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The impact of atmospheric acid deposition on tree growth and forest understory vegetation in the Athabasca Oil Sands Region

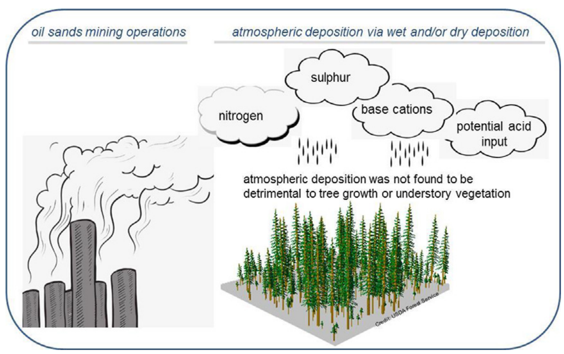
Samuel F. Bartels, Benoit Gendreau-Berthiaume [1], S. Ellen Macdonald *

Department of Renewable Resources, University of Alberta, Edmonton, Alberta T6G 2H1, Canada

HIGHLIGHTS

- Deposition was not found to be detrimental to tree growth or understory vegetation.
- Jack pine growth was higher in areas that later received higher deposition.
- The positive growth relationship with deposition weakened in the active mining era.
- Understory plant cover and richness increased with atmospheric N and S deposition.
- No effects of soil-mediated acidification on tree growth and understory vegetation

GRAPHICAL ABSTRACT



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ABSTRACT

Atmospheric acid deposition is of major concern in the Athabasca Oil Sands Region (AOSR) in northern Alberta, Canada, which is home to the third largest oil reserve in the world. After decades of oil sands production in the AOSR, the potential impact of deposition on forest health, including tree growth and understory biodiversity, is still not clear. We evaluated the relationship of modelled/interpolate atmospheric deposition of nitrogen (N), sulphur (S), base cations (BC), and derived potential acid input (PAI) from surface oil sands mining with: (1) the radial growth (i.e. basal area increment; BAI) of jack pine (*Pinus banksiana* Lamb.) trees using data from two decadal time periods, prior to (1957–1966) and during (2001—2010) active oil sands development in the AOSR; and (2) forest understory vegetation (abundance, diversity, and composition), which is an important component of forest biodiversity. BAI of jack pine trees varied with N, S, and BC deposition between the two time periods, and with the direction of the site relative to main emission sources. Growth was higher in areas close to the oil sands surface mining operations prior to and after oil sands development. BAI was also positively related to atmospheric deposition in the recent period, but these relationships were weaker in the active period versus the non-active period. Understory vegetation – including vascular plant cover, richness, and diversity – increased in relation to modelled atmospheric N and S deposition. There was limited correlation between soil pH or the BC:Al ratio (indicators of soil acidification) and BAI and understory vegetation responses. No evidence was found for detrimental effects of atmospheric emissions (and subsequent deposition) from oil sands production on tree

* Corresponding author.
E-mail address: ellen.macdonald@ualberta.ca (S.E. Macdonald).
[1] Current address: Institut des Sciences de la forêt tempérée, Université du Québec en Outaouais, Ripon, Québec, JOV 1VO.

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growth or forest understory vegetation. The results, if anything, suggest a fertilization effect due to enhanced atmospheric deposition of nitrogen compounds.

1. Introduction

Atmospheric deposition of nitrogen (N), sulphur (S), and inorganic elements from natural and anthropogenic sources via wet (rain, snow, and fog) and dry deposition (gases and particulates) is both a global and regional phenomenon that requires attention (Bouwman et al., 2002; Phoenix et al., 2006; Vitousek et al., 1997). The potential impacts on natural ecosystems are many including: acidification, eutrophication, alteration of nutrient availability, net primary productivity and carbon sequestration, and increased susceptibility to secondary stress and disturbances (Bobbink et al., 2010; Galloway, 1995; Simpson et al., 2006; Smith et al., 1999; Wei et al., 2012).

In forest ecosystems, the impacts of deposition are manifested as changes in the structure and function of the forests including the growth and development of trees and ground vegetation. Although the effect of atmospheric deposition of N and other acidifying compounds on forest ecosystems has received considerable attention, the outcomes are often debated. For instance, while some reports suggest forest decline or reduced tree growth is a consequence of soil acidification and chronic deposition levels and critical loads (e.g. Duchesne et al., 2002; Nellemann and Thomsen, 2001; Sverdrup et al., 1994), some other studies that assessed similar parameters in fertilized forests found little or no evidence for long-term growth or risks associated with deposition (e.g. Binkley and Hogberg, 1997; Binkley and Hogberg, 2016; Solberg et al., 2004).

Atmospheric deposition has also been linked to changes in forest plant biodiversity (Bobbink et al., 2010; Gilliam, 2006; Gilliam, 2019). For instance, N deposition has been suggested to result in increased abundance of nitrophilous species – including grasses, forbs and shrubs (e.g., Brunet et al., 1998; Gilliam et al., 2016; Nordin et al., 2005; Thimonier et al., 1992). In general terms this is often interpreted as a 'fertilization effect', whereby enhanced N availability stimulates plant growth. Excess N deposition can also result in decreased: herbaceous layer richness and diversity (Gilliam et al., 2016; Gilliam, 2019; Walter et al., 2017); and abundance and diversity of forest floor mosses, which are particularly sensitive to nitrogen and sulphur deposition (Mäkipää, 1995; Strengbom et al., 2003). The overall effects are often manifest as major shifts in the understory plant community composition driven primarily by increases in cover of a few nitrophilic species at the expense of many other species (Bobbink et al., 2010; Gilliam et al., 2016; Walter et al., 2016; Walter et al., 2017).

In addition to the general fertilization effects of atmospheric deposition, there is also the potential effect of soil-mediated acidification due to nitrification of excess N (Lu et al., 2014) and S deposition in soils (Nyborg et al., 1991). This acidification effect, which manifests as a change in soil acidity and depleted nutrient cation status, has been linked to reduced growth and vigour of trees (Duchesne et al., 2002; Sullivan et al., 2013), reduced herbaceous richness (Hallbacken and Zhang, 1998), and shifts in plant species community composition (Stevens et al., 2010).

In the Athabasca Oil Sands Region (AOSR) in northern Alberta, Canada, which has the third largest oil reserve in the world, much remains unknown about the potential impacts of deposition from these mining operations on forest ecosystems, including tree growth and plant biodiversity. The emissions and deposition of acidifying compounds and trace elements arising from industrial development in the AOSR have raised concerns about the state of the surrounding forest ecosystems. In the 1990's, as the oil sands industry was entering a period of expansion, a forest health monitoring program (focused on forest soils, tree growth, foliar nutrients, understory vegetation and lichen bio-indicators) was established to monitor for changes in jack pine forests, considered to be the most sensitive systems to acidic deposition (Foster et al., 2019). Although regional assessments of effects of emissions (e.g., SO_2, NO, NO_2) on air quality and terrestrial environments have been undertaken to varying degrees (Clair and Percy, 2015; Percy et al., 2012), the impacts on growth patterns of trees and forest biodiversity in the AOSR have not been extensively documented.

The present study was undertaken to assess the relationship of atmospheric acid deposition from industrial oil sands operations to the present state of the forest ecosystem in the AOSR. The specific questions posed were whether atmospheric deposition of acidifying substances, primarily N, S, and base cations (BC; Ca^{2+}, K^+, Mg^{2+}, Na^+), and potential acid input were related to (1) the radial (basal area) growth of jack pine (*Pinus banksiana* Lamb.), which is a relatively fast growing conifer, and (2) the abundance, diversity and composition of understory plant communities, which are an essential component of forest biodiversity. Earlier studies in the AOSR indicated limited potential for acidification of soils under current deposition levels (Jung et al., 2013; Whitfield et al., 2009) owing in part to base cation deposition that has the capacity to both neutralize acid input and function as a plant nutrient (Fenn et al., 2015; Watmough et al., 2014); therefore, we expected little or no influence of soil acidification. Rather, we hypothesized an increase in tree growth and understory vegetation abundance, richness, and diversity as a response to the nutrient availability, particularly N and BC deposition. For sites located at various distances and directions from the oil sands production emission sources, we compared the relation of atmospheric deposition with tree growth prior to initiation of industrial oil sands development to that in a more recent decade during which resource development activities had substantially expanded. We expected no relationship of current atmospheric deposition to tree growth prior to oil sands development.

2. Methods

2.1. Study region and sites

The primary sources of atmospheric deposition in the AOSR come from oil sands production activities, such as stack emissions, raw oil sands and processed materials, tailing sands, haul roads emissions, as well as limestone aggregates produced from Devonian bedrock exposed at the base of oil sands mines and from independent quarry operations in the core of the oil sands development. The Wood Buffalo Environmental Association (WBEA) has been monitoring air quality and effects on the terrestrial environment in this region since 1997. Reports on air quality indicate that atmospheric acid deposition amounts are enhanced within 30 km of the operations, decline with increasing distance from them, and reach background levels ~40–50 km away from main industrial emission sources (Clair and Percy, 2015). The present study forms part of WBEA's Terrestrial Environmental Effects Monitoring (TEEM) program designed to investigate the potential effects of air pollutants from oil extraction and upgrading operations on natural ecosystems (Foster et al., 2019). Field data were collected across a network of long-term monitoring sites established through the TEEM program for the purposes of forest health monitoring. Site selection for the monitoring program followed a protocol described in Foster et al. (2019). There are presently 27 forest interior and 25 forest edge research monitoring sites strategically located within ~100 km distance in all directions from major surface oil sands production emission sources in the region (see Fig. 7 in Foster et al., 2019); and these comprise a suite of jack pine-dominated forest stands growing on coarse-grained Eluviated Dystric Brunisol soils, which were considered to be most sensitive to atmospheric acid deposition from oil sands operations in the region (Clair and Percy, 2015). The sites were all classified as the a1.1 Pj/bearberry/lichen (*Pinus banksiana*/*Arctostaphylos uva-ursi*/*Cladina* spp.) ecosite phase (Beckingham and Archibald, 1996). The soils belong to the Mildred soil series and originated from coarse glaciofluvial parent materials of shallow calcareous bedrock, Aeolian deposits, or sand-textured glaciofluvial deposits.

The understory vegetation was heavily dominated by ground lichens – primarily *Cladonia* spp. – with low cover and diversity of vascular plants. The forest edge sites (situated at the interface between the forest and adjacent open areas, typically wetlands), were considered to be more exposed to air pollutants than the forest interior sites, and therefore established as early warning monitoring sites. These dry, nutrient poor jack pine sites had similar densities and ages (stand age ranged between 60 and 70 years at the time of sampling (2011−2013)), had never been managed, and were initiated following stand-replacing wildfire, which is the dominant natural disturbance in the region. The present study considered sites that had escaped recent wildfires in the region, and therefore included 19 forest interior and 24 forest edge sites; tree core data were also available from two sites that burned in a wildfire in 2011 (Appendix Table A1). Each site was categorized into one of four cardinal directions (north [N], east [E], south [S], and west [W] directions) from the center of major surface oil sands production emission sources in the AOSR and this was used to explore the existence of spatial patterns. Field data (tree cores, understory vegetation, and soil data) for this study came from the 2011–2012 major field season, as part of the ongoing forest health monitoring program (Foster et al., 2019).

2.2. Deposition measurements and estimates

Atmospheric deposition of N, S, and base cations were based on measurements of throughfall or bulk deposition of sulphate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), calcium (Ca^{2+}), potassium (K^+), magnesium (Mg^{2+}), and sodium (Na^+), plus dry deposition fluxes of nitrogen species (NO_2, HNO_3, NH_3, particulate-NH_4^+, and particulate-NO_3^-). Ion exchange resin collectors (IERs) were used to measure bulk deposition and throughfall deposition, while dry deposition fluxes of nitrogen species were calculated using the passive data and the Multi-Layer Dry Deposition Model (MLM). Deposition estimates were derived by spatially interpolating various time-based averages and model equations outlined below and described in detail in Edgerton et al. (2019) to arrive at specific quantities of interest. The present study used actual average (2009 to 2012) annual atmospheric deposition for sites where actual data were available and interpolated estimates for other sites for: total N (combined HNO_3^-, NH_3, NH_4^+, NO_2, and NO_3^- deposition estimates) and S (SO_4^{2-} throughfall deposition estimates), base cations (BC; sum of Ca^{2+}, K^+, Mg^{2+}, and Na^+), and potential acid input (PAI; defined as the between total (wet + dry) deposition of S + N species [$SO_x + NO_x + NH_x$] and estimated total deposition of neutralizing species [a.k.a. base cations: Ca, Mg, K Na]) (See base maps in Edgerton et al., 2019 Figs. 3, 14, and 21). The specific equations are as follows:

$$\text{Total sulfur deposition (Sdep)} = \text{Sdep}_W + \text{Sdep}_D \sim \text{Sdep}_B + \text{Sdep}_D \sim \text{Sdep}_{TF} \quad (1)$$

$$\text{Total nitrogen deposition (Ndep)} = \text{Ndep}_W + \text{Ndep}_D \sim \text{Ndep}_B + \text{Ndep}_D \quad (2)$$

$$\text{Base cation deposition (BCdep)} = \text{BCdep}_W + \text{BCdep}_D \sim \text{BCdep}_B + \text{BCdep}_D \sim \text{BCdep}_{TF} \quad (3)$$

$$\text{PAI} = \text{Sdep} + \text{Ndep} - \text{BCdep} \quad (4)$$

where, dep_W = measured or modelled wet deposition; dep_D = measured or modelled wet deposition; dep_B = measured bulk deposition; dep_{TF} = measured throughfall deposition.

2.3. Tree core sampling and measurements

In all sites, between 10 and 20 dominant or co-dominant (crown forming or belonging in the upper canopy) jack pine trees with diameter at breast height (DBH; 1.3 m above the root collar) > 10 cm were selected for tree core samples. For each selected tree an increment borer was used to obtain a core through the pith at 1.3 m height, which was then placed in a plastic straw with the ends closed, labelled and brought to the lab for ring growth analysis. In the lab, the cores were dried, securely mounted, and sanded to make the rings visible. Samples were then scanned, with a high-resolution scanner (Epson V750 Pro) at a resolution between 2400 and 4800 dpi (according to ring width) and rings were measured (to the nearest 0.01 mm) on the images using the CooRecorder Software (Larsson, 2008a). The CDendro software (Larsson, 2008b) was then used to cross date samples by comparing every sample to the average plot chronology to find measurement mistakes or potential missing rings. Cores were rejected when they could not be accurately cross-dated. Data from a total of 494 sample trees across all sites were used for subsequent analyses.

Ring width sequences, along with the DBH measurement, were used to determine tree age and to calculate average yearly growth rates in basal area (i.e. the cross-section of tree trunk or stem at the base) increment (BAI; expressed as cm^2/year). Utilizing both ring width data and DBH measurements this way allowed the determination of how much the tree grew, in terms of basal area, in each particular year by subtracting growth in the initial year from that of the current year. BAI was used to remove age-related growth trends while maintaining suppression or release events caused by potential disturbances or changes in deposition. Annual basal area incremental growth for each year was estimated as the difference in basal area of the present year (last year of growth) and the basal area of the tree up to the year previous. For each site, an average basal area increment chronology was generated. For the present study, basal area growth analysis was examined for two contrasting decadal timeframes; one that predates industrial oil sands mining (from 1957 to 1966; hereafter "non-active period") versus a recent period of active industrial oil sands mining and upgrading operations in the region (from 2001 to 2010; hereafter "active period"). For each sample tree within a site, basal area growth for the non-active and active periods were calculated as the average BAI over ten-year time intervals, 1957 to 1966, and 2001 to 2010, respectively. The trees were thus ~10 years old at the start of the non-active growth period and ~55 years old at the start of the active period. Five sites were excluded from analyses for tree growth because the trees were too young and one site was removed because it has an unusually low tree density and high diameter growth (Table A1).

2.4. Vegetation and soil sampling

Within each site, a vegetation plot measuring 10 m × 40 m was established for understory vegetation surveys. The plots were located within a minimum of three tree heights (approximately 50 m) distance from the forest edge, road, or other disturbances. Within the vegetation plot, 5–10 small (0.4 m × 1 m), 2 medium (1 m × 1 m), and 1 large (2 m × 20 m) subplots were delineated. In each subplot, the presence and percent cover of all vascular and nonvascular species were visually estimated and recorded. All vascular plants were identified to species, including tree species that occurred as seedlings or saplings. The dominant forest floor mosses and lichens were also identified to species, (except some *Cladonia* species other than *Cladonia uncialis* which were identified to the genus level).

At each sampling site, soil pits were dug to characterize soil physical and chemical properties at varying depth increments (Foster et al., 2019). Soil sampling included the forest floor organic (LFH) and mineral soil horizons for the determination of the following characteristics: pH, elemental carbon (C), N, and S, inorganic N (NH_4^+-N and NO_3^--N, mg kg^{-1}) and exchangeable cations (Al, Ca, Fe, K, Mg, Mn, and Na, cmol (+) kg^{-1}). Detailed chemical analyses and procedures are reported in MacKenzie and Dietrich (2019).

2.5. Data analyses

2.5.1. Basal area growth

The relationship between jack pine tree BAI and current atmospheric deposition was analyzed using linear mixed effect models that included the estimated deposition variables (total N, S, BC, and PAI), time period (non-active [1957–1966] versus active [2001−2010] period BAI), site location (N, E, S and W directions), and their interactions as fixed effects and trees nested in site as the random effect. Site type (forest edge or interior) was not included in the model since BAI did not differ significantly between the edge vs interior forest sites. The models were constructed using the *lme* function in the NLME package in R software version 3.2.1 (R Development Core Team, 2015). When the interaction term (two-way or three-way interaction) was significant (at $\alpha = 0.05$), i.e., the relationship with deposition differs between the two time periods and/or locations, the *lstrends* function in the LSMEANS package (Lenth, 2016) was used to test for significant differences in the slope of the growth versus deposition relationship between the two time periods and/or locations.

In the course of these analyses we determined that there was a spatial pattern of growth differences whereby trees in areas that receive higher deposition in the current period (i.e., in general, were closer to the oil sands surface mining, areas) were growing faster even before the start of oil sands mining activities (i.e., tree growth in the non-active period was correlated with deposition in the current period). We thus wanted to focus on the difference in growth between the two time periods while factoring out this pre-existing pattern; we did this by calculating the difference in growth between the two time periods for every tree ($\Delta BAI_{\text{difference}} = BAI_{\text{active}} - BAI_{\text{non-active}}$) and then including growth in the non-active period as a co-variate in a regression of the difference in growth on each deposition variable. This allowed us to assess how the change in growth over time relates to current levels of deposition. A two-sample *t*-test was used to compare BAI averages between the active and non-active period.

2.5.2. Understory vegetation

For richness we used the species list from all sub-plots combined. To get an idea of community composition we combined data from all the different sub-plots. Our rationale was that the smaller plots probably had more accurate measures of cover but larger plots had better representation of the species present and better cover values for larger plants (taller shrubs). So mean cover per species was calculated for each subplot (small, medium, and large) and then averaged across all subplots to obtain the best possible representation of cover per species at the site. Cover (% summed across taxa), species richness (number of species), and diversity (assessed as equivalent Simpson index following Hill numbers [Hill, 1973] to account for unevenness in species abundance) values were calculated from these data for the following understory vegetation categories: total (all vascular and nonvascular plants, including mosses and lichens); vascular plants; shrubs (all woody species, including tree seedlings or saplings); forbs (here including broadleaf non-woody species plus prostrate or trailing woody species); grasses (included graminoids and sedges); mosses; and lichens.

The relationships of understory vegetation cover and richness to the estimates of atmospheric deposition (total N, S, BC, and PAI) were assessed using linear regression models (analyzed separately for total, mosses, lichens, vascular plants, forbs, shrubs). The relationships of species richness and cover with each independent atmospheric deposition variable were assessed to first determine which variable(s) were significantly related. We had planned this screening to inform which variables to include in a subsequent multiple regression model. However, due to high multicollinearity among independent variables, particularly between N and S deposition ($r = 0.94$), we had to conduct separate analyses for each independent deposition variable. The model residuals were checked for conformity to the assumptions of normality and homogeneity of variance. No transformations were required to meet these assumptions. We also assessed whether there were linear or non-linear relationships between the response and predictor variables, and in all cases the linear relationship was best. Final linear regressions were conducted for those variables that were significant in the initial screening; these final regressions included the deposition variable along with location of the sites, and site type (forest edge or interior).

The relationship of atmospheric deposition to understory species composition was analyzed using redundancy analysis (RDA), which is a constrained ordination method that summarizes the variation in a set of response variables that can be explained by a set of explanatory variables (Legendre and Legendre, 1998). The species composition data consisted of Hellinger-transformed species by site cover values for each vascular and nonvascular species. The constraining variables consisted of each individual atmospheric deposition variable and the three-way interaction with site type and location, as described above. RDA was performed in R by specifying the Bray-Curtis distance in the VEGAN package (Oksanen et al., 2018), and statistical significance of model outputs was assessed using permutation procedures ($n = 999$).

2.5.3. Correlations with soil chemistry data

To determine whether there were any relationships between soil chemical properties and tree growth or understory vegetation, as well as to examine whether there was any evidence of soil-mediated acidity or alkalinity on basal area increment and understory response patterns, we analyzed correlations of soil variables (see MacKenzie and Dietrich, 2019 for details) with tree growth, the difference in growth between the two time periods, and understory vegetation variables using Spearman's rank correlations. This was done only for the forest interior sites because soils data were not available for the forest edge sites.

3. Results

3.1. Site characteristics

Estimated atmospheric deposition for total N ranged from 0.14 to 0.41 keq ha^{-1} yr^{-1}, S ranged from 0.12 to 0.87 keq ha^{-1} yr^{-1}, BC from 0.17 to 0.86 keq ha^{-1} yr^{-1}, and PAI from −0.46 to 0.79 keq ha^{-1} yr^{-1} (see also Edgerton et al., 2019). A total of 96 understory plant species (11 shrubs, 9 grasses, 39 forbs, 21 bryophytes, and 16 lichens) was recorded across all study plots (see species list in Table A2). Soil chemical properties, such as BC and cation exchange capacity (CEC) of the organic (LFH) soil horizon, respectively, ranged from 19.7 to 40.9 cmol (+) kg^{-1} and 18.1 to 39.6 cmol (+) kg^{-1}, pH from 3.5 to 4.3, and the BC:Al ratio from 11.8 to 51.5. Similarly, BC and CEC of the top mineral soil (0–5 cm) horizon, respectively, ranged from 0.4 to 2.6 cmol (+) kg^{-1} and 0.7 to 3.1 cmol (+) kg^{-1}, pH ranged from 3.4 to 4.9, and BC:Al from 1.5 to 24.1 (see also MacKenzie and Dietrich, 2019).

3.2. Relationship of atmospheric deposition to jack pine basal area growth

For BAI of jack pine the three-way interaction of deposition level, time period, and site location was significant for deposition of N, S, and BC (Table 1). Surprisingly, growth in the non-active period between 1957 and 1966, prior to oil sands industrial resource development in the AOSR, was positively related to estimated current atmospheric deposition of N, S, BC, and PAI, particularly for the north and south locations (Fig. 1). For sites east and west of the main oil sands surface mining operations there was no significant relationship with estimated current

Table 1
Results of linear regressions examining the influence of atmospheric deposition, as modelled for the current time period, on jack pine (*Pinus banksiana* Lamb.) basal area growth in the non-active and active periods of industrial oil sands mining development in the Athabasca Oil Sands Region, and for different directions relative to the oil sands mining activity.

Model terms	DF	*F*-value	*p*-value
a) Total nitrogen (TN)			
TN	1	1.790	0.191
$T_{ime\ period}$ (growth $_{non\text{-}active}$ vs. growth $_{active}$)[a]	1	319.038	<0.001
$D_{irection}$ (N, E, S, W)	3	3.251	0.035
$TN \times T_{ime\ period}$	1	10.830	0.001
$TN \times D_{irection}$	3	3.236	0.036
$T_{ime\ period} \times D_{irection}$	3	16.326	<0.001
$TN \times T_{ime\ period} \times D_{irection}$	3	22.960	<0.001
b) Sulphur (S)			
S	1	1.463	0.236
$T_{ime\ period}$ (growth $_{non\text{-}active}$ vs. growth $_{active}$)[a]	1	315.00	<0.001
$D_{irection}$ (N, E, S, W)	3	2.999	0.046
$S \times T_{ime\ period}$	1	7.737	0.006
$S \times D_{irection}$	3	2.979	0.047
$T_{ime\ period} \times D_{irection}$	3	15.486	<0.001
$S \times T_{ime\ period} \times D_{irection}$	3	22.235	<0.001
c) Base cations (BC)			
BC	1	6.382	0.017
$T_{ime\ period}$ (growth $_{non\text{-}active}$ vs. growth $_{active}$)[a]	1	301.932	<0.001
$L_{ocation}$ (N, E, S, W)	3	7.790	0.170
$BC \times T_{ime\ period}$	1	7.804	0.005
$BC \times D_{irection}$	3	1.081	0.371
$T_{ime\ period} \times D_{irection}$	3	14.303	<0.001
$BC \times T_{ime\ period} \times D_{irection}$	3	14.984	<0.001
d) Potential acid input (PAI)			
PAI	1	0.589	0.449
$T_{ime\ period}$ (growth $_{non\text{-}active}$ vs. growth $_{active}$)[a]	1	300.291	<0.001
$L_{ocation}$ (N, E, S, W)	3	1.760	0.175
$PAI \times T_{ime\ period}$	1	0.402	0.527
$PAI \times D_{irection}$	3	0.587	0.628
$T_{ime\ period} \times D_{irection}$	3	13.290	<0.001
$PAI \times T_{ime\ period} \times D_{irection}$	3	17.411	<0.001

[a] Reference level = active period (2001–2010) of oil sands mining.

deposition. Similar relationships were also observed for basal area growth in the active period; however, the slope of the positive relationship between growth and atmospheric deposition was lower for the active period than for the non-active period (north and south locations, Fig. 1A–D). Comparison of the growth patterns (based on *lstrends* tests for significant differences among the slopes; see Appendix Table A3) indicated no significant differences between the two time periods in the observed growth–atmospheric deposition relationship for N, S, and BC in the east and west locations, whereas the slope of the growth–deposition relationship was greater in the non-active period as compared to the active period for the north and south locations (Fig. 1A–D). The relationship between PAI and growth was weak or non-significant for all locations except for sites located south of the mining area, which showed a positive relationship of growth in the non-active period with estimated current PAI and a similar, but significantly weaker, relationship for the active period. There was no significant difference in the growth-PAI relationship between the two time periods for the north, east or west locations.

Overall, BAI was significantly higher in the non-active period (mean, $\overline{x} = 5.14$ cm^2/year) than the active period ($\overline{x} = 2.97$ cm^2/year) ($t = 11.97$, $p < 0.001$) and the trend in growth showed a decline from the non-active to the active period (Fig. 2). We found a strong negative correlation ($R^2 = 0.80$) between the difference in growth between the two time periods ($\Delta BAI_{difference} = BAI_{active} - BAI_{non\text{-}active}$) and growth in the non-active period; i.e., trees growing faster in the non-active period showed a greater decline in growth between the two time periods (Fig. 3). However, this difference in growth between the two periods was not significantly related to any of the deposition variables, when growth in the non-active period was included as co-variate (Appendix Table A4).

3.3. Responses of understory vegetation cover, richness and composition to atmospheric deposition

Understory vegetation cover, richness, and diversity were related to atmospheric deposition, with the specifics varying by species group (Table 2; Appendix Table A5). Cover of understory vascular plants generally increased with N deposition, but this varied among locations relative to emission source, with a much steeper slope for sites located south of the main emission sources as compared with north and east locations (Fig. 4A; see slope comparisons in Appendix Table A6). The cover of vascular plants, as well as shrubs also increased with S deposition (Fig. 4B–C). Shrub cover increased with PAI (Fig. 4D).

Species richness of understory vascular plants increased with estimated N deposition in all locations (Fig. 4E). Understory diversity for all plant groups combined varied with the interaction between N deposition and site type; it increased with N for sites located in the forest interior whereas it declined for those located at the forest edge (Fig. 4F, Appendix Table A6).

Redundancy analysis indicated no significant relationship of atmospheric deposition to understory species composition (Appendix Table A5). In other words, species composition did not change in response to atmospheric deposition for any particular species group; however, it differed significantly between the two site types, i.e. between forest edge vs forest interior sites (Appendix Table A7).

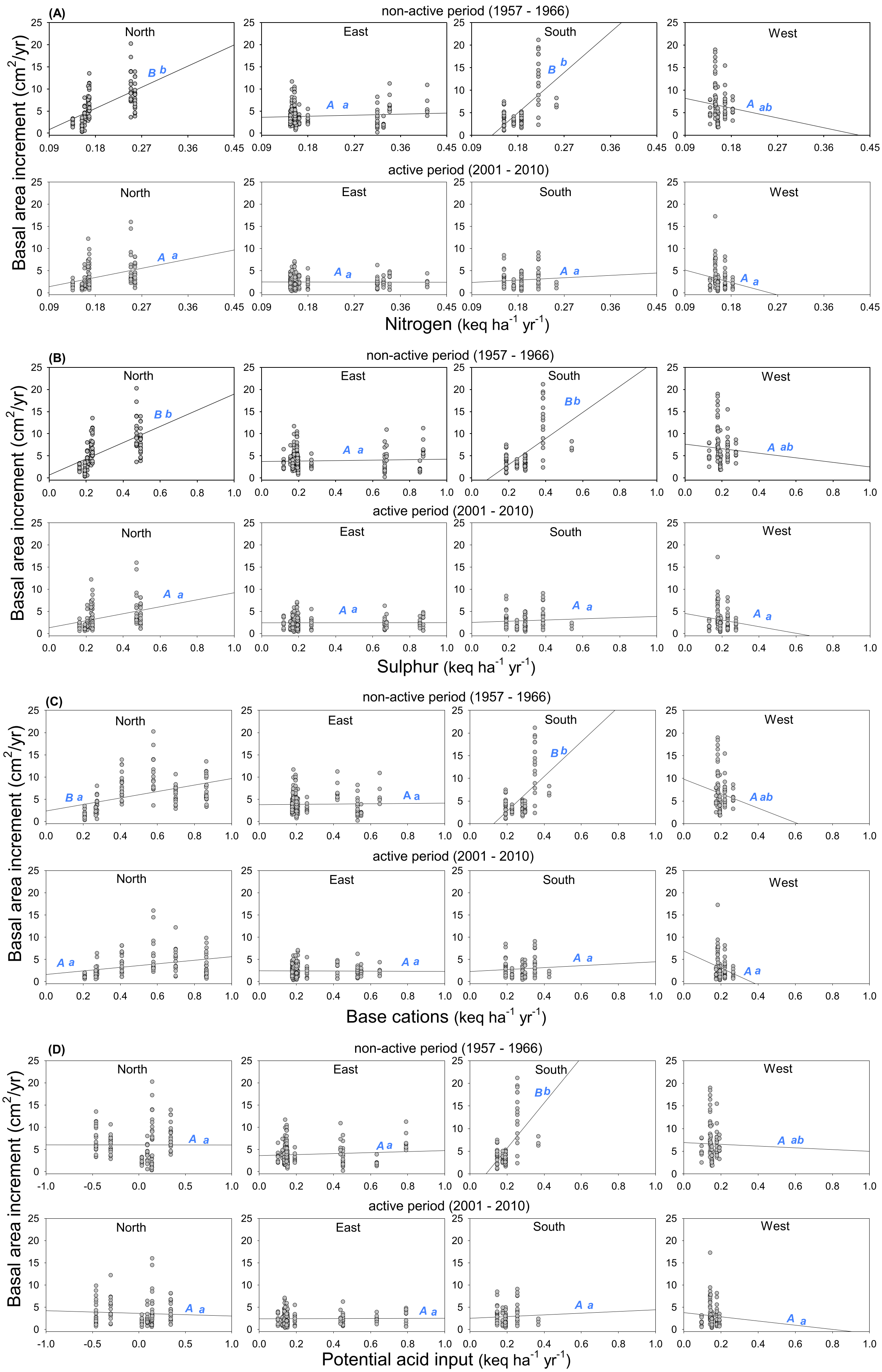
3.4. Correlations between soil variables, BAI, and understory vegetation

Basal area growth in the non-active period was not correlated with any of the (current) soil alkalinity or acidity variables, but growth in the active period was negatively correlated with the CEC of the organic soil horizon (Table 3). The difference in basal area increment between the active and non-active period was positively correlated with the BC:Al ratio and with pH of the surface (top 5 cm) mineral soil (Table 3), indicating that the trees on sites with higher BC:Al and pH showed less growth decline over time. Understory cover, particularly total cover, was negatively correlated with the BC:Al ratio of the top mineral soil, whereas vascular plant cover was positively correlated with the CEC of the LFH horizon. Understory species richness was not correlated with any of the organic or mineral soil variables, but diversity was positively correlated with BC in the LFH horizon (Table 3).

4. Discussion

4.1. Jack pine growth in relation to atmospheric deposition

Our results overall demonstrate a weak but predominantly positive relationship between atmospheric N, S, and BC deposition and basal area increment of jack pine trees, within the recent decadal time frame of active oil sands development in the AOSR. This was the case for sites north and south of the center of surface oil sands mining operations, but not for those to the east or west (which showed no significant relationship). In concordance with our results, data from this same monitoring program, collected on only 10 plots in 1998, showed that tree size (height, diameter, crown length – for stands with trees of similar age) was negatively related to distance from emission source (AMEC Earth and Environmental Limited, 2000). These results might be taken to suggest a positive (fertilization) effect of deposition on tree growth and this aligns with the fact that N deposition was positively related to foliar nitrogen (MacKenzie and Dietrich, 2019). However, this does not support a strong inference of fertilization effect as it is complicated by the fact that growth in the pre-mining (non-active) period was also positively correlated with estimates of current (active period) deposition. This was surprising, given that there could be no gradient of deposition related to industrial development in the AOSR in the non-active period; the region had no industrial activity and, in fact, was completely undeveloped at that time. However, data from earlier in



(A)
non-active period (1957 - 1966)
active period (2001 - 2010)
North
East
South
West
Basal area increment (cm²/yr)
Nitrogen (keq ha⁻¹ yr⁻¹)
(B)
Sulphur (keq ha⁻¹ yr⁻¹)
(C)
Base cations (keq ha⁻¹ yr⁻¹)
(D)
Potential acid input (keq ha⁻¹ yr⁻¹)

this monitoring program also support this trend; when they applied a standardization intended to correct for differences in growth that predated oil sands development, they found no difference in growth between high and low deposition areas (AMEC Earth and Environmental Limited, 2000). The results, therefore, show a pattern of spatial variation in growth within the region whereby in the period before surface oil sands mining began, trees on sites there were closer to what became the center of the mining activity, and that thus eventually experienced higher levels of atmospheric deposition, were growing faster than on sites farther away.

There was an overall decline in growth of trees over time and this can be attributed to natural stand dynamics (i.e., natural decline in growth as trees and stands age), long-term climatic changes, or direct and indirect effects of acid deposition. We found that the slopes of the growth–deposition relationships were, surprisingly, significant in the non-active but not in the active period. This suggests that trees on sites closer to what became the center of mining activity (and that thus later experienced higher levels of deposition) experienced more of a growth decline over time than those at other sites. There are a few possible explanations for this result. First, it might reflect the fact that these trees, which happened to be growing faster in the non-active period, more quickly reached a size at which growth decline naturally sets in or at which site conditions became more limiting. Second, the decline in slope of the growth-deposition relationship over time could be due to the fact that pre-existing spatial variation in site quality was reduced by regional deposition, which had a fertilization effect that benefitted the poorer sites, which were, coincidentally, more distant from the center of oil sands mining activity. This phenomenon is an example of what could be referred to as the (N) homogenization effect (sensu Gilliam, 2006). Finally, the greater decline in growth over time for sites receiving higher deposition might suggest some negative effects of atmospheric deposition on growth; however, our results do not support this hypothesis. When we examined the relationship of growth depression over time with deposition variables, factoring out the pre-existing differences in growth by including growth in the non-active period as a co-variate in analysis, there was no relationship between the change in growth over time and deposition.

Given the N-limitation of forests in the AOSR (Jung and Chang, 2012), we expected enhanced N availability from atmospheric deposition to stimulate tree growth, as has been found in studies elsewhere (Binkley and Hogberg, 1997; Binkley and Hogberg, 2016; Solberg et al., 2004), and as aligns with the general expectations of the effects of N fertilization in forestry practice (Bergh et al., 2014; Pettersson and Högbom, 2004). With foliar N concentrations ranging from 1.15 to 1.3% and foliar S from 65 to 120 ppm (MacKenzie and Dietrich, 2019) our sites appear to be at least moderately N and S deficient (Weetman et al., 1985) and thus might benefit from a fertilization effect of deposition. However, our results do not provide strong evidence of such an effect, neither do they suggest a negative, acidification, effect on growth.

We found no relationship of BAI in the active period with soil acidity, but there was a significant, weak, negative, relationship with CEC of the organic layer. Interestingly, there was a significant, positive correlation between the change in BAI over time and mineral soil pH and BC:Al. This suggests that alkalinity was associated with less growth decline over time.

4.2. Understory vegetation response to atmospheric deposition

Atmospheric N and S deposition and PAI were positively associated with understory plant cover, richness, and diversity, particularly

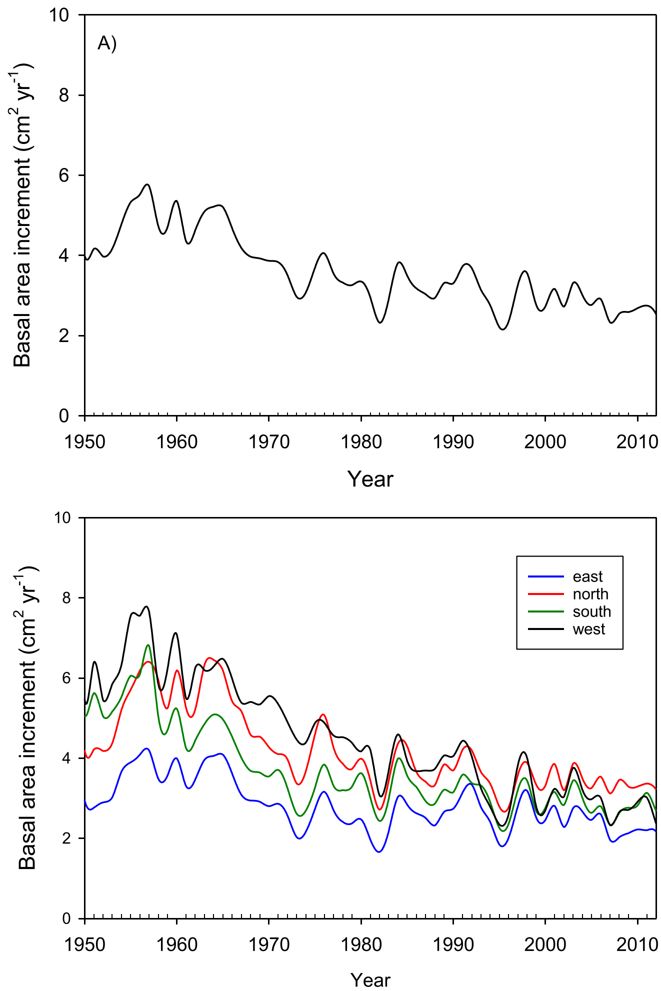


Fig. 2. Trend in basal increment (BAI) of jack pine trees across the entire study sites (A) and by direction (B).

vascular plants and shrubs and might suggest a possible fertilization effect of deposition on the understory vascular plant community. These results are generally consistent with previous observational and manipulative studies that also showed increased abundance of herb layer species in response to N loading (Bobbink et al., 2010; Bobbink et al., 1998), although such increases are often confined to nitrophilous species (Brunet et al., 1998; Gilliam et al., 2016; Nordin et al., 2005; Thimonier et al., 1992). The increase in species richness we observed, contradicts with studies that found no effect or reduced vascular plant richness with N addition (Bobbink, 2004; Gilliam et al., 2016; Gilliam, 2019; Hallbacken and Zhang, 1998; Walter et al., 2017). Sulphur, also being an essential element for plant growth, may stimulate growth and physiological responses especially in S deficient soils where SO_2 might be metabolized and taken up as nutrients. As mentioned above, the tree foliar nutrient data suggested moderate S deficiency on our sites.

Understory species diversity (the inverse of Simpson's index is sensitive to abundant species and therefore gives an indication of dominance in the community) increased with N deposition in sites located in the forest interior; however, there was no such relationship observed

Fig. 1. Basal area increments (BAI) of jack pine (*Pinus banksiana* Lamb.) prior to (1957 to 1966; i.e., the non-active period) and during an active period (2001 to 2010) of industrial oil sands development in the Athabasca Oil Sands Region (AOSR) as related to current atmospheric deposition estimates of nitrogen, sulphur, base cations, and potential acid input. Lower case letters compare significant differences ($\alpha = 0.05$) between the slopes of N, E, S, and W directions within each time period while upper case letters compare between the slopes of the non-active and active period for each direction. See Table A3 in Appendix for comparison of the slope of the relationships. See Edgerton et al. (2019) for details on the deposition estimates.

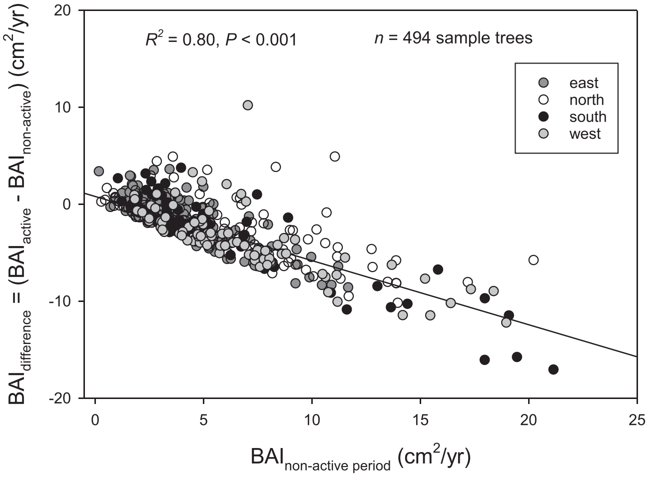


Fig. 3. Relationship of the difference in basal area growth between the two time periods (Δ BAI = BAI $_{active}$ − BAI $_{non\text{-}active}$) with growth in the non-active period.

for sites at the forest edge. This suggests increased dominance or greater unevenness in abundance among constituent species in response to N deposition in the forest interior. Thus N deposition impacts understory assemblages by promoting dominance of some species, mostly vascular plants, over others. This could result in shifts in the competitive balance of species in response to N (Langan, 2014). Others have also found evidence of initial increases in cover and decreases in species evenness as a result of increasing dominance of particularly N-demanding species (Gilliam, 2006; Walter et al., 2016).

We found no significant relationships of understory species composition to N, S, BC or PAI. This is contrary to the well-established sensitivity of especially mosses and lichens to atmospheric deposition (Mäkipää, 1995; Olsson and Kellner, 2006). Other studies also suggest that plant communities may show a tendency to shift toward compositions typical of high N availability (Bobbink et al., 1998) and that even small but chronic increases in deposition can cause noticeable changes in understory plant community composition (Nordin et al., 2005). We found no evidence of this in our case, and it is possible that current atmospheric deposition levels in the AOSR may be too low for these species to elicit a change in the species composition.

4.3. Limited evidence of soil-mediated effects of acidification

Earlier results from this monitoring program reported a positive relationship between tree height (in stands of similar age) and ammonium-nitrate concentration, and between tree diameter and phosphorous concentration in the organic (LF) soil layer (AMEC Earth and Environmental Limited, 2000). However, MacKenzie and Dietrich (2019) found no relationship between N deposition and LFH nitrogen and our results indicated limited evidence of soil-mediated effects of acidification on jack pine basal area growth and understory response to deposition. For instance, soil acidity indicators such as pH and BC:Al of the top mineral soil were positively but weakly related to growth depression (i.e., the difference in basal area growth between BAI of active versus the non-active period) of jack pine trees. Similarly, understory cover was negatively correlated with BC:Al. Due to minimal impact of deposition on soil base saturation, earlier predictive studies in the AOSR concluded that there is limited potential for acidification of soils under current deposition levels over the next century (Whitfield et al., 2009) or that soils may have recovered from acidification over time (Jung et al., 2013). Although soils in jack pine stands are naturally acidic with low base cation saturation, it is possible that the risk of soil acidification from deposition is mitigated by the deposition of base cations in industrially-generated fugitive dust (Fenn et al., 2015; Watmough et al., 2014).

5. Conclusions

We found no clear evidence of a negative effect of atmospheric deposition of N, S, or base cations on jack pine tree growth or understory vegetation in the Alberta Oil Sands Region. The positive relationship of atmospheric N and S deposition with forest understory vegetation cover and richness, might suggest a fertilization effect, although we did not observe the expected change in species composition to favour nitrophilous species. Additionally, there was limited evidence of soil-

Table 2
Relationship of modelled atmospheric deposition with forest understory vegetation cover, richness, diversity, and composition, and the interaction with site type (forest edge vs interior) and direction (N, E, S, W) from the center of oil sands mining activity. Significant p-values (at α = 0.05) are highlighted in bold.

Understory response	Deposition variable	Model summary	Model terms						
			$D_{eposition}$ DF = (1)	$S_{ite\ type}$ (1)	$D_{irection}$ (3)	$D_{ep.} \times S_{ite}$ (1)	$D_{ep.} \times D_{irection}$ (3)	$S_{ite} \times D_{irection}$ (3)	$D_{ep.} \times S_{ite} \times D_{irection}$ (2)
Vascular cover	Nitrogen								
		F	5.630	0.067	2.422	2.865	3.036	0.243	0.098
		P	**0.025**	0.798	0.087	0.102	**0.046**	0.865	0.907
		$R^2_{partial}$	0.22	0.01	0.18	0.14	0.22	0.03	0.01
	Sulphur	F	6.761	0.030	2.640	2.997	2.620	0.332	0.698
		P	**0.015**	0.865	0.069	0.094	0.070	0.802	0.506
		$R^2_{partial}$	0.25	0.01	0.19	0.13	0.20	0.03	0.05
Shrub cover	Sulphur	F	3.898	13.607	0.881	1.485	0.803	0.984	0.854
		P	0.058	**0.001**	0.463	0.233	0.503	0.415	0.437
		$R^2_{partial}$	0.08	0.30	0.07	0.03	0.05	0.09	0.06
	PAI	F	4.018	13.36	0.403	1.116	0.552	1.657	1.030
		P	0.055	**0.001**	0.752	0.300	0.651	0.199	0.370
		$R^2_{partial}$	0.05	0.27	0.05	0.02	0.03	0.15	0.07
Vascular richness	Nitrogen	F	4.978	0.912	7.013	0.078	3.022	2.651	1.058
		P	**0.034**	0.348	**0.001**	0.782	**0.046**	0.068	0.361
		$R^2_{partial}$	0.25	0.04	0.42	0.07	0.25	0.22	0.07
Total diversity	Nitrogen	F	5.009	2.212	1.150	4.314	1.431	0.266	1.156
		P	**0.033**	0.148	0.346	**0.047**	0.255	0.850	0.329
		$R^2_{partial}$	0.15	0.03	0.09	0.11	0.14	0.03	0.08

Notes: results are shown for only understory response variables that were significantly related to estimated atmospheric deposition based on initial regression analyses (see Table A5 in Appendix for these results and for other vegetation variables that were not significantly related to deposition).

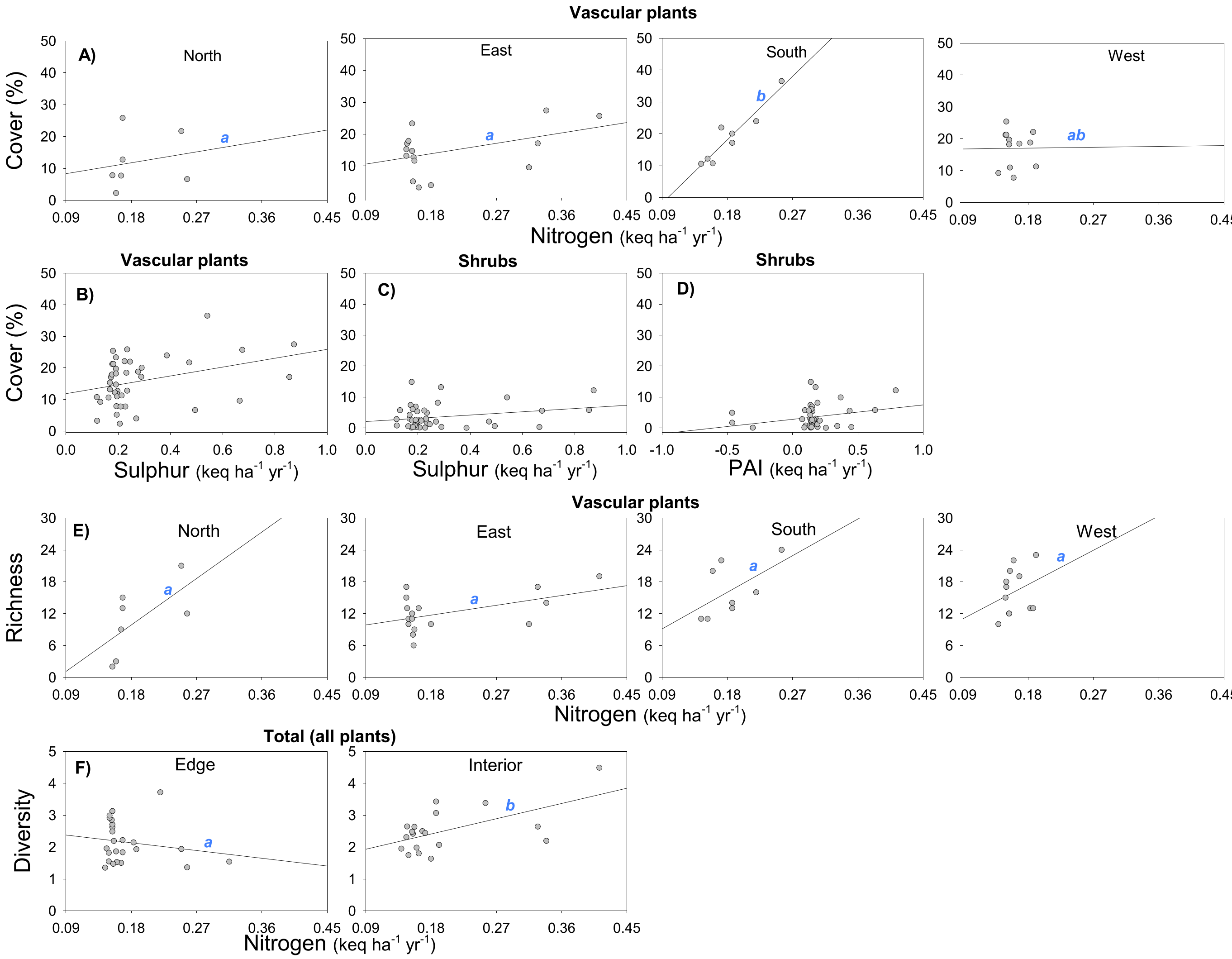


Fig. 4. Scatterplots of fitted regressions for the relationship of understory vegetation cover (a–d), richness (e), and diversity (f) to modelled atmospheric deposition of nitrogen, sulphur, and potential acid input (PAI) in the Athabasca Oil Sands Region. Letters compare significant differences ($\alpha = 0.05$) between the slopes of N, E, S, and W directions (A and E) and forest edge and interior sites (F). See Table A6 in Appendix for comparison of the slope of the relationships.

mediated effects of acidification on jack pine basal area growth and understory vegetation. We cannot exclude the possibility that the results reflect some underlying geographical patterns of intermediate to long-term changes in regional climate or the increasing trend in the number and size of wildfires in the region, as these were not considered in the present study. Ongoing oil sand operations continue to contribute to

Table 3
Spearman's rank correlation for organic (LFH) and top mineral (0–5 cm depth) soil variables with jack pine (*Pinus banksiana* Lamb.) growth (basal area increment: BAI) and understory vegetation response variables. Significant correlations ($p < 0.05$) are highlighted in bold.

Variable	Organic (LFH) soil				Mineral (0–5 cm) soil			
	BC	CEC	BC: Al	pH	BC	CEC	BC: Al	pH
a) Basal area increment (BAI)								
BAI $_{\text{non active period}}$	−0.07	−0.12	0.04	−0.01	0.08	0.11	−0.13	−0.07
BAI $_{\text{active period}}$	−0.15	**−0.18**	−0.14	−0.11	0.13	0.07	0.17	0.16
BAI $_{\text{difference}}$	−0.09	−0.03	−0.15	−0.09	0.09	0.03	**0.26**	**0.20**
b) Understory vegetation								
Cover (all plants)	−0.06	0.17	−0.04	0.04	−0.27	−0.15	**−0.52**	−0.31
Vascular cover	0.41	**0.58**	0.39	0.41	−0.14	−0.20	−0.30	−0.30
Shrub cover	−0.04	0.07	0.04	0.01	−0.13	−0.19	−0.27	−0.29
Richness (all plants)	0.23	0.19	0.23	0.37	−0.22	−0.19	−0.27	−0.26
Vascular richness	0.21	0.14	0.21	0.35	−0.06	−0.02	−0.16	−0.11
Diversity (all plants)	**0.49**	0.41	0.47	0.34	0.15	0.03	0.16	−0.23

Notes: the analyses were based on data from only the forest interior research monitoring sites due to limited soil chemical data for the edge forest sites at the time of analysis. BC: base cations; CEC: cation exchange capacity, BC: Al ratio of base cations to aluminium. See also MacKenzie and Dietrich (2019; this issue). BAI difference = BAI in the active period – BAI in the non-active period.

acidic and nitrogen deposition, and with an expected increase in oil sands production in the future, the role and effects of atmospheric deposition, acidification, and fertilization from emission sources merit continuous monitoring in WBEA's Forest Health Program.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.133877.

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