

Contemporary Issues in Québec's Temperate Forest — Part 3: Air Pollutants

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ABSTRACT

This paper is the third in a series documenting contemporary issues in Québec's temperate forest. It addresses air pollutants, including the acidifying air pollutants that cause acid rain, as well as ground-level ozone and trace elements. It briefly discusses the anthropogenic sources of these pollutants, their current status, their impacts on forest ecosystems and the issues they cause for Québec's temperate forest. Most air pollutants come from combustion of fossil fuels for energy production or transportation. Since the mid-1990s, thanks to pollutant emission reduction programs implemented in Canada and the United States, emissions and ambient air concentrations of most pollutants have declined significantly. Sugar maple (*Acer saccharum* Marsh.), a dominant species in the northern temperate zone, is especially sensitive to air pollutants and their impacts for ecosystems. Previous chronic pollution of these forests has resulted in significant loss of ecosystem services. However, environmental monitoring is ongoing with a view to documenting ecosystem reactions in the wake of contemporary decreases in anthropogenic emissions of air pollutants in North America.

Keywords: air pollution, acid rain, acidifying emissions, air quality, heavy metals

RÉSUMÉ

Cet article est le troisième d'une série qui vise à documenter les enjeux contemporains en forêt tempérée au Québec. Nous y abordons plus spécifiquement les polluants atmosphériques, notamment les polluants atmosphériques acidifiants à l'origine du phénomène des pluies acides, l'ozone troposphérique et les éléments traces. Nous discutons brièvement des sources anthropiques de ces polluants, de l'état de la situation, de leurs effets sur les écosystèmes forestiers et des enjeux qui en découlent pour la forêt tempérée au Québec. Les polluants atmosphériques proviennent majoritairement de la combustion de combustibles fossiles pour la production d'énergie et pour le secteur des transports. Depuis le milieu des années 1990, grâce aux programmes de réduction des émissions de polluants atmosphériques mis en œuvre au Canada et aux États-Unis, les émissions et les concentrations dans l'air ambiant de la plupart des polluants ont baissé significativement. L'érable à sucre (*Acer saccharum* Marsh.), une essence dominante dans la zone tempérée nordique, s'avère particulièrement sensible aux polluants atmosphériques et à leurs effets sur les écosystèmes. La pollution chronique historique de ces forêts engendre donc une perte de services écosystémiques importante. Des suivis environnementaux des écosystèmes forestiers se poursuivent afin de documenter la réaction des écosystèmes forestiers à la baisse contemporaine des émissions anthropiques de polluants atmosphériques en Amérique du Nord.

Mots-clés : pollution atmosphérique, pluies acides, émissions acidifiantes, qualité de l'air, métaux lourds

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Introduction

Other than carbon monoxide (CO) which, once in the atmosphere, is converted into carbon dioxide (CO₂) and plays a major role in climate change², there are several air pollutants that are harmful to forest ecosystems. They include the acidifying pollutants that cause acid rain (most notably sulphur dioxide [SO₂] and the nitrogen oxides [NO_x]), as well as tropospheric or ground-level ozone and trace elements.

In this third paper of a series on contemporary issues in Québec's temperate forests (see Part 1: Duchesne *et al.* (*In press*) for a definition of the area under study), we synthesize the existing literature on the anthropogenic sources of these pollutants, along with their current status, their impacts on forest ecosystems and the issues to which they give rise in Québec.

Acidifying air pollutants

Acidifying air pollutants (SO₂ and NO_x) are created mainly by fossil fuel combustion for energy production. In North America, the main SO₂ emitters are the petroleum and gas industry, the coal-fired energy sector and the non-ferrous metal smelting and refining industries. As for NO_x, the transportation sector is the main source, followed by the petroleum and gas industry and the coal-fired energy sector (ECCC 2023a). When these pollutants come into contact with atmospheric water vapour, they are converted respectively into sulphuric acid (H₂SO₄) and nitric acid (HNO₃), both of which can be transported over long distances by dominant winds before falling to earth in wet or dry form. These pollutants are the main causes of "acid rain", a phenomenon that has been shown to be harmful to terrestrial and aquatic ecosystems and infrastructures.

In recent decades, Canada and the provinces have entered into a series of national and international agreements aimed at reducing air pollutant emissions. They include: the *United Nations Economic Commission for Europe's*, initiated in 1979, and followed by the *Canada-US Air Quality Agreement* in 1991, the *Canada-Wide Acid Rain Strategy for Post-2000*, ratified in 1998, and the *Acid Rain Action Plan* adopted by the New England Governors and Eastern Canada Premiers, also in 1998. Various measures, including in the electrical energy sector, have been introduced since then and have resulted in a major reduction in air emissions and SO₂ and NO_x deposits in both Canada and the United States (ECCC 2023a). Measurements carried out by atmospheric precipitation chemistry monitoring networks showed a subsequent reduction of roughly 60% in sulphate deposits (SO₄), and 30% in nitrate deposits (NO₃) in eastern North America between 1990 and 2019 (Fig. 1, Cheng *et al.* 2022). Measurements taken at three stations operated by the Forest Ecosystem Research and Monitoring Network (RESEF) confirm these trends in Québec, with a significant reduction in SO₂ and NO₃ concentrations and an increase in precipitation pH values (see Fig. 2 showing trends at the station located in the northern temperate zone). Although the situation has improved, however, acidifying pollutants continue to cause concern and are responsible for significant environmental impacts, according to the conclusions of the last status report on the Air Quality Agreement published in 2023 (ECCC 2023a).

Acid rain damages the foliar organs of trees and disturbs the activities of enzymes, micro-organisms and mycorrhizal fungi (Zhang *et al.* 2023). It also impoverishes and acidifies forest ecosystem soils, increasing the availability of toxic metals in acid soils, and can cause nutrient deficiencies for trees, threatening long-term forest productivity (Likens *et al.* 1996; Houle *et al.* 1997; Driscoll *et al.* 2001; Duchesne and Houle 2006, 2008; Johnson *et al.* 2008; Zhang *et al.* 2023). The effects of acidification depend on the soil's pH, which determines its buffering capacity, and the Ca/Al ratio is a good indicator of aluminum toxicity, which, in large quantities, harms root cell division and is only minimally translocated within the plant.

The phenomenon is especially concerning for sugar maple, a species that is eminently sensitive to soil acidity and the availability of calcium (Ca) and magnesium (Mg) cations. This sensitivity has been shown in numerous studies linking growth, abundance, nutritional status, mortality, survival, photosynthesis and dieback symptoms in sugar maple to soil fertility and basic cation availability (Table 1). The impacts of soil fertility on the species' performance parameters have also been proved on several occasions by experiments in the forests and in controlled environments (Table 1). Some of these studies have shown that American beech is much less sensitive to soil fertility and basic cation availability than sugar maple (Table 1). Based on this, acid rain has been identified as one of the factors in the emergence of maple dieback in the early 1980s and the proliferation of American beech in declining maple stands.

Using the critical load for ecosystem acidity – i.e. the acidity load that an ecosystem can absorb over the long term without being damaged – the scientific community has been able to quantify the occurrence and severity of acid deposit effects in the forest ecosystems of eastern North America (Ouimet *et al.* 2001, 2006; Burns *et al.* 2008; Clark *et al.* 2018; Cathcart *et al.* 2024). The margin by which the critical load for ecosystem acidity has been exceeded has declined significantly since the early 2000s in line with the contemporary reduction in precipitation acidity (Cheng *et al.* 2022), and signs of improved soil fertility are beginning to appear (Lawrence *et al.* 2015; Hazlett *et al.* 2020; Lawrence and Bailey 2021). However, it is likely to be many decades before soil fertility is back to its pre-anthropogenic acidification level (Caputo *et al.* 2016; Watmough *et al.* 2016). At equilibrium with atmospheric CO₂, uncontaminated rain has a slightly acidic pH of approximately 5.6. Although the situation has improved, atmospheric inputs of nitrogen are still of concern (Pardo *et al.* 2019). While nitrogen is essential to plant growth, chronic inputs to forest ecosystems can saturate soils, leading to runoff and impoverishment of cations. Long-term experiments with chronic nitrogen inputs have shown that maple stands are especially sensitive (Moore and Houle 2023), while boreal forests appear to be extremely resilient (Houle *et al.* 2024a, 2024b).

Given the dominance of sugar maple in northeastern North America's temperate zone and the size of the area characterized by soils with low buffer capacity, past chronic pollution of the forests by acidifying air pollutants has caused significant loss of ecosystem services. For example, even in the early 1980s, it was estimated that two million tapholes (roughly 15% of the total

² Duchesne, L., F. Guillemette, S. Bédard and R. Ouimet. (Under review). Contemporary Issues in Québec's Temperate Forest — Part 4: Climate changes. Submitted to For. Chron.

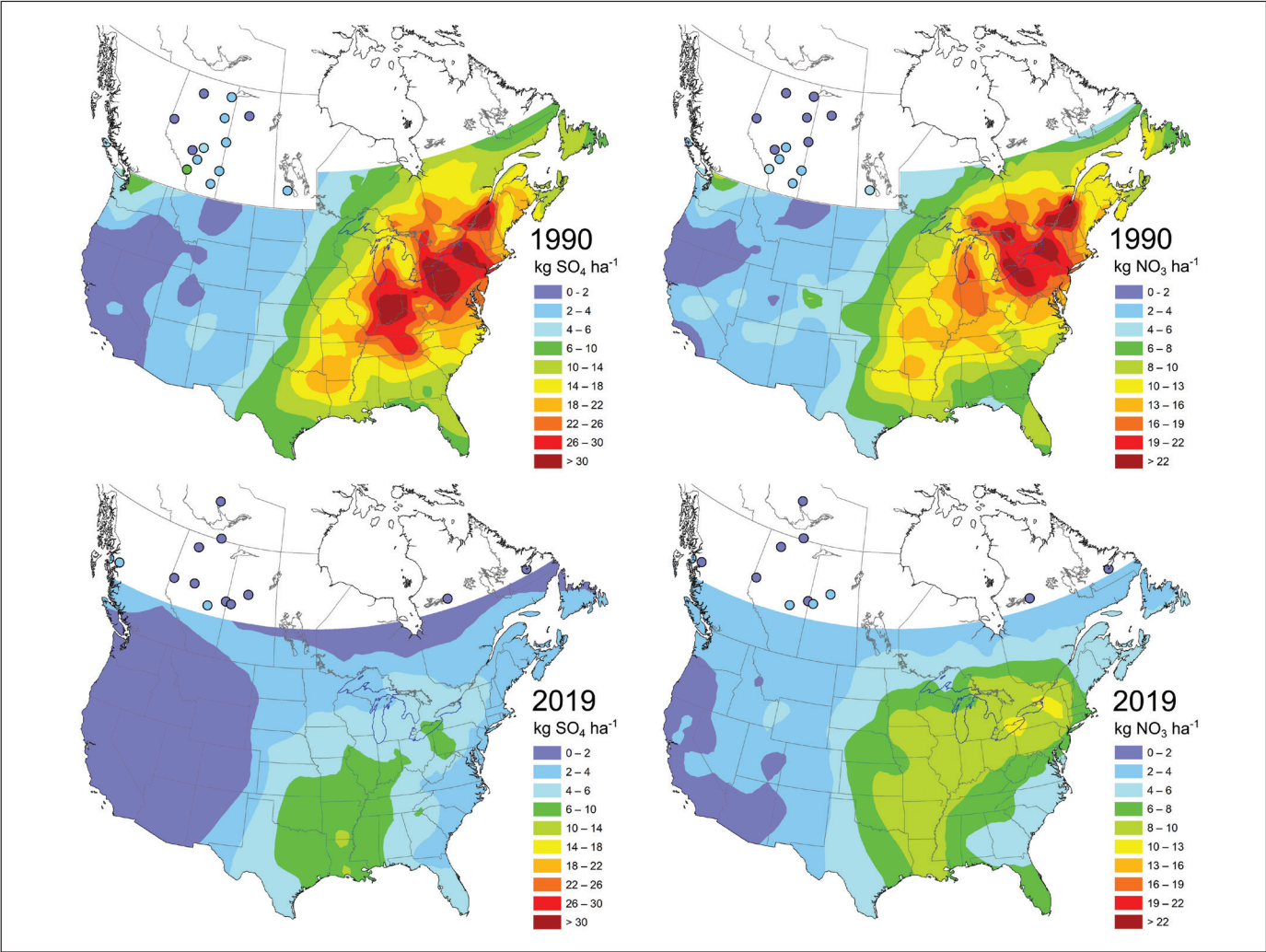


Fig. 1. Evolution of annual wet deposition of non-sea-salt sulphates (SO₄, left-hand column) and nitrates (NO₃, right-hand column) in North America from 1990 (top row) to 2019 (bottom row). Source: Air Quality Research Division, Environment and Climate Change Canada, Wet Deposition Maps – ECCC Data Catalogue.

Table 1. Examples of research documenting relationships between growth, abundance, nutritional status, mortality, survival, photosynthetic activity, sugar maple dieback symptoms and soil fertility, as well as sugar maple sensitivity to soil fertility, through experiments conducted in forests and controlled environments.

Study type	References (in ascending chronological order)
Relationship study for sugar maple	Bernier et Brazeau 1988a, 1988b, 1988c; Bernier <i>et al.</i> 1989; Ouimet et Camiré 1995, Ouimet <i>et al.</i> 1995; Wilmot <i>et al.</i> 1995; van Breemen <i>et al.</i> 1997; Horsley <i>et al.</i> 2000; McLaughlin <i>et al.</i> 2000; Arii <i>et al.</i> 2002, 2005; Drohan <i>et al.</i> 2002; Duchesne <i>et al.</i> 2002; Watmough 2002; St. Clair <i>et al.</i> 2005; Hallett <i>et al.</i> 2006; Kogelmann <i>et al.</i> 2006; Schaberg <i>et al.</i> 2006; Houle <i>et al.</i> 2007; Zaccherio <i>et al.</i> 2007; Page et Mitchell 2008; Long <i>et al.</i> 2009; Park et Yanai 2009; Bilodeau-Gauthier <i>et al.</i> 2011; Watmough <i>et al.</i> 2010; Sullivan <i>et al.</i> 2013; Pitel <i>et al.</i> 2014; Bishop <i>et al.</i> 2015; Cleavitt <i>et al.</i> 2018; Fernando <i>et al.</i> 2016; Lawrence <i>et al.</i> 2018; Soubeyrand <i>et al.</i> 2024
Experiments for sugar maple	Hendershot 1991; Ouimet et Fortin 1992; Paré <i>et al.</i> 1993; Ouimet <i>et al.</i> 1996a, 1996b; Wilmot <i>et al.</i> 1996; Long <i>et al.</i> 1997; Burke <i>et al.</i> 1998; Moore <i>et al.</i> 2000, 2008, 2012; Kobe <i>et al.</i> 2002; Wargo <i>et al.</i> 2002; Bailey <i>et al.</i> 2004; Juice <i>et al.</i> 2006; Moore et Ouimet 2006, 2014, 2021; Bigelow et Canham 2007; Huggett <i>et al.</i> 2007; Gradowski et Thomas 2008; Ouimet <i>et al.</i> 2008, 2017; Minocha <i>et al.</i> 2010; Cleavitt <i>et al.</i> 2011; Long <i>et al.</i> 2011; Duchesne <i>et al.</i> 2013; Halman <i>et al.</i> 2013, 2015; Battles <i>et al.</i> 2014; Marlow et Peart 2014; Long <i>et al.</i> 2015; Momen <i>et al.</i> 2015; Nolet <i>et al.</i> 2015; Collin <i>et al.</i> 2017, 2018; Bognounou <i>et al.</i> 2023
Comparison of the sensitivity of sugar maple and American beech	Long <i>et al.</i> 1997; van Breemen <i>et al.</i> 1997; Arii <i>et al.</i> 2002, 2005; Kobe <i>et al.</i> 2002; Bigelow et Canham 2007; Page et Mitchell 2008; Moore <i>et al.</i> 2008; Park et Yanai 2009; Minocha <i>et al.</i> 2010; Long <i>et al.</i> 2011; Duchesne <i>et al.</i> 2013; Marlow et Peart 2014; Battles <i>et al.</i> 2014; Halman <i>et al.</i> 2015; Nolet <i>et al.</i> 2015; Ouimet <i>et al.</i> 2017; Cleavitt <i>et al.</i> 2018; Lawrence <i>et al.</i> 2018; Bognounou <i>et al.</i> 2023; Tourville <i>et al.</i> 2023
Discussions and literature reviews	Jones et Hendershot 1989; St. Clair <i>et al.</i> 2008; Bal <i>et al.</i> 2015; West <i>et al.</i> 2023

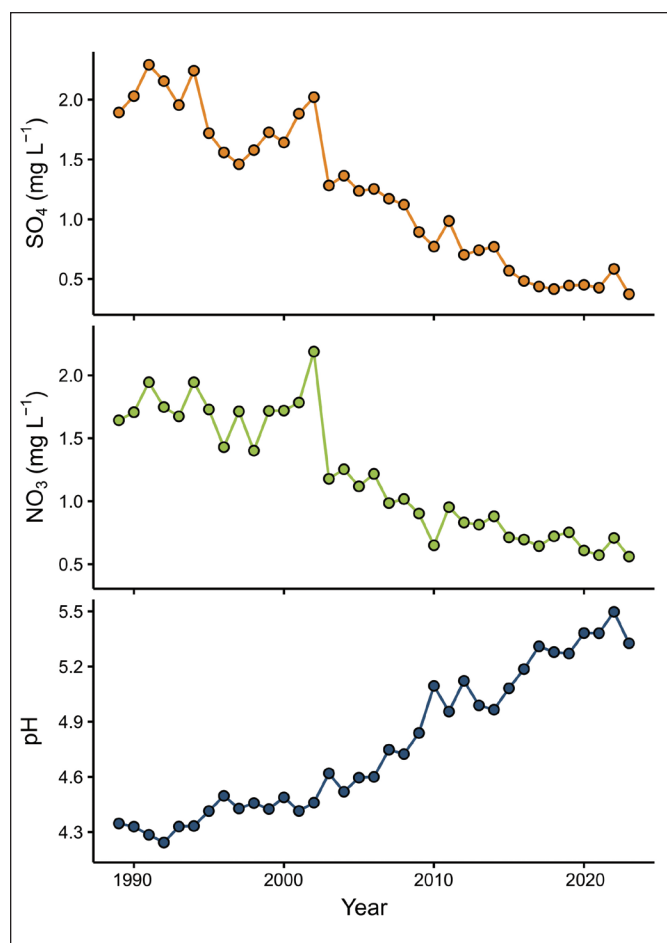


Fig. 2. Evolution in average annual concentrations of sulphates (SO_4) and nitrates (NO_3) and precipitation pH, measured at the Lac Clair station [Station touristique Duchesnay, Québec, Canada] by Québec's Forest Ecosystem Monitoring and Research Network for the period 1989–2023.

number of tapholes in Québec at the time) were lost due to sugar maple dieback, generating financial losses of tens of millions of dollars for maple syrup producers (Jones and Hendershot 1998). In the Adirondacks, deciduous forests damaged by acidity provide only half the potential benefits of forests on moderately to well-buffered soils (i.e., non-acidified soils) due to lost net present value for wood products, maple syrup, carbon sequestration and visual quality (Beier *et al.* 2017). In addition, the impacts of acid rain on the region's forest ecosystems limit the possibilities for sustainable ecosystem management and consequently reduce their potential economic and cultural value for present and future generations (Caputo *et al.* 2016). Among other things, although appropriate silviculture may be able to regenerate sugar maple trees on well-buffered soils, it is very unlikely that new natural maple cohorts will become established on acidified soils (Caputo *et al.* 2016). This suggests that the maple stands currently growing on acidified soils are an ecological legacy. Depending on the harvesting scenarios chosen, sugar maple is therefore likely to be replaced in the medium or longer term by species such as red maple and American beech. Given that poorly-buffered soils will take decades to recover completely from acidification, the change in composition will be a long-lasting legacy of acid rain (Caputo *et al.* 2016).

Tropospheric or ground-level ozone

Tropospheric or ground-level ozone (O_3) is not emitted as a pollutant but is what is referred to as a “secondary” pollutant that forms as a result of photochemical reactions between NO_x , volatile organic compounds (VOCs) and other pollutants present in the air. Triggered by ultraviolet rays, nitrogen dioxide (NO_2) reacts with the oxygen in the air to produce O_3 . However, nitrogen monoxide (NO) reacts quickly with O_3 , temporarily reducing O_3 concentrations. Because of this, O_3 concentrations are usually lower near urban areas due to the presence of traffic, and higher in rural areas and remote forests located far from NO emission points (Ainsworth *et al.* 2012; MELCCFP 2023, 2024).

The transportation sector is the main source of O_3 precursor pollutants, followed by industrial activity, residential heating, energy production and liquid fuel/solvent evaporation (ECCC 2023a). Thanks to the air pollutant emission reduction programs implemented in Canada and the United States, ambient air precursor pollutant concentrations and maximum O_3 values have tended to decline in the Canada-United States border zone since the mid-1990s (ECCC 2023a). In Québec, however, O_3 concentrations have increased in urban areas over the last decade but are stable or slightly lower in rural and forest stations (MELCCFP 2023). Data for Québec show that O_3 concentrations have not exceeded the eight-hour Canadian Ambient Air Quality Standard (CAAQS) of 62 ppb (parts per billion) since 2012–2014 but are very close to that level at numerous air quality measuring stations (MELCCFP 2024). However, there were 327 occurrences of levels exceeding the eight-hour O_3 standard (64 ppb) set out in the provincial *Clean Air Regulation* (CAR) at Québec's 48 air quality monitoring stations in 2021 (MELCCFP 2023). This is the highest number since the CAR came into force in 2012. It is due, among other things, to weather conditions conducive to O_3 accumulations, including high temperatures, that have affected both the northeastern United States and southern portions of Ontario and Québec. It is important to note that the apparent contradiction between compliance with the federal and provincial standards is due to the different statistical forms used. While the provincial standard refers to the average concentrations over an 8-hour period, the Canadian standard refers to the three-year average of the annual fourth highest of the daily maximum 8-hour average ozone concentrations.

O_3 toxicity for trees, especially sugar maple, has long been of concern (Hibben 1969). Generally speaking, hardwood species are more sensitive to high O_3 concentrations than softwood species (Wittig *et al.* 2009; Lee *et al.* 2022). The impacts of high O_3 concentrations for trees include reduced growth and a significant decrease in respiration, foliar surface, Rubisco (a key enzyme in photosynthesis) and chlorophyll, which may have caused photosynthetic capacity to decrease significantly (Wittig *et al.* 2009). O_3 is a powerful oxidant; tissues that are more fragile and less protected by waxes or layers of protective cells are more susceptible to damage, particularly the organs involved in photosynthesis. However, the O_3 action mechanism has yet to be clarified (Dizengremel *et al.* 2001).

For sugar maple, experiments have shown that high O_3 concentrations may reduce growth (Topa *et al.* 2001, 2004; Gaucher *et al.* 2003, 2006; Karnosky *et al.* 2005; King *et al.* 2005); alter structure, speed up leaf senescence and reduce photosynthesis, CO_2 absorption and stomatal conductance (Carlson 1979; Reich and Amundson 1985; Reich *et al.* 1986; Tjoelker *et al.* 1995; Bäck *et al.* 1999; Ainsworth *et al.* 2012); hinder mycorrhiza development (Duckmanton and Widden

1994); reduce biological performance and alter the food preferences of defoliating insects, including the forest tent caterpillar (Fortin *et al.* 1997); and compromise the ability of seedlings to acclimatize to cold conditions (Bertrand *et al.* 1999). Some research has shown that sugar maple seedling response is influenced by the light environment (Tjoelker *et al.* 1993, 1995; Bäck *et al.* 1999; Topa *et al.* 2001, 2004), suggesting that seedlings growing in shade, under the canopy, and shade leaves, are more sensitive to O_3 . However, other studies have suggested that the impact on sugar maple growth, biomass, photosynthesis, stomatal conductance and leaf or seedling senescence becomes weak or null after two to three years of exposure to high O_3 concentrations (Laurence *et al.* 1996; Rebbeck 1996; Rebbeck and Loats 1997; Pell *et al.* 1999) or have concluded that there is no evidence of O_3 being a factor in sugar maple decline (Hibben 1969; Manning 1989).

Sensitivity to high concentrations of O_3 has also been documented for other dominant species in Québec's northern temperate zone (red maple, yellow birch and American beech) (Jensen and Dochinger 1989; Davis and Skelly 1992; Schaub *et al.* 2003; Lee *et al.* 2022). Any generalization of observations concerning forest sensitivity to O_3 are complicated by the fact that it is dependent on tree development stage, microclimate, leaf phenology, in-species variations and other interactive stress factors. Tree sensitivity assessments are based on a limited number of species. In addition, most studies, whether carried out in natural or controlled environments, involve seedlings in non-competitive environments, and seedlings are not necessarily the best subjects for predicting mature tree reactions to O_3 . Lastly, the scope and extent of the response also depend on numerous edaphic and climate-related factors, as well as on exposure duration and dynamics and the number of successive years of exposure (Lefohn *et al.* 1997; Chappelka and Samuelson 1998; Lee *et al.* 2022). Because of this complexity, researchers generally use modelling to assess the impacts of high O_3 concentrations on forest ecosystems (Cailleret *et al.* 2018). Some studies have found that O_3 has only negligible impacts on forest productivity in North America, among other things, because of the preponderant role of natural and anthropogenic disturbances in forest dynamics (Landry *et al.* 2013), and also because of changes to composition and community-level compensatory processes that mitigate the negative impacts at tree level (Wang *et al.* 2016). On the other hand, some studies have shown a decline in productivity due to high O_3 concentrations (Ollinger *et al.* 1997; Felzer *et al.* 2004; Yue and Unger 2014). In these models, however, there is uncertainty as to some of the hypotheses, and it will be necessary to measure the impacts of O_3 on mature forests before knowing whether or not the predictions are accurate (Ollinger *et al.* 1997; Cailleret *et al.* 2018). This lack of information, combined with the complexity of the influential factors and their interactions, are likely to have resulted in these apparently contradictory conclusions about the effects of O_3 on forest productivity (Cailleret *et al.* 2018). The effects of ground-level O_3 on tree growth and forest productivity is nevertheless an issue in the northern temperate zone's deciduous forest.

Trace elements

Anthropogenic emissions of trace elements into the environment have been of concern for decades due to their toxicity, their persistence in the environment and their biological accumulation. Lead (Pb), mercury (Hg) and cadmium (Cd), the documented harmful substances used as Canadian indicators

of environmental sustainability, are all part the list of toxic substances in the *Canadian Environmental Protect Act* (1999) and in the CAR's air quality standards. The main sources of anthropogenic emissions of trace metals into the air include fossil fuels, the mining and metal processing industry and waste incineration, and there are significantly more anthropogenic emissions than natural emissions (Nriagu 1989; Pacyna and Pacyna 2001). Trace elements in the air are mainly concentrated in aerosols (or emitted as gases in the case of Hg), meaning that they can easily be transported over long distances (Simonetti *et al.* 2000).

Like acidifying pollutants and O_3 precursor pollutants, anthropogenic emissions of trace elements have declined considerably in North America in recent decades, thanks to the air pollutant emission reduction programs implemented in Canada and the United States. The removal of Pb from engine gasoline is just one of the measures contained in these programs (Dietrich and Filippelli 2023). In Canada, in the period from 1990 to 2021, emissions of Hg, Pb and Cd were reduced by 90%, 91% and 94%, respectively, (ECCC 2023b). In the case of Pb, lower emission levels have resulted in significantly better air quality (Fig. 3; Sullivan *et al.* 2018) and far fewer deposits (Dietrich and Filippelli 2023). These past trends in Pb emissions can be seen in tree growth rings, especially in sugar maple, where Pb concentrations increased in the first part of the 20th century and then decreased from the period 1970-1980 until today, to reach levels similar to or below those recorded at the beginning of the 20th century (Marty *et al.* 2024). Air quality measurements carried out at nine stations in Québec have shown that trace element detection frequency and measured concentrations are currently much lower at rural or forest stations than at urban stations. The rare cases in which levels exceeded CAR standards were observed at stations located close to known sources, such as factories or highways (MELCCFP 2023; Foucreault 2024).

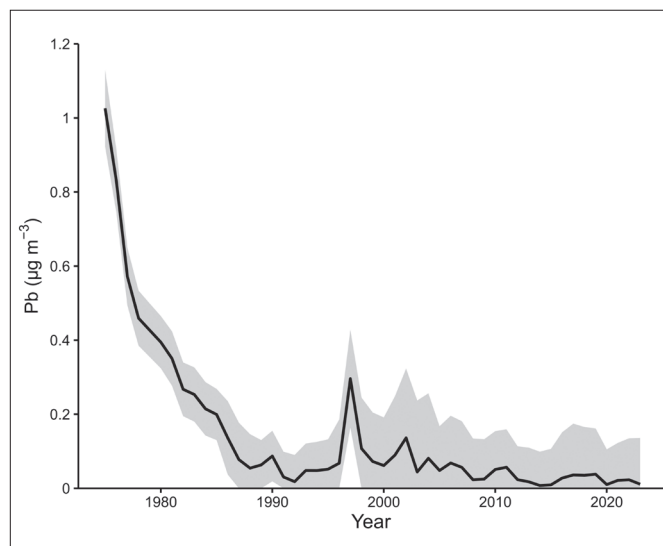


Fig. 3. Evolution of atmospheric lead (Pb) concentrations in Québec from 1975 to 2023. The data show the average annual median Pb concentration in total airborne particles measured at 107 stations by Québec's Air Quality Monitoring Network. The shaded portion shows the 95% confidence interval. Source: Direction de la qualité de l'air et du climat, Ministère de l'Environnement, de la Lutte contre les changements climatiques, de la Faune et des Parcs.

Trace metals enter the forest canopy via dry or wet deposition. Wet deposits include precipitation in the form of rain or snow, as well as fog and cloud droplets. Dry deposits include particle deposits and gas sorption. The portion of air deposits not intercepted by the forest canopy fall to the ground and are added to the elements generated by internal vegetation cycling, i.e., those absorbed by the roots and remobilized via trees (Bergkvist *et al.* 1989). Remobilization includes elements leached from various tree parts and those contained in forest litter and deadwood. Mobility within forest soils varies considerably among metals. Some trace metals form stable complexes with dissolved organic acids transported with percolating water when organic forest litter is mineralized (Bergkvist *et al.* 1989). As they percolate through the upper layer of mineral soil, organic acids become saturated in iron (Fe) and aluminum (Al) and end up being precipitated into the upper portion of the mineral horizons. Therefore, most of the mobilized trace metals accumulate in the upper part of the soil profile (Bergkvist *et al.* 1989; Watmough and Hutchinson 2004). The gradual release and increased concentration of trace metals from the mineral soil in the soil solution are influenced by soil acidity, which enhances their mobility (Watmough *et al.* 2005a). Metal mobility within the soil is also influenced by vegetation type, soil type and environmental variables influencing decomposition of organic matter (Bergkvist *et al.* 1989; Kaste *et al.* 2006).

Release and leaching of toxic trace metals have some major practical implications, in particular with respect to freshwater quality and community water supplies. Generally, trace metals are not particularly mobile and may be retained for years in the organic layer of forest soils and surface mineral horizons (Bergkvist *et al.* 1989; Watmough *et al.* 2005b). In the case of Pb, for example, organic horizons in forests in the northeastern United States can still contain up to 30 kg/ha of Pb derived from engine gasoline, despite legislation passed in the late 1970s which considerably reduced Pb emissions in North America (Kaste *et al.* 2006). Watmough and Hutchinson (2004) noted that roughly 90% of Pb pollution in a forest ecosystem in southern Ontario was situated in the upper layer (0–10 cm) of the mineral soil. Metal concentrations in forest ecosystem mineral horizons in northeastern North America are generally low, however, and are thought to be influenced more by soil properties and the geochemical composition of the rocks that form the soil parent material than by pollution from anthropogenic sources (Watmough 2008). Nevertheless, trace element pollution impacts biological activity and decomposition rates in organic forest litter, and this may negatively affect overall ecosystem productivity (Tyler 1972; Zwoliński 1994).

Pb accumulated in maple stand soils may be an issue for maple syrup production. Producers have made a considerable effort, among other things by modernizing their production equipment, to comply not only with California's drinking water standard, applicable to maple syrup, which allows for a maximum Pb level of 11 µg/L (Government of California 1986, also known as "Proposition 65"), but also with an agreement ratified by a judgment by consent handed down in 2014 by the California courts (Superior Court of the State of California 2014). The main goal of the agreement is to bring maple syrup production equipment up to standard, from tree to consumer, to ensure that there is no risk of Pb contamination in the process. For comparison, the Québec's Maple Syrup Marketing Agreement (PPAQ 2023) considers syrup to be contaminated when Pb levels are above 250 µg/L. The Health Canada threshold is 500 µg/L. Mobility of anthropogenically or naturally sourced Pb in maple stands may

hinder efforts to reduce Pb concentrations in maple syrup. However, following sampling carried out in the spring of 1995 in 90 maple stands across six farming regions of Québec, Pb was found to be present in maple sap (> 0.1 µg/L) in only 18% of cases. Although the maximum detected level was 52 µg/L, concentrations were generally low (average: 1 µg/L; 95th percentile: 5 µg/L), and ten times less, on average, than the California standard (Dumont *et al.* 1996; Renaud 1998). In Connecticut, a similar average level (1.1 µg/L) was measured in maple sap harvested in uncontaminated plastic tanks (Stilwell and Musantec 1996). Robinson *et al.* (1989) obtained higher values ranging from 3 to 2090 µg/L, with an average of 80 µg/L in 27 sugar bushes in Québec, New Brunswick and Nova Scotia. The fact that the sampled sap had in fact come into contact with production equipment may explain these high concentrations (Dumont *et al.* 1996). Given that sap is concentrated roughly 40 times in order to obtain maple syrup, a concentration of 0.275 µg/L would, in theory, be sufficient to obtain 11 µg/L in the syrup if all the Pb in sap were to transfer to the maple syrup. However, a significant amount of the Pb from the sap precipitates out of solution as the sap is processed into syrup, and the solid containing the Pb can be eliminated during filtering (Robinson *et al.* 1989; Dumont *et al.* 1996; Stilwell and Musantec 1996). Pb in maple syrup therefore appears to be mainly of technological origin, probably deriving from tin/Pb welding in maple syrup conditioning systems, storage in galvanized steel tanks, metal pipes and the use of pumps with bronze gear systems to collect and transfer sap (Dumont *et al.* 1996; Stilwell and Musantec 1996; Greenough *et al.* 2010; Mohammed *et al.* 2022). In Québec, however, these contamination risks should have been eliminated with the additional Pb reduction measures recently implemented in the *Maple Syrup Marketing Agreement* (e.g., PPAQ 2023, Appendix N). Exceptionally, for reasons that are as yet unknown, high concentrations of Pb in maple sap may nevertheless still be an issue for some producers (Dumont *et al.* 1996; Renaud 1998).

Trace metal pollution not only causes nutritional deficiencies, it is also harmful to the health of sensitive tree species (Gawel *et al.* 1996). Soil acidification resulting from acid rain affects toxic metal mobility and availability, and also absorption by tree roots and internal circulation of metals within trees and ecosystems. It can therefore have major nutritional and toxicological consequences for trees (Bergkvist *et al.* 1989; Kahle 1993; Watmough 2002; Bilodeau-Gauthier *et al.* 2011). For example, foliar concentrations of metals in sugar maple may be higher in acidic soil conditions but bear no relationship to concentrations in mineral soil (Watmough 2008). This shows that the metal cycle is improved in acidic soil conditions. In sugar maple, manganese (Mn) concentrations in leaves are higher on acidic soils, and these high concentrations are associated with dieback and reduced photosynthesis (see St. Clair *et al.* 2008 for a literature review). Trace metal pollution, interacting with soil acidification through acid rain, is therefore an issue for sugar maple health and productivity. In addition to interactions with acidifying air pollutants, exposure to metals may enhance water stress for trees and cause loss of conductivity due to embolisms (in particular for sugar maple), making them more vulnerable to drought (de Silva *et al.* 2012; Tanentzap and Ryser 2015).

Regarding the impacts of forest management, it has been shown that whole tree cutting in deciduous forests in northeastern North America increases mobility of some trace metals along with leaching from forest soils to surface water (Scott *et al.* 2001). Conversely, phytoextraction of trace metals appears to offer potential for depollution of soils, especially when

fast-growth tree species are used, for example willows or poplars, or species that collect large quantities of metals, such as birch (González-Oreja *et al.* 2008; Munro and Courchesne 2019).

In addition to Pb, Hg ranks among the most concerning of the trace metals. Because it is emitted in gaseous form, it remains in the air for longer and is more mobile than other metals. It can therefore be transported globally and be deposited far from its source. Like Pb, Hg is a toxic pollutant with harmful effects for human health throughout the world (Zhang *et al.* 2021). In addition, it accumulates in food chains, impacts biodiversity and upsets biological balance, thereby endangering ecosystems (Eagles-Smith *et al.* 2016). In boreal forests, Hg preferentially accumulates in the humus rather than in the mineral soil (Lindqvist *et al.*, 1991). As for CO₂, trees can absorb large quantities of atmospheric Hg and act as sinks in the Hg cycle (Obrist *et al.* 2021; Zhou *et al.* 2021). In a changing environment, interactions between climate change, vegetation dynamics and the processes governing the global Hg cycle have become a major environmental issue. In concrete terms, scientists anticipate a 60% decline in the terrestrial Hg sink compared to the current situation, due to reduced stomatal conductance caused by an increase in atmospheric CO₂ (Yuan *et al.* 2024).

Conclusion

In the last few centuries, industrialization in North America has generated air pollutant emissions mostly from combustion of fossil fuels used to produce energy or for transportation. Research has focused mainly on the harmful impacts of acidifying pollutants (i.e., SO₂ and NO_x), pollutants implicated in the formation of ground-level ozone (i.e., NO_x and COV) and trace elements (heavy metals) on forest ecosystems. As awareness grew about the harmful effects of these air pollutants for human health, wildlife and vegetation, ecosystem functions and infrastructures, several national and international agreements aimed at reducing emissions. As a result, emissions and ambient air concentrations of most atmospheric pollutants have declined significantly since the mid-1990s.

Although the situation has improved, air pollutants continue to cause concern and continue to have major harmful effects for forest ecosystems, and in particular for soil fertility and tree physiology. The situation is of particular concern for the sugar maple, a species that is eminently sensitive to exchangeable cation availability, soil acidity and high ozone concentrations. Sugar maple stands growing on acidified soils encounter problems with natural regeneration, and the sugar maple trees that make up these stands tend to exhibit low vigour. As a long-term legacy of acid rain, these stands may well evolve toward different compositions over time. Environmental monitoring of forest ecosystems continues in order to see how these ecosystems are reacting to the lower levels of anthropogenic air pollutant emissions in contemporary North America.

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