

Impacts of
harvesting
practices
on **nutrient
balances**
of forests under
high nitrogen deposition

Marleen **Vos**

Propositions

1. Forest harvesting practices should avoid large canopy openings and clearcuts.
(this thesis)
2. Forest soil acidification is more affected by the legacy of historic sulfur deposition than by current nitrogen deposition.
(this thesis)
3. Prioritizing grant acquisition skills in scientific funding will cause talented scientists to leave.
4. The current funding mechanisms of scientific research hinder long-term projects.
5. The impact of papers often mirrors the societal appeal of a subject rather than the scientific robustness of the research conducted.
6. Networking differs only from nepotism in that it considers people's abilities.
7. Organizations that restrict timber harvest should refrain from using wooden furniture.

Propositions belonging to the thesis entitled:

Impacts of harvesting practices on nutrient balances of forests under high nitrogen deposition

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Wageningen, 15 May 2024

Impacts of harvesting practices on nutrient balances of forests under high nitrogen deposition

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This research was conducted under the auspices of the C. T. de Wit Graduate School for Production Ecology and Resource Conservation (PE&RC)

Impacts of harvesting practices on nutrient balances of forests under high nitrogen deposition

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Thesis

submitted in fulfilment of the requirements for the degree of doctor at

Wageningen University

by the authority of the Rector Magnificus

Prof. Dr C. Kroeze

in the presence of the

Thesis Committee appointed by the Academic Board

to be defended in public

on Wednesday 15 May 2024

at 4 p.m. in Omnia Auditorium

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Impacts of harvesting practices on nutrient balances of forests under high nitrogen deposition

300 pages

PhD thesis, Wageningen University, Wageningen, the Netherlands (2024)

With references and English summary

DOI: <https://doi.org/10.18174/650894>

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Chapter I

General introduction

Challenges for sustainable forest biomass harvest

Forests play a key role in a sustainable bio-based economy by supplying biomass through tree harvesting (Antar et al., 2021). The use of biomass as a renewable raw material has promoted interest in forests, resulting in a notable increase in the utilization of biomass from European forests (EUROPE, 2020). This increased utilization of tree biomass is expected to continue in the future, driven by the growing demands for timber, raw materials for the industries and bioenergy (Egnell, 2017; Bell et al., 2018; Antar et al., 2021). However, the utilization of tree biomass exerts additional pressure on forests, as biomass harvesting can impact soil nutrient pools, site productivity, and overall forest health, including tree growth and survival (Thiffault et al., 2011; Evans, 2016; Börjesson et al., 2017; de Oliveira Garcia et al., 2018). Biomass harvest thus poses risks for forests which could threaten the renewability of the resource.

The nutritional status of European forests has declined over the last decades resulting from increased growth (Jonard et al., 2015; Penuelas et al., 2020) and the acidifying effects of nitrogen (N) and sulfur (S) deposition (de Vries et al., 2014a; Braun et al., 2020a). Increased growth is triggered by the higher atmospheric CO₂ levels and the fertilizing effects of N deposition (Hunter and Schuck, 2002; Etzold et al., 2020). This increased growth leads to immobilization of nutrients in tree biomass causing a decline in the soil nutrient pools. Eventually, growth is limited by the supply of essential nutrients like phosphorous (P) (Penuelas et al., 2020; Prietzel et al., 2020; Du et al., 2021). Furthermore, ongoing N deposition increases soil N availability, leading to acidification through elevated nitrification. This, in turn, accelerates the leaching of essential base cations, including calcium (Ca), magnesium (Mg), and potassium (K). As a result, nutrient imbalances and limitations are particularly pronounced in forests situated on nutrient-poor soils with low buffering capacities (De Vries et al., 1995b; Bowman et al., 2008). Many temperate forests are located on such soils influenced by the acidifying effects of N deposition and the legacy of elevated S deposition (Chuman et al., 2021).

Biomass harvest in forests is already under pressure by these negative effects of N deposition on soil nutrient availability and can potentially lead to lower site productivity (Achat et al., 2015; Vangansbeke et al., 2015; Egnell, 2017; de Vries et al., 2021). In addition biomass harvest can lead to a reduction of the soil nutrient stocks, which has been found even when only stems are harvested, hence referred to as stem only harvest (Akselsson et al., 2007b; de Vries et al., 2021), but specifically when alongside with the stem the crown is harvested, further referred to as whole tree harvest (Akselsson et al., 2007b; Vanguelova et al., 2010; Vangansbeke et al., 2015). Therefore, the effects of biomass harvest on soil nutrient stocks are a major concern, as biomass harvest, especially in the form whole tree harvest, can lead to nutrient limitations and imbalances (Walmsley et al., 2009; Vanguelova et al., 2010; Vangansbeke et al., 2015). These limitations and imbalances can reduce forest growth (Braun et al., 2010; Prietzel and Stetter, 2010; Fan et al., 2015; Hevia et al., 2019) and lower forest resilience due to increased sensitivity to drought, frost and

pests, and therefore, in turn, increase risks of forest dieback (de Vries et al., 2014b; Sardans and Peñuelas, 2015; Carter et al., 2017; Hevia et al., 2019; Scheel et al., 2022). Consequently, it is important to anticipate the long-term effects of different forest management practices on the ability of the forest to sustain tree biomass provision and maintain forest ecosystem functioning.

The nutrient budget: a decision support approach for sustainable biomass harvest

The ecological sustainability of tree biomass harvest, concerning soil nutrient stocks, can be evaluated through the nutrient budget approach, which assesses the balance of nutrient inputs and outputs (Ranger and Turpault, 1999; Pare and Thiffault, 2016), but this approach is often limited to few nutrients measured over rather short time periods. The most important nutrient input fluxes in temperate forests consist of atmospheric deposition and weathering while the major nutrient outputs are leaching, and the nutrients contained in exported tree parts by tree harvest (Fig. 1.1). Sustainable biomass harvest implies that the nutrient output via harvest and leaching should not exceed the nutrient input via deposition and weathering to avoid that nutrient stocks of the forest decline over subsequent rotations. Other nutrient inputs and outputs that can play a role in the nutrient balance are the supply of nutrients through groundwater (Hayes et al., 2019) and biological fixation and denitrification of nitrogen (N) from and to the atmosphere (Boring et al., 1988; Pare and Thiffault, 2016) and the loss of nutrients via runoff (Suescún et al., 2017) while flooding can be both a nutrient input as a nutrient output (Vourlitis et al., 2017; Talbot et al., 2021). Biological N fixation plays a role in N-limited environments (Pare and Thiffault, 2016) while flooding, runoff and nutrient supply via groundwater are related to the landscape geography. This thesis explicitly concentrates on low-land production forests situated on well drained poor sandy soils characterized by high nitrogen deposition, without groundwater access for roots and negligible runoff, and flooding, while biological N fixation is also very limited. The nutrient budget for these forests is therefore defined as the net amount of nutrients entering via deposition and weathering or leaving via export and leaching expressed in kg per hectare forest per year:

$$\text{Nutrient budget} = \text{Atmospheric deposition} + \text{Weathering} - \text{Leaching} - \text{Exported tree parts}$$

Evaluations of the effects of biomass harvest on the nutrient budget in forests are often limited to the effects of nutrient removal in exported tree parts (crown and stem), thereby often ignoring the feedbacks of harvest on atmospheric deposition and leaching (Fig. 1.1). Tree harvest causes changes in the forest structure which may temporarily increase or reduce the atmospheric deposition, due to differences in capturing gases or particulates as dry deposition (Lovett and Reiners, 1986; Bäumler and Zech, 1997; Gielis et al., 2009; Göttelein et al., 2023). Moreover, leaching may be temporarily increased as a result of reduced tree uptake and increased availability of nutrients through decomposed harvest residues (Katzensteiner,

2003; Rothe and Mellert, 2004; Gundersen et al., 2006; Titus et al., 2006; Piirainen et al., 2007; Sterck et al., 2021). Also weathering can be influenced by biomass harvest as harvest interferes with soil moisture availability, soil temperature, the capture of acid deposition and soil acidification by accelerated leaching (Olsson and Melkerud, 1991; Van der Salm et al., 1999; Guo et al., 2015; Dixon et al., 2016; Houle et al., 2020; Belyazid et al., 2022). However, the weathering flux itself is often highly uncertain, with uncertainties frequently reaching a magnitude comparable to or even greater than the mean estimated weathering rate, especially in systems with low weathering rates, such as poor sandy soils (Hodson et al., 1997; Klaminder et al., 2011; Futter et al., 2012; Simonsson et al., 2015).

Consequently, attempting to determine the feedback on weathering using conventional methods would have minimal impact on the balance's reliability. However, evaluating the post-harvest effects on deposition and leaching, which includes both the extent and duration of these effects, is crucial for improving the reliability of the nutrient balance.

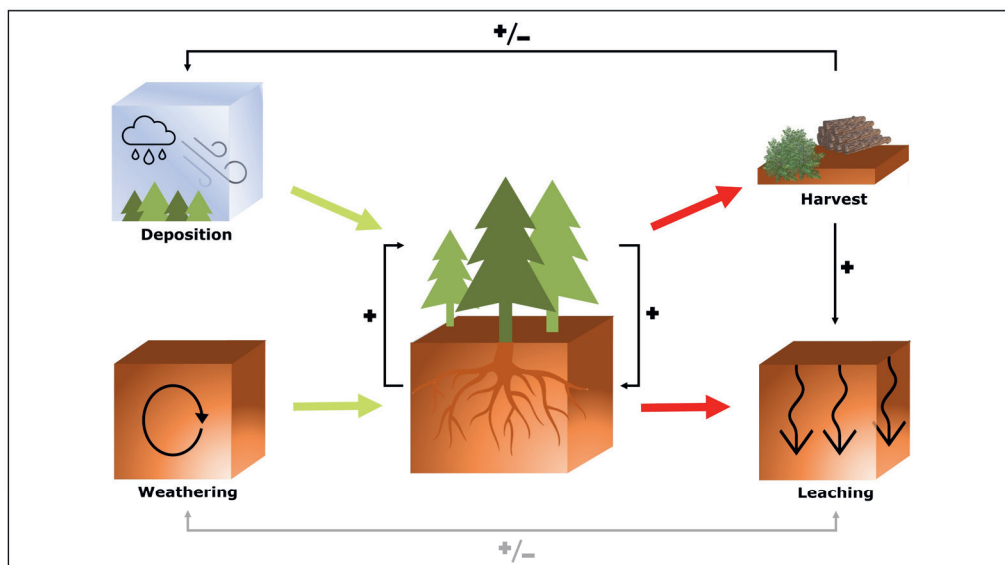


Figure 1.1 The nutrient balance primarily comprises deposition and weathering as nutrient inputs (indicated by green arrows), while leaching and harvest represent nutrient outputs (depicted by red arrows). Feedback mechanisms within this nutrient balance are denoted by black and grey arrows. Feedback is considered positive when it increases the flux, as expected for harvest increasing leaching, belowground nutrient stock increasing growth, and higher aboveground nutrient stock increasing the input of nutrients to the forest floor via litter. Such expected positive feedbacks are denoted by a '+', while uncertain feedbacks are indicated as '+/-'. Current methods provide sufficient certainty for evaluating the effects represented by black arrows. However, estimating the impact of the grey arrow is challenging due to considerable uncertainties associated with their estimation.

The challenges of forest nutrient budgets: addressing uncertainties and post-harvest dynamics

Even though the nutrient budget approach has been used as a guidance tool for sustainable forest management (de Vries et al., 2021; Titus et al., 2021; Ahrends et al., 2022; Vanguelova et al., 2022), it is not yet widely adopted due to several problems including: (i) large uncertainties in assessing or predicting deposition, weathering and leaching fluxes, (ii) the poor representation of nutrient dynamics following harvest (temporal variability) and (iii) the lack of data on specific tree and soil properties that would be required to predict forest nutrient budgets at regional scale (spatial variability) (Klaminder et al., 2011; Lucas et al., 2014; Pare and Thiffault, 2016; Johnson and Turner, 2019; Löfgren et al., 2021). The reported effects of tree harvesting on nutrients and therefore on soil productivity often lack consistency (Thiffault et al., 2011; Egnell, 2017; Garrett et al., 2021) and are difficult to quantify because of complex interactions between the forest biogeochemical cycle and tree growth (Pare and Thiffault, 2016; Rosenstock et al., 2019). First of all, the limited availability of measurements, leads to large uncertainties of the estimated nutrient fluxes at an annual time scale and at plot scale, especially over longer time periods (Pare and Thiffault, 2016; de Vries et al., 2021; Vanguelova et al., 2022). The uncertainties in the estimated nutrient fluxes further increase at regional scale caused by the lack of site specific data affecting those fluxes, such as tree height, canopy coverage and soil texture, which often necessitate the use of coarse deposition, weathering and leaching data (see for example de Vries et al. (2021), Rothstein and Gadoth-Goodman (2023) and Löfgren et al. (2021)). Therefore, some argue that nutrient budgets are inadequate for capturing the nutrient cycling of long-rotation forests in temperate biomes (Pare and Thiffault, 2016; Löfgren et al., 2021). To reduce uncertainties in nutrient budget predictions, it is essential to carry out intensive local measurements on nutrient fluxes in large-scale field experiments as the efficacy of biomass harvesting guidelines is hampered by the absence of site-specific data (Clarke et al., 2015; Pare and Thiffault, 2016; Vance et al., 2018; Löfgren et al., 2021; Titus et al., 2021).

Furthermore, intensive local measurements can contribute to reducing uncertainties related to the impacts of harvesting on leaching and deposition across varying harvest intensities, being essential for making informed decisions on forest management. Nutrient budget studies often rely on deposition and leaching estimates from mature forest stands, overlooking the post-harvest dynamics in deposition and leaching (for example Akselsson et al. (2007a) and Ahrends et al. (2022)). These post-harvest nutrient dynamics are highly influenced by the number of trees harvested (harvest intensity) and, in the case of leaching, the tree parts harvested (or harvest method, for example, stem only harvest versus whole tree harvest) and soil preparation. The extent and duration of these effects can vary among sites, depending on soil type, the geographic region and the tree species being harvested (Thiffault et al., 2011; Clarke et al., 2015). Depending on the forest and its management, these post-harvest dynamics can have a small (Gundersen et al., 2006; Berger et al., 2009; Talkner et al., 2010; Phillips and Watmough, 2012) to

substantial impact (Bäumler and Zech, 1997; Rothe et al., 2002a; Katzensteiner, 2003; Rothe and Mellert, 2004; Gundersen et al., 2006; Herrmann et al., 2006; Titus et al., 2006; Piirainen et al., 2007). The post-harvest dynamics may persist for only a few years (Huber et al., 2004; Wang et al., 2006; Huber et al., 2010) or extend over decades (Marcotte et al., 2008; Oda et al., 2021). Additionally, the post-harvest effects may vary between different nutrients (Bäumler and Zech, 1997; Martin et al., 2000; Berger et al., 2009; Webster et al., 2022). Therefore, to increase the reliability of the forest nutrient budget, it is of high importance to better quantify and understand post-harvest nutrient dynamics in relation to forest management practices, and account for possible effects of species and soil type. The effects of tree species and of forest management practices, including harvest intensity, harvest method, and post-harvest soil preparation which all affect the nutrient budget (Akselsson et al., 2007b; Piirainen et al., 2007; Thiffault et al., 2011; Egnell, 2017), are discussed in detail below.

The influence of tree species on forest nutrient budgets

Tree species can strongly influence the forest nutrient budget as tree species vary in the nutrient concentrations of their tissues, their total aboveground biomass and, therefore, in their nutrient stocks that will be exported by harvest (Weis and Göttlein, 2002; Andre et al., 2010; Palviainen and Finer, 2012; Paré et al., 2013; Pretzsch et al., 2014; Husmann et al., 2018; de Jong et al., 2022). Additionally, because of distinct species properties like deciduousness, canopy shape, leaf area index (LAI) and tree height, tree species also differ in their ability to intercept atmospheric particles (Lovett and Reiners, 1986; Beier and Gundersen, 1989; Aboal et al., 2000; Erisman and Draaijers, 2003; Staelens et al., 2006; Shapkalijevski et al., 2016; Yazbeck et al., 2021; Zhang et al., 2022). Moreover, tree species influence biogeochemical soil cycles by affecting litter decomposition rates through different lignin concentrations, consequently influencing nutrient cycling (Sariyildiz et al., 2005; Tao et al., 2019; Yu et al., 2021), soil acidity (Augusto et al., 2002; Mueller et al., 2012), water balance and microclimate (Augusto et al., 2002; Meinzer et al., 2013). These differences in biogeochemical cycling, water use, and the interception of acid atmospheric particles that cause soil acidification can result in variations in nutrient leaching (Rothe et al., 2002a; Kowalska et al., 2016; Pierret et al., 2019).

The impact of tree species on soil fertility is not unidirectional. It depends on factors, such as the mineral substrate, the role of soil organisms, climate, and forest management practices (Binkley and Giardina, 1998; Vesterdal and Raulund-Rasmussen, 1998; Augusto et al., 2002; Meier and Leuschner, 2014; Pretzsch et al., 2014). However, some general patterns of species effects on the nutrient balance emerge from the literature: broadleaved species generally have higher nutrient concentrations in the aboveground biomass (Augusto et al., 2000; Augusto et al., 2002; de Jong et al., 2022) and therefore a generally higher nutrient export following stem only harvest. Contrary, nutrient exports following whole tree harvest, where

the tree crown is also harvested, are generally higher for conifer species due to the export of needles (Pare and Thiffault, 2016). Furthermore, conifers species may be more effective in intercepting atmospheric particles compared to broadleaved species (Gundersen et al., 2009; Vanguelova and Pitman, 2019). There is no consistent effect observed on nutrient losses via leaching for coniferous or broadleaved species (Gundersen et al., 2009; Braun et al., 2020b), possibly because leaching may be more determined by soil properties like the buffer capacity, especially for stands under high N deposition input (Rothe and Mellert, 2004). Overall, the species effects on the nutrient budget remain ambiguous and species-specific data are necessary to understand the impact of the tree species on the forest nutrient balance.

Impact of harvest intensity on the nutrient budget

The harvest can either consist of a forest thinning or a final felling (or regeneration felling), each with different intensities. High-thinning (or crown-thinning) involves selectively removing individual trees to enhance the growth of the remaining trees by intervening in competitive relationships within the forest canopy (Den Ouden et al., 2010; Röhrig et al., 2020). In thinning, the forest canopy cover is mainly preserved as tree harvesting occurs at a low intensity, which is repeated frequently. Most commonly, thinning is classified as light thinning when less than 20% of the basal area is removed and as heavy thinning when > 35% of the basal area is removed (Mäkinen and Isomäki, 2004; del Río et al., 2008; del Río Gaztelurrutia et al., 2017; Gradel et al., 2017; Baah-Acheamfour et al., 2023). Thinning directly impacts the canopy structure by creating small irregular openings in the forest canopy which are gradually filled by either expanding mature trees or by forest regeneration. On the other hand, shelterwood and clearcut are considered final felling methods, where the shelterwood system aims to establish a new stand while preserving seed trees as shelter, whereas clear-cutting removes an entire stand of trees (Schönenberger and Brang, 2004; Röhrig et al., 2020). The basal area reduction is 100% in a clearcut, the reported basal area percentage removed in a shelterwood varies widely and is between 60-80% basal area removed when classified as sparse shelterwood (Loftis, 1990; Bradley et al., 2001; Holgén et al., 2003; Wardell-Johnson et al., 2004; Prévosto et al., 2011) while a more dense shelterwood can consist of a basal area reduction of 30-40%, thus approaching a heavy thinning (Brose et al., 1999; Holgén et al., 2003; Prévosto et al., 2011). However, despite the broad interpretation of the shelterwood system, there is a general consensus that, in shelterwood, small irregular patches of the canopy cover are left, creating a spacious canopy structure. This spacious canopy, however, reduces exposure and tempers the microclimate compared to a clearcut, thereby supporting seedling survival (Langvall and Löfvenius, 2002; Beadle and Sands, 2004; Paquette et al., 2006). Harvest intensity, and along with it, biomass export, increases from high-thinning to clearcut, making it a key determinant of nutrient export in harvested wood products. However, the extent of nutrient export in harvested wood products also depends on factors such as the rotation period or management interval, tree

species, forest age, the tree parts being exported, and the forest's management history (Wang et al., 2016; Achat et al., 2018a).

Impact of harvest intensity on atmospheric deposition

The harvest intensity also affects nutrient input through atmospheric deposition and nutrient losses via leaching. Atmospheric deposition into the forest is altered by harvest intensity due to reductions in canopy cover, changes in canopy structure and therefore in canopy roughness, and a decrease in LAI, all affecting the capture of gases and particulates and thereby dry deposition (Aboal et al., 2000; Yazbeck et al., 2021). For instance, the canopy roughness of a thinned stand is increased by small gaps in the forest canopy, which can increase the penetration of turbulent air (Russell et al., 2018). While increased canopy roughness potentially leads to higher interception of atmospheric deposition (Lovett and Reiners, 1986) this effect is often not observed (Stogsdill Jr et al., 1989; del Campo et al., 2022; Göttlein et al., 2023). Canopy roughness and air turbulence decrease in shelterwood and clearcut scenarios (Russell et al., 2018) coinciding with the reduction in canopy cover and LAI, frequently resulting in decreased atmospheric deposition (Bäumler and Zech, 1997; Weis et al., 2006; Berger et al., 2009; Yazbeck et al., 2021). However, the extent of this reduction strongly depends on the species-specific capacity to intercept deposition (Van Ek and Draaijers, 1994; Rothe et al., 2002a; Zhang et al., 2022), and the amount of the particles in the air. For example, the reduction of the atmospheric deposition in response to the reduction in canopy cover, canopy roughness, and LAI following harvest will be more pronounced in areas with high concentrations of particles, such as those near the sea or influenced by anthropogenic pollution, like agricultural areas (Semb et al., 1995; Ten Harkel, 1997; Tørseth et al., 1999; Balestrini et al., 2007; Hellsten, 2007). Therefore, classifying the extent to which deposition is influenced by canopy cover, structure, and roughness is challenging, as these effects can be modified by species and geographic location.

Impact of harvest intensity on nutrient leaching

Nutrient leaching can vary significantly over time and among different soil types, vegetation types, and climatic regions (Asano et al., 2006; Campbell et al., 2014; McDaniel et al., 2014; Fransson, 2018). Forest harvesting affects this process by reducing water uptake by trees, potentially increasing runoff and groundwater recharge, reducing nutrient uptake, causing soil disturbance, and releasing nutrients from decomposing harvesting residues (Parfitt et al., 1997; Nieminen, 2004; Piirainen et al., 2007; Devine et al., 2012; Sterck et al., 2021; del Campo et al., 2022). Nutrient losses via leaching often increase temporarily after tree harvest particularly with higher harvest intensities (Katzensteiner, 2003; Jerabkova et al., 2011; Göttlein et al., 2023). The extent of the increase in post-harvest leaching varies among tree species (Augusto et al., 2002; Rothe et al., 2002a; Jerabkova et al., 2011), N saturation and buffer capacity of the soil

(Gundersen et al., 2006; Piirainen et al., 2007; Huber et al., 2010; Olsson et al., 2022), and the recovery of the vegetation (Martin et al., 2000; Weis et al., 2001; Gundersen et al., 2006; Huber et al., 2010). Low harvest intensities, like high-thinning, typically result in a negligible to small increase in leaching, as they do not substantially alter the forest microclimate, water balance, and nutrient uptake (Weis et al., 2001; Jerabkova et al., 2011; Phillips and Watmough, 2012; del Campo et al., 2022). After a final felling, such as shelterwood and clearcut, increased mineralization due to higher soil temperature and soil moisture, along with reduced nutrient uptake by trees, can lead to increased nutrient leaching (Katzensteiner, 2003; Rothe and Mellert, 2004; Gundersen et al., 2006; Titus et al., 2006; Piirainen et al., 2007). However, the responses of leaching to tree harvest are highly variable and often site specific (Kreutzweiser et al., 2008; Achat et al., 2015). For example, leaching of N and K in a forest on a deep outwash substrate (sand/loamy sand) was higher underneath unharvested forests (Wilhelm et al., 2013) while for sandy soils usually higher N and P leaching can be found following a clearcut harvest (Piirainen et al., 2004; Richardson et al., 2017). Differences in leaching between sites may be related to the ongoing effects of acid deposition (N and S), which result in soil acidification and the release of base cations, which ultimately can result in accelerated base cation (Ca, Mg and K) leaching (Currie et al., 1999; Bowman et al., 2008; Braun et al., 2020b). Additionally, soil acidification is a site-specific effect, as it depends on the soil's buffer capacity, which is influenced by vegetation and soil N and base cation stocks (Ross et al., 2008; Lu et al., 2015; Jiang et al., 2016). Consequently, there is no unidirectional impact of harvest intensity on leaching because leaching is directed and influenced by many factors. This underscores the importance of experimental field sites to enhance our understanding of the leaching for different species, soil types and environmental conditions in response to harvest intensity.

Impact of biomass harvest method and soil preparation on the nutrient budget

When harvesting, the removed or exported tree parts may include only the stem (stem only harvest: SOH), both the stem and the crown (whole tree harvest: WTH), or only the stem without bark in case the bark is stripped in the forest (stem wood harvest: SWH). The most traditional harvest method is SOH, although there is recently increased interest in WTH due to growing demands for renewable energy (Levin and Eriksson, 2010; Briedis et al., 2011; Egnell et al., 2011; Berger et al., 2013; Ha et al., 2022). However, the sustainability of WTH is often debated because of the high nutrient exports (Walmsley et al., 2009; Thiffault et al., 2011; Palviainen and Finer, 2012; Pare and Thiffault, 2016; Zetterberg et al., 2016). Contrary, in-situ debarking in case of SWH, has the potential to mitigate the impact of harvest on the soil fertility (Raison et al., 1982; Andre et al., 2010; Achat et al., 2015). Despite the high extraction costs in current techniques, debarking in-situ on forest sites has the potential to be a valuable addition to existing harvesting methods (Koutsianitis and Tsioras, 2017; Heppelmann et al., 2019).

The biomass harvest method (SOH, WTH, and SWH) directly influences the nutrient budget through the nutrients in the exported tree parts and the nutrients in the harvest residues. The impact of the biomass harvest method on nutrient export depends on various factors, including tree species (Palviainen and Finer, 2012; Pare and Thiffault, 2016), stand age (Peri et al., 2006; Wang et al., 2016), management history and soil fertility. Management history determines, for example, the crown volume (Han et al., 2014; Georgi et al., 2018), and soil fertility influences the nutrient concentrations in the different tree compartments (Nordén, 1991; de Jong et al., 2022). In general, within mature stands, WTH can increase nutrient exports 2 to 4 fold compared to SOH (Palviainen and Finer, 2012; Vangansbeke et al., 2015) while SWH can decrease nutrient exports compared to SOH by up to 85% (Andre et al., 2010; Yan et al., 2017). These high exports are concerning as whole tree harvest has the potential to export more nutrients than available in the soil stocks (Merino et al., 2005; Vangansbeke et al., 2015) which could ultimately lead to a reduction of the site productivity.

Furthermore, nutrient conservation by leaving harvest residues in the forest may be counteracted by elevated nutrient losses via leaching. Harvest residues intentionally left in the forest for the SOH and SWH biomass harvest methods can result in an elevated post-harvest nutrient leaching (Rosén and Lundmark - Thelin, 1987; Kuehne et al., 2008; Wall, 2008; Devine et al., 2012). Again, the extent of this post-harvest leaching flux in relation to the harvest residues differs as a result of interactions with the amount of harvest residues, soil fertility, microbial activity, weather circumstances and vegetation (Stevens et al., 1995; Belleau et al., 2006; Smolander et al., 2008; Achat et al., 2015; Bergholm et al., 2015). These interactions can result in varying impacts of harvest residues on leaching, ranging from increased leaching following WTH (Fahey et al., 1991) or SOH (Valinia et al., 2021) to negligible differences observed between SOH and WTH (Mann et al., 1988; Sarkkola et al., 2016). Presence of harvest residues may also affect the microclimate and the vegetation which can either positively or negatively influence seedling growth, whereby seedling can reduce post-harvest leaching (Fahey et al., 1991; Thiffault et al., 2011). Furthermore, the amount of harvest debris can either stimulate or suppress microbial activity which has a direct effect on the soil nutrient availability and therefore on the nutrient leaching. Therefore, given the variability and complexity of factors influencing nutrient leaching in response to harvest debris, there is an urgent need for experimental data allowing for a better understanding of the effects of harvest residues on nutrient losses by leaching.

The common practice of the post-harvest soil preparation influences the decomposition of the harvest residues, alters soil communities and facilitates the establishment of regeneration (Lundmark-Thelin and Johansson, 1997; Piirainen et al., 2007; Kwaśna et al., 2019; Pitman and Peace, 2021; Smenderovac et al., 2023). Among the commonly used types of soil preparation is flail mulching, which disturbs the topsoil and fragments harvest residues into smaller pieces. Flail mulching increases the decomposition of harvest

residues, potentially leading to higher nutrient losses via post-harvest leaching (Lundmark-Thelin and Johansson, 1997; Piirainen et al., 2007; Sun et al., 2022). Despite the possible impact, the effects of flail mulching on the post-harvest leaching are hardly studied. The sparse literature on this topic provides an ambiguous overview: flail mulching can immobilize nutrients but still result in an elevated leaching in the temperate forest (Pitman and Peace, 2021) while in tropical regions, faster decomposition after mulching did not result in increased nutrient losses through leaching (Sommer et al., 2004). These varying effects might be attributed to factors such as soil type, soil communities, and site-specific properties, including the establishment of regeneration. Currently, information on the effect of flail mulching following SOH and WTH harvests is largely lacking, which hinders our ability to understand the impact of mulching on nutrient leaching. Therefore, experimental data is necessary incorporating the effects of soil preparation on the nutrient balance.

Overview of the knowledge gaps in forest nutrient balances

Currently, the uncertainty in forest nutrients budgets is often large because of limited data on nutrient dynamics of mature stands and the nutrient dynamics in the post-harvest period. Therefore, there is a clear need to assess forest nutrient budgets through utilizing large-scale field experiments (Clarke et al., 2015; Pare and Thiffault, 2016; Titus et al., 2021), for different tree species, soil types, climate zones and forest management practices (Augusto et al., 2002). Limited information of post-harvest nutrient dynamics in forests especially hampers accurate guidelines for sustainable management. As described above, management actions, such as harvest intensity, biomass harvest method, and soil preparation, may potentially have large impacts on nutrient budgets. Harvest intensity affects atmospheric deposition and leaching, but the extent varies by tree species, soil type, and location. Harvest method influences nutrient losses via harvested wood products and leaching, with inconsistent effects of harvest residues on leaching. Soil preparation can affect post-harvest leaching, but the impact depends on tree species and soil properties. To further develop guidelines for sustainable management thereby improving the accuracy of the forest nutrient balance, quantifying post-harvest nutrient dynamics in relation to soil type, tree species, and management practices is essential.

Objectives

The aim of this thesis is to understand the effects of different forest management practices on the deposition, uptake (harvest export), and leaching of nutrients, thereby influencing forest nutrient budgets. This research aims to contribute to the development of science-based guidelines for ecologically sustainable biomass harvest (Fig. 1.2). I tested the hypothesis that the main post-harvest nutrient input and output fluxes are

significantly influenced by tree species and management practices. The main research questions and outline of this thesis are:

1. How is the annual nutrient deposition input modified by forest structure, especially canopy openness (as driven by tree species and harvest intensity), for different nutrients?

In **Chapter 2**, I present and test a new method to measure atmospheric deposition of nutrients underneath forest canopies and in forest gaps. In **Chapter 3**, this method is applied to evaluate the effect of harvest modified canopy openness on the atmospheric deposition of nutrients into forests.

2. What are the present nutrient stocks in the forest and how does this relate to the total nutrient export following different harvest intensities and harvesting methods for different tree species?

In **Chapter 4**, I analyse the effect of tree canopy position on the distribution of biomass and nutrients within trees, and thus create the base for upscaling nutrients stock within trees to nutrient stocks per hectare forest. In **Chapter 5**, I use this upscaling approach to calculate the biomass and nutrients export for European beech, Douglas fir and Scots pine under different harvest intensities (high-thinning, shelterwood and a clearcut system) and harvest methods (stem only harvest, whole tree harvest).

3. How is the annual nutrient leaching in forest soils modified by forest structure (as driven by species and harvest intensity), harvest method and soil preparation for different nutrients?

In **Chapter 6**, I assess nutrient leaching from forest by combining monthly measurements of nutrient concentration in the soil moisture underneath the rooting zone with the soil drainage flux, which is quantified using a mechanistic forest model calibrated on site specific stand and soil data. Soil moisture concentrations were measured within all harvest intensity treatments and for the different harvest methods and soil preparations while the soil drainage was calculated on the level of the harvest intensity treatments.

4. What is the effect of harvesting practices on the nutrient balance shortly after harvest and what are the possible implications over a full rotation period?

In **Chapter 7**, I synthesise the findings of this thesis, creating annual nutrient balances for the second year after harvest for thinnings, shelterwood cut and clearcut following a SOH or WTH harvest and using soil preparation as compared to control plots without harvest. Furthermore, I evaluate if nutrient losses of harvest and leaching can be recovered using literature data describing how elevated post-harvest nutrient fluxes diminish over time.

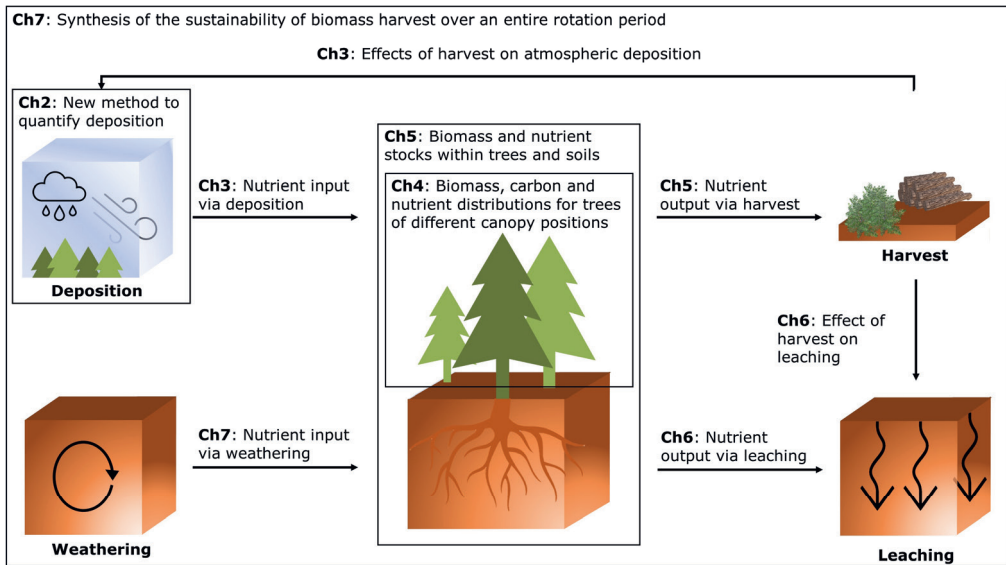


Figure 1.2 Conceptual diagram of this thesis, connecting the research topics of the different chapters. First, I further developed a new method to efficiently measure atmospheric deposition (Ch2). I used this method to measure the nutrient input via atmospheric deposition and the effects of harvest on this atmospheric deposition flux (Ch3). I quantified biomass, carbon and nutrient distributions of trees differing in canopy position (Ch4) in order to calculate precise aboveground biomass and nutrients stocks for trees (Ch5). I measured soil nutrient stocks and evaluated the nutrient output via harvest (Ch5). Next, I measured nutrient output via leaching including the effect of different harvest intensities on leaching (Ch6) and finally, I reviewed literature for nutrient input via chemical weathering (Ch7) and combined all the nutrient fluxes of the second year after harvest which were used to calculate the nutrient balances over entire rotation periods (Ch7).

Research approach

To evaluate the effect of forest harvest on the nutrient balance I installed a manipulative field study in forests on poor and acidified sandy soils in the Netherlands. Our sites represent an extreme case of production forest on low fertile soils receiving high nitrogen deposition inputs. Therefore I lead a team that installed fifteen experimental 1-ha plots in monoculture stands of European beech (*Fagus sylvatica*), Douglas fir (*Pseudotsuga Menziesii*) and Scots pine (*Pinus sylvestris*) which represent commonly harvested tree species in the Netherlands (Probos, 2013; Schelhaas et al., 2022). For each species, I selected 5 homogenous forest stands representing forests located on poor sandy soils in the middle and in the south of the Netherlands (Fig. 1.3), covering both the main aeolian sands in the Netherlands and representing a gradient of nitrogen deposition (RIVM, 2020). The selected forest plots were located on acidic sandy soils classified as Albic or

Entic Podzols or Dystric Cambisols within the texture classes fine sand to loamy medium sand (Table S1.1-S1.3). All study sites are subject to moderately high to high levels of N-deposition with annual atmospheric input ranging between 1200 to 2150 mol N/ha (RIVM, 2020), resulting in accelerated soil acidification, reducing nutrient availability (De Vries et al., 1995; de Vries et al., 2014). These soils are characterized by high nitrogen stocks, ranging between 1000 and 1100 kg ha⁻¹ in the organic layers (**Ch5**). The plots were characterized by a similar temperate, maritime climate with an interpolated 30-year average annual rainfall and temperature of 850 mm and 10.6 °C respectively (KNMI, 2021b). The forest in the plots consisted of relatively homogeneous, even-aged, single-tree species, planted forests between 50 and 120 years ago with a soil cover dominated by either foliar litter or mosses (Table S1.1). The dominant species within a stand took up more than 80% of the total crown cover and basal area. All plots had previously been managed following common silvicultural methods in the Netherlands. Thinning regimes started with thinning from below (removal of suppressed trees) and, in the last three decades, all stands were treated using high-thinning (removal of trees directly competing with future crop trees).

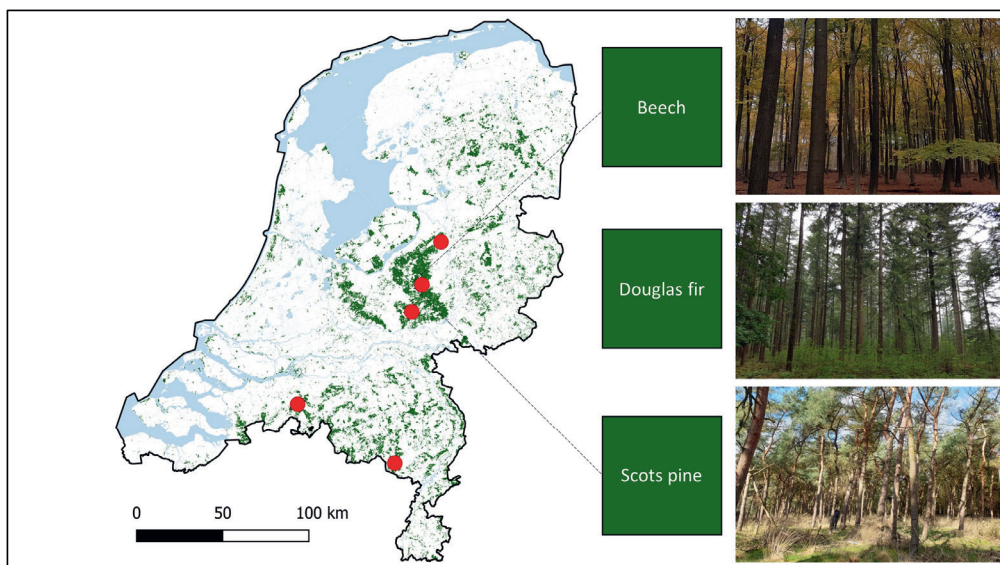


Figure 1.3 Locations of the experimental sites selected for measurements of the atmospheric deposition, tree and soil nutrient stocks, exports following harvest and leaching in this thesis. A) Map showing the locations of the experimental sites, different numbers denote the different study sites. The nationwide forest cover (in total 10% of the land area of the Netherlands) is shown in green (PDOK, 2015). B) In each location were three monoculture stands selected of respectively beech, Douglas fir and Scots pine. For each species, a picture representing such stands is shown.

In each of the 15 selected forest stands I installed a 1-ha plot in which 4 subplots were established, to which one of the four harvest intensities was assigned randomly (block design): control (no harvest), high-thinning (~20% of the basal area removed), shelterwood (~80% removed) and clearcut (100% removed). Harvest intensity was determined based on basal area reductions, whereby species-specific target basal areas per treatment were used (Table S1.4). Within the high-thinning plots two equal subplots (25 * 50m) were installed to which the harvest methods SOH and WTH were randomly assigned. For the shelterwood and clearcut harvest intensity plots a cross-design was installed consisting of SOH or WTH biomass harvest and flail mulching (yes/no) resulting in four equal subplots (25 * 25m). Harvest took place in February-March 2019, flail mulching was applied after harvest in March-April 2019. An example of the design of one of the experimental sites is shown in Fig. 1.4.

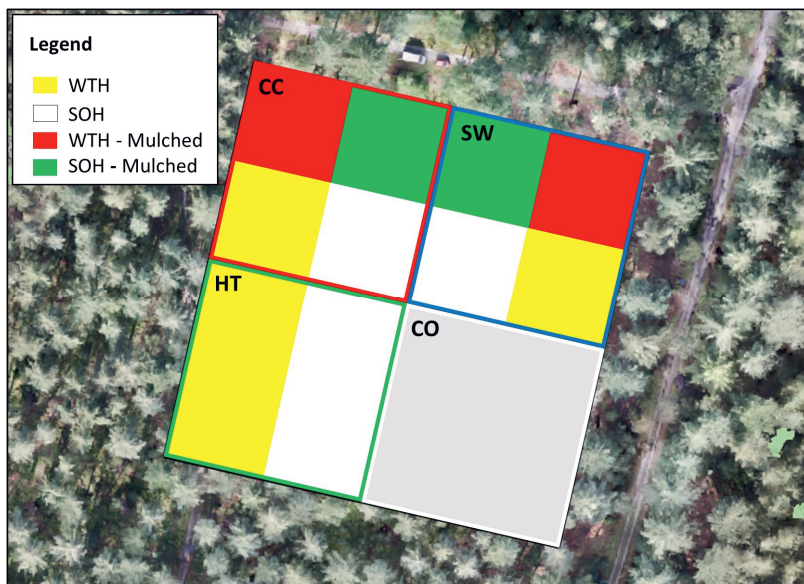


Figure 1.4 Overview of one of the 15 experimental plots. At each experimental site, a 1-ha plot was selected and the harvest intensity treatments high-thinning (HT), shelterwood (SW) and clearcut (CC) together with the unharvested control (CO) were randomly assigned. Within the HT harvest intensity, two equal subplots were installed for the harvest methods WTH and SOH. For the harvest intensities SW and CC, a cross-design of four plots was installed to which the treatments WTH, SOH and mulching (yes/no) were assigned.

Glossary

Definition of important concepts presented in this thesis.

General	
Biomass harvest	Harvest of the aboveground tree parts from forests.
Harvest intensity	The used silvicultural system with increasing biomass extraction ranging from high-thinning towards shelterwood and clearcut.
Harvest method	The tree parts extracted from the forests. Three different harvest methods are distinguished: stems are removed in case of stem only harvest (SOH) stems without bark are removed in stem wood harvest (SWH), and stems and crowns are removed in whole tree harvest (WTH).
Soil preparation	Post-harvest site preparation in order to facilitate natural regeneration or planting. The site preparation considered in this thesis consist of shredding the harvest residues to smaller pieces and is defined as flail mulching.
Canopy openness treatment	Related to the harvest intensity. High-thinning generally leads to a canopy openness of $\pm 20\%$ while canopy openness of the shelterwood and clearcut is respectively $\pm 80\%$ and 100% .
Experimental design	
Plot	The 1-ha experimental plot installed in 5 homogenous forest stands of beech (<i>Fagus sylvatica</i>), Douglas fir (<i>Pseudotsuga menziesii</i>) and Scots pine (<i>Pinus sylvestris</i>).
Subplot	Each plot was divided into four equal subplots for the harvest intensity treatments high-thinning (HT), shelterwood (SW) and clearcut (CC) and the unharvested control (CO)

Supplementary information

Table S1.1 Overview of stand and soil characteristics of the beech, Douglas fir and Scots Pine forests of this study. The forest stand property density includes all trees with DBH > 10 cm, Dg is the root mean square DBH and H is the height based on 16 dominant trees per stand. The soil cover values are coverage percentages for the foliar litter, mosses, graminoids and ferns. Mineral soil properties include the pH_{H2O}, the soil bulk density (BD), the average Al-S saturation of cation exchange capacity (CEC) by Al per hectare and the average soil base saturation (BS) per hectare. Both Al-S and BS are based on the unbuffered CEC, with "b.d." in the case where Ca, K, Mg and Na were below the detection limit and undetectable. Soil profiles were classified based on the field records according to the international standards (IUSS Working Group WRB 2015).

Site	Species	Forest stand			Soil cover					Mineral soil (0–30 cm depth)				
		Age	Density	Dg	H	Foliar litter	Mosses	Graminoids	Ferns	pH	BD	Al-S	BS	Soil type
		(yr.)	(n ha ⁻¹)	(cm)	(m)	%	%	%	%		g cm ⁻³	%	%	
1	Beech	94	260	34	22	100	0	0	0	4.3	1.0	96	2.1	Albic Podzol
2	Beech	100	140	47	26	98	2	0	0	4.3	1.0	100	b.d.	Entic/Albic podzol
3	Beech	82	220	38	25	100	0	0	0	4.2	0.98	94	1.3	Dystric Cambisol
4	Beech	98	240	34	24	97	3	0	0	4.5	1.3	83	10	Dystric Cambisol
5	Beech	46	1100	18	19	100	0	0	0	4.1	1.1	77	1.3	Dystric Cambisol
1	Douglas fir	74	120	54	41	28	72	0	0	4.2	1.3	68	3.3	Albic podzol
2	Douglas fir	59	170	50	37	37	63	0	0	4.0	1.1	75	2.0	Entic/Albic podzol
3	Douglas fir	60	140	52	37	26	69	2	0	4.2	1.2	100	3.8	Entic/Albic podzol
4	Douglas fir	66	150	51	36	93	5	0	0	4.0	1.1	74	4.0	Entic/Albic podzol
5	Douglas fir	60	240	44	28	3	2	0	95	4.2	1.2	91	7.1	Albic podzol
1	Scots pine	55	420	25	19	51	49	0	0	4.3	1.2	79	1.3	Albic podzol
2	Scots pine	48	430	24	18	54	37	9	0	4.1	0.97	88	3.2	Albic podzol
3	Scots pine	47	880	17	19	34	59	7	0	4.4	1.4	88	5.0	Dystric Cambisol
4	Scots pine	62	450	26	21	36	59	5	0	4.0	0.98	69	4.3	Albic podzol
5	Scots pine	73	470	27	16	31	24	45	0	4.2	1.1	92	5.0	Albic podzol



Table S1.2 Texture and soil type classified based on the WRB guidelines (WRB, 2015) for the five sites of beech (BE), Douglas fir (DG) and Scots pine (SP).

Site	Species	Depth	Silt	Very fine sand	Fine sand	Medium sand	Coarse sand	Type
			%	%	%	%	%	
			<63 µm	62.5-125 µm	125-250 µm	250-500 µm	500-1000 µm	
1	BE	0-30	17	10	24	35	13	Loamy medium sand
		40-50	25	7.5	20	31	16	
2	BE	0-30	26	15	20	29	9.8	Loamy medium sand
		40-50	29	9.5	18	31	12	
3	BE	0-30	35	20	16	21	6.9	Loamy medium sand
		40-50	45	12	15	21	7.8	
4	BE	0-30	12	24	45	18	0.5	Medium sand
		40-50	36	13	32	17	0.9	Loamy medium sand
5	BE	0-30	12	25	45	18	0.8	Medium sand
		40-50	21	17	39	21	1.8	Loamy medium sand
1	DG	0-30	12	11	31	35	10	Medium sand
		40-50	11	4.9	24	42	18	
2	DG	0-30	29	20	18	24	8.6	Loamy medium sand
		40-50	33	11	20	26	10	
3	DG	0-30	18	22	39	19	1.8	Loamy medium sand
		40-50	19	12	39	26	2.9	
4	DG	0-30	22	33	35	9.9	0.6	Loamy medium sand
		40-50	23	28	37	12	0.4	
5	DG	0-30	16	28	42	14	0.8	Medium sand
		40-50	13	24	47	15	0.2	
1	SP	0-30	17	16	29	30	8.2	Loamy medium sand
		40-50	18	11	26	33	12	
2	SP	0-30	28	26	25	17	2.9	Loamy medium sand
		40-50	34	19	26	18	1.9	
3	SP	0-30	6.2	26	50	17	0.3	Fine sand
		40-50	2.2	16	57	24	0.4	
4	SP	0-30	22	32	34	11	0.5	Loamy medium sand
		40-50	26	23	36	15	0.5	
5	SP	0-30	14	26	46	14	0.2	Medium sand
		40-50	4.9	19	58	19	0.1	Fine sand

Table S1.3 Total nutrient stocks (in kg ha⁻¹) in the mineral soil based on the total soil nutrient concentrations and bulk density of the top layer of the mineral soil (0 – 30 cm depth) and the sub layer of the mineral soil (40-50 cm depth). Data is shown for the five sites of beech (BE), Douglas fir (DG) and Scots pine (SP). The top layer (4 merged samples per site) was sampled in 2018-2019 (pre-harvest), the sub layer (1 merged sample per site) in 2020-2021 (post-harvest). Soil sampling procedure is described in **Ch5**.

Site	Species	Depth	P	S	K	Ca	Mg	Mn	Cu	Fe	Zn
1	BE	0-30	230 ± 18	300 ± 15	530 ± 48	250 ± 40	460 ± 77	68 ± 7	3.1 ± 0.072	8000 ± 760	14 ± 1.7
		40-50	250	290	960	480	1300	69	3.1	13000	22
2	BE	0-30	280 ± 26	410 ± 38	760 ± 35	290 ± 28	670 ± 92	58 ± 4.6	3.8 ± 0.62	14000 ± 1200	21 ± 2.5
		40-50	270	320	900	380	1100	85	3.1	15000	31
3	BE	0-30	380 ± 5.4	600 ± 11	960 ± 55	280 ± 28	740 ± 84	79 ± 5.9	5.8 ± 0.056	16000 ± 510	25 ± 1.5
		40-50	360	430	1100	370	1100	110	2.9	15000	29
4	BE	0-30	280 ± 24	350 ± 29	2100 ± 220	630 ± 74	2100 ± 380	120 ± 28	17 ± 3.2	14000 ± 2300	46 ± 9.6
		40-50	340	290	3000	1400	3400	220	23	20000	80
5	BE	0-30	240 ± 5.2	420 ± 26	440 ± 42	120 ± 6.8	180 ± 25	16 ± 1.1	10 ± 0.55	2200 ± 200	27 ± 2.1
		40-50	330	360	560	160	260	20	3.2	2700	16
1	DG	0-30	270 ± 30	270 ± 16	430 ± 24	200 ± 18	230 ± 39	19 ± 3.7	3.7 ± 0.035	4600 ± 1000	9.3 ± 1.9
		40-50	340	210	500	230	340	25	3.7	6700	11
2	DG	0-30	390 ± 57	500 ± 61	780 ± 120	330 ± 58	690 ± 200	110 ± 56	5.8 ± 1.7	15000 ± 2100	22 ± 5.9
		40-50	380	320	1100	550	1500	230	3.3	19000	40
3	DG	0-30	380 ± 31	400 ± 12	900 ± 32	590 ± 29	790 ± 25	140 ± 24	3.6 ± 0.14	16000 ± 2000	20 ± 0.92
		40-50	370	320	880	600	920	180	3.6	18000	25
4	DG	0-30	360 ± 78	460 ± 29	570 ± 32	250 ± 21	350 ± 50	38 ± 3.8	6.5 ± 1.4	5000 ± 590	14 ± 0.75
		40-50	380	350	640	250	410	32	3.2	6300	13
5	DG	0-30	550 ± 48	440 ± 29	910 ± 66	430 ± 43	570 ± 63	72 ± 20	33 ± 4.4	9200 ± 1300	155 ± 23
		40-50	550	310	1200	390	720	80	3.6	9900	68
1	SP	0-30	190 ± 32	300 ± 32	410 ± 34	160 ± 13	210 ± 83	24 ± 3.8	3.6 ± 0.19	6900 ± 2400	7.9 ± 1.4
		40-50	260	250	570	250	440	38	3.6	8700	11
2	SP	0-30	360 ± 25	530 ± 18	590 ± 56	260 ± 30	340 ± 75	38 ± 7.3	5 ± 0.75	11000 ± 1500	19 ± 3.0
		40-50	330	340	1100	510	1200	110	2.9	14000	26
3	SP	0-30	230 ± 5.9	250 ± 11	1600 ± 63	1000 ± 36	1500 ± 73	88 ± 2.8	4.1 ± 0.068	8700 ± 300	24 ± 1.1
		40-50	330	100	2300	1700	1800	150	4.1	8400	29
4	SP	0-30	250 ± 33	500 ± 29	470 ± 27	150 ± 13	220 ± 35	21 ± 4.2	5.1 ± 0.72	3000 ± 26	8.7 ± 0.11
		40-50	210	230	690	190	460	23	2.9	3400	8.7
5	SP	0-30	400 ± 25	370 ± 24	710 ± 66	200 ± 6.0	340 ± 49	29 ± 2.1	14 ± 0.53	8900 ± 1200	72 ± 3.1
		40-50	630	220	1400	340	830	45	3.4	12000	37

Table S1.4 Average basal area $\pm$ s.e. ($\text{m}^2 \text{ha}^{-1}$), realized target basal area ($\text{m}^2 \text{ha}^{-1}$) and basal area reduction for the timber harvest intensities high-thinning and shelterwood for European beech, Douglas fir and Scots pine. Target basal area of the clearcut was 0 for all species with a reduction of 100%. Basal area reductions per treatment per forest stand are in Table S1.1.

Species	Average BA	High-thinning			Shelterwood			Clearcut
		Target BA	Reduction		Target BA	Reduction		Reduction
	$\text{m}^2 \text{ha}^{-1}$	$\text{m}^2 \text{ha}^{-1}$	%	$\text{m}^2 \text{ha}^{-1}$	$\text{m}^2 \text{ha}^{-1}$	%	$\text{m}^2 \text{ha}^{-1}$	$\text{m}^2 \text{ha}^{-1}$
Beech	25 ± 0.86	17	18	4.6 ± 0.28	4.5	76	19 ± 0.71	24 ± 0.96
Douglas fir	32 ± 1.6	23	20	6.4 ± 0.40	5.0	78	25 ± 1.1	32 ± 1.5
Scots pine	23 ± 1.2	18	16	3.7 ± 0.47	4.1	83	19 ± 1.0	22 ± 0.65



Chapter 2

Testing Ion Exchange Resin for
quantifying bulk and throughfall
deposition of macro and
micro-elements on forests

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Marcel Hoosbeek & Frank J. Sterck.

Abstract

Atmospheric deposition is a major nutrient influx in ecosystems and high anthropogenic deposition of, for example, nitrogen may disrupt ecosystem functioning at large scales. Quantification of the deposition flux is required to understand the impact of such anthropogenic pollution on ecosystems. However, current methods to measure nutrient deposition are costly, labor intensive and potentially inaccurate, especially for nitrogen due to transformation processes. Ion Exchange Resin (IER) appears a promising cost-and labor-effective method that is reliable for nitrogen. The IER-method is potentially suited for deposition measurements on coarse time scales and for areas with little rainfall and/or low elemental concentrations. The accuracy of the IER-method is, however, hardly classified beyond nitrogen. We tested the IER-method for bulk deposition and throughfall measurements of macro and micro-elements by testing the resins adsorption capacity, recovery efficiency and behavior for field conditions.

We show that IER is able to adsorb 100% of Ca, Cu, Fe, K, Mg, Mn, P, S, Zn and NO_3 and >96% of P and Na. Loading the resin beyond the capacity resulted mainly in losses of Na, P, NH_4 while losses of Ca, Cu, Fe, Mg, Mn and Zn were hardly detected. Heat (40°C), drought and frost (-15°C) reduced the adsorption of P by 25%. Elemental recovery was close to 100% for NH_4 and NO_3 using KCl (1 or 2M) while high (83-93%) elemental recoveries of Ca, Cu, Fe, K, Mg, Mn and S were found using HCl as an extractant (2-4M). Drying the resin prior to extraction and using a shake-drip extraction method increased the recovery efficiencies. We found good agreement between the conventional bulk deposition method and the IER-method for field conditions although IER generally resulted in higher deposition estimates. These higher deposition estimates can be related to absence of biological reactions and lower uncertainties especially for elements with low deposition concentrations. Overall, IER is a powerful tool for the measurement of bulk deposition and throughfall of a broad range of elements.

1 Introduction

Atmospheric deposition is a major nutrient influx in many ecosystems and therefore crucial for ecosystem functioning (Van Langenhove et al., 2020). However, due to anthropogenic pollution, atmospheric deposition can potentially disrupt ecosystem nutrient balances, leading to exceedance of critical deposition thresholds of for example nitrogen which can in turn degrade ecosystem functioning (Rabalais, 2002; de Vries et al., 2011). Such degradation of ecosystems involves accelerated soil acidification and reduced availability of critical soil nutrients, such as base cations, which has detrimental impacts on biodiversity and water quality (Houdijk et al., 1993; Johansson et al., 2001; Stevens et al., 2004; Bowman et al., 2008; Horswill et al., 2008; Solberg et al., 2009; de Vries et al., 2014b; Lu et al., 2014). Atmospheric deposition is therefore of major importance to many ecosystems and monitoring deposition is necessary for policy, management, and conservation.

Measurements of atmospheric deposition are, however, costly and labor intensive. Direct measurements of dry deposition (i.e. input of elements as airborne particles) and wet-only deposition (i.e. input of elements via precipitation) (Lovett and Reiners, 1986; Balestrini et al., 2007) are scarce and current technology limits widespread measurements. For forests, the common method to assess total deposition (i.e., wet and dry deposition combined) is the collection of precipitation below forests, called throughfall, in collection devices of various shapes and sizes, while accounting for canopy exchange (Draaijers et al., 1996a; Thimonier, 1998), which is based on the additional measurement of precipitation outside the forest, known as bulk deposition. The combined measurement of nutrient inputs in precipitation below and outside forests, further called in this paper the bulk deposition method, is readopted in many monitoring networks (i.e. ICP forest network (Bleeker et al., 2003; de Vries et al., 2003; De Vries et al., 2007), the DONAIRE network (Pey et al., 2020) and the nationwide monitoring network in China (Xu et al., 2019)). However, the use of bulk deposition measurements requires frequent (up to weekly) sampling as ammonium in the collected rainwater may relative rapidly be transformed to nitrate by nitrification, with the speed being dependent on local weather conditions (Nicholas Clarke and König, 2016). The high sampling frequency and the high cost of traveling and laboratory analysis, limits the spatial and temporal scales at which this method can be applied. The alternative is larger sampling intervals, but this may cause inaccurate assessment of the input, especially of ammonium versus nitrate. An adequate assessment of both N compounds is especially crucial in regions where the allocation of N sources is highly sensitive (ammonium being caused by NH₃ emissions from agriculture and nitrate from NO_x emissions by traffic and industry). Better alternatives are needed to measure deposition efficiently in the field, improve the reliability of the measurements, reduce sampling effort and costs, and thus allow for more effective large-scale deposition monitoring programs.

The Ion Exchange Resin method (IER) was previously developed to measure bulk deposition at large spatial and temporal scales, but use of the method is yet limited to remote areas (Brumbaugh et al., 2016) the monitoring network of California (Fenn et al., 2018) or case studies (Clow et al., 2015; Garcia-Gomez et al., 2016; Hoffman et al., 2019). Wide-spread application of the IER-method is promising as the method allows to measure the accumulated deposition over long time periods (up to a year), which strongly reduces both the sampling effort in the field and the number of lab analysis, leading to major cost savings (Fenn and Poth, 2004; Kohler et al., 2012). Furthermore, the method is more reliable for nitrogen as the resin inhibits mineralization, nitrification and denitrification as affected by local weather conditions (Fenn and Poth, 2004; Kohler et al., 2012). Finally, the IER-method is able to measure the deposition in areas with low rainfall or low elemental concentrations, avoiding problems with detection limit and minimal sample size required in the bulk deposition method (Kohler et al., 2012). Because of these advantages, the IER-method is already more commonly used in other research fields ranging from analysis of available nutrients in soil water fluxes to purification of waste-water (Sibbesen, 1977; Binkley and Matson, 1983; Crabtree and Kirkby, 1985; Rengaraj et al., 2001; Verbych et al., 2005; Risch et al., 2020).

The IER-method is most commonly used for NH_4 and NO_3 measurements (Fenn et al., 2002; Fenn and Poth, 2004; Fang et al., 2011; Kohler et al., 2012; Clow et al., 2015; Garcia-Gomez et al., 2016; Hoffman et al., 2019), but few studies reported measurements of other elements (e.g. S, K, Ca, Mg, Na and Cl) (Van Dam et al., 1987; Simkin et al., 2004; Fenn et al., 2018). The applicability of the method to measure a broad range of elements depends on the performance of the resin, measured as the adsorption capacity (percentage of the total element flux bound to the resin) and the recovery efficiency of elements (percentage of the total element flux recovered from the resin) (Garcia-Gomez et al., 2016). Often though the adsorption capacity and recovery efficiencies are not reported (Fenn and Poth, 2004; Boutin et al., 2015; Fenn et al., 2015; Risch et al., 2020). Studies reporting the adsorption capacity (Simkin et al., 2004; Fang et al., 2011; Garcia-Gomez et al., 2016) describe only the adsorption of a limited number of elements under laboratory conditions. Recovery efficiency under laboratory conditions is more often reported, with in general high recovery efficiencies (87-100%) although the recovery of some macro elements (i.e. Ca and Mg) was below 50% (Simkin et al., 2004; Fang et al., 2011; Kohler et al., 2012; Clow et al., 2015; Wieder et al., 2016; Cerón et al., 2017). Despite the promising applicability, the adequacy of the IER-method to derive bulk deposition and throughfall under field conditions is hardly tested. The limited information on adsorption capacity combined with bad recoveries for some elements (i.e., Ca, Mg, Fe and Al) potentially limits the use of the IER-method for bulk deposition measurements.

The adsorption capacity and the recovery efficiency can be influenced by environmental field conditions like drought, frost or high temperatures (Qian and Schoenau, 2002; Bayar et al., 2012). However, there is hardly any study testing the influence of environmental field conditions on both the

adsorption capacity and the recovery efficiency of the resin. Furthermore, most tests refer to bulk deposition, whereas atmospheric deposition on forest is also measured as throughfall underneath vegetation canopies. Dissolved organic substances are higher in throughfall than in bulk deposition for which the adsorption capacity of the resin is lower (Langlois et al., 2003). Overall, recovery rates from resin exposed to environmental field conditions appear to be lower, urging the need for better evaluation of IER performance under field conditions (Krupa and Legge, 2000; Brumbaugh et al., 2016). Therefore, new tests are necessary to evaluate the effect of environmental conditions and organically-rich throughfall on the elemental recovery from the resin.

The recovery efficiency can be optimized by the use of different extraction methods. An often used extraction method is 2M KCl for nitrogen extraction (Fenn et al., 2002; Garcia-Gomez et al., 2016; Hoffman et al., 2019), but also combinations of either KI, HNO_3 , NaCl, H_2SO_4 and HCl were used (Van Dam et al., 1991; Kohler et al., 2012; Brumbaugh et al., 2016; Fenn et al., 2018). The KCl extraction method and the KI extraction method do not allow measurements of K deposition and are, as high dissolved salt solutions, problematic for measurements using inductively coupled plasma-atomic emission spectrometers (ICP-AES) (Hislop and Hornbeck, 2002; Brumbaugh et al., 2016). New tests are therefore needed to increase the recovery efficiency allowing to measure a broad range of elements including all macronutrients and micronutrients.

In this study we aim to test the capacity of IER as a method to quantify atmospheric deposition for a broad range of macro- and micro-elements, comparing results under laboratory and field conditions and in the latter case comparing bulk deposition and throughfall. We first tested the method under controlled laboratory conditions to indicate efficient resin volumes and to assess adsorption capacities and recovery efficiencies. Next, the behavior of the IER-method was tested under field conditions covering a gradient from closed forests to open areas to account for the effect of dissolved organic substances on the performance of the resin columns. From this, we provide different methodological protocols with accuracies for detecting different macro- and micro elements under field conditions such as forests.

2 *Methods*

2.1 *Preparation of the resin columns*

We prepared 45 resin columns for the laboratory tests of elemental adsorption and recovery, followed by the preparation of 30 columns for the field test of the IER-method. First, the resin columns were cleaned using 0.2M HCl and demineralized water. Then, the cleaned and dried resin columns were washed three times with demineralized water in the laboratory prior to filling the column with IER. To fill the columns, a plug of clean polyester fiber was placed inside the resin column and pushed to the bottom. A cleaned cap was screwed loosely at the bottom of the resin column, stabilizing the polyester plug. The resin column

was placed vertically above a container to collect the reagents. The ion exchange resin (Amberlite IRN 150, a 1:1 mixture of H^+ and OH^-) was washed with 8L demineralized water in batches of 500g of resin to remove small particles within the resin and to remove the resins smell which could attract animals. All liquids were drained from the resin using a vacuum pump, and for each resin column 9.8g of resin was weighted out and poured in the resin column using a pipette with demineralized water. When excess water had passed through, a second plug of polyester fiber was placed on top of the resin and both sides of the column were screwed tightly with cleaned caps. A schematic overview of these steps is in figure 2.1.

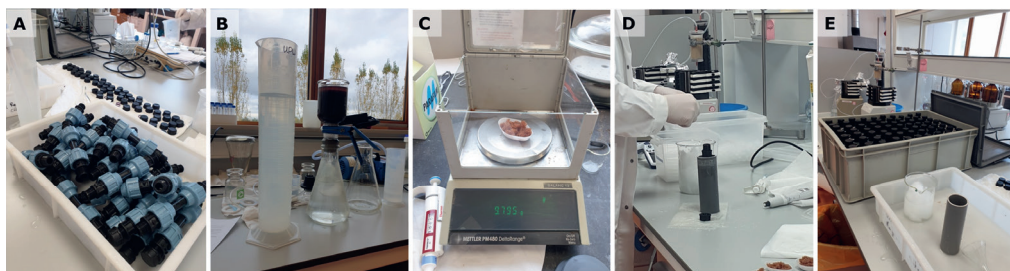


Figure 2.1 Preparation of the resin columns. A: Cleaned resin columns prior to filling with IER. B: cleaning of the Amberlite IRN-150 exchange resin using a vacuum pump. C: Weighing the resin prior to filling the resin column. D: Resin column stabilized in a holder during filling with resin. E: Overview of filled resin columns with the resin column stabilizer and the polyester plugs shown in the front.

2.2 Laboratory tests

The adsorption capacity and recovery efficiency of the IER (Amberlite IRN 150 H^+ and OH^- form) was tested at the Soil Chemistry Laboratory (CBLB), Wageningen University. First, based on existing deposition data (RIVM, 2015), we estimated the bulk deposition amounts ($kg\ ha^{-1}$) for different elements, and then used those to determine the needed molarity of the solution that was used to test the adsorption capacity of the resin. Both the adsorption capacity and recovery efficiency were subsequently tested for annual maximum bulk deposition rates across the Netherlands of the following elements: PO_4^{2-} , SO_4^{2-} , $N-NO_2^- + N-NO_3^-$, $N-NH_4^+$, Ca^{2+} , Mg^{2+} , K^+ , Na^+ , Fe^{2+} , Mn^{2+} , Cu^{2+} and Zn^{2+} .

To estimate the maximum bulk deposition values, the monthly measurements of existing bulk deposition data of representative stations ($\mu mol\ l^{-1}$) (RIVM, 2015), were summed to seasonal concentrations for the funnel surface and the stations were selected with the highest seasonal deposition, occurring during summer, for both macro- and micronutrients, based on the total molarity of the rainwater. Thereafter, the deposition of the summer was multiplied by 2, which is an average correction factor to convert bulk deposition to throughfall (Table S2.1). The concentration of this throughfall flux, multiplied by 4 (assumed that the summer values are representative of the entire year, which is a precautionary

approach), was dissolved in a 1 L solution separately for macro and micronutrients using stock solutions of Na_2SO_4 , NaCl , KNO_3 , KH_2PO_4 , NH_4NO_3 , NH_4Cl , MgSO_4 , CaCl_2 for the macronutrient solution and using stock solutions of FeCl_2 , $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$, $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and $\text{MnSO}_4 \cdot \text{H}_2\text{O}$ for the micronutrient solution. The solution used to test the adsorption and recovery capacity of the resin consisted of 400 μmol Ca, 10 μmol Cu, 2500 μmol Cl, 400 μmol Fe, 200 μmol K, 300 μmol Mg, 150 μmol Mn, 2300 μmol Na, 20 μmol PO_4 , 750 μmol SO_4 , 20 μmol Zn, 4000 μmol NH_4 and 2000 μmol NO_3 .

The adsorption capacity (i.e., percentage of total elemental influx adsorbed by the resin) was tested using 18 of the 45 resin columns for laboratory tests. Out of these 18 columns, 9 columns were used to mimic heat, drought, and frost conditions and 9 columns were used to test the columns capacity (Table S2.2). Heat, drought, and frost conditions were mimicked using 3 columns for each treatment which consisted of heating to 40°C, drying at 20°C to a constant weight and freezing at -19°C for 72 hours, respectively, followed by drip-wise loading with the macro- and micro solution. The resins capacity was simulated by dripping the macro- and the micro solutions through the resin columns using the normal concentration (3 columns and for the heat, drought and frost conditions), the double concentration (3 columns) and the triple concentration (3 columns), loading the columns up to respectively 70%, 140% and 210% of their capacity. Samples of the leachate were taken when all the solution was drained from the resin (after approximately 4 hours). Three loaded resin columns were thereafter flushed with demineralized water to test the stability of the adsorption. Both the samples of the leachate and the demineralized water used to wash the loaded columns, were analyzed for N-NH_4 , and $\text{N-NO}_2 + \text{N-NO}_3$ content using a Segmented Flow Analyzer (SFA type 4000, Skalar Analytical B.V., the Netherlands), and the content of Ca, Cu, Fe, Mg, Mn, Na, total-P, S and Zn using the ICP-AES (Thermo-Scientific iCAP 6500 DUO, USA).

The recovery efficiency (i.e., percentage of total elemental flux recovered from the resin) was tested using 39 out of the 45 resin columns for laboratory tests, re-using 12 columns from the adsorption capacity test. Only the columns loaded with the double and triple concentration of the macro- and micro solutions were excluded (Table S2.2). All unloaded columns were, similar to the previously loaded columns, drip-wise loaded with the macro- and micro solutions. Recovery efficiency was tested using a 2M KCl extraction for NH_4 and NO_3 based on previous reported high recovery rates (Fenn et al., 2002; Fenn and Poth, 2004; Fang et al., 2011; Kohler et al., 2012; Clow et al., 2015; Hoffman et al., 2019) and multiple molarities of HCl (ranging from 1 to 4M) for the other elements (Ca, Mg, K, Fe, Mn, Zn, Cu, Na, and S) since previous studies reported that low recovery efficiencies might be related to the molarity of the extraction solution used (Fenn et al., 2018). For both the KCl and HCl extractions, we varied the extraction volume, the extraction type, and the extraction method (Table 2.1).

Extraction volumes used were respectively 50 mL, 100 mL and 150 mL and extraction type was either single column extraction or batch extraction. Using the single column extraction type, the extractant

was applied on the entire column while in batch extraction the resin was divided into smaller samples. These subsamples of the resin were either fresh (i.e., solution drained resin) or dried at 28 °C to a constant weight (Table 2.1). Drying of the resin facilitates subsampling and the calculation of the deposition flux. The extraction method was either drip, in which the extractant was slowly dripped over the resin, or a shake-drip combination in which the resin was shaken in 50mL of the extractant for 1 hour and the remaining extractant was dripped over the resin.

Table 2.1 Overview of the test for effective extraction of the ion exchanger. The KCl extraction was used for the extraction of NH₄ and NO₃ while the HCl extraction is used for the extraction of Ca, Mg, K, Fe, Mn, Zn, Cu, Na, and S. The single column extraction included the entire loaded column (9.8g of resin) while for batch extraction a subsample (avg. 2.5g dried resin) was used which was extracted either fresh (i.e., solution drained) or dried.

Extraction fluid			Type	Resin	Samples	Method
	<i>M</i>	<i>mL</i>				
KCl	2	50	Single column	Fresh	3	Drip
	2	100	Single column	Fresh	3	Drip
	2	50	Batch	Fresh	2	Drip
	2	50	Batch	Dried	2	Drip
	2	50	Batch	Dried	2	Drip
HCl	1	50	Single column	Fresh	3	Drip
	1	100	Single column	Fresh	3	Drip
	2	100	Batch	Fresh	2	Drip
	2	100	Batch	Dried	2	Drip
	2	100	Batch	Fresh	2	Shake & drip
	2	100	Batch	Dried	2	Shake & drip
	4, 2 & 1	150	Batch	Fresh	2	Drip
	4, 2 & 1	150	Batch	Dried	2	Drip
	1 & 2	100	Batch	Fresh	1	Drip
	1 & 2	100	Batch	Dried	1	Drip
	2.5	100	Batch	Dried	2	Shake & drip
	3	100	Batch	Dried	2	Shake & drip
	3.5	100	Batch	Dried	2	Shake & drip
	4	100	Batch	Dried	1	Shake & drip

2.3 Field tests

To evaluate the accuracy of the IER-method to quantify bulk deposition and throughfall, a field study was carried out in the Netherlands (GPS 52.015745, 5.759924) in which we collected paired observations of bulk deposition and throughfall using water samples (referred to as the water-method) and the IER-method. The chosen field site consisted of a mature stand of European beech (*Fagus sylvatica*) which has

been harvested at different intensities in February 2019, which resulted in four ¼-ha plots within the same stand: an unharvested control (~0% canopy openness), a high-thinning (~25% canopy openness), a shelterwood (~75% canopy openness) and a clearcut (100% canopy openness) (Vos et al., 2023a; Vos et al., 2023b). The different harvest intensities allow to test the method for quantifying bulk deposition and throughfall including the effect of organic substances on the performance of the IER-method. The forest stand has a temperate maritime climate with a mean annual temperature of 10.4°C and a mean annual rainfall of 805mm (KNMI, 2022). An overview of the study site characteristics and the placement of the paired samples are in Table 2.2.

Table 2.2 Characterization of the study site and placement of the paired samplers in open gaps (bulk deposition) and underneath the forest canopy (throughfall).

Treatment	Canopy cover	Number of trees trees ha ⁻¹	Paired samplers	
	%		Bulk deposition	Throughfall
Control	94	245	6	1
High-thinning	72	180	5	2
Shelterwood	16	32	2	5
Clearcut	0	0	0	7

In each forest harvest treatment plot, 7 pairs of collectors were installed resulting in 28 common deposition collectors and 28 IER deposition collectors. These 7 collectors per plot had a collection surface >2000cm² above which the reliability of the measurement is significantly increased (Bleeker et al., 2003). The collectors consisted of a polyethylene funnel mounted to a resin column, which was filled with resin for the IER-method but left empty for the water-method, and a PVC hose connecting the resin column to a polypropylene water reservoir (Fig. 2.2A). The funnel had a surface of 288cm² (including half the rim, Fig. 2.2B) and the resin column (volume of 15.7ml) had an inner diameter and length of respectively 12.4 and 130mm. Both the funnel and the resin column were chemically resistant and not susceptible to damage through UV-light or low temperatures. Wire couplings, in which a mesh with the size of 2mm was mounted, were used to connect the resin column to the funnel and to a hose-tail (Fig. 2.2C, Fig. 2.2D). Prior to field installation, the funnel and the resin column including the wire couplings were cleaned from chemicals loosely bound to the surface by submerging into a 0.2M HCl solution for three hours, followed by a 15-hour immersion in demineralized water which was continuously refreshed. Afterwards, the compartments were allowed to dry in a clean room and stored in clean plastic bags.

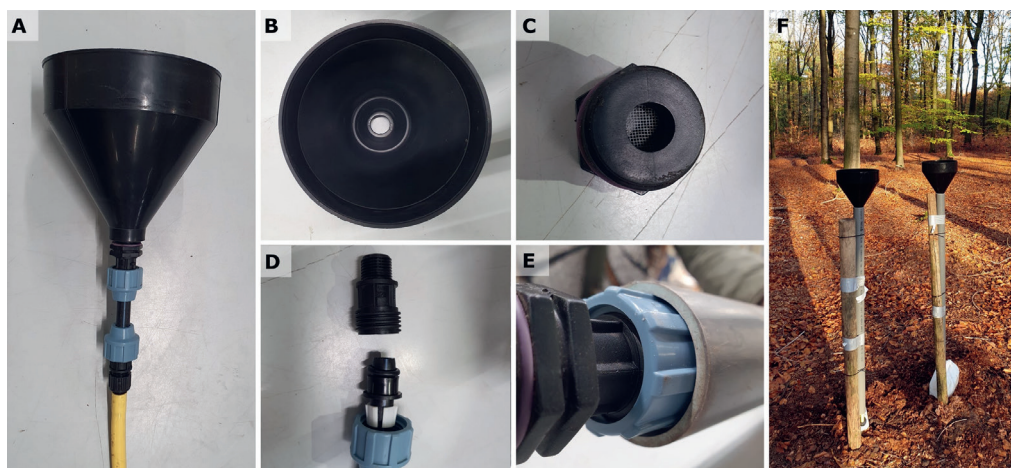


Figure 2.2 Construction of the deposition samplers. A: the connected sampler ready for use in the field. B: the used funnel with a collection surface of 288cm². C: The wire coupling between the funnel and the resin column containing a mesh to prevent larger objects entering the resin column. D: Overview of the resin column with the wire couplings. E: the resin column fitted tightly into the PVC tube which allowed easy installation of the resin columns in the field.

Field placement of the collectors was based on a digital elevation map of the canopy cover, assessed by drone-based photogrammetry (camera FC220). This digital elevation map was converted to a canopy cover map using 'reclassify' in ArcMap (version 10.6.1) in which all datapoints above 10m were assigned to be covered by canopy. Each plot was, thereafter, divided into an equal sized, seven block grid and the locations of the collectors were determined in each of these blocks using random points reflecting the canopy cover (%). Samplers were installed in the field using those random points on November 6th, 2019, by placing the clean, connected sampler in the holder (PVC-tube) and connecting the sample to the partly buried reservoir (Fig. 2.2F). The PVC tube was placed vertically so that the funnel, which was placed on top of the PVC tube, was aligned horizontal. The wire couplings of the resin column and the funnel fitted tightly into this PVC tube (Fig. 2.2E). Closed field blanks were installed simultaneously with the collectors, with one field blank in the clearcut (sun-exposed) and one field blank in the control (shade). Collectors and field blanks were operational for 10 weeks. Funnel contamination (leaf litter and bird droppings) was recorded, and contaminated funnels were cleaned weekly. For the water-method, the leachate was collected every week and send to the laboratory. In the laboratory, the sample volume was recorded and sample pH was measured followed by the contents of Al, Ca, Cu, Fe, K, Mg, Mn, Na, P, S and Zn using ICP-AES (Thermo-Scientific iCAP 6500 DUO, USA) and the contents of N-NH₄, N-NO₃+N-NO₂, and inorganic carbon (IC) and total carbon (TC) using a Segmented Flow Analyzer (SFA 4000, Skalar Analytical B.V., the

Netherlands) within 24h of sampling. The volume of the leachate of the IER collectors was collected monthly, the resin columns were collected on January 14th, 2020, dried together with lab blanks to a constant weight at 28°C and subsamples were taken for 2M KCl extraction followed by N-NH₄ and N-NO₂ + N-NO₃ content analysis using a Segmented Flow Analyzer (SFA 4000, Skalar Analytical B.V., the Netherlands) and for 3.5M HCl extraction followed by Ca, Cu, Fe, Mg, Mn, Na, P, S and Zn content analysis using the ICP-AES (Thermo-Scientific iCAP 6500 DUO, USA). As a result of contaminations by bird feces, only 18 out of the 28 paired collectors were used for the comparison. Uncontaminated paired collectors were evenly distributed over the canopy openness treatments.

2.4 Calculations and statistical analysis

The concentrations of the resin columns used within the laboratory and field test were corrected for the subsampling in case of batch extraction, corrected for field- and lab blanks and corrected for sample dilution prior to chemical analysis. To correct for subsampling, the concentration of the subsample was multiplied to the concentration of the entire column based on the weights of the subsample and the entire column respectively. Concentrations of the field and lab blanks were subtracted from the concentrations of the entire column to correct for field and lab contamination. For the samplers in the forest gaps the sunlight exposed field blank was used, for the samplers underneath the forest canopy, the shadow field blank was used. Subsequently, the concentrations of the resin columns used within the field test were converted to the amounts per ha⁻¹ for the entire measurement period. Thereafter, the deposition in kg ha⁻¹ was calculated based on the funnels surface. For the water-method, the precipitation in L ha⁻¹ was calculated based on the water volume per funnel (mL). Then the measured weekly concentrations were converted to kg L⁻¹ and multiplied with the precipitation (L ha⁻¹). Finally, for both methods, the samples were checked for bird droppings based on the P content, and samples with a P influx (in kg ha⁻¹) larger than the mean plus the 2 times the standard deviation were removed.

For the laboratory test, we calculated the adsorption capacity and the recovery efficiency. The adsorption capacity (i.e., percentage of total elemental influx adsorbed by the resin) was calculated as:

$$\text{Adsorption capacity} = \left(1 - \left(\frac{A_{out}}{A_{in}} \right) \right) * 100$$

In which A_{in} is the total amount of macro and micronutrients in the solution (in µmol) applied to the resin and A_{out} is the amount in the leachate (in µmol). The recovery efficiency (i.e., percentage of total elemental flux recovered from the resin) was calculated as:

$$\text{Recovery efficiency} = \frac{A_{ex}}{A_{in}} * 100$$

In which A_{ex} is the amount of macro and micronutrients in the leachate applied extract (in μmol) which was poured over the loaded resin.

Recovery efficiencies of lab extractions differing in molarity, resin pre-treatment and extraction type and the effect of element and canopy openness on field recovery were tested using ANOVA type I error for unbalanced data following construction of a generalized least squares model. Heterogeneity between groups was overcome using the varIdent weighting from the R package “nlme”. Tukey’s post-hoc (HSD) test was performed following ANOVA using the R package “emmeans” to test for differences between groups. Goodness of fit between the original and the IER-method of the field test were tested using linear models corrected for outliers in the data. Remaining outliers were removed from the linear models when the Cook’s distance was larger than $4/n$ in which n is the number of observations.

3 Results

3.1 Adsorption capacity

The adsorption capacity of the resin (i.e., % of elemental flux bound to the resin) under controlled laboratory conditions was 100% for all nutrients, with only Na and P being slightly lower (96-97%) (Fig. 2.3). The adsorption capacity was not influenced by the flushing of the resin with demineralized water.

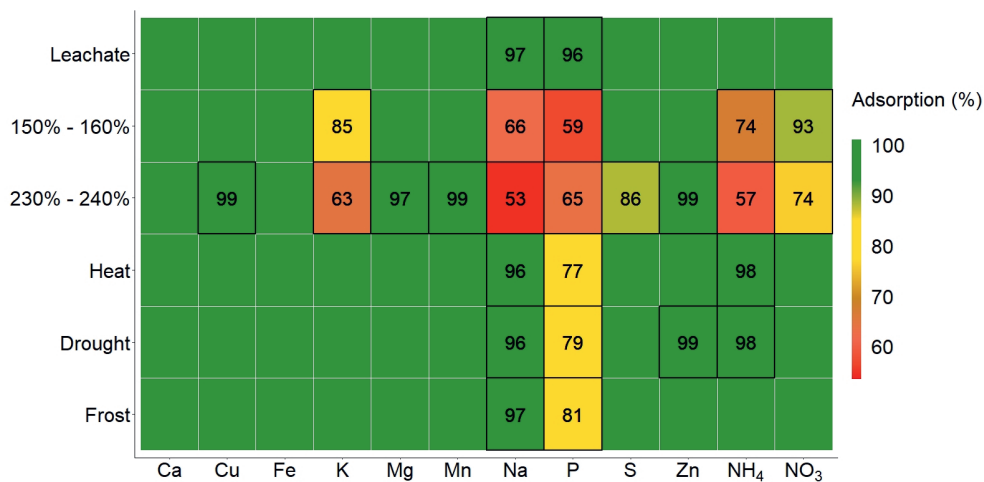


Figure 2.3 Overview of the adsorption capacity of the resin following different tests. Tests included the loading of the resin with known concentrations within the resins capacity (leachate), loading the resin beyond the resins capacity (150% - 160% of respectively cation and anion bed and 230% - 240% of respectively the cation and anion bed), and resin pre-treatments including warmth, drought, and frost. For adsorption capacities $\leq 99\%$ numbers of the adsorption capacity are given. Cells without numbers represent adsorption capacities of $>99\%$.

Overloading the resin up to 150% of the cation bed capacity resulted in decreased adsorption of $\text{Na} > \text{NH}_4 > \text{K}$ and a maximum loading of the cation bed of 115%. Overload of the anion bed up to 160% decreased the adsorption of $\text{P} > \text{NO}_3$. Increasing the elemental flux over the resin up to 230% of the cation bed capacity and 240% of the anion bed capacity resulted in lower adsorption of almost all elements except Ca and Zn (Fig. 2.3). Lab-controlled environmental conditions mimicking heat, drought and frost reduced the adsorption capacity of Na and P, and heat and drought slightly lowered the adsorption capacity of NH_4 . Elemental adsorption within the resins exchange capacity was thus close to 100% for all elements except P which was underestimated under extreme conditions.

3.2 Recovery efficiency

The recovery efficiency of NH_4 and NO_3 under laboratory conditions (i.e., % of the elements that can be extracted from the resin) was generally high (mostly 90-100%) with recovery depending on the molarity of the extraction (Table 2.3). The recovery efficiency of NH_4 and NO_3 was weakly but significantly higher with 1M KCl as an extractant compared to 2M KCl (Anova, F-value: 4.4, P-value: 0.048, Df: 22). We did not find differences between fresh or dry resin, or between drip or shake-drip treatments using KCl extractions.

Recovery efficiency of Ca, Cu, Fe, K, Mg, Mn, Na, P, S and Zn following HCl extraction was high (>90%) for Ca, K, Na and Mn, slightly lower (>80%) for Mg, S, Cu and Fe, relatively low for P (40-91%) and very low (6-25%) for Zn (Table 2.3). Because extraction of Zn was unreliable, this element is not further included in average recovery numbers. The average recovery efficiency was highest (90-100%) following either 2M HCl extraction or 4-2-1M HCl extraction. Recovery efficiency was significantly higher following an extraction on dried resin (avg. recovery 88%) compared to fresh resin (avg. recovery 80%), and recovery efficiency was slightly higher following a shake-drip treatment (avg. recovery 87%) compared to drip only treatment (avg. recovery 84%) (Table S2.3). We found an interaction effect between elements and pre-treatment, elements and molarity and elements and extraction type, indicating that different elements responded differently to the different treatments. Overall, highest average recoveries using HCl were found for the 2M dry weight shake and drip treatment, resulting in an average recovery of 100% whereas lowest average recoveries (72%) were found for the 1M fresh weight drip treatment (Table 2.3).

Table 2.3 Recovery efficiency of Ca, Cu, Fe, K, Mg, Mn, Na, P, S and Zn following HCl extractions and of NH₄ and NO₃ following KCl extractions. Recovery efficiencies are expressed as a % of the total elemental content poured over the resin. The efficiencies of the recovery were tested using extractions with different molarities, based on fresh (FW) or dried (DW) resin and based on drip or shake-drip treatments. The number of samples (n) for each extraction combination, the average recovery per extraction combination (Avg) and the average recovery for each element (Avg element) is given. Recovery percentages per element close to 100 are indicated in bold. Differences in recovery efficiencies between elements is indicated with small capital letters based on the average element recovery, test statistics are given in Table S2.3.

Mol	Resin	Method	n	Ca	Cu	Fe	K	Mg	Mn	Na	P	S	Zn	Avg*	NH ₄	NO ₃
1	FW	Drip	6	30	80	56	88	59	91	97	59	92	17	72	100	98
2	DW	Drip	2	130	94	99	100	98	100	93	40	91	21	94	97	94
2	DW	Shake-Drip	2	130	100	110	110	100	110	100	65	100	7.6	100	94	89
2	FW	Drip	2	85	81	88	86	84	91	82	43	96	25	82	80	95
2	FW	Shake-Drip	2	82	80	87	87	81	88	84	69	93	5.4	83		
1-2	DW	Drip	1	110	87	85	90	86	90	91	57	88	21	87		
1-2	FW	Drip	1	82	77	77	80	77	81	80	60	93	42	79		
2.5	DW	Shake-Drip	2	86	78	72	100	85	81	100	91	78	6.1	86		
3	DW	Shake-Drip	2	82	71	67	98	77	77	99	84	70	6.1	81		
3.5	DW	Shake-Drip	2	99	89	80	88	93	96	84	83	83	11	88		
4	DW	Shake-Drip	1	86	66	69	92	80	80	93	76	73	8.1	79		
4-2-1	DW	Drip	2	100	92	99	93	88	100	89	49	96	31	90		
4-2-1	FW	Drip	2	86	85	90	100	80	92	82	49	97	44	85		
Avg element				91 ^d	83 ^{bc}	83 ^c	93 ^{ab}	84 ^c	91 ^b	90 ^a	63 ^e	88 ^b	19 ^f			

* Average without Zn as this element was unreliable using our extraction method.

3.3 Performance under field conditions

There was a positive significant linear relationship between the deposition estimates of the water-method and the IER-method for all elements except for Ca, Zn and Fe (Table 2.5, Fig. 2.4). Absence of a relation for Ca, Zn and Fe was not related to the correction for contamination in blanks and for the lab-recovery (Table 2.5).

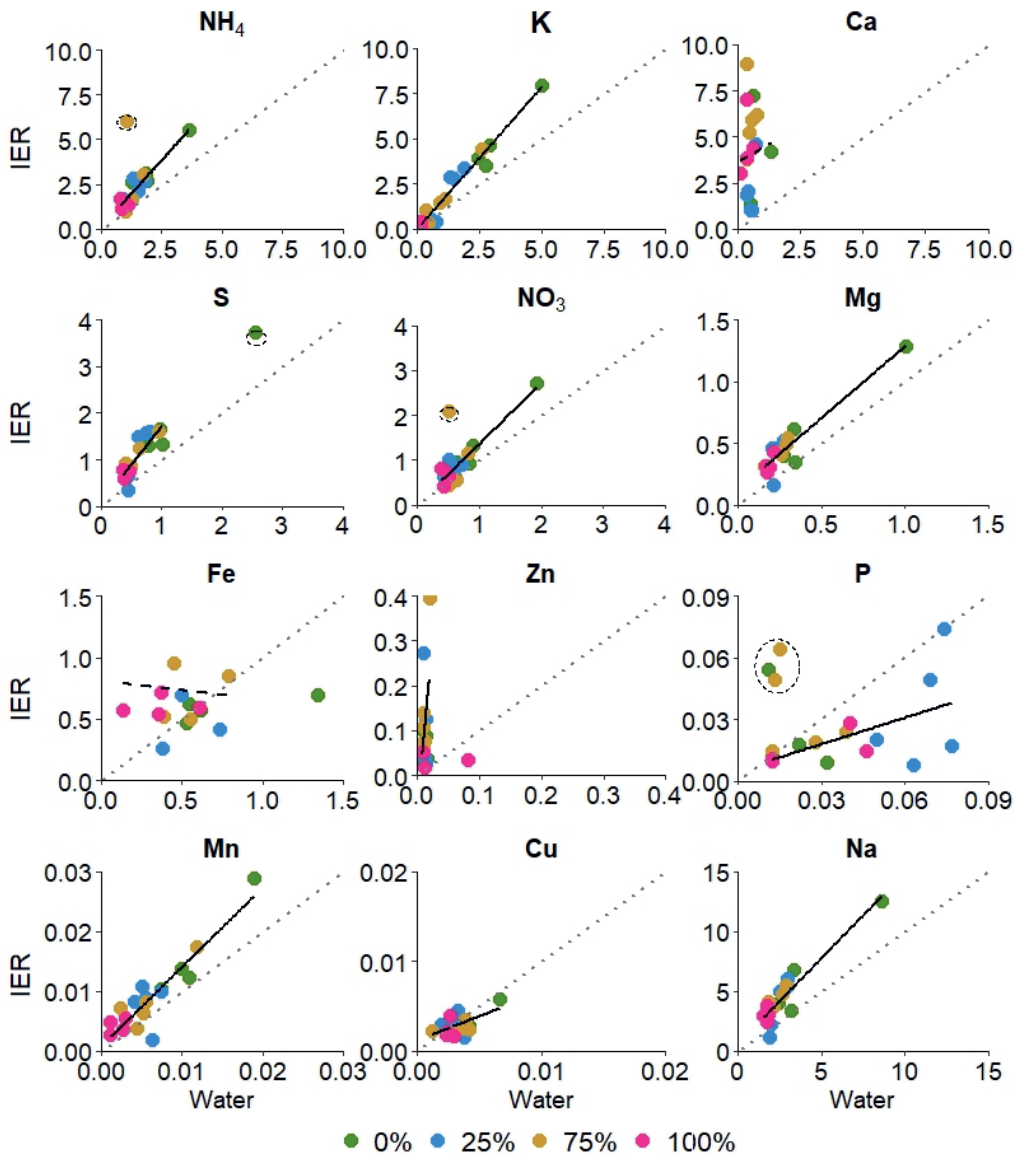


Figure 2.4 Relationship between the deposition estimates of the IER-method (kg ha⁻¹) and of the water-method (kg ha⁻¹) for the 10-week measurement period. Significant relationships are depicted with a solid black line, non-significant relationships with the dashed black line. The 1:1 line is shown as the dotted grey line.

Table 2.5 Regression coefficients and R² of models for the relation between IER columns and ORG columns including correction for blank and lab recovery (n = 18). Differences between corrected, blank corrected and recovery corrected is related to the corrections for contaminations (blank corrected) and for the recovery of the elements (recovery corrected). The corrected shows the data which is corrected for both the contamination and the recovery. The R²-adj. of the model with the best fit is highlighted in bold.

	NH ₄	NO ₃	S	P	K	Ca	Mg	Mn	Cu	Fe	Zn	Na
Corrected	Intercept	0.062 ±	-0.051 ±	0.0053 ±	0.051 ±	1.1 ±	-0.11 ±	-0.0031 ±	0.0012 ±	0.0020 ±	-0.13 ±	-1.5 ±
		0.21	0.098	0.20***	0.0078	0.15	0.042*	0.0012*	0.00066	0.0061	0.086	0.63*
	Slope	1.5 ±	1.4 ±	2.5 ±	0.43 ±	1.6 ±	1.3 ±	1.6 ±	0.54 ±	0.015 ±	16 ±	1.7 ±
Blank cor.		0.14***	0.13***	0.31***	0.17*	0.077***	0.13***	0.15***	0.19*	0.010	6.9*	0.20***
	R ² -adj	0.88	0.80	0.27	0.96	-0.036	0.86	0.87	0.29	0.069	0.21	0.82
	Intercept	0.11 ±	-0.02 ±	-1.39 ±	-2.6 ±	0.38 ±	-0.26 ±	-0.0049 ±	0.0040 ±	0.0067 ±	0.030 ±	-8.0 ±
Recov. Cor.		0.18	0.077	0.50*	0.32** ¹	0.36	0.12*	0.0031	0.0015*	0.013	0.025	2.6**
	Slope	1.1 ±	1.1 ±	5.2 ±	-3.8 ±	4.0 ±	3.6 ±	4.3 ±	1.2 ±	0.036 ±	4.4 ±	6.6 ±
		0.12***	0.11***	0.8***	8.0 ¹	0.20***	0.36***	0.43***	0.46*	0.022	2.1*	1.2***
Recov. Cor.	R ² -adj	0.82	0.84	0.70	-0.043 ¹	0.96	0.0028	0.84	0.84	0.081	0.16	0.64
	Intercept	0.15 ±	8.9 10 ⁻⁵ ±	0.070 ±	0.0053 ±	0.051 ±	0.13 ±	0.00087 ±	0.0012 ±	0.020 ±	-0.085 ±	-0.43 ±
		0.22	0.095	0.17	0.0078	0.15	0.039**	0.0011	0.00066	0.0061**	0.094	1.0
Recov. Cor.	Slope	1.5 ±	1.4 ±	1.6 ±	0.43 ±	1.6 ±	1.2 ±	1.3 ±	0.54 ±	-0.0025 ±	15 ±	1.9 ±
		0.14***	0.13***	0.26***	0.17*	0.077***	0.12***	0.15***	0.19*	0.010	7.6	0.43***
	R ² -adj	0.88	0.88	0.71	0.27	0.96	0.85	0.83	0.29	-0.063	0.16	0.54

*** < 0.001; ** 0.01 > < 0.001; * 0.05 < > 0.01; no star is not significant; ¹ log transformed values.

The IER data corrected for contamination of blanks and the lab-recovery overall resulted in the highest R^2 -adjusted, resulting in a corrected goodness of fit up to 0.96 (K) and between 0.8 to 0.9 for NH_4 , NO_3 , S, Mg, and Mn (Table 2.5). Canopy openness treatment significantly influenced the deposition estimates of the IER-method compared to the water-method (ANOVA, F-value: 6.9, P-value < 0.001, Df: 168) although between treatment differences were not significant following Tukey's post-hoc tests (Fig. S2.1). The IER-method tended to have lower deposition estimates in the 100% canopy openness treatment for Mg, Mn, Na and S, but overall, the IER-method resulted in higher deposition estimates for NH_4 , K, S, NO_3 , Mg, Mn, and Na compared to the water-method (Fig. 2.4). For Fe, P, and Cu, for which the water-method yielded higher deposition estimates, all values of the water-method were below detection limit (Table S2.4).

4 Discussion

4.1 Adsorption capacity

We aimed to test the capacity of IER as a method to quantify atmospheric deposition for a broad range of macro- and micro-elements, comparing results under laboratory and field conditions and in the latter case comparing bulk deposition and throughfall. First, the adsorption capacity of the IER when loaded within its capacity was generally high. High adsorption confirms earlier studies who found no elemental loss of NO_3 , NH_4 and SO_4 (Simkin et al., 2004; Sheibley et al., 2012; Sheng et al., 2013) or only slight losses of NH_4 and NO_3 (Fang et al., 2011) and contradicts findings of low resin adsorption (Langlois et al., 2003). We show that IER is also able to adsorb above 99% for a range of other elements including the base cations and some micronutrients. Therefore, IER can be loaded within the 70% of its exchange capacity without risking lower elemental adsorption. However, slightly lower adsorption capacities were found for Na and P. These lower adsorption capacities are caused by the lower cation-exchanger affinity for Na^+ and lower anion-exchanger affinity for HPO_4^{2-} (Skogley and Dobermann, 1996; Park et al., 2014). The lower adsorption capacities when using the resin within its capacity can lead to an underestimation of the total deposition of P by 4%, although other studies report no lower adsorption capacities for P (Tahovská et al., 2016). Despite the possible underestimation of the total deposition, studies using IER report P deposition values within the natural ranges (Decina et al., 2018; Hoffman et al., 2019), indicating that the method usually also works well for P. The lower adsorption capacity of Na, however, can result in lower estimates of the deposition for multiple elements when Na is used as an tracer for canopy exchange processes (Staelens et al., 2008b), and the use of Na as a tracer in IER-deposition studies is thus questionable.

To further test the affinity of the resin for the studied elements, the resin was loaded to approximately 160% and 240% of its capacity. Based on the adsorption capacity beyond the resins capacity, we found that the cation bed has an affinity of $\text{Ca} = \text{Fe} > \text{Cu} = \text{Mn} = \text{Zn} > \text{Mg} > \text{K} > \text{NH}_4 > \text{Na}$

which is in line with the previous reported resin affinity (Skogley and Dobermann, 1996). The anion bed has an affinity of $S > NO_3 > P$ which agrees with earlier studies (Skogley and Dobermann, 1996; Park et al., 2014). The resins affinity and the adsorption capacity for different levels of loading beyond the resins capacity is of importance for resin columns under suspicion of overloading. We did not find lower adsorption of Ca and Fe and only slightly lower adsorption of Cu, Mg, Mn and Zn, indicating that, when columns are slightly overloaded, these estimates are still reliable. When columns are loaded $> 100\%$ of the capacity, the estimates for K, Na, P, S, NH_4 and NO_3 are not reliable. Therefore, in case of suspicion of ion exchange overload, tests are recommended to check if stoichiometry between any element of Ca, Cu, Mg, Mn and Zn with K, Na, P, S, NH_4 and NO_3 falls within the stoichiometric range of natural deposition estimates. We strongly recommend collecting the resin columns prior to resin saturation as adsorption of Na and P can further decrease when saturating the resin up to 90 or 100%. The time period that the resin can stay in the field depends on the total atmospheric deposition and the volume of resin used. For remote areas with low deposition levels and low risk of sample contamination (e.g., by bird feces) the resin can stay for multiple months up to a year in the field as long as adequate resin volumes are used.

Heat, drought and frost treatments hardly influenced the absorption capacity of most elements, but decreased the P adsorption capacity and, in case of heat and drought, NH_4 and Zn (the latter only for drought) adsorption. These findings are in line with the adsorption behavior of some other IER types, where drying significantly reduced NH_4 adsorption while frost-thaw cycles did not (Hart and Binkley, 1984; Kjonaas, 1999). However, in other work extensive dry-wet cycles did not affect the adsorption of PO_4 , NO_3 and NH_4 (Mamo et al., 2004) indicating that the effect of environmental conditions differs per resin type. Application of the IER-method without an adsorption pre-test of the resin can therefore potentially underestimate NH_4 and P deposition when used in areas with temperatures above $40^\circ C$ and can potentially underestimate NH_4 , P and Zn deposition in areas with longer drought periods. Despite the effect on some elements, weather circumstances generally seem to have little effect indicating that the method is suitable under different climatic circumstances, like the boreal zone (Fenn et al., 2015), temperate zone (Fenn and Poth, 2004; Hoffman et al., 2019) and the tropics (Kohler et al., 2012; Ibrahim et al., 2022). The robustness of the method under different climatic circumstances implies that it can be used to compare deposition over large environmental gradients, which is essential to understand regional and global deposition patterns.

4.2 *Recovery efficiency*

The recovery efficiency was tested based on differences in molarity, resin pre-treatment and extraction type. Recovery of NH_4 and NO_3 was highest following a 1M KCl extraction based on controlled percolating of the extraction fluid through the resin. Although highest recovery following a 1M KCl extraction was

reported before (Hart and Binkley, 1984), most studies indicate that 2M KCl extractions will lead to higher recovery of both NO_3 and NH_4 (Kjonaas, 1999). However, the 2M KCl recovery efficiencies of this study were comparable to other studies using 2M KCl as an extractant (Fenn et al., 2002; Sheng et al., 2013; Tulloss and Cadenasso, 2015). The highest recoveries were obtained by using dried resin and the combined shake-drip methods.

Recovery efficiency following HCl extraction differed between elements and depended on the extraction itself. We choose HCl as an extractant as this extraction fluid allows measurements of a broad range of elements on the ICP-AES and this method was rarely tested. A limited number of studies used HCl as an IER extractant (Van Dam et al., 1987; Dobermann et al., 1997; Szillery et al., 2006; Yamashita et al., 2014) but only one study, testing only two elements, reported (high) elemental recoveries (Van Dam et al., 1987). Although H^+ has a relatively low affinity for the cation bed (Skogley and Dobermann, 1996), we expected that increasing molarities would increase recovery efficiency of both the cation and the anion bed. Surprisingly, recovery efficiency was highest using 2M HCl and 4-2-1M HCl, although highest recovery differed between elements (Table 2.3). Overall, we did find much higher recovery efficiencies for Ca and Mg using HCl extractions compared to KI and H_2SO_4 extractions (Kohler et al., 2012; Wieder et al., 2016), which can be related to a better extraction efficiency of HCl. Absence of higher recoveries using > 3M HCl can be caused by differences in extraction time between treatments (Zarrabi et al., 2014) although the overall differences in recovery efficiencies between extractants were rather small.

Recovery efficiency was higher when resin was dried prior to HCl extraction and when using the shake-drip extraction (Table S2.3). The mechanism behind higher recovery efficiency following pre-extraction drying remains speculative but might be related to a better accessibility of the extract to reach micropores when the resin was dried. Previously, it was argued that pre-loading drying resulted in lower recovery efficiencies because of unavailable micropores due to swelling of the resin after rewetting (Kjonaas, 1999) but this unavailability of micropores was contradicted by Mamo et al. (2004) who found that dry-wet cycles significantly increased the desorption of elements from the resin. Occurrence of dry-wet cycles under field conditions can therefore interfere with the recovery efficiency of elements from the resin which could possibly bias deposition estimates. This effect is, however, likely small as full drying resulted in only 8% more efficient recoveries. The higher recovery following shake-drip treatment can result from longer contact time with the extractant (Zarrabi et al., 2014) while still avoiding the equilibrium reaction which occurs when using the shake treatment only. However, the present paper was not designed to test the effect of extraction time on the recovery efficiency, a complete test of this hypothesis will have to await future experimentation.

Finally, the best extraction to use depends on the elements of interest. When studied elements are limited to the base cations, the 2M HCl extraction provides good recovery efficiencies. However, studies

including P and Zn should rather choose for a HCl extract with a higher molarity or choose another extractant. Overall, recovery efficiencies of P and Zn were rather low, which may result from the low initial concentrations (Zarrabi et al., 2014). We did not test for different extraction solutions as there are only limited options for extracting a broad range of macro- and micro-solutions. However, for P and Zn different extraction solutions should be tested to increase the recovery efficiency. Furthermore, using the recovery efficiencies, we found only limited evidence of a release of background levels of elements from the resin. Indications of the release of background levels were present for Ca (up to 130% recovery) and Na (up to 110% recovery). These indications were mainly present in the 2M HCl dry weight shake-drip extraction and could possibly be caused by lab contaminations. We did not find evidence for high background levels of NO_3 and NH_4 , contrary to Langlois et al. (2003) who argued that the IER-method was not suited for monitoring subtle patterns of NO_3 and NH_4 deposition. Together, our findings indicate that both KCl and HCl perform well as an extractant except for P and Zn for which new extraction methods should be tested.

4.3 Performance under field conditions

In general, deposition estimates based on the IER-method were positively related to the deposition estimates of the water-method, however, the IER-method often resulted in higher deposition estimates. Exceptions were Fe and Ca, for which we did not find a relation between the deposition estimates of the IER-method and the water-method. This could indicate pollution related to elevated Ca and Fe leaching from the sample materials. For example, in the sun-exposed field blank we found high Fe pollution causing the Fe deposition levels of all exposed collectors to be 0 (Fig. 2.4). For the collectors corrected for the shade-exposed field blank, we found good agreement between the deposition estimate of the IER-method ($0.68 \text{ kg ha}^{-1} \pm 0.12 \text{ s.e.}$) and the water-method ($0.66 \text{ kg ha}^{-1} \pm 0.09 \text{ s.e.}$) with the deposition estimates of both methods within the normal range of throughfall Fe deposition of the winter period (RIVM, 2015). For Zn we found much higher deposition values using the IER-method compared to the water-method in contrast to throughfall which was much higher than bulk deposition estimates multiplied by the throughfall correction factor (Table S2.1). It could be that the presence of organic particles interfered with the recovery efficiency of Zn, possibly leading to an overestimation of the Zn throughfall.

The higher deposition estimates of the IER-method compared to the water-method for NH_4 and NO_3 can be caused by absence of biochemical reactions which causes losses of these elements in the original samplers (Fenn and Poth, 2004; Kohler et al., 2012). Higher deposition estimates using the IER-method can also be related to the low concentration of elements in the water-method, which were often below detection limit (Table S2.4). Overall, slightly higher deposition values using IER columns were reported before (Fenn and Poth, 2004; Simkin et al., 2004; Kohler et al., 2012). Because of absence of biochemical reactions and higher reliability of the lab measurements for IER-samples, the IER-method is

likely more reliable to quantify both bulk deposition and throughfall compared to the water-method and the generally higher deposition estimates are likely a better representation of the actual atmospheric deposition.

The lower deposition estimates of P can be caused by a better adsorption of inorganic P compared to organic P to the resin (Zarrabi et al., 2014) which potentially reduces the recovery efficiency of P under field-conditions compared to lab-conditions. However, additional field tests are necessary for P to compare the difference between field and laboratory adsorption and recovery efficiencies. For other elements, the comparison of the IER-method and the water-method did not give evidence of lower adsorption or recovery efficiencies under field conditions. Absence of this effect might, however, be related to the winter-period in which the field measurements took place as, for example, pollen were hypothesized to reduce recovery of NH_4 , NO_3 and SO_4 from the IER (Brumbaugh et al., 2016). Lower field recovery might therefore, beside the resin type and the extraction method, be related to the amount of organic particles like pollen which was not included in this study.

5 Conclusions

We tested the suitability of the IER-method for quantifying bulk deposition and throughfall of macro- and micronutrients by assessing adsorption capacities and recovery efficiencies under controlled laboratory conditions, followed by an evaluation of the performance of the method under field conditions.

Results showed that (1) the adsorption capacity of the resin under controlled laboratory conditions was close to 100% for all nutrients; (2) Extraction using KCl (1 or 2 M) is effective for nitrogen (NH_4 and NO_3) with general high recoveries (mostly 90-100%) depending on the molarity of the extraction, while extraction using HCl is effective for Ca, K, Na, Mn, Mg, S, Cu and Fe but not for P and Zn for which testing other extraction methods or extraction fluids is recommended; (3) drying the resin prior to extraction and using a shake-drip extraction method increased the recovery efficiencies; (4) the IER-method is useful under a broad range of environmental conditions, since heat (40°C), drought and frost (-15°C) hardly affected the adsorption of nutrients except for P which was reduced up to 25%; and (5) the IER-method performed well under field conditions, resulting in similar but consistent higher deposition estimates compared to the water method.

Our results even imply a higher reliability of the IER-method than the water method since uncertainties related to biological reactions and the detection limit for lab measurements could be removed. We conclude that IER is a powerful tool for the monitoring the element input by bulk deposition and throughfall for a broad range of elements, across a broad range of environmental conditions.

Acknowledgements

This research is part of the Nutrient Balance project and was funded by the Dutch Research Council (NWO, No. ALWGS.2017.004). We acknowledge Henk van Roekel for his assistance with the construction of the deposition samplers, the field sampling and sample processing. We thank the CBLB laboratory for their cooperation in developing this method and we thank our partner, the National Forest Service for permission to work in their forest.

Supplementary information

Table S2.1 Conversion factor to calculate total deposition within a forest stand based on bulk precipitation data. Original data underneath the multiplication factors were corrected for canopy uptake of NH_4 and NO_3 and canopy leaching of Mg, Ca, K and Mn except for the inert ions Na, SO_4 , Zn and Cu. The ion Na is in all cases used to calculate canopy leaching of Ca, Mg and K. Stem flow was only included for beech forest and one Douglas fir stand.

Species	Na	NH_4	NO_3	K	Mg	SO_4	PO_4	Ca	Mn	Zn	Cu	Country	Stemflow	Source
Beech	1.2	1.2	1.2	1.2	1.2	1.2	1.3	1.2	1.2			Germany	Yes	(Talkner et al., 2010)
Beech						1.6								(Augusto et al., 2002)
Beech	2.4	3.6	2.0	3.1	2.4	4.8		2.4				Belgium	Yes	(Adriaenssens et al., 2012a)
Beech	1.2					1.6						Czech Republic	Yes	(Růžek et al., 2019)
Beech	1.2					1.2				1.1	2.1	Poland	Yes	(Kowalska et al., 2016)
Douglas fir	2.3	2.3	2.3	2.2	2.2	2.3	2.1	2.4				France	Yes	(Marques et al., 1997)
Douglas fir	2.3			1.4	1.8	3.3		1.5				Netherlands		(Van Ek and Draaijers, 1994)
Douglas fir	2.3	3.7	2.1	2.3	2.3	3.6	2.5	2.3				Netherlands	Yes	(Draaijers et al., 1997a)
Coniferous forest										2.4	2.6			(De Vries and Bakker, 1996)
Scots pine	1.1				1.2	1.2	1.6	1.1				Poland	No	(Kozłowski et al., 2020)
Scots pine	2.3			1.5	1.9			1.6				Netherlands	No	(Van Ek and Draaijers, 1994)
Scots pine	2.0					1.5				1.1	1.3	Poland	No	(Kowalska et al., 2016)
Corsican pine	1.5	6.5	3.5	3.4	1.9			1.4				Belgium	No	(De Schrijver et al., 2004)
Average														
Beech	1.5	2.4	1.6	2.2	1.8	2.3	1.3	1.8	1.2					
Douglas fir	2.3	3.0	2.2	2.0	2.1	3.1	2.3	2.1		2.4	2.6			
Scots pine	1.7	6.5	3.5	2.0	1.7	1.6		1.4						

N.B. The bulk throughfall deposition of SO_4^{2-} was assumed to be not influenced by canopy exchange as the stomatal uptake of SO_2 is balanced by foliar leaching of SO_4^{2-} (Draaijers and Erisman, 1995). However, Staelens et al. (2007) estimated that canopy leaching contributed 7% to the combined bulk throughfall and stemflow of SO_4^{2-} which was in line with the findings of Potter et al. (1991). Canopy exchange of Al and Cu are neglectable as both elements in deposition is found in a colloidal fraction and almost entirely complexed by DOC (Gandois et al., 2010b). The free metal ion forms of Zn (on average 30%) do interact with the canopy however concentration is only slightly increased or decreased (Gandois et al., 2010b).

¹ measured under a single tree, therefore not representative for a forest stand.

Table S2.2 Overview of the columns (n = 45) prepared for the different laboratory tests. The columns adsorption and extraction indicate if the columns were used for either the adsorption or the extraction test.

Pre-treatment	Loading	n	Adsorption	Extraction
Heat	1 * macro- and microfluid	3	Yes	Yes
Drought	1 * macro- and microfluid	3	Yes	Yes
Frost	1 * macro- and microfluid	3	Yes	Yes
None	1 * macro- and microfluid	30	Yes, 3 columns	Yes
None	2 * macro- and microfluid	3	Yes	No
None	3 * macro- and microfluid	3	Yes	No

Table S2.3 ANOVA F and P values for HCl extraction of different molarities, pre-treatments (DW or FW) and different extraction types (drip or shake-drip).

	DF	F-value	P-value
Element	9	250	< 0.0001
Pre-treatment	1	68	< 0.0001
Molarity	6	16	< 0.0001
Extraction type	1	4.2	0.043
Element * Pre-treatment	9	19	< 0.0001
Element * Molarity	54	7.8	< 0.0001
Element * Extraction type	9	2.2	0.025

Table S2.4 Elemental concentrations under detection limit (%) after 10-week long field sampling of the atmospheric bulk (throughfall) deposition in the Netherlands. Elemental concentrations were often under detection limit for the original method (Org), especially for the treatments (TM) shelterwood (SW) and clearcut (CC) and less often for the treatments control (CO) and high thinning (HT). For the Ion Exchange resin method (IER) values were less often under detection limit.

TM	Method	Ca	Cu	Fe	K	Mg	Mn	Na	P	S	Zn	NH ₄	NO ₃
CO	Org	83	100	100	0	0	50	0	100	0	83	0	0
HT	Org	100	100	100	0	0	100	0	100	0	100	0	0
SW	Org	100	100	100	50	17	100	0	100	0	100	0	0
CC	Org	100	100	100	100	33	100	0	100	0	83	0	0
CO	IER	0	0	20	0	0	0	0	40	0	0	0	0
HT	IER	0	0	40	0	0	0	0	20	0	0	0	0
SW	IER	0	0	17	0	0	0	0	0	0	17	0	0
CC	IER	0	0	0	0	0	0	0	60	0	20	0	0

Supplementary figures

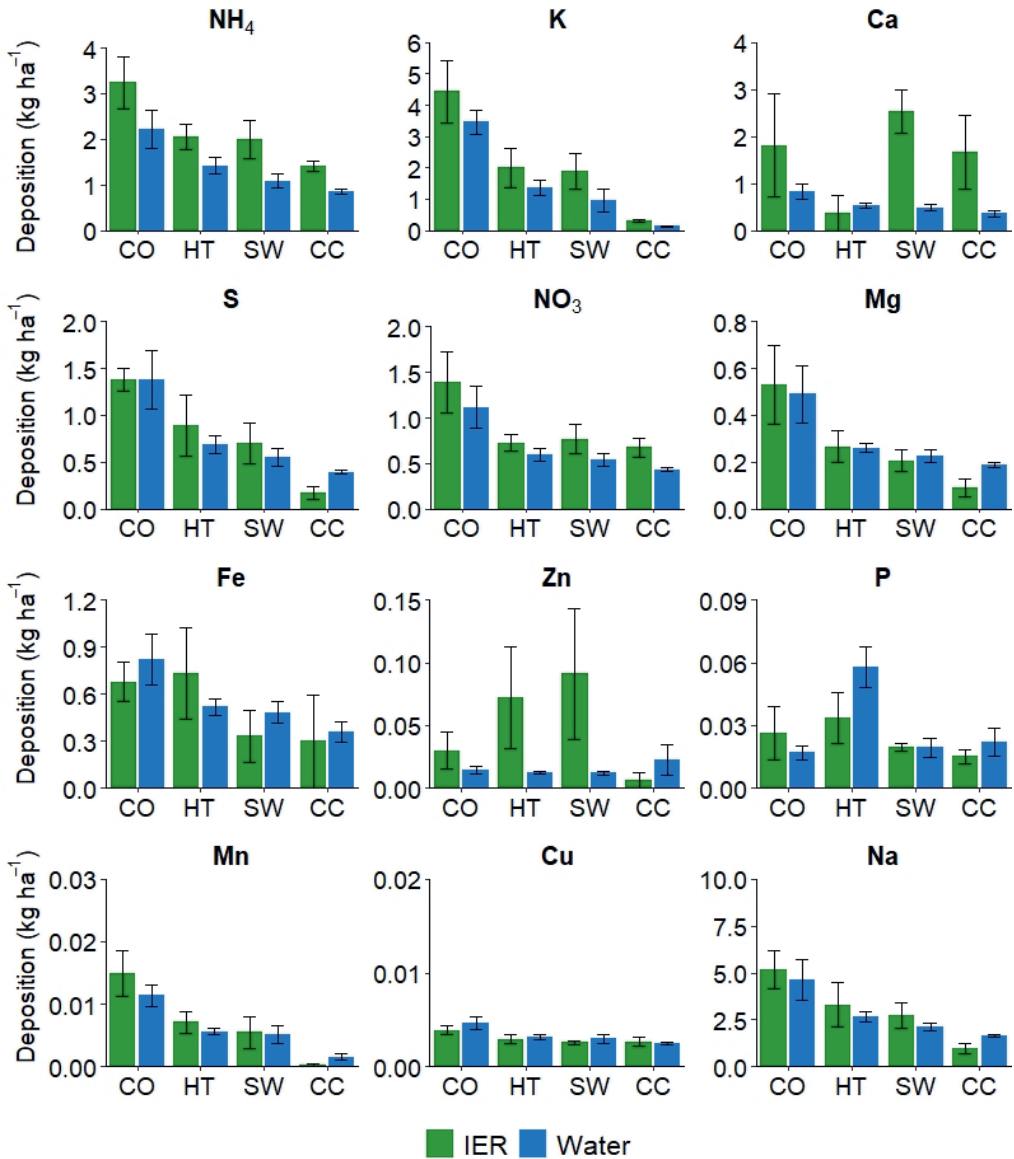


Figure S2.1 Deposition estimates of the IER-method (kg ha⁻¹) and the water-method (kg ha⁻¹) in the treatments control (CO), high-thinning (HT), shelterwood (SW) and clearcut (CC) for a 10-week measurement period.



Chapter 3

Canopy openness rather than tree species determine seasonal and annual atmospheric deposition into forests

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Abstract

Atmospheric nutrient deposition plays a crucial role in supplying nutrients to forest ecosystems and is therefore a key factor in maintaining the nutrient balance in forests. Deposition in forests is, besides the distance to emission sources, strongly influenced by tree species and stand properties, like tree height, leaf area index and canopy openness, which affect dry deposition. Consequently, tree harvesting can significantly influence atmospheric deposition by altering the forest structure, and these effects may vary among species. We compared seasonal and annual total atmospheric deposition in mature stands of European beech (*Fagus sylvatica*), Douglas fir (*Pseudotsuga menziesii*), and Scots pine (*Pinus sylvestris*). We assessed changes in total deposition resulting from the effects of high-thinning, shelterwood-cut and clearcut on forest structure in areas exposed to high nitrogen (N) deposition. Our study is one of the few that shows such trends not only for macronutrients but also for micronutrients, which have hardly been quantified before in deposition studies in forests.

We demonstrate that total deposition is generally highest in Douglas fir stands and lowest in Scots pine stands, with the most significant differences observed in nutrients primarily deposited as dry deposition (e.g., S, K). The total deposition presented in this study exceeds national estimates up to a factor 1.5 for N and up to 7.5 for the base cations (K, Ca, and Mg), indicating that deposition levels in forests are significantly higher than on low vegetation. Harvest intensity strongly influences the total deposition of all nutrients, except for P. Nutrient inputs through atmospheric deposition, on average, decrease 2.2 times when moving from mature stands to a clearcut. This decrease differs among tree species and between nutrients. While the relative decrease in base cations was higher than for N or S, the absolute base cation decrease was lower causing a reduction of the net acid input following thinning, especially for Douglas fir stands. Finally, total deposition fluctuates strongly between seasons: P deposition is higher during the growing season and S and Na deposition during the dormant season. Seasonal patterns were strongly present in both the throughfall and the canopy exchange. Due to pronounced seasonal patterns, long-term deposition data are essential for accurate nutrient budget estimations.

Our results highlight the importance of considering the harvest intensity effects on forest structure and, to a lesser extent, tree species when calculating nutrient inputs via atmospheric deposition. Furthermore, regular thinnings, especially in beech and Douglas fir, could provide a management tool to slow down soil acidification.

1 Introduction

Apart from water availability, the productivity of forests depends primarily on nutrient availability which in turn is mainly determined by nutrient inputs by deposition and 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) of which the quantity depends not only on the geographic region and tree species, but also on 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., 2007a; Aherne et al., 2012; Phillips and Watmough, 2012; Iwald et al., 2013), possibly over- or underestimating the nutrient input via atmospheric deposition up to 50% (Draaijers et al., 1997b). To reduce uncertainties in forest nutrient balance estimates for specific forest sites, a better understanding is required on the possible influence of tree species and stand properties on the nutrient input via atmospheric deposition.

The input of nutrients via atmospheric deposition consists of nutrients in precipitation (wet deposition) and gases and airborne particles deposited on rough surfaces like tree canopies (dry deposition) (Lovett and Reiners, 1986; Balestrini et al., 2007). In forests, atmospheric deposition is often measured as throughfall deposition, which consists of wet deposition that passes through the canopy (Draaijers et al., 1996a; Thimonier, 1998). Within the canopy, the nutrient concentration in rainfall is altered due to nutrient uptake by or leaching from the canopy (i.e. canopy exchange) and by wash-off of the airborne particles and gasses that were first deposited on tree crowns (i.e. dry deposition) (Verry and Timmons, 1977; Lovett and Lindberg, 1984; Lovett and Reiners, 1986; Lovett and Lindberg, 1992; Staelens et al., 2008b; Adriaenssens et al., 2012a). Furthermore, part of the intercepted precipitation reaches the forest floor via stemflow which highly differs between tree species (Silva and Rodríguez, 2001; Su et al., 2019; Houcai et al., 2021). The total deposition reaching the forest floor is the throughfall deposition and stemflow corrected for canopy exchange, but in open areas such as large clearcuts it only consists of bulk or wet deposition since possible effects of the canopy are excluded.

Throughfall deposition is influenced by tree species and stand properties, such as tree height and canopy openness, because of differences in canopy exchange and interception of dry deposition (André et al., 2008; Adriaenssens et al., 2011; Zhang et al., 2022). Differences in the throughfall deposition flux between species have been reported frequently although species differences are strongly related to the geographical area making it challenging to extrapolate findings across larger geographical regions (Van Ek and Draaijers, 1994; Adriaenssens et al., 2011; Shen et al., 2013; Zhang et al., 2022). The geographical area determines the dry deposition load as the dry deposition flux is related to the distance to the sea, especially for potassium (K), calcium (Ca) and magnesium (Mg) (Draaijers et al., 1997b; Balestrini et al., 2007) and to anthropogenic pollution sources, especially for ammonium (NH₄), nitrate (NO₃) and sulphate

(SO₄) (Nordén, 1991; Draaijers et al., 1997b). Furthermore, there is ambiguity on the extent of stand properties driving species differences. Generally, the dry deposition, and therefore the throughfall deposition, is influenced by the canopy structure and roughness like tree height, canopy architecture, canopy openness, and is thus related 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). Canopy exchange is often argued to be related to the canopy cover and the nutrient content of the foliage, which differs between tree species (Herrmann et al., 2006; André et al., 2008; Talkner et al., 2010), but canopy exchange can also differ within species growing on different soil types (Nordén, 1991). Because of the differences in deposition fluxes between species, and the interactions between throughfall and specific stand properties, including canopy structure and tree height, there is a clear need for using both regional and site information for accurately estimating nutrient inputs to forests.

Tree harvest modifies forest structure and airflow and, in turn, the interception of nutrient by forests. For example, harvesting 15% of the stem volume resulted in a 20% reduction of the throughfall deposition in Norway spruce stands in southern Germany (Göttlein et al., 2023), while harvesting 40% and 100% of the volume in Norway spruce stands in southern Germany resulted in a decrease of 45% to 60% of the deposition, respectively (Bäumler and Zech, 1997; Göttlein et al., 2023). As throughfall is related to basal area, canopy roughness and, in some cases, the leaf area index (Aboal et al., 2000), it is expected that different harvest intensities influence the throughfall deposition differently for species as basal area reductions and the related canopy openness are, at least partially, species specific stand properties. Therefore, to accurately estimate the nutrient inputs in forests, the effect of harvest on throughfall deposition should be quantified on a species level. However, this is rarely done.

The nutrient inputs by 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 deposition is strongly reduced in the winter compared to the growing season, in contrast to evergreen (coniferous) species where absolute dry deposition can increase during the winter (André et al., 2008; Adriaenssens et al., 2012b). It is not clear yet to which extent the effects of seasonality on the deposition flux and the total annual deposition flux are influenced by canopy openness. Increasing the canopy openness by different harvest intensities can change seasonal fluctuation of the total deposition and therefore modify species differences but such possible trends are poorly studied.

To fill these knowledge gaps, we quantified the effects of harvest intensity related changes in forest canopy openness on seasonal and annual atmospheric deposition of nutrients within forests and show how these effects are modified by tree species. We focused on stands of three major tree species in

the Netherlands, one deciduous species (European beech, *Fagus sylvatica*) and two evergreen conifers (Douglas fir, *Pseudotsuga menziesii* and Scots pine, *Pinus sylvestris*). We hypothesized that nutrient deposition in all tree species increases following a high-thinning because of increased canopy roughness causing higher dry deposition and decreases at high harvest intensities because of a decreased canopy roughness and therefore lower dry deposition. We further hypothesized that deposition over the year has stronger seasonal effects for deciduous trees (beech) compared to evergreen trees (both conifers). The results are discussed in view of the importance of deposition for forest nutrient budgets and possible implications for forest productivity and resilience. To address the hypotheses, we measured deposition in 5 experimental forest plots per tree species (with four subplots varying in harvest intensity) over a full year and estimated the total deposition by correcting for canopy exchange (based on Na deposition) and adding estimated stemflow. The nutrients considered involve macronutrients (N, S, Ca, K, Mg, P) and micronutrients (Mn, Cu, Fe, Zn) and the harvesting intensities consist of a high-thinning, shelterwood, clearcut and a non-harvested control.

2 Methods

2.1 Study sites and measurements

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*) in five regions across the Netherlands (Fig. S3.1). All fifteen stands have a temperate maritime climate with a mean annual temperature of 10.4°C and a mean annual rainfall of 805 mm (KNMI, 2022). In each of these fifteen stands, four 0.25 ha subplots were installed to which the harvest intensity treatments high-thinning (~20% of the basal area removed), shelterwood (80% removed), clearcut (100% removed), and unharvested control (0%) were randomly assigned (60 subplots in total). The main characteristics of the sampling sites can be found in Vos et al. (2023a) and (Vos et al., 2023b). In each subplot, 7 deposition samplers were installed in March 2020 (28 samplers per forest stand). The combined collection area of the seven samplers per plot totaled 2016 cm², surpassing the 2000 cm² threshold above which the reliability of the measurement substantially increases (Bleeker et al., 2003). The construction of the deposition samplers is described in detail in **Ch2**. Placement of the samplers in the high-thinning, shelterwood and unharvested control plots was based on canopy cover, assessed by drone-based photographs and a generated digital surface model to capture the canopy cover. Samplers were randomly placed using a stratified random point procedure, dividing the plot into seven equal grids and assigning one sampler to each grid. In each treatment, samplers were positioned either underneath the canopy or exposed, corresponding to the canopy cover of the treatment plot (Table S3.1). Regardless of placement in the control and high-thinning, all samplers were treated as throughfall

collectors which were corrected for canopy exchange, while exposed samplers in the shelterwood and clearcut were assumed to collect bulk deposition only.

Preparation of the resin columns

Measurements of throughfall and bulk deposition were based on the ion exchange resin method in which funnels are connected to a resin column which captures cations and anions from the solution that passes through the column. A total of 430 resin columns were prepared in the week before the installation of the deposition samplers in the field and, every three months, a week before replacement of the columns in the field. The resin that was used was the Amberlite IRN 150 (H^+ and OH^- form) which was washed with demineralized water following the resin preparation described in **Ch2**. 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. Resin columns were replaced every three months, replacement took place in June, September and December 2020 and were operational to March 2021.

Bulk deposition and throughfall measurements

Atmospheric inputs of nutrients by bulk deposition and throughfall were calculated by multiplying the water fluxes outside the forest canopy (i.e., precipitation) and below the forest canopy (i.e., throughfall) with the measured nutrient concentrations in those water fluxes. Water fluxes below the canopy and nutrient concentrations in throughfall were measured in high-thinning, shelterwood and the control, while water fluxes outside the canopy and nutrient concentrations in bulk deposition were measured in the clearcut and shelterwood treatments. Deposition measurements cover the deposition between 21st of March 2020 and the 21st of March 2021. The volume of the rainfall (mL) per funnel was recorded monthly as well as contaminations like presence of foliage in the funnel, organic coarse materials (branches bark) and the presence of bird feces. Funnels with contaminations were detached, rinsed with demineralized water, and re-attached to the resin columns. Resin columns were replaced every three months to capture the seasonal deposition. Parallel to the deposition measurements, blank resin columns were installed in a sun-exposed location and in a location underneath the forest canopy (shade) to correct for internal release of nutrients from the resin. These field blanks were installed in one of the forest sites. 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 columns was done following the procedure described in **Ch2**. First, the resin columns that were contaminated by bird feces were excluded from the extraction. Second, the field resin columns and additional lab blanks, included to correct for sample contamination within the laboratory, were dried to a constant weight at 28 °C. Subsequently, subsamples were taken for 2M KCl extraction followed

by NH_4 , NO_3 and NO_2 , total N and PO_4 concentration analysis using a Segmented Flow Analyzer (SFA type 4000, Skalar Analytical B.V., the Netherlands) and for 3.5 M HCl extraction followed by S, Ca, K, Mg, P, Mn, Cu, Fe and Zn concentration analysis using the ICP-AES (Thermo-Scientific iCAP 6500 DUO, USA).

Corrections for sample contamination

To calculate the nutrient input per field column, extracted concentrations were multiplied by the total weight of the field column for each subsample. To correct for background contamination in the field and in the laboratory, the concentrations of the field- and lab blanks were subtracted from the concentrations per column. For the samplers in the forest gaps, the sunlight-exposed field blank was used, for the samplers underneath the forest canopy, the shade field blank was used. Thereafter, corrected concentrations per funnel were scaled to kg ha^{-1} based on the funnel surface. Finally, for both methods, the samples were checked for contaminations, removing all values outside the 95% confidence interval. To do so, data were normalized, and the 95% confidence interval was calculated based on the mean $\pm$ 2 times the standard deviation. In total 23% of the values in the dataset were missing due to presence of bird feces in the funnels while the statistical check removed an additional 6% of the data resulting in 1200 real observations over the full year of sampling. The removed values and the missing values because of contaminations by bird feces were imputed using the R package *MICE* (multiple interpolations) based on Monte-Carlo simulations.

2.2 *Calculation of total nutrient deposition*

Total deposition calculation

To calculate the total deposition of each nutrient, corrections were made to include the effects of stemflow and canopy exchange for samplers beneath the forest canopy (Fig. 3.1).

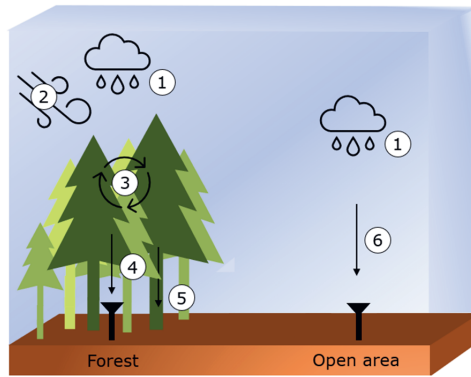


Figure 3.1 Flux Dynamics: Total Deposition in Forests (left) and Open Areas (right). In a forest, atmospheric deposition encompasses (1) wet deposition, (2) dry deposition, (3) alterations within the forest canopy (e.g., canopy exchange), resulting in throughfall flux (4) captured in the sampler (black shape), and stemflow (5). In open areas, deposition primarily comprises wet deposition (1), referred to as bulk deposition when captured in the sampler (6). Due to the absence of a canopy, dry deposition is minimal.

More specifically, the total deposition (X_{td}) of all nutrients (NH_4 , NO_3 , Ca, K, Mg, P, Mn, Cu, Fe, Zn and Na) was calculated as the sum of the measured throughfall (X_{tf}), and calculated stemflow (X_{sf}) minus canopy exchange (X_{ce}), according to:

$$X_{td} = X_{tf} + X_{sf} - X_{ce} \quad (1)$$

Stemflow calculations

Stemflow was calculated based on the assumption that (i) the amount of water entering the soil by stemflow is a function of the daily precipitation and (ii) nutrient concentrations in stemflow are equal to those in throughfall. The calculation of water entering the soil via stemflow was calculated in more detail for beech where stemflow is significant. For beech, stemflow was calculated based on the intensity of the rainfall event (P in mm day^{-1}) with differences in stemflow (SF) between the leaved (SF_L ; formula 2) and the leafless season (SF_{NL} ; formula 3) (Staelens et al., 2008a) multiplied by the canopy cover (CC; %):

$$SF_L = -2.09 + 0.098 * P * CC \quad (2)$$

$$SF_{NL} = -2.09 + 0.140 * P * CC \quad (3)$$

For Douglas fir and Scots pine, the amount of stemflow was calculated as a fraction of the daily rainfall multiplied by the canopy cover (%). The used rainfall fraction was 1% for stemflow in Douglas fir (Spittlehouse, 1998; Spencer and van Meerveld, 2016) and 3% for Scots pine (Pinos et al., 2021). The

daily precipitation of each forest site was collected from nearby weather stations in the network of the Royal Dutch Meteorological Institute (KNMI, 2021a). For each forest site, at least three nearby weather stations were selected, and the daily precipitation data was interpolated based on the distance to the forest site. An overview of the spatial location of these weather stations is provided in Fig. S3.1.

Canopy exchange of the base cations and micronutrients

Canopy exchange fluxes were calculated for nitrogen (NH_4 , NO_3), the base cations Ca, K and Mg and the micronutrients Mn, Cu, Fe and Zn. The canopy exchange of SO_4^{2-} and PO_4^{2-} was assumed negligible, implying that the total deposition was calculated as the sum of throughfall and stemflow. The estimation of the canopy exchange of the base cations and the micronutrients was based on the assumptions that (i) Na does not interact with the forest canopy (inert tracer) and (ii) the ratios of total deposition over bulk deposition are similar for Ca, K, Mg, Mn, Cu, Fe, Zn and Na. Specifically in coastal areas, the second assumption is not always valid (Baloutes. Greece, pers. comm.). Canopy exchange of the base cations and micronutrients was calculated by multiplying the bulk deposition of these cations with the ratio of the sodium input by both throughfall (measured in high-thinning, shelterwood and the control) and stemflow to the sodium input in bulk deposition (measured in the clearcut and shelterwood treatments), according to Ulrich (1983):

$$X_{ce} = (X_{tf} + X_{sf}) - \left(\frac{Na_{tf} + Na_{sf}}{Na_{bd}} * X_{bd} \right) \quad (4)$$

With X_{ce} is the canopy exchange of the base cations (Ca, K and Mg) and the micronutrients (Mn, Fe, Zn and Cu) in mg per funnel and per season and X_{tf} , X_{sf} and X_{bd} are respectively the throughfall deposition, stemflow and bulk deposition in mg per funnel and per season. By doing so, the canopy budget model, developed by Ulrich (1983) and further extended in multiple studies (Bredemeier, 1988; Draaijers and Erisman, 1995; De Vries et al., 1999; De Vries et al., 2001), was slightly extended by the inclusion of the canopy exchange of Mn, Cu, Fe and Zn (Rea et al., 2001; Gandois et al., 2010b).

Canopy exchange of NH_4 and NO_3

Canopy exchange of NH_4 was calculated as a fraction of the base cation canopy exchange as NH_4 and H^+ interact with the forest canopy by exchange with base cations (Roelofs et al., 1985; Draaijers et al., 1997a). We assumed that the total canopy uptake of H^+ (H_{ce}) and NH_4^+ (NH_{4ce}) is equal to the total canopy leaching of the base cations (BC_{ce}) taking place through ion exchange, corrected for the leaching of weak acids (WA_{ce}) (Van der Maas et al., 1991; Draaijers and Erisman, 1995):

$$\text{NH}_{4ce} = \text{BC}_{ce} - \text{WA}_{ce} - H_{ce} \quad (5)$$

There are three potential methods to estimate the weak acid (WA) concentration, i.e. (i) from the sum of HCO_3^- , derived from the pH and an assumed atmospheric CO_2 pressure, and RCOO^- , derived from DOC, (ii) from the measured alkalinity, while correcting for the pH and (iii) from the difference in concentration of the cations of H^+ and NH_4^+ , Ca^{2+} , K^+ , Mg^{2+} and Na^+ minus the strong acid anions SO_4^{2-} , NO_3^- and Cl^- (De Vries et al., 1999; De Vries et al., 2001). None of these methods were possible in our study because pH, DOC or alkalinity could not be measured using IER and Cl^- was not included in the analysis. Consequently we assumed that NH_4 exchange is $1/3$ of the base cation leaching based on De Vries et al. (1999) and De Vries et al. (2001) who found that H^+ exchange (uptake), NH_4^+ exchange (uptake) and WA exchange (leaching) are all similar and consist of $1/3$ of the base cation canopy leaching. Finally, the canopy exchange of NO_3 was calculated as the canopy exchange of N minus the canopy uptake of NH_4 (NH_{4ce}), in which the total N canopy exchange was calculated by accounting for the contribution of NH_4 and NO_3 to total N input by throughfall and stemflow, according to (De Vries et al., 2001; Adriaenssens et al., 2011):

$$\text{NO}_{3ce} = (\text{NH}_{4ce} * \left(\frac{(\text{NH}_{4tf} + \text{NH}_{4sf}) * x\text{NH}_4 + (\text{NO}_{3tf} + \text{NO}_{3sf})}{(\text{NH}_{4tf} + \text{NH}_{4sf}) * x\text{NH}_4} \right)) - \text{NH}_{4ce} \quad (6)$$

In which $x\text{NH}_4$ is a correction factor, which is assumed to be 5, implying that canopy uptake of NH_4 is much higher than of NO_3 .

2.3 Statistical analysis

All statistical analysis were performed in R version 4.1.0. Prior to statistical analysis, the total deposition was scaled to 1-ha deposition levels by multiplication of the funnel surface (288 cm^2) to the surface of a 1-ha plot. To compare the total seasonal deposition between different harvest intensity and species, two-way ANOVA tests were used. These analyses were performed by using linear mixed-effect models from the R package *nlme* using regions as a random structure. To meet the normality and homogeneity assumptions, skewed data were log transformed and, when necessary, additional VarComb and VarIdent variance structures were used which allow different variances between factor levels (Zuur et al., 2009). Tukey's post-hoc (HSD) test was performed following ANOVA based on the linear mixed-effect models using the R package *emmeans* to test for differences between seasons, harvest intensities and species. To test whether harvest intensity and tree species significantly explained the differences in seasonal total deposition, throughfall and canopy exchange of the different nutrients we conducted a partial Redundancy Analysis (p-RDA). This analysis calculates the variance explained by the different harvest intensities, the seasons, and the species. A fourth p-RDA was performed to distinguish between the factors driving the total annual deposition. Prior to the p-RDA, all data were log transformed to meet the linearity assumption and the p-RDA was conducted using the *vegan* package (Oksanen et al., 2022).

3 Results

3.1 Annual water fluxes and stand deposition

Average bulk precipitation measured from April 2020 to March 2021 in clearcut and shelterwood treatments of beech, Douglas fir and Scots pine was 580 mm (± 29 s.e.), 580 mm (± 40 s.e.) and 610 mm (± 36 s.e.) respectively. Bulk precipitation per study site is given in Table S3.2. Precipitation based on data of 21 nearby weather stations was on average 736 mm (± 17 s.e.) for the same period (Fig. S3.1, Table S3.2) being ≥ 100 mm higher than the bulk precipitation in the clearcuts indicating edge effects in the clearcut. Stemflow in the unharvested stands for beech, Douglas fir and Scots pine was estimated to be 47 mm (± 3.5 s.e.), 5.6 mm (± 0.34 s.e.) and 16 mm (± 0.93 s.e.), respectively. The annual interception, calculated as the difference between throughfall plus stemflow and the precipitation of nearby weather stations (bulk deposition was expected to be influenced by edge effects), was highest in the unharvested stands of Douglas fir (310 mm ± 6.0 s.e.) and lower in unharvested Scots pine and beech stands (respectively 240 mm ± 7.9 s.e. and 260 mm ± 6.5 s.e.). The throughfall and therefore the interception estimates varied across stands, season and harvest treatments (Table S3.2).

The nutrient concentration of the throughfall deposition was generally highest in Douglas fir (except for P), intermediate in Scots pine and lowest in beech (Table 3.1). The calculated nutrient input of the stemflow was highest in beech, adding up to 10% of the stand deposition (throughfall + stemflow) input (Table 3.1). The estimated stemflow in Douglas fir and Scots pine was relatively low. The annual canopy exchange, i.e., the uptake or release of nutrients by the foliage, was negative for NH_4 , NO_3 , Ca, Cu, Fe and Zn, implying that there was net uptake of these nutrients by the canopy (Table 3.1). In contrast, K, Mg (except for Scots pine) and Mn leached from the canopy whereby canopy leaching of K accounted for approximately 65% of the stand deposition reaching the forest floor in both beech and Scots pine and around 50% in Douglas fir. For Mn, canopy leaching accounted for 53% of the throughfall and stemflow flux in beech, and 22% and 30% for Douglas fir and Scots pine respectively (Table 3.1). Total deposition of N compounds, going from ca 27 kg ha⁻¹ yr⁻¹ in Scots pine to ca 31 kg ha⁻¹ yr⁻¹ in beech and ca 36 kg ha⁻¹ yr⁻¹ in Douglas fir, was 1.7 up to 2.5 times higher compared to bulk deposition (Table 3.1). The highest ratios of total deposition to bulk deposition were observed for base cations, S and Mn: base cations exhibited deposition levels 2 to 3 times greater than bulk deposition, with a fourfold increase observed for K in Douglas fir.

Table 3.1 The means and standard errors of the annual throughfall, stemflow, canopy exchange, total deposition and bulk deposition (all in kg ha⁻¹ yr⁻¹) and the ratio total deposition/bulk deposition of macro (NH₄, NO₃, total N, 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). Canopy exchange of S and P is assumed to be negligible (section 2.5.3). Data of the high-thinning and shelterwood are given in Table S3.3 and S3.4.

Species		NH ₄	NO ₃	S	Ca	K	Mg	P	Mn	Cu	Fe	Zn
Throughfall	BE	16 ± 3.0	5.8 ± 1.1	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
		24 ± 3.3	8.5 ± 1.2	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
	DG	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
		24 ± 0.29	8.5 ± 0.11	12 ± 0.037	9.9 ± 0.11	29 ± 0.19	4.7 ± 0.022	0.57 ± 0.0092	0.31 ± 0.005	0.021 ± 2.9 e ⁻⁴	0.17 ± 0.00067	0.62 ± 0.0023
	SP	18 ± 0.055	5.6 ± 0.014	6.1 ± 0.051	7.1 ± 0.031	19 ± 0.035	2.5 ± 0.011	0.49 ± 0.0022	0.17 ± 0.00098	0.014 ± 0.00061	0.13 ± 0.00068	0.3 ± 0.0069
		18 ± 0.051	5.6 ± 0.012	6.1 ± 0.048	7.1 ± 0.034	19 ± 0.098	2.5 ± 0.0082	0.49 ± 0.0039	0.17 ± 0.00073	0.014 ± 0.78 e ⁻⁴	0.13 ± 0.00061	0.3 ± 0.0077
Stemflow	BE	1.5 ± 0.29	0.54 ± 0.11	0.57 ± 0.037	0.91 ± 0.11	2.7 ± 0.19	0.27 ± 0.022	0.079 ± 0.0092	0.023 ± 0.005	9.4 e ⁻⁴ ± 2.9 e ⁻⁴	0.012 ± 0.00067	0.011 ± 0.0023
		0.32 ± 0.055	0.11 ± 0.014	0.17 ± 0.051	0.14 ± 0.031	0.4 ± 0.035	0.066 ± 0.011	0.0077 ± 0.0022	0.0043 ± 0.00098	0 ± 0	0.0022 ± 0.00068	0.0085 ± 0.0069
	DG	0.59 ± 0.051	0.19 ± 0.012	0.22 ± 0.048	0.25 ± 0.034	0.71 ± 0.098	0.088 ± 0.0082	0.018 ± 0.0039	0.0061 ± 0.00073	1.4 e ⁻⁴ ± 0.78 e ⁻⁴	0.0045 ± 0.00061	0.012 ± 0.0077
		0.53 ± 0.050	0.042 ± 0.036	0.042 ± 0.036	0.55 ± 0.69	1.6 ± 1.0	0.027 ± 0.29	0.010 ± 0.009	± 0.001	0.006 ± 0.015	± 0.012	± 0.037
	SP	-3.3 ± 0.27	-0.18 ± 0.012	-0.18 ± 0.012	-4.0 ± 0.51	14 ± 0.43	0.098 ± 0.044	0.068 ± 0.006	-0.016 ± 0.001	-0.18 ± 0.009	-0.31 ± 0.017	-0.2 ± 0.017
		-3.1 ± 0.27	-0.17 ± 0.012	-0.17 ± 0.012	-3.8 ± 0.51	13 ± 0.43	-0.3 ± 0.044	0.052 ± 0.006	-0.0098 ± 0.001	-0.16 ± 0.009	-0.2 ± 0.017	-0.2 ± 0.017
Canopy exchange	BE	24 ± 1.2	6.8 ± 3.2	6.0 ± 0.4	13 ± 1.3	9.6 ± 1.1	2.7 ± 0.23	0.97 ± 0.089	0.12 ± 0.010	0.025 ± 0.0034	0.25 ± 0.016	0.20 ± 0.043
		27 ± 1.2	8.8 ± 2.8	12 ± 3.2	14 ± 2.6	16 ± 2.8	4.8 ± 0.80	0.57 ± 0.13	0.24 ± 0.044	0.036 ± 0.0075	0.35 ± 0.069	0.94 ± 0.73
	DG	21 ± 0.19	6.0 ± 1.5	6.3 ± 1.1	11 ± 1.0	6.7 ± 0.26	2.9 ± 0.15	0.51 ± 0.066	0.12 ± 0.012	0.024 ± 0.0043	0.29 ± 0.043	0.51 ± 0.34
		21 ± 1.3	6.0 ± 0.63	6.3 ± 0.11	11 ± 0.48	6.7 ± 0.33	2.9 ± 0.1	0.51 ± 0.14	0.12 ± 0.0054	0.024 ± 0.0011	0.29 ± 0.0069	0.51 ± 0.012
	SP	11 ± 1.2	4.1 ± 1.0	3.9 ± 0.33	4.7 ± 0.51	3.9 ± 0.61	1.6 ± 0.11	0.46 ± 0.040	0.073 ± 0.0083	0.011 ± 0.0013	0.13 ± 0.017	0.29 ± 0.22
		13 ± 1.2	5.2 ± 0.76	3.6 ± 0.44	5.4 ± 0.64	2.7 ± 0.35	1.4 ± 0.21	0.56 ± 0.12	0.05 ± 0.0023	0.012 ± 0.0014	0.15 ± 0.016	0.19 ± 0.13
Total deposition	BE	14 ± 1.3	4.3 ± 0.63	3.4 ± 0.11	6.3 ± 0.48	3.4 ± 0.33	1.4 ± 0.1	0.80 ± 0.14	0.056 ± 0.0054	0.013 ± 0.0011	0.13 ± 0.0069	0.08 ± 0.012
		11 ± 1.2	4.1 ± 1.0	3.9 ± 0.33	4.7 ± 0.51	3.9 ± 0.61	1.6 ± 0.11	0.46 ± 0.040	0.073 ± 0.0083	0.011 ± 0.0013	0.13 ± 0.017	0.29 ± 0.22
	DG	13 ± 1.2	5.2 ± 0.76	3.6 ± 0.44	5.4 ± 0.64	2.7 ± 0.35	1.4 ± 0.21	0.56 ± 0.12	0.05 ± 0.0023	0.012 ± 0.0014	0.15 ± 0.016	0.19 ± 0.13
		13 ± 1.2	5.2 ± 0.76	3.6 ± 0.44	5.4 ± 0.64	2.7 ± 0.35	1.4 ± 0.21	0.56 ± 0.12	0.05 ± 0.0023	0.012 ± 0.0014	0.15 ± 0.016	0.19 ± 0.13
	SP	1.7 ± 2.5	1.6 ± 2.1	1.8 ± 3.1	2.1 ± 3.0	2.8 ± 4.1	1.9 ± 3.0	1.2 ± 1.2	2.1 ± 3.3	1.9 ± 3.3	1.9 ± 2.7	2.5 ± 3.2
		1.6 ± 1.6	1.2 ± 1.2	1.8 ± 1.8	2.0 ± 2.0	2.5 ± 2.5	2.1 ± 2.1	0.91 ± 0.91	2.4 ± 2.4	2.0 ± 2.0	1.9 ± 1.9	2.7 ± 2.7
Ratio	BE	1.7	1.6	1.8	2.1	2.8	1.9	1.2	2.1	1.9	1.9	2.5
	SP	1.6	1.2	1.8	2.0	2.5	2.1	0.91	2.4	2.0	1.9	2.7

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

The total annual deposition ranged from 0.01 kg ha⁻¹ yr⁻¹ (Cu) to 36 kg ha⁻¹ yr⁻¹ (N) across nutrients (Fig. 3.2). Across species, deposition levels were higher in Douglas fir stands compared to both beech and Scots pine, with the latter two showing only marginal differences (Fig. 3.2, Table S3.5). Higher total annual deposition in closed Douglas fir stands is related to the tree height while total annual deposition in closed beech stands is related to stand age and canopy cover and in closed Scots pine stands to LAI and stand density (partial-RDA, Var = 5.0, F = 3.0, p = 0.038, R₂-adj = 0.65, Fig. S3.2).

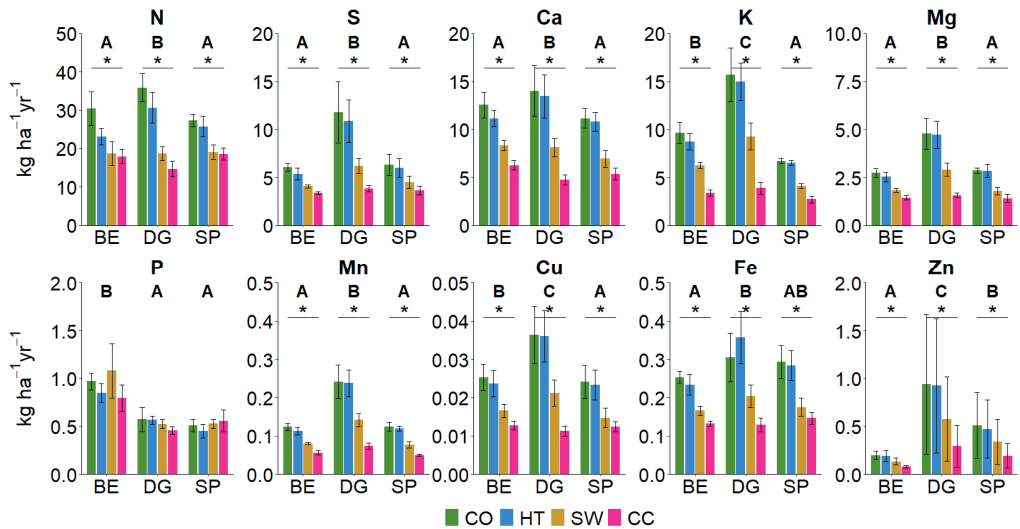


Figure 3.2 Total annual nutrient deposition (kg ha⁻¹ yr⁻¹) in the control (CO) and in the harvest intensities high-thinning (HT), shelterwood (SW) and clearcut (CC) for beech, Douglas fir and Scots pine. Different capital letters denote significant differences between species, asterisks indicate significant differences amongst different harvest intensities (nested-ANOVA, n=5, P<0.05). Differences between NO₃ and NH₄ deposition are given in Fig. S3.3 and differences between harvest intensities are given in Table S3.5.

The harvest intensity significantly influenced the total annual deposition (Fig. 3.2, Table S3.5): for most nutrients (P was an exception), deposition decreased with harvest intensity (thus with stand openness) from control forest to clearcut. Weak but consistent differences were present between control and high-thinning, while differences were more pronounced between high-thinning, shelterwood and clearcut (Table S3.5). Notwithstanding the similarity in these qualitative trends across species, species significantly differed in treatment effect sizes for NH₄, NO₃, K, S, Mg, Mn, and Cu, and not for Ca, P, Zn and Fe. Relatively large deposition differences between shelterwood and clearcut were observed for

Douglas (particularly for NO_3 , K, S, Mg, Mn and Cu), while such differences were much weaker for Scots pine and almost absent for beech (Fig. 3.2, Table S3.5).

The species-specific capacity to intercept deposition resulted in varying acid (sum of NH_4 , NO_3 and S) and base cation inputs. The acid input varied between $2.3 \text{ keq ha}^{-1} \text{ yr}^{-1}$ in Scots pine to $3.5 \text{ keq ha}^{-1} \text{ yr}^{-1}$ in Douglas fir and the base cation input varied from $0.96 \text{ keq ha}^{-1} \text{ yr}^{-1}$ in Scots pine to $1.5 \text{ keq ha}^{-1} \text{ yr}^{-1}$ in Douglas fir. Both the acid input and the base cation input decreased from control to clearcut (Fig. 3.3). The net acid input, which consists of the acid input minus the base cations, also declined from control to shelterwood and stabilized or slightly increased towards the clearcut. Strongest decline in the net acid input was observed for Douglas fir while in Scots pine only a slight decline was observed (Fig. 3.3).

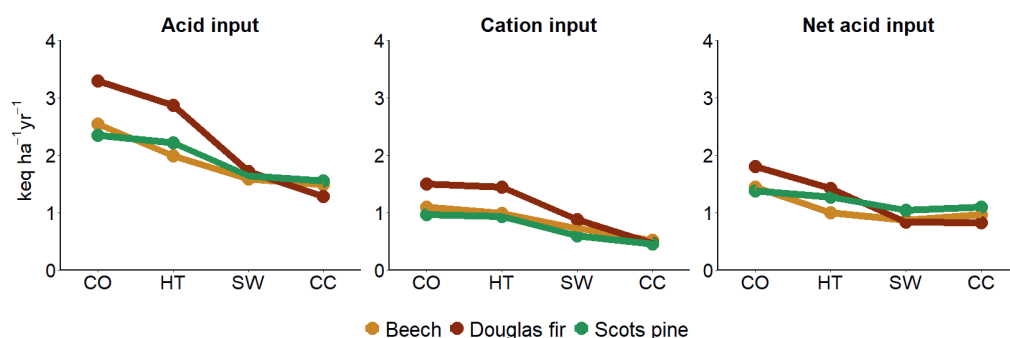


Figure 3.3 The total acid input by atmospheric deposition (sum of NH_4 , NO_3 and S), the base cation input (sum of Ca, K and Mg) and the total net acid input by atmospheric deposition (sum of NH_4 , NO_3 and S minus the sum of the base cations) in $\text{keq ha}^{-1} \text{ yr}^{-1}$ for the unharvested control (CO), high-thinning (HT), Shelterwood (SW) and clearcut (CC) harvest intensity.

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

Significant variations in total deposition of various nutrients were observed across seasons, with this seasonal variation varying among nutrients and across different species and harvest intensities. The percentage of variation within the total deposition explained by species, harvest intensity and season was 61% (partial-RDA, $\text{Var} = 6.4$, $F = 45$, $p < 0.001$, $R^2\text{-adj} = 0.69$, Fig. 3.4), with harvest intensity and season as primary drivers (respectively 29% and 23%), whereas species contributed a smaller proportion (8.7%).

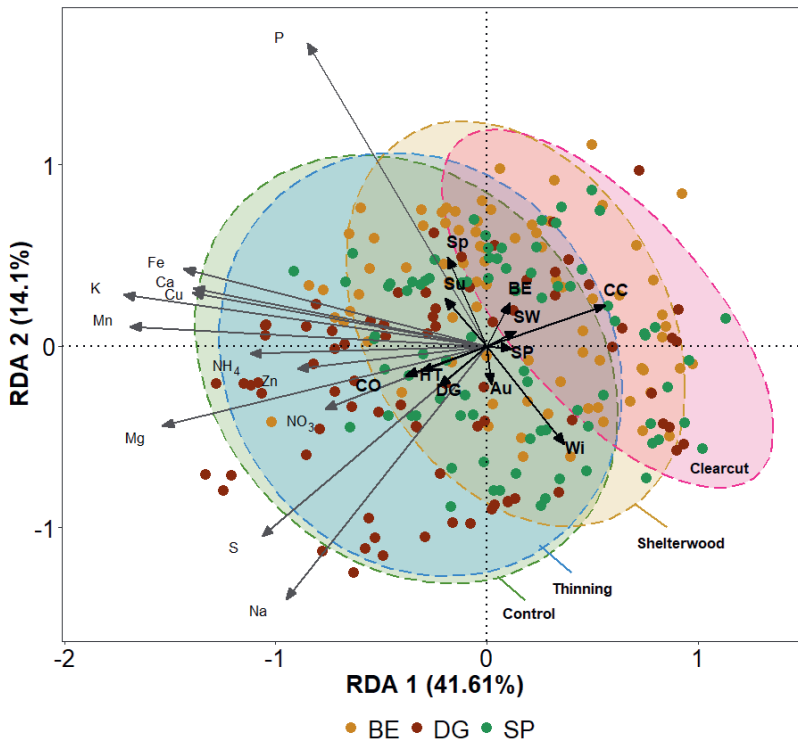


Figure 3.4 Total seasonal deposition of macro- (NH_4 , NO_3 , S, P, Ca, K and Mg) and micronutrients (Mn, Cu, Fe and Zn) (in kg ha^{-1}) explained by the centroids of the harvest intensities (control : clearcut) and species (BE: beech, DG: Douglas fir and SP: Scots pine)(RDA biplot). Total seasonal deposition of macro- and micronutrients is represented by the grey arrows, the effects of species, harvest intensity and seasons (Sp: spring, Su: Summer, Au: Autumn, Wi: Winter) by black arrows. T-distribution polygons are shown for the four harvest intensities, dots are colored by species. The length of arrows denotes the variation within the canopy exchange explained by species, treatments, and seasons.

Most of the variation of the total seasonal deposition as described by the first axis (explaining 42% of the variation) was determined by the difference between the stand openness moving from closed forest to a clearcut (Fig. 3.4). This axis also showed strong negative associations with most nutrients (Ca, Cu, Fe, K, Mn, NH_4 and Zn), confirming that deposition levels of those nutrients decrease with increasing harvest intensity (or stand openness). These nutrients do vary between different seasons and across species although this variation is subordinate to the variation caused by harvest intensity (Table 3.6, Table S3.5, Fig. S3.4).

Seasonal variations of the total deposition determined the variance explained by the second, third and fourth significant RDA axis. The second axis (explaining 14% of the variation) showed large differences

in total deposition between spring and winter, and to a lesser extend summer and winter, had only a strong association with a few nutrients: a positive association with P and a negative association with S and Na. The total P deposition varied strongly over the seasons, with higher deposition in spring and summer compared to autumn and winter (Fig. S3.4), while there was no effect of harvest intensity and only a minor effect of tree species (Table S3.5). The total deposition of S and Na increased from spring to winter, with deposition values decreasing from control to clearcut and consequently higher deposition in Douglas fir stands (Fig. S3.4, Table S3.5). The third and fourth significant RDA axis were of minor importance, explaining only 3.2% and 1.1% of the variance. Nevertheless, they were associated with the differences between summer and autumn (axis 3) and autumn and winter (axis 4). The variations in atmospheric deposition between species across different seasons were subtle. For example, there was a slightly higher seasonal difference in the total deposition in control and thinned stands in Douglas fir compared to Scots pine (Table S3.6). In beech and Scots pine, harvest intensity had a small effect on the total deposition of elements such as Fe, Zn and Cu in winter (Fig. S3.4). Overall, the analysis shows that harvest intensity and season have relatively independent effects on deposition, and that species effects are relatively minor when accounting for those harvest intensity and seasonal effects.

3.4 *Impacts of tree species and harvest intensity on throughfall and canopy exchange*

The total seasonal deposition in forests primarily consists of throughfall corrected for the canopy exchange, variations can therefore be attributed to either throughfall or canopy exchange fluxes. The variation within the throughfall (control : shelterwood) explained by species, harvest intensity, season, precipitation and precipitation interception was 64% (partial-RDA, $\text{Var} = 5.9$ F = 25, $p < 0.001$, $R^2\text{-adj} = 0.48$, Fig. 3.5A), with season as primary driver (25%) followed by species and harvest intensity (respectively 15% and 12%) while precipitation and precipitation interception together only explained 1.4%.

Canopy exchange of nitrogen (NH_4 , NO_3), the base cations (Ca, Mg, K) and the micronutrients (Mn, Cu, Fe and Zn) varied by season, and was affected by harvest intensity and tree species while precipitation and precipitation interception had hardly any effect on canopy exchange (38% of variation explained, p-RDA, $\text{Var} = 3.0$, F = 9.9, $p < 0.001$, $R^2\text{-adj} = 0.31$, Fig. 3.5B). Harvest intensity (from control to shelterwood-cut) accounted for 15% of the total variation, season for 12% and tree species for 4.6% of the total variation. The combined water fluxes (precipitation, and interception) accounted only for 1.4% of the total. In general, canopy uptake of Zn, Cu, Fe and Ca and canopy leaching of K and Mn were highest in the unharvested stand. Canopy leaching of NH_4 and NO_3 was highest in the autumn and lowest in the spring, furthermore, canopy leaching of NH_4 and NO_3 was highest in beech stands and lower in both Douglas fir and Scots pine. There was a slight net canopy uptake of Mg in beech, while in Scots pine canopy uptake only occurred in the autumn (Fig. 3.5B).

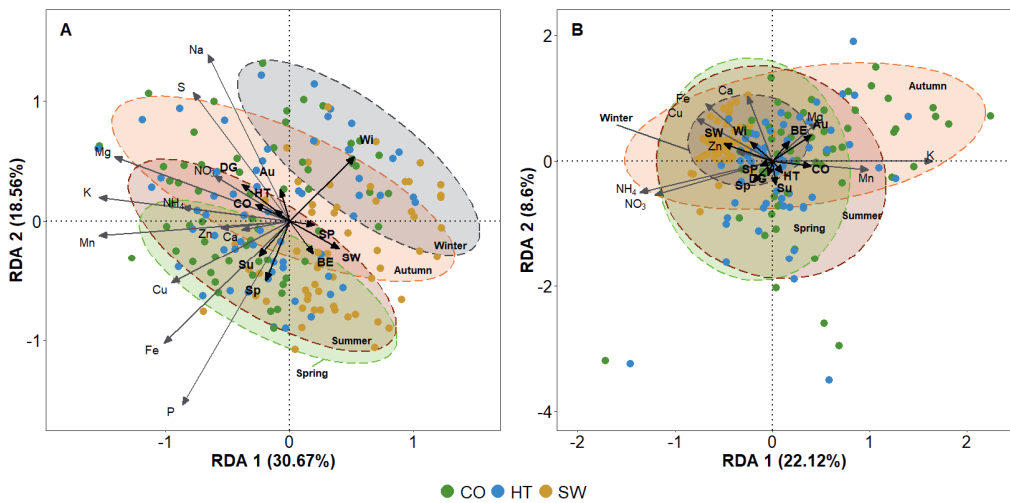


Figure 3.5 A: Seasonal throughfall of macro- (NH_4 , NO_3 , S, P, Ca, K and Mg) and micronutrients (Mn, Cu, Fe and Zn) and Na (in kg ha^{-1}) (A) and seasonal canopy exchange of macro- (NH_4 , NO_3 , Ca, K and Mg) and micronutrients (Mn, Cu, Fe and Zn) (B) explained by the centroids of harvest intensity (control : shelterwood), seasons and species (BE: beech, DG: Douglas fir and SP: Scots pine)(RDA biplot). Seasonal throughfall (A) and canopy exchange (B) are represented by the grey arrows, the effects of species, harvest intensity and seasons (Sp: spring, Su: Summer, Au: Autumn, Wi: Winter) by black arrows. T-distribution polygons are shown for the four seasons, dots are colored by the harvest intensity. The length of arrows denotes the variation explained by species, treatments, and seasons.

4 Discussion

4.1 The annual total nutrient inputs in Dutch forests: implications for soil acidification

In sustainable forestry, nutrient stocks should not decline over subsequent rotations, depending on both nutrient inputs through deposition and weathering, as well as nutrient losses due to biomass exports by harvesting trees and leaching. The total deposition of most nutrients in closed stands was within the range of deposition values 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 was considerably different from deposition values used in the current nutrient balance model predictions for forests in the Netherlands (de Vries et al., 2021). When comparing total deposition to the deposition maps of the Netherlands (RIVM, 2020; 2021), which is predominantly measured on short vegetations (Hoogerbrugge et al., 2022), through overlay of the locations, we found that average NH_4 and S deposition in forests is across species respectively 30-46% and 35-140% higher than the national estimate while the average NO_y deposition is between 11% lower to 21% higher (Fig. 3.2, Table S3.7) (RIVM, 2020; 2021). Base cation

input in this study was much higher compared to the values reported by Van Jaarsveld et al. (2010). We found that, across species, the average Ca deposition was 110-160% higher, K 260-750% higher and Mg 68-190% higher (Fig. 3.2, Table S3.8). We also found much higher P deposition levels compared to the expected P deposition in the Netherlands (de Vries et al., 2021). Comparative data on the deposition of Mn, Cu, Fe and Zn is lacking, however, we expect that forests receive more Mn, Cu, Fe and Zn because the deposition in the control stands was 2 to 3 times higher than in the clearcuts. Overall, this indicates that the current deposition rates in forests are much higher than anticipated.

The elevated acid deposition (N+S) in forests raises concerns, especially regarding the potential decrease of essential base cations (Ca, K, and Mg) in forests on sandy soils. This is a result of N and, historically, S deposition-induced soil acidification, intensifying leaching processes (Leeters et al., 2007; de Vries et al., 2021). Assessing the ratio of N and S to Ca, Mg, and K in total deposition provides an initial indicator of the acidification potential of the deposition (de Vries et al., 2003). Results indicate that total N inputs for the three tree species range from approximately 1.9 to 2.6 keq ha⁻¹ yr⁻¹, while S inputs vary from around 0.3 to 0.8 keq ha⁻¹ yr⁻¹, resulting in a total acid input of 2.2-3.4 keq ha⁻¹ yr⁻¹ (Fig. 3.3). Base cation inputs across species vary from approximately 0.6-0.7 keq ha⁻¹ yr⁻¹ for Ca, 0.2-0.4 keq ha⁻¹ yr⁻¹ for K, and 0.1-0.2 keq ha⁻¹ yr⁻¹ for Mg totaling a base cation input of 0.9-1.3 keq ha⁻¹ yr⁻¹ (Fig. 3.3). Despite these high base cation deposition inputs, the potential acid input is over twice as much, implying a potential loss of base cations due to leaching. However, the net acidic input (N + S – base cations), Fig. 3.3) is much lower than expected based on the average deposition data of the Netherlands (Table S3.7-S3.8) implying that, despite the higher N deposition, the potential loss of cations due to leaching might be lower than initially expected (Van Jaarsveld et al., 2010; RIVM, 2020; 2021).

The study's total deposition values, already partly exceeding the national average, may slightly underestimate actual values. Firstly, the calculation of stemflow volume in Douglas fir and Scots pine doesn't consider precipitation intensity, impacting stemflow in high-intensity rain. Additionally, the underestimation is linked to using throughfall concentrations to estimate stemflow nutrient concentrations. Stemflow nutrient concentrations can be significantly higher for, for example, K and Mg (up to three times) and Ca (up to two times) (Tan et al., 2018). While these discrepancies may be negligible for Douglas fir and Scots pine with relatively low stem flow contributions, they could be more relevant for beech trees.

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

The canopy openness of forests - resulting from different harvest intensities - has a larger effect on the total nutrient deposition than tree species or season (Fig. 3.4). Total annual deposition decreased with harvest intensity for all nutrients, except for P. Overall, reductions in deposition were relatively small from control forest to high-thinning, relatively large from high-thinning to shelterwood forest, and intermediate

from shelterwood to clearcut (Fig. 3.2, Table S3.5). These results are – at least partially - in agreement with other case studies (Bäumler and Zech, 1997; Aboal et al., 2000; Gielis et al., 2009; Göttlein et al., 2023). Yet, the expected positive effect of a rougher canopy on dry deposition in the high thinning, compared to a closed control forest, did not occur because dry deposition seemed to be lower. Initially, we hypothesized that thinnings would increase canopy roughness and, consequently, enhance the capture of particles by creating irregular, small openings in the forest canopy. However, we observed higher precipitation throughfall in the thinning (Table S3.2), yet a reduced capture of dry deposition, as also indicated by other studies (Stogsdill Jr et al., 1989; del Campo et al., 2022). Reduced dry deposition is caused by a decline in the aboveground surface area as dry deposition is largest at dense canopies with high LAI (Aboal et al., 2000; Yazbeck et al., 2021). The decrease in the dry deposition following a thinning therefore confirms that the tree canopy itself, rather than the irregularities in the forest canopy, determine the capture efficiency of the dry deposition.

A stronger decline of total deposition was observed after a shelterwood and a clearcut, corresponding to the greater losses in canopy cover (Fig. 3.2). The effects of shelterwood harvest on the total annual deposition was hardly studied before. The effect of a clearcut (100% basal area reduction) is comparable to the difference between bulk deposition and the total deposition of a closed forests (see for example Weis et al. (2006)). However, in our study the total deposition in the clearcut is not fully comparable to bulk deposition as nearby weather stations consistently recorded a 10% higher precipitation input, implying that the small size of the clearcut (circa 50x50 m) in our experiment underestimated the bulk deposition by 10% as a result of edge effects (Neal et al., 1993; Dam, 2001; den Ouden and Mohren, 2020). These edge effects could, in turn, enhance the throughfall deposition of the adjacent treatment plots (De Ridder et al., 2004; Wuyts et al., 2008; Wuyts et al., 2009). This indicates that the total deposition in small forest clearcuts might be consistently lower than the actual bulk deposition measured in large open areas.

Generally, the reduction in total deposition following the shelterwood-cut ($\pm$ 33-40% reduction) and clearcut ($\pm$ 50-68% reduction) compared to the closed stand can be attributed to lower interception of dry deposition due to a sharp decrease in canopy cover (Yazbeck et al., 2021). This decrease is not proportional to the canopy cover, as wet deposition (i.e., precipitation), except at the forest edges, is generally independent of the canopy cover (Dam, 2001; Erisman and Draaijers, 2003). The extent of the dry deposition and, therefore, the impact of harvest intensity differ strongly between regions. For example, heavy thinning in *Picea abies* in southern Germany decreased total deposition rates by $\pm$ 45% (Bäumler and Zech, 1997), while clearcuts in eastern Austria reduced deposition by $\pm$ 40% (Berger et al., 2009). In our study, dry deposition plays a key role, given the strong decline in total deposition from closed forest to clearcut as shown by the ratio of bulk to total deposition (Fig. 3.2). In other regions (e.g. the Netherlands

and France) (Draaijers et al., 1992; Marques and Ranger, 1997), the reductions from closed forest to clearcuts were similar to our study, or smaller (e.g. Austria) (Berger et al., 2009), or even larger (e.g. Germany) (Rothe et al., 2002a; Herrmann et al., 2006). These results imply that the effect of harvest intensity on the reduction of the total deposition is geographically determined. The results underscore the importance of considering geographical factors in understanding the effects of harvest intensity on atmospheric deposition, indicating that results cannot be directly extrapolated from our study to other areas.

4.3 *Harvest intensity effects differs between nutrients.*

The effects of harvest intensity differed strongly between nutrients. The lowest impact of harvest intensity was observed for P, NO₃, and NH₄, while substantial reductions were found for K, Mn, and Zn (Table S3.5). Generally, nutrients that are mainly deposited dry will show the largest reductions in response to tree harvest. The effects of harvest, therefore, more severely influence the total deposition of Na, Cl, Mg, and S in coastal areas (Ten Harkel, 1997; Tørseth et al., 1999; Hellsten, 2007), of N, P, S, and Zn in areas close to anthropogenic sources of pollution (Semb et al., 1995; Tørseth et al., 1999; Balestrini et al., 2007; Hellsten, 2007; Mamun et al., 2020), or other specific sources such as Ca-rich Sahara dust in southern Europe (Semb et al., 1995; Hellsten, 2007). The high share of the dry deposition for K in this study can originate from different sources, such as marine (Hellsten, 2007; Morselli et al., 2008), but also anthropogenic, including agricultural practices, traffic and wind-blown dust (Draaijers et al., 1996b; Tørseth et al., 1999). Dry deposition of Mn often comes from terrestrial sources including anthropogenic pollution (Navrátil et al., 2007; Hsu et al., 2010). The high deposition of Zn in this study can, in addition to the effects of traffic (Gunawardena et al., 2013), be related to a former Zinc factory in the Southern part of the Netherlands, resulting in notable differences between sites (Fig. 3.2). Generally, the reductions presented in this study across nutrients were in line with the literature, except for the 50% higher K throughfall and 50% lower Mg throughfall underneath, respectively, Scots pine and Douglas fir compared to similar stands in 1990 (Van Ek and Draaijers, 1994). These differences might be related to the canopy uptake of Mg in Scots pine (Table 3.1, Table S3.3-S3.4) and the low K content in needles of Douglas fir (Vos et al., 2023b), reducing canopy leaching due to lower soil fertility (Nordén, 1991).

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

The total deposition within different harvest intensities differs between species and nutrients which is related to a species' capacity to intercept dry deposition. The higher deposition in Douglas fir stands compared to beech and Scots pine stands (Fig. 3.2) is in agreement with other studies (Van Ek and Draaijers, 1994; Rothe et al., 2002a; Zhang et al., 2022), and related to Douglas stands often being taller

than the other two species (Fig. S3.2) increasing the dry deposition capacity (Lovett and Reiners, 1986; Erisman and Draaijers, 2003). Surprisingly, factors driving the dry deposition like 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) only explain 13% of the variation within the total deposition of closed stands in this study (Fig. S3.2). Absence of a clear effect of tree height might be related to the use of the absolute tree height instead of the relative height compared to surrounding stands. The p-RDA indicates that species differences in the interception of deposition in closed forests are mainly driven by stem density (explaining 44% of the variation), which was on average highest for Scots pine and lowest for Douglas fir (Table S1.1). We, however, consider the observed negative effect of stand density on dry deposition as indirect. Douglas firs have much lower density but can capture more dry deposition because they are taller and have a higher LAI compared to Scots pine, with beech taking an intermediate position. Overall, these species effects reduced with increasing harvest intensity. Thus, moving from closed to open stands, higher reductions were observed for Douglas fir compared to both beech and Scots pine (Fig. 3.2, Table S3.5). Similar species effects were observed in other studies, i.e. for example higher throughfall reductions in harvested *Picea abies* stands compared to mixed oak stands (Bäumler and Zech, 1997; Wheeler et al., 2000). With increasing tree harvest intensity, the species differences in dry deposition thus go down.

4.5 Harvest intensity and tree species in relation to soil acidification

Harvest intensity could potentially be a management tool to ease the effects of acidifying deposition since the effects of canopy openness (controlled by harvest) on deposition differed between acidifying nutrients (N and S) and the base cations (Ca, Mg and K) (Fig. 3.3). Generally, acidic inputs of atmospheric deposition are highest underneath closed stands of conifers due to the high capacity to intercept dry deposition as a result of enhanced capturing of particles and water by the needles (De Schrijver et al., 2007; Pierret et al., 2019). We however did not find evidence for higher acidic inputs underneath closed stands of conifers compared to broadleaved tree species, contrary to De Schrijver et al. (2007) and Rothe et al. (2002a). Nevertheless, when this acidic input is corrected for the base cations, conifers receive higher loads of net acidic inputs (Fig. 3.3). Higher neutralizing capacity of beech compared to conifers was reported before and is in line with the observation that conifers cause acidification of the precipitation (Kowalska et al., 2016; Pierret et al., 2019). These results underline the well-known higher risks of soil acidification in coniferous stands compared to broadleaved stands (De Schrijver et al., 2012). When moving from closed forests to more open stands, we found strong declines in the net acidic input towards the shelterwood and slight increase from shelterwood towards clearcuts (Fig. 3.3). This decline is present in all species but strongest for the shelterwood in Douglas fir followed by the high-thinning and shelterwood treatments in

beech for which the net acidic input declined by $\pm 50\%$. These results indicate that regular thinnings, especially in beech and Douglas fir, could provide a management tool to slow down soil acidification.

4.6 *Seasonal variations in total deposition*

The total deposition varies largely amongst seasons, indicating that seasonal variations is a major component in understanding annual deposition patterns, especially for P, S and N (Fig. 3.4). The total P deposition varies strongly between seasons, with higher deposition during the growing season compared to the dormant season, particularly in beech stands (Fig. S3.4). This elevated P deposition in the growing season contradicts expectations, as P is usually actively taken up by the canopy in oligotrophic ecosystems (Helmisaari and Mälkönen, 1989; Gordon et al., 2000; Houcai et al., 2021). Canopy leaching of P, a potential contributor to up to 67% of the total P deposition estimate (Duchesne et al., 2001; Talkner et al., 2010; Sohrt et al., 2019), is unlikely due to its peak in the autumn (Sohrt et al., 2019) and its correlation with foliar P content (Zhang et al., 2022), which exhibited signs of deficiency in the studied stands (Vos et al., 2023b). We, therefore, argue that the elevated P deposition during the growing season is more likely attributed to a notable contribution from sources such as pollen or local polluters, such as agricultural sources (Van Ek and Draaijers, 1994; Allen et al., 2010; Kopáček et al., 2011; Tipping et al., 2014). Given the absence of an effect of harvest intensity on P deposition, we argue that the interception of dry deposition of P is negligible, and the seasonal pattern of P is primarily driven by pollen (see also Rösel et al. (2012) and Doskey and Ugoagwu (1989)). The influence of pollen might not be limited to only P as tree pollen substantially increase K and NH_4 concentrations while reducing NO_3 concentrations with most pronounced effects in broadleaved trees (Verstraeten et al., 2023). However, while the pollen contribution may be the major source of the total deposition of P, they may be relatively small for K and N.

Furthermore, notable differences in Na and S deposition were present between seasons with higher deposition in the autumn in closed and thinned stands and higher deposition in the winter for the shelterwood and clearcut (Fig. 3.4). The seasonal difference in Na deposition is crucial to understand as Na has a central role in the determination of the canopy exchange (equation 4) and therefore in the total deposition of, amongst others, the base cations. Seasonal differences in Na deposition are therefore partly causing the seasonal patterns observed in the canopy exchange (Fig. 3.5B). For both Na and S, the higher throughfall in the autumn in the closed and thinned stands can be related to canopy leaching (Staelens et al., 2007; Thimonier et al., 2008; Adriaenssens et al., 2012a) while higher Na and S deposition during winter can be related to higher atmospheric concentrations (Van Ek and Draaijers, 1994; Adriaenssens et al., 2012a). Contrary to the suggestion of Thimonier et al. (2008) and Staelens et al. (2007) we did not find higher Na or S throughfall during spring (Fig. 3.5A) which contradicts the hypothesis that Na or S is

leached from the canopy during bud break and emerging leaves. More likely is that the higher deposition is related to the more efficient capture of Na and S in the control and thinned stands during autumn (Adriaenssens et al., 2012a). However, despite absence of evidence for canopy leaching of both Na and S, the use of those elements in the calculation of the canopy exchange can be questioned as both in throughfall and in total deposition, neither Ca, K and Mg show similar patterns consisting of higher deposition during the winter (Fig. 3.4, Fig. 3.5A and Fig. S3.4). However, the main factors causing the seasonal differences in the canopy leaching, which consist of elevated K and Mn leaching in autumn and winter, has been observed in earlier studies (Van Ek and Draaijers, 1994; Talkner et al., 2010) and is probably related to (drought induced) senescent leaves becoming more susceptible to ion leakage leading to substantial losses of both K and Mn (Schaefer and Reiners, 1990; Hagen-Thorn et al., 2006; Houle et al., 2016). Overall, this study does not find evidence against using Na as a tracer in the canopy exchange.

4.7 *Indications of canopy uptake of base cations*

Our results indicated a net canopy uptake of NH_4 , NO_3 , Ca, Mg (Scots pine only), Zn, Fe and Cu and a net canopy release of K and Mn. The net canopy release of K and Mn was lower than generally reported (Petty and Lindberg, 1990; Herrmann et al., 2006; Gandois et al., 2010b; Adriaenssens et al., 2012a) which might be caused by the low K and Mn concentrations in the foliage of our study trees (Talkner et al., 2010; Vos et al., 2023b). Canopy uptake of NH_4 and NO_3 with levels up to 90% of the throughfall deposition is widely accepted (Wilson and Tiley, 1998; Klopatek et al., 2006; Adriaenssens et al., 2011; Schwarz et al., 2014; Houle et al., 2015). Contrary, canopy uptake of Ca and Mg is less well-known while there is ample evidence of canopy leaching (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). However, canopy uptake of Ca and Mg was demonstrated before across a range of species (De Schrijver et al., 2004; Małek and Astel, 2008; Tan et al., 2018; Van Langenhove et al., 2020). We speculate that canopy uptake of Mg and Ca might be related to the high atmospheric deposition and to the lower foliar concentrations of these nutrients in our study sites (Vos et al., 2023b) and that the canopy for these nutrients turned from the often reported source role into a sink. Influence of foliar nutrient concentrations on canopy exchange was also proposed by Talkner et al. (2010) based on the work of Nordén (1991) who found increasing canopy leaching of base cations with increasing soil fertility. Our findings suggest that in highly acidified and nutrient-poor forest conditions, canopy adsorption of crucial nutrients such as Ca and Mg surpasses canopy leaching.

5 **Conclusions**

Sustainable management of forests on poor soils faces a challenge in maintaining base cation stocks (Ca, K, Mg) due to N and S deposition-induced soil acidification, causing base cation loss from the forest. The

inputs of N, S, Ca, K, and Mg in the non-harvested control plots of our study deviate significantly from local total deposition estimates. Despite a 29-140% higher NH_4 and SO_4 deposition, the net acidic input is lower than the national average because of higher base cation inputs (68-750% higher), indicating that the risk of soil acidification in forests is lower than expected based on nationwide models. This study urges, therefore, better calibration of deposition models, especially for countries facing challenges due to high N deposition loads.

We showed that the total annual nutrient deposition in stands of European beech, Douglas fir, and Scots pine consistently decreased with increasing tree harvest intensity and associated reduced canopy openness for all macro- and micronutrients, except for P which probably depended on pollen rather than deposition. Furthermore, the effect of harvest-induced canopy openness varied strongly between nutrients, with sharp decreases following harvest for nutrients that are mainly deposited dry (e.g., S, K). Across the study species, the highest deposition and highest acid inputs were observed in the relatively tall stands of Douglas fir and the lowest inputs in relative stem-dense, but short, stands of Scots pine. Moreover, our study showed that it is important to quantify deposition over the full annual cycle, to account for species differences in deposition across seasons. Overall, the species differences gradually reduced from closed forests to more open stands, and they were relatively small compared to the effects of harvest intensity.

Our results highlight the importance of considering the harvest intensity effects on forest structure and, to a lesser extent, tree species when calculating nutrient inputs via atmospheric deposition. Interestingly, harvest intensity may act as a management tool to reduce net acidic inputs into forests, but with current N deposition levels ongoing acidification and base cation loss cannot be avoided. Since deposition depends much on, for example, air quality, distance to the sea and local pollution sources, the reductions in total deposition with harvest intensity will vary geographically, emphasizing the need to consider location-specific factors for quantifying deposition-related inputs of nutrients in forests.

Acknowledgements

This research is part of the Nutrient Balance project and was funded by the Dutch Research Council (NWO, No. ALWGS.2017.004) and financially supported by Blom Ecology B.V. We acknowledge Henk van Roekel and many other members of the team for the monthly field sampling and the assistance with the sample handling. We thank our partners, National Forest Service, Union of private Forest Owner Groups, Het Loo Royal Estate, Staro nature and countryside, Borgman management consultants, National Park de Hoge Veluwe and Blom Ecology for financial support, permission to work in their forest or other provided services.

Supplementary information

Table S3.1 Overview of sampler placement in the control (CO), high-thinning (HT), shelterwood (SW) and clearcut (CC) for each forest site in relation to the canopy cover (%). In each treatment 7 samplers were placed either underneath the canopy (C) or exposed (E) in which the sampler placement reflected the canopy cover. The canopy cover of the clearcut was 0% as all trees were harvested. The deposition in the exposed sites of the shelterwood and clearcut was assumed to represent bulk deposition.

Site	Species	CO			HT			SW			CC	
		Cover (%)	C	E	Cover (%)	C	E	Cover (%)	C	E	C	E
1	Beech	85	6	1	75	5	2	26	2	5	0	7
2	Beech	91	6	1	69	5	2	28	2	5	0	7
3	Beech	95	6	1	72	5	2	24	2	5	0	7
4	Beech	82	6	1	64	4	3	23	2	5	0	7
5	Beech	93	7	0	79	6	1	21	2	5	0	7
1	Douglas fir	68	4	3	47	4	3	12	1	6	0	7
2	Douglas fir	82	6	1	66	5	2	11	1	6	0	7
3	Douglas fir	67	5	2	42	2	5	17	1	6	0	7
4	Douglas fir	81	6	1	67	5	2	9	1	6	0	7
5	Douglas fir	78	5	2	47	3	4	9	1	6	0	7
1	Scots pine	72	5	2	72	5	2	11	1	6	0	7
2	Scots pine	77	5	2	67	5	2	14	1	6	0	7
3	Scots pine	65	5	2	45	3	4	15	1	6	0	7
4	Scots pine	75	5	2	71	4	3	17	1	6	0	7
5	Scots pine	72	5	2	60	4	3	9	1	6	0	7

Table S3.2 Measured annual throughfall water fluxes (mm) $\pm$ standard error in the control (CO), high-thinning (HT) and shelterwood (SW) plots and bulk water fluxes in the shelterwood and clearcut (CC) plots compared to the interpolated precipitation of nearby weather stations (KNMI, 2021b). The stemflow fluxes are calculated. The standard error of throughfall in the shelterwood is missing as only one sampler was placed under the canopy (Table S3.1).

Site	Species	Throughfall			Bulk		Stemflow			KNMI
		CO	HT	SW	SW	CC	CO	HT	SW	
1	BE	507 $\pm$ 22	432 $\pm$ 29	580 $\pm$ 7.1	557 $\pm$ 28	638 $\pm$ 20	52	46	16	782
2	BE	474 $\pm$ 25	500 $\pm$ 33	584 $\pm$ 66	633 $\pm$ 18	643 $\pm$ 31	57	43	18	787
3	BE	423 $\pm$ 33	473 $\pm$ 25	417 $\pm$ 34	511 $\pm$ 16	605 $\pm$ 14	46	35	12	702
4	BE	392 $\pm$ 11	402 $\pm$ 27	520 $\pm$ 71	524 $\pm$ 23	525 $\pm$ 22	42	33	12	731
5	BE	376 $\pm$ 21	410 $\pm$ 49	521 $\pm$ 12	536 $\pm$ 27	506 $\pm$ 32	38	32	8.6	605
1	DG	408 $\pm$ 21	372 $\pm$ 28	469	520 $\pm$ 31	536 $\pm$ 30	5.3	3.7	0.94	751
2	DG	500 $\pm$ 24	520 $\pm$ 39	579	621 $\pm$ 26	701 $\pm$ 18	6.5	5.3	0.88	798
3	DG	435 $\pm$ 23	475 $\pm$ 30	579	614 $\pm$ 35	621 $\pm$ 28	4.9	3.1	1.2	735
4	DG	411 $\pm$ 34	441 $\pm$ 20	545	563 $\pm$ 19	580 $\pm$ 20	6.3	5.2	0.7	774
5	DG	330 $\pm$ 26	328 $\pm$ 41	478	355 $\pm$ 23	460 $\pm$ 14	4.9	3	0.57	629
1	SP	512 $\pm$ 36	536 $\pm$ 29	560	615 $\pm$ 14	608 $\pm$ 16	17	17	2.6	781
2	SP	491 $\pm$ 26	503 $\pm$ 53	537	634 $\pm$ 15	652 $\pm$ 28	18	16	3.3	797
3	SP	506 $\pm$ 22	535 $\pm$ 12	573	580 $\pm$ 23	595 $\pm$ 18	14	9.9	3.3	734
4	SP	473 $\pm$ 39	509 $\pm$ 24	455	678 $\pm$ 3.7	698 $\pm$ 4.2	17	17	4	778
5	SP	378 $\pm$ 30	428 $\pm$ 29	406	460 $\pm$ 27	482 $\pm$ 19	14	11	1.7	629

Table S3.3 The means and standard errors of the annual throughfall, stemflow and canopy exchange, all in ($\text{kg ha}^{-1} \text{yr}^{-1}$) of macro (NH_4 , NO_3 , Ca, K, S, Mg and P) and micronutrients (Zn, Mn, Fe and Cu) in high-thinning of beech, Douglas fir and Scots pine. Canopy exchange of S and P is assumed to be negligible (section 3.2.5.3).

	Species	NH_4	NO_3	Ca	K	S	Mg	P	Zn	Mn	Fe	Cu
Throughfall	Beech	13 ± 1.7	4.3 ± 0.45					$0.79 \pm$	$0.14 \pm$	$0.15 \pm$	$0.12 \pm$	$0.015 \pm$
				8.3 ± 1.1	22 ± 1	4.9 ± 0.58	2.2 ± 0.28	0.095	0.033	0.023	0.007	0.0027
	Douglas fir	21 ± 2.8	7.8 ± 1.2					$0.56 \pm$	$0.83 \pm$	$0.26 \pm$	$0.16 \pm$	$0.021 \pm$
Stemflow	Scots pine	17 ± 1.7	5.8 ± 0.61	8.8 ± 1.3	25 ± 1.3	11 ± 2.2	4.5 ± 0.52	0.042	0.64	0.043	0.025	0.0049
				6.8 ± 0.38	16 ± 1.7	5.8 ± 0.96	2.3 ± 0.18	0.069	0.3 ± 0.18	0.014	0.013	0.0025
	Beech	$0.96 \pm$	$0.32 \pm$	$0.72 \pm$		$0.41 \pm$	$0.18 \pm$	$0.059 \pm$	$0.01 \pm$	$0.011 \pm$	$0.01 \pm$	$5.4 \text{ e}^{-4} \pm$
Canopy exchange		0.10	0.033	0.077	1.7 ± 0.15	0.045	0.024	0.0088	0.0025	0.0023	0.001	3.0 e^{-5}
	Douglas fir	$0.20 \pm$	$0.072 \pm$	$0.087 \pm$	$0.26 \pm$	$0.11 \pm$	$0.048 \pm$	$0.005 \pm$	$0.0074 \pm$	$0.0023 \pm$	$0.001 \pm$	
	Scots pine	0.037	0.014	0.019	0.03	0.024	0.0091	0.00071	0.0062	0.00048	0.00037	0 ± 0
		$0.50 \pm$	$0.17 \pm$	$0.21 \pm$	$0.48 \pm$	$0.18 \pm$	$0.069 \pm$	$0.013 \pm$	$0.0083 \pm$	$0.0041 \pm$	$0.0039 \pm$	$2.9 \text{ e}^{-5} \pm$
		0.086	0.031	0.03	0.087	0.036	0.011	0.0035	0.0049	0.00038	0.00066	2.9 e^{-5}
	Beech	$-4.3 \pm$	$-0.33 \pm$	$-2.1 \pm$			$-0.15 \pm$		$-0.041 \pm$	$0.045 \pm$	$-0.1 \pm$	$-0.0079 \pm$
		0.18	0.035	0.84	15 ± 0.98		0.08		0.041	0.022	0.024	0.0019
	Douglas fir	$-1.9 \pm$	$-0.11 \pm$				$-0.17 \pm$		$-0.089 \pm$	$0.028 \pm$	$-0.19 \pm$	$-0.015 \pm$
	Scots pine	0.49	0.057	-4.6 ± 1.2	10 ± 0.88		0.21		0.067	0.022	0.049	0.0029
		$-1.8 \pm$	$-0.11 \pm$				$-0.51 \pm$		$-0.17 \pm$	$0.037 \pm$	$-0.15 \pm$	$-0.0084 \pm$
		0.63	0.04	-3.8 ± 0.8	9.7 ± 1.7		0.17		0.12	0.0087	0.027	0.0022

Table S3.4 The means and standard errors of the annual throughfall, stemflow and canopy exchange, all in ($\text{kg ha}^{-1} \text{yr}^{-1}$) of macro (NH_4 , NO_3 , Ca, K, S, Mg and P) and micronutrients (Zn, Mn, Fe and Cu) in the shelterwood of beech, Douglas fir and Scots pine. Canopy exchange of S and P is assumed to be negligible (section 3.2.5.3).

	Species	NH_4	NO_3	Ca	K	S	Mg	P	Zn	Mn	Fe	Cu
Throughfall	Beech	13 ± 2.7	4.6 ± 0.66	7.8 ± 0.77	9.4 ± 0.88	4.0 ± 0.17	1.7 ± 0.11		0.15 ± 0.28	0.09 ± 0.0061	0.13 ± 0.011	0.015 ± 0.0018
	Douglas fir	14 ± 1.2	5.4 ± 0.51	6.4 ± 0.58	9.4 ± 1.2	6.2 ± 0.79	2.8 ± 0.26	1.1 ± 0.53	0.53 ± 0.42	0.13 ± 0.018	0.13 ± 0.014	0.016 ± 0.0017
	Scots pine	14 ± 1.3	5.0 ± 0.52	6.5 ± 0.74	4.7 ± 0.37	4.5 ± 0.65	1.7 ± 0.18	0.53 ± 0.049	0.31 ± 0.19	0.079 ± 0.0096	0.14 ± 0.016	0.013 ± 0.0013
	Beech	0.26 ± 0.041	0.11 ± 0.023	0.17 ± 0.02	0.2 ± 0.028	0.09 ± 0.005	0.038 ± 0.0056	0.019 ± 0.0045	0.0021 ± 0.00042	0.0015 ± 0.00033	0.0025 ± 0.0004	0 ± 0
Stemflow	Douglas fir	0.024 ± 0.0060	0.00 ± 0.00	0.0046 ± 0.0014	0.012 ± 0.0037	0.0046 ± 0.00083	8.6 e ⁻⁴ ± 5.7 e ⁻⁵	3.7 e ⁻⁴ ± 1.5 e ⁻⁴	6.6 e ⁻⁴ ± 6.2 e ⁻⁴	0 ± 0	0 ± 0	0 ± 0
	Scots pine	0.076 ± 0.0098	0.03 ± 0.0071	0.032 ± 0.0076	0.02 ± 0.0053	0.02 ± 0.0069	0.0023 ± 0.00097	0.0024 ± 0.00049	0.0011 ± 0.00084	8.6 e ⁻⁵ ± 3.5 e ⁻⁵	5.7 e ⁻⁵ ± 3.5 e ⁻⁵	0 ± 0
	Beech	-0.52 ± 0.16	-0.057 ± 0.012	-0.45 ± 0.36	2.0 ± 0.64	-0.021 ± 0.016			-0.0052 ± 0.014	0.0052 ± 0.0033	-0.023 ± 0.0082	-0.0018 ± 0.0004
Canopy exchange	Douglas fir	0.68 ± 0.27	0.046 ± 0.02	-1.3 ± 0.28	-0.66 ± 0.55	-0.076 ± 0.081			-0.024 ± 0.0076	-0.0086 ± 0.0089	-0.049 ± 0.015	-0.0046 ± 0.0017
	Scots pine	0.11 ± 0.12	0.008 ± 0.0092	-0.49 ± 0.31	0.21 ± 0.17	-0.044 ± 0.018			-0.059 ± 0.049	-0.0037 ± 0.0028	-0.023 ± 0.0099	-0.0014 ± 0.0011

Table S3.5 The means and the standard errors of the total annual deposition ($\text{kg ha}^{-1} \text{yr}^{-1}$) of the macro- (NO_3 , NH_4 , S, P, Ca, K and Mg) and micronutrients (Mn, Cu, Fe and Zn) in the harvest intensities high-thinning (HT), shelterwood (SW) and clearcut (CC) and in the control (CO) for beech (BE), Douglas fir (DG) and Scots pine (SP). Total deposition of the unharvested control, high-thinning and shelterwood consist of the throughfall plus stemflow minus the canopy exchange (Table 3.1, Table S3.3, Table S3.4), while total deposition of the clearcut equals the bulk deposition. Differences between species within treatments are given in the column 'Sp', differences between treatments within species are denoted with small letters.

	Control	Sp	High-thinning	Sp	Shelterwood	Sp	Clearcut	Sp	Ratio
NO_3									
BE	6.8 ± 1.2^b	a	4.9 ± 0.46^b	a	4.8 ± 0.69^{ab}	a	4.3 ± 0.63^a	a	1.6
DG	8.8 ± 1.2^c	b	8.0 ± 1.2^{bc}	b	5.3 ± 0.52^b	a	4.1 ± 1.0^a	a	2.1
SP	6.0 ± 0.19^b	a	6.1 ± 0.66^b	ab	5.1 ± 0.53^{ab}	a	5.2 ± 0.76^a	a	1.2
NH_4									
BE	24 ± 3.2^b	a	18 ± 1.7^b	a	14 ± 2.9^a	a	14 ± 1.3^a	a	1.7
DG	27 ± 2.8^b	a	23 ± 2.7^b	b	13 ± 1.5^a	a	11 ± 1.2^a	a	2.5
SP	21 ± 1.5^b	a	20 ± 2.2^b	ab	14 ± 1.4^a	a	13 ± 1.2^a	a	1.6
S									
BE	6.0 ± 0.4^c	a	5.4 ± 0.62^{bc}	a	4.1 ± 0.16^{ab}	a	3.4 ± 0.11^a	a	1.8
DG	12 ± 3.2^c	b	11 ± 2.2^c	b	6.2 ± 0.79^b	b	3.9 ± 0.33^a	a	3.1
SP	6.3 ± 1.1^b	a	6.0 ± 1.0^b	a	4.5 ± 0.66^{ab}	a	3.6 ± 0.44^a	a	1.8
P									
BE	0.97 ± 0.089^a	b	0.85 ± 0.099^a	b	1.1 ± 0.29^a	a	0.80 ± 0.14^a	a	1.2
DG	0.57 ± 0.13^a	a	0.57 ± 0.042^a	ab	0.53 ± 0.047^a	a	0.46 ± 0.04^a	a	1.2
SP	0.51 ± 0.066^a	a	0.45 ± 0.072^a	a	0.53 ± 0.049^a	a	0.56 ± 0.12^a	a	0.91
Ca									
BE	13 ± 1.3^b	a	11 ± 0.83^{ab}	a	8.3 ± 0.52^a	a	6.3 ± 0.48^a	a	2.1
DG	14 ± 2.6^c	b	13 ± 2.2^{bc}	b	8.1 ± 0.93^b	a	4.7 ± 0.51^a	a	3.0
SP	11 ± 1.0^b	a	11 ± 0.98^b	ab	6.9 ± 0.90^a	a	5.4 ± 0.64^a	a	2.0
K									
BE	9.6 ± 1.1^c	b	8.7 ± 0.82^{bc}	b	6.2 ± 0.31^b	b	3.4 ± 0.33^a	b	2.8
DG	16 ± 2.8^b	c	15 ± 1.9^b	c	9.3 ± 1.4^b	c	3.9 ± 0.61^a	c	4.1
SP	6.7 ± 0.26^c	a	6.5 ± 0.26^c	a	4.1 ± 0.26^b	a	2.7 ± 0.35^a	a	2.5
Mg									
BE	2.7 ± 0.23^c	a	2.5 ± 0.24^{bc}	a	1.8 ± 0.097^{ab}	a	1.4 ± 0.10^a	a	1.9
DG	4.8 ± 0.80^c	b	4.7 ± 0.70^c	b	2.9 ± 0.34^b	b	1.6 ± 0.11^a	a	3.0
SP	2.9 ± 0.15^c	a	2.8 ± 0.34^c	a	1.8 ± 0.19^b	a	1.4 ± 0.21^a	a	2.1
Mn									
BE	0.12 ± 0.0097^c	a	0.11 ± 0.010^{bc}	a	0.081 ± 0.0031^{ab}	a	0.056 ± 0.0054^a	a	2.4
DG	0.24 ± 0.044^c	b	0.24 ± 0.035^{bc}	b	0.14 ± 0.017^b	b	0.073 ± 0.0083^a	a	3.4
SP	0.12 ± 0.012^c	a	0.12 ± 0.0068^c	a	0.076 ± 0.0082^b	a	0.05 ± 0.0023^a	a	2.4
Cu									
BE	0.025 ± 0.0034^b	b	0.024 ± 0.0035^b	b	0.017 ± 0.0017^{ab}	b	0.013 ± 0.0011^a	b	1.9
DG	0.036 ± 0.0075^b	c	0.036 ± 0.0067^b	c	0.021 ± 0.0034^b	c	0.011 ± 0.0013^a	a	3.3
SP	0.024 ± 0.0043^c	a	0.023 ± 0.0039^{bc}	a	0.015 ± 0.0025^{ab}	a	0.012 ± 0.0014^a	b	2.0
Fe									
BE	0.25 ± 0.016^b	a	0.23 ± 0.027^b	a	0.17 ± 0.012^b	a	0.13 ± 0.0069^a	a	1.9
DG	0.35 ± 0.069^c	a	0.36 ± 0.069^c	a	0.2 ± 0.030^b	a	0.13 ± 0.017^a	a	2.7
SP	0.29 ± 0.043^b	a	0.28 ± 0.039^b	a	0.18 ± 0.023^a	a	0.15 ± 0.016^a	a	1.9
Zn									
BE	0.20 ± 0.043^a	a	0.19 ± 0.057^a	a	0.13 ± 0.037^a	a	0.08 ± 0.012^a	a	2.5
DG	0.94 ± 0.73^b	a	0.92 ± 0.70^b	a	0.58 ± 0.44^{ab}	a	0.29 ± 0.22^a	a	3.2
SP	0.51 ± 0.34^b	a	0.47 ± 0.30^b	a	0.34 ± 0.24^{ab}	a	0.19 ± 0.13^a	a	2.7
Na									
BE	17 ± 1.7^b	a	16 ± 2.1^b	a	11 ± 0.78^a	a	8.9 ± 0.69^a	a	1.9
DG	31 ± 5.0^c	b	32 ± 5.3^c	b	20 ± 2.5^b	b	10 ± 1.1^a	a	3.1
SP	20 ± 1.8^b	a	20 ± 3.0^b	a	13 ± 1.5^a	a	10 ± 1.5^a	a	2.0

Table S3.6 Significance of seasonal variations of concentrations in bulk precipitation (in CC: clearcut) and throughfall (CO: control, HT: high-thinning and SW: Shelterwood), tested by One-Way Anova. Anova F-value and significance level (***) $p \leq 0.001$, ** $p \leq 0.01$, * $p \leq 0.05$, n.s. = not significant) are given (n = 60).

	Beech				Douglas fir				Scots pine			
	CO	HT	SW	CC	CO	HT	SW	CC	CO	HT	SW	CC
NH ₄	0.97 ^{n.s.}	69***	7.9**	34***	2.4 ^{n.s.}	1.5 ^{n.s.}	0.64 ^{n.s.}	7.8**	63***	5.1*	1.3 ^{n.s.}	12***
NO ₃	2.9 ^{n.s.}	2.4 ^{n.s.}	2.7 ^{n.s.}	1.1 ^{n.s.}	1.4 ^{n.s.}	0.64 ^{n.s.}	8.2**	3.5*	4.0*	0.11 ^{n.s.}	6.1**	6.4**
Ca	2.2 ^{n.s.}	22***	2.9 ^{n.s.}	1.5 ^{n.s.}	57***	9.9***	3.7*	8.4**	1.9 ^{n.s.}	1.1 ^{n.s.}	2.6 ^{n.s.}	7.3***
Mg	17***	2.8 ^{n.s.}	3.3 ^{n.s.}	3.2 ^{n.s.}	3.6*	5.3*	12***	1.9 ^{n.s.}	4.8*	2.1 ^{n.s.}	1.2 ^{n.s.}	1.4 ^{n.s.}
K	89***	17***	180***	17***	4.8*	19***	7.4**	17***	16***	15***	8.3**	36***
S	5.3*	0.83 ^{n.s.}	6.2**	1.7 ^{n.s.}	8.8**	3.1 ^{n.s.}	3.3 ^{n.s.}	4.1*	2.6 ^{n.s.}	3.3 ^{n.s.}	2.1 ^{n.s.}	9.4**
P	310***	26***	48***	30***	75***	63***	22***	57***	53***	470***	32***	16***
Mn	14***	10***	22***	6.3**	9.6**	11***	8.2**	3.9*	15***	9.5**	8.1**	8.8**
Cu	9.3**	2.9 ^{n.s.}	18***	4.1*	2.8 ^{n.s.}	9.9***	5.4*	2.3 ^{n.s.}	5.6*	3.3 ^{n.s.}	14***	9.0**
Fe	5.5*	3.2 ^{n.s.}	12***	5.5*	9.5**	5.4*	3.4 ^{n.s.}	5.9*	11***	8.4**	11***	11***
Zn	4.1*	9.8**	20***	1.1 ^{n.s.}	13***	2.1 ^{n.s.}	0.55 ^{n.s.}	1.3 ^{n.s.}	1.9 ^{n.s.}	2.5 ^{n.s.}	0.30 ^{n.s.}	1.7 ^{n.s.}
Na	78***	4.3*	24***	17***	4.3*	6.4**	59***	3.2 ^{n.s.}	57***	18***	26***	19***

Table S3.7 Total deposition of NH_x and NO_y , potential acidity (Pot.A) and SO_4 in $\text{eq ha}^{-1} \text{yr}^{-1}$ for the forest sites, as derived from the large-scale deposition maps of the Netherlands (RIVM, 2020; 2021), referred to as 'modelled' and measured at the sites, referred to as 'observed'. The SO_4 deposition is calculated based on the potential acid deposition minus the NH_x and NO_y deposition according to $\text{SO}_{4i} = (\text{Pot.A}_i - \text{NH}_{xi} - \text{NO}_{yi} - 280)/2$ (in $\text{eq ha}^{-1} \text{yr}^{-1}$). The modelled data for each forest site is the mean for the years 2020 and 2021, covering the period of the deposition measurements of this study, on a 1 km x 1 km grid cell where the study site was located .

Site	Species	NH_x		NO_y		Pot.A	SO_4	
		modelled	observed	modelled	observed	modelled	modelled	observed
1	BE	1179	1499	458	407	2146	229	337
2	BE	1240	2070	618	593	2384	246	312
3	BE	1255	2356	556	785	2361	270	449
4	BE	1644	1214	542	307	2796	330	412
5	BE	1372	1285	547	350	2547	347	374
1	DG	1278	1214	485	357	2278	235	368
2	DG	1245	2213	532	635	2310	253	443
3	DG	1286	1999	501	857	2322	255	461
4	DG	1764	2427	599	650	2995	352	1060
5	DG	1133	1856	460	600	2221	348	1372
1	SP	1278	1285	485	400	2278	235	274
2	SP	820	1642	378	471	1706	228	306
3	SP	1089	1214	477	400	2099	253	268
4	SP	1537	1713	539	428	2696	340	524
5	SP	1133	1713	460	421	2221	348	599

Table S3.8 Total modelled deposition of Ca, K and Mg in mol ha⁻¹ yr⁻¹ for the forest sites as derived from the deposition maps published in Van Jaarsveld et al. (2010) and measured at the sites, referred to as 'observed'. The modelled data for each forest site are mean values for the period 2000 -2005 and values for each forest sites are based on an overlay with the 5 km x 5 km grid cell where the study site was located.

Site	Species	Ca		K		Mg	
		modelled	observed	modelled	observed	modelled	observed
1	BE	130	374	52	248	78	119
2	BE	131	274	47	212	71	99
3	BE	142	399	46	358	63	136
4	BE	128	299	52	215	85	128
5	BE	111	217	41	202	45	82
1	DG	130	207	52	332	78	144
2	DG	131	217	47	210	71	144
3	DG	133	349	45	332	68	193
4	DG	135	574	53	512	98	321
5	DG	134	399	42	614	40	181
1	SP	130	242	52	184	78	115
2	SP	131	200	47	161	71	119
3	SP	133	299	45	189	68	111
4	SP	135	299	53	153	98	140
5	SP	134	324	42	171	40	103

Supplementary figures

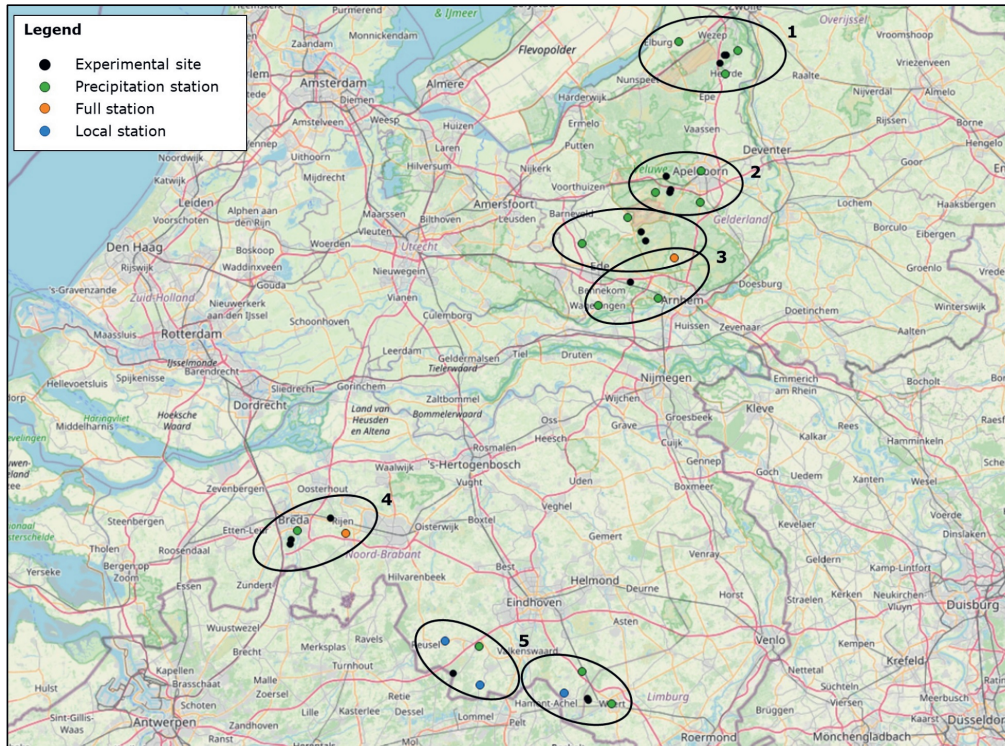


Figure S3.1 Location of the sites (1-5) in which the experimental sites are shown with black dots. The daily precipitation of each site is interpolated based on nearby weather stations which are grouped by the ellipses. Data from the KNMI precipitation stations is collected from (KNMI, 2021a), data from the KNMI full stations is collected from (KNMI, 2021b) and data from the local stations is collected from either weather station Bladel¹, weather station Luyksgestel² or weather station Budel³.

¹ <https://www.hetweeractueel.nl/weer/bladel/actueel/>

² <https://www.weerstationluyksgestel.nl/weather28/index.php>

³ <https://www.hetweeractueel.nl/weer/budel/actueel/>

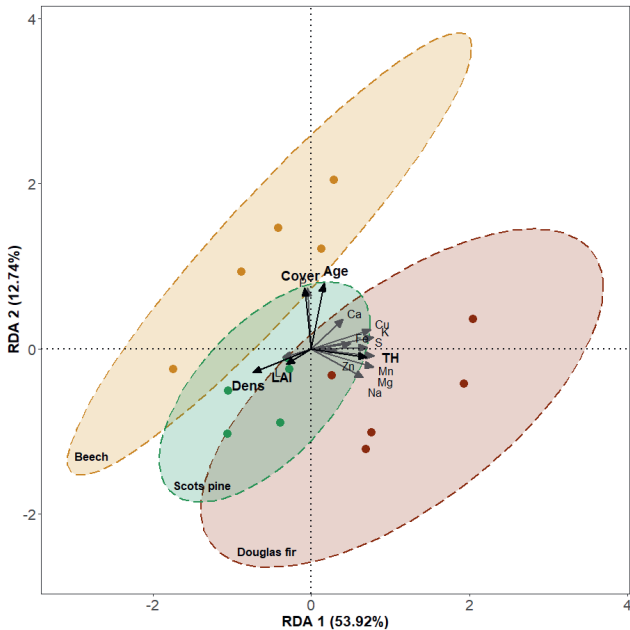


Figure S3.2 Total annual deposition of macro- (NH₄, NO₃, Ca, K and Mg) and micronutrients (Mn, Cu, Fe and Zn) in unharvested stands grouped by the centroids of the species (RDA biplot). Canopy exchange of macro- and micronutrients and precipitation interception is represented by the grey arrows, the effects of species tree density (Dens), leaf area index (LAI), canopy cover (Cover), tree height (TH) and stand age (Age) by black arrows. T-distribution polygons are shown for the four seasons, dots are colored by harvest intensity. The length of arrows denotes the variation explained.

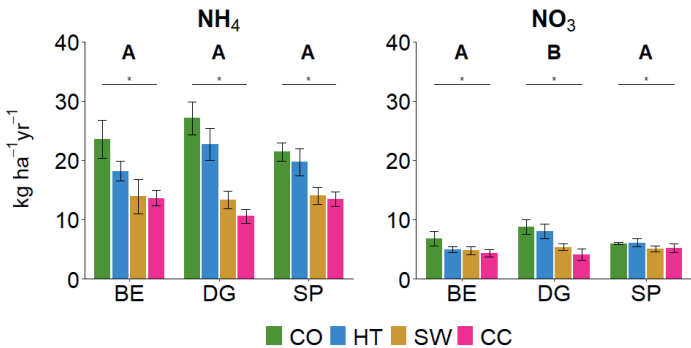


Figure S3.3 Total annual nutrient deposition (kg ha⁻¹ yr⁻¹) in the harvest intensities high-thinning (HT), shelterwood (SW) and clearcut (CC) and in the control (CO) for beech, Douglas fir and Scots pine. Different capital letters denote significant differences between species, asterisks indicate significant differences amongst different harvest intensities (nested-ANOVA, n=5, P<0.05). Differences between harvest intensities are given in Table S3.5.



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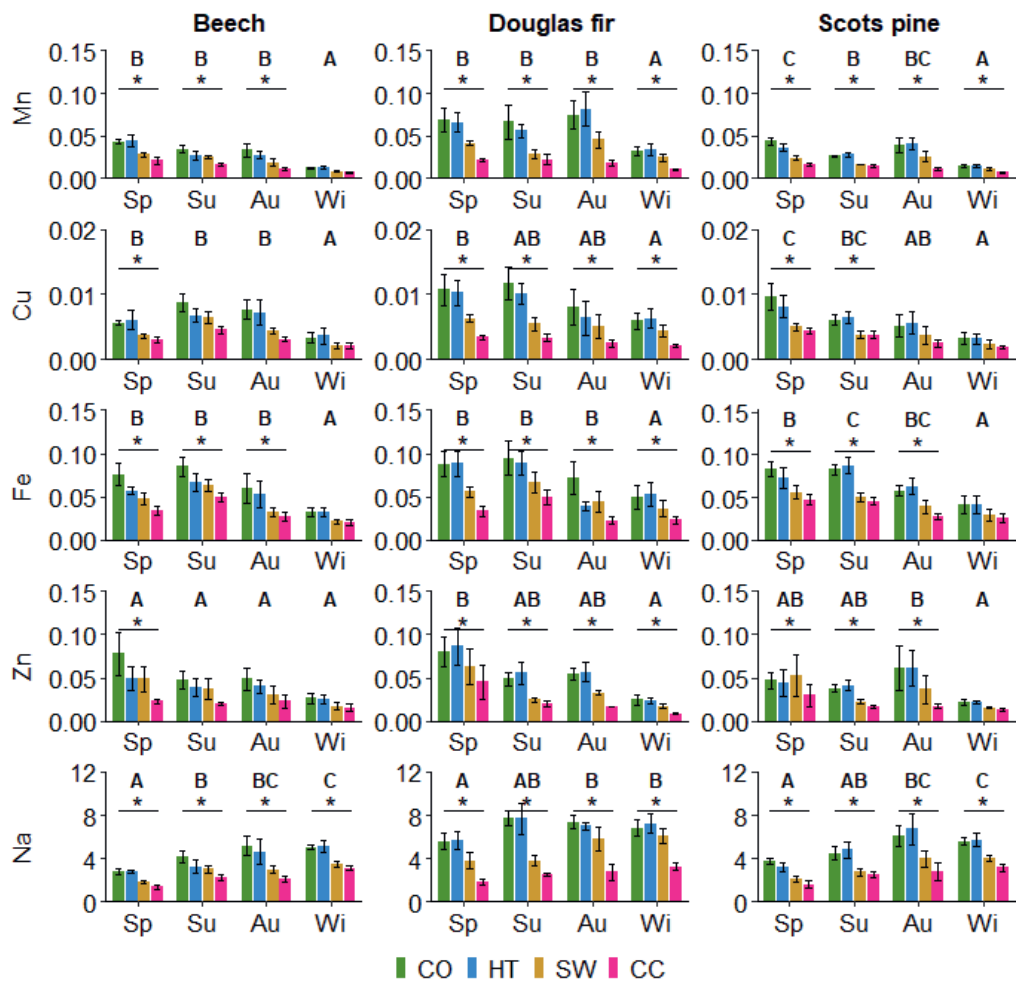


Figure S3.4 Total nutrient deposition (kg ha⁻¹) in the spring (Sp), summer (Su), autumn (Au) and winter (Wi) in the harvest intensities high-thinning (HT), shelterwood (SW) and clearcut (CC) and in the control (CO) for beech, Douglas fir and Scots pine. Different capital letters denote significant differences between seasons (nested-ANOVA, n=5, P<0.05). Differences between seasons within harvest intensities are in Table S3.6.



Chapter 4

Aboveground carbon and nutrient distributions are hardly associated with canopy position for trees in temperate forests on poor and acidified sandy soils

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*Forest ecology and management (2023),
Volume 529, 1 February 2023*

Abstract

High demands on forest for carbon storage and provision of timber and biofuel require precise and reliable estimates of the biomass, carbon and nutrient stocks in different tree compartments. Whether the fraction of biomass distributed in aboveground tree compartments and the carbon and nutrient concentrations varies systematically across trees in different canopy positions remains unclear despite its importance for understanding forest ecology. Here, we compared the distribution of biomass, carbon and nutrients from underlying carbon and nutrient concentrations between different aboveground tree compartments for 15 mature trees of European beech (*Fagus sylvatica*), Douglas fir (*Pseudotsuga menziesii*) and Scots pine (*Pinus sylvestris*) with dominant, intermediate and suppressed canopy position.

We show that carbon concentrations were relatively constant across tree compartments while nutrient concentrations increased from stem, bark, branches towards needles. Canopy position had only minor effects on carbon and nutrient concentrations and on the distribution of biomass, carbon and nutrient between aboveground tree components. Nutrient concentrations and stoichiometric results confirm that the forests were affected by high N deposition and low availability of P and base cations.

Our results imply that predictions from allometric scaling theory better apply to aboveground tree components than from functional equilibrium theory. Models aiming for estimating tree and forest biomass and carbon and nutrient stocks can apply equal biomass, carbon and nutrient stocks for trees independent of canopy position as a valid assumption but testing this assumption for a broader range of species and site conditions remains recommended.

1 Introduction

Forests cover approximately 31% of the global land area and provide many ecosystem services including carbon sequestration, nutrient and water cycling, and the production of timber and biomass (UNEP, 2020). At global scale, forests sequester approximately 30% of the anthropogenic CO₂ emissions (Pan et al., 2011; Quéré et al., 2018) and thus act as a net carbon sink. However, a growing demand for commodities (timber, biofuel and fiber) have intensified forest harvesting (Mantau et al., 2010; Nabuurs, 2015), with uncertain implications for future carbon sequestration and nutrient cycling by forests.

Forest models are used to estimate the stocks and fluxes of carbon (Liski et al., 2006; Akselsson et al., 2007a; Franklin et al., 2012) and nutrients (Akselsson et al., 2007a; Vangansbeke et al., 2015; Pare and Thiffault, 2016; de Vries et al., 2021). Good estimates of carbon and nutrient stocks in forests require data on forest biomass, and the distribution of carbon and nutrients over the different tree compartments (Poorter and Sack, 2012; Wertz et al., 2020). Such information is still poorly quantified for large adult forest trees (Schippers et al., 2015), particularly for tree compartments other than foliage. Another caveat in such estimates is the role of canopy position of trees, ranging from fully exposed, large, dominant trees in the upper canopy to shaded, small, suppressed trees in the understory. Canopy position differences involve large differences in access to light, tree metabolism, carbon gain and transpiration across trees in the same forest, and potentially inflates the uncertainty in estimating nutrient and carbon stocks in forest models (Franklin et al., 2012) but this has hardly been quantified.

Two theories dominating the literature come with different predictions for resource distribution within plants. The functional equilibrium theory (Brouwer, 1962), also called the optimal partitioning theory (McCarthy and Enquist, 2007), predicts that the resource allocation in trees is driven by priority and demand, whereby resources are allocated to the organ that acquires the most limiting resource. Contrastingly, the theory of allometric scaling, predicts that resource allocation is driven by scaling relationships between organs that vary with individual size, and not with the environment (Shinozaki et al., 1964; Enquist and Niklas, 2002; McCarthy and Enquist, 2007). In most forest models, the distribution of biomass within trees is predicted based on allometric scaling from DBH or tree height (Bartelink, 1997; Li and Zhao, 2013; Pretzsch et al., 2014). Models based on the functional equilibrium theory are hardly used, although sometimes stem density measures, pointing towards effects of competition on biomass allocation, are included in the models (Xue et al., 2012; Schepaschenko et al., 2018). In this study the predictions of both theories will be tested for the aboveground biomass distribution in trees differing in canopy positions, creating a framework for including the effects of competition on resource allocation.

Trees with a dominant canopy position grow faster (D'Amato and Puettmann, 2004; Reid et al., 2004; Castagneri et al., 2008) and may increase the share of branches over the stem (Krejza et al., 2017; Wertz et al., 2020) in response to high light levels. Suppressed trees growing at lower light availability may

increase height growth over radial growth (Naidu et al., 1998; Zhou et al., 2018; Yang et al., 2019), and invest more in stem wood (Wertz et al., 2020) at the costs of investment in the crown (Naidu et al., 1998; Reid et al., 2004; Vanninen and Mäkelä, 2005; Sterck and Schieving, 2007; Krejza et al., 2017). Such responses nevertheless differ between species differing in shade tolerance (Van de Peer et al., 2017; del Río et al., 2019), or environment (Lines et al., 2012). Despite the differences in biomass allocation between dominant and suppressed trees, there is no consensus on the magnitude of this effect. Trees can adjust their allocation to maintain remarkable constant biomass distributions between different components (e.g. crown versus stem) to maintain major functions in very different environments (Anfodillo et al., 2016; Petit et al., 2018). Yet, whether such relative biomass distributions also hold for trees in different canopy positions amongst different environments remains, as far as we know, poorly quantified.

Canopy position may also affect tree carbon concentrations, but in most forest, carbon models, tree carbon concentrations are assumed to be constant and approximately 50% of the biomass (Litton et al., 2007; Zhang et al., 2009; Thomas and Martin, 2012). Such strong assumptions potentially add uncertainty in carbon stock estimates since carbon concentrations differ across compartments and organs (Bert and Danjon, 2006; Zhang et al., 2009; Thomas and Martin, 2012). Some studies showed that intraspecific competition alters carbon concentrations (Peri et al., 2010), but other imply that this is not the case (Zhang et al., 2009). It has been suggested that trees invest more in structural carbohydrates and lignin compounds under less favourable conditions, but more in lipid and protein compounds under favourable conditions (Lambers et al., 2008). In this study, we will test whether carbon concentrations can indeed be assumed constant across tree compartments for trees in different canopy positions.

For nutrient concentrations, the biogeochemical niche hypothesis (Peñuelas et al., 2008; Penuelas et al., 2010) predicts that species maintain a constant nutrient stoichiometry in their compartments. Nevertheless, species retain a certain degree of plasticity in nutrient concentration and allocation to a change in competitive conditions (Peñuelas et al., 2008; Sardans et al., 2015), both belowground (Peri et al., 2006; Yang et al., 2009) and aboveground (Reid et al., 2004). Dominant, rapidly growing trees may maintain higher metabolic rates (D'Amato and Puettmann, 2004; Reid et al., 2004; Castagneri et al., 2008), and acquire water and nutrients more rapidly than suppressed trees (Martin et al., 1997; Granier et al., 2000; Aranda et al., 2012). This is in line with higher nutrient concentrations observed for more dominant relative to suppressed trees (Peri et al., 2006; Peri et al., 2010; Wu et al., 2020b) but, ambiguously, reverse patterns have also been reported (Reid et al., 2004; Peri et al., 2006; Couto-Vazquez and Gonzalez-Prieto, 2010). An additional complication is that higher nutrient concentrations in dominant trees were reported for N, P (Peri et al., 2006; Wu et al., 2020b), K, Mg, and S (Peri et al., 2006), while Ca concentrations were found to be higher in suppressed trees (Peri et al., 2006). Furthermore, nutrient concentrations depend also on the compartment, with higher concentrations of N (Reid et al., 2004; Couto-

Vazquez and Gonzalez-Prieto, 2010) and P (Reid et al., 2004) reported for foliage of suppressed trees relative to dominant trees. Data regarding the effects of canopy position on nutrient concentrations in other tree compartments (e.g., stem wood, stem bark, or branches of different size) are however very limited and, to our best knowledge, almost absent in combination with real measures of total biomasses of these compartments within mature trees. This means that possible effects of canopy position on the biomass, carbon and nutrient stocks within trees cannot yet be generalized, which is required for reducing the uncertainty of forest model predictions on carbon and nutrient stocks in forests.

In this study, we aim at assessing the impact of canopy position on the distributions of tree biomass, carbon and nutrients amongst different tree compartments. We therefore quantified the biomass and the carbon and nutrient concentrations within tree compartments in dominant, intermediate and suppressed trees of European beech (*Fagus sylvatica*), Douglas fir (*Pseudotsuga menziesii*), and Scots pine (*Pinus sylvestris* L.) growing in forests on poor sandy soils in the Netherlands. The aboveground compartments that were considered include needles, small branches, coarse branches and stem bark, stem sapwood and stem heartwood. The nutrients considered involve macronutrients (N, P, K, Ca, S, Mg) and micronutrients (Cu, Fe, Mn, Zn). We specifically addressed the following questions:

- (1) Is carbon concentration indeed rather constant across tree compartments?
- (2) Do nutrient concentrations within trees reflect the low cation and phosphorous availability of acidified sandy soils and the relatively levels of nitrogen deposition over the past decades?
- (3) What is the distribution of biomass, carbon and nutrients among different tree compartments?
- (4) What is the effect of canopy position on total amounts and the distribution of biomass, carbon and nutrient stocks among different tree compartments?

2 Method

2.1 Study area

In 2018, we selected one 1-ha forest plot dominated by European beech (*Fagus sylvatica*), one by Douglas fir (*Pseudotsuga menziesii*) and one by Scots pine (*Pinus sylvestris* L.) in five study areas (Fig. 4.1), resulting in 15 forest plots in total. These three species represent important timber species in the Netherlands and other parts of Western and Central Europe. The selected forest plots were located on acidic sandy soils classified as Albic or Entic Podzols or Dystric Cambisols (WRB, 2015)(Table S4.1). These soils are characterized by high nitrogen stocks, ranging between 42-54 kg ha⁻¹ in the top 30 cm of the mineral soil and between 1000-1100 kg ha⁻¹ in the organic layers. Dissolved organic carbon in the top 30 cm of the mineral soil ranged between 1000-1100 kg ha⁻¹ (Vos et al., 2023a). The plots were characterized by a similar temperate, maritime climate with an interpolated 30-year average annual rainfall and temperature of 850 mm and 10.6°C respectively (KNMI, 2021b). The forest in the plots consisted of

relatively homogeneous, even-aged, single-tree species planted forests between 50–120-year-old. All dominant species within a stand took up more than 80% of the total crown cover and basal area. All plots had previously been managed following common silvicultural methods in the Netherlands. Thinning regimes started with thinning from below (removal of suppressed trees) and, in the last three decades, all stands were treated using high-thinning (removal of trees directly competing with future crop trees). All study sites are subject to moderately high to high levels of N-deposition with annual atmospheric input ranging between 1200 to 2150 mol N/ha (RIVM, 2020), resulting in accelerated soil acidification, reducing nutrient availability (De Vries et al., 1995a; de Vries et al., 2014a).

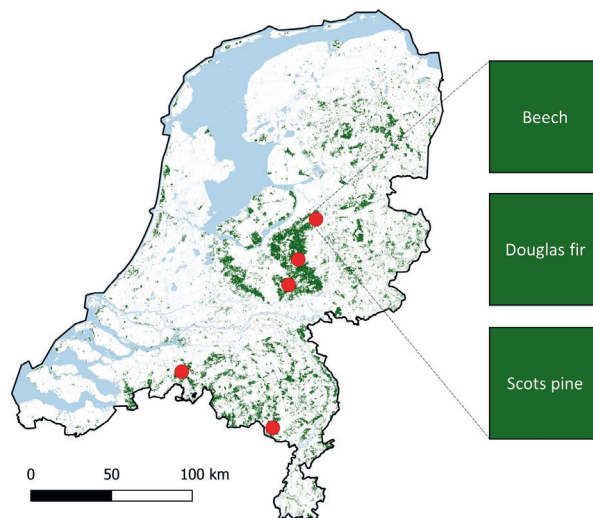


Figure 4.1 Locations of sites selected for biomass, carbon and nutrient measurements in this study. The numbers denote the study sites, shapes the different plots (overlapping plots not shown). The nationwide forest cover (in total 10% of the land area of the Netherlands) is shown in green (PDOK, 2015).

In October 2018, forest stand properties were measured in each of the 15 plots before harvest of the trees in February–March 2019. The stem diameters at breast height (DBH) were measured for all trees in the 1-ha plot (Table 4.1). For 16 sampling points, dominant tree height was measured for 5 dominant trees using a digital measuring device (Nikon Forestry Pro laser, Japan). Each sampling point was in a 4 by 4 grid across the plot with 20 m distance between the points. Forest biomass stock was calculated based on the dry weight of the trees and the DBH of all trees using plot-specific biomass expansion factors (Vos et al., 2023a). We cored 20 bulked soil samples from the mineral layer (0–30 cm depth) separately via systematic sampling, with equal distances between sampling points covering the whole plot. Samples were dried at 40 °C to a constant weight and sieved (< 2 mm). Unbuffered cation exchange capacity (CEC) was

measured according to Varian Vista with ICP-AES (Thermo-Scientific iCAP 6500 DUO, USA) (Houba, 1997). The unbuffered cation exchange capacity in the mineral soil for all sites was dominated by exchangeable aluminum. Concentrations of exchangeable base cations (Ca, K and Mg) were below detection limit indicating almost an absence of any base saturation (Table 4.1).

Table 4.1 Overview of stand and soil characteristics of the selected European beech, Douglas fir and Scots pine forests of this study.

Site	Species	Age (yr.)	Density (tree ha ⁻¹)	Dg (cm)	BA (m ² ha ⁻¹)	Stock (t ha ⁻¹)	Hdom (m)	Soil pH	Al-CEC (%)	Soil BS (%)
1	Beech	94	248	34	22.8	195	22.1	4.3	91	2.1
2	Beech	101	140	47	24.8	194	25.6	4.3	97	0
3	Beech	82	197	37	21.6	194	24.5	4.2	94	1.3
4	Beech	98	219	34	21.2	201	24.3	4.5	83	10
5	Beech	46	840	17	21.5	129	19.4	4.1	77	1.3
1	Douglas fir	74	119	54	28.0	203	41.0	4.2	68	3.3
2	Douglas fir	59	170	50	32.8	233	36.5	4.0	75	2.0
3	Douglas fir	60	138	52	29.7	216	37.3	4.2	96	3.8
4	Douglas fir	66	127	52	27.0	197	36.0	4.0	74	4.0
5	Douglas fir	60	239	44	37.1	247	28.3	4.2	88	7.1
1	Scots pine	55	406	25	21.0	83	18.8	4.3	79	1.3
2	Scots pine	48	425	24	19.8	87	18.3	4.1	88	3.2
3	Scots pine	47	835	17	20.3	77	18.7	4.4	88	5.0
4	Scots pine	62	400	26	22.4	97	20.9	4.0	69	4.3
5	Scots pine	73	466	27	26.6	118	15.8	4.2	91	5.0

Notes: Density includes all trees with DBH > 10 cm; Dg is the average arithmetic DBH; BA is the forest basal area; Stock is the biomass stock per hectare based on plot specific biomass expansion factors (Vos et al., 2023a); Hdom the dominant height based on height of 16 dominant trees per hectare; Soil pH is pH H₂O of mineral soil 0-30 cm depth; Al-CEC is the percentage of the unbuffered CEC occupied by Al; Soil BS is the base saturation of the mineral soil based on the percentage of the unbuffered CEC occupied by the sum of Ca, K, Mg and Na (all under detection limit).

2.2 Biomass, carbon and nutrient measurements

To estimate the aboveground biomass, and carbon and nutrient stocks for trees differing in canopy position, one dominant, one intermediate and one suppressed tree was selected in each plot (Table 4.2). Those trees were picked from three equally-spaced DBH classes, covering the entire observed DBH range in each plot (Fig. S4.1). The total of three trees per plot in five study areas resulted in 15 sampled trees per species. Tree compartments included were needles (no leaves for beech, since trees were harvested in winter, February/March 2019), small branches (≤ 2 cm diameter) and coarse branches (2-10 cm diameter), and for the stem (> 10 cm diameter), bark and sapwood and, if present, heartwood. The stem was defined

as the main axis from the stem base upwards until the point where the stem diameter became smaller than 10 cm. The crown was defined as branches < 10 cm diameter, including needles. To estimate the total stem and branch volume of the tree, the total tree height, stem length and the branch base diameters and lengths for all living branches (> 1 cm diameter) were measured. Whole tree fresh weight and stem fresh weight were measured directly after harvest in the field, using a tractive scale (Allscales Europe, cap. 3000 kg $\pm$ 2 kg) attached to an excavator (Fig. 4.2). Difference between whole tree fresh weight and stem fresh weight resulted in the crown fresh weight. To estimate the biomass of needles, small branches and coarse branches 4 representative branches (not severely damaged due to the felling) per tree were selected covering the observed range of measured branch diameters. For each of those branches, the diameter, the total length and the length of the coarse branch were measured. Total fresh weight and fresh weight of the coarse branch were measured by weighing the whole branch and the coarse branch with a tractive scale (crane scale SF-918, cap. 150 kg $\pm$ 0.1kg, Fig. 4.2). All small branches per sampled branch were collected in sealed plastic bags, labelled and stored at 4°C until measurement of: 1) total fresh weight; 2) dry weight of branch wood; 3) dry weight of needles (except for beech); and 4) dry weight of cones (except for beech), with dry weight defined as the constant weight after drying samples at 70 °C. Fresh and dry weights were also determined separately for a subsample of needles to calculate moisture loss allowing the calculation of fresh needle mass.

Table 4.2 Mean $\pm$ standard error (n=5) of DBH (cm), tree height (m) and stem length (m) for the sampled dominant, intermediate and suppressed trees per species. Tree height is defined as the vertical distance between stem base to highest crown part, and stem length as the distance from stem base to the point along the stem with a stem diameter <10 cm. This latter point was a cutoff point, where we distinguished between stem and crown.

Species	Canopy position	DBH (cm)	Tree height (m)	Stem length (m)
Beech	Dominant	48.4 $\pm$ 5.5	23.9 $\pm$ 1.2	18.6 $\pm$ 1.4
	Intermediate	34.5 $\pm$ 4.9	22.6 $\pm$ 1.6	16.3 $\pm$ 2.0
	Suppressed	26.6 $\pm$ 4.2	20.8 $\pm$ 1.1	13.7 $\pm$ 2.4
Douglas fir	Dominant	62.8 $\pm$ 2.4	34.7 $\pm$ 1.9	29.9 $\pm$ 1.9
	Intermediate	48.3 $\pm$ 2.0	33.8 $\pm$ 2.9	29.1 $\pm$ 2.6
	Suppressed	34.7 $\pm$ 3.4	27.0 $\pm$ 3.0	22.0 $\pm$ 0.9
Scots pine	Dominant	33.7 $\pm$ 2.6	20.2 $\pm$ 1.2	15.6 $\pm$ 0.6
	Intermediate	25.2 $\pm$ 2.4	18.0 $\pm$ 0.5	13.4 $\pm$ 0.7
	Suppressed	16.6 $\pm$ 1.1	14.9 $\pm$ 0.7	8.2 $\pm$ 0.9



Figure 4.2 Pictures of measurements in the field (top) and in the laboratory (bottom). Fieldwork included measurement of the fresh weight of the entire tree (top left), fresh weight of the stem (top middle left), fresh weight of branches (top middle right) and fresh weight of the coarse branch (top left). Laboratory work included separation of the stem disk into bark, sapwood and, if present, heartwood (bottom left), extracting slices for determination of volume (bottom middle) and separation of the dried branch material into needles (bottom middle right) and small branches (bottom right).

To estimate the biomass of the stem bark, stem sapwood and stem heartwood and the coarse branches, various disk samples were taken from each stem: close to the stem base, at the cut-off point of 10 cm stem diameter, and at 1/4th, 2/4th and 3/4th of the stem length, and for the branches at 2 cm stem diameter cut-off point, and halfway the 10-cm a 2-cm cut-off point. All disks were labelled, stored in plastic bags at 4°C to prevent drying until further processing. Prior to destructive subsampling, the diameter, thickness and perimeter of the disks were measured. Subsequently, the whole disk was separated into bark, sapwood and, if present, heartwood (Fig. 4.2). Measurements conducted on separated compartments included measurements of 1) diameter; 2) perimeter; 3) fresh weight; 4) fresh weight density; 5) dry weight; and 6) dry weight density.

Samples for chemical analysis were based on a mass weighted sample along the tree compartments (Supplement 1, Formulas S1-S11). The material was ground in a mill containing 1.5 mm stainless steel screen for nutrient analysis and analysed for N and carbon content by using a CN-analyzer (LECO TruSpec CHN, USA). Concentrations of P, S, K, Ca, Mg, Mn, Cu, Fe and Zn were analysed following 0.43M HNO₃ extraction and by using an ICP-AES (Thermo-Scientific iCAP 6500 DUO, USA) (Houba, 1997). Details on the method are in Supplement 1.

2.3 Biomass quantification models

The biomass of the stem bark, stem sapwood and stem heartwood were calculated based on the volume and density of the tissues along different segments of the stem. Stem length was divided into four segments, and for each segment the volume was calculated based on the means of the stem disks at the top and bottom of the segment, treating the stem segment as a truncated cone. Accuracy of the calculation was evaluated based on the calculated and measured fresh aboveground biomass (Fig. S4.5 - S4.6). Highest accuracy for volume calculations of the stem wood was achieved by calculating the radius of the stem wood and heartwood as a function of the perimeter of the disk, instead of calculations based on measured disk diameter. Bark volume was calculated as a function of the dry weight, density and thickness of the bark per disk to correct for the heterogenous nature of the bark. Volume corrections for bark, sapwood and heartwood were executed for beech and Scots pine, based on the number of ramifications within the stem (formula S18). The specific density of the bark, sapwood and heartwood per stem disk was used to calculate the fresh and dry biomass per compartment. A stepwise overview of the calculations to derive stem volume, stem dry weight and stem fresh weight and the validation of the calculations are in Supplement 1.

The dry and fresh biomass of needles, small branches and coarse branches was estimated for the entire crown based on the four sampled branches per crown. To derive whole crown estimates, linear mixed effect models were fitted per species using the restricted maximum likelihood method and nested within the tree following the procedure as described by (Zuur et al., 2009). Branch models were formulated as a compromise between the best possible estimates and the simplicity of the model. Therefore, models were based on the volume of branches as this proved to be a better estimate than length and diameter separately. Overview of the fitted regression models is in Table S4.2 – S4.3.

2.4 Statistical analysis

Biomass, carbon and nutrient distributions between the different tree compartments were expressed as the mass fraction (%) of the total aboveground biomass, carbon stock and nutrient stock, respectively. The aboveground biomass excluded the foliage mass to harmonize the biomass fractions of the coniferous trees (Douglas fir and Scots pine) with the deciduous European beech. Differences in the biomass, carbon and nutrient distributions and carbon and nutrient concentrations between canopy positions and tree compartments were analysed by using a two-factor nested ANOVA. Nesting was carried out to correct for the dependency between compartments within the tree and site. The used statistical was:

$$Y_{itk} = \mu + \alpha_i + B_{t(i)} + \varepsilon_{iz}$$

Where Y is the biomass distribution, carbon or nutrient concentration or mass distribution per tree (k), α is the fixed effect of canopy position, β is the fixed effect of the tree compartment (t) and ϵ is the residual error within tree (i) and study site (z). All data were checked for normality and homogeneity of variances. The constant variant function `varIdent` (nlme package) was used when variances were heterogenous. All statistical analyses were performed with R version 3.6.1. based on the nlme package followed by pairwise comparison with Tukey's *posthoc* test (Emmeans package). To reduce the number of type-I error results in the multiple test comparison, statistical tests were considered significant at $P < 0.01$.

3 Results

3.1 Biomass distribution, carbon concentration and mass distribution.

The total aboveground dry biomass per tree ranged between 108-3117 kg for beech, 255-2913 kg for Douglas fir and 54-698 kg for Scots pine. Average mass-based total tree carbon concentrations were 487 g kg⁻¹ biomass for beech, 503 g kg⁻¹ for Douglas fir and 507 g kg⁻¹ for Scots pine. Trees from different canopy positions varied substantially in total biomass; relative to suppressed trees, intermediate and dominant trees had 59-204% and 255-441% more biomass, respectively.

Biomass and carbon mass fractions differed strongly between tree compartments, with highest (up to 75%) values for stem wood, followed by coarse branches, small branches and bark for beech and Scots pine, and by bark, coarse branches and small branches for Douglas fir (Fig. 4.3). Carbon concentrations varied little between tree compartments. However, carbon concentrations were slightly lower in stem wood and higher in small branches for beech, and lower in stem wood and coarse branches but higher in stem bark for both conifers (Fig. 4.3).

Trees from different canopy positions were remarkably similar in biomass distribution, carbon concentration, and carbon distribution, despite few significant trends for Scots pine (Table S4.4). Canopy position influenced biomass distribution and carbon mass distribution in the sapwood of Scots pine (ANOVA test, $P < 0.01$), but the differences between tree components were not significant probably because the canopy position effects were relatively inferior (Fig. 4.3). Overall, suppressed Scots pine trees had a lower, but insignificant, biomass (and carbon) fraction in the sapwood, which was mainly compensated by higher, but insignificant, mass fractions in coarse branches. On tissue level canopy position caused a higher needle carbon concentration in intermediate trees compared to suppressed trees, but effect sizes were small (Fig. 4.3).

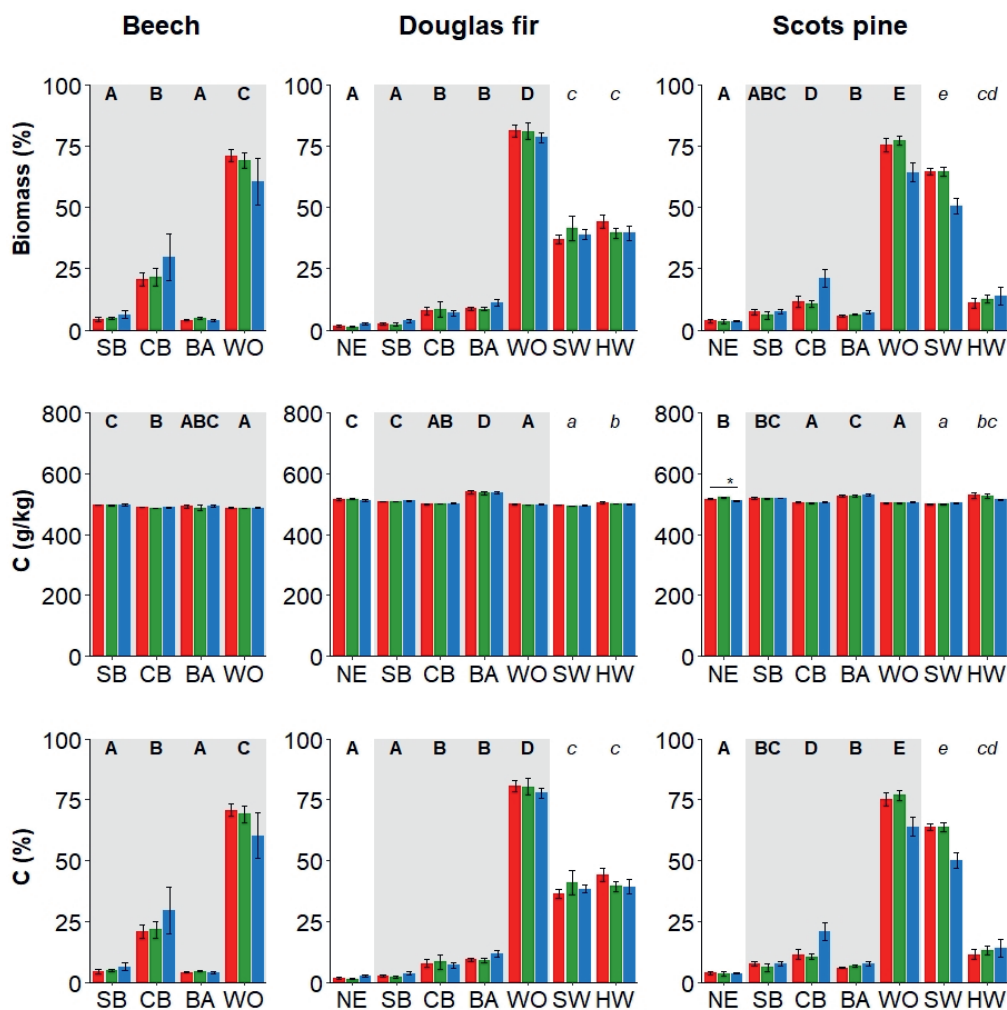


Figure 4.3 Biomass distribution (%), carbon (C) concentrations (g kg^{-1}) and carbon mass distribution (%) in needles (NE), small branches (SB), coarse branches (CB), stem bark (BA), stem wood (WO), and for both conifers separately stem sapwood (SW) and stem heartwood (HW), for trees of European beech, Douglas fir and Scots pine in dominant position (red bars), intermediate position (green bars) and suppressed position (blue bars). Error bars indicate standard error from the mean value ($n=5$). Different capital letters indicate significant differences ($p < 0.01$) among NE, SB, CB, BA and WO, and small letters between sapwood (SW) and heartwood (HW) for Douglas fir and Scots pine. We did not find significant differences for European beech and Douglas fir between canopy positions but did so for all variables for Scots pine ($p < 0.01$). The within compartment interaction with canopy position is marked with * when $p < 0.01$. Without an * the main effects of canopy position and interactions with compartments were insignificant.

3.2 *Nutrient concentrations.*

Nutrient concentrations differed between tree compartments. Overall, nutrient concentrations increased from stem wood < coarse branches < stem bark < small branches < needles (Fig. 4.4). Calcium, however, showed the highest concentrations in the stem bark for both beech and Scots pine (Fig. 4.4). Patterns were less obvious for micronutrients, such as Mn, Cu, Zn and Fe (Fig. S4.8).

Significant effects of canopy position on macronutrient concentrations were absent in European beech and Douglas fir and hardly observed in Scots pine. Suppressed Scots pine trees had higher Ca concentrations compared to dominant trees with strongest effects in the stem wood (Fig. 4.4). Micronutrient concentration in Douglas fir was hardly influenced, effects were absent in both beech and Scots pine (Table 4.3, Fig. S4.8). Significant interactions between tree compartments and canopy positions were scarce, only in Scots pine weak significances were found.

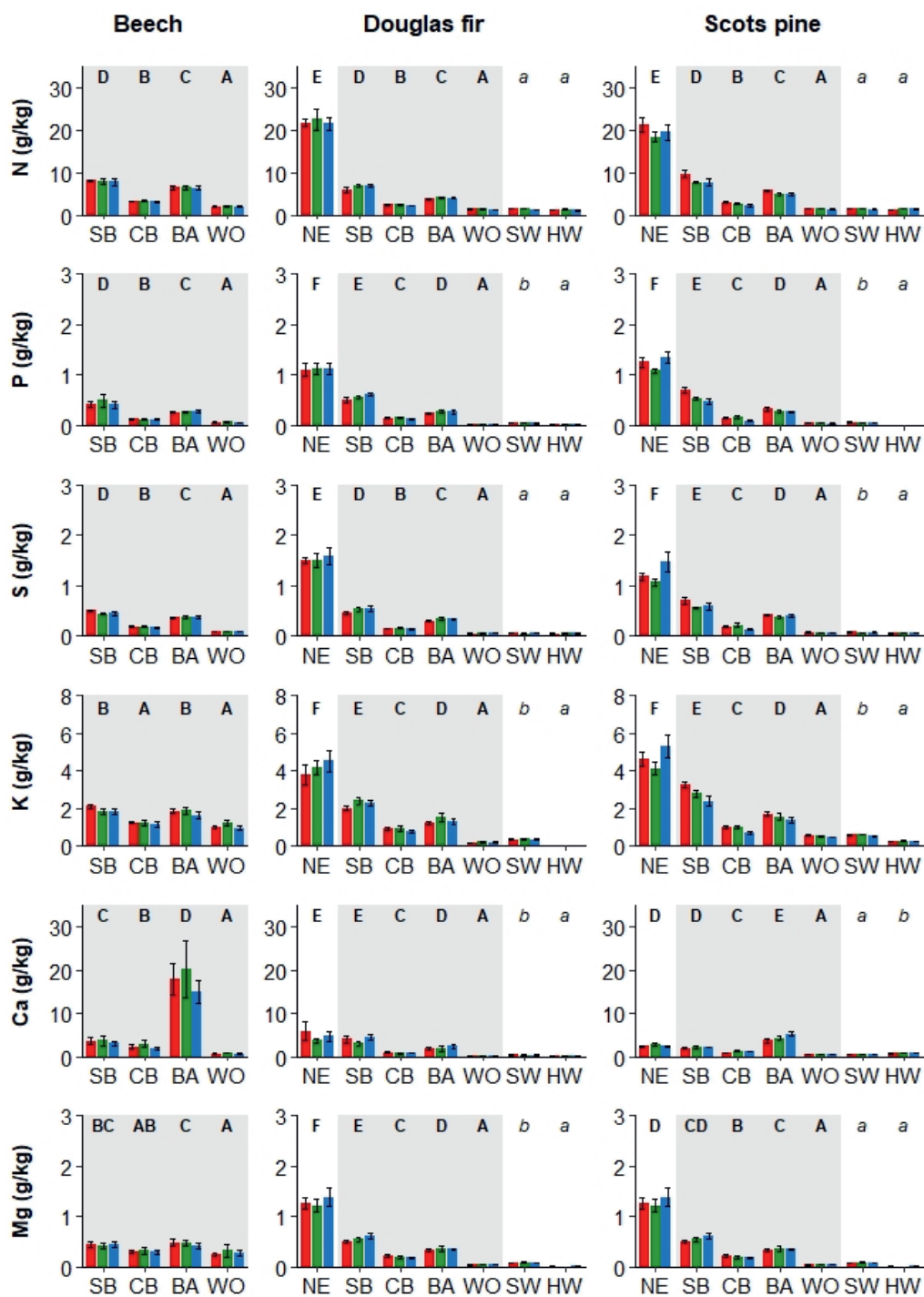


Figure 4.4 Concentrations of macronutrients in needles (NE), small branches (SB), coarse branches (CB), stem bark (BA), stem wood (WO), and for both conifers separately for stem sapwood (SW) and stem heartwood (HW) for trees in dominant position (red bars), intermediate position (green bars) and

suppressed position (blue bars). Error bars indicate standard error from the mean value (n=5). Different capital letters indicate significant differences ($p < 0.01$) among NE, SB, CB, BA and WO, and small letters between sapwood (SW) and heartwood (HW) for Douglas fir and Scots pine. All interactions between canopy position and tree compartments were insignificant ($p > 0.01$).

Table 4.3 Significance levels (P-values) of the analysis of variance for the effect of canopy position on nutrient concentrations and nutrient mass fractions. Mean values, S.E. and test statistics of Anova are in Table S4.5 and Table S4.6. To reduce the number of type-I error results in our multiple test comparison, we only highlight (in bold) the significant results with $P < 0.01$.

Species	N	P	S	K	Ca	Mg	Mn	Cu	Zn	Fe
Concentration										
Beech	0.87	0.67	0.32	0.45	0.67	0.90	0.099	0.22	0.30	0.14
Douglas fir	0.31	0.90	0.12	0.69	0.78	0.39	<.001^a	0.34	0.65	0.12
Scots pine	0.21	0.083	0.88	0.84	<.001^a	0.063	0.18	0.34	0.32	0.049
Mass fraction										
Beech	0.70	0.81	0.69	0.89	0.60	0.63	0.93	0.51	0.38	0.41
Douglas fir	0.15	0.85	0.76	0.31	0.55	0.50	0.60	0.14	0.70	0.028
Scots pine	0.29	0.70	0.73	0.84	0.97	0.67	0.94	0.96	0.078	0.44

Notes: Significant P-values are given in bold. To reduce the number of type-I error results in our multiple test comparison, statistical tests were considered significant at $p < 0.01$. ^a not different in post-hoc test.

3.3 Nutrient mass fractions

All tree compartments contributed substantially (c. >10%) to nutrient stocks in trees, regardless of low biomass fractions or low nutrient concentrations (Fig. 4.5, Fig. S4.10). Stem wood in general contained the highest stocks, up to 50%, but not in all cases. For example, beech trees stored up to 40% of the Ca mass in the stem bark and only 30% in the stem wood. Douglas fir trees stored relatively similar nutrient amounts in stem wood and stem bark, except for N. Scots pine trees stored 40% of the total P mass in needles, while for other macronutrients the highest stocks were present in the sapwood. On average, woody branches hold 42% of the nutrient mass in beech, 30% of the nutrient mass in Douglas fir and 40% of the nutrient mass in Scots pine. Overall, the analyses imply that all tree components contribute substantially to overall nutrient stocks, but that nutrient stocks per tree compartment differ largely between species.

Effects of canopy position on the nutrient mass distributions were non-significant (Table 4.3). Significant interactions between tree compartments and canopy positions were scarce, only in Scots pine weak significances were found (Table S4.6). Overall, canopy position effects were thus marginal or absent.

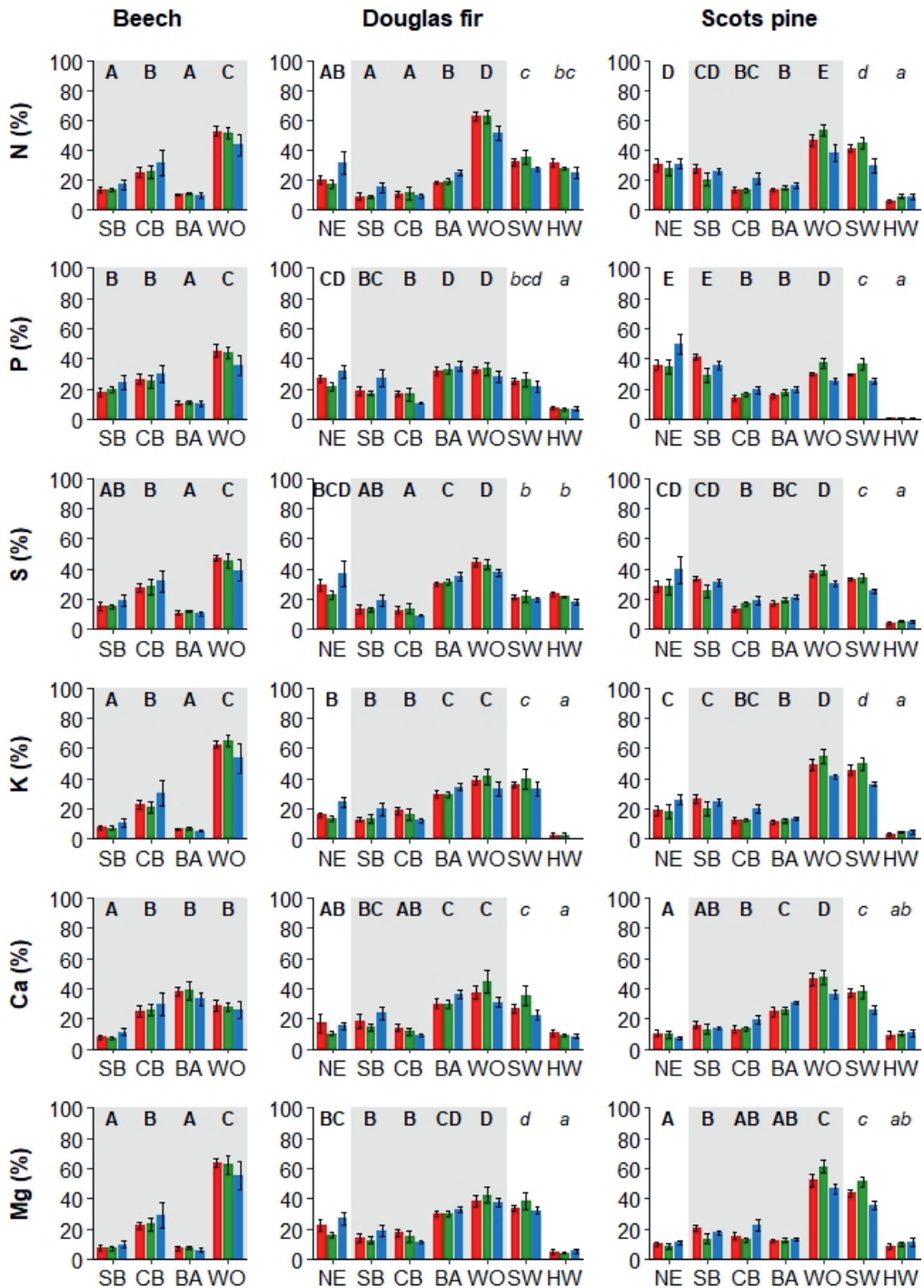


Figure 4.5 Mass distribution expressed as 100% of total above ground mass for macronutrients in needles (NE), small branches (SB), coarse branches (CB), stem bark (BA), stem wood (WO), and for both conifers separately stem for sapwood (SW) and stem heartwood (HW), for trees in dominant position (red

bars), intermediate position (green bars) and suppressed position (blue bars). Error bars indicate standard error from the mean value ($n=5$). Different capital letters indicate significant differences ($p < 0.01$) among NE, SB, CB, BA and WO, and small letters between sapwood (SW) and heartwood (HW) for Douglas fir and Scots pine. All interactions between canopy position and tree compartments were insignificant ($p > 0.01$).

4 Discussion

In this study, we compared the biomass stocks and the carbon and nutrient concentrations within tree compartments between dominant, intermediate and suppressed trees of European beech, Douglas fir and Scots pine growing in forests on poor sandy soils in the Netherlands. We show that the effects of canopy position on the carbon and nutrient concentrations and on the distributions of biomass, carbon and nutrients between compartments are often not significant. In line with our questions, we put this major result in context by discussing the carbon concentrations within tree compartments, the nutrient concentrations within tree compartments and the distribution of biomass, carbon and nutrients among different tree compartments. Ultimately, the implications for the effect of canopy position on total amounts and distribution of biomass, carbon and nutrient stocks are discussed.

4.1 Carbon concentrations slightly deviated from 50%

Our study results confirm that carbon concentrations are indeed relatively constant and close to 50% but highlights nevertheless subtle differences across species and tree compartments. Averaged carbon concentrations in this study were above 50% in both conifers (Douglas fir: 50.3%, Scots pine 50.7%) and below 50% in beech (48.8%). These values are similar to those reported in the literature for beech (Joosten et al., 2004), Douglas fir (Canary et al., 2000; Jain et al., 2010) and Scots pine (Janssens et al., 1999; de Aza et al., 2011). Our study thus confirms that carbon concentrations are close to the 50% estimate, but that angiosperms may have slightly lower carbon concentrations in plant tissues than gymnosperms (Thomas and Martin, 2012).

For all three species, a subtle increase in carbon concentrations was observed from stem wood, coarse branches, small branches towards needles. This slight increase can be explained by the proximity towards foliage where sugars are produced (Woodruff and Meinzer, 2011) and, in case of beech, related to the storage of non-structural carbohydrates for spring growth (Barbaroux et al., 2003). The observed carbon concentrations per tree compartment were similar to those reported by others (Laiho and Laine, 1997; Tolunay, 2009; Armolaitis et al., 2013; Hernández-Vera et al., 2017; Beets and Garrett, 2018; Husmann et al., 2018; Węgiel and Polowy, 2020). Subtle differences were found between different tree compartments with relatively low carbon concentrations in the sapwood and, for both conifers, high carbon concentrations in the bark (Fig. 4.3). High carbon concentrations in the bark have also been observed

among other species (Bert and Danjon, 2006; Tolunay, 2009; Martin et al., 2015; Pompa-Garcia et al., 2017), and might be related to high levels of lignin (Franceschi et al., 2005; Bert and Danjon, 2006), non-structural carbohydrates (Zhang et al., 2014), and defense chemicals (Franceschi et al., 2005; Graça, 2015). In contrast, the carbon concentration in the thin bark of beech was similar to the carbon concentrations of other tree compartments (Fig. 4.3). The higher carbon concentration in the heartwood compared to the sapwood of both conifers, most strikingly for Scots pine, has also been observed in other coniferous tree species (Jain et al., 2010; de Aza et al., 2011), and may be attributed to higher concentrations of lignin (Scheffer, 1966; Bertaud and Holmbom, 2004; Benouadah et al., 2019), cellulose (Bertaud and Holmbom, 2004; Campbell et al., 2007; Benouadah et al., 2019), resin acids (Piispanen and Saranpää, 2002; Bergström, 2003), and various kinds of lipophilic and hydrophilic extractives (Bertaud and Holmbom, 2004; Benouadah et al., 2019) all with different carbon concentrations. These results imply that the presence of heartwood affects the observed carbon concentrations within stems. Overall, our study confirms that small differences exist in carbon concentrations between aboveground tree compartments whereby variation within conifers was higher compared to beech, and that assumption of 50% carbon concentrations would lead to an overestimation of the carbon stock of 3.6% in European beech and an underestimation of 1.2% in Scots pine.

4.2 *Nutrient concentrations indicate N surplus and P limitation*

We compared nutrient concentrations within different aboveground tree compartments with other studies to show possible effects of the nutrient availability in the acidified sandy soils, and the relatively high levels of nitrogen deposition. Nutrient concentrations differed between tree compartments: macronutrients were highest in the needles and lowest in the stem wood, as was expected based on the different physiological demands of tree compartments and shown by multiple other studies (Clayton and Kennedy, 1980; Ranger et al., 1995; Mussche et al., 1998; Knust et al., 2016; Husmann et al., 2018; Węgiel et al., 2018; de Vries et al., 2019). For micronutrient concentrations we did however not observe such trends amongst tree compartments (Table S4.8) suggesting that physiological demands are of limited importance and possibly overruled by age related nutrient accumulation (Caritat and Terradas, 1990; Li et al., 2020), and the relative mobility of micro-nutrients (Ots and Mandre, 2012) which can be influenced by a surplus of N as well as P-limitation (Wu et al., 2021).

The nutrient concentrations of our study trees differed from the values reported in literature, with low concentrations of P, K, Ca, Mg and Mn, especially in both conifer species (Table 4.4). The N concentrations were high, which agrees with the high levels of nitrogen deposition for our study sites. These high N concentrations were also reflected by low foliar C:N ratios in Douglas fir and Scots pine, with values (resp. 24 and 27%) only half the ratios reported for other temperate conifers (McGroddy et al.,

2004; Sardans et al., 2011). Foliar concentrations indicated low nutritional status of P and K in Douglas fir while P only was latent deficient in Scots pine (Van den Burg and Schaap, 1995; Mellert and Gottlein, 2012). The observed foliar N:P ratios (17-20%) exceeded the N:P thresholds of 14.1 (Scots pine) and 16 (general threshold), which is indicative of P limitation (Koerselman and Meuleman, 1996; Aerts and Chapin III, 1999; Mellert and Gottlein, 2012). Such P limitation is also echoed by the very low foliar C:P ratios of Douglas fir and Scots pine, which was only 1/3th of the average foliar C:P ratio reported for a set of temperate conifers (McGroddy et al., 2004). The observed differences in nutrient concentrations are probably driven by surplus of N and limitation of P and base-cations, reflecting the soil acidity (pH between 4.0 and 4.5) of the studied forests.

Our study trees are representative of forest with low tree nutritional status. Tree nutritional status is decreasing over Europe with N deposition as the hypothesized trigger (Jonard et al., 2015). Surplus of N and the related soil acidification causes leaching of base cations and releases of aluminum from the soil (Bowman et al., 2008). The average occupation of aluminum on the cation exchange complex in this study is $84 \pm 2.5 \%$ (Table 4.1) indicating a nutrient poor and strongly acidified soil. High soil N and soil acidification can impair the uptake of P, K and Mg (Braun et al., 2020a) but still increases tree productivity (de Vries et al., 2014a; Jonard et al., 2015; Sardans et al., 2016), which results in limitation of nutrients like P (Braun et al., 2010; Sardans et al., 2016; Du et al., 2021). Signs of deficiencies of P in Douglas fir stands and shortages of Mg and Ca in Scots pine stands was already detected in 1986 in forests in the Netherlands and related to high levels of N deposition (Mohren et al., 1986; Houdijk and Roelofs, 1993). Low K concentrations, especially in Douglas fir, is known to be negatively correlated with increasing levels of ammonium (Van Dijk et al., 1990). Also, this study confirms low macro nutrient concentrations in the aboveground tree compartments indicating the persistent effects of N deposition on the forest ecosystem.

Table 4.4 Average nutrient concentrations of N, P, S, K, Ca and Mg (g/kg) in literature and the difference (%) with values observed in this study. Percentage of difference (Diff) is calculated as the mean concentration of this study divided by the mean concentration in literature minus 100%. Difference of $\geq 20\%$ are highlighted. The reference studies used (n: number of studies) are in Table S4.7.

		Beech			Douglas fir			Scots pine		
		Mean		Diff	Mean		Diff	Mean		Diff
		n	g/kg	%	n	g/kg	%	n	g/kg	%
N	NE				7	13	63	4	13	50
	SB	3	7.4	-3.0	4	6.7	-2.1	2	6.2	35
	CB	2	2.8	15	2	4.1	-43	2	2.3	16
	BA	6	7.8	-18	6	3.6	10	4	4.1	27
	SW	5	2.0	2.6	3	1.7	-18	4	0.93	57
	HW				3	1.5	-20	4	0.72	87
P	NE				7	1.8	-37	4	1.3	-5.8
	SB	4	0.59	-25	4	0.71	-21	2	0.56	1.3
	CB	3	0.25	-52	2	0.35	-59	2	0.25	-46
	BA	7	0.37	-29	6	0.34	-23	6	0.44	-33
	SW	6	0.09	-33	2	0.06	-31	4	0.07	-28
	HW				2	0.01	2.9	4	0.02	-75
S	NE				2	0.99	55	2	1.1	11
	SB	1	0.48	-4.0	0	n.d.	n.d.	1	0.73	-16
	CB	1	0.15	22	1	0.12	26	1	0.25	-31
	BA	3	0.51	-27	3	0.19	64	4	0.55	-27
	SW	2	0.10	-7.6	2	0.07	-35	0	n.d.	n.d.
	HW				2	0.06	-23	0	n.d.	n.d.
K	NE				7	6.1	-32	5	4.6	1.0
	SB	4	2.2	-12	4	2.6	-13	3	2.7	4.9
	CB	3	1.4	-14	2	1.4	-37	2	1.0	-13
	BA	7	2.3	-23	6	1.6	-17	6	1.6	-2.5
	SW	6	1.1	-1.4	3	0.42	-14	5	0.44	32
	HW				3	0.14	-90	5	0.17	55
Ca	NE				7	6.2	-23	5	3.1	-18
	SB	4	4.3	-21	4	6.1	-37	3	2.2	-3.4
	CB	3	2.5	-5.0	2	5.3	-84	2	1.7	-32
	BA	7	18	0.28	6	4.3	-52	6	7.3	-39
	SW	6	0.97	-24	3	0.43	-14	5	0.61	-7.3
	HW				3	0.22	-47	5	0.77	7.6
Mg	NE				6	1.3	-0.5	5	0.75	-9.3
	SB	4	0.44	-2.3	3	0.73	-23	3	0.60	1.9
	CB	3	0.32	-2.8	2	0.33	-38	2	0.39	-22
	BA	6	0.51	-11	6	0.33	4.9	4	0.57	-15
	SW	6	0.25	14	3	0.09	3.7	5	0.16	8.3
	HW				3	0.03	-61	5	0.16	14

4.3 *Distributions were similar for biomass and carbon, but differed for nutrients*

The distribution of biomass and carbon amongst tree compartments was – as expected from the relatively constant carbon concentrations – highly similar (and hence we focus on only biomass in this discussion) but differed from the distributions of nutrients. As expected, most biomass is stored in stem wood (range: 67% - 80%) and these observed stem wood mass fractions fell within the range of reported values in earlier studies (Nihlgård, 1972; Grier and Logan, 1977; Pellinen, 1986; Ranger et al., 1995; Vanninen et al., 1996; Andre et al., 2010; Husmann et al., 2018). The sapwood fraction (Douglas fir 39%, Scots pine 60%) was relatively close to the heartwood fraction (41%) in Douglas fir, while the heartwood fraction (12%) was lower in Scots pine, but such fractions are typically age dependent (Vanninen et al., 1996; Gjerdrum, 2003). Stem bark biomass fractions in beech (4.2%) and Douglas fir (9.3%) were close to those reported by earlier studies for beech (Nihlgård, 1972; Husmann et al., 2018) and Douglas fir (Ranger et al., 1995), but lower for Scots pine (6.4%) compared to an earlier study on 40-80 year old trees (DBH 16–33 cm) in southern Finland (Vanninen et al., 1996). The lower stem bark fraction was probably caused by a lower volume of the bark since the density of the bark (0.32 g cm^{-3}) was within the normal range ($0.27\text{--}0.36 \text{ g cm}^{-3}$) (Dibdiakova and Wang, 2015). The observed biomass fractions in branches (range: 10% - 29%) were consistent with branch mass fractions reported by previous studies (Nihlgård, 1972; Ranger et al., 1995; Skovsgaard and Nord-Larsen, 2012; Wertz et al., 2020). The observed needle mass fractions in both Douglas fir (1.8%) and Scots pine (3.6%) were low, only half the needle mass percentages reported in literature (Ranger et al., 1995; Vanninen et al., 1996). Since our study trees were harvested in the winter of 2019, we speculate that these low needle masses were caused the severe 2018 summer drought, causing defoliation both in Douglas fir and Scots pine (Rebetez and Dobbertin, 2004; Galiano et al., 2010; Sergent et al., 2014). We thus conclude that biomass and carbon distributions were rather similar to values reported from other sites, expect for some differences caused by age (heartwood – sapwood) or recent weather conditions (needle mass).

Whereas the highest stocks of biomass and carbon (>67%) were within stems and much lower for the other tree compartments (range: 1.8% - 24%), stem stocks were relatively low for nutrients (range: 23% - 60%) while other tree compartment had stocks of 18% - 19%. These differences result from the (in most cases) much higher nutrient concentrations in bark, branches and needles, reflecting high physiological demands compared to the stem. The nutrient stocks in the stem wood are compared to the other components still higher (average: 44%), which thus resulted from the large stem wood biomass fractions. The implications of these nutrient stocks in bark and crown should be considered in forestry practices, moving from stem only harvest to biomass harvest including crown.

4.4 *Canopy position had only minor effects on aboveground carbon and biomass distributions*

Our results show that the aboveground distribution of carbon and biomass among different tree compartments was hardly affected by the canopy position of trees. First, canopy position had no effect on carbon concentrations except in Scots pine where the needles of suppressed trees had slightly lower carbon concentrations than the needles of dominant or intermediate trees (Fig. 4.3). Since trees were harvested in winter, it remains very speculative whether such differences in needle carbon concentrations between canopy positions result from a lower respiration: gross assimilation - ratio in trees with higher canopy position (Lebaube et al., 2000). Earlier studies also find hardly any significant effect of canopy position on tree carbon concentrations (Naidu et al., 1998; Xing et al., 2005; Zhang et al., 2009; Zhou et al., 2018). We therefore conclude that carbon concentrations are not affected by the canopy position of trees.

Second, the distribution of biomass and carbon hardly differed between suppressed, intermediate and dominant trees, except for some minor effects observed for Scots pine. This result seems remarkable since it is well known that dominant trees grow much faster than suppressed trees (D'Amato and Puettmann, 2004; Reid et al., 2004; Castagneri et al., 2008) and develop wider crowns and thicker stems than more suppressed trees (Dieler and Pretzsch, 2013; Pretzsch, 2014). While such differences were also apparent for our study trees confirming that trees are plastic in crown shapes, our results show that trees can be highly plastic in shape while controlling the distributed biomass amongst tree compartments within narrow ranges. This result is consistent with observations of similar biomass distributions between twigs and leaves for trees in temperate to boreal conditions (Petit et al., 2018), by the similar distributions of leaf versus stem biomass in trees of dry versus wet sites (Anfodillo et al., 2016) and by other studies on aboveground biomass distribution (Gargaglione et al., 2010; Skovsgaard and Nord-Larsen, 2012; Van de Peer et al., 2017). Our results thus imply that aboveground biomass distributions are more in line with the theory of fixed scaling relationships (Shinozaki et al., 1964; Enquist and Niklas, 2002), and less with the functional equilibrium theory (Brouwer, 1962; Reynolds and Thornley, 1982).

Remarkably, the functional equilibrium theory – predicting that resource allocation is driven by priority and demand whereby trees adapt the biomass distribution in response to competition - was supported by other studies reporting a higher share of crown biomass for dominant trees (Bartelink, 1996; 1997; Naidu et al., 1998; Vanninen and Mäkelä, 2005; Krejza et al., 2017; Wertz et al., 2020) and increased height growth for suppressed trees (Naidu et al., 1998). The effects of canopy position on the biomass distribution in Scots pine did not involve an increase of crown biomass fraction for dominant trees (Fig. 4.3) nor increased height growth for suppressed trees (Fig. S4.7) providing no support for the functional equilibrium theory. Absence of an effect of canopy position on the aboveground biomass distribution might be related to the environment (Lines et al., 2012) and the exposure to limited resources (Schall et al., 2012; Slot et al., 2012). For example, beech showed remarkable plasticity in response to

competition (Dieler and Pretzsch, 2013; Pretzsch, 2014) although low site fertility weakened the effect (Dieler and Pretzsch, 2013). Also for Scots pine smaller effects of competition on biomass distribution were observed on poor sites (Vanninen and Mäkelä, 2005). We therefore hypothesize that the absence of an effect of canopy position on biomass distribution, nutrient concentrations and nutrient stocks is related to the nutrient poor and acidic site conditions resulting from the effects of N deposition.

4.5 *Canopy position is not the main driver of nutrient concentrations and stocks*

Canopy position was hypothesized to alter nutrient concentration as dominant trees have higher metabolic rates and therefore acquire water and nutrients more rapidly (Martin et al., 1997; Granier et al., 2000; D'Amato and Puettmann, 2004; Reid et al., 2004; Castagneri et al., 2008; Aranda et al., 2012). Surprisingly, this study showed hardly any effect of canopy position on nutrient concentrations. Absence of an effect of canopy position on nutrient concentrations was observed by multiple studies (Höhne, 1964; Son and Gower, 1992; Naidu et al., 1998; Sette et al., 2013). The higher levels of Ca in Scots pine trees with a suppressed canopy position are in line with observations for *Nothofagus antarctica* (Peri et al., 2006). Remarkably there is no consistency in the nutrient concentrations between trees of different canopy position in Scots pine as lower concentrations of N, P, K and S were observed in dominant trees compared to suppressed trees (Wright and Will, 1958; Węgiel et al., 2018). The mechanisms behind these differences in nutrient concentrations remains speculative. Immobile nutrients, like Ca, can accumulate in older woody parts due to low translocation rates which could cause higher concentrations in suppressed trees (Finér and Kaunisto, 2000; Prasolova and Xu, 2003). The minor effects of canopy position on biomass distributions and nutrient concentrations explain the absence of strong effects of canopy position on nutrient stocks. While we cannot exclude a mitigating role of the poor soils in our study sites on divergent nutrient concentrations within trees, our results and those from the reported literature imply that canopy position does not act as the main driver of tree nutrient concentrations for supporting divergent metabolic rates between trees differing in canopy position.

5 **Conclusions**

The distribution of biomass, carbon and nutrient differs among tree compartments and tree species. The canopy position does have no or minor effects on the aboveground distribution of biomass and carbon and on nutrient concentration and distributions between aboveground tree compartments. These results are better in line with the allometric scaling theory than the functional equilibrium theory.

Our study implies that models aiming for estimating tree and forest biomass and carbon and nutrient stocks should apply species specific biomass, carbon and nutrient stocks with equal biomass, carbon and nutrient

stocks for trees independent of canopy position as a valid assumption, but we nevertheless recommend testing this assumption for a broader range of species and site conditions.

Acknowledgements

This research is part of the Nutrient Balance project and was funded by the Dutch Research Council (NWO, No. ALWGS.2017.004). We acknowledge Henk van Roekel, Leo Goudzwaard and many other members of the harvest team for their valuable help with establishing the experiment, the harvest and the sample processing. We thank our partners, National Forest Service, Union of private Forest Owner Groups, Het Loo Royal Estate, Staro nature and countryside, Borgman management consultants, National Park de Hoge Veluwe and Blom Ecology for financial support, permission to work in their forest or other provided services.

Supplementary information

Supplementary methods

Destructive biomass sampling

Trees selected for destructive biomass sampling were harvested in February and March 2019 with a chainsaw at approximately 20 cm above the ground surface. Directly after harvest, the total tree height, length of the stem, crown length, and the diameter and length of all living branches were measured with measuring tape and caliper. The stem was defined as the main axis extending from the butt until the point where the stem diameter reached 10 cm. The crown was defined as branches < 10 cm diameter and, if present, needles. Crown length was measured from the point where the stem diameter reached 10 cm till the bud.

The fresh weights of the whole tree and the stem were determined in the field by weighing the tree with a tractive scale (Allscales Europe, cap. 3000 kg $\pm$ 2kg) attached to a mechanical crane. For dominant trees of Douglas fir (*Pseudotsuga menziesii*) and dominant and intermediate beech trees (*Fagus sylvatica*), the trees were weighted in separate parts while for Scots pine (*Pinus sylvestris*) and the smaller Douglas fir trees the whole trees were weighted as a whole. The weight of the tree crown was calculated as the difference between the weight of the whole tree and stem mass after the branches of the tree crown were removed.

Stem disks with an average thickness of 3.5 cm were taken at the butt, at the top point where the stem diameter was 10 cm, and at 1/4th, 1/2nd and 3/4th of the stem length. Disks samples were also taken at the point where the main crown axis had a diameter of 2 cm and at the mid-point of the crown length. All disks were labelled, wrapped in plastic bags to avoid water loss, and stored at 4°C until further processing.

To estimate the biomass of needles and branches 4 representative branches (not severely damaged due to the felling) per tree were selected, covering the observed range of measured branch diameters. Branch wood was divided in two subcategories: small branches containing all branch wood with a diameter less than 2 cm and coarse branches containing all branch parts with diameter above 2 cm. For each branch, the diameter (close to branch junction), total length and length of the coarse branch were measured. The total branch fresh weight and the fresh weight of the coarse branch were weighted using a tractive scale (crane scale SF-918, cap. 150 kg $\pm$ 0.1kg), the latter after removing all sub-branches at the point where these reached 2 cm in diameter. These sub-branches were collected in sealed plastic bags, labelled per sample branch and stored at 4°C until further processing.

Stem and crown disks were further processed in the laboratory. Initial measurements include opposing diameters covering the highest within-disk variability, perimeter and disk thickness measurements. Perimeter and diameter measurements were repeated after separation of bark, sapwood and if present, heartwood. For each disk the total fresh weight was determined, and the fresh weight of the disk wood and disk bark were weighted after peeling the bark of the disk. The dry weight of the bark was determined after drying the bark for at least 48 hours at 70°C. The wet and dry density of the bark (g cm^{-3}) were determined by weighting a representative wet and dry bark piece and submerging in water on a balance to measure the weight and the volume respectively.

Disks of Scots pine were marked with safranin to separate sapwood from heartwood. For Scots pine and Douglas fir, sapwood and heartwood, if present, was split manually with the use of a chisel (Fig. S4.2). Both the fresh weights (g) of the sapwood and the heartwood were determined. For determination of dry weight of the sapwood and heartwood, two pieces of these tissues were extracted with a chisel from opposing sites on the disks, covering the highest variability. The fresh weight of the extracted sapwood and heartwood pieces was determined prior to submergence in water on a balance to measure the fresh volume. Dry weight and dry volume were determined after the pieces were dried for at least 48 hours at 70°C following the same protocol. The dry weight of the sapwood and heartwood was calculated based on the percentage water loss in both pieces.

For all sampled branches with diameter smaller than 2 cm, the total fresh weight was measured in the laboratory. Needle subsamples were taken from the biggest and smallest branch to determine the water loss after drying at 70°C to a constant weight. Dry weight of the branch material was recorded before branch wood and needles were split manually. For beech, no further separation had to be made between branch material and foliage because the trees did not have leaves. After separation, small branch wood and needles were dried at 70°C to a constant weight and their dry weights were determined.

Preparation of nutrient samples

Composite nutrient samples were prepared for stem bark, sapwood and heartwood and for coarse branches (diameter > 2 cm), small branches (diameter < 2 cm) and needles. All material for nutrient analysis was grinded to 1.5 mm and dried at 70°C to a constant weight before the composite sample was taken.

Composite samples for needles were based on randomly taken needles from the biggest and smallest sampled branch per tree and mixed in equal proportions. For the composite sample of the small branch, all branch parts with a diameter < 2 cm were grinded and a composite sample was taken after the material was thoroughly mixed.

The composite samples of the stem bark, sapwood and heartwood and the coarse branches were based on the stem and crown disks. Stem and crown disks were processed following the same procedure:

for each disk the bark was sampled as a whole, and for both sapwood and heartwood sample material was taken from the sawdust left after drilling holes covering the complete disk to correct for differences in nutrient concentrations between younger and older tissues. Only in case of small disks the entire sapwood and heartwood were sampled. The procedure for the composite samples differed between coarse branches and stem bark, sapwood and heartwood. For both the nutrient samples was a composite sample between disk but additional steps were needed for coarse branches to mix the bark, sapwood and heartwood within disks.

The proportions of the composite sample per disk for respectively stem bark, sapwood and heartwood assumed that the disks were representing the whole stem in equal proportions except for disk 1 and disk 5. These disks represent only half of the stem length compared to disk 2, 3 and 4 (Fig. S4.3). To correct for a difference in tissue density along the stem the surface of the tissue (cm^2) was multiplied by a thickness of 1 cm, which allowed to calculate the disk weigh based on the tissue density (cm^3). The weight (W) of the tissue t per disk i for the composite sample was calculated as a function of the tissues surface per disk (S_{ti}) and the tissues density (ρ_{ti}):

$$W_{ti} = (y * l_i * S_{ti} * \rho_{ti}) / TW_t \quad (\text{S1})$$

Where y is the desired sample weight (g), l is the segment length for disk i (Fig. S4.3) and TW_t is the total weight of tissue t along the stem. TW_t is calculated as:

$$TW_t = \sum_{i=1}^5 (l_i * S_{ti} * \rho_{ti}) \quad (\text{S2})$$

The subsamples of bark, sapwood and heartwood per disk were merged per tissue to the composite samples of stem bark, sapwood and heartwood.

The composite sample of the coarse branch consists of a mix of bark, sapwood and if present, heartwood from disks taken from crown base, at the mid-point of the crown length and at the point where the main crown axis was 2 cm diameter. First a composite sample with a representative composition of bark, sapwood and, if present, heartwood was taken per disk. This within-disk composite sample was based on the dry weight of the tissues, where the dry weight of the sapwood and heartwood was calculated as a function of the fresh weight and the fresh and dry weights of the extracted sapwood and heartwood pieces (Fig. S4.2):

$$DW_{ti} = FW_{ti} * \left(\frac{DW_{ti,1}}{FW_{ti,1}} + \frac{DW_{ti,n}}{FW_{ti,n}} \right) / n \quad (\text{S3})$$

Where DW is the dry weight of tissue t on disk i , FW is the fresh weight and numbers denote fresh and dry weights of the two extracted sapwood and heartwood pieces. The dry weight per tissue per disk was used to calculate the weight of the separate tissues for the within disk composite sample (W_{ti}):

$$W_{ti} = (DW_{ti} * y) / DW_i \quad (S4)$$

where y is the desired sample weight (g) and DW_i is the sum of the dry weights of bark, sapwood and, if present, heartwood. The within disk composite sample was the result of the merged subsamples of the tissues.

Second the three within disk composite samples were merged to one composite sample representing the coarse branch. To do so, the weight of the three crown disks was averaged to a 1 cm thick disk by dividing the weight by the disk thickness. The length of the branch represented by each crown disk is, comparable to the stem disk, only half the length for the disk at branch base and branch top compared to the disk at mid-point of the branch length. The weight (W) of the sample per disk i for the composite sample was calculated as the weight of the 1 cm thick disk times the segment length divided by the sum of the standardized weights of the three disks:

$$W_i = (l_i * y * AW_i) / TW \quad (S5)$$

where l is the length represented by disk i , y is the desired sample weight (g) and AW_i is the averaged weight of disk i . The total weigh (TW) is the sum of the averaged weights of the three disks. The composite sample for the coarse branch is the product of the weights calculated in formula S5.

Stem volume and stem dry weight calculations

The stem was defined as the main axis extending from the butt until the point where the stem was 10 cm in diameter. The fresh and dry weights of bark, heartwood and sapwood were measured based on 5 stem disks taken with equal spacing along the stem. R code is available upon request.

Calculations of the stem volume

The calculation of the volume of the bark, sapwood and heartwood within the stem was based on the perimeter (cm) of the disk wood and heartwood and based on dry weight, density and disk thickness for bark. Perimeter data of disk wood and heartwood was used to calculate the average radius for these tissues t on disk i (R_{ti}) via:

$$R_{ti} = (PM_{ti} / \pi / 2) \quad (S6)$$

Because of the heterogenous nature of the bark, a different method was used to calculate the bark radius. This method corrected for the coarse structure of the outer bark which was pronounced for both Douglas fir and Scots pine. Bark radius for disk i was calculated based on the fresh weight (g) of the bark (FW_{BA}), the density of the bark (ρ_{BA} ; g cm⁻³) and the average thickness of the bark (Th_{BA} ; cm):

$$R_{BAi} = \sqrt{FW_{BAi} / \rho_{BAi} / Th_{BAi} / \pi} \quad (S7)$$

Every disk represented a part of the stem which is expressed as stem section length (SSL). Disks 2, 3 and 4 represent twice the stem section length compared to disk 1 and 5. In order to keep the stem section length equal between the disk, the stem is subdivided into 8 equal sections (Fig. S4.4). The height of each section is calculated via:

$$SSL = SL/8 \quad (S8)$$

in which both stem length (SL) and the stem segment length (SSL) were expressed in cm. For each stem section the volume of the bark (based on R_{BAi}), for the stem wood (sapwood and heartwood) and for the heartwood was calculated separately. The volume per tissue t and segment s was based on the formula of a truncated cone:

$$V_{ts} = \left(\frac{1}{3}\right) * \pi * (R_{ti}^2 + R_{ti} * R_{t(i+1)} + R_{t(i+1)}^2) * SSL \quad (S9)$$

The volume of the sapwood is calculated as the dry weight of the stem wood minus the heartwood.

Additional volume corrections were executed for beech and Scots pine based on the number of ramifications (SR) within the stem. The volume calculations were entirely based on the main stem axis, ignoring all ramifications within the stem with a thickness of > 10 cm diameter at junction (Fig. S4.5). The volume of stem ramifications was calculated for bark, sapwood and heartwood (indicated as tissue t) via:

$$V_{SRt} = SR * V_{t(s=5)} * Scaling \quad (S10)$$

The scaling, derived from the best fit between calculated and measured weights, is 1 for Scots pine and 0.55 for beech. The extra volume of V_{SRt} is assigned to the volume of section 5 (Fig. S4.4).

Calculation of the dry weight of the stem

The dry weight of the stem was calculated as the volume of the separate tissues times the density. The dry mass per unit of fresh volume, e.g., the density of tissue t on disk i (ρ_{ti}), was calculated per disk based on dry weight (DW) and fresh volume (FV) measurements of the bark, sapwood and heartwood pieces (Fig. S4.2):

$$\rho_{ti} = \left(\frac{DW_{ti1}}{FV_{ti1}} + \frac{DW_{tin}}{FV_{tin}} \right) / n \quad (S11)$$

The dry weight of the stem is calculated by:

$$DW_{ti} = V_{ts} * \rho_{ti} \quad (S12)$$

$$DW_t = \sum_{i=1}^5 DW_{ti} \quad (S13)$$

$$DW = \sum_{t=1}^3 DW_t \quad (S14)$$

In which the dry weight of tissue t for disk i (DW_{ti}) is calculated based on the volume of the tissue t for section s times the density of tissue t on disk i (formula 12). The dry weight of the tissue along the stem is the sum of the dry weight of the separate stem sections (formula S13) while the dry weight of the entire stem is the sum of the three stem tissues (formula S14).

Calculations of the stems fresh weight

The fresh weight of the stem was calculated in a comparable way to the calculation of the dry weight. The volume of tissue t on section s was multiplied by the fresh density of tissue t on disk i. The fresh density of tissue t on disk i (ρ_{ti}) was calculated based on fresh weight (FW) and fresh volume (FV) measurement of pieces p representing the tissue t on disk i:

$$\rho_{ti} = \left(\frac{FW_{ti1}}{FV_{ti1}} + \frac{FW_{tin}}{FV_{tin}} \right) / n \quad (S15)$$

The fresh weight of the stem was calculated by:

$$FW_{ti} = V_{ts} * \rho_{ti} \quad (S16)$$

$$FW_t = \sum_{i=1}^5 FW_{ti} \quad (S17)$$

$$FW = \sum_{t=1}^3 FW_t \quad (S18)$$

Volume and dry weight calculations of crown compartments

The tree crown was defined as the part of the central tree bole above the 10 cm diameter point including all branches with diameter less than 10 cm at stem junction. The tree crown was further divided into coarse branches (diameter between 2 and 10 cm), small branches (diameter ≤ 2 cm) and needles (only for Douglas fir and Scots pine). For all tissues in the crown, linear mixed effect models were developed as described by (Zuur et al., 2009) with parameters estimated by the restricted maximum likelihood method. In order to avoid the influence of day-to-day variations in moisture content of the tissue, the fresh weight of the separate tissues was converted to dry weight on branch level. All models were based on the volume of branches as this proved to be a better estimate than branch length and diameter as separate response variables. Branch volume of branch i on tree z was calculated as a function of branch-base radius (R) and length (L) of branch i on tree z using the formula of a cone:

$$V_{zi} = (1/3) * \pi * R_{iz}^2 * L_{iz} \quad (S19)$$

The dry weight (DW) of the coarse branches (CB), small branches (SB) and needles (NE) of branch *i* on tree *z* per species was modelled as a function of branch volume (*V*). For all models, a random intercept slope model with different intercepts for volume and different slopes per tree improved model AIC considerably ($AIC \geq \Delta 2$). The used model for all tissues was:

$$Y_{iz} = \mu + \alpha_i + \varepsilon_{iz} \quad (S20)$$

Where *Y* is the dry mass of branch *i* on tree *z*, μ is the intercept, α is the fixed effect of branch volume and ε is the residual error within branch (*i*) and tree (*z*). Details on the models are in Table S4.1.

To obtain mass predictions for each crown tissue per tree, the formulas for coarse branch, small branch and needle were applied for all branches per tree. The total dry crown biomass consisted of the sum of the biomass of coarse branches, small branches and needles of all branches per tree. For Douglas fir and Scots pine the biomass of the main axis above the 10 cm diameter point was calculated separately and added to the dry crown biomass.

The biomass of the main axis above the 10 cm diameter point (the leading branch) was calculated based on measurements of the 3 stem disks taken with equal spacing along this leading branch. Leading branch biomass calculations were similar with the stem biomass calculations following the formulas S6 to S18. The total biomass of the leading branch was the sum of the weight of all separate tissues.

To evaluate the accuracy of the crown calculations, the dry weight of tissues *t* was multiplied by the ratio of the fresh weight to dry weight:

$$FW_t = DW_t * \left(\frac{FW_{t1}}{DW_{t1}} + \frac{FW_{tn}}{DW_{tn}} \right) / n \quad (S21)$$

The fresh mass of the crown consisted of the sum of the fresh weight of the tissues:

$$FCM = \sum_{t=1}^4 FW_t \quad (S22)$$

Validation of results

The calculation of the fresh stem mass (formulas S15-S18) and the fresh crown mass (formulas S21-S22) allowed for a direct comparison between the calculated and measured fresh stem mass (Fig. S4.6) and fresh crown mass (Fig. S4.7).

The relation between the measured and weighted fresh mass of the stem and crown was highly related, yielding R^2 adjusted above 0.93 for all models and a slope ≥ 0.93 (Table S4.2). The relation between the measured and weighted fresh stem and crown mass was highest for Douglas fir in which the weighted fresh stem mass explained 99% of the variation in the calculated fresh stem mass.

Supplementary Tables**Table S4.1** Soil classification for each plot based on the soil classification guidelines of IUSS Working Group WRB (2015).

Site	Beech	Douglas fir	Scots pine
1	Albic Podzols (Arenic)	Albic Podzols (Arenic)	Albic Podzols (Arenic)
2	Entic or Albic Podzols (Arenic)	Entic or Albic Podzols (Arenic)	Albic Podzols (Arenic)
3	Dystric Cambisols (Arenic)	Entic or Albic Podzols (Arenic)	Dystric Cambisols (Arenic) or Albic Podzols (Arenic)
4	Dystric Cambisols (Arenic, Drainic)	Entic or Albic Podzols (Arenic, Drainic)	Albic Podzols (Arenic, Drainic), or Albic Arenosols (Humic)
5	Dystric Cambisols (Arenic) or Albic Podzols (Arenic)	Albic Podzols (Arenic) or Albic Arenosols (Protospodic)	Albic Podzols (Arenic) or Albic Arenosols (Protospodic)

Table S4.2 Parameter estimates $\pm$ standard error ($n = 60$), the marginal R^2 (R^2_m), t-value and p-value of the intercept and branch volume of the fixed structure of the models for the dry weights of coarse branches (CB), small branches (SB) and needles for beech (BE), Douglas fir (DG) and Scots pine (SP). The random structure was equal between the models and consisted of a random intercept and slope model in which the branch volume was nested within the tree. For the random structure, the standard deviation (S.D.) of the intercept, branch volume and the residual standard deviations are given. The random structure considerably improved the model fit, with higher values of the conditional R^2 (R^2_c) compared to the marginal R^2 .

			Fixed structure						Random structure				
Species	Part	Trans ¹	R^2_c	R^2_m	Intercept		Branch volume		Corr.	S.D.			Corr.
					Estimate	t-value	p-value	Slope		Int.	Vol.	Residual	
BE	CB	Exp.	0.99	0.89	1.1 ± 0.19	5.9	< 0.001	$0.1 E^{-6} \pm 0.01 E^{-6}$	0.24	0.33	$2.1 E^{-8}$	0.99	1.0
	SB	None	0.95	0.79	280 ± 63	4.5	< 0.001	0.21 ± 0.019	0.082	110	0.062	300	0.98
DG	CB	None	0.99	0.98	-0.095 ± 0.034	-2.8	0.0079	$8.4 E^{-4} \pm 0.20 E^{-4}$	-0.34	0.042	$5.1 E^{-5}$	0.14	0.997
	SB	Sqrt	0.96	0.67	-58 ± 33	-1.7	0.092	15 ± 19	-0.88	91	6.8	80	-0.999
SP	NE	Sqrt	0.98	0.64	-91 ± 23	-4.0	< 0.001	13 ± 1.6	-0.88	67	6.1	49	-1.0
	CB	None	0.99	0.96	-0.12 ± 0.034	-3.4	0.0029	$7.1 E^{-4} \pm 0.40 E^{-4}$	-0.73	0.061	$1.1 E^{-4}$	0.12	1.0
	SB	Sqrt	0.95	0.80	-170 ± 50	-3.3	0.0018	28 ± 3.1	-0.88	140	11	150	-0.999
	NE	Sqrt	0.97	0.70	-54 ± 27	-2.0	0.053	14 ± 2.0	-0.90	86	7.2	63	-0.97

Table S4.3 Parameter estimates $\pm$ standard error ($n = 15$), t-value and p-value for the intercept and the measured fresh mass of the models validating the goodness of fit between the measured and the calculated fresh mass for the tree stem and crown of beech (BE), Douglas fir (DG) and Scots pine (SP). The transformation (Trans) was done both on the response and explanatory variable. Intercept was in all cases not-significant. The slope, indicating the goodness of fit, was in all cases > 0.9 .

Species	Part	Trans	R ² -adj.	p-value	Intercept			Measured fresh mass		
					Estimate	t-value	p-value	Slope	t-value	p-value
BE	Stem	None	0.94	< 0.001	26 $\pm$ 110	0.23	0.82	0.94 $\pm$ 0.069	14	< 0.001
	Crown	Sqrt	0.95	< 0.001	-0.66 $\pm$ 1.3	-0.50	0.63	1.1 $\pm$ 0.070	15	< 0.001
DG	Stem	None	0.99	< 0.001	27 $\pm$ 53	0.52	0.62	0.99 $\pm$ 0.021	45	< 0.001
	Crown	Sqrt	0.96	< 0.001	1.0 $\pm$ 0.93	1.1	0.29	0.94 $\pm$ 0.055	17	< 0.001
SP	Stem	Log	0.94	< 0.001	0.30 $\pm$ 0.35	0.86	0.41	0.95 $\pm$ 0.061	15	< 0.001
	Crown	Log	0.93	< 0.001	-0.17 $\pm$ 0.36	-0.48	0.64	1.0 $\pm$ 0.079	13	< 0.001

Table S4.4 Mean values $\pm$ standard error ($n = 15$) of biomass distribution (%), C concentration (g g^{-1}) and C mass distribution (%) of the tree compartments needles (NE), small branches (SB), coarse branches (CB), stem bark (BA), stem sapwood (SW) and heartwood (HW) for beech (BE), Douglas fir (DG) and Scots pine (SP). The F value and significance of the two-way Anova based on canopy position (CP) and tree compartment (TC) and their interaction (Int) are displayed.

	Biomass distribution (%)			C concentration g g^{-1}			C mass distribution		
	BE	DG	SP	BE	DG	SP	BE	DG	SP
NE	-	1.8 ± 0.24^a	3.6 ± 0.37^a	-	51 ± 1.6^c	52 ± 1.4^b	-	1.9 ± 0.25^a	3.6 ± 0.38^a
SB	5.1 ± 0.60^a	2.8 ± 0.38^a	7.0 ± 0.65^{abc}	50 ± 1.3^c	51 ± 0.63^c	52 ± 1.0^{bc}	5.2 ± 0.60^a	2.8 ± 0.39^a	7.1 ± 0.66^{bc}
CB	24 ± 3.4^b	7.7 ± 1.1^b	14 ± 1.9^d	49 ± 0.82^b	50 ± 1.0^{ab}	50 ± 0.87^a	24 ± 3.4^b	7.6 ± 1.1^b	14 ± 1.9^d
BA	4.2 ± 0.25^a	9.3 ± 0.58^b	6.4 ± 0.31^b	49 ± 3.4^{abc}	54 ± 2.6^d	53 ± 2.1^c	4.2 ± 0.25^a	10 ± 0.63^b	6.7 ± 0.34^b
SW	67 ± 3.4^c	39 ± 1.8^c	60 ± 2.1^e	49 ± 0.53^a	50 ± 0.72^a	50 ± 1.1^a	67 ± 3.4^c	39 ± 1.8^c	59 ± 2.1^d
HW	-	41 ± 1.5^c	12 ± 1.4^{cd}	-	50 ± 1.1^b	52 ± 3.8^{bc}	-	41 ± 1.5^c	13 ± 1.4^{cd}
<i>Anova</i>									
CP	0.53 <i>n.s.</i>	6.3 <i>n.s.</i>	3.4 <i>n.s.</i>	0.3 <i>n.s.</i>	0.4 <i>n.s.</i>	3.7 <i>n.s.</i>	0.53 <i>n.s.</i>	6.2 <i>n.s.</i>	3.1 <i>n.s.</i>
TC	54 E^{-1} ***	37 E^{-1} ***	60 E^{-1} ***	25 ***	10 E^{-1} ***	63 ***	54 E^{-1} ***	37 E^{-1} ***	56 E^{-2} ***
Int	0.62 <i>n.s.</i>	0.67 <i>n.s.</i>	3.6 ***	1.5 <i>n.s.</i>	0.9 <i>n.s.</i>	3.7 ***	0.61 <i>n.s.</i>	0.68 <i>n.s.</i>	3.5 **

*** $P \leq 0.001$, ** $0.005 \leq P < 0.001$ * $0.01 \leq P < 0.005$, *n.s.* $P > 0.01$. Different letters denote significant

differences among canopy positions according to Tukey's *posthoc* test with a significance level of $P < 0.01$.

The statistical model used for BE, DG and SP is $Y_{it} = \mu + \alpha_i + B_{t(i)} + \varepsilon_{it}$ where α is the fixed effect of CP, β the fixed effect of TC and ε the residual error within tree (i) and tissue (t).

Table S4.5 Mean values $\pm$ standard error ($n = 15$) of the nutrient concentrations of N, P, S, K, Ca, Mg (g/kg) and Mn, Cu, Zn and Fe (mg/kg) of needles (NE), small branches (SB), coarse branches (CB), stem bark (BA), stem sapwood (SW) and heartwood (HW) for beech (BE), Douglas fir (DG) and Scots pine (SP). The F value and significance of the two-way Anova based on canopy position (CP) and tree compartment (TC) and their interaction (Int) are displayed.

	NE	SB	CB	BA	SW	HW	CP	TC	Int
N									
BE	-	7.9 $\pm$ 0.37 ^d	3.2 $\pm$ 0.09 ^b	6.4 $\pm$ 0.22 ^c	2.0 $\pm$ 0.05 ^a	-	0.14 ^{n.s.}	46 E ⁻¹ ***	0.16 ^{n.s.}
DG	22 $\pm$ 0.99 ^e	6.6 $\pm$ 0.27 ^d	2.4 $\pm$ 0.09 ^b	3.9 $\pm$ 0.12 ^c	1.4 $\pm$ 0.06 ^a	1.2 $\pm$ 0.06 ^a	1.4 ^{n.s.}	52 E ⁻¹ ***	1.6 ^{n.s.}
SP	20 $\pm$ 0.92 ^e	8.4 $\pm$ 0.44 ^d	2.6 $\pm$ 0.13 ^b	5.2 $\pm$ 0.19 ^c	1.5 $\pm$ 0.07 ^a	1.4 $\pm$ 0.07 ^a	1.9 ^{n.s.}	26 E ⁻¹ ***	2.8 [*]
P									
BE	-	0.44 $\pm$ 0.05 ^d	0.12 $\pm$ 0.01 ^b	0.26 $\pm$ 0.01 ^c	0.06 $\pm$ 0.01 ^a	-	0.43 ^{n.s.}	20 E ⁻¹ ***	0.18 ^{n.s.}
DG	1.1 $\pm$ 0.06 ^f	0.55 $\pm$ 0.03 ^e	0.14 $\pm$ 0.01 ^c	0.26 $\pm$ 0.02 ^d	0.04 $\pm$ 0.00 ^b	0.01 $\pm$ 0.00 ^a	0.10 ^{n.s.}	54 E ⁻¹ ***	0.70 ^{n.s.}
SP	1.2 $\pm$ 0.06 ^f	0.57 $\pm$ 0.04 ^e	0.14 $\pm$ 0.01 ^c	0.29 $\pm$ 0.02 ^d	0.05 $\pm$ 0.00 ^b	0.00 $\pm$ 0.00 ^a	3.5 ^{n.s.}	35 E ⁻¹ ***	3.2 ^{**}
S									
BE	-	0.46 $\pm$ 0.02 ^d	0.18 $\pm$ 0.01 ^b	0.37 $\pm$ 0.01 ^c	0.09 $\pm$ 0.00 ^a	-	1.3 ^{n.s.}	54 E ⁻¹ ***	0.53 ^{n.s.}
DG	1.5 $\pm$ 0.07 ^e	0.51 $\pm$ 0.02 ^d	0.15 $\pm$ 0.01 ^b	0.32 $\pm$ 0.01 ^c	0.05 $\pm$ 0.00 ^a	0.05 $\pm$ 0.00 ^a	2.9 ^{n.s.}	30 E ⁻¹ ***	1.3 ^{n.s.}
SP	1.2 $\pm$ 0.08 ^f	0.61 $\pm$ 0.03 ^e	0.17 $\pm$ 0.02 ^c	0.40 $\pm$ 0.01 ^d	0.07 $\pm$ 0.00 ^b	0.05 $\pm$ 0.00 ^a	0.13 ^{n.s.}	57 E ⁻¹ ***	2.1 ^{n.s.}
K									
BE	-	1.9 $\pm$ 0.08 ^b	1.2 $\pm$ 0.06 ^a	1.8 $\pm$ 0.09 ^b	1.05 $\pm$ 0.07 ^a	-	0.89 ^{n.s.}	53 ^{***}	0.81 ^{n.s.}
DG	4.2 $\pm$ 0.28 ^f	2.2 $\pm$ 0.10 ^e	0.88 $\pm$ 0.06 ^c	1.4 $\pm$ 0.09 ^d	0.37 $\pm$ 0.02 ^b	0.01 $\pm$ 0.01 ^a	0.38 ^{n.s.}	43 E ⁻¹ ***	1.1 ^{n.s.}
SP	4.7 $\pm$ 0.27 ^f	2.8 $\pm$ 0.14 ^e	0.90 $\pm$ 0.05 ^c	1.5 $\pm$ 0.08 ^d	0.58 $\pm$ 0.02 ^b	0.27 $\pm$ 0.01 ^a	3.2 ^{n.s.}	61 E ⁻¹ ***	2.4 ^{n.s.}
Ca									
BE	-	3.4 $\pm$ 0.45 ^c	2.4 $\pm$ 0.35 ^b	18 $\pm$ 2.5 ^d	0.74 $\pm$ 0.03 ^a	-	0.43 ^{n.s.}	67 E ⁻¹ ***	1.4 ^{n.s.}
DG	4.8 $\pm$ 0.82 ^e	3.8 $\pm$ 0.41 ^e	0.86 $\pm$ 0.06 ^c	2.1 $\pm$ 0.29 ^d	0.37 $\pm$ 0.02 ^b	0.12 $\pm$ 0.01 ^a	0.26 ^{n.s.}	56 E ⁻¹ ***	2.1 ^{n.s.}
SP	2.5 $\pm$ 0.12 ^d	2.1 $\pm$ 0.10 ^d	1.2 $\pm$ 0.05 ^c	4.4 $\pm$ 0.31 ^e	0.57 $\pm$ 0.02 ^a	0.83 $\pm$ 0.03 ^b	20 ^{***}	43 E ⁻¹ ***	1.9 ^{n.s.}
Mg									
BE	-	0.43 $\pm$ 0.03 ^b	0.31 $\pm$ 0.03 ^a	0.46 $\pm$ 0.03 ^c	0.29 $\pm$ 0.04 ^a	-	0.10 ^{n.s.}	21 ^{***}	0.34 ^{n.s.}
DG	1.3 $\pm$ 0.08 ^f	0.56 $\pm$ 0.02 ^e	0.20 $\pm$ 0.01 ^c	0.35 $\pm$ 0.02 ^d	0.09 $\pm$ 0.00 ^b	0.01 $\pm$ 0.00 ^a	1.1 ^{n.s.}	36 E ⁻¹ ***	0.77 ^{n.s.}
SP	0.68 $\pm$ 0.03 ^d	0.61 $\pm$ 0.03 ^c	0.30 $\pm$ 0.01 ^b	0.48 $\pm$ 0.03 ^c	0.18 $\pm$ 0.01 ^a	0.19 $\pm$ 0.01 ^a	4.0 ^{n.s.}	39 E ⁻¹ ***	3.8 ^{***}
Mn									
BE	-	140 $\pm$ 19 ^c	75 $\pm$ 9.0 ^b	230 $\pm$ 23 ^d	43 $\pm$ 4.4 ^a	-	3.1 ^{n.s.}	49 E ⁻¹ ***	1.3 ^{n.s.}
DG	450 $\pm$ 76 ^e	150 $\pm$ 17 ^d	62 $\pm$ 9.5 ^c	80 $\pm$ 9.7 ^c	27 $\pm$ 4.7 ^b	6.2 $\pm$ 1.1 ^a	76 ^{***}	38 E ⁻¹ ***	1.4 ^{n.s.}
SP	210 $\pm$ 21 ^d	87 $\pm$ 8.7 ^c	48 $\pm$ 4.8 ^b	75 $\pm$ 7.3 ^c	34 $\pm$ 3.5 ^a	48 $\pm$ 5.5 ^b	2.1 ^{n.s.}	77 E ⁻¹ ***	0.85 ^{n.s.}
Cu									
BE	-	4.2 $\pm$ 0.24 ^b	3.5 $\pm$ 0.9 ^b	3.6 $\pm$ 0.17 ^b	1.2 $\pm$ 0.09 ^a	-	1.8 ^{n.s.}	13 E ⁻¹ ***	0.48 ^{n.s.}
DG	4.6 $\pm$ 0.18 ^c	6.9 $\pm$ 0.66 ^d	3.3 $\pm$ 0.38 ^b	4.3 $\pm$ 0.29 ^c	3.1 $\pm$ 1.5 ^{abc}	1.0 $\pm$ 0.17 ^a	1.2 ^{n.s.}	40 ^{***}	1.1 ^{n.s.}
SP	4.5 $\pm$ 0.43 ^{bc}	5.2 $\pm$ 0.26 ^c	5.3 $\pm$ 1.1 ^{abc}	3.1 $\pm$ 0.11 ^a	2.6 $\pm$ 0.50 ^{ab}	3.7 $\pm$ 0.99 ^{abc}	1.2 ^{n.s.}	23 ^{***}	1.5 ^{n.s.}
Zn									
BE	-	28 $\pm$ 3.1 ^c	10 $\pm$ 1.7 ^{ab}	16 $\pm$ 3.6 ^b	6.5 $\pm$ 1.0 ^a	-	1.4 ^{n.s.}	52 ^{***}	1.4 ^{n.s.}
DG	78 $\pm$ 33 ^c	120 $\pm$ 42 ^d	21 $\pm$ 4.8 ^b	68 $\pm$ 24 ^c	10 $\pm$ 2.8 ^a	9.9 $\pm$ 3.3 ^a	0.46 ^{n.s.}	15 E ⁻¹ ***	0.91 ^{n.s.}
SP	69 $\pm$ 9.7 ^c	64 $\pm$ 14 ^c	25 $\pm$ 2.9 ^b	47 $\pm$ 6.3 ^c	15 $\pm$ 2.3 ^a	17 $\pm$ 1.7 ^{ab}	1.3 ^{n.s.}	11 E ⁻¹ ***	1.6 ^{n.s.}
Fe									
BE	-	55 $\pm$ 6.5 ^c	19 $\pm$ 6.3 ^b	37 $\pm$ 3.3 ^c	5.3 $\pm$ 2.3 ^a	-	2.5 ^{n.s.}	10 E ⁻¹ ***	1.3 ^{n.s.}
DG	140 $\pm$ 12 ^d	150 $\pm$ 24 ^d	15 $\pm$ 2.0 ^b	41 $\pm$ 4.1 ^c	3.3 $\pm$ 0.36 ^a	3.5 $\pm$ 0.58 ^a	2.7 ^{n.s.}	58 E ⁻¹ ***	2.2 ^{n.s.}
SP	80 $\pm$ 6.5 ^d	95 $\pm$ 10 ^d	16 $\pm$ 4.1 ^b	30 $\pm$ 2.3 ^c	5.9 $\pm$ 2.2 ^a	3.3 $\pm$ 0.93 ^a	4.5 ^{n.s.}	13 E ⁻¹ ***	0.68 ^{n.s.}

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*** $P \leq 0.001$, ** $0.005 \leq P < 0.01$ * $0.01 \leq P < 0.05$, ^{n.s.} $P > 0.01$. Different letters denote significant differences among canopy positions according to Tukey's *posthoc* test with a significance level of $P < 0.05$. The statistical model used was $Y_{it} = \mu + \alpha_i + B_{t(i)} + \varepsilon_{iz}$ where α is the fixed effect of canopy position, β the fixed effect of tree compartment (t) and ε the residual error within tree (i) and site (z).

Table S4.6 Mean values $\pm$ standard error ($n = 15$) of the nutrient mass distribution of N, P, S, K, Ca, Mg, Mn, Cu, Zn and Fe (%) within needles (NE), small branches (SB), coarse branches (CB), stem bark (BA), stem sapwood (SW) and heartwood (HW) for beech (BE), Douglas fir (DG) and Scots pine (SP). The F value and significance of the two-way Anova based on canopy position (CP) and tree compartment (TC) and their interaction (Int) are displayed.

	Tree compartment						Anova		
	NE	SB	CB	BA	SW	HW	CP	TC	Int
N									
BE	-	14 $\pm$ 1.4 ^a	27 $\pm$ 3.1 ^b	9.7 $\pm$ 0.61 ^a	49 $\pm$ 2.9 ^c	-	0.37	12 E ⁻¹ ***	0.49 n.s.
DG	23 $\pm$ 3.1 ^{ab}	10 $\pm$ 1.5 ^a	9.9 $\pm$ 1.4 ^a	20 $\pm$ 1.2 ^b	31 $\pm$ 2.0 ^{bc}	28 $\pm$ 1.8 ^{bc}	2.4 n.s.	28 ***	1.9 n.s.
SP	29 $\pm$ 2.3 ^d	24 $\pm$ 1.9 ^{cd}	15 $\pm$ 1.7 ^{bc}	14 $\pm$ 0.84 ^b	38 $\pm$ 2.6 ^d	7.4 $\pm$ 0.95 ^a	1.4 n.s.	48 ***	1.9 n.s.
P									
BE	-	20 $\pm$ 1.9 ^b	27 $\pm$ 2.4 ^b	11 $\pm$ 0.69 ^a	42 $\pm$ 3.0 ^c	-	0.21	58 ***	0.63 n.s.
DG	27 $\pm$ 2.0 ^{cd}	21 $\pm$ 2.3 ^{bc}	15 $\pm$ 1.7 ^b	33 $\pm$ 1.7 ^d	24 $\pm$ 2.0 ^{bcd}	7.0 $\pm$ 0.69 ^a	0.17	58 ***	1.4 n.s.
SP	41 $\pm$ 3.7 ^e	36 $\pm$ 2.3 ^e	17 $\pm$ 1.3 ^b	18 $\pm$ 0.95 ^b	30 $\pm$ 1.9 ^c	0.20 $\pm$ 0.20 ^a	0.37	34 E ⁻¹ ***	3.3 **
S									
BE	-	16 $\pm$ 1.6 ^{ab}	29 $\pm$ 2.9 ^b	11 $\pm$ 0.62 ^a	44 $\pm$ 2.8 ^c	-	0.38	72 ***	0.58 n.s.
DG	30 $\pm$ 3.4 ^{bcd}	15 $\pm$ 1.7 ^{ab}	12 $\pm$ 1.3 ^a	32 $\pm$ 1.3 ^c	21 $\pm$ 1.3 ^b	21 $\pm$ 1.0 ^b	0.29	30 ***	1.8 n.s.
SP	32 $\pm$ 3.6 ^{cd}	30 $\pm$ 1.9 ^{cd}	16 $\pm$ 1.3 ^b	19 $\pm$ 0.85 ^{bc}	31 $\pm$ 1.5 ^c	4.5 $\pm$ 0.50 ^a	0.33	66 ***	2.8 **
K									
BE	-	8.6 $\pm$ 1.1 ^a	25 $\pm$ 3.1 ^b	6.3 $\pm$ 0.37 ^a	60 $\pm$ 3.6 ^c	-	0.12	18 E ⁻¹ ***	0.83 n.s.
DG	18 $\pm$ 1.8 ^b	15 $\pm$ 1.8 ^b	16 $\pm$ 1.6 ^b	31 $\pm$ 1.3 ^c	36 $\pm$ 2.6 ^c	1.4 $\pm$ 0.95 ^a	1.3 n.s.	82 ***	2.0 n.s.
SP	21 $\pm$ 2.3 ^c	24 $\pm$ 2.0 ^c	15 $\pm$ 1.4 ^{bc}	12 $\pm$ 0.64 ^b	44 $\pm$ 2.2 ^d	4.3 $\pm$ 0.60 ^a	0.18	12 E ⁻¹ ***	3.6 ***
Ca									
BE	-	8.8 $\pm$ 1.0 ^a	27 $\pm$ 2.8 ^b	37 $\pm$ 2.6 ^b	27 $\pm$ 2.2 ^b	-	0.55	60 ***	0.37 n.s.
DG	14 $\pm$ 2.1 ^{ab}	19 $\pm$ 2.3 ^{bc}	12 $\pm$ 1.2 ^{ab}	32 $\pm$ 1.8 ^c	28 $\pm$ 2.8 ^c	9.4 $\pm$ 0.98 ^a	0.43	35 ***	1.6 n.s.
SP	9.0 $\pm$ 1.2 ^a	14 $\pm$ 1.3 ^{ab}	15 $\pm$ 1.5 ^b	27 $\pm$ 1.5 ^c	34 $\pm$ 2.2 ^c	10 $\pm$ 1.1 ^{ab}	0.03	48 ***	2.6 n.s.
Mg									
BE	-	8.1 $\pm$ 1.1 ^a	25 $\pm$ 3.2 ^b	6.8 $\pm$ 0.64 ^a	60 $\pm$ 3.7 ^c	-	0.49	11 E ⁻¹ ***	0.47 n.s.
DG	22 $\pm$ 2.2 ^{bc}	15 $\pm$ 1.8 ^b	14 $\pm$ 1.6 ^b	31 $\pm$ 1.0 ^{cd}	35 $\pm$ 2.0 ^d	4.7 $\pm$ 0.75 ^a	0.75	10 E ⁻¹ ***	1.3 n.s.
SP	9.9 $\pm$ 0.92 ^a	17 $\pm$ 1.4 ^b	17 $\pm$ 1.8 ^{ab}	13 $\pm$ 0.59 ^{ab}	44 $\pm$ 2.3 ^c	10 $\pm$ 1.2 ^{ab}	0.42	76 ***	2.9 **
Mn									
BE	-	11 $\pm$ 1.2 ^a	27 $\pm$ 3.0 ^b	15 $\pm$ 1.0 ^b	46 $\pm$ 2.6 ^c	-	0.07	13 E ⁻¹ ***	0.66 n.s.
DG	26 $\pm$ 3.4 ^{bcd}	15 $\pm$ 2.0 ^{ab}	15 $\pm$ 1.6 ^{ab}	26 $\pm$ 1.2 ^c	35 $\pm$ 2.7 ^c	8.8 $\pm$ 0.95 ^a	0.55	43 ***	1.3 n.s.
SP	17 $\pm$ 1.8 ^a	14 $\pm$ 1.2 ^a	15 $\pm$ 1.8 ^a	11 $\pm$ 0.64 ^a	47 $\pm$ 2.2 ^b	14 $\pm$ 1.7 ^a	0.07	66 ***	1.7 n.s.
Cu									
BE	-	12 $\pm$ 1.6 ^{ab}	36 $\pm$ 4.8 ^{bc}	8.7 $\pm$ 0.89 ^a	44 $\pm$ 3.7 ^c	-	0.73	40 ***	0.80 n.s.
DG	4.7 $\pm$ 0.85 ^a	11 $\pm$ 2.2 ^b	13 $\pm$ 2.1 ^b	22 $\pm$ 2.6 ^{cd}	34 $\pm$ 5.2 ^{bc}	20 $\pm$ 2.2 ^{bc}	2.5 n.s.	49 ***	1.6 n.s.
SP	5.9 $\pm$ 0.91 ^a	14 $\pm$ 2.0 ^{ab}	21 $\pm$ 2.1 ^b	8.3 $\pm$ 1.4 ^a	46 $\pm$ 4.1 ^c	11 $\pm$ 2.6 ^{ab}	0.04	50 ***	1.7 n.s.
Zn									
BE	-	18 $\pm$ 2.2 ^b	26 $\pm$ 2.9 ^b	7.2 $\pm$ 0.78 ^a	49 $\pm$ 3.1 ^c	-	1.1 n.s.	62 ***	0.56 n.s.
DG	5.4 $\pm$ 0.95 ^a	17 $\pm$ 2.4 ^{bc}	10 $\pm$ 1.4 ^a	30 $\pm$ 2.4 ^{cd}	24 $\pm$ 3.4 ^c	20 $\pm$ 2.8 ^{bc}	0.38	23 ***	1.3 n.s.
SP	11 $\pm$ 1.1 ^a	19 $\pm$ 2.4 ^a	16 $\pm$ 1.9 ^a	14 $\pm$ 0.86 ^a	42 $\pm$ 3.0 ^b	9.8 $\pm$ 1.2 ^a	3.6 n.s.	32 ***	1.8 n.s.
Fe									
BE	-	27 $\pm$ 3.5 ^a	32 $\pm$ 5.2 ^a	15 $\pm$ 1.8 ^a	26 $\pm$ 4.8 ^a	-	1.0 n.s.	5.3 **	0.46 n.s.
DG	21 $\pm$ 2.0 ^{bcd}	32 $\pm$ 4.7 ^{cd}	10 $\pm$ 1.4 ^a	33 $\pm$ 3.0 ^d	12 $\pm$ 1.4 ^{ab}	13 $\pm$ 1.8 ^{abc}	5.8 n.s.	16 ***	1.2 n.s.
SP	23 $\pm$ 2.7 ^b	48 $\pm$ 4.0 ^c	13 $\pm$ 2.3 ^b	15 $\pm$ 1.3 ^b	21 $\pm$ 3.7 ^b	2.2 $\pm$ 0.32 ^a	0.91	11 E ⁻¹ ***	1.7 n.s.

Chapter 4

*** $P \leq 0.001$, ** $0.01 \leq P < 0.001$, * $0.05 \leq P \leq 0.01$, ^{n.s.} $P > 0.05$. Different letters denote significant differences among canopy positions according to Tukey's *posthoc* test at a significance level of $P < 0.05$. The statistical model used was $Y_{it} = \mu + \alpha_i + B_{t(i)} + \varepsilon_{iz}$ where α is the fixed effect of canopy position, β the fixed effect of tree compartment (t) and ε the residual error within tree (i) and site (z).

Table S4.7 Mean values of N, P, S, K, Ca, Mg, Mn, Cu, Fe and Zn in the different tree compartments (TC) in beech (BE), Douglas fir (DG) and Scots pine (SP). Selected data provided nutrient concentrations for needles (NE), small branches (SB), coarse branches (CB), stem bark (BA), stem sapwood (SW) and/or stem heartwood (HW).

Species	TC	N	P	S	K	Ca	Mg	Mn	Cu	Fe	Zn	Reference				
		g/kg						mg/kg								
BE	SB	8.4	0.79	0.48	3.0	6.9	0.52	200	3.9	120	43	(Husmann et al., 2018)				
	SB	8.0	0.36		2.1	3.4	0.38					(Mussche et al., 1998)				
	SB											(Caritat and Terradas, 1990)				
	SB											(Weis and Göttlein, 2002)				
	SB		0.74		2.2	4.7	0.46					(Calvaruso et al., 2017)				
	SB	5.8	0.46		1.5	2.3	0.40					(de Vries et al., 2019)				
	CB	2.8	0.24	0.15	1.6	3.2	0.37	320				(Husmann et al., 2018)				
	CB	2.8	0.21		1.1	1.7	0.22					(Mussche et al., 1998)				
	CB											(Weis and Göttlein, 2002)				
	CB		0.30		1.6	2.6	0.36					(Calvaruso et al., 2017)				
	BA	10	0.36	0.35	2.1	9.3	0.42	1800				(Andre et al., 2010)				
	BA	6.9	0.35	0.33	2.4	23	0.52					(Husmann et al., 2018)				
	BA	7.7	0.20		1.7	8.4	0.39					(Mussche et al., 1998)				
	BA	7.4	0.43		2.5	26	0.54					(Rademacher et al., 2009)				
	BA	6.1	0.39	0.84	3.0	21	0.76					1000	3	8	20	(Jönsson, 2000)
	BA															(Weis and Göttlein, 2002)
	BA		0.45		2.7	23	0.44									(Calvaruso et al., 2017)
	BA	8.4	0.39		2.0	13	0.55									(de Vries et al., 2019)
	SW	3.2	0.07	0.10	1.0	0.67	0.20	330				(Andre et al., 2010)				
	SW	1.5	0.10	0.09	1.1	0.97	0.30					(Husmann et al., 2018)				
	SW	1.7	0.05		1.1	1.4	0.21					(Mussche et al., 1998)				
	SW											140	6.9	48	28	(Caritat and Terradas, 1990)
	SW	1.5	0.11		1.1	1.2	0.29									(Rademacher et al., 2009)
	SW											79	2.0	23	4.5	(Hagen-Thorn and Stjernquist, 2005)
	SW															(Weis and Göttlein, 2002)
	SW		0.12		1.1	0.83	0.27									(Calvaruso et al., 2017)
	SW	2.0	0.09		0.93	0.75	0.25									
DG	NE	16	1.0		4.7	6.1	1.3					340				(Ponette et al., 2001)
	NE							(Vittori Antisari et al., 2018)								
	NE	9.6	4.0		6.0	7.1	1.6	(Mitchell et al., 1996)								
	NE	16	1.3		5.6	3.8	1.2	(Ranger et al., 1995)								
	NE	8.1	2.2		7.8	11		(Overton et al., 1973)								
	NE	15	1.7	1.1	6.6	4.2	0.93	100	3	75	24	(Harrison et al., 1994)				
	NE	18	1.0		5.7	3.3	1.4					(Olsthoorn et al., 2006)				

	NE	12	1.2	0.87	6.6	7.9	1.4				(Clayton and Kennedy, 1980)	
	SB	11	1.4		4.5	4.9	1.1				(Ranger et al., 1995)	
	SB	2.4	0.46		2.3	8.7					(Overton et al., 1973)	
	SB	4.3	0.35		1.9	3.3	0.54				(de Vries et al., 2019)	
	SB	8.7	0.61		1.7	7.5	0.56				(Clayton and Kennedy, 1980)	
	CB	4.2	0.5		2.1	4.6	0.4				(Ranger et al., 1995)	
	CB	4.0	0.19	0.12	0.71	5.9	0.26				(Clayton and Kennedy, 1980)	
	BA	3.4	0.3		1.4	2.6	0.27				(Ponette et al., 2001)	
	BA	2.3	0.36	0.09	1.4	2.6	0.35	58	27	19	(Schowalter and Morrell, 2002)	
	BA	2.1	0.49		2.8	2.0	0.44				(Mitchell et al., 1996)	
	BA	3.3	0.42	0.36	2.2	3.1	0.32	390			(Ranger et al., 1995)	
	BA	3.6	0.17		0.95	1.6	0.25				(de Vries et al., 2019)	
	BA	6.8	0.31	0.13	1.2	14	0.38				(Clayton and Kennedy, 1980)	
	SW	0.55	0.06	0.03	0.35	0.38	0.09	22	7.0	6.0	(Schowalter and Morrell, 2002)	
	SW	1.3	0.06		0.56	0.34	0.10				(de Vries et al., 2019)	
	SW	3.4		0.12	0.36	0.56	0.07				(Clayton and Kennedy, 1980)	
	HW	0.48	0.01	0.02	0.05	0.18	0.02	9.3	6.3	5.3	(Schowalter and Morrell, 2002)	
	HW	1.4	0.01		0.23	0.23	0.04				(de Vries et al., 2019)	
	HW	2.7		0.10	0.13	0.26	0.03				(Clayton and Kennedy, 1980)	
SP	NE	13	1.4	0.95	3.7	2.8	0.95				(Armolaitis et al., 2013)	
	NE	12	1.2	1.3	4.5	3.2	0.65				(Węgiel et al., 2018)	
	NE	11	1.3		4.5	2.1	0.89	500	2.0	24	40	(Saarsalmi et al., 2006)
	NE							1100	2.7	34	47	(Węgiel et al., 2019)
	NE							380		60	30	(Varnagirytė-Kabašinskienė et al., 2014)
	NE	16	1.4		6.1	4.0	0.74					(Knust et al., 2016)
	NE				4.4	3.4	0.53	250				(Gielen et al., 2016)
	SB	7.1	0.78	0.73	2.7	2.3	0.63					(Węgiel et al., 2018)
	SB							340	3.6	33	45	(Węgiel et al., 2019)
	SB				3.4	2.3	0.66	90				(Gielen et al., 2016)
	SB	5.4	0.34		1.9	2.0	0.50					(de Vries et al., 2019)
	CB	2.5	0.33	0.25	1.2	2.1	0.42					(Węgiel et al., 2018)
	CB							320	1.5	23	35	(Węgiel et al., 2019)
	CB	2.0	0.17		0.9	1.3	0.35					(Knust et al., 2016)
	BA	4.2	0.5	0.58	1.4	6.5	0.68					(Armolaitis et al., 2013)

	BA	4.9	0.62	0.43	1.5	7.9	0.99	650 2.4 22 67 160 44 29 31 3.5 33 15 432 3.4 162 45				(Węgiel et al., 2018)
	BA											(Wegiel et al., 2019)
	BA											(Varnagirytė-Kabašinskiene et al., 2014)
	BA	2.8	0.26		1.3	6.9	0.46					(Knust et al., 2016)
	BA	5.6	0.11		0.60	2.6	0.15					(de Vries et al., 2019)
	BA		0.38	0.35	1.6	7.2						(Saarela et al., 2005)
	BA		0.75	0.85	3.2	13					(Saarela et al., 2005)	
	SW	1.1	0.1		0.74	0.58	0.24	30				(Wright and Will, 1958)
	SW	0.79	0.07		0.47	0.80	0.11					(Häsänen and Huttunen, 1989)
	SW	0.47	0.06		0.3	0.5	0.15					(Helmisaari and Siltala, 1989)
	SW				0.2	0.57	0.16					(Gielen et al., 2016)
	SW	1.4	0.07		0.49	0.62	0.16					(de Vries et al., 2019)
	HW	0.64	0.02		0.13	0.71	0.23	41				(Wright and Will, 1958)
	HW	0.6	0.03		0.39	0.97	0.14					(Häsänen and Huttunen, 1989)
	HW	0.4	0.00		0.00	0.60	0.15					(Helmisaari and Siltala, 1989)
	HW				0.07	0.81	0.13					(Gielen et al., 2016)
	HW	1.3	0.01		0.27	0.79	0.17					(de Vries et al., 2019)

Table S4.8 Average nutrient concentrations of Mn, Cu, Fe and Zn (mg/kg) in literature and the difference (%) with values observed in this study. Percentage of difference (Diff) is calculated as the mean concentration of this study divided by the mean concentration in literature minus 100%. Difference of $\geq 20\%$ are highlighted. The reference studies used (n: number of studies) are in Table S4.6.

		Beech		Douglas fir			Scots pine		
		Mean		Mean		Diff	Mean		Diff
		n	mg/kg	n	mg/kg	%	n	mg/kg	%
Mn	NE			2	220	100	4	560	-63
	SB	2	450	0	n.d.	n.d.	2	220	-60
	CB	1	320	0	n.d.	n.d.	1	320	-85
	BA	3	1300	2	220	-64	4	320	-76
	SW	4	180	1	22	24	1	30	15
	HW			1	9.3	-34	1	41	17
Cu	NE			2	2.5	88	2	2.4	93
	SB	1	3.9	0	n.d.	n.d.	1	3.6	44
	CB	0	n.d.	0	n.d.	n.d.	1	1.5	260
	BA	1	3.0	0	n.d.	n.d.	3	3.1	0.1
	SW	2	4.5	0	n.d.	n.d.	0	n.d.	n.d.
	HW			0	n.d.	n.d.	0	n.d.	n.d.
Fe	NE			2	160	-10	3	39	110
	SB	1	120	0	n.d.	n.d.	1	33	190
	CB	0	n.d.	0	n.d.	n.d.	1	23	-32
	BA	1	8	1	27	51	4	65	-56
	SW	2	35	1	7.0	-53	0	n.d.	n.d.
	HW			1	6.3	-44	0	n.d.	n.d.
Zn	NE			1	24	230	3	39	75
	SB	1	43	0	n.d.	n.d.	1	45	43
	CB	0	n.d.	0	n.d.	n.d.	1	35	-29
	BA	1	20	1	19	260	3	42	11
	SW	2	16	1	6.0	70	0	n.d.	n.d.
	HW			1	5.3	86	0	n.d.	n.d.

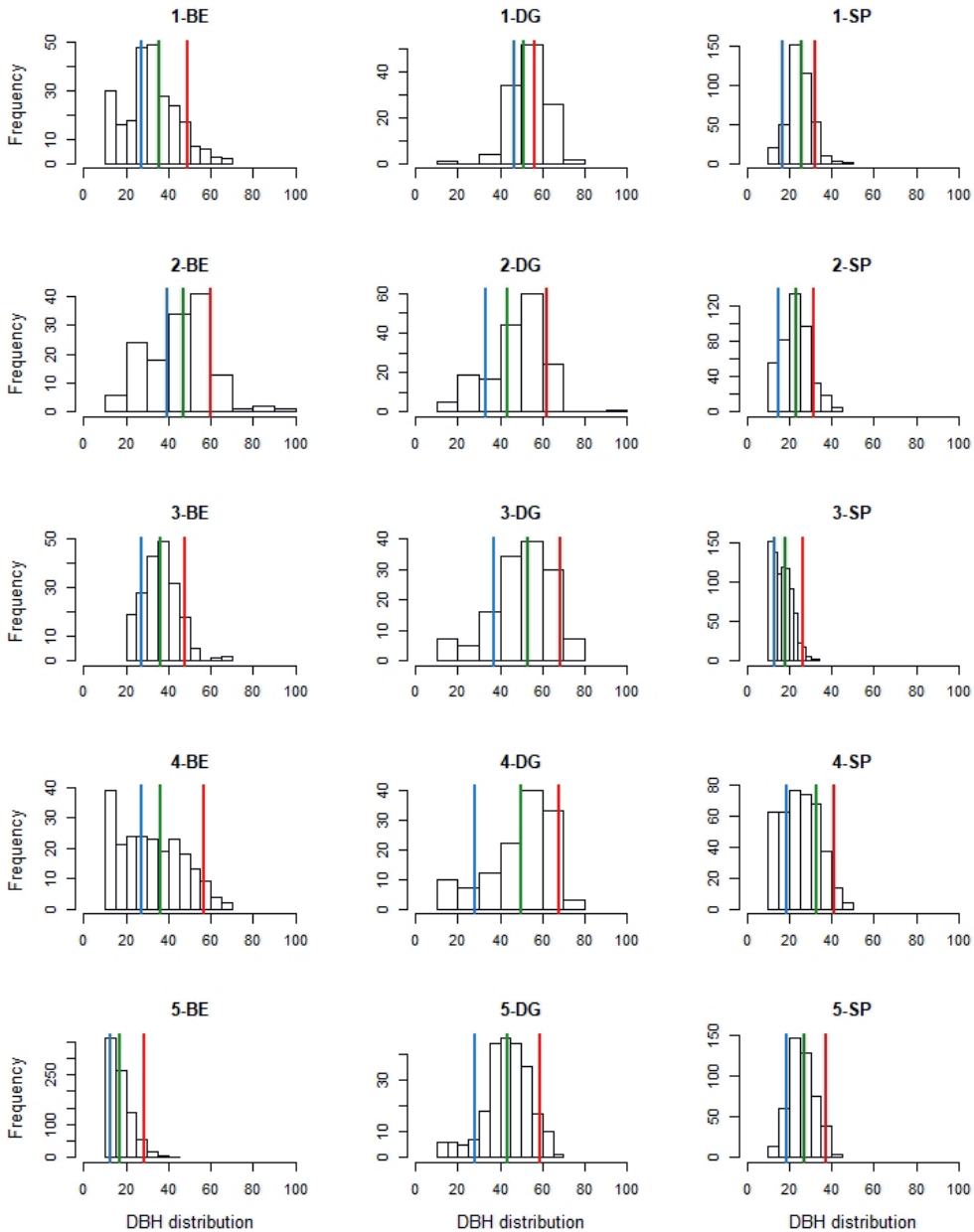
Supplementary figures

Figure S4.1 Frequency (number of trees per ha) of DBH (in cm) per study site. The letters refer to the location of the stand in the Netherlands (Fig.1) of the beech (BE), Douglas fir (DG) and Scots pine (SP) sites. Vertical lines indicate the DBH of the trees sampled for biomass, carbon and nutrients. Red lines represent the DBH of the selected dominant tree, green of the selected intermediate tree and blue for the selected suppressed tree. Diameter distribution per site is derived by DBH measurement of all trees within the 1-ha plot.



Figure S4.2 Separation of the bark, sapwood and heartwood (left) and the extraction of sapwood and heartwood pieces for the fresh weight to dry weight conversion and the density measurements.

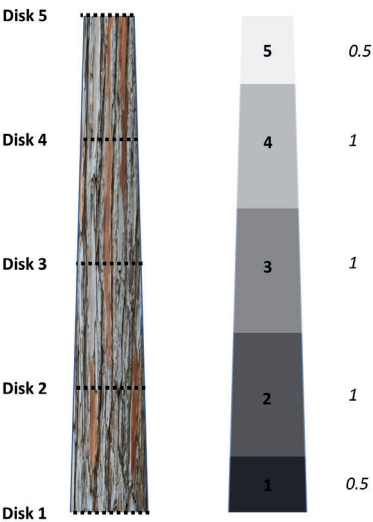


Figure S4.3 Overview of the relative weight of the nutrient samples based on disk locations and the stem parts represented by the disks. The numbers (right) denote the length of the stem bole represented by a disk whereby section 1 and 5 represent only half the length of the other stem segments.

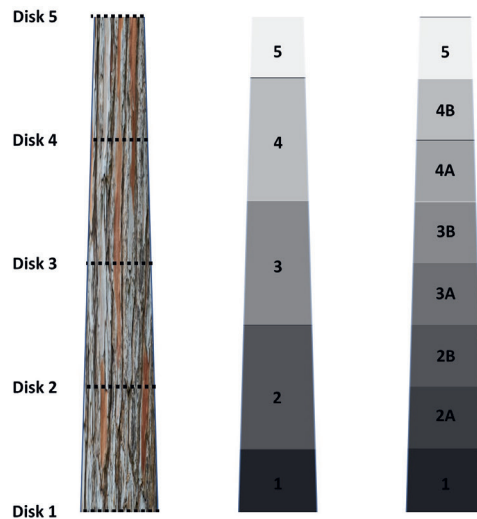


Figure S4.4 Overview of stem disks and the part of the stem represented by the disks. Left: location of the stem disks. Middle: part of the stem represented per disk. The numbers denote the different stem sections. Right: the stem is further divided in smaller pieces with an equal stem segment length.



Figure S4.5 Visualization of stem ramification for one of the beech trees prior to felling. In red the stem, defined as the main axis extending from the butt until the point where the stem diameter reached 10 cm. In blue the stem ramifications. These stem ramifications were classified as branches when the diameter reached 10 cm.

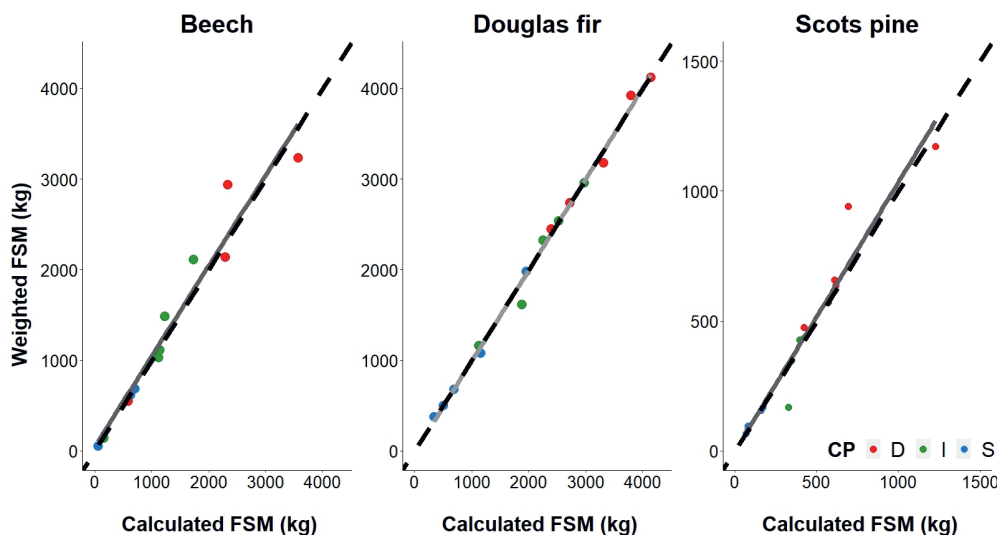


Figure S4.6 Comparison between weighted fresh stem mass (FSM) and the calculated FSM for beech, Douglas fir and Scots pine. The relation between the weighted and calculated FSM (grey line) equals almost the 1:1 line (black dashed line) and is highly significant (Table S4.2).

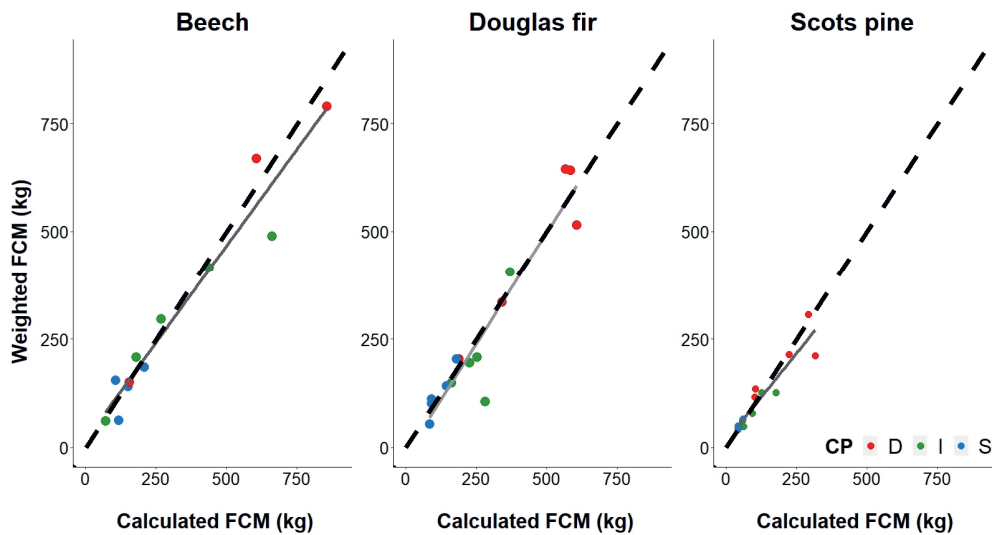


Figure S4.7 Comparison between weighted fresh crown mass (FCM) and the calculated FCM for beech, Douglas fir and Scots pine. The relation between the weighted and calculated FSM (grey line) equals almost the 1:1 line (black dashed line) and is highly significant (Table S4.2).

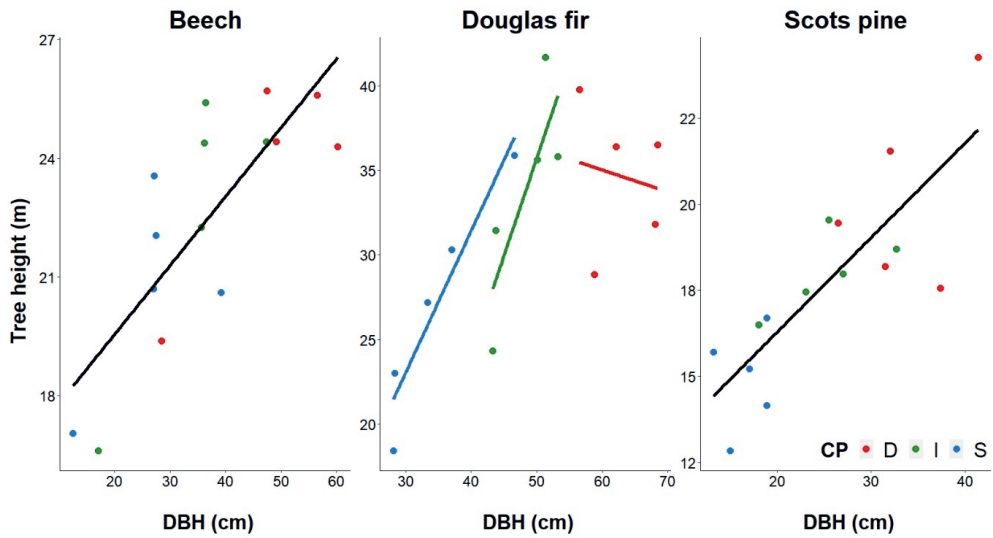


Figure S4.8 Diameter breast height (DBH) to tree height relation for dominant (D), intermediate (I) and suppressed (S) trees of beech, Douglas fir and Scots pine. Solid lines show significant models. Impact of canopy position on the DBH tree height relationship is only significant (< 0.05) for Douglas fir.

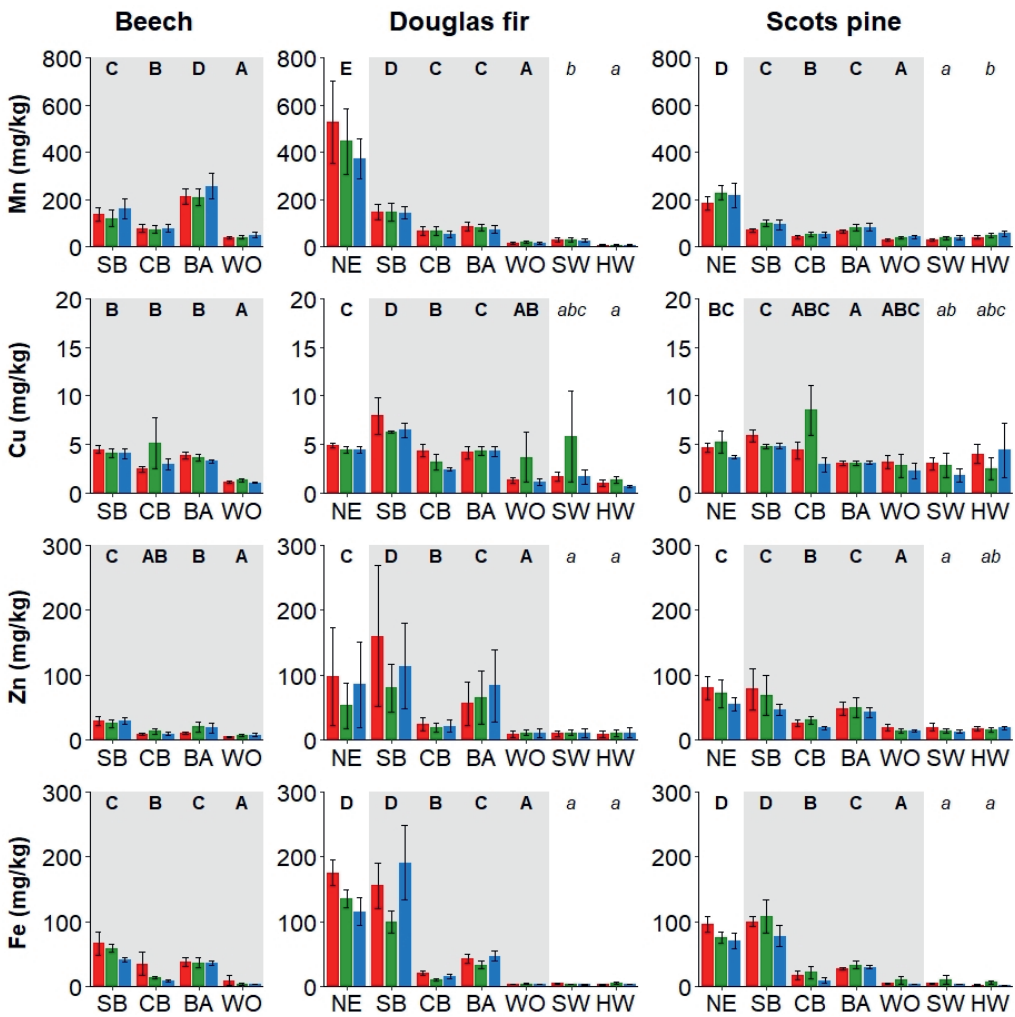


Figure S4.9 Concentrations of micronutrients in needles (NE), small branches (SB), coarse branches (CB), stem bark (BA), stem wood (WO), and for both conifers separately stem sapwood (SW) and stem heartwood (HW), for dominant (red bars), intermediate (green bars) and suppressed (blue bars) trees of beech, Douglas fir and Scots pine. Error bars indicate standard error from the mean value ($n = 5$). Different capital letters indicate significant differences ($p < 0.01$) among NE, SB, CB, BA and WO, and small letters among sapwood (SW) and heartwood (HW) of Douglas fir and Scots pine. All interactions between canopy position and tree compartments were insignificant ($p > 0.01$).

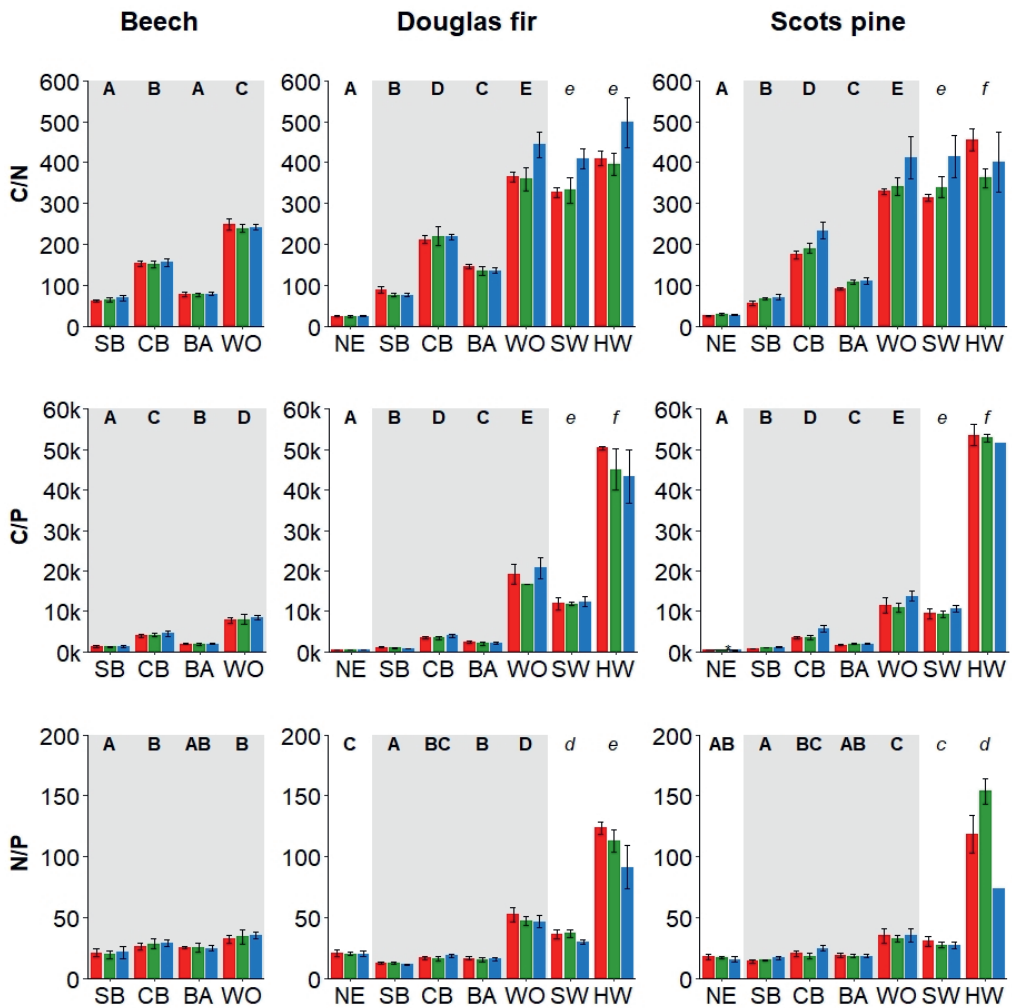


Figure S4.10 C/N, C/P and N/P ratios in needles (NE), small branches (SB), coarse branches (CB), stem bark (BA), stem wood (WO), and for both conifers separately stem sapwood (SW) and stem heartwood (HW), for dominant (red bars), intermediate (green bars) and suppressed (blue bars) trees of beech, Douglas fir and Scots pine. Error bars indicate standard error from the mean value ($n=5$). Different capital letters indicate significant differences ($p < 0.01$) among NE, SB, CB, BA and WO, and small letters among sapwood (SW) and heartwood (HW) of Douglas fir and Scots pine. All interactions between canopy position and tree compartments were insignificant ($p > 0.01$).

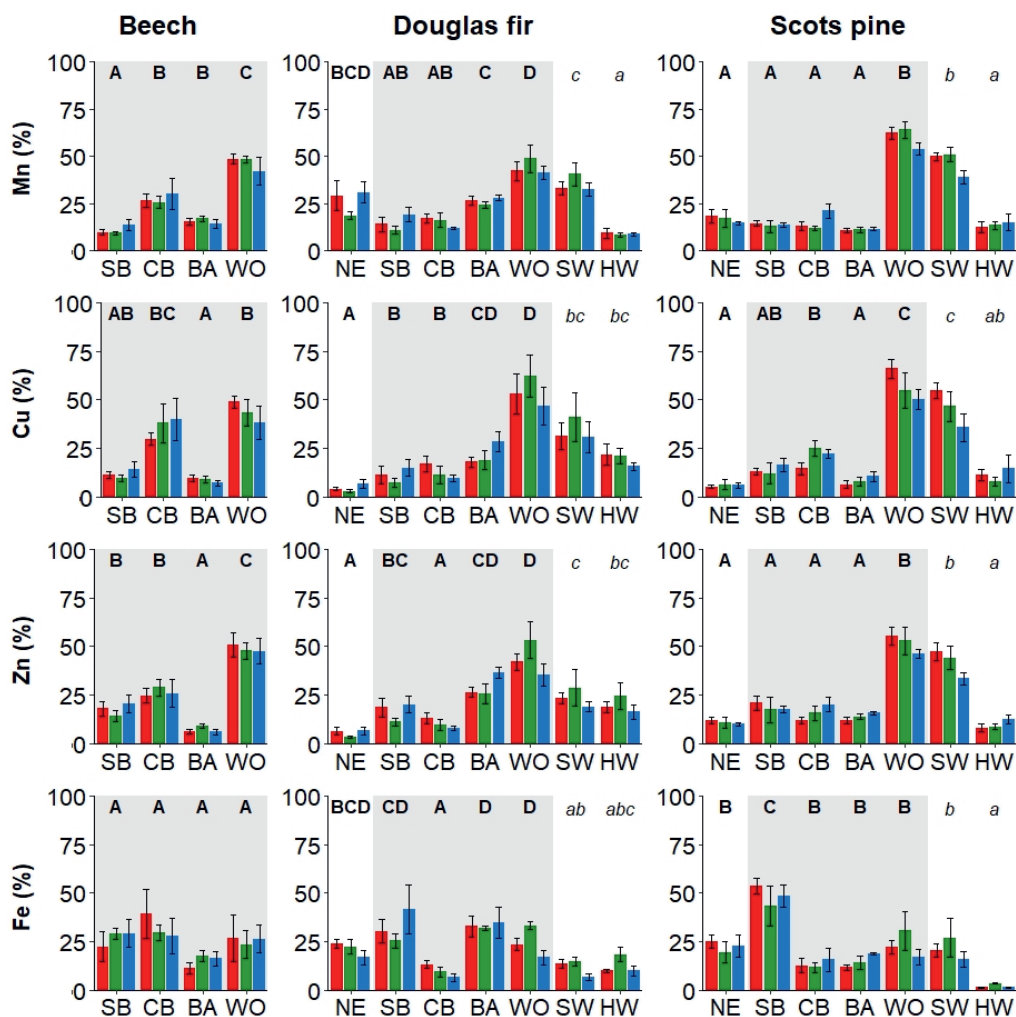


Figure S4.11 Mass fractions of macronutrients in needles (NE), small branches (SB), coarse branches (CB), stem bark (BA), stem wood (WO), and for both conifers separately stem sapwood (SW) and stem heartwood (HW), for dominant (red bars), intermediate (green bars) and suppressed (blue bars) trees of beech, Douglas fir and Scots pine. Error bars indicate standard error from the mean value ($n = 5$). Different capital letters indicate significant differences ($p < 0.01$) among NE, SB, CB, BA and WO, and small letters among sapwood (SW) and heartwood (HW) of Douglas fir and Scots pine. All interactions between canopy position and tree compartments were insignificant ($p > 0.01$).



Chapter 5

The sustainability of timber and
biomass harvest in perspective of forest
nutrient uptake and nutrient stocks

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*Forest ecology and management (2023),
Volume 530, 15 February 2023*

Abstract

The sustainability of tree harvest is questioned since harvest results in increased nutrient losses which may reduce nutrient stocks in forest soils, particularly in forests on acidified and poor soils with low base saturation. We used a new forest experiment to quantify nutrient stocks and nutrient uptake rates in mature forest stands, and to assess the forest nutrient balance in relation to different forest management scenarios: clearcut, shelterwood and thinning; and whole-tree harvest (WTH), stem-only harvest (SOH) and wood only harvest (WOH, with on-site bark stripping). Forests were dominated by trees of *Fagus sylvatica*, *Pseudotsuga Menziesii* or *Pinus sylvestris*, all situated on poor, acidified soils.

We measured forest biomass and nutrient stocks based on destructive sampling of fifteen mature trees per species and by using new, calibrated allometric relationships. Aboveground stocks of N, P, S, K, Ca, Mg, Mn, Cu, Fe and Zn were calculated for foliage, branches, stem bark and stem wood. Annual forest growth and nutrient uptake were determined using tree ring measures and allometric relationships. Organic layer nutrient stocks and available nutrients in the mineral soil were determined following intensive soil sampling.

Stands of beech, Douglas fir and Scots pine differed in aboveground biomass and nutrient stocks, with highest biomass stocks in Douglas fir. However, beech stands had the highest aboveground nutrient stocks, nutrient uptake rates and nutrient losses following harvest, followed by Scots pine. Organic layer nutrient stocks generally exceeded aboveground nutrient stocks, except for the base cations and Mn. Compared to SOH, WTH increased nutrient export between 66% (Douglas fir) up to 100% (Scots pine), while WOH decreased the nutrient export between 23% (beech) up to 41% (Douglas fir).

High aboveground base cation and Mn stocks indicate potential long-term threats to forest nutrition if trees are harvested. However, in Douglas fir stands, nutrient losses through SOH may fully recover when using rotation periods of 80 years. Contrary, negative Ca balances are predicted when applying SOH in beech and Scots pine, since Ca stocks are potentially depleted within 2 final fellings. WTH poses, regardless of the species, potential threats for sustainable biomass harvest as nutrients cannot be recovered using common rotation periods. WOH conserves nutrients within the forest posing opportunities for sustainable biomass harvest. For similar temperate forest on acidified, sandy soils, we therefore recommend limiting tree harvest depending on the tree species, and to avoid WTH and consider WOH to better conserve critical nutrients required for long-term forest recovery.

1 Introduction

Forest biomass can play a major role in the European bioeconomy in phasing-out the use of fossil-based raw materials and products (Wolfslehner, 2020; Jonsson et al., 2021). The value of forest resources leads to an increased interest in timber harvest, and the harvest of logging residues, such as crown material and trees not suited for timber production (Ericsson and Nilsson, 2006; Verkerk et al., 2019; Kumar et al., 2020). Forest harvest is therefore expected to increase over the coming decades, whereby the demand for biomass (stem wood and branches) may exceed the sustainable supply (Börjesson et al., 2017).

The sustainability of the increased harvest of forest biomass as a renewable resource is questioned since it results in increased nutrient losses which may reduce the forest nutritional status (de Oliveira Garcia et al., 2018). Nowadays, nutritional status of many forests in Europe is already deteriorating as high N deposition and CO₂ fertilization are triggering nutrient imbalance in trees (Sardans et al., 2015; Waldner et al., 2015; Du et al., 2021). These nutrient imbalances are partly induced by increased forest growth caused by CO₂ fertilization (Jonard et al., 2015; Penuelas et al., 2020), which increases biomass export and therefore nutrient export through harvest (Achat et al., 2018a), and partly by the loss of base-cations due to soil acidification caused by N deposition (Bowman et al., 2008). Even though N deposition is slowly decreasing, recovery of the nutrient balance remains limited in European forests (Schmitz et al., 2019).

Biomass harvest has been argued to increase forest P limitation (Sardans et al., 2015; Du et al., 2021) and has the potential to result in negative balances of Ca, Mg and K (de Oliveira Garcia et al., 2018; de Vries et al., 2021). The effect, however, depends on the soil's capacity to counteract the negative effects of harvest and N deposition through internal nutrient supply. Effects of increased biomass harvest are therefore more pronounced on nutrient poor soils (Thiffault et al., 2011; de Vries et al., 2021). In these soils the increased biomass harvest, in combination with ongoing N deposition, may eventually limit forest growth.

Besides of soil type and fertility, harvest intensity and tree species composition also influence the effect of biomass harvest on the forest nutrient balance. Nutrient losses of timber harvest are higher in clearcut systems compared to shelterwood systems and selection forests. Nutrient pools have been suggested to decrease over a century after a clearcut (Richardson et al., 2017) while shelterwood systems could recover the loss of nutrients through harvest within 25 years (Carpenter et al., 2021). However, in nutrient poor systems, clearcutting decreased soil concentrations of P and Ca while other nutrients were replenished within a couple of decades (Vangansbeke et al., 2015). Selection cutting had a limited impact on the forest nutrient balance in the USA (Briggs et al., 2000), but substantial negative balances were detected following thinnings in Scots pine and Norway spruce stands in Germany (Knust et al., 2016).

The nutrient export through harvest depends on the harvested tree species. In general, harvesting of broadleaf trees results in higher nutrient exports than harvesting of coniferous species (Augusto et al., 2000; Palviainen and Finer, 2012). The effects of nutrient removal under different harvest intensities thus strongly depends on the species but also on stand age, basal area and stand productivity (Augusto et al., 2000; Soalleiro et al., 2007). The magnitude of the effect, however, is not clear. Inconsistent effects of biomass harvest are reported for both forest productivity and soil responses (Vance et al., 2018), indicating the need for species and site specific data on the effect of nutrient export through different levels of biomass harvest.

Increased harvest of forest biomass may also imply a shift from conventional stem only harvest to whole tree harvest. Whole tree harvest increases the biomass export through extraction of crown material and logging residues by up to 26% (Mantau et al., 2010). The increase in biomass removal is dependent on the tree species with e.g. 15-20% biomass gain for Scots pine (Mikšys et al., 2007) up to 60% biomass gain for European beech (Andre et al., 2010). The nutrient export, however, may increase up to 5 times for P with large differences between nutrients and species (Palviainen and Finer, 2012). Because of the higher nutrient export, whole tree harvest can result in greater soil nutrient reductions compared to stem only harvest (Clarke et al., 2021) which may cause reductions of forest productivity. However, whole tree harvest did not alter forest productivity in *Pinus radiata* stands in New Zealand (Garrett et al., 2021) and further empirical evidence for lower forest productivity is lacking. Although the sustainability of whole tree harvest is debated, especially for stands on poor and acidified soils, there is not much evidence regarding the export of nutrients, the biomass gains and the remaining forest nutrient stocks allowing for forest recovery and consequences for forest productivity.

The aim of the present paper is to experimentally quantify the nutrient stocks and annual nutrient uptake of mature forest stands of three major tree species in the Netherlands: *Fagus sylvatica*, *Pseudotsuga Menziesii* and *Pinus sylvestris* on poor and acidified soils, and compare those stocks to the nutrient export by applying different tree harvest strategies. More specifically, we (i) investigated nutrient stocks in different crown and aboveground stem parts of trees and in the organic soil layers using an intensive field and lab campaign to measure biomass and nutrient concentrations in different tree and soil parts and newly established allometric equations for upscaling those measurements to entire forest nutrient stocks and intensive soil sampling; (ii) estimated the annual nutrient uptake of a forest stand from tree ring measurements and tree nutrient concentrations; (iii) investigated the biomass and nutrient exports of different harvest intensities based on a field experiment and (iv) calculated the nutrient exports of different biomass harvest methods: stem only harvest (SOH), whole tree harvest (WTH) and wood only harvest (WOH), which differs from SOH by stripping and leaving stem bark in the forest. The results on forest nutrient budgets are discussed in view of the long-term sustainability of biomass harvest.

2 Materials and methods

2.1 Experimental design

A forest experiment was established in February and March 2019 in the Netherlands. This experiment consists of monoculture stands of European beech (*Fagus sylvatica*), Douglas fir (*Pseudotsuga menziesii*) and Scots pine (*Pinus sylvestris*) in five regions (Fig. S5.1). In each of these fifteen stands, four 0.25 ha-subplots were installed to which the harvest intensity treatments high thinning, shelterwood, clearcut, and unharvested control were randomly assigned. Harvest intensity was determined based on basal area reductions, whereby species-specific target basal areas per treatment were used (Table 5.1). All stands have a temperate maritime climate with a mean annual temperature of 10.4°C and a mean annual rainfall of 805 mm (KNMI, 2022). The stands are located on acidic sandy soils classified as Albic or Entic Podzols or Dystric Cambisols (WRB, 2015). A general description of the study sites including stand properties, soil cover and soil classification is provided in Table 5.2.

Table 5.1 Average basal area $\pm$ s.e. ($\text{m}^2 \text{ha}^{-1}$), realized target basal area ($\text{m}^2 \text{ha}^{-1}$) and basal area reduction for the timber harvest intensities high-thinning and shelterwood for European beech, Douglas fir and Scots pine. Target basal area of the clearcut was 0 for all species with a reduction of 100%. Basal area reductions per treatment per forest stand are in Table S5.1.

Species	Average BA $\text{m}^2 \text{ha}^{-1}$	High-thinning			Shelterwood			Clearcut
		Target BA $\text{m}^2 \text{ha}^{-1}$	Reduction %	$\text{m}^2 \text{ha}^{-1}$	Target BA $\text{m}^2 \text{ha}^{-1}$	Reduction %	$\text{m}^2 \text{ha}^{-1}$	Reduction $\text{m}^2 \text{ha}^{-1}$
Beech	25 ± 0.86	17	18	4.6 ± 0.28	4.5	76	19 ± 0.71	24 ± 0.96
Douglas fir	32 ± 1.6	23	20	6.4 ± 0.40	5.0	78	25 ± 1.1	32 ± 1.5
Scots pine	23 ± 1.2	18	16	3.7 ± 0.47	4.1	83	19 ± 1.0	22 ± 0.65

2.2 Biomass sampling

To determine the dry biomass of the aboveground tree compartments in forests, allometric relationships were constructed to scale biomass measures from three harvested trees per stand and thus fifteen trees per species in total to the entire above-ground forest. In each stand, a dominant, intermediate and suppressed tree representing the average DBH within the canopy position class was felled in February or March 2019. Per tree, the dry biomass was determined for small branches (up to $\varnothing$ 2 cm), coarse branches ($2 \text{ cm} < \varnothing < 10 \text{ cm}$), stem bark, stem sapwood and, in the case of Douglas fir and Scots pine, stem heartwood and needles. Total dry biomass was calculated based on within-tree crown allometric relationships (for branches and needles) and calculations of stem volume and tissue densities (for stem bark, sapwood and heartwood) (Vos et al., 2023b). Nutrient samples per tree compartment were taken for the analysis of nitrogen (N) and carbon (C) contents using a CN-analyzer (LECO TruSpec CHN, USA).

Table 5.2 Stand and soil characteristics of the beech, Douglas fir and Scots pine forests of this study. The stand density includes all trees with DBH > 10 cm, Dg is the root mean square DBH and H is the height based on 16 dominant trees per stand. The soil cover are coverage percentages for the foliar litter, mosses, graminoids and ferns. Mineral soil properties include the pH_{H2O}, the soil bulk density (BD), the average Al-S saturation of cation exchange capacity (CEC) by Al per hectare and the average soil base saturation (BS) per hectare. Both Al-S and BS are based on the unbuffered CEC, with "b.d." in the case where Ca, K, Mg and Na were undetectable. Soil profiles were classified according to the international standards (IUSS Working Group WRB 2015).

Site	Species	Forest stand			Soil cover					Mineral soil (0–30 cm depth)				
		Age	Density	Dg	H	Foliar litter	Mosses	Graminoids	Ferns	pH	BD	Al-S	BS	Soil type
		(yr.)	(n ha ⁻¹)	(cm)	(m)	%	%	%	%		g cm ⁻³	%	%	
1	Beech	94	260	34	22	100	0	0	0	4.3	1.0	96	2.1	Albic Podzol
2	Beech	100	140	47	26	98	2	0	0	4.3	1.0	100	b.d.	Entic/Albic podzol
3	Beech	82	220	38	25	100	0	0	0	4.2	0.98	94	1.3	Dystric Cambisol
4	Beech	98	240	34	24	97	3	0	0	4.5	1.3	83	10	Dystric Cambisol
5	Beech	46	1100	18	19	100	0	0	0	4.1	1.1	77	1.3	Dystric Cambisol
1	Douglas fir	74	120	54	41	28	72	0	0	4.2	1.3	68	3.3	Albic podzol
2	Douglas fir	59	170	50	37	37	63	0	0	4.0	1.1	75	2.0	Entic/Albic podzol
3	Douglas fir	60	140	52	37	26	69	2	0	4.2	1.2	100	3.8	Entic/Albic podzol
4	Douglas fir	66	150	51	36	93	5	0	0	4.0	1.1	74	4.0	Entic/Albic podzol
5	Douglas fir	60	240	44	28	3	2	0	95	4.2	1.2	91	7.1	Albic podzol
1	Scots pine	55	420	25	19	51	49	0	0	4.3	1.2	79	1.3	Albic podzol
2	Scots pine	48	430	24	18	54	37	9	0	4.1	0.97	88	3.2	Albic podzol
3	Scots pine	47	880	17	19	34	59	7	0	4.4	1.4	88	5.0	Dystric Cambisol
4	Scots pine	62	450	26	21	36	59	5	0	4.0	0.98	69	4.3	Albic podzol
5	Scots pine	73	470	27	16	31	24	45	0	4.2	1.1	92	5.0	Albic podzol

Concentrations of phosphorus (P), sulfur (S), potassium (K), calcium (Ca), magnesium (Mg), manganese (Mn), copper (Cu), iron (Fe) and zinc (Zn) following HNO_3 extraction were analysed with ICP-AES (Thermo-Scientific iCAP 6500 DUO, USA). The selection of the sampled trees, destructive biomass sampling, calculation of the dry weight and the determination of the nutrient concentration per compartment is described in more detail in (Vos et al., 2023b).

2.3 Calculation of tree biomass and nutrient stocks

To estimate above-ground biomass for all trees in each subplot, allometric relationships for the aboveground biomass were developed based on these three intensively measured trees per stand summing to 15 trees per tree species. The dry weights of small branches, coarse branches, stem bark, stem sapwood, and, in the case of Douglas fir and Scots pine, stem heartwood and needles were modelled as a function of the DBH. The following statistical model was constructed:

$$\ln(DW_{TC}) = \alpha_{[l]} + \beta_{[l]} * \ln(DBH) + \varepsilon_{[l]} \quad (1)$$

where DW_{TC} is the dry weight of a tree compartment (needle, small branch, coarse branch, stem bark, stem sapwood and stem heartwood). The models were nested per location (l) using random intercept and random intercept slope models. Parameters were estimated in linear form by using logarithmic transformation to increase model performance. Model performance was evaluated based on Pearson's correlation between the log-transformed measured and fitted values, yielding average correlations of 0.95 for total stem weight and 0.89 for total tree crown weights. The constructed allometric relationships are given in Table 5.3.

Table 5.3 Allometric relationships for the biomass of needles, branches, stem bark and stem wood for beech, Douglas fir and Scots pine. For nested models, chosen when nesting improved model AIC by $\Delta 2$ (Zuur et al., 2009), the average model is reported. Stand specific models are in Table S5.2. Goodness of fit of the models was assessed with Pearson's correlations of the log-transformed observed values versus the log-transformed predicted values.

Treepart	Sub model	Species	Nested	Model	Pearson
Needle	None	Douglas fir	No	$\ln(NE) = -3.36 + 1.64 * \ln(DBH)$	0.80
		Scots pine	Yes	$\ln(NE) = -5.44 + 2.29 * \ln(DBH)$	0.98
Branches	Small branches	Beech	No	$\ln(SB) = -2.68 + 1.77 * \ln(DBH)$	0.82
		Douglas fir	No	$\ln(SB) = -2.93 + 1.63 * \ln(DBH)$	0.82
		Scots pine	Yes	$\ln(SB) = -5.39 + 2.47 * \ln(DBH)$	0.98
	Coarse branches	Beech	No	$\ln(CB) = -3.47 + 2.40 * \ln(DBH)$	0.82
		Douglas fir	No	$\ln(CB) = -5.93 + 2.65 * \ln(DBH)$	0.86
		Scots pine	Yes	$\ln(CB) = -9.69 + 3.79 * \ln(DBH)$	1.00
Bark	None	Beech	Yes	$\ln(BA) = -4.30 + 2.20 * \ln(DBH)$	1.00
		Douglas fir	No	$\ln(BA) = -3.33 + 2.08 * \ln(DBH)$	0.93
		Scots pine	No	$\ln(BA) = -3.96 + 2.03 * \ln(DBH)$	0.97
Stem wood	Sapwood	Beech	Yes	$\ln(SW) = -2.09 + 2.37 * \ln(DBH)$	1.00
		Douglas fir	No	$\ln(SW) = -3.36 + 2.46 * \ln(DBH)$	0.93
		Scots pine	No	$\ln(SW) = -2.95 + 2.42 * \ln(DBH)$	0.93
	Heartwood	Douglas fir	No	$\ln(HW) = -4.19 + 2.69 * \ln(DBH)$	0.98
		Scots pine	No	$\ln(HW) = -3.94 + 2.21 * \ln(DBH)$	0.86

To estimate the foliar biomass of the beech stands, published allometric models were fitted to the data. Allometric relationships were selected when based solely on European beech, using only tree DBH as a predictor and different allometric relationships were available for the aboveground biomass, stem biomass, branch biomass and foliar biomass. Based on the review by Zianis et al. (2005), these allometric relationships were available for beech trees in the Netherlands (Bartelink, 1997), France (Le Goff and Ottorini, 2000) and Spain (Santa Regina and Tarazona, 2001). The allometric relationships of Bartelink (1997) and Le Goff and Ottorini (2000) structurally overestimated the aboveground biomass of large trees, particularly for branches. The allometric equations of Santa Regina and Tarazona (2001) provided a good fit for the aboveground biomass and stem biomass and a reasonable fit for branch biomass (r^2 in range 0.93-1.0). The allometric relationship for beech foliage of Santa Regina and Tarazona (2001) were therefore used to estimate beech foliar biomass.

The allometric relationships were used to model the biomass of the different aboveground tree compartments for each tree with a DBH > 10 cm within the 1-ha stand. To avoid bias due to back transformation of log-transformed data, we applied a correction factor to minimize mean squared error according to the method described by Shen and Zhu (2008). This correction factor resulted in the smallest

bias for predicting biomass of non-sampled trees (Clifford et al., 2013). The total biomass stock per stand was the sum of the biomass of all aboveground tree compartments, while the biomass export under high-thinning, shelterwood and clearcut was calculated as the biomass of the harvested trees per treatment:

$$B_{tj} = \sum DW_{cts} * 4 \quad (2)$$

where B is the biomass in kg ha⁻¹ for treatment t and stand s, and DW is the dry weight of tree compartment c for treatment t and stand s. Because measurements were done in a 0.25ha subplot, the total biomass was multiplied by 4 to retrieve biomass in kg ha⁻¹. Nutrient stocks were calculated per tree compartment using:

$$NS_{cs} = DW_{cs} * [Nutrient]_{cs} \quad (3)$$

where the nutrient stock (NS) is the result of the dry weight of tree compartment c within stand s times the nutrient concentration [Nutrient] of tree compartment c within stand s. The total nutrient stock per stand was the sum of the nutrient stocks per tree compartment per tree, where different tree compartments and different trees were included dependent on the treatment.

The treatments distinguished were harvest intensity, distinguishing high thinning (HT), shelterwood (SW) and clearcut (CC) in which different basal areas were removed– and harvest method consisting of whole tree harvest, stem only harvest and wood only harvest. In the “whole tree harvest” treatment all aboveground tree biomass is harvested; in the “stem only harvest” treatment, all the stem biomass is harvested (stem is defined as stem base until Ø 10 cm); and in the “wood only harvest” treatment the wood of the stem, without the stem bark, is harvested. The biomass and nutrient exports were calculated based on these harvest intensity treatments and harvest methods from five stands per species. All export data per treatment in each of the 0.25 ha subplots were scaled up to 1 ha.

2.4 Annual nutrient uptake

Annual nutrient uptake rates were estimated for the period 2008-2018 based on diameter increment, which were linked to changes in DBH and then to changes in total aboveground biomass and nutrient stock. Stem disks from the stem base (at 30 cm height) from the 15 trees used for biomass sampling, were polished and tree-ring widths were measured to the nearest 1/100 mm on two perpendicular radii using dendrochronological measuring equipment (LINTAB, TSAP; Rinn, 2003). The COFECHA ver. 6.02P software (Grissino-Mayer, 2001) was used to assess the data quality and accuracy after cross dating. The ring width data of the last 10 years were used to calculate the average yearly diameter increase using the formula:

$$ADI = (\sum \left(\frac{RW_i}{d_{i-1}} \right) * 100) / 10 \quad (4)$$

where ADI is the average relative annual diameter increment (%), RW_i is the ring width in year i which ranged from 2008 to 2018, d is the diameter of the stem wood of year $i - 1$ and 10 is the number of years included.

The annual diameter increase of Douglas fir and Scots pine was influenced by the stem diameter (Table S5.3). Therefore, all trees per stand were assigned to annual diameter increment (%) of the sampled tree with the nearest DBH. The diameter after one year of annual growth was calculated by multiplying the ADI of the nearest assigned tree per forest with the DBH using:

$$DBH_{new\ is} = DBH_{is} + \left(DBH_{if} * \frac{ADI_s}{100} \right) \quad (5)$$

Where the new DBH per tree i and stand s is the sum of the measured DBH per tree and forest times the ADI of the sampled tree with the nearest DBH per forest. The total biomass and nutrient stock per stand were calculated based on DBH_{new} by implementing the allometric relationships for foliage, small branches, coarse branches, stem bark and stem wood (Table 5.3). The uptake of nutrients over 1 year is the result the nutrient stocks based on DBH_{new} minus the nutrient stocks based on DBH. This calculation assumes that the annual diameter increment is primarily due to stem wood increment by ignoring the often-neglectable annual diameter increment caused by bark growth.

2.5 Soil sampling

To obtain soil nutrient stocks, the mineral soil and the organic soil layers were sampled between November 2018 and January 2019, prior to forest harvest. Five soil samples were taken in each of four subplots (one subplot per treatment), resulting in 20 (sub-)samples per stand. Sampling points were determined systematically in a cross design with the central sampling point in the geometrical center of the subplot. For each central sampling point, the thickness of the litter layer, fragmented layer and humified layer was noted and the soil profile was described according to international standards (WRB, 2015). Samples of the organic soil layers and mineral soil were taken at each sampling point. Organic soil samples consisted of bulked samples of the ectorganic OL, OF and OH layers and were collected within a Ø 14.5 cm ring allowing to calculate the mass per unit of surface ($g\ cm^{-2}$). Bulk samples of the mineral soil were taken from the 0 to 30 cm depth directly underneath the organic soil layers sampling point using a split tube sampler (Eijkelkamp Soil & Water, Giesbeek, The Netherlands) at each sampling point. Samples were stored at 4 °C directly after the sampling before drying to a constant weight at 40°C.

Dried samples of the organic soil layers were ground to homogenize the sample in a mill containing a 1.5 mm stainless steel screen. The weight of the organic soil sample was corrected for the admixture with mineral soil by using loss on ignition (550°C). Samples were merged per subplot and total contents of C and N were measured using a CN-analyzer (LECO TruSpec CHN, USA). The contents of P, S, K, Ca,

Mg, Mn, Cu, Fe and Zn were determined after extraction with 0.43M HNO₃ (Groenenberg et al., 2017) on the ash of ignition (550°C) via ICP-AES (Thermo-Scientific iCAP 6500 DUO, USA).

Mineral soil samples were sieved to 2 mm to separate gravel from the fine earth fraction. Samples were merged per subplot and direct available nutrients (P, S, K, Ca, Mg, Mn, Cu, Fe and Zn) and soil pH were measured after H₂O extraction (1:10 soil water ratio); the unbuffered cation-exchange capacity (CEC) was measured by using 0.1M BaCl extraction. The contents of extractable nutrients and cations were determined with ICP-AES (Thermo-Scientific iCAP 6500 DUO, USA). Contents of N and P-PO₄ in the fine earth were determined with a Segmented Flow Analyzer (SFA type 4000, Skalar Analytical B.V., the Netherlands).

Soil nutrient stocks were calculated for the organic soil layers and mineral soil separately. The dry mass of the organic soil layers was corrected for the admixture with mineral soil particles before the pseudo total nutrients stocks were calculated by multiplication of the dry mass (kg ha⁻¹) and the nutrient concentration. For the mineral soil, the bulk density of the fine earth (g cm⁻³) was multiplied with the extractable available nutrient contents and the sample depth to calculate the available nutrient stocks per hectare in the top 30-cm mineral soil.

2.6 Statistical analysis

Prior to statistical analysis, data on aboveground biomass and nutrient stocks, belowground nutrient stocks and nutrient uptake were scaled to a 1-ha forest using data on subplot level. To compare biomass and nutrient stocks and nutrient uptake between species, one-way ANOVA tests were performed. To test biomass increment for the different tree compartments a two-way ANOVA test was used. The analyses were performed by using mixed-effect linear models from the R package “nlme”. Paired t-test was used to test the differences in the aboveground and below ground nutrient stocks by using the R package “stats”. Left- or right-skewed data were log or square root transformed, respectively, to meet the normality and homogeneity assumptions. Tukey’s post-hoc (HSD) test was performed following One-way ANOVA using the R package “emmeans” to test for differences between species. Spatial independence of the stands within the locations was tested using random structures. The added random structure did not improve AIC ($\Delta 2$ AIC) for any of the models.

3 Results

3.1 Aboveground biomass and nutrient stocks and soil organic layer nutrient stocks

The average total aboveground stock of dry biomass per hectare was on average 190 ± 13 (s.e.) tons in beech, 230 ± 10 (s.e.) tons in Douglas fir and 100 ± 8.1 (s.e.) tons in Scots pine stands (Table 5.4). The aboveground nutrient stocks were nutrient and species dependent. The nutrient stocks decreased in the

order of $N > Ca > K > Mg > S > P > Mn > Zn > Fe > Cu$. Beech had generally higher nutrient stocks compared to Douglas fir and Scots pine, while Douglas fir had the highest aboveground biomass (Fig. 5.1, Table 5.4). Beech had significantly higher aboveground nutrient stocks for Ca, K, Mg and Mn, with the Ca stock nearly 4 times higher compared to Douglas fir and Scots pine. Scots pine stands had in general the lowest nutrient stocks, with significantly lower stocks of N and Fe compared to both beech and Douglas fir. Stocks of S and P only differed significantly between beech and Scots pine, while species did not differ significantly in the aboveground stocks of Zn and Cu.

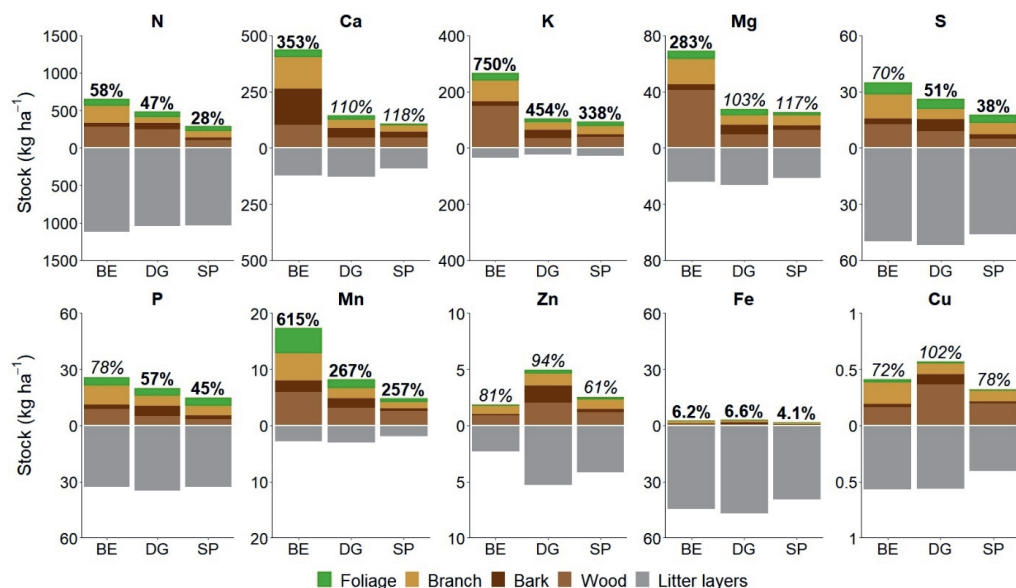


Figure 5.1 Nutrient stocks (kg ha^{-1}) in the foliage, branches, stem bark, stem wood and organic layers in forest stands dominated by European beech (BE), Douglas fir (DG) and Scots pine (SP). All nutrient stock values are based on measurements, except for the foliage of European beech which were calculated based on allometric relationships and nutrient concentrations derived from literature, see method section 2.3. The percentage values present the ratios of the above-ground nutrient stocks in trees divided by the nutrient stocks in the organic layers. Bold percentages indicate statistical differences between the above ground and the organic layer nutrient stocks according to Paired t-test statistics (Table S5.6).

The total nutrient stocks in the organic soil layers (thickness 78 – 97 mm; Table S5.5) did not significantly differ between tree species (Table 5.5). Largest nutrient stocks in the organic soil layers were observed for N and lowest nutrient stocks were observed for Cu (Fig. 5.1). The total nutrient stocks in the organic soil layers were larger than aboveground nutrient stocks, except for the base cations (Ca, K, Mg)

and Mn (Fig. 5.1, Table S5.6). The organic soil stock of Ca, K, Mg and Mn were smaller than the aboveground stocks, but differences were only large and significant for K and Mn, and for Ca and Mg in beech (Fig. 5.1). The highest biomass nutrient stock to soil nutrient stock was observed for K, with an aboveground nutrient stock between 338% to 750% of the stock present in the organic soil layers (Fig. 5.1). In contrast, high stocks in the organic soil layers as compared to the aboveground tree stock were observed for Fe, where 4 to 6% of the organic soil layers stock was present in the aboveground tree biomass. The organic layer nutrient stocks of major nutrients N, P and S were similar to (in case of beech) or larger than the aboveground nutrient stocks. Overall, these results hint to potential growth limitations by base cations and Mn, but in different amounts for different species.

Table 5.4 Exports of biomass (ton ha⁻¹) and nutrients (kg ha⁻¹) in a clearcut following whole tree harvest, stem only harvest and stem wood harvest. Whole tree harvest depicts the total aboveground biomass and nutrient stocks of the forests. ANOVA F and P values are presented to compare differences in biomass and nutrient exports between species. The F value and significance of the one-way Anova based on species is displayed per harvest method. Different letters denote significant differences among species according to Tukey's *posthoc* test with a significance level of P < 0.05.

	Biomass ton ha ⁻¹	N kg ha ⁻¹	Ca kg ha ⁻¹	K kg ha ⁻¹	Mg kg ha ⁻¹	S kg ha ⁻¹	P kg ha ⁻¹	Mn kg ha ⁻¹	Zn kg ha ⁻¹	Fe kg ha ⁻¹	Cu kg ha ⁻¹	
Whole tree	Beech	190 ± 17 ^b	520 ± 54 ^b	380 ± 76 ^b	220 ± 25 ^b	59 ± 13 ^a	27 ± 2.1 ^b	20 ± 2.1	12 ± 2.7 ^a	1.5 ± 0.27	2.3 ± 0.33 ^{ab}	0.35 ± 0.026
	Douglas fir	240 ± 19 ^b	490 ± 50 ^b	150 ± 34 ^a	100 ± 6.2 ^a	28 ± 1.9 ^a	27 ± 2.8 ^b	20 ± 1.6	8.6 ± 2.5 ^a	5.2 ± 3.3	3.1 ± 0.53 ^b	0.53 ± 0.10
	Scots pine	99 ± 7.8 ^a	290 ± 32 ^a	110 ± 11 ^a	93 ± 12 ^a	25 ± 0.93 ^a	17 ± 2.2 ^a	15 ± 2.7	4.8 ± 0.60 ^a	2.5 ± 0.84	1.6 ± 0.16 ^a	0.32 ± 0.035
Stem only	<i>F</i> -value	27	8.7	12	11	4.6	4.9	2.6	2.4	5.0	3.8	
	<i>P</i> -value	<0.001	< 0.005	< 0.001	< 0.005	0.047	0.027	0.11	0.15	0.026	0.053	
	Beech	130 ± 13 ^b	310 ± 33 ^b	240 ± 50 ^b	150 ± 20 ^b	42 ± 10 ^b	14 ± 1.2 ^b	10 ± 1.3 ^b	7.5 ± 1.6	0.88 ± 0.19	0.85 ± 0.16 ^{ab}	0.17 ± 0.012 ^a
Stem wood	Douglas fir	210 ± 17 ^c	330 ± 31 ^b	89 ± 19 ^a	63 ± 3.4 ^a	16 ± 1.1 ^a	16 ± 1.5 ^b	10 ± 0.79 ^b	5.0 ± 1.4	3.6 ± 2.3	1.5 ± 0.33 ^b	0.42 ± 0.11 ^b
	Scots pine	78 ± 5.7 ^a	140 ± 16 ^a	71 ± 5.9 ^a	48 ± 4.4 ^a	16 ± 0.54 ^a	7.2 ± 0.64 ^a	5.2 ± 0.83 ^a	3.0 ± 0.31	1.4 ± 0.39	0.56 ± 0.12 ^a	0.21 ± 0.027 ^a
	<i>F</i> -value	27	17	13	24	6.3	14	12	2.6	3.4	6.9	6.8
Stem wood	<i>P</i> -value	<0.001	< 0.001	0.001	< 0.001	0.014	< 0.001	0.001	0.11	0.087	0.010	0.011
	Beech	130 ± 120 ^b	260 ± 28 ^b	94 ± 11 ^b	140 ± 19 ^b	38 ± 9.9 ^c	11 ± 1.1 ^b	8.1 ± 1.2 ^b	5.6 ± 1.2	0.76 ± 0.17	0.57 ± 0.17	0.14 ± 0.0096
	Douglas fir	190 ± 150 ^c	250 ± 24 ^b	46 ± 5.4 ^a	35 ± 2.6 ^a	9.3 ± 0.63 ^a	8.9 ± 0.92 ^b	5.1 ± 0.53 ^{ab}	3.3 ± 1.1	2.0 ± 1.2	0.67 ± 0.14	0.33 ± 0.12
Stem wood	Scots pine	72 ± 54 ^a	110 ± 14 ^a	44 ± 3.7 ^a	38 ± 3.7 ^a	13 ± 0.52 ^b	4.8 ± 0.46 ^a	3.4 ± 0.65 ^a	2.5 ± 0.27	1.2 ± 0.32	0.38 ± 0.11	0.19 ± 0.028
	<i>F</i> -value	27	17	14	15	12	16	9.6	2.1	1.4	1.6	2.4
	<i>P</i> -value	<0.001	< 0.001	< 0.001	0.002	0.004	< 0.001	0.003	0.16	0.29	0.25	0.14

Nutrient stocks per stand and per tree compartment are in Table S5.4.

3.2 Annual nutrient uptake and nutrient availability

The estimated annual above-ground biomass increment was $4700 \pm 430 \text{ kg ha}^{-1} \text{ yr}^{-1}$ in beech stands, $7800 \pm 910 \text{ kg ha}^{-1} \text{ yr}^{-1}$ in Douglas fir stands and $3900 \pm 430 \text{ kg ha}^{-1} \text{ yr}^{-1}$ in Scots pine stands. The biomass increment is significantly higher in Douglas fir compared to both beech and Scots pine and is driven by a higher biomass increment of the stem wood and bark (Table S5.7). Annual nutrient uptake was largest for N and lowest for Cu (Fig. 5.2). Base cation uptake differed between species, beech had 2.4 times higher uptake rates than Douglas fir and Scots pine stands for Ca, and 1.5 and 1.9 times higher for K and Mg respectively (Fig. 5.2, Table S5.8). There was no significant difference in Mn uptake between the species, although beech tended to have a higher uptake than Douglas fir and Scots pine.

The concentrations of available nutrients in the mineral soil did not differ between species and decreased in the order of $\text{N} > \text{S} > \text{K} > \text{Fe} > \text{Mg} > \text{Ca} > \text{P} > \text{Zn} > \text{Mn} > \text{Cu}$. The base saturation was on average 3.6% while the average CEC-Al equaled 85%. The annual nutrient uptake was generally lower than the available nutrients in the mineral soil which was measured during wintertime (Table 5.5). Only the annual uptake of Ca in beech and Mn in Scots pine exceeded the available nutrients in the mineral soil (Fig. 5.2, Table 5.5). The nutrient stock of the organic soil layers was over 50 times higher than the annual nutrient uptake for N, P and S, and less than 10 times bigger than the annual nutrient uptake of K and Mn (in case of beech).

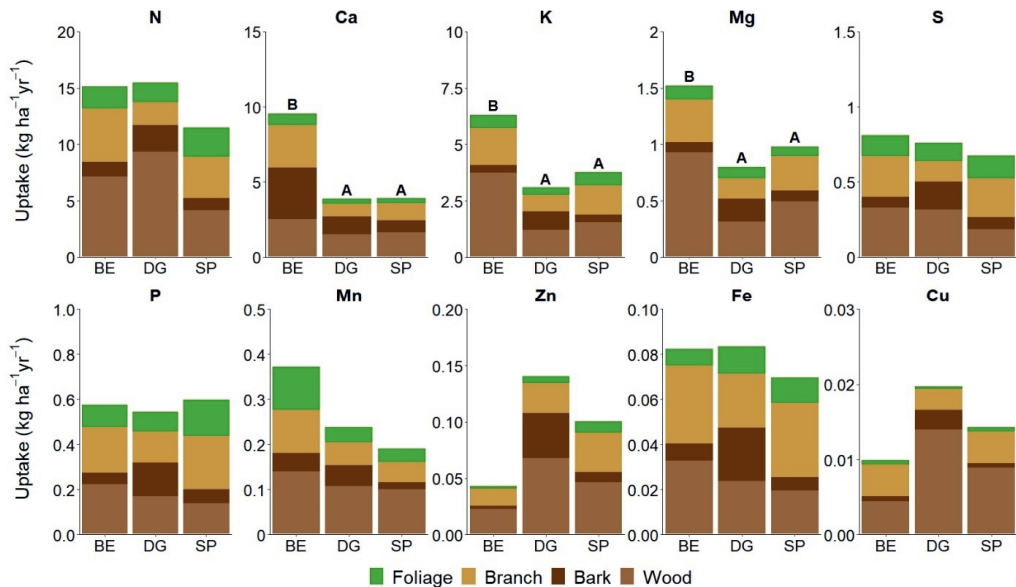


Figure 5.2 Estimated mean annual nutrient uptake rates in the foliage, branches, stem bark and stem wood for forest stands dominated by European beech (BE), Douglas fir (DG) and Scots pine (SP) over the period 2008-2018 ($\text{kg ha}^{-1} \text{ yr}^{-1}$). Different capital letters indicate significant differences (ANOVA, $p < 0.05$) among species. Absence of capital letters indicate no significant differences between species.

Table 5.5 The means (and standard errors) of the total nutrient stocks of macro (N, Ca, K, Mg, S and P) and micro (Mn, Zn, Fe and Cu) nutrients in the organic soil layers (kg ha⁻¹) and the available stock of macro and micronutrients (kg ha⁻¹) in the upper 30 cm of the mineral soil. The F value and significance of the one-way Anova based on species per soil layer (organic layers and mineral soil) is displayed.

	N	Ca	K	Mg	S	P	Mn	Zn	Fe	Cu	
	kg ha ⁻¹	kg ha ⁻¹	kg ha ⁻¹	kg ha ⁻¹	kg ha ⁻¹	kg ha ⁻¹	kg ha ⁻¹	kg ha ⁻¹	kg ha ⁻¹	kg ha ⁻¹	
Organic layers	Beech	1100 ± 190	120 ± 15	36 ± 4.2	24 ± 2.3	50 ± 6.5	33 ± 4.0	2.8 ± 0.40	2.3 ± 0.82	45 ± 6.5	0.57 ± 0.09
	Douglas fir	1000 ± 100	130 ± 8.7	23 ± 2.7	27 ± 4.3	52 ± 2.8	35 ± 2.6	3.1 ± 0.40	5.3 ± 3.7	47 ± 2.7	0.56 ± 0.05
	Scots pine	1000 ± 110	92 ± 10	28 ± 2.5	22 ± 2.6	46 ± 5.5	33 ± 2.9	1.9 ± 0.28	4.2 ± 2.1	40 ± 5.9	0.41 ± 0.05
	<i>F-value</i>	0.12	2.9	3.9	0.64	0.37	0.14	3.08	1.1	0.50	2.0
	<i>P-value</i>	0.89	0.09	0.05	0.54	0.70	0.87	0.08	0.39	0.62	0.18
Mineral soil	Beech	42 ± 1.8	5.2 ± 1.7	26 ± 3.7	8.0 ± 1.7	18 ± 0.78	4.3 ± 0.43	0.49 ± 0.19	0.42 ± 0.045	16 ± 1.9	0.10 ± 0.028
	Douglas fir	54 ± 2.8	8.7 ± 1.8	22 ± 1.4	6.9 ± 0.69	28 ± 2.2	7.4 ± 1.3	0.71 ± 0.19	2.1 ± 0.89	21 ± 2.3	0.12 ± 0.032
	Scots pine	46 ± 2.4	4.0 ± 0.50	27 ± 2.3	4.8 ± 0.86	19 ± 1.1	5.8 ± 0.72	0.15 ± 0.039	0.70 ± 0.23	15 ± 1.7	0.065 ± 0.0090
	<i>F-value</i>	2.0	1.3	0.18	0.76	3.3	1.9	2.1	0.19	0.69	0.11
	<i>P-value</i>	0.18	0.30	0.84	0.49	0.07	0.19	0.17	0.83	0.52	0.89

The nutrient stocks of the organic layers per stand are in Table S5.4, the available concentration of the mineral soil during wintertime per stand are in Table S5.8.

3.3 *Impact of harvest intensity on biomass and nutrient export*

The reduction to the target basal area for the treatments yielded an average basal area export between 3.7 to 6.4 m² for high thinning, 19 to 25 m² for shelterwood and 22 to 34 m² for clearcut (Table 5.2). Basal area reductions were comparable between beech and Scots pine but higher for Douglas fir corresponding to much higher absolute biomass exports in Douglas fir compared to beech and Scots pine (Fig. 5.3, Table 5.4). Biomass reductions for the different timber- and biomass harvest intensities ranged from 12% (SOH) - 21% (WTH) in high thinning, 55% (SOH) - 86% (WTH) in shelterwood and 67% (SOH) - 100% (WTH) in a clearcut (Table S5.9).

Nutrient export was highest for N and lowest for Cu for all timber harvest intensities, proportional to nutrient stocks (Table 5.4). Differences in biomass and nutrient export were substantial between high thinning and shelterwood and high thinning and clearcut, while shelterwood and clearcut resulted only in a slight difference in biomass and nutrient export (Fig. 5.3).

Nutrient export differed between species. In a conventional stem only harvest clearcut of beech significantly higher stocks of Ca and K and, although not significant, higher stocks of Mg were exported. In a Scots pine stem only clearcut, significant lower stocks of N, P and S were exported (Fig. 5.3). Differences in biomass and nutrient exports were more pronounced when comparing whole-tree harvest but less when comparing stem-wood harvest (Fig. S5.2, Table 5.4).

3.4 *Impact of harvest method on biomass and nutrient export*

The biomass and nutrient export were compared between stem only harvest and whole tree harvest, and between stem only harvest and wood only harvest. Compared to stem only harvest, whole tree harvest increased the biomass export on average by 35% (52 tons ha⁻¹) for beech, 12% (26 tons ha⁻¹) for Douglas fir and 27% (21 tons ha⁻¹) for Scots pine following clearcut harvest. The average increase of nutrient export was highest in Scots pine (100%), intermediate in beech (87%) and lowest in Douglas fir (66%). Highest increase of nutrient export was observed for Fe where whole tree harvest increased export up to 220% (Fig. 5.4). Large increase in export was also observed for P, whole tree harvest resulted in an increased P export of 92% in Douglas fir, 96% in beech and 180% in Scots pine. The average increase of the base cations (Ca, K, Mg) and Mn export resulting from whole tree harvest was 55% in beech, 66% in Scots pine and 68% in Douglas fir.

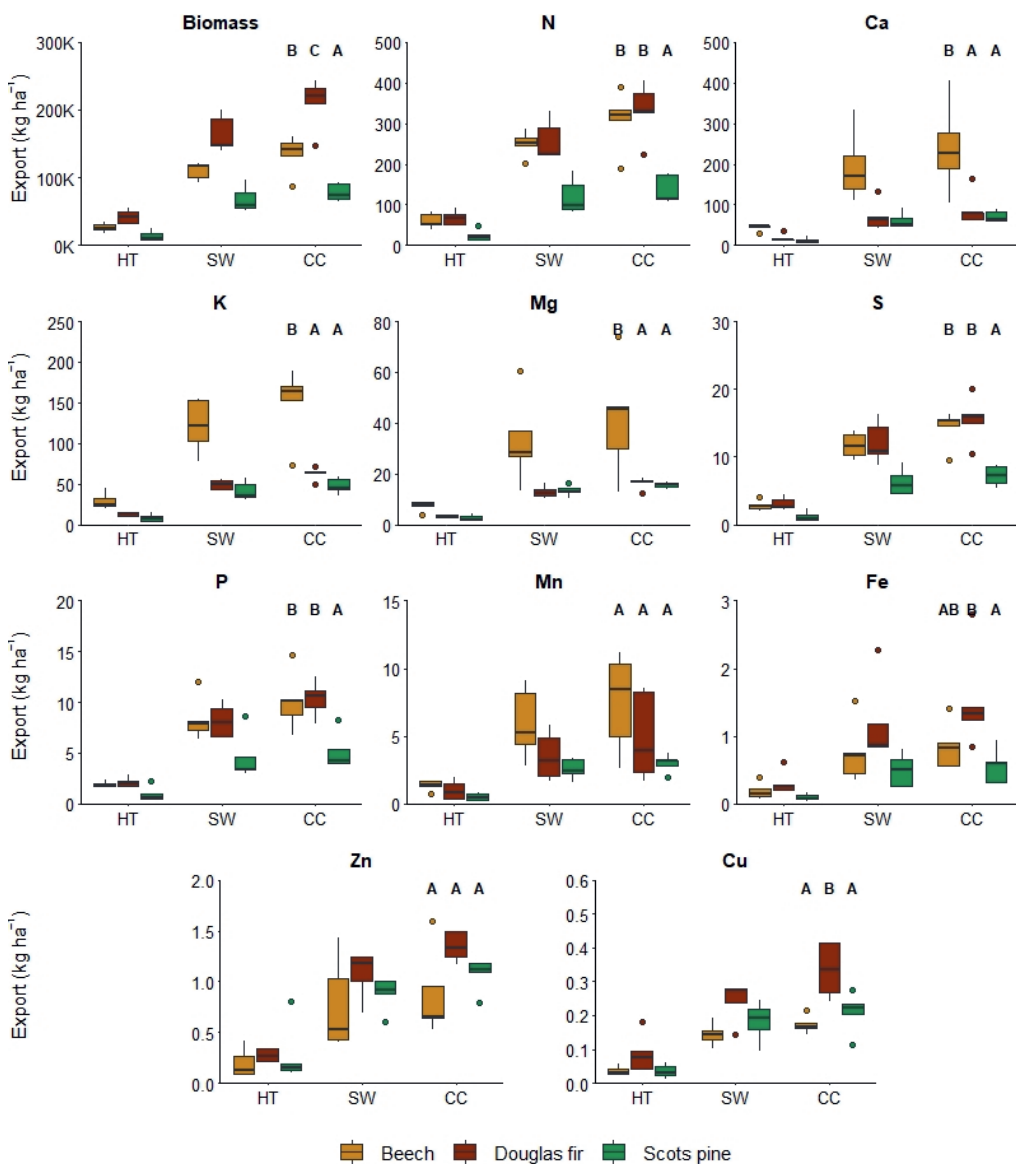


Figure 5.3 Export of biomass, macronutrients (N, P, K, S, Ca and Mg) and micronutrients (Mn, Cu, Fe, Zn) under the different timber harvest intensities: high-thinning (HT), shelterwood (SW) and clearcut (CC). The harvest method considered here is stem only harvest, the biomass harvest intensities whole tree harvest and stem wood harvested are in Fig. S5.2. Colored dots are outliers. Different capital letters indicate significant differences (ANOVA, $p < 0.05$) among species for the clearcut treatment. Same magnitude, but with less pronounced differences is expected for the high-thinning and shelterwood treatments.

Compared to stem only harvest, wood only harvest (excluding the bark) in a clearcut decreased the biomass export by 5.9% in beech (7 tons ha⁻¹), 9.8% in Douglas fir (21 tons ha⁻¹) and 7.5% in Scots pine (6 tons ha⁻¹) (Fig. 5.4). Yet, the decrease of nutrient losses was much higher: wood only harvest resulted in an average decrease in nutrient export of 23% in beech, 25% in Scots pine and 41% in Douglas fir compared to stem only harvest. The highest decrease of nutrient export was observed for Ca in beech (58%) and P and Fe in Douglas fir (resp. 51% and 56%). The export of base cations Ca, K, Mg and Mn following wood only harvest decreased by 23% in Scots pine, 26% in beech and 43% in Douglas fir.

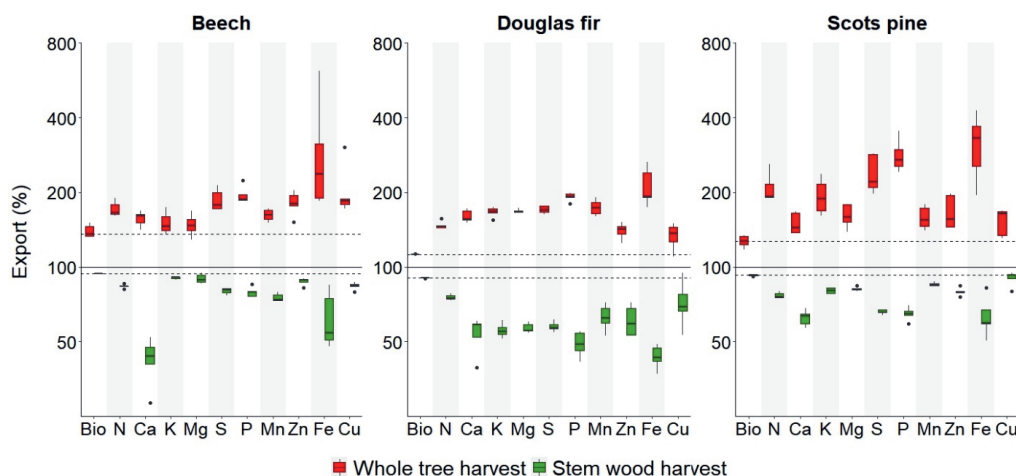


Figure 5.4 Effects of whole tree harvest (WTH) and wood only harvest (WOH, with bark removed and left in forest) on biomass and nutrient export relative to stem only harvest (SOH, wood and bark). The total export per nutrient in SOH is set to 100% and is denoted with a solid line. The red bars indicate the increased losses due to WTH, and the green bars the reduction in losses following WOH. The dashed lines refer to the average biomass lost in WTH and WOH relative to SOH. Note that biomass loss (Bio) is also indicated by the first bar in each plot.

4 Discussion

4.1 The potential of forest regrowth from a soil nutrient stock perspective.

The potential of forest regrowth after tree harvest depends largely on soil nutrient stocks. The nutrient stocks in organic soil layers are particularly important for long-term site nutrition for forest on low fertility sites, such as acidic soils (Prietzl and Stetter, 2010; Garrett et al., 2021). Many forest soils are sensitive to acidification resulting from long lasting acid deposition. The sampled forest soils were strongly acidic with a low base saturation (Table 5.2), which represent the conditions of many other European forests on acidified soils (Riek et al., 2012; Binkley and Högberg, 2016).

We found that the nutrient stocks of the organic soil layers were higher than in (above-ground) trees for most nutrients, except for Ca, K, Mg (base cations) and Mn, regardless of the tree species (Fig. 5.1). Aboveground stocks, however, differed between tree species with higher base cation stocks in beech. Overall, comparing annual nutrient uptake in aboveground woody tree biomass (i.e., immobilization) and the nutrient stocks of the organic layers, we observed that the current organic layer nutrient stock could support up to 20 years of annual K and Mn uptake and up to 50 years of Ca and Mg uptake. This indicates that, with too limited external nutrient supplies, the base cation stocks of organic soil layers may be insufficient for long-term site nutrition. High nutrient uptake demands by roots caused by high turnover rates (Brunner et al., 2013), which is not taken into account in this study, could further limit the period of growth supported by the organic layers. As the organic soil layers are the major source of base cations (Kuehne et al., 2008), base cation nutrition is a potential limiting factor for long-term forest growth when harvests are continued. Organic soil layers provide sufficient stocks of other nutrients to supply long-term forest growth. The current organic soil layers stocks potentially supply the current annual uptake in woody tree biomass of N, P and S for forest growth for more than 80 years, which is a common rotation period.

The dependency on the organic soil layers as a nutrient source, however, differs per species. For example, the superficial fine root distribution of Scots pine suggests a high dependency on the organic soil layers for nutrient uptake (Vanninen and Mäkelä, 1999; Helmisaari et al., 2007), while Douglas fir roots also appear in the top of the mineral soil (Nnyamah and Black, 1977; Olsthoorn, 1991). Deep soil uptake has been demonstrated for beech which reduces the dependency on the organic soil layers as a nutrient buffer (Berger et al., 2006; Turpault et al., 2019). Comparison of the nutrient stocks of the organic layers solely may therefore underestimate the potential of the soil nutrient stock to support forest growth.

The top of the mineral soil (down to 30 cm depth) however, hardly provides an additional nutrient stock for base cations as base saturation levels are generally below 4% (Table 5.2). The overall influence of the deep mineral soil (>30 cm depth) as a nutrient source in beech remains ambiguous. Generally, the biochemical cycling is inferior to nutrient uptake from biological nutrient cycling (Berger et al., 2006; van der Heijden et al., 2015). The uptake from organic soil layers and the biological cycling of nutrients accounts for a large part of the base cation uptake in beech (45-60%) (Göransson et al., 2006; van der Heijden et al., 2015; Turpault et al., 2019). This is reflected by the low organic layer stock of Ca, K and Mg stock of beech, which equaled the organic layer stocks in Douglas fir and Scots pine despite the higher aboveground nutrient stocks of beech. Therefore, despite of the deep soil uptake of beech, beech stands, as well as Douglas fir and Scots pine stands, depend on external nutrient supply of base cations for forest regrowth after harvest on the long term.

Finally, remarkable high stocks of N, S and Fe were present in the organic soil layers, with the Fe stock up to 500 times the annual Fe uptake. These high stocks are clear indicators of the ongoing effects

of S and N deposition. Reduction of the N deposition is expected to result in only a marginal response of forest stands (Schmitz et al., 2019), while accumulation of S in the organic soil layers is linked to a 50% reduction of the soil Ca and Mg pool (Prietz et al., 2004). The organic soil layers therefore hold legacies with negative impacts on (base cation) nutrition for the coming decades.

4.2 Growth and nutrient uptake

Species differ in both growth rate and nutrient uptake rates. Douglas fir grew faster in aboveground biomass than beech and Scots pine (Table S5.7) but was similar in nutrient uptake compared to Scots pine (Fig. 5.2). This higher nutrient use efficiency, resulting from the lower nutrient concentration of Douglas fir (Vos et al., 2023b), may contribute to the competitive superiority of Douglas fir over beech and Scots pine on low fertile sites.

Uptake of the macronutrients N, P and S did not differ between species. The annual uptake of base cations in aboveground tree biomass (including foliage), however, was up to 3 times higher in beech stands compared to Douglas fir and Scots pine stands (Fig. 5.2). This high base cation uptake was not reflected in high annual biomass growth, which implies a low base cation use efficiency in beech. This low efficiency may lead to more rapid depletion of soil exchangeable base cation stocks and nutrient imbalances, which result in growth reductions on nutrient poor sites (Balcar et al., 2011; Calvaruso et al., 2017; Cremer and Prietz et al., 2017; Court et al., 2018). Such growth reductions may already occur as the studied beech stands were of medium to poor yield contrary to Douglas fir and Scots pine stands, that were of excellent and good yield, respectively, according to traditional yield tables for such sites (Jansen et al., 2018). Also the within tree nutrient imbalances are likely to occur in beech stands as we found 24% lower annual aboveground uptake rates for K, 30% lower for Mg and around 50% lower for P and Ca compared to a forest stand with similar biomass production ($5.1 \text{ tons ha}^{-1} \text{ yr}^{-1}$) in France (Calvaruso et al., 2017). From this, we hypothesize that the growth of beech might be impaired by low mineral supply of base cations. Such impaired growth may limit further use of beech for timber and biomass production, also because nutrient imbalances have been previously related to increased sensitivity to drought and forest dieback (Bal et al., 2015; González de Andrés et al., 2021). From this we speculate that the sustainability of beech stands on soils with low reservoirs of base cations (K, Ca, Mg) and Mn may be at risk with respect to the mineral supply.

Surprisingly, despite lower concentrations of base cations and P in aboveground tree biomass (Vos et al., 2023b), the direct available nutrient stock in wintertime (Table 5.5) was overall higher than the annual nutrient uptake (Fig. 5.2) indicating no direct nutrient limitation for base cations, Mn and P in the short term. Caution for interpretation is however required since we did not take annual nutrient uptake rates in roots into account, therefore potentially underestimating the actual annual nutrient uptake rate by

trees. Nevertheless, available P in wintertime was 10 times greater than the annual P uptake although this available P was far below the optimum for tree growth (Van den Burg and Schaap, 1995). On an European level, P limitation is increasing as indicated by foliar concentrations (Du et al., 2021). Nonetheless, the relatively high available P concentrations in the mineral soil relative to tree uptake indicated no P limitation. Foliar concentrations did however indicate P limitation but not base cation limitation (Vos et al., 2023b). We do not know how these ambiguous responses emerge from underlying mechanisms, which may include decreased mycorrhizal uptake efficiency (Braun et al., 2010), the preferential uptake of N over other elements (Vanguelova and Pitman, 2019) or a mismatch in decomposition and uptake. Based on the soil organic stock and the availability of nutrients in the mineral soil, we argue that P limitation is not caused by a small total soil P stock contrary to base cations which are at risk of soil stock limitation.

4.3 *Base cation balance for forest growth*

The long-term recovery after harvest not only depends on the nutrient stocks in the soil, but also on the fluxes driving the dynamics of these stocks. The fluxes include losses by tree uptake and leaching from the soils on the one hand and the gain via deposition and weathering on the other hand. To quantify the nutrient balances of base cations for our forest stands, we compared the estimated nutrient uptake rates in aboveground woody biomass (including bark) with the estimated total deposition based on wet-only deposition (RIVM, 2015) and leaching and weathering data of comparable forest stands (de Vries et al., 2021). We focused on nutrient uptake of the woody biomass and bark and not the foliage, because the stem accumulates nutrients whereas the foliage recycles nutrients to soil within few years. This assumption is reasonable for mature forest of this study, which are supposed to have relatively constant annual needle production and low natural tree mortality rates (Turner and Long, 1975; Flower-Ellis, 1985; Albrektson, 1988). The leaching and weathering fluxes, however, add considerable uncertainty to the balance as site specific leaching and weathering fluxes are needed for balance calculations (Pare and Thiffault, 2016) and conclusions on balances remain therefore uncertain too. Nevertheless, the thus calculated nutrient balances were predominantly negative for K, Ca and Mg, indicating an annual reduction of the soil nutrient stock up to $4.2 \text{ kg ha}^{-1} \text{ yr}^{-1}$ for K, $1.1 \text{ kg ha}^{-1} \text{ yr}^{-1}$ for Mg and $8.6 \text{ kg ha}^{-1} \text{ yr}^{-1}$ for Ca (Table 5.6). The most negative balances were present for beech, which had the highest uptake rates, while balances for Douglas fir were least negative.

Table 5.6 Annual nutrient uptake in stem wood, bark and branches ($\text{kg ha}^{-1} \text{ yr}^{-1}$) and the nutrient inputs via weathering ($\text{kg ha}^{-1} \text{ yr}^{-1}$) and leaching ($\text{kg ha}^{-1} \text{ yr}^{-1}$). Weathering and leaching rates are based on published data of forest nutrient budgets in the Netherlands (de Vries et al., 2021), deposition is based on bulk deposition data (RIVM, Table S5.10) multiplied by the correction factor to calculate total deposition within a forest stand (Table S5.11). The balance is the sum of weathering and deposition minus uptake and leaching. The net external nutrient input is the sum of nutrients coming in by soil weathering and atmospheric deposition minus leaching, and thus excludes the nutrient uptake by trees.

Flux		K	Mg	Ca
Uptake ¹ (-)	Beech	5.7 ± 0.53	1.4 ± 0.16	8.8 ± 1.2
	Douglas fir	2.8 ± 0.27	0.70 ± 0.094	3.5 ± 0.63
	Scots pine	3.2 ± 0.47	0.89 ± 0.091	3.6 ± 0.48
Leaching (-)	Beech	2.3	2.9	5.8
	Douglas fir	1.3	1.6	3.2
	Scots pine	1.9	2.4	4.8
Weathering (+)		2.0	1.8	3.0
Deposition (+)	Beech	1.1	1.2	2.0
	Douglas fir	1.0	1.4	2.3
	Scots pine	1.0	1.2	1.5
Balance	Beech	-4.9	-1.3	-9.6
	Douglas fir	-1.1	0.90	-1.4
	Scots pine	-2.1	-0.29	-3.9
External nutrient input	Beech	0.8	0.1	-0.8
	Douglas fir	1.7	1.6	2.1
	Scots pine	1.1	0.6	-0.3

¹ uptake in wooden parts (stem wood, stem bark and branches).

Negative annual balances for base cations in beech, together with P limitation (Sverdrup et al., 2006; Schmidt et al., 2015) and negative balances of Ca in Douglas fir (Sverdrup et al., 2006) have been observed throughout Europe. Deficiencies, resulting from long-term negative balances, were observed for Mg and K in needles of Douglas fir in Czech Republic (Šrámek et al., 2019), indicating that negative balances of base cations in both beech and Douglas fir are widespread. Even on a global scale, K fertilization was found to increase tree growth in 69% of the forest ecosystems (Tripler et al., 2006), indicating that base cation nutrition is potentially limiting forest growth on continental to global scale. Base cation nutrition is currently still deteriorating due to high N deposition. The uptake of K is negatively affected by N uptake

in beech stands (Vanguelova and Pitman, 2019) and even though N deposition is decreasing, recovery of the nutrient imbalance remains limited in European forests (Schmitz et al., 2019). Although, the loss of base cations can be (partly) mitigated by deep soil uptake in beech, no such mechanisms can possibly compensate for nutrient losses in Douglas fir and Scots pine (Nnyamah and Black, 1977; Olsthoorn, 1991; Vanninen and Mäkelä, 1999; Helmisaari et al., 2007). Overall, despite high uncertainty in the nutrient balance, there are multiple indicators suggesting possible growth limitation due to negative balances of K, Ca, Mg and Mn. These negative balances may result in long-term growth reductions and can be a risk for timber and biomass production as well as forest health.

4.4 *Effect of harvest intensity*

Nowadays, current forest management tends to shift towards less intense harvest intensities in which a continuous cover is maintained. These less intense forest management practices are favoured because of the greater resistance of forest to biotic and abiotic damages (Knoke, 2009). The biomass export in high thinning, a low intensity forest management, leads to limited biomass ($53000\text{--}14000\text{ kg ha}^{-1}$) and nutrient exports ($0.12\text{--}220\text{ kg}$, Fig. 5.3). However, effects of low intensity forest management will be comparable to nutrient exports in high intensity forest management as the frequency of the low forest management is higher, diminishing possible advantages for forest nutrient balances.

Regardless of harvest intensity, we expected the species in our study to have large export differences since they differ in biomass growth, biomass distribution and nutrient concentrations. Our results confirm earlier work on Douglas fir, showing higher biomass yield than beech and Scots pine on well-drained nutrient poor sites (Fig. 5.3) (Bastien, 2019; Thomas et al., 2022). Despite this higher yield, nutrient export in all harvest intensities was generally lower compared to the other two species. Due to the low nutrient export, there is no direct threat of negative nutrient balances for Douglas fir timber harvest. For example, loss of base cations can be recovered within 9 years after a stem only high thinning up to 42 years after a stem only clearcut (Table 5.4, Table 5.6). Contrary, negative nutrient balances were reported previously for Douglas fir stands (Ranger et al., 2002; de Vries et al., 2021), with more negative balances during stand development due to higher nutrient uptake and leaching (Ranger et al., 2002). The nutrient dynamics during stand development were not considered in this study. In the nutrient export balance, the annual uptake of nutrients in foliage was not considered, although this short-term uptake flux is known to result in negative balances (Table 5.6). It is therefore possible that nutrient budgets become temporarily negative during stand development following harvest which may decrease tree growth.

Timber harvest in beech and Scots pine is likely to impede forest nutrition within two final felling's using common rotation periods. These final felling's could be either two clearcut harvests or a series of thinning's, both resulting in the harvest of all stems. Timber harvest, regardless of harvest intensity, in

both beech and Scots pine resulted in negative balances of base cations with no natural refill of exported Ca due to the negative external supply (Table 5.6). To fully replace the loss of the other base cations in beech, rotation periods of 70 years should be used for a stem only high thinning up to a rotation period of 420 years for a stem only clearcut. These rotation periods can be calculated by dividing the nutrient export (Fig. 5.3) by the external nutrient supply (Table 5.6). For Scots pine, the Mg and K stock will be recovered within 30 years following clearcut harvest up to 60 years following stem only clearcut harvest. Although deep layer uptake could provide another nutrient influx for beech, this influx may have only a limited effect on tree nutrition (Berger et al., 2006; van der Heijden et al., 2015). Negative nutrient budgets for base cations due to harvest were reported previously (Růžek et al., 2019; de Vries et al., 2021), together with possible P limitation (de Vries et al., 2021). We did not find risks of declining P stocks as P stocks of all species following stem only harvest in a clearcut can recover within 50 years considering leaching, weathering (de Vries et al., 2021) and deposition (RIVM, 2016). Negative balances of Ca and P have been found previously (Vangansbeke et al., 2015; de Vries et al., 2021) as well as negative balances of K and Mg which were linked to elevated S deposition (Schaaf et al., 1995) and risks for negative K balances were mentioned for Scandinavia (Palviainen and Finer, 2012). Finally, we predict that harvest in beech stands leads to negative nutrient balances, regardless of harvest intensity. We also have indications that the nutrient balances following stem only harvest in Scots pine seem solely hampered by negative Ca inputs. The annual decreasing soil nutrients stocks, resulting from negative external nutrient input, imply that additional measures are necessary to counteract the loss of base cations if trees will be harvested in these forests in the long term.

4.5 Effect of biomass harvest method

The biomass harvest method, i.e., harvest of crown materials in case of whole tree harvest (WTH) or stripping the bark in case of wood only harvest (WOH), strongly influenced the nutrient export by harvest but had relatively small effects on biomass exports. The highest biomass gain (beech: 140%, taking stem only harvest as the 100%-reference) after WTH was accompanied by nutrient losses up to 310%. The biomass gain following whole tree harvest for Scots pine (130%) is in line with earlier reports for Scots pine (Palviainen and Finer, 2012; Węgiel et al., 2018) but the export in beech in this study was higher which could be caused by wider crowns (Göttlein et al., 2012; Ulbricht et al., 2016). Whole tree harvest caused considerable increase in base cation (150-190%) and P (190-280%) export. These increased losses of scarce nutrients will cause more negative nutrient balances or unrealistic long rotation periods for both beech and Scots pine. Whole tree harvest in Douglas fir seems more sustainable, since base cation losses due to whole tree harvest are replenished by external nutrient input within 70 years. However, because of low stocks of especially K and Mn in the organic soil layers, there seems hardly any nutrient buffer in the

system to recover from disturbances. Such disturbances impact soil nutrient stocks as, for example, leaching temporarily increases after harvest (Katzensteiner, 2003; Rothe and Mellert, 2004; Gundersen et al., 2006; Piirainen et al., 2007). Therefore, although nutrient stocks can be replenished by external nutrient supply assuming stable conditions, the poor soil nutrient buffers for base cations, Mn and P makes whole tree harvest potentially unsustainable and a risk for nutrition and reduced growth (Thiffault et al., 2011) within a single rotation period for such forests on poor and acidified soils.

Wood only harvest, thus removing stem without stem bark, resulted in a 6-10% lower biomass export (again compared to stem only harvest as the 100%-reference) but saves up to 50% of the base cation and P export. Highest reductions in nutrient exports were shown for Douglas fir while reductions were lowest for beech (Fig. 5.4). Wood only harvest will lower the base cation export with 10% to 60% and the P export by 30 to 50% compared to stem only harvest. High stocks of base cations in the bark are observed across multiple species, including an up to 50% decrease of base cation export following wood only harvest (Andre et al., 2010; Achat et al., 2015). Although this study confirms that leaving the bark in the forest is a sustainable management practice (Pyttel et al., 2015; Manolis et al., 2019), the effects differ per nutrient and per species. Wood only harvest will allow rotation periods of 50 years following a clearcut in Douglas fir but cannot counteract the negative external nutrient input in beech and Scots pine, indicating that harvest in beech and Scots pine will still lead to negative Ca balances. However, wood only harvest can prevent depletion of soil K and Ca stocks in both beech and Douglas fir which has previously been observed for coppice oak systems (Pyttel et al., 2015). Furthermore, wood only harvest will keep the main stock of micronutrients in the forest (Manolis et al., 2019). We strongly advocate to shift conventional stem only harvest to wood only harvest. In-situ debarking has been done for Spruce where 91% of the bark was left in the forest in the final felling (Mergl et al., 2021). High debarking efficiencies using harvesters were reported in multiple studies, concluding that in-situ debarking is a potential addition to existing harvesting methods (Heppelmann et al., 2019; Holzleitner and Kanzian, 2022). Debarking of trees may therefore not lead to technical impossibilities but will considerably improve nutrient balances for forests on low-fertile soils.

5 Conclusions

Many forests occur on acidified, poor and well-drained forest soils, i.e., soils with low base saturation that are at risk of base cation and Mn limitation. For 15 Dutch forest stands on such soils, we measured nutrient stocks for macro- and micronutrients and calculated potential limitations in nutrient supply for forest recovery. The aboveground base cation and Mn stocks are generally larger than the soil stocks, posing an immediate threat to forest nutrition if the trees are harvested. Even without harvest, negative external nutrient inputs like the negative input of Ca in both beech and Scots pine forests poses threats to forest

growth and vitality. This negative external Ca balance limits also timber harvest in both beech and Scots pine as exported nutrients cannot be recovered, resulting in a depletion of the Ca nutrient stock within 2 final fellings. Contrary, timber harvest in Douglas fir stands will not result in negative base cation balances using rotation periods of 80 years as base cation losses by harvest are fully replaced by external input. However, temporary negative nutrient balances resulting in lower growth might occur as aboveground K and Mn stocks are larger than the soil stocks. We show that on such nutrient poor forest soils, whole tree harvest should be avoided as harvesting crown materials results in negative nutrient balances, with particularly base cation nutrient removal exceeding the base cation nutrient stocks. Whole tree harvest also resulted in extraordinary high export losses of P, which may limit future forest growth. Instead, wood only harvest, where the bark is stripped in the field, may pose opportunities for sustainable biomass harvest as it can conserve up to 50% of the nutrients in the system compared to regular timber harvest. We therefore recommend that use of such forests on acidified, poor soils will be limited to low intensity harvesting, and recommend debarking trees to conserve large quantities of nutrients within the forest system upon harvest.

Acknowledgements

This research is part of the Nutrient Balance project and was funded by the Dutch Research Council (NWO, No. ALWGS.2017.004). The contribution of M. Valtera was supported from the European Regional Development Fund (project no. CZ.02.2.69/0.0/0.0/16_027/0007953). We acknowledge Henk van Roekel, Leo Goudzwaard and many other members of the harvest team for their valuable help with establishing the experiment, the harvest and the sample processing. We thank our partners, National Forest Service, Union of private Forest Owner Groups, Het Loo Royal Estate, Staro nature and countryside, Borgman management consultants, National Park de Hoge Veluwe and Blom Ecology for financial support, permission to work in their forest or other provided services.

Supplementary information

Table S5.1 Basal area ($\text{m}^2 \text{ha}^{-1}$) removed in the timber harvest intensities high-thinning, shelterwood and clearcut in beech, Douglas fir and Scots pine. Target basal areas per harvest intensity and the reduction percentage are in Table 5.1.

Stand	Species	High-thinning	Shelterwood	Clearcut
		$\text{m}^2 \text{ha}^{-1}$	$\text{m}^2 \text{ha}^{-1}$	$\text{m}^2 \text{ha}^{-1}$
1	Beech	4.0	19	24
2	Beech	5.7	18	29
3	Beech	5.0	18	19
4	Beech	2.9	16	20
5	Beech	5.5	24	26
1	Douglas fir	4.7	22	22
2	Douglas fir	7.4	23	32
3	Douglas fir	6.4	20	35
4	Douglas fir	4.9	27	31
5	Douglas fir	8.6	32	40
1	Scots pine	1.8	16	21
2	Scots pine	4.6	15	20
3	Scots pine	2.6	18	22
4	Scots pine	2.8	21	25
5	Scots pine	6.8	26	25

Table S5.2 Overview of the specific stand models for the nested allometric relationships in needles, small branches, coarse branches, bark and sapwood. The population models are described in Table 5.3.

Treepart	Species	Stand	Model
Needle	Scots pine	1	$\ln(NF) = -5.71 + 2.29 * \ln(DBH)$
		2	$\ln(NF) = -5.15 + 2.29 * \ln(DBH)$
		3	$\ln(NF) = -5.46 + 2.29 * \ln(DBH)$
		4	$\ln(NF) = -5.81 + 2.29 * \ln(DBH)$
		5	$\ln(NF) = -5.07 + 2.29 * \ln(DBH)$
Small branches	Scots pine	1	$\ln(SB) = -5.68 + 2.47 * \ln(DBH)$
		2	$\ln(SB) = -5.03 + 2.47 * \ln(DBH)$
		3	$\ln(SB) = -5.46 + 2.47 * \ln(DBH)$
		4	$\ln(SB) = -5.74 + 2.47 * \ln(DBH)$
		5	$\ln(SB) = -5.05 + 2.47 * \ln(DBH)$
Coarse branches	Scots pine	1	$\ln(CB) = -13.95 + 4.86 * \ln(DBH)$
		2	$\ln(CB) = -7.40 + 3.22 * \ln(DBH)$
		3	$\ln(CB) = -10.46 + 4.00 * \ln(DBH)$
		4	$\ln(CB) = -8.25 + 3.40 * \ln(DBH)$
		5	$\ln(CB) = -8.37 + 3.49 * \ln(DBH)$
Bark	Beech	1	$\ln(BA) = -2.18 + 1.61 * \ln(DBH)$
		2	$\ln(BA) = -3.52 + 1.97 * \ln(DBH)$
		3	$\ln(BA) = -4.06 + 2.17 * \ln(DBH)$
		4	$\ln(BA) = -5.95 + 2.70 * \ln(DBH)$
		5	$\ln(BA) = -5.78 + 2.58 * \ln(DBH)$
Sapwood	Beech	1	$\ln(SW) = -1.72 + 2.27 * \ln(DBH)$
		2	$\ln(SW) = -0.41 + 1.88 * \ln(DBH)$
		3	$\ln(SW) = -1.75 + 2.29 * \ln(DBH)$
		4	$\ln(SW) = -2.83 + 2.59 * \ln(DBH)$
		5	$\ln(SW) = -3.72 + 2.83 * \ln(DBH)$

Table S5.3 The DBH and the annual diameter increase (ADI) of sampled trees of beech, Douglas fir and Scots pine. The annual diameter increase is calculated as the average of the diameter increase (%) in the period 2009-2018. The ADI of Douglas fir (t-test: t value = -5.3, p = 0.004) and Scots pine (t-test: t value = -6.4, p = 0.002) was related to the DBH.

Stand	Beech		Douglas fir		Scots pine	
	DBH cm	ADI %	DBH cm	ADI %	DBH cm	ADI %
1	49	1.9	57	1.1	32	1.0
1	36	0.7	51	2.3	26	1.4
1	27	1.0	47	1.4	17	0.2
2	60	0.9	62	2.0	32	3.0
2	47	0.5	44	1.4	23	1.7
2	39	1.8	33	1.7	15	1.5
3	48	1.2	69	0.5	27	2.8
3	36	1.0	53	1.1	18	2.2
3	27	0.8	37	1.5	13	1.8
4	57	0.8	68	1.1	41	2.5
4	36	0.4	50	2.1	33	1.4
4	27	1.6	28	2.0	19	0.8
5	29	2.5	59	1.6	37	3.0
5	17	0.7	43	1.4	27	0.7
5	13	1.7	28	1.0	19	0.8
Average ± se	37 ± 3.5	1.5 ± 0.32	49 ± 3.4	1.3 ± 0.25	25 ± 2.2	2.5 ± 0.38

Table S5.4 Stocks in aboveground biomass compartments and in forest-floor dry mass of macro (N, Ca, K, Mg, S and P) and micro (Mn, Zn, Fe and Cu) nutrients (kg ha⁻¹) for European beech, Douglas fir and Scots pine prior to harvest. Percentages indicating the relative nutrient stock of the aboveground biomass compared to the forest-floor were calculated as the above-ground tree stock divided by the stocks in the organic layers.

Scots pine																			
Douglas fir										Beech									
Nutrient	Site	Beech					Douglas fir					Scots pine					%		
		Foliage kg ha ⁻¹	Branch kg ha ⁻¹	Bark kg ha ⁻¹	Wood kg ha ⁻¹	Organic layers kg ha ⁻¹	%	Foliage kg ha ⁻¹	Branch kg ha ⁻¹	Bark kg ha ⁻¹	Wood kg ha ⁻¹	Organic layers kg ha ⁻¹	%	Foliage kg ha ⁻¹	Branch kg ha ⁻¹	Bark kg ha ⁻¹		Wood kg ha ⁻¹	Organic layers kg ha ⁻¹
N	1	86	200	47	280	800	77	67	64	73	210	1300	33	51	56	29	87	1200	19
	2	92	260	55	280	1400	49	80	72	79	250	1100	44	78	100	28	84	1200	24
	3	81	230	66	290	1700	40	67	69	69	260	1000	46	45	59	27	88	830	26
	4	80	210	55	280	660	94	61	69	72	220	1100	36	59	88	35	130	1200	25
	5	85	160	32	160	1000	43	120	95	100	280	700	88	96	120	38	150	710	57
Ca	1	33	160	160	110	87	537	13	28	38	43	150	83	5.9	17	20	43	95	91
	2	36	130	110	85	120	289	8	26	25	36	110	86	11	33	28	37	120	91
	3	32	87	110	100	140	233	15	33	28	42	110	103	6.6	20	22	40	77	115
	4	31	220	320	130	170	411	11	28	29	41	120	88	5.0	24	30	45	110	95
	5	33	64	51	58	96	214	43	61	93	61	160	166	18	44	34	60	61	256
K	1	25	69	14	140	27	917	17	28	31	33	32	347	10	18	8.2	38	29	263
	2	27	83	16	130	44	579	14	29	25	38	23	453	17	36	8.8	36	36	274
	3	24	66	17	170	45	629	16	28	26	35	24	446	11	20	7.6	28	24	271
	4	23	82	18	190	25	1265	10	19	26	30	22	388	12	26	9.4	46	30	309
	5	25	52	7.4	69	38	401	15	26	28	34	15	702	34	47	13	46	21	657
Mg	1	5.3	18	3.2	42	22	308	4.6	5.6	7.0	9.0	40	66	1.7	5.0	2.7	14	25	93
	2	5.7	22	5.1	34	30	220	3.8	7.5	7.1	9.6	26	110	3.0	8.4	2.7	12	25	102
	3	5.0	16	4.7	29	30	183	4.2	6.4	5.9	9.2	26	99	2.0	6.8	3.0	12	23	104
	4	4.9	23	4.3	77	19	574	4.8	6.3	7.4	8.9	29	94	1.6	6.5	2.8	13	23	103
	5	5.3	8.3	1.4	12	21	131	4.6	5.8	7.1	8.9	13	202	3.3	9.9	3.2	14	11	266

Continued on the next page.

S	1	6.0	11	2.6	12	36	86	4.7	4.4	6.0	7.4	60	38	2.8	4.7	2.5	5.1	48	32
	2	6.5	15	2.8	10	61	56	5.2	5.2	6.3	9.6	52	51	4.9	7.0	2.2	4.2	59	31
	3	5.7	12	3.4	14	69	51	4.5	5.0	5.2	7.6	50	45	2.7	3.8	2.0	3.5	34	35
	4	5.6	12	3.3	15	38	94	4.4	5.0	6.2	8.1	55	43	3.2	5.6	2.7	5.3	57	29
	5	6.0	9.2	2.3	7.7	45	57	8.4	5.7	8.1	11	43	77	8.6	8.4	3.0	6.3	33	80
P	1	4.3	9.6	2.0	8.0	27	90	4.4	5.2	5.9	4.2	43	46	2.5	3.6	1.5	2.8	31	34
	2	4.6	7.4	1.8	7.0	38	54	3.2	4.8	4.4	5.2	34	52	4.8	5.7	1.5	2.6	42	35
	3	4.0	8.5	2.4	7.5	44	51	3.4	5.2	4.3	5.4	34	54	3.1	3.6	1.6	2.4	29	37
	4	3.9	13	2.4	14	22	151	3.3	5.1	5.0	4.3	37	47	3.1	4.7	1.6	3.3	37	34
	5	4.2	7.9	1.6	5.4	33	58	5.2	6.0	6.0	5.8	27	85	8.4	9.2	2.7	6.2	26	101
Mn	1	4.2	5.7	2.1	8.0	2.7	754	0.67	0.89	0.87	1.3	2.2	166	0.45	0.72	0.41	2.5	1.8	227
	2	4.5	5.1	1.9	5.4	3.7	462	2.4	3.0	2.5	6.1	3.9	357	1.0	1.6	0.55	2.8	2.9	203
	3	4.0	2.8	1.5	4.0	2.4	515	2.8	2.8	2.2	4.9	4.1	314	0.84	1.3	0.48	3.4	2.0	299
	4	3.9	7.5	3.3	9.0	3.7	641	0.46	0.85	0.77	1.3	2.1	157	0.27	0.56	0.27	1.5	1.5	173
	5	4.2	1.7	0.6	2.1	1.6	539	1.8	1.5	1.7	1.9	3.1	225	1.0	1.5	0.52	2.9	1.3	453
Zn	1	0.10	0.41	0.053	0.47	0.86	121	0.044	0.34	0.44	1.2	1.6	123	0.15	0.36	0.24	0.92	2.1	81
	2	0.11	0.41	0.071	0.48	1.9	57	0.071	0.48	0.49	0.99	1.7	121	0.16	0.63	0.20	0.62	2.2	74
	3	0.094	0.92	0.20	1.6	2.2	125	0.085	0.43	0.46	0.55	1.3	115	0.13	0.39	0.19	1.0	1.6	104
	4	0.093	0.72	0.078	0.64	1.2	127	0.059	0.47	0.48	0.69	2.0	85	0.14	0.42	0.21	0.80	2.2	71
	5	0.10	0.87	0.17	0.83	5.5	36	1.4	3.7	5.7	6.5	2.0	87	0.76	2.3	0.63	2.5	1.3	49
Fe	1	0.32	0.79	0.22	0.66	32	6	0.36	0.60	0.67	0.40	41	5	0.16	0.74	0.14	0.20	37	3
	2	0.34	2.4	0.22	0.26	57	6	0.47	1.4	0.73	0.53	53	6	0.31	0.76	0.16	0.16	60	2
	3	0.30	1.3	0.45	0.47	63	4	0.39	0.68	0.62	0.61	41	6	0.17	0.71	0.17	0.79	24	8
	4	0.30	1.3	0.32	0.29	39	6	0.60	1.3	0.61	0.56	53	6	0.25	0.61	0.18	0.37	41	3
	5	0.32	1.1	0.22	1.3	33	9	0.66	1.2	1.5	1.1	47	10	0.51	1.0	0.27	0.39	35	6
Cu	1	0.026	0.13	0.023	0.14	0.24	129	0.014	0.091	0.057	1.0	0.47	253	0.014	0.075	0.015	0.27	0.34	109
	2	0.028	0.12	0.030	0.11	0.54	55	0.018	0.095	0.094	0.32	0.56	93	0.017	0.13	0.016	0.20	0.44	82
	3	0.024	0.17	0.039	0.20	0.76	58	0.014	0.077	0.069	0.16	0.44	68	0.010	0.065	0.017	0.21	0.24	124
	4	0.024	0.16	0.032	0.16	0.62	61	0.013	0.072	0.072	0.16	0.65	49	0.012	0.059	0.021	0.08	0.52	33
	5	0.026	0.29	0.019	0.13	0.68	68	0.024	0.13	0.15	0.17	0.70	67	0.024	0.14	0.026	0.22	0.48	84

Table S5.5 Average thickness ($\pm$ standard deviation) of the litter layer, fragmented layer and humified layer (mm) and the total thickness of the organic layer for each site of European beech, Douglas fir and Scots pine prior to harvest.

Site	Species	Litter layer	Fragmented layer	Humified layer	Organic layer
1	Beech	24 $\pm$ 8.3	30 $\pm$ 13	37 $\pm$ 19	91 $\pm$ 25
2	Beech	27 $\pm$ 9.7	25 $\pm$ 8.1	45 $\pm$ 13	97 $\pm$ 18
3	Beech	24 $\pm$ 9.0	43 $\pm$ 13	44 $\pm$ 17	110 $\pm$ 24
4	Beech	26 $\pm$ 24	11 $\pm$ 10	24 $\pm$ 18	62 $\pm$ 47
5	Beech	38 $\pm$ 7.5	38 $\pm$ 15	51 $\pm$ 29	130 $\pm$ 33
1	Douglas fir	18 $\pm$ 14	39 $\pm$ 14	39 $\pm$ 16	96 $\pm$ 24
2	Douglas fir	20 $\pm$ 8.3	29 $\pm$ 14	31 $\pm$ 14	80 $\pm$ 24
3	Douglas fir	20 $\pm$ 12	26 $\pm$ 11	38 $\pm$ 14	84 $\pm$ 28
4	Douglas fir	17 $\pm$ 4.1	21 $\pm$ 9.6	40 $\pm$ 20	79 $\pm$ 28
5	Douglas fir	17 $\pm$ 9.4	13 $\pm$ 7.0	19 $\pm$ 13	49 $\pm$ 23
1	Scots pine	15 $\pm$ 4.1	33 $\pm$ 15	44 $\pm$ 11	92 $\pm$ 19
2	Scots pine	14 $\pm$ 5.6	25 $\pm$ 10	51 $\pm$ 25	90 $\pm$ 27
3	Scots pine	16 $\pm$ 5.1	22 $\pm$ 7.7	30 $\pm$ 11	68 $\pm$ 13
4	Scots pine	22 $\pm$ 5.7	32 $\pm$ 9.4	49 $\pm$ 16	100 $\pm$ 23
5	Scots pine	22 $\pm$ 8.9	30 $\pm$ 10	34 $\pm$ 19	86 $\pm$ 23

Table S5.6 Comparison of the aboveground stock and the organic layer stock of N, Ca, K, Mg, S, P, Mn, Zn, Fe and Cu based on 5 stands per species (df = 5). Paired T-test t and P values are presented.

Species	N	Ca	K	Mg	S	P	Mn	Zn	Fe	Cu
Beech	-2.8*	4.2*	8.0**	3.1*	-2.7 ^{n.s.}	-1.2 ^{n.s.}	6.5**	-0.65 ^{n.s.}	-6.6**	-2.6 ^{n.s.}
Douglas fir	-4.3*	0.59 ^{n.s.}	40***	0.21 ^{n.s.}	-5.8**	-4.9**	2.9*	-0.61 ^{n.s.}	-18***	-0.032 ^{n.s.}
Scots pine	-5.8**	0.82 ^{n.s.}	4.9**	1.0 ^{n.s.}	-4.4*	-3.7*	4.9**	-1.3 ^{n.s.}	-6.4**	-1.2 ^{n.s.}

*** $P \leq 0.001$, ** $0.01 \leq P < 0.001$ * $0.05 \leq P < 0.01$, ^{n.s.} $P > 0.05$.

Table S5.7 Total biomass (kg ha⁻¹) and annual biomass increment (kg ha⁻¹ yr⁻¹) of foliage, branches, bark and wood in beech, Douglas fir and Scots pine. The F value and significance of the two-way Anova based on species, tree compartment (foliage, branch, bark and wood) and the interaction between species and tree part are displayed for the model based on annual biomass and the annual biomass increment. Different capital letters denote significant differences among species for the different tree compartments, small letters denote significant differences between tree compartments within a tree species, both according to Tukey's *posthoc* test with a significance level of $P < 0.05$.

	Beech			Douglas fir			Scots pine		
	Biomass kg ha ⁻¹	Biomass increment kg ha ⁻¹ yr ⁻¹		Biomass kg ha ⁻¹	Biomass increment kg ha ⁻¹ yr ⁻¹		Biomass kg ha ⁻¹	Biomass increment kg ha ⁻¹ yr ⁻¹	
Foliage	3500 ± 90 ^{Aa}	82 ± 12 ^{Aa}		3500 ± 260 ^{Aa}	78 ± 9.3 ^{Aa}		3400 ± 650 ^{Aa}	130 ± 27 ^{Aa}	
Branch	51000 ± 3100 ^{Bc}	1200 ± 120 ^{Bc}		22000 ± 1100 ^{Ab}	650 ± 80 ^{Ac}		18000 ± 2600 ^{Ab}	750 ± 150 ^{Ab}	
Bark	7900 ± 800 ^{Ab}	200 ± 20 ^{Ab}		20000 ± 1130 ^{Bb}	580 ± 68 ^{Cb}		6000 ± 330 ^{Aa}	200 ± 21 ^{Ba}	
Wood	130000 ± 11000 ^{Bd}	3600 ± 440 ^{Ad}		190000 ± 7500 ^{Cc}	6700 ± 790 ^{Bd}		73000 ± 5300 ^{Ac}	2800 ± 260 ^{Ac}	
Total average	190000 ± 14000 ^A	5100 ± 560 ^A		230000 ± 10000 ^B	8000 ± 940 ^B		100000 ± 8200 ^A	3900 ± 430 ^A	
Anova	Biomass kg ha ⁻¹	Biomass increment kg ha ⁻¹ yr ⁻¹		Biomass kg ha ⁻¹	Biomass increment kg ha ⁻¹ yr ⁻¹		Biomass kg ha ⁻¹	Biomass increment kg ha ⁻¹ yr ⁻¹	
F-Value P-Value	Species	Species		Tree part	Tree part		Interaction	Interaction	
	12 < 0.001	9.9 < 0.001		42 E ⁻¹ < 0.001	32 E ⁻³ < 0.001		60 < 0.001	15 < 0.001	

Table S5.8 Estimated annual nutrient uptake rates over the period 2008–2018 ($\text{kg ha}^{-1} \text{ yr}^{-1}$) in the foliage, branches, stem bark and stem wood for European beech, Douglas fir and Scots pine. The averaged data per species is shown in figure 5.2. The soil nutrient concentrations depict the available nutrient concentration in the top layer of the mineral soil (0 – 30 cm depth) in winter 2018–2019 before harvest.

Scots pine																
Douglas fir										Beech						
	Site	Foliage kg ha ⁻¹ yr ⁻¹	Branch kg ha ⁻¹ yr ⁻¹	Bark kg ha ⁻¹ yr ⁻¹	Wood kg ha ⁻¹ yr ⁻¹	Soil kg ha ⁻¹	Foliage kg ha ⁻¹ yr ⁻¹	Branch kg ha ⁻¹ yr ⁻¹	Bark kg ha ⁻¹ yr ⁻¹	Wood kg ha ⁻¹ yr ⁻¹	Soil kg ha ⁻¹	Foliage kg ha ⁻¹ yr ⁻¹	Branch kg ha ⁻¹ yr ⁻¹	Bark kg ha ⁻¹ yr ⁻¹	Wood kg ha ⁻¹ yr ⁻¹	Soil kg ha ⁻¹
N	1	2.2	5.1	1.0	8.0	32	1.5	1.7	2.0	7.5	46	1.2	1.5	0.69	2.5	27
	2	1.6	4.4	0.94	4.7	37	2.3	2.4	2.9	12	72	4.2	5.9	1.3	4.5	55
	3	1.6	4.6	1.5	7.3	52	0.90	1.2	1.3	5.8	49	2.4	2.7	1.3	4.9	49
	4	1.3	3.1	1.1	5.4	47	1.5	2.1	2.4	9.5	56	2.1	3.5	1.0	4.6	47
	5	2.5	4.8	1.3	7.6	43	2.7	2.7	3.0	12	49	3.4	5.2	1.2	4.8	52
Ca	1	0.84	3.9	3.2	3.2	2.0	0.27	0.70	1.0	1.5	2.6	0.16	0.49	0.43	1.0	1.8
	2	0.62	2.5	2.0	1.4	2.4	0.22	0.76	0.77	1.4	6.5	0.54	1.8	1.2	1.8	5.5
	3	0.64	1.8	2.7	2.5	2.2	0.20	0.56	0.52	0.88	6.0	0.36	0.78	0.93	2.1	5.0
	4	0.49	3.4	6.3	2.4	18	0.26	0.78	0.94	1.6	5.6	0.18	0.92	0.84	1.5	2.3
	5	0.96	1.8	1.8	2.3	1.2	0.89	1.4	2.5	2.0	23	0.56	1.7	0.90	1.9	5.5
K	1	0.63	1.9	0.29	4.0	16	0.36	0.82	0.87	1.2	14	0.28	0.58	0.22	1.0	11
	2	0.47	1.5	0.27	2.0	14	0.36	1.1	0.80	1.5	23	0.74	2.2	0.43	2.0	32
	3	0.48	1.4	0.40	4.1	21	0.24	0.50	0.50	0.72	21	0.57	0.95	0.34	1.6	36
	4	0.37	1.3	0.35	3.6	56	0.27	0.61	0.97	1.2	23	0.39	1.0	0.27	1.7	24
	5	0.73	1.6	0.32	3.5	26	0.35	0.75	0.81	1.4	27	1.0	1.9	0.38	1.6	29
Mg	1	0.13	0.46	0.064	1.3	5.0	0.10	0.16	0.20	0.31	2.3	0.039	0.14	0.073	0.41	1.5
	2	0.10	0.39	0.090	0.60	4.9	0.11	0.29	0.25	0.41	9.5	0.14	0.45	0.12	0.57	5.4
	3	0.10	0.32	0.11	0.68	6.3	0.057	0.11	0.11	0.20	8.4	0.10	0.29	0.13	0.66	11
	4	0.078	0.35	0.087	1.3	22	0.12	0.18	0.27	0.34	6.9	0.057	0.26	0.078	0.45	2.2
	5	0.15	0.26	0.058	0.56	2.4	0.11	0.15	0.19	0.28	7.4	0.10	0.41	0.097	0.44	3.9

Continued on the next page.

S	1	0.15	0.29	0.053	0.37	14	0.10	0.12	0.16	0.26	13	0.069	0.13	0.054	0.12	12
	2	0.11	0.25	0.049	0.18	16	0.15	0.16	0.21	0.40	38	0.22	0.42	0.10	0.23	23
	3	0.12	0.24	0.075	0.34	22	0.062	0.088	0.099	0.16	24	0.14	0.19	0.090	0.19	16
	4	0.089	0.19	0.065	0.29	19	0.11	0.15	0.21	0.33	28	0.11	0.22	0.076	0.18	20
	5	0.17	0.29	0.089	0.35	19	0.18	0.15	0.25	0.41	37	0.24	0.36	0.091	0.24	23
P	1	0.11	0.23	0.040	0.22	3.4	0.10	0.14	0.16	0.14	7.0	0.065	0.11	0.039	0.074	3.0
	2	0.080	0.13	0.033	0.12	2.9	0.086	0.15	0.14	0.19	6.4	0.22	0.34	0.072	0.15	6.2
	3	0.081	0.17	0.055	0.18	4.9	0.049	0.094	0.090	0.11	3.9	0.16	0.18	0.071	0.13	3.6
	4	0.063	0.19	0.048	0.26	3.0	0.084	0.15	0.18	0.18	13	0.11	0.18	0.047	0.12	9.8
	5	0.12	0.22	0.059	0.24	7.1	0.12	0.15	0.17	0.22	6.4	0.27	0.40	0.085	0.24	6.3
Mn	1	0.11	0.14	0.041	0.22	0.16	0.014	0.025	0.025	0.044	0.075	0.014	0.024	0.013	0.078	0.025
	2	0.079	0.096	0.035	0.091	0.14	0.068	0.11	0.087	0.27	1.1	0.051	0.079	0.024	0.12	0.080
	3	0.081	0.056	0.032	0.094	0.12	0.039	0.051	0.042	0.11	0.89	0.043	0.048	0.020	0.17	0.47
	4	0.062	0.12	0.064	0.17	2.0	0.011	0.026	0.029	0.049	0.19	0.0098	0.022	0.0076	0.053	0.044
	5	0.12	0.049	0.022	0.087	0.04	0.039	0.042	0.055	0.068	1.3	0.032	0.057	0.014	0.091	0.13
Zn	1	0.0025	0.0094	0.00095	0.015	0.25	0.0010	0.0095	0.013	0.040	0.084	0.0041	0.011	0.0060	0.026	0.069
	2	0.0019	0.0071	0.0012	0.0081	0.27	0.0021	0.016	0.015	0.048	0.35	0.0093	0.034	0.0088	0.032	0.29
	3	0.0019	0.017	0.0036	0.036	0.35	0.0012	0.0073	0.0094	0.011	0.35	0.0067	0.017	0.0082	0.054	0.35
	4	0.0015	0.011	0.0016	0.011	0.62	0.0015	0.014	0.015	0.029	0.30	0.0051	0.016	0.0059	0.027	0.12
	5	0.0029	0.024	0.0050	0.030	0.62	0.028	0.088	0.15	0.22	9.4	0.027	0.10	0.019	0.099	2.7
Fe	1	0.0081	0.019	0.0042	0.016	13	0.0080	0.013	0.016	0.012	7.9	0.0046	0.023	0.0026	0.0059	6.6
	2	0.0060	0.061	0.0038	0.0042	17	0.016	0.037	0.030	0.023	32	0.018	0.045	0.0061	0.010	15
	3	0.0061	0.027	0.011	0.0093	18	0.0053	0.011	0.010	0.012	31	0.0093	0.036	0.0088	0.058	25
	4	0.0047	0.019	0.0067	0.0061	27	0.015	0.029	0.020	0.022	17	0.0095	0.024	0.0053	0.013	8.5
	5	0.0093	0.037	0.0088	0.11	5.0	0.015	0.028	0.043	0.049	17	0.016	0.040	0.0076	0.016	18
Cu	1	0.0065	0.0034	0.00050	0.0044	0.023	0.00031	0.0029	0.0016	0.035	0.037	0.00051	0.0027	0.00036	0.011	0.020
	2	0.00048	0.0022	0.00053	0.0019	0.038	0.00048	0.0038	0.0033	0.019	0.033	0.00092	0.0065	0.00066	0.010	0.043
	3	0.00050	0.0034	0.00094	0.0049	0.043	0.00020	0.0013	0.0013	0.0028	0.046	0.00049	0.0027	0.00079	0.010	0.093
	4	0.00038	0.0024	0.00063	0.0030	0.34	0.00031	0.0021	0.0023	0.0072	0.089	0.00044	0.0024	0.00063	0.0028	0.051
	5	0.00075	0.0074	0.00077	0.0058	0.071	0.00054	0.0040	0.0044	0.0069	0.38	0.00079	0.0071	0.00079	0.011	0.12

Table S5.9 Biomass reductions (%) for different harvest intensities (high-thinning, shelterwood and clearcut) and harvest methods (WOH: wood only harvest; SOH: stem only harvest; WTH: whole tree harvest). The reduction percentage is calculated as the biomass exported divided by the total aboveground biomass.

	Beech			Douglas fir			Scots pine		
	WOH	SOH	WTH	WOH	SOH	WTH	WOH	SOH	WTH
High-thinning	13%	14%	19%	17%	18%	21%	12%	13%	17%
Shelterwood	55%	58%	80%	64%	71%	80%	62%	67%	86%
Clearcut	67%	72%	100%	82%	91%	100%	72%	78%	100%

Table S5.10 Wet only deposition rates in the Netherlands ($\text{kg ha}^{-1} \text{yr}^{-1}$) as measured in 2016 in the RIVM rainwater monitoring network for the stations representing the deposition nearby the study sites.

Station	NH ₄	NO ₃	K	Mg	SO ₄	PO ₄	Ca	Zn	Fe	Cu
Biest-Houtakker	8.1	10	0.51	0.70	6.8	0.15	1.1			
De Zilk								0.034	0.11	0.011
Speuld	8.4	13	0.58	0.93	7.3	0.068	1.1			
Vredepeel	8.3	9.7	0.37	0.41	5.1	0.27	1.2	0.067	0.50	0.019
avg	8.3	11	0.49	0.68	6.4	0.16	1.1	0.051	0.31	0.015
s.e.	0.088	1.1	0.062	0.15	0.67	0.059	0.033	0.013	0.16	0.0033

Table S5.11 Conversion factor to calculate total deposition within a forest stand based on bulk precipitation data. Original data underneath the multiplication factors were corrected for canopy uptake of NH_4 and NO_3 and canopy leaching of Mg, Ca, K and Mn except for the inert ions Na, SO_4 , Zn and Cu. The ion Na is in all cases used to calculate canopy leaching of Ca, Mg and K. Stem flow was only included for beech forest and one Douglas fir stand.

Species	Na	NH_4	NO_3	K	Mg	SO_4	PO_4	Ca	Mn	Zn	Cu	Country	Stemflow	Source
Beech	1.2	1.2	1.2	1.2	1.2	1.2	1.3	1.2	1.2			Germany	Yes	(Talkner et al., 2010)
Beech						1.6								(Augusto et al., 2002)
Beech	2.4	3.6	2.0	3.1	2.4	4.8		2.4				Belgium	Yes	(Adriaenssens et al., 2012a)
Beech	1.2					1.6						Czech Republic	Yes	(Růžek et al., 2019)
Beech	1.2					1.2				1.1	2.1	Poland	Yes	(Kowalska et al., 2016)
Douglas fir	2.3	2.3	2.3	2.2	2.2	2.3	2.1	2.4				France	Yes	(Marques et al., 1997)
Douglas fir	2.3			1.4	1.8	3.3		1.5				Netherlands		(Van Ek and Draaijers, 1994)
Douglas fir	2.3	3.7	2.1	2.3	2.3	3.6	2.5	2.3				Netherlands	Yes	(Draaijers et al., 1997a)
Coniferous										2.4	2.6			(De Vries and Bakker, 1996)
Scots pine	1.1			1.2	1.2	1.6		1.1				Poland	No	(Kozłowski et al., 2020)
Scots pine	2.3			1.5	1.9			1.6				Netherlands	No	(Van Ek and Draaijers, 1994)
Scots pine	2.0					1.5				1.1	1.3	Poland	No	(Kowalska et al., 2016)
Corsican pine	1.5	6.5	3.5	3.4	1.9			1.4				Belgium	No	(De Schrijver et al., 2004)
Average														
Beech	1.5	2.4	1.6	2.2	1.8	2.3	1.3	1.8	1.2					
Douglas fir	2.3	3.0	2.2	2.0	2.1	3.1	2.3	2.1		2.4	2.6			
Scots pine	1.7	6.5	3.5	2.0	1.7	1.6		1.4						

N.B. The bulk throughfall deposition of SO_4^{2-} was assumed to be not influenced by canopy exchange as the stomatal uptake of SO_2 is balanced by foliar leaching of SO_4^{2-} (Draaijers and Erisman, 1995). However, Staelens et al. (2007) estimated that canopy leaching contributed 7% to the combined bulk throughfall and stemflow of SO_4^{2-} which was in line with the findings of Potter et al. (1991). Canopy exchange of Al and Cu are neglectable as both elements in deposition is found in a colloidal fraction and almost entirely complexed by DOC (Gandois et al., 2010b). The free metal ion forms of Zn (on average 30%) do interact with the canopy however concentration is only slightly increased or decreased (Gandois et al., 2010b).¹ measured under a single tree, therefore not representative for a forest stand.

Supplementary figures

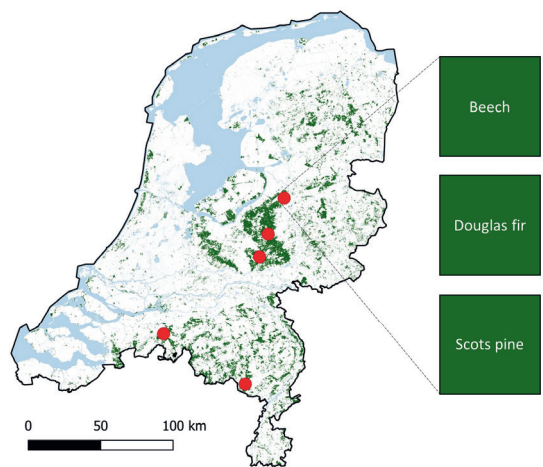


Figure S5.1 Locations of forest stands selected for biomass, carbon and nutrient measurements in this study. The nationwide forest cover (in total 10% of the land area of the Netherlands) is shown in green.

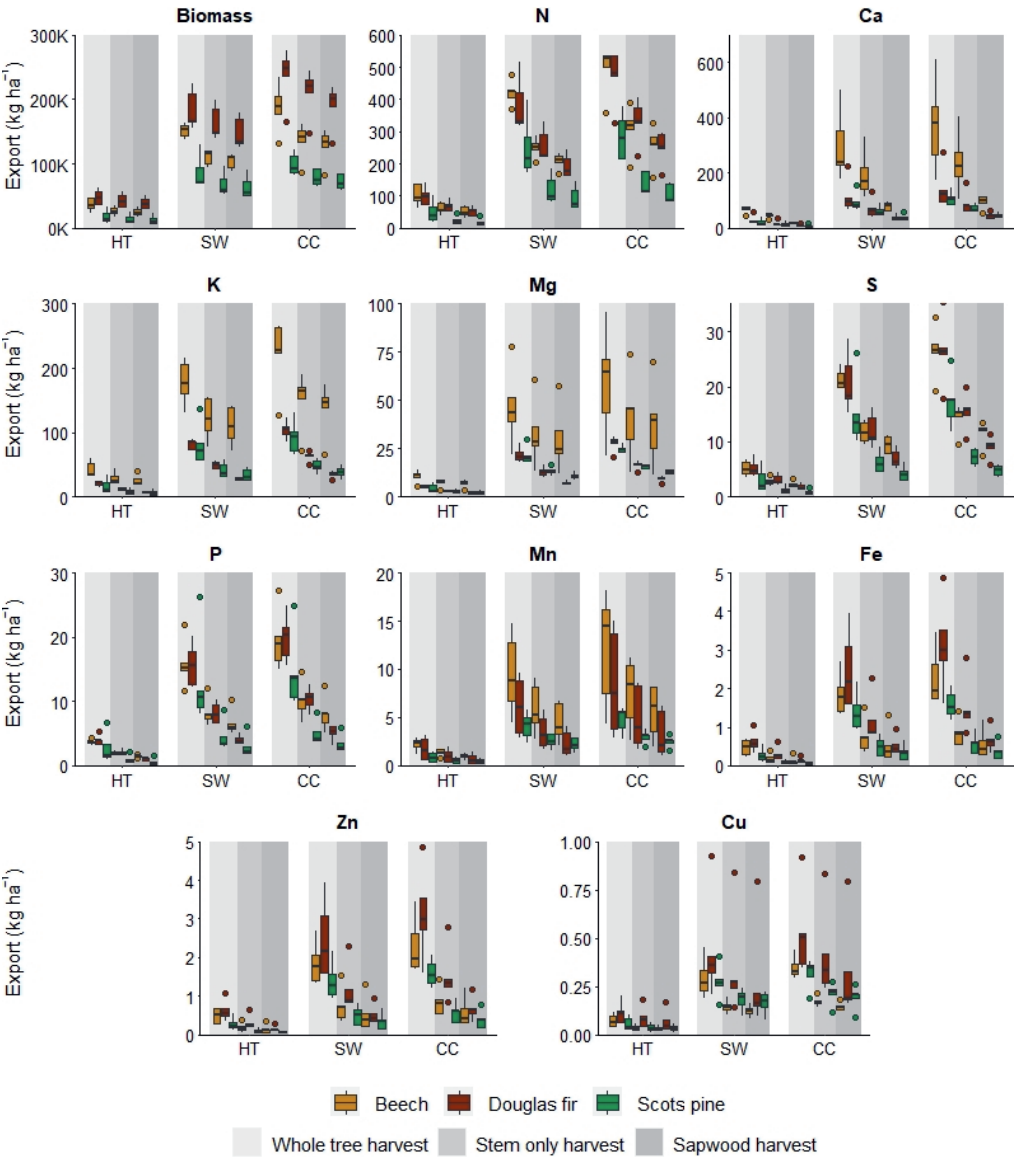


Figure S5.2 Export of biomass, macronutrients (N, P, K, S, Ca and Mg) and micronutrients (Mn, Cu, Fe, Zn) under different harvest scenarios: high thinning (HT), shelterwood (SW) and clearcut (CC) and different harvest intensities: whole tree harvest, stem only harvest and stem wood harvest.



Chapter 6

Effects of forest management on
dissolved nutrient concentrations,
water fluxes and nutrient leaching

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Abstract

The increasing focus on forest timber and biomass production in Europe raises concerns about the sustainability of various harvesting practices, as the forest nutritional status has deteriorated due to enhanced leaching of base cations in response to soil acidification induced by elevated nitrogen (N) and sulfur (S) deposition. In response to this concern, forest nutrient budgets have been employed to ensure that nutrient losses, mainly from harvest and leaching, remain within the limits of nutrient gains from atmospheric deposition and soil weathering. Forest harvesting potentially influences the nutrient leaching through the harvest intensity, harvest method and soil preparation. This study aims to quantify the impact of these forest management practices on post-harvest nutrient leaching, considering three different harvest intensities (high-thinning ~20%, shelterwood ~80%, and clearcut), two harvest methods (stem only and whole tree harvest), and soil preparation (mulching) versus unharvested control plots for three major tree species in the Netherlands (beech, Douglas fir, and Scots pine). The study combines an annual cycle of monthly measurements of dissolved nutrient concentrations over five experimental sites per species with a mechanistic model simulating monthly water fluxes to calculate nutrient leaching.

In the unharvested control plots, nutrient leaching was generally higher in Douglas fir than in Scots pine and beech. A clearcut, and to a lesser extent shelterwood, strongly increased dissolved nutrient concentrations, especially nitrate (NO_3), indicating a rapid mobilization of large N stocks, and to a lesser extent S, associated with accelerated losses of the base cations calcium (Ca), magnesium (Mg) and potassium (K) and of the acid cations aluminum (Al), iron (Fe) and manganese (Mn). Thinning had small effects on leaching, which implies that, at least in the short term, thinning prevents accelerated soil acidification. The effects of harvest methods on leaching appeared to be marginal, and mulching had a negligible impact on post-harvest leaching. Our results highlight the importance of forest structure, affected by harvest intensity and, to a lesser extent, tree species, on nutrient losses via leaching. We call for long term studies, including at least 10 years, to quantify the effects of harvesting practices on forest nutrient balances during a rotation period. These studies are essential since the dynamics in leaching after a final felling (clearcut or shelterwood) or thinning are highly uncertain but essential for understanding long term forest nutrient budgets over a full rotation.

1 Introduction

The production of forest timber and biomass receives increasing attention as forest policies encourage the use of forest products in view of the EU's bioeconomy (Lerink et al., 2023). There is a potential for increasing harvest rates and wood mobilization, according to an updated Bioeconomy Strategy (Strategy, 2018). The sustainability of increased harvest intensity, however, is a major concern as the forest nutritional status has been deteriorating across Europe following soil acidification and climate change (Jonard et al., 2015; Braun et al., 2020a; Penuelas et al., 2020). This concern has promoted the use of forest nutrient budgets, utilizing a mass balance approach aiming to ensure that nutrient losses, primarily through harvest and leaching, do not surpass the nutrient gains from atmospheric deposition and soil weathering (Pare and Thiffault, 2016; de Oliveira Garcia et al., 2018; de Vries et al., 2021). However, this approach has been criticized (Klaminder et al., 2011; Lucas et al., 2014; Pare and Thiffault, 2016; Johnson and Turner, 2019; Löfgren et al., 2021) due to its lack of high quality input data, resulting in substantial uncertainties in estimated nutrient budgets that hinder its ability to guide sustainable forest management practices. Apart from poor data availability, uncertainties are also related to a lack of representation of nutrient dynamics in the post-harvest period, particularly concerning the nutrient losses by leaching, which can be comparable to or greater than the nutrient losses via harvest (Akselsson et al., 2007b; Pare and Thiffault, 2016; de Vries et al., 2021; Löfgren et al., 2021).

Leaching is often highly variable (Akselsson et al., 2007b; Rothwell et al., 2008; De Vries et al., 2010) and controlled by numerous factors including weather conditions, soil type and buffer capacity, acid deposition, tree species and canopy openness (Lovett et al., 2002; Rothe et al., 2002a; Asano et al., 2006; Christiansen et al., 2006; Rothwell et al., 2008; Fröberg et al., 2011; Jiang et al., 2018; Braun et al., 2020b). These factors can form complex interactions that can strengthen or weaken each other. For example, acid deposition negatively influences the soils buffer capacity (Bowman et al., 2008), and tree species composition influences the drainage flux and the nutrient concentrations, with effects differing between sites and across nutrients (Augusto et al., 2002; Rothe et al., 2002a). Furthermore, tree species differ in the interception of acid deposition, causing differences in SO_4 and NO_3 leaching which strongly influences the leaching of base cations such as calcium, potassium and magnesium (De Vries et al., 1995b; De Vries et al., 2007). This effect might further differ between soil types as buffer capacities differ. Many studies focus mainly on N leaching in response to N deposition (Jussy et al., 2004b; Van der Salm et al., 2007a; Dise et al., 2009; Gundersen et al., 2009; Chiwa et al., 2019; Lucander et al., 2021) and large knowledge gaps still exist for leaching of other nutrients than N, including base cations and especially P and micronutrients (Zhou et al., 2018) hampering the use of forest nutrient budgets to guide forest management (de Vries et al., 2021).

Harvest intensity can significantly influence leaching, especially through altered nutrient deposition (**Ch3**), modified water fluxes, reduced nutrient uptake by vegetation and disturbance of the topsoil. Generally, leaching tends to increase with increasing harvest intensity. Experimental results hardly show an increase following a forest thinning (> 80% of the trees remaining) (Gundersen et al., 2006; Phillips and Watmough, 2012), while increased leaching levels have been found following a shelterwood harvest where only sporadic big seed trees are left, to highest levels of leaching following a clearcut harvest (Katzensteiner, 2003; Rothe and Mellert, 2004; Gundersen et al., 2006; Titus et al., 2006; Piirainen et al., 2007). Differences in leaching flux following different harvest intensities can be related to the residual tree cover (Gundersen et al., 2006; Titus et al., 2006) but also to soil fertility, soil structure, seasonality and (micro)climate (Helmisaari and Mälkönen, 1989; Connolly, 1998; Bélanger et al., 2003; Ring, 2007). The duration of the increased leaching flux following harvest strongly depends on the recovery of the vegetation, the soil type and the weather conditions. Because of many interacting factors determining both the increase in leaching following harvest and the duration of this increase, the actual extent of the post-harvest leaching remains ambiguous.

Post-harvest leaching may also be influenced by the harvest method ('stem only harvest' or 'whole tree harvest') and post-harvest soil preparation such as mulching. Residuals from stem-only harvest can lead to elevated decomposition, consequently increasing nutrient leaching (Devine et al., 2012). Mulching disrupts the topsoil, breaking down harvest residues into smaller pieces and thereby enhances the decomposition of these residues, ultimately resulting in increased nutrient losses through post-harvest leaching (Lundmark-Thelin and Johansson, 1997; Piirainen et al., 2007). However, effects of harvest method and soil preparation are difficult to interpret because of interactions with soil type and buffer capacity, (micro)climate and vegetation (Titus et al., 1997; Wall, 2008). For example, mulching was shown to immobilize nutrients but still resulted in elevated leaching in a temperate forest (Pitman and Peace, 2021) while in a tropical forest the faster decomposition after mulching did not increase the losses of nutrients by leaching (Sommer et al., 2004). In forests situated on acidic soils, faster decomposition resulting from stem-only harvest and mulching can significantly increase leaching compared to whole tree harvest, primarily due to a low soil buffer capacity (Bélanger et al., 2003). However, contrasting findings suggest that stem-only harvest may lead to lower nutrient leaching from soils due to nutrient immobilization by microbes (Gundersen et al., 2006). Furthermore, Harvest residues left on the site can hinder the establishment of regeneration while soil preparation may stimulate the establishment of the regeneration (Collins et al., 2011; Rhoades et al., 2020; Kampherbeek et al., 2021). However, when regeneration has established, harvest residues can have a positive influence on seedling survival and growth (Heinemann and Kitzberger, 2006; Rhoades et al., 2020). These conflicting responses underscore

the necessity for more experimental data across diverse forest systems to better comprehend the effects of harvest methods and soil preparation on post-harvest leaching.

The aim of the present paper is to quantify the effect of forest management on post-harvest nutrient leaching as a function of harvest intensity, harvest method and soil preparation for different tree species. We therefore investigated the impact of different harvest intensities, harvest methods and soil preparation in monocultures of three major tree species in the Netherlands: *F. sylvatica*, *P. Menziesii* and *P. sylvestris*. We used a unique forest experiment, in which different harvest intensities were applied and replicated within 15 experimental plots of 47-100 year old forests on poor, acidified soils; a clearcut (100% of trees harvested), shelterwood (~80% of trees harvested), thinning (~20% of trees harvested) and control (no harvest) as described in Vos et al. (2023a) and Vos et al. (2023b). Additionally, two harvest methods (stem only harvest and whole tree harvest) were applied for thinning, shelterwood, and clearcut treatments, and mulching was applied to shelterwood and clearcut. We quantified the dissolved nutrient concentrations in soil solution below the rooting zone on a monthly basis over a full year for the different treatment combinations. We combined the collected data with a mechanistic modelling approach that simulates the water flux to calculate nutrient leaching.

2 Methods

2.1 Study area

The study was carried out in monoculture stands of European beech (*Fagus sylvatica*), Douglas fir (*Pseudotsuga menziesii*) and Scots pine (*Pinus sylvestris*) across the Netherlands with a temperate maritime climate with a mean annual temperature of 10.6°C and a mean long-term annual rainfall of 850 mm (KNMI, 2022). For each of these three species, we established five experimental forest plots distributed in different areas, thus summing to 15 experimental forest plots in total. Selected stands had a minimum size of 1 hectare and represent an extreme case of production forest on low fertile soils receiving high nitrogen deposition inputs. The selected stands were located on acidic sandy soils which classified as Albic or Entic Podzols or Dystric Cambisols within texture classes fine sand to loamy medium sand and were characterized by high nitrogen stocks. Further description of the study area can be found in **Ch1.7** and in (Vos et al., 2023a; Vos et al., 2023b).

2.2 Experimental design

The forest experiment was established in February to April 2019 by implementing the harvest intensities, harvest methods and soil preparation in each stand. First, each stand was divided into four equal plots of 0.25-ha to which the harvest intensity treatments high thinning, shelterwood, clearcut, and unharvested control were randomly assigned. Tree density and basal area were measured on plot level and harvest

intensity was determined based on species-specific target basal areas per treatment, reflecting common management practices in the Netherlands (Table 1.3). Each plot, except the unharvested control, was divided into two equal subplots of 25 * 50 m to which the harvest methods 'stem only harvest' and 'whole tree harvest' were randomly assigned. The shelterwood and clearcut plots were further divided into four equal subplots whereby half of the stem only harvest subplot and half of the whole tree harvest subplot were mulched. Mulching as soil preparation consisted of shallow flail mulching in which the harvest leftovers were shredded and mixed with the top part of the forest floor. An overview of the design including the four harvest intensity plots and the 7 sub-plots related to harvest method and soil preparation is given in Fig. S6.1. The stands were harvested in February and March 2019 and soil preparation was carried out in April 2019.

In January to February 2020, 41 macrorhizons (Rhizosphere Research Products, the Netherlands) were installed in each of the fifteen stands to collect soil solution samples thus summing to 615 sampling sites in total. These macrorhizons consisted of a 2.5 mm rhizon tip with a mean pore size of 0.15 μm mounted to a PVC tube (Fig. 6.1). Macrorhizons were installed at a depth of 50cm in the mineral soil. Prior to macrorhizon installation, a small patch of the forest floor was removed to determine the top of the mineral soil. Macrorhizons were inserted under an angle of 30°, by using a gouge ($\varnothing 15\text{ mm}$) and a macrorhizon inserter ensuring a close fit between soil and the device. The macrorhizons were placed following a systematic design with equal distances between the macrorhizons and the plot edge. Five macrorhizons were placed in each of the unharvested controls and in the whole tree harvest plots in high-thinning, shelterwood and clearcut in a cross design. In the other 7 subplots, 3 macrorhizons were placed in a diagonal line (Fig. S6.1). Macrorhizons were left to settle in the soil profile for approximately 6 weeks.

2.3 Soil solution sampling

Soil solution was sampled on regular monthly intervals from April 2020 to March 2021. The samples were collected by applying negative pressure to the macrorhizons generated by a 30 mL syringe. Syringes were covered and left overnight in the field to ensure that all pressure was used to collect the soil moisture. Samples were collected the next day, directly cooled and processed within 24h by determining the number of syringes with soil moisture, creating composited samples per subplot and determining the sample volumes and storing the samples at -19°C.

Soil moisture samples with a volume $\geq 2.5\text{ mL}$ were taken for the analysis of total nitrogen (Nts), ammonium (N-NH_4), nitrate ($\text{N-NO}_3+\text{NO}_2$) and phosphate (P-PO_4) by using the Segmented Flow Analyser (SFA, Skalar 4000, the Netherlands). Concentrations of aluminum (Al), phosphorus (P), sulfur (S), potassium (K), calcium (Ca), magnesium (Mg), manganese (Mn), copper (Cu), iron (Fe) and zinc (Zn) were analyzed with ICP-AES (Thermo-Scientific iCAP 6500 DUO, USA).



Figure 6.1 Overview of the macrorhizons used (top left) and the installation of the macrorhizons in the field. First, a small part of the organic layer was removed till the mineral soil (top middle left) and then the macrorhizon was placed under an angle of 30°, by using a gouge (ø15 mm) (top right). Soil moisture was sampled by attaching a syringe to the macrorhizon (bottom left), which was covered by organic matter in the field. Samples within each plot showed large heterogeneity as shown by three syringes which were taken from a stem only harvest plot in a clearcut (bottom right).

2.4 Water flux estimation

To estimate drainage fluxes over the sampling period we used a spatially explicit individual-based forest model (hereafter referred to as model) that simulates the daily growth of individual trees in a forest plot based on their morphology and physiology (de Vries, In prep.). This model is ideally suited for the estimation of the drainage and therefore leaching fluxes in this study. Firstly, the spatially explicit individual-tree based approach allows the model to explicitly simulate the impact of thinning interventions on forest structure and subsequently on the light environment and water fluxes. Additionally, dendrochronology's (see **Ch5**), dendrometer data (from 2022), and soil water potential measurements (from 2022) from our experimental plots have been used to validate model predictions on inter-annual growth, intra-annual growth and soil water potential, respectively. The model implements a tipping bucket approach to simulate daily soil water dynamics from the sum of daily rainfall, interception, transpiration, evaporation, and drainage. Daily rainfall is partially intercepted by the canopy, and stored up to a canopy

storage capacity, beyond which additionally intercepted rainfall will reach the soil through stemflow and canopy drip. The rainfall that is intercepted and captured by the canopy is assumed to fully evaporate over the course of the day. The remaining rain is then added to the soil water content of the organic layers (i.e., the litter layer) as throughfall. Following the tipping bucket approach, the water that exceeds a layer's field capacity flows into the next soil layer, until eventually the water leaches out of the rooting zone at a depth of 60 cm below the litter layer. During the day, the internal water content of the tree is decreased through transpiration to support photosynthetic activity, which is calculated by the Farquhar photosynthesis model (Farquhar et al., 1980), adapted by Yin and Struik (2017). To calculate transpiration, the model assumes that leaves are either light limited and stomata are opened to maintain the potential rate of photosynthesis, or they are water limited and stomata are closed, leading to strong reductions in transpiration and photosynthesis. The model calculates how much water is available for transpiration before the leaves reach the critical water potential for stomatal closure based on canopy light capture, weather conditions and tree water status (i.e., total tree water content in roots, stem, branches and foliage). The deficit in tree water content that is built up during the day is then refilled during the night through water uptake from the soil. Finally, soil evaporation reduces soil water content of the litter layer and first 10 centimeters of the mineral soil. Evaporation is calculated using the Priestley-Taylor equation (Priestley and Taylor, 1972) calibrated for a forest clearcut (Flint and Childs, 1991), accounting for microclimatic effects on temperature and relative humidity dependent on canopy leaf area. For a more elaborate model description, see de Vries (In prep.).

In the model, we made a virtual replicate of the experimental plots, using DBH data from 2018 and long-term growth estimates per DBH class from dendrochronological data to reconstruct each individual tree in the experimental plot, placed at random locations since tree location data were not available (Fig. S6.2). In addition to the individual trees, we added a layer of undergrowth with a height of 1m and a LAI of $2 \text{ m}^2 \text{ m}^{-2}$. The leaves in this undergrowth layers are not linked to individuals but are assumed to be homogeneously distributed in space and display the same phenology as the dominant tree species in the plot. Daily meteorological data from the nearest KNMI weather stations were used as model input (described in **Ch3**). The thickness of the litter layers were measured at plot level (**Ch5**) and used for model parameterization. Specific soil parameters like field capacity, wilting point and saturation were based on parameters for podzolic soils as described in Heinen et al. (2020). The simulated water balance was then compared to measured values of: (i) rainfall interception (ii) the difference in Na deposition and Na leaching, assuming no interaction of Na with the soil (tracer behavior).

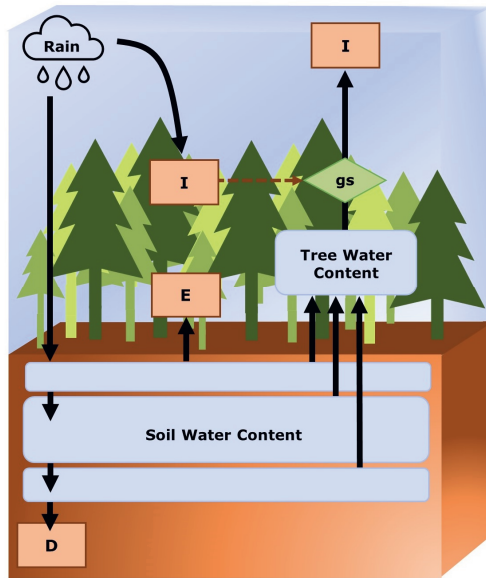


Figure 6.2 Summary of the water pools (blue rectangles), processes (green rhombi), and inputs/outputs (orange parallelograms) of the model's water dynamics module. The model input consists of daily rainfall, with a portion intercepted by the canopy (I), and the remainder added to the topsoil. When rainfall surpasses a soil layer's capacity, the excess infiltrates subsequent layers, with the top layer being the forest floor, the second layer being the mineral soil (0-40 cm), and the third layer from which dissolved nutrient concentrations are sampled (40-60 cm), until drainage occurs (D). Water evaporates (E) from the topsoil based on temperature, humidity, and irradiance. Tree water content (TWC) decreases during the day through transpiration (T), with the rate determined by stomatal conductance (gs), a function of potential photosynthesis in the absence of water limitation, tree water content, rainfall interception, and vapor pressure deficit.

2.5 Soil moisture corrections and calculation of the leaching

Our sampling strategy was designed for the comparative collection of data from 11 treatments over 12 months, with 5 replicate plots for each of the three studied tree species. The goal was to generate a total of 1980 samples for laboratory analyses, utilizing 7380 macrorhizon samples, which were pooled for each treatment within each plot. The monthly collection of soil moisture resulted in 1218 composited samples for laboratory analysis, indicating that 762 samples were missing, primarily due the dry summer of 2020. Considering the heterogeneous soil properties, the nutrient concentration data was first screened on the presence of outliers by converting the data to a gaussian distribution and calculating the mean and standard deviation of each element for each harvest intensity treatment per site. All values outside of the 95%

confidence interval were subsequently removed resulting in a 2% increase of missing values in the data. The values that were eliminated as well as the missing data were subsequently imputed using MICE (Multiple Imputations by Chained Equations) based on predictive mean matching in R which relies on Monte Carlo simulations.

The nutrient leaching fluxes were calculated by multiplying the drainage flux (L m^{-2}) as simulated by the model with the measured nutrient concentrations (mg L^{-1}) in the soil moisture at 50 cm depth in the mineral soil. To derive kg ha^{-1} the resulting concentration (mg m^{-2}) was divided by 100. As there was no relationship between nutrient concentration and the amount of collected soil moisture, the leaching flux was aggregated into consecutive time periods, each represented by the nutrient concentration measured in the middle of that time period. The aggregated leaching flux was multiplied by the corresponding nutrient concentration of the month and scaled to a monthly kg ha^{-1} .

2.6 Statistical analysis

All statistical analysis were performed in R version 4.1.0. To evaluate the percentage of variance in dissolved nutrient concentrations explained by harvest intensity, harvest method, soil preparation, the month, and various environmental variables such as total monthly precipitation (mm), average temperature ($^{\circ}\text{C}$), diverse proxies for soil moisture, and drainage flux (L m^{-2}), we conducted a Partial Redundancy Analysis (p-RDA). Prior to the p-RDA, all data were log transformed to meet the linearity assumption and the p-RDA was conducted using the *vegan* package (Oksanen et al., 2022).

To compare the model predictions with field estimations and to explore the effects of species, harvest intensity, harvest method and soil preparation on the monthly dissolved nutrient concentrations and on the total annual leaching, five different types of statistical models were constructed. First, the effects of species and harvest intensity on the dissolved nutrient concentrations were tested on a monthly basis using the dissolved concentrations in un-mulched whole tree harvest subplots within high-thinning, shelterwood, and clearcut harvest intensities against an unharvested control. Second, the effects of harvest method and the interactions with species and harvest intensity were tested on a monthly basis using the dissolved nutrient concentrations of the un-mulched plots in the high-thinning, shelterwood and clearcut. Third, the effects of soil preparation and the interactions with species, harvest intensity and harvest methods were explored using the dissolved nutrient concentrations of the shelterwood and clearcut. Fourth, the effects of tree species and harvest intensity on the annual nutrient leaching were tested and finally, the effects of tree harvest method and the interactions with tree species and harvest intensities on the annual nutrient leaching were tested. These five different statistical models were constructed using the nested *glmm* models from the *glmmTMB* package (Magnusson et al., 2017). Temporal autocorrelation within the *glmm* based on monthly data was overcome by adding a cubic B-spline smoother which was

constructed using the *splines* package and the prediction wrapper for GAM smooth terms from the *mgcv* package (R Core Team, 2013; Wood and Wood, 2015) while spatial dependency within sites was corrected using random effects. The *glmm* models were constructed using the gamma distribution or the zero-inflated gamma distribution with a log or square root link. Statistical model performance was tested using the *DHARMA* package (Hartig and Hartig, 2017) using the Kolmogorov-Smirnov test for uniformity, the *DHARMA* nonparametric test for dispersion and the *DHARMA* outlier test. Additionally, a second outlier test was performed using cook's distance based on the influence of specific points as provided by the *car* package (Fox and Weisberg, 2018). Observations with a cook's distance > 0.05 were considered to be influential for the monthly statistical models and were replaced by the mean of the nutrient concentration in the previous and the next month. For the annual statistical models, a threshold of 0.5 was used for cook's distance and influential points were set to a missing value. Dispersion formulas were added when necessary. The effects of the conditional statistical model part was tested with a *glmm* adapted two way ANOVA followed by *Tukey's posthoc* from the *emmeans* package (Lenth et al., 2019).

3 Results

3.1 Comparison of the performance of the model used for drainage estimation

The performance of the hydrological model was evaluated by comparing predicted rainfall water interception with measurement-based estimates and by contrasting observed Na deposition with predicted Na leaching (Fig. 6.3). Field interception shows a significant non-linear relationship with modeled interception, indicating that the model tends to underestimate interception when monthly field interception exceeds 25 mm ha⁻¹ and overestimate it below 25 mm ha⁻¹ (Fig. 6.3, Table S6.1). The Na balance, in which Na deposition is assumed to be equal to Na leaching in the undisturbed control stands, was indeed relatively close to equilibrium for Douglas fir and Scots pine, with a difference of only ± 1 kg ha⁻¹ yr⁻¹ between the annual deposition and leaching flux. In beech, however, Na leaching was approximately 30% higher than Na deposition, indicating that leaching in beech might be slightly overestimated. For the thinning, shelterwood and clearcut plots, leaching is higher than deposition reflecting that the decrease in Na deposition is not fully reflected within one year by the same reduction in dissolved Na concentrations (Fig. 6.3).

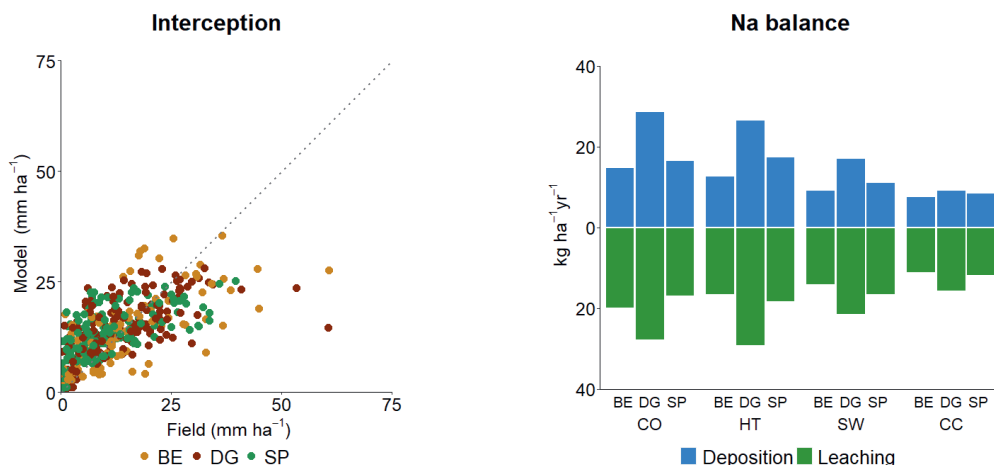


Figure 6.3 Comparison of model predictions and field estimations of rainfall interception and the Na balance. The interception compares the field precipitation interception as determined in **Ch3** and the modelled interception (both mm ha^{-1}) of beech (BE), Douglas fir (DG) and Scots pine (SP). The significant relation is shown with the black line, the dashed line represents the 1:1 line. The Na balance is based on the total deposition of Na for the control (CO), high-thinning (HT), shelterwood (SW), and clearcut (CC) in BE, DG, and SP, which is taken from **Ch3** and compared to the total Na leaching (both in $\text{kg ha}^{-1} \text{yr}^{-1}$).

3.2 Annual water balances for the different tree species

The water balance was computed for a 12-month period (April 2020 – March 2021) which was a relatively dry period with an average rainfall of $730 \pm 17 \text{ mm yr}^{-1}$ across all sites, with significantly less rain observed in the southern sites compared to the northern sites (approximately a 200 mm yr^{-1} difference) (Table S6.2). The modeled annual drainage flux varied across harvest intensities, being lowest in the control of Douglas fir and highest for the clearcut in Scots pine (Table 6.1). Across all tree species, an increase in forest openness (from control to clearcut) resulted in higher drainage, reflecting changes in interception, transpiration, and evaporation. The drainage in Douglas and Scots pine was relatively similar across the harvest intensities with slightly higher drainage observed for Scots pine. These species, however, differed in their interception and, for the control forest, in the transpiration (Table 6.1). Beech consistently exhibited lower interception and transpiration but significantly higher evaporation, leading to higher drainage compared to Douglas fir and Scots pine. Considering a rainfall of $730 \pm 17 \text{ mm}$ over the measurement period, approximately 45% of rainwater in control plots and high thinning beech stands leaves the rootzone as surplus through drainage, compared to around 35% in Douglas fir and Scots pine. In shelterwood and clearcut plots, this is respectively $\pm 60\%$ and 65% irrespective of tree species (Table 6.1).

Table 6.1 Annual means $\pm$ s.e. of the interception (Int), transpiration (Trans), evaporation (Evap) and drainage (Drain) of water (all mm yr⁻¹) for the harvest intensity treatments high-thinning (HT), shelterwood (SW), clearcut (CC) and the unharvested control (CO) as modelled for beech, Douglas fir and Scots Pine. The means and standard error are based on five different sites per species. Precipitation per site is given in Table S6.2. Differences in precipitation with the combined interception, transpiration, evaporation and drainage is the result of changes in the soil water content.

	Beech				Douglas fir				Scots pine			
	CO	HT	SW	CC	CO	HT	SW	CC	CO	HT	SW	CC
Int	136 $\pm$ 8.2	126 $\pm$ 4.9	40 $\pm$ 2.9	5.9 $\pm$ 0.092	199 $\pm$ 8.0	192 $\pm$ 3.1	127 $\pm$ 5.1	105 $\pm$ 1.8	181 $\pm$ 1.8	170 $\pm$ 4.3	120 $\pm$ 3.4	104 $\pm$ 1.7
Trans	156 $\pm$ 6.6	151 $\pm$ 5.6	48 $\pm$ 2.8	2.8 $\pm$ 0.055	211 $\pm$ 15	201 $\pm$ 8.7	90 $\pm$ 5.2	49 $\pm$ 1.0	225 $\pm$ 11	201 $\pm$ 9.2	88 $\pm$ 3.3	48 $\pm$ 1.1
Evap	78 $\pm$ 4.5	85 $\pm$ 7.5	204 $\pm$ 6.4	256 $\pm$ 7.0	25 $\pm$ 5.1	26 $\pm$ 2.5	52 $\pm$ 2.8	68 $\pm$ 3.1	21 $\pm$ 1.8	26 $\pm$ 2.0	54 $\pm$ 2.3	62 $\pm$ 3.5
Drain	329 $\pm$ 1.7	336 $\pm$ 5.9	402 $\pm$ 7.9	428 $\pm$ 13	258 $\pm$ 24	275 $\pm$ 17	431 $\pm$ 6.8	480 $\pm$ 7.1	266 $\pm$ 22	297 $\pm$ 18	438 $\pm$ 8.2	487 $\pm$ 8.6

3.3 Patterns in dissolved nutrient concentrations

The variation in dissolved nutrient concentrations throughout the year was partially explained by harvest intensity, tree species, and the volume of collected soil moisture, while harvest method and soil preparation had minimal impact (p-RDA, Var = 3.2, F = 25, $p \leq 0.001$, R²-adj = 0.31, Fig. 6.4). Harvest intensity (from control to clearcut) accounted for 12% of the variation in the dissolved monthly nutrient concentrations, while species and the volume of collected soil moisture explained 4.6% and 3.7% respectively. The first RDA-axis was determined by the difference between the unharvested control and the clearcut while the second RDA-axis was driven by the difference between Douglas fir and Scots pine (Fig. 6.4). Both harvest method and soil preparation significantly explained the variation in the dissolved nutrient concentrations although the variation explained by either one of them was $\leq 0.5\%$. The cumulative proportion explained by the other significant terms (month, monthly precipitation, average temperature and diverse proxies for soil moisture) was 5.1%. The unconstrained CA axis explained higher percentages of variation within soil moisture concentration. The first unconstrained axis was driven by the difference between dissolved Na and K concentrations, explaining 16% of the total variation. The second unconstrained axis was related to the difference between Ca and Mg compared to Al, explaining 12% of the total variation. The Ca/Al and Mg/Al ratios are both between 0.5 to below 0.25, demonstrating a declining pattern from control to clearcut for beech and Scots pine, while no clear pattern exists for Douglas fir (Fig. S6.3).

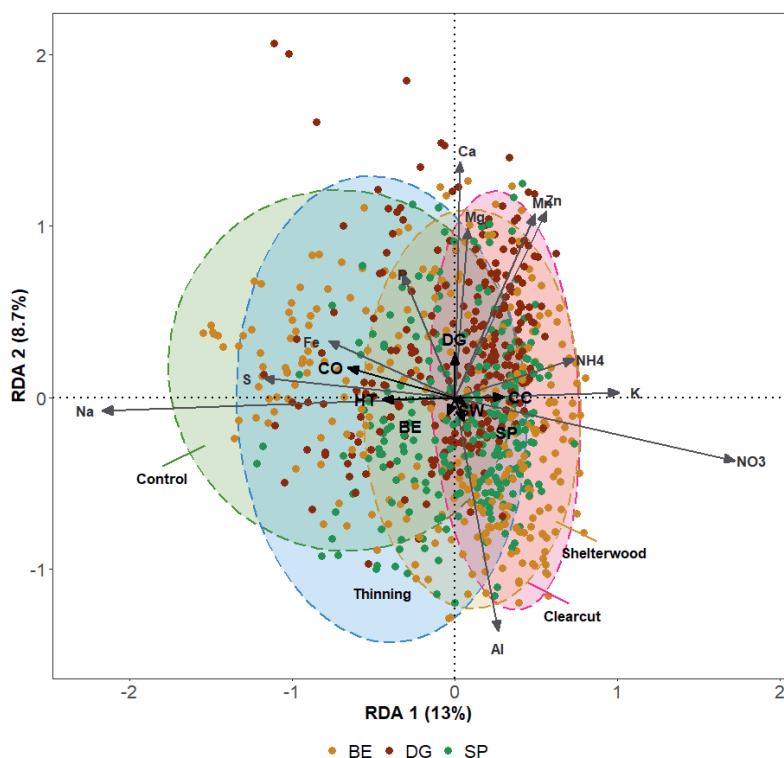


Figure 6.4 Explanation of the monthly concentrations of macro- (NH_4 , NO_3 , S, P, Ca, K and Mg) and micronutrients (Mn, Cu, Fe and Zn) in soil moisture by the centroids of harvest intensity treatments (control : clearcut), tree species (BE: beech, DG: Douglas fir and SP: Scots pine), months grouped by seasons and soil moisture content (RDA biplot). Effects of the spring and autumn, soil preparation, harvest method (SOH vs. WTH), precipitation, temperature, drainage and water content of the sampled soil layer are not included in this figure as the contribution of these elements was marginal. The concentrations of the nutrients in the soil moisture are represented by the grey arrows, the effects of species, canopy openness treatments, seasons and soil moisture by black arrows. The length of arrows denotes the variation explained by respectively the nutrients, treatments, seasons, species and soil moisture.

3.4 Impacts of tree species and management on dissolved nutrient concentrations

Tree species had a significant effect on the concentrations of all dissolved nutrients except for NH_4 , total N and Al, with most pronounced effects on the S, Mn and Mg concentrations (Table 6.2, Table S6.3, Fig. S6.4). Harvest intensity significantly influenced the dissolved nutrient concentrations for all nutrients except for P and Cu. Interaction effects between species and harvest intensity was diverse amongst elements. For K, for example, the concentration increased from control to clearcut in beech while for Douglas fir the concentration was significantly higher in the control (Table S6.3). However, overall, there

was a tendency of increasing nutrient concentration with harvest intensity in beech for NO₃, NH₄, total N, K, Mn, Zn and Al and in Scots pine for NO₃, total N, K, Mg and Mn while for Douglas fir only an increase was observed for NH₄, total N and Mn (Table S6.3, Fig. S6.4). In Douglas fir, the nutrient concentration of S, Ca, K and Na decreased with increasing harvest intensities while such a decrease was only found for S and Na in beech and Scots pine (Table S6.3, Fig. S6.4).

Harvest method did not significantly influence the dissolved Ca, Mg, Mn, Al and Na concentrations, and for other elements, except NO₃, K and Fe, the patterns were rather weak (Table 6.2, Table S6.4). Consistently higher dissolved NO₃ concentrations were observed following a SOH high-thinning, whereas K concentrations were consistently elevated following SOH across different harvest intensities. Conversely, Fe concentrations were consistently higher following WTH (Table S6.4). No consistent effects of harvest method on NH₄, total N, S, P, Cu and Zn were found (Table S6.4).

The soil preparation had hardly any influence on the dissolved nutrient concentrations (Table 6.2). Dissolved P concentrations tended to be higher following mulching treatments in both the SOH and WTH treatments in Scots pine, while dissolved Mg concentrations were higher in un-mulched WTH treatments compared to mulched WTH treatments (Table S6.4). An overall effect of mulching on dissolved nutrient concentrations seems lacking. Hence, the drainage flux and therefore the annual leaching was not computed separately for soil preparation due to the absence of differences in concentration, and no difference in the drainage flux between mulched and non-mulched plots.

Table 6.2 The influence of tree species, harvest intensity (HI), harvest method (HM) and soil preparation (SP, mulching) and their interactions on dissolved nutrient concentrations as denoted by the chi-square and significance levels or the analysis of deviance (Type II Wald chi-square tests). Within species, treatment, harvest method and soil preparation mean values, S.E. and the Tukey's post-hoc test statistics are given in Table S6.3 and S6.4.

	Statistical model 1			Statistical model 2			Statistical model 3			
	Species	HI	Species * HI	HM	HM * Species	HM * HI	SP	SP * Species	SP * HM	SP * HI
NO ₃	8.5 *	86 ***	48 ***	8.1 **	1.5 n.s.	5.9 n.s.	0.85 n.s.	4.0 n.s.	0.46 n.s.	0.43 n.s.
NH ₄	1.9 n.s.	54 ***	13 *	5.9 *	1.2 n.s.	5.0 n.s.	0.10 n.s.	3.4 n.s.	2.6 n.s.	3.3 n.s.
N _{tot}	0.86 n.s.	300 ***	85 ***	8.8 **	0.63 n.s.	2.4 n.s.	0.40 n.s.	7.6 *	1.1 n.s.	1.6 n.s.
S	130 ***	31 ***	25 ***	5.1 *	9.9 **	2.0 n.s.	0.01 n.s.	4.7 n.s.	4.1 *	0.54 n.s.
P	18 ***	6.5 n.s.	8.3 n.s.	5.0 *	4.0 n.s.	13 **	11 **	11 **	0.73 n.s.	0.00 ns.
Ca	86 ***	55 ***	7.1 n.s.	0.71 n.s.	12 **	4.2 **	0.74 n.s.	4.4 n.s.	0.03 n.s.	0.52 ns.
K	66 ***	82 ***	103 ***	41 ***	2.0 n.s.	2.0 n.s.	0.75 n.s.	8.4 *	2.8 n.s.	0.05 n.s.
Mg	160 ***	32 ***	27 ***	3.3 n.s.	10 **	3.7 n.s.	19 ***	0.08 n.s.	0.39 n.s.	0.02 n.s.
Mn	230 ***	77 ***	16 **	0.93 n.s.	25 ***	6.6 *	2.6 n.s.	3.6 n.s.	2.0 n.s.	0.80 n.s.
Cu	0.90 n.s.	0.99 n.s.	32 ***	4.8 *	4.0 n.s.	4.7 n.s.	0.02 n.s.	2.2 n.s.	0.08 n.s.	0.05 n.s.
Fe	41 ***	13 **	21 **	41 **	15 ***	2.2 n.s.	2.3 n.s.	3.6 n.s.	16 ***	6.6 **
Zn	30 ***	35 ***	22 **	15 ***	1.8 n.s.	0.47 n.s.	18 n.s.	0.26 n.s.	5.7 *	7.1 **
Al	5.6 n.s.	50 ***	80 ***	2.2 n.s.	1.3 n.s.	3.6 n.s.	0.51 n.s.	13 **	5.1 n.s.	0.01 n.s.
Na	49 ***	460 ***	16 *	0.14 n.s.	1.6 n.s.	11 **	0.16 n.s.	2.9 n.s.	12 ***	0.03 n.s.

*** $P \leq 0.001$, ** $0.01 \leq P < 0.001$, * $0.05 \leq P \leq 0.01$, n.s. $P > 0.05$. Different letters denote significant differences among canopy positions according to Tukey's posthoc test at a significance level of $P < 0.05$.

3.5 Impacts of tree species and management on annual nutrient leaching

The annual leaching of all elements, except NO_3 and Cu, was influenced by tree species (Table 6.3). In unharvested forests, there was a tendency for higher leaching of macronutrients and Na in Douglas fir. Moreover, in those control plots, the leaching of NO_3 was lower than S in beech ($6.2 \text{ kg ha}^{-1} \text{ yr}^{-1}$ versus $5.7 \text{ kg ha}^{-1} \text{ yr}^{-1}$). This patterns was reversed for Douglas fir ($10 \text{ kg ha}^{-1} \text{ yr}^{-1}$ versus $10 \text{ kg ha}^{-1} \text{ yr}^{-1}$) and Scots pine ($8.9 \text{ kg ha}^{-1} \text{ yr}^{-1}$ versus $4.5 \text{ kg ha}^{-1} \text{ yr}^{-1}$), resulting in S leaching being more acidifying in beech ($0.3 \text{ meq ha}^{-1} \text{ yr}^{-1}$) while in Douglas fir and Scots pine the NO_3 leaching is more acidifying (respectively $0.21 \text{ meq ha}^{-1} \text{ yr}^{-1}$ and $0.28 \text{ meq ha}^{-1} \text{ yr}^{-1}$). The effects of species on the leaching changed towards the clearcut. For mobile elements like NO_3 and K a sharp increase was observed moving from control to clearcut either changing the species effects (for NO_3) or diminishing the species effects (K). For the clearcut, there is no consistent species trend for annual leaching since, for example, N and Al leaching were highest for beech, while for the other macronutrients (S, P, Mg and Ca), leaching was generally highest for Douglas fir (Table S6.5).

Table 6.3 The influence of tree species, harvest intensity (HI), harvest method (HM) and their interactions on annual nutrient leaching fluxes as denoted by the chi-square and P values of the analysis of deviance (Type II Wald chi-square tests). Differences in leaching following mulching was not tested as mulching did hardly influence the dissolved nutrient concentrations (Table 6.2). Within species, treatment, harvest method and soil preparation mean values, S.E. and the Tukey's post-hoc test statistics are in Table S6.4 and S6.5.

	Statistical model 1			Statistical model 2		
	Species	HI	Species * HI	HM	HM * Species	HM * HI
NO_3	1.3 n.s.	170 ***	20 **	2.6 n.s.	1.7 n.s.	1.2 n.s.
NH_4	7.2 *	66 ***	21 **	1.6 n.s.	1.5 n.s.	1.4 n.s.
N_{tot}	10 *	220 ***	23 ***	2.5 n.s.	2.8 n.s.	0.38 n.s.
S	56 ***	22 **	9.2 n.s.	1.3 n.s.	9.6 **	1.6 n.s.
P	620 ***	320 ***	240 ***	1.7 n.s.	9.2 **	0.52 n.s.
Ca	28 ***	43 ***	8.3 n.s.	5.8 *	2.4 n.s.	0.16 n.s.
K	31 ***	100 ***	27 ***	19 ***	0.36 n.s.	1.4 n.s.
Mg	74 ***	79 ***	10 **	5.8 *	11 **	1.1 n.s.
Mn	110 ***	160 ***	8.9 n.s.	0.04 n.s.	9.4 **	3.8 n.s.
Cu	1.9 n.s.	11 *	22 **	0.89 n.s.	2.0 n.s.	0.60 n.s.
Fe	31 ***	13 **	7.3 n.s.	4.6 *	2.9 n.s.	4.1 n.s.
Zn	36 ***	200 ***	29 ***	0.026 n.s.	7.5 *	2.2 n.s.
Al	1.9 n.s.	130 ***	28 ***	0.00 n.s.	1.5 n.s.	0.94 n.s.
Na	48 ***	95 ***	12 n.s.	2.6 n.s.	2.7 n.s.	0.76 n.s.

*** $P \leq 0.001$, ** $0.01 \leq P < 0.001$, * $0.05 \leq P \leq 0.01$, n.s. $P > 0.05$. Different letters denote significant differences among canopy positions according to Tukey's posthoc test at a significance level of $P < 0.05$.

As with dissolved nutrient concentrations, nutrient leaching strongly increases moving from control stands to clearcut (Fig. 6.5) although patterns are different amongst nutrients and across species. The strongest increase from control to clearcut was observed for NO_3 , which increased up to 15 times across

species (Table S6.5). In beech stands, the leaching increasing strongest when moving from control to clearcut, are total N (mostly determined by the increase in NO_3) followed by Al (6 times increase) and the base cations (particularly K and Mg). In Douglas fir, the increase in dissolved concentrations was the highest for Mn (6 times increase) while total N increased 3.3 times. The concentrations in Scots pine increased most for total N (6.6 times) followed by the base cations (particularly K and Mg). When comparing the different harvest intensities, small but consistent differences were found between control and high-thinning and between shelterwood and clearcut (Fig. 6.5, Table S6.5).

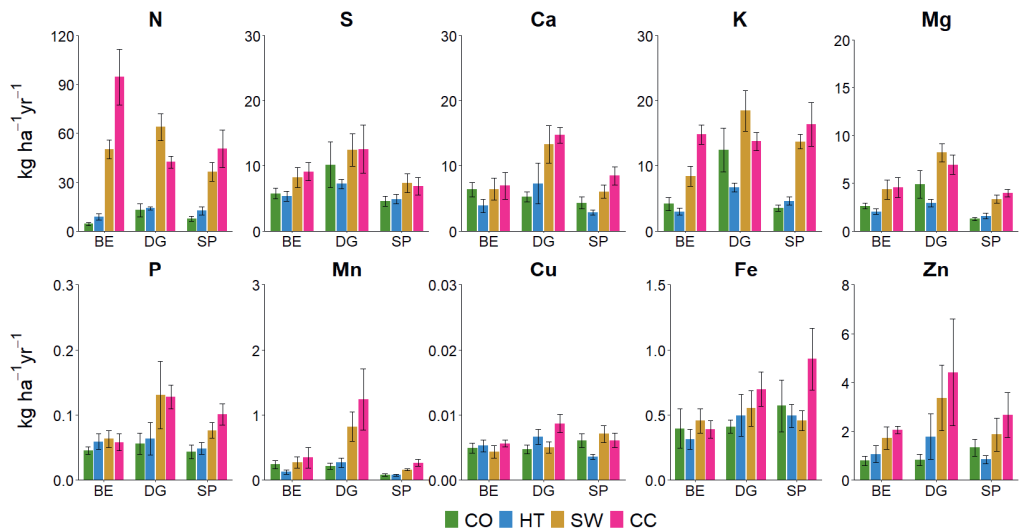


Figure 6.5 Mean annual leaching fluxes of macro- (N, S, P, Ca, K and Mg) and micronutrients (Mn, Cu, Fe and Zn) in forest stands dominated by beech (BE), Douglas fir (DG) and Scots pine (SP) as a function of the harvest intensity (CO: control, HT: high-thinning, SW: Shelterwood and CC: Clearcut). For the high-thinning, shelterwood and clearcut the data of the non-mulched whole tree harvest plots are shown.

3.6 Effects of harvest method on annual nutrient leaching

The harvest method had only marginal impact on annual nutrient leaching, with the few significant effects being relatively small compared to the influences of harvest intensity and tree species (Table 6.3, Table S6.6). There is a weak indication of higher dissolved nutrient concentrations of base cations following SOH in beech (only for K), Douglas fir (Ca and K), and Scots pine (Ca, K, Mg, and Mn, Fig. 6.6). Conversely, for S, there is a tendency of higher concentrations following WTH in beech, while in Douglas fir, the concentrations tended to be higher following SOH, and no general patterns can be distinguished for dissolved P and Fe concentrations.

The effect of harvest method on annual leaching differed slightly from the influence on nutrient concentrations in soil moisture (Table 6.2 and Table 6.3). The generally larger effects of harvest method on soil moisture concentration compared to leaching could be attributed to the season, with a higher influence of harvest method on concentration in the summer when leaching fluxes are very low.

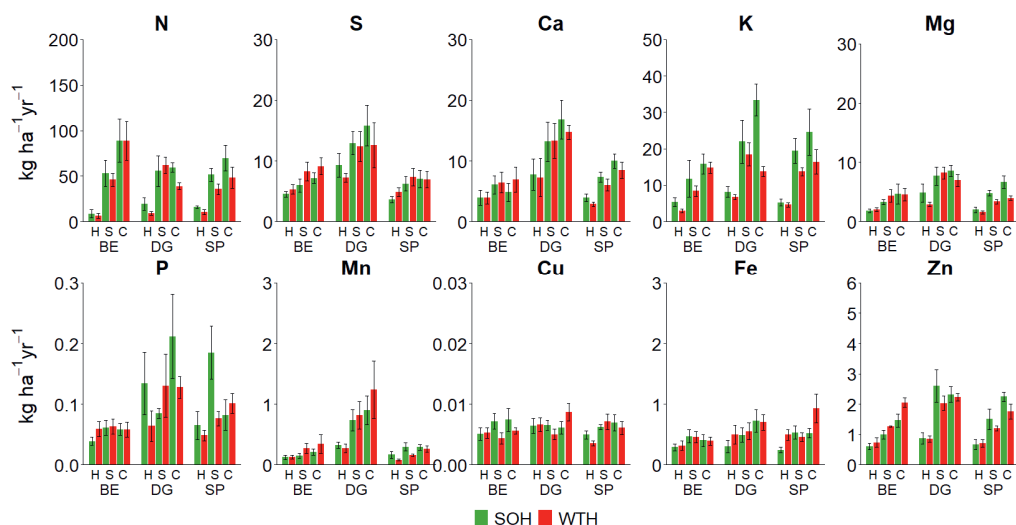


Figure 6.6 Mean annual leaching fluxes of macro- (N, S, P, Ca, K and Mg) and micronutrients (Mn, Cu, Fe and Zn) in beech (BE), Douglas fir (DG) and Scots pine (SP) as a function of the harvest method (SOH: stem only harvest and WTH: Whole tree harvest). Effects of harvest method are shown per harvest intensity (H: high-thinning, S: Shelterwood and C: Clearcut). Between groups differences for the elements in which harvest method has an effect (Table 6.2) are given in Table S6.6.

4 Discussion

4.1 Water fluxes in forests

The forest water balance is crucial for estimating drainage and nutrient leaching, but assessing this balance is challenging because drainage depends on a multitude of factors, including forest structure, forest composition, soil type, climate, small-scale spatial heterogeneity, preferential flow paths, and intra-annual patterns in water fluxes (Legout et al., 2009; Van Der Heijden et al., 2013). The drainage flux can be estimated from a range of methods with varying levels of detail; from average annual estimates, quantifying interception as a fraction of rainfall and evapotranspiration from literature based values (de Vries et al., 2021), one-dimensional water balance models with a static single leaf canopy (Christiansen et al., 2006), to the spatially explicit individual-tree based and mechanism-based modelling approach used here. This mechanistic modelling approach involves simulating a 3D forest structure, determining water

interception and throughfall, calculating transpiration and evaporation demands based on 3D light conditions, and employing validated sub-models that are widely applied in mechanistic plant growth models (Jansson, 2004; Merganičová et al., 2005; Tian et al., 2012; Van Der Heijden et al., 2013; Leppä et al., 2020). These sub models are used to estimate the 3D light environment (Evers and Bastiaans, 2016), stomatal behavior, canopy photosynthesis, and transpiration through physiological mechanisms (Monteith, 1965; Priestley and Taylor, 1972; Farquhar et al., 1980; Yin and Struik, 2017). As such, a mechanistic modelling approach is theoretically able to go beyond the data to which the model is validated and simulate across different forest structures. Such a model can thus be applied in different situations and, in our case, could be easily adapted for effectively capturing the spatial and temporal variations in the forest water balance, and drainage in particular, as triggered by the effects of different harvest intensities on forest structure.

The annually simulated drainage differed slightly between tree species and significantly varied among harvest intensity treatments (Table 6.1). The drainage fluxes were two times larger than the median drainage fluxes for intensively managed forests in Europe, while the estimated transpiration fluxes were 100 to 200 mm yr⁻¹ lower (Van der Salm et al., 2007b). However, other studies in areas with comparable rainfall reported more similar values for drainage in beech (Christiansen et al., 2006), Douglas fir and Scots pine (Gielen et al., 2010; Prietzel et al., 2023) or even higher values (Gielen et al., 2010; Legout et al., 2016). Our model simulation of drainage thus falls well within the range of reported drainage values for similar forests. The broad range of reported values may both imply dependency on various factors (e.g., climate, soil) but also uncertainty of the methods used in different studies.

Comparing model results with our field data, we observed a reasonable to good fit in the precipitation interception and total N balance (Fig. 6.3). The slight under- and overestimation of interception is probably attributed to heterogeneity in the field data, given the highly heterogeneous nature of throughfall volume and, consequently, interception estimates (Bleeker et al., 2003). Such variations also affect the representativity of field measurements and make it difficult to validate hydrological models (van Der Heijden et al., 2019). Furthermore, in Douglas fir and Scots pine stands, N deposition is almost entirely balanced by N leaching while in beech stands, N leaching exceeds N deposition by approximately 30%. This suggests a potential overestimation of leaching in these stands though still below estimates from other beech stands (Prietzel et al., 2023). Despite these remaining uncertainties, we conclude that our drainage estimates fall within a realistic range, providing a basis for inter-species comparisons and assessing forest management effects on drainage and, consequently, leaching.

4.2 *Dissolved nutrient concentrations and leaching in unharvested control*

The annual leaching in unharvested closed control plots provide valuable insights into the impacts of tree species (Rothe et al., 2002b; Oulehle and Hruška, 2005; Legout et al., 2016), the nutritional status of the forests (Smethurst, 2000; Waldner et al., 2015), and anthropogenic soil acidification (Schaaf et al., 1995; Braun et al., 2020b). We found large variability in the leaching between sites and over time which is caused by high variability in the dissolved nutrient concentrations as also observed in other studies (Legout et al., 2016; Paul et al., 2022). Tree species explains 17% of the variation within the dissolved nutrient concentrations in unharvested stands (Fig. S6.6), while the majority of the variation remains unexplained. This variation can, for example, be attributed to differences in soil drainage, root proximity, and microbial decompositions (Baeumler and Zech, 1998; Högberg et al., 2013; McGahan et al., 2014) but also to the season, which accounts for 17% of the variation. The significance of the sampling month confirms the more often reported variation in dissolved nutrients over the year with generally higher nutrient concentrations in summer caused by an interplay of lower soil water fluxes, biological activity and biological uptake (Kristensen et al., 2004; Vestin et al., 2008; Fetzer et al., 2022).

Tree species largely impact the leaching of nutrients in closed stands, as shown by the species differences in the annual leaching of N, S, P, K, Mg, Mn, Zn and Na. Leaching in Douglas fir was generally higher, exceeding leaching of N, S and base cations (but only K and Mg, not Ca for beech) by 1.8 to 3.0-fold for beech and by 1.1 to 3.8-fold for Scots pine (Table 6.3, Fig. 6.5). The high leaching in Douglas fir can be attributed to the elevated dissolved concentration of the acid nutrients (NO_3 , S) which were accompanied by elevated concentrations of the base cations (Ca, Mg and K) and of Mn, Zn, Al and Na (Fig. 6.5, Table S6.3) confirming the results of Prietzel et al. (2023) that Douglas fir at N-rich sites has elevated NO_3 , Ca, Mg and Al concentrations. The elevated concentrations of these nutrients reflect the overall higher deposition interception by tall Douglas fir stands compared to shorter beech and Scots pine stands (**Ch3**) (Augusto and Ranger, 2001; Oulehle and Hruška, 2005). Differences in nutrient leaching between beech and Scots pine were less pronounced; nutrient leaching and the dissolved concentrations tended to be higher for some nutrients in beech (S, Ca and Mg) but lower compared to Scots pine for other (NO_3 , K). These differences reflect differences in nitrification, mineralization and adsorption from deeper layers of, for example N and S, between species as previously observed (Kaiser et al., 2001; van der Heijden et al., 2011; Legout et al., 2016) as the deposition of these nutrients is rather similar between the species. Overall, the impact of species on the annual nutrient leaching clearly indicates the need to use species specific leaching estimates.

Due to elevated NO_3 and S concentrations, related to the past and current elevated acid deposition, we expected elevated Fe and Al concentrations and low base cation concentrations in the soil solution. The dissolved nutrient concentrations were well in line with observations in Douglas fir (Paul et al., 2022) and

Scots pine stands (Bārdule et al., 2021) while concentrations of amongst others NO_3 , S, Fe and Al in beech were higher compared to a previous study (Jochheim et al., 2022), reflecting the ongoing soil acidification driven by N and historical S deposition. The elevated concentrations of NO_3 in the soil solution are often related to a less favorable nutritional status including soil acidification and growth reductions (Aber et al., 1989; Waldner et al., 2015; de Vries and Schulte-Uebbing, 2019). Our result do not indicate that there is a direct nutrient limitation in these forests although N concentrations are clearly above the threshold for N saturation (Gundersen et al., 2006). Quantitatively, we found annual base cation losses comparable to the values reported in the literature (Berger et al., 2009; Prietzel et al., 2023), and the relatively high N leaching fell in the range reported for N saturated systems (Gundersen et al., 2006; Verstraeten et al., 2012). Ultimately, the high NO_3 and S concentrations reflect the ongoing soil acidification.

The results suggest that in beech stands there is higher N retention compared to Douglas fir and Scots pine. In beech the annual N leaching was only 28% of the annual N deposition while this fraction was 56% in Douglas fir and 63% in Scots pine (**Ch3**), which explain the slightly higher above- and belowground N stocks in beech stands (**Ch5**). Contrary, there might be S retention in Douglas fir and Scots pine as deposition minus the tree uptake exceeds the S leaching (**Ch3, Ch5**) contradicting that these forest soils are saturated with S causing S leaching to equal deposition (De Vries et al., 1995b; De Vries et al., 2007). This S retention for these conifers might however be temporary, as observed in spruce stands (Meesenburg et al., 2016) since elevated S stocks were not observed for Douglas fir and Scots pine (**Ch5**) or might be related to the greater uptake and recycling of S in conifers (Legout et al., 2016) causing temporal variations in the ratio between S deposition and S leaching.

It is generally assumed that the high rate of N deposition is the main driver of soil acidification in many European forests, causing cation losses and hampering sustainable nutrient balances for tree nutrition (Braun et al., 2020b). Surprisingly, we found that S is the main driver for soil acidification in beech while NO_3 is the main driver for Douglas fir and Scots pine implying the long-term legacy effects of high S deposition prior to 2000 for high current S leaching and soil acidification in beech stands. Prior studies have reported higher SO_4 to NO_3 concentrations in beech stands (Legout et al., 2016) but also in coniferous stands (Oulehle and Hruška, 2005) indicating that in various systems the acidification is still largely driven by S. Nutrient budgets for 121 forested plots in Europe prior to 2000 suggest that S was the dominant source of soil acidification as S behaves as a near tracer while for N there is strong retention, most likely due to N immobilization, apart from N uptake (De Vries et al., 2007), causing much higher sulphate than nitrate losses. The high retention of N in control plots is also found in other studies (de Jong et al., 2023).

The level of soil acidification is often evaluated using the base cation to Al ratio (Bc/Al) (Sverdrup, 1993) in which a Bc/Al ratio of 10 is recommended as critical limit (Ouimet et al., 2006) while ratios below

1.0 have been proposed as threshold values below which there is risk of significant damage to plants (Sverdrup, 1993; Sverdrup and De Vries, 1994). In the studied stands, rather the K/Al ratio is a key determinant for the dissolved nutrient concentrations, explaining 16% of the variation (Fig. S6.6). In control stands, the Bc/Al ratio is 1.4 in beech, 2.1 in Douglas fir and 1.0 in Scots pine (Fig. S6.3) indicating that the stands are close to the risk of plant damage, far beyond the critical limit of 10 and that base cation concentrations have declined relative to Al concentrations (De Vries and Leeters, 2001). Comparable low Bc/Al ratios are observed throughout Europe (Oulehle and Hruška, 2005; Verstraeten et al., 2012; Meesenburg et al., 2016; Johnson et al., 2018) but proof that such low ratios are related to declining forest growth or health is missing (Iost et al., 2012; Meesenburg et al., 2016; Hruška et al., 2023). Yet, the use of this ratio for assessing forest damage risks have led to a strong debate on the strict use of this critical load concept (Løkke et al., 1996), especially for forests in areas with elevated deposition of sea salts (Hansen et al., 2007). However, despite that the link between forest health and Bc/al ratios is missing, the high dissolved Al concentration can still be problematic as Al can reduce Ca and Mg uptake and therefore harm tree nutrition (Vanguelova et al., 2005; de Wit et al., 2010), which can be problematic as pools and concentrations of Ca and Mg in the branches and foliage are already low (**Ch4**, **Ch5**).

4.3 *Impacts of harvest intensity on nutrient leaching*

As expected, tree harvest intensity increased nutrient leaching because of both increased drainage and nutrient concentrations from control forest to clearcut due to reduced uptake of water and nutrients. The absence of large trees reduced water and nutrient uptake, while the mobilization of a significant nitrogen stock (**Ch5**) contributed to increased nitrate and associated cation leaching, as reflected by higher concentrations in soil moisture (Aber, 1992; Horswill et al., 2008; Jerabkova et al., 2011; Lucas et al., 2014; Cusack et al., 2016). The observed substantial increase in nutrient leaching from low (control stands) to high harvest intensity (Fig. 6.5) underscores the significant impact of forest management practices on nutrient dynamics. The strong impact of harvest intensity on nutrient leaching, dissolved nutrient concentrations and drainage than was most pronounced for NO₃ (especially in beech), S, the base cations (Ca, Mg and K) and Al. For P and Cu, the increase in leaching was only driven by an increase in the drainage as dissolved nutrient concentrations were not influenced by harvest intensity. For P, this result is not surprising considering the strong buffering of dissolved P, even though increasing concentrations following clearcuts were reported before (Pirainen et al., 2004; Siebers and Kruse, 2019). The strong buffering is confirmed in this study as P uptake in control stands was not reflected in P leaching in clearcuts, implying a significant decrease in net P mineralization in the forest floor as also argued by (Yanai, 1998). Overall, increasing harvest intensity significantly impacted leaching in the second-year post-harvest. However, the

sustainability of the harvest intensity depends not only on the magnitude but also on the duration of this increased leaching which was not considered in this study.

The strongly elevated NO_3 losses accompanied by elevated S losses of a clearcut indicate the increasing soil acidification following disturbances. Extremely high NO_3 losses following harvest, which can be observed following clearcuts across Europe (Weis et al., 2006; Göttlein et al., 2023), only result in the loss of a fraction of the total N stock (Ch5) as post-harvest leaching is temporary. The post-harvest elevation of the leaching commonly lasts for one up to six years indicating a very limited time frame of elevated losses (Jewett et al., 1995; Carignan and Steedman, 2000; Martin et al., 2000; Swank et al., 2001; Katzensteiner, 2003; Huber et al., 2004; Wang et al., 2006; Hope, 2009; Huber et al., 2010; Jerabkova et al., 2011). The high increase in NO_3 leaching in beech is likely related to its forest floor C/N ratio of 24, which falls below the threshold for elevated N leaching (MacDonald et al., 2002; Gundersen et al., 2006; Dise et al., 2009), whereas both Douglas fir and Scots pine had C/N ratios of respectively 26 and 28 (Ch5). These C/N ratios show that the system is close to N saturation, which is also reflected in the increase in NO_3 concentrations as in non-saturated systems NO_3 concentrations decrease following a clearcut contrary to saturated systems (Ring et al., 2001; Jussy et al., 2004a; Ranger et al., 2007). The observed increase in NO_3 and S leaching in beech and Scots pine resulted in only a minor rise in base cation and Mn leaching, leading to an overall decrease in the acid load buffered by the base cations and Mn. In contrast, Al concentrations strongly increased (Table S6.3), suggesting that the rise in NO_3 concentrations toward clearcut is primarily buffered by an increase in Al concentrations, as reflected in the BC/Al ratios (Fig. S6.3) which was also observed by Göttlein et al. (2023). This result underlines that NO_3 leaching in these stands is not fully reflected in accelerated base cation losses and that the soils are currently in the Al buffering phase. As the Bc/Al ratios are strongly debated, the effect of this Al buffering mechanism on tree growth and health remains hypothetical (Binkley and Högberg, 2016) although there are some pointers that this ongoing soil acidification influences tree fine roots (Braun et al., 2005).

For all nutrients, the effects of thinning (~20% of biomass removal) on leaching was small, indicating that thinning preserves nutrients in the systems and can contribute to sustainable use of forests on poor soils (see also Gundersen et al. (2006)). The practice of thinning, commonly repeated over 4-8 year periods (Den Ouden et al., 2010), seems to be a safe way of harvesting trees since it allows for conserving soil nutrients by reducing leaching, contrary to shelterwood and clearcut. However, as elevated N deposition is still ongoing, there is an annual increase in N stocks (de Jong et al., 2023) which can cause higher leaching in both control and thinned stands when the soil becomes fully saturated and causing leaching to approach deposition levels. From the perspective of sustainable forest use, our leaching results show the potential of low intensity tree harvest, such a high thinning in our study, for long term forest use with acceptable low losses of critical nutrients.

4.4 *Impacts of harvest method and soil preparation on nutrient leaching*

The drainage fluxes were simulated for different harvest intensities, but the possible effects of harvest methods (SOH, WTH) and soil preparation treatments were not accounted for, assuming that these practices would not affect hydrology. We did not find differences in the collected amount of soil moisture between SOH and WTH treatments confirming the assumption. Mulching resulted in significantly lower collected soil moisture in spring and summer months (Apr-Oct, Table S6.7, Fig. S6.5), but not in the moist autumn and winter months when leaching mainly occurs, indicating that the differences in soil moisture only occur when soils are not saturated. Despite these seasonal effects the implications for effects on annual leaching estimates were only inferior.

The SOH harvest method in either shelterwood or clearcut, was expected to cause higher nutrient leaching than the WTH method since the harvest residues left on the soil in SOH enhance decomposition (Smolander et al., 2010; Ojanen et al., 2017). We found the impact of harvest method on annual leaching, however, to be small compared to the influences of harvest intensity and tree species (Table 6.3, Table S6.6), and the few observed effects of harvest method on dissolved nutrient concentrations were not consistent. A weak indication of enhanced leaching following SOH was observed for K, Ca (only in Douglas fir and Scots pine), Mg and Mn (only in Scots pine, Fig. 6.6). Elevated dissolved K concentrations, and therefore elevated nutrient leaching was reported before, although this coincides with higher Ca, Mg, NH_4 and NO_3 concentrations and leaching (Hedwall et al., 2013; Clarke et al., 2018). The lack of an impact of harvest method on, for example NO_3 leaching, may be attributed to forest regeneration, the substantial existing N reservoir in the soil, being much higher than N in harvest residues (Thiffault et al., 2011; Devine et al., 2012), or the emission of N_2O from N in harvest residues (Ineson et al., 1991; Tate et al., 2006; Törmänen et al., 2020). The lack of an effect of the harvest method on Mg and Ca is likely related to the low base saturation of these acid sites, with the effects of harvest residue retention being more pronounced in well-buffered sites (Zetterberg et al., 2016). Our results align with the theory that high levels of Al saturation in coarse-textured soils are the reason that harvest residue retention has no significant effect on soil cation levels in mineral soils (Bélanger et al., 2003; Thiffault et al., 2011). Absence of a clear effect of harvest method on P might be caused by large fluctuations in the dissolved nutrient concentrations, causing large variation in the leaching estimates for the SOH treatments in Douglas fir and Scots pine (Fig. 6.6, Fig. S6.4). Absence of an effect on Fe and Zn is due to the low stock of these nutrients in the harvest residues, comprising < 2% of the annual leaching for Zn and between 5-10% of the annual leaching in Fe for the clearcut (**Ch5**). In conclusion, the overall effects of different harvest methods are marginal, because the potential effects are probably offset by other processes like immobilization or local leaching hotspots.

We observed hardly any effect of mulching on the soil solution chemistry, which was surprising given the expectation that mulching would enhance decomposition, leading to a larger mobilization of

nutrients and consequently higher dissolved nutrient concentrations (Lundmark-Thelin and Johansson, 1997; Piirainen et al., 2007). The slight impact on P could be attributed to disturbed decomposition processes in the soil, altering organic matter and organic P, resulting in a shift from organic to (adsorbed) inorganic P while hardly changing total P concentrations (Cade-Menun et al., 2000). Overall, we found no evidence for faster decomposition leading to increased leaching following mulching, as reported by Bélanger et al. (2003), nor for nutrient immobilization or elevated leaching, as reported by Pitman and Peace (2021). These findings align with research on chipping, considered comparable to mulching, which has been shown not to affect enzyme activities or decomposition (Waldrop et al., 2003) and not to influence nutrient availability and leaching (Sommer et al., 2004; Belleau et al., 2006). Although we cannot rule out stronger mulching effects directly after harvesting (since our leaching measurements were performed 3 years after harvest), our results nevertheless imply that such effects are limited in time (<2 years) and therefore in the longer run small in comparison with the effects of the high tree harvest intensity in clearcut or shelterwood systems.

5 Conclusions and outlook

An intense one-year monitoring period revealed substantial variations in dissolved nutrient concentrations and annual nutrient leaching in response to tree species, harvest intensity and to a lesser extent harvest method. Tree species affected leaching substantially in unharvested control plots, generally being higher in Douglas fir stands than in Scots pine and beech stands, but impacts differed for different nutrients. Harvest intensity strongly affected leaching as leaching increased at high harvest intensities (clearcut and shelterwood) but hardly at low harvest intensity (high-thinning). Clearcutting, and to a lesser extent shelterwood, increased nutrient concentrations, especially NO_3 , indicating a rapid mobilization of large N stocks induced by high (current and past) N deposition. This mobilization was associated with accelerated losses of Al, Fe and Mn in these acid forest soils, while impacts on base cation leaching were more limited. Additionally, we also show that NO_3 leaching and related acidification effects become specifically stronger in intensively harvested forests (clearcut and shelterwood) compared to control forest and high-thinning (with ~20% biomass removal). Other forest management actions (harvest method and soil preparation) had only small to even negligible effects on leaching in the second year after harvest. These results imply that sustainable forest use on infertile soils benefits from implementing continuous cover forestry practices characterized by low harvest intensity for extracting trees, and that sustainable forest use is at high risk when applying final fellings in the form of clearcut or shelterwood. To assess the extent of the risks of a final felling, we call for studies assessing leaching effects over longer time spans following tree harvest to even better underpin sustainability criteria of forest management.

Acknowledgements

This research is part of the Nutrient Balance project and was funded by the Dutch Research Council (NWO, No. ALWGS.2017.004). We acknowledge Leo Goudzwaard, Henk van Roekel, Flora van Eupen and many others for their valuable help with sample processing and lab analyses. We thank our partners, National Forest Service, Union of private Forest Owner Groups, Het Loo Royal Estate, Staro nature and countryside, Borgman management consultants, National Park de Hoge Veluwe and Blom Ecology for financial support, permission to work in their forest or other provided services.

Supplementary information

Table S6.1 Results of the generalized linear mixed model analysis showing the relationship between the modelled interception and the field interception in interaction with harvest intensity and tree species. Temporal autocorrelation is captured using a bspline smoother (not shown).

Explaining term	Estimate	S.E.	z	Pr(> z)
Intercept	5.6	1.2	4.9	< 0.001
Field interception	9.9	1.0	9.6	< 0.001
High-thinning	-0.24	1.2	-0.20	n.s.
Shelterwood	0.61	1.1	0.55	n.s.
Douglas fir	1.6	1.1	1.5	n.s.
Scots pine	2.3	1.0	2.3	< 0.05
Scaled modelled soil moisture * High-thinning	-0.46	1.1	-0.42	n.s.
Scaled modelled soil moisture * Shelterwood	-6.8	1.1	-0.63	< 0.001
Scaled modelled soil moisture * Douglas fir	-1.1	1.0	-1.1	n.s.
Scaled modelled soil moisture * Scots pine	-2.5	1.0	-2.6	< 0.01

Table S6.2 Interpolated precipitation in mm over the measurement period (April 2020 – Mart 2021) of nearby weather stations for the three species and the five sites. The data is derived from KNMI KNMI (2021b), the interpolation method is described in **Ch3**.

Site	Beech	Douglas fir	Scots pine
1	782	751	781
2	787	798	797
3	702	735	734
4	731	774	778
5	605	629	629

Table S6.3 Mean monthly values $\pm$ standard error ($n = 60$) of the concentrations of NO_3 , NH_4 , N_{tot} , S, P, Ca, K, Mg, Mn, Cu, Fe, Zn, Al and Na (mg L^{-1}) in soil moisture at 50 cm depth. The concentrations are measured for the mineral soil of beech (BE), Douglas fir (DG) and Scots pine (SP) plots for the harvest intensity treatments high-thinning, shelterwood and clearcut and the unharvested control. The mean values are averaged over the year based on 5 replications (plots). The dissolved concentrations of the high-thinning, shelterwood and clearcut are based on the non-mulched whole tree harvest plots as these plots best approximate the soil conditions of the unharvested control. For each species, the differences between the harvest treatments are denoted with different letters according to Tukey's *posthoc* test at a significance level of $P < 0.05$. The differences between species within the same harvest intensity are given in the columns *Sp* based on Tukey's *posthoc* test at a significance level of $P < 0.05$. Anova test-statistics for the used statistical model (model 1) are given in Table 6.1, the within treatment differences between whole tree harvest, stem only harvest and the post-harvest soil preparation are given in Table S6.2.

	Control	<i>Sp</i>	High-thinning	<i>Sp</i>	Shelterwood	<i>Sp</i>	Clearcut	<i>Sp</i>
NO_3								
BE	3.8 ± 1.0^a	a	3.8 ± 0.90^a	a	11 ± 1.2^b	a	18 ± 1.8^c	b
DG	8.8 ± 1.6^{bc}	b	3.8 ± 0.67^a	a	12 ± 1.5^d	a	7.8 ± 0.75^b	a
SP	5.5 ± 0.9^{ab}	ab	5.7 ± 0.92^a	a	8.7 ± 0.98^b	a	9.0 ± 1.2^{ab}	a
NH_4								
BE	0.25 ± 0.072^a	a	0.33 ± 0.082^a	a	0.77 ± 0.15^b	b	0.67 ± 0.14^b	a
DG	0.66 ± 0.21^{ab}	a	0.32 ± 0.11^a	a	0.28 ± 0.057^{bc}	ab	0.37 ± 0.053^c	a
SP	0.34 ± 0.095^b	a	0.24 ± 0.064^a	a	0.29 ± 0.052^{ab}	a	0.38 ± 0.11^b	a
N_{tot}								
BE	4.0 ± 0.9^a	a	5.0 ± 0.94^a	a	13 ± 1.3^b	b	19 ± 1.8^b	b
DG	10 ± 1.6^a	b	6.7 ± 0.95^a	b	13 ± 1.5^b	ab	9.0 ± 0.79^b	a
SP	6.2 ± 0.89^a	b	6.7 ± 0.95^a	b	9.2 ± 0.94^b	a	10 ± 1.1^b	a
S								
BE	4.7 ± 0.66^b	b	1.7 ± 0.093^a	a	2.1 ± 0.17^a	a	2.1 ± 0.13^{ab}	b
DG	6.7 ± 1.1^b	c	5.5 ± 0.83^a	b	2.8 ± 0.23^a	b	2.8 ± 0.28^a	b
SP	1.8 ± 0.12^b	a	1.7 ± 0.097^{ab}	a	1.9 ± 0.13^{ab}	a	1.5 ± 0.1^a	a
P								
BE	0.017 ± 0.0027^a	a	0.018 ± 0.0023^a	a	0.021 ± 0.0033^a	a	0.015 ± 0.0019^a	a
DG	0.067 ± 0.024^a	a	0.28 ± 0.11^a	a	0.031 ± 0.0044^a	a	0.045 ± 0.0079^a	ab
SP	0.022 ± 0.0031^{ab}	a	0.018 ± 0.002^{ab}	a	0.018 ± 0.0027^a	a	0.019 ± 0.0017^b	b
Ca								
BE	2.2 ± 0.18^c	b	1.4 ± 0.17^a	a	1.9 ± 0.26^{ab}	a	3.3 ± 0.49^{bc}	a
DG	3.8 ± 0.56^{ab}	b	4.2 ± 0.71^b	b	3.1 ± 0.39^{ab}	b	3.1 ± 0.26^a	b
SP	1.8 ± 0.18^b	a	1.3 ± 0.16^a	a	1.4 ± 0.16^{ab}	a	1.7 ± 0.14^b	a
K								
BE	1.6 ± 0.21^a	a	1.6 ± 0.22^a	a	2.3 ± 0.28^b	a	5.0 ± 0.53^c	b
DG	5.7 ± 0.78^b	b	2.8 ± 0.4^a	b	4.5 ± 0.57^b	b	3.1 ± 0.29^a	a
SP	2.0 ± 0.26^a	a	2.0 ± 0.25^a	b	3.4 ± 0.31^b	b	3.7 ± 0.35^b	ab
Mg								
BE	0.95 ± 0.083^b	b	0.74 ± 0.084^a	b	1.1 ± 0.10^b	b	1.3 ± 0.16^b	ab
DG	2.4 ± 0.27^a	c	2.4 ± 0.43^{ab}	c	1.7 ± 0.18^{ab}	c	1.4 ± 0.14^a	b
SP	0.63 ± 0.066^a	a	0.65 ± 0.084^a	a	0.81 ± 0.086^b	a	0.82 ± 0.054^b	a
Mn								
BE	0.074 ± 0.0091^{ab}	a	0.051 ± 0.0065^a	b	0.092 ± 0.017^{ab}	a	0.13 ± 0.020^b	a
DG	0.21 ± 0.039^a	a	0.20 ± 0.035^a	c	0.17 ± 0.023^a	b	0.27 ± 0.039^b	b
SP	0.041 ± 0.0045^{1b}	b	0.036 ± 0.0044^a	a	0.043 ± 0.0051^b	a	0.057 ± 0.005^c	a

Continued on the next page.

Cu								
BE	0.0015±0.00022 ^a	a	0.002 ± 0.00027 ^{ab}	ab	0.0018 ± 0.0003 ^b	b	0.0011±0.00018 ^a	a
DG	0.002 ± 0.00025 ^{ab}	ab	0.002 ± 0.00028 ^b	b	0.0011±0.00018 ^a	a	0.0019±0.00025 ^b	b
SP	0.002 ± 0.00023 ^a	a	0.0016±0.00018 ^a	a	0.0017 ± 0.0002 ^a	a	0.0015±0.00022 ^a	a
Fe								
BE	1.3 ± 0.35 ^b	a	0.087 ± 0.0082 ^a	a	0.11 ± 0.0098 ^{ab}	a	0.093 ± 0.006 ^{ab}	a
DG	0.13 ± 0.013 ^{ab}	a	0.17 ± 0.016 ^{ab}	b	0.16 ± 0.019 ^b	a	0.16 ± 0.014 ^a	b
SP	0.24 ± 0.043 ^a	a	0.15 ± 0.014 ^a	b	0.12 ± 0.0081 ^a	a	0.18 ± 0.017 ^a	b
Zn								
BE	0.4 ± 0.047 ^a	a	0.45 ± 0.068 ^a	a	0.58 ± 0.066 ^b	a	0.73 ± 0.061 ^b	ab
DG	1.4 ± 0.32 ^a	b	0.81 ± 0.088 ^a	b	0.85 ± 0.11 ^a	a	0.98 ± 0.15 ^a	b
SP	0.67 ± 0.078 ^a	b	0.47 ± 0.047 ^a	a	0.56 ± 0.062 ^a	a	0.57 ± 0.058 ^a	a
Al								
BE	2.9 ± 0.38 ^a	a	2.5 ± 0.32 ^a	a	6.3 ± 0.63 ^b	b	8.7 ± 0.75 ^c	b
DG	5.3 ± 0.71 ^{ab}	b	4.5 ± 0.43 ^{ab}	b	5.9 ± 0.64 ^b	ab	3.9 ± 0.34 ^a	a
SP	3.4 ± 0.37 ^a	a	3.7 ± 0.40 ^a	b	4.7 ± 0.49 ^a	a	4.5 ± 0.56 ^a	a
Na								
BE	9.7 ± 0.99 ^d	b	4.5 ± 0.19 ^c	a	4.3 ± 0.48 ^b	a	2.6 ± 0.14 ^a	a
DG	17 ± 2.8 ^b	c	10 ± 0.98 ^b	b	5.1 ± 0.55 ^a	a	3.4 ± 0.21 ^a	b
SP	6.0 ± 0.38 ^d	a	5.1 ± 0.36 ^c	a	4.1 ± 0.34 ^b	a	2.5 ± 0.14 ^a	a

Table S6.4 Mean values $\pm$ standard error ($n = 60$) of the monthly dissolved concentrations of NO_3 , NH_4 , N_{tot} , S , P , Ca , K , Mg , Mn , Cu , Fe , Zn , Al and Na (mg L^{-1}) in the soil moisture at 50 cm depth in the mineral soil. Dissolved concentrations are provided for beech (BE), Douglas fir (DG), and Scots pine (SP) plots under the harvest intensities of high-thinning, shelterwood, and clearcut, as well as the harvest methods stem-only harvest (SOH) and whole tree harvest (WTH). The value 'O' represents non-mulched plots, while 'K' represents mulched plots. The mean values are averaged over the year based on 5 replications (plots). Within each harvest intensity treatment, differences per species for the two harvest methods are denoted with different letters (a and b) according to Tukey's *posthoc* test at a significance level of $P < 0.05$. Differences within harvest intensity treatment per species for soil preparation are denoted with the letters A and B according to Tukey's *posthoc* test at a significance level of $P < 0.05$. No post-hoc test was conducted when the ANOVA was not significant (Table 6.1). Differences between treatments and species are in Table S6.1. The different statistical models used (resp. model 2 and 3) are described in the methods.

	Statistical model 2				Statistical model 3				Statistical model 3				Clearcut			
	High-thinning				Shelterwood				WTH							
	SOH	O	WTH	O	SOH	O	K	O	SOH	O	K	O	SOH	O	K	WTH
NO_3	BE	4.9 $\pm$ 0.97 ^b	3.8 $\pm$ 0.90 ^a	12 $\pm$ 1.3 ^a	10 $\pm$ 1.6	11 $\pm$ 1.2 ^a	9.0 $\pm$ 1.3	17 $\pm$ 1.9 ^a	17 $\pm$ 1.7	18 $\pm$ 1.8 ^a	15 $\pm$ 1.5	18 $\pm$ 1.8 ^a	17 $\pm$ 1.9 ^a	17 $\pm$ 1.7	18 $\pm$ 1.8 ^a	15 $\pm$ 1.5
	DG	9.2 $\pm$ 1.2 ^b	3.8 $\pm$ 0.67 ^a	11 $\pm$ 1.7 ^a	16 $\pm$ 2.2	12 $\pm$ 1.5 ^a	13 $\pm$ 1.3	11 $\pm$ 1.5 ^b	11 $\pm$ 1.3	7.8 $\pm$ 0.75 ^a	8.4 $\pm$ 1.3	7.8 $\pm$ 0.75 ^a	11 $\pm$ 1.5 ^b	11 $\pm$ 1.3	7.8 $\pm$ 0.75 ^a	8.4 $\pm$ 1.3
	SP	6.2 $\pm$ 0.92 ^b	5.7 $\pm$ 0.92 ^a	10 $\pm$ 1.0 ^a	8.4 $\pm$ 1.1	8.7 $\pm$ 1.0 ^a	7.1 $\pm$ 0.9	12 $\pm$ 1.2 ^a	12 $\pm$ 1.4	9.0 $\pm$ 1.2 ^a	8.9 $\pm$ 1.0	9.0 $\pm$ 1.2 ^a	12 $\pm$ 1.2 ^a	12 $\pm$ 1.4	9.0 $\pm$ 1.2 ^a	8.9 $\pm$ 1.0
	NH_4	BE	0.38 $\pm$ 0.14 ^a	0.33 $\pm$ 0.082 ^a	0.76 $\pm$ 0.17 ^a	0.7 $\pm$ 0.17	0.77 $\pm$ 0.15 ^a	0.51 $\pm$ 0.13	1.1 $\pm$ 0.2 ^a	0.87 $\pm$ 0.16	0.67 $\pm$ 0.14 ^a	1 $\pm$ 0.18	1.1 $\pm$ 0.2 ^a	0.87 $\pm$ 0.16	0.67 $\pm$ 0.14 ^a	1 $\pm$ 0.18
N_{tot}	DG	0.28 $\pm$ 0.069 ^a	0.32 $\pm$ 0.11 ^a	0.3 $\pm$ 0.11 ^b	0.47 $\pm$ 0.12	0.28 $\pm$ 0.06 ^a	0.4 $\pm$ 0.06	0.63 $\pm$ 0.15 ^b	0.56 $\pm$ 0.17	0.37 $\pm$ 0.05 ^a	0.25 $\pm$ 0.04	0.37 $\pm$ 0.05 ^a	0.63 $\pm$ 0.15 ^b	0.56 $\pm$ 0.17	0.37 $\pm$ 0.05 ^a	0.25 $\pm$ 0.04
	SP	0.35 $\pm$ 0.086 ^a	0.24 $\pm$ 0.064 ^a	0.37 $\pm$ 0.1 ^a	0.45 $\pm$ 0.12	0.29 $\pm$ 0.05 ^a	0.44 $\pm$ 0.12	0.19 $\pm$ 0.04 ^a	0.72 $\pm$ 0.2	0.38 $\pm$ 0.11 ^a	0.38 $\pm$ 0.09	0.38 $\pm$ 0.11 ^a	0.19 $\pm$ 0.04 ^a	0.72 $\pm$ 0.2	0.38 $\pm$ 0.11 ^a	0.38 $\pm$ 0.09
	BE	6.0 $\pm$ 1.1 ^a	5.0 $\pm$ 0.94 ^a	14 $\pm$ 1.4 ^a	12 $\pm$ 1.7	13 $\pm$ 1.3 ^a	10 $\pm$ 1.4	18 $\pm$ 1.9 ^a	19 $\pm$ 1.7	19 $\pm$ 1.8 ^a	16 $\pm$ 1.6	19 $\pm$ 1.8 ^a	18 $\pm$ 1.9 ^a	19 $\pm$ 1.7	19 $\pm$ 1.8 ^a	16 $\pm$ 1.6
	DG	10 $\pm$ 1.2 ^b	6.7 $\pm$ 0.95 ^a	12 $\pm$ 1.7 ^a	17 $\pm$ 2.1	13 $\pm$ 1.5 ^a	14 $\pm$ 1.3	12 $\pm$ 1.5 ^a	13 $\pm$ 1.4	9.0 $\pm$ 0.79 ^a	9.4 $\pm$ 1.3	9.0 $\pm$ 0.79 ^a	12 $\pm$ 1.5 ^a	13 $\pm$ 1.4	9.0 $\pm$ 0.79 ^a	9.4 $\pm$ 1.3
S	SP	7.4 $\pm$ 1.0 ^b	6.7 $\pm$ 0.95 ^a	11 $\pm$ 1.1 ^a	10 $\pm$ 1.1	9.2 $\pm$ 0.94 ^a	8.4 $\pm$ 1.0	12 $\pm$ 1.2 ^a	14 $\pm$ 1.5	10 $\pm$ 1.1 ^a	10 $\pm$ 1.0	10 $\pm$ 1.1 ^a	12 $\pm$ 1.2 ^a	14 $\pm$ 1.5	10 $\pm$ 1.1 ^a	10 $\pm$ 1.0
	BE	2.0 $\pm$ 0.29 ^a	1.7 $\pm$ 0.093 ^a	1.9 $\pm$ 0.14 ^a	2.0 $\pm$ 0.15	2.1 $\pm$ 0.17 ^b	2.2 $\pm$ 0.40	1.8 $\pm$ 0.11 ^a	1.9 $\pm$ 0.20	2.1 $\pm$ 0.13 ^a	1.9 $\pm$ 0.15	2.1 $\pm$ 0.13 ^a	1.8 $\pm$ 0.11 ^a	1.9 $\pm$ 0.20	2.1 $\pm$ 0.13 ^a	1.9 $\pm$ 0.15
	DG	3.5 $\pm$ 0.36 ^a	5.5 $\pm$ 0.83 ^a	2.9 $\pm$ 0.21 ^a	2.7 $\pm$ 0.18	2.8 $\pm$ 0.23 ^{ab}	2.6 $\pm$ 0.19 ^a	3.2 $\pm$ 0.41 ^b	3.3 $\pm$ 0.33	2.8 $\pm$ 0.28 ^a	2.5 $\pm$ 0.24	2.8 $\pm$ 0.28 ^a	3.2 $\pm$ 0.41 ^b	3.3 $\pm$ 0.33	2.8 $\pm$ 0.28 ^a	2.5 $\pm$ 0.24
	SP	1.5 $\pm$ 0.13 ^a	1.7 $\pm$ 0.097 ^b	1.6 $\pm$ 0.13 ^a	1.8 $\pm$ 0.26	1.9 $\pm$ 0.13 ^b	1.9 $\pm$ 0.40	1.8 $\pm$ 0.21 ^{aa}	1.9 $\pm$ 0.21 ^B	1.5 $\pm$ 0.10 ^a	2.0 $\pm$ 0.16	1.5 $\pm$ 0.10 ^a	1.8 $\pm$ 0.21 ^{aa}	1.9 $\pm$ 0.21 ^B	1.5 $\pm$ 0.10 ^a	2.0 $\pm$ 0.16
P	BE	0.019 $\pm$	0.018 $\pm$	0.017 $\pm$	0.024 $\pm$	0.021 $\pm$	0.019 $\pm$	0.013 $\pm$	0.020 $\pm$	0.015 $\pm$	0.016 $\pm$	0.015 $\pm$	0.013 $\pm$	0.020 $\pm$	0.015 $\pm$	0.016 $\pm$
	DG	0.0041 ^b	0.0023 ^a	0.002 $\pm$	0.0041	0.0033 ^a	0.0029	0.0013 ^a	0.0042	0.0019 ^b	0.0024	0.0019 ^b	0.0013 ^a	0.0042	0.0019 ^b	0.0024
	SP	0.035 $\pm$	0.28 $\pm$	0.021 $\pm$	0.017 $\pm$	0.031 $\pm$	0.031 $\pm$	0.082 $\pm$	0.026 $\pm$	0.045 $\pm$	0.026 $\pm$	0.045 $\pm$	0.082 $\pm$	0.026 $\pm$	0.045 $\pm$	0.026 $\pm$
	SP	0.0063 ^a	0.11 ^a	0.0022 ^a	0.0018	0.0044 ^a	0.0049	0.0287 ^b	0.0051	0.0079 ^a	0.0039	0.0079 ^a	0.0287 ^b	0.0051	0.0079 ^a	0.0039
P	SP	0.038 $\pm$	0.018 $\pm$	0.036 $\pm$	0.025 $\pm$	0.018 $\pm$	0.047 $\pm$	0.020 $\pm$	0.075 $\pm$	0.019 $\pm$	0.022 $\pm$	0.019 $\pm$	0.020 $\pm$	0.075 $\pm$	0.019 $\pm$	0.022 $\pm$
	SP	0.012 ^a	0.002 ^a	0.0049 ^{ba}	0.0059 ^B	0.0027 ^{aA}	0.0252 ^B	0.0041 ^{aA}	0.0576 ^B	0.0017 ^{aA}	0.0039 ^B	0.0017 ^{aA}	0.0041 ^{aA}	0.0576 ^B	0.0017 ^{aA}	0.0039 ^B

Ca	BE	1.5 ± 0.2 ^a	1.4 ± 0.17 ^a	2.7 ± 0.41 ^a	2.5 ± 0.34	1.9 ± 0.26 ^a	1.9 ± 0.26	2.0 ± 0.34 ^a	2.8 ± 0.34	3.3 ± 0.49 ^a	2.9 ± 0.43
	DG	3 ± 0.39 ^a	4.2 ± 0.71 ^a	2.5 ± 0.27 ^a	2.8 ± 0.3	3.1 ± 0.39 ^a	2.8 ± 0.23	2.9 ± 0.34 ^a	3.1 ± 0.38	3.1 ± 0.26 ^a	2.5 ± 0.28
	SP	1.3 ± 0.13 ^a	1.3 ± 0.16 ^a	1.6 ± 0.11 ^b	1.4 ± 0.14	1.4 ± 0.16 ^a	1.7 ± 0.23	2.1 ± 0.17 ^a	2.1 ± 0.18	1.7 ± 0.14 ^a	1.7 ± 0.16
K	BE	2.1 ± 0.30 ^b	1.6 ± 0.22 ^a	4.3 ± 0.58 ^b	4.6 ± 0.67	2.3 ± 0.28 ^a	2.9 ± 0.4	5.3 ± 0.78 ^b	5.2 ± 0.64	5.0 ± 0.53 ^a	4.7 ± 0.54
	DG	3.3 ± 0.43 ^b	2.8 ± 0.4 ^a	4.3 ± 0.52 ^a	5.7 ± 0.59	4.5 ± 0.57 ^b	4.9 ± 0.47	6.4 ± 0.63 ^b	4.9 ± 0.46	3.1 ± 0.29 ^a	3.8 ± 0.43
	SP	2.8 ± 0.35 ^b	2.0 ± 0.25 ^a	4.0 ± 0.37 ^{ab}	2.5 ± 0.28 ^a	3.4 ± 0.31 ^a	2.6 ± 0.26	4.8 ± 0.63 ^{bb}	3.9 ± 0.30 ^a	3.7 ± 0.35 ^a	3.6 ± 0.33
Mg	BE	0.76 ± 0.083 ^a	0.74 ± 0.084 ^a	1.2 ± 0.17 ^a	1.2 ± 0.17	1.1 ± 0.10 ^{ab}	1.0 ± 0.14 ^a	1.3 ± 0.16 ^a	1.3 ± 0.16	1.3 ± 0.16 ^{ab}	1.1 ± 0.14 ^a
	DG	2.2 ± 0.21 ^a	2.4 ± 0.43 ^b	1.5 ± 0.13 ^a	1.7 ± 0.19	1.7 ± 0.18 ^{ab}	1.6 ± 0.14 ^a	1.7 ± 0.19 ^a	1.4 ± 0.13	1.4 ± 0.14 ^{ab}	1.2 ± 0.13 ^a
	SP	0.85 ± 0.082 ^b	0.65 ± 0.084 ^a	1.0 ± 0.08 ^a	0.72 ± 0.072	0.81 ± 0.086 ^{ab}	0.83 ± 0.112 ^a	1.1 ± 0.10 ^a	1.2 ± 0.15	0.82 ± 0.054 ^{ab}	0.8 ± 0.078 ^a
Mn	BE	0.048 ±	0.051 ±	0.071 ±	0.076 ±	0.092 ±	0.066 ±	0.05 ±	0.089 ±	0.125 ±	0.12 ±
	DG	0.0069 ^a	0.0065 ^a	0.011 ^a	0.013	0.017 ^b	0.009	0.007 ^a	0.014	0.02 ^b	0.019
	SP	0.11 ±	0.20 ±	0.14 ±	0.16 ±	0.17 ±	0.18 ±	0.20 ±	0.20 ±	0.27 ±	0.19 ±
Cu	BE	0.0012 ±	0.0020 ±	0.0018 ±	0.0014 ±	0.0018 ±	0.002 ±	0.0012 ±	0.0015 ±	0.0011 ±	0.0014 ±
	DG	0.0017 ^a	0.00027 ^b	0.0003 ^a	0.0003	0.0003 ^a	0.0004	0.0002 ^b	0.0003	0.0002 ^a	0.0002
	SP	0.0016 ±	0.002 ±	0.0014 ±	0.0012 ±	0.0011 ±	0.0014 ±	0.0008 ±	0.0013 ±	0.0019 ±	0.0013 ±
Fe	BE	0.00024 ^a	0.00028 ^a	0.0002 ^a	0.0002	0.0002 ^a	0.0002	0.0002 ^a	0.0002	0.0002 ^b	0.0002
	DG	0.0018 ±	0.0016 ±	0.0014 ±	0.0011 ±	0.0017 ±	0.0012 ±	0.0011 ±	0.0011 ±	0.0015 ±	0.0014 ±
	SP	0.00024 ^a	0.00018 ^a	0.0002 ^a	0.0002	0.0002 ^a	0.0002	0.0002 ^a	0.0002	0.0002 ^a	0.0002
Zn	BE	0.10 ± 0.011 ^a	0.087 ± 0.008 ^a	0.11 ± 0.011 ^{ab}	0.13 ± 0.014 ^b	0.11 ± 0.010 ^a	0.10 ± 0.012	0.09 ± 0.009 ^a	0.1 ± 0.009	0.090 ± 0.006 ^a	0.12 ± 0.012 ^b
	DG	0.14 ± 0.017 ^a	0.17 ± 0.016 ^b	0.11 ± 0.012 ^{ab}	0.17 ± 0.016 ^b	0.16 ± 0.019 ^b	0.18 ± 0.022	0.14 ± 0.014 ^a	0.14 ± 0.015	0.16 ± 0.014 ^{ab}	0.13 ± 0.013 ^a
	SP	0.091 ± 0.001 ^a	0.15 ± 0.014 ^b	0.11 ± 0.011 ^a	0.12 ± 0.01	0.12 ± 0.008 ^b	0.12 ± 0.013	0.11 ± 0.010 ^a	0.13 ± 0.017	0.18 ± 0.017 ^{ab}	0.13 ± 0.011 ^a
Al	BE	0.42 ± 0.048 ^a	0.45 ± 0.068 ^b	0.65 ± 0.10 ^b	0.53 ± 0.063	0.58 ± 0.066 ^{ab}	0.56 ± 0.091 ^a	0.47 ± 0.039 ^{ab}	0.53 ± 0.048 ^b	0.73 ± 0.061 ^{bb}	0.53 ± 0.049 ^a
	DG	0.70 ± 0.14 ^a	0.81 ± 0.088 ^a	0.80 ± 0.10 ^a	0.68 ± 0.097	0.85 ± 0.11 ^a	0.70 ± 0.07	0.81 ± 0.145 ^{ab}	1.01 ± 0.215	0.98 ± 0.151 ^{bb}	0.62 ± 0.074 ^a
	SP	0.43 ± 0.049 ^a	0.47 ± 0.047 ^a	0.55 ± 0.061 ^a	0.5 ± 0.055	0.56 ± 0.062 ^a	0.63 ± 0.068	0.70 ± 0.093 ^{ab}	0.57 ± 0.089 ^a	0.57 ± 0.058 ^{ab}	0.50 ± 0.056 ^a
Na	BE	3.3 ± 0.41	2.5 ± 0.32	5.5 ± 0.53	4.9 ± 0.5	6.3 ± 0.63 ^b	5.1 ± 0.66 ^a	7.9 ± 0.91	8.9 ± 0.91	8.7 ± 0.75 ^b	6.5 ± 0.66 ^a
	DG	5.8 ± 0.64	4.5 ± 0.43	5.8 ± 0.69 ^a	8.2 ± 0.87 ^b	5.9 ± 0.64	6 ± 0.51	5.3 ± 0.56 ^a	6.0 ± 0.69 ^b	3.9 ± 0.34	5.0 ± 0.55
	SP	3.5 ± 0.52	3.7 ± 0.40	4.8 ± 0.48	5.3 ± 0.64	4.7 ± 0.49	4.2 ± 0.51	6.1 ± 0.63	6.5 ± 0.71	4.5 ± 0.56	5.0 ± 0.55
	BE	5.5 ± 0.74	4.5 ± 0.19	4.0 ± 0.34 ^a	4.3 ± 0.33	4.3 ± 0.48 ^b	4.5 ± 0.65	2.5 ± 0.13 ^a	3.1 ± 0.18 ^b	2.6 ± 0.14	2.8 ± 0.17
	DG	12 ± 1.1	10 ± 0.98	5.3 ± 0.39	4.9 ± 0.31	5.1 ± 0.55 ^b	4.0 ± 0.33 ^a	3.3 ± 0.34	3.7 ± 0.3	3.4 ± 0.21 ^b	3.0 ± 0.24 ^a
	SP	5.7 ± 0.33	5.1 ± 0.36	3.2 ± 0.17	4 ± 0.43	4.1 ± 0.34	3.5 ± 0.19	3 ± 0.3	2.9 ± 0.18	2.5 ± 0.14	2.7 ± 0.22

Table S6.5 Mean values $\pm$ standard error ($n = 60$) of the annual leaching flux of NO_3 , NH_4 , N_{tot} , S, P, Ca, K, Mg, Mn, Cu, Fe, Zn, Al and Na ($\text{kg ha}^{-1} \text{ yr}^{-1}$) for beech (BE), Douglas fir (DG) and Scots pine (SP) plots and the harvest intensity treatments high-thinning, shelterwood and clearcut and the unharvested control. The annual mean values are based on 5 replications (plots). The dissolved nutrient concentrations of the high-thinning, shelterwood and clearcut that were used to calculate the annual fluxes are based on the non-mulched whole tree harvest plots as these plots best approximates the soil conditions of the unharvested control. For each species, the differences between the harvest treatments are denoted with different letters according to Tukey's *posthoc* test at a significance level of $P < 0.05$. The differences between species within the same harvest intensity are given in the columns *Sp* based on Tukey's *posthoc* test at a significance level of $P < 0.05$.

	Control	<i>Sp</i>	High-thinning	<i>Sp</i>	Shelterwood	<i>Sp</i>	Clearcut	<i>Sp</i>
NO_3								
BE	6.2 ± 2.8^a	a	5.8 ± 2.2^a	a	45 ± 5.3^b	ab	91 ± 17^c	b
DG	10 ± 4.0^a	a	8.4 ± 2.2^a	a	61 ± 8.7^b	b	37 ± 3.7^b	a
SP	8.9 ± 3.3^a	a	10 ± 2.8^a	a	35 ± 5.7^b	a	47 ± 12^b	a
NH_4								
BE	0.36 ± 0.064^a	a	0.37 ± 0.15^a	a	3.3 ± 1.2^b	b	1.6 ± 0.63^b	a
DG	0.72 ± 0.24^a	a	0.87 ± 0.22^a	b	1.2 ± 0.39^a	a	1.5 ± 0.025^a	a
SP	0.33 ± 0.02^{ab}	a	0.30 ± 0.042^a	a	0.92 ± 0.23^{bc}	a	1.5 ± 0.49^c	a
N_{tot}								
BE	4.4 ± 0.92^a	a	8.8 ± 2.0^b	a	50 ± 5.7^c	a	94 ± 17^c	b
DG	13 ± 4.0^a	b	14 ± 1.0^a	a	64 ± 8.3^b	a	42 ± 3.6^b	a
SP	7.7 ± 1.4^a	ab	12 ± 2.6^a	a	36 ± 5.7^b	a	51 ± 12^b	a
S								
BE	5.7 ± 0.81^{ab}	ab	5.3 ± 0.79^a	a	8.2 ± 1.6^{bc}	ab	9.1 ± 1.4^c	b
DG	10 ± 3.5^a	b	13 ± 5.6^a	b	12 ± 2.5^a	b	13 ± 3.7^a	b
SP	4.5 ± 0.76^a	a	4.9 ± 0.72^{ab}	a	7.4 ± 1.4^b	a	6.9 ± 1.4^{ab}	a
P								
BE	0.045 ± 0.0059^a	a	0.059 ± 0.012^a	a	0.063 ± 0.013^a	a	0.058 ± 0.013^a	a
DG	0.056 ± 0.016^b	b	0.064 ± 0.025^{bc}	b	0.13 ± 0.052^a	b	0.13 ± 0.018^c	b
SP	0.043 ± 0.011^a	b	0.049 ± 0.0088^a	b	0.076 ± 0.012^b	ab	0.10 ± 0.017^c	a
Ca								
BE	6.4 ± 1.1^{ab}	a	3.9 ± 1.0^a	a	6.4 ± 1.7^{ab}	a	6.9 ± 2.0^b	a
DG	5.3 ± 0.76^a	a	7.3 ± 3.1^{ab}	b	13 ± 2.9^{bc}	b	15 ± 1.2^c	b
SP	4.3 ± 0.88^{ab}	a	2.9 ± 0.35^a	a	6.0 ± 0.99^{bc}	a	8.5 ± 1.4^c	a
K								
BE	4.2 ± 0.98^a	a	3.0 ± 0.54^a	a	8.4 ± 1.5^b	a	15 ± 1.5^c	a
DG	12 ± 3.4^b	b	6.7 ± 0.67^a	b	18 ± 3.1^b	b	14 ± 1.4^b	a
SP	3.5 ± 0.48^a	a	4.6 ± 0.64^a	ab	14 ± 1.1^b	ab	16 ± 3.3^b	a
Mg								
BE	2.6 ± 0.29^{ab}	b	2.0 ± 0.30^a	a	4.3 ± 1.0^{bc}	a	4.5 ± 1.0^c	b
DG	4.9 ± 1.4^{ab}	c	3.4 ± 0.53^a	b	8.2 ± 0.96^b	b	6.9 ± 1.0^b	ab
SP	1.3 ± 0.17^a	a	1.6 ± 0.29^a	a	3.3 ± 0.40^b	a	4.0 ± 0.41^b	a
Mn								
BE	0.24 ± 0.059^a	b	0.12 ± 0.032^a	ab	0.27 ± 0.085^a	a	0.34 ± 0.16^a	a
DG	0.21 ± 0.046^a	b	0.27 ± 0.071^{ab}	b	0.82 ± 0.22^{bc}	b	1.2 ± 0.47^c	b
SP	0.081 ± 0.02^a	a	0.076 ± 0.015^a	a	0.16 ± 0.016^b	a	0.26 ± 0.052^c	a
Cu								
BE	0.0049 ± 0.0008^a	a	0.0053 ± 0.0009^a	ab	0.0064 ± 0.0022^a	a	0.0056 ± 0.00052^a	a
DG	0.0047 ± 0.00071^a	a	0.0067 ± 0.0011^{ab}	b	0.005 ± 0.00088^a	a	0.0087 ± 0.0014^b	a
SP	0.0061 ± 0.0010^{ab}	a	0.0036 ± 0.00042^a	a	0.0071 ± 0.0013^b	a	0.0061 ± 0.0011^{ab}	a

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Fe						
BE	0.40 ± 0.15^a	a	0.31 ± 0.077^a	a	0.45 ± 0.095^a	a
DG	0.41 ± 0.049^a	a	0.5 ± 0.16^a	a	0.81 ± 0.28^a	b
SP	0.57 ± 0.20^a	a	0.49 ± 0.089^a	a	0.46 ± 0.075^a	ab
Zn						
BE	0.79 ± 0.19^a	a	1.1 ± 0.35^a	a	1.7 ± 0.46^b	a
DG	0.83 ± 0.23^a	ab	1.8 ± 0.93^a	a	3.4 ± 1.3^b	b
SP	1.3 ± 0.35^a	b	0.9 ± 0.14^a	a	1.9 ± 0.67^a	a
Al						
BE	7.1 ± 1.4^a	a	5.7 ± 2.1^a	a	26 ± 3.9^b	a
DG	9.0 ± 3.2^a	a	12 ± 1.8^{ab}	b	27 ± 3.2^c	a
SP	7.9 ± 0.83^a	a	8.6 ± 2.2^a	ab	19 ± 3.7^b	a
Na						
BE	20 ± 1.6^b	a	17 ± 1.6^b	a	18 ± 4.1^b	ab
DG	28 ± 3.6^c	b	32 ± 4.3^{bc}	b	21 ± 2.6^{ab}	b
SP	17 ± 2.3^b	a	16 ± 3.4^{ab}	a	16 ± 1.0^b	a

Table S6.6 Mean values $\pm$ standard error ($n = 60$) of the annual leaching of NO_3 , NH_4 , N_{tot} , S , P , Ca , K , Mg , Mn , Cu , Fe , Zn , Al and Na ($\text{kg ha}^{-1} \text{ yr}^{-1}$). The mean values are averaged over the year based on 5 replications (plots) and are given for beech (BE), Douglas fir (DG) and Scots pine (SP) for the harvest intensities high-thinning, shelterwood and clearcut. Within harvest intensity treatment differences per species for the harvest methods stem only harvest (SOH) and whole tree harvest (WTH) are denoted with different letters according to Tukey's *posthoc* test at a significance level of $P < 0.05$. Anova test statistics are given in Table 6.3, the differences between treatments and species are given in Table S6.4. The different statistical models (resp. model 2 and 3) are described in the methods.

	Statistical model 2					Statistical model 3				
	High-thinning					Shelterwood				
	SOH	WTH	O	K		SOH	WTH	O	K	
NO_3	BE	8.3 $\pm$ 4.3	5.8 $\pm$ 2.2	42 $\pm$ 13	45 $\pm$ 5.3	35 $\pm$ 7.8	85 $\pm$ 16	76 $\pm$ 24	91 $\pm$ 17	62 $\pm$ 9.9
	DG	19 $\pm$ 6.7	8.4 $\pm$ 2.2	54 $\pm$ 21	61 $\pm$ 8.7	54 $\pm$ 9.8	56 $\pm$ 9.3	60 $\pm$ 6.1	37 $\pm$ 3.7	40 $\pm$ 8.5
	SP	16 $\pm$ 1.2	10 $\pm$ 2.8	49 $\pm$ 8.1	35 $\pm$ 5.7	33 $\pm$ 7.0	69 $\pm$ 9.9	60 $\pm$ 14	47 $\pm$ 12	48 $\pm$ 9.9
NH_4	BE	0.3 $\pm$ 0.049	0.37 $\pm$ 0.15	3.1 $\pm$ 1.2	2 $\pm$ 1.6	1.2 $\pm$ 0.43	4 $\pm$ 1.9	3 $\pm$ 2.6	1.6 $\pm$ 0.63	5.0 $\pm$ 2.1
	DG	0.45 $\pm$ 0.069	0.87 $\pm$ 0.22	1 $\pm$ 1.4	2.4 $\pm$ 0.3	1.6 $\pm$ 0.55	3.1 $\pm$ 1.3	2.2 $\pm$ 0.97	1.5 $\pm$ 0.025	0.84 $\pm$ 0.17
	SP	0.52 $\pm$ 0.11	0.3 $\pm$ 0.042	2.3 $\pm$ 1.1	1.9 $\pm$ 0.81	2.2 $\pm$ 1.0	0.73 $\pm$ 0.97	3 $\pm$ 0.24	1.5 $\pm$ 0.49	1.6 $\pm$ 0.42
N_{tot}	BE	6.7 $\pm$ 2.4	8.8 $\pm$ 2	56 $\pm$ 22	46 $\pm$ 15	39 $\pm$ 8.1	89 $\pm$ 14	82 $\pm$ 23	94 $\pm$ 17	69 $\pm$ 12
	DG	22 $\pm$ 7.5	14 $\pm$ 1	57 $\pm$ 20	84 $\pm$ 17	58 $\pm$ 8.6	60 $\pm$ 9.6	66 $\pm$ 4.3	42 $\pm$ 3.6	44 $\pm$ 8.0
	SP	18 $\pm$ 1.2	12 $\pm$ 2.6	54 $\pm$ 8	44 $\pm$ 7.5	39 $\pm$ 8.5	71 $\pm$ 11	68 $\pm$ 14	51 $\pm$ 12	52 $\pm$ 10
S	BE	4.5 $\pm$ 0.45 ^a	5.3 $\pm$ 0.79 ^b	6.0 $\pm$ 1.6 ^a	8.1 $\pm$ 1.0	8.0 $\pm$ 1.3	7.2 $\pm$ 1.5 ^a	7.9 $\pm$ 0.89	9.1 $\pm$ 1.4 ^a	9.2 $\pm$ 0.85
	DG	9.3 $\pm$ 1.9 ^a	7.2 $\pm$ 0.73 ^a	13 $\pm$ 1.4 ^a	12 $\pm$ 1.9	10 $\pm$ 0.93	16 $\pm$ 4.6 ^b	16 $\pm$ 3.4	13 $\pm$ 3.7 ^a	11 $\pm$ 1.3
	SP	3.6 $\pm$ 0.51 ^a	4.9 $\pm$ 0.72 ^a	6.2 $\pm$ 1.6 ^a	8.3 $\pm$ 1.2	9.8 $\pm$ 3.6	7.0 $\pm$ 0.92 ^a	8.3 $\pm$ 1.4	6.9 $\pm$ 1.4 ^a	9.9 $\pm$ 1.7
P	BE	0.039 $\pm$	0.059 $\pm$	0.061 $\pm$	0.093 $\pm$	0.086 $\pm$	0.058 $\pm$	0.059 $\pm$	0.058 $\pm$	0.087 $\pm$
	DG	0.0069 ^a	0.012 ^a	0.019 ^a	0.012	0.019	0.01 ^a	0.0097	0.013 ^a	0.013
	SP	0.13 $\pm$	0.064 $\pm$	0.085 $\pm$	0.068 $\pm$	0.17 $\pm$	0.39 $\pm$	0.13 $\pm$	0.13 $\pm$	0.11 $\pm$
Ca	BE	0.051 ^a	0.025 ^a	0.0079 ^a	0.0083	0.040	0.049 ^a	0.13	0.038 ^a	0.018
	DG	0.065 $\pm$	0.049 $\pm$	0.18 $\pm$	0.12 $\pm$	0.30 $\pm$	0.082 $\pm$	0.15 $\pm$	0.10 $\pm$	0.11 $\pm$
	SP	0.023 ^a	0.0088 ^a	0.026 ^b	0.044	0.22	0.097 ^a	0.025	0.024 ^a	0.017
Fe	BE	4.0 $\pm$ 1.2 ^a	3.9 $\pm$ 1.0 ^a	6.1 $\pm$ 4.2 ^a	10 $\pm$ 1.5	8.9 $\pm$ 3.7	4.8 $\pm$ 6.1 ^a	13 $\pm$ 1.5	6.9 $\pm$ 2.0 ^a	13 $\pm$ 5.2
	DG	7.7 $\pm$ 2.5 ^a	7.3 $\pm$ 3.1 ^a	13 $\pm$ 1.6 ^a	12 $\pm$ 3.2	13 $\pm$ 2.0	17 $\pm$ 3.7 ^a	15 $\pm$ 3.2	15 $\pm$ 1.2 ^a	13 $\pm$ 2.0
	SP	3.9 $\pm$ 0.6 ^a	2.9 $\pm$ 0.35 ^a	7.3 $\pm$ 1.1 ^a	6.1 $\pm$ 0.81	8.3 $\pm$ 2.2	10 $\pm$ 1.7 ^b	11 $\pm$ 1.2	8.5 $\pm$ 1.4 ^a	9.3 $\pm$ 1.7

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K	BE	5.4 ± 1.2 ^a	3.0 ± 0.54 ^a	12 ± 8.2 ^a	18 ± 5.1	8.4 ± 1.5 ^a	12 ± 3.2	16 ± 7.5 ^b	26 ± 2.8	15 ± 1.5 ^a	22 ± 4.8
	DG	8.2 ± 1.4 ^a	6.7 ± 0.67 ^a	22 ± 6.8 ^a	27 ± 5.9	18 ± 3.1 ^a	18 ± 4.2	33 ± 3.5 ^b	23 ± 4.3	14 ± 1.4 ^a	19 ± 3.8
	SP	5.2 ± 0.9 ^a	4.6 ± 0.64 ^a	19 ± 2.4 ^a	12 ± 3.4	14 ± 1.1 ^a	12 ± 2.3	25 ± 2.5 ^b	19 ± 6.4	16 ± 3.3 ^a	18 ± 3.1
Mg	BE	1.8 ± 0.32 ^a	2.0 ± 0.3 ^a	3.3 ± 2.2 ^a	5.0 ± 0.43	4.3 ± 1.0 ^a	4.5 ± 1.9	4.7 ± 1.7 ^a	6.1 ± 1.7	4.5 ± 1.0 ^a	4.6 ± 0.83
	DG	4.8 ± 1.5 ^a	2.9 ± 0.39 ^a	7.7 ± 2.1 ^a	8.5 ± 1.5	8.2 ± 0.96 ^a	7.0 ± 1.3	8.6 ± 1.1 ^a	7.2 ± 0.93	6.9 ± 1.0 ^a	5.5 ± 0.96
	SP	2.0 ± 0.48 ^b	1.6 ± 0.29 ^a	4.8 ± 0.33 ^b	3.5 ± 0.5	3.3 ± 0.40 ^a	4.1 ± 0.54	6.7 ± 1.2 ^b	5.8 ± 1	4.0 ± 0.41 ^a	4.3 ± 0.84
Mn	BE	0.12 ± 0.035 ^a	0.12 ± 0.032 ^a	0.15 ± 0.17 ^a	0.32 ± 0.04	0.27 ± 0.085 ^b	0.29 ± 0.13	0.21 ± 0.17 ^a	0.42 ± 0.056	0.34 ± 0.16 ^b	0.48 ± 0.12
	DG	0.32 ± 0.054 ^a	0.27 ± 0.071 ^a	0.73 ± 0.22 ^a	0.71 ± 0.17	0.82 ± 0.22 ^a	0.84 ± 0.26	0.90 ± 0.22 ^a	0.94 ± 0.23	1.2 ± 0.47 ^a	0.87 ± 0.36
	SP	0.16 ± 0.055 ^b	0.076 ± 0.015 ^a	0.29 ± 0.035 ^a	0.2 ± 0.066	0.16 ± 0.016 ^a	0.26 ± 0.053	0.29 ± 0.096 ^a	0.36 ± 0.047	0.26 ± 0.052 ^a	0.30 ± 0.080
Cu	BE	0.0051 ± 0.0053 ±	0.0053 ±	0.0072 ±	0.0076 ±	0.0043 ±	0.011 ±	0.0074 ±	0.0076 ±	0.0056 ±	0.0075 ±
	DG	0.0099	0.0088	0.0035	0.0013	0.00095	0.0035	0.002	0.0019	0.00052	0.0016
	SP	0.0064 ±	0.0067 ±	0.0065 ±	0.0063 ±	0.005 ±	0.0076 ±	0.0061 ±	0.008 ±	0.0087 ±	0.0074 ±
Fe	BE	0.0012	0.0011	0.0011	0.00082	0.00088	0.0015	0.0013	0.001	0.0014	0.0011
	DG	0.0049 ±	0.0036 ±	0.0062 ±	0.0051 ±	0.0071 ±	0.0051 ±	0.0069 ±	0.0061 ±	0.0061 ±	0.0077 ±
	SP	0.00069	0.00042	0.0016	0.00041	0.0013	0.0012	0.00086	0.0013	0.0011	0.0018
Zn	BE	0.29 ± 0.061 ^a	0.31 ± 0.077 ^a	0.47 ± 0.11 ^a	0.51 ± 0.11	0.45 ± 0.095 ^a	0.44 ± 0.14	0.40 ± 0.11 ^a	0.43 ± 0.096	0.39 ± 0.069 ^a	0.54 ± 0.064
	DG	0.30 ± 0.10 ^a	0.50 ± 0.16 ^b	0.49 ± 0.13 ^a	0.68 ± 0.12	0.55 ± 0.14 ^a	0.79 ± 0.24	0.72 ± 0.14 ^a	0.68 ± 0.18	0.70 ± 0.13 ^a	0.64 ± 0.14
	SP	0.24 ± 0.049 ^a	0.49 ± 0.089 ^b	0.53 ± 0.078 ^a	0.50 ± 0.11	0.46 ± 0.075 ^a	0.53 ± 0.094	0.52 ± 0.16 ^a	0.63 ± 0.079	0.93 ± 0.24 ^a	0.68 ± 0.096
Al	BE	0.83 ± 0.24 ^a	1.1 ± 0.35 ^b	2.0 ± 0.75	2.0 ± 1.0	1.7 ± 0.46 ^a	1.8 ± 0.44	1.7 ± 0.43 ^a	2.2 ± 0.33	2.0 ± 0.15 ^b	2.0 ± 0.41
	DG	1.9 ± 1.0 ^a	1.8 ± 0.93 ^a	3.7 ± 0.8 ^a	2.8 ± 1.2	3.4 ± 1.3 ^a	2.7 ± 0.39	4.5 ± 2.1 ^a	4.1 ± 2.2	4.4 ± 2.2 ^a	2.8 ± 0.92
	SP	1.1 ± 0.43 ^a	0.84 ± 0.16 ^a	2.4 ± 0.54 ^a	1.9 ± 0.92	1.9 ± 0.67 ^a	2.7 ± 0.75	3.1 ± 0.59 ^a	2.4 ± 0.84	2.7 ± 0.93 ^a	2.4 ± 0.64
Na	BE	7.7 ± 2.3	5.7 ± 2.1	20 ± 5.3	20 ± 3.3	26 ± 3.9	19 ± 3.9	40 ± 10	40 ± 11	45 ± 6.0	28 ± 5.4
	DG	9.1 ± 2.7	12 ± 1.8	26 ± 5.6	41 ± 8.1	27 ± 3.2	25 ± 2.2	23 ± 4.3	31 ± 2.2	19 ± 3.6	25 ± 4.4
	SP	8.5 ± 0.96	8.6 ± 2.2	22 ± 4.8	26 ± 4.6	19 ± 3.7	18 ± 3.9	35 ± 4.5	31 ± 8.2	23 ± 6.2	26 ± 4.9
	BE	16 ± 2.1	17 ± 1.6	16 ± 3.9	20 ± 3.8	14 ± 0.86	18 ± 4.5	11 ± 0.81	14 ± 1.3	11 ± 0.88	10 ± 0.97
	DG	35 ± 7.9	29 ± 4.5	26 ± 2	22 ± 3.4	21 ± 2.6	17 ± 1.0	16 ± 2.4	17 ± 1.6	16 ± 2.1	13 ± 1.3
	SP	20 ± 1.9	18 ± 3.1	15 ± 2.3	19 ± 0.47	16 ± 1.0	16 ± 1.0	13 ± 1.7	14 ± 2.2	12 ± 1.3	14 ± 1.9

Table S6.7 Results of the generalized linear mixed model analysis showing the relationship between the scaled, observed amounts of collected water and modelled scaled soil moisture indicators in interaction with harvest intensity and tree species. Temporal autocorrelation is captured using a bspline smoother (not shown). Only the results of the conditional model are shown; effects of the dispersion model are not included.

Explaining term	Estimate	S.E.	z	Pr(> z)
Intercept	0.65	0.033	20	< 0.001
Scaled modelled soil moisture	-0.55	0.047	-12	< 0.001
High-thinning	-0.18	0.036	-5.0	< 0.001
Shelterwood	-0.48	0.031	-16	< 0.001
Clearcut	-0.53	0.031	-17	< 0.001
Douglas fir	0.03	0.0094	3.3	< 0.001
Scots pine	0.03	0.0096	2.7	< 0.01
Scaled modelled soil moisture * High-thinning	0.15	0.061	2.4	< 0.01
Scaled modelled soil moisture * Shelterwood	0.52	0.046	11	< 0.001
Scaled modelled soil moisture * Clearcut	0.58	0.046	13	< 0.001
Scaled modelled soil moisture * Douglas fir	-0.001	0.0025	-0.263	n.s.
Scaled modelled soil moisture * Scots pine	-0.000	0.0019	-0.005	n.s.

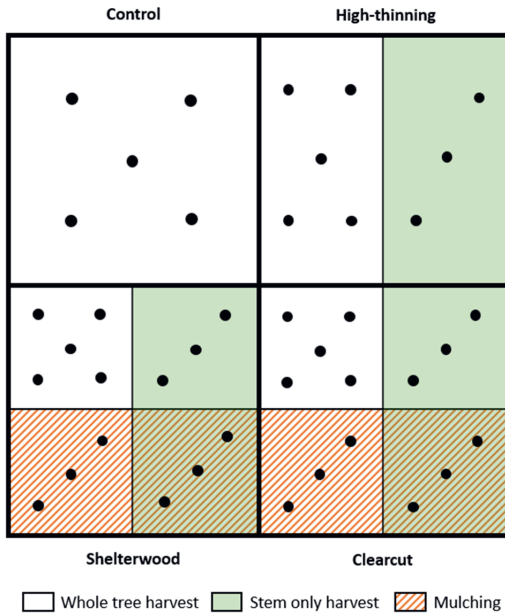
Supplementary figures

Figure S6.1 Example of an experimental site. Each site was divided into four equal subplots for the harvest intensities high-thinning, shelterwood and clearcut and the unharvested control (outlined in blue). The high-thinning was divided into two equal subplots for whole tree harvest and stem only harvest respectively while in the shelterwood and clearcut plots half of the whole tree harvest and stem only harvest plots was mulched. The position of the macrorhizons is indicated with the black dots which were placed in a cross design for the control and the whole tree harvest plots and in a linear line for the other plots.

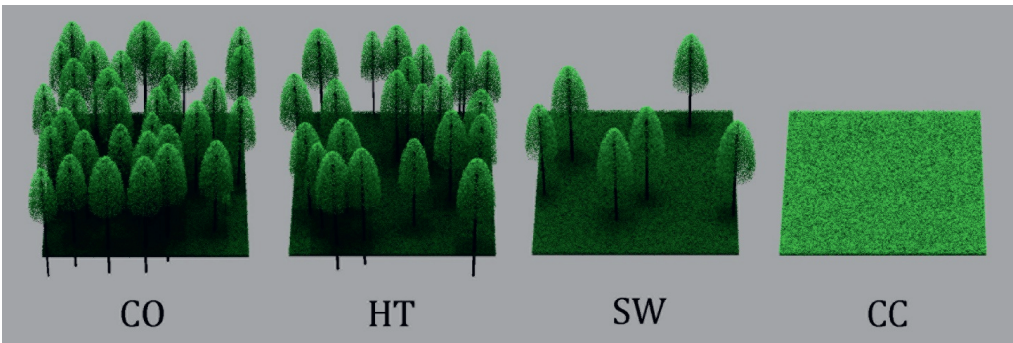


Figure S6.2 Virtual replicate of the stand properties of one of the study sites as simulated with the model used to calculate drainage. The tree number and DBH is based on measurements, the trees are placed at random locations since location data was not available.

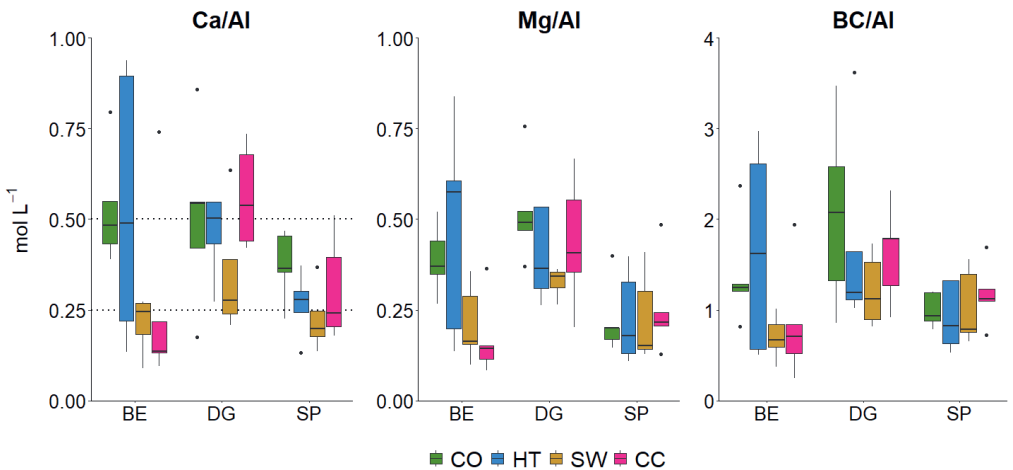


Figure S6.3 The molar ratio of Ca/Al and of Mg/Al in control (CO), high-thinning (HT), shelterwood (SW) and clearcut (CC) harvest intensities in beech (BE), Douglas fir (DG) and Scots pine (SP). The molar ratio is calculated based on annual mean concentrations of Ca, Mg and Al in the soil moisture.

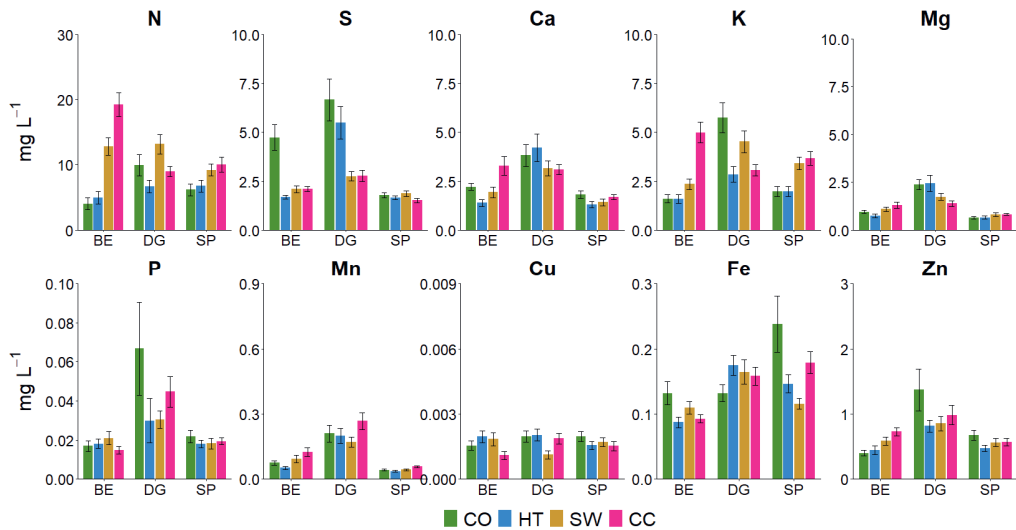


Figure S6.4 Mean monthly dissolved concentrations of macro- (N, S, P, Ca, K and Mg) and micronutrients (Mn, Cu, Fe and Zn) in beech (BE), Douglas fir (DG) and Scots pine (SP) as a function of the harvest intensity (CO: control, HT: high-thinning, SW: Shelterwood and CC: Clearcut). For the high-thinning, shelterwood and clearcut the data of the non-mulched whole tree harvest plots are shown in this figure.

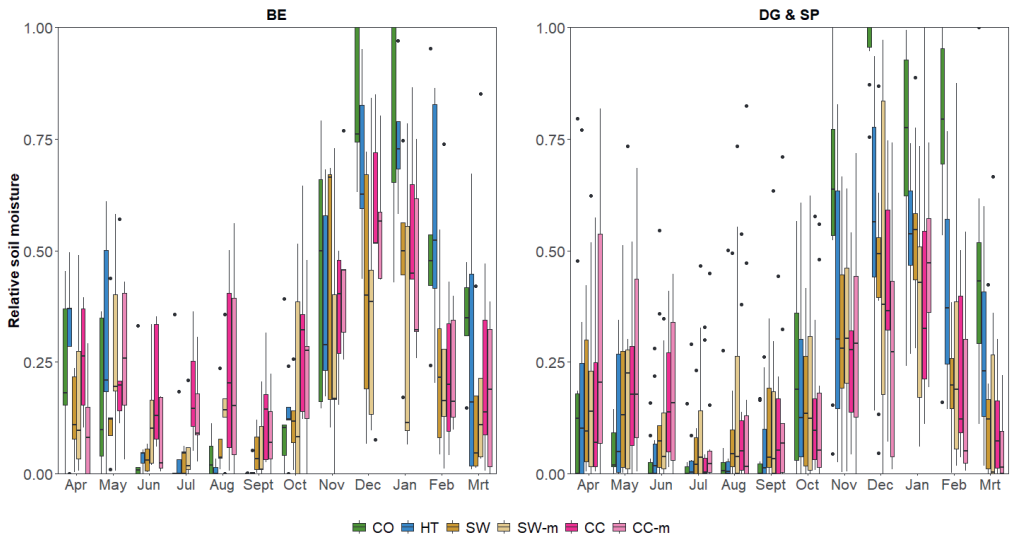


Figure S6.5 Relative soil moisture (0-1) collected within the harvest intensity treatments high-thinning (HT), shelterwood (SW) and clearcut (CC) and in the unharvested control (CO). For the SW and CC treatment the relative soil moisture for the mulching soil preparation is included (indicated by the addition of -m) as soil moisture was significantly influenced by the mulching treatment.

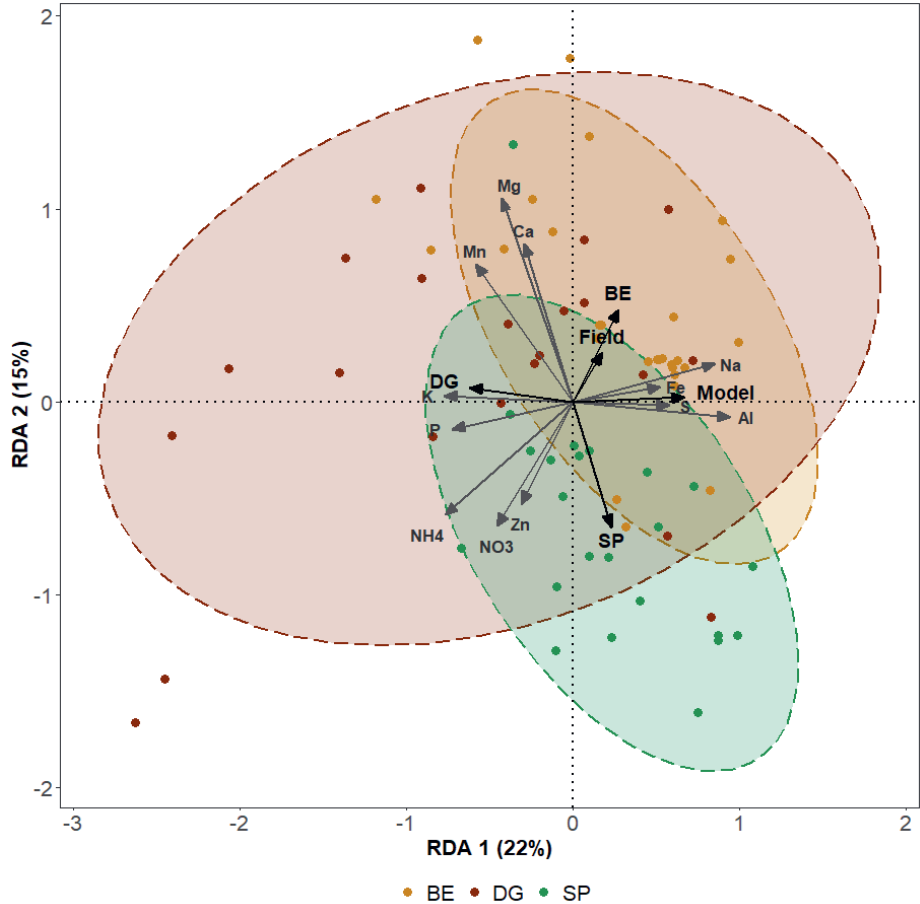


Figure S6.6 Explanation of the monthly concentrations of macro- (NH₄, NO₃, S, P, Ca, K and Mg) and micronutrients (Mn, Cu, Fe and Zn) in soil moisture of unharvested stands of beech (BE), Douglas fir (DG) and Scots pine (SP). The terms Field and Model relate to the soil moisture proxy measured in the field and to the modelled soil moisture. Effects of the months is for visual reasons not included in this figure. The concentrations of the nutrients in the soil moisture are represented by the grey arrows, the effects of species, canopy openness treatments, seasons and soil moisture by black arrows. The length of arrows denotes the variation explained by respectively the nutrients, treatments, seasons, species and soil moisture.



Chapter 7

General discussion

7.1 Introduction

The construction of accurate, data-driven forest nutrient budgets using large-scale field experiments is crucial for the understanding of nutrient dynamics on low fertility soils, both for mature, unharvested stands and for the post-harvest dynamics in harvested stands (Augusto et al., 2002; Clarke et al., 2015; Pare and Thiffault, 2016; Titus et al., 2021). Additionally, accurate measurements of the nutrient fluxes to and from the forest system, deposition, weathering, uptake, and leaching, enhance the reliability of forest nutrient budgets as decision-making tools. This thesis aims to establish such accurate forest nutrient budgets to contribute to the establishment of science-based guidelines for managing production forests on sandy soils facing high acidifying deposition, which constitute a significant portion of European forests. In this thesis I quantified forest nutrient fluxes required for assessing an accurate forest nutrient budget for Dutch forest in the Netherlands. These forests can be seen as an extreme case of production forest on low fertility soils receiving high N deposition inputs. The studied soil types are acid sandy soils with pH values below 4.5 and base saturation values below 10%. These soil types are prevalent in up to 40% of the plots within the European-wide monitoring network ICP (The International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests) (Fleck et al., 2016). Forests on comparable soils, with similar substrates which are moderately to strongly acidified, represent 31-58% of the forests in the Czech Republic (Šantrůčková et al., 2019), as well as substantial forest areas in countries such as Germany (e.g., the Black Forest, Bavarian Forest, Ore Mountains, Harz Mountains, and North German lowlands) (Meesenburg et al., 2019), Belgium (De Schrijver et al., 2006), and Poland (Mill, 2006).

In this chapter, I synthesize the main findings of my thesis by constructing forest nutrient budgets based on detailed measurements of the atmospheric nutrient deposition (**Ch3**), nutrient uptake (**Ch5**) and nutrient leaching (**Ch6**) and by adding the chemical weathering derived by literature review (**Ch7.2**). These balances are then compared to the measured nutrient stocks (**Ch5**). The effects of species and management on these fluxes is summarized in box 7.1.

With this synthesis I aim to answer the fourth research question: *"What is the effect of harvesting practices on the nutrient balance shortly after harvest and what are the possible implications over a full rotation period."* I first discuss the nutrient budgets of mature stands (**Ch7.3**) and, after that, the post-harvest nutrient budgets of stands harvested at different intensities (**Ch7.4**), using various harvest methods and post-harvest soil preparations (**Ch7.5**). Negative nutrient budgets are compared to the nutrient stocks in the organic soil layers as these are particularly important for long-term site nutrition for forest on acidic soils (Prietzl and Stetter, 2010; Garrett et al., 2021). Subsequently, I propose general assumptions about the period required for post-harvest nutrient fluxes to return to pre-harvest levels following literature review (**Ch7.6**). These assumptions guide the construction of forest nutrient balances over the entire rotation period, aiming to evaluate the time needed to restore the soil nutrient pool. The

implications of these nutrient budgets are discussed in the context of sustainable forest management of forests on poor sandy soils (**Ch7.7**). Finally, I present conclusions and define the main research lines required for improving forest management practice under high anthropogenic pressure (**Ch7.8**).

1. How is the annual nutrient deposition input modified by forest structure, especially canopy openness (as driven by tree species and harvest intensity), for different nutrients?

Annual nutrient deposition, measured using the Ion Exchange Resin method (**Ch2**), varied significantly across tree species, being highest in Douglas fir and lowest in Scots pine stands. Unharvested forests showed deposition estimates surpassing national estimates by 30-140% for NH_4 and S and by 68-750% for the base cations (Ca, Mg and K), implying a lower net acidic input than that based on nationwide models. Effects of harvest intensity on the interception of total deposition was stronger than the effects of tree species, as a clearcut resulted in a 1.2-4.1-fold decrease in total deposition. Our findings emphasize the crucial role of considering harvest intensity effects on forest structure and, to a lesser extent, tree species when assessing nutrient inputs via atmospheric deposition. Additionally, regular thinnings, particularly in beech and Douglas fir, emerged as potential tools to slow down soil acidification (**Ch3**).

2. What are the present nutrient stocks in the forest and what is the total nutrient export following different harvest intensities and harvesting methods for different species?

Beech forests have highest aboveground nutrient stocks while Douglas fir forests have the highest biomass stocks. Nutrient stocks of the organic layer surpass aboveground stocks, except for base cations and Mn, indicating potential long-term threats to forest nutrition if trees are harvested. Thinning results in relatively low nutrient exports, but frequent low-intensity forest management reduces potential advantages for nutrient balances by increasing the nutrient export in harvested wood. Compared to stem-only harvest (SOH), whole-tree harvest (WTH) increases nutrient export by 66% to 100% and poses potential threats to sustainable biomass harvest due to unrecoverable nutrients. Wood-only harvest (WOH), in which the bark is removed in the field, decreases nutrient export by 23% to 41%, emerging as a powerful tool to retain nutrients within the forest (**Ch5**).

3. How is the annual nutrient leaching in forest soils modified by forest structure (as driven by species and harvest intensity), harvest method and soil preparation for different nutrients?

Annual leaching is generally highest in Douglas fir with species effects fading out moving from control to clearcut. Clearcut, and to a lesser extent shelterwood, increased water fluxes and dissolved nutrient concentrations, especially NO_3 , indicating a rapid mobilization of large N stocks associated with accelerated losses of the acid cations Al, Fe and Mn and to a lesser extent the base cations Ca, Mg and K. Thinning had relatively small effects on leaching, the effects of harvest methods on leaching appeared to be marginal and impacts of mulching were negligible. Our results highlight that forest structure, mainly impacted by harvest intensity and, to a lesser extent, tree species, have large impacts on nutrient losses by leaching (**Ch6**).

7.2 Weathering

The mineral soil's chemical weathering was not measured in this thesis. The focus was on external inputs (deposition) and outputs (uptake and leaching), causing changes in soil pools. However, understanding weathering is crucial to assess the extent to which the net soil release of (base) cations, assuming higher outputs than inputs, is buffered by the release of primary minerals, thereby affecting soil buffer capacity.

For the base cations Ca, Mg and K, weathering can be a major flux of the nutrient budget possibly equalling the losses of those nutrients through harvest (Klaminder et al., 2011). However, despite the overall importance of the weathering in the nutrient budgets there is no common agreement on the most appropriate method to estimate weathering. Commonly used methods include (1) historical weathering based on elemental depletion in soil profiles using e.g. zirconium (Olsson and Melkerud, 1991; Olsson and Melkerud, 2000); (2) current rates from input-output budgets (Drahota et al., 2006; Simonsson et al., 2015); (3) strontium isotope ratios (Åberg, 1995; Perakis et al., 2006); (4) modelling using process-based weathering models like PROFILE (Sverdrup and Warfvinge, 1993; Akselsson et al., 2006) and (5) laboratory experimental methods (de Vries, 1994; Bain and Langan, 1995; van der Salm et al., 1998). Often, large differences in the estimation of the weathering rates between these methods are found with differences up to a factor of 20 (Bain and Langan, 1995; Kolka et al., 1996; Whitfield et al., 2006; Casetou-Gustafson et al., 2020). Because of the huge variability in weathering estimates between these methods I used literature values for the weathering estimation as application of either one of these commonly used methods will not significantly reduce the uncertainty of the nutrient balance, which was the main aim of this thesis.

The weathering estimates of the base cations (Ca, K and Mg) are based on de Vries et al. (2021), weathering estimates of P on a literature review of Newman (1995) while weathering estimates of Mn, Cu, Fe, Zn and Al were calculated using a scaling approach based on base cation weathering (De Vries and Bakker, 1996; Vrabel and Paces, 1996). The base cation weathering was determined using laboratory experiments of van der Salm et al. (1998) and de Vries (1994), classified by pH and texture class in Van der Salm et al. (1999) and slightly adapted following a literature review in de Vries et al. (2021). These weathering estimates generally correspond well with literature in which the cumulative weathering of the base cations in a 1 m deep soil profile varies roughly between 0.6 to 11 kg ha⁻¹ yr⁻¹ (Hyman et al., 1998; Klaminder et al., 2011; Yang et al., 2013; Starr et al., 2014; Johnson et al., 2015). Based on the soil texture, I selected the weathering estimates of the base cation-poor sandy soils (low silt fraction) and the base cation-rich sandy soils (higher silt fraction) (Hyman et al., 1998; Yang et al., 2013). The weathering estimates of the poor sandy soils were thereafter appointed to the soil layer with lowest silt fraction and the estimates of the rich sandy soil to the soil layer with highest silt fraction, thereby ignoring possible effects of tree species on the weathering rate (Table S1.2). The base cation weathering estimates of all

other soil layers were interpolated based on the silt fraction and corrected for a mineral soil column of 50 cm depth assuming, based on the tree root distribution (**Ch5**), assuming that all elements lower than 50 cm depth are leached (**Ch6**). This assumption, however, is not always valid as tree uptake from > 50 cm depth mineral soil is not entirely absent (**Ch5**). For P, Newman (1995) provided a range of 0.04 – 0.2 kg P ha⁻¹ yr⁻¹ for Europe from which, in line with de Vries et al. (2021), I selected an average weathering estimate of 0.1 kg P ha⁻¹ yr⁻¹. The weathering of Mn, Cu, Zn and Al were calculated using the molar ratio of the total metal content to the base cation content in the parent material according to (De Vries and Bakker, 1996; Vruble and Paces, 1996) :

$$M_{we} = 5.10^{-5} * BC_{we} * \frac{ctM_p}{ctBC_p}$$

Here, the weathering rate of the heavy metal M (M_{we} in mg m⁻² yr⁻¹) is a function of the weathering rate of base cations (BC_{we} in mol_c ha⁻¹ yr⁻¹) times the ratio of the total heavy metal content (ctM_p in mg kg⁻¹) to the total base cation content in the parent material ($ctBC_p$ in mol_c kg⁻¹). Fe weathering could not be determined as a result of sesquioxide (iron)-coated sand grains, causing a large overestimation of the weathering rate due to dissolution of secondary sesquioxides during analysis. The total content of base cations and heavy metals in the soil is derived from total analysis of the soil layer at 40-60 cm depth (Table S1.3). The weathering rates per study site that are used in the post-harvest nutrient balance (Fig. 7.1) are given in Table 7.1.

Table 7.1 Calculated weathering rates (kg ha⁻¹ yr⁻¹) for the different study sites. The mineral composition of the soil is given in Table S1.1-S1.3 in **Ch1**.

Site	Species	Ca	Mg	K	Na	Mn	Cu	Zn
1	BE	0.89	0.93	1.1	0.60	0.046	0.0021	0.014
2	BE	0.95	1.1	1.1	0.66	0.073	0.0027	0.027
3	BE	1.0	1.4	1.2	0.75	0.10	0.0027	0.027
4	BE	0.89	0.93	1.1	0.60	0.052	0.0053	0.019
5	BE	0.85	0.80	1.1	0.56	0.031	0.0050	0.025
1	DG	0.83	0.72	1.0	0.54	0.035	0.0052	0.016
2	DG	0.98	1.2	1.2	0.69	0.15	0.0023	0.027
3	DG	0.88	0.90	1.1	0.59	0.13	0.0026	0.018
4	DG	0.91	1.0	1.1	0.62	0.044	0.0044	0.018
5	DG	0.86	0.81	1.1	0.56	0.055	0.0025	0.047
1	SP	0.87	0.87	1.1	0.58	0.050	0.0047	0.014
2	SP	0.98	1.2	1.2	0.68	0.086	0.0022	0.020
3	SP	0.77	0.53	1.0	0.48	0.032	0.00089	0.0062
4	SP	0.92	1.0	1.1	0.63	0.031	0.0040	0.012
5	SP	0.82	0.71	1.0	0.53	0.027	0.0020	0.022

The size of the weathering flux, however, is often prone to large uncertainties. For example, the uncertainties are often comparable or larger than the mean estimated weathering rates which can hamper the use of nutrient balances for sustainable forest management (Klaminder et al., 2011; Simonsson et al.,

2015). Especially, since weathering is influenced by changes in climate (Crawford et al., 2019). Furthermore, harvest itself can further increase the uncertainty related to the weathering estimates as weathering not only depends on the mineralogy of the soil but also on availability of moisture (Dixon et al., 2016; Belyazid et al., 2022), temperature (Olsson and Melkerud, 1991; Bhatti et al., 2000; Clarke et al., 2015; Houle et al., 2020), and input of strong acids from anthropogenic sources (Van der Salm et al., 1999; Guo et al., 2015). Some of these properties, like soil moisture and temperature, even differ between SOH and WTH (Ballard, 2000; Achat et al., 2015) indicating that this harvest method might result in temporary different weathering. Weathering is even argued to increase following forest regeneration (Vadeboncoeur et al., 2014), as biological demand can influence the size of the weathering flux (Van Schöll et al., 2006; Pare and Thiffault, 2016), indicating that weathering is a dynamic process influenced by forest management via various mechanisms.

7.3 Tree species impact on nutrient balances in unharvested stands

Tree species play a crucial role in the nutrient balance of unharvested stands as tree species differ in the capacity to intercept deposition, in nutrient uptake and in nutrient leaching (Fig. 7.1) (**Ch3, Ch5, Ch6**). In unharvested stands, both nutrient deposition and nutrient leaching are highest in Douglas fir. Atmospheric deposition is the major input of all nutrients, as weathering plays a relatively minor role in our forests. Tree uptake plus leaching are generally comparable to deposition, therefore, the annual balance of mature, unharvested Douglas fir stands is neutral to positive for almost all nutrients, except for P, Mn, and Fe. The Mn and Fe balances are negative across species. (Fig. 7.2). For P there are large uncertainties related to the balance across species (Fig. 7.2), which can partly be caused by the methodology (**Ch2, Ch3**). Despite some uncertainty, there is no indication of a decline of the organic layer P stock in Douglas fir as this stock is slightly higher compared to beech and Scots pine (**Ch. 5**). Beech generally had the highest nutrient uptake in unharvested forest stands which was especially pronounced for the base cations and Mn (**Ch5**). These high nutrient uptake rates are not compensated by higher deposition or by a reduction in leaching which results in negative nutrient balances in beech (Fig. 7.1, Fig. 7.2). The nutrient uptake could be, to some extent, altered by internal translocation, which was implicitly included in the nutrient uptake estimates (**Ch5**). Although deep layer uptake could provide another nutrient influx for beech, this influx is expected to have only a limited effect on tree nutrition (Berger et al., 2006; van der Heijden et al., 2015). The negative base cation balances, only observed in beech stands, point to imbalanced tree nutrition and related decline in forest vitality, which could affect growth (**Ch5**). Finally, Scots pine stands generally have lowest total deposition and showed a tendency of the lowest leaching of the base cations and Mn compared to beech and Douglas fir while showing rather comparable nutrient uptake rates compared to Douglas fir. The combination of these generally low fluxes in Scots pine compared to beech and Douglas fir overall

results in neutral to positive nutrient budgets except for Mn, Fe and Zn which are consistently negative across the species.

The negative balances of Mn and Zn observed in all species indicate substantial losses, posing a potential threat to forest nutrition within a rotation period. The significant annual losses of Mn and Zn in beech forests (respectively 23% and 28% of the stock in the organic layers), suggest a potential risk of limitation for beech which is primarily caused by high nutrient uptake. Losses of Zn were substantial for Scots pine (21% of the stock in the organic layer), while Mn losses in Douglas fir and Scots pine are comparatively smaller (respectively 3.5% and 4.9% of the organic layer stock) (Fig. 7.1, Fig. 7.2). Despite substantial losses, tree nutrition showed no deficiency; although foliar Mn concentrations are lower, they remain above the limitation threshold (Bergmann and Wrazidlo, 1976)(**Ch4**). There is limited evidence for negative Mn and Zn budgets and declining soil pools in temperate forests (Falkengren-Grerup et al., 1987; Bergkvist et al., 1989; Navrátil et al., 2007; Gandois et al., 2010a; Swathi A et al., 2013). However, since soil acidification triggers Mn and Zn mobilization thereby increasing risks of leaching (Bergkvist et al., 1989; Watmough et al., 2005; Navrátil et al., 2007; Watmough et al., 2007), the negative balances in these high N deposition regions are to be expected. Negative Mn and Zn budgets are concerning as Mn limitation could lead to foliage mortality, reduced photosynthetic rates and tree growth, reduced biomass allocation to roots and even tree death (De Ronde et al., 1988; Göransson, 1994; Morales et al., 2018) and Zn limitation could lead to malformed trunks, dieback or limit regeneration (Boardman and McGuire, 1990; von Arnold et al., 2011). Nutrient limitation, particularly Mn deficiency, can also cause tree dehydration by affecting transpiration, water use efficiency, and root exploration (Hajiboland, 2012). This increased sensitivity to drought adds to existing concerns in these systems, where drought already poses a significant threat to growth and mortality (Sterck et al., 2021). Interestingly, Mn limitation may also limit soil acidification due to the Mn-peroxidase enzyme restriction which is involved in the breakdown of lignin (Roth et al., 2022). This constrains N availability through organic matter decomposition, potentially contributing to the observed high (**Ch5**) and increasing organic layer nitrogen stocks in Dutch stands (de Jong et al., 2023), and thus reducing nitrate leaching and related acidification. The implications of these negative Mn and Zn balances, however, remain unclear as there still are large total pools (Table S1.3) which can sustain forest nutrition over longer term as argued for the base cations (Rosenstock et al., 2019).

In contrast to Mn and Zn, the negative balance of Fe results in minor impacts on total nutrient stocks, with annual losses being less than 1% of the organic layer Fe stock. The negative Fe balance suggests the onset of the Fe buffering stage as a response to long-term N and S deposition while the Al buffering may be slowing down (**Ch6**). This aligns with soil pH nearing 4, signifying the initiation of the Fe buffering stage (**Ch1**)(Bowman et al., 2008). There is a noted increase in Fe losses across various northern

regions (Sarkkola et al., 2013; Björnerås et al., 2017). While this rise isn't definitively linked to soil acidification, it is associated with areas covered by coniferous trees (Škerlep et al., 2022), which are known to amplify soil acidification by more efficient interception of N and S (acid deposition), and by their humus quality and litter decomposition rate (Augusto et al., 2002; Cremer and Prietzel, 2017) (**Ch3**). Overall, elevated Fe concentrations and leaching is concerning given they can be associated with decreased forest growth (Elias et al., 2009).

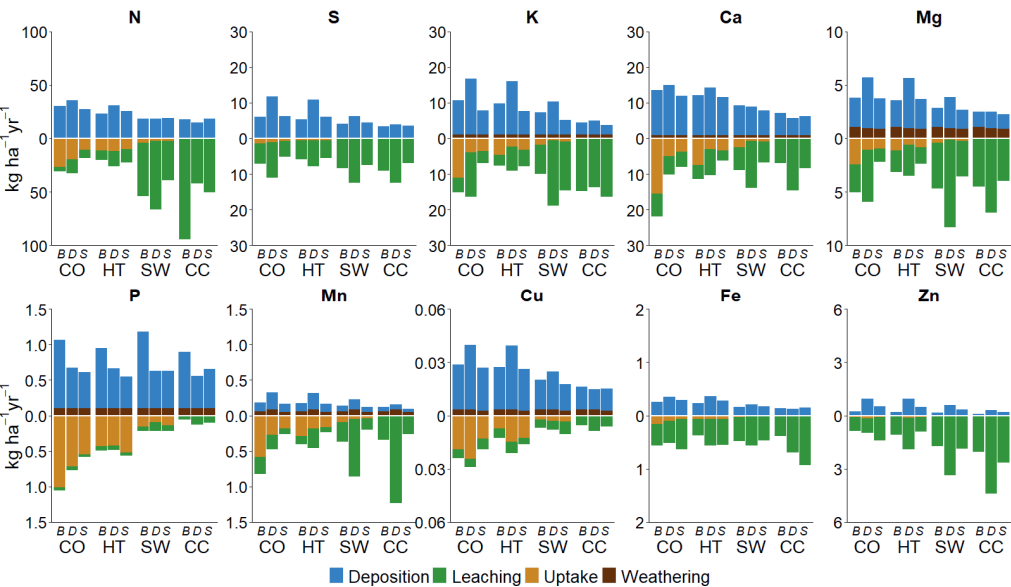


Figure 7.1 Nutrient balance for the unharvested control (CO) and the harvest intensities high-thinning (HT), shelterwood (SW) and clearcut (CC) for beech (*B*), Douglas fir (*D*) and Scots pine (*S*) using WTH for the second year after harvest. The WTH scenario was chosen to facilitate the comparison of the leaching between different harvest intensities. The input fluxes (deposition and weathering, not shown for Fe) are shown above the zero line, the output fluxes (annual nutrient uptake and leaching) are shown below the zero line.

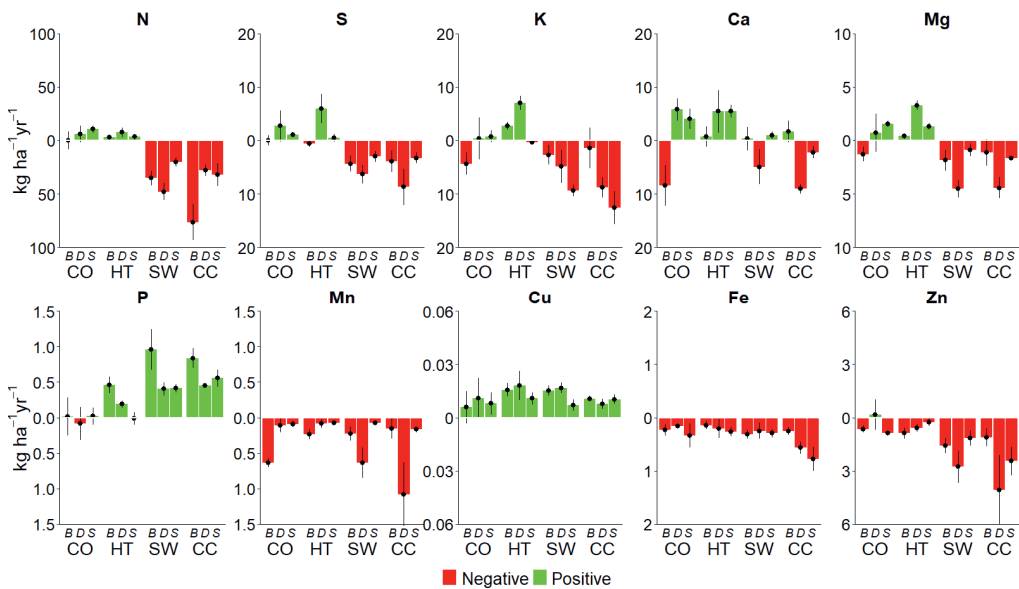


Figure 7.2 Final nutrient balance showing higher nutrient inputs compared to outputs (positive) and lower nutrient inputs compared to outputs (negative) for the unharvested control (CO) and the harvest intensities high-thinning (HT), shelterwood (SW) and clearcut (CC) for beech (B), Douglas fir (D) and Scots pine (S) using WTH for the second year after harvest. Overview of the nutrient inputs and outputs are in figure 7.2.

7.4 Effects of harvesting intensity on nutrient budgets

The harvest intensity largely influences all fluxes of the nutrient budgets except the weathering for which no effect of harvest intensity was assumed (**Ch7.2**). Except for P, atmospheric deposition was reduced with increasing harvest intensity as a result of increasing canopy openness towards clearcuts causing a general decrease of 2.2 times towards the clearcut with absolute reductions (range 1.2 – 4.1 times) differing between species and nutrients (**Ch3**). The decrease in atmospheric deposition induced by higher harvest intensities largely impacts the nutrient balance especially since leaching increased with increasing harvest intensity, particularly for N and base cations (Fig. 7.1). With increasing tree harvest intensity, tree uptake is directly reduced by the selected harvested tree volume. Despite such generic qualitative trends, there were remarkable quantitative differences between the species. When tree cover becomes smaller (from control to clearcut), the nutrient uptake in beech is reduced more than the deposition, suggesting an overall increase in post-harvest nutrient stocks. For Douglas fir and Scots pine, the opposite was observed, indicating an overall decrease in post-harvest nutrient stocks. Lastly, the leaching of most nutrients strongly increases towards clearcuts leading to nutrient losses exceeding the total losses of unharvested stands. When considering whole tree harvest, thus without the nutrient input from

decomposing harvest residues, nutrient losses from the clearcut exceeded the nutrient losses of the combined uptake and leaching for N, S and Zn in control stands across the three species. For Ca, Mg, Fe and Zn, the larger losses in the clearcut compared to the control were observed in both Douglas fir and Scots pine while for K this was only observed in Scots pine. The larger losses in the clearcut compared to the control indicates a (large) mobilization of these elements from the soil nutrient stocks in response to increasing harvest intensities.

In the clearcut, the substantial nutrient mobilization, particularly for N, led to a shift in the post-harvest nutrient balance. N balances changed from positive in the control to losses of approximately 30 kg ha⁻¹ yr⁻¹ in Douglas fir and Scots pine, escalating to 80 kg ha⁻¹ yr⁻¹ in beech (Fig. 7.2). These losses translated to a 3% reduction in the organic layer nutrient stock for Douglas fir and Scots pine and a 7% loss for beech. Despite the relatively modest N losses, when combined with increased S losses, they result in a net acid output of 5.7 keq ha⁻¹ in beech and 2.5 keq ha⁻¹ in Douglas fir and Scots pine clearcuts, thereby increasing soil acidification. An increase in post-harvest soil acidification has been demonstrated before, resulting in a pH drop and a decline of the base saturation pools (Roy et al., 2021). However, the extent of this acid output was higher than expected, possibly due to the high forest N stocks. No effect of harvest on soil acidification was observed in forests not impacted by anthropogenic soil acidification (Grand et al., 2014), emphasizing the importance of considering existing nutrient stocks in predicting post-harvest effects.

The N mineralization and S mobilization are associated with the mobilization and loss of base cations (K, Ca, and Mg), Mn, Fe (mostly for the clearcuts), and Zn. Generally, the highest losses of base cations are observed for clearcuts of Douglas fir stands, with approximately 25% of the acid output buffered by base cation losses, compared to 18% in Scots pine and only 1% for beech stands. These differences in net acid output buffered by base cations across species are surprising given that the forest floor nutrient stocks are rather comparable (**Ch5**). This suggests that beech is better able to conserve the base cations in the system because leaching hardly exceeds deposition and, without uptake by remaining trees, results in only small negative to positive nutrient balances in the clearcuts. For Douglas fir, a clearcut, and to a lesser extent a shelterwood harvest, results in nutrient losses by leaching which exceed nutrient influx by deposition and therefore cause absolute losses of base cations that are larger than for beech and pine. Remarkably, the losses of Ca and Mg in Douglas fir are bigger than the total loss (uptake and leaching) in unharvested stands, indicating a rapid mobilization. This mobilization was, besides the cations, also observed for Mn and Zn (Fig. 7.1) for which the same responses to harvest were described before (Olsson et al., 1996). For Scots pine, the highest mobilization is observed for K. Studies have emphasized the sensitivity of K to depletion by any harvesting intensity, particularly in conifers, as harvesting leads to substantial K losses (Olsson et al., 1996; Katzensteiner, 2003; Duchesne and Houle, 2006). Post-harvest

base cation losses, varying with harvest intensity among species, don't directly imply that harvesting Douglas fir (with the highest losses) leads to the largest nutrient exports, as these dynamics don't incorporate the absolute amount of nutrients exported in harvested wood products (**Ch5**). In conclusion, the dynamics of nutrient mobilization and losses in harvested stands reveal species-specific variations. Beech shows better conservation of cations in the clearcut but not in the control, while Douglas fir and to a lesser extent Scots pine, experiences substantial losses following clearcuts but not in control forests.

In summary, my findings indicate a potentially strong negative impact of high-intensity harvesting, particularly in shelterwood and clearcut scenarios. Interestingly, such adverse effects were minimal in high thinning, even with approximately 20% biomass harvested. This suggests that regular high thinning, applied every 4-8 years, results in positive or neutral post-harvest nutrient balances for most elements, except Mn, Fe, and Zn. Importantly, regular thinnings do not contribute to accelerated acidification, as there is no mobilization of N and S from the soil stocks. The losses of Mn, Fe, and Zn in high thinning are comparable to those in closed forests indicating no direct added effect on nutrient limitation. Therefore, based on the post-harvest nutrient balance, harvesting in continuous cover systems emerges as a promising approach to sustain the forest nutrient balances.

7.5 Effects of harvest method and soil preparation on nutrient budgets

Harvest methods can alter forest nutrient budgets through nutrient export in harvested wood products and by influencing leaching, while soil preparation has a potential influence on nutrient leaching only. I demonstrated that whole tree harvest increases average nutrient exports by 87% in beech, 66% in Douglas fir, and 100% in Scots pine relative to stem only harvest (with the crown left in the forest) (**Ch5**). Since stem-only harvest results in these nutrients being added to the forest floor as harvest debris, I expected higher leaching compared to the whole tree harvest treatment, particularly in the clearcut with the highest debris accumulation and no tree uptake. However, measurements show that, except for K, effects on leaching were minor (S, Ca, Mg, Mn, Zn), not consistent (P, Fe) or even absent (N, Cu). The minor effects on leaching hardly influence the postharvest nutrient budget for S, Ca, Mn, and Zn as no general pattern can be distinguished (Fig. 7.3). Minor effects on leaching of most nutrients can be attributed to low mobility, strong retention in the system or differences in release rates from decomposing harvest debris, causing elevated leaching patterns over longer time periods than measured in this thesis. Strong retention can be observed, for instance, in the case of P, as P stocks in harvest debris decreased by 49% three years after harvest (Palviainen et al., 2004a), which was not reflected in our leaching estimates. Similarly, Ca showed accumulation in branches (Palviainen et al., 2004b) explaining the absence of elevated Ca leaching following SOH.

The nutrient budget for K is more negative following SOH solely due to an increase in leaching. Accelerated K losses following SOH are consistent across species for shelterwood and clearcut, with effects most pronounced for Douglas fir. In a clearcut, the K stock in the harvest debris (crown material left in the forest) equals 71 kg ha⁻¹ in beech, and 42 and 45 kg ha⁻¹ in Douglas fir and Scots pine. Post-harvest leaching following SOH results in an additional loss of 6.7, 19.2, and 8.0 kg ha⁻¹ yr⁻¹ for beech, Douglas fir, and Scots pine, respectively. Based on one year of measurements it is unclear if SOH leads to higher K stocks in the forest floor as the duration of the elevated post-harvest leaching and the course of this post-harvest leaching determine the total K losses. Both the duration and the course of this post-harvest leaching is expected to be related to the settlement of the regeneration (**Ch7.6**). As earlier studies report elevated K losses up to 27 years after harvest (Martin et al., 2000; Webster et al., 2022) it might be that total K losses in both export and leaching following SOH equals the total losses following WTH. Elevated K losses can be related to the high mobility and release rate, causing almost all K to be released from logging residues in the first year after harvest (Palviainen et al., 2004b; Chen et al., 2023) indicating a large flux available for leaching. Another nutrient that showed increased leaching following SOH, though not consistently across species, is Mg. Also Mg is known to be released more quickly from harvest debris compared to, for example Ca (Osono and Takeda, 2004). The budget shows a more negative trend following a SOH in shelterwood and clearcut in Scots pine and after a SOH clearcut in Douglas fir (Fig. 7.3). In Scots pine, post-harvest Mg losses through leaching magnify the negative budget by 2 to 3 times. In the second-year post-harvest, the additional Mg losses after SOH make up 18% to 29% of the Mg stock in the harvest debris for shelterwood and clearcut, respectively. The elevated leaching losses following SOH indicate that the substantial difference in nutrient export between SOH and WTH is partly offset by elevated leaching following SOH. Therefore, it is important to include elevated leaching losses when evaluating SOH harvest scenario's in nutrient balances.

Soil preparation was expected to alter the forest nutrient budget by influencing decomposition, leading to a larger mobilization of nutrients and consequently higher dissolved nutrient concentrations and nutrient leaching (Lundmark-Thelin and Johansson, 1997; Piirainen et al., 2007). However, mulching did not result in an alteration of the dissolved nutrient concentrations nor leaching, indicating that mulching has no influence on the nutrient budget in the second year after harvest (**Ch6**). As previously indicated, stronger mulching effects directly after harvest can currently not be ruled out as mulching effects on leaching have hardly been studied before, but the results still imply that the effects of mulching will be limited to a small-time window, thereby being definitely less important for whole rotation nutrient budgets.

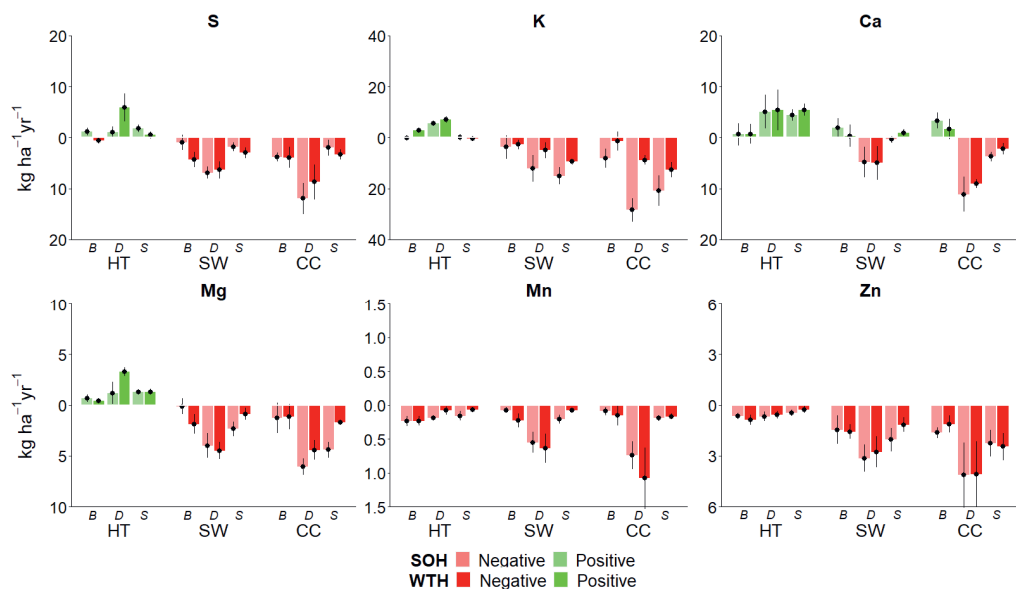


Figure 7.3 Final nutrient balance showing higher nutrient inputs compared to outputs (positive) and lower nutrient inputs compared to outputs (negative) for the high-thinning (HT), shelterwood (SW) and clearcut (CC) for beech (B), Douglas fir (D) and Scots pine (S) using SOH and WTH for the second year after harvest. As deposition, nutrient uptake and weathering are independent of the harvest method, only the balances are shown of the nutrients of which the leaching was influenced by the harvest method (**Ch6**).

7.6 Duration of post-harvest effects

The temporal changes in magnitude of post-harvest effects are crucial for determining the impact of harvest (intensity) effects on nutrient budgets throughout the entire rotation period. Various mechanisms regulate the different nutrient fluxes (deposition, leaching and uptake), influencing the time these fluxes require to return to pre-harvest levels. Here, I outline how such mechanisms may influence the dynamics of these fluxes during an entire rotation and, from this, I estimate the number of years needed for the deposition, uptake, and leaching fluxes to revert to pre-harvest levels in a high-thinning, shelterwood and clearcut system. My data on post-harvest fluxes (**Ch7.4**) and these hypothesized years to pre-harvest flux levels are the base for evaluating possible long-term effects of harvest (intensity) on nutrient budgets over rotations of 80 years (**Ch7.7**).

Deposition

Recovery of nutrient deposition to pre-harvest levels following a thinning is assumed to happen fast as the main forest structure is preserved (**Ch4**) and recovery of the deposition is mainly related the recovery of the canopy, especially in terms of the LAI (Granier et al., 2008). Depending on the thinning intensity, LAI

can be restored within 1 to 2 years in young forest stands (Misson et al., 2005; Granier et al., 2008) while no LAI recovery was observed two years post-thinning in forest stands with mature trees (Davi et al., 2008). This can be related to the increase in recovery time of the canopy as trees age (Den Ouden et al., 2010). In this context, I assume that canopy cover and deposition will return to pre-thinning levels within six years. This assumption is based on the average thinning interval (between 4-8 years) applied in these forests (Den Ouden et al., 2010), overlooking potential age-related effects on LAI recovery.

Recovery following a shelterwood or clearcut is mainly influenced by tree height (Lovett and Reiners, 1986; Augusto et al., 2002), particularly the relative tree height compared to the surroundings as tree height plays a crucial role in canopy turbulence, driving dry deposition (Beier et al., 1993; Yazbeck et al., 2021). The absence of studies linking atmospheric deposition to (relative) tree height poses challenges in incorporating the post-harvest effects of shelterwood and clearcut on atmospheric deposition into nutrient budgets. Research on the impact of clearcuttings on canopy interception of precipitation, which can be related to interception of dry deposition, is scarce and suggests varying results. Some studies indicate a 50% lower interception in clearcuts compared to unharvested stands 10 years post-clearcut, while others report only a 7% decrease 14 years after clearcut harvest (Marcotte et al., 2008; Oda et al., 2021). Due to high uncertainty in atmospheric deposition returning to pre-harvest levels in shelterwood and clearcut, I assume a proportional return based on the height growth of regeneration. Height growth data is derived from yield tables by Jansen et al. (2018), with regeneration expected to start in the first year and reach canopy closure around 6 years, corresponding to heights of 2.5, 4.7, and 1.5 m for beech, Douglas fir, and Scots pine, respectively. This assumption may, however, underestimate the impact of deposition on rotation period balances due to high ungulate densities negatively affecting regeneration in Dutch forests (Ramirez et al., 2019; Ramirez et al., 2021). This negative effect is also observed in some of the study sites showing slow settlement of the regeneration (Kampherbeek et al., 2021).

Uptake

Nutrient uptake holds significant importance in assessing nutrient budgets, particularly as they may turn negative over time as nutrient uptake increase up to 17 times during stand development (Ranger et al., 2002). However, generalizing about the rotation dynamics in uptake is challenging because those dynamics are poorly studied and affected by multiple factors such as management and disturbance history, soil differences and regeneration method (natural or planted). The nutrient uptake of regenerating stands remains poorly classified, yet these stands display unique patterns, accumulating biomass rapidly, with relative high biomass allocation to foliage and root development, and maintaining higher nutrient concentrations in their tissues (Sprugel, 1984; Miller, 1995; Bouvet and Melun, 2013; Rodríguez-Soalleiro et al., 2018). Moreover, after harvest, nutrient uptake by remaining trees may rise due to increased

nutrient, light, and water availability (Burgess and Wetzel, 2000; Sterck et al., 2021), potentially enhancing tree growth in residual trees (Mitchell et al., 1996; Carlyle, 1998). Moreover, competing vegetation can significantly increase the nutrient uptake in regenerating stands, possibly leading to negative feedbacks for seedling growth (Ferreira et al., 2021). Additionally, nutrient uptake depends on the soil's capacity to supply nutrients, which for poor soils may lead to increasing nutrient limitations and differences in nutrient resorption over time (Yan et al., 2018; Zhang et al., 2018; Wu et al., 2020a). Given these uncertainties in the uptake dynamics during the rotation, I did not estimate the nutrient uptake over a rotation period. Instead, I used the estimated nutrient losses in the exported biomass (**Ch5**) as the nutrient losses for the whole rotation period balances.

Leaching

The duration of the post-harvest effect of leaching for the different harvest intensities depends on a variety of factors including the dominant species (Jewett et al., 1995; Jerabkova et al., 2011), regeneration presence or time until regeneration establishment (Martin et al., 2000; Swank et al., 2001; Katzensteiner, 2003; Piirainen et al., 2007; Huber et al., 2010), initial soil nutrient stocks (Jerabkova et al., 2011), N saturation, soil buffer capacity (Piirainen et al., 2007; Huber et al., 2010), annual variations in rainwater surplus (de Vries et al., 2003; Webster et al., 2022), increased organic matter decomposition (Titus et al., 2006), and nutrient stocks in harvest residues (**Ch6**) (Parfitt et al., 1997). Post-harvest dynamics in soil solution chemistry are known to vary significantly in both duration and the time when nutrients reach peak concentrations (Jewett et al., 1995; Carignan and Steedman, 2000; Martin et al., 2000; Swank et al., 2001; Katzensteiner, 2003; Huber et al., 2004; Wang et al., 2006; Hope, 2009; Huber et al., 2010; Jerabkova et al., 2011). For instance, nutrient losses returned to preharvest levels within 4 to 6 years for NO₃, Ca, Mg, and Na, while for K, nutrient losses were still elevated 21-27 years after a clearcutting (Martin et al., 2000; Webster et al., 2022). In contrast, following a partial clear-felling, no elevated leaching of Na and Ca was reported for Sitka Spruce dominated catchments in Central Scotland (Tetzlaff et al., 2007). Generally, studies reporting post-harvest nutrient losses suggest that elevated leaching returns to post-harvest levels within 3 to 6 years (Jewett et al., 1995; Carignan and Steedman, 2000; Martin et al., 2000; Swank et al., 2001; Katzensteiner, 2003; Hope, 2009; Jerabkova et al., 2011) although much shorter timeframes like 1 to 2 years have also been reported (Huber et al., 2004; Wang et al., 2006; Huber et al., 2010). Furthermore, it remains ambiguous how post-harvest effects fade within this timeframe, as the greatest post-harvest effects in some studies are found six months to a year following harvest (Wang et al., 2006), while no elevated leaching was observed in this first year by Hope (2009). Instead, Hope (2009) found, in agreement with Swank et al. (2001), the highest leaching in the 2-3 years after harvest. Currently, mechanisms driving the fadeout of post-harvest leaching are poorly studied and highly

uncertain. Given that the majority of studies indicate a 3–6-year timeframe for post-harvest leaching and given my own results showing significant harvest effects on leaching 2 years post-harvest, I presume that leaching in the studied forests reverts to pre-harvest levels within six years, irrespective of harvest intensity, and that leaching remains at the measured level for the first three years and declines afterwards linearly. This assumption is based on the idea that leaching remains elevated until canopy closure, expected within six years for high-thinning, or until the establishment of regeneration. Due to the high ungulate density negatively impacting the regeneration (Ramirez et al., 2019; Kampherbeek et al., 2021; Ramirez et al., 2021), I expect leaching to persist for a longer duration after shelterwood and clearcutting. Therefore, I assumed that leaching for these harvest intensities remains elevated until 10 years post-harvest and then decreases linearly from year 3 to year 10 post-harvest.

7.7 Implications of harvest intensity for rotation period budgets

Here, I developed rotation period nutrient budgets assuming the SOH method for thinning, shelterwood, and clearcut. These budgets provide a comprehensive view of the impact of the post-harvest dynamics on rotation period budgets. I first outline the assumptions and calculations for these budgets, followed by descriptions and comparisons of the thinning, shelterwood, and clearcut balances with nutrient balances during rotations reported in the literature.

Assumptions and calculations underlying full rotation nutrient budgets.

For high-thinning, the impact on the rotation period was limited to an 8-year period, reflecting the assumed thinning frequency. Over this period, weathering was assumed stable. Leaching and deposition were assumed constant at post-harvest levels for the first three years, and linearly returning to pre-harvest levels from the third to the sixth year (Fig. 7.4A). Uptake was not incorporated in this balance; instead, nutrient export in stems and bark was used to assess nutrient recovery following stem only harvest under the assumed thinning frequency (**Ch7.6**). In this first exploration of possible full rotation impacts, I did not evaluate the implications of whole tree harvest or stem wood harvest (**Ch5**).

For the shelterwood and clearcut, I assumed a stable weathering rate, and deposition returning to pre-harvest levels assuming a linear relation with stand height. Leaching was assumed constant over the first three years at the measured post-harvest level, and then returning to control stand levels by the 10th year post-harvest (Fig. 7.4B). In the shelterwood treatment, I assumed that shelter trees would not be harvested, resulting in continuously higher deposition and lower leaching compared to a clearcut, and particularly so in the first 10 years post-harvest (Fig. 7.4B). For both shelterwood and clearcut, nutrient uptake was not considered; instead, nutrient export in stem bark and wood (SOH) was considered under the assumption that all harvested trees are exported. By doing so, I treat the nutrient uptake as an internal

flux, and I ignore possible feedbacks between nutrient uptake and the size of the available soil pool. This assumption was made as nutrient uptake over rotation periods can hardly be determined. For thinning interventions, the first two thinnings (in years 20 and 28) were considered non-commercial, with wood left in the forests. Nutrient export in subsequent thinnings between years 30 and 80 was calculated based on the extracted volume in this period, as given in the yield tables (Jansen et al., 2018). This volume was divided into fractions for bark, sapwood, and, in the case of Douglas fir and Scots pine, heartwood, based on reported distribution fractions (**Ch4**). The volume of extracted bark, sapwood, and heartwood (if applicable) was converted to mass based on wood density (**Ch4**), then multiplied by the average nutrient concentration of these tissues (**Ch4**) to obtain nutrient loss with stem export. The nutrient export from these regular thinnings was finally added to the nutrient export of the shelterwood and clearcut, as calculated in **Ch5**. For the calculation of the nutrient balance of the entire rotation, I subtracted the loss by leaching and stem export from the inputs by deposition and weathering (Fig. 7.5).

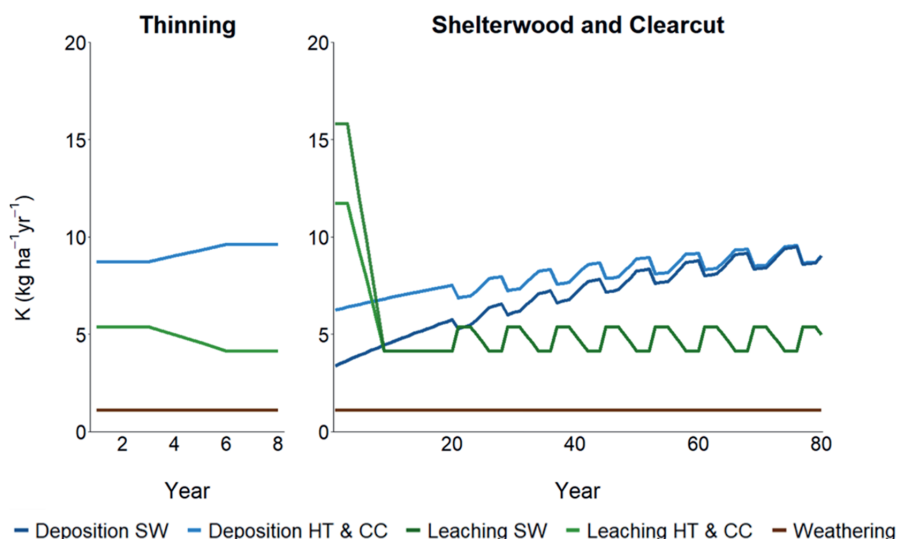


Figure 7.4 Visualization of assumptions for rotation period nutrient budget calculations for high-thinning (HT) (A) and shelterwood (SW) and clearcut (CC) (B) based on dynamics in the deposition and leaching for K. The harvest intervention was in year 1, with thinning added from year 20 onwards in shelterwood and clearcut, occurring every 8 years. Weathering rate (brown line) was assumed stable. Deposition (blue line) and leaching (green line) were assumed constant at post-harvest levels for three years after thinning and returned from year 3 linearly to pre-harvest levels in the sixth year. For shelterwood and clearcut, deposition was linearly related to tree height, and leaching stayed at post-harvest levels for three years, returning to pre-harvest levels in the 10th year post-harvest. Thinning interventions in shelterwood and clearcut followed the same assumptions as in figure A.

Thinning

A repeated thinning intervention every 8 years generally results in a positive nutrient balance, indicating that gains from deposition and weathering surpass losses from leaching and nutrient export in harvested wood (Fig. 7.5). However, Mn, Fe, and Zn balances couldn't be restored, indicating a potential risk of nutrient limitation if rates of deposition, weathering, uptake, and leaching remain stable. While the risk of Mn depletion is associated with tree harvest, regular thinning may deplete the Mn stocks in the organic soil layer within three interventions. The total Mn soil pool, however, could sustain thinning for decades, similar arguments were made for base cation stocks (Rosenstock et al., 2019). The negative Fe balance suggests soil Fe mobilization and a decline in the total soil nutrient stock, with unclear effects on the available stock as Fe weathering couldn't be determined. However, Fe input through weathering of primary minerals is generally low or neglectable, but ample Fe will be available through dissolution of secondary Fe-oxides deposited on quartz sand grains indicating no risk of shortage of available Fe. The negative balances of Zn stem from high leaching rates, indicating mobilization. Substantial Zn losses, compared to both available (**Ch5**) and total soil stocks (Table 1.3), suggest a risk of depletion within decades.

The thinning balance supports the adoption of continuous thinnings in the form of continuous cover forestry over the conventional model involving final felling. However, this balance should be interpreted cautiously, considering that the studied stands were not managed as a continuous cover system but as a conventional even-aged rotation forest. In continuous cover forestry, small scale interventions (e.g., cutting small groups of trees) may be additionally required to allow for the successful establishment of regeneration and thus turning even-aged stands into uneven-aged stands. Consequently, flux magnitudes may thus differ for continuous cover forestry compared to the explored thinning regime, and likely result in slightly different, and potentially more negative, nutrient balances.

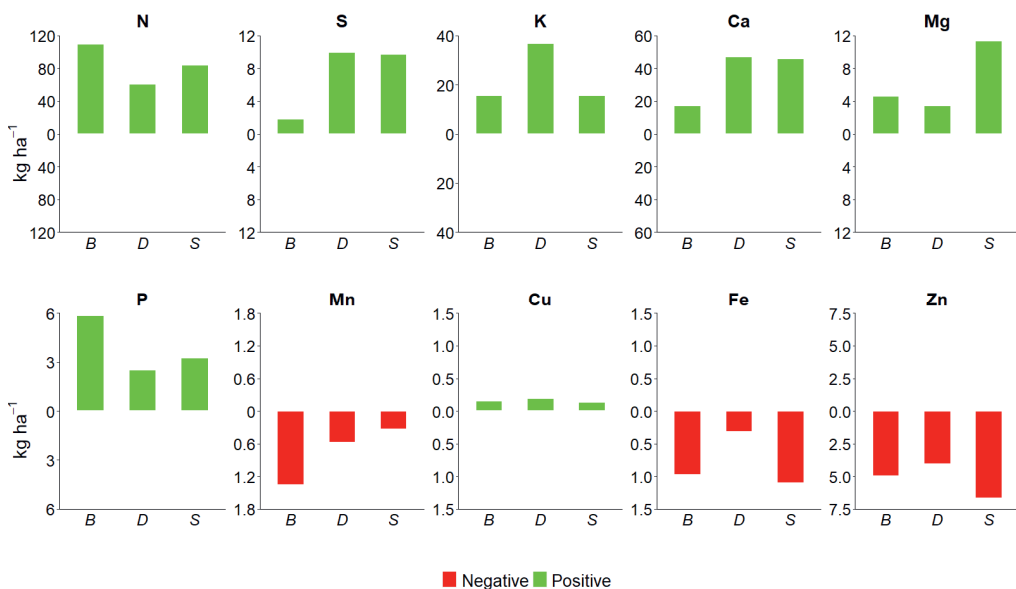


Figure 7.5 Nutrient balance showing higher nutrient inputs compared to outputs (positive) and lower nutrient inputs compared to outputs (negative) for the high-thinning using SOH for beech (B), Douglas fir (D) and Scots pine (S). This nutrient balance is constructed for a period of 8 years in which the cumulative of deposition and weathering is compared with the cumulative of leaching and the total nutrient export in stems. A positive balance indicates stable or increasing soil nutrient stocks, while a negative balance suggests nutrient depletion.

Shelterwood and clearcut

Rotation period balances for conventional, even-aged rotation forests using shelterwood or clearcut as a final felling and continuous thinnings during stand development reveal significant nutrient losses for S, K, Mg, Mn, Fe, and Zn (Fig. 7.6). Generally, nutrient losses are more pronounced when opting for clearcut as a final felling, although Mn, Fe, and Zn balances are negative regardless of the final felling type. Possible consequences of negative Mn, Fe and Zn balances are already discussed for the control stands (**Ch7.3**). The negative balances of S for beech and Douglas fir in both shelterwood and clearcut suggest ongoing effects of historical S deposition. While negative S budgets are observed, S limitation is not expected due to the system being S saturated, causing leaching to mirror deposition (De Vries et al., 1995b; De Vries et al., 2007). The rotation period balances indicate that S leaching exceeds S deposition, which is caused by the high S leaching, reflecting the control stand leaching, while the deposition has not yet returned to pre-harvest levels (Fig. 7.4). Since I assumed constant S leaching based on my measurements in clearcut and shelterwood (**Ch6**), the forecast for this negative S balance may weaken when S soil stock gradually decline over time.

When comparing the species, I expected negative cation balances for the three species, especially for beech, as harvest of stems results in high base cation exports (**Ch5**). The rotation period balances show negative budgets of K and Mg, while balances were surprisingly positive for Ca. The negative balances for K are only predicted following clearcut, with losses ($80 - 100 \text{ kg ha}^{-1}$ across species) far exceeding the available soil K stock (**Ch5**). A critical note to the available soil stocks is that the conventional methods currently underestimate the plant-available pools of Ca, Mg and K in forest soils (Bel et al., 2020), indicating that the impact of negative nutrient balances on available soil stocks might be lower than initially expected. The total K stock could support up to five rotation periods of this conventional rotation forest management using a clearcut as a final felling, indicating no direct risk for nutrient limitation (Table S1.3). Long term nutrition based on total soil stocks is in line with Rosenstock et al. (2019) who stated that total soil stocks could sustain centuries of harvest. Overall, the strong negative balances for K should be considered when planning forest management, as K deficiency could limit gross primary productivity by up to over 50% (Ouimet et al., 2013; Cornut et al., 2023). Furthermore, the negative budgets of Mg following shelterwood and clearcut in Douglas fir were surprising, as negative Mg balances in Douglas fir were not expected (de Vries et al., 2021). Moreover, Douglas fir had the highest Mg stock in the organic layers, which was previously argued to be related to a build-up of the Mg stock originating from atmospheric deposition (Cremer and Prietzel, 2017). The negative Mg balances are caused by high leaching compared to deposition (Fig. 7.7). However, the time period over which leaching returns to pre-harvest levels and the time period over which deposition returns to pre-harvest levels are both highly uncertain, which could result in the absence of negative Mg balances in the field. However, the risk of Mg deficiency in Douglas fir cannot be ignored, especially since Mg deficiency in Douglas fir has been demonstrated before (Ranger et al., 2002; Šrámek et al., 2019). Overall, the species differences were surprising, especially the positive balances in beech, as harvest in beech was expected to result in limitation even at the low harvest intensity level (de Vries et al., 2021).

The rotation period balance shows strong retention of both N and P. The strong N retention was also shown by other studies on N dynamics in Dutch forests (de Jong et al., 2023), and imply that the large amounts of deposited N over the past decades is largely stored in the forest stands. However, the level of N retention, as shown in the balance, may change in the future since N leaching is expected to increase when the soils become fully N saturated. The large and still increasing N stock may create risks for the sustainability of biomass harvest, as when N leaching starts to equal N deposition, there will be further depletion of the base cation stock, Mn and Zn stock, and increasing mobilization of Al and Fe, leading to direct risks for forest health and functioning. The strong P retention was surprising, as P was expected to become limiting even under low harvest intensities (de Vries et al., 2021). However, the large

P retention is probably caused by the significant impacts of pollen which can be seen as an internal flux, leading to an overestimation of the P input.

Finally, the rotation period balances clearly show that clearcut harvest is not sustainable due to high losses, especially of K, Mn, and Zn. Shelterwood is a more sustainable option for a final felling, although this should be done with caution, especially in Douglas fir, since Mg budgets are negative. Lastly, regarding Mn and Zn, the losses following a shelterwood are substantial, even when compared to the total soil stocks, indicating that even a shelterwood system could lead to Mn and Zn deficiencies. Therefore, continuous cover forest with small interventions to promote regeneration added to continuous thinning regimes may show the lowest Mn and Zn losses in production forests on poor sandy, acidified soils, and should be preferred over conventional, even-aged rotation forests using shelterwood or clearcut as a final felling.

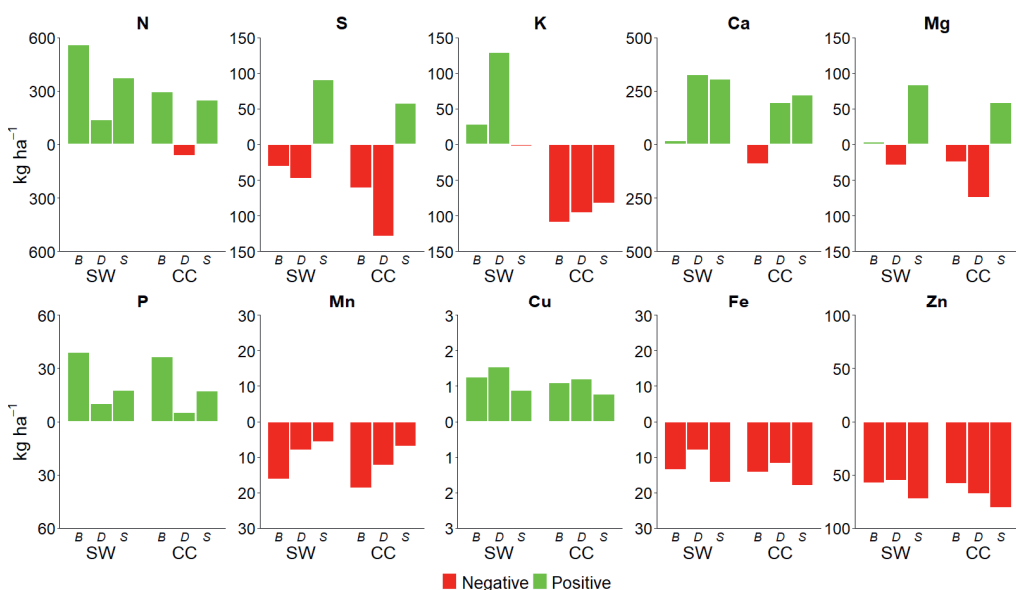


Figure 7.6 Nutrient balances over full rotations showing higher nutrient inputs compared to outputs (green and positive) and lower nutrient inputs compared to outputs (red and negative) for the shelterwood (SW) and clearcut (CC), assuming stem only harvest (SOH) for beech (B), Douglas fir (D) and Scots pine (S). This nutrient balance is constructed for a period of 80 years in which the cumulative effects of deposition and weathering are compared with the cumulative effects of leaching and the total nutrient export in stems removed following SOH. A positive balance indicates stable or increasing soil nutrient stocks, while a negative balance suggests trends towards nutrient depletion.

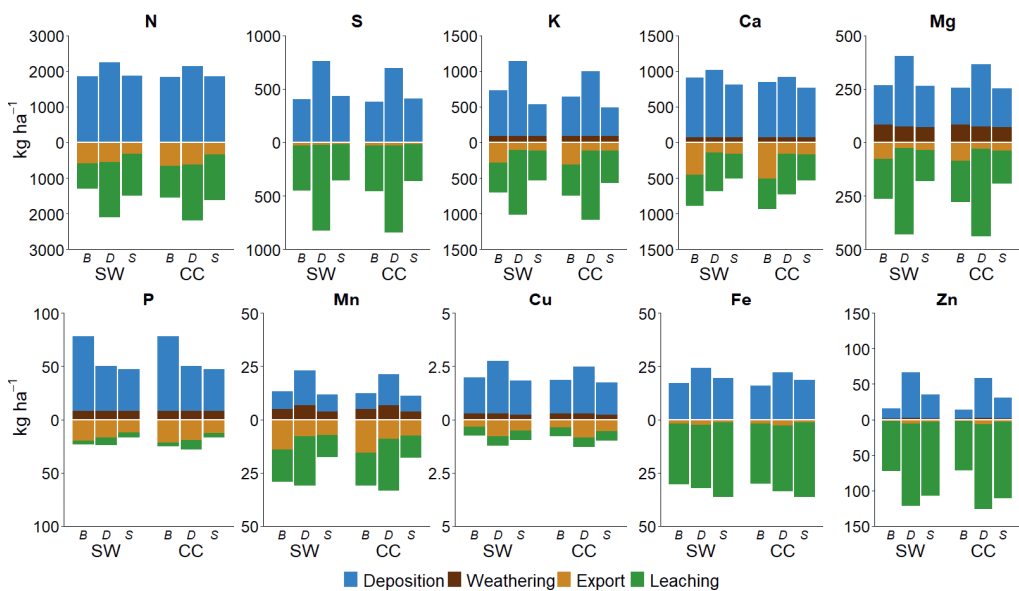


Figure 7.7 Nutrient balances over full rotations for the harvest intensities shelterwood (SW) and clearcut (CC) for beech (B), Douglas fir (D) and Scots pine (S) using SOH. The considered rotation period is 80 years. Visual representation of the main assumptions is given in Fig. 7.4. The total input fluxes (deposition and weathering, not shown for Fe) are shown above the zero line, the output fluxes (annual nutrient uptake and leaching) are shown below the zero line.

7.8 Conclusions

In this dissertation, the impact of harvest intensity on nutrient balances were assessed in untreated (control) stands and after a high-thinning, shelterwood, and clearcut. The main fluxes of the nutrient balance —atmospheric deposition, nutrient uptake and export, and leaching — revealed substantial responses to increasing harvest intensity (from control to clearcut). For deposition, increasing harvest intensity and, consequently, increasing canopy openness decreased the nutrient deposition of various macro- and micronutrients, apart from P, with the most significant reductions primarily observed for nutrients deposited in dry deposition. The study highlights the necessity of considering harvest intensity effects on forest structure when calculating nutrient inputs via atmospheric deposition. Additionally, it challenges existing nationwide deposition models by demonstrating that forests experience approximately 50% higher deposition as compared to short vegetation, emphasizing the urgent need for model calibrations. The comparison of nutrient uptake and export with nutrient stocks in the organic layers revealed immediate threats of post-harvest base cation and Mn limitations, posing risks to tree nutrition. Whole tree harvest on nutrient-poor soils should be avoided, while wood-only harvest with debarking on the site can conserve up to 50% of nutrients compared to more traditional stem only harvest. Annual

leaching was significantly affected by tree species and harvest intensity, with a strong increase in leaching at high tree harvest intensities (clearcut and shelterwood) and minimal impact at low tree harvest intensity (thinning). Remarkably, harvest method and soil preparation showed relatively weak or no effects two years after harvest. I observed strong and long-lasting effects of current and past N and S deposition, leading to elevated concentrations and leaching the base cations and of Al, Fe, Mn, and Zn, emphasizing the high risk for long term nutrient depletion associated with final cuts in the form of clearcut or shelterwood.

The trends in annual nutrient balances from low (control forest and high thinning) to high harvest intensity (shelterwood and clearcut) show a decrease in deposition, nutrient uptake and an increase in leaching, resulting in generally positive nutrient balances in control stands to strongly negative nutrient balances following clearcut. This trend, however, is not observed for P and Cu, which showed a minimal increase in leaching, and in the case of P, no decrease in deposition following a clearcut, indicating that the reduction of nutrient uptake is followed by immobilization and, therefore, positive nutrient balances. Base cation balances, expected to be growth limiting, were generally positive in control and thinned stands, except for beech controls with high uptake. Negative Mn, Fe, and Zn balances across species and harvest intensities indicate ongoing soil acidification effects and potential limitations in these elements. The full rotation period balances suggest that some of the negative balances are offset in the longer term, but still imply significant losses of K, Ca (only for beech), and Mg (beech and Douglas fir) following clearcuts, emphasizing a preference for shelterwood over clearcut in final fellings. Yet, the continuous high-thinning showed more positive balances than the shelterwood system. From this I suggest that continuous cover forestry, adopting a regular thinning regime with possibly small-scale harvests added to promote local regeneration, may be the best way forward to develop sustainable forest management practices for forests on poor, sandy and acidified soils and, as such, maintain a sustainable production of forest biomass.

7.9 Outlook

This thesis showed that the use of detailed and site-specific data coming from a large-scale forest experiment resulted in a substantial change in the assessment of the nutrient budget for forests on sandy, acidified soils. These results are currently used to modify the practical guidelines for sustainable forest management use by Dutch foresters. In comparison with the currently used decision support model for biomass harvest (de Vries et al., 2021), this study showed no risk of P depletion across the species and no Ca and K depletion except when using a clearcut as a final felling. Conversely, this study suggests potential Mn and Zn depletion, emphasizing the need for further investigation in future studies. However, there is an urgent need to enhance knowledge of (post-harvest) nutrient dynamics, especially over longer periods, and for forests in different geographical areas with variations in deposition inputs, soil conditions, tree

species, and forest management regimes. Additionally, future predictions will need to consider the ongoing effects of climate change on tree performance in various tree harvest regimes (Sterck et al., 2021), but this was beyond the scope of my thesis. In the context of this thesis, I propose further research into ten different aspects of the nutrient balance:

1. There is a necessity for improved estimation of weathering rates, given the potential influence of factors such as harvest, climate change, and soil acidification (**Ch7.2**).
2. The focus was on the nutrient balance of conventional even-aged rotation forests and post-harvest dynamics within such stands. Given the emphasis on continuous thinnings, it is essential to assess nutrient dynamics in uneven-aged and mixed species stands managed under a continuous cover forest management regime.
3. Post-harvest dynamics were exclusively evaluated for the second year after harvest. The duration required for post-harvest nutrient dynamics in atmospheric deposition and leaching to stabilize to mature forest dynamics is unknown, leading to substantial uncertainties in rotation period balances. Moving from these large assumptions to field data on longer term forest recovery is essential to enhance the reliability of the rotation period nutrient balance.
4. I did not explore nutrient dynamics over a rotation period. Leaching dynamics are likely unstable, varying during self-thinning stages and evolving differently across species and soil types (Johnson et al., 1995; Swank et al., 2001). Post-harvest dynamics in soil solution chemistry are known to vary strongly both in duration and in the timing when nutrients reach their highest concentrations (**Ch6**). For this study, it remains speculative whether we sampled the soil solution chemistry during the peak like observed by Swank et al. (2001) or that the greatest post-harvest effects happened in the first year or mainly after three years post-harvest (Wang et al., 2006). Studying leaching over the entire rotation period, considering post-harvest leaching duration for different species, soil types, and climates, and linking these fluxes to specific soil properties and regeneration establishment is essential for a comprehensive understanding.
5. The intensive sampling of deposition and leaching was conducted in a year that was drier than normal (approximately 150 mm below annual average) which could have influenced the results especially for leaching. Therefore, monitoring soil solution chemistry in unharvested stands over extended periods is crucial for reliable incorporation into nutrient budgets, considering variations between years (Fahey and Yavitt, 1988) and annual hydrological differences (de Vries et al., 2003), which may lead to substantial differences in leaching estimates between years (Webster et al., 2022).
6. Current legislation in the Netherlands targets a significant reduction in N deposition, which could alter nutrient leaching over a rotation period, leading to different nutrient balances. Conversely, if N deposition remains elevated, the system may reach full saturation, impacting leaching dynamics.

Hence, continuous, long-term monitoring of leaching is crucial for a comprehensive understanding of nutrient dynamics.

7. Nutrient balances potentially turn negative over the rotation period. Consequently, investigating the nutrient uptake of the regeneration and the remaining forest is essential, along with assessing possible effects of temporary nutrient depletion to understand the overall sustainability of the forest ecosystem.
8. I did not consider the substantial uptake of nutrients by belowground biomass (e.g., (Helmisaari, 1995; Leuschner et al., 2004; Vanguelova et al., 2005; Yuste et al., 2005)). Neglecting nutrient immobilization by roots leads to an underestimation of nutrient uptake, with effects varying among nutrients and species. It is crucial to incorporate belowground nutrient immobilization in roots for the annual nutrient balances, especially since trees tend to increase root investment when facing nutrient limitations (Yuste et al., 2005).
9. The nutrient balance is determined for mature forests growing on nutrient-poor sites, as such, the results cannot be extrapolated to younger stands or stands on richer substrates as there is ample evidence of within-species variability in nutrient concentrations likely influenced by soil fertility, soil acidity and tree age (Boerner, 1984; Bouvet and Melun, 2013; Heineman et al., 2016; Achat et al., 2018b; Rodríguez-Soalleiro et al., 2018). It is therefore recommended to use tree nutrient concentration data representative for the age class and soil fertility of the specific site.
10. Ideally, when estimating nutrient exports, biomass expansion factors specific to species, age, region, and site index (Jalkanen et al., 2005; Teobaldelli et al., 2009) should be used. However, this data is often lacking, increasing uncertainty in the nutrient budget. When lacking, it is strongly advised to use biomass expansion factors accounting for species and age, as the proportion of stem and branch biomass increases with tree age, and young stands often exhibit high variance and heterogeneity in structure (Lehtonen et al., 2004; Jalkanen et al., 2005; Pajtik et al., 2008). Therefore, standing stock estimates should be developed with care, considering specific stand properties.

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Summary

Forests play a key role in achieving a fully bio-based economy by supplying woody biomass through tree harvesting. The growing interest in biomass utilization in Europe is driving an increased demand for tree biomass, primarily for timber and other wood products, as well as bioenergy. This utilization, however, puts additional pressure on forests, impacting soil, nutrient pools, site productivity, and overall forest health, including tree growth and survival rates.

The sustainability of biomass harvest and removals, concerning tree and soil nutrient stocks, can be evaluated through the nutrient budget approach, where nutrient input fluxes include atmospheric deposition and weathering, while major nutrient outputs comprise leaching and nutrients in removed tree parts after harvest (Fig. 1). Sustainable biomass harvest requires that nutrient output from harvest and leaching does not exceed nutrient input from deposition and weathering, preventing a decline in available forest nutrient stocks over successive rotations or felling cycles. Additional nutrient inputs and outputs, such as groundwater supply, biological fixation, denitrification, and nutrient loss through rainwater runoff and river flooding, can also contribute to this balance but are considered negligible in the study plots.

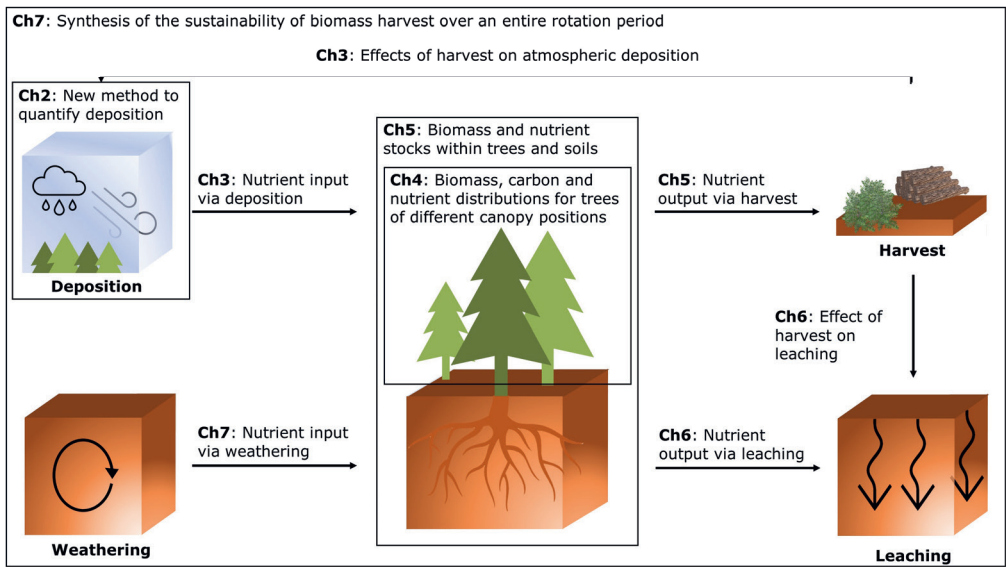


Figure 1 Overarching diagram of forest nutrient budgets, also connecting the research topics per chapter of the PhD thesis.

The aim of this thesis is to understand the effects of different forest management practices on the deposition, export through harvest, and the leaching of nutrients, thereby influencing forest nutrient budgets (Fig. 1). First, I tested the hypothesis that the nutrient input and output fluxes in unharvested plots are significantly influenced by tree species. Next, I tested the hypothesis that the main post-harvest nutrient input and output fluxes are significantly influenced by forest management practices, specifically

Summary

harvest intensity, and also harvest methods, while tree species differences are more limited. Finally, I calculated the post-harvest nutrient balances and used these in a first attempt to upscale the post-harvest dynamics to a whole rotation period nutrient budget of 80 years. By doing so, the research outcome contributes to the development of science-based guidelines for ecologically sustainable biomass harvest.

The experiments are conducted in monoculture forest stands of European beech (*Fagus sylvatica*), Douglas fir (*Pseudotsuga menziesii*), and Scots pine (*Pinus sylvestris*) on well-drained, poor, and acidic sandy soils in five regions in the Netherlands. The study sites have a temperate maritime climate (annual temperature of 10.4°C and rainfall of 805 mm), are exposed to high nitrogen (N) deposition, lack groundwater access for roots, have negligible runoff and biological N fixation, and no flooding. The experiment was initiated in 2018, with initial measurements, including standing stock and intensive soil sampling, carried out in winter 2018/2019. Tree harvest took place in February-March 2019. Harvest intensities include control (no harvest), high thinning (HT, where 15 to 20% of the basal area is felled), shelterwood (SW; 76-83%), and clearcut (CC; 100%). The harvest methods considered are whole tree harvest (WTH), where the crown is also harvested; stem-only harvest (SOH), where crowns are left in the forest; and wood-only harvest (WOH), in which the bark is stripped in the field. Flail mulching, carried out in March-April 2019, is the predominant practice for soil preparation after harvest and is used to facilitate the establishment of the regeneration. The type of mulching considered is shallow mulching, where harvest residues like branches and tops are cut into smaller pieces, without disturbing the mineral soil.

In **Chapter 2**, a new method for measuring atmospheric nutrient deposition of macro- and micronutrients (N, P, S, Ca, Mg, K, Mn, Cu, Fe and Zn) beneath forest canopies and in gaps is presented and tested. The Ion Exchange Resin (IER)-method's suitability for quantifying deposition of macro- and micro-nutrients was assessed. Results indicate that the IER-method is effective for all nutrients (except Phosphorus or P) under various laboratory conditions, with minimal impact from factors like heat (up to 40°C), drought, and frost (down to -15°C). Additionally, the IER-method performed well under field conditions, providing a more consistent estimation of deposition compared to conventional approaches. This method proves to be a powerful tool for monitoring atmospheric deposition in managed forests.

In **Chapter 3**, the IER-method is applied to evaluate the effect of harvest-modified canopy openness on the atmospheric deposition of macro- and micronutrients in forests. We show that the total annual nutrient deposition of macro- and micronutrients in beech, Douglas fir, and Scots pine stands consistently decreased with increasing tree harvest intensity and associated increased canopy openness. An exception is P, which probably depends on pollen dispersal rather than atmospheric deposition. Across species, the highest deposition of acid N and S compounds but also of the base cation Ca, Mg and K were observed in the relatively tall stands of Douglas fir, and the lowest in the relatively short and denser stands of Scots pine. Forest stand deposition surpassed nationwide model estimates, with NH₄ and S deposition

exceeding estimates by 29-140%, and neutralizing base cation deposition exceeding estimates by 68-750%, suggesting a lower risk of soil acidification in forests than expected. Moreover, the experiments showed the importance of accounting for seasonal differences in nutrient deposition. The results also highlight the importance of including the effects of harvest intensity on forest structure and, to a lesser extent, the selected tree species to make a scientifically sound calculation of nutrient inputs via atmospheric deposition. Interestingly, harvest intensity may function as a management tool to partially reduce the net acidic inputs into Dutch forests as thinned forests intercept highest base cations (K, Ca, Mg) in comparison to acids (NH_4 , S). This compensating effect, however, remains rather limited.

In **Chapter 4**, the effect of tree canopy position and, consequently, tree competition on the distribution of biomass, carbon, and nutrients within trees was analysed, creating the base for upscaling the nutrient stock of a single tree to the nutrient stocks per hectare of managed forest. Carbon concentrations were relatively constant across tree compartments, while nutrient concentrations increased from stem, bark, branches towards needles. Canopy position had only minor effects on carbon and nutrient concentrations and on the distribution of biomass, carbon, and nutrients between aboveground tree components. This effect was overall negligible, indicating that models aiming to estimate tree and forest biomass, carbon, and nutrient stocks can apply equal biomass, carbon, and nutrient stocks for trees independent of canopy position as a valid assumption. Hence, our results are in line with the allometric scaling theory that assumes a constant distribution of biomass, carbon, and nutrients regardless of tree competition.

In **Chapter 5**, the biomass and nutrient data from individual trees, as described in Chapter 4, were used to create allometric relationships and calculate nutrient stocks for macro- (N, P, S, Ca, Mg, K) and micronutrients (Mn, Fe, Cu, Zn) in aboveground tree tissues. Annual nutrient uptake was determined based on tree ring measurements indicating tree growth, utilizing the constructed allometric relationships. Organic layer nutrient stocks and available nutrients in the mineral soil were assessed through intensive soil sampling. Beech, Douglas fir, and Scots pine stands showed substantial variations in aboveground biomass and nutrient stocks, with Douglas fir having the highest biomass stocks. Beech stands demonstrated the highest aboveground nutrient stocks, nutrient uptake rates, and nutrient losses after harvest, followed by Scots pine. Organic layer nutrient stocks generally surpassed aboveground nutrient stocks, except for the base cations and Mn, which suggest potential long-term threats to forest nutrition when harvested. In comparison to SOH, WTH increased nutrient export by 66-100%, while WOH decreased nutrient export by 23-41%. Considering the differences in nutrient stocks, uptake rates, and nutrient exports following harvest methods, it is recommended to limit tree harvest based on tree species, avoid WTH, and consider WOH to better preserve critical nutrients essential for long-term forest recovery.

In **Chapter 6**, the impact of forest management practices on post-harvest nutrient leaching was assessed for beech, Douglas fir, and Scots pine, considering different harvest intensities (CO, HT, SW, and CC), various harvest methods (SOH, WTH), and soil preparation (mulching). Nutrient leaching calculations involved a yearly cycle of monthly measurements of dissolved nutrient concentrations beneath the rooting zone, coupled with a mechanistic model simulating monthly water fluxes. In control stands, Douglas fir exhibited generally higher nutrient leaching compared to Scots pine and beech stands. Clearcutting, and to a lesser extent shelterwood, led to increased dissolved nutrient concentrations, indicating rapid mobilization of substantial nitrogen stocks, resulting in accelerated losses of nitrate, base cations, and aluminium, contributing to accelerated soil acidification. Thinning showed minor effects on leaching, acting as a preventive measure against accelerated soil acidification. Harvest methods had marginal effects on leaching, with mulching showing negligible impact. The results underscore the significance of forest structure, influenced by harvest intensity and, to a lesser extent, tree species, in nutrient losses via leaching. Additionally, they indicate that soil acidification effects from nitrogen or sulphur deposition become more pronounced in intensively harvested forests compared to high thinning and control plots.

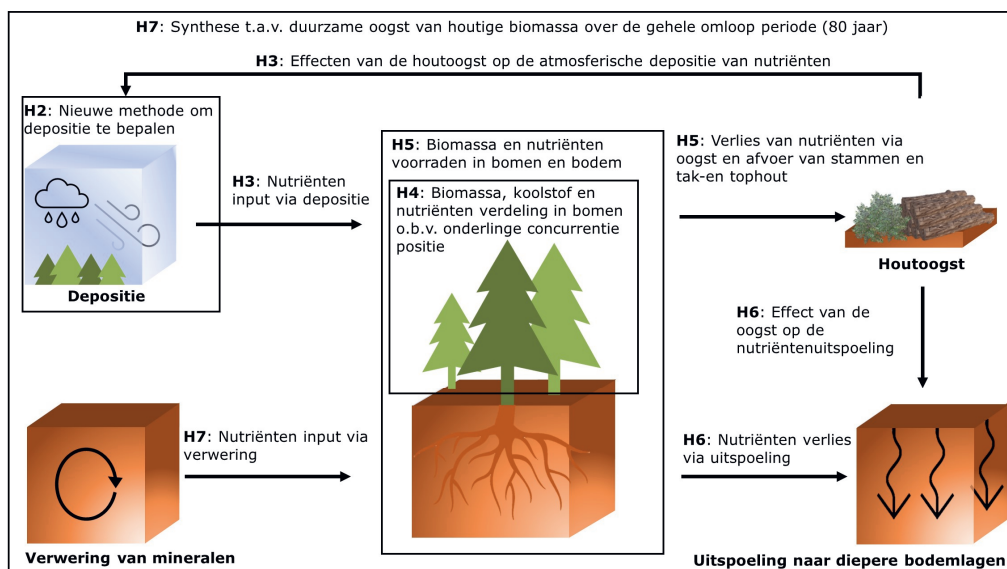
In **Chapter 7**, the evaluation of this thesis led to the compilation of annual nutrient balances for the second-year post-harvest, considering three harvest intensities (HT, SW, CC) and two harvest methods (SOH, WTH) in comparison to control plots without harvest. A literature review determined the weathering flux, revealing a modest nutrient input flux compared to deposition. Post-harvest nutrient balances for the second year indicated strongly negative budgets for shelterwood and clearcut, while high-thinning showed slightly negative to positive balances, indicating that continuous forest cover management, including thinning and possibly small-scale interventions, should be preferred over a final cut. Regardless of harvest intensity, Mn, Fe, and Zn balances were negative across species, indicating potential risks of nutrient limitation, particularly for Mn and Zn, while the Fe balance indicated mobilization and the initiation of the Fe soil buffering mechanism. Additionally, I assessed the duration over which post-harvest nutrient dynamics would diminish, constructing full-rotation nutrient balances using continuous thinnings and a final cut (SW or CC). Assumptions included deposition returning to pre-harvest levels linearly with tree height and elevated leaching within 10 years post-harvest, after which it fades linearly. Rotation period balances suggested significant long-term losses of K, Ca (only for beech), and Mg (beech and Douglas fir) following clearcuts, emphasizing a preference for shelterwood over clearcut in final fellings. In conclusion, achieving sustainable forest harvest on poor, sandy soils for the studied species is best realized through low-intensity practices, for example by focussing on continuous cover management with regular thinning and small-scale interventions (e.g., small group cuts creating small open spots) added for promoting regeneration, and thus turning even-aged forests to uneven-aged forests.

Samenvatting

Bossen vervullen een cruciale rol in de transitie naar een koolstofarme en circulaire bio-economie vanwege de levering van biomassa door houtoogst. De groeiende belangstelling voor het gebruik van biomassa in Europa leidt tot een grotere vraag naar hout voor zowel duurzame toepassingen als voor het opwekken van warmte en elektriciteit. Een volledig circulaire bio-economie kan daarom leiden tot een toenemende vraag naar hout en houtige biomassa wat kan resulteren in een toenemende druk op bossen. Deze toenemende druk kan effecten hebben op de bodem, de nutriëntenvoorraden, boomgroei, bosherstel en biodiversiteit.

Eén van de belangrijkste aspecten voor duurzaam bosbeheer is een evenwichtige nutriënten balans. De nutriëntenbalans bestaat uit de verhouding tussen enerzijds de nutriënteninstroom in de vorm van atmosferische depositie en verwerking van mineralen en anderzijds de nutriëntenuitstroom via uitspoeling en export van nutriënten na de houtoogst (Fig. 1). Duurzame houtoogst impliceert dat het verlies van nutriënten door houtoogst en uitspoeling niet groter is dan de nutriënteninput door depositie en verwerking. Ook andere stromen of fluxen kunnen invloed hebben op dit evenwicht. Denk dan aan de toevoer van voedingsstoffen via het grondwater en mogelijke overstromingen, biologische fixatie en de afvoer via regenwaterafvloeiing. Voor onze proefopzet met Nederlandse bossen op hogere zandgronden (zie hierna) worden deze andere stromen als verwaarloosbaar beschouwd.

Het doel van deze scriptie is in beeld brengen van de effecten van bosbeheer op de atmosferische depositie, de nutriënten export door de houtoogst en de uitspoeling van nutriënten en deze effecten te vertalen in een nutriëntenbalans van bossen onder verschillende beheersscenario's (Fig. 1). Als eerste is de hypothese getest dat de toe- en afvoer van nutriënten in bossen significant beïnvloed wordt door de hoofdboomsoort. Vervolgens is de hypothese getest dat de belangrijkste nutriënten in- en outputstromen significant worden beïnvloed door het bosbeheer. Het bosbeheer is hierbij gedefinieerd de oogstintensiteit, oogstmethode en de bodembehandeling. Ten slotte zijn de nutriëntenbalansen na de houtoogst berekend. Deze berekening is gebruikt om de dynamiek van de depositie, opname en uitspoeling van nutriënten op te schalen naar een rotatieperiode van 80 jaar. Hierdoor draagt deze studie bij aan de ontwikkeling van wetenschappelijk onderbouwde richtlijnen voor een duurzame houtoogst in de Nederlandse bossen.



Figuur 1 Overzicht van de nutriëntenbalans met daarin de nutriënteninput via depositie en verwerking en de nutriëntenoutput door houtoogst en uitspoeling. De pijlen geven de onderliggende verbanden aan, ook wordt verwezen naar de desbetreffende hoofdstukken in de scriptie.

De experimenten zijn uitgevoerd in gelijkjarige monoculturen van Europese beuk (*Fagus sylvatica*), Douglasspar (*Pseudotsuga menziesii*) en grove den (*Pinus sylvestris*) op goed doorlatende, arme en zure zandgronden in vijf regio's in Nederland. De geselecteerde opstanden hebben een gematigd zeeklimaat (gemiddelde jaartemperatuur van 10,4°C en regenval van 805 mm), zijn blootgesteld aan hoge stikstofdepositie (N), en liggen buiten het bereik van grondwater. De biologische N-fixatie is zeer beperkt. Het experiment is in 2018 gestart en de nulmetingen van de oorspronkelijke situatie, inclusief staande voorraad en nutriëntenvoorraden in de bodem, zijn uitgevoerd in de winter van 2018/2019. De houtoogst is uitgevoerd in februari-maart 2019. De toegepaste oogstintensiteiten betreffen controle (geen oogst), hoogdunning (waarbij 15 tot 20% van het grondvlak is gekapt), schermkap (76-83% van het grondvlak gekapt) en kaalkap (100% gekapt). Naast de oogstintensiteit is ook de oogstmethode onderzocht, waarbij onderscheid gemaakt is tussen het vellen en weghalen (oogsten) van hele bomen, inclusief tak- en tophout, het oogsten van de stam (tak- en tophout blijft in het bos) en het oogsten van het stamhout zonder schors (stam wordt ter plekke gestript; schors blijft achter). Ondiep klepelen is uitgevoerd in maart-april 2019 op de helft van de schermkap en kaalkapplots. Deze vorm van klepelen, waarbij oogstresten zoals het tak- en tophout, in kleinere stukken wordt gebroken, is de algemene praktijk voor groundbewerking na de oogst en wordt gebruikt om de (natuurlijke) verjonging te faciliteren.

In **Hoofdstuk 2** is een nieuwe methode getest om de atmosferische depositie van macro- en micronutriënten (N, P, S, Ca, Mg, K, Mn, Cu, Fe en Zn) in bossen en open gebieden te meten. De Ion Exchange Resin (IER)-methode filtert opgevangen neerslag (natte depositie) door een gemengde anion- en kationenwisselaar. De metingen zijn hierdoor minder gevoelig voor weersinvloeden en biologische omzettingen. De resultaten tonen aan dat de IER-methode effectief is voor het meten van de atmosferische depositie van alle nutriënten (behalve fosfor of P) onder diverse laboratoriumomstandigheden, met minimale invloed van factoren zoals hitte (tot 40°C), droogte en vorst (tot -15°C). De IER-methode presteerde bovendien goed onder veldomstandigheden, wat resulteerde in een consistentere schatting van de depositie in vergelijking met conventionele benaderingen. Deze methode blijkt een krachtig instrument te zijn voor langdurige metingen van atmosferische depositie in bossen.

In **Hoofdstuk 3** wordt de IER-methode toegepast om het effect van de toenemende openheid van het kronendak ten gevolge van houtoogst op de atmosferische depositie van macro- en micronutriënten in bossen te meten. De resultaten tonen aan dat de totale jaarlijkse depositie van macro- en micronutriënten in opstanden van de beuk, Douglasspar en de grove den consistent afnam met toenemende oogstintensiteit en de daarmee samenhangende grotere openheid van het kronendak. Een uitzondering is P, waarbij de input waarschijnlijk bepaald werd door de depositie van pollen. De hoogste depositie van NH₄ en S (zure depositie) en van K, Ca en Mg (basen) is waargenomen in de relatief hoge opstanden van de Douglasspar, en de laagste in de relatief korte en dichtere opstanden van de grove den. De totale depositie in bosopstanden overtrof de landelijke modelschattingen, waarbij de NH₄ en S-depositie de schattingen met 29-140% overtroffen. De depositie van basenkationen overtrof de schattingen met 68-750%, wat erop wijst dat het risico op bodemverzuring in bossen lager is dan verwacht. De resultaten benadrukken het belang van het integreren van de effecten van houtoogst en de effecten van de boomsoort in de schattingen van de jaarlijkse totale depositie op bossen. Daarnaast kan houtoogst in een lage intensiteit fungeren als een beheerinstrument om de huidige relatief hoge N-depositie en bijbehorende zuur input in Nederlandse bossen deels te verminderen, hoewel het compenserende effect beperkt blijft.

In **Hoofdstuk 4** wordt het effect van boomkroonpositie, wat een maat is van de onderlinge concurrentie tussen bomen, op de verdeling van biomassa, koolstof en nutriënten binnen bomen geanalyseerd. De verdeling van biomassa en nutriënten vormde de basis voor het opschalen van de nutriëntenvoorraad van een enkele boom naar de nutriënten voorraden per hectare beheerd bos. Koolstofconcentraties waren relatief constant over boomcompartimenten, terwijl nutriëntenconcentraties toenamen van stam, schors, takken naar naalden. Boomkroonpositie had slechts een klein effect op koolstof- en nutriënten concentraties en op de verdeling van biomassa, koolstof en nutriënten tussen bovengrondse boomcomponenten. Dit effect was over het algemeen verwaarloosbaar, wat aangeeft dat modellen die gericht zijn op schattingen van boom- en bosbiomassa, koolstof en nutriëntenvoorraden,

gelijke biomassa, koolstof en nutriëntenvoorraden voor bomen kunnen gebruiken ongeacht de kroonpositie. Onze resultaten zijn in lijn met de theorie van de allometrische schaling die uitgaat van een constante verdeling van biomassa, koolstof en nutriënten ongeacht de concurrentiepositie van een boom.

In **Hoofdstuk 5** worden de biomassa en de nutriëntendata van individuele bomen, zoals beschreven in Hoofdstuk 4, gebruikt voor het opstellen van allometrische relaties. Allometrische relaties zijn essentieel voor het voor het opschalen van de biomassa en daardoor de nutriëntenvoorraden van boomniveau naar bosniveau. De bovengrondse nutriëntenvoorraad werd hierbij bepaald voor macro- (N, P, S, Ca, Mg, K) en micronutriënten (Mn, Fe, Cu, Zn). De jaarlijkse opname van nutriënten door bomen werd bepaald op basis van metingen van de jaarringen in de stam als indicator voor boomgroei in combinatie met de geconstrueerde allometrische relaties. De voorraden van nutriënten in de organische laag en de beschikbare nutriënten in de minerale bodem werden bepaald via intensieve bodembemonstering. Opstanden van de beuk, Douglasspar en de grove den vertoonden aanzienlijke variaties in bovengrondse biomassa en nutriëntenvoorraden, waarbij de Douglassparren de hoogste biomassa hadden. Beukenopstanden hadden echter de hoogste bovengrondse nutriëntenvoorraden, opnamepercentages en nutriëntenverliezen na de oogst, gevolgd door grove den opstanden. Over het algemeen overtroffen de nutriëntenvoorraden in de organische laag de bovengrondse nutriëntenvoorraden, behalve voor de basenkationen (K, Ca en Mg) en Mn, wat wijst op mogelijke nutriëntentekorten bij herhaalde houtoogst. In vergelijking met het oogsten van alleen het stamhout verhoogde het oogsten van de hele boom, dus inclusief tak- en top hout, de export van nutriënten met 66-100%, terwijl het oogsten van alleen stamhout zonder bast (WOH) de export van nutriënten met 23-41% verminderde. Gezien de verschillen in nutriëntenvoorraden, opnamepercentages en export van nutriënten, wordt aanbevolen om de houtoogst te beperken op basis van boomsoort, oogst van tak- en top hout te vermijden en het oogsten van enkel de stammen (dus zonder bast) te overwegen om essentiële nutriënten te behouden.

In **Hoofdstuk 6** wordt de impact van bosbeheer op het uitspoelen van nutriënten na de oogst bepaald voor opstanden van de beuk, Douglasspar en de grove den voor verschillende oogstintensiteiten, diverse oogstmethoden en bodembehandelingen (klepelen). De bepaling van de nutriëntenuitspoeling is gebaseerd op een jaarlijkse cyclus van maandelijkse metingen van de nutriëntenconcentraties in het bodemvocht onder de wortelzone, gekoppeld met een mechanistisch model dat maandelijkse waterfluxen simuleert. In controlebossen vertoonde de Douglasspar over het algemeen hogere nutriëntenuitspoeling in vergelijking met opstanden van de beuk en de grove den. Kaalkap, en in mindere mate schermkap, leiden tot verhoogde nutriëntenconcentraties in het bodemvocht wat wijst op een snelle mobilisatie van de grote stikstofvoorraden. Deze mobilisatie resulteert in versnelde verliezen van nitraat, basenkationen en aluminium, wat bijdroeg aan versnelde bodemverzuring. Dunningen leiden tot geringe effecten op de nutriëntenuitspoeling en leiden dus niet tot versnelde bodemverzuring. De verschillende oogstmethoden

beïnvloeden de uitspoeling maar voor enkele nutriënten waarbij de invloed meestal gering is. Klepelen had geen effecten op de nutriëntenuitspoeling. De resultaten benadrukken het belang van de bosstructuur, beïnvloed door oogstintensiteit en, in mindere mate, boomsoorten, bij nutriëntenverliezen via uitspoeling. Daarnaast zijn de effecten van bodemverzuring door stikstof- of zwaveldepositie sterker in intensief geogoste bossen vergeleken met gedunde en controle opstanden.

In **Hoofdstuk 7** zijn de onderzoeksresultaten van de eerdere hoofdstukken gecombineerd en verwerkt in nutriëntenbalansen voor het tweede jaar na de houtoogst. Hierbij zijn de effecten van de drie oogstintensiteiten (hoogdunning, schermkap en kaalkap) en twee oogstmethoden (oogsten van stamhout en oogsten van stamhout plus tak- en tophout) vergeleken met controlepercelen zonder oogst. De nutriënteninput via verwerking, bepaald op basis van een literatuuronderzoek, was slechts bescheiden ten opzichte van de depositie. Nutriëntenbalansen van de schermkap en kaalkap waren sterk negatief, terwijl die van de hoogdunning licht negatief tot positief waren. Dit geeft aan dat continue dunningen een beter alternatief zijn dan een eindkapsysteem. Ongeacht de oogstintensiteit waren de Mn, Fe en Zn balansen negatief voor alle boomsoorten, wat wijst op potentiële risico's van nutriëntenlimitatie, met name voor Mn en Zn, terwijl het consistente verlies van Fe duidt op mobilisatie en het Fe-bodembuffermechanisme. Daarnaast is beoordeeld over welke periode de nutriëntendynamiek na de oogst zou afnemen. Op basis van literatuuronderzoek zijn aannames gedaan met betrekking tot het uitdoven van de effecten van de houtoogst op de depositie en de uitspoeling. Aangenomen wordt dat de depositie lineair schaaft met de boomhoogte en dat de uitspoeling tot 10 jaar na de oogst verhoogd is, waarbij de afname lineair is. Rotatieperiode-balansen, opgesteld voor een periode van 80 jaar waarbij bossen vanaf 30 jaar elke 8 jaar worden gedund en waarbij de eindkap bestaat uit schermkap of kaalkap, suggereren aanzienlijke verliezen van K, Ca (alleen voor beuk), en Mg (beuk en Douglasspar) na kaalkap, waarbij de voorkeur werd gegeven aan schermkap boven kaalkap in geval een eindkap noodzakelijk is. Concluderend kan gesteld worden dat duurzame oogst van bossen op arme, zanderige bodems het beste bereikt kan worden door het toepassen van een uitkapsystemen met continue dunning, eventueel aangevuld met kleinschalige ingrepen om regeneratie te bevorderen.

Acknowledgements

As I sit down to write these acknowledgements, a wave of nostalgia washes over me. It's a peculiar feeling, one of melancholy mixed with gratitude, as I reflect on the years that have led me to this moment. Pursuing a PhD was never a central aspiration in my life, however, now at the end of this journey I can say that these years have shaped me, and they will be amongst the most beautiful years of my life. During this PhD trajectory, I learned to overcome challenges, tested the limits of my perseverance and I experienced the joy of working in an environment in which almost everything seemed possible. This environment consisted of the knowledge and passion of the FEM-group, the valuable help and practical insights of UNIFARM and of course of partner-organizations that initiated this research, gave permission for experimental work in their forests and provided practical insights and help in the establishment of the experiment. Overall, I want to express my appreciation to everyone who made this PhD journey possible. It's a collective effort, and I owe my gratitude to my supervisors, colleagues, family and friends who offered guidance and support.

First and foremost, I would like to thank Frank Sterck without whom I would probably never have started this PhD journey. Frank, our paths first crossed when we discussed the possibility of a master's thesis in collaboration with WSL as it was my wish to do field research in the Alps. Through that master's thesis and the subsequent internship, I not only began to appreciate science but truly started to love doing science. Alongside Georg von Arx, the both of you encouraged me and provided me with the freedom to explore and inquire. It was this taste of scientific freedom that encouraged me to dive deeper into the world of science. I want to thank you for entrusting me to contribute to the development of the PhD proposal, ultimately resulting in the NWO grant within the top sector of Horticulture and Starting Materials. As my daily supervisor and first promotor, your advice, feedback and support were very valuable throughout my PhD. By affording me the scientific freedom you enabled me to explore new methods and expand the PhD to the research project it is now. I have appreciated all the help and support, also in the form of practical help with the monthly sampling rounds. Frank, without your initial guidance and ongoing support I would neither have started nor successfully ended this remarkable PhD journey, therefore many thanks.

I would also like to thank my other supervisors Wim de Vries, Jan den Ouden and Marcel Hoosbeek which gave indispensable feedback, advice and support. Wim, you have been an enthusiastic supervisor with a keen eye for detail and a realistic perspective on the feasibility of the research plans. Your involvement in the PhD trajectory began during the proposal development, and your insights played a crucial role in shaping the proposal and refining the research methods, especially those related to atmospheric deposition and leaching. Your knowledge and feedback have contributed significantly to the quality and extend of this thesis. Many thanks for your dedication and valuable contributions. Jan, your

expertise and support in all forestry-related matters were of crucial importance especially during the most challenging phase of my PhD – the installation of the experiment. I had limited practical experience in forestry, and your critical, down-to-earth approach guided me through this crucial phase. You not only taught me essential skills but also encouraged me to think beyond conventional boundaries. It's safe to say that without your input, I would not have been able to install the experiment. Also further in the PhD journey, your feedback, although sometimes challenging to incorporate, significantly improved the scientific output. Jan, you exemplify the scientist I aspire to be – as you are deeply connected to applied research and practical applications of this research. Marcel, when I started this PhD, I had little knowledge and experience in the field of soil sciences which posed a major challenge as soil chemistry was a major part of this PhD. In your kind and patient way, you have provided me with the needed knowledge for this part of the PhD for which I'm grateful. You took the time to help me understand complex concepts and provided essential insights as well as facilitating connections with the CBLB laboratory. Your contributions are much appreciated.

With so many remarkable persons involved in this journey, it's challenging to set the order of acknowledgement. However, when I reflect on the person who played a key role in the practical aspects of this PhD – the countless hours of fieldwork and lab work – I definitely think of Henk van Roekel. Henk, we shared countless hours together in the field, at the Unifarm facilities and within the CBLB laboratory and I enjoyed working with you. From the start of the experiment, you became my right hand and soon you became indispensable. You coordinated all the assistance we received from UNIFARM, spanning from the provision of materials for weighing the trees in the field (including that impressive crane!) to the assistance of various individuals during the demanding fieldwork phases. Furthermore, you were a big help in the development of the deposition samplers and indispensable by the installation of these samplers and the leaching samplers. Because of your efforts, it was easy for me to trust you with the monthly sampling of the deposition which allowed us to simultaneously collect deposition and leaching samples, which significantly advanced our research. The fieldwork was often demanding, involving long days that stretched from dawn till dusk, occasionally in unfavorable weather conditions. It also encompassed a great deal of monotonous work, such as the preparation, cleaning, and weighing of approximately 2000 resin columns—a task that spanned weeks. However, it was always a joy working together. Henk, thanks a lot for all your efforts!

I also would like to thank the organizations and their representatives who not only initiated this research but also made our ambitious field experiment a reality. Many thanks to Erwin All and Leon Hahn of State Forestry, Leontien Krul of National Park Hoge Veluwe, Arno Willems of Kroondomein het Loo, David Borgman of Borgman consultancy, Erik van de Staak of Staro Nature and Countryside, Gerard Koopmans of Bosgroep Midden Nederland and Johan Blom of Blom Ecology. Through your support, both in terms of

Acknowledgements

in-kind contributions – granting us permission to work in their forests and offering support during the experiment - and cash contributions, we were able to conduct the scientific work. I extend my gratitude to State Forestry, which generously provided 10 of the experimental sites across various management units. I also wish to thank the different management units for accommodating this experiment, recognizing that it created additional communication and coordination efforts. To Kroondomein het Loo, National Park de Hoge Veluwe, and Staro Nature and Countryside, I am thankful for providing the remaining 5 experimental sites, a crucial contribution to the success of this PhD research. I also want to recognize the support provided by Borgman consultancy, as Nico Spliethof assisted with the installation of the plots and the measurements of the GPS coordinates. Their expertise and techniques were valuable in reliably outlining the plots even in densely regenerating areas. Gerard Koopmans and Erik van de Staak played a crucial role in organizing the timber harvest and handling of the associated challenges, all within a stringent deadline. Lastly, I want to express my gratitude to Blom Ecology for their generous funding, which facilitated the development of the deposition method and subsequent deposition sampling. Thanks a lot!

I further extend my gratitude to the multitude of individuals who lent their expertise and support throughout this remarkable PhD journey. Your contributions during the installation of the experiment, field sampling and laboratory sample processing were very valuable. My sincere thanks to Leo Goudzwaard, whose extensive experience was very valuable during the installation of the experiment and the field sampling. Leo, you were the first to extend practical support and continued to do so from the preparation of the fieldwork to the sampling of the trial trees and the monthly field sampling in which you were often accompanied by Ellen Wilderink. A special acknowledgement goes to Martin Valtera who carried out the pre-harvest soil sampling under huge time pressure what made it necessary to work even in challenging weather conditions. Looking back, I realize that the task was monumental, yet you managed to complete the entire soil sampling before the harvest. Your dedication was exemplary, and I am profoundly grateful. A sincere thank you to the forest managers who assisted with the pre-harvest marking of trees. Also, many thanks to all the Unifarm people for their help, John van de Lippe for the flexibility to provide manpower, but also Wim van de Slikke, Dafydd Timmermans, Gerrit Huisman, Berry Onderstal (providing all the drone images of the forests!), Ed and many more – your flexibility, assistance, and conviviality made the (field)work not just productive but also enjoyable.

Special recognition is due to Dieke de Boer, the first MSc thesis student within this research project. Dieke, your contributions during the busiest phase of the fieldwork, the trial tree sampling, were invaluable. Your commitment to data quality, even under immense pressure, played a great role in the successful determination of the biomass distributions and biomass stocks of the forests. I also want to acknowledge the dedication of all the other MSc students: Walter Baas and Assif Friedman for their help with post-harvest soil sampling and their study on the effects of harvest on enzyme activity, Daniël van de

Gaal for his assistance with post-harvest soil sampling, Loes Kampherbeek for her work on regeneration, and Flora van Eupen for her work on organic soil stocks and leaching. Additionally, I want to express my thanks to BSc students Paul van Duinhoven and Marit Derks for measuring the annual rings of Douglas fir and Scots pine, Ruben Baan Hofman for his work on regeneration and Sander Verhage for his work on wildlife pressure in the different plots.

Furthermore, I want to thank the people of the CBLB laboratory for their cooperation and assistance. For this PhD, you analyzed more than 8000 samples, a huge task that was further complicated by the challenging circumstances brought about by the COVID-19 pandemic. Furthermore, our samples were not always straightforward; we pushed the boundaries, aiming to measure beyond and far above detection limits, often contending with minute sample quantities. Thanks for pulling it off!

I want to thank my husband, family and friends. Bart, you encouraged me and supported me from the moment we met. You were there for me in the most difficult time when the pressure to submit this thesis was enormous. Mom and dad, there are no words to express my gratitude. I'm grateful for how you raised me, providing a safe home with love. Many thanks for the freedom you provided me when I choose my studies and career path although I choose a direction that was not conventional. Also, many thanks for how you raised me, you taught me to be responsible, to not give up despite when things are sometimes not easy. Thanks to my brothers and sisters and their families, even though my research was often vague for you and the related work unimaginable, you provided a very welcome distraction from my work and relativism as you showed that family, rather than work, is the most important thing in life. To my friends, you all enrich my life and without you I would not have been the person I am today. Diliaan, thanks for the distraction you provided and all our endless games of Colonist of Catan. Thanks for providing a listening ear when I was completely fed up due to unforeseen setbacks or when I was enthusiastic because of nice results! Richard Sikkema, we started as colleagues around the same time at the FEM group. You were always interested which was refreshing as I sometimes felt as the odd one out with my Dutch oriented research. Many thanks for the numerous times you helped out with field sampling and many thanks for all the coffee meetings. Now at the end of this PhD trajectory I consider you not only as an esteemed former colleague but also as a friend.

Finally, I acknowledge that completing this thesis would not have been possible without the support of God Almighty, who granted me strength, perseverance, and reason. Soli Deo Gloria!

Short Biography



Marleen Vos was born on the 3rd of July 1993 in Nederhemert (Gelderland), The Netherlands. From 2010 to 2011, she studied Applied Biology at CAH University of Applied Sciences (nowadays the AERUS university of applied sciences) in Almere. In 2011 she continued the study Applied Biology at HAS university of Applied sciences in 's Hertogenbosch, the Netherlands from which she graduated in 2014. She enrolled in the master program of Forest and Nature conservation at Wageningen University in 2014 where she specialized in the direction of Ecology and followed a minor in spatial, statistical and hydrological

modelling. During this master program she studied tree anatomy to investigate hydraulic bottlenecks in tree branches to understand mechanisms that exist in trees which could prevent tree death due to severe droughts. This research was carried out in the long-term irrigation experiment Pfywald, Switzerland, and was a collaboration between Wageningen University and the Swiss Federal Institute for Forest, Snow and Landscape Research WSL in Birmensdorf, Switzerland. During her master study, especially during the master thesis and internship, she became fascinated with all aspects of both forest ecology and forest management and with the impacts of scientific research on forest management practices. She finished her master's in 2016 and started to work as an ecological consultant at Blom Ecologie in 2017. In the summer of 2017, she developed in close collaboration with Frank Sterck and Wim de Vries a PhD proposal for the NWO grant 'Horticulture and Starting Materials' which was honored on October 27th, 2017. She started her PhD in January 2018 at the Forest Ecology and Management group of Wageningen University under the supervision of Frank Sterck, Wim de Vries, Marcel Hoosbeek and Jan den Ouden. Alongside this PhD project, she remained to work as an ecological consultant. She carried out her PhD project to contribute to the development of scientific guidelines for sustainable biomass harvest in which she focused on the dynamic period after harvest. Her work offers novel insights in the effects of tree harvest on the nutrient balance via altered nutrient gains in deposition and altered nutrient losses via leaching.

List of publications

Sikkema, R., Thomassen, E. A. H., **Vos, M.A.E.**, Bouwman, M., Limpens, J., & Nabuurs, G. J. (2020). Nederlandse boseigenaar ervaart gevolgen klimaatverandering en pleit voor steun. *Vakblad Natuur Bos Landschap*, 169, 18-23.

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Vos, M.A.E., de Boer, D., de Vries, W., den Ouden, J., & Sterck, F.J. (2023). Aboveground carbon and nutrient distributions are hardly associated with canopy position for trees in temperate forests on poor and acidified sandy soils. *Forest Ecology and Management*, 529, 120731.

Vos, M.A.E., den Ouden, J., Hoosbeek, M., Valtera, M., de Vries, W., & Sterck, F. (2023). The sustainability of timber and biomass harvest in perspective of forest nutrient uptake and nutrient stocks. *Forest Ecology and Management*, 530, 120791.

Submitted

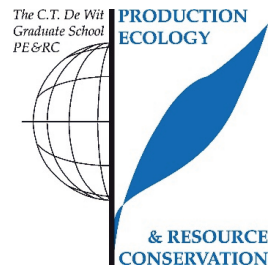
Vos, M.A.E., de Vries, W., den Ouden, J. & Sterck, F.J. (submitted). Canopy openness rather than tree species determine seasonal and annual atmospheric deposition into forests.

Vos, M.A.E., de Vries, W., Veen, G.F., Hoosbeek, M.R., & Sterck, F.J. (submitted). Testing Ion Exchange Resin for quantifying bulk and throughfall deposition of macro and micro-elements on forests.

van der Woude, S., Reiche, J., Sterck, F., Nabuurs, G.J., **Vos, M.** & Herold, M. (submitted). Sensitivity of Sentinel-1 Backscatter to Management-related Disturbances in Temperate Forests.

PE&RC Training and Education Statement

With the training and education activities listed below the PhD candidate has complied with the requirements set by the C.T. de Wit Graduate School for Production Ecology and Resource Conservation (PE&RC) which comprises of a minimum total of 32 ECTS (= 22 weeks of activities)



Review/project proposal (6 ECTS)

- Ecological boundaries for sustainable biomass harvest in production forests

Post-graduate courses (7.5 ECTS)

- Conflicting demands in European forests, a wicked problem; PE&RC (2018)
- Structural equation modelling; PE&RC (2020)
- Forest management across Europe; PE&RC (2021)

Invited review of journal manuscripts (1 ECTS)

- Agricultural and forest meteorology: forest inventory, climate extremes, net primary production and tree growth (2022)

Competence, skills and career-oriented activities (3.2 ECTS)

- Competence assessment; WGS (2018)
- Supervising BSc and MSc thesis students; WGS (2019)
- Reviewing a scientific paper; WGS (2021)
- Scientific writing; WGS (2022)

Scientific integrity/ethics in science activities (0.3 ECTS)

- Scientific integrity; WGS (2018)

PE&RC Annual meetings, seminars and PE&RC weekend/retreat (1.5 ECTS)

- PE&RC Day (2019)
- PE&RC Midterm weekend (2020)
- PE&RC Last year retreat (2022)

Discussion groups/local seminars or scientific meetings (7.5 ECTS)

- FEM Journal club (2018)
- Project presentation Unifarm (2018)
- FEM Writing club (2023)
- Beheerdersdag (2018, 2022)
- Aardhuis symposium (2023)
- Life-resilience symposium (2023)
- Netherlands annual ecology meeting (2018)
- Studiedag bosbodemon ontwikkeling (2019)
- FEM R club (2020)
- Klankbordgroep nutriëntenbalans (2022)
- Annual meeting with project partners (2018-2022)

International symposia, workshops and conferences (3.2 ECTS)

- 9th ICP Forest scientific conference; Zürich (2021)
- Workshop Gent University; Gent & Hoog Buurlo (2023)

Societally relevant exposure (0.4 ECTS)

- Participation in the TV program Kennis van nu (2020)
- Two publications in vakblad natuur en bos (2020, 2022)

Lecturing/supervision of practicals/tutorials (9.6 ECTS)

- Resource dynamics (2018-2021)
- Ecologie (2021)

BSc/MSc thesis supervision (6 ECTS)

- Nutrient content in Douglas fir and Scots pine on soils affected by nitrogen deposition
- The effect of forest management on natural regeneration density on poor, sandy soils in the Netherlands
- Diversity of tree species regeneration two years after various harvest treatments in The Netherlands
- The effect of forest management on Extracellular Enzyme Activity as an indicator for nutrient availability in poor sandy soils
- Mineralization of Organic Matter in Forest Soils
- Analysis of Douglas fir growth rings on Dutch sandy soils
- Annual growth ring analysis of Scots pine trees on Dutch sandy soils

Training and Education Statement

- Preventie verstoring depositie en uitspoeling metingen door grofwild
- Forest management strategies and their influence on leaching in Dutch sandy forests
- The impact of forest management on nutrient stocks

The research described in this thesis was financially supported by NWO Science Domain (NWO-ENW) project ALWGS.2017.004, which is financed by the Dutch Research Council (NWO).

Financial support from Wageningen University for printing this thesis is gratefully acknowledged.

Cover design

Edwin Seda

Photography

Marleen Vos

Printed by

Proefschriftmaken.nl

