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Soil and tree chemistry reflected the cumulative impact of acid deposition in *Pinus banksiana* and *Populus tremuloides* stands in the Athabasca oil sands region in western Canada

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ABSTRACT

The spatial variability of soil chemistry and Ca/Al ratios of soil solution and fine roots were investigated in jack pine (*Pinus banksiana*) and trembling aspen (*Populus tremuloides*, aspen) stands to assess the impact of chronic acid deposition on boreal forest ecosystems in the Athabasca oil sands region (AOSR) in Alberta, Canada. Available SO_4^{2-} (as the sum of soluble and adsorbed SO_4^{2-}) accumulated in the soil near tree boles of both species, reflecting the influence of canopy intercepted SO_4^{2-}. In jack pine stands, pH and soluble base cation concentrations decreased towards tree boles due to increased SO_4^{2-} leaching; the reverse was found in aspen stands due to deposition of base cations leached from the canopy. As a result, Ca/Al ratios in the soluble fraction in soils near jack pine boles were 5–20 times lower than that near aspen boles. The Ca/Al ratio did not reach the critical limits of 1.0 for soil solution (ranged from 1.0 to 4.1) or 0.5 for fine roots (0.7–7.9) in the studied watersheds. However, Al^{n+} concentrations in the soil solution ranged from 0.2 to 4.1 mg L^{-1} in NE7 and from 0.1 to 8.5 mg L^{-1} in SM8 that can inhibit the growth of white spruce (*Picea glauca*) seedlings that commonly succeed aspen in upland sites in the AOSR. We suggest that the spatial variation caused by tree canopies/stems will affect forest regeneration and the effect of acid deposition on forest succession in the AOSR should be further studied.

1. Introduction

The Athabasca oil sands region (AOSR) is the largest area for open-pit oil sands mining in Alberta, Canada (Humphries, 2008) and a large amount of acid forming compounds has been released to the atmosphere over the past several decades (Hazewinkel et al., 2008). As acid deposition can induce soil acidification in terrestrial ecosystems, adverse effects of acid deposition and subsequent soil acidification on forest ecosystems are of concern in the AOSR, especially in acid-sensitive watersheds with soils having low capacities for pH buffering (AMEC, 2009) and SO_4^{2-} adsorption (Jung et al., 2011b). As the impact of acid deposition on forest ecosystems is the result of long-term chronic exposure, accumulative indicator can be more useful to show the long-term impact of acid deposition than indicators that respond quickly to recent changes in air quality (Vanguelova et al., 2007a).

Acid deposition and subsequent leaching of strong acid anions such as SO_4^{2-} increase soil acidity and reduce the availability of cationic nutrients such as Ca^{2+} in the ecosystem (Likens and Bormann, 1995). Increasing soil acidity raises Al solubility and Al concentrations (particularly in the forms of Al^{3+} and $AlOH^{2+}$) in the soil (Tejnecký et al., 2010). In acidic soils major forms of Al ions such as Al^{3+} and $AlOH^{2+}$ are toxic to plants (Delhaize and Ryan, 1995). Therefore, soil Al and Ca concentrations are affected by acid deposition, and Al toxicity and Ca deficiency can be common in acidic soils (Clark and Baligar, 1995). In addition to Al^{n+} and Ca^{2+} concentrations in the soil solution, Ca/Al ratios in soil solution and fine roots have been suggested as sensitive indicators for acidity stress in forest ecosystems under elevated levels of acid deposition (Hirano et al., 2007; Innes, 1995; Shortle and Smith, 1988) due to antagonism between Al and Ca uptake (Delhaize and Ryan, 1995). The Ca/Al ratio of soil solution results from changes in soil chemistry due to acid deposition, and the Ca/Al ratios of fine roots can reflect the cumulative adsorption of toxic Al in fine roots to exclude Al transport to aboveground plants in acidic soils.

Spatial patterns of soil chemistry such as pH and SO_4^{2-} concentrations around trees can reflect the interaction between acid deposition and tree canopies. Atmospheric deposition occurs through two processes: precipitation deposition (=bulk deposition) and interception deposition (Ulrich, 1983). Bulk deposition

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occurs vertically by gravity while interception deposition involves aerosols or particulate matters being intercepted through fogs, clouds, or dews by obstacles. Interception deposition leads to increasing atmospheric deposition around trees by throughfall and stemflow, especially because acid materials such as SO_2 mainly travel as aerosols. For example, interception deposition was similar to bulk deposition for base cations but more than twice that of bulk deposition for SO_4^{2-} in Douglas-fir (*Pseudotsuga menziesii*) stands in the Netherlands (Draaijers and Erisman, 1995). Similar trends were found in European beech (*Fagus sylvatica* L.) stands in Belgium (Staelens et al., 2008), and jack pine (*Pinus banksiana*) and trembling aspen (*Populus tremuloides*) stands in the AOSR (Jung et al., 2011a), indicating that soils around trees are exposed to greater amounts of acid deposition and experience greater changes in soil chemistry than those in open fields.

In addition to differences in depositional processes, tree species affects soil chemistry through a number of other processes: canopy uptake and leaching, organic acid exudation, nutrient uptake and turnover, and litterfall decomposition as affected by the quality and quantity of the litterfall (Augusto et al., 2002; Binkley and Giardina, 1998; Knops et al., 2002; Rhoades, 1996), typically resulting in more acidic soils in conifer stands than in deciduous stands. Decreasing patterns of soil pH and concentrations of base cations in the soil and/or soil solution towards the tree bole have been reported in conifer stands in industrial areas (Zinke, 1962; Riha et al., 1986a; Skeffington, 1983), accompanying decreasing pH and increasing SO_4^{2-} concentrations in stemflow (and throughfall) as compared with bulk deposition due to accumulated acid deposition and canopy leaching of organic acids (Staelens et al., 2008; Jung et al., 2011a). On the other hand, reports on spatial patterns of soil chemistry in deciduous stands have not been consistent, with both lower (Sato and Wakamatsu, 2001) and greater acidity towards a tree bole (Skeffington, 1983; Chang and Matzner, 2000) being reported. The deciduous canopy can raise pH of throughfall and stemflow through canopy uptake of H^+ associated with canopy leaching of base cations. Soil acidity in deciduous stands can decrease towards tree boles in the non- or less-polluted areas due to canopy leaching of base cations, but can increase in highly polluted areas due to interception deposition of acid substances.

In this study, we tested two hypotheses: (1) soil acidification by acid deposition was accelerated surrounding tree boles due to interception deposition, which would be indicated by the spatial variability of soil chemistry, and (2) trees in the AOSR were exposed to acid stress due to chronic acid deposition, which would be shown by Ca/Al ratios of soil solution and fine roots. A better understanding of soil solution chemistry and the spatial variability in soil properties caused by tree species and canopy interaction will substantially help us better assess and understand the impact of atmospheric deposition on terrestrial ecosystems.

2. Materials and methods

2.1. Site description

Two watersheds, NE7 and SM8, in the AOSR were selected for this research. Watershed NE7 (57.15°N, 110.86°W) is located northeast of Fort McMurray, Alberta, Canada, while SM8 (56.21°N, 111.20°W) is located south of Fort McMurray. As most oil sands mining industries in the AOSR were located north of Fort McMurray and prevailing wind direction was east and east-southeast (Vickers et al., 2002), NE7 would have been exposed to greater air pollution than SM8 (WBEA, 2010). Climate conditions were similar in both watersheds. The mean annual temperature is 0.7 °C with a mean relative humidity of about 67%. The mean annual precipitation and evaporation are 456 and 486 mm, respectively (Ok et al., 2007). Dominant tree species were approximately 60-year-old trembling aspen (aspen) and jack pinetrees in upland forests in both watersheds. Stand density was 1385 trees ha^{-1} in NE7 and 1170 trees ha^{-1} in SM8. The mean diameter at breast height (DBH) of dominant trees and stand height, based on average height of dominant trees, were 10.6 cm and 11.2 m, respectively, in NE7, and 13.1 cm and 12.2 m, respectively, in SM8. Dominant tree species in low-lying areas and wetlands was black spruce (*Picea mariana*). Common soil types in upland forests of both watersheds were coarse textured Elluviated Brunisolic soils and Luvisolic soils (Bertsch and Bloom, 1996) in the Canadian system of soil classification (Soil Classification Working Group, 1998) and they belong to Boralf in US Soil Taxonomy (Soil Survey Staff, 1994). Basic properties of soils in NE7 and SM8 are provided in Table 1. For this study, five 20 m × 20 m plots were established in each watershed in 2005. Three plots were jack pine dominated stands and the other two plots were aspen dominated stands distributed across the landscape in each watershed.

2.2. Water sampling and analysis

Bulk precipitation, throughfall and stemflow collectors were installed in November 2005 to collect the first sample in May 2006. Chemical properties of bulk deposition, throughfall, and stemflow were used to compare changes in precipitation by canopy trees between watersheds and between tree species. The collectors for bulk deposition and throughfall consisted of a 1 L bottle, a funnel (10 cm radius) with a screen (1 mm × 1 mm opening size), and a PVC tube covered by aluminum foil. The PVC tube was pushed about 5 cm into the soil and served as a stand for the funnel and the bottle for collecting the water sample was housed within the PVC tube. Three bulk deposition collectors were installed in open areas near experimental plots for collecting bulk deposition in each watershed. Three throughfall collectors were installed in each plot by placing the collectors below the canopy of dominant trees with various sized canopy cover area. Collectors were located at about 2/3 points from tree trunks to canopy cover boundary. This means the collectors were typically located approximately 70–110 cm from the tree trunk. Three stemflow sample collectors were also set up in each plot. Stemflow was gathered in collecting bottles of 4 L capacity through an opened up tygon tube glued around a bole from breast height to collectors on the ground, placed alongside the tree bole. Collectors for bulk deposition, throughfall, and stemflow were replaced every month during the growing season from May to October from 2006 to 2009 and in May 2010.

To collect soil solution samples, two stainless steel zero-tension lysimeters were installed at each of the 15 (within the main rooting zone) and 45 cm (beyond the main rooting zone) soil depths in May 2008. Distance between a zero-tension lysimeter and its closest tree was similar to the arrangement for throughfall collectors. The water collection area of each lysimeter was 500 cm^2. Soil solution was collected every month during the growing season from June to October in 2008, from May to October in 2009, and in May 2010. All collecting bottles were washed with 20% HCl solution and properly rinsed with deionized water in the laboratory. A few 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 water samples in between samplings.

All samples were stored in a refrigerator before analysis. A portion of each sample was filtered with 0.22 μm syringe filters before chemical analysis. The pH was measured with an AR20 pH meter (Fisher Scientific Ltd., USA). Concentrations of Ca^{2+}, Mg^{2+}, K^+, and Al^{n+} were measured using an ICP-MS (Elan 6000 quadrupole, Perkin-Elmer, Inc., USA). Concentrations of SO_4^{2-}, NO_3^-, and NH_4^+ ions were analyzed with ion chromatography (DX 600, Dionex