



# Soil solution chemistry weak response to long-term N addition points towards a strong resilience of northeastern American forests to past and future N deposition

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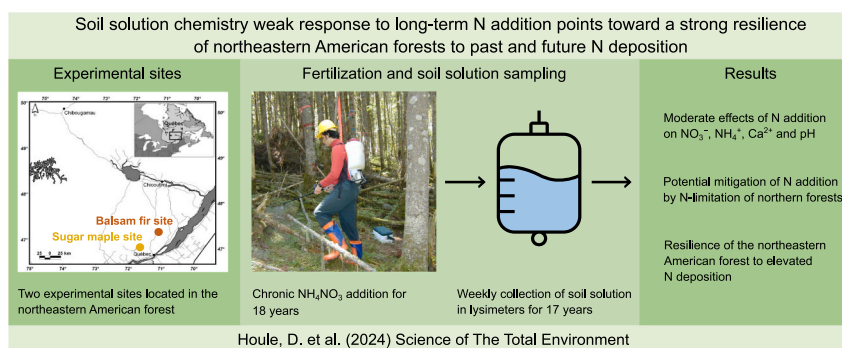
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## HIGHLIGHTS

- Following 18 years of treatment, N addition caused soil solution pH to decrease.
- N addition also induced higher  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  and  $\text{Ca}^{2+}$  concentrations in soil solution.
- Considering its duration and dose, N treatment effects on soil solution are weak.
- The impacts of N addition could be mitigated by N limitation of northern forests.
- Northeastern American forests seem to be resilient to elevated N deposition.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Northern temperate and boreal forests are large biomes playing crucial ecological and environmental roles, such as carbon sequestration. Despite being generally remote, these forests were exposed to anthropogenic nitrogen (N) deposition over the last two centuries and may still experience elevated N deposition as human activities expand towards high latitudes. However, the impacts of long-term high N deposition on these N-limited forest ecosystems remain unclear. For 18 years, we simulated N deposition by chronically adding ammonium nitrate at rates of 3 (LN treatment) and 10 (HN treatment) times the ambient N deposition estimated at the beginning of the experiment at a temperate sugar maple and a boreal balsam fir forest site, both located in northeastern America. LN and HN treatments corresponded respectively to addition of  $26 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  and  $85 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  at the temperate site and  $17 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  and  $57 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  at the boreal site. Between 2002 and 2018, soil solution was collected weekly during summer and concentrations of  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{Ca}^{2+}$  and pH were measured, totalling  $\sim 12,700$ – $13,500$  observations per variable on the study period. N treatments caused soil solution  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  and  $\text{Ca}^{2+}$  concentrations to increase while reducing its pH. However, ion responses manifested through punctual high concentration events (predominantly on the HN plots) that were very rare and leached N quantity

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was extremely low at both sites. Therefore, N addition corresponding to 54 years (LN treatment) and 180 years (HN treatment) of accelerated ambient N deposition had overall small impacts on soil solution chemistry. Our results indicate an important N retention of northeastern American forests and an unexpected strong resilience of their soil solution chemistry to long-term simulated N deposition, potentially explained by the widespread N-limitation in high latitude ecosystems. This finding can help predict the future productivity of N-limited forests and improve forest management strategies in northeastern America.

## 1. Introduction

During the 19th and 20th centuries, worldwide atmospheric deposition of nitrogen (N) considerably increased with the expansion of human activities (Galloway et al., 2004). More recently, since the 2000s, deposition of oxidized forms of N (e.g.,  $\text{NO}_3^-$ ) globally decreased due to emission regulation policies implemented mostly in North America and Europe (Ackerman et al., 2019; Engardt et al., 2017; Li et al., 2016). On the other hand, global atmospheric deposition of reduced N, such as  $\text{NH}_4^+$ , increased with intensity varying among regions. In northeastern Canada,  $\text{NO}_3^-$  deposition significantly decreased since 2000 as well, but  $\text{NH}_4^+$  deposition remained stable (Houle et al., 2015).

Northern temperate and boreal forests of North America, despite being generally remote, were exposed to relatively high N deposition from the past centuries which can still impact these ecosystems through cumulative effects as N is highly retained in forest soils (Templer et al., 2012). High N deposition can strongly affect northern forests productivity by fertilising soils or modifying their chemistry (Yan et al., 2017). Soil chemistry changes can cause foliar nutrient imbalance and potentially be deleterious for understory vegetation and tree growth (Jung et al., 2018; Xu et al., 2021). Simulated N deposition experiments often report N accumulation in forest soils, associated with soil acidification and base cations leaching (e.g.,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ; Tian and Niu, 2015). In temperate and boreal forests, 4–6 years of N addition was also showed to induce higher  $\text{NO}_3^-$  and  $\text{Ca}^{2+}$  concentrations in soil solution which translates to higher N and cations leaching (Jung and Chang, 2012; Geng et al., 2020). These studies also indicate that artificial N addition caused N leaching mainly under the form of  $\text{NO}_3^-$ . However, N leaching rate greatly varies with geographic location as it depends on the forest type and N status (Braakhekke et al., 2017).

Concentrations of inorganic forms of N in soil solution, usually measured with belowground lysimeters, are good indicators of ecosystem N saturation status (Evans et al., 2008; Schmitz et al., 2019). Unlike soil sampling, soil solution collection is non-destructive and provides a dynamic view of long-term changes in soil nutrient concentrations and pH. Lysimeters enable the collection of many observations through time which facilitates accurately assessing soil chemistry status and helps better estimate soil nutrient availability and leaching. Despite significant advantages of assessing soil solution chemistry, only a few N fertilization experiments include long-term data of soil solution inorganic N and major cations concentrations in northern temperate or boreal forests, as they usually focus on dissolved organic carbon (DOC) or are located in tropical forests (see Cusack et al., 2016; Guo et al., 2020; Liu et al., 2017b). Moreover, N addition experiments often occurred in locations already receiving relatively high ambient N deposition, but northern forests, especially in the boreal zone, are generally N-limited (Du et al., 2020) and may have a different response to elevated N deposition compared to more polluted forests. Studies using more realistic experimental designs, especially relative to N addition dose and chemical form, are also needed to draw more robust conclusions on the influence of elevated N deposition on forest ecosystems. For instance, exogenous N applied at rates of 20–30 times the ambient N deposition is extremely unlikely to match past or future N deposition but is often performed in fertilization studies. Therefore, work on the effects of elevated N deposition on N-limited forest soils using more realistic experimental duration and treatment doses are lacking and the long-term impacts of higher N deposition on northern

forests soil solution chemistry remain unclear. However, assessing the consequences of elevated N deposition on forest soil solution nutrient status is essential to plan sustainable forest management as soil solution chemistry is linked to vegetation growth and health.

The goal of this work was to assess the long-term impacts of elevated N deposition on the soil solution chemistry in two major bioclimatic domains of the northeastern American forest (i.e., the sugar maple-yellow birch domain located in the temperate zone and the balsam fir-white birch domain located in the boreal zone) using a more realistic experimental design (regular treatment applications approaching natural wet deposition chemical form and stoichiometry). For 18 years, we simulated N deposition by chronically adding ammonium nitrate during the growing season at the surface of a temperate and a boreal experimental forest site and measured weekly concentrations of  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{Ca}^{2+}$ , and pH in soil solution collected in lysimeters placed at depths of 30 cm and 60 cm. At the boreal forest site, previous work showed that, despite punctual increases,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  concentrations in soil solution were not significantly higher after 3–8 years of N addition compared to the control (Houle and Moore, 2008, 2019). However, the temperate forest site began to show some signs of N saturation (i.e., higher  $\text{NO}_3^-$  leaching) after 3 years of N addition at rate of  $85 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  (Moore and Houle, 2009). Based on these previous results, we predicted that (i) the long duration of the treatment would finally induce N saturation and increased N leaching, detectable with higher soil solution  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and  $\text{Ca}^{2+}$  concentrations as well as lower pH, and that (ii) the effects of N treatments would be less pronounced at the boreal forest site than at the temperate forest site, as the latter already indicates potential N saturation.

## 2. Methods

### 2.1. Experimental sites description

In this study, we used two experimental forest sites located in the province of Quebec, Canada. The most southern site ( $46^{\circ}57'N$   $71^{\circ}40'W$ ) is in the northern temperate forest where the dominant tree species is the sugar maple (*Acer saccharum* Marsh.) associated with yellow birch (*Betula alleghaniensis* Britt.) and American beech (*Fagus grandifolia* Ehrh.), while the other site ( $47^{\circ}17'N$   $71^{\circ}14'W$ ) is in the boreal forest and is dominated by balsam fir (*Abies balsamea* [L.] Mill.) trees. Hereafter, study sites will be referred as per their dominating tree species (i.e., sugar maple or balsam fir).

The understory vegetation mainly comprises yew shrubs (*Taxus baccata* [L.]) at the sugar maple site and feather mosses (e.g., *Pleurozium schreberi* [Willd. ex Brid.] Mitt.) and ericaceous shrubs (e.g., *Rhododendron groenlandicum* [Oeder] Kron & Judd) at the balsam fir site. Soils are classified as Orthic Humo-Ferric Podzols, as well as Typic Haplorthod at the balsam fir site (Soil Classification Working Group, 1998). Humus is a mor-moder type at the sugar maple site and mor type at the balsam fir site. Organic FH horizons are acid with a pH of 3.72 at the sugar maple site and 4.08 at the balsam fir site.

Ambient N deposition is estimated to be  $8.5 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  at the sugar maple site and  $5.7 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  at the balsam fir site (Ste-Marie and Houle, 2006), which is considered relatively low compared to other N fertilization experimental forest sites in the U.S. and Europe (Magill et al., 2004).

Additional information about the characteristics of the study sites is

presented in Table S1.

## 2.2. Fertilization experimental design

The N fertilization procedure was described in detail in Renaudin et al. (2023). Briefly, 9 experimental units of  $15 \times 15$  m (at the sugar maple site) or  $10 \times 10$  m (at the balsam fir site), separated by at least 10–15 m, were delimited on each site (Fig. S1). Control (C), low N addition (LN), and high N addition (HN) treatments were randomly assigned to blocks of three experimental unit replicates. LN treatment corresponded to a 3-fold increase of ambient N deposition received by each site at the beginning of the experiment (i.e., applications of  $26 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  at the sugar maple site and  $17 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  at the balsam fir site), while HN treatment corresponded to a 10-fold increase (i.e., applications of  $85 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  at the sugar maple site and  $57 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  at the balsam fir site). LN and HN treatments consisted of applications of a  $\text{NH}_4\text{NO}_3$  solution supplied as four passes using a backpack sprayer (Solo, Newport News, VA, USA). Treatment application started in June 2001 and finished in October 2018, and was performed once a month between June and October.

## 2.3. Soil solution sampling and chemical analyses

In October 2000, two tension lysimeters per experimental unit (Soil moisture Equipment Corp., Model 1911) were placed at depths of 30 and 60 cm, respectively. For 17 years (2002–2018), soil solution was collected each week from May to November and stored at  $4^\circ\text{C}$  until analysis.

In total, from 2002 to 2018, we collected 7279 samples at the sugar maple site and 6238 samples at the balsam fir site.

After collection, soil solution samples were all filtered at  $0.45 \mu\text{m}$  (Nucleopore) and pH was measured directly with a probe.  $\text{NH}_4^+$  and  $\text{NO}_3^-$  concentrations were respectively measured by colorimetry (Technicon AA2) and ion chromatography, while  $\text{Ca}^{2+}$  concentration was measured by plasma emission spectrometry.

## 2.4. Statistical analyses and leached $\text{NO}_3\text{-N}$ calculations

All statistical analyses were performed with R Studio v.4.3.1. (R Core Team, 2023) and figures were built with ggplot2 package (Wickham, 2016). Data normality was assessed visually by building Q-Q plots and by using the Anderson-Darling test (nortest package v.1.0–4; Gross and Ligges, 2015).

To evaluate linear time trends (2002–2018) of soil solution  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{Ca}^{2+}$  concentrations, and pH, for each sampling site, lysimeter depth, and N treatment, Mann-Kendall tests were performed (Kendall package v.2.2.1; McLeod, 2022) and Sen slopes were calculated (zyp package v.0.10–1.1; Bronaugh and Schoeneberg, 2023). For  $\text{NO}_3^-$  only, cumulative frequencies of every concentration observation were calculated using the cumsum function of the R base package.

Differences of soil solution  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{Ca}^{2+}$  concentrations, and pH between sampling sites, lysimeter depths, and N treatments were assessed with Kruskal-Wallis tests followed by post-hoc Dunn's tests (adjusted with the Benjamini-Hochberg method), as data was not normally distributed.

Finally, to assess the effects of sampling site, lysimeter depth, treatment, and time since beginning of treatment (i.e., sampling date) on soil solution  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{Ca}^{2+}$  concentrations and pH, we performed regression type random forest models (randomforest package v.4.7–1.1; Liaw and Wiener, 2002) with mtry (i.e., number of predictors randomly picked at each split) set as 2 and ntree (i.e., number of trees to build in the model) set as 5000. We ran 6 iterations of each model, combined the iterations into a single model, and classified the predictors according to their variable importance score.

The quantity of leached  $\text{NO}_3\text{-N}$  for each site and N treatment was estimated by calculating monthly  $\text{NO}_3^-$  concentrations in soil solution

collected at 60 cm depth (i.e., outside the rooting zone) and measuring monthly bulk precipitation from 2002 to 2018. Bulk precipitation was measured using collectors placed on each site and monthly volumes were corrected for estimated evapotranspiration ( $\sim 30\%$  of total precipitation amounts) to estimate leached water fluxes. Fluxes of leached  $\text{NO}_3\text{-N}$  were then calculated by multiplying  $\text{NO}_3\text{-N}$  concentrations to water fluxes and were subsequently converted to  $\text{kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ .

## 3. Results

### 3.1. Time trends of forest soil solution $\text{NO}_3^-$ , $\text{NH}_4^+$ , $\text{Ca}^{2+}$ concentrations, and pH

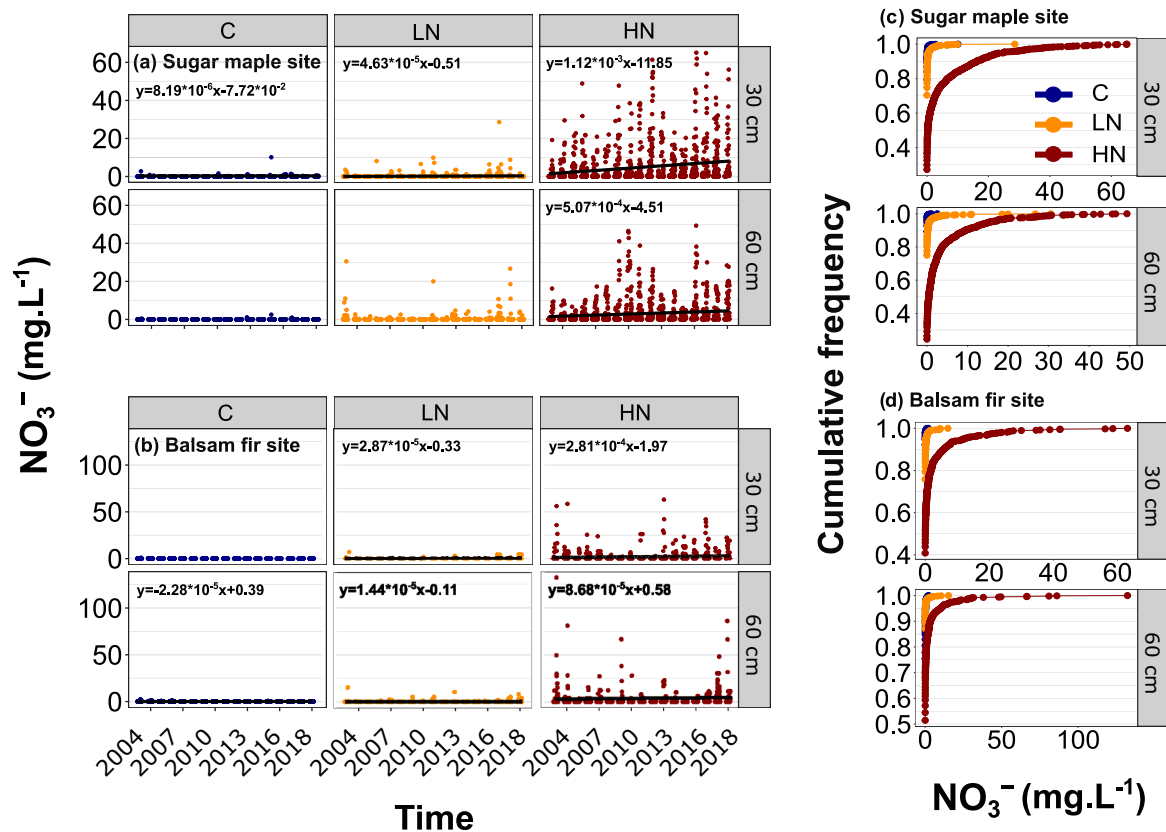
Between 2002 and 2018, soil solution chemistry fluctuated across the two experimental sites.  $\text{NO}_3^-$  and  $\text{NH}_4^+$  concentrations were generally low (means  $< 1.5 \text{ mg}\cdot\text{L}^{-1}$ ) in control units and underwent minor changes over time ( $-0.08 < \tau < 0.08$ ; Table S2). At the sugar maple site (30 cm depth),  $\text{NO}_3^-$  concentration measured in controls increased while it decreased at the balsam fir site (60 cm depth) (Fig. 1; Table S2). On the contrary, control  $\text{NH}_4^+$  concentration at 30 cm increased at the balsam fir site (Fig. 2; Table S2). Linear time trends of  $\text{Ca}^{2+}$  concentration (Fig. S2) and pH (Fig. 3) in control units were similar across all study sites and respectively decreased and increased over 17 years at both 30 and 60 cm depths (Table S2).

N addition had significant effects on soil solution elemental time trends. At the sugar maple site, both Mann-Kendall tests and linear regressions revealed that  $\text{NO}_3^-$  concentration significantly increased over time in LN and HN treatments at 30 cm ( $\tau = 0.12$  and  $0.18$ , respectively) and in HN treatment at 60 cm ( $\tau = 0.16$ ). Similarly, soil solution  $\text{NO}_3^-$  concentration increased in LN and HN treatments at 30 cm at the balsam fir site. However, N addition had less pronounced effects on temporal variations of soil solution  $\text{NH}_4^+$  concentration ( $-0.10 < \tau < 0.10$ ). For the three treatments,  $\text{NH}_4^+$  concentration did not vary significantly over time at the sugar maple site. At the balsam fir site,  $\text{NH}_4^+$  concentration in HN treatment decreased at 30 cm while increasing in LN treatment at 60 cm. For all treatments, sites, and lysimeter depths,  $\text{Ca}^{2+}$  concentration showed the strongest significant decreasing trends during the 17 years of experiment ( $-0.72 < \tau < -0.26$ ). However, tau and Sen's slopes were respectively 1.3 and 1.1 times higher for N treatments than for the control, suggesting higher average  $\text{Ca}^{2+}$  concentrations under N addition. The effects of N addition on soil solution pH differed among experimental sites. At the balsam fir site, pH increased between 2002 and 2018 for all treatments and lysimeter depths ( $0.12 < \tau < 0.33$ ). Soil solution pH followed a similar pattern at the sugar maple site, except for HN treatment units where no significant trend was detected.

### 3.2. Effects of N addition on forest soil solution chemistry

Soil elemental concentrations and pH globally differed among experimental sites, lysimeter depths, and N treatments (Table S3). The sugar maple site displayed the highest  $\text{NO}_3^-$  concentration (i.e., mean =  $1.38 \text{ mg}\cdot\text{L}^{-1}$ ; Fig. 4) and the lowest pH (i.e., mean = 4.83; Fig. S4) while the balsam fir site displayed the highest  $\text{Ca}^{2+}$  concentration (i.e., mean =  $0.42 \text{ mg}\cdot\text{L}^{-1}$ ; Fig. S3) and the lowest  $\text{NH}_4^+$  concentration (i.e., mean =  $0.023 \text{ mg}\cdot\text{L}^{-1}$ ; Fig. 5). Mean  $\text{NO}_3^-$  concentration was 1.5 times higher at 30 cm than at 60 cm at the sugar maple site while there was no significant difference with depth at the balsam fir site. At the two experimental sites,  $\text{NH}_4^+$  concentration was higher at 30 cm than at 60 cm whereas  $\text{Ca}^{2+}$  and pH were both higher at 60 cm than at 30 cm.

N treatment also had contrasting impacts on soil solution chemistry. At all sites,  $\text{NO}_3^-$  concentration was significantly higher for HN treatment than for LN and control (Fig. 4). At the sugar maple site, mean  $\text{NO}_3^-$  concentration was 4 times higher in LN and 120 times higher in HN compared to control at 30 cm, and 11 times higher in LN and 150 times higher in HN compared to control at 60 cm. However, while N addition



**Fig. 1.** Temporal variations (2002–2018) and cumulative frequencies of soil solution  $\text{NO}_3^-$  concentration collected at 30 and 60 cm depths at the sugar maple (a, c) and balsam fir (b, d) sites submitted to three treatments (control: C, low N addition: LN, and high N addition: HN). Dot colors correspond to treatments and solid black lines represent linear regressions. Temporal trends statistical significances were assessed with Mann-Kendall tests and linear regressions were only represented when tests were significant.

impacts on  $\text{NO}_3^-$  concentration were more noticeable at the sugar maple site than at the balsam fir site (Fig. 1a, b), overall treatment effects were mild at both study sites, as most  $\text{NO}_3^-$  observations (86–94 % for LN treatment and 37–67 % for HN treatment) were below  $0.10 \text{ mg}\cdot\text{L}^{-1}$  (Fig. 1c, d).

In agreement with this result, we determined that the quantity of leached  $\text{NO}_3^-$ -N was extremely low on LN and HN experimental units at both study sites. Leached  $\text{NO}_3^-$ -N corresponded of losses of  $0.087 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  on LN treatment units and  $1.16 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  on HN treatment units at the sugar maple site, as well as  $0.029 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  on LN treatment units and  $0.55 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  on HN treatment units at the balsam fir site. At the sugar maple site, leached  $\text{NO}_3^-$ -N corresponded to 0.33–1.36 % of N treatment application rates and 0.25–1.24 % of the total N load received on N treatment units (i.e., N provided by treatments + ambient N deposition). At the balsam fir site, leached  $\text{NO}_3^-$ -N corresponded to 0.17–0.96 % of N treatment application rates and 0.12–0.87 % of the total N load received on N treatment units.

Following the pattern observed for  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  concentration was significantly higher in HN compared to LN and control at the balsam fir site (Fig. 5). With respective means of  $0.06 \text{ mg}\cdot\text{L}^{-1}$  and  $0.008 \text{ mg}\cdot\text{L}^{-1}$ ,  $\text{NH}_4^+$  concentration was 7.5 times higher in HN and 1.05 times higher in LN than in control. However,  $\text{NH}_4^+$  concentration did not significantly differ between the three treatments at the sugar maple site. At both study sites, LN treatment displayed the lowest soil solution  $\text{Ca}^{2+}$  concentrations (Fig. S3). Compared to the control,  $\text{Ca}^{2+}$  concentration in HN treatment was significantly higher at sugar maple site but lower at the balsam fir site. LN treatment had contrasted effects on soil solution pH across sites as it led to significantly higher pH at the sugar maple site but significantly lower pH at the balsam fir site, compared to the two other treatments (Fig. S4). However, pH was 0.07 and 0.12 units lower in HN

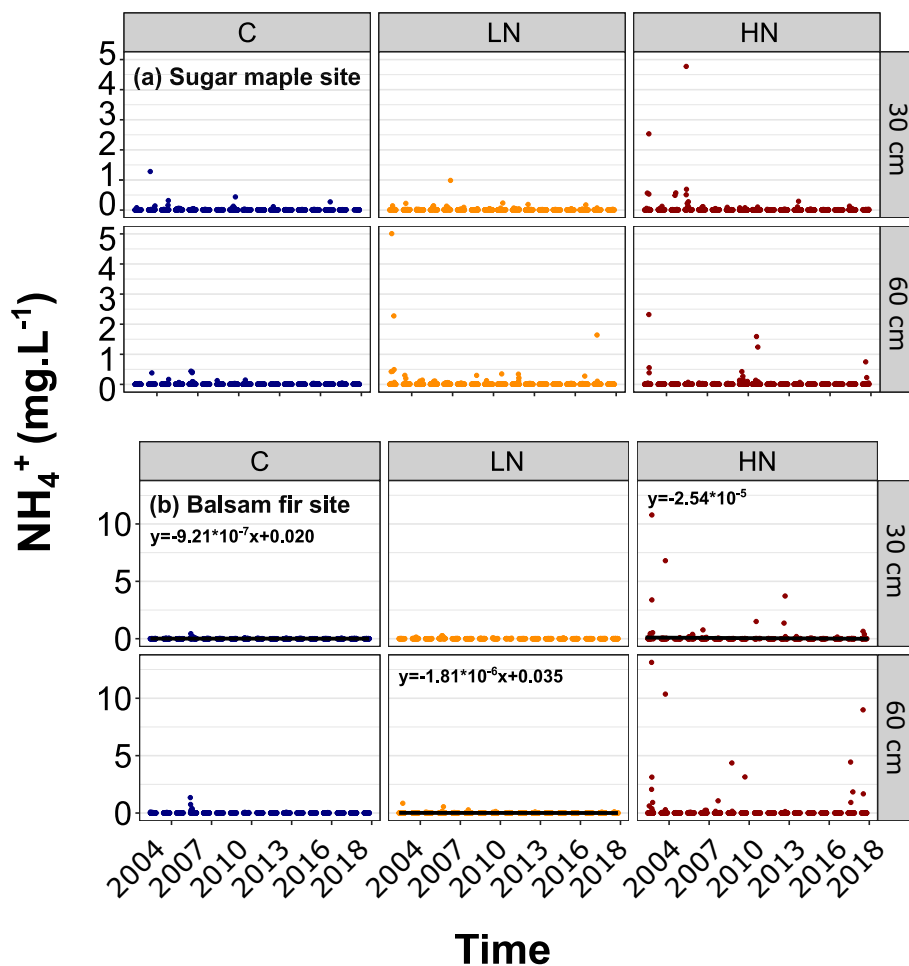
than in control at the sugar maple and balsam fir site, respectively.

Random forest models revealed that the sampling site, lysimeter depth, and time (i.e., sampling date) were all significant predictors of soil solution  $\text{NO}_3^-$ ,  $\text{Ca}^{2+}$ , and pH (Fig. S5). These predictors all together respectively explained 21 %, 35 %, and 41 % of  $\text{NO}_3^-$ ,  $\text{Ca}^{2+}$ , and pH variations. On the other hand, only treatment and time were significant predictors of  $\text{NH}_4^+$  concentration, the model explaining 3 % of its variation. Treatment was the most important predictor of soil solution  $\text{NO}_3^-$  and  $\text{NH}_4^+$  and second most important predictor of  $\text{Ca}^{2+}$  but, despite being significant, it was the least important predictor of pH.

## 4. Discussion

### 4.1. Impacts of 18 years of N addition on soil solution chemistry

High concentration of inorganic N in soil solution (particularly  $\text{NO}_3^-$ ) is generally seen as an indicator of soil N saturation (i.e., N availability exceeding total biological demand) (Aber et al., 2003). At both study sites,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  concentrations were low and stable in the soil solution of control units at 30 and 60 cm depths (Figs. 1 and 2; Table S2). Overall, our data suggest that, on the study period, experimental sites' control units were not N-saturated according to their relatively high organic soil C:N ratios (i.e., C:N = 23 at the sugar maple site and C:N = 28 at the balsam fir site; Table S1). This is supported by previous reports of important N retention at the tree canopy (Houle et al., 2015) and at the watershed scale (Duchesne and Houle, 2006, 2008). HN treatment had a noticeable impact on  $\text{NO}_3^-$  concentration which significantly increased in soil solution and, in agreement with our hypothesis, the impacts of N addition were much stronger at the sugar maple site than at the balsam fir site, potentially because the sugar maple site is located more south



**Fig. 2.** Temporal variations (2002–2018) of soil solution  $\text{NH}_4^+$  concentration collected at 30 and 60 cm depths at the sugar maple (a) and balsam fir (b) sites submitted to three treatments (control: C, low N addition: LN, and high N addition: HN). Dot colors correspond to treatments and solid black lines represent linear regressions. Temporal trends statistical significances were assessed with Mann-Kendall tests and linear regressions were only represented when tests were significant.

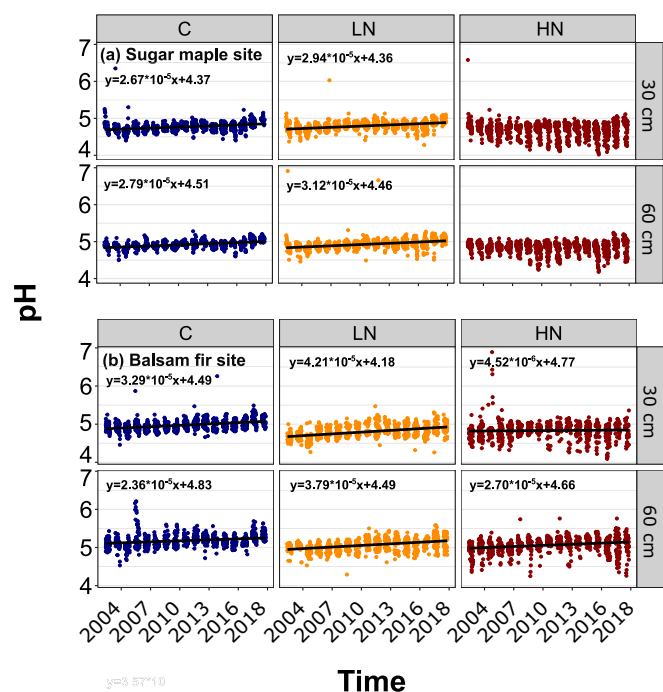
and receives about 1.5 times more ambient N deposition than the balsam fir site.

However, despite visible effects of N treatments, the amount of leached  $\text{NO}_3^-$  was impressively low and corresponded to <1.5 % of the added N and total N load at the two study sites, indicating extreme N retention by these forest ecosystems even under high rates of N inputs. This is in line with previous soil and soil solution analyses performed in 2003 and 2008 showing that 3 and 8 years of N addition did not induce N saturation at the experimental sites (Houle and Moore, 2008, 2019). A meta-analysis that included 65 forest ecosystems across Europe (primarily from the mid-1980s) showed that leaching of  $\text{NO}_3^-$  became important at N inputs above  $25 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  (Dise and Wright, 1995). In northern and western Europe, Erisman and de Vries (2000) suggested that  $\text{NO}_3^-$  leaching started to occur when throughfall inputs exceeded  $10 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ , but more specifically in soils with  $\text{C:N} < 25$ . Based on results of N addition experiments mainly localized in Europe, Xie et al. (2024) also proposed a similar N saturation threshold of  $5\text{--}10 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  in boreal forests and  $5\text{--}46 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  in temperate forests. Given the factorial design of our experiment (i.e., treatments being scaled on site's ambient total N load), equivalent treatment units do not receive the same amount of N between sites. However, if we consider the total received N load (i.e., natural N deposition plus the experimentally added N), all the treated units received  $>10 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  but more noticeable  $\text{NO}_3^-$  leaching was observed only in the HN units, which received  $\sim 99 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  at the sugar maple site and  $\sim 66 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  at the balsam fir site, questioning the  $5\text{--}10$

$\text{kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  threshold mentioned earlier. We suggest that a universal N saturation threshold does not exist and that bioclimatic domain, forest and soil type, and past N deposition (being linked with site's N status and limitation) are important drivers of  $\text{NO}_3^-$  leaching response.

On the other hand, overall  $\text{NH}_4^+$  concentrations remained low at all sites and treatments compared to  $\text{NO}_3^-$ , and mostly varied with geographic location and lysimeter depth.  $\text{NH}_4^+$  is generally taken up preferentially by micro-organisms and trees (see Geisseler et al., 2010; Lupi et al., 2013; Liu et al., 2017a) and can also be transformed into  $\text{NO}_3^-$  by nitrifying bacteria at the pH range found in our studied soils (see Table S1; Norton and Stark, 2011). Moreover, because of its diffusion coefficient in soil,  $\text{NO}_3^-$  is also more mobile and more prone to leaching via soil solution compared to  $\text{NH}_4^+$  (Owen and Jones, 2001). All together, these biogeochemical mechanisms may explain the low concentrations of  $\text{NH}_4^+$  in soil solution and the weak effects of N addition treatments, especially compared to  $\text{NO}_3^-$ .

Since the beginning of the 2000s, an overall recovery of lakes and streams acid-base status from acid rains was observed in Europe and North America (de Wit et al., 2021; Houle et al., 2020; Marty et al., 2021), which could have resulted from pH increase of the soil solution feeding surface water. Not many results exist for soil solution but, in good agreement with aquatic systems' response, pH increased in control and LN units of the sugar maple and balsam fir sites over our study period (Fig. 3). However, HN treatment visibly countered soil solution pH recovery, leading to no or decreasing pH trends and much higher  $\text{Ca}^{2+}$  leaching (Fig. S2; Table S2), which could ultimately impact tree



**Fig. 3.** Temporal variations (2002–2018) of soil solution pH collected at 30 and 60 cm depths at the sugar maple (a) and balsam fir (b) sites submitted to three treatments (control: C, low N addition: LN, and high N addition: HN). Dot colors correspond to treatments and solid black lines represent linear regressions. Temporal trends statistical significances were assessed with Mann-Kendall tests and linear regressions were only represented when tests were significant.

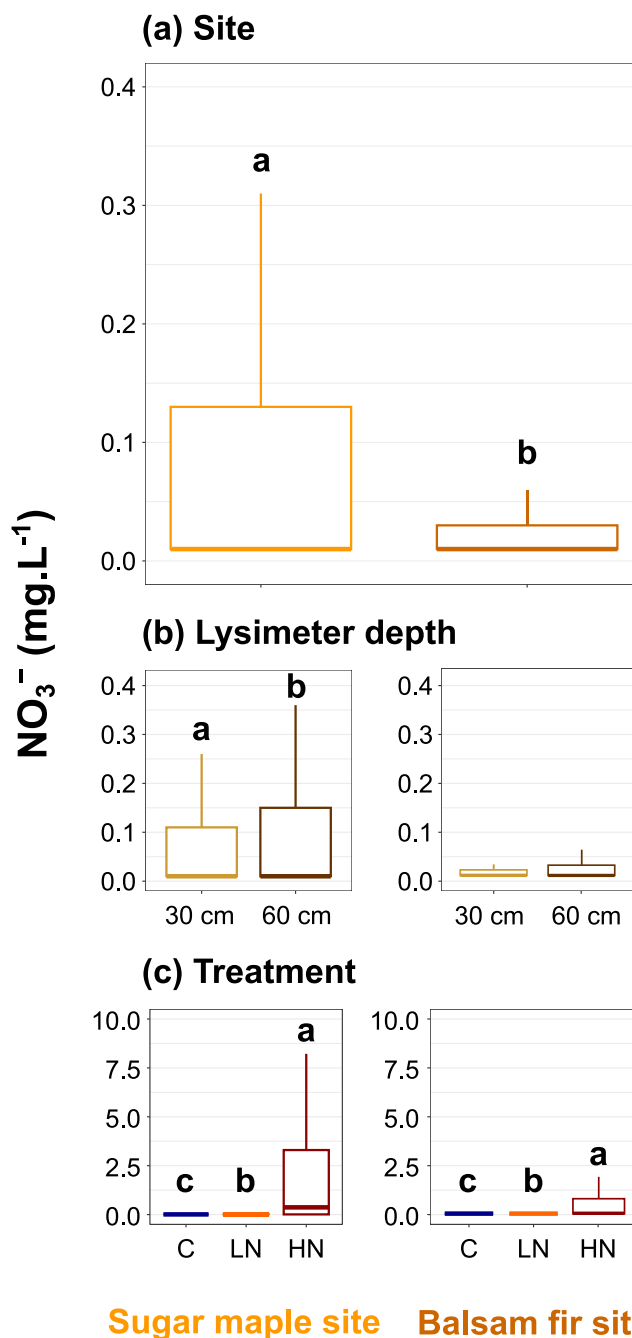
nutrition especially at the sugar maple site (see Moore and Houle, 2023).

#### 4.2. Resilience of northeastern American forests to past and future N deposition

N treatments had noticeable impacts on soil solution chemistry. These impacts were more pronounced for  $\text{NO}_3^-$  concentrations, which were 120–150 times higher in HN treatment compared to the control at the sugar maple site. However, soil solution ion responses manifested through punctual high concentration events (especially in HN treatment) rather than consistent and more permanent concentration increases, and N retention rates were extremely high at both sites. This shows that N addition had globally weak to moderate effects on soil solution, especially when considering the duration and the application dose used for this experiment.

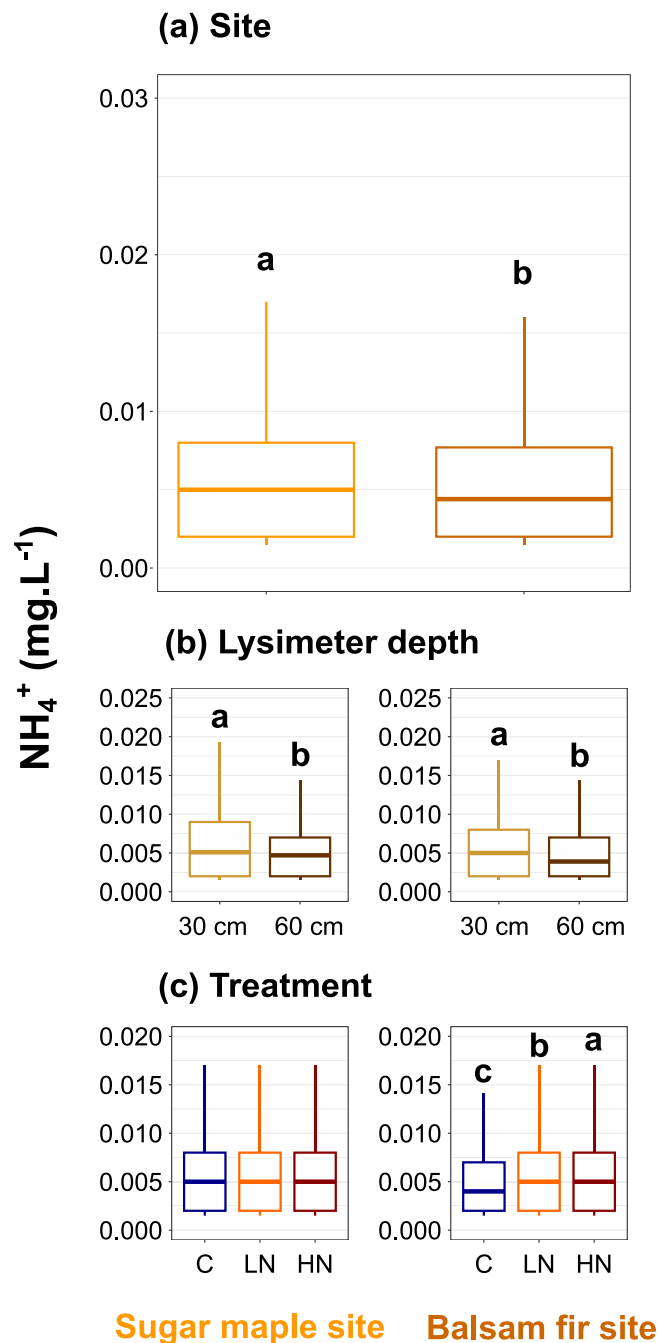
Recently, similar resilience to N treatments was observed for organic and mineral soil chemistry at the study sites (Houle et al., 2024). All together, these elements point towards a strong resilience of northeastern American forest ecosystems to elevated N deposition. Similar observations were made in Sweden by Högberg et al. (2014), who showed that a coniferous boreal forest receiving high amounts of added N still returned to its N limited state 20 years after the end of N addition. Because N limitation is widespread in northern forests (Du et al., 2020), especially in boreal forests, extra N is accumulated in soil organic horizons and rapidly taken up by vegetation (Moore and Houle, 2023; Houle et al., 2024) and microbes (e.g., mycorrhizal fungi; Högberg et al., 2017). Therefore, we suggest that the observed resilience of high-latitude forests to high N deposition is due, at least partially, to its N limitation status causing high ecosystem N retention.

In our study, LN treatment represents a relatively realistic rate of N deposition, amounting to four times the ambient N deposition in the years preceding the beginning of the experiment, while HN treatment represents an extreme N deposition rate. Past N deposition rates at our



**Fig. 4.**  $\text{NO}_3^-$  concentration in soil solution collected between 2002 and 2018 at the sugar maple and balsam fir forest sites, at 30 and 60 cm depths, and submitted to three treatments (control: C, low N addition: LN, and high N addition: HN). For each boxplot, the box corresponds to the data interquartile range (from first to third quartile), the bold horizontal line in the box represents the median, the lower whisker extends from the first quartile to the minimum and the upper whisker extends from the third quartile to the maximum. Letters show significantly different concentrations between sites, lysimeter depths, and treatments.

experimental sites likely never reached LN treatment application rates (see Houle et al., 2015) and certainly not for a period as long as in our study. N deposition will also most likely not reach LN treatment level given current  $\text{NO}_3^-$  deposition decrease in North America (Gilliam et al., 2019). Considering the near absence of LN treatment effects discussed earlier, we suggest that soil solution chemistry of northern temperate and boreal forests in North America was not or barely affected by N deposition in the preceding decades, which also implies minimal



**Fig. 5.**  $\text{NH}_4^+$  concentration in soil solution collected between 2002 and 2018 at the sugar maple and balsam fir forest sites, at 30 and 60 cm depths, and submitted to three treatments (control: C, low N addition: LN, and high N addition: HN). For each boxplot, the box corresponds to the data interquartile range (from first to third quartile), the bold horizontal line in the box represents the median, the lower whisker extends from the first quartile to the minimum and the upper whisker extends from the third quartile to the maximum. Letters show significantly different concentrations between sites, lysimeter depths, and treatments.

impacts on vegetation N nutrition.

## 5. Conclusions

Experimental N addition has been extensively used to study atmospheric N deposition impacts on forest ecosystems. Most of these impacts have been characterised through measurements of vegetation growth,

foliar chemistry, and soil chemistry, but long-term surveys of soil solution elemental composition are very scarce, especially in northern remote forests. Here, we showed that, in agreement with our first hypothesis, artificial N addition induced higher  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  and  $\text{Ca}^{2+}$  leaching and soil solution acidification. Except for  $\text{NH}_4^+$ , these effects were more visible at the sugar maple site than at the balsam fir site, in agreement with our second hypothesis. Overall, the most noticeable impact of elevated N was an increase of soil solution  $\text{NO}_3^-$  concentration, especially under HN treatment at the sugar maple site, which was associated with lower pH visibly counteracting pH recovery from past acid rains. However,  $\text{NO}_3^-$  response was mainly driven by a greater frequency of high concentration events as most of the observations remained low ( $< 0.10 \text{ mg.L}^{-1}$ ), and the amount of  $\text{NO}_3^-$ -N leached was extremely low at both sites ( $< 1.5\%$  of total N load).

In summary, we show in this study that 18 years of chronic N addition corresponding to 54 years (LN treatment) and 180 years (HN treatment) of accelerated ambient N deposition had moderate impacts on soil solution chemistry of northeastern American temperate and boreal forests. Other extensive work performed on the same sites since 2000, which gathered information relative to soil and foliar chemistry, soil microbiome, and tree growth (Houle and Moore, 2008, 2019; Moore and Houle, 2023; Renaudin et al., 2023; Houle et al., 2024), also all indicate a resilience of northeastern American forests to elevated N deposition, possibly explained by the strong N-limitation widespread in high-latitude ecosystems and causing high N uptake and retention by vegetation and microbes. This suggests that northern temperate and boreal forests of North America won't be significantly affected by potential future high N deposition, especially given the current decreases in  $\text{NO}_3^-$  deposition detected at our experimental sites (Houle et al., 2015) and globally in eastern North America (Gilliam et al., 2019). This finding can help predict the future productivity of N-limited forests and implement more sustainable forest management strategies in northeastern Canada and U.S, where forestry represents an important economic sector. However, further work is needed to evaluate the impacts of other anthropogenic contaminants (e.g., organic compounds, metals) on high-latitude forest ecosystems, especially with human activities developing progressively further in northern regions.

## CRedit authorship contribution statement

**Daniel Houle:** Writing – review & editing, Resources, Methodology, Investigation, Funding acquisition, Data curation. **Marie Renaudin:** Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Formal analysis, Data curation. **Louis Duchesne:** Writing – review & editing, Resources, Investigation, Data curation. **Jean-David Moore:** Writing – review & editing, Resources, Methodology, Conceptualization. **Apolline Benoit:** Writing – review & editing, Software.

## Declaration of competing interest

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

## Data availability

The data that support the findings of this study is available upon request.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2024.174387>.

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