Dissertationes Forestales 293

Nitrogen cycling from the perspective of boreal Scots pine trees

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Academic dissertation

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ABSTRACT

Nitrogen (N) and associated carbon (C) cycling were studied in an N-limited boreal Scots pine (*Pinus sylvestris* L.) forest in Hyytiälä, southern Finland and were compared to two N-rich temperate forests, the Speulderbos Douglas fir (*Pseudotsuga menziesii* (Mirb.) Franco) forest in the Netherlands and the Sorø European beech (*Fagus sylvatica* L.) forest in Denmark.

Nitrogen and carbon cycling in the Scots pine forest were modelled. These results were compared to continuous year-round observations to obtain an overall understanding of nutrient cycling in the forest. The N balance of the Scots pine forest was calculated based on direct measurements, measurement-based estimations and model results. Nitrogen uptake and resorption by trees were estimated based on continuous measurements. Litter fall dynamics of the Scots pine and Douglas fir forest were compared. Scots pine needle N dynamics were compared between the three forests.

Soil was the main N storage in the boreal Scots pine forest and most of this N was in recalcitrant form. Scots pine trees were very efficient at saving and recycling N. This together with atmospheric N deposition, potential N uptake by the canopy and organic N uptake mean that the importance of mineralization as the process driving N cycling may have been overestimated.

Most of the N was allocated simply to replace dead tissue in the Scots pine forest. This means that the additional N received via N deposition may significantly increase the N pool size that trees have for extending their biomass N (net growth). Because Scots pine trees were found to be dependent on efficient N use and recycling, this adversely also means that even slight snow and storm damages may cause foliar biomass to decrease due to reduced relocation on top of the direct effect of losing the foliage due to damage, affecting forest carbon sink strength.

Keywords: nitrogen balance, resorption, litter fall, nitrogen deposition, premature abscission.

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LIST OF ORIGINAL ARTICLES

The thesis is based on the following four articles, which have been published in peer-reviewed journals. The articles are referred to in the text by their Roman numerals:

I. Grote R., Korhonen J. F. J. and Mammarella I. Challenges for evaluating process-based models of gas exchange at forest sites with fetches of various species. Forest Systems, 20(3), 389-406, 2011. https://doi.org/10.5424/fs/20112003-11084.

II. Korhonen, J. F. J., Pihlatie, M., Pumpanen, J., Aaltonen H., Hari P., Levula J., Kieloaho A.J., Nikinmaa E., Vesala T., Ilvesniemi H. Nitrogen balance of a boreal Scots pine forest. Biogeosciences, 10, 1083–1095, 2013. https://doi.org/10.5194/bg-10-1083-2013.

III. Portillo-Estrada M., Korhonen J. F. J., Pihlatie M., Pumpanen J., Frumau A., Morillas L., Tosens T., Niinemets Ü. Annual and interannual variations in soil C and N input from canopy litterfall in two European coniferous forests. Annals of Forest Science, 70 (4), 367-379, 2013. https://doi.org/10.1007/s13595-013-0273-0.

IV. Wang L., Ibrom A., Korhonen J. F. J., Frumau A., Wu J., Pihlatie M., Schjoerring, J. K. Interactions between foliar nitrogen status and leaf longevity in relation to N cycling in three contrasting European forest canopies. Biogeosciences, 10, 999-1011, 2013. https://doi.org/10.5194/bg-10-999-2013.

AUTHOR’S CONTRIBUTION

Janne Korhonen alone was responsible for writing the introduction to the thesis, was the main author in paper **II** and participated in writing manuscripts **I**, **III** and **IV**. Janne Korhonen was responsible for data analysis in paper **II** and participated in data analysis for papers **I**, **III** and **IV**. Janne Korhonen participated in planning the research for all the manuscripts. Janne Korhonen participated in the measurements for papers **II**, **III**, and **IV**. In paper **I**, Janne Korhonen participated in the model initialization, development and interpretation of the results.

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LIST OF SYMBOLS AND ABBREVIATIONS

anammox	anaerobic ammonium reduction
C	carbon
CO_2	carbon dioxide
Γ	ratio between bulk foliar concentrations of NH_4^+ and H^+
GHG	greenhouse gas
GPP	gross primary production
N	nitrogen
N_2	dinitrogen, nitrogen gas
N_2O	nitrous oxide
NEE	net ecosystem exchange
NH_4^+	ammonium
NH_3	ammonia
NO	nitric oxide (nitrogen oxide)
NO_2	nitrogen dioxide
NO_2^-	nitrite
NO_3^-	nitrate
NO_x	NO and NO_2
Nr	reactive nitrogen
Pg	petagram (10^{12} kg, 10^9 t, 10^{15} g)
TER	total ecosystem respiration

1. GLOBAL AND REGIONAL PERSPECTIVE

1.1. Reactive nitrogen in the environment

Nitrogen (N) pollution, originating mostly from N fertilization and combustion processes, may have both beneficial and harmful effects on ecosystems. For example, forest productivity is typically limited by low N availability, whereas N pollution increases eutrophication and decreases biodiversity. Nitrogen cycling is linked to carbon (C) cycling and N pollution therefore affects the ability of forests to act as sinks for atmospheric C. Forests absorb and bind N pollution and stop it from spreading and polluting aquatic systems such as lakes, the Baltic Sea or groundwater.

1.1.1. Dinitrogen and reactive nitrogen

Approximately 78% of the atmospheric dry air volume is N. This equals ca. 3 900 000 Pg N (3.9×10^{18} kg N). Lithosphere N storage is approximately half of that, i.e. 2 000 000 Pg N (2×10^{18} kg N). Of this, the N level in the Earth's soils is 133–140 Pg N for the upper 1-m layer (pedosphere) (Batjes 2014). Based on the studies of Bar-On et al. (2018) and Elser et al. (2000), the estimated N level in biomass is very roughly 20 Pg N, roughly 80% of which is in terrestrial plants, ca. 20% in microbes and 2% in animals.

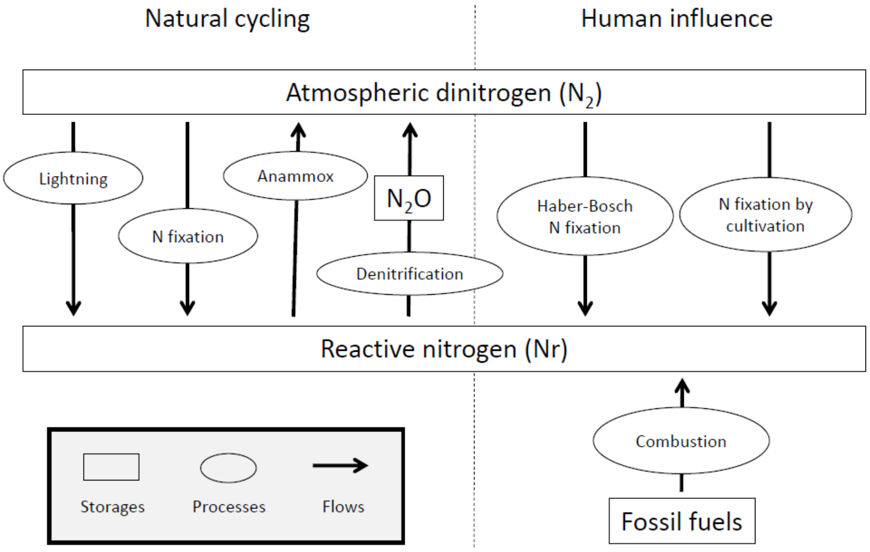


Figure 1. Schematic figure of the main processes contributing to global nitrogen (N) cycling. The dashed line divides the processes into natural and anthropogenic parts. Anammox and denitrification are both natural processes affected by anthropogenic reactive nitrogen (Nr) increase. Anammox = anaerobic ammonium oxidation. Both nitrous oxide (N_2O) and dinitrogen (N_2) are produced during denitrification.

Nearly all atmospheric N is gaseous dinitrogen (N_2). The dinitrogen molecule has a covalent triple bond between the two N atoms, meaning it is extremely inert (non-reactive). For this reason, plants or other living organisms cannot directly utilize it. The vast majority of lithosphere N cannot be used by living plants because it is physically inaccessible.

All other N besides N_2 is called reactive nitrogen (Nr; Sutton et al. 2011). These N compounds are typically gaseous or solid at room temperature and some are water-soluble. The reactivity of these substances varies from almost non-reactive to extremely reactive. Reactive N is essential to all known life; it has an important role in atmospheric chemistry and in many cases, the availability of N largely determines the fertility of an ecosystem at a regional scale.

1.1.2. Formation and removal of reactive nitrogen

Global N cycling is briefly described in Figure 1. The main natural pathways for transforming N_2 into Nr are N_2 fixation (transformation of N_2 to NH_4^+) by N-fixing organisms and by N_2 molecules in the atmosphere being broken down by lightning. Global natural N_2 fixation is ca. 0.11 Pg N yr^{-1} and 0.12 Pg N yr^{-1} for terrestrial and marine ecosystems, respectively, and lightning produces approximately 0.005 Pg N yr^{-1} of nitrogen oxides (NO_x) (Schumann and Huntrieser 2007). This Nr goes into the active cycle and occurs in several processes in the atmosphere, and in terrestrial and aquatic ecosystems.

Reactive N mainly exits from the active cycle either via N_2 transformation during denitrification or anaerobic ammonium oxidation (anammox) or through physical isolation by sedimentation processes. In denitrification, both N_2 and nitrous oxide (N_2O) are produced, and N_2O is also produced by other processes (Otte et al. 2019). Nitrous oxide is a strong greenhouse gas and has an atmospheric lifetime of approximately 120 years (Prather et al. 2015). It is mainly transformed into N_2 in the stratosphere by photolysis and oxidative processes (Montzka et al. 2011). Natural N_2 formation is approximately 0.4 Pg N yr^{-1}, 0.1 Pg N yr^{-1} of which originates from terrestrial ecosystems. Sedimentation in oceans is estimated to be ca. 0.025 Pg N yr^{-1} (Canfield et al. 2010).

The Nr level in active cycling is increasing because of industrial N_2 fixation, fossil fuel burning and biological N_2 fixation associated with land cultivation. Industrial N_2 fixation, fossil fuel combustion and increased N_2 fixation by cultivated land are approximately 0.1 Pg N yr^{-1}, 0.025 Pg N yr^{-1} and 0.035 Pg N yr^{-1}, respectively (Gruber and Galloway 2008). Nitrogen released during bedrock weathering equals ca. 0.025 Pg N yr^{-1} (Houlton et al. 2018).

In industrial N fixing, N_2 is transformed into Nr, which may be used as a plant fertilizer or in industrial processes. The process was first successfully conducted in the laboratory in the early 20th century by Fritz Haber and a few years later was developed into an industrial process by a team lead by Carl Bosch. This process is known as the Haber-Bosch process (Erisman et al. 2008). In fossil fuel combustion, Nr is released from fossil storages into active cycling in the atmosphere and in terrestrial and aquatic ecosystems. Legume production for fodder is the most important source of Nr related to land cultivation (Smil 1999).

1.1.3. The grand challenge of excess nitrogen

Industrial N fixing (Haber-Bosch process) is needed for producing N fertilizers. Approximately half of the human population is estimated to be fed by food produced using

N fertilizers based on industrial N fixing and the global population would be significantly smaller without them (Smil 2002; Stewart et al. 2005; Erisman et al. 2008). Supporting human life on Earth is therefore currently dependent of producing new Nr.

A major portion of the Nr used as a fertilizer is not taken up by plants and is lost to the atmosphere mostly as ammonia gas (NH_3) or to aquatic systems mainly as nitrate (NO_3^-) (Galloway and Cowling 2002). Energy production is another anthropogenic source of Nr in the environment and it is currently largely based on the combustion of fossil fuels containing N. In our study, this Nr is called excess Nr.

Reactive N spreads via the atmosphere and is deposited into vegetation, soils, aquatic ecosystems etc. typically within some hundreds of kilometres from the source, but may also be transported thousands of kilometres (Sanderson et al. 2008). It also spreads via water flows within terrestrial ecosystems, from terrestrial ecosystems to aquatic ecosystems and within aquatic ecosystems. Reactive N in terrestrial or aquatic ecosystems may also be lost back to the atmosphere in gaseous forms. In the atmosphere, Nr may react and alter atmospheric chemistry. From the atmosphere, Nr is eventually deposited back to aquatic or terrestrial ecosystems, where it once again occurs in many processes. This phenomenon is called the nitrogen cascade (Galloway et al. 2003).

Excess Nr causes several environmental problems (Fig 2). Nitrous oxide in the troposphere is a very strong and stable greenhouse gas (GHG). In the stratosphere, it breaks down into nitrogen oxides (NO_x; NO and NO_2), which participate in ozone (O_3) depletion. Nitrous oxide has become the most important O_3-depleting emission (Ravishankara et al. 2009). Nitrogen oxides are poisonous gases that participate in many chemical processes in the atmosphere. Unlike in the stratosphere, NO_x in the troposphere are precursors for O_3 (Fishman and Crutzen 1978). NO_x also affects secondary aerosol formation in the atmosphere. In terrestrial and aquatic ecosystems, Nr causes eutrophication, which leads to biodiversity loss and anoxia. For example, anoxia is a major challenge in the Baltic Sea. Ammonium (NH_4^+) causes acidification of soils in heavily fertilized agricultural areas, while NO_3^- pollutes groundwaters. For example, the use of groundwater as drinking water is restricted in parts of Central Europe because it is polluted by NO_3^- (Sutton et al. 2011).

Reducing the Nr level in the environment is difficult because it is spread throughout ecosystems. Two major processes, i.e. denitrification and anammox, transform Nr into N_2 (Stein and Klotz 2016). In the denitrification process, a major fraction of the Nr is transformed into N_2O, an extremely strong and stable GHG (see chapter 2.5.1). Therefore, favouring environmental conditions in which denitrification occur is not a sustainable solution. Anammox is an important process transforming Nr to N_2 in marine ecosystems (van de Vossenberg et al. 2013) and it is widely used to remove N from wastewaters. Nitrous oxide is not produced in this process.

1.2. Forests

Forests cover 31% of the global land area, which is divided into boreal (31%), temperate (17%), subtropical (8%) and tropical forests (44%) of the global forest area. The area is decreasing at an approximate annual rate of 0.1% (Keenan et al. 2015).

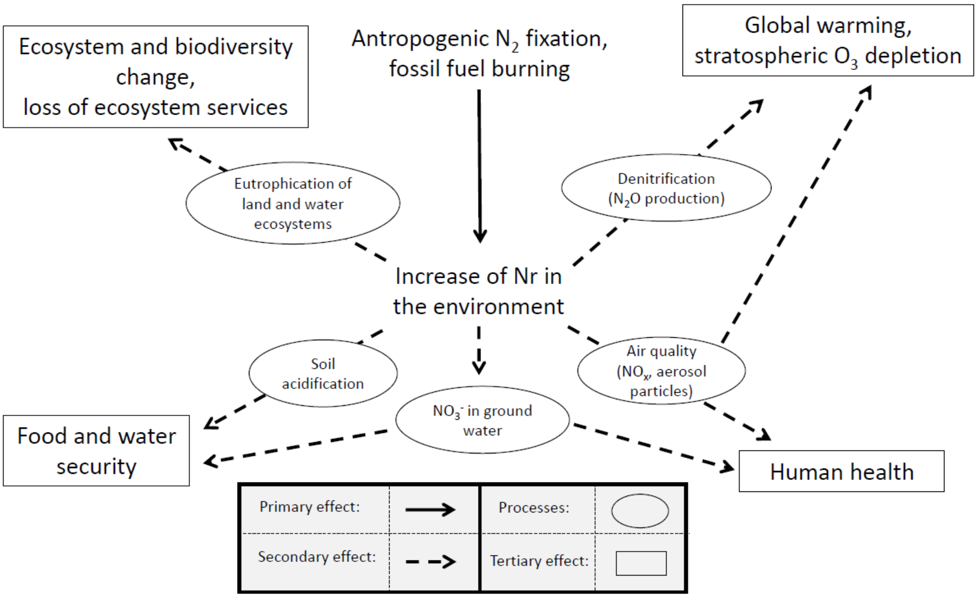


Figure 2. The major environmental effects of anthropogenic reactive nitrogen (N_r) increase at a global scale.

In a forest ecosystem, trees alter the microclimate such as temperature, light, wind and air humidity. Trees and undergrowth plants also strongly alter the physical and chemical properties of soil. They fix atmospheric C into solid plant matter, which is then deposited in the soil as litter. This litter is decomposed to form soil organic matter. After a disturbance, such as a forest fire or clear cutting, new forest is grown by so-called successive tree species such as Scots pine or birches. These trees may alter the environment so strongly that the successive species may be unable to regenerate at later stages of forest succession. In other words, they render their growing environment unsuitable for their regrowth, and other tree species, such as spruces, take over the role of dominant species.

1.2.1. Boreal and temperate forests

Boreal forests are forests in the Northern Hemisphere in a boreal (subarctic) climate, between the tundra and temperate ecosystems. Boreal forests roughly cover Fennoscandia (Norway, Sweden, Finland, the Kola Peninsula and Karelia), the majority of Russia and a large part of Canada. The boreal forest is mostly characterized by coniferous trees, which often, but not always, are evergreen. Scots pine is the most dominant species in Fennoscandia. Boreal forests generally receive relatively small amounts of atmospheric N deposition (Jia et al. 2016). The climate in the boreal forest region varies but is typically characterized by a humid climate with distinct dormant and growing seasons. Based on the Köppen-Geiger climate classification, the average temperature during the coldest month in the boreal climate is 0 °C or lower and the average temperatures of the warmest one to three months are +10 °C or higher. Apart for the southern coast, Finland is completely located within the boreal climate

(Southern coast is Dfb, and the rest is Dfc in Köpper-Geiger classification; Peel et al. 2007).

Temperate forests grow in the temperate zone, between the boreal and subtropical zones. Temperate forests may be broadleaf, coniferous, mixed or tropical. The temperate forests in West and Central Europe are typically broadleaf or mixed forests. According to the Köppen-Geiger climate classification, the average temperature of the coldest month in the temperate climate is between 0 °C and 18 °C and the average temperature of the warmest month is over 10 °C, (Peel et al. 2007). The average temperature of the coldest month in the oceanic (temperate) climate is over 0 °C (Cfb, Cfc, Cwb and Cwc in the Köppen-Geiger classification), while the average temperature of the warmest month is below 22 °C (Peel et al. 2007).

Boreal and temperate forests store 270 Pg C and 120 Pg C, respectively, and sequester C from the atmosphere at rates of 0.5 Pg C yr^{-1} and 0.7 Pg C yr^{-1}, respectively (Pan et al. 2011).

1.2.2. The importance of forests

Forests control the global climate by regulating atmospheric CO_2 concentrations (Pan et al. 2011) and regional and local climates by affecting the Earth’s surface reflectance (albedo; Betts and Ball 1997; Kuusinen 2014) and atmospheric composition, for example via volatile organic compound (VOC) emissions (Kulmala et al. 2004). Forests absorb and buffer pollutants (pollution retention), sequester atmospheric C and Nr, transform them into tree biomass and less harmful substances, and prevent them and organic particles from being transported into aquatic ecosystems such as lakes and the Baltic Sea.

Aside from being beneficial for the climate and aquatic systems, forests provide other ecosystem services. They act as sources of wood, berries, mushrooms and game. Forests are important because of their cultural, recreational and aesthetic values. They are also important for retaining biodiversity. Environmental change, most importantly global warming and increased N deposition, alter the functioning of boreal forests and inevitably also the ability of forests to provide these ecosystem services.

1.3. Plant nutrients

Plants benefit from several chemical elements. Elements essential for normal plant functioning are called plant nutrients. Plant nutrients are typically divided into macronutrients and micronutrients based on their abundance in plants. Nitrogen, phosphorus (P), potassium (K), calcium (Ca), sulfur (S), magnesium (Mg), C, oxygen (O) and hydrogen (H) make up the macronutrients, while iron (Fe), boron (B), chlorine (Cl), manganese (Mn), zinc (Zn), copper (Cu), molybdenum (Mo) and nickel (Ni) make up the micronutrients. Approximately 50% of the dry plant mass is C. The N content of foliar litter varies depending on species and environment. For boreal and temperate tree species, N content is typically ca.0.8–2.5% of the dry mass, being generally higher in broadleaved species than evergreen coniferous species. Concentrations of P, S, K and Ca are typically approximately 0.1% of the leaf dry mass (Berg and McClaugherty 2003).

1.3.1. Origin of plant nutrients

Plants get C from atmospheric carbon dioxide (CO_2) in photosynthesis. Photosynthesis is also responsible for plants getting O and H from water. Water circulates through the atmosphere, which may therefore be seen as the source of water. Plants take N from the soil, but in most cases this N also originates from the atmosphere, only in centurial or millennial time scales. Other nutrients are mostly released from the mineral soil and bedrock through weathering. Nitrogen may also originate from bedrock, but this is not particularly significant in Finland or Sweden (Morford et al. 2011; Houlton et al. 2018).

1.3.2. Nutrient demand of plants

The fractions in which plants require nutrients depend mainly on the nutrient concentrations of the tissue being grown by the plant is growing. However, a major fraction of certain nutrients, such as K, are not fixed within the tissue but are used for example to regulate the osmotic potential of fluids inside the plant. Some nutrients are lost to the atmosphere in gaseous forms, for example as VOCs, although generally in small amounts.

Ecological stoichiometry studies the relation between how much nutrients plants take up and how much they require. The fraction in which nutrients are needed is almost always different than the fraction that is available. This means that one nutrient is usually more scarcely available than the others, and the availability of this nutrient limits the usability of nearly all the other nutrients. This limiting nutrient is called the minimum nutrient. The concept is known as the Sprengel-Liebig Law of the Minimum (van der Ploeg et al. 1999).

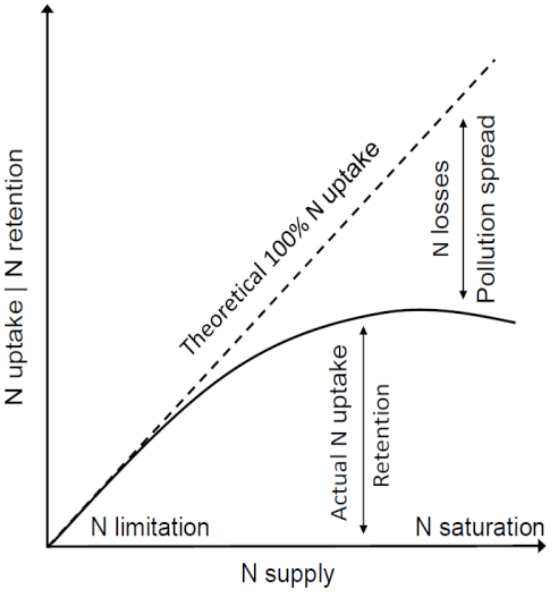


Figure 3. Conceptual image of nitrogen (N) retention in forest ecosystems as a function of N supply such as atmospheric N deposition and mineralization. Nitrogen-limited forest ecosystems act as efficient sinks for N, preventing the spread of N into e.g. aquatic systems. Retention capacity depends on the ability of plants and microbes to take up and utilize N. The same principle applies for crop farming, in which case the N supply refers to N fertilization.

1.3.3. Nitrogen availability

Nitrogen availability mainly depends on the history and current inputs and outputs of N. Nitrogen is considered the most common minimum nutrient (Vitousek and Howarth 1991; LeBauer and Treseder 2008), meaning that the vitality of forests, especially boreal forests, are limited by low N availability (Hyvönen et al. 2008). In these ecosystems, plants and microbes take up and use available N efficiently. These ecosystems are called *nitrogen limited*. Increased N availability in these ecosystems means increased biomass production.

An excess of N may be harmful for trees and the entire ecosystem, for example because excess NH_4^+ is toxic to plants (Gerendas et al. 1997; Pan et al. 2016). These ecosystems also lose their capacity to buffer Nr from spreading further in the atmosphere and aquatic ecosystems. Ecosystems in a state of excess N are called nitrogen saturated. Nitrogen saturation has several definitions and criteria, including a lack of growth in response to additional N, increased N losses from the ecosystem and the equivalence of N inputs and N losses (Aber 1992; De Schrijver et al. 2008). The concepts of N limitation and N saturation are illustrated in Figure 3. Increased N losses from the ecosystem mean N_2O and NO_x emissions to the atmosphere and NO_3^- leaching to the groundwater and other aquatic ecosystems (Aber 1992). It is noteworthy that these ecosystems may still be net sinks for N; however, their ability to buffer and clean pollution is reduced.

Boreal forests are typically deprived of N, whereas many forests, notably temperate forests in Western and Central Europe and subtropical forests in the eastern U.S. and eastern China receive N in excess quantities (Jia et al. 2016). Although N as such is typically abundant in forest soils, its availability heavily depends on the physical and chemical properties of the compound it is bound to, on the physical and chemical environment and on the activity of the structures responsible for decomposition and nutrient uptake. Nitrogen stored in the foliage is referred to as active and N stored in other tissue, such as wood, is referred to as structural N.

2. NUTRIENT CYCLING AND ENERGY FLOWS IN FORESTS

The main C and N cycling processes are shown in Figure 4. Most N processes are described separately, but C and N cycling are described simultaneously in the case of litter fall and decomposition (chapters 2.2 and 2.3). Energy does not cycle in forest ecosystems, as energy flows are unidirectional, but energy flows and C cycling are treated together in chapter 2.1.

2.1. Energy flows in forests

2.1.1. Photosynthesis

The uptake of energy and C to the forest ecosystem happens simultaneously via photosynthesis. This makes C a special nutrient in addition to being an important building material for living organisms. When energy is used, C is lost to the atmosphere.

In photosynthesis, solar energy is used to detach C atoms from a CO_2 molecule and this C together with water (H_2O) is used to form sugars. The energy capture occurs in the chloroplasts, where solar energy (light) is absorbed by chlorophyll. This energy is temporarily stored in NADPH by reducing nicotinamide adenine dinucleotide phosphate ($NADP^+$) and in adenosine triphosphate (ATP) converted from adenosine diphosphate (ADP). These processes are generally called the light reactions of photosynthesis. Atmospheric CO_2 is then fixed and used to bind this temporally stored energy to sugars, which are more stable compounds. This chain of processes is called the Calvin cycle or the dark reactions of photosynthesis. Carbon dioxide needed for photosynthesis is transported to the chloroplasts through stomata (singular stoma) via simple diffusion. In this diffusive process, water evaporates to the atmosphere through transpiration.

Gross primary productivity (GPP) is a measure of ecosystem photosynthetic production. The annual GPP of typical European forests is rather constant, generally ranging between 10 000 and 15 000 kg C ha^{-1} yr^{-1} (Valentini et al. 2000). This thesis covers three forests. The average GPPs of a boreal Scots pine forest in Hyytiälä, Finland, a temperate Douglas fir forest in Speulderbos, the Netherlands and a temperate European beech forest in Sorø, Denmark are 1000 kg C ha^{-1} yr^{-1} (Ilvesniemi et al. 2009), ca. 2 200 kg C ha^{-1} yr^{-1} (van Wijk et al. 2001) and 1900 kg C ha^{-1} yr^{-1} (Wu et al. 2013), respectively.

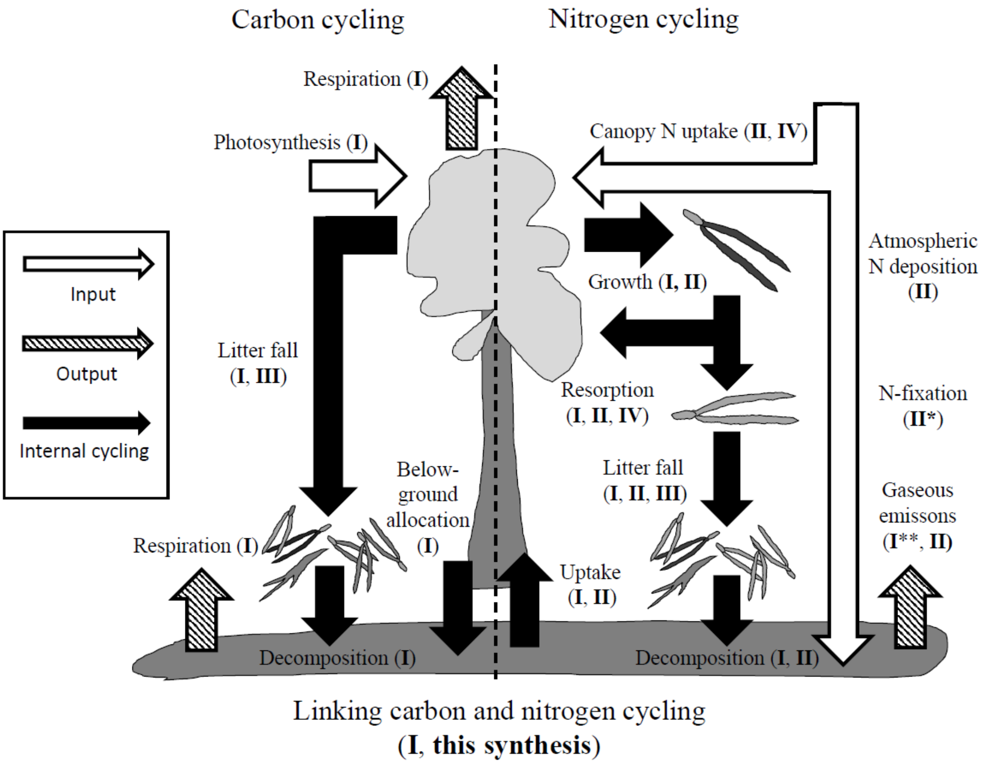


Figure 4. Key processes of carbon (C) and nitrogen (N) cycling in boreal forests. Roman numerals represent the articles in which the processes have been studied. *Only the range for N fixation was given, based on literature. **Gaseous N losses were also studied as part of the modelling work (**I**) but these results have not been published.

2.1.2. Respiration

The oxidation of substrates, typically sugars, is called respiration. Molecular oxygen (O_2) is usually required as an electron acceptor, but anaerobic respiration may also occur. The release of energy is the main purpose of respiration. In aerobic respiration, CO_2 is released to the atmosphere. In plants, respiration is usually divided into maintenance respiration and growth respiration. Maintenance respiration is vital for the survival of living cells and is highly temperature dependent. Growth respiration occurs when new plant tissue is built. It is typically limited by nutrient availability and is controlled for example by plant hormones and temperature.

Respiration is often divided into autotrophic and heterotrophic respiration, although sometimes the border between these two is unclear. Autotrophic respiration occurs in organisms that produce the substrate for respiration themselves. This includes plants in general. Photoautotrophs derive their energy from photosynthesis and mainly use sugars for respiration. Heterotrophic respiration is dependent on an external substrate source and decomposition and mineralization result from it.

Soil respiration consists of both autotrophic and heterotrophic soil respiration. Heterotrophic soil respiration is usually higher in boreal forests than autotrophic soil respiration, although autotrophic soil respiration may be as high as heterotrophic soil respiration during the growing season (Bond-Lamberty et al. 2004; Subke et al. 2006).

Total ecosystem respiration (TER) includes the maintenance and growth respiration of aboveground trees, respiration in the roots and rhizosphere, heterotrophic soil respiration, the respiration of ground vegetation etc. The difference between GPP and TER is called net ecosystem exchange (NEE). Total ecosystem respiration of the boreal Scots pine forest is approximately 800 kg C ha^{-1} yr^{-1} (Ilvesniemi et al. 2009), while being ca. 1400 kg C ha^{-1} yr^{-1} in the temperate Douglas fir forest (van Wijk et al. 2001) and an average 1600 kg C ha^{-1} yr^{-1} in the temperate European beech forest (Wu et al. 2013). In European forests, TER is typically ca. 80% of GPP (Janssens et al. 2001).

2.2. Senescence and litter fall

Living cells and organs have limited lifetimes and at a certain point they lose their efficiency and eventually stop functioning. Longevity is the measure of the average lifetime of living organs. Higher longevity means that trees need to annually allocate fewer resources, such as N, to the foliage. The ageing and dying of plant organs is called senescence. Senescence is the beginning of the stage in which nutrients turn from nutrient-cycling drivers into nutrients that are in the cycle. In senescence, nutrients may either be directly mobilized or are at the beginning of the decomposition process. Whether a nutrient is directly mobilizable mainly depends on the chemical structure it is bound to. Mobile nutrients, such as N, P and K may be at least partly directly mobilized and translocated to other parts of the organisms. For example, N may partly be taken back from dying leaves. This process is often referred to as resorption, relocation or retranslocation (Aerts 1996).

Senescence is typically a controlled process. In boreal forests, foliar senescence follows a clear annual pattern (**III**). This is observed as a high level of brown foliar litter shedding in the autumn. The brown colour is a consequence of plants breaking down chlorophyll and resorbing the nutrients from it.

Disturbances, such as storms and heavy snow events, lead to premature abscission (uncontrolled or premature litter fall) of plant organs. This means litter fall occurring without resorption. Whether these nutrients are lost from the ecosystem, whether they remain unavailable in the soil or whether they quickly become available for plant use depends on many factors.

2.3. Decomposition

Nutrients in forests are in constant cycling. Plant litter, such as leaves, branches and roots, along with dead organisms contain nutrients that are not directly in organism-available form. To be usable for organisms as building materials, these nutrients have to be released. The whole process of this breakdown is called decomposition, which includes breaking down both the physical and chemical structure of the litter. Soil fauna is mainly responsible for the breakdown of physical structures and soil mixing. With the help of extra-cellar enzymes (exoenzymes), microbes break down the chemical structure of litter. The process of making nutrients organism-available is called mineralization or mobilization. In chemistry, the breakdown process of the molecular structure of organic material closer to monomer or mineral forms is often referred to as depolymerization.

Decomposition rate is dependent on the availability of substrate for decomposition, chemical and physical quality of the substrate, oxygen availability, soil moisture, pH, the availability of free energy, faunal, microbiological and enzymatic activity and on other environmental factors. Not all nutrients in the litter are mobilized. A part may also be lost to the atmosphere as gases, remain immobilized or even become more recalcitrant. The decomposition rate varies from days to centuries or even longer time periods (Trumbore 2000; Karhu 2010).

2.4. Nitrogen balance of forests

Nitrogen balance considers the inputs and outputs of N to the ecosystem. When the inputs of N exceed the outputs, N accumulates to the forest and the forest acts as an N sink.

2.4.1. Nitrogen inputs to forests

Nitrogen in forest ecosystems originates from the atmosphere via atmospheric Nr deposition or N_2 fixation. The global pre-industrial level of N_2 deposition is estimated at ca. 1 kg N ha^{-1} (Bala et al. 2013). N_2 fixation in boreal forests is estimated to be in the order of magnitude of 2 kg N ha^{-1} (Wardle et al. 1997) and is not assumed to have changed substantially because of industrialization. At these rates, the N accumulation takes centuries to several millennia. Atmospheric N deposition to forests currently varies between 1 kg N ha^{-1} yr^{-1} (for example Lapland) and ca. 40 kg ha^{-1} yr^{-1} in Western Europe (Jia et al. 2016). N_2 fixation is usually assumed to be at pre-industrial levels or possibly even lower in N-saturated forests.

Dinitrogen fixation is a process in which atmospheric N_2 is transformed into organism-available NH_3. This is carried out by nitrogenase enzyme and certain microbes that are collectively called diazotrophs. This process requires high levels of energy, which is often obtained from photosynthesizing plants in the form of sugars. Diazotrohic microbes may live in symbiotic or less intense associative relationships with the host plant or microbes may be free-living with no clear relationship. The borderline between these classifications is often unclear and the terms ‘free-living’ and ‘associative’ are both used occasionally, mainly to separate them from a symbiotic relationship. In boreal forests, *Frankia* sp. are important N_2-fixing organisms, and they are associated with trees, especially alders (*Alnus* sp.), with which *Frankia* sp. form a symbiotic relationship with. In boreal forests, an associative relationship between cyanobacterium (*Nostoc* sp.) and the ubiquitous feather moss (*Pleurozium schreberi* (Brid.) Mitt.) reportedly fixes up to 2 kg N ha^{-1} yr^{-1} in northern Fennoscandian forests (DeLuca et al. 2002).

Bedrock may be a significant source of N (Morford et al. 2011). Modelling shows that weathering may create an input of over 10 kg N ha^{-1} yr^{-1}. Ecosystems in Europe are estimated to produce a couple of kg N ha^{-1} yr^{-1}, whereas no significant N input from bedrock is estimated in Finland and Sweden (Houlton et al. 2018).

Atmospheric deposition is typically divided into dry, wet and occult deposition, the last of which mainly has importance in mountainous regions. In dry deposition, aerosol particles or gas molecules are deposited to surfaces after hitting them. The rate of aerosol particle dry deposition is dependent on atmospheric particle concentrations and size distributions, windiness, and the physical structure, orientation, distribution and area of the surface. The rate of N deposition also depends on the N content of the aerosol particle, of which the soluble NH_4^+ and NO_3^- residing at the surface of the aerosol particle can be measured separately.

Nitrogen dry deposition of gases depends on chemical properties of the gases and surfaces, such as electric charges, surface wetness and gas concentration gradients of N-containing gases. Ammonium, nitrogen dioxide (NO_2), nitric acid (HNO_3) and nitrous acid (HNO_2 or HONO) are the most important molecules contributing to N dry deposition. Other N-containing molecules, such as organic molecules, e.g. peroxyacetyl nitrate (PAN), and amines, contribute to N deposition, but they are not typically quantified separately (Flechard et al. 2011; Ge et al. 2011; Wu et al. 2012).

In wet deposition, water droplets scavenge material from the atmosphere and transport it to the forest during precipitation. So-called bulk deposition is usually measured instead of wet deposition. It consists of wet deposition and some dry deposition as well (Korhonen et al. 2012). Nitrogen in wet deposition is typically measured as NO_3^-, NH_4^+ and total N.

Most of the N is deposited in the canopy. This is especially true for dry deposition. Part of the N absorbed by the canopy can be transported to the ground via throughfall. Throughfall is low during light rain events, and the canopy intercepts most precipitation. In this case, the canopy may also intercept wet N deposition efficiently. In heavier rain events, N may accumulate to the canopy or previously deposited N may be rinsed to the ground. At the SMEAR II research forest in Hyytiälä, Finland, canopy interception is approximately one-third of the total precipitation (Ilvesniemi et al. 2010).

2.4.2. Nitrogen outputs from a forest

Nitrogen naturally exits a forest ecosystem either via gaseous N losses or discharge. In both cases, the chemical composition of the N compound largely determines whether it will escape the ecosystem or not. The known mechanisms of N losses to the atmosphere are mostly related to the denitrification process.

Boreal forests are located in a humid climate, meaning that at an annual scale, excess water escapes the ecosystem as discharge. The escaping water contains nutrients either in molecular or particulate form and thus water flowing out from the system also transports nutrients. Podzolized soils of boreal forests are acidic, whereas soil mineral particles have a negative charge. This means that free cations in the soil solution are efficiently bound to soil particles, whereas anions are soluble to the soil liquids and are in danger of being transported from the system via runoff. Nitrogen anions are NO_2^- and NO_3^-, and they are produced in the nitrification process. Solid particles in water are another pathway of nutrient loss in discharge. These solid particles themselves may consist of nutrients or cations may reside on the surface of the particles.

2.5. Nitrogen cycling within a forest

2.5.1. Nitrification and denitrification

Nitrification and denitrification have important roles in soil N cycling and they also determine how much N is lost as gases to the atmosphere or in discharge. The general availability of mineral N affects how much nitrification and denitrification occurs, and the rate of these processes is used to determine whether an ecosystem is N limited or N saturated.

In decomposition, organic matter is oxidized, energy is released and CO_2 is produced. Normally during decomposition, O acts as the electron acceptor. In an anaerobic environment, including anaerobic microsites within aerobic soils, inorganic N compounds with N in a positive oxidation state may also act as electron acceptors. This chain of processes is called denitrification. In denitrification, the oxidation state of N decreases. In the first step, NO_3^- is reduced to NO_2^-, then to NO, next to N_2O and finally to N_2. The intermediate products NO and N_2O are gases that may easily escape the system to the atmosphere. Dinitrogen is lost to the atmosphere, as it is no longer Nr.

Nitrification is an aerobic process that occurs in soils. In the process, NH_4^+ or NH_3 is oxidized to nitrite (NO_2^-) or typically further to NO_3^- by either ammonia-oxidizing bacteria or archaea. Nitrification is an important process in agricultural fields and in many forest ecosystems. However, nitrification is reportedly small in boreal forests compared to the other processes of N cycling (Ambus et al. 2006). One suggested reason is the relatively high acidity of the soils (De Boer and Kowalchuk 2001).

2.5.2. Plant–microbe interactions in soil

Plants exude sugars, a source of C and energy, to soil microorganisms, mainly mycorrhizal fungi (*mychorrhiza*) and *rhizobacteria*. These mycorrhizal fungi and rhizobacteria have symbiotic relationships with plants. Often, this relationship is mutualistic, meaning that it is beneficial for both the plant and the microorganism. In exchange, plants obtain nutrients, such as N, P K from fungi. The symbiosis enhances the decomposition process, which is usually classified as heterotrophic respiration (Fontaine et al. 2007; Chen et al. 2014; Adamczyk et al. 2019). This is often referred to as the priming effect (Bingeman et al. 1953). Soil respiration that is directly dependent on sugars produced by plants is called root and rhizosphere respiration and it includes belowground autotrophic respiration, but depending on the definition, it may also include some heterotrophic respiration. The symbiosis with mychorriza and rhizobacteria helps plants overcome the reality that initially the fractions of available nutrients in the soil is not the same as the fraction of nutrients they need.

Mychorrhizal fungi may be divided into *endo-* and *ectomycorrhiza*. Endomycorrhiza have hyphae that grow into the cell membrane of plant roots, whereas in most cases the hyphae of ectomycorrhiza only surround the roots. The dominant tree species in Fennoscandian forests, i.e. Scots pine and Norway spruce (*Picea abies* (L.) H.Karst.), are known to form mutualistic symbiosis with ectomycorrhiza (Korkama et al. 2006; Pickles et al. 2010).

2.5.3. Nitrogen uptake by plants

Nitrogen may be taken up via the roots or leaves. The root pathway occurs in association with mycorrhiza as NH_4^+ or NO_3^- (Courty et al. 2015) or as organic N, such as with amino acids (Chapin et al. 1993; Näsholm et al. 1998; Persson et al. 2003; Näsholm et al. 2009). Certain plants prefer to take up NH_4^+, whereas others prefer NO_3^-. In N-limited systems, NH_4^+ uptake is more common. The advantage of NO_3^- is that it is easier to transport within the plant, the major disadvantage being that it needs to be reduced to NH_4^+ before it may be used in the synthesis of more complex molecules that are required by the plant (Marschner 1995, Marzluf 1997). The main advantages and disadvantages for NO_3^- are the opposite than for NH_4^+ uptake. In boreal forests, NH_4^+ is the main mineral form of N and boreal forest plants typically prefer NH_4^+ uptake.

Plants are known to take up nutrients, including N, via their leaves (Burkhardt et al. 2012). High NH_3 concentrations in N-polluted areas also means that plants may take up NH_3 gas directly via the foliage (**IV**). Organisms living on trees are called epibionts, or epiphytes when the organism is also a plant. Epibionts and epiphytes can also take N directly from the atmosphere (Elbert et al. 2012). It is also possible that NH_3 is lost to the atmosphere from the foliage, because NH_3 is produced in photorespiration.

Mobilized nutrients may be used as building blocks for plant tissue. The nutrients become immobilized in this process, meaning that they need to be mineralized before they can be re-used. At this stage, these nutrients work as functioning organisms driving nutrient cycling. Nitrogen has several roles in the life of plants. It is a constituent of proteins, chlorophyll, nucleic acids (DNA and RNA) and cell membranes. Proteins are the polymers of amino acids, which in turn are derivatives of amines, which in turn are derivatives of NH_3. One hundred g of NH_3 contains 82.4 g of N, whereas 100 g of proteins contain an average 16 g of N. All known enzymes are proteins, excluding certain RNA molecules.

2.5.4. Strategies to cope with low nitrogen availability

Trees have several mechanisms to cope with limited N availability. The Roman numerals indicate the papers included in this thesis that focused on these strategies:

1. Trees may affect how much N they bind in their active and structural tissues. For example, a relatively sparse canopy with low N concentration automatically means a relatively low N requirement but usually also lower carbohydrate production (**I**, **IV**).
2. Trees can increase the longevity of their tissue such as leaves. For example, evergreen coniferous trees do not need to regrow their entire foliage each year (**I**, **IV**).
3. Trees can recycle N by resorbing nutrients from dying tissue. This is especially important in the case of foliage (**II**, **IV**).

4. Trees can increase N uptake belowground by allocating resources to the roots and rhizosphere, especially carbohydrates produced in the canopy (**I**).
5. Trees can take up N via the canopy, assuming that the environment and chemical composition of the atmosphere is favourable for atmospheric N deposition (**II**, **IV**).

Near-perfect resource allocation between the canopy and roots is assumed in the *functional equilibrium* -hypothesis (Brouwer 1963; Lambers 1983; van Noordwijk and Dewilligen 1987). The applicability of the concept as an existing property of nature is supported by studies showing that trees growing in nutrient-poor environments have a higher fraction of roots compared to foliage than trees growing in more fertile environments (Ericsson 1995; Helmisaari et al. 2007). Assuming N as a minimum nutrient, functional equilibrium means that if the ratio between C and N is too high, more resources are allocated belowground for root growth, root maintenance and carbohydrate exudates to obtain more N. If the ratio is too low, more of the resources are allocated to growing chlorophyll in the foliage to assimilate more carbohydrates. In functional equilibrium, trees are close to a perfect compromise between options 1 and 4 described above. The concept of functional equilibrium implies that trees can sacrifice one nutrient, mostly C, to increase the availability of another nutrient such as N.

2.5.5. Effect of nitrogen availability to carbon sequestration

Atmospheric N deposition has increased the availability of N in boreal and temperate forests and this has been found to increase carbon sequestration to the forests. The importance of the effect has been debated, but recent literature estimates the increase to be between 10 and 100 kg (C) kg^{-1} (N) in boreal and temperate forests (de Vries et al. 2009; Liu and Greaver 2009; Thomas et al. 2010; Erisman et al. 2011; de Vries et al. 2014; Schulte-Uebbing and de Vries 2018). However, much higher effects have been proposed previously (Magnani et al. 2007) and certain studies question the importance of the effect (Gundale et al. 2014; Fernandez-Martinez et al. 2017). Based on fertilization experiments, the effect was approximately 30 kg (C) kg^{-1} (N) for Scots pine in Sweden (Hyvönen et al. 2008).

Janssens et al. (2010) lists mechanisms of increased Nr availability reducing soil respiration, such as a decrease in belowground allocation by trees, a decrease in the activity of soil microbes, a decrease of the priming effect, a decrease in saprotrophic microbial communities leading to lower decomposition rates, and increased stabilization of the soil organic material.

3. AIMS

The specific aims of this study were:

1. Forming an overall view of C and N cycling in boreal Scots pine forests (**I**, **II**)
2. Quantifying all the relevant N pools and fluxes in a boreal Scots pine forest ecosystem (**II**)
3. Understanding how boreal Scots pine trees take up, use and recycle N (**II**, **III**, **IV**)
4. Comparing N cycling in boreal Scots pine forests to N cycling in other forests with higher N availability (**III**, **IV**)

4. MATERIALS AND METHODS

4.1. Measurement sites

The main site of this study represents an N-deprived Scots pine-dominated forest at the SMEAR II station (Hari and Kulmala 2005) in Hyytiälä, southern Finland. The forest was regenerated in 1962 by sowing after clear-cutting, prescribed burning and soil preparation. The soil is haplic podzol, and soil depth varies from 0 to 200 cm. The organic soil layer is approximately 5 cm thick. The measurement station was established in 1995. At the time of the measurements (1998–2010), the site represented a medium fertile even-aged young boreal forest. Part of the forest was thinned in 2002 (Vesala et al. 2005). The measurement site is divided into two overlapping areas. The meteorological footprint area is the larger of the two, covering a circle with a 200-m radius (in total approx. 126 000 m^2). Most of this area is even-aged Scots pine forest, but approximately 14% of the tree basal area is Norway spruce and 12% is broadleaved trees (Ilvesniemi et al. 2009). The intensive measurement area is ca. 1200 m^2, formed by two micro-catchments that only receive water through precipitation (Ilvesniemi et al. 2010). The measurements used in papers **II**, **III** and **IV** were conducted at the intensive measurement area, whereas measurements used in paper **I** represent the meteorological footprint area. For more details about the areas and timing of the measurements used in this study, see Table 1.

Boreal Scots pine sites in Hyytiälä located in Dfc climate in the Köppen-Geiger climate classification were compared to two temperate oceanic sites located in Cfb climate (Fig. 5; Peel et al. 2007). The Danish Sorø European beech (*Fagus sylvatica* L.) site has similar precipitation as the Hyytiälä site. The Douglas fir (*Pseudotsuga menziesii* (Mirb.) Franco) site in Speulderbos (often referred to as Speuld), the Netherlands, shares a similar age, leaf type and leaf longevity as the Hyytiälä forest. The Sorø forest was planted with European beech in 1921 and is managed by thinning various fractions of the forest by 20% every 10th year, thinning averaging 2% per year (Pilegaard et al. 2011). The Speulderbos forest was planted with two-year old Douglas fir seedlings in 1962 (Su et al. 2009) and was thinned in the winter of 1995–1996 (Raat et al. 2010). The temperate sites receive high levels of atmospheric N deposition compared to the boreal site. See Tables 2 and 3 for characteristics of the physical and ecological aspects of the forests, respectively. In this study, the Finnish Hyytiälä site is referred to as the boreal Scots pine forest, the Danish Sorø site is referred to as the temperate European beech forest and the Speulderbos site is referred to as the temperate Douglas fir forest.

4.2. Evaluating the overall nutrient cycling using process-based modelling

Dynamic process-based models are a tool for investigating complex systems and the level of existing knowledge and understanding of these systems. A combination of process-based models were used to model N and C cycling at the boreal Scots pine-dominated Hyytiälä site. Ecological cycling was modelled with PSIM (Physiological SImulation Model; Grote 2007) and geochemical processes with modified DNDC (DeNitrification-DeComposition; Li et al. 1992). The models were coupled by modelling the framework MoBiLE (Modular Biosphere simuLation Environment) (Grote et al. 2009a; Grote et al. 2009b; Holst et al. 2010). Landscape-DNDC is a model developed further based on MoBiLE (Haas et al. 2013).

Figure 5. The location of the measurement sites in the northern part of Europe. Hyytiälä represents a boreal Scots pine forest in Finland, Sorø represents a temperate European beech forest in Denmark and Speulderbos represents a temperate Douglas fir forest in the Netherlands.

Table 1. Data used in the manuscripts of this study. In Hyytiälä, data were collected from two areas, the smaller catchment area being part of the larger circular footprint area with a 200-m radius. In manuscript **II**, some data measured in the campaign measurements were from different years.

Paper	I	II	III	IV
Hyytiälä, Finland data years	1998–2007	2006–2010	1999–2010	2008–2009
Hyytiälä catchment (0.12 ha)	X	X	X	X
Hyytiälä footprint (12.6 ha)	X			
Sorø, Denmark				X
Speulderbos, The Netherlands			X	X
All tree species	X	X		
Main tree species	X		X	X
Continuous measurements	X	X	X	
Campaign measurements		X		X
Process-based modelling	X	X		

MoBiLE was parametrized based on soil survey and vegetation inventories. Meteorological data were used to run the model, and the results were compared against flux measurements conducted by chambers and eddy covariance. Certain improvements were made to the model such as implementing a phenology model that has been tested to work well with the Hyytiälä forest (Mäkelä et al. 2004). The patchiness of the tree species cover at the 200-m meteorological footprint area was tested using three simulation setups: 1) one tree species (Scots pine), 2) three individual simulations with each tree species separately (Scots pine, Norway spruce and silver birch (*Betula pendula* Roth.)) and 3) one simulation of mixed forest with all tree species combined.

4.3. Continuous measurements in a boreal Scots pine forest

Continuous year-round measurements of relevant material and energy flows have been measured at the SMEAR II station in Hyytiälä since 1995 (Hari and Kulmala 2005) and were extensively used in this thesis. In this study, the boreal forest ecosystem was defined to consist of everything between the canopy top down to the soil depth where the deepest plant roots reach, excluding the bedrock.

Biomass accumulation in trees was based on biomass inventories. For the inventory, the stem shapes of the sample trees were measured and modelled based on breast height diameter and other variables. Breast height diameter was periodically measured from all the trees in the intensive measurement area, and other variables used to model the biomass fractions were based on the measurements of sample trees. Biomass inventories in the 200-m radius footprint area were based on periodic measurements on sample trees.

Table 2. Physical environment of the measurement sites (**II**; **IV**; Raat et al. 2010; Pilegaard et al. 2011).

Site name	Hyytiälä	Sorø	Speulderbos
Country	Finland	Denmark	the Netherlands
Coordinates	61° 50′ N, 24°17′ E	55° 29′ N, 11° 38′ E	52° 15′ N, 5° 45′ E
Climatic region	boreal (Dfc)	temperate oceanic (Cfb)	temperate oceanic (Cfb)
Mean T (°C)	2.9	8.6	9.4
Mean annual precipitation	709	730	900
Altitude from sea (m)	181	40	52
N deposition (kg N ha^{-1})	7.4	20	42 (throughfall)
Soil	Haplic podzol	Orthic podzol	Oxyaquic hapludalf

Table 3. Ecological characteristics of the measurement sites (IV; Su et al. 2009; Launiainen 2010; Pilegaard et al. 2011; Weligepolage et al. 2012).

Site name	Hyytiälä	Sorø	Speulderbos
Tree species	Scots pine	European beech	Douglas fir
Leaf morphology	coniferous	broadleaf	coniferous
Leaf type	evergreen	deciduous	evergreen
Stand age in 2009 (yr)	47	88	47
Dominant stand height (m)	16	26	32 (in 2006)
Leaf longevity (yr)	2.7	0.5 (1)*	2.6
Included in papers	I, II, III, IV	IV	III, IV

*Average time span of complete canopy renewal is one year for European beech and the same as the longevity for the studied Scots pine and Douglas fir trees.

Assimilation of CO_2 to the ecosystem was measured with the eddy covariance method, as described in Vesala et al. (2005). In this method, air gas concentrations and 3-dimensional wind velocity were automatically measured approximately 10 times per second. This method results in NEE of CO_2 between the forest and the atmosphere in a time scale of approximately 30 minutes. Net ecosystem exchange may be separated into GPP and TER, for example by utilizing existing knowledge of the temperature dependency of respiration (Kolari et al. 2009). Eddy covariance measurement gives the overall fluxes from a relatively large area, whereas most of the other measurements used in this study are based on a much smaller but more intensively studied area.

Nitrogen deposition to the system was based on bulk-deposition measurements and measurement-based modelling of dry deposition (Flechard et al. 2011). Bulk deposition includes wet deposition and some dry deposition. Wet-only deposition was estimated based on the method described in Korhonen et al. (2012). Altogether, the result was a measurement-based estimation of nine deposition fractions that may simply be summed up for total deposition. This represents the best knowledge of the actual total N deposition to the forest because dry deposition is not counted twice. Organic deposition was also measured to get the best possible measurement-based estimation of the total N deposition to the forest. Bulk deposition was measured above and below the canopy. This allowed the measurement of canopy N interception, along with studying the transformations of N in the canopy.

Hyytiälä station has been established on two microcatchment areas over granite bedrock. Discharge water is forced to exit these areas via two weirs. Waterflow was automatically measured at both weirs, and the nutrient contents were sampled daily when flowage passed through the weirs. Nutrient output in the discharge was based on these measurements.

Gaseous emissions of N_2O and NO were measured based on automatic and manual chamber measurements (Pihlatie et al. 2007). In this method, gas concentration change over time is measured in a chamber closing a known area and volume of the soil. NO fluxes were measured using three automated dynamic chambers in 2011. Nitrous oxide fluxes were measured typically twice a month from six manual chambers. Gas samples were collected from the chamber airspace during approximately 30-min enclosure and were later analysed using a gas chromatograph (Pihlatie et al. 2007).

Many processes of internal N cycling within the forest, such as N allocated for growth, N resorption and N uptake by plants, could not be directly measured. These processes were estimated based on mass balance.

4.4. Litter fall measurements

Tree litter fall was measured at Hyytiälä and Speulderbos based on litter traps that were collected approximately once a month. The litter was separated into compartments, weighed and an elemental analysis was conducted of the compartments. As a result, a time series was constructed of litter fall quantities of various components. These data were compared to meteorological data in paper **III**. Based on the time series, we calculated the C and N levels in annual litter production. These values represent the nutrient input to soil along with the nutrient levels that trees lose annually because of dying tissue.

4.5. Physiological activity and degree of leaf senescence

Leaves of the dominant tree species from Hyytiälä, Sorø and Speulderbos forests were collected once or twice a month in 2008 and 2009 from the canopy top and base. The samples were immediately stored in liquid N and dried for analysis. Carbon, N, chlorophyll A and B, bulk tissue NH_4^+ concentrations and pH were measured from the samples (**IV**). Chlorophyll concentrations were used as a proxy of physiological activity and degree of senescence. The ratio between bulk foliar concentrations of NH_4^+ and H^+ (Γ) was calculated. It was used as an indicator for NH_3 emission potential (**IV**).

4.6. Description of the methods used in the articles

Article **I** is a combination of theoretical process-based modelling, data analysis of measured field data from the boreal Scots pine forest and interpretation of the model results and the measured data and theoretical model development. This manuscript combines C, N and energy cycling in a boreal forest.

Article **II** is a combination of extensive long-term field measurements, data analysis and mass balance calculations. This manuscript is solely concentrated on N cycling and storages in the boreal Scots pine forest.

Article **III** concentrates on a single process in internal N cycling, i.e. litter fall. It is based on field measurements of two coniferous forest sites, data analysis and data interpretation. The study is relevant for understanding nutrient cycling, especially C and N.

Article **IV** concentrates on physicochemical properties of tree canopy related to canopy N uptake and plant strategies in conserving and recycling N. This article is based on laboratory analysis of the leaves of dominant tree species from three forest sites with various tree species and different N deposition.

The data used in the articles are very briefly shown in Table 1.

5. RESULTS AND DISCUSSION

5.1. Nitrogen balance of a boreal Scots pine forest

Nitrogen cycling and main N storages in a boreal Scots pine forest from the perspective of Scots pine trees are presented in Figure 6. Nitrogen accumulation to the ecosystem equals the atmospheric N deposition and biomass N increase. This means no change in the soil N storage size.

5.1.1. Nitrogen storages

The total N storage in the studied 46–48-year-old boreal Scots pine forest at the SMEAR II station in Finland was in the order of 2000 kg N ha^{-1}. The vast majority (almost 90%) of this was bound to the soil matrix. Mineral nutrient storage in the soil (0.3 kg N ha^{-1}) was ca. four orders of magnitude lower than the whole soil N storage. The ammonium-N level (NH_4^+-N; 0.3kg N ha^{-1}) was two orders of magnitude higher than that of nitrate-N (NO_3^--N; 0.005 kg N ha^{-1}), which in turn was approximately one order of magnitude higher than the nitrite-N level (NO_2^-–N; **II**). Nitrogen availability is known to generally be limited in boreal forests and plants can only use mineral or at most very small organic N compounds, and therefore most of the N in a forest is in a form that is not available for plants.

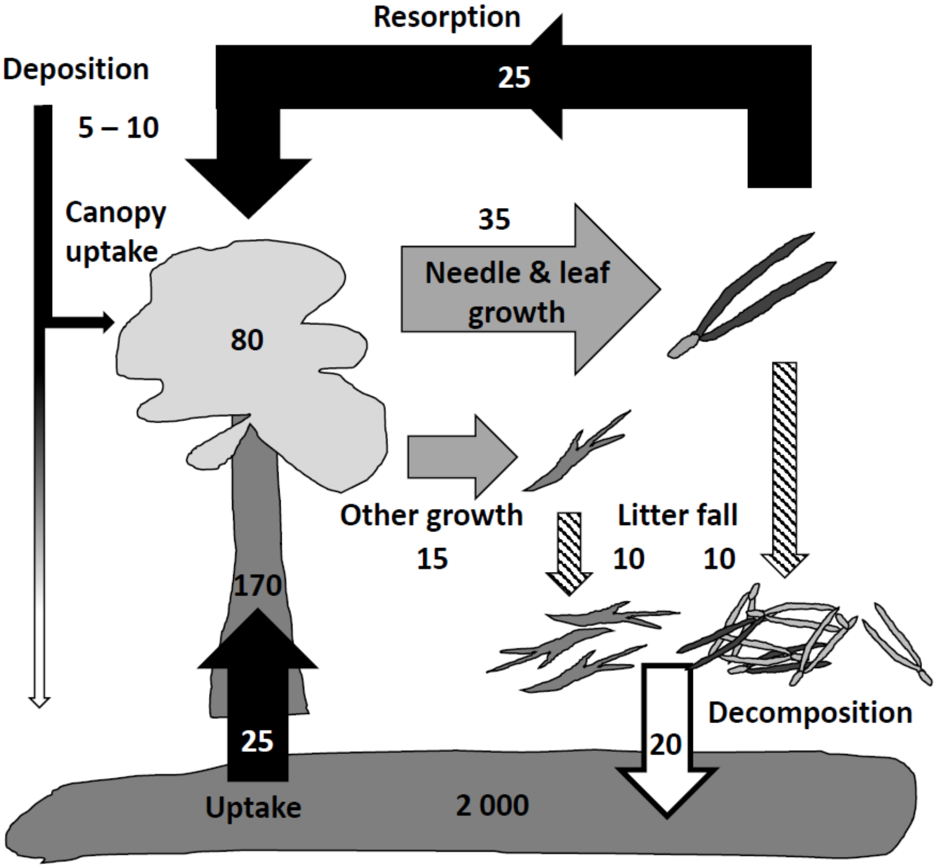
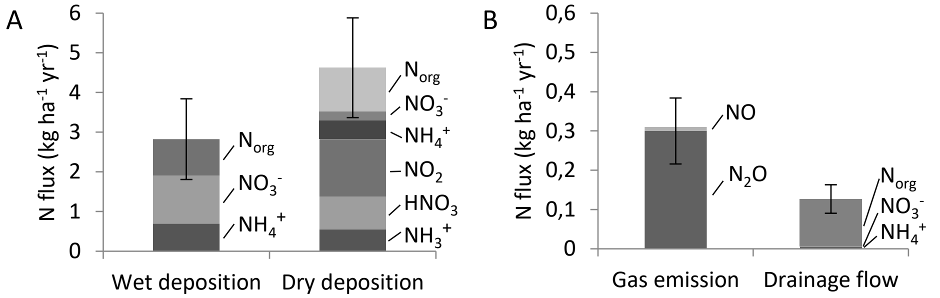


Figure 6. Simplified image of nitrogen (N) cycling and main N storages in a boreal Scots pine forest from the perspective of Scots pine trees. The unit is kg N ha^{-1} yr^{-1}, except for storages of foliar biomass N (80), other biomass N (150) and soil N (2000), which are all in kg N ha^{-1}. White arrows: input to soil, black arrows: input to growth pool, grey arrows: growth, arrows with diagonal pattern: litter fall. For a more detailed image, see publication **II**.



Figures 7A and **7B**. Annual nitrogen (N) inputs (A) and outputs (B) from the ecosystem. Note that the vertical scale is 10-fold in Figure A compared to Figure B. Modified from paper **II**.

The accumulation rate of N to the boreal Scots pine forest was estimated at 5 to 10 kg N ha^{-1} yr^{-1}. This coincides with the accumulation of N in living plant biomass, including wood (7.5 kg N ha^{-1} yr^{-1}). Soil N storage seemed to remain rather unchanged (-1 +/- 8 kg N ha^{-1} yr^{-1}) although the uncertainty is relatively high (**II**).

Nitrogen storage in plant biomass (250 kg N ha^{-1}) was ca. one order of magnitude lower than in the soil. Approximately 25% of this storage was structural N in wood and ca. 30% was active N in the foliage (**II**). Based on the results of Zechmeister-Boltenstern et al. (2011), soil microbial biomass in the studied boreal Scots pine forest was estimated to be roughly 50 kg N ha^{-1}. This means that the ratio between N in living biomass to N in dead biomass is around 1:7. Assuming that forest soil N storage does not markedly change during succession (see below), younger forests generally have a lower, and older forests a higher living to dead biomass ratio.

5.1.2. Nitrogen inputs

Nitrogen inputs to the boreal Scots pine forest are presented in Figure 7A. Nitrogen inputs to the forest ecosystem consist of atmospheric N deposition and biological N_2 fixation. The latter was not measured but it was estimated to contribute between 0 to 25% of the total N input. The measurement-based estimation of total atmospheric N deposition ranged from 5 to 10 kg N ha^{-1} yr^{-}1 (**II**). The value is slightly higher than usually reported, although the reported values are typically not comparable. This is because often 1) organic deposition is not measured, 2) dry deposition is not measured or estimated and 3) so-called bulk deposition is reported. Bulk deposition is a value that is relatively easy to measure and it includes wet deposition and some dry deposition. Because bulk deposition already contains some of the dry deposition, they cannot simply be summed up to get total N deposition. Deposition is sometimes measured below the canopy, which typically underestimates the total deposition. An atmospheric deposition rate of 5 to 10 kg N ha^{-1} yr^{-1} is considered elevated, yet still relatively low.

5.1.3. Nitrogen outputs

Nitrogen outputs from the boreal Scots pine forest are presented in Figure 7B. Total N outputs from the system were low compared to both the storages and inputs. The total measured output from the system was ca. 0.3 kg N ha^{-1} yr^{-1} (**II**). Denitrification is the main process responsible for N outputs from the ecosystem and it requires anaerobic conditions. Although the forest soil at the measurement site is quite porous and water penetrates the soil relatively easily, it is till, i.e. a mixture of various particle sizes. This allows microsites in the soil to be filled with water and anaerobic conditions to occur at small scales enabling denitrification. However, the low NO_3^- levels compared to NH_4^+ suggest that denitrification is not a dominant process in the soil of the studied boreal Scots pine forest.

Nitric oxide (NO) emissions were low, approximately 0.01 kg N $ha^{-1}yr^{-1}$, but N_2O emission was the most important measured single N output from the ecosystem, 0.2 kg N ha^{-1} yr^{-1}. Dinitrogen was not measured but is estimated to range from 0 to 2 kg N ha^{-1} yr^{-1}, which happens to be in the same range as the estimation of N input in N_2 fixation (**II**). The relatively low NO emissions are attributed to low nitrification rates, which are often, but not always linked to high soil acidity (low pH) (De Boer and Kowalchuk 2001).

Water flows from the system were another N output from the system. Organic N was the main form of N in this form, ca. 0.1 kg N ha^{-1} yr^{-1}. The combined output of NH_4^+, NO_3^- and NO_2^- was at least two orders of magnitude smaller (**II**).

Overall, the low outputs from the system are associated with the low levels of organism-available N, meaning that organisms take up almost all available mineral N before it can escape to the atmosphere.

5.2. Controlled and uncontrolled litter fall

The total average annual litter fall, including needles, branches etc., was approximately 2900 kg ha^{-1} and 4300 kg N ha^{-1} in the boreal Scots pine forest and temperate Douglas fir forest, respectively (**III**). Needle litter fall was the main difference between the forests. Needle litter production was more than double in the Douglas fir forest. The Scots pine forest produced ca. 10 times more bark than the Douglas fir forest but otherwise the litter production was comparable. Foliar litter fall was the most important single source of litter in both forests (**II**, **III**). The C concentration of litter was approximately 50% in both forests (**III**). In the boreal Scots pine forest, when both broadleaved litter and large branches were taken into account, foliar litter fall contributed to half of the total litter fall in terms of mass N and C (**II**, data not shown). In the Douglas fir forest, over 80% of the N in litter fall originated from the needles.

Most autumnal foliar litter fall is controlled in the boreal Scots pine forest, as part of the nutrients are being resorbed (in senescence) from the needles and leaves before they shed. This was observed visually from the colour of the foliar litter and from the litter N content during the year. Nitrogen resorption efficiency in boreal Scots pine trees was high, averaging 71%, including needle litter that was shed while green. The more foliar litter fall is controlled, the more N the tree is conserving. Therefore, controlled shedding of litter in the autumn is clearly advantageous for trees with limited N availability.

Contrary to the autumnal foliar litter fall, premature abscission (uncontrolled litter fall) is responsible for a significant part of the total litter fall (**II**, **III;** data not shown). In premature abscission, all of the nutrients in the leaves or needles are lost, at least temporarily. In the controlled case, approximately two-thirds of N is retained in the tree. Losing three green needles by premature abscission equates to losing as much N as is needed to grow two new needles the next year. Because needle longevity is relatively high (2.7 years for Scots pine in

Hyytiälä; **IV**), the effect may last for several years. Therefore, if the fraction of premature abscission increases, it may have severe effects on tree productivity.

The N utilization of trees was estimated based on the net increment of tree biomass and the quantity of N needed to replace dying tissue. The net increment of aboveground N in the tissues of trees in the boreal Scots pine forest was 7.5 kg N ha^{-1} yr^{-1}, whereas approximately 43 kg N ha^{-1} yr^{-1} of plant biomass turned into litter, mainly into needles and branches. This gives a net use of N of 50 kg N ha^{-1} yr^{-1}**.** Eighty-six per cent of this was used to replace dead tissue, and only 14% was used for actual growth (**II**, Fig. 8). This means that a 14% increase in foliar litter fall would lead to a situation with insufficient N levels for new tree growth. The net increment of biomass N coincides with the N accumulation rate to the ecosystem (Fig. 8).

Premature abscission is related to harsh environmental conditions such as storms, early frost in autumn or heavy snow (**III**). During these conditions, plants lose both their functioning tissue, structural nutrients, such as C in woody tissue and the nutrients that could have been resorbed. Therefore, climatic factors causing premature abscission may strongly decrease tree growth.

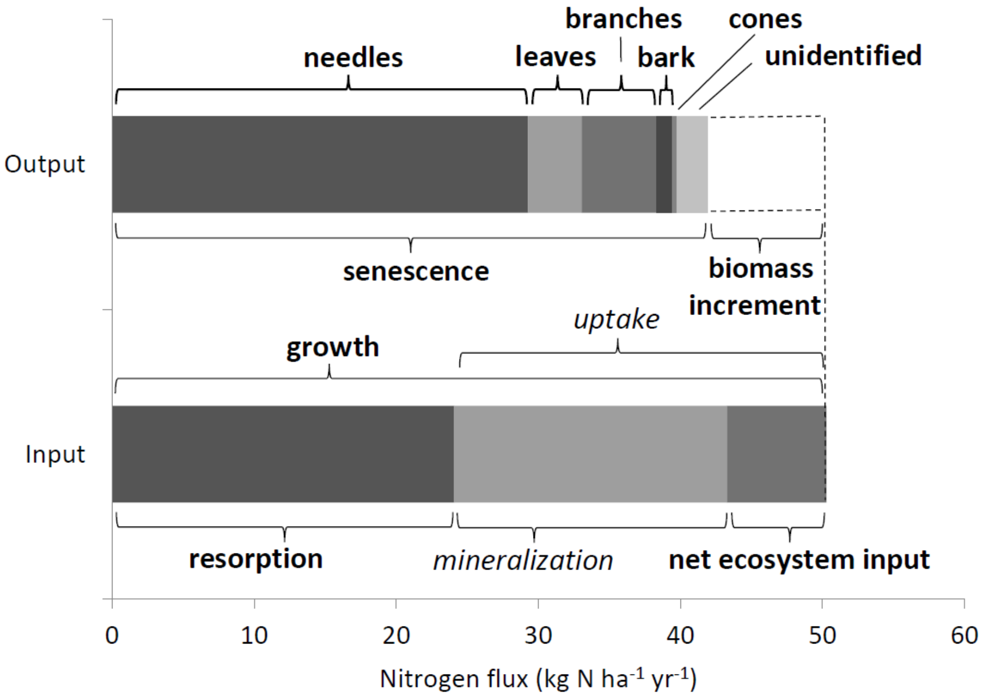


Figure 8. Annual inputs and outputs of nitrogen (N) from the perspective of trees in a boreal Scots pine forest. The bar shown with dotted lines represents the annual increment of N in the biomass. The variables in bold are primary measurements and the ones in italics are calculated from the other variables based on mass balance as follows: Growth = Senescence + Biomass increment; Uptake = Growth – Retranslocation; Release from soil = Uptake – Net ecosystem input; Net ecosystem input = Deposition – Gaseous emissions – Drainage flow. Modified from paper **II**.

Nutrients lost from dead foliage are not lost forever. Nutrients from dead needles and leaves may be mineralized for plant use in decomposition. Mineralization is supposedly more rapid for green needles and leaves compared to brown needles and leaves, as larger fractions of the nutrients in fresh foliage are in mobile form. However, a significant fraction of labile mineral N deposited to the forest floor is reportedly not directly used by plants (Nadelhoffer et al. 1999). Strong winds and heavy snow events may potentially cause oscillation of the needle biomass and photosynthesis, because if a large quantity of needles is lost one year, a supposedly larger quantity of plant-available N will be available in the soil for uptake the following year. A connection between needle litter fall and tree growth based on empirical observations was observed in Lehtonen et al. (2008).

5.3. Dynamics of litter fall and foliar nitrogen concentration

The quantity and constituents of the litter fall varied throughout the year in the boreal Scots pine forest, and peaked in autumn (**III**). The litter fall from December to March was relatively small. Branch litter fall was highest from January to May, cone litter fall was highest in June and July, bark litter fall was highest in spring and summer and foliar litter fall was highest in September and October.

At the temperate Douglas fir forest, cone litter fall was highest in June, whereas needle litter fall had two peaks, in May and October (**III**). The seasonal pattern of branch and cone litter fall was similar to the Scots pine forest and no major variation was observed in bark litter fall.

The N concentration of litter varied during the year in boreal Scots pine and temperate Douglas fir forests. In the boreal Scots pine forest, N content was highest in the late summer, whereas the concentration in the temperate Douglas fir forest was highest in winter (**III**). On the other hand, the N content in the green needles was lowest in late summer in the boreal Scots pine forest, while no systematic variation was observed in the temperate Douglas fir forest. The N content in the deciduous temperate European beech forest was rather constant after the leaves were fully developed but the N content dropped in autumn (**IV)**.

5.4. The role of canopy nitrogen uptake in boreal forests

Over half (4.4 of 7.4 kg N ha^{-1} yr^{-1}) of the N atmospheric deposition was retained in the canopy, defining the upper boundary for canopy N uptake. The fate of this N remains unclear, but canopy N uptake by trees is a potential option and this has been reported for coniferous trees (e.g. Wortman et al. 2012). The upper limit of canopy N uptake would be that all the N retained in the canopy is taken up by trees. This corresponds to approximately one-fourth of the total N uptake of the trees.

Epibiont biomass is one possible sink for N retaining in the canopy. Epibiont biomass in the forest has not been estimated, but based on visual observations, trees are not covered by epibionts, with the exception of certain lichen in the lower parts of tree stems. Unidentified litter fall of 2 kg N ha^{-1} yr^{-1} was measured on the ground. The N concentration of this litter was relatively high, approximately 1.2%, which is in the N concentration range reported for lichen (Johansson et al. 2011). Because no significant numbers of epibionts were observed in the canopy, N accumulation to epibionts cannot be large. This means that if epibionts were a major sink for N retaining in the canopy, the turnover rate of this N would be rapid. This would mean that before trees have access to this N, the N immobilized by epibionts would need to be remobilized by soil microbes.

If we assume that epibiont biomass is somewhat constant and that this N originates from epibionts, we obtain an estimation that N uptake by trees is ca. 3 kg N ha^{-1} yr^{-1}. In the N balance calculation (**II**), the unidentified litter was assumed to originate from the trees. If it actually originates from epibionts, both the N uptake level by plants and the quantity of litter produced would be slight overestimations. It is also possible that N is lost back to the atmosphere in gaseous form. Tree stems and shoots have recently been reported to potentially act as sources (Machacova et al. 2016; Lenhart et al. 2019; Machacova et al. 2019) but sometimes also as sinks (Machacova et al. 2017) of atmospheric N_2O. The gaseous loss of NH_3 from the leaves was concluded to be insignificant due to the low ratio between bulk foliar concentrations of NH_4^+ and H^+ (Γ) (IV; Flechard et al. 2013).

5.5. Strategies to cope with low nitrogen availability

Allocating less N to the foliage is an important way to conserve N. This depends on the N concentration of the foliage and on the total foliage biomass. In the boreal Scots pine forest, needle N concentration increased from 1% in winter to 1.3% in the growing season, while averaging 1.2%. This is low compared to the rather constant N concentration of 2% in Douglas fir needles and 2.6% in European beech leaves in midsummer (**IV**). The total maximum N storage in the foliage was similar to the boreal Hyytiälä Scots pine forest and temperate European beech forest in Sorø, 70 kg N ha^{-1} and 62 kg ha^{-1}, respectively (**IV**), despite the different leaf types. The temperate Douglas fir forest in Speulderbos had a much larger N storage, 240 kg ha^{-1} (**IV**). The N concentration and total N content of the foliage reflects the N availability difference between coniferous trees, but a comparison of N content between the coniferous species and broadleaved European beech is not feasible without looking at other factors as well.

Boreal Scots pine trees conserved N by having relatively long-living needles, longevity averaging 2.7 years. This is similar to the longevity of 2.6 years in Douglas fir needles in Speulderbos. In contrast, the deciduous European beech forest in Sorø grows leaves every year, with longevity being approximately half a year and the time for complete canopy renewal being one year (**IV**). Leaf longevity is mostly dependent on tree species and does not necessarily mean that these results have anything to do with N availability.

Nitrogen resorption efficiency (measured from total litter fall) was highest in the boreal Scots pine forest and in the temperate European beech forest, being around 70% (**IV**). The resorption efficiency of the temperate Douglas fir forest was 37% (**IV**). Determined by canopy foliar mass, leaf longevity and resorption efficiency, the N flux in the foliar litter fall was by far the highest in the temperate Douglas fir forest, 58 kg N ha^{-1} yr^{-1}, compared to 18 kg N ha^{-1} yr^{-1} in the temperate European beech forest and to the modest 8 kg N ha^{-1} yr^{-1} in the boreal Scots pine forest (**IV**). Foliar litter fall defines the lower boundary for N uptake that is needed to maintain the canopy. The differences between the forests are drastic, and the results are very well in line with the N availability of the three forests.

In relative terms, the Hyytiälä Scots pine forest annually only needed to take up 11% of the N content of the foliage, whereas Douglas fir and European beech trees needed to take up much more, 24% and 30%, respectively. In absolute terms, the difference is even more drastic when taking the peak N content in the foliage into account.

The belowground allocation in the boreal Scots pine forest was a significant sink for C

(**I;** data not shown), as also seen from the data of Korhonen et al. (2009), suggesting that roughly 20% of the GPP was used in root and rhizosphere respiration. This is linked to N mineralization and uptake, as described in (Janssens et al. 2010). Root and rhizosphere respiration is reportedly as high as heterotrophic soil respiration in summer (Högberg et al. 2001; Bhupinderpal-Singh et al. 2003) but is smaller in temperate and boreal zones at an annual scale (Subke et al. 2006).

Canopy N uptake was at most 20% of the total N uptake by the trees and ca. 9% of the annual N use in the boreal Scots pine forests. It may have some importance in boreal Scots pine forests. Normally N uptake is associated with belowground C allocation. Both the N and C economy of trees are improved when N is taken up directly by the canopy. However, even if N is deposited to the ground, it is still in plant-available form, so that the exact benefit of canopy N uptake remains unclear. It seems possible that N retained in the canopy was immobilized by epibionts. Overall, N availability was reflected to the N use of trees, but certain traits, such as leaf longevity, set boundaries for acclimation to N availability.

5.6. Nitrogen availability and carbon sequestration

Based on functional equilibrium, the uptake of deposited N either by the canopy or roots means that less C needs to be allocated belowground. This partly explains the findings that atmospheric N deposition and increased Nr availability increase C sequestration. The allocation belowground means smaller root and rhizosphere respiration and less C exuded to symbiotic microbes. This in turn decreases heterotrophic respiration due to the reduced priming effect and may cause increased stabilization of soil organic matter, as explained in Janssens et al. (2010).

Root uptake was estimated at 26 kg N ha^{-1} yr^{-1}, the N mineralization rate was estimated at 19 +/- 7 kg N ha^{-1} yr^{-1}, whereas total N use by trees was 50 +/- 6 kg N ha^{-1} yr^{-1}. This means that N mineralization ranged from 23% to 52% of the total N use by the trees and shows that most of the N that plants use for growth does not immediately originate from N mineralization, emphasizing the importance of N resorption and N deposition. Furthermore, plants taking up organic N deflates the idea that N mineralization alone drives N cycling in boreal forests.

Resorption, atmospheric N deposition and canopy N uptake decrease autotrophic and heterotrophic soil respiration and control C sequestration to the forests. With the concept of functional equilibrium, quantitative estimations of these processes to C sequestration may perhaps be made. However, that is a topic for another study.

6. CONCLUSIONS

The thesis summarizes extensive research efforts towards a comprehensive N balance of a boreal forest ecosystem. The work demonstrates that it is possible to quantify most of the N and C storages and flows in the boreal Scots pine forest in Hyytiälä, Finland. Although directly measuring certain variables, such as root biomass increase or N deposition, was not practical or possible, these variables were estimated based on a combination of available measurements and modelling. Using various measurement methods to calculate the N balance challenges the comparability of the results and adds uncertainties, whereas process-based modelling may be used to evaluate the reliability of the measurements. Results based on process-based modelling provide confidence to the observations.

Without anthropogenic effects, the vitality of boreal and temperate forests are generally limited by low N availability. Industrial N_2 fixation and fossil fuel burning have increased and continue to increase Nr levels in the environment. This additional N affects the C and N cycling in forests. In certain areas, forests have become N-saturated, meaning that the excess N reduces their vitality. The N deposition of the studied temperate forests in Denmark and the Netherlands is so high that they could be N-saturated, but this was not observed in this study. The boreal Scots pine forest is clearly limited by N availability. This means that additional N increases forest vitality and increases the forest as a sink for atmospheric C. Boreal forest trees in a rapid growing stage absorb most of the additional N to their own biomass, meaning that the soil N storages seem to be somewhat unchanged.

By efficiently coping with low N availability, trees may absorb more solar energy and forests may act as stronger sinks for atmospheric C. Understanding how trees control both C and N cycling in the forests is essential for predicting how forests act as C sinks in the future. One of the key principles controlling these cycles is how trees cope with limited N availability.

Boreal Scots pine trees are experts at conserving and recycling N. The level of N stored in the canopy is relatively low because the canopy is not very dense and needle N concentration is relatively low. Furthermore, needles have relatively long lifetimes, meaning that the annual N demand for growing new needles remains at a moderate level. Boreal Scots pine trees are also very efficient at recycling N from the old needles before they shed to the ground. Approximately two-thirds of needle N is resorbed in the litter fall and this covers ca. half of the total N use of Scots pine trees.

Coniferous trees have adapted to saving N by growing needles with relatively long lifetimes. This happens even though they may have plenty of Nr available, as seen in the temperate Speulderbos Douglas fir forest. Deciduous European beech with relatively high N availability also conserve N by resorbing it from old leaves. Unlike boreal Scots pine trees, trees in the studied temperate forests do not markedly conserve N when it comes to growing canopies.

Boreal Scots pine trees use most of the N available for allocation for maintaining their structures and only a fraction is used for increasing their biomass. Because resorption is a major N source for trees, premature abscission may have a remarkable impact on forest growth. If heavy storm events and snow damages increase in the future, this may have a significant impact on the productivity of boreal forests and their ability to act as C sinks. Nitrogen is not easily lost from boreal forest ecosystems, but as Scots pine needles have a lifetime of several years, the effect of disturbances may be relatively long.

Atmospheric N deposition is challenging to assess. Based on novel methods that include organic N and only take dry deposition into account once, the N deposition in boreal forests seems higher than previously estimated. A significant part of the deposited N remains in the canopy. Lichen and other epibionts with high turnover rates appear to take up and immobilize this N after it is deposited to the ground.

When estimating the net mineralization rate of forest soil based on the mass balance (or top-down) approach, the net mineralization of soil N in N-limited forests is often assumed to be in the same order of magnitude as plant N uptake. Taking N resorption, N deposition, foliar N uptake and organic N uptake properly in account makes the estimates for net N mineralization rates needed to sustain plant growth markedly smaller. These estimations should be critically compared to other estimations of net mineralization rate, such as the ones based on soil CO_2 emissions or laboratory net mineralization measurements. Correctly determining the mineralization rate of soil is essential for estimating C sinks of boreal Scots pine forests in the future climate. A slower net mineralization rate may mean that boreal forests are less prone to turn into massive sources of atmospheric CO_2.

Considering future research, the sensitivity of forest productivity to abiotic damage from the perspective of N cycling could be studied further. Comparisons could be made of top-down and bottom-up estimations of mineralization rates of forest soils. Dinitrogen fixation and N_2 losses from forests could be studied further, as this could turn out to be important for also better quantifying the global N cycling. Atmospheric N wet deposition could be separated from bulk deposition measurements to conduct better estimation of the actual N deposition. In addition, the approach in this study may help quantify how much C sequestration increases with N deposition in N-limited ecosystems.

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