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Canopy openness rather than tree species determines atmospheric deposition into forests

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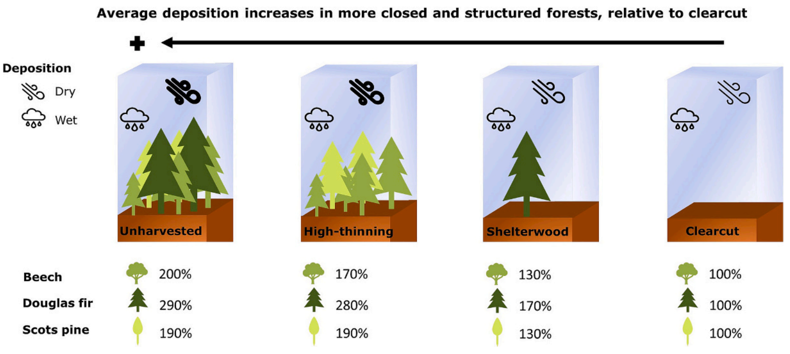
HIGHLIGHTS

- Canopy openness rather than tree species determines atmospheric deposition into forests.
- Acidifying sulphur and nitrogen deposition are ca. 50 % higher in forests than in open sites.
- Base cation deposition into forests is ca. 100 % higher than in open sites.
- Net acid inputs can be modified by forest management.
- Seasonal variation in total deposition is hardly affected by the leaf phenology of the species.

GRAPHICAL ABSTRACT



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ABSTRACT

Atmospheric nutrient deposition plays a crucial role in supplying nutrients to forests on poor soils, making it a key factor in maintaining nutrient stocks and forest productivity. We compared total atmospheric deposition in production forests of European beech (*Fagus sylvatica*), Douglas fir (*Pseudotsuga menziesii*), and Scots pine (*Pinus sylvestris*) by measuring bulk deposition and throughfall while accounting for canopy exchange. We assessed the differences in total deposition resulting from forest management practices such as high-thinning, shelterwood and clearcutting, on forest structure for both macronutrients and micronutrients in areas exposed to high nutrient deposition.

We demonstrate that total nutrient deposition is highest in Douglas fir stands and lowest in Scots pine stands, primarily due to differences in dry deposition. The total deposition in forest exceeds national estimates because nutrient deposition in forests was higher than expected compared to more open areas. Canopy openness strongly influences the total deposition of all nutrients except phosphorus (P). Total deposition increases by a factor of 2.2 when moving from clearcuts to closed forests, with the extent of the increase varying among tree species and nutrients. Additionally, total deposition fluctuates between seasons, revealing clear seasonal patterns in both throughfall and canopy exchange.

Our results suggest that the effective capture of potentially growth-limiting nutrients (such as K, Ca, Mg and Mn) in closed and thinned production forests significantly contributes to the resilience of forests on nutrient-

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poor, acidified soils. Our results underscore the importance of considering the effects of harvest intensity on canopy openness and forest structure and, to a lesser extent, tree species when calculating nutrient inputs from atmospheric deposition.

1. Introduction

Forest productivity depends on nutrient availability in the soil, which is mainly determined by nutrient inputs from deposition, weathering, and nutrient retention capacity. In European forests, atmospheric deposition represents a significant source of nutrient input into the ecosystem (Van Langenhove et al., 2020), but varies among forests depending on geographic region, tree species and stand properties such as tree height and canopy openness (Kowalska et al., 2016; Zhang et al., 2022). Nevertheless, the impact of stand properties on deposition is neglected in many forest nutrient budget studies (Akselsson et al., 2007; Aherne et al., 2012; Phillips and Watmough, 2012; Iwald et al., 2013), potentially leading to over- or underestimations of nutrient input via atmospheric deposition of up to 50 % (Draaijers et al., 1997b). To reduce uncertainties in forest nutrient balance and forest growth, better understanding of the possible influence of tree species and stand properties on nutrient input via atmospheric deposition is essential.

Nutrient input via atmospheric deposition consists of nutrients in precipitation (wet deposition) and gases and airborne particles deposited within tree crowns (dry deposition) (Lovett and Reiners, 1986; Balestrini et al., 2007). In forests, atmospheric deposition is often measured as throughfall deposition, which includes wet deposition that passes through the canopy (Draaijers et al., 1996a; Thimonier, 1998). Within the canopy, nutrient concentrations in rainfall change due to uptake by or leaching from the canopy (i.e. canopy exchange) and wash-off of airborne particles and gasses deposited on tree crowns (i.e. dry deposition) (Lovett and Lindberg, 1984; Lovett and Lindberg, 1992; Staelens et al., 2008b; Adriaenssens et al., 2012). Additionally, part of the intercepted precipitation reaches the forest floor via stemflow, which varies significantly among tree species (Silva and Rodríguez, 2001; Su et al., 2019; Houcai et al., 2021). Total deposition on the forest floor thus comprises throughfall and stemflow, corrected for canopy exchange. In open areas, it consists only of bulk deposition as stem and canopy effects are excluded.

Throughfall deposition in forests is influenced by geographic location, tree species and forest structure. The geographic location determines the dry deposition load, as the dry deposition flux relates to distance from the sea - especially for potassium (K), calcium (Ca) and magnesium (Mg) (Draaijers et al., 1997b; Balestrini et al., 2007)- and to anthropogenic pollution sources, particularly for ammonium (NH_4), nitrate (NO_3) and sulphate (SO_4) (Nordén, 1991; Draaijers et al., 1997b). Furthermore, dry deposition and, therefore, throughfall deposition are influenced by canopy structure and roughness, including tree height, canopy architecture, and openness, which relate to basal area and stand age (Lovett and Lindberg, 1984; Nordén, 1991; Aboal et al., 2000; Erisman and Draaijers, 2003; Herrmann et al., 2006; Klopatek et al., 2006; De Schrijver et al., 2008; Griffith et al., 2015; Zhang et al., 2022). Tree harvest modifies such forest structural properties by creating a more open canopy and modifying air flow, and generally results in significant reductions in throughfall deposition: for example, harvesting 15 % of stem volume led to 20 % reduction in throughfall deposition in Norway spruce stands in southern Germany (Göttlein et al., 2023), while harvesting 40 % and 100 % of stem volume resulted in 45 % to 60 % reductions, respectively (Bäumler and Zech, 1997; Göttlein et al., 2023). Canopy exchange is often considered to be related to canopy cover and nutrient content of foliage, which varies among tree species (Herrmann et al., 2006; André et al., 2008; Talkner et al., 2010), but it can also differ within species growing on different soil types (Nordén, 1991). These complex interactions affecting throughfall should be accounted for in order to reduce the current uncertainly in nutrient input estimates in forests in local sites and at regional scale.

Nutrient inputs from total deposition fluctuate strongly over the growing season (Herrmann et al., 2006; Klopatek et al., 2006; Su et al., 2019), and even within the period of leaf fall (Garten Jr et al., 1988; Adriaenssens et al., 2012b). For deciduous species, nutrient input through deposition is greatly reduced in winter compared to the growing season. In contrast, for evergreen (coniferous) species, absolute dry deposition can increase during winter (André et al., 2008; Adriaenssens et al., 2012b). The extent to which seasonality affects total annual deposition flux, particularly in relation to canopy openness, remains unclear. Increased canopy openness due to different harvest intensities may alter seasonal fluctuations in total deposition and therefore modify species differences, but such impacts are poorly studied.

Aside from the limited research on how forest canopy openness and harvest intensity affect seasonal and annual atmospheric nutrient deposition, most studies have focused solely on macronutrients, overlooking micronutrients or trace elements like manganese (Mn), copper (Cu), iron (Fe), and zinc (Zn). Although some studies have addressed deposition of micronutrients for temperate Asian and North America forests (Hou et al., 2005; Landre et al., 2010; Zhang et al., 2021; Richardson et al., 2024), such data for European temperate forests remain scarce (Ukonmaanaho et al., 2001). Micronutrients are essential for tree growth, enzymatic functions, and metabolic processes such as photosynthesis and respiration (Broadley et al., 2012). They also influence forest health and productivity, enhancing resilience against diseases, pests, and environmental stressors (Weinmann et al., 2023; Lim-Hing et al., 2024). Deficiencies in both macro- and micronutrients or imbalances can lead to poor growth, reduced vitality (Vogel and Jokela, 2011; Ivanov et al., 2022), and disruptions in nutrient cycling and ecosystem function. Given their role in tree vitality and wood quality, assessing the total deposition of both macro- and micronutrients is required for understanding the implication of nutrient dynamics for forest.

To address these knowledge gaps, we quantified the effects of harvest-induced changes in forest canopy openness on seasonal and annual atmospheric nutrient deposition, and examined how these effects vary by tree species. We focused on three major tree species in the Netherlands: one deciduous (European beech, *Fagus sylvatica*) and two evergreen conifers (Douglas fir, *Pseudotsuga menziesii* and Scots pine, *Pinus sylvestris*). In this region, nutrient deposition mainly originates from agriculture (NH_3), industry (SO_2, NO_x), traffic (NO_x), and sea salt aerosols (Ca, Mg and K) (Vet et al., 2014). We hypothesized that nutrient deposition increases after high-thinning due to greater canopy roughness, which enhances dry deposition, but decreases at higher harvest intensities because of reduced canopy roughness and lower dry deposition. We further hypothesized that deposition shows stronger seasonal effects in deciduous trees (beech) compared to evergreen conifers. The results are discussed in the context of their implications for forest nutrient budgets, productivity, and resilience. We measured deposition in five experimental forest plots per tree species, each with four subplots varying in harvest intensity, over a full year. Total deposition was estimated by adjusting for canopy exchange (using Na as a proxy). Nutrients analyzed included macronutrients (N, S, Ca, K, Mg, P) and micronutrients (Mn, Cu, Fe, Zn), while the harvesting intensities included high-thinning, shelterwood, clearcut and a non-harvested control.

2. Methods

2.1. Study sites and measurements

2.1.1. Study sites and field placement of deposition samplers

Atmospheric deposition was measured in monoculture stands of European beech (*Fagus sylvatica*), Douglas fir (*Pseudotsuga menziesii*) and Scots pine (*Pinus sylvestris*) across five regions in the Netherlands (Fig. S1). All 15 stands are located in a temperate maritime climate, with a 20-year mean annual temperature of 10.5 °C (range 10.3–10.8 °C) and mean annual rainfall of 790 mm (range 700–870 mm) (KNMI, 2021). Monthly temperature and precipitation data for the regions are provided in Table S1. The forest consisted of relatively homogeneous, even-aged, single-tree species stands, planted between 50 and 120 years ago. The dominant species in each stand accounted for over 80 % of the total crown cover and basal area. All plots had been managed according to typical silvicultural practices in the Netherlands. Initial thinning followed a low-thinning regime (removal of suppressed trees), while high-thinning (removal of trees competing with future crop trees) was applied over the past three decades. The selected stands are located on acidic sandy soils, classified as Albic or Entic Podzols, or Dystric Cambisols, within texture classes ranging from fine sand to loamy medium sand. The soils were characterized by high nitrogen stocks and very low base saturation (de Jong et al., 2023; Vos et al., 2023a; Vos et al., 2023b).

In each of the 15 stands, four 0.25 ha subplots were established and randomly assigned one of four harvest intensity treatments: high-thinning (~20 % of the basal area removed), shelterwood (80 % removed), clearcut (100 % removed), or unharvested control (0 % removed), resulting in 60 subplots in total. In March 2020, seven deposition samplers were installed in each subplot (28 samplers per forest stand) using a new method recently tested by Vos et al. (2024). The combined collection area of the samplers per plot totaled 2016 cm^2, exceeding the 2000 cm^2 threshold for reliable measurements (Bleeker et al., 2003). The samplers consisted of a polyethylene funnel mounted to a resin column, connected by a PVC hose to a polypropylene water reservoir. The funnel had a surface of 288 cm^2 (including half the rim). Both the funnel and resin column were chemically resistant and not vulnerable to UV-light or low temperature damage. A wire coupling with a 0.51 mm mesh was used to connect the resin column to the funnel and the hose-tail. Before field installation, the funnel, resin column, and wire couplings were cleaned by submerging them into a 0.2 M HCl solution for 3 h, followed by a 15-hour immersion in demineralized water, which was continuously refreshed. The compartments were then dried in a clean room and stored in clean plastic bags.

Sampler placement in the high-thinning, shelterwood and unharvested control plots was based on canopy cover, assessed using drone photographs and a digital surface model. A stratified random point procedure was used to randomly place samplers, dividing each plot into seven equal grids, with one sampler assigned to each grid. Samplers were positioned either under the canopy or in exposed areas, according to the canopy cover of each treatment plot (Table S2). In both the control and high-thinning plots, all samplers were treated as throughfall collectors. Samplers in the shelterwood plots that were not under the canopy, as well as those in the clearcut, were assumed to collect only forest gap bulk deposition.

2.1.2. Preparation of resin columns

Throughfall and forest gap bulk deposition were measured using the ion exchange resin (IER) method, where funnels are connected to resin columns that capture cations and anions from the solution passing through them. This method was thoroughly tested and published in Vos et al. (2024). A total of 430 resin columns were prepared in the week before installing the deposition samplers. Every three months, new columns were prepared one week before field replacement, which occurred in June, September and December 2020, with columns remaining operational to March 2021.

The resin columns, made of chemically resistant HDPE, had a volume of 15.7 mL, an inner diameter of 12.4 mm, and a length of 130 mm. After drying, the columns were washed three times with demineralized water before being filled with IER (Amberlite IRN 150, H+ and OH— forms). The IER was pre-washed with 8 L of demineralized water to remove particles and odor, and liquids were drained using a vacuum pump. Each resin column was filled with 10 g of resin resulting in a total exchange capacity of 0.011 and 0.009 mol l^{-1} for the cation and anion bed respectively. Tests of the IER's adsorption capacity and recovery efficiency (Amberlite IRN 150 H+ and OH— form), as described in Vos et al. (2024), showed 100 % adsorption for Ca, Cu, Fe, K, Mg, Mn, P, S, Zn and NO_3 and >96 % for P and Na. Elemental recovery was nearly 100 % for NH_4 and NO_3 using KCl, and high (83–93 %) recovery rates for Ca, Cu, Fe, K, Mg, Mn and S were obtained using HCl as an extractant.

2.1.3. Deposition measurements

Atmospheric nutrient inputs from forest gap bulk deposition (in a small forest clearing, not under a canopy) and throughfall were calculated by multiplying the water fluxes outside and below the canopy with the corresponding nutrient concentrations. Water fluxes and nutrient concentrations in throughfall were measured in high-thinning, shelterwood and the control, while forest gap bulk deposition was measured in the clearcut and shelterwood treatments. Deposition measurements cover the period from March 21, 2020, to March 21, 2021.

Rainfall volume (mL) per funnel was recorded monthly, and contamination (e.g., bird feces, vandalism) was noted. Funnels were positioned horizontally to prevent capture capacity reduction, and contaminated funnels were detached, rinsed with demineralized water, and reattached to resin columns. Contaminated columns were excluded from analysis. Resin columns were replaced every three months to capture the seasonal deposition. Additionally, blank resin columns were installed in sun-exposed and shaded locations (under the canopy) to correct for internal release of nutrients, with these blanks set up at one forest site. Upon collection, all resin columns that had been in the field for three months were sealed and stored in dark boxes at 4 °C until further extraction.

Extraction of the resin followed the procedure in Vos et al. (2024). Resin columns contaminated with bird feces were excluded. Resin from field columns and lab blanks (for lab contamination correction), was dried to a constant weight at 28 °C, and the weight (in g) per column was recorded. Subsamples were taken, their weight (in g) recorded, and used for 2 M KCl extraction and analyzed for NH_4, NO_3 and NO_2, total N and PO_4 concentrations using a Segmented Flow Analyzer (SFA type 4000, Skalar Analytical B.V., the Netherlands). Additionally, 3.5 M HCl extractions were analyzed for S, Ca, K, Mg, P, Mn, Cu, Fe and Zn concentration using ICP-AES (Thermo-Scientific iCAP 6500 DUO, USA).

2.1.4. Corrections for sample contamination

Nutrient input for each field column was calculated by multiplying the extracted concentrations (mg L^{-1}) of the KCl and HCl extraction from each resin subsample by the total resin weight in the corresponding column. Background contamination was corrected by subtracting concentrations from field and lab blanks. For samplers in forest gaps, the sunlight-exposed field blank was used, for those under the canopy, the shaded blank was applied. Corrected concentrations per funnel were scaled to kg ha^{-1} based on the funnel surface area. Each sample was checked for contamination, and values outside the 95 % confidence interval were removed. To do so, the data were normalized, and confidence intervals were calculated as the mean ± 2 standard deviations for each nutrient per treatment. In total, 23 % of the dataset was excluded due to bird feces contamination, and an additional 6 % was removed based on the statistical check. This left 1200 valid observations over the full year of sampling. Missing values were imputed using the R package *MICE* (multiple interpolations) based on Monte-Carlo simulations (Buuren and Groothuis-Oudshoorn, 2011).

2.2. Calculation of total nutrient deposition

2.2.1. Total deposition calculation

To calculate total deposition (kg ha^{-1}), corrections were applied to account for canopy exchange for samplers beneath the forest canopy. The total deposition of nutrients (X_{td}, kg ha^{-1}) – including NH_4, NO_3, Ca, K, Mg, Mn, Cu, Fe and Zn - was calculated as the sum of measured throughfall (X_{tf}, kg ha^{-1}) and calculated canopy exchange (X_{ce}, kg ha^{-1}), according to:

$$X_{td} = X_{tf} \pm X_{ce} \tag{1}$$

2.2.2. Canopy exchange of base cations and micronutrients

Canopy exchange fluxes were calculated for nitrogen (NH_4, NO_3), base cations (Ca, K, Mg), and micronutrients (Mn, Cu, Fe, Zn). Canopy exchange for SO_4^{2-} and PO_4^{2-} was assumed to be negligible, meaning total deposition was estimated as the throughfall. The estimation of canopy exchange for base cations and micronutrients relied on two key assumptions: (i) Na does not interact with the forest canopy (inert tracer) and (ii) the ratio of total deposition to bulk deposition is similar for Ca, K, Mg, Mn, Cu, Fe, Zn and Na. However, this second assumption is not always valid in coastal areas (Baloutes. Greece, pers. comm.). Canopy exchange of base cations and micronutrients was calculated by multiplying their bulk deposition by the ratio of Na input from throughfall, to Na input in forest gap bulk deposition, following Ulrich (1983):

$$X_{ce} = X_{tf} - \left(\frac{Na_{tf}}{Na_{bd}} * X_{bd}\right) \tag{2}$$

where X_{ce} is the canopy exchange of base cations (Ca, K, Mg) and micronutrients (Mn, Fe, Zn, Cu) in kg ha^{-1} $season^{-1}$ and X_{tf}, and X_{bd} represent throughfall deposition and forest gap bulk deposition, respectively, in kg ha^{-1} $season^{-1}$. This approach slightly extends the canopy budget model developed by Ulrich (1983) and further refined by multiple studies (Bredemeier, 1988; Draaijers and Erisman, 1995; De Vries et al., 1999; De Vries et al., 2001) to include canopy exchange of Mn, Cu, Fe and Zn (Rea et al., 2001; Gandois et al., 2010).

2.2.3. Canopy exchange of NH_4 and NO_3

Canopy exchange of NH_4 was calculated as a fraction of the base cation canopy exchange, based on NH_4 and H^+ interacting with the forest canopy through exchange with base cations (Roelofs et al., 1985; Draaijers et al., 1997a). We assumed the total canopy uptake of H^+ (H_{ce}) and NH_4^+ (NH_{4ce}) equals the total canopy leaching of base cations (BC_{ce}), adjusted for weak acid leaching (WA_{ce}) (Van der Maas et al., 1991; Draaijers and Erisman, 1995):

$$NH_{4ce} = BC_{ce} - WA_{ce} - H_{ce} \tag{3}$$

There are three methods to estimate weak acid (WA) concentration: (i) summing HCO_3 (from pH and assumed atmospheric CO_2 pressure) and $RCOO^-$ (from DOC), (ii) using measured alkalinity adjusted for pH, or (iii) calculating the difference between the concentrations of cations (H^+ and NH_4^+, Ca^{2+}, K^+, Mg^{2+} and Na^+) and strong acid anions (SO_4^{2-}, NO_3^- and Cl^-) (De Vries et al., 1999; De Vries et al., 2001). However, since pH, DOC and alkalinity could not be measured using IER and Cl^- was excluded from the analysis, we assumed that NH_4 exchange is $^1/_3$ of base cation leaching. This assumption is based on De Vries et al. (1999) and De Vries et al. (2001) who reported that H^+ uptake, NH_4^+ uptake, and WA leaching are all roughly $^1/_3$ of base cation canopy leaching. Finally, NO_3 canopy exchange was calculated as total N canopy exchange minus NH_4 uptake (NH_{4ce}). The total N canopy exchange was calculated by accounting for the contribution of NH_4 and NO_3 to total N input by throughfall, according to (De Vries et al., 2001; Adriaenssens et al., 2011):

$$NO_{3ce} = \left(NH_{4ce} * \left(\frac{NH_{4tf} * xNH_4 + NO_{3tf}}{NH_{4tf} * xNH_4}\right)\right) - NH_{4ce} \tag{4}$$

In which xNH_4 is a correction factor, assumed to be 5, indicating that canopy uptake of NH_4 is much higher than that of NO_3.

2.2.4. Bulk deposition

The forest gap bulk deposition measured in this study underestimated true bulk deposition due to lower precipitation in the forest clearings compared to open fields (Table S3). To estimate true bulk deposition at each site, we adjusted the forest gap bulk deposition using rainfall data from nearby weather stations, assuming nutrient concentrations in both were equal.

2.3. Statistical analysis

All statistical analysis were performed in R version 4.1.0. Two-way ANOVA tests were used to compare total seasonal deposition and net acid input across harvest intensities and species. These analyses used linear mixed-effect models from the R package *nlme*, with regions as a random structure (Pinheiro et al., 2017). Seasonality was assessed using one-way ANOVAs for each element, treatment, and species. Skewed data were log-transformed to meet normality and homogeneity assumptions. When necessary, VarComb and VarIdent variance structures were applied to account for different variances between factor levels (Zuur et al., 2009). Tukey's post-hoc (HSD) test was used, based on the linear mixed-effects models via the *emmeans* package, to test for differences among seasons, harvest intensities and species (Lenth et al., 2019).

To assess whether harvest intensity and tree species significantly explained variations in seasonal and total deposition, throughfall, and canopy exchange of different nutrients, we performed a partial Redundancy Analysis (p-RDA). This analysis quantified the variance explained by harvest intensity, seasons, and species. A fourth p-RDA was used to identify the main factors driving total annual deposition. Prior to p-RDA, all data were log-transformed to meet linearity assumptions, and the analysis was conducted using the *vegan* package (Oksanen et al., 2022).

3. Results

3.1. Annual water fluxes and stand deposition

Average gap bulk precipitation from April 2020 to March 2021 in clearcut and shelterwood treatments for beech, Douglas fir, and Scots pine was 580 mm (±29 s.e.), 580 mm (±40 s.e.) and 610 mm (±36 s.e.) respectively. Precipitation data from 21 nearby weather stations averaged 736 mm (±17 s.e.) for the same period (Fig. S1, Table S3), which was ≥100 mm higher than the gap bulk precipitation indicating edge effects. Annual interception in unharvested stands, calculated as the difference between throughfall and precipitation from nearby weather stations, was highest in Douglas fir stands (310 mm ± 17 s.e.) followed by Scots pine (260 mm ± 14 s.e.) and beech (270 mm ± 20 s.e.). Throughfall and interception estimates varied across stands, season and harvest treatments (Table S3).

The nutrient content of throughfall deposition was highest in Douglas fir (except for P), intermediate in Scots pine, and lowest in beech (Table 1). Annual canopy exchange – defined as nutrient uptake or release by foliage - was negative for NH_4, NO_3, Ca, Cu, Fe, and Zn, indicating net uptake of these nutrients (Table 1). In contrast, K, Mg (except in Scots pine), and Mn leached from the canopy. Canopy leaching of K accounted for approximately 65 % of total deposition in both beech and Scots pine and around 50 % in Douglas fir. For Mn, canopy leaching comprised 71 % of throughfall in beech, and 48 % and 68 % for Douglas fir and Scots pine, respectively (Table 1). Total deposition of N compounds ranged from ca 23 kg ha^{-1} yr^{-1} in beech to about 27 kg ha^{-1} yr^{-1} in Scots pine and 36 kg ha^{-1} yr^{-1} in Douglas fir,

Table 1
The means and standard errors of the annual throughfall, canopy exchange, total deposition and bulk deposition (clearcut bulk deposition corrected for the open field rainfall) (all in kg ha^{-1} yr^{-1}) and the ratio total deposition/bulk deposition of macro (NH_4, NO_3, S, Ca, K, Mg and P) and micronutrients (Zn, Mn, Fe and Cu) in control stands (no harvest) of beech (BE), Douglas fir (DG) and Scots pine (SP). In addition, data are shown for the tracer Na. Canopy exchange of S and P is assumed to be negligible (Section 2.2.3). Data of the high-thinning and shelterwood are given in Tables S4 and S5.

		NH_4	NO_3	S	Ca	K	Mg	P	Mn	Cu	Fe	Zn	Na
Throughfall	BE	13 ± 1.0	4.2 ± 0.66	5.5 ± 0.36	8.4 ± 1.1	28 ± 1.4	2.6 ± 0.22	0.89 ± 0.083	0.24 ± 0.047	0.016 ± 0.0018	0.13 ± 0.0075	0.11 ± 0.024	16 ± 1.6
	DG	24 ± 3.3	9.0 ± 1.4	12 ± 3.1	9.9 ± 1.6	29 ± 0.92	4.7 ± 0.54	0.57 ± 0.12	0.31 ± 0.048	0.021 ± 0.0035	0.17 ± 0.027	0.62 ± 0.47	31 ± 4.9
	SP	18 ± 0.80	5.6 ± 0.15	6.1 ± 1.1	7.1 ± 0.7	19 ± 1.6	2.5 ± 0.13	0.49 ± 0.062	0.17 ± 0.018	0.014 ± 0.0021	0.13 ± 0.01	0.3 ± 0.18	19 ± 1.7
Canopy exchange	BE	−5.6 ± 0.29	−0.40 ± 0.028		−3.0 ± 1.1	20 ± 1.3	0.15 ± 0.088		0.12 ± 0.044	−0.0074 ± 0.0017	−0.10 ± 0.012	−0.072 ± 0.025	
	DG	−3.2 ± 1.1	−0.17 ± 0.10		−4.0 ± 1.5	14 ± 2.2	0.015 ± 0.28		0.067 ± 0.027	−0.015 ± 0.0042	−0.18 ± 0.043	−0.31 ± 0.25	
	SP	−3.0 ± 0.75	−0.17 ± 0.038		−3.7 ± 0.63	13 ± 1.8	−0.29 ± 0.067		0.050 ± 0.013	−0.0091 ± 0.0022	−0.15 ± 0.035	−0.19 ± 0.15	
Total deposition	BE	18 ± 0.90	4.6 ± 0.66	5.5 ± 0.36	11 ± 1.2	8.8 ± 1.0	2.5 ± 0.21	0.89 ± 0.083	0.11 ± 0.0089	0.023 ± 0.0031	0.23 ± 0.015	0.18 ± 0.041	16 ± 1.6
	DG	27 ± 2.8	9.2 ± 1.3	12 ± 3.1	14 ± 2.6	15 ± 2.8	4.7 ± 0.78	0.57 ± 0.12	0.24 ± 0.043	0.036 ± 0.0074	0.35 ± 0.068	0.92 ± 0.72	31 ± 4.9
	SP	21 ± 1.5	5.8 ± 0.18	6.1 ± 1.1	11 ± 0.97	6.5 ± 0.27	2.8 ± 0.14	0.49 ± 0.062	0.12 ± 0.012	0.023 ± 0.0041	0.28 ± 0.041	0.49 ± 0.33	19 ± 1.7
Bulk deposition	BE	11 ± 1.7	3.2 ± 0.27	4.2 ± 0.27	7.8 ± 0.68	4.2 ± 0.38	1.8 ± 0.17	1.0 ± 0.18	0.070 ± 0.0067	0.016 ± 0.0016	0.16 ± 0.0091	0.099 ± 0.014	11 ± 1.1
	DG	8.9 ± 0.37	3.2 ± 0.15	5.0 ± 0.53	6.1 ± 0.69	5.1 ± 0.94	2.0 ± 0.15	0.59 ± 0.061	0.094 ± 0.011	0.015 ± 0.0020	0.17 ± 0.026	0.39 ± 0.30	13 ± 1.4
	SP	11 ± 0.92	3.6 ± 0.15	4.4 ± 0.44	6.5 ± 0.63	3.3 ± 0.34	1.7 ± 0.20	0.68 ± 0.12	0.061 ± 0.0036	0.015 ± 0.0018	0.18 ± 0.018	0.24 ± 0.17	12 ± 1.5
Ratio	BE	1.6	1.4	1.3	1.4	2.1	1.4	0.89	1.6	1.4	1.4	1.8	1.5
	DG	3.0	2.9	2.4	2.3	2.9	2.4	1.0	2.6	2.4	2.1	2.4	2.4
	SP	1.9	1.6	1.4	1.7	2.0	1.6	0.72	2.0	1.5	1.6	2.0	1.6

representing 1.3 to 2.5 times higher values compared to bulk deposition (Table 1). The highest ratios of total deposition to bulk deposition were found for base cations, S and Mn: base cations exhibited deposition levels 2 to 3 times greater than bulk deposition, with a threefold increase observed for K in Douglas fir.

3.2. Impacts of tree species and harvest intensity on total annual deposition

Total annual deposition ranged from 0.02 kg ha^{-1} yr^{-1} (Cu) to 36 kg ha^{-1} yr^{-1} (N) across nutrients (Fig. 1). Deposition levels were higher in

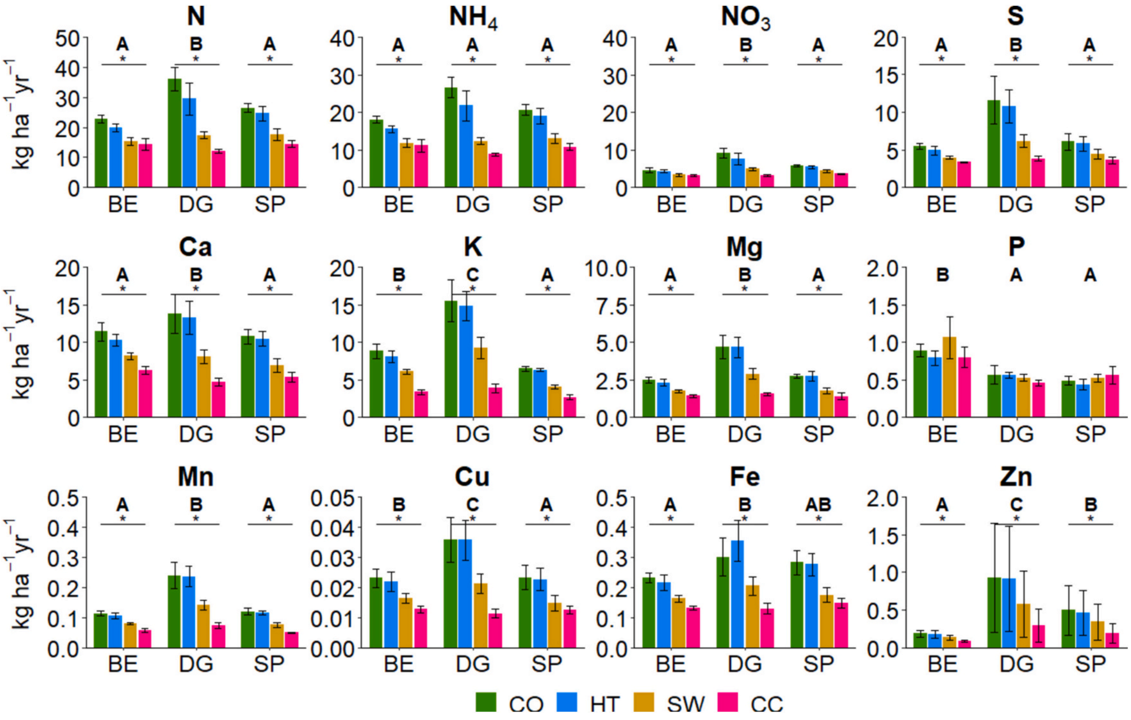

Fig. 1. Total annual nutrient deposition (kg ha^{-1} yr^{-1}) in the control (CO) and in the harvest intensities high-thinning (HT), shelterwood (SW) and clearcut (CC) for beech (BE), Douglas fir (DG) and Scots pine (SP). Different capital letters denote significant differences among species, asterisks indicate significant differences among different harvest intensities (nested-ANOVA, $n = 5$, $P < 0.05$). The tracer Na is given in Fig. S3. Differences among harvest intensities are given in Table S6.

Douglas fir stands than in beech and Scots pine, with only marginal differences between the latter two (Fig. 1, Table S6). The higher total annual deposition in closed Douglas fir stands is attributed to tree height, while in closed beech stands, it relates to stand age and canopy cover, and in closed Scots pine stands, to LAI and stand density (partial-RDA, Var = 6.0, F = 3.3, $p = 0.023$, R_2-adj = 0.64, Fig. S2).

Harvest intensity significantly influenced total annual deposition (Fig. 1, Table S6): for most nutrients (P being an exception), deposition decreased with harvest intensity and stand openness, from control forest to clearcut. Weak but consistent differences were present between control and high-thinning, while differences were more pronounced among high-thinning, shelterwood and clearcut (Table S6). Although the qualitative trends were similar across species, they differed significantly in treatment effect sizes for NH_4, NO_3, K, S, Mg, Mn, and Cu, but not for Ca, P, Zn and Fe. Notable differences between shelterwood and clearcut were observed in Douglas fir (particularly for NO_3, K, S, Mg, Mn and Cu), while such differences were weaker for Scots pine and nearly absent for beech (Fig. 1, Table S6).

The species-specific capacity to intercept deposition led to varying inputs of acids (sum of NH_4, NO_3 and S) and base cations. Acid input ranged from 2.3 keq ha^{-1} yr^{-1} in Scots pine to 3.3 keq ha^{-1} yr^{-1} in Douglas fir, while base cation input varied from 0.93 keq ha^{-1} yr^{-1} in Scots pine to 1.5 keq ha^{-1} yr^{-1} in Douglas fir. Both acid and base cation inputs decreased from control to clearcut (Fig. 2). The net acid input, calculated as acid input minus base cations, significantly declined from control to shelterwood and stabilized or slightly increased toward the clearcut. The strongest decline in net acid input occurred in Douglas fir, while in Scots pine showed only a slight decline (Fig. 2, Table S7).

3.3. *Impacts of tree species and harvest intensity on total seasonal deposition*

Significant variations in total deposition, throughfall and canopy exchange were observed across seasons, with differences among nutrients, species and harvest intensities. The percentage of variation in total deposition explained by species, harvest intensity and season was 61 % (partial-RDA, Var = 6.6, F = 48, $p = 0.001$, R_2-adj = 0.70, Fig. S4). Harvest intensity and season were the primary drivers (28 % and 25 %, respectively), while species contributed a smaller proportion (10 %). The variation in throughfall (control: shelterwood) explained by species, harvest intensity, season, precipitation and interception was 60 % (partial-RDA, Var = 6.2, F = 28, p = 0.001, R_2-adj = 0.51, Fig. S6A). Season was the primary driver (28 %), followed by species (17 %) and harvest intensity (13 %) while precipitation and interception together explained only 2.2 %. Canopy exchange of N (NH_4, NO_3), base cations (Ca, Mg, K) and micronutrients (Mn, Cu, Fe, Zn) varied by season and was influenced by harvest intensity and tree species, with minimal impact from precipitation and interception (35 % of variation explained, p-RDA, Var = 3.0, F = 9.8, p = 0.001, R2-adj = 0.30, Fig. S6B). Harvest intensity (control: shelterwood) accounted for 16 % of the variation, season for 1 %, and tree species for 4.6 %, while the combined water fluxes (precipitation and interception) contributed only 1.5 %.

Seasonal variations in total deposition were primarily related to large differences in total deposition between spring and winter, and to a lesser extent, between summer and winter, with strong association with only a few nutrients: a positive association with P and negative associations with S and Na. Total P deposition was higher in spring and summer compared to autumn and winter (Fig. S5), with no significant effect from harvest intensity and only a minor influence from tree species (Table S6). The total deposition of S and Na increased from spring to winter, with values decreasing from control to clearcut (Fig. S5, Table S6). Seasonal variations in atmospheric deposition among species were subtle, with slightly greater seasonal differences in total deposition observed in control and thinned Douglas fir stands compared to Scots pine (Table S8). Overall, the analysis shows that harvest intensity and season have relatively independent effects on deposition, while species effects are minor in comparison to these harvest intensity and seasonal influences.

4. Discussion

4.1. *Annual total nutrient inputs in Dutch forests: implications for soil acidification*

The total nutrient input in the investigated forest stands is assessed by a combination of measured throughfall and calculated canopy exchange while assuming no nutrient input through stemflow. Rough stemflow estimates for our study sites, based on rainfall partitioning depending on rainfall intensity for beech (Staelens et al., 2008a) and fixed rainfall partitioning to stemflow, i.e. 1 % and 3 % of rainfall, for Douglas fir and Scots pine (Spittlehouse, 1998; Spencer and van Meerveld, 2016) show a general pattern of a contribution of 11 % in unharvested beech and low contribution in Douglas fir and Scots pine stands (Tables 1, S9). This result is in line with literature, indicating that stemflow, however, can be a significant nutrient input in beech stands (Staelens et al., 2007) while it is negligible in Douglas fir and Scots pine stands (Ranger et al., 2002; Cayuela et al., 2018). Because of the high uncertainty of these stemflow estimates, we excluded them from our total deposition estimates. This may lead to some underestimation for total deposition in beech but did not affect our major findings for harvest intensity effects on deposition.

The total deposition, thus without stemflow, in closed stands fell within the range commonly observed in European forests (Van Ek and Draaijers, 1994; Herrmann et al., 2006; Kopáček et al., 2011; Adriaenssens et al., 2012a; Zhang et al., 2020) but differed considerably from the values used in current nutrient balance models for Dutch forests (de Vries et al., 2021). Comparing our results to national deposition maps (RIVM, 2020, 2021), which are largely based on short vegetation

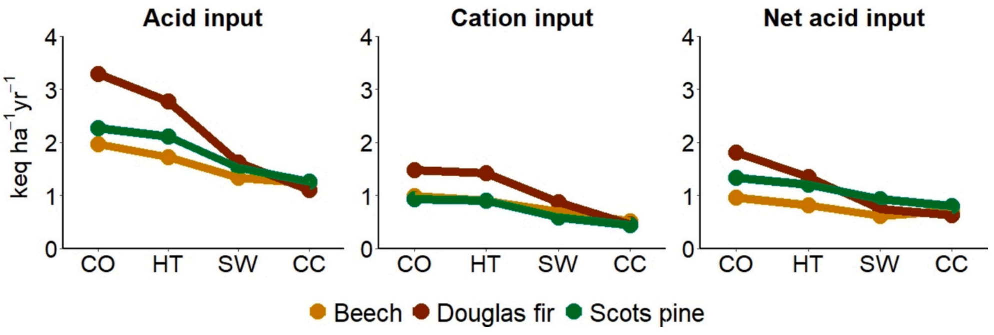

Fig. 2. The total acid input by atmospheric deposition (sum of NH_4, NO_3 and S), the base cation input (sum of Ca, K and Mg) and the total net acid input by atmospheric deposition (sum of NH_4, NO_3 and S minus the sum of the base cations) in keq ha^{-1} yr^{-1} for the unharvested control (CO), high-thinning (HT), shelterwood (SW) and clearcut (CC) harvest intensity. Two-way Anova results of the net acid input are given in Table S7.

measurements (Hoogerbrugge et al., 2022), showed that average NH_4 and S deposition in forests is 0–43 % and 22–38 % higher, respectively, than national estimates across species. Average NO_y deposition was 23 % to 40 % lower (Fig. 1, Table S10) (RIVM, 2020, 2021). Base cation and P inputs were also much higher than expected (Van Jaarsveld et al., 2010; de Vries et al., 2021), with Ca deposition 100–160 % higher, K 250–750 % higher, and Mg 50–190 % higher (Fig. 1, Table S10). This indicates that current deposition rates in forests are higher than assumed.

Elevated acid deposition (N + S) raises concerns, particularly regarding the potential decrease of base cations (Ca, K, and Mg) in forests on sandy soils, where N and historical S deposition induced soil acidification, intensifying leaching processes (Leeters et al., 2007; de Vries et al., 2021). The ratio of N and S to Ca, Mg, and K in total deposition is an indicator of the acidification potential (de Vries et al., 2003). Our results show a total acid input of 2.3–3.3 keq ha^{-1} yr^{-1} and a base cation input of 0.93–1.5 keq ha^{-1} yr^{-1} (Fig. 2). Despite high base cation inputs, acid input remains more than double, suggesting potential cations loss through leaching especially in N-saturated systems or after shelterwood or clearcut harvesting (Vos, 2024). However, the net acidic input (N + S – base cations, Fig. 2) is lower than expected based on national data (Table S10), indicating that the potential for cations loss might be lower than initially thought (Van Jaarsveld et al., 2010; RIVM, 2020, 2021).

The study's total deposition values, which partly exceed the national average, may slightly underestimate actual values. First, stemflow was not measured but may contribute to approximately 11 % of the total deposition for beech but is considered negligible (1–2 %) for Douglas fir and Scots pine, as discussed above. Second, dissolved organic N, dissolved organic P, and particulates were not measured because they are uncharged and thus not captured by the resin. This omission likely leads to an underestimation of N and P deposition across all treatments, by up to 20 % for N and 12 % for P (Ham and Tamiya, 2007; Mahowald et al., 2008; Sleutel et al., 2009).

4.2. Harvest-related reduction of atmospheric deposition is driven by a reduction in dry deposition

Canopy openness, resulting from varying harvest intensities, has a larger impact on total nutrient deposition than tree species or season (Fig. S4). Total annual deposition decreased with increasing harvest intensity for all nutrients except P. Reductions were minor from control forest to high-thinning, larger from high-thinning to shelterwood, and intermediate from shelterwood to clearcut (Fig. 1, Table S6). These results partially align with other studies (Bäumler and Zech, 1997; Aboal et al., 2000; Gielis et al., 2009; Göttlein et al., 2023). However, the expected increase in dry deposition from a rougher canopy in high thinning was absent. Although thinning increased precipitation throughfall (Table S3), it reduced capture of dry deposition, lowering total deposition compared to the control, as noted in other studies (Stogsdill Jr et al., 1989; del Campo et al., 2022). This reduction is likely due to a smaller aboveground surface area, as dense canopies with high LAI capture more dry deposition (Aboal et al., 2000; Yazbeck et al., 2021). Thus, the tree canopy itself, rather than the irregularities in the forest canopy, determines the capture efficiency of dry deposition. Whether structurally complex stands, such as old-growth forests (Parker et al., 2004), could capture nutrients more effectively remains an open question, which is relevant to forest policies promoting closer-to-nature management and stricter protection of natural forests (Larsen et al., 2022).

A sharper decline in total deposition occurred after shelterwood harvesting and clearcutting, corresponding with greater canopy loss (Fig. 1). Few studies have examined shelterwood harvest effects on deposition. Clearcutting (100 % basal area reduction) mirrors the difference between bulk deposition and deposition in closed forests (e.g., Weis et al. (2006)). However, our study's clearcut data isn't directly comparable to bulk deposition, as nearby weather stations recorded ca. 20 % more precipitation, suggesting edge effects in the small clearcut (circa 50 × 50 m) in our study may have led to an underestimation (Neal et al., 1993; Dam, 2001; den Ouden and Mohren, 2020). These edge effects might have also increased throughfall deposition in adjacent plots (De Ridder et al., 2004; Wuyts et al., 2008; Wuyts et al., 2009). As modern forestry increasingly favors smaller clearings, data from small clearings should guide forest management, as total deposition in small clearcuts may consistently be lower than bulk deposition in large open areas.

The reduction in total deposition following shelterwood and clearcutting can be attributed to decreased dry deposition interception due to significant canopy cover loss (Yazbeck et al., 2021). This decrease is not proportional to canopy cover, as wet deposition generally remains unaffected, except at forest edges (Dam, 2001; Erisman and Draaijers, 2003). The impact of harvest intensity on dry deposition varies regionally. For example, heavy thinning in *Picea abies* in southern Germany reduced total deposition by ±45 % (Bäumler and Zech, 1997), while clearcuts in eastern Austria reduced deposition by ±40 % (Berger et al., 2009). In our study, dry deposition played a key role, given the sharp decline in total deposition from closed forest to clearcut (Fig. 1). Similar reductions have been observed in regions, like the Netherlands, France and Canada (Draaijers et al., 1992; Marques and Ranger, 1997; Edgerton et al., 2020), while Austria and Germany showed smaller and larger reductions, respectively (Rothe et al., 2002; Herrmann et al., 2006; Berger et al., 2009). These findings underscore the geographic variability in how harvest intensity affects deposition, highlighting the importance of considering local factors before generalizing results.

4.3. Harvest intensity effects differs among nutrients

Harvest intensity effects varied strongly across nutrients. The lowest impact was observed for P, NO_3, and NH_4, while larger reductions were found for K, Mn, and Zn (Table S6). Nutrients primarily deposited via dry deposition showed the greatest declines after tree harvest. Consequently, impacts of harvest are more pronounced for Na, Cl, Ca, K Mg, and S in coastal areas (Ten Harkel, 1997; Tørseth et al., 1999; Hellsten et al., 2007) and for N, P, S, Mn, Cu, and Zn in regions near pollution sources (Semb et al., 1995; Tørseth et al., 1999; Balestrini et al., 2007; Hellsten et al., 2007; Navrátil et al., 2007; Hsu et al., 2010; Mamun et al., 2020), or other specific sources such as Sahara dust (rich in Ca and Fe) in southern Europe (Semb et al., 1995; Hellsten et al., 2007; Bergas-Massó et al., 2023). The high proportion of dry K deposition in this study may come from multiple sources, including marine (Hellsten et al., 2007; Morselli et al., 2008), agriculture, traffic and wind-blown dust (Draaijers et al., 1996b; Tørseth et al., 1999). The elevated Zn deposition could be linked to traffic (Gunawardena et al., 2013) and the presence of a former Zinc factory in the southern Netherlands, causing notable site differences (Fig. 1).

The reductions in nutrient deposition in this study are consistent with the literature, except for the 50 % higher K throughfall and 50 % lower Mg throughfall beneath Scots pine and Douglas fir compared to similar stands in 1990 (Van Ek and Draaijers, 1994). These differences could be due to Mg uptake in Scots pine (Table 1, Tables S4–S5) and the low K content in Douglas fir needles (Vos et al., 2023b), which reduces canopy leaching (Nordén, 1991).

4.4. Species-driven variation in deposition decrease from closed to open forests

Total deposition varies slightly among species due to their capacity to intercept dry deposition. Douglas fir stands show higher deposition than beech and Scots pine (Fig. 1), consistent with other studies (Van Ek and Draaijers, 1994; Rothe et al., 2002; Zhang et al., 2022). This is likely because Douglas firs are generally taller (Fig. S2) (Lovett and Reiners, 1986; Erisman and Draaijers, 2003). Surprisingly, factors driving the dry deposition such as canopy cover, LAI and tree height (Lovett and

Reiners, 1986; Beier and Gundersen, 1989; Aboal et al., 2000; Erisman and Draaijers, 2003; Staelens et al., 2006; Yazbeck et al., 2021) explain only 13 % of the total deposition variation in unharvested control stands (Fig. S2). This lack of a clear tree height effect may stem from using absolute tree height instead of relative height to surrounding stands.

In this study, species differences are primarily driven by stem density (explaining 43 % of the variation), with Scots pine having the highest and Douglas fir the lowest density (Fig. S2). However, negative effect of stand density on dry deposition appears to be indirect. Douglas fir stands, despite lower density, capture more dry deposition because they are taller and have a higher LAI than Scots pine, with beech falling in between. These species effects diminish with increasing harvest intensity, with Douglas fir showing greater reductions in dry deposition than beech or Scots pine as stands transition from closed to open (Fig. 1, Table S6). Similar patterns have been observed in other studies, such as greater throughfall reductions in harvested *Picea abies* compared to mixed oak stands (Bäumler and Zech, 1997; Wheeler et al., 2000). As harvest intensity increases, species differences in dry deposition lessen.

4.5. Harvest intensity and tree species in relation to acidifying deposition

Harvest intensity could potentially be a management tool to ease the effects of acidifying deposition, as canopy openness (controlled by harvest) affects the deposition of acidifying nutrients (N and S) and base cations differently (Ca, Mg and K) (Fig. 2). Acidic inputs from atmospheric deposition are highest under closed conifer stands due to their enhanced ability to intercept dry deposition by capturing particles and water via their needles (De Schrijver et al., 2007; Pierret et al., 2019). However, we found no evidence of higher acidic inputs under conifers compared to broadleaved species, contrary to De Schrijver et al. (2007) and Rothe et al. (2002). Nevertheless, when corrected for base cations, conifers still receive higher net acidic inputs (Fig. 2), consistent with reports of beech having a higher neutralizing capacity and the tendency of conifers to acidify precipitation (Kowalska et al., 2016; Pierret et al., 2019). These findings reaffirm the higher risk of soil acidification in coniferous versus broadleaved stands (De Schrijver et al., 2012). As forests transition from closed to more open stands, we found strong declines in net acidic input, especially toward shelterwood systems, with a slight decrease from shelterwood to clearcuts (Fig. 2). The decline was most pronounced in Douglas fir shelterwood, followed by high-thinning and shelterwood in beech, where net acidic input dropped by ±50 %. These results suggest that single-tree selective harvesting (comparable to our thinning treatment), particularly in beech and Douglas fir, could be an effective harvesting strategy to slow down soil acidification considering that the impacts of single-tree harvest by thinning on base cation removal and base cation leaching rates is limited (Vos, 2024). Unlike thinning, shelterwood and clearcut systems accelerate soil acidification due to the mobilization and leaching of stored nitrogen and base cations, thereby counteracting the lower net acid input (Vos, 2024). The effects of reduced net acid input following low-intensity harvesting are challenging to predict, as there may be a time lag between the decrease in acid inputs and the system's response (Gilliam et al., 2019).

4.6. Seasonal variations in total deposition

We hypothesized that deposition would vary more seasonally for deciduous than for evergreen Scots pine and Douglas fir, but this was not confirmed. Although total deposition fluctuated significantly across seasons, especially for P, S and N, no notable differences among species were observed. Instead, seasonal deposition was primarily influenced by canopy structure, transitioning from closed forest to clearcuts (Fig. S4; Table S8).

P deposition was higher during the growing season, particularly in beech stands (Fig. S5). This contradicts expectations, as P is typically absorbed by the canopy in nutrient-poor systems (Helmisaari and Mälkönen, 1989; Gordon et al., 2000; Houcai et al., 2021). Canopy leaching of P is unlikely since it peaks in autumn (Sohrt et al., 2019) and correlates with foliar P content (Zhang et al., 2022), where signs of deficiency was noted in our study (Vos et al., 2023b). We attribute the increased P deposition during growing seasons to external sources like pollen or agricultural pollution (Van Ek and Draaijers, 1994; Allen et al., 2010; Kopáček et al., 2011; Tipping et al., 2014). Since harvest intensity had no effect on P deposition, the dry deposition of P appears negligible, with pollen likely driving the seasonal pattern (Doskey and Ugoagwu, 1989; Rösel et al., 2012). Pollen may also contribute to K and NH_4 increases, but its influence on these nutrients is relatively minor compared to P (Verstraeten et al., 2023).

Na deposition showed significant seasonal variation, with higher deposition in autumn for closed and thinned stands, and in winter for shelterwood and clearcut areas (Fig. S4). Na deposition is particularly important due to its role in canopy exchange (eq. 2), which partially explains seasonal changes in base cation deposition (Fig. S6B). Higher throughfall of Na in autumn for closed and thinned stands may result from canopy leaching (Staelens et al., 2007; Thimonier et al., 2008; Adriaenssens et al., 2012a) or from more efficient capture of Na in autumn (Adriaenssens et al., 2012a) as increased winter deposition may be related to elevated atmospheric concentrations (Van Ek and Draaijers, 1994; Adriaenssens et al., 2012a). Contrary to previous suggestions (Staelens et al., 2007; Thimonier et al., 2008), we found no evidence of higher Na throughfall in spring (Fig. S6A), challenging the idea of canopy leaching during bud break. While the elevated Na content in autumn may indicate some degree of canopy leaching, this effect appears minor. Given the seasonal variations of the Na deposition, using Na in canopy exchange calculations for Ca, K, and Mg may have introduced uncertainties, as it assumes these elements behave similarly to Na with respect to contributions from wet and dry deposition.

The main factors behind seasonal differences in canopy leaching, including increased K and Mn leaching in autumn and winter, have been observed in earlier studies (Van Ek and Draaijers, 1994; Talkner et al., 2010). These are likely linked to (drought-induced) senescence, which makes leaves becoming more prone to ion leakage, resulting in substantial losses of K and Mn (Schaefer and Reiners, 1990; Hagen-Thorn et al., 2006; Houle et al., 2016). Overall, this study finds no evidence against using Na as a tracer in canopy exchange.

4.7. Indications of canopy uptake of base cations

Our results show net canopy uptake of NH_4, NO_3, Ca, Mg (in Scots pine), Zn, Fe and Cu along with net release of K and Mn. The release of K and Mn was lower than generally reported (Petty and Lindberg, 1990; Herrmann et al., 2006; Gandois et al., 2010; Adriaenssens et al., 2012a) possibly due to the low foliar concentrations of these elements in our study trees (Talkner et al., 2010; Vos et al., 2023b). Canopy uptake of NH_4 and NO_3, reaching up to 90 % of throughfall deposition, is well-established (Wilson and Tiley, 1998; Klopatek et al., 2006; Adriaenssens et al., 2011; Schwarz et al., 2014; Houle et al., 2015). While Ca and Mg leaching from canopies is well-documented (Draaijers et al., 1997a; Moreno et al., 2001; De Schrijver et al., 2007; Talkner et al., 2010; Adriaenssens et al., 2012a; Shen et al., 2013; Edgerton et al., 2020), canopy uptake of these elements is less known but has been observed across various species (De Schrijver et al., 2004; Małek and Astel, 2008; Tan et al., 2018; Van Langenhove et al., 2020). We hypothesize that the high atmospheric deposition, strongly acidified soils, and low soil Ca and Mg availability – resulting in low foliar Ca and Mg concentrations in the trees at our study sites – caused the canopy to shift from being a source to a sink for these nutrients (Vos et al., 2023b). This aligns with Talkner et al. (2010) and Nordén (1991), who suggested that canopy leaching of base cations increases with soil fertility. Various reports document the beneficial effects of foliar Ca sprays on Ca uptake and crop growth, particularly where soil Ca availability is limited; this also applies to minor nutrients (e.g., Bons and Sharma, 2023). However, it is important to note that these findings are uncertain, as they assume that

Na behaves as a tracer and that the total deposition/bulk deposition ratio is similar for Na, Ca, K, and Mg. While Na leaching seems unlikely, it is possible that the total deposition/bulk deposition ratio is lower for Ca than for Mg, which would imply reduced Ca uptake. Our results suggest that in acidified, nutrient-poor forests, canopy adsorption of essential nutrients like Ca and Mg can exceed canopy leaching, highlighting the need for caution when extrapolating deposition data, as these processes can vary even in similar forests (Edgerton et al., 2020).

5. Conclusions

Sustainable management of forests on poor soils faces challenges from base cation losses (Ca, K, Mg) due to N and S deposition-induced soil acidification. Our non-harvested control plots showed significantly higher N, S, Ca, K, and Mg inputs compared to open sites, which are usually monitored by national policy. The net input of NH_4 and SO_4 on closed or thinned forests were 26–140 % higher than open sites, and this was even higher (68 %–750 %) for base cations and some micronutrients. When harvest intensity increases, the reduction in net acid input exceeds the reduction in base cations. This highlights the potential of forest management practices that influence forest structure and canopy openness to mitigate soil acidification in areas with high N deposition, provided leaching remains controlled.

We found that nutrient deposition in European beech, Douglas fir, and Scots pine stands consistently decreased with increased canopy openness via increased harvest intensity, except for P, likely due to pollen rather than atmospheric deposition. Harvest-induced canopy openness had varied effects on nutrients, with sharp declines for those deposited mainly through dry deposition (e.g., S, K). The highest nutrient and acid inputs occurred in taller Douglas fir stands, while shorter, dense Scots pine stands had the lowest inputs. As stands became more open, species differences diminished, making harvest intensity the dominant factor influencing deposition patterns.

Our results highlight the importance of considering harvest intensity effects on canopy openness, and to a lesser extent tree species, when estimating nutrient inputs from atmospheric deposition. While tree harvest can help reduce acid inputs, continued N deposition makes soil acidification and base cation loss inevitable. Since deposition is influence by factors like air quality, proximity to the sea, and local pollution, the effects of harvesting on total deposition will vary by location. Importantly, the efficient capture of potentially growth-limiting macronutrients (e.g., K, Ca, Mg) and micronutrients (such as Mn) in closed and thinned forests (canopy openness $<$20 %) contribute to the resilience on forests on nutrient-poor, acidified soils, and implies that intensive tree harvest (e.g. shelterwood or clearcut) are better avoided in those conditions.

CRediT authorship contribution statement

Marleen A.E. Vos: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Wim de Vries:** Writing – review & editing, Methodology, Funding acquisition, Conceptualization. **Jan den Ouden:** Writing – review & editing, Conceptualization. **Frank J. Sterck:** Writing – review & editing, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: M.A.E. Vos reports financial support was provided by Dutch Research Council. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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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.2024.177933.

Data availability

Data will be made available on request.

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