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Modelling carbon isotopes in spruce trees reproduces air quality changes due to oil sands operations

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A B S T R A C T

Direct monitoring of air quality does not cover more than the last three decades in most industrialized countries. For that reason studies using growth-ring carbon isotopes ($\delta^{13}C$) of several species of trees have recently investigated isotopic responses in the contexts of stationary and diffuse pollution in humid continental conditions. Here, the growth-ring $\delta^{13}C$ series (1880–2009) of spruce trees living in sub-humid subarctic conditions were measured to assess if they represent indicators for air quality changes near oil sands (OS) developments initiated in northeastern Alberta in 1967. The measured $\delta^{13}C$ pre-operation rings at two forest sites were analyzed along local climatic conditions to develop response-to-climate statistical models and predict the natural isotopic behaviour for the most recent part of the ring series. The measured trends and climate-modelled (natural) $\delta^{13}C$ values strongly depart during the operation period, depicting anomalies which can be nicely reproduced by multiple regression models combining climate and a proxy for OS airborne emissions. This research allows envisioning the use of carbon dendroisotopic indicators to compensate for the lack of long-term air quality measurement, and monitor environmental conditions in the sub-humid terrestrial ecosystem exposed to emissions from oil sands operations which are predicted to increase in the future.

1. Introduction

Atmospheric and NOx deposition may acidify surface waters and remove nutrients essential for the maintenance of forest soil fertility. In North America, the direct measurement of these air pollutants is generally limited to the last 20–30 years in rural regions where stationary sources are active or in urban areas typified by diffuse pollution. To obtain a longer perspective, past emissions are generally modelled on the basis of productivity reported by the industries. The context of the oil sands districts requires attention as the current level of developments generates significant SOx and NOx emissions. In northwestern Canada, the production of oil is expected to triple in the next few years, and a consequent increase in acidic emissions is expected (Alberta Environment, 2008; CAPP, 2012a,b). Direct air quality monitoring has been initiated in 1997 (WBEA, 2007). However, to help predict potential impacts in this broad region, a pre-operation perspective is needed.

Recently, isotopic indicators in trees have been tentatively used to palliate such lack of long-term records given that widespread species have the potential to provide annual records of environmental changes over several decades, even centuries (e.g., Farquhar et al., 1989; Lipp et al., 1996; McCarroll and Loader, 2004). As trees absorb CO_2 and some air contaminants through their foliar system, their growth-ring carbon stable isotopes ($\delta^{13}C$) reflect local climatic conditions (e.g., Young et al., 2010) and changes in ambient air chemistry at the time of their formation (Savard, 2010 and references therein). For instance, several studies have successfully inferred long-term trends of point source air pollution (e.g., Freyer, 1979; Sakata and Suzuki, 2000; Savard et al., 2004; Wagner and Wagner, 2006). But only a few studies have addressed the question of diffuse atmospheric pollution from urban centres (Bukata and Kyser, 2007; Savard et al., 2009; Doucet et al., 2012), where reconstructing environmental changes constitutes a real challenge due to the multitude of emitting points, their varied history, and the overall complex chemistry of the emissions. The particular context of the oil sands developments sits between the point source and urban centre types, as its regional conditions share the simplicity of the contamination chemistry with point sources and the varied history and diffuses emission characters of urban areas.

One of the main difficulties in trying to reconstruct changes in air quality using isotopic indicators of living natural archives, particularly trees, lies with the need to separate natural effects (climate, disease, population dynamics) from anthropogenic impacts (stress

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due to pollution) that affect the indicators. Interestingly, natural effects such as disease and population dynamics can be generally avoided by rigorously selecting sites and tree stands (see Section 3). A central point to the rationale of this research is the fact that the climatic and anthropogenic effects can be separated if consistent climatic series extending several decades prior to the onset of industrial operations are available for the region of interest (Doucet et al., 2012). Such climatic series are available for the northeastern Albertan oil sands region. But on the other hand, the sensitivity of the dendroisotopic systems to climate variations and pollution stress vary with the species of trees (Black, 1982), and greatly depends on the climatic regime. If the isotopes of spruce trees which are widespread in the boreal ecozone have been successfully shown to record climate and air quality conditions of humid continental regions, their utility for such purpose in a sub-humid subarctic area has never been assessed.

The main purpose of this research was to test the hypothesis that ring $\delta^{13}C$ series of white and black spruce trees growing under sub-humid subarctic conditions can record pollution effects due to oil sands extraction. This study specifically aimed at: (1) verifying if it is possible to objectively separate, on 129 years-long $\delta^{13}C$ series, the effects due to climatic variations from those generated by airborne emissions from the Lower Athabasca oil sands operations; and ultimately (2), providing a historical perspective on changes in air quality, and acidic deposition, that may affect the terrestrial ecosystem. The approach advocated here for separating the natural and anthropogenic effects involves using response functions for tree rings of the pre-operation period, and predicting the natural $\delta^{13}C$ behaviour for the recent period.

2. Region of investigation

2.1. History of emissions in the Lower Athabasca oil sands region

Oil production from bitumen extraction in Alberta was initiated in 1967, and expanded significantly in 1978 and 2003. Through these developments large amounts of atmospheric N and S have been released in the Athabasca oil sands region where truck fleets, oil upgraders, desulphurization and hydrogen plants, boilers, heaters and turbines have been expanding in a complex temporal and spatial fashion (Fig. 1). The N and S emissions from these sources have not been monitored prior to 1997, therefore SO_2 calculated using hydrocarbon productivity in the region is used as a proxy for air quality (CEMA, 2003) The proxy for NOx emissions increased steadily up to 320 T/d in 2010, while the SO_2 proxy increased from being negligible before 1967, to >450 T/day in 1993–1994, followed by substantial decreases after 1995 due to technological advances (Hazewinkel et al., 2008; Morrison, 2006). Current depositional rates of SO_4^{2-}-S have been measured in the region using ion exchange resin collectors which yielded throughfall depositional rates between 7.4 and 39.2 kg/ha/year for fifteen sites within a radius of 27 km from operations (Proemse et al., 2012), and an average rate of 1.1 kg/ha/year (maximum of 3.7 kg/ha/year) for 11 sites distributed over the broad region (26–150 km from operations; Wieder et al., 2010).

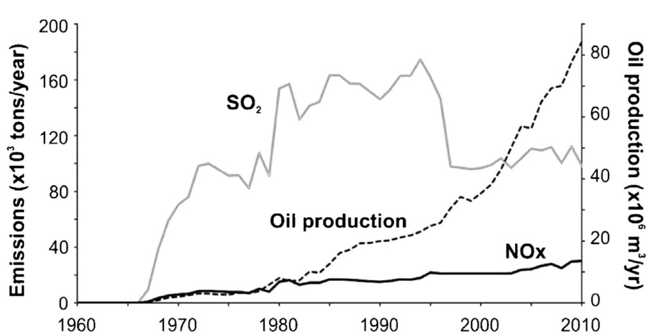

Fig. 1. Modelled annual production (right vertical axis) and measured acidifying emissions for the Lower Athabasca Region in northeastern Alberta (CEMA, 2003; EC, 2013).

2.2. Climate data and meteorological stations

The investigated area is located north of Fort McMurray and belongs to the boreal forest of the interior plains physiographic region of Canada. Forests are largely dominated by jack pine, a species prone to regeneration after fires, and by spruce stands, in areas preserved from wild fires. Note that in this sector of the boreal forest, natural fires represent one of the most important ecological disturbances. The investigated region has a generally flat topography, largely covered with muskegs, and is characterized by a sub-humid subarctic climate (Phillips et al., 1990), seeing an average total precipitation of 455 mm/year, with contrasted seasons where winters and summers have an average temperature of −16.3 and 15.6 °C, respectively.

The climatic parameters used for the statistical analyses are extracted from the Climate Research Unit (version TS3.10.1), at the nearest points to the studied sites (Mitchell and Jones, 2005). The temperature and precipitation records are continuous over the 1910–2009 period.

Sites 1 and 2 are located at 12 km N–E and 37 km S–E from the heart of the open mining operations, which is arbitrarily positioned halfway between the Suncor Millennium and Syncrude Mildred Lake upgraders (Fig. 2). However, specifically site 2 is nearly as close as site 1 to open mining operations and their sources of emissions (about 13 km).

3. Material and methods

3.1. Site selection and sampling of trees

Ideal forest sites for air and soil pollution study using dendrogeochemistry must hold healthy old trees growing on well drained soils (Bégin et al., 2010). One reason for this last point is that drainage influences the dendrogeochemical responses to climatic conditions. In the case of $\delta^{13}C$ series, soil drainage characteristics will not necessarily alter trees sensitivity to changes in air quality. The three stands were selected where healthy and old dominating trees showed no apparent ecological perturbations: black spruce trees (*Picea mariana* (Mill) BSP) at site 1, and white spruce trees (*Picea glauca* (Moench) Voss) at site 2 (Table 1 and Fig. 2). Site 1 lies on a podzol developed on flat and poorly drained sandy deposits undergoing paludification. The site 2 stand grows on a well drained brunisol developed on a gentle slope cutting through a glacial till.

Sixteen trees were carefully selected at site 1, and 17, at site 2, and sampled with 5 mm diameter increment borers at breast height for dendrochronology (dating rings, measuring growth rates, studying stand dynamics). Among them, three specimens were sectioned with a chain saw and sampled for geochemical analyses. All samples were measured using standard dendrochronological methods; the COFECHA programme was used to verify dating (Holmes, 1992). Rings were manually separated with clean stainless blades at a two-year resolution for the 1880–1909 and 1962–2009 periods (n = 39), and at one-year resolution for the pre-operation 1910–1961 episode (n = 52), which will constitute the statistical calibration period. The rings of individual trees were ground with a Wiley mill at 4 mesh. Then sub-samples of corresponding years and of exact same weight from the three different trees at each site were pooled together for the $\delta^{13}C$ results to be representative of the site.