was the shortest (<55 cm) but with the highest LAI (Table 3.4). In terms of soil properties in the fen, Carex plots had the highest OM (67%) and moisture content (82%), likely owing to its larger belowground biomass (Popović et al., unpublished). Juncus and Typha were both similar in terms of OM and moisture content. Bulk density was similar between species plots, but lower for Typha. Porosity was the same for Juncus and Carex (0.9), but higher for Typha (1).

In the upland, deciduous trees were almost 1.5 times taller than conifers, with an average height >285 cm (Table 3.4). Although deciduous trees were tallest, they had the lowest LAI (approximately half the LAI of conifers). Conversely, coniferous trees were smaller but had higher LAI, due to greater density of needles, increasing their surface area. In terms of soil properties, OM was similarly low between both tree types, but slightly greater overall for deciduous trees (6%), likely owing to annual leaf-drop in the fall. Moisture content was slightly higher for deciduous trees (8%), due to higher OM content. Soil bulk density (1.2 g cm^{-3}) and porosity (0.5) were the same for both tree types.

Table 3.4: Soil and vegetation properties for the dominant fen species and upland tree types at Nikanotee fen and upland for summer 2019 growing season. Soil chemical properties were determined from 15 cm peat cores (fen) and 10 cm mineral cores (upland). Standard deviation provided in parentheses.

Landscape	Species	Soil Properties				Vegetation Properties	
		Bulk density (g cm ⁻³)	Porosity	Moisture content (% grav.)	OM (%)	Median Height (cm)	LAI
Fen	Juncus	0.2 (0.08)	0.9 (0.2)	77 (6)	60.3 (3.8)	54.4 (2.2)	1.6
	Carex	0.2 (0.08)	0.9 (0.06)	82 (4)	66.6 (17.1)	61.2 (12.6)	1.3
	Typha	0.1 (0.04)	1.0 (0.04)	79 (5)	61.6 (3.0)	83.6 (8.3)	1.2
Upland	Deciduous	1.2 (0.09)	0.5 (0.03)	8 (1.9)	5.7 (1.1)	285.9 (45.9)	1.6
	Coniferous	1.2 (0.06)	0.5 (0.04)	6 (1.4)	5.3 (0.6)	198.6 (56.3)	3.0

Total nitrogen (TN) concentrations were not significantly different among fen species for soil ($p>0.05$) or vegetation ($p>0.05$). However, a significant difference was observed between soil and vegetation TN ($p<0.05$). Soil TN concentrations were almost identical for both *Carex* and *Typha* with 9.9 mg g^{-1} and 9.8 mg g^{-1} , respectively, and lower for *Juncus* (8.4 mg g^{-1}) (Table 3.5). C:N ratio in soil were the same for *Carex* and *Typha* (26) and slightly higher for *Juncus* (27). Vegetation TN concentration was highest for *Typha* (14.8 mg g^{-1}) and lowest for *Juncus* (12.4 mg g^{-1}), with *Carex* TN falling in between (13.9 mg g^{-1}). C:N ratio for vegetation was lowest for *Typha* (25) and highest for *Juncus* (29) with C:N ratio of *Carex* in between (27).

Total N concentrations in soil beneath both tree types did not differ ($p>0.05$, deciduous = 0.09 mg g^{-1} , conifers = 0.08 mg g^{-1} ; Table 3.5). However, TN concentrations in deciduous vegetation (15.8 mg g^{-1}) were more than twice those of conifers (7.2 mg g^{-1}) ($p<0.001$). A significant difference was also observed between soil TN and vegetation TN ($p<0.001$). C:N ratios in soil were again slightly higher for deciduous (21) than coniferous trees (20). C:N ratios of vegetation were lower for deciduous (25) and more than two times higher for coniferous trees (59).

Table 3.5: Soil and vegetation macronutrient C, N concentrations and C:N nutrient ratios for the dominant fen vegetation species and upland tree types at Nikanotee fen and upland during summer 2019 season. Standard deviation provided in parentheses.

Landscape	Cascade	Species/Tree Type	Macronutrient Concentrations		Nutrient Ratio
			C (mg g ⁻¹)	N	C:N ratio
Fen	Soil	Juncus	228.1 (24.9)	8.4 (1.3)	27 (1.6)
		Carex	261.3 (57.5)	9.9 (2.5)	26 (1.6)
		Typha	255.5 (17.8)	9.8 (0.9)	26 (0.7)
	Vegetation	Juncus	364.7 (10.2)	12.4 (1.7)	29 (3.4)
		Carex	374.5 (11.2)	13.9 (0.1)	27 (1.0)
		Typha	365.4 (9.8)	14.8 (2.0)	25 (2.9)
Upland	Soil	Deciduous	1.9 (0.3)	0.09 (0.02)	21 (1.6)
		Coniferous	1.6 (0.3)	0.08 (0.01)	20 (1.6)
	Vegetation	Deciduous	387.4 (13)	15.8 (1.1)	25 (2.3)
		Coniferous	423.7 (15)	7.2 (2.0)	59 (17.4)

Soil extractable N & net N-cycling

Soil extractable (ext-) $\text{NO}_3\text{-N}$ concentrations were low for both landscape units and did not differ significantly between vegetation species (Figure 3.5A) or tree type (Figure 3.6A) ($p>0.05$). In the fen, ext-TIN concentrations were similar between Juncus and Typha, and slightly greater for Carex (Figure 3.5B) but did not differ significantly among species ($p>0.05$). In the upland, ext-TIN concentrations were similar for both tree types and did not differ ($p>0.05$) (Figure 3.6B).

Net N-cycling processes were low for both landscape units. In the fen, net nitrification hovered around zero (Figure 3.5C), whereas both net ammonification and mineralization (Figure 3.5 D & E) were slightly higher. In the upland, all net N-cycling processes were close to zero (Figure 3.6C, D & E). No statistically significant differences were observed between vegetation species ($p>0.05$) or tree type ($p>0.05$).

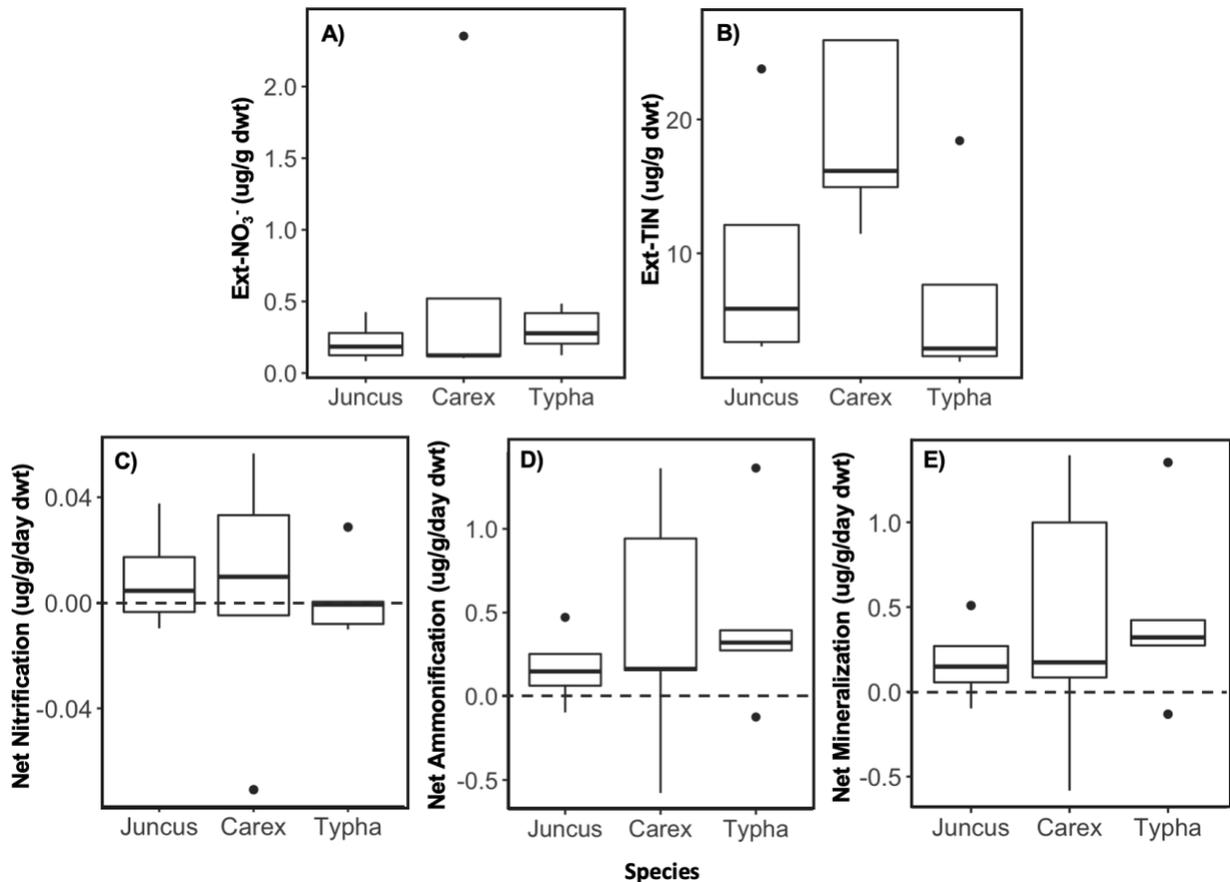


Figure 3.5: Soil extractable (A) nitrate (ext-NO₃⁻) and (B) total inorganic N (ext-TIN) and net soil N-cycling processes for (C) net nitrification, (D) net ammonification and (E) net mineralization for the dominant vegetation species plots at Nikanotee fen.

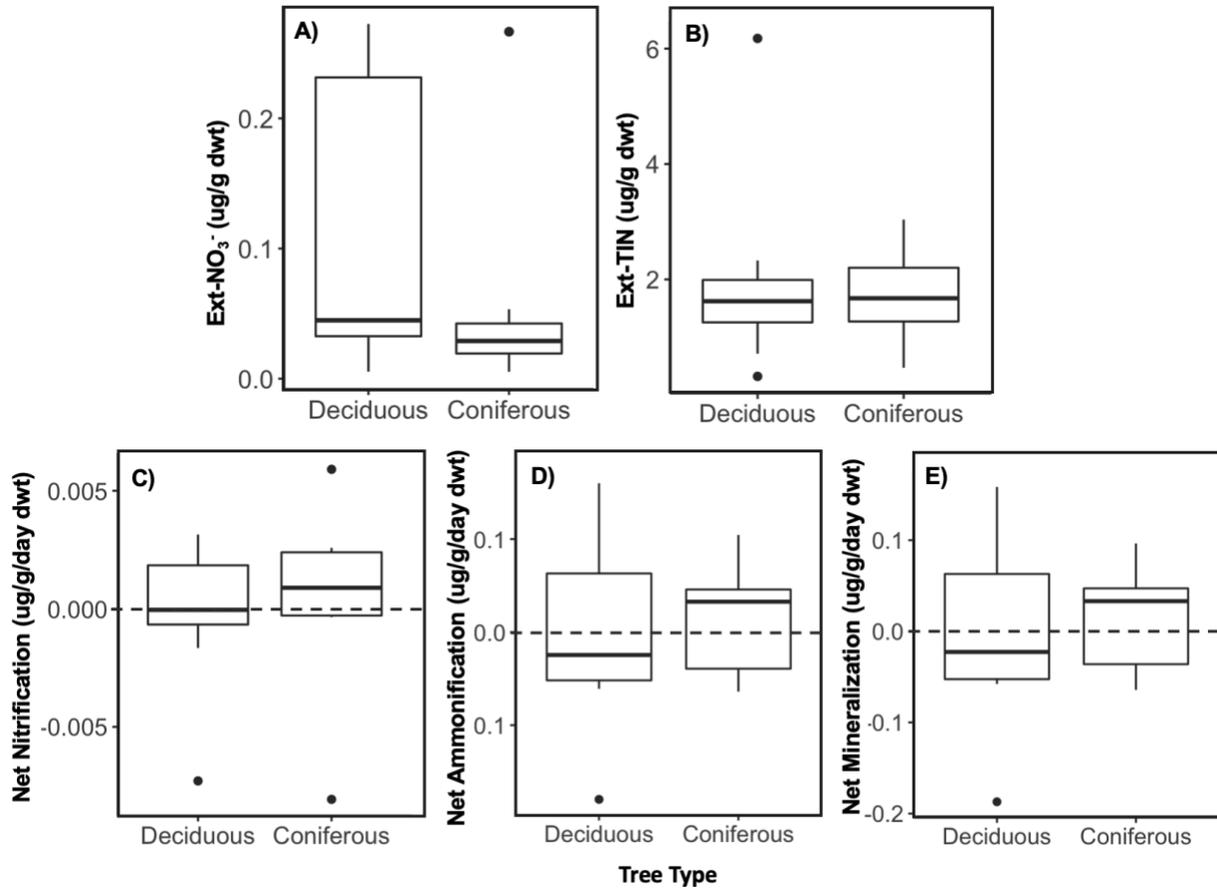


Figure 3.6: Soil extractable (A) nitrate (ext-NO₃⁻) and (B) total inorganic N (ext-TIN) and net soil N-cycling processes for (C) net nitrification, (D) net ammonification and (E) net mineralization for the two tree type plots in Nikanotee upland.

3.5 Discussion

Nitrogen Deposition

Quantifying atmospheric N deposition loads to any system is a prerequisite for understanding the impacts of exogenous N to ecosystem nutrient dynamics (Bobbink et al., 1992). Atmospheric N deposition loads received over the 3-month summer period by the reclaimed fen-upland were greater-than-background N deposition loads in the AOSR (~1 kg N ha⁻¹ yr⁻¹) (Vitt et al., 2003).

However, AOSR N deposition loads are very low compared to those in Europe, Asia, and other regions of North America. Nitrogen deposition in Europe ranges between 1-60 kg N ha⁻¹ yr⁻¹ but averages 16.8 kg N ha⁻¹ yr⁻¹ (MacDonald et al., 2002). In China, N deposition averages 20.4 kg N ha⁻¹ yr⁻¹ (Yu et al., 2019), with maximum values of 63.5 kg N ha⁻¹ yr⁻¹ (Lü & Tian, 2007). In Western USA, N deposition typically ranges between 1-4 kg N ha⁻¹ yr⁻¹, but can be as high as 30-90 kg N ha⁻¹ yr⁻¹ downwind from major agricultural and urban areas in southern California (Fenn et al., 2003).

Nitrogen deposition loads fall within the range previously quantified for industrial sites in the AOSR, which have quantified a wide range of total N loads from 4.4 kg N ha⁻¹ yr⁻¹ (Fenn et al., 2015) to 17 kg N ha⁻¹ yr⁻¹ in the centre of mining activities (Wieder et al., 2019), and as high as 24.3 kg N ha⁻¹ yr⁻¹ on other reconstructed forests (Hemsley et al., 2019). Differences in annual deposition loads are reflective of multiple factors including location (i.e., downwind from emission sources), meteorological conditions, and varying emission mixtures and quantities of N emitted by stacks, vehicles, and machinery (Fenn et al., 2015). These factors affect how much deposition reaches the landscape, resulting in annual variations in N deposition loads in the AOSR.

Studies that have analyzed partitioning of wet and dry deposition within the vicinity of oil sands activities estimate that bulk deposition is largely comprised of the dry component (~80%), resulting in elevated N loads (Fenn et al., 2015; Proemse et al., 2013; Hsu et al., 2016). A preliminary dust study (using the marble method) conducted on the reclaimed fen-upland in June 2014 found that both landscape units received >155 kg ha⁻¹ of dry dust deposition (fen>upland) (E. Kessel, unpublished, University of Waterloo). Dust data from 2014 showed NH₄-N and NO₃-N concentrations of approximately 10 and 5 mg L⁻¹, respectively, for both landscape units (E. Kessel, unpublished, University of Waterloo). The reclaimed site is situated directly within the

hub of industrial activities and only a few hundred metres from an active haul-road to the east, with a less active, but relevant, dirt road to the west. Thus, considerable dry deposition from surrounding activities is inevitable and likely makes up most of the site's bulk N deposition.

Nitrogen deposition to the environment is controlled by atmospheric turbulence, surface roughness, physical/chemical properties of N compounds, chemical potential gradient between the atmosphere and receptors, and the nature of vegetative surfaces (Hanson & Lindberg, 1991; Bobbink et al., 2012). The fen received significantly more bulk N deposition compared to the upland, due to the openness of the fen and difference in vegetation structure. The fen is composed of low-lying vegetation with lower average LAI (1.4; Table 3.4) compared to the upland with a taller treed canopy and higher average LAI (2.3; Table 3.4). Although the distribution of trees in the upland is sparse, the structure of the vegetation increases the overall roughness of the landscape, resulting in increased turbulence above the canopy and greater retention of N deposition (Bobbink et al., 2012; Pelster et al., 2009). Increased turbulence should keep pollutants suspended in the air longer, making them more likely to be transported away. However, due to the layered structure of trees and increased LAI, the chance of deposition to foliage is high and therefore N deposition loads are typically greater in mature forests (Bobbink et al., 2012). In this study, bulk samplers were installed close to the top of the canopy, thereby missing the collection of N deposition intercepted by the forest canopy, resulting in an underestimation of deposition (Pelster et al., 2009; Balestrini et al., 2007). Another characteristic that likely influenced the amount of N deposition to the upland is sheltering by the surrounding forested slopes, most notably the denser East and South slopes. This forest sheltering (or edge effect) helps intercept deposition to the upland (Wuyts et al., 2008). In contrast, the fen is open, less sheltered and situated adjacent to the site access road (to the north), resulting in increased dry deposition.

Nitrogen deposition to the fen-upland is not as high as other sites located in the vicinity of major oil sands activities. However, N deposition is higher than background AOSR levels, and thus should be monitored as the site continues to develop. Nitrogen deposition could be acting as a chronic fertilizing agent, favouring growth of vascular fen species (Gerdol et al., 2006; Wieder et al., 2020) and preventing establishment of a more diverse ecosystem. Although biological N₂-fixation is another source of new N to peatland ecosystems, it is likely low in the reclaimed fen, as Wieder et al. (2019, 2020) suggest that N₂-fixation is progressively inhibited at N deposition >3 kg N ha⁻¹ yr⁻¹. As the site continues to grow and the upland canopy closes, deposition in the upland will increase, particularly that of throughfall (TF) deposition (Fenn et al., 2015; Proemse et al., 2013). This will result in the accumulation of N deposition on trees, which will increase the hydrological flux of N ions to the forest floor during rain events (Fenn et al., 2015; Pelster et al., 2009), which will become increasingly important as the system develops.

Nitrogen Dynamics of a Reclaimed Fen

The N solution chemistry profile of an ecosystem provides an indicator of where N is being utilized and stored within the soil-water-vegetation continuum and is highly indicative of N-status (Fenn & Poth, 1998; Binkley et al., 1982). Atmospheric N deposition concentrations largely consist of NH₄⁺, owing to greater emissions of reduced N forms (i.e., NH₃) in the region (Fenn et al., 2015). There is no bryophyte layer at Nikanotee fen, that would typically act as a N deposition filter, slowing N from reaching the rhizosphere in natural fens and bogs (Wieder et al., 2019; Lamers et al., 2000). Thus, all atmospherically deposited N to the reclaimed Nikanotee fen that is not intercepted by vegetation is deposited directly onto the water or peat surface. Within the water column, N concentrations were highest in surface water (SW), due to close interaction with incoming N deposition and little plant uptake. Concentrations of SW N at Nikanotee fen were

greater compared to natural fens in Alberta. Vitt & Chee (1990) determined SW N concentrations across three fen types (poor, moderate- and extreme-rich) in central Alberta and found TIN to average 0.06 mg L^{-1} across all fens ($\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ was 4). The difference in SW N at these sites compared to Nikanotee fen is likely due to dilution in pools and dugout pits where samples were taken. Whitfield et al. (2010) compared SW N concentrations between two fen sites in the AOSR; one close to oil sands activities and the other more remote in Stony Mountain. The remote fen SW TIN was 0.067 mg L^{-1} with $\text{NH}_4\text{-N}:\text{NO}_3\text{-N} < 1$ (NO_3^- dominance at this natural fen), whereas SW TIN at the fen close to industrial activities was 0.29 mg L^{-1} with $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ of 10, which is similar to Nikanotee. High $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ at the more industrial fen could be due to increased deposition of NH_4^+ , since SW NO_3^- was not very different between both of their sites.

Nitrogen concentrations decreased in porewater (PW) and $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ increased, indicating uptake of NO_3^- in the rhizosphere by vascular vegetation (Wiedermann et al., 2009). Wieder et al. (2019) conducted an N fertilization study at an ombrotrophic bog located south of Fort McMurray, Alberta, and found that PW N was unaffected by experimental N additions. In their study, NH_4^+ and NO_3^- concentrations were slightly higher at the beginning of the 5-year study period with 1.37 and 0.2 mg L^{-1} , respectively, but proceeded to decrease with increasing N addition over the rest of the study to 0.13 and 0.01 mg L^{-1} , respectively ($\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ of 13). Porewater N concentrations from Nikanotee fen were almost identical to Wieder et al. (2019) in the years following their first sampling season, and $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ was similar. Even considering N additions of 1.5 to $27.6 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, Wieder et al. (2019) observed no increase in PW N, which confirms that Nikanotee fen PW N is consistent with natural peatlands in the region and that N deposition may not influence PW N. An identical N addition study by Wieder et al. (2020) was conducted on a poor fen situated in the same area south of Fort McMurray. In both cases, Wieder

et al. (2019, 2020) did not observe any response in PW N concentrations, suggesting that added N was taken up immediately to support growth of bryophytes and vascular vegetation. Very low PW N concentrations at Nikanotee fen and high $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ compared to SW, indicates uptake of N by vascular vegetation, particularly NO_3^- uptake.

Groundwater (GW) TIN increased (due to NO_3^-) and $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ decreased to 1 (similar to SW). Increasing GW NO_3^- suggests leaching, which typically indicates N saturation (Aber et al., 1989). However, since vegetation is assimilating N in PW, this increase in NO_3^- (and overall GW TIN) is likely natural downward percolation of this highly mobile N form. Groundwater NO_3^- concentrations fell within the mid-range for natural fens ($0.01\text{-}0.2 \text{ mg L}^{-1}$) in the AOSR but was most similar to poor fens (0.05 mg L^{-1}) (Nwaishi et al., Under review). However, NH_4^+ was low in comparison to natural fens ($0.2\text{-}0.5 \text{ mg L}^{-1}$) (Nwaishi et al., Under review). Overall GW TIN was low compared to natural fens in the AOSR, indicating low N conditions at the reclaimed fen.

One indication of high N-status (N saturation) in peatlands is increased soil extractable N (ext-N), increased soil mineralization processes (linked to stimulated decomposition rates) and a lowering of the $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio (due to increased nitrification) (Lamers et al., 2000; Wieder et al., 2019). During the first summer following peat transplant in 2013, peat extractable NH_4^+ (ext- NH_4^+) was slightly higher ($\sim 20 \text{ ug g}^{-1} \text{ dry weight}^{-1}$ (dwt) on average), but overall similar to 2019 (Nwaishi et al., 2015). Peat ext- NH_4^+ in 2013 was almost the same as peat ext- NH_4^+ for *Carex* species (18.3 ug g^{-1}) in 2019, due to greater OM and belowground biomass of *Carex* (Popović et al., unpublished), but greater than both *Juncus* and *Typha* ($<10 \text{ ug g}^{-1}$). Conversely, peat extractable NO_3^- (ext- NO_3^-) in 2013 was $8.23 \text{ ug g}^{-1} \text{ dwt}^{-1}$ (Nwaishi et al., 2015). Six years later, peat ext- NO_3^- across the fen declined to $<0.7 \text{ ug g}^{-1} \text{ dwt}^{-1}$ and $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ increased for each of the three dominant vegetation species. Peat ext- NO_3^- was greater in the first summer post-construction due

to retention of NO_3^- in the peat from the donor fen, since NO_3^- was the dominant form of TIN in the donor fen as a result of dewatering (Nwaishi et al., 2015; Macrae et al., 2013). After Nikanotee fen was seeded and vegetation began to grow, vascular species have since become the dominant plant community, which are highly efficient at utilizing NO_3^- (Wiedermann et al., 2009). However, the dominant anoxic conditions in the fen can also be responsible for the reduction of NO_3^- to NH_4^+ . As of 2019, soil NO_3^- has depleted significantly, resulting in a greater soil $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio, even with a N deposition load higher than background levels for the AOSR. The additional atmospheric N added to the fen is likely being immediately consumed by microbes and vascular vegetation and is not (currently) increasing the N-status of the fen.

Soil ext-N and net mineralization at Nikanotee fen were low compared to natural boreal fens. Soil ext-TIN in natural fens in northern Alberta is $\sim 50 \text{ ug g}^{-1} \text{ dwt}^{-1}$ (Nwaishi et al., 2015) with concentrations as high as 200 and 600 $\text{ug g}^{-1} \text{ dwt}^{-1}$ (17-50x greater than Nikanotee fen) (Bayley et al., 2005). Net nitrification rates were low (typical of natural fens) (Nwaishi et al., 2015; Bayley et al., 2005), but not as low at Nikanotee fen where they are considered negligible. Very low or negative (immobilization) net nitrification rates influence soil NO_3^- concentrations in peatlands (increased uptake by soil microbes) and lower the potential for NO_3^- leaching (Westbrook & Devito, 2004), suggesting a low N system. Soil C:N greater than 15-20 can also be used to determine net N immobilization (Weintraub & Schimel, 2003). However, net mineralization generally dominated the reclaimed fen for each species, thus C:N ratio cannot be relied upon solely to assess immobilization (Macrae et al., 2013).

Soil net mineralization rates at Nikanotee fen in 2013 were comparable to 2019 rates. However net nitrification was far lower in 2019 (Nwaishi et al., 2016) due to low ext- NO_3^- . Although Nikanotee fen is situated in a high N deposition zone for the AOSR, receiving 3-5 times

more NH_4^+ than NO_3^- deposition, and an overall N deposition load greater than background levels, net mineralization processes have not increased since the first summer post-reclamation. Rather, soil N and net mineralization processes have decreased due to highly productive microbes and vascular vegetation, which appear to be efficiently utilizing N and preventing atmospherically derived N from saturating the site.

Vegetation uptake is one of the major controls on water chemistry and is responsible for storing large amounts of atmospheric N in fen ecosystems (Whitfield et al., 2010). Vegetation contained the highest N concentrations compared to water and soil in the reclaimed fen. The efficiency in which vegetation stores N is evident by the lower $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio compared to soil, especially greater NO_3^- content. Fenn et al. (1996) found foliar TN to be greater at sites with high N pollution compared to low pollution sites (in forest systems). Total N content in vascular vegetation at Nikanotee fen fell within the range (and slightly above) that of natural fens in the region. At Nikanotee fen, vegetation TN ranged between 12.4-14.8 mg g^{-1} compared to natural fens that had vascular vegetation TN concentrations of 11-13 mg g^{-1} (Nwaishi et al., Under review). Considering all other ecosystem receptors indicate Nikanotee fen is a low N environment, slightly greater TN concentrations here could be due to different vascular species storing more TN than those at the natural fens, particularly Typha. Typha is more commonly found in reclaimed fens than natural fens and has a greater aboveground biomass that can store larger amounts of TN (Roy et al., 2016).

For increased ecosystem N content (or N saturation) to begin, vegetation demand for N must be satisfied before soil, or any other component of the ecosystem continuum (Stoddard et al., 1994; Aber et al., 1989). At Nikanotee fen, N content in vegetation was significantly greater than N content in soil for all species, however no differences were observed between species. Carex

contained the greatest N content in soil, due to its greater OM and belowground biomass (Popović et al., unpublished), whereas Typha contained the greatest TN concentration in vegetation, owing to its greater height and aboveground biomass (Popović et al., unpublished). An assessment of the productivity of each species will assist in our understanding of the competitive nature of the dominant vascular vegetation at Nikanotee fen.

Nitrogen Dynamics of a Reclaimed Upland

Similar to the fen, N deposition consisted largely of NH_4^+ in the upland, with NH_4^+ in bulk and wet deposition being 5 and 3 times greater than NO_3^- , respectively. As previously mentioned, this is due to greater NH_x forms in the AOSR (Fenn et al., 2015). From N deposition to stemflow (SF), the $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio flipped to <1 in SF of both tree types. It would be expected that SF would be more enriched in NH_4^+ (since NH_4^+ is the larger depositional input to the system). However, nutrient interactions within the canopy can alter the $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio and are important to forest nutrient cycling (Houle et al., 2015; Pelster et al., 2009). The notable reduction of NH_4^+ over NO_3^- suggests canopy consumption of NH_4^+ , likely due to high affinity of the canopy to the positive charge in foliar and bark surfaces (Houle et al., 2015). Canopy uptake of N is beneficial in low N soils and is especially advantageous for young trees to avoid N competition in the soil and understory (Houle et al., 2015). Canopy preference of NH_4^+ or NO_3^- differs among the literature with deciduous and coniferous forests in the WBP retaining $\text{NH}_4^+ > \text{NO}_3^-$ (Pelster et al., 2009), a N fertilized forest also retained greater NH_4^+ (Gaige et al., 2007), while studies in the Northwest USA and Maine reported greater NO_3^- retention (Fenn et al., 2013; Lovett & Lindberg, 1993).

Upland trees may also be leaching or flushing excess NO_3^- , which depends on the ability of trees to replenish their foliar N pools (Lindberg et al., 1986). This indicates that young upland trees may already be full of NO_3^- due to canopy uptake and soil stocks. Since there are no epiphytic

lichen on the trunks to benefit from excess NO_3^- (Lindberg et al., 1986), this increases overall SF NO_3^- concentrations within the canopy. Additionally, wash-off of dry-deposited NO_3^- adsorbed to the trees may also be incorporated into SF (Lovett & Lindberg, 1993). However, considering that bulk deposition was mostly composed of NH_4^+ , wash-off of NH_4^+ should also be occurring. Since this is not the case, strong canopy uptake of NH_4^+ is likely occurring in the upland. Stemflow N concentrations differed between both tree types with conifer SF being almost 3 times higher than deciduous trees, due to conifers being more efficient at scavenging N pollution on their needles (higher density and LAI) (De Schrijver et al., 2007).

As the upland canopy closes, SF (and TF) N will increase as both tree types become more efficient at intercepting wet and dry N deposition, increasing N flux to the soil surface (Pelster et al., 2009). Stemflow and TF N are expected to be greater below fully-grown conifers as they are more efficient at scavenging N pollution (De Schrijver et al., 2007). However, conifers typically have a lower demand for N, which could increase N leaching from these trees (Campbell et al., 2004).

Similar to Gingras-Hill et al. (2018), NH_4^+ dominated GW TIN in the upland, and was approximately 5 times lower than it was in 2015. In June 2015, newly planted saplings were fertilized, but rapid loss of N through run-off (although a rare occurrence) and leaching was observed during the next rain event after fertilization (Gingras-Hill et al., 2018). Increased GW N in 2015 was due to fertilization, however considering that the upland had been subjected to N deposition for the following four years, GW N declined. Interactions between vegetation uptake and N deposition in high N deposition regions, may be effective at controlling N leaching into GW (Emmett et al., 1993; Devito et al., 1999). The high GW $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio (20) suggests uptake of NO_3^- by vegetation and/or microbes, or reduction of NO_3^- into NH_4^+ within the anoxic

subsurface layers. Although NO_3^- is more mobile than NH_4^+ , the hydrophobic properties of the tailings sand result in NH_4^+ leaching into GW (Gingras-Hill et al., 2018). Depending on the depth of tree roots in the reclaimed upland, vegetation may have difficulty accessing GW N due to lack of micropores (limited capillary rise and nutrient movement) in tailings sand (Jung et al., 2014). Thus, N deposition may be an important source of N for microbial processes and vegetation growth in the reclaimed upland if GW N is inaccessible or low.

Soil C:N has been suggested as an indicator of soil N leaching, with C:N<25 proposed as being indicative of high NO_3^- leaching in upland forests (Gundersen et al., 1998). However, sites receiving TF deposition <10 kg N ha yr⁻¹ have been shown to have low soil NO_3^- concentrations regardless of C:N ratio (Kristensen et al., 2004). Nikanotee upland demonstrated low soil NO_3^- concentrations even with soil C:N<25. However, as the canopy develops, soil N may accumulate as TF N increases >10 kg N ha yr⁻¹, which will subsequently increase N turnover rates (Fenn & Poth, 1998).

Net soil mineralization in the upland was low (~0 ug g⁻¹ day⁻¹ dwt⁻¹) for both tree types. Other AOSR reclamation sites built using similar topsoil amendments, had higher overall soil net mineralization rates (McMillan et al., 2007). At a silviculture project near Peace River, Alberta, net nitrification and mineralization rates for mineral soil were greater than Nikanotee upland for deciduous, mixed and coniferous stands (Jerabkova et al., 2006). Additionally, TN in mineral soil was between 1.91-2.99 mg g⁻¹, but ext- NO_3^- and - NH_4^+ were only slightly higher than Nikanotee upland with 0.2 and 2.7 ug g⁻¹ on average, respectively (Jerabkova et al., 2006). At the reclaimed sites in McMillan et al. (2007), soil TN was greater than Nikanotee upland with TN between 2.2-4.5 mg g⁻¹ (and 9.2 mg g⁻¹ for their natural aspen site). However, soil C:N ratios were identical to Nikanotee upland (soil C:N was lower for the natural aspen site). Thus, Nikanotee upland has

highly depleted soil TN concentrations and lower net mineralization rates. However, ext-N may be approaching that of natural boreal forests, which are typically low N environments. Thus, N deposition does not appear to have increased soil net mineralization in the reclaimed upland and rather N deposition may (currently) be beneficial to the upland as it develops.

Lower net nitrification and net mineralization in the upland (despite more aerobic conditions) (Macrae et al., 2013, 2006), suggests microbial suppression may be occurring in the upland. Soil stockpiling, erosion, low OM content, and contamination can compromise or even destroy soil biota and impact microbially facilitated nutrient cycling (McMillan et al., 2007; Nkongolo et al., 2016). Stockpiling soils for reclamation can be detrimental to mycorrhizal fungi that trees rely on for nutrient cycling (McMillan et al., 2007). Runoff events are small in the sub-humid climate of the WBP, although infrequent hydrological flushing events that carry fluxes of N ions to the fen may occur about once every 20 years (Devito et al., 2005b). Nitrogen runoff rates were low in the upland in 2015 (Gingras-Hill et al., 2018) and have likely continued to decrease with vegetation establishment and litter cover. Organic matter content, however, is very low in the reclaimed upland. In 2015, OM content was ~19% (top 10 cm) (Gingras-Hill et al., 2018), and decreased to <6% in 2019. With time, litterfall accumulation and decomposition will increase OM, subsequently increasing net mineralization processes. Finally, tailings sand contains high hydrocarbon content, which may reduce or inhibit the inherent microbial activity of the soil capping layer (Rowland et al., 2009). Although negligible net nitrification may reflect efficient microbial immobilization of current N deposition (Laxton et al., 2010), low OM suggests otherwise. Future studies should consider soil microbial analysis in the upland (particularly nitrifying bacteria), since microbial communities are one of the key attributes for assessing the biogeochemical functioning of upland soils (Quideau et al., 2013). Additionally, low soil

biodiversity (and low OM) can impair biological activity (Nkongolo et al., 2016), which could have implications on C sequestration (McMillan et al., 2007).

Young forest stands depend largely on soil as an N source and are highly effective at retaining N in their biomass (Fenn et al., 1998; Stoddard, 1994). Extractable N of both tree types was greater in litter than soil, and the litter $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ decreased, reflecting accumulation of both inorganic N forms in vegetation. This was even more apparent in TN concentrations between soil and vegetation. Deciduous trees had significantly higher TN in their foliage than conifers, which is typical of broadleaf species (Kristensen et al., 2004; Laxton et al., 2012). Reclaimed upland trees have high N demand as they are rapidly developing and accumulating biomass, which makes nutrient loss low and N saturation unlikely (Fenn et al., 1998).

Potential Implications of N Deposition on a Reclaimed Fen-upland

One of the main objectives of the Nikanotee Fen Watershed is to create a biodiverse ecosystem that consists of typical boreal species (Daly et al., 2012). Fen ecosystems are naturally composed of a mix of shrubs, vascular vegetation, and bryophyte species. Bryophytes are important to fen ecosystems because they contribute to C accumulation and sequestration, but they are also beneficial at filtering N deposition (Wieder et al., 2019). Critical loads of 10-20 kg N ha⁻¹ yr⁻¹ and 15-35 kg N ha⁻¹ yr⁻¹ are recommended for European poor and rich fens, respectively (Bobbink et al., 2010). However, a critical load of 3 kg N ha⁻¹ yr⁻¹ is recommended for poor fens in Alberta (Wieder et al., 2020). It is expected that surpassing this critical load in natural fens would result in a decrease of bryophytes and an increase in vascular vegetation, with a decrease in overall fen diversity (Bobbink et al., 2010). In summer 2018, N deposition to Nikanotee fen was 4.07 kg N ha⁻¹, which can be estimated to be >10 kg N ha⁻¹ yr⁻¹ (Cameron et al., 2021), and in summer 2019 it was 2.1 kg N ha⁻¹ (8.5 kg N ha⁻¹ yr⁻¹). As such, bryophytes at Nikanotee fen may find it difficult

to establish and survive on the reclaimed site, restricting the potential for a diverse fen ecosystem. However, other factors may also restrict establishment of bryophytes, including shading by vascular vegetation (and litter) (Limpens et al., 2003; Bubier et al., 2007) as well as elevated salinity levels from oil sands process water (Pouliot et al., 2013). Nitrogen deposition is likely fueling productivity of vascular vegetation in Nikanotee fen, which could have long-term implications on water-use, especially with a warming climate. Fen water-levels in addition to N deposition loads affect vegetation growth responses (Thormann & Bayley, 1997). Thus, future research in the reclaimed fen should assess the response of experimental N addition on the productivity of the dominant vegetation species in coordination with water-level fluctuations.

Considering soil OM content, soil N availability, and net mineralization processes are all low in the reclaimed upland, tree growth is likely benefiting from additional N inputs. Nitrogen deposition to mature forests has been found to be far greater in TF deposition compared to open deposition, resulting in increased N fluxes to the forest floor from the accumulation of deposition within the canopy (Fenn et al., 2015; Proemse et al., 2013). This could result in N enrichment to the forest soil (Pelster et al., 2009), increasing net mineralization processes (Fenn & Poth, 1998). However, low OM and low soil biota may inhibit microbially facilitated N mineralization (Nkongolo et al., 2016), resulting in excess N leaching. It is imperative that future studies assess soil microbial communities in the upland to better understand net mineralization of the site and how it may react to N deposition.

When considering soil bulk density and vegetation biomass, N content in vegetation (for both the fen and upland) is small relative to total bulk N deposition. Indeed, there is more N in the top 0-15 cm of soil than is deposited, but at the same time, the constructed fen-upland still receives a considerable bulk N deposition input. However, overall, the fen and upland appear to be low N

environments (based off the ecosystem indicators within the soil-water-vegetation continuum of both landscape units) even though the reclaimed system is situated within an increased N deposition zone in the AOSR. Nitrogen deposition on the reclaimed site is chronic low-level fertilization that is likely stimulating plant growth. In the reclaimed fen, this is beneficial to vascular vegetation growth, but is unfavourable to the diversity of the fen; and in the reclaimed upland, N deposition may be beneficial to tree development, considering low OM and low soil net mineralization processes.

3.6 Conclusion

This is the first study to quantify N deposition to a post-mined constructed fen-upland in the AOSR to assess the N-status of a reclaimed ecosystem subjected to exogenous N inputs. Although the reclaimed system is situated directly within a high N deposition zone in the AOSR, the N-status of the fen and upland do not suggest that the reclaimed site is N saturated or in the early stages of N saturation. According to the stages of N saturation laid out by Aber et al. (1989), the reclaimed site is likely between stage 0-1 (closer to 0), where despite higher-than-background N deposition inputs, the enriching impacts of N deposition on the ecosystem are not evident (based off various ecosystem receptor indicators) and N losses are negligible. Since N is a limiting nutrient in the boreal environment, high N deposition is likely acting as a fertilization agent, boosting ecosystem productivity and growth of the reclaimed system (Stoddard, 1994). Nitrogen deposition inputs have not yet increased N availability of the reclaimed fen-upland (indeed N availability is sub-optimal), thus N retention is efficient. Studies conducted on natural ecosystems in the AOSR suggest that these ecosystems may be in the early stages of N saturation (i.e., Stage 1) (Laxton et al., 2010), therefore N-status of the reclaimed site should be continually monitored as both the fen

and upland continue to establish. It may only be a matter of time for chronic N deposition accumulation to occur that could reduce N-retention mechanisms (i.e., vegetation and microbial uptake) and increase N-status of the system, which could threaten N saturation and ultimately result in detrimental vegetative and ecosystem community responses.

4.0 Conclusions, Limitations & Future Research

4.1 Conclusions

This study assessed nitrogen (N) deposition loads across four boreal ecosystems moving southwards from the industrial centre in the Athabasca Oil Sands Region (AOSR), and the implications of N deposition on the N-status of a reclaimed fen-upland located directly within the industrial centre. In general, N deposition loads decreased southwards from the industrial centre, following previous patterns of decreasing N deposition with distance outwards from mining operations. The susceptibility of ecosystems to the nutrient enriching impacts of N deposition are expected to align with the spatial pattern of N deposition, making the reclaimed fen-upland (situated in a heavily industrial area) the most susceptible to N enrichment. However, steam-assisted gravity drainage (SAGD) activities and other industrial activities may create isolated N patches within the AOSR, separate from the main industrial centre. Excessive N inputs to typically low N boreal ecosystems can result in the accumulation of nitrogenous compounds, leading to enhanced N availability and increased ecosystem N-status. This may help boost plant productivity (initially) but may ultimately result in shifts in community composition and a decrease in ecosystem diversity (Bobbink et al., 1998; Macdonald, 2015; Greaver et al., 2012). Although N deposition may also result in ecosystem acidification, this is of little concern to the reclaimed fen-upland as the reclaimed site likely receives ample amounts of base cation (BC) deposition from fugitive dust due to nearby haul road activity (see Figure A.1), buffering the effects of acid deposition (Watmough et al., 2014). Thus, the N enriching impact of N deposition is the top concern to the reclaimed fen-upland. Since the goal of the Nikanotee fen-upland reclamation project is to construct a self-sustaining, carbon accumulating and biodiverse boreal ecosystem (Daly et al., 2012), it is important to examine exogenous N inputs and assess whether this N source

is increasing the N-status of the fen-upland, as ecosystem N enrichment could impact overall reclamation goals and objectives. Furthermore, N enrichment, particularly in fen peatlands, may increase plant productivity (especially of vascular species) (Bubier et al., 2007; Bragazza et al., 2006), which could increase plant water-use and have long-term impacts on the hydrology of the system.

The first objective of this thesis assessed N deposition from a large-scale perspective to quantify N deposition loads up to 65 km to the south from the industrial oil sands centre in the AOSR. In general, N deposition was greatest near the industrial centre and decreased with distance southward. Nitrate ($\text{NO}_3\text{-N}$) deposition was significantly higher at the reclaimed fen-upland than all reference sites. Both ammonium ($\text{NH}_4\text{-N}$) and total inorganic N (TIN) deposition were only significantly different than the reference site located 65 km away, but were greatest overall at the reclaimed fen-upland. Quantification of N deposition loads and conclusions from Manuscript 1 will assist in the creation of an integrative nutrient framework that will further our understanding of the biogeochemical functioning of disturbed and natural ecosystems in the AOSR (Nwaishi et al., Under review). Overall, results from Manuscript 1 show that the reclaimed site received the greatest N deposition loads due to close proximity to mining activities and N emission sources. This highlights the susceptibility of the reclaimed ecosystem to exogenous N and the potential for excess N inputs to impact the soil-water-vegetation continuum of the system. Manuscript 2 (and the next two objectives) assessed the impact of N deposition on the soil-water-vegetation continuum specifically of the reclaimed Nikanotee fen-upland situated directly within industrial oil sands activities (small-scale perspective).

In the fen, N availability was greatest in vegetation (and litter) of each of the three dominant species (where N is typically largely stored within the ecosystem continuum) ~7 years post-

construction. No significant differences in N storage were observed between the three species. Within the water column, $\text{NH}_4\text{-N}$ and $\text{NO}_3\text{-N}$ were balanced in both surface- and groundwater (based off $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio). However, $\text{NH}_4\text{-N}$ and $\text{NO}_3\text{-N}$ were unbalanced in porewater, due to a large decrease in NO_3^- concentration, resulting in a higher ratio. This indicates active uptake of NO_3^- by vascular vegetation (Wiedermann et al., 2009). Nitrogen availability in soil was low compared to natural fens in the region and compared to the first summer post-construction (in 2013). Soil NO_3^- was very low, again reflecting active uptake of NO_3^- by vegetation and soil microbes. Furthermore, soil net mineralization rates were low, especially for nitrification, which was much lower than 2013 nitrification rates. Although the reclaimed fen is situated in a high N deposition zone in the AOSR, N deposition does not appear to be increasing N availability or net mineralization processes, suggesting that the fen is a low N environment.

In the upland, N availability was greatest in groundwater, likely due to leaching of NH_4^+ through the tailings sand. However, vegetation N was still high and the $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio was low, reflecting storage of both inorganic N forms. Furthermore, greater total N concentration (TN) in vegetation over soil also reflects N storage in vegetation, especially since young forests greatly depend on soil as an N source (Fenn et al., 1998). Between both tree types, vegetation TN was significantly greater in deciduous compared to coniferous trees, which is typical of broadleaf species. In terms of stemflow (SF), both tree types were more concentrated in $\text{NO}_3\text{-N} > \text{NH}_4\text{-N}$ likely due to canopy consumption of $\text{NH}_4\text{-N}$, which is beneficial to vegetation growth in low N soils. Soil N availability was low, but only slightly lower than other reclaimed uplands. However, soil TN concentrations were highly depleted. Furthermore, soil net mineralization rates were very low (lower than the anaerobic fen), suggesting potential microbial suppression in the upland. Other factors such as low organic matter (OM) content also suggest compromised microbial

mineralization. Although the reclaimed upland is situated in a high N deposition zone in the AOSR, N deposition does not appear to be increasing N availability and net mineralization processes, suggesting that the upland is also a low N environment.

The third objective was to determine the N-status of the fen and upland, and assess the potential implications of N deposition on the development of the reclaimed system. Based on multiple ecosystem receptor indicators (i.e., $\text{NH}_4\text{-N}:\text{NO}_3\text{-N}$ ratio, N solution chemistry profile, soil and foliar TN, C:N ratio and soil net mineralization processes), N deposition does not appear to be influencing the N-status of either landscape unit. Although the reclaimed site received higher-than-background N deposition loads, the fen and upland are both low N environments. In the fen, N deposition is likely fueling the growth of vascular vegetation, which will make it difficult for a diverse vegetation community consisting of bryophytes to establish in the future. In the upland, N deposition is likely beneficial to tree development especially if substrates used in reclamation cannot sustain microbial communities and provide adequate N for plant growth. As the upland canopy closes, N deposition will accumulate within the foliage, resulting in increased SF (and throughfall) N fluxes below the canopy. This may help in increasing soil net mineralization but should be monitored as N enrichment could occur with time.

4.2 Limitations & Future Research

The reclaimed fen and upland were assessed as two separate landscape units in this thesis rather than a joined landscape. Runoff events are typically low in the sub-humid WBP climate (Devito et al., 2005b) and N concentrations in runoff were not assessed in summer 2019. However, the upland regularly provides the fen with water via groundwater and thus groundwater N will migrate from the upland into the fen. Future research should connect N fluxes between both landscape

units and surrounding slopes to create an N budget that thoroughly quantifies and examines all N inputs and outputs (i.e., N₂-fixation and denitrification) of the reclaimed system.

In the fen, lysimeters had to be elevated half-way through summer because of heavy rainfall events increasing surface water levels. Elevating the samplers may have resulted in greater N concentrations in surface water leachate due to peat drying in-between rain events. Future research should consider alternative methods to collect surface water leachate particularly in pooled sample plots.

In the upland, surface- and porewater N were not determined due to compact mineral soil and difficulty installing instruments. Due to this limitation, determining near-surface bioavailable N concentrations in the upland was not possible. Future research should develop methodologies to examine this portion of the soil-water-vegetation continuum, which will further our understanding of how much N is readily available for root uptake of developing trees.

Deposition studies conducted in the AOSR show that N is not the only compound emitted by industrial activities and deposited onto surrounding ecosystems. Compounds such as inorganic sulphur are also a concern to the environment within the vicinity of industrial activities and are likely being deposited onto the reclaimed ecosystem (Proemse et al., 2012; Wieder et al., 2010). Future research should investigate the biogeochemical and ecological impacts of these other prevalent compounds on reclaimed landscapes in the AOSR.

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Appendix

Images



Figure A.1: Nikanotee fen-upland during A) clear-sky conditions and B) dusty conditions in summer 2019.