Four years of simulated N and S depositions did not cause N saturation in a mixedwood boreal forest ecosystem in the oil sands region in northern Alberta, Canada

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Abstract: We conducted a simulated nitrogen (N) and sulfur (S) deposition experiment in the Athabasca oil sands region (AOSR) where NO_x and SO_2 have been emitted from oil sands mining/extracting and upgrading activities and then deposited to the surrounding ecosystems for decades. To evaluate changes in tree growth rates, N pool sizes, and nutrient losses by S and N deposition, the following four treatments were applied: control (CK), N addition (+N, 30 kg N ha^{-1} yr^{-1}), S addition (+S, 30 kg S ha^{-1} yr^{-1}), and +NS additions (+NS, 30 kg N and 30 kg S ha^{-1}), from 2006 through 2009. Nitrogen addition increased ($p<0.05$) tree growth in the +N and +NS treatments, indicating N-limitation in the studied forest, while none of the treatments affected understory growth or soil microbial biomass. The treatments affected inorganic N concentrations in the soil only immediately following N addition. Minimal amounts of NO_3^- were leached below 45 cm (considered to be below the main rooting zone) of the soil profile in any of the treatments. Decreases in exchangeable Ca^{2+} and Mg^{2+} by N and S additions were likely due to increased tree uptake following increased tree growth in the former and increased leaching with sulfate in the latter. Although the lack of significant N leaching indicates that the risk of N saturation was low after four years of elevated N deposition, reduction of exchangeable base cations implies that nutrient imbalance remains a concern in AOSR in the long term.

Key words: nitrogen cycling, acid deposition, N saturation, base cation leaching, boreal forest, Athabasca oil sands region.

1. Introduction

Long-term atmospheric nitrogen (N) and sulfur (S) depositions have been shown to cause changes in elemental biogeochemical cycles, soil acidification, and nutrient deficiency or imbalance in forest ecosystems (Aber et al., 1989; Johnson and Siccama, 1983). Compared with historical emission rates, the total emission of acid-causing materials in North America is now substantially lower due to efforts such as the Eastern Acid Rain Program in Canada and the US Clean Air Act (Morrison, 2006). However, while SO_2 emissions have decreased globally, NO_x emissions in industrialized regions remain elevated (Galloway et al., 2004). The Athabasca oil sands region (AOSR) is the world's second largest reservoir of recoverable oil (>130 billion barrels) in the form of bitumen and is the largest area for open-pit oil sands mining in Alberta, Canada (Humphries, 2008). The SO_2 emission peaked at approximately 400 Mg day^{-1} in the 1980s followed by a substantial reduction in the mid-1990s and remained between 250 and 300 Mg day^{-1} in the 2000s (Hazewinkel et al., 2008). On the other hand, NO_x emission has increased since the beginning of commercial oil production and is expected to increase due to expansion of oil sands mining/extraction and upgrading activities and increase in the population (Aherne and Shaw, 2010; Hazewinkel et al., 2008). Therefore, impacts of S and N deposition on boreal forests in AOSR are of great concern and, furthermore, influence of N deposition will increase in the future.

The N status of forest ecosystems can be divided into different stages in relation to responses of ecosystems to chronic N deposition: N limitation, alleviation of N limitation, and N saturation (Aber et al., 1998). In boreal forests, the initial stage has been normally known to be N-limited followed by a stage of alleviation of N limitation as N availability increases through chronic N deposition (Hari and Kulmala, 2008). In the second stage, external N input may be taken up by plants and thus provide a beneficial effect on tree growth. Furthermore, the N may move into the soil as organic N through litterfall and fine root turnover (Aber et al., 1998; Nadelhoffer et al., 1999). Net N mineralization rates may increase in response to increased soil N content, which may increase inorganic N concentrations in the soil and, subsequently, N leaching and N emissions as the ecosystem becomes N saturated (Aber et al., 1998; Matson et al., 2002). Chronic N deposition affects the cycling of both N and other nutrients (Aber et al., 2003; Likens and Bormann, 1995). While the N deposited plays the role of a fertilizer in N-limited forests, increased uptake in combination with leaching of cationic nutrients with nitrate may cause cationic nutrient deficiencies or nutrient imbalances for trees, which may ultimately cause forest decline (Erisman and De Vries, 2000; Skeffington and Wilson, 1988).

Even though chronic N and S depositions may induce detrimental effects on forest ecosystems, the response time of ecosystems to such effects may vary, depending on a number of factors such as deposition rates and nutrient demand by the biota, especially for the response to N deposition (Gundersen et al., 1998). In North America, N saturation has been often found in forests in eastern U.S. that have been impacted by high N deposition rates (Jeffries, 1995). Although several monitoring sites in eastern Canada, e.g., the Turkey Lake watersheds, have been shown to be N-saturated (Foster et al., 1989), most forests in Canada did not show any evidence of N saturation due to relatively low rates of N deposition (Houle et al., 1999). Simulated studies with elevated deposition rates have been conducted in forests less impacted by acid deposition to evaluate potential changes in ecosystem properties and processes. Simulated studies in N-limited forests could show short-term changes in the N cycle and conditions of other nutrients such as Ca and Mg uptake (Aber et al., 1998; Moore and Houle, 2009).

Sulfur deposition in AOSR has been a major concern for soil acidification due to greater emission rates of S than N (even after the curtailment of S emission rates as discussed earlier), a low capacity for sulfate adsorption of soils (Jung et al., 2011), and N limitation of the ecosystems in the region (NO_x-SO_2 Management Working Group, 2004). Meanwhile, the concern regarding N deposition has increased due to the expected increase in N emission rates in AOSR and in western Canada (Aherne and Shaw, 2010) though N saturation, based on our literature review, has not been reported in western Canada and boreal forests in AOSR. Increasing atmospheric NO_2 concentrations and N deposition rates, however, have influenced N cycling in forest ecosystems in AOSR (Laxton et al., 2010), suggesting that there was potential progressing of the N cycle towards N saturation.

To evaluate the impact of elevated levels of N and S depositions on forest ecosystems in AOSR, we conducted a simulated N and S deposition experiment examining changes in tree and understory growth rates, in N cycling (N leaching and N storage in microbial biomass), and in the leaching loss of cationic nutrients from soils. We hypothesized that 1) boreal forests surrounding AOSR are N but not S limited and thus the growth rates of dominant tree species will be increased by elevated levels of N deposition, but not by elevated levels of S deposition. One of the questions that needs to be answered is how do boreal forests in AOSR respond to increased levels of N and S depositions, as such responses are site specific, 2) N leaching will not be increased in the short-term by the elevated levels of N and S depositions in the N-limited boreal forest, and 3) cationic nutrient leaching will not be increased in the short-term by the elevated levels of N deposition, consistent with the lack of N leaching.

2. Material and Methods

2.1 Site description and experimental design

The research plots were established in a boreal forest stand (56.1° N 110.9° W) located about 100 km southeast of Fort McMurray in AOSR in northern Alberta, Canada. Most of the mining sites and upgrading facilities in AOSR were distributed within about a 100 km distance north of Fort McMurray. In AOSR, the mean annual temperature is 0.7 °C with mean relative humidity of 68% and mean annual precipitation of 456 mm (Environment Canada, 2010). The main canopy tree species were approximately 60-year-old trembling aspen (*Populus tremuloides*, referred to as aspen below) followed by white spruce (*Picea glauca*) aged between 25 and 55years old, based on increment cores collected in 2011. Aspen and white spruce constituted 71 and 22% of the canopy trees by stem density, respectively, and most of the dominant trees (based on the tree's relative height) were aspens. White spruce trees were also found as seedlings growing in the understory as this species is very shade tolerant. Other canopy trees found in the plots were balsam fir (*Abies balsamea*), balsam poplar (*Populus balsamifera*), black spruce (*Picea mariana*), and paper birch (*Betula Papyrifera*). The dominant understory species were prickly rose (*Rosa acicularis*) and wild sarsaparilla (*Aralia nudicaulis*) based on coverage. The ecosite phase was d1 with medium nutrient regime and mesic moisture regime based on ecological site classification (Beckingham and Archibald, 1996). Soils were mostly Gray Luvisols classified based on the Canadian system of soil classification (Soil Classification Working Group, 1998) or Boralf in Soil Taxonomy (Soil Survey Staff, 1994).

The experiment used a 2×2 factorial design with blocking. One factor studied was N addition (with two levels) and the other was S addition also with two levels to simulate elevated levels of N and S deposition in the oil sands region. Thus, four treatments were set up: control (CK), N addition (+N, 30 kg N $ha^{-1}yr^{-1}$ as NH_4NO_3), S addition (+S, 30 kg S $ha^{-1}yr^{-1}$ as Na_2SO_4), and +NS additions (+NS), with addition of N and/or S started in 2006 and through 2009. For this experiment, four blocks were set up based on topographic position to ensure relatively uniform soil/site conditions within each block and four plots of 20 × 20 m were established in each block. Within each block, the treatments were randomly assigned to the plots. The proportion of canopy tree species was variable between blocks with between 56 and 91% aspen and between 5 and 29% white spruce. The blocking design allowed relatively similar composition of canopy trees between treatments within each block. Soil texture of surface mineral soil (0-15 cm) was sandy loam in two blocks and silt loam in the other two blocks. The deeper soil (15-45 cm) had greater clay content than surface mineral soil but texture was more variable between blocks.

Fertilizers were broadcast applied using a spreader in granule forms. Between 2006 and 2008, the N and S were applied once in early summer because more than two thirds of deposition falls down in AOSR in the growing season approximately between June to September (Wieder et al., 2010). Since 2009, the N and S were applied in three equal splits on July 2, July 22, and August 11. We changed the N and S addition method in 2009 because the split application can imitate deposition processes in the natural system that happen continuously with seasonal variation than one-shot application even though a method to simulate the exact way N and S were deposited in the natural system was not possible.

2.2 Plant sampling and analysis

Diameter at breast height (DBH, measured at 1.3 m above ground) of all trees in each plot was measured in June 2006 (with the exception noted below) and in June 2009. Trees with DBH less than 0.5 cm in 2006 were excluded from DBH measurement. Tree growth was assessed by evaluating the annual increment of aboveground biomass from 2006 to 2009, presented as kg ha^{-1} yr^{-1} rather than the annual increment of basal area (m^2 ha^{-1} yr^{-1}) to compare the effects of treatments on biomass between trees, understory and litterfall. Tree aboveground biomass was calculated with a DBH-based set of equations established for each of the tree species found in this study (Lambert et al., 2005). The aboveground portion of the understory vegetation was sampled from four quadrats of 30 × 30 cm in each plot in late July, 2009 to determine the aboveground biomass of the understory vegetation. Foliar samples were collected in late July, 2009 from healthy aspen trees that did not display any visual symptom of insect or disease damage. Three litterfall traps of 10 cm diameter and 10 cm depth were set up in each plot in late October, 2008, and litterfall samples were collected in late October, 2009.All plant samples were dried in an oven for 72 hours at 70 °C and weighed to determine dry biomass. The samples were ground to pass through a 0.42-mm sieve and homogenized. Nitrogen concentrations of plant samples were determined using a Carlo Erba NA 1500 elemental analyzer (Carlo Erba Instruments, Milano, Italy).

2.3 Soil sampling and analysis

Soil samples were collected from the forest floor and the surface mineral soil (0-15 cm) in each plot. The first sampling was on July 2, 2009 before the first addition of N and S in that growing season and samples were collected again on 10, 20, 40, 80 and 320 days after July 2, 2009. After each sampling, the soil samples were placed in a cooler, transported back to the laboratory, and stored in a refrigerator. Fresh soil samples were crushed to pass through a 2-mm sieve, with coarse fragments, roots, and debris removed. Soil moisture content was determined in a forced air oven at 105°C. Soil microbial biomass C (MBC) and N (MBN) were determined by the chloroform fumigation-extraction method (Voroney et al., 2008) within a week after sampling. A 0.5 mol L^{-1} K_2SO_4 solution was used to extract C and N from fumigated and non-fumigated samples at a 1:10 (w:v) ratio. Then a 2 mol L^{-1} KCl was used to extract (at the 1:10 w:v ratio) available ammonium and nitrate (Kalra and Maynard, 1991), with the ammonium and nitrate concentrations determined on a Dionex DX-600 ion chromatography (Dionex Corp., Sunnyvale, CA).

The following soil properties were analyzed using air-dried samples collected 320 days after the first addition of N and S in the 2009 growing season. Soil pH was measured using 10 g of air-dried soil in 40 mL of water for forest floor samples or 20 mL of water for surface mineral soil samples. Each soil sample was further ground with a ball mill and used for total C and N analysis on a Carlo Erba NA 1500 elemental analyzer (Carlo Erba Instruments, Milano, Italy). Exchangeable cations, including Ca^{2+}, Mg^{2+}, K^+, Na^+ and Al^{3+}, were determined after extraction with 1 mol L^{-1} NH_4Cl at a ratio of 5 g soil to 100 mL extractant. After filtration, the filtrates were analyzed using a Perkin-Elmer Elan 6000 quadrupole ICP-MS (Shelton, CT). The Ca/Al ratio was calculated with exchangeable Ca^{2+} and Al^{3+} based on a molar unit.

2.4 Soil solution sampling and analysis

Two stainless steel zero-tension lysimeters (manufactured in house) installed at each of the 15 cm (within the main rooting zone) and 45 cm soil depths (below the main rooting zone) in 2007 to collect soil solution samples. Soil solution samples were collected every month from May to October, 2009, in the growing season, and then in May, 2010, after spring thaw. The water collection area of each lysimeter was 500 cm^2. For samples collected in May, collecting bottles connected to lysimeters were put in place in October the year before. The bottles were washed with 20% HCl and rinsed with deionized water in the laboratory and 3-4 drops of 0.1 g L^{-1} phenyl mercury acetate were added to each container prior to deployment to the field to minimize microbial activities in the water samples between sampling intervals. All samples were measured for their volume and kept in a refrigerator. A portion of each sample was filtered with 0.22 μm syringe filters before chemical analysis. Concentrations of Ca^{2+}, Mg^{2+}, K^+ and Na^+ were measured using a Perkin-Elmer ICP-MS (Shelton, CT). Concentrations of SO_4^{2-}, NO_3^- and NH_4^+ ions were analyzed with a Dionex Corp DX 600 ion chromatography (Sunnyvale, CA). Leaching loss below each depth of each ion was determined by using collected water volume and concentration of each ion.

2.5 Statistical analysis

All statistical analyses were performed using version 9.02 of the SAS software (SAS Institute Inc, Cary, NC). The datasets were checked for normality of distribution and they were all normally distributed. Analysis of variances (ANOVA) was performed to determine the effects of N and S addition and sampling time on the measured parameters. To analyze the effects of N and S additions on MBN, MBC and inorganic N, the compound symmetry covariance structure was selected to consider the effects of time-repeated measurement. All pair-wise comparisons among treatments were done with the Bonferroni correction. Tree biomass in 2006 was used as a covariate to compare tree biomass increment and litterfall amount between treatments. An α value of 0.1 was chosen to indicate statistical significance due to the high spatial variability of the measured parameters.

3. Results

3.1 Effects of N and S additions on plant growth and foliar N

Nitrogen addition increased tree biomass ($p<0.05$) and litterfall amount ($p<0.05$) (Fig. 1a and 1b) but aboveground understory biomass was not affected by any of N and S additions (Fig. 1c). The annual increase of tree biomass was the greatest in the +NS treatment which was greater than that in the +N treatment ($p<0.1$), indicating a synergistic interaction between N and S additions ($p<0.1$). However, no synergistic response between N and S additions was found for litterfall. None of the treatments affected N concentrations in foliar and litterfall of trees (Fig. 1a and b). Nitrogen addition increased ($p<0.01$) whereas S addition decreased ($p<0.1$) N concentrations in the aboveground component of the understory vegetation (Fig. 1c). No difference was found between the CK and +NS treatments (Fig. 1c).

3.2 Effects of N and S additions on soil properties

In the forest floor, N addition increased exchangeable Ca^{2+} and K^+ ($p<0.1$ and $p<0.05$, respectively) while S addition increased exchangeable Na^+ ($p<0.05$) (Table 1). In the surface mineral soil, both N and S additions decreased exchangeable Ca^{2+} ($p<0.1$), and exchangeable

Ca^{2+} and Mg^{2+} was the lowest in the +NS and the greatest in the CK treatments. Exchangeable K^+ in the surface mineral soil was not affected by any of the treatments and exchangeable Na^+ increased in the +S and +NS treatments ($p<0.01$) compared to that in the CK. Both N and S additions decreased Ca/Al ($p<0.1$) with greater Ca/Al in the CK than in any other treatments in the surface mineral soil ($p<0.1$), but was not statistically different in the forest floor.

3.3 Effects of N and S additions on MBC, MBN, and inorganic N

The treatments did not affect MBC and MBN in the forest floor or the surface mineral soil (Fig. 2). The MBC and MBN in the surface mineral soil were about one tenth of that in the forest floor. After the summer season, MBC and MBN decreased in the forest floor but increased in the surface mineral soil ($p<0.01$). The NH_4-N concentration in the forest floor was also one tenth of that in the mineral soil (Fig. 2). In the forest floor, N addition increased NH_4-N concentration from the first addition ($p<0.05$), but the treatment effect disappeared 80 days after the first application (= 40 days after the third application). By the following spring, NH_4-N concentrations in the forest floor were lower ($p<0.05$) in the +S and +NS than in the CK and +N treatments, suggesting that there might be a negative effect of S addition on N mineralization in early spring. The NO_3-N concentrations in the surface mineral soil, on average, were 3.1 times those in the forest floor (Fig. 2). The NO_3-N concentrations in the forest floor were not affected by the treatments and were relatively stable across the sampling dates. The NO_3-N concentrations in the surface mineral soil were much higher in the +N and +NS treatments 10 and 20 days after the first addition ($p<0.05$) than those in the CK while no difference was found between treatments in the other sampling periods.

3.4 Effects of N and S additions on leaching loss of cations and anions

Soil solution at 15 cm below the mineral soil surface was collected in May, July, and August while minimal amounts of soil solution were collected at 45 cm below the mineral soil surface except in May, when large quantities of water from melting snow presumably leached through the soil profile. The leaching of NO_3^- below 15 cm was increased by N addition ($p<0.05$) while that of NH_4^+ was not affected by any of the treatments (Table 2 and Fig. 3). However, annual leaching loss of inorganic N below 15 cm depth was low in all treatments, with losses of NH_4^+<1.0 mmol$_c$ m^{-2} yr^{-1} and those of NO_3^- ranging between 0.3 and 1.5 mmol$_c$ m^{-2} yr^{-1} (Table 2). For both NH_4^+ and NO_3^-, leaching loss below 45 cm was negligible (Table 2). Annual leaching loss of SO_4^{2-} was increased by S addition at both 15 cm and 45 cm ($p<0.001$ for both) (Table 3). Most SO_4^{2-} leaching took place in May (Fig. 3). Leaching losses of Ca^{2+}, Mg^{2+} and Na^+ had a similar trend to that of SO_4^{2-} (Fig. 3). However, leaching loss of K^+ in May (data not shown) was similar to that in August. Sodium ion was dominant in soil solutions in the two S addition (+S and +NS) treatments, but minor in non-S added treatments (Table 2).

4. Discussion

4.1 Plant growth responses to simulated N and S depositions

Nitrogen addition increased tree growth in this study, indicating that N was a limiting factor in the studied forest ecosystem (Fig. 1). This result supports our first hypothesis and is also consistent with the N-limited ecosystems hypothesis of Aber et al. (1998) and results of short-term fertilization studies in Canadian boreal forests (e.g. Newton and Amponsah, 2006). This implies that tree growth in the studied forest ecosystem would be improved by elevated atmospheric N deposition in near future. Nitrogen addition, however, induced reduction of exchangeable Ca^{2+} and Mg^{2+} likely due to increasing uptake of nutrients by increasing tree growth (Table 1). Moreover, combination with N and S addition intensified decrease of exchangeable Ca^{2+} and Mg^{2+} (Table 1). The long-term studies have reported that repeated N fertilization or chronic N deposition with excessive amount of N may induce nutrient imbalance in the soil and plant (Aber et al., 1989; Skeffington and Wilson, 1988) as increased tree growth by N deposition may demand additional amounts of other nutrients. Furthermore, decreasing exchangeable cation concentrations associated with soil acidification caused by N and S deposition can exacerbate nutrient imbalance such as deficiency of Mg and K for tree growth (Erisman and De Vries, 2000). Based on our results, the studied ecosystem did not show any signs of N saturation after four years of N addition, which was consistent with previous research in other study sites with similar application rates (Moore and Houle, 2009; Sogn and Abrahamsen, 1998). Meanwhile, significant reduction of exchangeable cation concentrations in the mineral soil caused by N and/or S additions (Table 1) may imply potential long-term risk of nutrient imbalance in the studied boreal forest.

In the study, tree biomass increment in NS+ was greater than that in N+, implying that S could be a limiting factor when N limitation was alleviated. Another possibility is that base cations may be a second limiting factor in the studied site and released base cations such as Ca^{2+} and Mg^{2+} from exchange sites by Na^{+} from the added Na_2SO_4. Repeated N fertilization studies have reported potential limitation other nutrients as increasing tree growth demands more nutrients (Ampohsah et al., 2005; Kishchuk et al., 2002). However, we should be careful to make a conclusion that N and S additions had a synergistic effect on tree growth in the studied forest because the litterfall amount data did not support the synergistic interaction between N and S additions. Other possible supposition is a potential bias at assessing tree biomass using allometric equation, combined with variation between treatments though the block designed experiment and statistical analysis including a covariate of initial tree biomass were applied to reduce effects of variation between plots. For example, stand density was 0.22±0.04 m^{-2} in +N but 0.26±0.04m^{-2} in +NS; 0.21±0.03 in CK and 0.22±0.04 in +S. Relatively great stand density in +NS might exaggerate the assessed tree biomass.

Improved overstory tree growth following elevated levels of N deposition may decrease the growth of the understory vegetation (Thomas et al., 1999). For example, increased canopy cover may lead to reduced light condition on the ground causing decreases in understory growth (Lieffers et al., 1999). On the other hand, N addition may not increase tree growth when competitive understory vegetation competes for the limited N supply in the soil (Matsushima and Chang, 2006). In this study, N addition did not affect understory growth even though it increased N concentrations in the understory vegetation (Fig. 1c), implying that the growth of the understory vegetation was likely limited by other factors, such as light and water availabilities. However, chronic N deposition can affect understory community structure. Increased N availability may facilitate growth of nitrophilic species and affect the competitive interaction between plants (Gilliam, 2006). In addition, increased N availability may affect plant community structure indirectly by influencing biotic conditions such as infestation of insects and pathogens (Nordin et al., 2009). In the studied forest ecosystem, N addition increased the percent cover of nitrophilic understory species such as wild sarsaparilla and Canada mayflower (*Maianthemum* *canadense*) while S addition increased the percent cover of bunchberry dogwood (*Comus Canadensis*) but decreased that of prickly rose after 3-year simulated N and S addition (Neufeld, unpublished data); though the total understory biomass was not affected by N addition and/or S addition (Fig. 1c).Therefore, further research is needed to better understand the effects of N and S depositions on plant communities as well as nutrient cycling in AOSR.

4.2 Soil N and nitrate leaching responses to simulated N and S depositions

Time required for significant responses to N deposition to occur varies with the specific N pool of concern. Soil inorganic N concentrations typically respond quickly to N deposition compared with other N pools such as soil organic N (Gundersen et al., 1998). Increased soil inorganic N concentration reflects the alleviation of N limitation and may subsequently be followed (likely after many years of continued N deposition) by N leaching, a sign considered to indicate N saturation (Aber et al., 1998). In this study, N addition induced immediate increase of soil NH_4^+ and NO_3^- concentrations but that were not sustained (Fig. 2), possibly related to several mechanisms. First, the added N may be transferred from inorganic to other N forms/pools in the ecosystem, such as being taken up by plants to support increased tree growth (see section 3.2). Second, inorganic N may become immobilized by soil microbes that can increase with increased N deposition (Tietema, 1998; Goulding et al., 1998). Soil microbial biomass in this study was not affected by N and S treatments but by sampling time (Fig. 2), likely due to seasonal changes of the abiotic factors such as soil temperature and water availability (Tate, 1995; Yang et al., 2010). Another potential is N leaching loss to the surrounding environment. However, our data indicate that leaching of NO_3^- below 45 cm of the soil profile was negligible in all treatments, due to the low annual precipitation, relatively low permeability of the illuviated B horizon in Luvisolic soils and the tight N cycling even after artificial addition of 30 kg N ha^{-1} yr^{-1} for 4 years, implying that the studied ecosystem had not exceeded its N retention capacity.

Our result therefore supports the second hypothesis. It is consistent with results in the accelerated N addition study in eastern Canada reporting no N saturation with deposition rates ranging from 9 to 85 kg N $ha^{-1}yr^{-1}$ (Houle, 2006; Moore and Houle, 2009). On a long-term basis, however, 10 kg N ha^{-1} of deposition has been regarded as a threshold likely to lead to significant N leaching based on data from 126 forest sites in Europe and North America (Dise and Wright, 1995; Stoddard et al., 2001). In Canada, the highest deposition rates were reported for south-central Ontario and south-western Quebec with about 13 kg N ha^{-1} yr^{-1} in the 1990s and currently normally less than 10 kg N ha^{-1} yr^{-1} based on the Canadian Air and Precipitation Monitoring Network (Vet et al., 2004). Those were moderate levels of deposition as compared to the much higher deposition rates in some areas in Europe and the United States (Jeffries, 1995). So far only a few watersheds such as the Turkey Lake watershed have been reported to experience N saturation (Foster et al., 1989; Houle, 2006). Current N deposition rates in AOSR have been reported to be low at about 2 kg N ha^{-1} yr^{-1} (Jung et al., 2011b; Wieder et al., 2010). Based on the reported N deposition rates in AOSR and our results from this short-term simulated N and S deposition study, the risk of N saturation appears to be low in the studied forest ecosystem.

Care should be taken when applying the result from this study to the whole boreal forest ecosystems in AOSR. Considering that N concentration in the atmosphere changes with distance from emission sources, N deposition would range widely depending on the location in relation to the emission sources; for example, in 2009, annual averages of N_2O concentration in the air around industry sites ranged from 4.8 to 29.7 ppb and were much higher than the 2.5 ppb near

Anzac, a site located about 35 km southeast of Fort McMurray (WBEA, 2010). Furthermore, NO_x emissions have increased and are expected to further increase in the region. For example, emission rates of NO_x in the early 2000s were more than twice that of the rates in the early 1990s (Hazewinkel et al., 2008). Therefore, the effects of N deposition on ecosystems in AOSR remain a major concern.

4.3 Soil exchangeable cations and cation leaching

In this study, simulated N and S depositions decreased exchangeable Ca^{2+} and/or Mg^{2+} in the surface mineral soil whereas N addition increased exchangeable Ca^{2+} in the forest floor (Table 1), likely due to calcium recycling from greater deposition of litterfall (Likens and Bormann, 1995). Two possible processes may have caused the lower exchangeable cation concentrations in the N and/or S added treatments: leaching and plant uptake. Base cation leaching and subsequent deficiency of cationic nutrients have been described as harmful effects of acid deposition (Aber et al., 1989; Binkley and Richter, 1987). Numerous factors affect base cation leaching such as weather conditions and soil physical and chemical properties (Likens and Bormann, 1995; De Schrijver et al., 2008). In AOSR, a quarter of the annual precipitation (456 mm) (Environment Canada, 2010) falls down to the soil as snow during the non-growing season and the majority of it may infiltrate into the soil or lost as surface runoff over a short snow-melting period with minimal transpiration. In contrast, water infiltration into the soil in the growing season is relatively low with the region having low annual precipitation and about half of the annual precipitation occurring between June and August (Environment Canada, 2010). In this study, leaching loss in May following snowmelt was responsible for most of the annual loss for SO_4^{2-}

and cations. Increased SO_4^{2-} leaching in early spring in the +S and +NS treatments accompanied the increased leaching of cations and the extent of SO_4^{2-} leaching loss was similar to the sum of cation leaching loss (Fig. 3).Considering that H^+ deposition is normally paired with SO_4^{2-} deposition under the natural condition and more strongly adsorbed to cation exchange sites than Na^+ (Bohn et al., 2001) that was used in this study, the risk of Ca^{2+}, Mg^{2+} and K^+ leaching with SO_4^{2-} was probably greater than what was determined in this study at the given S addition level.

Nitrogen addition effects on exchangeable $Ca^{2+}$and Mg^{2+}concentrationsin the surface mineral soil were significant. Depletion of exchangeable Ca^{2+} and Mg^{2+}in the surface mineral soil could be caused by increased storage (through plant uptake) of Ca and Mg in plant biomass as well as increased leaching out of a particular soil layer or decreased deposition of those cations (Likens et al., 1998). However, when the rooting zone was considered as a unit for evaluation, the third hypothesis is also supported by results from this study, that cationic nutrient leaching was not affected by the elevated level of simulated N deposition. Increased Ca and Mg uptake accompanying increased tree growth by N fertilization likely contributed to the reduction of exchangeable Ca^{2+} and Mg^{2+} in the mineral soil in the N addition treatments. Soil acidification in forest ecosystems involves a variety of internal processes to produce or consume H^+. Uptake of base cations by plants has been regarded as a major process of soil acidification in unpolluted or less polluted forest ecosystems (Marcos and Lancho, 2002; van Breemen et al., 1983), suggesting that increased uptake of base cations by trees in the N addition treatment may have contributed to soil acidification in the studied forest without N saturation occurring. As S deposition is expected to have accelerated soil acidification in AOSR, which has soils with coarse texture and low S adsorption capacity (Aherne and Shaw, 2010; Jung et al., 2011a), reduction of the soil exchangeable cationic nutrient pool caused by increased N deposition as discussed earlier can exacerbate nutrient imbalance or deficiency in combination with soil acidification (Erisman and De Vries, 2000), even though significant nitrate leaching does not occur in AOSR.

As discussed in section 4.2, there have not been many reports on N saturation in N-limited Canadian forests. In addition, global warming coupled with increased chronic N deposition will likely increase the net primary productivity of boreal forests in the oil sands region in the future (Hari and Kulmala, 2008; Magnani et al., 2007). However, the increased net primary productivity does not eliminate the risk of N saturation and N leaching and/or emission can occur with a relatively low N deposition rate (De Schrijver et al., 2008). Therefore, the long-term effect of elevated levels of N deposition, especially coupled with significant S deposition, remains a concern, as nutrient imbalance or deficiency in AOSR due to reductions in soil cationic nutrient availabilities caused by increased tree growth/uptake and cation loss that accompanies SO_4^{2-} leaching.

5. Conclusions

There was no evidence of N saturation in the studied forest ecosystem in AOSR after four years of elevated levels of simulated N and S depositions (with a total of 120 kg ha^{-1} of N or S added alone or in combination). There was no long-term increase of inorganic N concentrations in the soil, leaching of N beyond the main rooting zone in the soil profile was minimal, and tree growth was increased by simulated N deposition, all indications of N limitation in the studied forest stand. However, exchangeable Ca^{2+} and Mg^{2+} concentrations in the surface mineral soil layer were found to be reduced by N and S additions because of increased cation leaching associated with increased SO_4^{2-} leaching caused by S addition and increased nutrient uptake associated with increased tree growth resulting from N addition. This has implications for potential risk of induced nutrient imbalance in boreal forests in AOSR by long-term chronic N and S depositions. The long-term effects of N and S depositions on the cycling of N and other nutrients in the ecosystems in AOSR remain to be studied, even though the immediate risk of N saturation or causing negative environmental consequences in the boreal forest is low if the rates of N and S depositions can be controlled under the current level.

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Table 1. Basic properties of soils sampled in 2010 from the studied site in a mixedwood forest in the Athabasca oil sands region. Different lowercase letters indicate a significant difference between the treatments at α=0.1. Values in parentheses are standard errors of the means.

Soil layer	Treatment	pH	Total C	Total N	Exchangeable cations					Ca/Al[1]
					Ca^{2+}	Mg^{2+}	K^{+}	Na^{+}	Al^{3+}	
			g kg^{-1}		mmol$_c$ kg^{-1}					
Forest floor	CK	5.5	296	13.7	41.2ab	5.5	7.5c	0.09c	0.06	1207
		(0.2)	(36)	(2.1)	(6.8)	(1.1)	(0.4)	(0.01)	(0.03)	(655)
	+N	5.5	308	14.4	47.5a	6.7	8.5b	0.11c	0.07	1140
		(0.1)	(37)	(1.2)	(6.1)	(1.0)	(0.8)	(0.01)	(0.04)	(397)
	+S	5.5	311	13.4	37.5b	5.5	8.2b	0.66b	0.06	953
		(0.2)	(52)	(2.2)	(5.6)	(1.2)	(0.8)	(0.2)	(0.03)	(298)
	+NS	5.5	311	13.9	45.8ab	6.6	8.9a	1.6a	0.05	1630
		(0.2)	(24)	(1.8)	(9.1)	(3.1)	(1.1)	(1.0)	(0.02)	(891)
Surface mineral soil (0-15 cm)	CK	5.0a	5.7	0.35	3.5a	0.67a	0.56	0.06c	0.23	75.7a
		(0.1)	(0.8)	(0.07)	(0.7)	(0.11)	(0.05)	(0.01)	(0.28)	(89.8)
	+N	4.5b	6.2	0.37	2.5ab	0.55ab	0.56	0.06c	0.38	12.9b
		(0.2)	(0.6)	(0.06)	(0.2)	(0.11)	(0.04)	(0.00)	(0.21)	(7.7)
	+S	4.7ab	5.8	0.36	2.8ab	0.51ab	0.56	0.31a	0.35	14.3b
		(0.3)	(0.7)	(0.04)	(0.3)	(0.15)	(0.04)	(0.07)	(0.10)	(16.3)
	+NS	4.8ab	6.3	0.37	2.0b	0.39b	0.55	0.20b	0.40	13.0b
		(0.1)	(0.3)	(0.03)	(0.6)	(0.08)	(0.05)	(0.02)	(0.26)	(4.6)

[1] Ca/Alis the ratio of exchangeable Ca^{2+} to exchangeable Al^{3+} based on a molar unit.

Table 2. Annual leaching loss of NO_3^-, SO_4^{2-}, $NH_4^+$and base cations in 2009 in a mixedwood forest in the Athabasca oil sands region, Alberta, Canada. Different lowercase letters indicate a significant difference between treatments at α=0.1. Values in parentheses are standard errors of the means.

Depth	Treat-ment	SO_4^{2-}	NO_3^-	NH_4^+	Ca^{2+}	Mg^{2+}	K^+	Na^+
		---------- mmol$_c$ m^{-2} yr^{-1} ----------						
Below 15 cm soil depth	CK	6.9b	0.3b	0.7	7.9b	3.9ab	2.8bc	1.2c
		(2.4)	(0.1)	(0.1)	(1.4)	(1.1)	(0.8)	(0.3)
	+N	3.9b	1.5a	0.9	9.0ab	3.6b	6.1a	2.9b
		(2.4)	(0.7)	(0.3)	(1.3)	(0.4)	(2.1)	(0.6)
	+S	59.1a	0.1b	0.8	11.2a	4.9a	3.1b	46.5a
		(22.1)	(0.0)	(0.2)	(2.1)	(1.1)	(0.7)	(10.7)
	+NS	55.0a	1.3a	0.8	8.2ab	3.8ab	2.2c	41.3a
		(23.4)	(0.5)	(0.2)	(1.4)	(0.5)	(0.2)	(11.3)
Below 45 cm soil depth	CK	2.3c	0.03	0.2b	0.8c	0.2c	0.03	0.1b
		(1.2)	(0.01)	(0.1)	(0.1)	(0.1)	(0.01)	(0.1)
	+N	0.3d	0.02	0.1b	0.4d	0.1c	0.1	0.1b
		(0.2)	(0.01)	(0.05)	(0.1)	(0.1)	(0.01)	(0.2)
	+S	12.4a	0.02	0.1b	4.1a	0.9a	0.1	1.9a
		(3.5)	(0.02)	(0.04)	(0.6)	(0.6)	(0.1)	(1.2)
	+NS	6.1b	0.04	0.5a	1.6b	0.6b	0.1	1.3a
		(1.0)	(0.02)	(0.2)	(0.6)	(0.2)	(0.1)	(0.4)

Figure captions

Fig. 1. Effects of N and S additions on the forest vegetation: a) canopy trees (*Populus tremuloides*), b) litterfall, and c) understory in 2009 in a mixedwood forest in the Athabasca oil sands region, Alberta, Canada. Nitrogen and S were applied at the rate of 30 kg ha yr^{-1} as N and/or S from 2006. The CK, +N, +S and +NS refer to control, N addition, S addition and N+S addition, respectively. Error bars are standard errors of means.

Fig. 2. Effects of N and S additions on soil microbial biomass C (MBC), N (MBN), and inorganic N in a) the forest floor and b) the surface mineral soil in 2009 in a mixedwood forest in the Athabasca oil sands region, Alberta, Canada. The CK, +N, +S and +NS refer to control, N addition, S addition and N+S addition, respectively. First addition in 2009 was on July 2, with a rate of 10 kg N and/or S ha^{-1}, and the 2nd and 3rd additions occurred 20 and 40 days after the 1st addition, respectively, with the same rate. Error bars are standard errors of means.

Fig. 3. Effects of N and S additions on leaching loss a) below 15 cm and b) below 45 cm of NH_4^+, NO_3^-, SO_4^{2-}, Na^+, Ca^{2+}, and Mg^{2+} in 2009 in a mixedwood forest in the Athabasca oil sands region, Alberta, Canada. The CK, + N, +S and +NS refer to control, N addition, S addition and N+S addition, respectively. Error bars are standard errors of means.

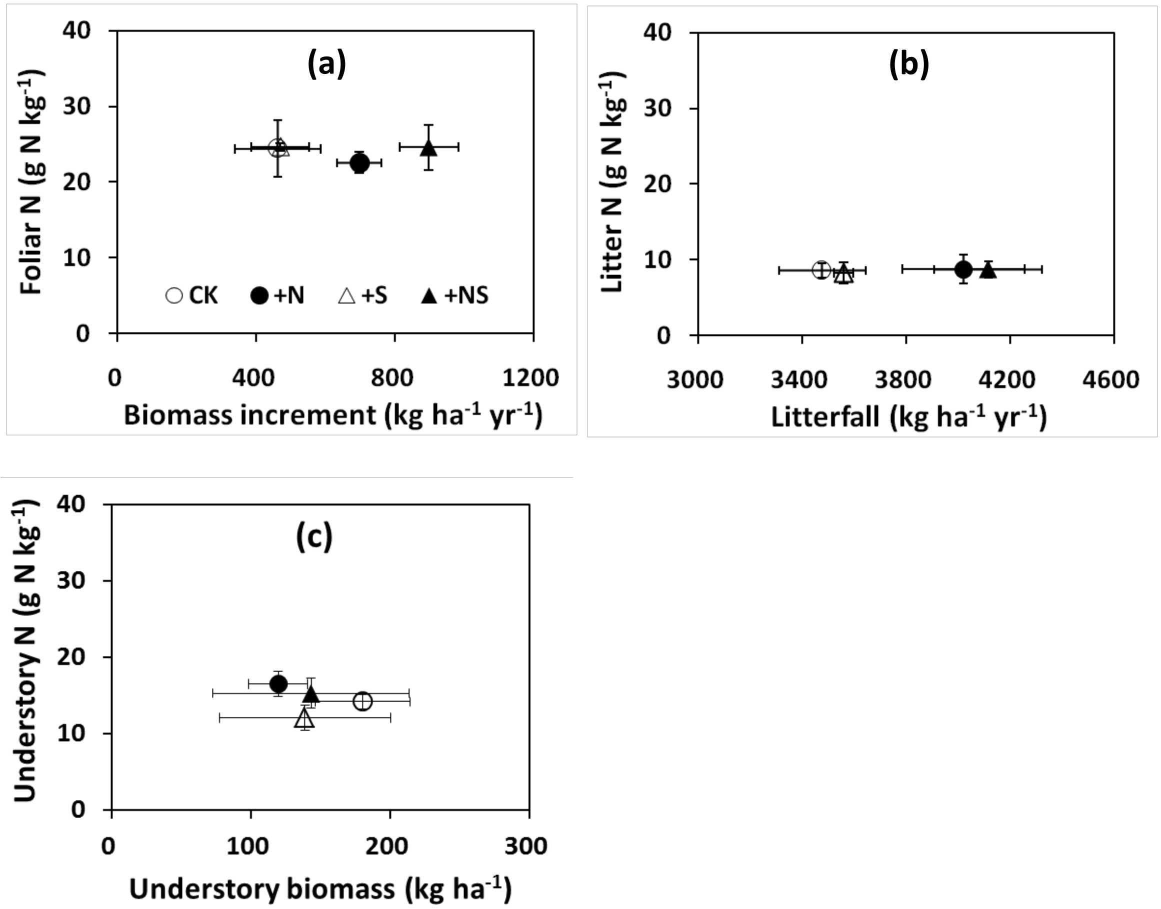
(a)
Foliar N (g N kg^{-1})
Biomass increment (kg ha^{-1} yr^{-1})
○ CK ● +N △ +S ▲ +NS
0
10
20
30
40
0
400
800
1200
(b)
Litter N (g N kg^{-1})
Litterfall (kg ha^{-1} yr^{-1})
0
10
20
30
40
3000
3400
3800
4200
4600
(c)
Understory N (g N kg^{-1})
Understory biomass (kg ha^{-1})
0
10
20
30
40
0
100
200
300

Fig. 1.

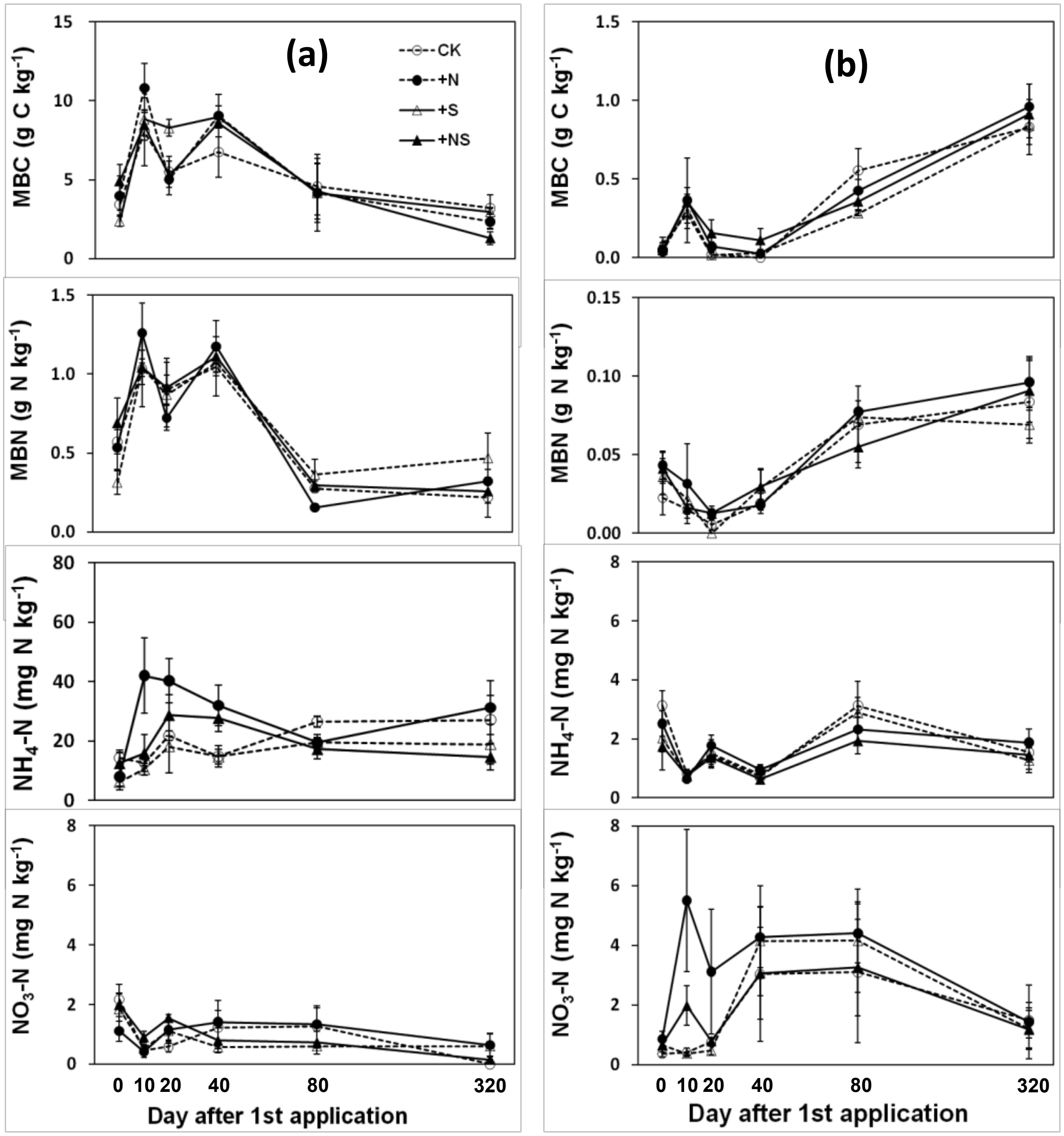
(a)
(b)
CK
+N
+S
+NS
MBC (g C kg^{-1})
MBN (g N kg^{-1})
NH$_4$-N (mg N kg^{-1})
NO$_3$-N (mg N kg^{-1})
0
10
20
40
80
320
Day after 1st application

Fig. 2.

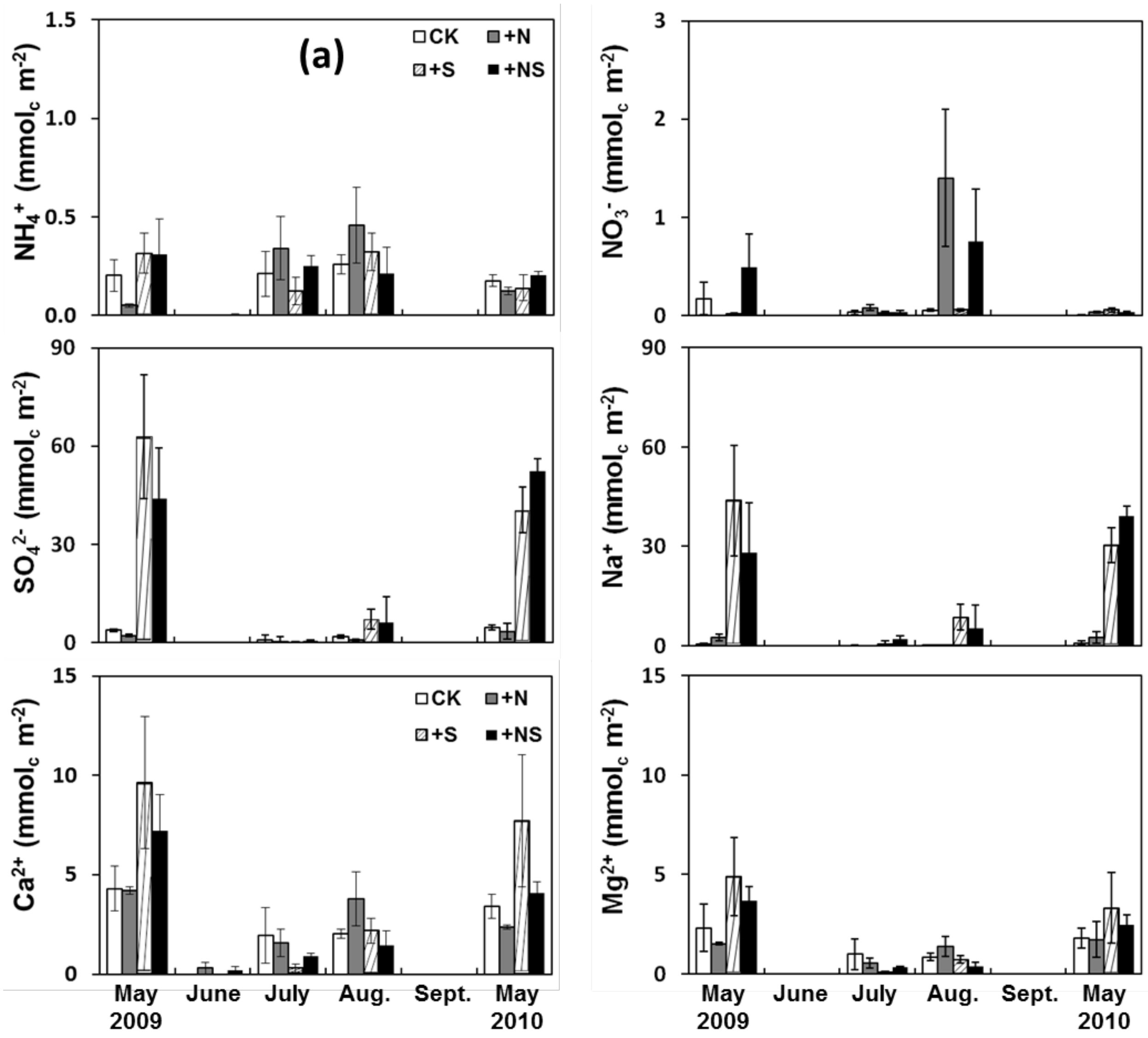
(a)
□CK
▨+S
■+N
■+NS
NH_4^+ (mmol$_c$ m^{-2})
0.0
0.5
1.0
1.5
NO_3^- (mmol$_c$ m^{-2})
0
1
2
3
SO_4^{2-} (mmol$_c$ m^{-2})
0
30
60
90
Na^+ (mmol$_c$ m^{-2})
0
30
60
90
Ca^{2+} (mmol$_c$ m^{-2})
0
5
10
15
Mg^{2+} (mmol$_c$ m^{-2})
0
5
10
15
May 2009
June
July
Aug.
Sept.
May 2010

Fig. 3.

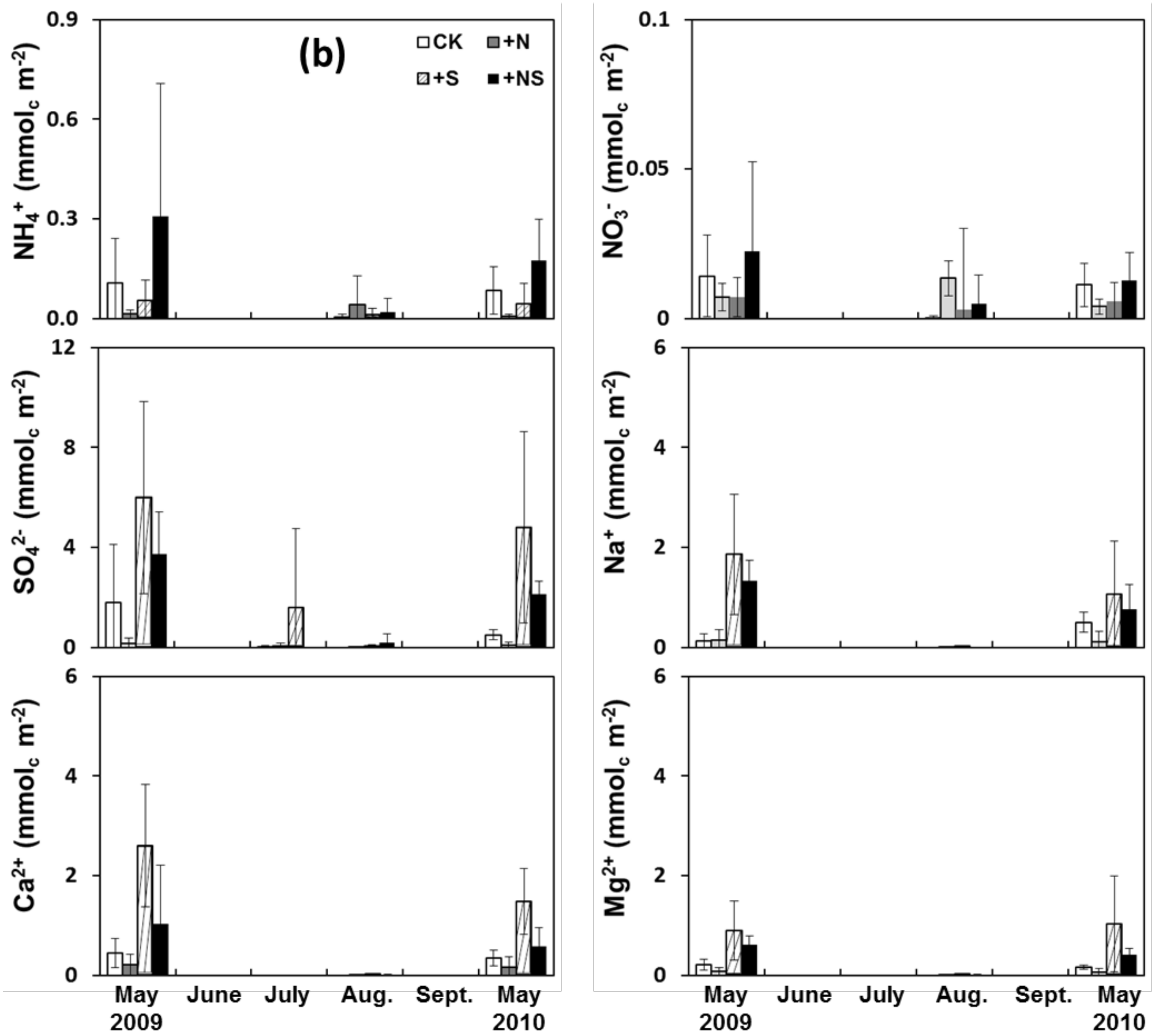

Fig. 3. Continued